

The Early Evolution of Small
Terrestrial Bodies:
A Palaeomagnetic and Geochemical
Study of Meteorites



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Abstract

Meteorites act as windows into the early solar system, providing a wealth of information on the composition, formation, and evolution of planetary bodies. After their formation, planetary bodies are reworked by several secondary processes which modify their original chemical and physical properties. In this thesis, I study two such processes (aqueous alteration and impacts), using magnetic and geochemical techniques on stony-iron, iron, and chondritic meteorites.

The formation of stony-iron meteorites is a topic of intense research and both mesosiderites and pallasites are hypothesised to have formed through impact mixing of metal and silicates. I use palaeomagnetic and microscopy techniques on the mesosiderite Estherville and show that Estherville does not record a high-coercivity natural remanence. However, Estherville has the ability to record strong ($>150 \mu\text{T}$) fields. The slow cooling rates recorded by mesosiderites ($<1 \text{ K/Myr}$) imply a formation close to the core-mantle boundary, suggesting that they would have been exposed to strong fields if a dynamo was active. However, they do not carry a high-coercivity remanence, which may indicate that the mesosiderite-forming impact shut off the dynamo.

To investigate the formation of pallasites, I use *in situ* chemical analysis and find that all metal in Seymchan has the same origin. Based on this result and textural observations, I propose a pallasite formation model involving two or more impacts, and provide additional evidence for the

previously proposed link between Main Group pallasites and IIIAB meteorites.

Finally, I study aqueous alteration using quantum diamond microscope magnetic mapping to spatially resolve the magnetic signals of the CM chondrite MCY 05231. I suggest that magnetite forms via different reaction pathways in this sample, and that magnetite formed via pseudomorphic replacement of metal leads to the inheritance of magnetic remanence from the precursor grain. I use these results to suggest a magnetic history for MCY 05231, and to discuss implications for bulk magnetic studies of chondrites.

Extended Abstract

Meteorites act as windows into the early solar system, providing a wealth of information on the composition, formation, and evolution of planetary bodies, including terrestrial planets, planetesimals and asteroids. After their formation, planetary bodies are reworked by several secondary processes which modify their original chemical and physical properties. In this thesis, I present magnetic and geochemical data to advance our understanding of secondary processes affecting small terrestrial bodies in the early solar system. I focus on two types of processes (impacts and aqueous alteration) by studying stony-iron, iron and chondritic meteorites.

While impacts are known to have been very common in the early solar system, many details are missing from the formation of meteorites whose lithologies and petrological textures have been heavily modified by impact mixing (for example, stony-iron meteorites). Mesosiderites are a group of stony-iron meteorites containing fragments of crust, mantle, and core material. Their formation is debated, but it is generally understood that they require highly energetic impacts to mix such a diversity of material. I use palaeomagnetic techniques on the mesosiderite Estherville to investigate whether the parent body retains its differentiated structure. Given that low-fidelity samples are not well suited to analysis using traditional palaeomagnetic techniques, I also study the crystallography of the cloudy zone islands, to test the hypothesis that cloudy zone formation in a field causes a bias in the c-axis orientation of tetrataenite in the cloudy zone. I find that the mesosiderite cloudy zone contains crystals with uniform crystallographic orientation, and I suggest that this is due to the slow cooling rates of mesosiderites, causing all cloudy zone islands to become connected, and making it energetically difficult for them to align with a field. However, palaeomagnetic results indicate that Estherville can record fields stronger than $150 \mu\text{T}$.

Therefore, mesosiderites are able to record strong fields, and they were likely to have experienced them because of their slow cooling rate, implying formation at depth in a parent body. However, they do not carry a high-coercivity remanence, which may suggest that the mesosiderite-forming impact shut off the dynamo.

The second science chapter explores the formation of pallasites, the other stony-iron meteorite group. The pallasite meteorites are composed of olivine crystals in an FeNi matrix. Traditionally, they were thought to have formed at the core-mantle boundary, but palaeomagnetic evidence has shown that this is not the case. Many alternative formation mechanisms have been proposed since then. One such mechanism is partial differentiation followed by an impact event, which predicts different metal compositions preserved in the pallasites. I use *in situ* trace element analysis of the FeNi metal in the meteorite Seymchan to test this hypothesis, and I find no systematic compositional difference in the metal in Seymchan, which suggests that all metal has the same origin. I also measure a IIIAB iron meteorite (Henbury) and find compositional similarities to Seymchan, lending further support to the hypothesised genetic link between Main Group pallasites and IIIAB iron meteorites. To explain chemical and textural observations from this study and previous work, I propose a complex collisional history, involving 2 or more impacts on the pallasite parent body.

Finally, in the third science chapter I turn my attention to aqueous alteration, by studying chondritic meteorites. Chondrites are used to constrain the strength of the magnetic field in the nebula, which is thought to play an important role in disk dynamics and planetary formation. However, different magnetic carriers report different palaeointensity estimates, and recent work has highlighted the importance of understanding aqueous alteration and its effects on magnetic mineralogy in order to interpret the solar nebula palaeomagnetic record. I use QDM (quantum diamond microscope) magnetic mapping to spatially resolve the magnetic signals of the CM chondrite MCY 05231, as well as petrological observations to identify the corresponding magnetic sources in the sample. I find two uniform magnetisations in the matrix:

one carried by magnetite and pyrrhotite, and another carried by the primary metal. At the same time, the magnetite in the chondrules is randomly magnetised from one chondrule to another, but uniform within a chondrule. I suggest that the magnetite in MCY 05231 formed via different pathways, which influenced whether it inherited its magnetisation from a precursor grain or acquired a new remanence at the time of alteration. I provide evidence that magnetite formed via pseudomorphic replacement of metal may inherit the remanence of the parent metal grain, and I discuss implications for bulk magnetic studies of the solar nebula.

These studies highlight the importance of high-resolution, *in situ* techniques in the study of meteorites, whose complicated formation histories and intricate mineralogies and textures make them challenging, yet highly impactful targets for research.

Statement of Originality

I declare that this thesis is entirely my own work, and except where otherwise stated, describes my own research. All data associated with this thesis are included in the Oxford Research Archive.

Ana Pagu

October 2023

Acknowledgements

After learning all about the wonderful world of rocks during my undergraduate degree, I decided to do a DPhil out of a passion for rocks, space, and the very niche intersection between the two. Five years, one pandemic and many personal challenges later, it turned out to be a bit more than I had signed up for and it is difficult to imagine how I could have completed my DPhil without the support, guidance and understanding of the people who helped me along the way.

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Dedicated to my grandma
(1940 - 2019)

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Chapter 1

Introduction

The formation of terrestrial bodies is a topic of intense research in planetary science, and one of its ultimate goals is to understand the formation of the Earth and the origin of life. These questions have motivated the emergence of numerous multidisciplinary approaches to shed light on the early solar system. Using spacecraft observations, modelling, and meteorite studies, we are closer than ever to understanding how our solar system came to be. However, many questions remain open. In this thesis, I tackle some of these questions using multifaceted array of methods applied to the study of meteorites.

1.1 Planetary accretion

"The planetesimal theory is often portrayed as a sequence of steps rather like acts in a play. The characters and scenery change with time, and the audience applauds at the end of each act."

- Chambers (2004)

The planetesimal theory describes the formation of planets in three stages: formation of planetesimals, formation of planetary embryos, and formation of planets (Chambers 2004). Through a combination of collisions, surface-sticking (Dullemond & Dominik 2005), instability due to gas drag (known as streaming instability; Youdin & Goodman 2005, Johansen et al. 2007, 2015), and possibly gravitational instability

(Youdin & Shu 2002), the dust of the solar nebula forms small (1-1000 km) early solar system bodies known as planetesimals. Once planetesimals reach a few km in diameter, they are able to gravitationally perturb their neighbourhoods, and further collisions, aided by dynamical friction and gravitational focusing, lead to the formation of planetary embryos and eventually, planets (Thommes et al. 2003).

Following the analogy used by Chambers (2004), much like acts in a play, the different steps and processes involved in the formation of a terrestrial body are often portrayed as sequential, following one another with little to no overlap. However, unlike acts in a play, these processes overlap in both time and space, and sometimes the sequence changes or it is cut short – not all planetesimals collide to form planetary embryos (present-day remnants of planetesimals exist, as evidenced by asteroids, comets, and Kuiper belt objects; Weiss 2017), and the accretion of embryos to become planets is not always complete (e.g., Mercury and Mars; Asphaug & Reufer 2014).

These incomplete processes are very helpful in understanding the intermediate stages of planetary formation, and the secondary processes affecting planetesimals. Because of their large size, planets are thermally processed so much that it is difficult to understand their early evolution, as evidence of early processes had long been erased by subsequent processing. On the other hand, planetesimals often preserve more pristine material and evidence of incomplete processes (e.g., weakly altered chondrites, partially differentiated meteorites). Furthermore, because of their smaller sizes, collisions created meteorites which preserve core and mantle material (Section 1.2.1.1), providing a unique insight into planetary interiors. For these reasons, planetesimals offer a window into the processes operating during the early stages of planetary accretion and differentiation on bodies such as the Earth.

1.2 Planetary processes

After a terrestrial body forms, it is often affected by one or more processes reworking the parent body (e.g., differentiation, impacts etc.). Due to being widespread and

often acting at planetary scales, all meteorites are affected by at least one such process but more often a combination of several processes. As such, interpretations of primary features and/or the early solar system environment hinge on understanding the formation history of a meteorite. Here I present a broad overview of the most common secondary processes affecting early solar system bodies: magmatic differentiation, impacts, aqueous alteration, and planetary dynamos.

1.2.1 Magmatic differentiation

When a terrestrial body forms, it heats up primarily due to the decay of the radioactive material that it accretes (mostly short-lived nuclides such as ^{26}Al and ^{60}Fe ; Urey 1955). The amount of radiogenic material in a terrestrial body depends on the timing of formation, as early formation leads to the accretion of more radiogenic material which has yet to decay (Kleine & Wadhwa 2017). This radiogenic material decays within the body and leads to heating. In terrestrial bodies that accreted early, the heating results in widespread melting, and gravitational separation causes differentiation into a core, mantle and crust.

In smaller bodies such as planetesimals, full differentiation does not always occur due to the variable onsets of accretion (late accretion leading to less radioactive material and less heating) and their smaller sizes, as they may not accrete enough radioactive material and may not be able to retain radiogenic heat. Therefore, planetesimals experienced a significantly wider range of thermal histories than planets – while planets melted and differentiated completely, planetesimals exhibit a variety of intermediate melting states (Elkins-Tanton et al. 2011, Weiss 2017), allowing evidence of early processes to be preserved and studied.

1.2.1.1 Types of meteorites

Differentiation is a helpful context in which to understand the different types of meteorites. As fragments of asteroids, meteorites have traditionally been divided into four

groups: undifferentiated meteorites (chondrites), and differentiated meteorites sampling the crust (stony meteorites, also known as achondrites), core (iron meteorites) and a combination of core material and crust and/or mantle material (stony-iron meteorites) (Weisberg et al. 2006). This classification is based on petrography and chemistry.

Chondrites are disequilibrium mixtures of silicates and metal, which do not show signs of widespread melting. However, there is substantial evidence for aqueous alteration due to the accretion and subsequent melting of ice (e.g., Suttle et al. 2021; Section 1.2.3), and heating leading to thermal metamorphism (Tonui et al. 2014). Chondrites are further divided into 12 subtypes based on their chemistry, and some chondrites preserve the bulk non-volatile composition of the solar system (Russell 1929, Palme & Beer 1993, Lodders 2003). The most notable feature of chondrites are their chondrules, small (μm - to mm-sized) igneous particles formed in the solar nebula (Rubin 2000, Connolly Jr & Jones 2016). Chondrites also contain Calcium-Aluminium Inclusions (CAIs), which are widely considered to be the oldest objects to form in the solar nebula (4567.60 ± 0.36 Ma; Jacobsen et al. 2008). Because of the absence of melting and similarity to the bulk solar system composition, chondrites are believed to sample undifferentiated material. This material could originate from undifferentiated, rubble pile-type asteroids (e.g., C-type asteroids), which likely accreted later in the history of the solar system (as demonstrated by chondrule formation ages; e.g., Krot et al. 2009, Connelly et al. 2012) and contained too little ^{26}Al to drive melting and differentiation (Kunihiro et al. 2004). Alternatively, it has been proposed that some chondrites (e.g., CV chondrites; Carporzen et al. 2011) originate on partially differentiated planetesimals, which have differentiated interiors and unmelted, chondritic lids (Elkins-Tanton et al. 2011).

On the other hand, non-chondritic (differentiated) meteorites have undergone large degrees of thermal processing on the parent body. Achondrites are generally considered to sample the crusts and/or mantles of differentiated bodies, as they are

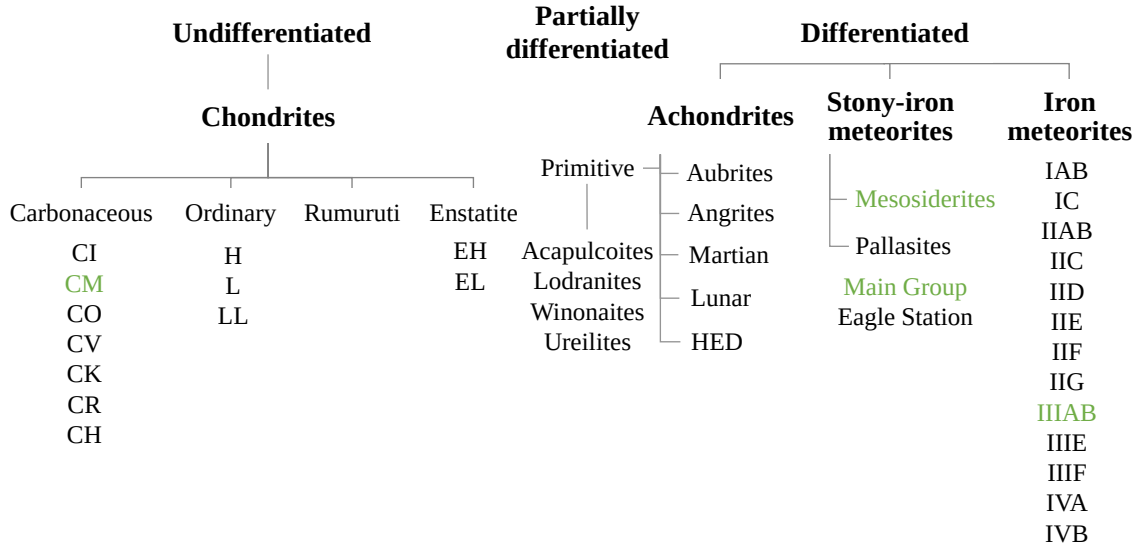


Figure 1.1: Meteorite classification, showing the meteorites studied in this thesis in green. This classification is based on petrological observations, not palaeomagnetic interpretations.

highly thermally processed and contain little to no metal. They are further subdivided into 9 subgroups, some of which originate on the Moon (Korotev 2005), Mars (Nyquist et al. 2001), and possibly the asteroid Vesta (McSween Jr et al. 2013).

Iron meteorites generally sample planetary cores and they are composed primarily of FeNi metal, although sometimes they also contain silicate inclusions of chondritic and achondritic material (Ruzicka 2014). They are further subdivided into 14 chemically distinct groups, based on their trace element concentrations (Goldstein et al. 2009). Initially, iron meteorites were classified in groups I-IV in order of decreasing Ga and Ge concentrations (Goldberg et al. 1951, Lovering et al. 1957). Subsequently, compositional analysis of other elements led to a further classification of A-G within each pre-existing (I-IV) subgroup (Wasson & Kimbrell 1967, Wasson et al. 1998). Approximately 15% of iron meteorites remain ungrouped (Goldstein et al. 2009).

Finally, there are two types of stony-iron meteorites (mesosiderites and pallasites), which contain approximately equal proportions of metal and silicates, and whose formation histories likely involved impact mixing (Section 1.2.2). This classification is schematically shown in Fig. 1.1.

The composition of iron meteorites, as well as other meteorite types, is firstly determined by the affinity of the constituent elements, which depends on the temperature at which the elements condense from the solar nebula. Following the Goldschmidt element classification (Goldschmidt 1937), elements are classified as lithophile, siderophile, chalcophile, or atmophile (Fig. 1.2). In the context of differentiated meteorites, iron meteorites tend to be richer in siderophile elements (elements which partition into FeNi metal), whereas achondrites are richer in lithophile elements (elements which partition into silicate minerals). Some iron meteorites and the metallic parts of stony-iron meteorites are thought to be fractionally crystallised (also known as magmatic; Wasson 1985), because they came from differentiated parent bodies. As their cores crystallised, the composition of the liquid metal evolved to become enriched in the elements that favour the liquid phase, and depleted in the elements that favour the solid. This leads to large compositional variations in some elements within the same meteorite group (e.g., Ir, a highly siderophile element that is incompatible in the liquid, shows four orders of magnitude variation within the IIIAB magmatic iron group; Goldstein et al. 2009). Finally, subsolidus partitioning between different phases in metal (kamacite and taenite) also influences the composition of the individual phases (Rasmussen et al. 1988, Mullane et al. 2004), but not the bulk composition.

In this thesis, I study both differentiated (stony-iron and iron) and undifferentiated (carbonaceous chondrite) meteorites (Fig. 1.1), spanning a broad range of lithologies and formation mechanisms.

1.2.2 Impacts

Due to the dynamic environment of the early solar system, impacts were common and their effects are seen in most meteorite groups (e.g., the stony-iron meteorites, silicate-bearing iron meteorites, and brecciated chondrites and achondrites).

In chondritic meteorites, impacts have been found to play an important role in compaction (Bland et al. 2014, Cambioni et al. 2021) and brecciation (Bischoff et al.

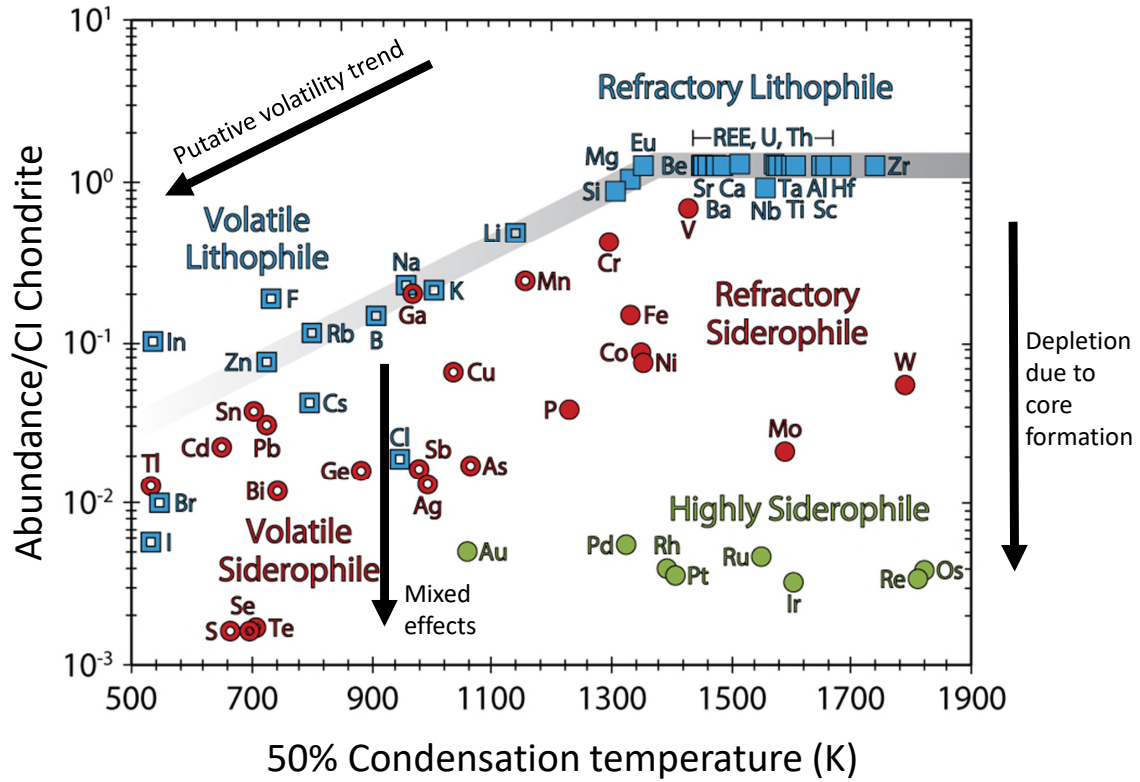


Figure 1.2: Abundance of elements in the bulk silicate Earth, plotted against the 50% condensation temperature (temperature at which 50% of the element condensed from the solar nebular gas) calculated by Lodders (2003) (figure adapted from Wood et al. 2019). The compositions are relative to CI chondrites (Palme & O'Neill 2014) and normalised by Mg. The elements are coloured based on affinity. Volatile elements have low condensation temperatures, whereas refractory elements have higher condensation temperatures. Three trends are annotated: core formation (leading to depletion in siderophile elements in the bulk silicate Earth, because siderophile elements partition into the core), increasing volatility (leading to depletion in the most volatile elements at low condensation temperatures, due to nebula dissipation by the time the condensation temperature is reached), mixed effects (both core formation and volatility lead to the depletion of volatile siderophile elements in the bulk silicate Earth).

2006). In differentiated meteorites, impacts have created a remarkable diversity in lithologies, in particular silicate-bearing iron meteorites (IIE, IAB, and IVA) and stony-iron meteorites (mesosiderites and pallasites). For silicate-bearing iron meteorites and mesosiderites, there is general agreement that impact-related processes led to metal-silicate mixing (e.g., Ruzicka et al. 1999, Ruzicka & Hutson 2010, Tomkins et al. 2013, Ruzicka 2014, Haba et al. 2019). The origin of pallasites has been subject to intense debate, with some formation models involving cumulate layers at the base of the mantle (Boesenberg et al. 2012), dykes extruded from the core during core crystallisation (Johnson et al. 2020), and different types of impacts (Yang et al. 2010, Kruijer et al. 2022, Walte & Golabek 2022), potentially linked with partial differentiation (Walte et al. 2020). The formation of mesosiderites and pallasites is explored in detail in Chapters 2 and 3.

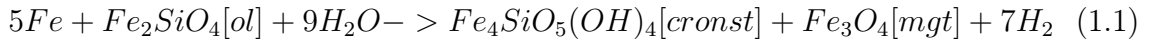
A type of collision with particular relevance to small planetary bodies is the hit-and-run collision (Asphaug et al. 2006), in which two bodies collide without merging. Instead, the smaller body escapes from the collision, often deformed and/or without its outer layers. This type of collision has been used to explain the diversity of planetary bodies in the solar system – hit-and-run collisions can produce mantle-stripped asteroids such as asteroid (16) Psyche (Asphaug 2017), and may have been responsible for the metal-silicate mixing that formed the mesosiderites (Haba et al. 2019). Hit-and-run collisions are common during accretion, having been estimated to make up 42% of all collisions (Chambers 2013).

1.2.3 Aqueous alteration

Aqueous alteration refers to the chemical and textural changes that a rock undergoes as a result of hydration. When planetesimals form at large heliocentric distances (beyond the snow line, assumed to be 5 AU; Cieza et al. 2016), they accrete ice as well as nebular gas and dust. Accreted ice melts due to heating from radioactive decay or impacts, and it is generally believed to be the source of the water that led to

the aqueous alteration widely experienced by carbonaceous chondrites (e.g., Grimm & McSween Jr 1989). Volatile-rich (C-type) asteroids are the most common asteroid type (42% of the asteroid population; DeMeo & Carry 2014), so aqueous alteration was likely a very common process in the early solar system.

For carbonaceous chondrites, it is generally agreed that the alteration took place on the chondritic parent body (McSween Jr 1979, Tomeoka & Buseck 1985, Trigo-Rodriguez et al. 2006), or in uncompact precursor planetesimals (Metzler 2004). Alteration proceeds in several stages, and starts with the alteration of anhydrous silicates (olivine and pyroxene) to serpentine (particularly cronstedtite, $\text{Fe}_4\text{SiO}_5(\text{OH})_4$), and alteration of primary metal and sulfides to magnetite, pyrrhotite and tochilinite ($2(\text{Fe},\text{Ni},\text{Cu})_{1-x}\text{S} \times n(\text{Fe},\text{Mg})(\text{OH})_2$, where x ranges between 0.08 and 0.28; Organova et al. 1974). Castillo-Rogez & Young (2017) describe the initial stages of alteration using the reaction:



where *ol* is olivine, *cronst* is cronstedtite, and *mgt* is magnetite. More advanced alteration produces increasingly higher-Mg serpentine (Castillo-Rogez & Young 2017, Suttle et al. 2021).

Aqueous alteration acted on volatile-rich planetesimals early in their history (2.3 – 8 Myr after CAI formation for CM chondrites, Suttle et al. 2021). Some aqueous alteration took place before the solar nebula dissipated (current estimates for nebula dissipation are 3.94 Myr after CAI in the inner solar system and 4.89 Myr after CAI in the outer solar system; Wang et al. 2017, Borlina et al. 2022), and since alteration can lead to the formation of magnetic minerals (magnetite and pyrrhotite), aqueously altered meteorites have been found to record the nebula magnetic field present during alteration. Although many details about the initial stages of accretion are still poorly constrained (Chambers 2004), it is likely that the dynamics of the nebular gas and

dust were influenced by the magnetic field of the protoplanetary disk (Bai & Goodman 2009, Bai 2017, Weiss et al. 2021), which likely had a role in early planet-building by influencing the distribution of gas and dust (Hayashi 1981). The nebula field may have also influenced the isotopic reservoirs in the early solar system (Kruijer et al. 2020), with consequences for the building blocks of planets. As a result, understanding the effects of aqueous alteration on the magnetic mineralogy of chondrites is a valuable piece of the puzzle in elucidating the early history of the solar system.

1.2.4 Planetary dynamos

Dynamo theory describes the process through which a rotating, convecting, electrically conducting fluid can produce and maintain a magnetic field over long time scales. In terrestrial planets, the fluid is usually the molten iron core, which cools through convection (Stevenson 2003).

Both thermal and compositional convection are believed to be capable of producing a field. Purely thermal convection takes place when hotter material rises buoyantly through the core, followed by sinking once it cools down (Weiss et al. 2010). Thermal convection requires a high heat flux to maintain, so it is likely to be short-lived (Sternberg & Crowley 2013). Compositional convection starts after the core begins to crystallise, because the crystallisation process excludes the light elements present in the core (e.g., sulfur, Chabot 2004), and they rise through the liquid core. This process was found to be highly efficient at driving dynamos over billions of years, for Earth-like planets (Nimmo 2009). While thermal convection dominates when the entire core is molten, compositional convection is believed to be the main driver for convection after the core begins to crystallise. It is also possible that there may be a period of dynamo quiescence between the two periods of convection, as suggested by Nichols et al. (2016, 2021). Based on thermal modelling and previous meteorite studies (Fig. 1.3), Bryson et al. (2019b) identify different magnetic epochs defined by the mechanism of dynamo generation (thermal or compositional). They find a low field

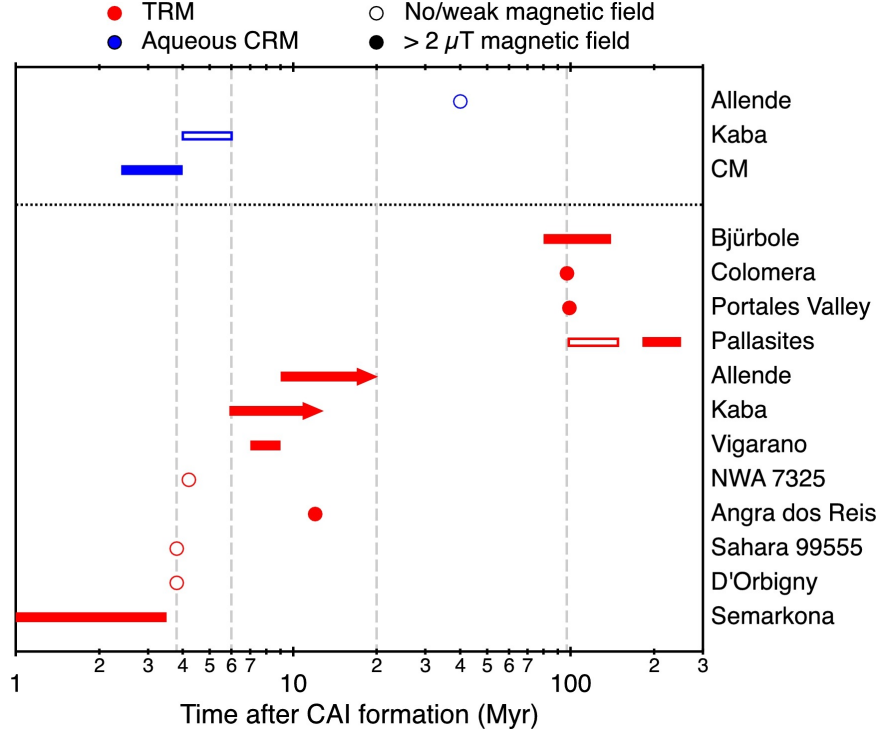


Figure 1.3: Asteroid magnetic record compiled from previous magnetic studies of meteorites (Bryson et al. 2019b and references therein). Grey dashed lines show epochs of high and low recovered palaeointensities.

epoch between 4 and 6 Myr (before thermal convection started) and between ~ 20 and 100 Myr after CAI (between thermal and compositional convection), although their model showed that the timing of dynamo activity depends strongly on the radius of the body.

Magnetic field generation depends on numerous factors, such as heat flux out of the core, parent body size, timing of accretion (thus influencing the extent of radiogenic heating), and the chemistry of the core. In turn, establishing the presence and intensity of a magnetic field at a certain time can provide valuable insight into the formation and evolution of early solar system bodies. Magnetic studies on meteorites offer a window into the early solar system, allowing us to understand the magnetic fields that were operating when the magnetic particles in meteorites acquired their remanence. Therefore, magnetic techniques and results from previous magnetic studies are used throughout this thesis to shed light on the early evolution of meteorite

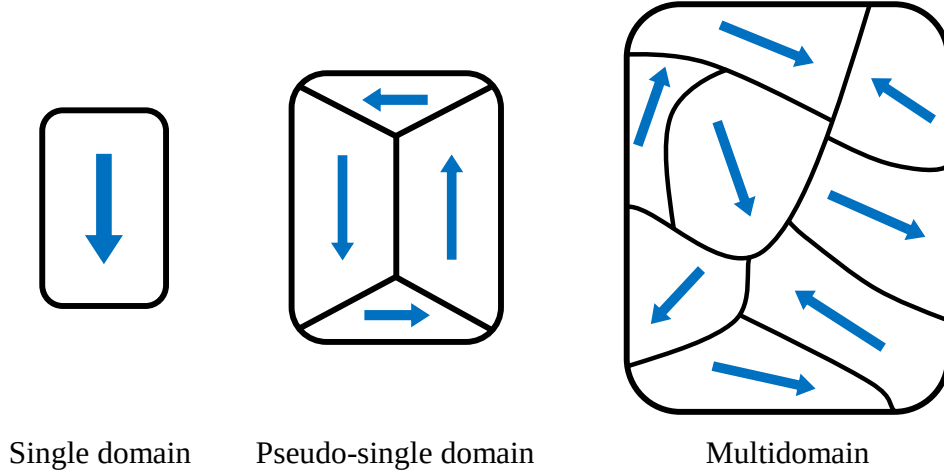


Figure 1.4: Schematic showing different magnetic domain states.

parent bodies.

1.2.4.1 Meteoritic magnetic mineralogy

Meteorites have been widely used for magnetic studies because of their abundance of iron and associated metal and sulfide minerals. Magnetic minerals often found in meteorites include: FeNi metals (kamacite, taenite, tetrataenite, awaruite), sulfides (pyrrhotite), phosphides (schreibersite), and oxides (magnetite, chromite). Of these, the only minerals that are ferromagnetic or ferrimagnetic at room temperature are the FeNi metals, schreibersite, pyrrhotite and magnetite (Gattacceca et al. 2014). The magnetic properties of these minerals are summarised in Table 1.1.

One of the main properties influencing whether a mineral is a reliable palaeomagnetic recorder is the size of the crystal, and its associated domain state. A magnetic domain is a region of a crystal where its magnetisation is unidirectional, and a crystal can have one domain ("single-domain state" or SD) or several ("pseudo-single domain state", PSD, or "multidomain state", MD; Fig. 1.4). As grain size increases, it becomes energetically favourable to form multiple domains rather than keep increasing the size of one domain, which leads to the the PSD and MD states. For a crystal to be able to record a high-stability magnetic signal, it needs to be very small, ideally

in SD or PSD state — the exact size requirement for a domain state depends on the properties of each crystal (primarily, composition and shape). SD crystals are traditionally considered the ideal domain state for palaeomagnetism, since it leads to high-stability signals that can be retained over long periods of time. However, some PSD crystals have also been shown to carry very high-stability fields in magnetite (Nagy et al. 2017) and kamacite (Nagy et al. 2019).

In meteorites, small magnetic particles usually occur in three forms: as fine grains within a silicate matrix (e.g., in chondrites), as inclusions in silicate crystals (e.g., dusty olivines in pallasites and chondrites), or as part of the cloudy zone (e.g., iron and stony-iron meteorites). The cloudy zone is a nanometric region that forms part of the Widmanstätten intergrowth between kamacite and taenite in iron meteorites. As a meteorite cools, kamacite lamellae form within taenite, leading to an excess of Ni in taenite. The cloudy zone then forms through spinodal decomposition in "islands" of tetrataenite (Ni-rich) in a Ni-poor matrix (Yang et al. 1997). Tetrataenite (tetragonal FeNi with 50-55% Ni) is the ordered form of taenite (cubic FeNi), having a $L1_0$ ordered structure and a small tetragonal distortion ($c/a = 1.0036 \pm 0.00005$; Albersen 1981). Very slow cooling through the tetrataenite ordering temperature (320°C) leads to a slight elongation of the c-axis of the crystal, along one of the existing crystallographic axes of the cubic precursor. As a result of the ordering and tetragonal distortion, tetrataenite dramatically increases its magnetic anisotropy (Wasilewski 1988) and coercivity (up to 2 T when in single-domain state; Uehara et al. 2011). Its uniaxial magnetic easy axis is oriented along the crystallographic c-axis (Néel et al. 1964), which lies parallel to one of the axes of the precursor taenite. Tetrataenite becomes magnetised along its magnetic easy axis, and over the length scale of the cloudy zone, a bias towards the field direction develops if a field is present during ordering. In other words, the cloudy zone acquires a chemical transformation remanent magnetisation (CTRM; Dunlop & Özdemir 1997) during atomic ordering (Wasilewski 1988, Uehara et al. 2011), which forms the basis for cloudy zone magnetic studies (Bryson et al.

Mineral	Composition	Crystal structure	Magnetic behaviour	Curie T (°C)	Coercivity	SD size	Vortex size
Kamacite	FeNi, 0-12%Ni	bcc	Ferromagnetic	770 ¹	MD: <18 mT ² ; SD: <600 mT ³	25 nm ⁴ 28 nm ³ 10-25 nm ⁵	SV: 25 - 50 nm ⁵
Taenite	FeNi, 25-55% Ni	fcc	Ferromagnetic above 45%Ni, otherwise paramagnetic ⁶)	350-400 (for ferromagnetic), 310 ⁷	1.3 - 3.4 mT ⁷	14-50 nm ⁸	>50 nm ⁹
Awaruite	FeNi, 65-90%Ni	fcc	Ferromagnetic	610 ¹⁰	MD: ~100 mT ⁹ ; SD: 490 mT ¹¹ , up to 2T ¹²		
Tetrataenite	FeNi, 48-55%Ni	Tetragonal	Ferromagnetic	550°, but orders at 320°		6 - 160 nm ¹³	No SV state ¹³
Chromite*	FeCr ₂ O ₄	Hexoctahedral	Ferrimagnetic	66 K ¹⁴ 70 K ¹⁵ 71 K ¹⁶	335-505 mT at 10 K ¹⁷		
Schreibersite	(Fe,Ni) ₃ P	Tetragonal	Ferromagnetic	310 ¹⁸			
Magnetite	Fe ₃ O ₄	Hexoctahedral	Ferrimagnetic	580 ¹⁹	10 - 50 mT ²⁰ 25 - 30 mT ²¹	28-75 nm ²²	0.1 - 5 μ m ²³
Pyrrhotite	Fe _{1-x} S, x = 0-0.2	Monoclinic or Hexagonal	Antiferromagnetic (hexagonal), ferrimagnetic (monoclinic) ²⁴	325 (monoclinic) ²⁴ 275 (hexagonal) ²⁵	70-85 mT ²⁶ , can be >100 ²⁷	<3 μ m ²⁸	
Greigite	Fe ₃ S ₄	Hexoctahedral	Ferrimagnetic	330 ²⁹	60 to >100 mT ²⁹	$\leq$ 56 nm ³⁰	70 - 300 nm ³⁰

Table 1.1: Properties of magnetic minerals commonly found in meteorites. The coercivities shown in this table are examples of previously determined coercivities for each mineral, which may vary due to grain shape and size. SD = single-domain, SV = single-vortex; *Ferritchromite (Fe²⁺(Fe_{1-n}Cr_n)₂O₄, n = 0.2-0.6; Hodel et al. 2020) is magnetic with decreasing Cr and increasing Fe content (Robbins et al. 1971, Ziemiak & Castelli 2003). 1 - Nagata (1973), 2 - Fuller (1974), 3 - Lappe et al. (2011), 4 - Muxworthy & Williams (2015), 5 - Nagy et al. (2019), 6 - Uehara et al. (2011), 7 - Funaki et al. (1986), 8 - Devienne et al. (2023), 9 - Gattacceca et al. (2014), 10 - Butler (1972). (*Continues on the next page*)

Table 1.1: (*Continued*) 11 - Nagata (1982), 12 - Einsle et al. (2018), 13 - Mansbach et al. (2022), 14 - Gattacceca et al. (2011), 15 - Gattacceca et al. (2013), 16 - Horen et al. (2014), 17 - Bezaeva et al. (2014), 18 - Lovering & Parry (1962), 19 - Dunlop & Özdemir (1997), 20 - Hodych et al. (1998), 21 - Dunlop (1986), 22 - Muxworthy & Williams (2009), 23 - Sridhar et al. (2021), 24 - Dekkers (1989), 25 - Dekkers (1988), 26 - Roberts et al. (2006), 27 - O'Reilly (1984), 28 - Clark (1984), 29 - Roberts (1995), 30 - Valdez-Grijalva et al. (2018).

2014a, 2015, Nichols et al. 2016, Maurel et al. 2020).

Atomic ordering leading to tetrataenite formation is difficult to replicate in the laboratory due to the slow cooling rate required (e.g., most pallasites have 1-10 K/Myr cooling rates; Yang et al. 1997), leading to uncertainties in the exact nature of tetrataenite NRM and the extent to which the NRM direction reflects the magnetising field direction. However, the CTRM acquisition process described above is supported by the presence of magnetic biases in the cloudy zones of Main Group pallasites (Bryson et al. 2015, Nichols et al. 2016, 2021), IIE irons (Maurel et al. 2020), and H chondrites (Bryson et al. 2019a). Conversely, the cloudy zone in the IAB irons shows no bias towards a field direction, suggesting that no field was present during tetrataenite formation (Nichols et al. 2018). Micromagnetic simulations of tetrataenite formation, combined with scanning precession electron diffraction (SPED) studies also confirmed the link between the crystallographic c-axis and the magnetic easy axis of cloudy zone tetrataenite (Einsle et al. 2018), giving confidence that the cloudy zone is a reliable palaeomagnetic recorder.

1.2.4.2 Recent magnetic studies

Meteorite palaeomagnetism is a field which grew steadily over the past 50 years, before a more rapid growth in the last decade was driven by advances in traditional magnetic techniques and the advent of novel *in situ* techniques, permitting a more in-depth investigation and understanding of meteorite formation and acquisition of magnetic remanence.

Meteorite studies have primarily been used to learn about two types of magnetic fields: ancient dynamo fields acting on planetesimals and nebular fields acting in the very early solar system. An internally-generated magnetic field has been proposed on several chondritic the parent bodies (CV chondrites: Carporzen et al. 2011, Gattac-
ceca et al. 2016; ordinary chondrites: Bryson et al. 2019a; R chondrites: Cournède et al. 2020), achondrites (HED meteorites: Fu et al. 2012; angrites: Weiss et al. 2008, Wang et al. 2017), Main Group pallasites (Tarduno et al. 2012, Bryson et al. 2015, Nichols et al. 2021) and iron meteorites (IVA: Bryson et al. 2017; IIE: Maurel et al. 2020, 2021). Thus, the early solar system appears significantly more magmatically and magnetically active than previously thought.

However, not every differentiated body generated a dynamo, as some studies find no field present (e.g., the ungrouped achondrites NWA 7325, Weiss & Tikoo 2014; the Main Group pallasites, Nichols et al. 2016). These studies show that the absence of a dynamo can also have important implications for our understanding of the parent body’s evolution. In the pallasite example, this observation was used to infer a period of quiescence between the two stages of dynamo generation (thermal and compositional convection; Nichols et al. 2016, 2021).

Meteorite palaeomagnetism has also been used to place constraints on the nebular magnetic field that was still active before the nebula dispersed in the early solar system. Recovered palaeointensities are attributed to a nebular field based on the age of the sample, as well as thermal evolution modelling — if a magnetisation was acquired before a planetesimal would be expected to have differentiated and operated a dynamo, then it is likely to be due to a nebular field (after ruling out impact-generated fields, Crawford & Schultz 1999, and solar wind magnetic fields, Oran et al. 2018). This has been shown to be the case for the chondrules in Semarkona, an LL chondrite (Fu et al. 2014), the chondrules in CO chondrites (Borlina et al. 2021), the chondrules in CR chondrites (Fu et al. 2020b), as well as CM chondrites (Cournède et al. 2015) and C2-ungrouped chondrites (Bryson et al. 2020a,b). At the

same time, younger meteorites which show no evidence of a nebular field magnetic record have been used to constrain the timing of the dispersal of the solar nebula, based on the decay of the nebular field (NWA 7325: Weiss & Tikoo 2014; Kaba, a CV chondrite: Gattacceca et al. 2016; three angrites: Wang et al. 2017; and the matrix of CO chondrites: Borlina et al. 2022).

In this thesis, both differentiated (stony-iron; Chapter 2) and undifferentiated (chondritic; Chapter 4) meteorites were studied using magnetic methods, to learn about internally-generated magnetic fields and the field acting early in the solar nebula.

1.3 Aims & Thesis outline

The primary aim of this thesis is to understand the secondary processes acting on planetesimals, and their effects on meteorites. This thesis focuses on impacts (Chapters 2, 3) and aqueous alteration (Chapter 4). While impacts are known to have been very common in the early solar system, many details are missing from the formation of meteorites whose lithologies have been heavily affected by impact mixing (for example, the stony-iron meteorites). At the same time, recent work has highlighted the importance of understanding aqueous alteration and its effects on magnetic mineralogy (Borlina et al. 2022, Bryson et al. 2023), in order to improve the credibility of palaeointensity estimates from chondritic meteorites. In this thesis, I use high-resolution magnetic and geochemical techniques to study stony-iron (mesosiderites and pallasites), iron (IIIAB) and chondritic (CM) meteorites, in order to improve our understanding of the early evolution of planetesimals and increase the credibility of meteorite palaeomagnetic studies.

Chapter 2: Mesosiderite Palaeomagnetism In this chapter, I conduct a thorough palaeomagnetic investigation of mesosiderites, to constrain their formation and early evolution. I use bulk palaeomagnetic techniques coupled with electron mi-

croscopy, to find that Estherville does not record a bulk high-coercivity (HC) magnetic signal, but that it is able to record a field stronger than $150 \mu\text{T}$. I use these observations to place constraints on the formation of mesosiderites. Mesosiderites are known to be abundant in tetrataenite, a ferromagnet with exceptionally high coercivity, so I explore a number of different methods to identify it and I discuss their relative efficiency. Finally, because low-fidelity samples are not well suited to analysis using traditional palaeomagnetic techniques, I also use EBSD (electron backscatter diffraction) to study the crystallography of the cloudy zone islands. I find that the cloudy zone islands are uniformly oriented, and I discuss this result in connection with the effect of cooling rate on cloudy zone formation.

Chapter 3: The Origin of Pallasites In this chapter, I investigate the formation of pallasites using LA-ICP-MS trace element analysis of FeNi metal, using the meteorites Seymchan (a Main Group pallasite) and Henbury (a IIIAB iron). I test the hypothesis that two distinct metal textures (matrix metal and metal pockets trapped in olivines) contain metal of different origins, by resolving their respective compositions. I find that all metal in Seymchan has the same source, and I propose a pallasite formation model involving two or more impacts to satisfy the chemical and textural constraints from this study and previous work. Finally, I discuss the previously proposed link between Main Group pallasites and IIIAB irons.

Chapter 4: The effect of aqueous alteration on chondrite magnetic mineralogy In this chapter, I use QDM (quantum diamond microscope) magnetic mapping to spatially resolve the magnetic signals of the CM chondrite MCY 05231, to investigate how aqueous alteration affects its magnetisation. I also use optical and electron microscopy to identify the source of each magnetic signal. I resolve different magnetic behaviours in the matrix and the chondrules, as well as two uniform magnetisation directions within the matrix – alteration products (pyrrhotite and magnetite) in the matrix are magnetised uniformly, in a distinct direction from the magnetisation of

the primary metal. At the same time, the chondrules are randomly magnetised. I use these results to suggest that the magnetite in MCY 05231 forms via different pathways, which influence whether it inherits its magnetisation from a previous grain, or whether it acquires a new remanence in the local field. Finally, I suggest a magnetic history for the meteorite and discuss implications for bulk magnetic studies of the solar nebula magnetic field.

Chapter 5: Summary and future work In this chapter, I summarise the findings of previous chapters and suggest directions for future research.

Appendix A This appendix contains supplementary data for Chapter 3 (sulfide composition, and mean major and trace element compositions for each metal pocket sampled in Seymchan).

Appendix B This appendix details the mathematical basis for two cluster analysis criteria (the silhouette criterion and the gap criterion) used in Chapter 3.

Appendix C This appendix contains supplementary data for Chapter 4 (results of the net moment inversion algorithm to fit QDM magnetic sources to dipoles, and supplementary figures).

Chapter 2

Mesosiderite Palaeomagnetism

2.1 Introduction

2.1.1 Mesosiderites

Mesosiderites are stony-iron meteorites composed of approximately 50% FeNi metal and sulfides, and 50% silicates (primarily pyroxene, plagioclase, olivine, silica (usually trydimite); Mittlefehldt et al. 1998). As the only meteorites that combine crustal and mantle material (the silicates) with core material (the metal) (Mittlefehldt et al. 1998), it is widely believed that their asteroid parent body experienced an impact early in its history, and that at least one of the impacting bodies was fully differentiated. However, the nature of the impact and origin of the metal are unclear: the metal could have originated entirely from the impactor, a differentiated body (Powell 1971, Wood 1979, Hassanzadeh et al. 1990), or it could have originated in the parent body's own core, which got mixed with the crust and mantle following the impact (Greenberg & Chapman 1984, Scott et al. 2001, Haba et al. 2019, Sugiura et al. 2022).

While it is generally accepted that an impact was involved in the formation of mesosiderites, the timing of the impact is uncertain. Bogard & Garrison (1998) identify an Ar-release event interpreted as a collisional event <4 Gyr ago, based on ^{39}Ar - ^{40}Ar dating. However, the young Ar-Ar ages could also be attributed to slow cooling (Haack et al. 1996). Haba et al. (2017) date zircons in Estherville using U-Pb systematics, and obtain an age of 46 ± 26 Myr after CAI formation for the

silicate-metal mixing event, which is expected to reset the U-Pb decay system in zircons (Hopkins et al. 2015). Furthermore, Haba et al. (2019) propose that the metal-silicate mixing occurred at $4,525.39 \pm 0.85$ Myr based on U-Pb dating, and they propose that mesosiderites formed through a hit-and-run collision of asteroid 4 Vesta.

The possibility that mesosiderites and the HED (Howardite-Eucrite-Diogenite) meteorites have the same parent body (asteroid Vesta) is a current open question (Greenwood et al. 2015). Evidence for a common origin includes oxygen isotopes (Greenwood et al. 2006, 2017), similarities in metal composition (Yamaguchi et al. 2006), synchronous Lu-Hf ages for cumulate eucrites and mesosiderites (Haack et al. 2003), similar parent body sizes (Haack et al. 1996, 2003), and similar Co, Ni abundances and Pt/Ir ratios (Shirai et al. 2016). Evidence against a common origin includes differences in olivine compositional ranges and FeO/MnO ratios (Greenwood et al. 2015), platinum-group element abundances (Shirai et al. 2016), compositional diversity in clasts (Greenwood et al. 2017), cosmic ray exposure ages (Terribilini et al. 2000), and differentiation ages obtained from Mn-Cr systematics (Wadhwa et al. 2003).

Mesosiderites are widely recognised as the slowest-cooled meteorites identified, and previous studies indicate a two-stage cooling history. At high temperatures (1150 – 900°C), Fe-Mg profile measurements in pyroxenes indicate a cooling rate of 1 – 100°C/year (Ganguly et al. 1994). Below 400°C, the cooling rate slows to 0.2 – 0.5°C/Ma, calculated from Ni and Co concentrations in the Widmanstätten pattern (Goldstein et al. 2014, Wasson & Hoppe 2014). This is also consistent with studies on the magnetic microstructure of mesosiderites (Nichols et al. 2020), $^{40}\text{Ar}/^{39}\text{Ar}$ age (Bogard & Garrison 1998) and Fe-Mg ordering in orthopyroxene (Ganguly et al. 1994). The initial fast cooling is attributed to thermal re-equilibration following the mixing of cool silicates and hot metal. To explain the subsequent slow cooling, it has been suggested that the meteorites were buried to a significant depth within the

parent body after they formed (Haack et al. 1996, Greenwood et al. 2015).

The large number of open questions surrounding mesosiderite formation and their extremely slow cooling rates make these meteorites an interesting target for research. A useful way to learn about the formation of a terrestrial body is to study the internal processes active early in its history, including whether it generated a dynamo, as an active dynamo would have implications for the size of a planetesimal, when it accreted, and if/how an inner core crystallised (Section 1.2.4).

At the same time, due to their early formation, meteorites often act as snapshots into the early solar system, and studying the magnetic recording of a meteorite can provide a wealth of information on the history and internal dynamics of the parent body (Section 1.2.4.2). While the origin of mesosiderites on Vesta is not certain, it is likely that mesosiderites came from a parent body similar to Vesta, which could have produced a dynamo in its early history. Fu et al. (2012) identified a remanence of $2\ \mu\text{T}$ in an HED meteorite and attributed it to crustal remanence caused by an earlier dynamo on Vesta, and Formisano et al. (2016) created a model showing that Vesta could have produced a dynamo lasting 150 – 500 Ma, which would be capable of imparting the hypothesised crustal remanence. This suggests that the mesosiderite parent body could have produced a dynamo early in its history. Identifying a magnetic remanence in mesosiderites would represent strong evidence for this, providing valuable information on the internal dynamics and formation of the mesosiderite parent body.

The long cooling history of mesosiderites also has the potential to offer insights into the late stages of a dynamo if the field is recorded by tetrataenite (see Section 2.1.2), as this mineral forms at relatively low temperatures reached late in the cooling process. Furthermore, studying the palaeomagnetic record of mesosiderites could also provide information on the nature of the metal-silicate mixing event and the extent to which it scrambled the mesosiderite parent body – if a field is recorded by mesosiderites, that field would have operated after the impact, which would indicate that the impact

did not scramble the entire parent body (i.e., at least the core remained intact). This would be consistent with a less destructive impact such as a hit-and-run collision (Haba et al. 2019).

Whether a dynamo was active during the impact depends on the timing of the impact and the characteristics of the mesosiderite parent body – according to the modelling on the case study of Vesta (Formisano et al. 2016), an internally-generated dynamo could last up to 500 Myr, so it would be active if the mesosiderite-forming impact was early (4.525 Gyr; Haba et al. 2019), but not if the impact was late (<4 Gyr; Bogard & Garrison 1998).

Mesosiderites are petrologically similar to the other stony-iron meteorite group (the pallasites), because they have a similar history involving metal-silicate mixing through an impact. Magnetic methods have been applied to the Main Group pallasites and identified a magnetic remanence imparted by fields generated by core dynamo activity, and these studies have enhanced our understanding of planetary magnetic fields by resolving two stages of dynamo generation in the early history of the pallasite parent body, separated by a period of quiescence (Tarduno et al. 2012, Bryson et al. 2015, Nichols et al. 2016, 2021). Given the similarity between pallasites and mesosiderites, there is scope for investigating mesosiderites to explore the nature of the impact and the internal dynamics of its parent body.

Several magnetic studies have previously been performed on mesosiderites. Larson et al. (1973) obtained preliminary bulk demagnetisation data that suggests an unstable magnetic signal. In addition, Kukkonen & Pesonen (1983) measured the NRM and magnetic susceptibility of several meteorite groups, including mesosiderites, to classify them based on their petrophysical properties. They found that the magnetic moment of mesosiderites ranges between $55 \times 10^{-5} \text{ Am}^2\text{kg}^{-1}$ and $2350 \times 10^{-5} \text{ Am}^2\text{kg}^{-1}$, and the magnetic susceptibility ranges between $5550 \times 10^{-7} \text{ m}^3\text{kg}^{-1}$ and $8970 \times 10^{-7} \text{ m}^3\text{kg}^{-1}$. Collinson (1991) analysed the magnetic record of oriented subsamples of Estherville and found their magnetisations to be spatially heterogeneous and inconsistent among

subsamples. This led them to suggest that the metal present in Estherville acquired its remanence before its mixing with the silicates, leading to the scatter of NRM directions in each subsample. More recently, Bono & Tarduno (2015) performed a preliminary single-crystal magnetic study on metal and sulfide inclusions in olivines from the mesosiderite NWA 8368, and found a stable high-temperature signal in two mutually-oriented samples. Finally, Nichols et al. (2020) studied the reliability of the cloudy zone as a magnetic recorder in several meteorite groups, including the mesosiderites. They found that the cloudy zone in Estherville does not preserve a stable magnetic signal, since the tetrataenite islands are multidomain due to the slow cooling rates experienced while tetrataenite was forming.

2.1.2 Tetrataenite

Tetrataenite is an FeNi mineral (50-55% Ni) which forms through atomic ordering of taenite as it slowly cools through 320°C. Tetrataenite has been shown to be a very hard ferromagnet, with high magnetic anisotropy and coercivity up to 2 T when in single-domain state (Uehara et al. 2011). Because of its high coercivity, tetrataenite is very difficult to remagnetise. Furthermore, its formation temperature is below its Curie temperature, so if tetrataenite retains a signal, that rules out reheating above 320°C (Dos Santos et al. 2015). These features make it an attractive target for palaeomagnetic studies, having previously been used to extract magnetic information from several metal-bearing meteorite groups (Section 1.2.4.1).

It has been suggested that tetrataenite formation in a magnetic field causes it to adopt one of three c-axis orientations, with an overall directional bias towards the field direction, since the magnetic easy axis aligns with the crystallographic c-axis of tetrataenite (Einsle et al. 2018). This is based on observations of clustering of neighboring tetrataenite island orientations in the cloudy zone (Bryson et al. 2014b, Einsle et al. 2018). Therefore, studying the crystallography of tetrataenite in the cloudy zone may be another way to partly constrain the magnetic field active dur-

ing cloudy zone formation, with the caveat that this method cannot distinguish the polarity (positive or negative orientation) along the c-axis, only that the field was aligned with the axis.

2.1.3 Chapter outline

Since previous magnetic studies on mesosiderites report inconsistent data and do not agree on a magnetic history for the parent body, the main aim of this chapter is to constrain the recording fidelity of Estherville to assess its quality as a palaeomagnetic recorder, and use it to further constrain the formation of the mesosiderite parent body.

In this chapter, I outline a petrologic (Section 2.3.1) and bulk magnetic study (Section 2.3.3) of the mesosiderite Estherville, which reveals that it does not have a stable, high-coercivity component of natural remanent magnetisation, but that it has the fidelity to recover strong magnetic fields ($\sim 150 \mu\text{T}$; Section 2.4.1). I then discuss the likelihood of the mesosiderites having experienced strong, dynamo-generated magnetic fields on the parent body (Section 2.4.5)

Since traditional palaeomagnetic methods are not suited to analysing such challenging samples, I also adopt a different method to obtain magnetic information from Estherville, by studying the crystallography of the cloudy zone to test the hypothesis that tetrataenite formation within a magnetic field leads to a c-axis orientation bias towards the field direction (Einsle et al. 2018; Section 2.3.2). I find that all tetrataenite in the cloudy zone adopts the same crystallographic orientation and I suggest that the slow cooling rates experienced by mesosiderites resulted in large, interconnected cloudy zone islands, which made it energetically favourable for all islands to have the same alignment, instead of alignments dictated by an external magnetic field (Section 2.4.2).

Since tetrataenite is known to be abundant in mesosiderites (Clarke & Scott 1980), I also use this study to compare and contrast different methods to identify tetrataen-

ite, finding that magnetic methods can more readily establish the presence of single-domain tetrataenite (Section 2.4.4).

2.2 Methods

2.2.1 Sample

This study focuses on the mesosiderite Estherville, which fell in Iowa in 1879. Its total recovered mass was 320 kg and the 37.4 g piece of Estherville used in this study was borrowed from the Oxford University Museum of Natural History (sample Mt.051). Prior to magnetic measurement, the sample was stored in a magnetically shielded room (background field <500 nT) in the Department of Earth Sciences, University of Oxford, for ~ 6 months.

2.2.2 Sample preparation

The bulk sample was first cut with a 4-inch diamond blade rock saw, which produced two subsamples and the remainder of the bulk sample. One of the subsamples was then cut with a different saw (low-speed, equipped with a diamond-impregnated blade) using oil for lubrication, as water could promote the formation of terrestrially magnetised iron oxides. Four samples of Estherville were created (Table 2.1), one of which (EV1_4) was then mounted in epoxy resin using a Struers EpoFix kit and polished for analysis using electron microscopy. The number of cuts through the sample was minimised in order to preserve meteorite material, and as a result, all subsamples include some of the fusion crust. The subsamples were stored in a magnetically shielded room after the cutting, and they were only exposed to the Earth's field for approximately 18h prior to the magnetic measurements.

First, the meteorite was polished by hand to a smoothness of $1\text{ }\mu\text{m}$ and cleaned with petroleum ether between steps. Prior to starting the EBSD analysis, the sample surface was impregnated with resin to prevent silicate crystals coming loose and leading to scratches on the softer metal. It was then polished to $0.25\text{ }\mu\text{m}$ using a diamond

Sample ID	Sample preparation & Dimensions (mm)	Weight (g)	Methods applied
EV1_1	Chip, 12 x 7 x 8 mm	1.9 ± 0.005	NRM measurement, AF demagnetisation, IRM + ARM acquisition
EV1_2	Chip, 16 x 13 x 8 mm	2.7 ± 0.005	NRM measurement
EV1_3	Remainder of bulk sample after cutting, 20 x 20 x 30 mm	29.5 ± 0.05	NRM measurement
EV1_4	Polished sample mounted in resin, 11 x 7 x 4 mm	1.3 ± 0.1	NRM measurement, AF demagnetisation, IRM acquisition, electron microscopy

Table 2.1: Summary of Estherville subsamples and methods applied to each of them.

suspension for 15 minutes, then to $0.05 \mu\text{m}$ for 2h on a Vibromet vibrating polisher, cleaning with petroleum ether in an ultrasonic bath between stages. The sample was placed in vacuum for 12h after each of these stages to remove any water left from the polishing suspension, to prevent oxidation.

To dissipate surface charge during electron microscopy, a 20 nm carbon coat was deposited prior to the SEM work, using an Edwards-type vacuum coater. For EBSD, a thin Ir coat (~ 2.5 nm) was used together with Cu tape between the sample and the holder, and two lines of silver dag paint connecting the tape and the two main metallic regions of the sample. The Ir coat was applied using a Leica EM ACE600 metal coater. All sample preparation was performed in the Department of Earth Sciences in the University of Oxford.

2.2.3 X-Ray Microtomography

Prior to cutting the meteorite, X-ray microtomography was used to produce a 3D scan of the bulk sample. I used a Nikon XTH 225ST X-ray tomography scanner at the XTM Facility, Palaeobiology Research Group, University of Bristol, at 223 kV voltage and $80 \mu\text{A}$ intensity, with a 0.5 mm Sn X-Ray filter.

X-Ray microtomography served a dual purpose: to show the distribution of the

metal in Estherville and to help identify the best line of section along which to cut the sample. The software used for visualisation of the image stack and analysis is Fiji (ImageJ; Schindelin et al. 2012).

2.2.4 Electron Microscopy

The aim of the microscopy is to characterise the known magnetic minerals in terms of their size and distribution. The magnetic minerals in Estherville are metal (kamacite, taenite, tetrataenite), schreibersite and chromite, and their properties are presented in Table 1.1. To image small magnetic grains, I used electron microscopy techniques: scanning electron microscopy (SEM), backscatter electron and energy-dispersive spectroscopy (EDS) for semi-quantitative element characterisation, as well as electron backscatter diffraction (EBSD) to determine crystal structure.

Initially, SEM was used for general imaging and to obtain a complete petrographic description of the sample. I used the FEI Quanta 650 SEM based in the Department of Earth Sciences at 20 kV current, using Aztec for data analysis. For a more thorough quantitative analysis of the metal, I then used the CAMECA SX-5 FE electron microprobe (EMPA) in the Earth Sciences Department, which is equipped with five wavelength-dispersive spectroscopy (WDS) detectors, to obtain absolute concentrations of Fe, Ni, Si, S, and P at 15 kV and 20 nA, with a spot size of 1 μm . The measurement times are 60 s (Si), 30 s (P, S) and 20 s (Fe, Ni). The detection limits were approximately 180 ppm (Si), 370 ppm (S), 320 ppm (P), 600 ppm (Fe) and 900 ppm (Ni).

Finally, I used Electron Backscatter Diffraction (EBSD) to assess the crystallographic orientations of the cloudy zone. EBSD was performed in the Material Sciences Department at the University of Oxford, on a Zeiss Merlin SEM equipped with a Bruker QUANTAX eFlash HD EBSD detector. The SEM is operated by the Bruker Esprit software, which was also used for the data analysis, together with Channel 5 Tango.

EBSD was used to produce a map of the cloudy zone, tetrataenite rim and kamacite (900x magnification, 250 nm step size), and a subset of the map re-measured at higher magnification and resolution (3500x magnification, 70 nm step size). Both maps are analysed at 20 kV and 35 ms exposure time. The high-resolution pixel size was chosen to be significantly lower than the average cloudy zone island size in Estherville (463 ± 32 nm; Yang et al. 1997).

The crystallographic orientations determined by EBSD are displayed using Inverse Pole Figures (IPFs), which assign a colour to each crystal orientation on a map, generating IPF maps that show how the orientation changes throughout a sample.

Furthermore, EBSD was also used on some silicate minerals and the metal grains, in order to index them. These measurements were taken in the Department of Earth Sciences, University of Oxford, using an Oxford Instruments EBSD detector in the FEI Quanta 650 SEM, at 30 kV current in a low-vacuum environment.

2.2.4.1 EBSD reference files

The raw EBSD data is indexed according to reference files containing the lattice parameters and diffraction data of a known crystal, allowing identification of a crystal based on its structure. The tetrataenite reference file is based on the lattice parameters reported by Albertsen (1981) ($a = b = 3.5761$ Å, $c = 3.589$ Å, space group P4/mmm). A crystal file containing these lattice parameters was also created in TWIST, and used to analyse the results. The site occupation for used for the TWIST file is given in Table 2.2.

To identify kamacite and taenite, I used a reference file for body-centered cubic (bcc) Fe (AMCDB ID 0012931) for kamacite, and a file for body-centered cubic (bcc) Fe (AMCDB ID 0012931) for taenite. As Ni contents in both kamacite and taenite can vary widely (approximately 0% and 7% in kamacite and 7% to 55% in taenite) without altering the crystal structure, the Fe substitution to Ni does not affect the lattice. This implies that it would not lead to a difference in the Kikuchi bands detected by EBSD, as they depend on the crystallography, not the composition of a

Atom	x	y	z
Fe	0	0	0
Ni	0.5	0.5	0.5

Table 2.2: Site occupation within the tetrataenite unit cell, based on data from Albertsen (1981), used to create a crystal file in TWIST.

mineral. Therefore, pure Fe in the fcc and bcc crystal structures is suitable to index kamacite and taenite through EBSD.

2.2.5 Palaeomagnetism

2.2.5.1 Alternating field demagnetisation

The four Estherville subsamples listed in Table 2.1 were mutually oriented manually and mounted in plastic boxes using blue tack to prevent movement. The Natural Remanent Magnetisation (NRM) was measured on each subsample, and the NRM was then removed in two subsamples (EV1_1 and EV1_4) using alternating field (AF) demagnetisation. Only two subsamples were demagnetised in order to minimise the destructive analysis performed on the meteorite. Increasingly higher intensity alternating fields were applied in three orthogonal directions, in an 18-step run up to 110 mT. The remaining magnetisation was measured after every step. The measurements were conducted on a 2G Enterprises Superconducting Rock Magnetometer (SRM) in a magnetically shielded room (<200 nT background field) in the University of Southampton Palaeomagnetism Laboratory. Zijdeveld diagrams are plotted using pmagpy 4.2.50 (Tauxe et al. 2016).

2.2.5.2 Palaeointensity determination

To determine the palaeointensity recorded by Estherville, I use the ARM (Anhysteretic Remanent Magnetisation) and IRM (Isothermal Remanent Magnetisation) methods, normalising the sample’s NRM by a 200 μ T ARM (200 mT AF field) and a 950 mT IRM. The methods are given by:

$$P_{ARM} = \frac{\Delta NRM}{\Delta ARM} \frac{b}{f'} \quad (2.1)$$

$$P_{IRM} = \frac{\Delta NRM}{\Delta IRM} \times a \quad (2.2)$$

where ΔNRM , ΔARM and ΔIRM are the NRM, ARM and IRM lost through AF demagnetisation, b is the DC field (here, 200 μT), f' is the experimentally calibrated ratio between TRM (thermoremanent magnetisation) and ARM, and a is the calibration factor between TRM and IRM.

I use $f' = 1.34$ and $a = 3000 \mu\text{T}$, which have been calibrated for kamacite-bearing samples (Tikoo et al. 2012, Lappe et al. 2013, Weiss & Tikoo 2014). While other magnetic carriers are present in mesosiderites (e.g., tetrataenite, schreibersite, taenite), their TRM/ARM and TRM/IRM ratios are not well constrained. However, the coercivity of taenite is similar to kamacite while in multidomain state (up to 18 mT; Table 1.1 and references therein), suggesting that their f' and a values will be similar (Bryson et al. 2017). The magnetic properties of schreibersite are not well understood, but its low abundance in mesosiderites (<2 vol%, Powell 1971) indicates a limited contribution to remanence. Finally, tetrataenite likely carries a CTRM (Section 1.2.4.1), which is more difficult to approximate using an ARM or IRM than a TRM. This is because previous work suggests that CTRMs are weaker than TRMs by a factor of 1-10 (McClelland 1996, Smirnov & Tarduno 2005), leading to a likely underestimated palaeointensity using the TRM-calibrated factors f' and a defined above. Furthermore, the high coercivity of tetrataenite means that the AF field used during ARM acquisition may not be able to reach the tetrataenite coercivity range, such that the ARM might not approximate the entire tetrataenite population in the sample. Here, an AF field of 200 mT is used, which is the maximum field possible on the D-2000 AF coil in the Oxford Magnetism Laboratory. This field exceeds the coercivity of multidomain tetrataenite (100 mT; Gattacceca et al. 2014), but not the coercivity of SD tetrataenite, which would likely exceed 300 mT (Bryson et al. 2017).

As a result, an ARM could approximate multidomain tetrataenite, but not single domain, which is where a stable remanence is more likely to be stored. However, this effect is not expected to represent a problem for the IRM method (Equation 2.2), because demagnetisation is not required and the direct fields are much stronger (up to 950 mT; Section 2.2.5.3). Furthermore, this study shows that the sample contains multidomain tetrataenite, but no significant single-domain tetrataenite (Section 2.3.3), so uncertainties in the ARM/TRM and IRM/TRM calibration for tetrataenite are unlikely to introduce a major source of error to the palaeointensity determination.

2.2.5.3 IRM acquisition

Samples EV1_1 and EV1_4 were imparted with increasingly higher IRMs in a 23-step run up to 950 mT, and the acquired magnetisation was measured after each step. The 950 mT IRM was partially removed through AF demagnetisation in a 13-step run up to 110 mT. The IRM acquisition provides information on the spectrum of coercivities present in the rock, even if the minerals with those coercivities did not record any component of the NRM. Any mineral will gain an IRM as long as the applied field is stronger than the mineral's coercivity, which allows access to minerals that may not have been magnetised originally. This makes it possible to obtain and analyse a full coercivity spectrum, contributing to the identification of the magnetic carriers present in the sample. Additionally, since the fields applied this way are much higher than the alternating fields used in demagnetisation (950 mT rather than 110 mT), it is possible reach the high intensities that saturate the sample, even if it did not reach saturation naturally.

The IRMs were imparted using an in-line IRM coil in the Southampton Palaeomagnetism Laboratory, and subsequently measured and demagnetised using the SRM.

2.2.5.4 Magnetic fidelity experiments

To determine the palaeomagnetic fidelity of the samples, I used ARM fidelity experiments as described by Tikoo et al. (2012), Bryson et al. (2017) and Nichols et al.

(2021). This protocol involves applying a laboratory magnetic field and obtaining a palaeointensity estimate from the sample, by removing the acquired magnetisation. Based on the accuracy of the estimate (how close the palaeointensity value is to the true value of the applied field), it is possible to determine the minimum field that can be recorded by the magnetic carriers in Estherville.

Magnetic fidelity experiments were performed by applying and demagnetising a series of ARMs (AC field of 200 mT, and DC bias fields of 150 and 200 μ T), and attempting to retrieve the bias field values through the palaeointensity method described above (Eq. 2.2), through normalisation to an IRM (Nichols et al. 2021). The ARMs were imparted using a D-2000 AF coil, and subsequently measured and demagnetised on a 2G Enterprises SRM located in a magnetically shielded room in the Oxford Magnetism Laboratory. The ARMs are normalised by an IRM in order to retrieve the highest ARM (200 μ T) that could be applied on the D-2000 AF coil. The modified palaeointensity formula is:

$$I = \frac{\Delta ARM_b}{\Delta IRM} \times a \quad (2.3)$$

where ΔARM_b is the ARM lost through demagnetisation at the bias field b to be retrieved, ΔIRM is the applied 950 mT IRM, and a is the same calibration factor of 3000 μ T in Eq. 2.1.

Following Bryson et al. (2017), the difference (D') and error (E) metrics are used to evaluate the fidelity of the sample:

$$D' = \frac{I - L}{L} \quad (2.4)$$

$$E = \frac{W}{L} \quad (2.5)$$

where L is the applied laboratory intensity (equivalent to b in Eq. 2.1 and 2.3), I is the retrieved intensity from Eq. 2.3, and W is the width of the 95% confidence interval. The fidelity test succeeds if D' is between -0.5 and 1 and E is below 1, following Tikoo

et al. (2012) and Nichols et al. (2021). However, some authors also suggested 0.5 as an upper limit for E (Bryson et al. 2017). These fidelity criteria are very generous, making it very likely that recorders that fail the criteria are not able to record a field. Furthermore, E is the error strictly associated with the linear fit, without accounting for other sources of error (e.g., uncertainties in the palaeointensity determination due to the calibration factor a , both formal as determined by Tikoo et al. 2012, and due to the presence of magnetic phases other than kamacite, as detailed in Section 2.2.5.2). Here, these criteria are adopted in order to rule out poor palaeomagnetic recorders and to quantify the improvement in fidelity with higher fields, in the absence of more robust statistical tests for magnetic fidelity.

2.3 Results

The results section is separated into three parts: petrography (Section 2.3.1), cloudy zone crystallography (Section 2.3.2), and rock and palaeomagnetism (Section 2.3.3).

2.3.1 Petrography

2.3.1.1 X-Ray microtomography

X-Ray microtomography was used to scan the bulk sample and produce a 3D stack of 2002 images, and an animation going through the stack.

The microtomography reveals three primary components in Estherville: metal (highest density phase, appearing bright white), silicates (low density, dark grey) and sulfides (intermediate) (Fig. 2.1). Minor metal grains are also visible within the silicate matrix. The higher-Ni phases in the metal are also visible due to their higher density, which allows the Widmanstätten pattern to be traced in 3D.

The sample has almost equal proportions of silicates and metal, but some slices are more metal-rich than others. Before cutting, a section with an equal split between metal and silicates (with some sulfides) was identified using this method. Due to the widespread presence of metal and its large density contrast to the silicates, the images

contain X-Ray residuals where the waves were not able to pass through the metal. Therefore, image resolution in the metal-rich slices is poor (Fig. 2.1B), but the slices with less metal are able to resolve individual silicate crystals (Fig. 2.1A).

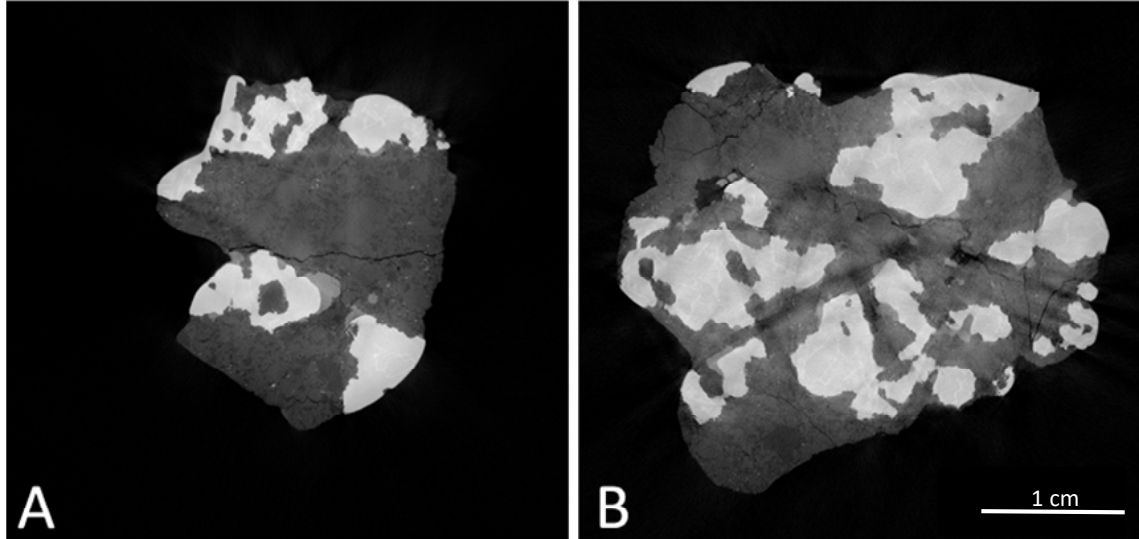


Figure 2.1: CT images taken at 223 kV voltage and 80 μ A intensity, with a 0.5mm Sn X-Ray filter. The bright areas are metal and the dark areas are silicates. A. Slice 448, resolving individual silicate crystals; B. Slice 1030, with dark stripes caused by metal blocking some of the X-Rays.

2.3.1.2 Electron microscopy

Estherville sample EV1_4 is a mixture of a mafic silicate rock and metal (Fig. 2.2). The silicate part contains pyroxenes (enstatite and inverted pigeonite) and plagioclase (anorthite) with accessory tridymite (SiO_2), merrillite ($\text{Ca}_9\text{NaMg}(\text{PO}_4)_7$) and chromite ($\text{Fe}(\text{Cr,Al})_2\text{O}_4$). The metallic part contains FeNi alloys (kamacite, taenite, tetrataenite), sulfides (troilite, FeS) and phosphides (schreibersite, $(\text{Fe,Ni})_3\text{P}$). An oxidised rim is present around some of the metal, with non-stoichiometric iron oxides, and the sample is covered in glassy fusion crust from atmospheric heating, melting and subsequent quenching. There is no structural evidence for shock deformation (e.g., rock fabric, shocked veins).

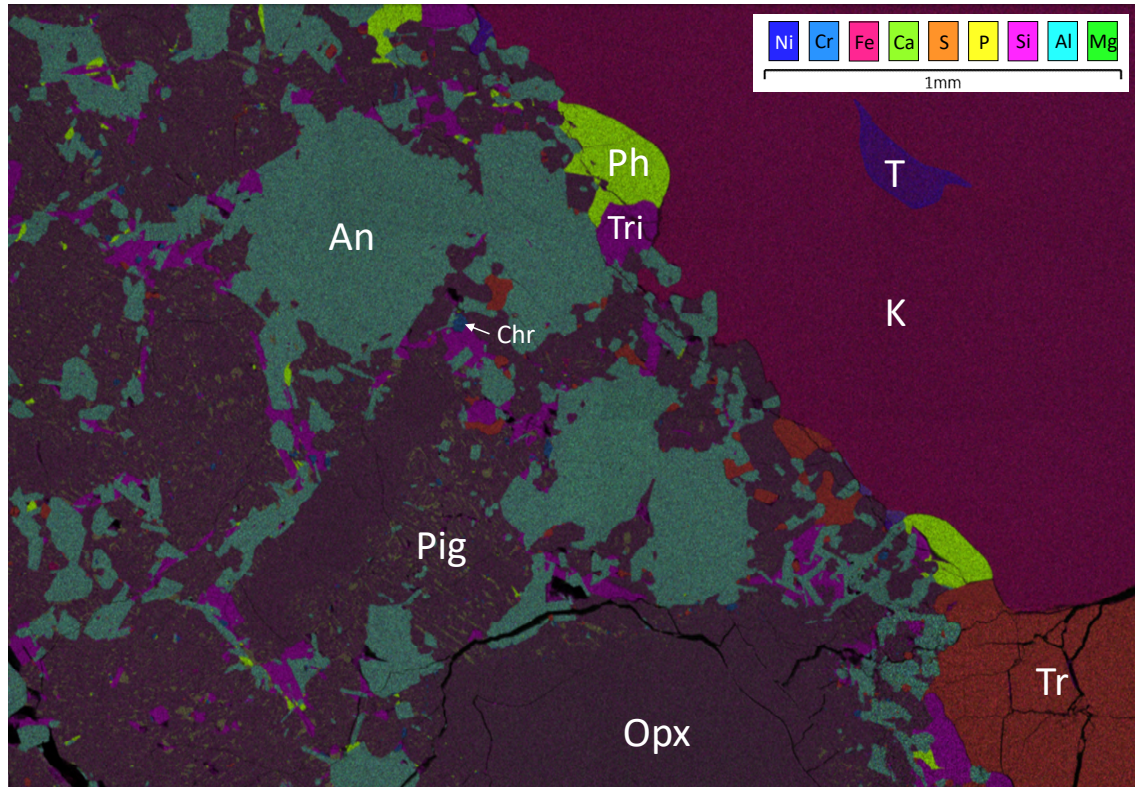


Figure 2.2: EDS elemental map, measured at 20 kV. The background was corrected using the Oxford Instruments Aztec software. K = kamacite, T = taenite (and other high-Ni phases), Tr = troilite, Opx = orthopyroxene, Pig = Inverted Pigeonite (orthopyroxene + clinopyroxene lamellae), An = anorthite, Chr = chromite, Tri = tridymite, Ph = phosphate (merrillite).

Silicates The pyroxenes are orthopyroxene (enstatite) and inverted pigeonite (now enstatite with augite lamellae exsolved upon slow cooling, occasionally with surviving pigeonite cores - Fig. 2.3D). Texturally, the two pyroxenes differ in crystal size (the enstatite being coarser) and in inclusion abundance and composition. While the inverted pigeonite can contain inclusions of sulfide, metal, tridymite, chromite and occasionally anorthite, the coarse orthopyroxene crystals lack inclusions, except for metal and sulfide inclusions which form trails that trace former crystal boundaries (Fig. 2.3A). Locally, the pyroxene also contains symplectic structures of vermicular chromite in a tridymite matrix (Fig. 2.3B) and it has rims of finer inverted pigeonite crystals (Fig. 2.3C). The silica is tridymite, as confirmed through EBSD analysis of crystal structure. Tridymite is a high-temperature, low-pressure silica polymorph,

indicative of thermal metamorphism or very high magmatic temperature followed by quenching. Tridymite often forms euhedral needles or laths as an interstitial phase (Fig. 2.2). No olivine is present on the exposed surface of this sample. This mineral assemblage suggests that the silicate part of the rock is an HT-LP (high-temperature, low-pressure) metamorphosed basalt.

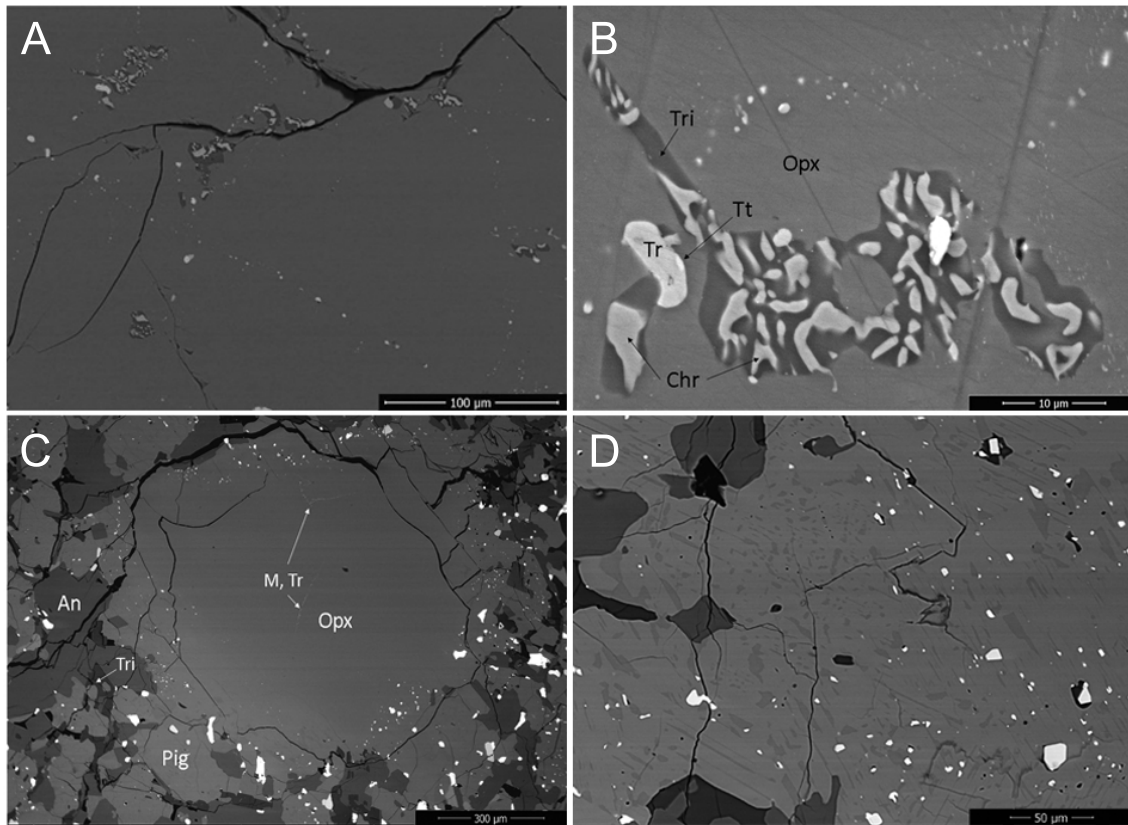


Figure 2.3: A. Trails of inclusions and chromite-tridymite intergrowths in a coarse pyroxene crystal (BSE image); B. Close up BSE image of the intergrowth; C. Different pyroxene exhibiting similar inclusion trails. Note how the pyroxene crystal is surrounded by finer inverted pigeonite, anorthite and tridymite crystals, and how the coarse pyroxene crystals are featureless apart from the inclusion trails (BSE image). D. BSE image of inverted pigeonite crystals, showing inclusions and lamellae of augite within enstatite, and a pigeonite core.

Metals Metal phases form approximately 50% of the meteorite. Two coarse (~ 0.5 cm diameter) metal nodules were identified, along with smaller crystals ($1\text{--}50\ \mu\text{m}$ diameter) mixed in with the silicates. Generally, the metal appears to fill in the

gaps in the silicate crystals, particularly around the edges of the main metal nodules (Fig. 2.2), although some of the smaller metal crystals mixed with the silicates are euhedral.

Several metallic phases were identified. The most common phase is kamacite (bcc FeNi, 0-7% Ni) which forms the majority of the metal mass. High-Ni lamellae are present within the kamacite, in the characteristic meteoritic Widmanstätten pattern, which is characterised by thick, irregularly shaped high-Ni lamellae (Fig. 2.4B). The lamellae are zoned, with high Ni along the margins and lower Ni in the middle, forming an M-shaped profile. The shape of the profile depends on the width of the lamella and on the cooling rate (Fig. 2.5). No plessite was observed in this sample.

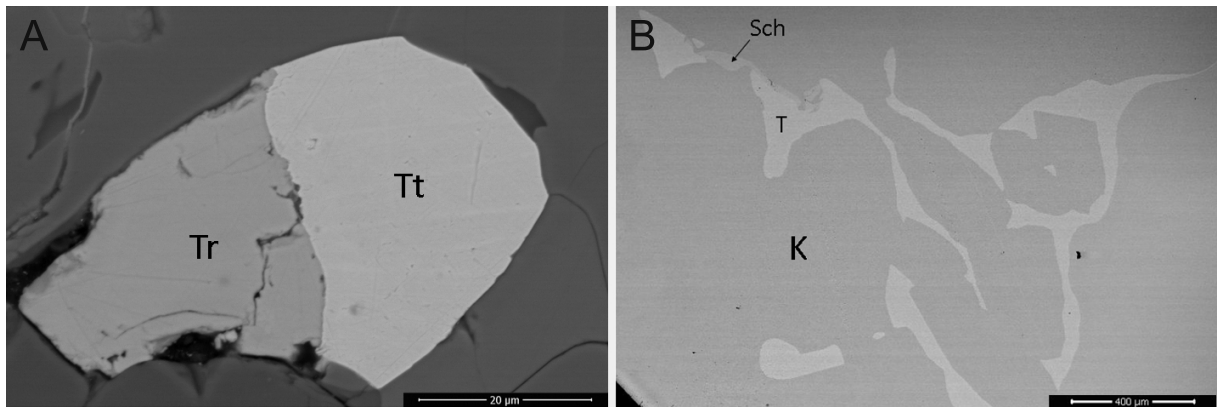


Figure 2.4: A. Fine metal/sulfide grain in the silicate matrix, composed of troilite (Tr) and tetrataenite (Tt) (BSE image); B. Widmanstätten pattern within the metal. K = kamacite, T = taenite or tetrataenite, Sch = schreibersite (BSE image).

The results from the electron probe analysis of the metal are summarised in Figure 2.6. Kamacite is characterised by constant Ni ($\sim 7\%$) in the interior of the metal nodules, and lower Ni when isolated in fine crystals within the silicate matrix (0-5%). There is some variation in Ni content within the coarse kamacite in the vicinity of other phases (taenite, tetrataenite and schreibersite), tending towards lower Ni and higher Fe. Generally, the fine kamacite falls on a Fe-Ni line ($\text{Fe} + \text{Ni} = 100\%$, Fig. 2.6), and the coarse kamacite in the metal nodules forms a near-horizontal line of constant Ni ($\sim 7\%$) and variable Fe (90-92%).

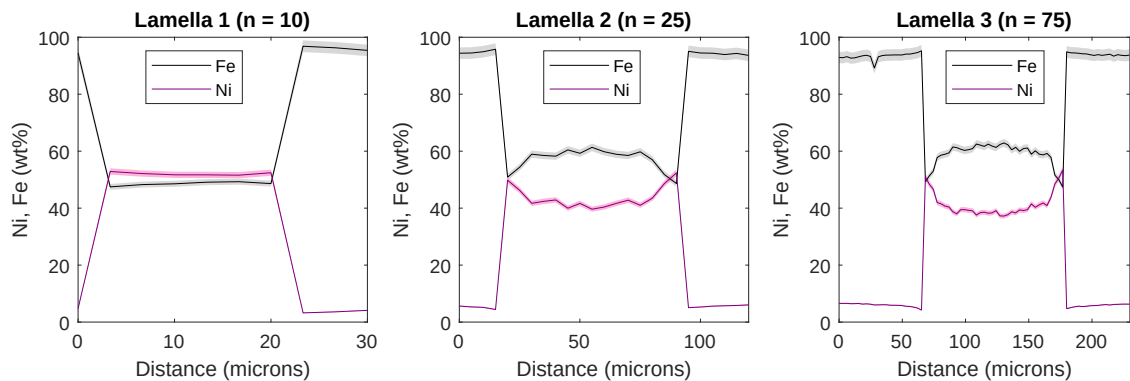


Figure 2.5: Fe and Ni profiles through taenite lamellae, obtained using EMPA. The shaded regions correspond to the 95% confidence intervals ($\pm 1\sigma$) of the measurements.

SEM-EDS and EMPA analysis revealed three candidates for tetrataenite, identified based on nickel content (50-55%). In Estherville, high-Ni FeNi occurs within the cloudy zone (Nichols et al. 2020), in the silicates as micron-sized grains, usually in association with kamacite and/or troilite (Fig. 2.4A), and as nanometric inclusions within pyroxene crystals (Fig. 2.3A). Given the compositional overlap between taenite and tetrataenite (Fig. 2.15), chemical analysis alone is not enough to distinguish between the two crystals. However, taenite and tetrataenite have different crystal systems (cubic and tetragonal), so EBSD was also used to try to differentiate between them (Sections 2.3.2, 2.4.4).

Other minerals Troilite is ubiquitous in Estherville and almost always associated with metal phases. Chromite typically contains 3-4% Al in addition to the typical Fe, Cr and O contents. It often occurs as inclusions in troilite but it is also present as an accessory phase within the silicate part. Merrillite is a common phosphate in meteoritic materials, and in Estherville it occurs in association with the metal, as protrusions into kamacite (Fig. 2.2), and more rarely as small isolated grains in the silicates. Schreibersite is also present in very small quantities near the taenite lamellae within the kamacite (Fig. 2.4B), and it contains approximately 33 wt% Fe and 51 wt% Ni.

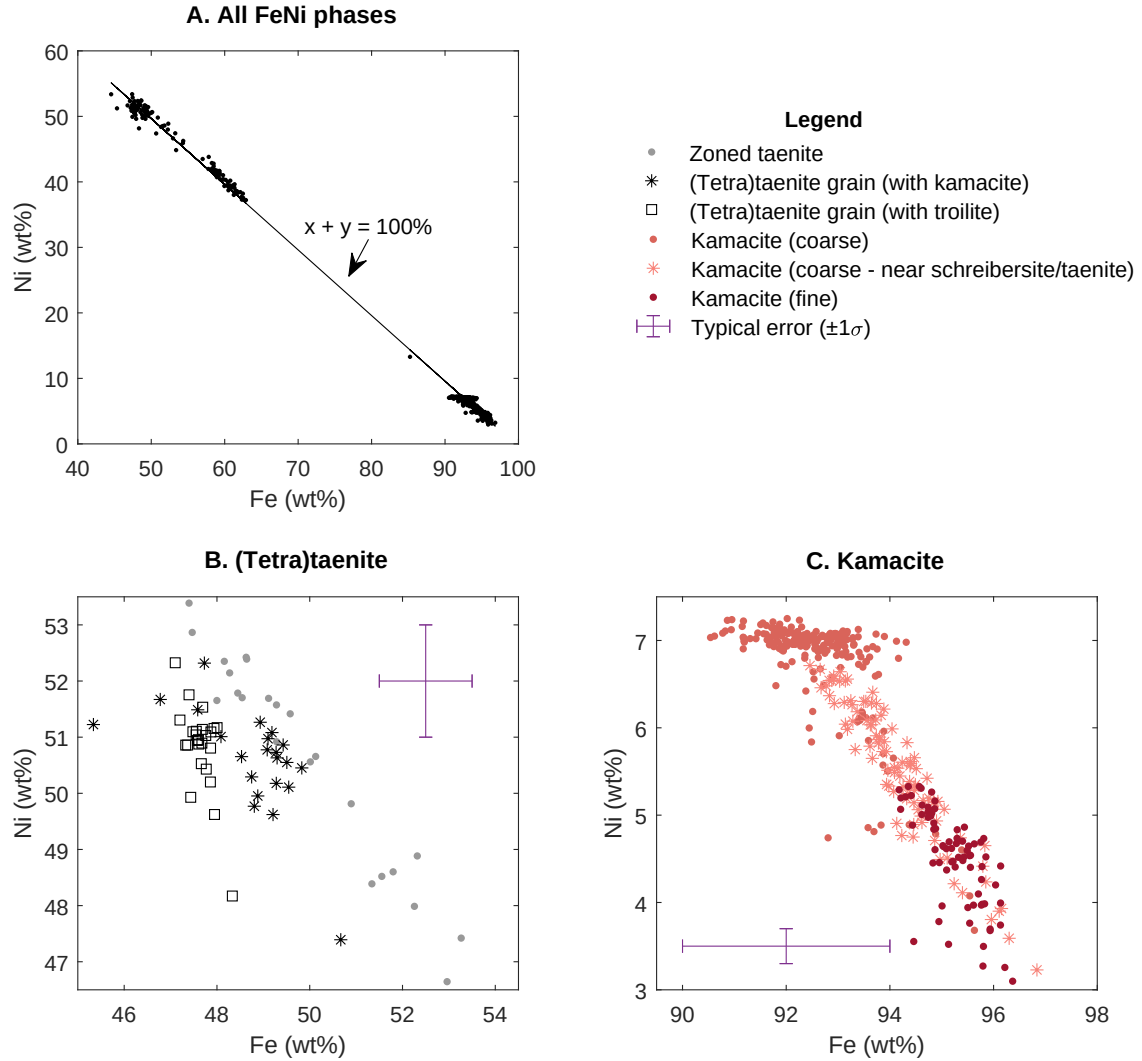


Figure 2.6: A. Fe, Ni contents (in weight %) for all FeNi alloys in Estherville, obtained using EMPA. The data is fitted through by the line $x + y = 100\%$. B. Close up on high-Ni end (taenite and tetrataenite). Note the Fe and Ni offsets from the line, depending on which other phase the (tetra)taenite shares a grain with in the silicate matrix (sharing with troilite leads to be increased Fe depletion, whereas sharing with kamacite leads to broadly equal Ni and Fe depletion). C. Close up on low-Ni end (kamacite). Note the flat trend of the high-Ni kamacite and that the low-Ni end is occupied exclusively by the fine kamacite in the silicate matrix. The error bars show typical errors ($\pm 1\sigma$) for the measurements.

The petrological analysis of Estherville confirms that the studied clast is an HT-LP metamorphosed basalt, consistent with the precursor to the silicate portion of the mesosiderites being an achondrite.

2.3.2 EBSD analysis of the Cloudy Zone

Two sets of EBSD maps were produced in the same area, in a high-Ni lamella within kamacite (Fig 2.7A). The first map (hereinafter referred to as map 1) shows kamacite, tetrataenite rim and cloudy zone (Fig 2.7B), and the second map (map 2) is a higher resolution, higher magnification version of the first one, showing only some of the tetrataenite rim and the cloudy zone (Fig 2.7C).

2.3.2.1 Phase identification

Phase identification relies on Kikuchi band contrast (among other factors), which is a measure of EBSD pattern quality. In map 2, the band contrast reveals the mesosiderite cloudy zone, with bright areas representing the tetrataenite rim and islands, and dark areas representing the cloudy zone matrix and some polishing imperfections (Fig. 2.8 B). The EBSD phase maps are shown in Fig. 2.8 C, D. The cloudy zone islands are indexed as fcc Fe, and in isolated places, the cloudy zone matrix is indexed as bcc Fe.

2.3.2.2 Crystallographic orientation

IPF maps for the cloudy zone were created based on map 2 (Fig. 2.9). Fig. 2.9 is indexed using bcc Fe and fcc Fe, and shows a uniform crystallographic orientation across both the cloudy zone and the tetrataenite rim. Furthermore, visual inspection of the raw EBSD patterns reveals that they are identical throughout the cloudy zone of map 2, even when they are indexed as different crystal orientations. Likewise, kamacite (indexed as bcc Fe) also shows uniform crystal orientation.

2.3.3 Rock and palaeomagnetism

2.3.3.1 Alternating Field Demagnetisation

The NRM measurement results are summarised in Table 2.3. The magnetic moments are high, reflecting the large metal content in all samples (approx. 50 vol%).

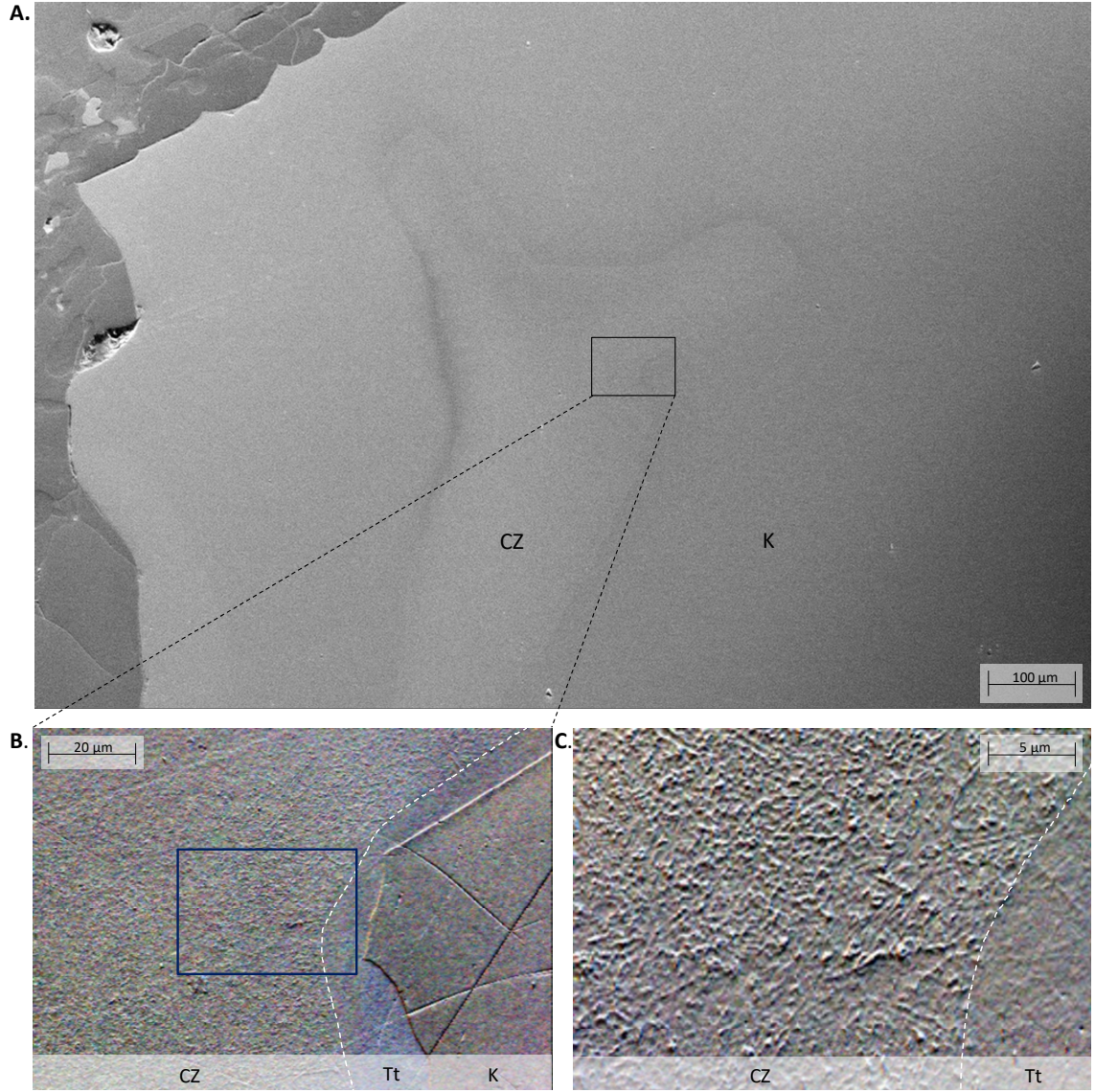


Figure 2.7: Electron images showing the location of each EBSD map. A. Secondary electron image showing a taenite lamella surrounded by kamacite. The box delineates the region where map 1 was measured. B. Forward Scatter Detector (FSD) image of the region for map 1, with a box delineating the area covered by map 2; C. FSD image of the area covered by map 2. CZ = Cloudy Zone, Tt = Tetrataenite rim, K = kamacite.

The results of the AF demagnetisation of the NRM are summarised in Table 2.4 and in the orthogonal projection diagrams in Fig. 2.10. The demagnetisation reveals a stable low-coercivity (LC) component in both samples, which was removed by 12 mT. The LC-HC transition is low (12 mT) and there is no recoverable HC component, as

the vectors become scattered and clustered around the origin with increasing applied field. The background noise was consistently low during the demagnetisation (up to 10^{-10} Am²), so the scatter was likely due to low palaeointensities and randomisation of magnetic domains with each AF step.

2.3.3.2 Palaeointensity determination

A palaeointensity was recovered from the low-coercivity component of the magnetisation (up to 12 mT) using the slope calculated through least squares fitting of five datapoints, normalising the NRM by a 200 μ T ARM (Fig. 2.11). The recovered palaeointensity is 110 ± 123 μ T, where the error corresponds to the 95% confidence

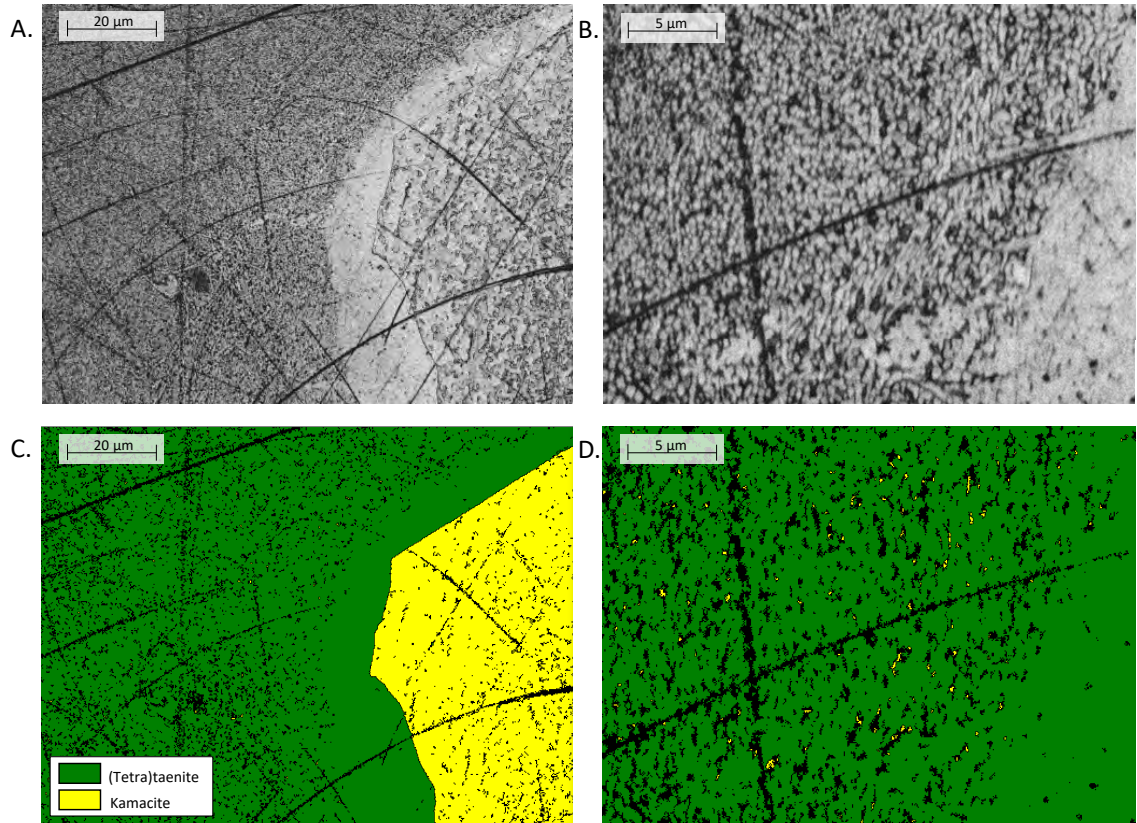
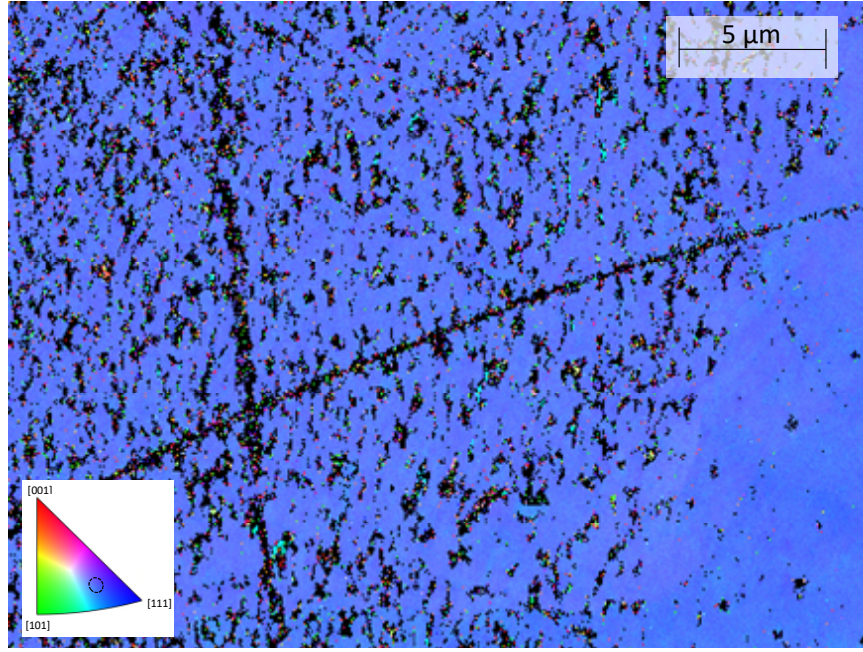


Figure 2.8: A. Band contrast on map 1; B. Band contrast on map 2; C. Phase map of map 1, where green = tetrataenite or taenite, and yellow = kamacite. D. Phase map of map 2, where green = tetrataenite or taenite. The black areas in all maps represent areas which could not be indexed (cloudy zone matrix and scratches across the sample).

A. Crystal orientation map



B. Inverse Pole Figures

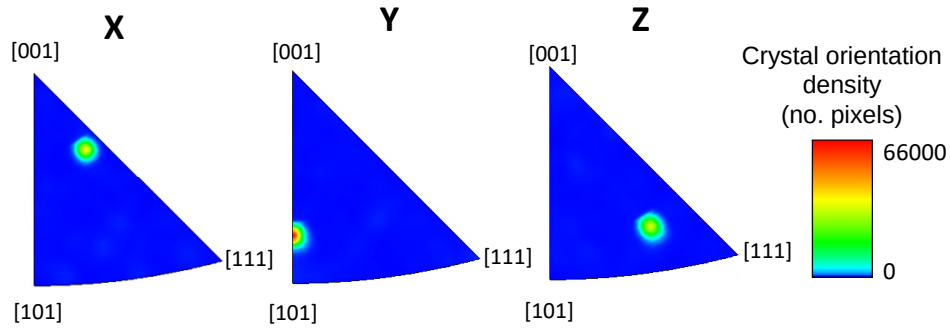
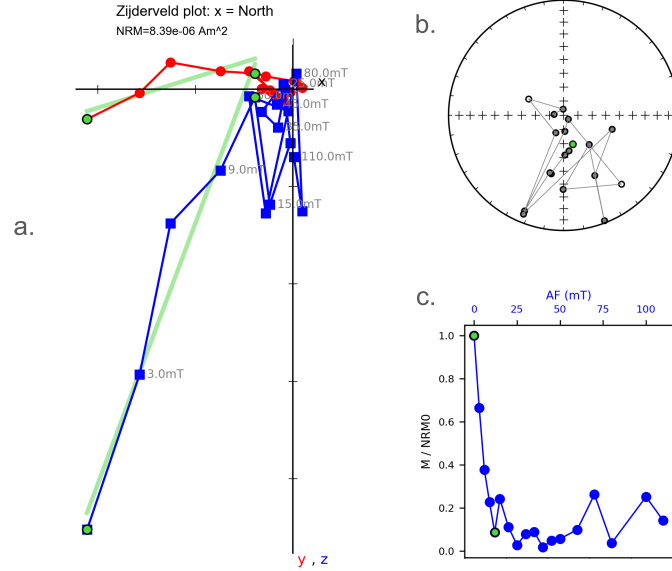


Figure 2.9: A. Crystallographic orientation in the cloudy zone and rim, indexed as fcc Fe, based on the Inverse Pole Figure (IPF) relative to the z-axis of the sample. The uniform colour shows that the cloudy zone and tetrataenite rim are indexed as one crystal with the same orientation throughout. The orientation is shown by the dashed circle on the inset IPF figure. B. IPFs for fcc Fe, relative to the three sample axes. The figures show the directions of each sample axis (x, y and z) relative to three crystallographic axes [001],[101] and [111]. The orientations are tightly clustered in each IPF, further showing that the crystal orientation is the same on the entire map. The IPF figure relative to the z-axis of the sample is used to produce the map in A.

A



B

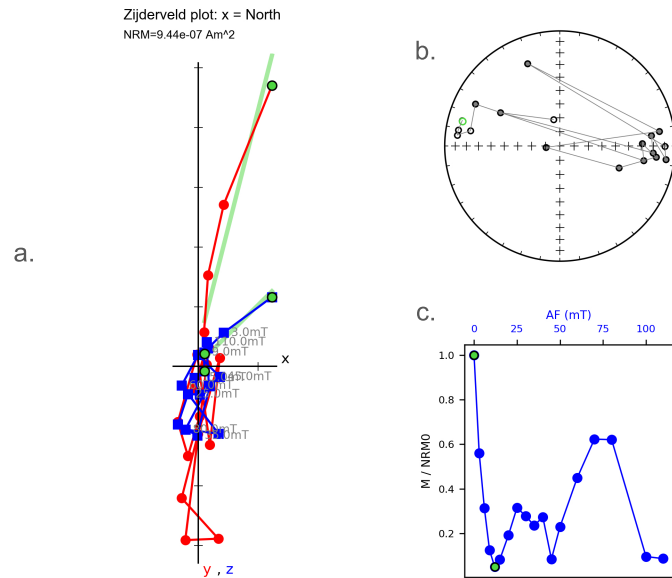


Figure 2.10: Demagnetisation of samples EV1_1 (A) and EV1_4 (B). a. Orthogonal (Zijderveld) projections showing the magnetic vectors of the samples, demagnetised up to 110 mT. The light green lines show the low-coercivity trends. b. Lower hemisphere, equal area stereonet showing each measurement in the demagnetisation sequence. c. Magnetic moment normalised by its initial value, plotted against applied AF field. The green circles show the low-coercivity trend plotted in the orthogonal projections (a).

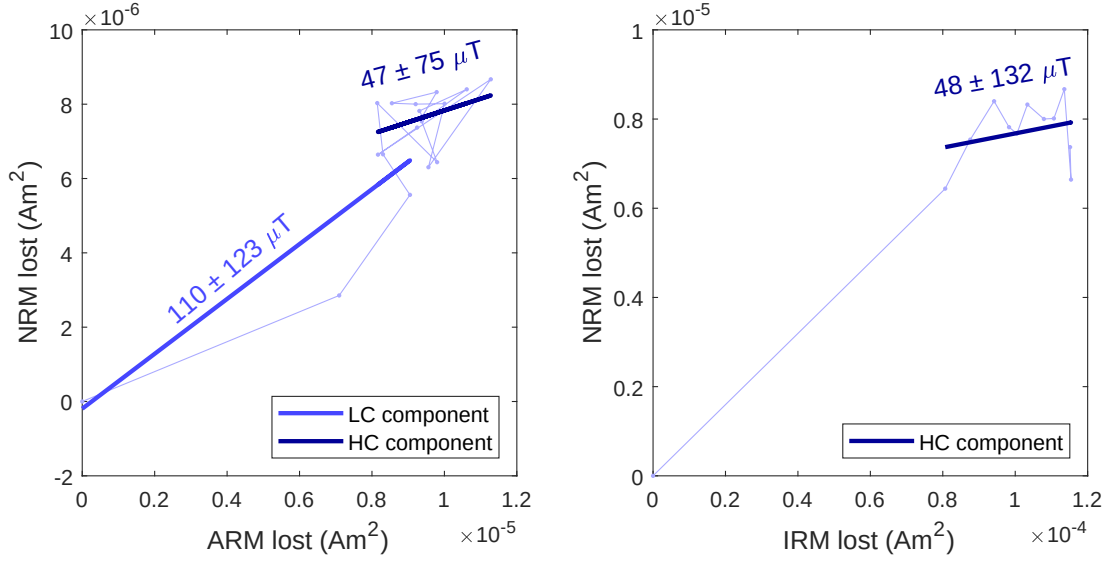


Figure 2.11: NRM lost vs ARM lost (left) and NRM lost vs IRM lost (right) for palaeointensity determination using the ARM method, with least square fit lines, separated into LC and HC components. The resulting palaeointensities are shown on each line.

interval of the fit, which is high due to the low number of datapoints. The palaeointensities recovered from the HC segment using the ARM and IRM methods are $47 \pm 75 \mu\text{T}$ and $47 \pm 132 \mu\text{T}$, which are consistent with the lack of a stable HC component.

2.3.3.3 IRM acquisition

The IRM acquisition curve and the coercivity spectrum demonstrate that the majority of the IRM is acquired at low coercivities (Fig. 2.12). The main peak at 15 mT corresponds to multidomain metal, and the secondary peak at 100 mT has previously been identified as multidomain tetrataenite (Gattacceca et al. 2014). The MD tetrataenite peak does not immediately drop to 0 Am^2 , but instead slopes down until

Sample ID	NRM Intensity (Am^2)
EV1_1	8.37×10^{-6}
EV1_2	1.01×10^{-5}
EV1_3	1.90×10^{-5}
EV1_4	9.45×10^{-7}

Table 2.3: Summary of NRM intensity measurements

Sam- ple ID	NRM (Am ²)	Com- ponent	AF range (mT)	Mean Dec (°)	Mean Inc (°)	MAD (°)	dANG (°)
EV1_1	8.37×10^{-6}	All	0 - 110	175.1	64.8	13.8	3.2
		LC	0 - 12	162.6	68.6	5.2	13.7
		HC	12 - 110	152.6	-87.1	21.4	158.0
EV1_4	9.45×10^{-7}	All	0 - 110	278.1	-15.1	12.6	137.4
		LC	0 - 12	284.3	-13.7	4.9	2.7
		HC	12 - 110	273.6	-13.0	22.2	167.6

Table 2.4: Summary of AF demagnetisation results for samples EV1_1 and EV1_4. Samples EV1_2 and EV1_3 were not demagnetised in order to minimise the destructive analysis (Table 2.1). MAD = Mean angular deviation, dANG = Deviation Angle

it reaches 0 Am² at 800 mT, which indicates the presence of some high-coercivity minerals in Estherville. Single-domain metal can reach coercivities of 600 mT (Lappe et al. 2011), making it the likely source of the signal between 100 mT and 600 mT, in the absence of observed pyrrhotite in Estherville (Section 2.3.1). Other potential high-coercivity carriers are discussed in Section 2.4.3.

The AF demagnetisation of the IRM is shown in Fig 2.13E. The IRM was demagnetised up to 110 mT, which fails to completely demagnetise the sample. This is also consistent with high coercivity (>110 mT) minerals being present.

2.3.3.4 ARM fidelity experiments

Successive ARMs were imparted and then removed from sample EV1_1, and the demagnetisation behaviour is shown in Fig. 2.13 B-D. Palaeointensities were calculated by normalisation to IRM, in order to constrain the magnetic fidelity of the sample. The results from the fidelity experiments are shown in Fig. 2.14. According to the criteria outlined in Section 2.2.5, the only component that passes the fidelity test is the HC component of the 200 μ T ARM (149.2 μ T TRM-equivalent field, using $f' = 1.34$ for kamacite-bearing samples; Lappe et al. 2013, Tikoo et al. 2014, Weiss & Tikoo 2014). The palaeointensity recovered from this component is $130 \pm 62 \mu$ T ($D' = -0.13$, $E = 0.82$), suggesting that the sample has the fidelity to record a 149.2

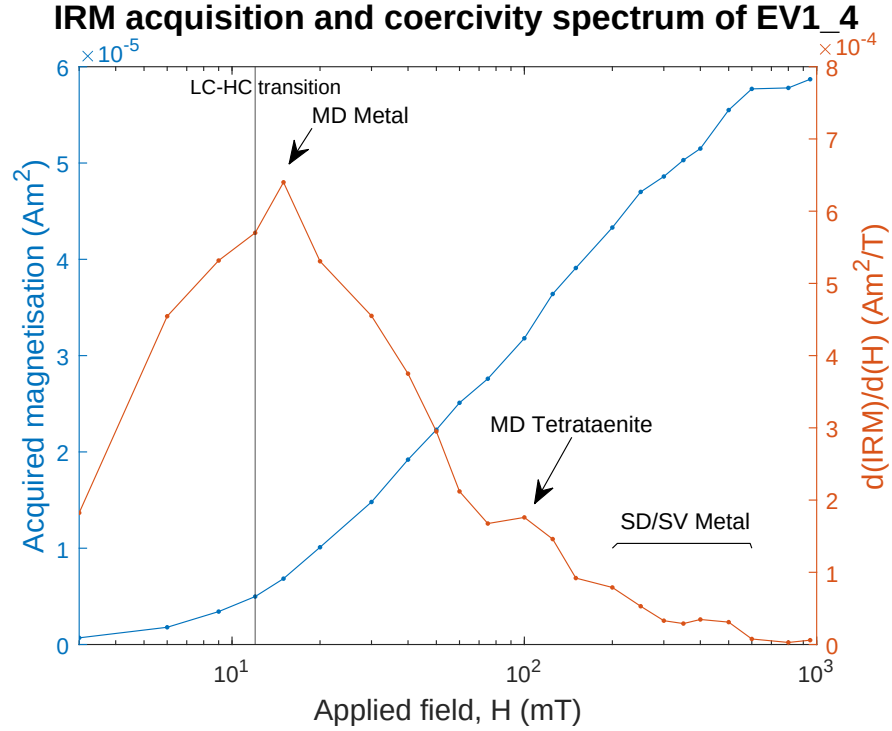


Figure 2.12: IRM acquisition (blue) and coercivity spectrum (orange) for sample EV1_4. The black vertical line marks the LC-HC transition at 12 mT.

μT field.

2.4 Discussion

In this section, I discuss the bulk magnetic record and fidelity of Estherville (Section 2.4.1), and since the sample has low magnetic fidelity, I then discuss an alternative way to extract magnetic information (crystallography of the mesosiderite cloudy zone, Section 2.4.2). I summarise the magnetic carriers present (Section 2.4.3) with an emphasis on tetraetaenite and the relative effectiveness of the different methods used to detect it (Section 2.4.4), and end by discussing implications for the field which could have operated on the mesosiderite parent body (Section 2.4.5).

2.4.1 Magnetic remanence and fidelity

The AF demagnetisation of the sample reveals a stable LC component, blocked to 12 mT (Fig. 2.10). The LC component directions in the two mutually oriented

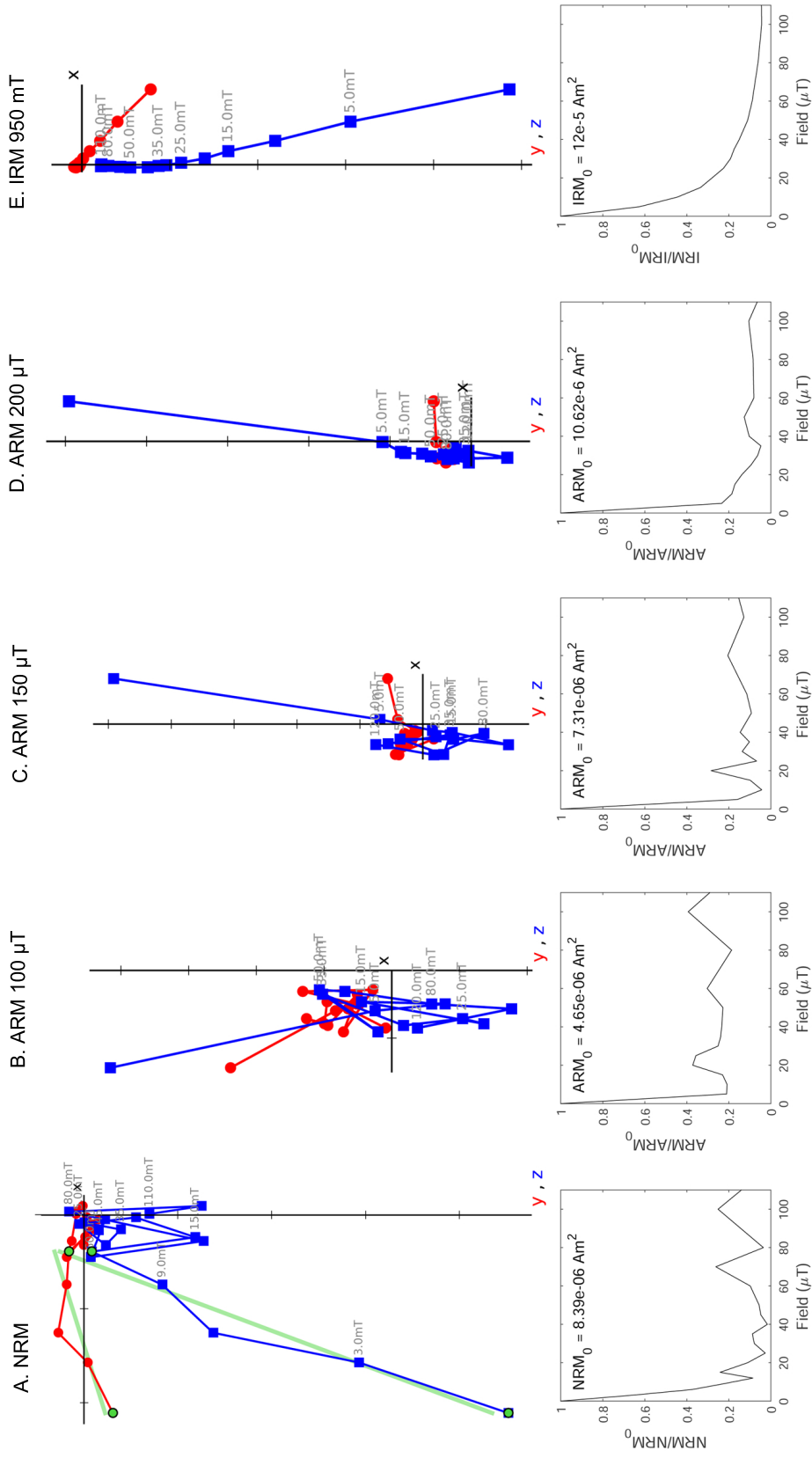


Figure 2.13: Demagnetisation behaviour of the NRM of Estherville, as well as three successive ARMs (100, 150 and 200 μT and a 950 mT IRM, shown on Zijderveld diagrams where $x = \text{East}$. The bottom plots show the remaining moment (normalised by the initial moment, as shown on each plot) at every AF step.

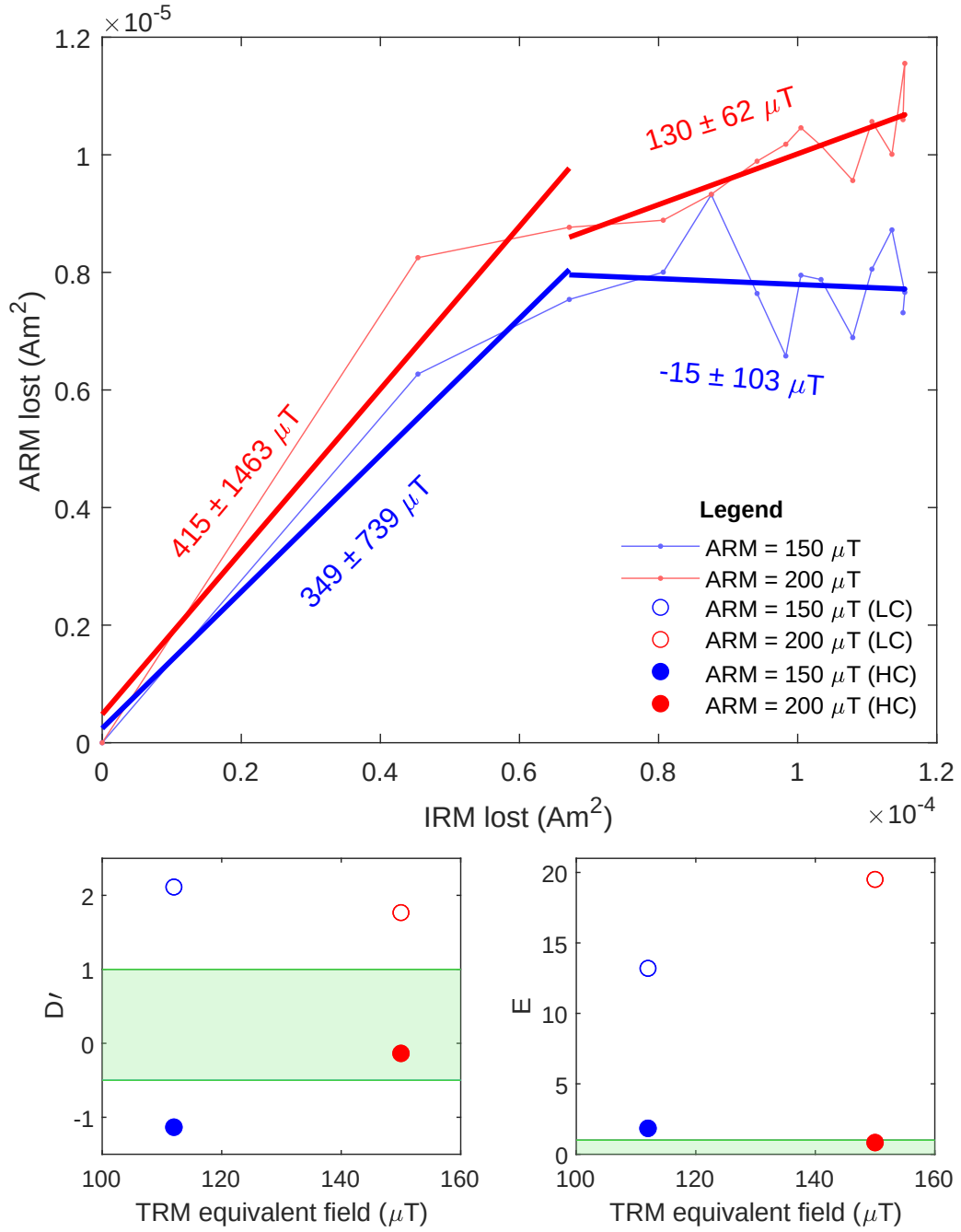


Figure 2.14: Top: Plot showing 150 μT and 200 μT ARM lost vs 950 mT IRM lost for sample EV1_1, with least square fit lines, separated into LC and HC components for both datasets. The recovered palaeointensities are shown on each line. Bottom: D' and E metrics for each fit line in the top figure, plotted against the TRM-equivalent field calculated using $f' = 1.34$ (Weiss & Tikoo 2014). The permissible intervals as established in Section 2.2.5 are shaded in green. The only component that passes the fidelity test is the HC component of the 200 μT ARM, with $D' = -0.13$, $E = 0.82$.

subsamples are not consistent with each other, which could indicate that the signal is a Viscous Remanent Magnetisation (VRM) acquired since the samples were cut approximately 6 months prior to the magnetic measurements. However, since the samples were stored in a magnetically shielded room after the preparation and only removed approximately 18h before the measurements (Section 2.2.2), it is unlikely that they would have time to acquire a 12 mT remanence. It is more likely that the LC component is a pTRM recorded by the fusion crust during atmospheric passage. All samples contain some fusion crust (Section 2.2.2) and tumbling through the atmosphere could explain the difference in directions between oriented subsamples.

The demagnetisation behaviour also indicates that Estherville does not record a HC signal (Fig. 2.10). This could be explained in two ways: either there was no field to be recorded, or the field was low intensity and the rock does not have the magnetic fidelity to record it. The ARM fidelity experiments show that Estherville has the ability to record a 150 μ T TRM-equivalent field (Fig. 2.14). This is based on the permissible error range being less than 1 (Tikoo et al. 2012, Nichols et al. 2021), although some authors have used a maximum error of 0.5 instead (Bryson et al. 2017). Adopting a limit of 0.5 for E would suggest that the recording fidelity of Estherville is >150 μ T. Since these ranges are arbitrary and the majority of previous studies adopt a limit of 1, and because it is apparent from Fig. 2.13 that the 200 μ T ARM (equivalent to a 150 μ T TRM) demagnetises neatly, I also adopt a limit of 1 for E and suggest that the recording fidelity of the bulk sample is 150 μ T.

2.4.2 Cloudy Zone crystallography

In this section, I discuss the results of the EBSD analysis of the metal microstructures observed in Estherville. The cloudy zone and tetrataenite rim are successfully indexed using fcc Fe for reference, in both the phase maps (Fig. 2.8) and IPF maps (Fig. 2.9). However, the cloudy zone of mesosiderites is known to be composed of tetrataenite (Novotny et al. 1981) and there is no evidence for reheating (e.g., kamacite breakdown

to martensite as observed by Clarke & Mason 1982). Its indexing as taenite instead of tetrataenite suggests that the tetragonal distortion caused by atomic ordering ($c/a = 1.0036 \pm 0.00005$; Albertsen 1981) is too negligible to be detected by EBSD.

Kamacite is successfully indexed as bcc Fe outside of the high-Ni lamellae. Furthermore, minor kamacite is detected in the cloudy zone matrix, but this should be treated cautiously given the low pattern quality associated with all areas between islands (Fig. 2.8). The low band contrast could be caused by relief in the polishing, if the polishing preferentially removes the matrix, or it could be due to reduced lateral resolution caused by the change in phase, which is a particularly important consideration if the boundary between the phases is not perpendicular to the tilt angle of the sample (Zaefferer 2007). If the volume of matrix is reduced, the effects of sub-surface grain boundaries may appear to affect all or most of the matrix. However, it has previously been suggested that the cloudy zone matrix is composed of kamacite instead of antitaenite based on Mössbauer data (Nichols et al. 2020), so it is possible that this signal is real despite the lower band contrast.

The IPF map corresponding to the cloudy zone (Fig. 2.9) shows that the islands in the mesosiderite cloudy zone all have the same crystallographic orientation. By contrast, Einsle et al. (2018) used SPED (scanning precession electron diffraction) on the cloudy zone of the Tazewell meteorite (IAB iron) and found that groups of neighboring tetrataenite islands show uniform c-axis orientations, forming three clusters of crystallographic orientations, one for each of the three choices of c-axis orientation for tetrataenite. The discrepancy could be due to the different cooling rates of Estherville (0.2 - 0.5°C/Ma; Goldstein et al. 2014) and Tazewell ($\sim 10^\circ\text{C}/\text{Ma}$; Yang et al. 1997), as the Estherville cloudy zone had a much longer time to cool. Following spinodal decomposition at $\sim 400^\circ\text{C}$ (exact temperature depending on Ni content), the precursor taenite in the cloudy zone region decomposes into high-Ni taenite and a low-Ni phase (points F and G on the "zoned taenite path" in Fig. 2.15). The high-Ni taenite forms as islands, which have time to grow until atomic ordering

at 320°C causes taenite to become tetrataenite (point H in Fig. 2.15). Because of the slow cooling rate, by the time the rock cools to 320°C, the islands may already be interconnected. Nichols (2017) simulated spinodal decomposition and found that high-Ni taenite is all connected next to the tetrataenite rim, while the low-Ni phase forms islands. Further from the rim, the matrix phase forms a network, isolating tetrataenite islands. This results in a pure high-Ni taenite rim (which will become the tetrataenite rim after atomic ordering at 320°C – "rim path" in Fig. 2.15), as well as an interconnected network next to the rim – islands only become isolated further from the rim, where there is a higher abundance of low-Ni matrix phase (Fig. 2.16). Since the high-Ni taenite precursor to tetrataenite is all connected following spinodal decomposition, it would be energetically difficult for tetrataenite to adopt different c-axis orientations based on the ambient magnetic field, which explains why the cloudy zone has one crystallographic orientation in Fig. 2.9. The cloudy zone effectively behaves in the same way as the tetrataenite rim, despite having undergone spinodal decomposition, which is consistent with the observation of no change in magnetic behaviour between tetrataenite rim and islands in Estherville (Nichols 2017).

Therefore, cloudy zone crystallography does not provide any insight into whether a magnetic field was present during cloudy zone formation in mesosiderites. Even if a field were present, no information about it can be extracted from the mesosiderite cloudy zone, which is consistent with the conclusion from the traditional palaeomagnetism methods (Section 2.4.5). This could be specific to mesosiderites because of their extremely slow cooling rates, which could still allow the c-axis orientations of faster-cooled meteorites to provide magnetic information, as suggested by Einsle et al. (2018).

It was previously suggested that the cooling rate in mesosiderites leads to further breakdown to plessite within each island (Yang et al. 1996), based on observations on the mesosiderite Chinguetti. The uniform cloudy zone orientation observed in this study strongly suggests that no further breakdown took place, unless the length-scale

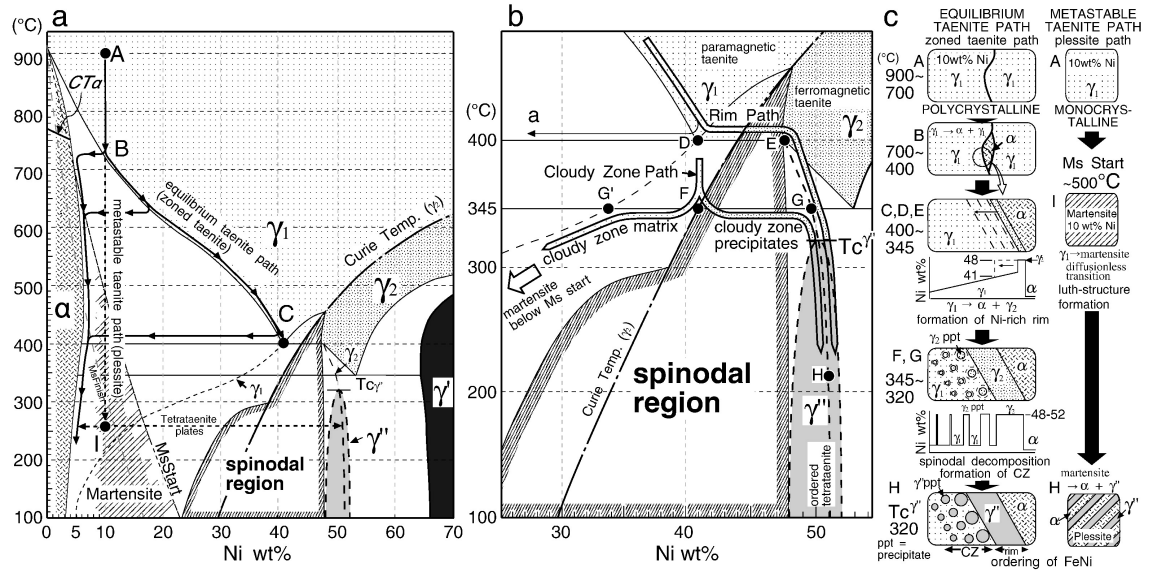


Figure 2.15: a. FeNi phase diagram (Uehara et al. 2011). b. Close up on the spinodal and tetrataenite regions, showing the evolution of the cloudy zone. As kamacite lamellae grow and reject Ni, taenite becomes compositionally zoned with high Ni next to the interface with kamacite (Fig. 2.5, points B and C on the phase diagram) due to Ni diffusion being much slower in taenite than it is in kamacite (Rasmussen et al. 1988). The high-Ni composition cools and reaches the spinodal region (point F), where it splits into high-Ni taenite (which will become the cloudy zone islands, point G) and low-Ni taenite (which will become the cloudy zone matrix, point G'). At point G, the mineral is still disordered taenite, and it orders to tetrataenite upon slow cooling through 320 °C ($T_c^{\gamma''}$) to form tetrataenite islands (point H). At the same time, the cloudy zone matrix evolves to a low-Ni composition, hypothesised to be antitaenite (Rancourt & Scorzelli 1995) or kamacite (Nichols et al. 2020). c. Schematics of cloudy zone and plessite formation. α = kamacite; γ_1 = paramagnetic taenite; γ_2 = ferromagnetic taenite; γ' = awaruite; γ'' = tetrataenite; Ms = martensite; $T_c^{\gamma''}$ = tetrataenite ordering temperature (320°C); $CT\alpha$ = Curie temperature of kamacite (750-770°C); CZ = cloudy zone.

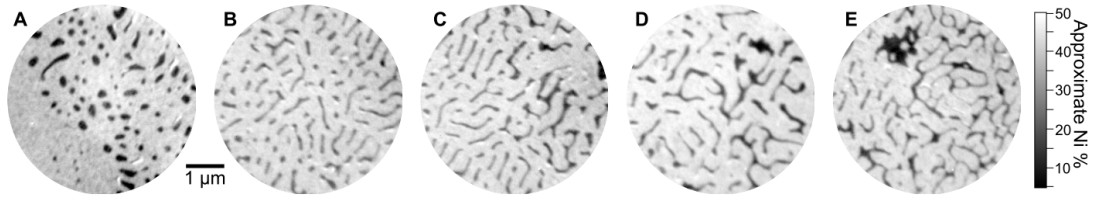


Figure 2.16: Compositional images of the Estherville cloudy zone taken with X-PEEM, showing cloudy zone formation through spinodal decomposition (Nichols 2017).

of the heterogeneity is below the spatial resolution of the map (70 nm). Even if that were the case, at least some pixels would be expected to show a different orientation, which is not observed.

2.4.3 Magnetic carriers

The coercivity spectrum in Fig. 2.12 provides evidence for high-coercivity material in Estherville, within the 200 - 600 mT range (Fig. 2.12). This material could carry a reliable magnetic signal that is not currently included in the magnetic fidelity determination, because any mineral with a coercivity above 200 mT would not be accessed by the ARM used to constrain the fidelity (Section 2.2.5.4). Several minerals in Estherville have the potential to carry this signal: metal (kamacite and taenite), tetrataenite, and schreibersite, and their magnetic properties are summarised in Table 1.1. In this section, I evaluate these minerals to determine which ones are the likely high-coercivity carriers in the sample, and then I review the efficiency of the different methods used to identify SD tetrataenite.

Metal Kamacite is the most abundant metal phase in mesosiderites, making up most of the metal portion of the meteorite (Fig. 2.2). As a result, it almost always occurs as multidomain, having low coercivity as a result. However, the coercivity of SD/SV metal (<28 nm) has been found to extend to 600 mT (Lappe et al. 2011), which suggests that the metal inclusions in pyroxene (Fig. 2.3B) could be a candidate for the 200 - 600 mT signal on the coercivity spectrum.

Tetrataenite The presence of multidomain tetrataenite was confirmed by the coercivity spectrum produced through IRM acquisition, as a peak on 100 mT previously identified by Gattacceca et al. (2014) and Bryson et al. (2017) (Fig. 2.12). Given the high magnetic coercivity of single-domain tetrataenite (ranging between 490 mT, determined by Nagata 1982 and 2 T, determined by Einsle et al. 2018), if it were present in the sample in detectable amounts, a prominent peak in the coercivity spectrum in Fig. 2.12 would be expected above 500 mT (e.g., Fig 3d from Bryson et al. 2017), which is not observed. Furthermore, the HC noise in the Zijdeveld diagrams is centered on the origin (Fig. 2.10). If SD tetrataenite were present, the noise in the Zijdeveld diagrams would be offset from the origin, as was observed in the silicate portions of the meteorite Steinbach (IVA), which were not magnetised but contained SD tetrataenite (Fig. 4b in Bryson et al. 2017). The coercivity spectrum and demagnetisation behaviour of Estherville both confirm that no SD tetrataenite is present.

Schreibersite In the exposed section described in Section 2.3.1, schreibersite was observed as a 200 μm exsolution lens within the metal (Fig. 2.4). This size indicates that it is in multidomain state, and therefore unlikely to carry a stable magnetisation. However, the presence of SD or vortex schreibersite cannot be ruled out elsewhere in the sample. Furthermore, since its coercivity has not been accurately constrained yet, it is not currently clear how schreibersite contributes to the coercivity spectrum of the sample. However, due to its low abundance (<2 vol.%, Powell 1971) and common occurrence in multidomain grains (Fig. 2.4B), it is unlikely to contribute significantly to either coercivity spectrum or magnetic record of mesosiderites.

2.4.4 Identification of tetrataenite

As tetrataenite is a very hard ferromagnet when present in single-domain or single-vortex state, reliable magnetic information can be extracted from it. This study

employed different methods to detect tetrataenite, and here I discuss their effectiveness.

Since tetrataenite has a narrow compositional range (50-55% Ni), EDS and EMPA were used to identify minerals with this composition. Tetrataenite forms the high-Ni end of the Fe-Ni line in Fig. 2.6B, being slightly offset towards lower Fe and Ni. This offset varies based on which other phase it shares a grain with (kamacite or troilite) within the silicates, suggesting potential contamination due to diffusion from the other phases, or error introduced by the instrument measuring signal from both phases.

As outlined in Section 2.3.1, high-Ni metal was identified in three forms in Estherville: within the cloudy zone present in high-Ni lamellae in the metal (Fig. 2.8), as micron-sized grains in the silicate portion of the sample (Fig. 2.4A), and as nanometric inclusions in pyroxene (Fig. 2.3B).

Since mesosiderites have very slow cooling rates, the high-Ni lamellae in kamacite are composed almost entirely of cloudy zone (with tetrataenite islands in a ‘matrix’ of a low-Ni phase) rather than the usual cloudy zone and plessite, and the cloudy zone tetrataenite islands grew into multidomain range (Nichols et al. 2020). Furthermore, all observed high-Ni crystals within the silicate mass are also too large to be SD (1-50 μm). However, as mentioned previously, pyroxene crystals contain arrays of inclusions. The inclusions are primarily troilite, but there is evidence for <100 nm high-Ni material present. Both EDS and EMPA spot sizes are too large to measure the composition of nanometric inclusions, so the measured composition includes some of the neighboring pyroxene. Since the composition of pyroxene can be independently measured, and since the data includes some elements which belong to pyroxene only (e.g., Mg and Si), the relative contributions from each phase can be broadly approximated, which then allows the isolation of the high-Ni grain only. A FeNi phase with 55% Ni and approx. 70 nm grain size was identified this way, but a higher resolution technique would be required to determine the definitive size and composition of the

phase. If the phase is tetrataenite, this size has been shown to carry a reliable magnetic signal within the cloudy zone (Bryson et al. 2014a). However, the resolution challenges make this approach far from ideal. Furthermore, even at higher resolution, the compositional overlap between taenite and tetrataenite would remain a problem, making chemical analysis techniques challenging to use to identify tetrataenite.

As outlined in Section 2.4.2, the tetrataenite and taenite lattices cannot be structurally distinguished from each other through EBSD because their lattice parameters are too similar, so EBSD is also not able to identify tetrataenite. Higher-resolution structural techniques such as TEM (transmission electron microscopy) have previously been used to establish the presence of the ordered tetrataenite structure (e.g., Lewis et al. 2014), but the extensive sample preparation makes it difficult to use routinely.

On the other hand, magnetic techniques have proven effective in establishing the absence of SD/SV tetrataenite, as described in the previous section. Therefore, the presence (or absence) of this mineral was readily established using magnetic techniques, making them more efficient at identifying tetrataenite than microscopy techniques.

As discussed in the section above, the coercivity spectrum (Fig. 2.12) and demagnetisation behaviour of Estherville (Fig. 2.10) together confirm that no SD/SV tetrataenite is present, which suggests that the pyroxene inclusions are likely to be SD/SV taenite. Since an ARM only approximates minerals with coercivities below the AF field used (200 mT), it does not take these inclusions into account, so the low magnetic fidelity of the bulk meteorite (Section 2.4.1) may not apply to the taenite inclusions. Future work could therefore target these inclusions by isolating their magnetic signal (e.g., using *in situ* magnetic mapping) to understand their remanence.

2.4.5 Implications for planetary magnetism

If Estherville is able to record a $150\ \mu\text{T}$ field (Section 2.4.1), the lack of stable HC component suggests that the mesosiderite parent body was not producing a field of this intensity when mesosiderites were cooling. There are two possible explanations for this: either the mesosiderite parent body did not have the physical properties required to produce a $150\ \mu\text{T}$ field, or the mesosiderite-forming impact prevented a field from being recorded (either because it was destructive and it shut off the dynamo, or because it occurred after the dynamo had already stopped). These possibilities are discussed below.

Firstly, I evaluate the circumstances that would allow a $150\ \mu\text{T}$ field to be produced in a planetesimal, since such a strong field is higher than would be expected from most planetary dynamos, unless the meteorite was located close to the field source. A summary of planetary bodies in our solar system and their respective field strengths is presented in Table 2.5, and it is noted that none of the terrestrial bodies listed generates a field strong enough to be recorded by mesosiderites. However, the field strengths in Table 2.5 were recorded close to or on the surface of the body, whereas mesosiderites are hypothesised to have been buried deeply into the parent body post-impact (Scott et al. 2001), exposing them to stronger fields. Furthermore, the present-day measurements presented for Mercury, Ganymede and Io may not be representative for the fields that these bodies generated in the early solar system. Detailed dynamo and thermal modelling for the mesosiderite parent body would be required, but this is largely unconstrained due to the lack of time-resolved palaeointensities allowing us to link the cooling rate in the mantle to dynamo activity in the core. Instead, I review existing models for dynamo generation on terrestrial bodies similar to the mesosiderite parent body and/or bodies capable of generating strong fields, to evaluate if they would be able to produce fields strong enough to be recorded by mesosiderites.

Terrestrial body	Field strength (μT)	Depth	Method	Reference
Mercury	0.45	Surface	Spacecraft measurement	Christensen (2006)
Mars	up to 77	Surface	Spacecraft measurement & Modelling*	Arkani-Hamed (2012)
Moon	up to 111	Regolith	Palaeointensity	Cournède et al. (2012)
Ganymede	0.75	Surface	Spacecraft measurement	Kivelson et al. (1996a)
Io	1.3	Surface	Spacecraft measurement	Kivelson et al. (1996b)
Pallasite parent body	up to 140	20-30 km depth	Palaeointensity	Nichols et al. (2021), Tarduno et al. (2012)

Table 2.5: Field intensities recorded in different terrestrial bodies in the solar system. *For Mars, measurements of crustal remanence were combined with modelling to estimate the strength of the dynamo that could have magnetised the crust.

Mantle-stripped asteroids Strong fields could be produced on terrestrial bodies that cooled very quickly, such as mantle-stripped asteroids and stripped cores, since the fast cooling would drive vigorous core convection. On an asteroid with a very thin mantle, the meteorites would necessarily be located close to the core, experiencing strong fields as a result. Modelling of dynamo evolution on such bodies was performed by Scheinberg et al. (2016) and found that a field strength up to $270 \mu\text{T}$ could be reached. However, a very thin mantle implies fast cooling, which is not compatible with the formation of mesosiderites, despite previous studies suggesting that the mesosiderite parent body could be asteroid (16) Psyche (Davis et al. 1999, Viikinkoski et al. 2018).

A large-cored, thin-mantled body A large-cored, thin-mantled planetary body was hypothesised for the pallasites, as they require strong fields to be produced to explain their palaeointensities and cooling rates (Nichols et al. 2021). This case is considered separately from mantle-stripped asteroids despite potentially being a result

of mantle stripping, because the thicker mantle (70 km; Nichols et al. 2021) could allow slow cooling. This theoretical body would generate up to 95 μT intensities in the mid-mantle, but it is possible that higher intensities could be achieved closer to the core-mantle boundary, which may be recorded by slower-cooled meteorites such as the mesosiderites, assuming that the slower cooling rate is due to formation at greater depth.

Vesta Modelling based on the case study of Vesta suggests that very strong fields (up to 1000 μT in the core) could be produced for a very short amount of time (~ 0.5 Myr), and that the field intensity would go down to 100 μT on the surface (Formisano et al. 2016). It is therefore possible that a dynamo could produce a field strong enough to be recorded by mesosiderites, provided that they were buried close to the core-mantle boundary of their parent body following the impact that injected the metal, which is consistent with their extremely slow cooling rates. In turn, this would require that the parent body has a relatively thin mantle, and/or that the impact energy scrambled the mantle such that the mesosiderites were buried close to the core. The lack of a remanence in mesosiderites then requires that these conditions were not met, since mesosiderites did not experience a strong field during cooling.

Alternatively, the absence of a field could be explained if core convection was shut off by a destructive impact that would scramble the entire body. The earliest estimate of the mesosiderite-forming impact is 4.525 Gyr as a hit-and-run collision of Vesta (Haba et al. 2019), which would be later than the period of intense field generation modelled by Formisano et al. (2016). The impact is therefore unlikely to have prevented the dynamo from operating in the first place, but it may have stopped it, thus preventing the metallic part of mesosiderites (inserted by the impact) from becoming magnetised. In this case, the thermal energy from the impact may have also reset a potential remanence recorded by the silicate parts.

Finally, the mesosiderite-forming impact may have occurred too late for the mesosiderites to record a field. Both U-Pb estimates of the age of the impact are within 42-46 Myr

after CAI (Haba et al. 2017, 2019), during a time when no palaeointensity data were recovered from any meteorite. This was hypothesised to be due to a period of no dynamo activity between thermal and compositional convection on planetesimals (Fig. 1.3; Bryson et al. 2019b, Dodds et al. 2021). The duration of thermally-generated dynamo activity depends strongly on the radius of the parent body (Bryson et al. 2019b), which is currently unconstrained for the mesosiderite parent body. Nevertheless, palaeomagnetic evidence from other meteorites supports the possibility that the absence of a field could be due to metal-silicate mixing (and therefore, mesosiderite formation) taking place after thermal convection stopped being able to generate a field. However, the possibility of a later, compositional dynamo remains open.

In summary, fields stronger than $150 \mu\text{T}$ could be experienced by rocks close to the core-mantle boundary of a planetesimal, according to dynamo models based on bodies similar to the mesosiderite parent body. No information on lower fields can be extracted from mesosiderites due to their low fidelity, but it is likely that mesosiderites were able to record strong fields, and they were also likely to have experienced them due to their slow cooling rates and inferred deep burial. The fact that they did not experience such fields could mean that the core was too small (not being able to generate a field strong enough), or that they were not buried closely enough to the core. Alternatively, it could be due to the timing or strength of the mesosiderite forming impact — a destructive impact could have stopped the magnetic field generation, while the timing places mesosiderite formation during a period when no palaeointensities could be recovered from any meteorite, possibly due to dynamo quiescence between thermal and composition convection (Bryson et al. 2019b, Dodds et al. 2021).

2.5 Conclusions

- The islands in the mesosiderite cloudy zone all have the same crystallographic orientation, suggesting that their orientation was not influenced by a magnetic

field if one was present at the time of tetrataenite ordering. This may be specific to mesosiderites because of their slow cooling rate and resulting coarse islands, so it may still be a viable mechanism to extract magnetic information from the cloudy zones of faster-cooled meteorites, as supported by data from Einsle et al. (2018).

- Tetrataenite is important in palaeomagnetism because of its high coercivity, so it would be valuable to be able to identify it efficiently in natural samples ahead of analyses. Chemical analysis using SEM/EDS and EMPA cannot identify tetrataenite due to its compositional overlap with taenite and due to resolution issues in the size range occupied by SD tetrataenite. Furthermore, structural analysis using EBSD may not be able to distinguish tetrataenite and taenite due to the small tetragonal distortion that takes place during atomic ordering. Higher-resolution techniques such as TEM may be able to identify it, but the extensive sample preparation prevents its routine use. On the other hand, magnetic methods which distinguish phases based on their coercivity (e.g., IRM acquisition) can readily establish whether tetrataenite is present, making them an easier and less expensive method to identify this mineral.
- Bulk palaeomagnetic measurements of Estherville show that the recording fidelity of mesosiderites is $\sim 150 \mu\text{T}$, and the lack of a stable HC signal suggests that the meteorite has not experienced this field while it was cooling. Such a strong intensity could be achieved on terrestrial bodies similar to the mesosiderite parent body (e.g., Vesta; Formisano et al. 2016) close to the core-mantle boundary. However, mesosiderites did not record this field, which could mean that the parent body was too small to generate it, that mesosiderites did not form close to the core-mantle boundary where the fields are strongest, or that the timing or strength of the mesosiderite-forming impact prevented a remanence from being recorded.

- The coercivity spectrum of Estherville suggests that high-coercivity (200 - 600 mT) magnetic carriers are present in the sample, likely in the form of taenite inclusions within pyroxene. Future work could employ *in situ* magnetic imaging techniques such as SQUID microscopy or quantum diamond microscopy to access the magnetic recordings of these grains.

Chapter 3

The origin of pallasites

3.1 Introduction

“If one were to write a geological Alice in Wonderland and try to think of the most handsome and yet improbable specimens, pallasite meteorites would be strong contenders.”

- Buseck (1977)

3.1.1 Pallasites

Pallasites are stony-iron meteorites composed of olivine crystals encased in a predominantly FeNi metal matrix with minor sulfides and phosphides. Pallasites are further classified into Main Group pallasites and Eagle Station pallasites.

Although they are primarily composed of only two components (metal and olivines), a wide variety of textures have been identified in Main Group pallasites (referred to as "pallasites" from here on). McKibbin et al. (2019) summarise the textures based on a study of 30 pallasites, and they find three types of olivines: rounded, angular and fragmental. Olivine crystals can also form aggregates. Angular and fragmental olivines form through fracturing of olivine aggregates, which is commonly accepted to have occurred due to a deformation event (Scott 1977). On the other hand, olivine rounding occurs during high-temperature annealing leading to the reduction of olivine surface area to minimise interfacial energy between olivine and the metal melt (Scott

1977). A pallasite usually displays one or two of these olivine morphologies, and they can sometimes display all three, as is the case in the Seymchan (Boesenberg et al. 2012) and Brehm meteorites (Buseck 1977, Scott 1977). A complex formation history is required to explain why olivines that are sometimes found next to each other seem to have experienced such different formation mechanisms and thermal histories.

Following the olivine morphologies, the metal is either found as large veins that often separate olivine aggregates, or as small, isolated pockets within the aggregates. Walte et al. (2020) identified three metal pocket morphologies: (1) round metal inclusions in olivines, typically mm-sized, (2) small ($\sim 1\text{-}5$ mm), equant, high-dihedral angle pockets at olivine triple/quadruple junctions, and (3) larger ($\sim 2\text{-}10$ mm), irregularly shaped pockets with four or more adjacent olivines. They find that the shape of each pocket is determined by the surface energy of the metal melt, which is minimised in order to achieve the most energetically favourable configuration. The low surface-to-volume ratio of large pockets makes them more likely to be distorted by ongoing grain growth (Walte et al. 2003), resulting in an irregular shape bounded by four or more olivine crystals, in contrast to the equant shape of the small pockets, which are often found at olivine triple or quadruple junctions (Walte et al. 2020). Locally, the pockets are connected by veinlets, which can also be truncated and smoothed out by annealing in some pallasites (e.g., Esquel and Fukang; Walte & Golabek 2022).

The metal/olivine association has led to the traditional view that Main Group pallasites formed at the core-mantle boundary (CMB) of a planetesimal (e.g., Wood 1978, Boesenberg et al. 2012). However, this theory has been difficult to reconcile with the density contrast between olivine and metal, which is expected to lead to gravitational segregation (this is known as “the pallasite problem”, e.g., Mittlefehldt 1999). Furthermore, recent palaeomagnetic evidence has shown that a strong magnetic field is recorded by the pallasites, both in taenite inclusions within olivines (Tarduno et al. 2012) and in the tetrataenite islands within the metallic matrix (Bryson et al. 2015, Nichols et al. 2016, 2021), which indicates the presence of an internally-generated

magnetic field. This is not consistent with a CMB origin for pallasites because a dynamo requires a molten core (Section 1.2.4), with a temperature above the melting point of FeNi (between 988 and 1538°C, depending on sulfur content; Scheinberg et al. 2016). The tetrataenite ordering temperature (320°C), and taenite blocking temperature (350 - 400°C; Funaki et al. 1986) are impossible to reconcile with a molten core if pallasites formed at the core-mantle boundary, due to the unrealistically large temperature gradient that would be required across the boundary. Furthermore, the spread in cooling rates measured in pallasites precludes an origin at the same depth in the parent body (2.5 – 18 K/Myr; Yang et al. 2010). Since pallasites did not form at the CMB, intensive research has been conducted to understand their origin.

Another MG pallasite formation theory is that of an impact with another differentiated body (Scott 1977, Tarduno et al. 2012, Kruijer et al. 2022), which could be a hit-and-run collision where the mantle of the smaller body is removed resulting in a series of metal-enriched small bodies (Asphaug et al. 2006), or where the impactor’s metal core is inserted into the silicate mantle of the larger body (Kruijer et al. 2022) (Fig. 3.1A, B). Alternatively, Yang et al. (2010) suggested that a destructive impact formed a separate pallasite planetesimal, but such an impact would be difficult to reconcile with a convecting outer core, to satisfy the constraints from palaeomagnetism.

A hybrid formation scenario was recently proposed by Walte et al. (2020). Based on textural evidence and deformation experiments, they argue that pallasites formed in a partially differentiated mantle, where the metal melted and started forming a core (Fig. 3.1E). As the temperature increased due to radiogenic heating, the silicates partially melted and started migrating towards shallower depths, closing off the metal network (Cerantola et al. 2015), and leaving behind olivines with trapped metal pockets. An impact with a differentiated body then took place and caused an influx of external core metal (Fig. 3.1F), creating the matrix metal, and leading to two generations of non-mixing metal in pallasites: a first generation of primary

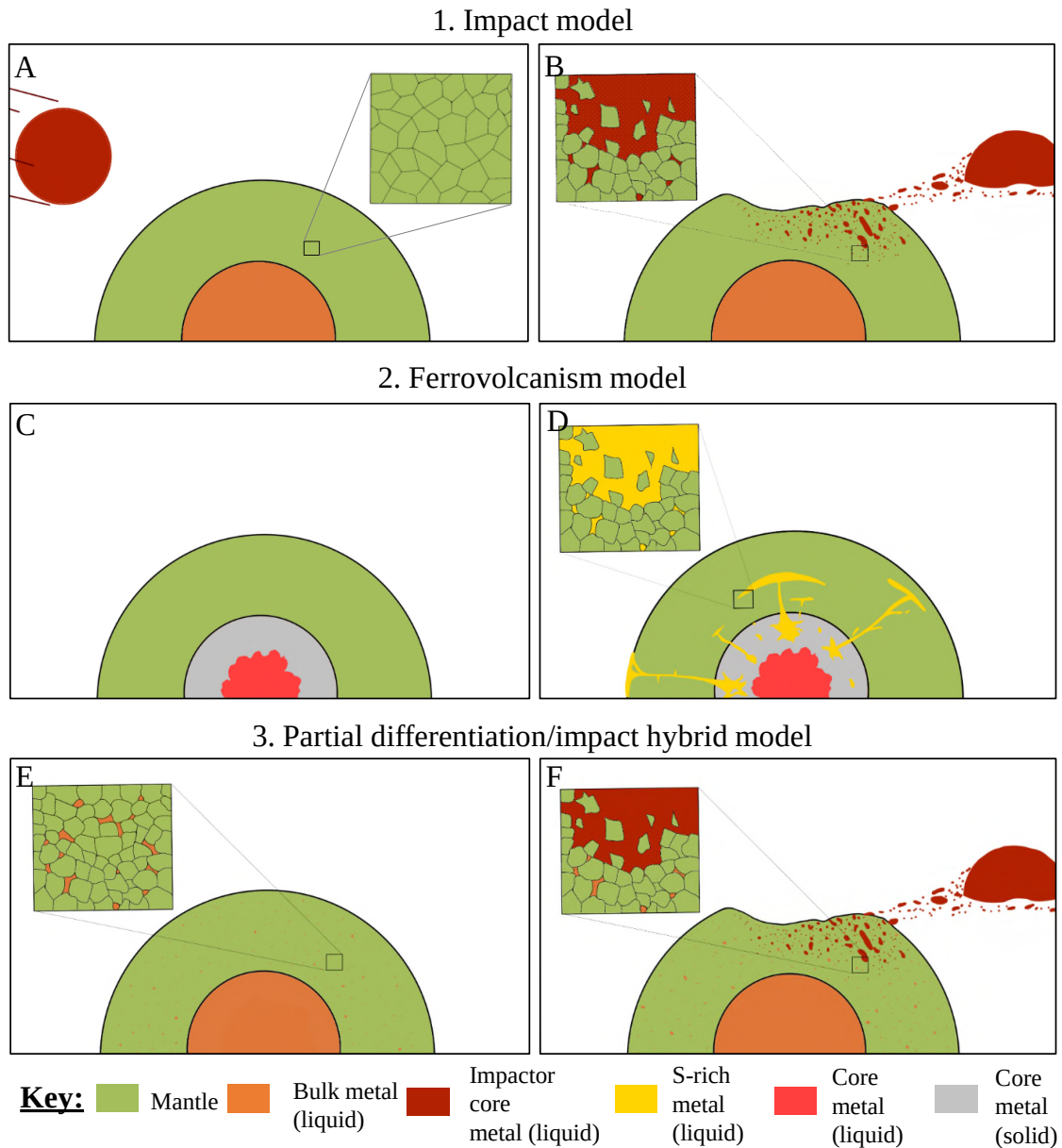


Figure 3.1: Summary of formation models proposed for Main Group pallasites. A, B. Impact model (here pictured as a hit-and-run collision) which would likely lead to all metal in pallasites having the composition of the impactor's metal. C, D. Ferrovolcanism model, where volatile-enriched metal is extruded from a core crystallising from the top down. This is also likely to lead to the same composition in all metal in pallasites. E, F. Partial differentiation/impact hybrid model, where the mantle of a partially crystallised planetesimal contains metal of chondritic composition that did not take part in core formation (E). An impact causes influx of metal of a different composition, leading to two metal compositions in pallasites (F).

metal, trapped in isolated pockets within olivine aggregates, and a second generation of external core metal brought by an impact (Walte et al. 2020, 2023). Alternatively, the two generations of metal could also come from multiple impacts (Walte et al. 2020, Walte & Golabek 2022). The aim of this chapter is to test this hypothesis by measuring the composition of the metal in pallasites, since two metal populations would be expected to have different compositions (one of which is the chondritic composition, in the partial differentiation scenario).

Finally, Johnson et al. (2020) proposed ferrovolcanism as a formation scenario for pallasites (Fig. 3.1C, D). In small planetary bodies, top-down core crystallisation may occur, trapping liquid metal in the middle (Williams 2009). The liquid becomes lighter than the solid due to rejection of light elements from the growing solid core, and it can buoyantly rise out of the core, as long as the overburden from the mantle is small enough (Abrahams & Nimmo 2019). This leads to the formation of metallic dykes in the mantle or crust of a planetesimal (Johnson et al. 2020).

3.1.2 Pallasite-IIIAB link

Main Group Pallasites have been proposed to be genetically linked with IIIAB iron meteorites. The evidence for this link includes similar isotopic signatures (oxygen isotopes: Clayton & Mayeda 1996, Greenwood et al. 2015; Mo isotopes, Burkhardt et al. 2011, Kruijer et al. 2022; S isotopes: Dottin III et al. 2018, Kruijer et al. 2022), metal composition (Wasson & Choi 2003), and HSE systematics (Mullane et al. 2004). One example of similar metal and HSE systematics is shown in Fig. 3.2, which shows the Ir and Ni contents of pallasites and different iron groups, with good overlap between pallasites and IIIAB irons.

On the other hand, Scott et al. (2009) argue that the isotopic similarities are not definitive evidence, because this isotopic space is very crowded, and pallasites and IIIABs are also overlapped by other groups (e.g., angrites, an ungrouped eucrite). Yang et al. (2010) argue that the similarity in metal composition is also not definitive

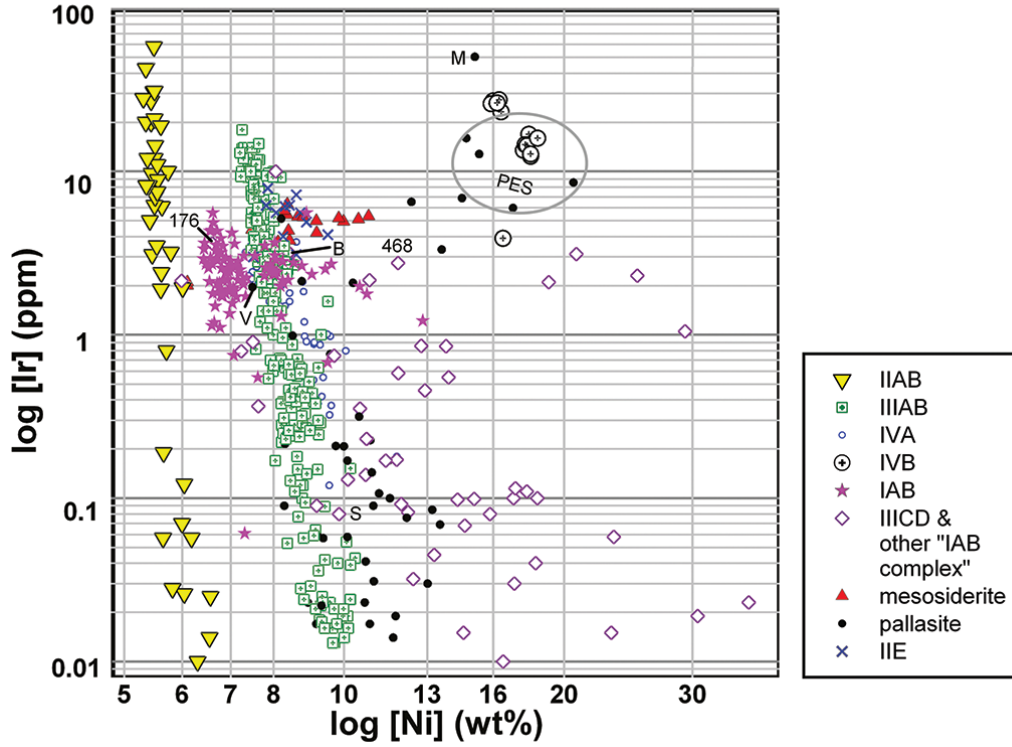


Figure 3.2: Ni vs. Ir plot of different iron and stony-iron meteorite groups (Ruzicka et al. 2017 and references therein)

evidence, since the pallasites occupy a wide range of compositions, especially in Ge and Ga. Evidence against this genetic link further includes shock stages and reheating indicators (many IIIABs are reheated and/or shocked (Buchwald 1975), which is not the case for pallasites, Yang et al. 2010). Cosmic ray exposure ages (Eugster et al. 2006) and cooling rates (2.5 - 18 K/Myr in pallasites, 50 - 350 K/Myr in IIIAB, Yang et al. 2010) also argue against the link. However, the cooling rates of the two groups have been reconciled in the recent study by Kruijer et al. (2022), who explain the distinct cooling rates through a hit-and-run collision where the impactor is the IIIAB parent body, whose metal is inserted into the mantle of a different asteroid.

3.1.3 Open questions & Chapter outline

As outlined above, the primary open question surrounding pallasites is their formation mechanism, which must satisfy the diversity of textural features found in pallasites.

Any formation model must also address why the density contrast between olivines and metal did not lead to gravitational separation (the "pallasite problem"). Furthermore, the link to the IIIAB iron meteorites remains an open question (Section 3.1.2).

In this chapter, I present a textural and chemical study of the metal in Seymchan, a Main Group pallasite, aiming to answer these questions. To distinguish between the proposed formation mechanisms for pallasites, I identify the two proposed populations of metal (Section 3.3.1) and test the hypothesis that they formed on different parent bodies, using high-precision *in situ* measurements of trace element composition.

I show that all metal measured in Seymchan has identical composition (Sections 3.3.2 and 3.4.1), which is not consistent with the formation model proposed by Walte et al. (2020), as their model predicts distinct compositions between matrix and pockets. I also suggest that the metal has undergone magmatic fractionation leading to a deviation from chondritic element abundance, which is not consistent with metal pockets being primary (Section 3.4.1.1). Furthermore, I measure the composition of a IIIAB iron meteorite (Section 3.3.3) and show that it has compositional overlap with the metal in Seymchan, lending further support to the proposed link between pallasites and IIIAB irons (Section 3.4.2). Finally, I discuss implications for the formation of pallasites and suggest that pallasites formed through at least two impacts, the first of which caused an influx of metal (likely of IIIAB composition) in the mantle of the pallasite parent body (Section 3.4.1). The second impact caused the secondary deformation features observed in this study and in previous work (e.g., Walte & Golabek 2022) without any influx of material.

3.2 Methods

3.2.1 Sample preparation

A slab of the meteorite Seymchan was provided by the Oxford University Museum of Natural History (specimen number Mt.519) and two slices were cut using a Buehler

Isomet low-speed saw with a 400 μm diamond-impregnated brass blade, using ethane-
diol for lubrication. The slices were cleaned with isopropanol following the cutting,
and they were then cut into a total of five segments (Fig. 3.3), which were mounted in
epoxy resin using a Struers EpoFix kit. One extra chip containing a metal pocket was
also mounted with one of the segments. The subsampling was designed to maximise
the number of individual metal pockets that can be measured, while also including
some of the matrix metal. The rock cutting was performed by Matthew Beverly-
Smith in the Earth Sciences Department, and I did the mounting in resin and further
sample preparation, unless otherwise stated.

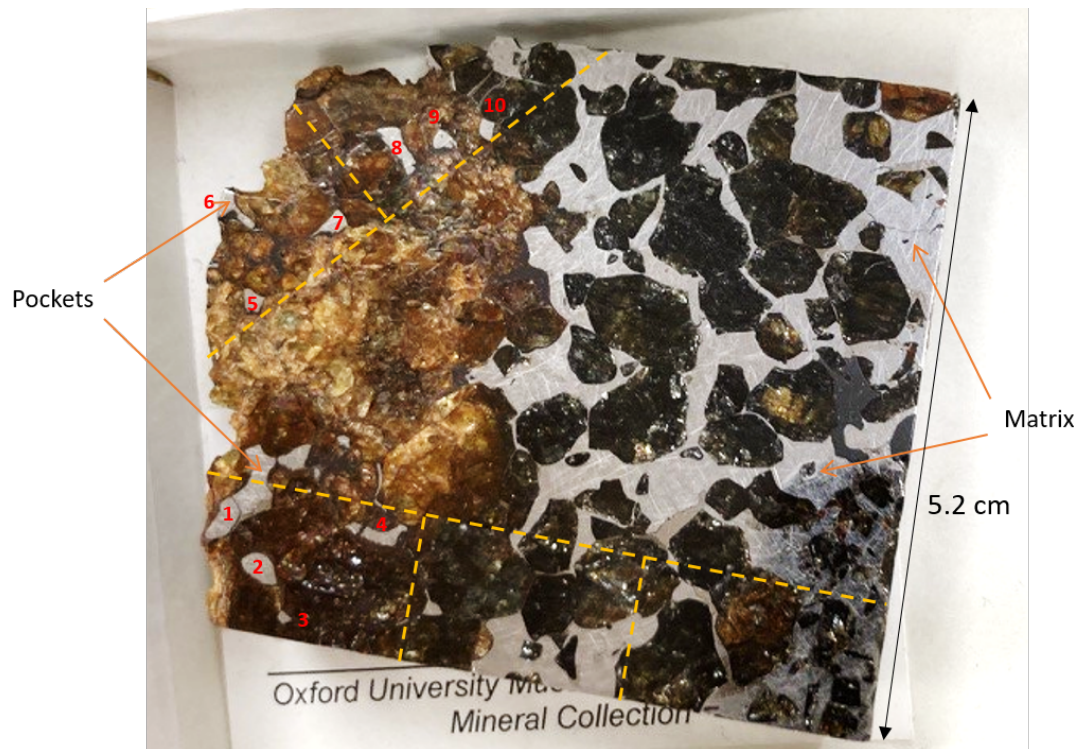


Figure 3.3: Photograph of Seymchan before sample preparation, courtesy of the Oxford University Museum of Natural History, showing the two main metal textures (matrix and pockets). The yellow lines show where the cuts were made to create five subsamples.

The mounted samples were polished to 1 μm using a SupaFinish 1 diamond compound with oil for lubrication on a standard polishing pad, cleaning with ethanol in between polishing stages. At the end of the polish, I cleaned the sample with ethanol

in an ultrasonic bath for 60 seconds. Before electron probe analysis, the sample was coated with a 20 nm carbon coat. The coating was removed before LA-ICP-MS analysis by repolishing down to 1 μm , using the same steps as above.

A polished and mounted sample of the meteorite Coahuila (IIAB) was also borrowed from the Oxford University Museum of Natural History (specimen number Mt.179) to use as internal standard for LA-ICP-MS (Section 3.2.2.3), and repolished using the same steps as above. Before electron probe analysis, Coahuila was also carbon coated.

A sample of Henbury (IIIAB) was also borrowed from the same museum (specimen number Mt.164), to use as compositional reference for the IIIAB irons, and to compare it to the composition of the metal in Seymchan. The sample is covered in fusion crust, and a corner was cut and mounted in an epoxy resin mount using the same method as above. In addition, to prevent scratching from fragments of the fusion crust coming loose, the sample was surface impregnated with resin, ground using a diamond abrasive plate and hand-lapped with a silicon abrasive powder. This sample preparation was also performed by Matthew Beverly-Smith in the Earth Sciences Department. Henbury was then polished down to 0.05 μm in two steps: 0.3 μm with aluminium oxide for 10 minutes, then colloidal silica polishing for 2h on a Vibromet vibrating polisher.

The mounted sample of Henbury was later etched with nital (solution containing 95% ethanol and 5% nitric acid) to emphasise the Widmanstätten pattern of the meteorite and aid the identification of different phases when sampling using the LA-ICP-MS. A fine polish was required ahead of the etching, to prevent surface imperfections obscuring the underlying features. Before electron probe analysis, this sample was also carbon coated.

3.2.2 Laser Ablation Inductively-Coupled Plasma Mass Spectrometry (LA-ICP-MS)

3.2.2.1 Background

Laser Ablation Inductively-Coupled Plasma Mass Spectrometry (LA-ICP-MS) is an *in situ* chemical analysis method, able to measure the composition of trace elements to part-per-billion precision (Gray 1985). This analysis was performed in the Department of Earth Sciences at the University of Cambridge, with help from Dr. Jason Day, who advised on the method and set up the measurements.

LA-ICP-MS starts with ablation of material at a chosen spot using a laser. The ablated and vapourised material is moved from the laser unit using He, and transported to the ICP-MS unit, where Ar plasma created by the ICP torch ionises the components of the ablated material. The newly formed ions are transported to the quadrupole mass spectrometer using charged metal plates, and the ions are filtered based on their mass-to-charge ratio. Since most ICP ions are singly charged, ICP-MS produces a mass spectrum.

The ability of LA-ICP-MS to resolve spatial heterogeneities in chemical composition to high precision makes it ideal for an *in situ* study of meteoritic metal, allowing the distinction between different metal textures to be maintained in the data. The two previously identified metal textures mentioned above (metal pockets and matrix) are sampled individually, distinguishing between kamacite and taenite in each case.

3.2.2.2 Isotopic analysis

The isotopes measured were ^{57}Fe , ^{59}Co , ^{60}Ni , ^{62}Ni , ^{63}Cu , ^{69}Ga , ^{72}Ge , ^{182}W , ^{185}Re , ^{189}Os , ^{193}Ir and ^{197}Au . These elements were chosen as they have all previously been used to distinguish different iron meteorite groups (e.g., Scott & Wasson 1975). When choosing which isotopes to measure for elements where more than one stable isotope is available, several factors were considered, including isotope abundance (where possible, the isotopes with the highest natural abundance were measured for the trace elements), as well as avoiding different types of interferences:

1. Isobaric interferences, where two isotopes with the same atomic mass cause interference in the mass spectrometer (no two isotopes with the same atomic number were measured).

2. Polyatomic interferences, where elements react to form compounds of higher mass inside the plasma, which could cause interference with higher-mass isotopes (e.g., ^{56}Fe has the same mass as $^{40}\text{Ar}^{16}\text{O}$, so ^{57}Fe was measured instead).

3. Double charged ions, where a portion of the atoms with lower ionisation potential (e.g., rare earth elements, alkaline metals) are doubly charged and could be filtered into the wrong collector based on their mass/charge ratio. No evidence for interference due to double charging was detected.

After known interferences were avoided, the isotopes with the highest natural occurrence were chosen in order to maximise the signal detected. One interference was unavoidable (^{72}Ge with 2 atoms of ^{36}Ar), so we briefly measured ^{73}Ge as well, despite its low natural abundance (approx. 7%). The results from ^{72}Ge and ^{73}Ge were indistinguishable from each other, and since ^{36}Ar has very low natural abundance (<1%), it was concluded that the interference was negligible and that the ^{72}Ge results are reliable. ^{73}Ge was therefore removed from the main set of measurements to reduce measurement time and increase the number of data points.

3.2.2.3 Calibration

I used a sample of Coahuila (IIAB iron meteorite) for external calibration, using previously reported data on its trace element composition for the minor elements (Table 3.1), and independently acquired electron probe data for the major elements. As a hexahedrite, Coahuila is composed almost entirely of kamacite and it has a relatively homogenous composition, making it ideal for calibration of metallic materials. Samples of Coahuila have previously been used for external calibration of LA-ICP-MS studies on iron meteorites by Ash et al. (2007), Hilton et al. (2020), Tornabene et al. (2020) and others. Furthermore, as an iron meteorite, Coahuila has a similar composition to the sample of interest.

Furthermore, independent electron probe measurements of the Ni content of each laser spot were taken after the LA-ICP-MS analysis (Section 3.2.3), to be used for internal calibration. The internal calibration uses known Ni concentrations (measured in ppm) to convert the LA-ICP-MS output (measured in counts-per-second) to concentration. This serves a dual purpose: firstly, to correct for heterogeneities within a sample (especially due to kamacite/taenite partitioning); and secondly, to correct for fluctuations in the laser power during a measurement run. The Ni content for each LA-ICP-MS measurement was averaged using 3-4 probe measurements, as shown in Fig 3.4. Two isotopes of Ni were measured using LA-ICP-MS, the first of which (^{60}Ni) was used as internal standard and adopted averaged measurements taken on the electron probe. The second isotope (^{62}Ni) was used to validate the calibration, as its measured values are expected to be very similar to the internally calibrated ^{60}Ni values.

3.2.2.4 Measurement parameters

The LA-ICP-MS measurements were taken with an 80 μm laser spot size, with 30% laser fluence. The large spot size was required to obtain measurable signals for trace elements in metal, due to the high thermal conductivity of metal dispersing the heat from the laser. I used 1 ms dwell time for Fe and Co, 10 ms for ^{62}Ni , Cu and Ga, 20 ms for ^{60}Ni , and 100 ms for all other elements. To ensure the accuracy of ^{60}Ni measurements used for calibration, a longer dwell time was used to measure this isotope. The detection limits vary between different measurements of each element, but can be approximated as follows: 207 ± 109 ppm (Fe), 14 ± 6 (^{60}Ni), 28 ± 15 (^{62}Ni), 58 ± 64 ppm (Co), 2.2 ± 0.8 ppm (Cu), 1.1 ± 0.5 (Ga), 3.6 ± 1.3 (Ge), 0.5 ± 0.2 (W), 0.45 ± 0.51 (Os), 0.8 ± 0.2 (Re), 0.1 ± 0.2 (Ir), 0.2 ± 0.4 (Au). The individual detection limit for each measurement can be found in the dataset included with this thesis. Most of the Re measurements were below detection limit, so Re was excluded from the analysis. For the other elements, only the specific measurements below detection limit were excluded from the analysis and from the averaging method

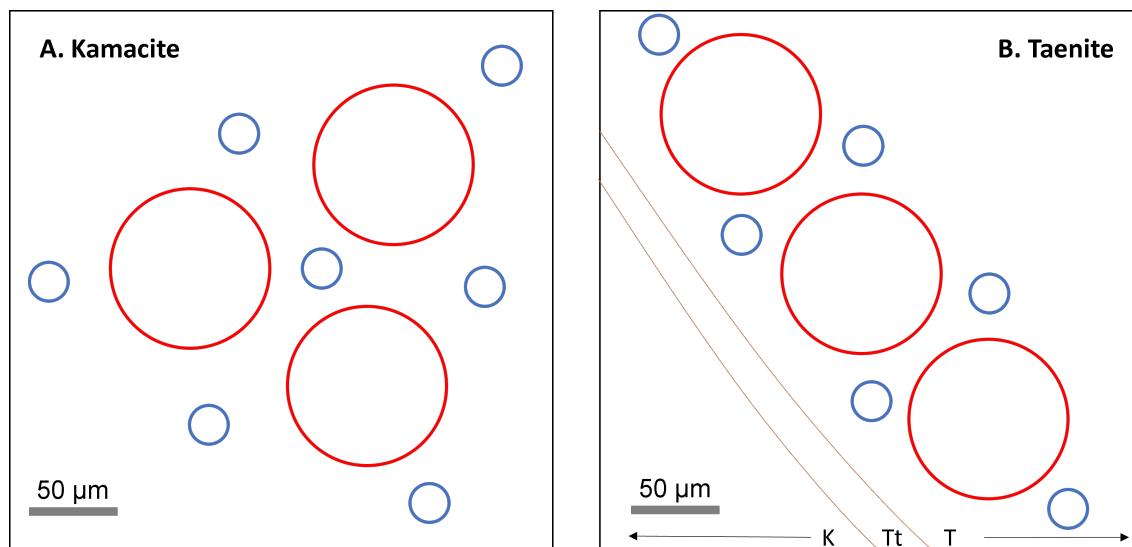


Figure 3.4: Schematic of the general sampling strategy in kamacite (A) and taenite (B), where red circles represent LA-ICP-MS spots (80 μm diameter) and blue circles represent EMPA spots (20 μm diameter). Laser spots were generally arranged as a triangle in kamacite, as kamacite areas are large. The spatial extent of taenite is more limited, so spots in taenite were usually arranged as a line parallel to the tetrataenite rim. Changes to both of these configurations were occasionally made, in order to match the morphology of the sample. The EMPA spots surround the laser spots and the Ni composition for calibration was obtained by averaging the EMPA spots adjacent to each laser spot.

described in Section 3.2.4.5. One exception was made for cluster analysis, because cluster analysis requires all measurements from a laser spot to be above detection limit – detection limits in this case are treated as described in Section 3.2.4.1.

An instrumental attenuation was applied to the major elements, to allow them to be measured without oversaturating the detectors. The same attenuation is used for measurements on the samples of interest and on the external standard, so it does not affect the results, because the counts-per-second to concentration ratios remain the same.

3.2.2.5 Sampling strategy

A total of 222 spots were measured in Seymchan, along with 36 in Henbury (31 in kamacite and 5 in taenite) and 51 in Coahuila. In Seymchan, 49 spots were taken in the matrix (36 in kamacite and 13 in taenite), and 173 spots were taken in the

Element	Concentration	Reference
Fe	$93.215 \pm 0.652\%$	This study
Ni	$5.229 \pm 0.428\%$	This study
Co	$0.615 \pm 0.014\%$	This study
Cu	122 ppm	Smales et al. (1967)
Ga	60.7 ppm	Wasson (1969)
Ge	189 ppm	Wasson (1969)
Ir	15 ppm	Wasson (1969)
Au	0.489 ppm	Petaev & Jacobsen (2004)
W	2.869 ppm	Petaev & Jacobsen (2004)
Re	1.203 ppm	Petaev & Jacobsen (2004)
Os	9.48 ppm	Petaev & Jacobsen (2004)

Table 3.1: Bulk composition of Coahuila used for LA-ICP-MS calibration. The major elements (Fe, Ni, Co) were independently measured on the electron probe, but the other elements could not be measured due to their small concentrations, so literature values were used instead. Since Coahuila is a hexahedrite, it is composed entirely of kamacite and generally considered very homogeneous. Therefore, these values are unlikely to be different among different samples.

pockets (91 in kamacite and 82 in taenite). The different number of measurements in the pockets and matrix is partly due to the subsampling strategy, which was designed to maximise the number of exposed pockets, allowing 11 individual pockets to be sampled (Section 3.2.1), and partly due to the requirement to have a statistically significant number of measurements in each pocket, to distinguish different trace element compositions if they exist.

Three laser spots were taken in each location, arranged as a triangle in kamacite and as a line in taenite (Fig. 3.4). This method aims to increase redundancy in case some of the measurements fail, for example due to the unknown 3D structure of the sample, if subsurface chemical heterogeneity is sampled by the laser. This could introduce scatter in the data, since the EMPA internal calibration is based on the surface composition (as the interaction volume of electrons is smaller than the laser interaction volume), so subsurface deviations from the surface composition are not calibrated for. The time-resolved LA-ICP-MS signal was monitored in each spot, and if a change in phase was detected as the laser advanced through the sample, the change would be filtered out by adjusting the interval that the signal is averaged over.

If the interval was too small to allow this, the measurement was discarded.

3.2.3 Electron Microprobe Analysis (EMPA)

For quantitative element analysis of the metal in both Seymchan and Coahuila, I used the CAMECA SX-5 FE electron microprobe at the Department of Earth Sciences, University of Oxford. This analysis was performed with assistance from Dr. Andrew Matzen, particularly for calibration. EMPA was performed after LA-ICP-MS analysis on Seymchan, Coahuila and Henbury. On Coahuila, EMPA was used to determine the major element composition (Fe, Ni and Co; Table 3.1) for external calibration. On Seymchan and Henbury, EMPA was used after the LA-ICP-MS analysis to measure the Ni content in each laser spot, for internal LA-ICP-MS calibration (Section 3.2.2.3).

To measure Ni post-LA-ICP-MS analysis, I used the following operating conditions: 15 kV accelerating voltage, 40 nA beam current, 20 μm spot size and 30 s counting time. The detection limits were approximately 350 ppm (Fe), 500 ppm (Ni), and 270 ppm (Co).

The electron probe cannot be used to probe inside a laser spot, as the signal from a laser spot would not be reliable due to the difference in relief and lack of polish inside the laser spot. Therefore, an averaging method was devised, which involves surrounding a laser spot with several probe spots, and averaging between them (Fig. 3.4). A large electron spot size (20 μm) was adopted in order to average the signal over a wider area, to avoid small-scale heterogeneities that are not representative of the large area that a laser spot covers. The location of the probe spots was taken a few microns from the laser spots, to maximise similarity between the metal composition inside and outside the laser spot, while minimising the risk of the electron beam drifting and hitting the interior of a laser spot, and avoiding potential effects of heating and volatile element migration.

An overlap correction between Co and Fe was applied due to the slight overlap of the Fe $k\beta$ 1 and Co $k\alpha$ peaks, using Fe metal for the correction. The calibration was

performed using natural and synthetic reference materials.

3.2.4 Statistical methods

3.2.4.1 Cluster analysis

To determine whether all the metal forms one compositional cluster, or if any compositional substructures are present, the cluster analysis method of k-means clustering was used to partition the chemical data into k mutually exclusive clusters (Lloyd 1982). In this method, the data is partitioned by minimising within-cluster variance (the sum of the squares of distances between each data point and the centroid of the cluster). K-means clustering was performed using 5 replicates, meaning that the clustering is repeated 5 times with new initial centroid positions, and the clustering configuration with the lowest within-cluster variance is returned. This gives greater confidence in the results as it allows the function to improve the fit after each replicate. The clustering was performed using in-built functions included in the MATLAB Statistics and Machine Learning Toolbox.

In order for cluster analysis to be applied on the entire dataset, all elements must have an equal number of measurements. This is not always the case due to the measurements of some elements (e.g., Re, W, Os) often being below detection limit, leading to fewer measurements. To mitigate this problem, Re was removed from the dataset for cluster analysis, since the number of Re measurements below detection limit exceeds 50% of all measurements. All other elements were kept in the dataset even though some still contain measurements below detection limit (W, Os, Ir, Au), but the laser spots with any measurement below the limit were removed (e.g., if a laser spot only has Au below detection limit, the spot is removed, which also removes its measurements of the other elements). This is to maintain an equal number of measurements for all elements, and it leads to a reduction in data points from 195 to 84, for 10 elements (this is referred to as the "reduced" dataset). In addition to analysis of the reduced data set, cluster analysis was also performed only on the

elements which have all their measurements above detection limit (Fe, Ni, Co, Cu, Ga, Ge), where the total number of data points is 195 – this is referred to as the "6-element" dataset.

Finally, cluster analysis is applied on data for kamacite and taenite individually, to check for clustering of data within each phase, and to calculate individual pocket means.

3.2.4.2 Clustering evaluation

K-means clustering requires the number of clusters to be specified ahead of clustering, as opposed to being the result of the algorithm. Two additional methods are used to determine the optimal number of clusters:

- Silhouette statistics (Rousseeuw 1987)
- Gap statistics (Tibshirani et al. 2001)

Silhouette statistics measure how similar a datapoint is to other points within its own cluster, combined with how dissimilar it is to points in other clusters. Values range between -1 and 1, and a high value suggests that a datapoint is well matched to other data in the same cluster, and poorly matched to data in other clusters. One silhouette is computed per cluster and shown on a silhouette plot, which is a histogram of silhouette values for a given number of clusters. On silhouette plots, all silhouettes having positive values indicates a good clustering configuration, whereas low or negative values indicate poor clustering. The arithmetic mean of all silhouette values in a clustering configuration is reported as an overall metric for the quality of clustering. An example of k-means clustering evaluated using silhouette statistics is shown in Fig. 3.5, and the mathematical details of this method are given in Appendix B.1.1.

K-means clustering and silhouette statistics evaluate at least two clusters, and in this study it is sometimes helpful to establish if there is no clustering (i.e., if the

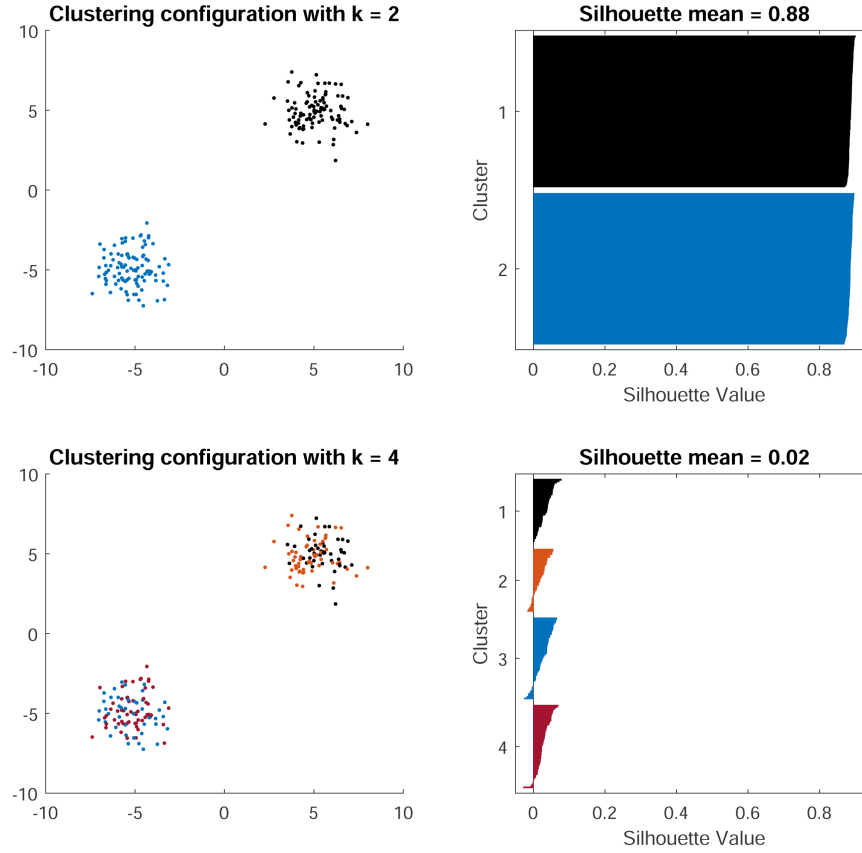


Figure 3.5: Example of k-means clustering evaluated using silhouette statistics. K-means clustering is performed on a dataset containing two randomly generated normal distributions with means of -5 and 5, and standard deviations of 1. The top set of plots shows k-means clustering with $k = 2$ (2 clusters), and the associated silhouette plot with a mean of 0.88. The bottom set of plots shows the same dataset, partitioned into 4 clusters ($k = 4$), with a resulting silhouette mean of 0.02. The silhouette plot for the second case has much lower values, as well as negative values, which indicates overlap between clusters. The silhouette means indicate that the optimal number of clusters is 2. In both cases, the colours on the plots correspond to each cluster.

data partitions into only one cluster), which is not supported by silhouette statistics. For this reason, I also use gap statistics, which are able to evaluate a clustering configuration with only one cluster (Tibshirani et al. 2001).

Gap statistics formalise the traditional "elbow" method for identifying optimal clustering. The elbow method involves plotting the within-cluster variance of the data against the number of clusters in a configuration, to identify the break in the slope of

the curve, which is assumed to correspond to the optimal number of clusters. Since the break in slope is not always clear (often sloping gradually rather than changing sharply in one point), the elbow method is generally considered unreliable. Gap statistics improve this method by comparing the real variance to the variance of data that does not contain any clustering, generated through Monte Carlo simulations. The optimal number of clusters is the point where the difference between the real variance and the reference variance is greatest. The mathematical details of this method are given in Appendix B.1.2.

Silhouette and gap statistics are used to determine the optimal number of clusters for all the datasets, evaluating configurations of 1 to 6 clusters.

3.2.4.3 Data standardisation

Before cluster analysis, the data were scaled to remove the bias which would otherwise be introduced by some elements having much higher concentrations than others (e.g., Fe could make up 95% of the sample, while Au makes up 1 ppm). Scaling was achieved by subtracting the mean of the dataset from each point, and then dividing each result by the standard deviation of the dataset. This results in a mean of 0 and a standard deviation of 1 for each element, giving each element equal weight in the cluster analysis.

3.2.4.4 Additional tests to evaluate similarity in the data

Cluster analysis is useful for multi-dimensional datasets because it allows all elements measured in a single laser spot to be analysed at once, providing an overall view on metal composition. In addition, I also compared data on each element individually, to determine whether any element behaves differently.

The compositional data for each element were interpreted using two statistical tests (the Two-Sample Student T-Test and the Wilcoxon Rank Sum Test) to establish if the datasets for pockets and matrix metal are subsets of the same population (in other words, if they are statistically identical). For both populations, I perform the

tests to 95% and 99% confidence level, and the null hypothesis is that the populations are identical. Both tests are performed using MATLAB built-in functions.

The Two-Sample T-Test The Two-Sample Student T-Test (Student 1908) computes the means of two datasets, assuming that they are normally distributed and independent of each other. The means are then compared to standard values based on the chosen confidence level.

The Wilcoxon Rank Sum Test The Wilcoxon Rank Sum Test (Mann & Whitney 1947) also compares two populations, without the need to assume that they are normally distributed. Each data point is assigned a number based on its rank within both datasets combined (for example, the lowest number in either dataset has rank 1, the second in either dataset has rank 2, etc). The ranks are then summed up and compared between the two datasets, then compared to standard values.

3.2.4.5 Determination of average compositions

The data in this study are divided into distinct groups (matrix kamacite, matrix taenite, pocket kamacite and pocket taenite) with different abundances and associated contributions to the bulk composition of the sample. To account of their different abundances, weighted means and standard deviations are used to report average compositions.

The individual compositions of each group are calculated as mean $\pm$ standard deviation. To obtain the average compositions of the matrix and pockets, the kamacite (f_k) and taenite fractions (f_t) of the metal need to be known, which requires an assumption of the bulk Ni content of the metal (Ni_{bulk}). I assume a bulk Ni content (in metal) of 9.51% following Van Niekerk et al. (2007), who used neutron activation analysis on the metal in Seymchan. This method does not distinguish between different phases or microstructures within the metal (e.g., kamacite, taenite, tetrataenite

or plessite), instead calculating the Ni content of the bulk metal. Here, the assumption that all Ni in the Seymchan metal is stored in either kamacite or taenite may therefore introduce an error, because other metal phases present are not accounted for (e.g., tetrataenite and plessite). To mitigate this, measurements of taenite include tetrataenite and plessite, as they are all usually present in zoned taenite lamellae. Other Ni-bearing minerals in pallasites (e.g., schreibersite, olivine, etc.) do not introduce an error because their Ni concentration is not included in the value reported by Van Niekerk et al. (2007).

Using the value for Ni_{bulk} above, I then calculate the kamacite and taenite fractions using a mass balance, assuming that all Ni in the metal is present in either kamacite or (zoned) taenite:

$$f_k = \frac{Ni_{bulk} - Ni_t}{Ni_k - Ni_t} \times 100 \quad (3.1)$$

$$f_t = 1 - f_k \quad (3.2)$$

where Ni_k and Ni_t are the average Ni contents in kamacite and taenite, respectively. Ni_k and Ni_t are calculated by averaging all data from EMPA in the matrix and pockets respectively, after outliers were removed. This distinction is maintained in order to preserve potential compositional differences between the matrix and pockets. The calculation results in 73% kamacite and 27% taenite in the matrix, and 71% kamacite and 29% taenite in the pockets.

After the kamacite and taenite fractions are calculated, assuming that the metal consists entirely of kamacite and taenite (Equation 3.2), I use weighted means to calculate the average compositions of the matrix and pocket metal:

$$W = \frac{\sum_{i=1}^n w_i \times x_i}{\sum_{i=1}^n w_i} \quad (3.3)$$

where x_i is each datapoint, w_i is the weight assigned to it (here, the kamacite or taenite fraction corresponding to each datapoint) and n is the total number of datapoints.

The error on the resulting average values is calculated using a weighted standard deviation:

$$S = \sqrt{\frac{\sum_{i=1}^n w_i (x_i - W)^2}{\frac{M-1}{M} \sum_{i=1}^n w_i}} \quad (3.4)$$

where W is the weighted mean defined in Equation 3.3, x_i is each datapoint, w_i is the weight assigned to it, n is the number of datapoints and M is the number of non-zero weights.

To calculate the bulk composition of the metal, a further weighting is applied, because the pockets represent approximately 10% of the metal (Walte et al. 2020). Equation 3.4 is unchanged, but w_i is now the product between the kamacite/taenite fraction and matrix/pocket fraction (f_m and f_p) for each point, based on the divisions established at the start of this subsection. For example, matrix kamacite is very abundant, because its weight is $f_k \times f_m = 73\% \times 90\% = 65.7\%$, whereas pocket taenite only makes up $29\% \times 10\% = 2.9\%$ of the sample.

When averaging kamacite and taenite compositions to estimate the bulk composition, the averaging does not account for subsolidus fractionation between kamacite and taenite, which may introduce error in the bulk compositions. The error is addressed in Section 3.4.1.1.

3.3 Results

3.3.1 Textural analysis

This sample of Seymchan is characterised by a prominent olivine aggregate (approximately 50% of the sample, including a small amount of metal in pockets), as well as isolated angular olivines surrounded by matrix metal (Fig. 3.3). There is also a

smaller olivine aggregate fragmented into 2 (top right of Fig. 3.3), containing a small metal pocket.

The two metal textures identified by Walte et al. (2020) – pockets of metal trapped in olivine cumulates and matrix metal in between cumulates – are also identified in this sample. I have also identified two of the three pocket types outlined in the above studies: type 2 (small, equant, high-dihedral angle pockets; Fig. 3.6A) and type 3 (large, irregularly-shaped pockets; Fig. 3.6B). Pockets of type 1 (submicron rounded inclusions in olivines) were not found in this sample. A total of 11 pockets are analysed texturally and chemically, shown by the numbers in Fig. 3.3. In this study, I refer to metal as pocket and matrix metal, to maintain the distinction between the two main textures.

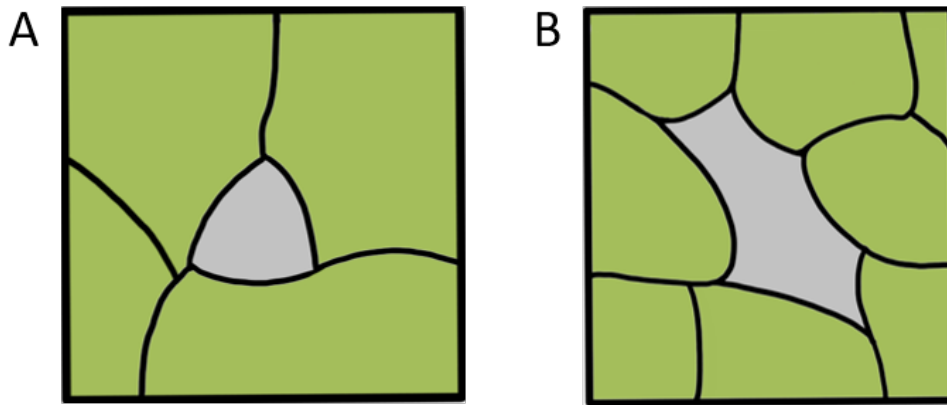


Figure 3.6: Pocket metal textures in Seymchan, first identified by Walte et al. (2020). A. Type 2 pocket (small, equant, high-dihedral angles, bounded by 3-4 olivines with convex boundaries); B. Type 3 pockets (large, irregularly-shaped, bounded by more than 4 olivines). The images are 1 cm across, but pocket sizes can vary. Grey = metal; Green = olivine.

An important consideration in determining whether a pocket contains primary metal is whether it was isolated when the metal was still molten, or in other words, whether flow between the pocket and the matrix metal was possible. The primary/isolated pocket criteria outlined by Walte et al. (2020) can be summarised as follows:

- Shape of the pocket: type 2 pockets are equant, whereas type 3 pockets have irregular shapes, often shaped by the growth of the neighboring olivines;
- Shape of the contact with olivine: convex towards the olivine for type 2, either convex or concave for type 3;
- Number of neighbouring olivines: owing to their small size, type 2 pockets are surrounded by 3-4 olivines, while type 3 olivines are surrounded by >4 olivines;
- Dihedral angles: type 2 pockets have high ($>60^\circ$) dihedral angles, while type 3 can have any dihedral angle;

Fig. 3.7 shows photomicrographs of the 11 pockets. Pockets 2, 3, 5, 7, 9, 10 belong to type 2, whereas pockets 1, 4, 6 and 11 belong to type 3. Pocket 8 has characteristics from both types (intermediate size, non-equant shape, high dihedral angle) and it could belong to either type. Based on the high dihedral angles and the regular shape (despite the fact that it is elongated rather than equant), pocket 8 is considered to belong to type 2.

The 3D extent of the pockets is uncertain, but the thickness of the analysed slab was ~ 1 cm before cutting and the olivine aggregate occupies the same space on both sides of the slab. It is therefore safe to assume that the thickness of the olivine aggregate is $\gg 1$ cm, giving confidence that the pockets are isolated from the matrix. The pockets do not extent to the other side of the slab, with the exception of pockets 7, 8 and 9, where the slab is thinner ($\sim 0.3 - 0.5$ cm). It should further be noted that pockets 6 and 11 are on the edge of the sample, which could mean that they sample the matrix, rather than an isolated pocket. This is especially true for pocket 11, which came from the underside of the sample, having come loose during the cutting process.

Table 3.2 summarises the criteria for a primary pocket outlined by Walte et al. (2020), and shows which pockets sampled in this study meet the criteria. All criteria are met by pockets 2, 3 and 10 (type 2) and 1 and 4 (type 3) only. Pockets 6

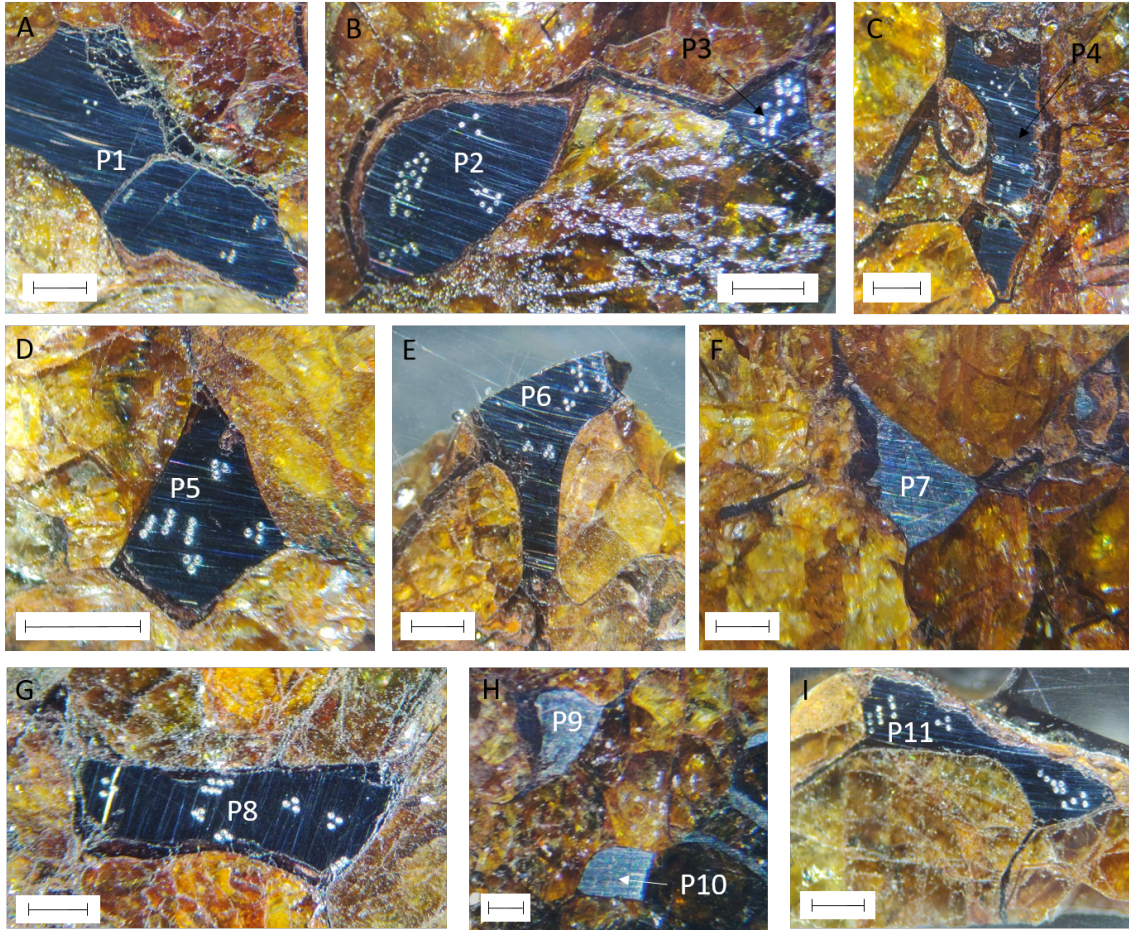


Figure 3.7: A – I. Optical images of the pockets trapped within olivine, labelled according to the pocket number. Images F and H are taken pre-LA-ICP-MS, thus no laser spots are visible. Scale bar = 1 mm.

and 11 may meet the criteria, but since the pockets are not fully exposed, they are treated as non-isolated. When determining the spatial extent of a pocket, sulfides and phosphides are taken together with the metal, since they likely exsolved out of the metal as it solidified.

Along with primary features, Seymchan exhibits a variety of secondary features. Firstly, it is apparent in several places in Fig. 3.3 that the main olivine cluster on the left side of the sample was fractured, and the gaps were infilled with metal, leading to the olivine cluster looking as though it was pulled apart. This is a well-documented feature in pallasites with angular and fragmental olivines, as it is generally attributed to a deformation event (Walte et al. 2020). Further secondary features are repre-

Pocket type	Pocket number	Shape of pocket	Shape of boundaries with olivine	Number of olivine neighbours	Dihedral angle
		Type 2 = Equant	Type 2 = convex	Type 2 = 3-4	Type 2 = all over 60°
		Type 3 = Irregular	Type 3 = any	Type 3 > 4	Type 3 = any
2	2	✓ ¹	✓	✓	✓
	3	✓	✓	✓	✓ ²
	5	✓	✓	✓	x
	7	✓	x	✓	x
	8	✓	x	x	x
	9	✓	✓ ³	✓ ⁴	x
	10	✓	✓	✓	✓
3	1	✓	✓	✓	✓
	4	✓	✓	✓	✓
	6	✓	✓	unknown	✓
	11	✓	✓	unknown	✓

Table 3.2: Summary of textural criteria for the isolation of metal pockets in pallasites, with check marks indicating a met criterion. All type 3 pockets have at least one low dihedral angle, but high dihedral angles are not a requirement for this type of pocket being isolated, so the criterion is marked as met. ¹ Pocket would be equant, but two sides merged together to form a longer, convex side. ² All dihedral angles of pocket 3 are over 60°, but a veinlet appears to merge it to pocket 2. ³ Shape of the boundaries difficult to distinguish due to 2-3 boundaries merging to become one curved (convex) boundary. ⁴ Number of neighboring olivines unclear.

sented by veinlets within the olivine aggregate (e.g., between pockets 2 and 3 in Fig. 3.7B, and radiating away from pocket 11 in Fig. 3.7I). The veinlets often contain sulfides and phosphides (Appendix A.1) and they form sharp angles, as though formed between angular olivines. The veinlets intersect the pockets at sharp angles, which was also observed in different samples of Seymchan and to a smaller extent, in Imilac (Walte & Golabek 2022), confirming the secondary character of these features.

3.3.2 Composition of Seymchan

The trace element composition of metal in the pockets and matrix of Seymchan is reported in Table 3.3. The data are separated into kamacite and zoned taenite

(including plessite, tetrataenite and cloudy zone), which are measured separately in both pockets and matrix, and then compared directly. Some measurements were below detection limit (35% of W measurements, 38% of Os measurements, 1.4% of Ir measurements and 1.4% of Au measurements) and they were excluded from the mean compositions in Table 3.3, which may artificially increase the reported mean values for these elements.

This section is composed of three main subsections: firstly, I use cluster analysis on the entire dataset to distinguish different clusters based on all measured elements (Section 3.3.2.1). Secondly, I break down the data into matrix and pocket metal, and discuss differences in individual elements between the two subgroups (Section 3.3.2.2). Finally, I present compositional differences between individual pockets (Section 3.3.2.3).

3.3.2.1 Cluster analysis results

Results from the k-means clustering are shown in Fig. 3.8. K-means clustering was performed for a reduced data set (removing measurements below detection limit in any element), as well as a subset of six elements (Fe, Ni, Co, Cu, Ga and Ge; referred to as "6-element dataset") to minimise zero values, as explained in Section 3.2.4.1. The plots on the first column of Fig. 3.8 are scatter plots of Fe vs. Ni after they have been scaled according to the method described in Section 3.2.4.3, with colours corresponding to each cluster, as determined through cluster analysis. The plots on the second column are silhouette plots, where each silhouette corresponds to a cluster. The mean value for each silhouette plot is also shown, as a measure of how well the clustering fits the data (the higher the silhouette mean, the better the fit).

In both the reduced and 6-element dataset, two clusters are preferred over three, both from quantitative (higher silhouette means for the two cluster plots) and qualitative analysis (the three-cluster silhouette plots contain smaller or negative values). When partitioning the data into two clusters, the clusters represent kamacite and taenite. When the data is grouped into three clusters, the taenite cluster is resolved

	Fe (%)	Ni (%)	Co (%)	Cu (ppm)	Ga (ppm)	Ge (ppm)	W (ppm)	Os (ppm)	Ir (ppm)	Au (ppm)
Kamacite	Matrix	6.61	0.59	80.28	26.04	77.51	0.52	1.00	0.72	1.77
	σ	0.74	0.03	11.15	2.52	4.27	0.17	0.51	0.28	0.20
	Range n	4.83 - 8.49 107	0.47 - 0.64 107	54.5 - 96.92 39	20.25 - 30.04 39	68.54 - 87.48 39	0.26 - 0.83 30	0.24 - 1.94 29	0.22 - 1.23 39	1.34 - 2.21 38
	Pockets	6.92	0.58	79.78	26.78	78.64	0.51	0.78	0.63	1.75
	σ	0.25	0.04	8.33	2.53	4.00	0.27	0.35	0.23	0.28
	Range n	5.58 - 7.92 164	0.48 - 0.66 164	58.96 - 98.63 86	19.96 - 32.26 86	68.13 - 89.19 86	0.1 - 1.87 53	0.17 - 1.73 47	0.19 - 1.44 85	1.24 - 2.43 86
Kamacite mean σ		6.65 0.80	0.58 0.03	80.18 10.50	26.19 2.50	77.73 4.19	0.52 0.19	0.97 0.49	0.70 0.27	1.77 0.22
Taenite	Matrix	17.24	0.34	286.66	26.31	62.75	0.55	1.36	1.25	2.87
	σ	2.77	0.02	71.01	6.15	7.72	0.20	0.49	0.24	0.43
	Range n	13.72 - 23.39 29	0.29 - 0.38 29	199.78 - 397.56 9	20.88 - 39.44 9	54.42 - 74.67 9	0.23 - 0.82 9	0.69 - 2.28 8	0.92 - 1.7 9	2.32 - 3.66 9
	Pockets	15.79	0.37	260.74	26.45	61.91	0.51	0.94	0.64	2.68
	σ	2.39	0.03	53.16	3.22	6.56	0.68	0.52	0.31	0.61
	Range n	10.38 - 28.5 157	0.25 - 0.43 157	186.53 - 409.58 73	20.32 - 36.65 73	50.61 - 86.05 73	0.09 - 0.9 42	0.18 - 2.87 45	0.12 - 1.5 71	1.55 - 4.67 71
Taenite mean σ		16.70 2.79	0.35 0.03	274.38 62.03	26.38 4.75	62.35 6.96	0.54 0.18	1.20 0.52	0.97 0.40	2.78 0.52
Matrix mean*		7.58	0.56	96.51	26.06	76.35	0.52	1.04	0.76	1.86
	σ	3.25	0.08	59.58	2.88	6.02	0.17	0.51	0.31	0.37
	Pockets mean*	9.41	0.52	126.37	26.70	74.33	0.51	0.82	0.63	1.98
Bulk*		4.07	0.10	83.84	2.71	8.73	0.25	0.41	0.25	0.56
		91.32	7.89	103.33	26.21	75.89	0.52	1.00	0.73	1.89
	σ	3.47	3.50	67.09	2.86	6.79	0.19	0.50	0.30	0.43

Table 3.3: Mean and standard deviation values for the concentration of the measured elements for kamacite and taenite in Seymchan (separated into matrix and pocket metal) and Henbury. Major elements (Fe, Ni, Co) are obtained using EMPA, and minor elements (Cu, Ga, Ge, W, Os, Ir, Au) are obtained using LA-ICP-MS. The matrix and pocket means were calculated by assuming the bulk Ni of the metal (9.51%, Van Niekerk et al. 2007) and using the kamacite and taenite means listed above to calculate the kamacite and taenite fractions of the metal, resulting in 73% kamacite and 27% taenite in the matrix, and 71% kamacite and 29 % taenite in the pockets. The matrix and pocket averages were then calculated as weighted means (Section 3.2.4.5). The bulk concentrations were calculated by further assuming that the pockets make up 10% of the metal in Seymchan (Walte et al. 2020). The means marked by * do not account for subsolidus fractionation between kamacite and taenite, which may produce errors in the bulk compositions. The measurements below detection limit (35% of W measurements, 38% of Os measurements, 1.4% of Ir measurements and 1.4% of Au measurements) were excluded from the mean compositions, which may artificially increase the reported values for these elements. These errors are addressed in the text (Section 3.4.1.1). n = the number of measurements included in the averaging, after measurements below detection limits are removed; σ = standard deviation.

into two halves, for high Ni and low Ni (Fig. 3.9). Internal variation is expected in taenite since it is zoned, so breaking down taenite based on Ni does not provide any additional information about the origin of the metal. Configurations with more than three clusters are also evaluated and the results show that two clusters is the optimal configuration (Fig. 3.10).

To check for further patterns within kamacite and taenite, k-means clustering is also applied individually to each phase. Fig. 3.10 shows the silhouette values of different clustering configurations (up to 6 clusters) for the entire dataset, as well as kamacite and taenite individually. In all cases, two clusters represent the optimal configuration using silhouette statistics.

An additional cluster evaluation criterion is also introduced in Fig 3.10 – the gap criterion (Tibshirani et al. 2001) is unique because it allows the evaluation of different clustering configurations (2 to 6 clusters) against no clustering (all the data form one cluster), which is not supported by the silhouette criterion. The gap criterion indicates that there is no clustering within kamacite, since one cluster is preferred over any higher clustering configuration. For taenite, the gap criterion supports the presence of three clusters. However, as previously explained, a range of values is expected in taenite due to the zoning, so clustering within taenite is not very informative in this case.

3.3.2.2 Matrix/Pockets comparison

The data are further broken down into matrix and pocket metal measurements, to investigate the difference in composition in each element. From visual inspection, there is overlap between pocket kamacite and matrix kamacite, and between pocket taenite and matrix taenite, in any element plotted against Ni (Fig. 3.11). As Ni varies throughout the width of a taenite lamella from $\sim 10\%$ to 20% , clusters of elements in taenite form elongated clusters or lines, as opposed to the kamacite clusters, which are more circular. Fig. 3.11 and Table 3.3 both exclude the measurements that were below detection limit.

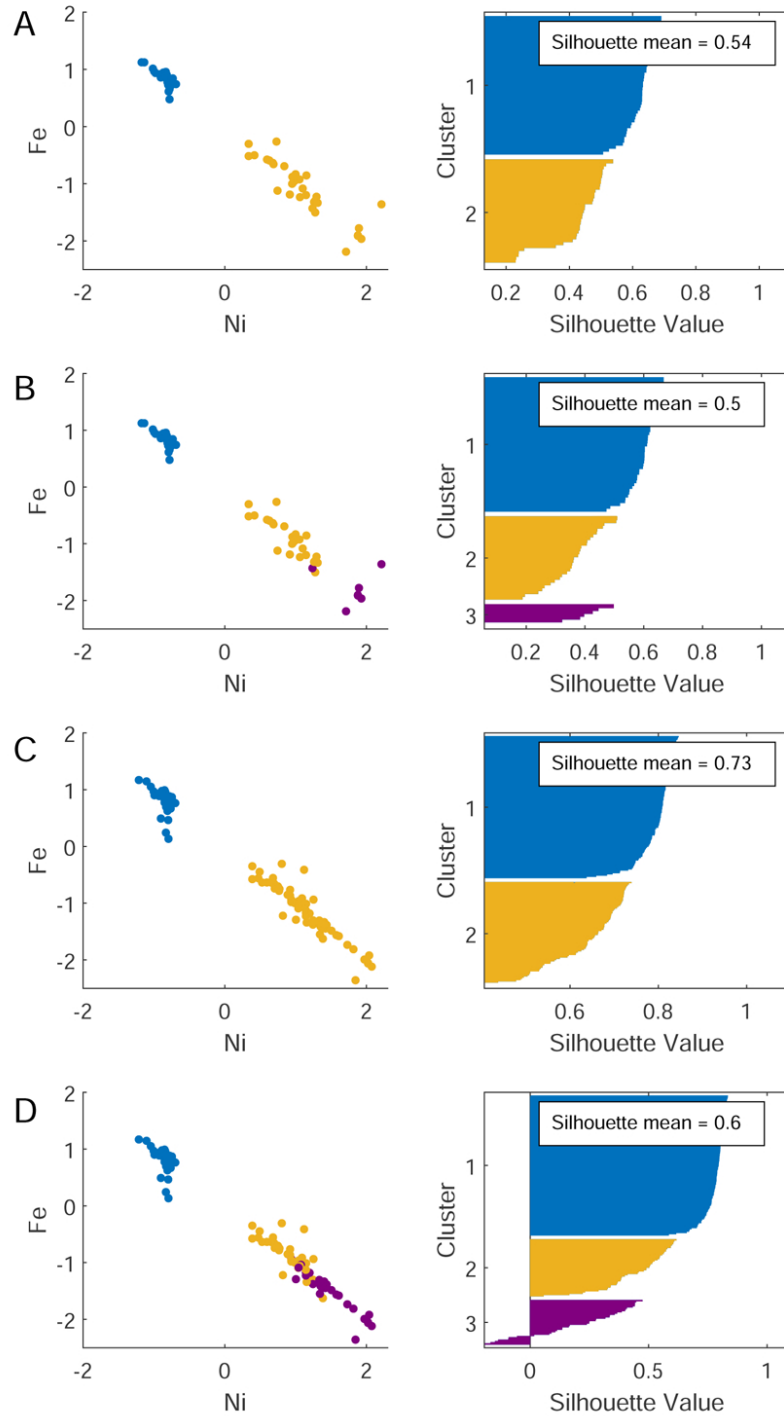


Figure 3.8: Results of cluster analysis on Seymchan. Left: Scaled Fe vs. Ni scatter plots of the data, with colours corresponding to the clusters obtained using k-means clustering. Right: Silhouette plots corresponding to each clustering, including the silhouette mean value for the clustering. A. Reduced dataset, 2 clusters; B. Reduced dataset, 3 clusters; C. 6-element dataset (Fe, Ni, Co, Cu, Ga, Ge), 2 clusters. D. 6-element dataset, 3 clusters.

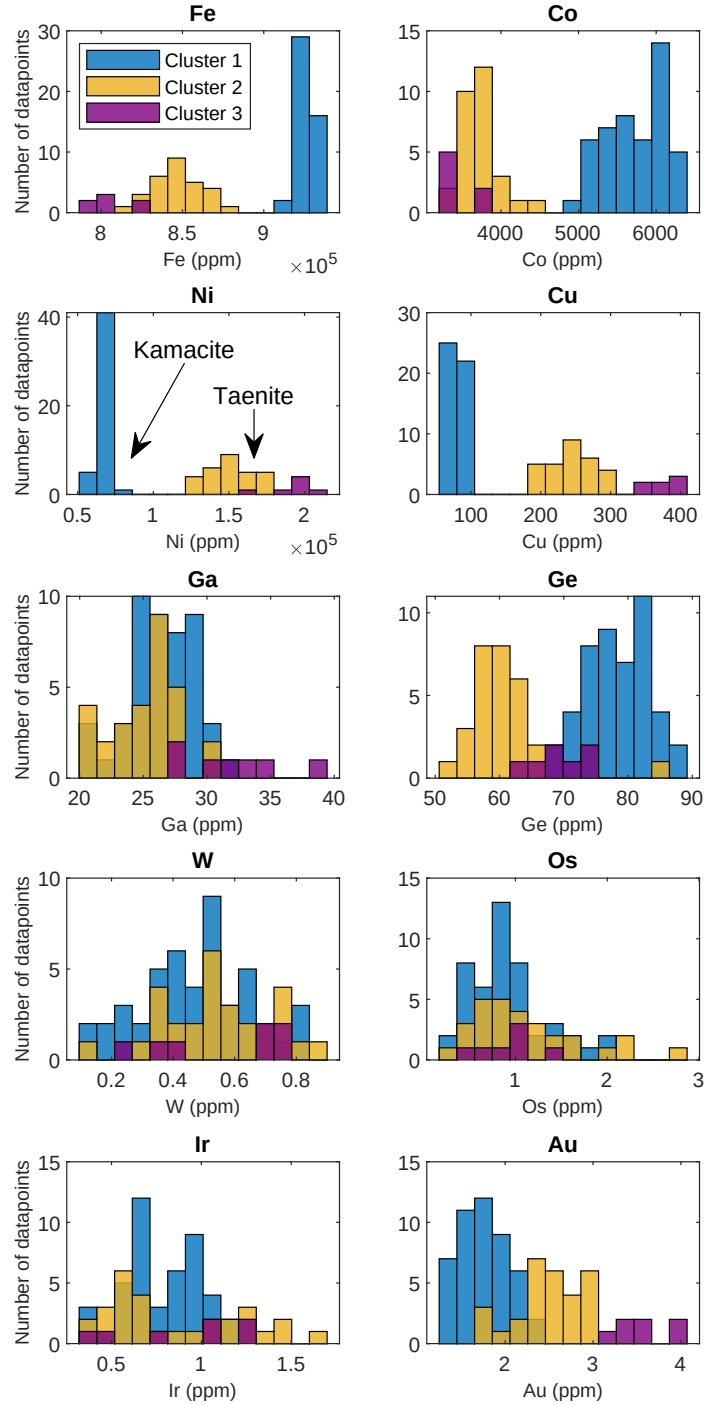


Figure 3.9: Histograms showing how the measurements are clustered based on the clustering shown in Fig. 3.8B. From visual inspection of the Ni histogram, cluster 1 corresponds to kamacite, and clusters 2 and 3 correspond to taenite. When clustering using only two clusters (e.g. Fig 3.8A), clusters 2 and 3 are merged together.

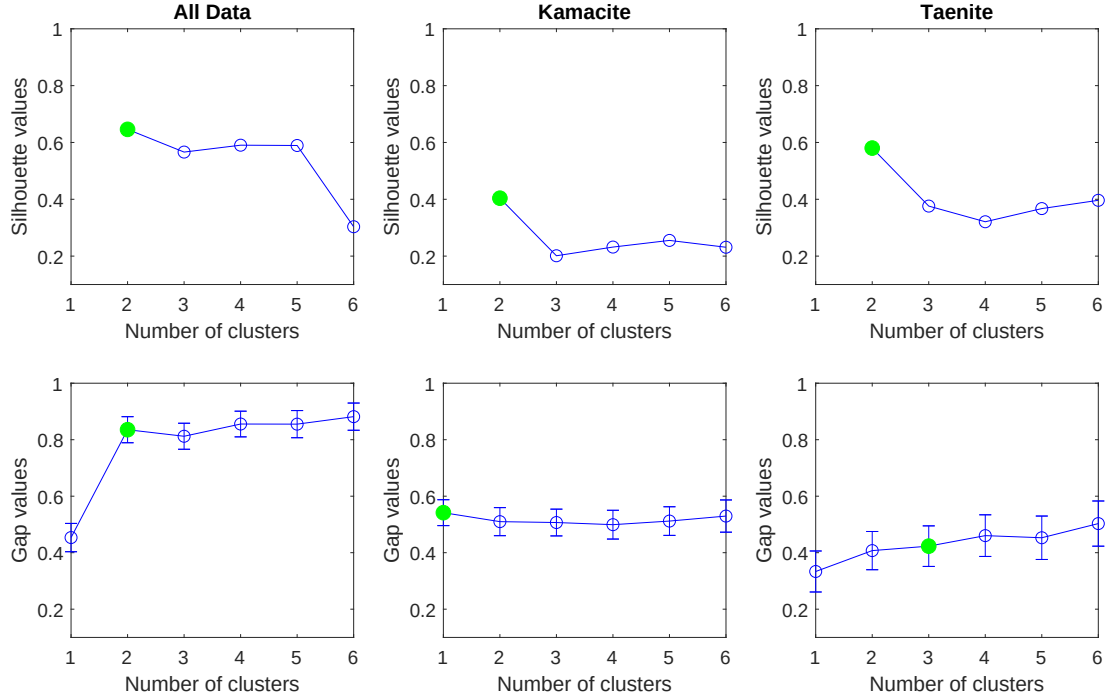


Figure 3.10: Evaluation of different clustering configurations using silhouette values and gap values, calculated for configurations of 1 up to 6 clusters, for the reduced dataset, as well as kamacite and taenite individually. The green circle indicates the optimal clustering configuration in each case. According to the silhouette criterion, the preferred clustering configuration is the number of clusters with the highest silhouette value. The gap criterion aims to identify an "elbow" formally, by calculating the greatest change between successive clustering configurations. The standard deviation of the gap value (shown by the error bars) also plays a role in the result, and the preferred configuration is the lowest number of clusters for which the gap value for k clusters is greater than the gap value for $k + 1$ clusters plus the error. The error depends on the standard deviation and the number of Monte Carlo simulations performed (see Section B.1.2).

		Fe	Co	Ni	Cu	Ga	Ge	W	Os	Ir	Au
T-test (95%)	K								x		
	T	x								x	
T-test (99%)	K										
	T									x	
Wilcoxon test (95%)	K										
	T	x		x					x	x	
Wilcoxon test (99%)	K										
	T									x	

Table 3.4: The results of the two-sample T-test and Wilcoxon test comparing the datasets for kamacite (in pockets vs. matrix) and taenite (in pockets vs. matrix) at 95% and 99% confidence level. The fields marked with ‘x’ show the elements that passed the test, indicating distinct concentrations in the pockets and matrix. K = kamacite; T = taenite.

The histograms in Fig. 3.9 were also used to determine if the datasets are normally distributed. The measurements display normal distributions for most elements in both kamacite and taenite (considering both clusters 2 and 3 as taenite), but the taenite measurements of some elements reveal several peaks (e.g., Ni, Cu, Ir), or an asymmetrical distribution (e.g., Os). Because of the zoning in taenite, a spread in values is expected over a normal distribution, which would indicate one mean value surrounded by noise. Furthermore, taenite is less abundant than kamacite, which could introduce further scatter. Since normality could not be rigorously verified in every case, both statistical tests (two-sample student T-test and the Wilcoxon rank sum test) were applied to all datasets.

The test results are shown in Table 3.4. For kamacite, Os passes the T-test at 95% confidence, suggesting distinct Os concentration between pockets and matrix, while the other elements fail, suggesting identical concentration. At 99% confidence, all elements pass the tests. For taenite, Ir fails at both 95 and 99% confidence, while Fe fails at 95% confidence and all other elements pass.

The Wilcoxon tests suggests distinct Co concentrations in kamacite, but identical concentrations for all other elements, including Os. For taenite, the tests pass for Fe,

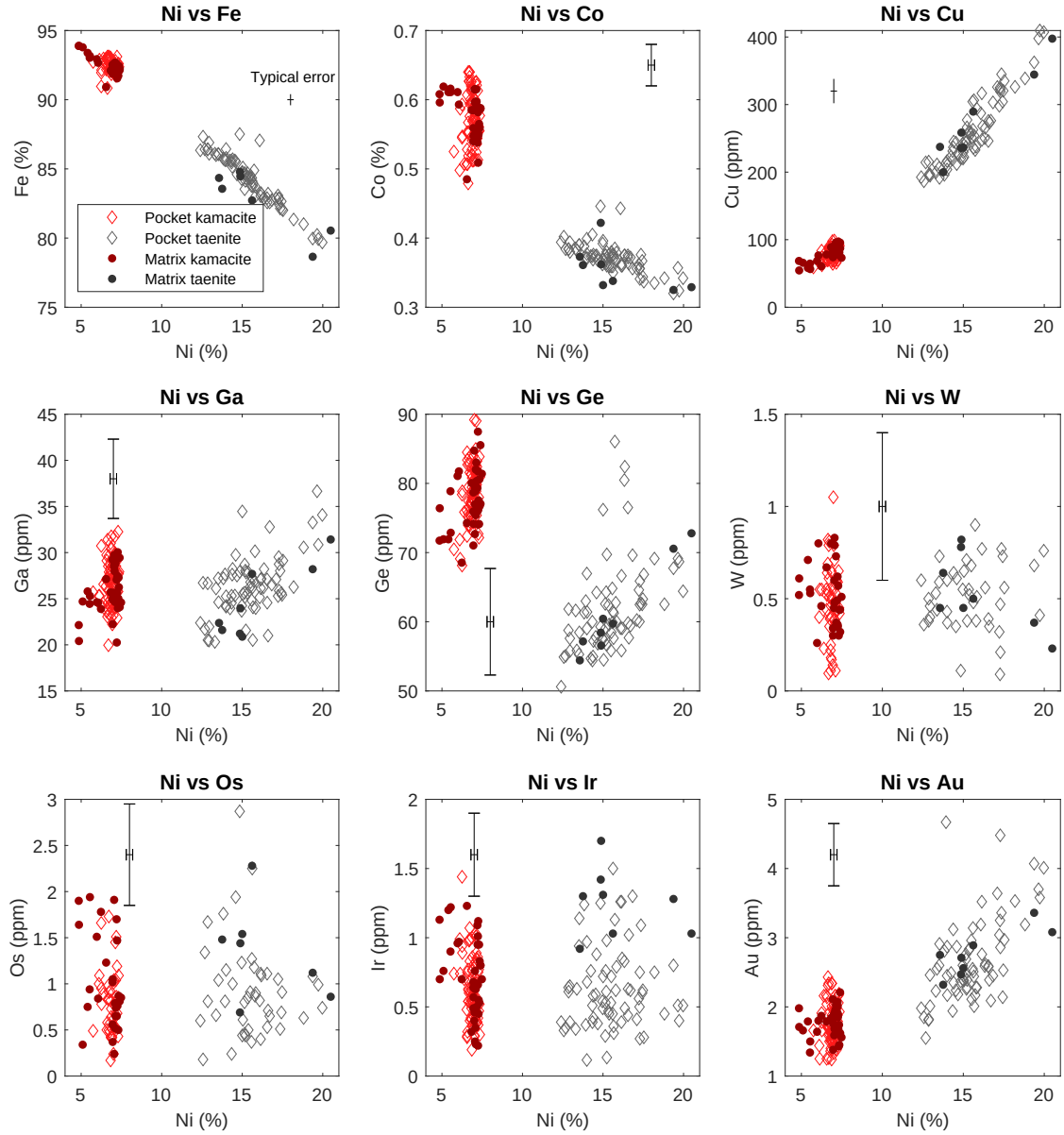


Figure 3.11: Scatter plots showing Fe, Co, Cu, Ga, Ge, W, Os, Ir and Au plotted against Ni in both matrix metal (circles) and pockets (diamonds), distinguishing between kamacite (red) and taenite (grey). The major element data (Fe, Ni, Co) is measured using EMPA, while the other elements are measured using LA-ICP-MS. The error bars show the typical error on each plot.

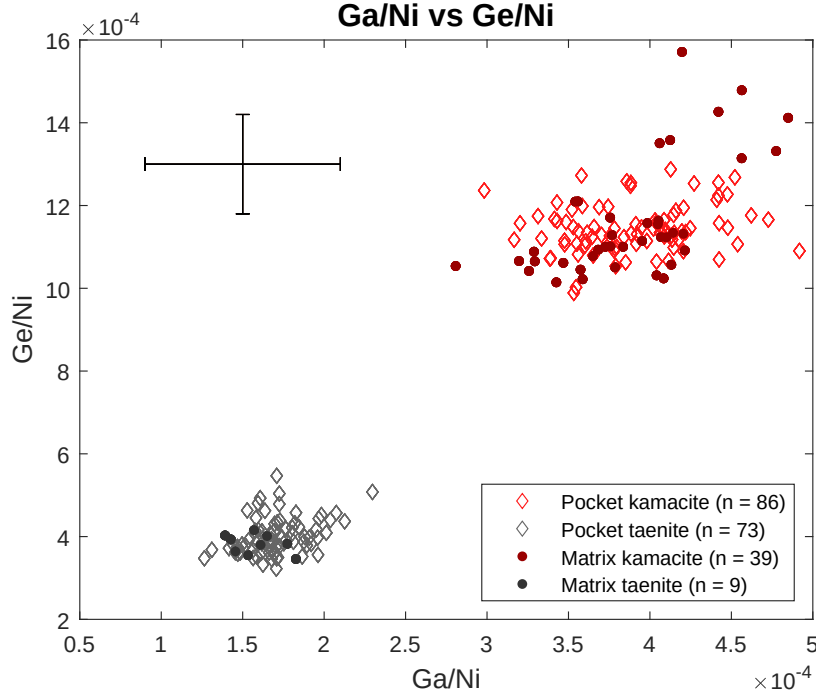


Figure 3.12: Ga/Ni plotted against Ge/Ni in Seymchan. Note the different sample sizes (specified in the legend) which could explain the lower mean value for matrix taenite. The datasets for both taenite and kamacite are identical to 99% confidence, suggesting that the Ga/Ni and Ge/Ni ratios are indistinguishable between matrix and pocket metal. The error bars show the representative error ($\pm 1\sigma$) for Ga and Ge normalised by Ni, assuming no error in Ni content. Because Ni is internally calibrated with EPMA data, the error associated with Ni measurements is significantly lower than Ga and Ge.

Ni, Os and Ir (distinct concentration) and fail for all other elements.

Fig. 3.12 plots Ga/Ni against Ge/Ni in both datasets, showing excellent overlap between matrix and pockets. The clusters are statistically indistinguishable to 99% confidence, which further suggests that the Ga and Ge compositions are identical.

3.3.2.3 Individual pocket compositions

The individual composition of each pocket is shown in Fig. 3.13, normalised by the average composition of both kamacite and taenite, and in Table A.2. The data for Ni, Fe and Co are based on EMPA results, and they show much less scatter than LA-ICP-MS data used for the other elements. In general, the most scatter is observed for W, Os, Ir, and to a smaller extent Au, which are also the elements found in the smallest

concentrations, and which have the most measurements below detection limit. Some pockets did not have taenite exposed, hence the fewer pockets plotted on the second set of graphs in Fig 3.13.

The average composition of kamacite and taenite in the matrix is also plotted in Fig 3.13 for reference, normalised by the overall composition. Despite the majority of measurements being taken in the pockets, the matrix average shows a remarkably flat trend and values close to 1 when normalised by the overall average, particularly in kamacite. This suggests that the matrix composition is representative for all the metal sampled from Seymchan. Fig. 3.13 A and C show all pocket compositions, whereas Fig. 3.13 B and D only show the compositions of the pockets which meet all the criteria to be isolated (Section 3.3.1), along with the errors on each composition (Table A.2).

Cluster analysis was also performed on a dataset that includes all pocket averages (both kamacite and taenite) and the results are shown in Fig. 3.14. The silhouette plots on the second column reveal that the optimal clustering is with two clusters instead of three (silhouette means of 0.59 vs. 0.49). As with the overall data (Fig. 3.8), these clusters correspond to kamacite and taenite, and there is no evidence for further clustering within them.

3.3.3 Composition of Henbury

The composition of Henbury (IIIAB iron) is reported in Table 3.5. As IIIAB iron meteorites have faster cooling rates than pallasites (Yang & Goldstein 2006), the taenite lamellae are significantly thinner, which led to difficulties sampling them with the laser. The laser spot size in taenite was 50-70 μm , adjusted to match the width of the widest taenite lamellae, which led to smaller counts-per-second values, resulting in higher detection limits and more uncertainty in the results. Furthermore, only 4 lamellae were thick enough to be sampled this way. This led to significant spread in the data from taenite and high standard deviations (Table 3.5). Table 3.5 excludes

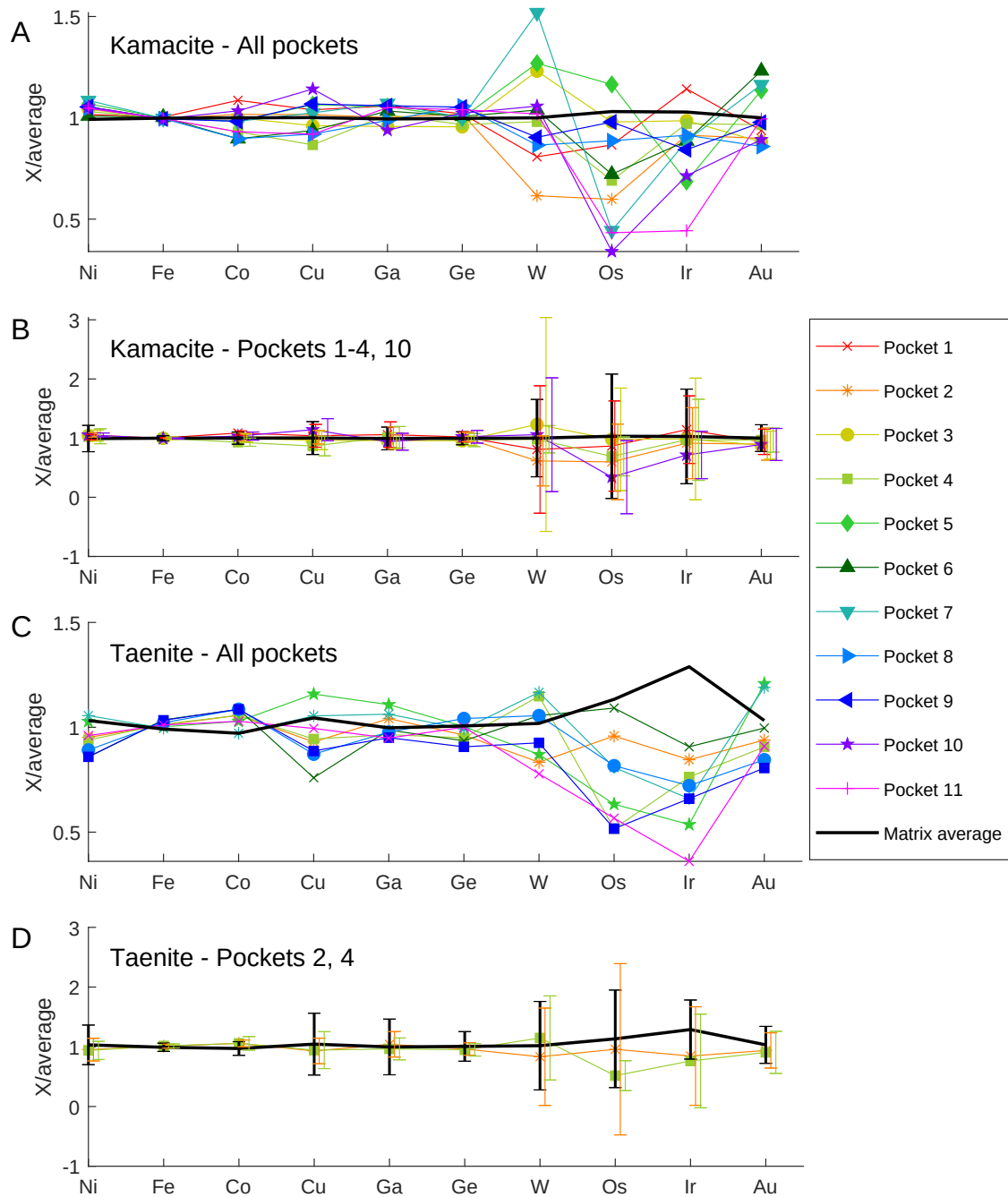


Figure 3.13: Spider diagrams showing the average composition of each element in each pocket, normalised by the average composition of all kamacite and taenite in the sample. The coloured lines correspond to pockets, and the black line corresponds to the average value of the matrix. The errors are given as $\pm 1\sigma$. A. Kamacite, all pockets; B. Kamacite, reliable pockets only (pockets 1-4 and 10); C. Taenite, all pockets; D. Taenite, reliable pockets only (pockets 2, 4).

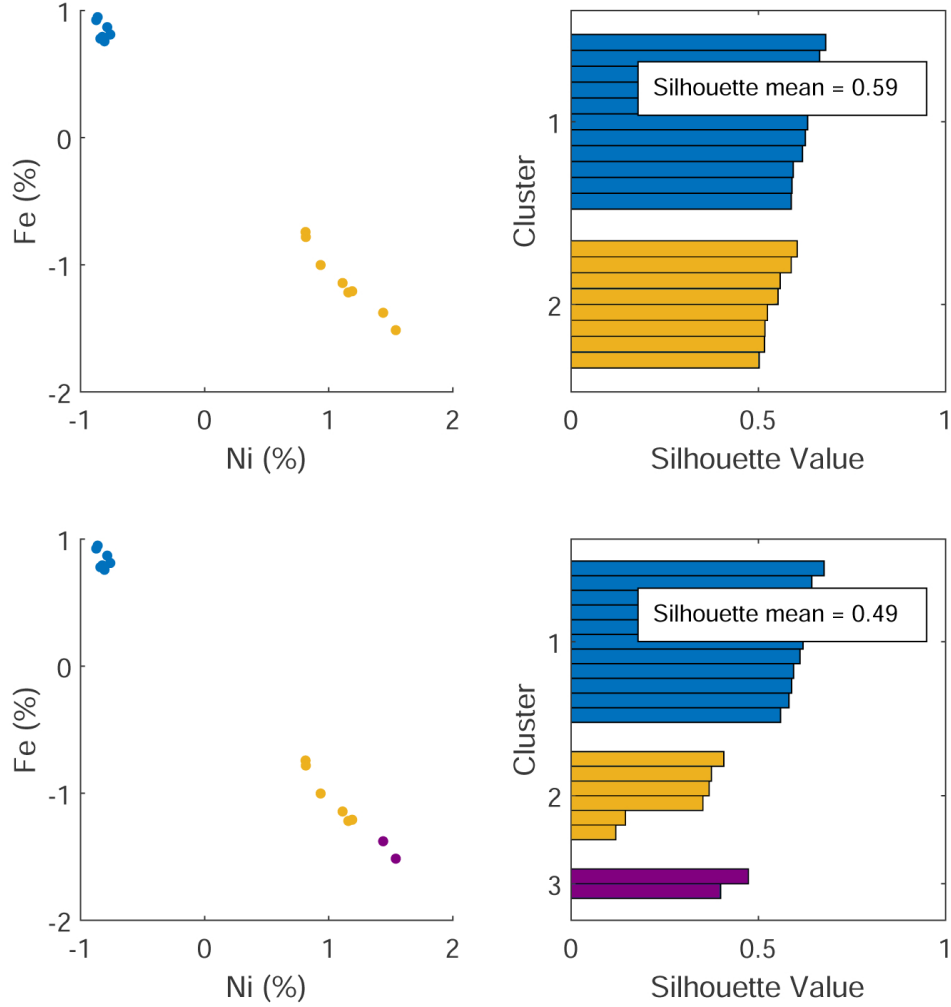


Figure 3.14: Results of cluster analysis on the individual pocket averages in Seymchan. Left: Scaled Fe vs. Ni compositions, with colours showing the clustering obtained from k-means clustering into 2 clusters (top) and 3 clusters (bottom). Right: Silhouette plots corresponding to each clustering, showing a preference for 2 clusters (mean = 0.59) over 3 clusters (mean = 0.49).

one kamacite Au measurement below the detection limit of 2.82 ppm, and two taenite W measurements below the detection limits of 1.06 and 0.45 ppm, respectively.

3.4 Discussion

In this section, I discuss the origin of the metal in Seymchan by first comparing it to chondritic compositions (Section 3.4.1.1). I then investigate potential differences between the matrix and pockets (Section 3.4.1.2), and between individual pockets

	Fe (%)	Ni (%)	Co (%)	Cu (ppm)	Ga (ppm)	Ge (ppm)	W (ppm)	Os (ppm)	Ir (ppm)	Au (ppm)
Kamacite mean	92.10	6.95	0.50	140.25	19.14	37.49	0.66	7.02	6.62	0.41
σ	1.30	0.17	0.02	8.12	2.49	3.69	0.22	1.55	1.17	0.09
Range	85.6 -	6.4 -	0.5 -	118.7 -	14.2 -	28.7 -	0.4 -	4.6 -	4.3 -	0.2 -
n	93.2	7.3	0.5	153.3	24.8	44.4	1.2	10.6	8.8	0.6
	66	66	66	30	30	30	30	30	30	29
Taenite mean	80.88	17.54	0.39	529.93	16.67	33.16	0.49	4.83	5.67	0.64
σ	5.36	5.71	0.12	70.48	5.76	12.54	0.04	2.44	2.52	0.15
Range	70.9 -	10.9 -	0.2 -	441.5 -	13.1 -	22.1 -	0.5 -	2 - 7.6	2.2 -	0.5 -
n	87	29.1	0.5	613.3	25.3	45.6	0.5	7.8	7.8	0.9
	11	11	11	4	4	4	2	4	4	4
Bulk	92	7.03	0.50	142.79	19.04	37.34	0.66	6.98	6.57	0.40
σ	1.69	1.08	0.02	31.73	2.44	3.65	0.21	1.49	1.14	0.09

Table 3.5: Mean values for Fe, Ni, Co, Cu, Ga, Ge, W, Os, Ir and Au, for the kamacite and taenite in Henbury. Fe, Ni and Co data were acquired using EMPA, while the other elements were measured using LA-ICP-MS. The bulk concentrations are calculated using a mass balance between taenite and kamacite, and assuming a bulk Ni content of 7.44% (Wasson & Kimbrell 1967), and they do not account for sub-solidus partitioning between kamacite and taenite. Any measurements below detection limit were removed from the averaging (see main text). σ = standard deviation; n = sample size.

(Section 3.4.1.3), and I discuss the hypothesised link to the IIIAB iron meteorites (Section 3.4.2). Finally, I propose a formation model for pallasites (Section 3.4.3).

3.4.1 Origin of the metal

3.4.1.1 Iridium and Gold

Iridium is a highly siderophile element that is incompatible in liquid (the solid-liquid fractionation coefficient D_0 is 1.49 ± 0.07 ; Chabot et al. 2017), which causes it to fractionate into the metal core during planetary differentiation, and into the solid as the core crystallises. Fractionation between liquid and solid metal causes large variations in Ir concentration within single iron groups (four orders of magnitude variation in IIIABs; Fig. 3.2), which is usually visible in Ir vs Ni plots as a near-vertical line.

In Seymchan, Ir concentration is 0.7 ± 0.27 ppm in kamacite and 0.97 ± 0.4 ppm in taenite, while Ni averages to $6.65 \pm 0.7\%$ in kamacite and $16.70 \pm 2.69\%$ in taenite, assuming that the pockets make up 10% of the total volume of metal, as measured by Walte et al. (2020). Using a mass balance calculation between kamacite and taenite, and assuming that the bulk Ni content in pallasite metal is 9.51% (Van Niekerk et al. 2007), this leads to a bulk Ir content of 0.73 ± 0.3 ppm (Table 3.3). This value is below the reported Ir contents of the metal in chondritic groups, as shown in Fig. 3.15. Ir concentration in chondritic metal was previously reported as 2 - 3 ppm in carbonaceous chondrites (van Kooten et al. 2022), 2.1 - 2.8 ppm in enstatite chondrites (Kong et al. 1997), and ~ 2.5 ppm, up to 4.5 ppm in ordinary chondrites (Kadlag & Becker 2017). The Ir value reported in this study is an upper limit, since the calibration is based on the meteorite Coahuila, which has a higher Ir content than Seymchan (15 ppm; Wasson 1969). This could lead to an error in the results, but this error would only have increased the values reported here, which further indicates that Ir in the metal measured here shows a negative deviation from chondritic abundance that could be attributed to magmatic fractionation.

A way to test this hypothesis is to analyse a siderophile element with opposite fractionation behaviour to Ir, which would preferentially stay in molten metal instead of partitioning into the solid core. Au has a solid-liquid partition coefficient of 0.37 ± 0.05 (Chabot et al. 2017), which indicates a strong preference for the liquid phase. Chondritic abundances of Au have been reported as 0.1 - 0.3 ppm in carbonaceous chondrites (van Kooten et al. 2022), 0.6 ppm in ordinary chondrites (Kadlag & Becker 2017), and 0.6 - 1.5 ppm in enstatite chondrites (lower estimate by Kadlag et al. 2019, upper estimate by Kong et al. 1997). By contrast, Au in this study varies between 1.77 ± 0.22 ppm in kamacite, and 2.78 ± 0.52 ppm in taenite. Assuming 9.51% bulk Ni content, the bulk Au content of Seymchan metal is 1.89 ± 0.43 ppm (Table 3.3). The Au concentration in pallasite metal is generally higher than chondritic metal abundances (Fig. 3.15), which further suggests that fractionation is the likely cause for the deviation in both Au and Ir.

The bulk compositions of Au and Ir in this study are calculated without accounting for subsolidus partitioning between kamacite and taenite during cooling, which may introduce errors in the bulk compositions reported above. However, the taenite/kamacite partitioning coefficient of Ir is close to 1 (1.33 ± 0.34 ; Mullane et al. 2004), and the measured Ir contents of kamacite and taenite are very similar (0.7 ± 0.27 ppm in kamacite vs. 0.97 ± 0.4 ppm in taenite). Therefore, kamacite-taenite partitioning did not affect Ir significantly in this sample. In the case of Au, the partitioning coefficient is higher (1.76 ± 0.39 ; Mullane et al. 2004), suggesting a stronger tendency to partition into taenite. This is observed in the measured bulk Au contents in kamacite and taenite (1.77 ± 0.22 vs. 2.78 ± 0.52 ppm, respectively). Since there is much less taenite than kamacite in the sample, weighting by kamacite/taenite fraction (Section 3.2.4.5) effectively decreases the influence of taenite on the bulk concentration. Because Au partitions into taenite, this translates into diminishing the effect of Au being more abundant in taenite. Therefore, not accounting for subsolidus fractionation in this case underestimates the real bulk content of Au. However, a higher

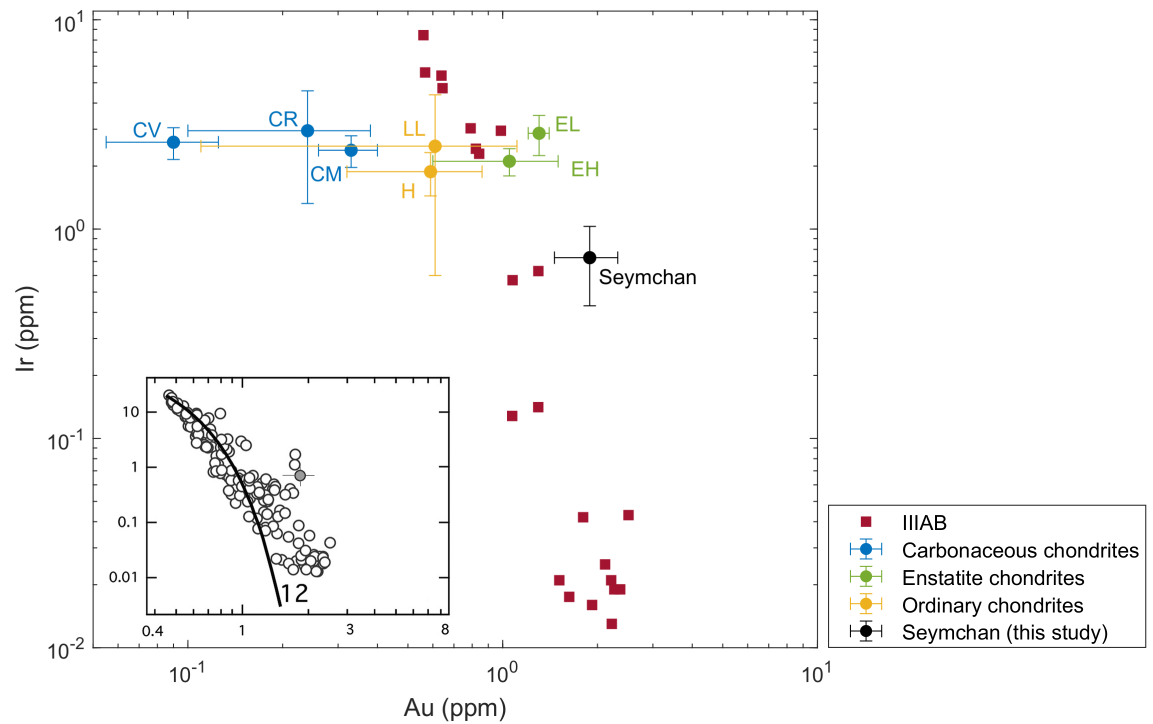


Figure 3.15: Au vs. Ir plot showing the difference between IIIAB irons (Wasson 1999), different types of chondrites (carbonaceous, van Kooten et al. 2022; ordinary, Kadlag & Becker 2017; enstatite, Kong et al. 1997, Kadlag et al. 2019) and Seymchan (this study). The inset is a figure from Goldstein et al. (2009), showing Ir vs. Au concentration in IIIAB meteorites, matched to a fractional crystallisation model which corresponds to 12 wt% S. The bulk concentration in Seymchan is shown by the grey circle on the inset.

Au content makes the distinction between pallasite metal and chondritic metal more pronounced in this case, strengthening the argument against a chondritic origin for the metal in Seymchan.

A small error may also be introduced due to some Au and Ir measurements being below the detection limit of ~ 0.1 ppm (1.4% of measurements in both cases; Table 3.3). These measurements were removed from the mean compositions, which may have increased the means by a small amount. However, 1.4% is within the existing errors for both elements, so this does not influence the argument against chondritic origin.

The similarity of Ir and Au contents in pockets and matrix, together with the overall low Ir and high Au abundances, strongly suggest that the metal in the pockets is not chondritic. The deviation from chondritic composition is consistent with magmatic fractionation, suggesting that the metal took part in core formation, which does not support the metal pockets being primary and isolated since metal insertion.

3.4.1.2 Chemical differences between matrix and pockets

From the statistical results outlined in Section 3.3.2.2, the element compositions of the two groups are identical, with the exceptions of Os in kamacite, and Fe, Ni, Os and Ir in taenite. However, Table 3.3 shows that the means of these elements are within error of each other (kamacite: $Os_{matrix} = 1 \pm 0.51$ ppm vs $Os_{pocket} = 0.78 \pm 0.35$ ppm; taenite: $Fe_{matrix} = 81.96 \pm 2.85\%$ vs $Fe_{pocket} = 83.86 \pm 2.79\%$, $Ni_{matrix} = 17.24 \pm 2.77\%$ vs $Ni_{pocket} = 15.79 \pm 2.39\%$, and $Os_{matrix} = 1.36 \pm 0.49$ ppm vs $Os_{pocket} = 0.94 \pm 0.52$ ppm), indicating that the concentrations are similar enough to be indicative of the same iron meteorite class.

Ir in taenite is more distinct ($Ir_{matrix} = 1.25 \pm 0.24$ ppm vs $Ir_{pocket} = 0.64 \pm 0.31$ ppm), but this could be due to the low number of datapoints in taenite, as there is overlap between the two Ir datasets in Fig. 3.11. Furthermore, the mean Ir values in matrix and pockets, which account for the different volumes of kamacite and taenite present in the sample, are within error of each other (Table 3.3). Finally, taenite

lamellae have varying widths (from tens of μm to mm) and they are zoned, which could lead to scatter in the data.

If the Ir difference measured here is an accurate representation of the mean Ir content of taenite, a difference of 0.6 ppm is unlikely to be significant for the formation mechanism of pallasites, since Ir varies by several orders of magnitude in iron meteorite groups as a result of magmatic fractionation (Table 3.6). In IIIAB irons alone, Ir varies by three orders of magnitude, so a difference in Ir caused by mixing of different iron reservoirs would be expected to be more significant than 0.6 ppm difference.

When evaluating chemical differences in differentiated meteorites, it is helpful to consider the scale of the variation expected to result from magmatic fractionation in all elements. Table 3.6 shows previously reported variations in the trace elements considered in this study, which cover at least two orders of magnitude in all elements, with the exception of Cu, which varies only by a factor of 50 (Scott 1972). This lends further support to the previous suggestion that the small statistical differences between metal and pockets in some elements are very unlikely to be due to the presence of a different iron metal class, and more likely to be due to instrumental error.

3.4.1.3 Chemical differences between individual pockets

Fig. 3.13 and Table A.2 show that all pockets have very similar trace element compositions, with some deviations in W in pocket 7, Os in pocket 10 and Os and Ir in pocket 11. Removing the pockets that do not meet the isolation criteria reviewed in Section 3.3.1 removes some of these element deviations, leaving only the low Os content in pocket 10 (Fig. 3.13C). However, the Os concentration is within error of the other pocket means and the matrix, suggesting that all pockets have compositions indistinguishable from each other.

Since cluster analysis on all pocket compositions does not detect any clustering beyond kamacite vs. taenite (Fig. 3.14), and because of the similarity in mean

Element	Number of orders of magnitude of variation	Iron meteorite groups measured	Reference
Ir	4	All	Goldstein et al. (2009)
Ge	5	All	Goldstein et al. (2009)
Ga	3	All	Scott & Wasson (1975)
Au	2	All	Scott & Wasson (1975)
Os	4	IIAB, IIIAB, IIF, IID	Pernicka & Wasson (1987)
Re	4	IIAB, IIIAB, IIF, IID	Pernicka & Wasson (1987)
W	2	All	Scott (1978)
Cu	1	All	Scott (1972)

Table 3.6: Reported magnitude of variation in the trace elements included in this study.

compositions outlined above and in Fig. 3.13, it does not appear necessary to maintain the distinction between the pockets that strictly meet the criteria outlined in Table 3.2 and those that do not, since they all have the same composition. For these reasons, further discussion refers to data from all metal pockets.

3.4.2 Pallasite-IIIAB link

To compare pallasite and IIIAB metal, I consider elements which do not undergo significant fractional crystallisation during core formation, such that the comparison is possible without the need to model fractionation. According to experimentally determined D_0 coefficients for solid-liquid partitioning in the Fe-Ni system (Chabot et al. 2017), the elements used in this study which are not expected to have fractionated strongly (D_0 close to 1) are Co ($D_0 = 0.95 \pm 0.03$) and W ($D_0 = 0.95 \pm 0.04$).

Fig. 3.16 shows Co and W plotted against Ni for Seymchan and Henbury (Fig. 3.16A), accompanied by histograms for the data for kamacite and taenite, respectively

(Fig. 3.16B, C). W shows good overlap between Seymchan and Henbury in both kamacite and taenite, which supports a genetic link between pallasites and IIIAB irons.

Co values are more distinct, which could be due to the kamacite-taenite partitioning of Co being affected by cooling rate. Co partitions into kamacite, as suggested by the ratios between Co content in kamacite over taenite (Co_k/Co_t) determined by Rasmussen et al. (1988) and Goldstein et al. (2014), which are consistently >1 in all iron meteorite groups studied. In Fig. 3.16, kamacite Co (Co_k) is lower in Henbury relative to Seymchan, while the taenite Co (Co_t) is slightly higher in Henbury (Tables 3.3, 3.5). This leads to a lower Co_k/Co_t ratio in Henbury than in Seymchan, which indicates less partitioning of Co into kamacite in Henbury. Goldstein et al. (2014) measured the Co and Ni contents of several iron and stony-iron groups, and showed a systematic decrease in the Co_k/Co_t ratio with increasing cooling rate (see Fig. 14 in Goldstein et al. 2014), since slow cooling allows more time for the kamacite phase to grow, and for Co to partition into kamacite. This explains the observed difference in Co contents in the kamacite and taenite of the two studied meteorites as a consequence of the difference in cooling rates between pallasites and IIIAB irons, and lends further support to a genetic link. It should also be noted that while the Co_t datapoints for Henbury do not overlap Seymchan, the means of the two datasets do ($0.39 \pm 0.12\%$ in Henbury vs $0.35 \pm 0.03\%$ in Seymchan).

The bulk Au and Ir concentrations in Seymchan were plotted on top of a fractional crystallisation trend in IIIABs modelled by Goldstein et al. (2009) (Fig. 3.15 inset), showing good agreement with the expected Au/Ir fractionation trend. Au and Ir fractionate strongly in the liquid and solid respectively, having very different partitioning coefficients (Section 3.4.1.1). Therefore, the Au and Ir contents of Seymchan also support the link between Main Group Pallasites and IIIABs.

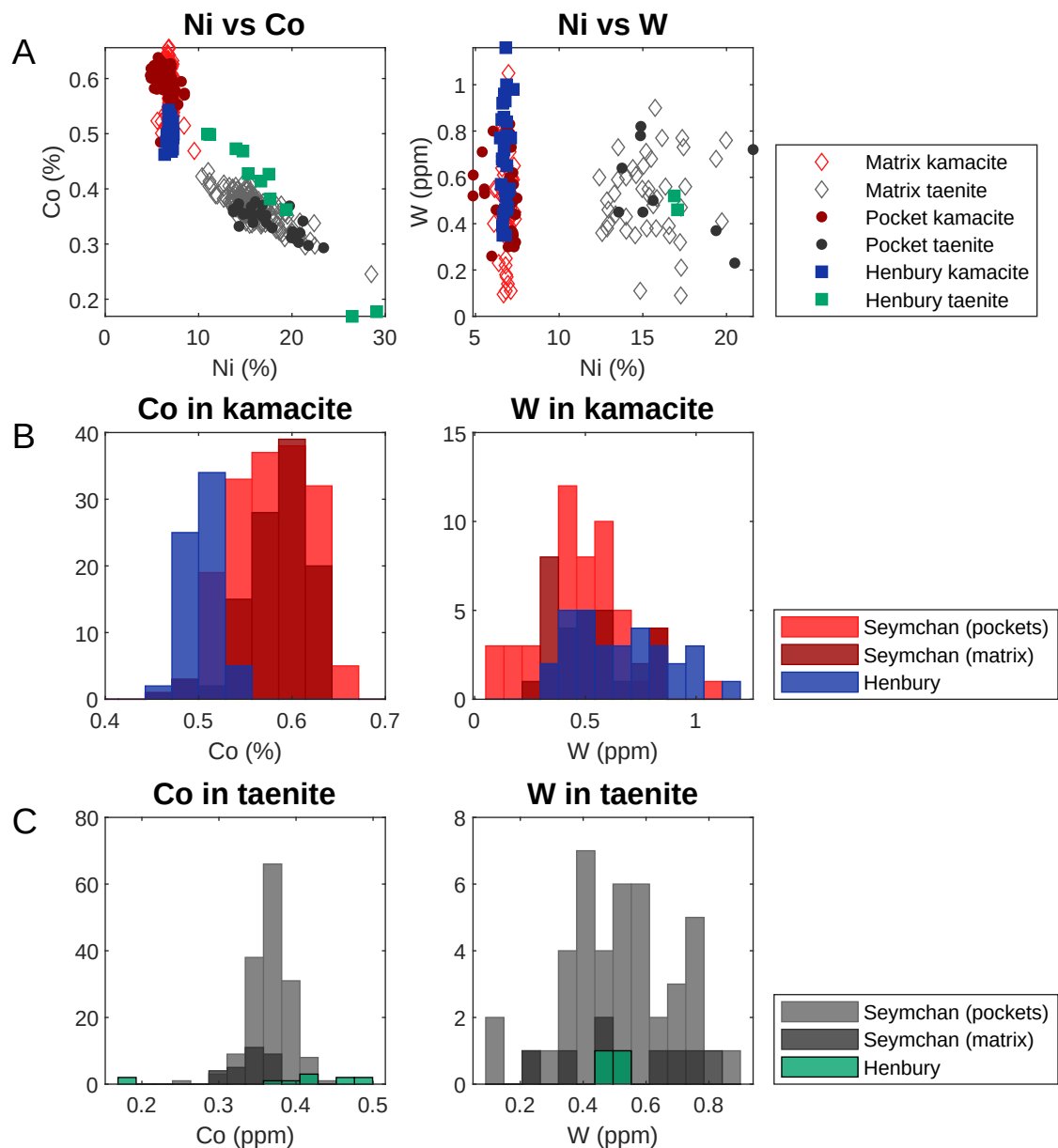


Figure 3.16: A. Scatter plots showing the Co and W compositions plotted against Ni, for Henbury and Seymchan (distinguishing between pocket and matrix for Seymchan). EMPA data is used for Co and Ni, and LA-ICP-MS data is used for W. Representative 1σ errors: 0.1% (Ni), 0.03% (Co), 0.4 ppm (W). B. Histograms showing overlap in Co and W between Henbury and Seymchan, for kamacite only. C. Histograms showing Co and W data for Henbury and Seymchan, showing good overlap for W. For Co, the scatter in Henbury taenite could be due to sampling difficulties outlined in the results section.

3.4.3 Formation of pallasites

The trace element analysis of the metal in Seymchan suggests no chemical difference between the matrix and the pockets (Section 3.3.2.2), as well as no difference among individual pockets (Section 3.3.2.3). Furthermore, Fig. 3.13 shows that the average composition of the kamacite in the matrix is representative of the average composition of all sampled kamacite. Taken together, the data in this study demonstrate that all the metal in Seymchan has the same composition, which is most readily explained by all metal having the same source. In this section, I discuss possible formation mechanisms that consider all metal having one source.

Based on the Ir and Au contents discussed in Section 3.4.1.1, the difference between the measured metal composition and the chondritic composition is consistent with magmatic fractionation. It is therefore very unlikely that any metal in Seymchan is primary (chondritic) or left over from partial differentiation, as suggested by Walte et al. (2020).

The data presented here provide further evidence for the hypothesised pallasite-IIIAB link (Section 3.4.2), in agreement with previous work (Burkhardt et al. 2011, Dottin III et al. 2018, Kruijer et al. 2022). The IIIAB link makes ferrovolcanism a very unlikely formation model for pallasites, since the only way to reconcile the different cooling rates experienced by the two groups (2.5-18 K/Myr in pallasites vs. 50-350 K/Myr in IIIAB; Yang et al. 2010) is through the impact history suggested by Kruijer et al. (2022). In this scenario, the IIIAB parent body collides with the pallasite parent body injecting IIIAB metal into the pallasite mantle, which leaves the IIIAB core exposed and able to cool very quickly. Kruijer et al. (2022) further suggest that without prior stripping of the core, the IIIABs would not have been able to cool as quickly as suggested by Matthes et al. (2020), who used Pd-Ag chronometry to estimate that the IIIAB core crystallised by 2.6 ± 1.3 Ma, in agreement with the fast cooling rate determined by Yang et al. (2010).

The compositional analysis of the metal therefore indicates a likely impact origin for pallasites. Textural observations from this study and previous work suggest that more than one impact contributed to the current textures in pallasites, since some secondary features are not texturally equilibrated in Seymchan (e.g., the veinlet linking pockets 2 and 3 in Fig. 3.3B), and they sometimes appear to cross-cut equilibrated features (the pockets). Experimental work performed by Walte et al. (2020) and Walte & Golabek (2022) showed that the metal pockets are texturally equilibrated with the surrounding olivines, as suggested by the high dihedral angles of the small pockets and the irregular shapes of the large pockets, both shaped by the static grain growth of the olivines. Based on the cross-cutting relationships between equilibrated and non-equilibrated features, and since the data presented here suggest that the metal in pockets has an impact origin, the fracturing and veinlet formation must postdate the metal-silicate mixing impact that brought the metal which eventually became pockets.

The unequilibrated nature of the fractures and veinlets is also supported by the textural study of other pallasites, which found annealed veinlets in Esquel, Fukang, and to some extent, Imilac (Walte & Golabek 2022). The different textures were interpreted as the result of burial to different depths, leading to different cooling rates and degrees of annealing that texturally equilibrate the secondary features in deeper meteorites. This is supported by the relatively fast cooling rate of Seymchan (7.1 ± 1.2 K/Myr; Yang et al. 2010), compared to the slower cooling rates of the other meteorites above (3.3 - 4.0 K/Myr for Esquel and Imilac, 5.8 K/Myr for Fukang, based on cloudy zone island size; Yang et al. 2010).

I therefore suggest the following order of events leading to the formation of pallasites:

1. The pallasite parent body is fully differentiated, such that little to no residual primary metal is left in the mantle from incomplete core formation (Fig. 3.17A). The IIIAB parent body impacts the pallasite parent body, injecting core metal into the

mantle of the pallasite parent body (Fig. 3.17B), as also suggested by Kruijer et al. (2022). Rapid cooling is then required in order to prevent the metal and olivines from separating due to their high density contrast.

2. A combination of annealing and static grain growth leads to metal pockets being trapped in olivine aggregates. The annealing results in textural equilibration of metal pockets and olivines (Fig. 3.17C).

3. Another deformation event (likely another impact) fractures the olivine aggregates, while the metal injected during stage 1 is still molten. This leads to secondary fractures in the olivine aggregates, as well as veinlets within the aggregates (Fig. 3.17D). The deformation event could be another impact elsewhere on the parent body, causing shockwaves that reach the pallasite-forming region, but this impact is unlikely to contribute any metal to the pallasites, since all metal has the same composition.

4. Annealing following the impacts leads to different extents of textural equilibration of the secondary features, resulting in the range of features observed by Walte & Golabek (2022) in different pallasites (Fig. 3.17 E). Since Seymchan has relatively fast cooling (7.1 ± 1.2 K/Myr; Yang et al. 2010), this stage does not lead to changes in its textures, but may affect other MG pallasites (Walte & Golabek 2022).

5. The pallasite parent body is fragmented by one or more impacts more recently (20 - 230 Myr, based on the cosmic ray exposure ages reported by Eugster et al. 2006), leading to loose pieces, some of which reached the Earth as meteorites.

In this model, the olivine aggregates are mantle restites which contain little to no metal left behind by incomplete core formation. If the pockets contain any primary metal, it would mix with the impactor's metal and change its composition, such that the current composition is a mixture between the original, chondritic composition of the metal on the pallasite parent body, and the impactor metal. However, this seems unlikely given the evidence for a genetic link between pallasite metal and IIIABs, where observed deviations between the two compositions can be explained through

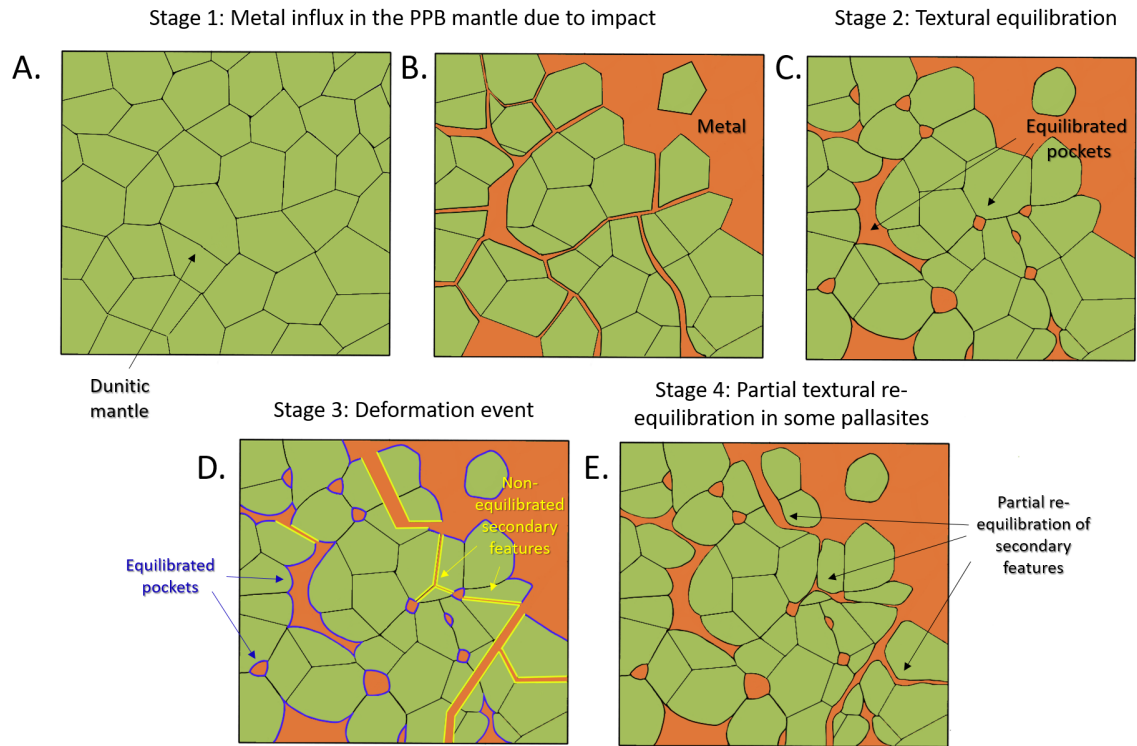


Figure 3.17: Stages 1-4 of the pallasite formation model presented here. A, B. An impact causes influx of molten metal in the mantle of the pallasite parent body, creating metallic veins in dunite and occasionally fracturing the olivines. The influx of metal is spatially heterogenous, such that olivine-rich areas can be preserved and separated by large veins of metal (e.g., top right of panel B). C. The olivine-metal mixture equilibrates texturally through annealing and static grain growth, leading to the types of pockets identified by Walte et al. (2020). D. A new deformation event (potentially due to shockwaves from another impact) creates secondary features as shown by the yellow lines, which are angular and show cross-cutting relationships to the previously equilibrated features shown by the blue lines. E. In some pallasites (e.g., Esquel, Fukang; Walte & Golabek 2022), further annealing leads to partial textural equilibration of the secondary features.

magmatic fractionation (Section 3.4.1.1). Furthermore, Fig. 3.15 shows that no mixture between chondritic and IIIAB metal could explain the composition of Seymchan, perhaps with the exception of a mixture between the most Ir-poor, Au-rich IIIAB composition and enstatite chondrites. However, the chondritic precursors to pallasites are likely to be ordinary chondrites (Sunshine et al. 2007), rather than enstatite chondrites. Furthermore, the amount of enstatite chondrite material needed to mix with IIIAB material to form the measured Seymchan composition is $\sim 76\%$, calculated based on the respective Ir contents of the two meteorites, since the measured pallasite composition plots so closely to the IIIAB trend. This value is significantly higher than the expected proportion of primary metal, even in Seymchan, which has the highest percentage of pockets of any known pallasite (up to 15%; Walte et al. 2020). Therefore, it is very unlikely that pockets of chondritic metal could have mixed with IIIAB metal to create the measured metal composition, further suggesting that no primary pockets existed in the mantle prior to the pallasite-forming impact.

3.4.3.1 Timescales

In terms of the relative timings of stages 1-3 above, fast cooling of the newly formed olivine-metal mixture is required to prevent the metal from separating from the olivines. At the same time, the textural equilibration of the pockets requires high-temperature annealing ($\sim 1300^\circ\text{C}$; Walte et al. 2020). To reconcile these seemingly contradictory requirements, I investigate the timescales required to form the annealed features while avoiding metal-olivine separation.

A model aiming to quantify the timescale of textural equilibration of metal pockets is not available, but the olivine growth model published by Solferino et al. (2015) achieves texturally equilibrated pockets (Fig. 1 in Solferino et al. 2015) as well as olivine growth and rounding, so I refer to this model to estimate the timescale required for pocket equilibration. According to Solferino et al. (2015), the time taken for an olivine crystal with an initial size of $0.1\ \mu\text{m}$ to grow to the average olivine grain size found in Seymchan (3.8 mm) at 1300°C would be approximately 6000 years. This is

calculated using the growth law and experimentally determined parameters reported by Solferino et al. (2015), for a 20 vol.% metal fraction (the growth law is given by Equation 1, and the parameter values can be found in Section 3.3 in their paper). The growth rate strongly depends on annealing temperature (1400°C leads to ~ 1600 years needed for olivine growth, whereas 1200°C leads to $\sim 27,000$ years), and on the metal fraction (more metal leads to slower olivine growth). Here, the metal fraction in pockets is $\sim 10\%$ (Walte et al. 2020), so the time estimates above represent upper bounds. Therefore, olivine growth to the grain size found in Seymchan is expected to be a geologically fast process (< 6000 years at 1300°C). Although olivine growth is not strictly needed for the model presented here (the mantle is fully differentiated in Fig. 3.17A, so the olivines are already coarse), since the annealing that causes growth also leads to equilibrated pockets, the timescale for pocket equilibration can be assumed to be either similar, or faster than the timescale for olivine growth.

It is unclear how long separation between metal and olivines would take, but Scott (1977) point out that separation would be impeded if the metal is partially molten, instead of fully molten. If the temperature of the metal was close to the liquidus of Fe-Ni-S melt (1400 - 1500°C) before an impact intruded metal into a slightly colder mantle, immediate cooling of the metal would produce a partial metal melt, while maintaining the high temperatures required for textural equilibration (1200 - 1300°C). The relative viscosity (viscosity divided by melt viscosity) of a partial melt increases rapidly during crystallisation (Fig. 3.18, from Costa et al. 2009). For example, Scott & Kohlstedt (2006) calculate seven orders of magnitude increase in relative viscosity between 50% and 65% solid fraction (Fig. 3.18). The curves in Fig 3.18 are calculated for magmas, but since only relative viscosity is plotted (instead of absolute viscosity), the same trend is expected to apply to metal. It is therefore likely that the viscosity increase during cooling below the Fe-Ni-S liquidus would inhibit metal flow, as suggested by Scott (1977). Furthermore, the presence of S lowers the solidus of the metal to $\sim 1000^\circ\text{C}$ (Scott 1977, Fei et al. 1997), which allows

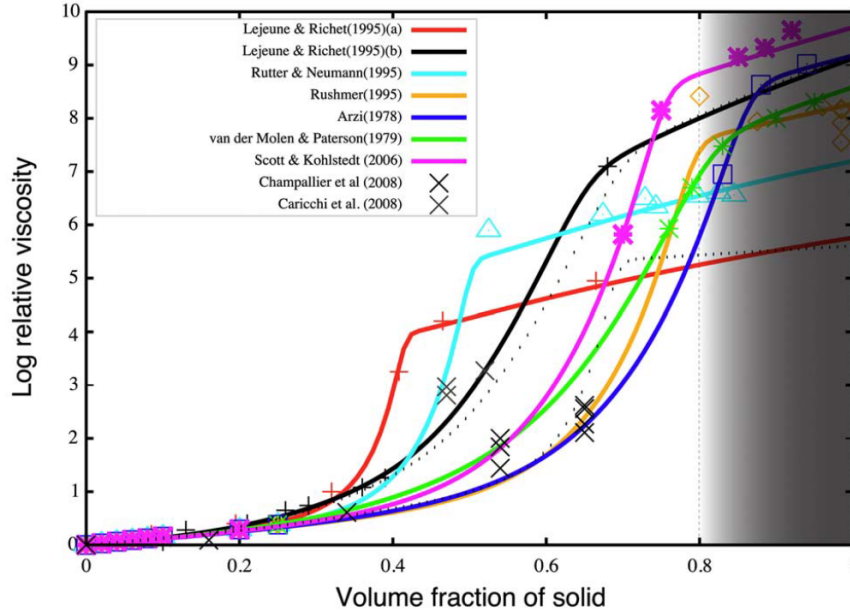


Figure 3.18: Relative viscosity as a function of volume fraction of solids in a crystallising melt (Costa et al. 2009).

the pockets to remain at least partially molten until the second deformation event in Stage 3 (Fig. 3.17D) creates unequilibrated metal textures such as fractures and veinlets. Given that equilibration of pockets was likely geologically fast, and since the metal was partially molten during annealing, the "pallasite problem" of olivine/metal separation is likely to be avoided in the formation model presented here.

Stage 4 occurs in pallasites that continue to be annealed after the second impact, leading to partial re-equilibration of the new (unequilibrated) deformation features formed during Stage 3 (Fig. 3.17E). The extent of partial re-equilibration was hypothesised by Walte & Golabek (2022) to be related to the final depth that a meteorite is buried at, since high temperatures persist for longer at greater depth. This is based on the study of four pallasites (Seymchan, Esquel, Imilac and Fukang), where the extent of post-impact annealing approximately correlates with the cooling rate. However, it should be noted that this correlation does not extend to all pallasites – for example, Glorieta Mountain is one of the slowest cooled MG pallasites measured (2.5 ± 0.3 K/Myr; Yang et al. 2010), and it still contains angular fragments, based on visual inspection of the images of this meteorite included in the LPI meteorite

database (accessed 23/09/2023). At the same time, Finmarken (fastest cooled MG pallasite, 18.2 ± 8.7 K/Myr; Yang et al. 2010) looks texturally similar to Seymchan, based on images in the same database. Although more annealing certainly leads to smoother features with time, this simple relationship may not be enough to explain the diversity of textures in pallasites. Future work could be directed at evaluating the influence of initial (pre-impact) olivine shapes and sizes, proximity to the impact sites, and complex cooling histories.

3.5 Conclusions

- Textural analysis of the pallasite Seymchan revealed a variety of metal and olivine textures which have previously been used to understand the formation of pallasites. I identified 11 equilibrated metal pockets trapped within olivine aggregates, as well as unequilibrated deformation features (fractures and veinlets within the olivine aggregates), likely indicative of an impact. The cross-cutting relationship between the texturally equilibrated pockets and angular, non-equilibrated veinlets suggests that the impact which formed the veinlets postdates pocket formation.
- In-situ measurements of Fe, Ni, Co, Cu, Ga, Ge, W, Os, Ir and Au contents of the pockets and matrix in Seymchan, performed using EMPA and LA-ICP-MS, show that the pockets are chemically identical to the matrix. Furthermore, the individual pocket compositions are within error of each other. This indicates that all the metal in Seymchan likely has the same source.
- The composition of the metal in Seymchan is distinct from previously reported chondritic metal compositions and it is instead consistent with a magmatic origin. Furthermore, direct comparison to the composition of Henbury (IIIAB iron) lends further support to a genetic link between pallasites and IIIAB irons, as suggested by previous work (e.g., Kruijer et al. 2022).

- All the metal having the same source is inconsistent with the partial differentiation formation model proposed by Walte et al. (2020). Furthermore, ferrovolcanism remains an unlikely formation scenario due to the IIIAB link and difficulties reconciling upward diking from the core with the different cooling rates of pallasites and IIIAB irons. This leaves impact origin as the most likely formation model for pallasites. However, the impact history is complex owing to the complicated textures found in pallasites. Since the metal has one (external) source, the metal pockets must have formed after the first impact. The pockets then equilibrated texturally with the olivines, before a second impact caused the secondary deformation features seen in the olivine aggregates, without any further influx of metal. Further annealing may lead to re-equilibration of the secondary features in some pallasites. Finally, one or more impacts created loose pallasite pieces, some of which reached the Earth as meteorites. This model reconciles previous textural observations with *in situ* chemical data presented in this chapter, by suggesting a complex impact history involving two or more impacts on the pallasite parent body.

Chapter 4

The effects of aqueous alteration on the magnetic mineralogy of CM chondrites

4.1 Introduction

4.1.1 Studies of the solar nebula magnetic field

The first stages of terrestrial body formation involve the very earliest processes in the solar system, when the first solids formed in the protoplanetary disk and started accreting to form planetesimals, planetary embryos and eventually, planets (Section 1.1). Although many details about the initial stages of accretion are still poorly constrained (Chambers 2004), it is likely that the dynamics of the nebular gas and dust were governed by the magnetic field acting on the protoplanetary disk (Bai & Goodman 2009, Bai 2017, Weiss et al. 2021).

Information on the duration and strength of the nebular magnetic field can be obtained by studying primitive meteorites such as chondrites, as they have avoided widespread melting, and their components formed early enough that they could potentially record a magnetic remanence attributable to the nebula field. For example, the strength of the field in the outer (e.g., CV chondrites, Fu et al. 2021a, CO chondrites, Borlina et al. 2021), and inner nebula (LL chondrites, Fu et al. 2014) have been used to argue for heterogenous magnetic fields throughout the nebula (Fu et al. 2021a) and disk mass loss between the inner and outer nebula (Borlina et al. 2021), reinforcing

the view that the early solar system contained two distinct reservoirs (carbonaceous and non-carbonaceous; Bermingham et al. 2020, Kleine et al. 2020). Furthermore, several studies have been used to infer the timing of solar nebula dissipation based on the absence of a magnetic field during the aqueous alteration of meteorites, when new minerals form and would be expected to record the nebula field if the field is still active (Gattacceca et al. 2016, Borlina et al. 2022).

Two types of measurements have previously been used to recover the strength of the nebula field: studies of chondrules, which can record pre-accretionary TRMs of the nebula field present when they form (Fu et al. 2014, Borlina et al. 2021), and bulk studies of aqueously altered meteorites. Aqueous alteration (Section 1.2.3) involves the formation of relatively high concentrations of magnetite and pyrrhotite, and consumption of metal (Palmer & Lauretta 2011, King et al. 2017), which has led to carbonaceous chondrites being used extensively for palaeomagnetism (Fig. 4.1). Given that alteration took place early in the history of these meteorites (2.3 - 8 Myr after CAI formation for CM chondrites; Fujiya et al. 2012, Jilly et al. 2014, Visser et al. 2020; see review by Suttle et al. 2021) and that metal is one of the first minerals to alter (Suttle et al. 2021, 2022), these palaeointensities have been attributed to the nebular magnetic field.

The field recorded by a meteorite depends strongly on the timing of magnetic acquisition and the heliocentric distance of the meteorite, because the nebula field is expected to decay both in time and with distance from the Sun (Weiss et al. 2021). Even so, there are wide variations in measured palaeointensities, both among different chondrite groups, and within each subgroup. Fig. 4.1 shows previous studies on carbonaceous chondrites that attribute the remanence of a chondrite to the nebular field, and the recovered palaeointensities are colour-coded based on magnetic carrier. It is noted that palaeointensities recovered from magnetite-bearing meteorites (in particular, CM chondrites) are lower than palaeointensities recovered from pyrrhotite and metal, with the exception of Winchcombe (Bryson et al. 2023), where the presence

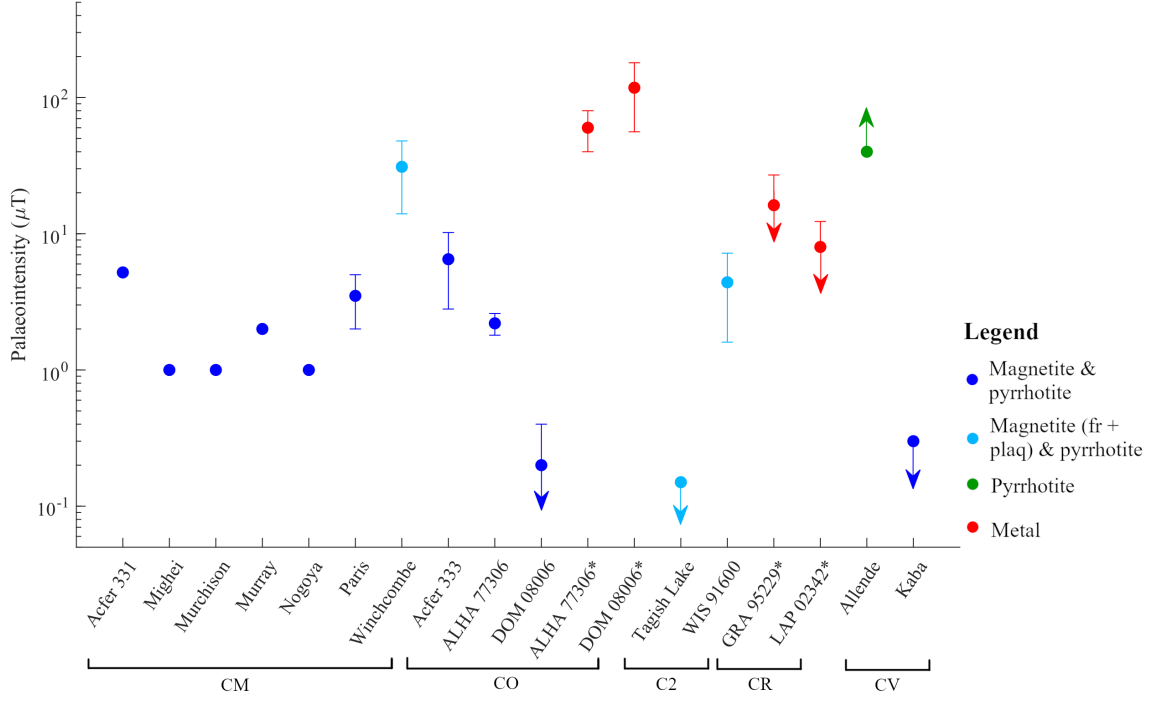


Figure 4.1: Magnetic palaeointensities recovered from carbonaceous chondrites (CM, CO, C2-ungrouped, CV, CR), with colours corresponding to the magnetic carriers in each meteorite. The blue circles indicate meteorites which contain magnetite, whether or not it contributes to the remanence. In addition, light blue circles are known to contain framboidal and plaquette magnetite. The measurements for meteorites marked with * are measurements from individual chondrules. Only the most recent palaeointensity estimate is included for each meteorite. References: all CM chondrites (except Acfer 331 and Winchcombe) – Cournède et al. (2015); Acfer 331 – Gattacceca & Rochette (2004); Winchcombe – Bryson et al. (2023); Acfer 333 – Gattacceca & Rochette (2004); DOM 08006 & ALHA 77306 chondrules – Borlina et al. (2021); and matrix – Borlina et al. (2022); Tagish Lake – Bryson et al. (2020a); WIS 91600 – Bryson et al. (2020b); CR chondrites – Fu et al. (2020b); Allende – Fu et al. (2021a); Kaba – Gattacceca et al. (2016).

of framboidal and plaquette magnetite likely indicates a different formation mechanism for magnetite (Sridhar et al. 2021). The low palaeointensities recorded by most CM chondrites are not consistent with measurements from other meteorites formed in the outer nebula (e.g., the chondrules in CO chondrites; Borlina et al. 2021), or with those formed in the inner nebula (the chondrules in LL chondrites; Fu et al. 2014). Together with the recently discovered discrepancies within the CM chondrite group (Bryson et al. 2023), these observations highlight the importance of studying the alteration pathways leading to the formation of the magnetite in order to inform palaeomagnetic interpretations.

In CM chondrites, the remanent magnetisation seems to be carried predominantly by pyrrhotite, and magnetite does not appear to carry a remanence, except in Murchison (Cournède et al. 2015). This is based on thermoremanent curves showing a drop at 200 - 300°C for all meteorites measured (except Murchison), which is indicative of the remanence being carried primarily by pyrrhotite. For Murchison, the drop is at 550°C, which was interpreted as magnetite. In all samples analysed, Cournède et al. (2015) found that the magnetic susceptibility is dominated by magnetite, regardless of whether the remanence is carried by magnetite or pyrrhotite.

Furthermore, Sridhar et al. (2021) found that the magnetic mineralogy of CM chondrites is dominated by $<0.1 \mu\text{m}$ single-domain and $0.1 - 5 \mu\text{m}$ vortex state magnetite, using First Order Reversal Curves (FORC) diagrams. Therefore, magnetite is present in the size range that allows it to carry a stable signal.

The question of why magnetite does not appear to be magnetised in most CM chondrites is interesting because it may affect palaeointensity recovery in chondrites. If a sample contains both pyrrhotite and magnetite, but only pyrrhotite carries a remanence, then the sample's NRM will be weaker than it would have been if all carriers were magnetised. Palaeointensity methods require the entire sample to be magnetised by a lab-induced field (e.g., an ARM; Section 2.3.3.4), which magnetises all phases in the sample, regardless of whether they were originally uniformly mag-

netised. Therefore, if the NRM of a sample is only carried by pyrrhotite, then the ARM lost vs. NRM lost line will have a shallow slope, leading to an underestimated palaeointensity (Bryson et al. 2023). This could have implications for the strength and timescale of dissipation of the solar nebula (for example, the nebula may have dissipated later than inferred; Borlina et al. 2022).

4.1.2 Magnetite formation

Magnetite can form via three alteration pathways in meteorites: oxidation of metal (Palmer & Lauretta 2011), oxidation of troilite (Sridhar et al. 2021), and oxidation of pyrrhotite, to form magnetite-pentlandite intergrowths (Singerling & Brearley 2020). When magnetite forms from metal, it can form either through pseudomorphic replacement of the metal, or precipitation from Fe-rich solution (Palmer & Lauretta 2011). Complete pseudomorphic replacement leads to grains composed entirely of magnetite, which may contain trace Ni inherited from the metal. Incomplete replacement leads to grains with metal cores surrounded by magnetite rims. By contrast, precipitated magnetite contains little/no Ni, because Ni stays behind when the metal dissolves (Palmer & Lauretta 2011), as measured in framboidal magnetite by Kimura et al. (2013). Magnetite grains formed through precipitation tend to be much smaller (100-200 nm in C2 chondrites; Tonui et al. 2014, Bryson et al. 2020b) than pseudomorphic magnetite (tens of μm ; Palmer & Lauretta 2011). Framboids and plaquettes are types of magnetite formed through precipitation (Greshake et al. 2005), but precipitative magnetite can also occur as isolated matrix grains (Render et al. 2022).

4.1.3 Explaining the weak magnetite remanence

There are a few possible explanations for magnetite carrying a weak magnetisation, and they are schematically presented in Fig. 4.2. The metal precursor of magnetite is assumed to be magnetised within an individual chondrule, based on previous work on CO chondrites (Borlina et al. 2021) and LL chondrites (Fu et al. 2014). During

accretion, magnetised chondrules are incorporated in a chondritic parent body in random orientations, such that their pre-accretionary remanences cancel out, producing no net remanence (Borlina et al. 2022). Smaller primary metal grains are also incorporated into the matrix of the chondrite. When magnetite forms from metal, it can either become magnetised by the local field (assuming that a field is present), or it can inherit the magnetisation of the metal (e.g., Heider & Dunlop 1987, Jiang et al. 2017, Ge et al. 2021). These two cases (here referred to as "new magnetisation" and "inheritance") are end-points, and the same sample could theoretically contain a mixture of magnetite that acquires a new magnetisation, and magnetite that inherits the metal's magnetisation (Sridhar et al. 2021). The three cases (new magnetisation, inheritance, and the intermediate case) are detailed below.

1. New magnetisation (Fig. 4.2.1) If magnetite does not inherit its remanence from metal, it would likely be able to record the field present during alteration, leading to a uniform magnetisation in the sample (Fig. 4.2.1). Both the chondrules and the matrix would be uniformly magnetised, although unaltered metal may still display different magnetisation directions.

2. Inheritance (Fig. 4.2.2) If magnetite is able to inherit the remanence of the metal when it oxidises, it would appear unmagnetised, because it would preserve the random orientations of the pre-accretionary remanence (Borlina et al. 2022, Bryson et al. 2023). Therefore, if a field is present during alteration, it would likely not be recorded by magnetite (Fig. 4.2.2).

3. Intermediate (Fig. 4.2.3) If different reactions contribute to the formation of magnetite in the same sample (for example a combination of pseudomorphic replacement of metal, where the remanence may be inherited, and oxidation of troilite to form framboidal magnetite, where the remanence is newly acquired), the two types of magnetite could be magnetised in different ways and/or have different recording

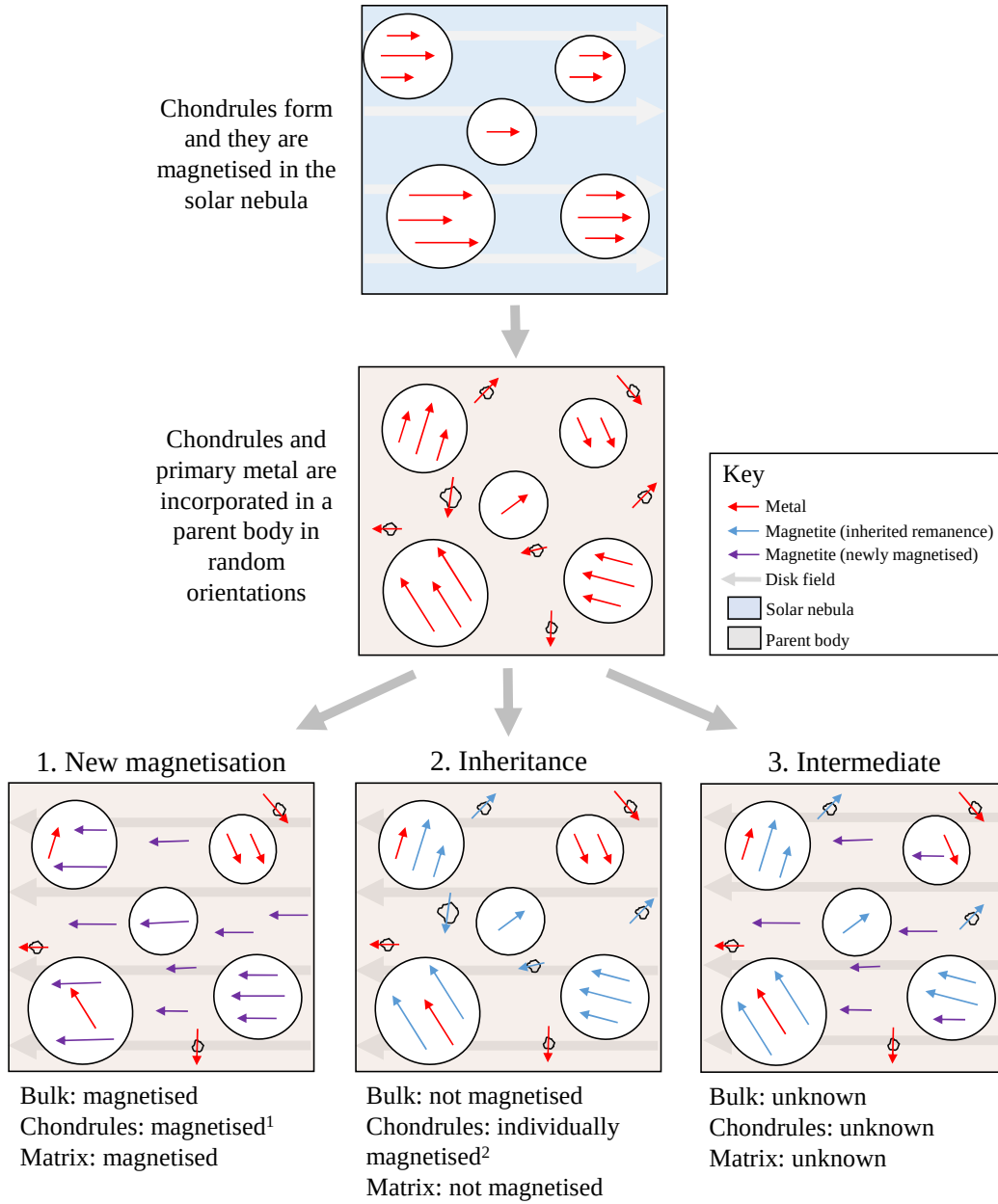


Figure 4.2: Schematic showing how different magnetite formation pathways could affect the bulk magnetisation of a meteorite. Top 2 panels: The chondrules are assumed to be magnetised in the solar nebula (Borlina et al., 2021) and to accrete in the parent body in random orientations, leading to randomly oriented chondrules that each carry a uniform pre-accretionary remanence. 1. New magnetisation – if magnetite can become magnetised by the local field, the bulk signal would be uniformly magnetised. The chondrules would also be magnetised in this case, but some chondrules where the metal remains unaltered may still carry a different, pre-accretionary remanence (¹). 2. Inheritance – if magnetite inherits the pre-accretionary remanence of the metal, the sample would remain unmagnetised in bulk, but each chondrule would be carrying a uniform pre-accretionary remanence(²). (*Continues on the next page*)

Figure 4.2: (*Continued*) 3. Intermediate case – if some magnetite acquires a new magnetisation and some inherits the metal’s magnetisation, the resulting bulk signal would be complex. The bulk signal may have a weak net remanence if enough magnetite became magnetised, but this is difficult to predict. Some chondrules may remain uniformly magnetised if all magnetite in a chondrule inherited the metal’s magnetisation. Note that in panels 1 and 3, the location of the newly formed magnetite does not have to coincide with the location of metal, since magnetite formed through precipitation does not have to be physically associated with metal (Palmer & Lauretta 2011).

fidelities (Sridhar et al. 2021). This would lead to a complicated bulk signal where only some magnetite is uniformly magnetised, which might result in a weaker-than-expected bulk magnetite remanence (Bryson et al. 2023).

If no field is present during alteration, magnetite would be unmagnetised regardless of whether it inherits a remanence from metal. However, if it inherits its signal from metal, the chondrules would still be expected to be magnetised even if no field is present. By contrast, if all magnetite forms via a different mechanism, chondrules where most metal has been replaced by magnetite would also appear unmagnetised, which may be a way to distinguish between these two scenarios. However, it is likely that a field was present during alteration, given that the alteration took place early (2.3 - 8 Myr after CAI formation; Suttle et al. 2021), while the solar nebula is believed to have dissipated by 3.94 and 4.89 Myr in the inner and outer nebula, respectively (Wang et al. 2017, Weiss et al. 2021). Furthermore, FeNi metal is one of the first minerals to oxidise to magnetite during aqueous alteration (Suttle et al. 2021, 2022), as suggested by the high abundance of magnetite in the least altered CO chondrites (e.g., DOM 08006; Davidson et al. 2019), strongly suggesting that the nebula field was still active during magnetite formation.

The key to distinguishing between these scenarios is to spatially resolve the magnetic remanence of the different phases in CM chondrites, and to link the remanence to the individual magnetic grains. If magnetite is magnetised differently than pyrrhotite but similarly to metal, that would constitute strong evidence for magnetic inheri-

tance. By contrast, if some magnetite appears aligned with pyrrhotite, and some aligned with metal within a chondrule, that could suggest that the two populations may have formed in different ways.

4.1.4 Chapter outline

In this chapter, I outline a petrologic and magnetic study of the CM chondrite MCY 05231 using QDM (quantum diamond microscope) maps of spatially resolved magnetic signals (Glenn et al. 2017, Fu et al. 2020a), combined with a petrologic study on the formation of magnetite. I find that magnetite likely forms through three pathways (pseudomorphic replacement of metal, precipitation from Fe-rich solution, and pseudomorphic replacement of pyrrhotite; Section 4.4.1) and that the chondrule magnetic signals are random in both metal and magnetite, consistent with inheritance of remanence from metal to magnetite (Section 4.4.2). This contrasts to the matrix signal, which is unidirectional in both pyrrhotite and magnetite. I also show that the metal in the matrix is uniformly magnetised in a different direction than pyrrhotite and magnetite, suggesting that the matrix magnetite did not inherit its remanence from metal, but instead acquired it during alteration (Section 4.4.2). Finally, I discuss implications for the formation of CM chondrites (Section 4.4.5) and for bulk magnetic studies on chondrites (Section 4.4.6).

4.2 Methods

4.2.1 Sample

This study uses a thin section sample of MCY 05231, a highly altered CM chondrite (petrologic grade 1/2, where 1 is the most altered, and 3 is pristine). MCY 05321 was measured in bulk by King et al. (2017), who found that it contains 19.6% olivine, 3.4% enstatite, 1% calcite, 1.6% magnetite, 1.3% sulfides, and 73.1% phyllosilicates. Like all CM chondrites, the components of MCY 05321 are a fine phyllosilicate matrix, chondrules and chondrule fragments, rare AOAs (Amoeiboid Olivine Aggregates) and

CAIs, all of which are aqueously altered (Suttle et al. 2021). The sample size is $\sim 1 \times 1.5$ cm and it contains a small (~ 1.5 mm length $\times \sim 150$ μm width) fragment of fusion crust. The sample was provided by Dr. Ashley King and NASA’s ANSMET programme.

4.2.2 Microscopy

Microscopy was used to determine the mineralogy of the sample, and to identify regions of interest for further study. The regions of interest were imaged in order to link the magnetic signals from the QDM to the magnetic carriers.

The optical images were taken on a Leica DM750P microscope in the Earth Sciences Department at the University of Oxford, equipped with an incident light unit and a Leica ICC50 W optical camera, imaging up to 20x magnification. In addition, I used the Zeiss EVO SEM equipped with EDS and a backscatter detector in the Department of Material Sciences, University of Oxford, for imaging and semi-quantitative chemical analysis. The SEM was operated at 20 kV and 2.5 nA, in high-vacuum mode (pressure $\sim 10^{-6}$ mbar).

4.2.3 Quantum diamond microscopy

The Oxford geo-QDM (quantum diamond microscope) uses a diamond with ^{15}N nitrogen-vacancy (NV) centres, and measures 51 microwave frequencies within two ranges (2830 – 2842 MHz and 2898 – 2910 MHz), using 20 frame pairs per frequency, and averaging 10 sweeps each time. The QDM operates 900 μT bias fields cancelled to < 0.7 μT (Glenn et al. 2017), and produces maps with dimensions 2.496 x 1.560 mm, with pixel size 5.2 μm and 4x4 pixel binning. The data are processed using the QDMLab MATLAB toolbox (Volk et al. 2022) to produce and analyse magnetic maps. The QDM is also equipped with an optical camera, which creates images that allow petrologic features on each map to be identified and compared to reflected light optical images.

I identified 11 regions of interest (ROIs) in the sample, and the corresponding QDM maps covered approximately 25% of the exposed surface. The ROIs were chosen in advance through optical microscopy, focusing on regions containing magnetite and/or chondrules. Any grain within the main body of a chondrule or on the edge of a chondrule (within or before the fine-grained rim (FGR), if applicable) is considered as part of the chondrule. Where possible, the mapped regions are on the edge of the sample, which helped to locate them again in subsequent QDM runs.

The measurements were taken in contact with the diamond for maximum resolution and minimal noise, and sometimes with a higher sample-to-sensor distance (known as lift-off distance) in cases where highly reflective grains (for example, metal grains and large magnetite grains) cause scatter in the signal. This scatter leads to high-intensity noise over the grain, which can only be removed by measuring the sample at lift-off distance of 10 - 70 μm , with the exact value depending primarily on the size of the grain (larger grains cause more noise and require higher lift-off). This reduces resolution and increases the source-to-noise ratio, but it provides access to the highly reflective grains, at the expense of weaker signals which are no longer visible. Wherever possible, the quantitative analysis was performed on maps measured in contact, except for analysis of the reflective grains, which was performed on the minimum possible lift-off distance. This sometimes led to measurements at several lift-off distances in a single area. For example, location 8 has grains A and B well resolved in contact, grains C and D well resolved at 30 μm lift off, and grain E well resolved at 70 μm , so the quantitative analysis is performed on maps of the corresponding lift-off.

4.2.3.1 Data processing

To quantify the strength and orientation of the magnetic signals in the sample, I use the in-built QDMLab processing tools (unless otherwise specified below), to perform the following tasks:

- if needed, apply a threshold to the map to filter out hot pixels caused by highly reflective grains
- subtract the background field value using quadratic background subtraction
- upward continue the map by a small distance (Section 4.2.3.2) to increase the dipolarity of the sources and improve the dipole fit
- convert the resulting B_{111} map from the $\langle 111 \rangle$ diamond direction to a B_z map, with signals in the z direction
- align the B_z maps for the AF steps and ARM with the NRM map, to correct for misalignment or rotations between measurements
- select individual sources, or clusters of sources when they are too close to each other to be separated, such that the sources are surrounded by as much source-free space as possible
- perform least squares fitting of the resulting (upward continued) dipole with 10 repetitions to ensure accuracy, using the code provided by Fu et al. (2020a).

This workflow was repeated for 53 sources, for the NRM, every AF step and the ARM (Section 4.2.4), and the QDMLab scripts were slightly modified to support batch operations. In each case, the dipolarity parameter D was calculated as defined by Fu et al. (2020a):

$$D = 1 - \frac{RMS_{residual}}{RMS_{original}} \quad (4.1)$$

where $RMS_{residual}$ is the root-mean-square of the residual map (original map minus fitted dipole) divided by $RMS_{original}$, the root-mean-square of the original map. D approaches 1 for a magnetic source that can fully be described by a dipole, but will have low values for more complex magnetic sources (e.g., with quadrupole or octupole components). The workflow is shown in Fig. 4.3.

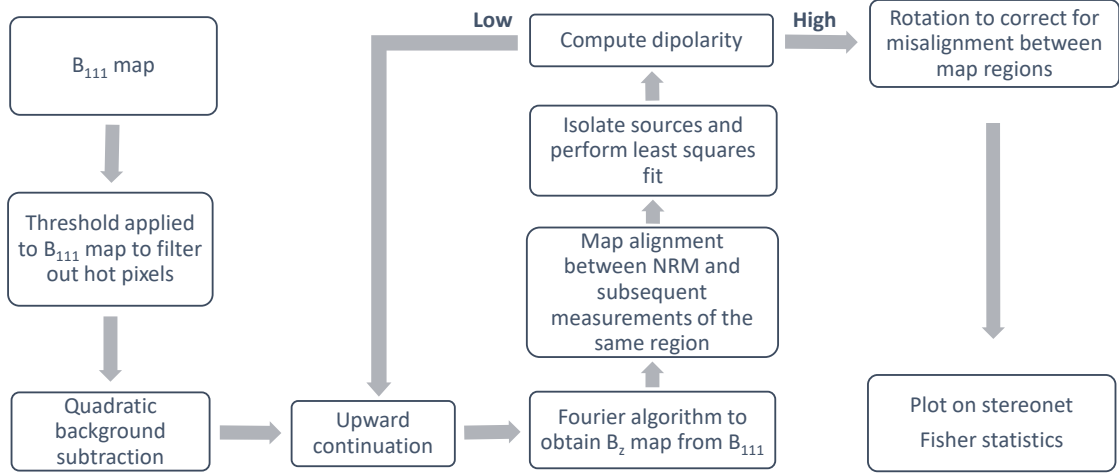


Figure 4.3: Workflow for net moment inversion for each magnetic source analysed, modified from Fu et al. (2020a).

The errors associated with the net moment inversion depend primarily on the dipolarity of the source. Fu et al. (2020a) found that for $D > 85\%$, the magnitude error is $\leq 10\%$ and angular error $\leq 8^\circ$, and that the errors gradually increase with decreasing D . For D between 50% and 60%, the magnitude error is $< 30\%$ and the angular error is $< 20^\circ$ (Fig. 5c and 5d from Fu et al. 2020a).

Only dipolar sources were fitted using the algorithm above (e.g., Fig. 4.4A), and any magnetic signal with a quadrupole component (e.g., Fig. 4.4B) and/or with a dipolarity parameter below 50% (e.g., Fig. 4.4C) was excluded from the analysis. Additional reasons to exclude a fit from the analysis included noise on the highly reflective grains which cannot be removed through measurement at high lift-off distance (e.g., Fig. 4.4D), NRM signal splitting into several distinct signals (e.g., Fig. 4.4E) or becoming too faint during demagnetisation (e.g., Fig. 4.4F), and sources on the edge of a map being partially or completely cut off by misalignment between measurements (e.g., Fig. 4.4G). In the ARM, some dipoles were also excluded because of high background signal caused by the applied field, making it difficult to isolate the original source (e.g., Fig. 4.4H). In addition, two dipoles in each set could not be fitted by the algorithm. The criteria outlined above resulted in 23% of the NRM fits being excluded, along with 36% of the 25 mT step fits, 40% of the 50 mT step

fits, 36% of the 100 mT step fits, and 40% of the ARM step fits. The declinations and inclinations of the remaining dipoles were plotted on equal area stereonet using Stereonet (Allmendinger et al. 2011, Cardozo & Allmendinger 2013), and used to perform Fisher statistics (Section 4.2.6).

The QDM images were overlain on top of the SEM images in order to match each magnetic signal to its source, and to mutually orient the QDM maps, since the SEM stage was not rotated between images. Any misorientation among different QDM maps were corrected for by rotating the dipoles $\sim$ since the analysis was performed on thin sections, any misalignment is limited to the plane of the section, so a rotation can be corrected for by simply adjusting the declination of the B_z signals by the rotation angle.

4.2.3.2 Upward continuation

Upward continuation (UC) is a data processing algorithm which extrapolates the magnetic signals on a map to a specified distance above the map, effectively averaging out signal of wavelengths below the UC distance. UC is used to increase the dipolarity of complex (non-dipolar) sources, reduce noise, or average several dipolar signals. The choice of UC distance is important, as too much can introduce error due to edge artefacts. It is therefore recommended that the upward continuation distance does not exceed 25% of the characteristic map dimension (Fu et al. 2020a), which here corresponds to 390 μm .

The upward continuation distance used here varied depending on the nature of the signal. All signals were upward continued by 5 μm in the first instance, to reduce measurement noise without altering the source signal. For complex sources, a higher UC (usually 30 μm) was used to increase dipolarity and improve the dipole fitting. In three instances, very high UC (150 – 200 μm) was used to calculate the average signal of an entire chondrule. UC was applied on the full B_{111} maps (2.50×1.56 mm), so 200 μm is below 25% of the map dimensions, preventing edge artefacts from introducing errors in the upward continued maps.

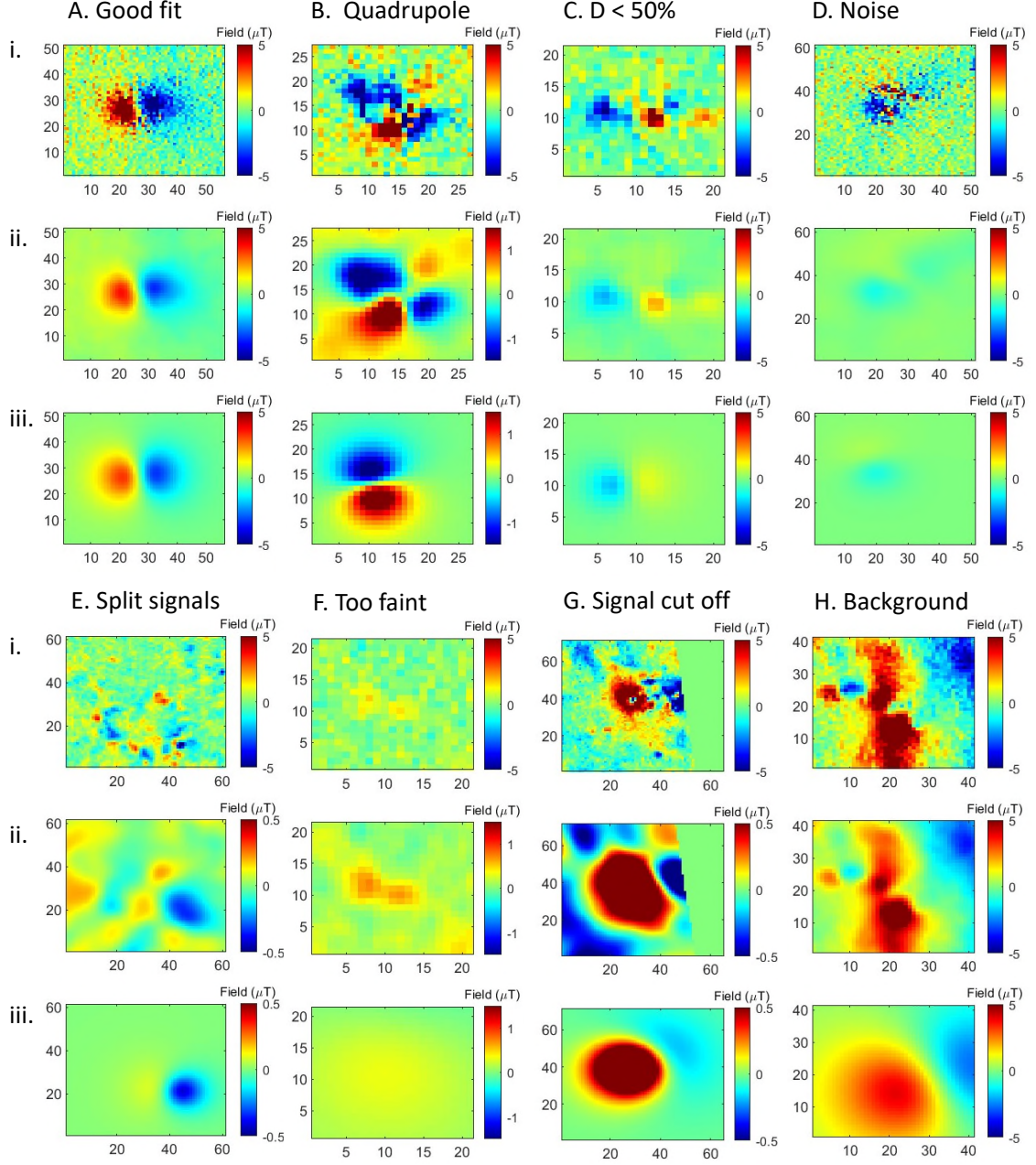


Figure 4.4: Examples of reasons to reject dipole fits from the analysis. Each set of maps A-H shows i. the cropped B_z map, ii. the upward continued B_z map, and iii. the dipole fitted to the upward continued map. A. Example of a good fit (datapoint 44 of the NRM, $D = 83\%$ at $UC = 10 \mu\text{m}$); B. Signal rejected as it needs a quadrupole fit (datapoint 10 of the NRM, $D = 39\%$ at $UC = 10 \mu\text{m}$). C. Signal rejected due to low dipolarity (datapoint 25 of the ARM, $D = 44\%$ at $UC = 5 \mu\text{m}$); D. Signal rejected due to noise on a highly reflective grain that could not be removed by increasing lift-off (datapoint 34 of the NRM, $D = 39\%$ at $UC = 20 \mu\text{m}$); E. Signal rejected due to splitting into several signals during demagnetisation (datapoint 45 of the 50 mT AF step, $D = 25\%$ at $UC = 30 \mu\text{m}$); (*Continues on the next page*)

Figure 4.4: (*Continued*) F. Signal rejected due to becoming too faint during demagnetisation (datapoint of the 25 mT AF step, $D = 43\%$ at $UC = 5$). G. Signal rejected as it is partially cut off due to misalignment during measurement (datapoint 1 of the ARM, $D = 52\%$ at $UC = 30$); H. Signal rejected due to background signal becoming too strong in the ARM (datapoint 53 of the ARM, $D = 48\%$ at $UC = 5$). UC = upward continuation.

If dipole fitting was unsuccessful (low dipolarity parameter and/or a fit that does not appear representative of the original signal), the algorithm was repeated with a different upward continuation distance, sometimes together with different cropping coordinates for the source region being fitted (Fig. 4.3).

4.2.4 AF demagnetisation

MCY 05231 was demagnetised using alternating field demagnetisation, in three steps: 25 mT, 50 mT and 100 mT, demagnetising in three orthogonal directions each time. The demagnetisation was performed using a D-2000 AF coil in the Earth Sciences Department, University of Oxford. The ROIs were then re-measured on the QDM after every AF step.

4.2.5 ARM experiments

After removal of the NRM by the AF steps, the sample was also given an ARM at a $200\ \mu\text{T}$ bias DC field in a 150 mT AC field, using a D-2000 AF coil, to determine how the magnetisation is acquired by different minerals. Using an ARM-TRM calibration factor of 3.33 for magnetite-bearing samples (Weiss & Tikoo 2014), this ARM is equivalent to a $60\ \mu\text{T}$ TRM. QDM maps in the same locations were then measured and compared with the maps of the NRM.

4.2.6 Fisher statistics

Mean directions and corresponding 95% confidence ellipses are calculated for the magnetic signals in each phase, for the NRM and every AF step. Furthermore, mean directions are calculated for all signals in the matrix and chondrules.

In addition, Watson’s randomness test (Watson 1956) is used to determine if there is directionality within the signals, or if they are random. The null hypothesis (random directions) is rejected if the resultant vector R of a set of vectors is higher than a critical value, which depends on the number of vectors in a dataset. The test is performed at 95% confidence level.

Finally, the F test (Eq. 4.2; Watson & Williams 1956) was applied to determine whether mean vector directions are statistically identical. Two datasets with sample sizes N_1 and N_2 , and resultant vectors R_1 and R_2 are compared using the equation:

$$F = (N - 2) \frac{R_1 + R_2 - R}{N - R_1 - R_2} \quad (4.2)$$

where N is the total sample size of the two datasets, and R is the resultant vector of all vectors in the two datasets. The resulting F value is compared to tabulated F values corresponding to 2 and $2(N-2)$ degrees of freedom, and if the computed F is below the tabulated value, the directions are statistically identical. The test was performed at 95% confidence level.

4.3 Results

4.3.1 Petrology

Optical microscopy and SEM analysis were used to identify the mineral phases present in the sample (Section 4.2.1), and here I focus on the morphology and distribution of the magnetic phases (magnetite, pyrrhotite and metal), and non-magnetic phases commonly associated with them (pentlandite, $(\text{Fe,Ni})_9\text{S}_8$, and tochilinite, $2(\text{Fe,Ni,Cu})_{1-x}\text{S} \times n(\text{Fe,Mg})(\text{OH})_2$, where x ranges between 0.08 and 0.28; Organova et al. 1974). In addition, I describe the chondrules and chondrule fragments encountered, and the magnetic inclusions that they contain.

The three magnetic phases were identified in the first instance based on their colour in reflective light (grey for magnetite, white for metal, Fig. 4.5A, gold for sulfides, Fig. 4.5E), and SEM-EDS was used to distinguish between pyrrhotite and

pentlandite, between magnetite and tochilinite, and to image the fine mineral intergrowths. In addition, magnetite is distinguished from tochilinite based on magnetism, as tochilinite is non-magnetic.

The metal in MCY 05231 is kamacite ($\text{Fe}_{93-95}\text{Ni}_{5-7}$), and it is most commonly found as circular chondrule inclusions (up to a few μm in size; Fig. 4.5E), or surrounding chondrules as larger grains (up to 100 μm , but most commonly 30 - 50 μm diameter; Fig. 4.5A). Metal grains are more rarely found in the matrix, within the 20 - 50 μm size range.

Pyrrhotite and pentlandite are usually found in the matrix, in grains up to $\sim 50 \mu\text{m}$. When they are intergrown, they can be distinguished based on colour in reflected light (pentlandite is lighter gold, while pyrrhotite is a darker gold, closer to bronze; Fig. 4.5E), and based on brightness in electron images (pentlandite is slightly brighter as its atomic weight is higher due to the presence of Ni; Fig. 4.5B). When the two phases are not found in close proximity, they were distinguished based on composition and magnetic signal, as pentlandite is non-magnetic. Pyrrhotite-pentlandite intergrowths (PPIs), as seen altered in Fig. 4.5B, are well documented in chondrites, and hypothesised to be primary features (Singerling & Brearley 2018, 2020). PPIs sometimes occur in association with micron-sized metal, and they are termed "micron-sized metal inclusion-bearing pyrrhotite-pentlandite intergrowth" (MMI-PPI) grains, as seen in Fig. 4.5E (Singerling & Brearley 2018). The primary pyrrhotite composition identified by Singerling & Brearley (2018) is very close to stoichiometric FeS (62 - 63 wt% Fe, corresponding to 48 - 49.5 atomic%, close to the ideal 50 atomic% Fe in troilite). In the altered PPI in Fig. 4.5B, a minor part of the pyrrhotite has the same composition as the primary pyrrhotite identified by Singerling & Brearley (2018), but the majority of the pyrrhotite contains 60 wt% Fe and 40 wt% S, which is consistent with monoclinic pyrrhotite (Fe_7S_8) formed through aqueous alteration. This confirms that the original PPI has been aqueously altered.

Magnetite occurs as rims mantling metal grains, indicating progressive oxidation

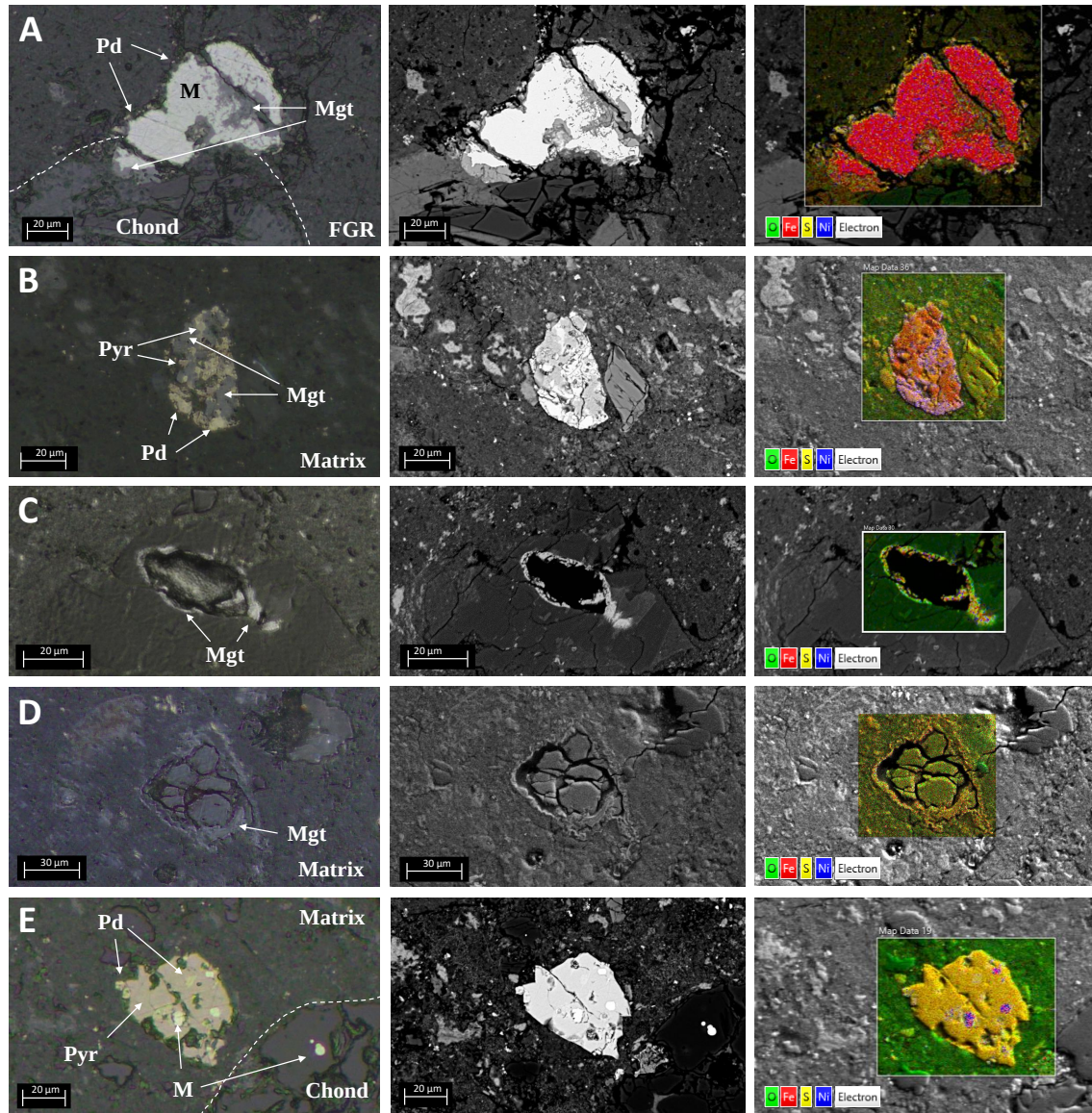


Figure 4.5: Occurrence of different magnetic phases in MCY 05231. From left to right, each grain is shown in a reflected light image, BSE image and EDS map. A. Metal grain at the edge of a chondrule, displaying magnetite mantling around edges and cracks, which is also surrounded by accessory pentlandite grains. B. Magnetite-pentlandite intergrowth in the matrix, with minor pyrrhotite, possibly as an altered PPI grain (Singerling & Brearley 2020). C. Magnetite rim around a hole in the matrix. D. Magnetite outline around a silicate grain in the matrix. E. Sulfide-metal intergrowth showing metal droplets surrounded by pyrrhotite, with minor pentlandite, suspected to be a MMI-PPI grain (Singerling & Brearley 2018). To the bottom right of the image, metal also occurs as micron-sized droplets in a chondrule. M = metal, Mgt = magnetite, Pyr = pyrrhotite, Pd = pentlandite, Chond = chondrule, FGR = fine-grained rim

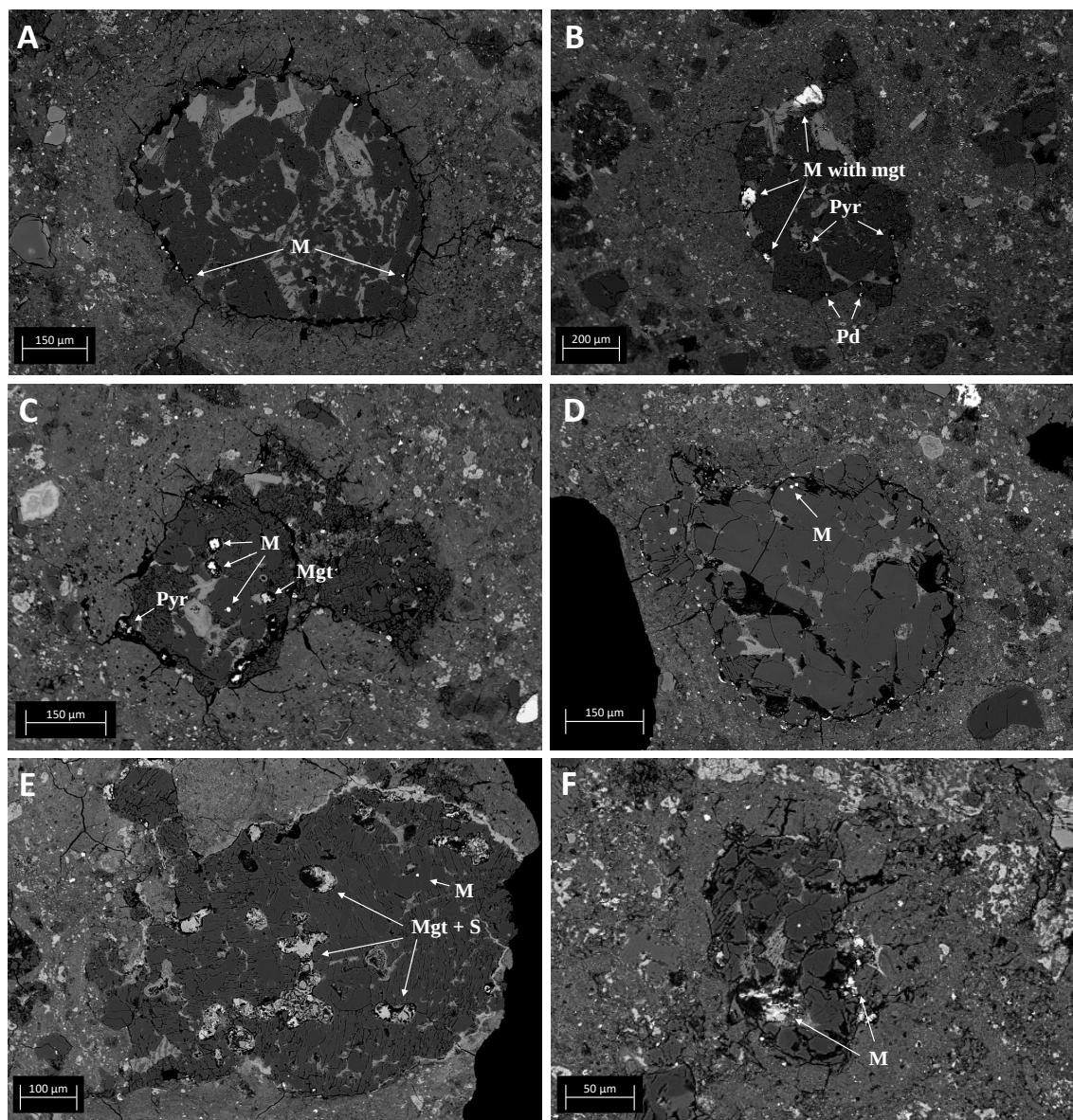


Figure 4.6: BSE images of chondrules in MCY 05231, and their magnetic inclusions. M = metal, Mgt = magnetite, Pyr = pyrrhotite, Pd = pentlandite, S = sulfides.

of metal to form magnetite (e.g., Palmer & Lauretta 2011) (Fig. 4.5A). Magnetite can also occur on its own, generally as smaller grains (e.g., 20 μm inclusion of magnetite in Fig. 4.6C). As metal is more common in chondrules than in the matrix, magnetite formed directly from metal is also more common in chondrules. Magnetite was identified in four grains in the matrix: intergrown with pentlandite and pyrrhotite (two grains, one of which is seen in Fig. 4.5B), and as outlines around other grains (Fig. 4.5C, D). The second outline (Fig. 4.5D) is less certain, as it was too thin to analyse with EDS without beam overlap with the adjacent phases. However, since it is rich in Fe and strongly magnetised (as seen in the top left map in Fig. 4.8A), it is likely to be magnetite. Finally, dendritic magnetite was found in the 150 μm fusion crust fragment on the sample (Section 4.2.1). No framboid or plaquette magnetite was observed, in line with previous studies outlining their scarcity in CM chondrites (e.g., Sridhar et al. 2021).

The matrix also contains very small (submicron) particles and sulfides and possibly magnetite, although complete identification was not possible due to the small sizes.

The chondrules in MCY 05321 range in size between 100 μm and 700 μm . They are usually composed of olivine and/or pyroxene and they are generally well preserved – the olivines are not completely altered to phyllosilicates and the fine-grained rims (FGRs) are still well-defined. Fig. 4.6 shows electron images of different chondrules imaged in MCY 05231 and the inclusions they contain. Their shapes are circular (Fig. 4.6A, D), but can also be flattened (Fig. 4.6B, F) or irregular (Fig. 4.6C). They contain inclusions of metal and/or magnetite which may be intergrown with sulfides, and rarely, inclusions of pyrrhotite on its own.

4.3.2 Magnetic description

The distribution of the areas mapped using the QDM is shown in Fig. 4.7. All magnetic phases are visible in the QDM maps. The strongest fields are produced by the larger grains of metal, exceeding 12 μT , and by magnetite in the fusion crust,

which locally exceeds $30 \mu\text{T}$ (location 10 in Fig. 4.7 and Fig. 4.9D). The upper limit of the fields in both cases is difficult to establish due to noise caused by the high reflectivity of the grains (Section 4.2.3). Small grains of metal, magnetite and pyrrhotite all produce signals in the $5\text{--}8 \mu\text{T}$ range, although magnetite signals tend to be stronger than pyrrhotite signals. Locally, magnetite can reach $12 \mu\text{T}$, such as in the chondrule in Fig. 4.12 (detailed below).

The strongest signals are produced by large isolated grains, and most of these signals are associated with the chondrules, because coarse metal and magnetite grains are more frequent. The background is also weakly magnetic ($\pm 2 \mu\text{T}$) in most of the sample – Figure 4.8 shows two examples of magnetic background: surrounding chondrules, both within the FGR and outside of it (Fig. 4.8A) and not associated with chondrules (Fig. 4.8B). The signal is demagnetised by a 25 mT AF field and likely corresponds to grains which are too small and uniformly distributed to produce resolvable dipolar signals, instead creating a ubiquitous faint background.

4.3.3 Net moment analysis

4.3.3.1 NRM

The results of the net moment analysis are shown in Fig. 4.10 (for the matrix) and 4.11 (for chondrules), and in Appendix C.

Fig 4.10 shows stereonet for the NRM (Fig. 4.10A) and demagnetisation behaviour (Fig. 4.10B) for the matrix, separated by the phase that makes up the majority of the surface of the grain. The NRM for the matrix reveals two distinct clusters. Pyrrhotite and magnetite are magnetised the same direction, with mean vectors at $D = 320.5^\circ$ and $I = 15.3^\circ$ ($\alpha_{95} = 38.1^\circ$) for pyrrhotite and $D = 335.6^\circ$ and $I = 10.6^\circ$ ($\alpha_{95} = 49^\circ$) for magnetite, within error of each other (the two α_{95} ellipses are overlapping and they contain both mean vectors). The F test (Watson & Williams 1956) was applied to these directions and resulted in an F value of 0.26, below the critical value (3.49), indicating that the directions are identical at a 95%

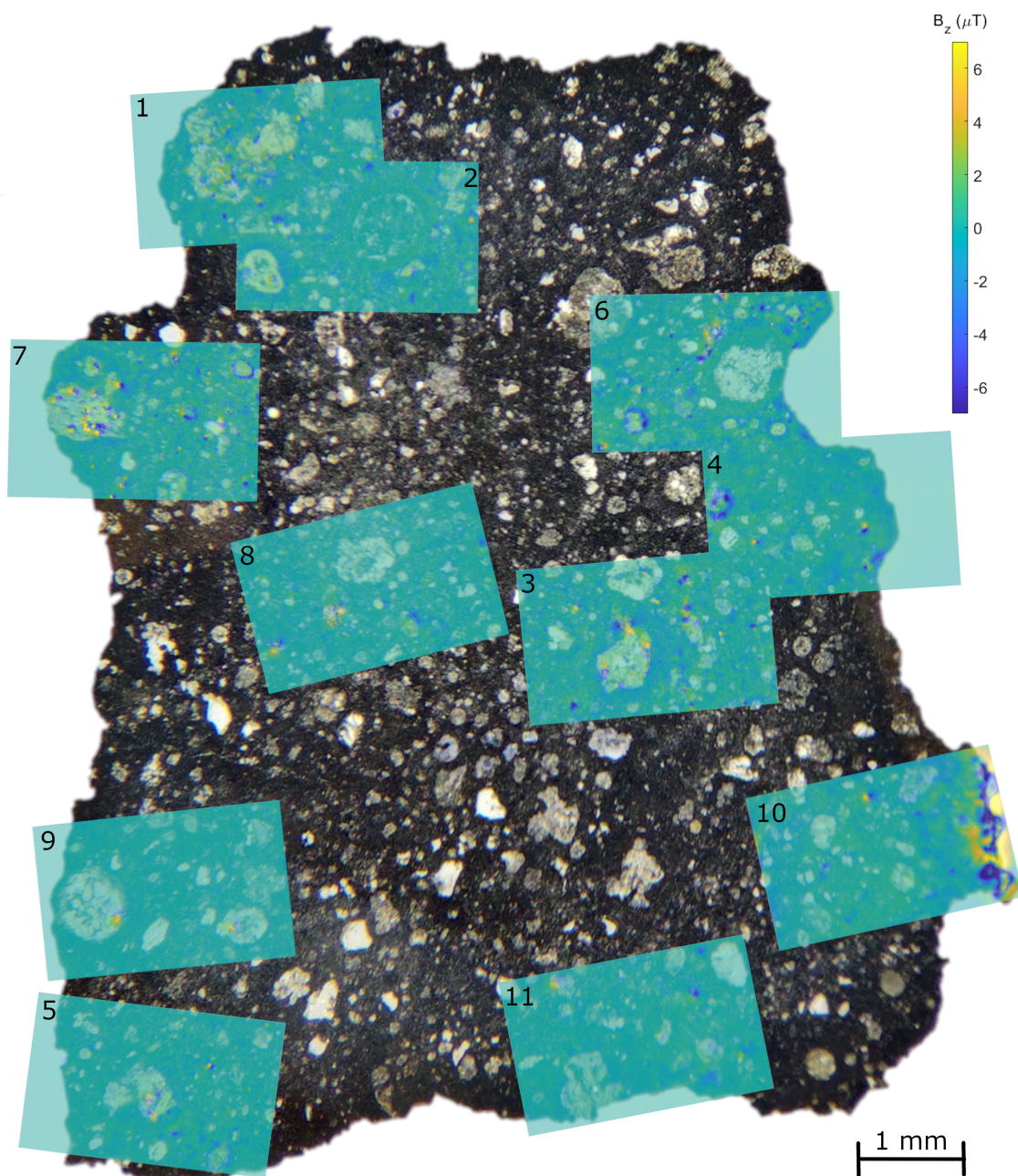


Figure 4.7: All QDM maps measured of the sample's NRM, overlain on an optical image of the MCY 05231 thin section. All maps have the same colourbar, shown on the right.

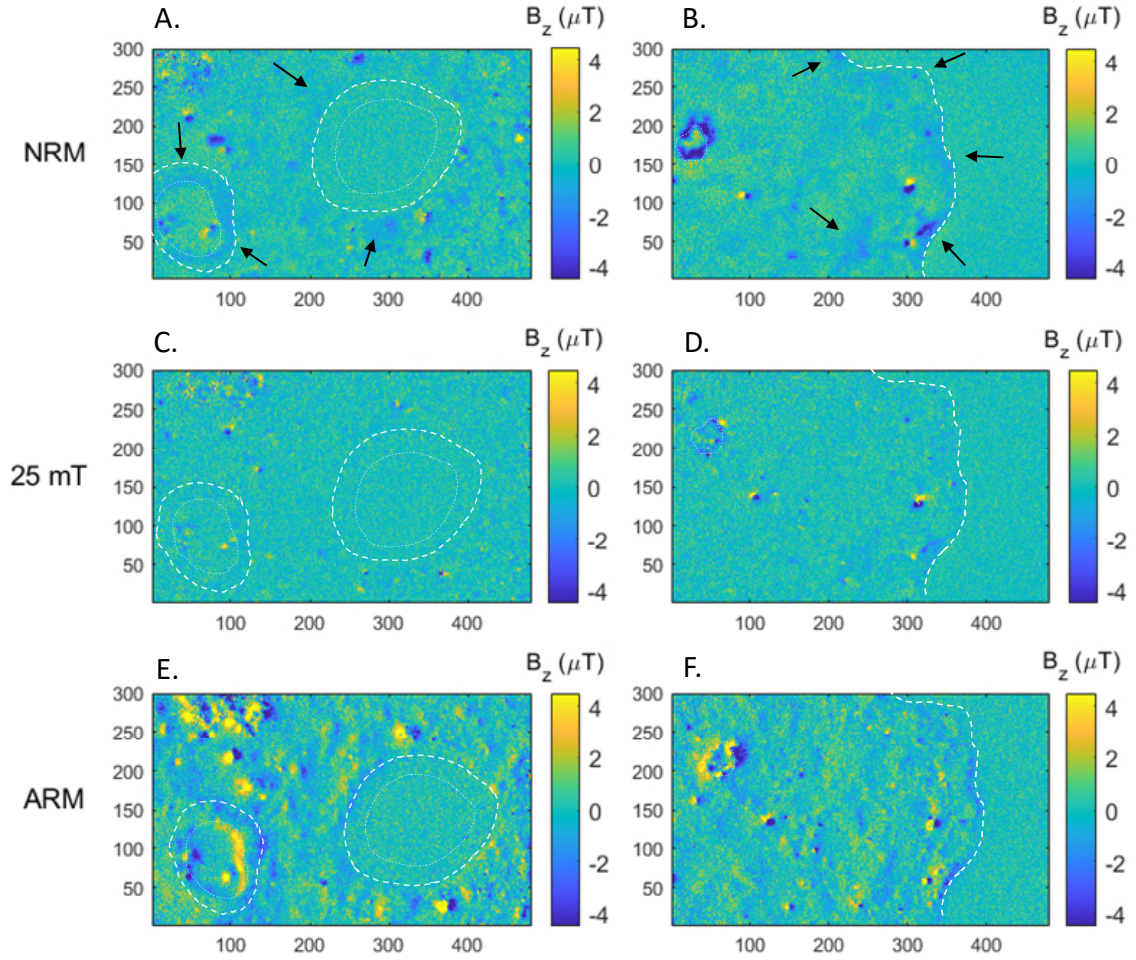


Figure 4.8: NRM, 25 mT AF and ARM of location 2 (left) and 4 (right), showing faint blue outlines around chondrules (in loc 2) and around the edge of the sample (loc 4) in the NRM. The background signals are demagnetised by a 25 mT AF field but become magnetised again by the ARM. The dashed lines outline chondrules (thin lines), their fine-grained rims (thick lines), and the edge of the sample. The black arrows indicate magnetised background.

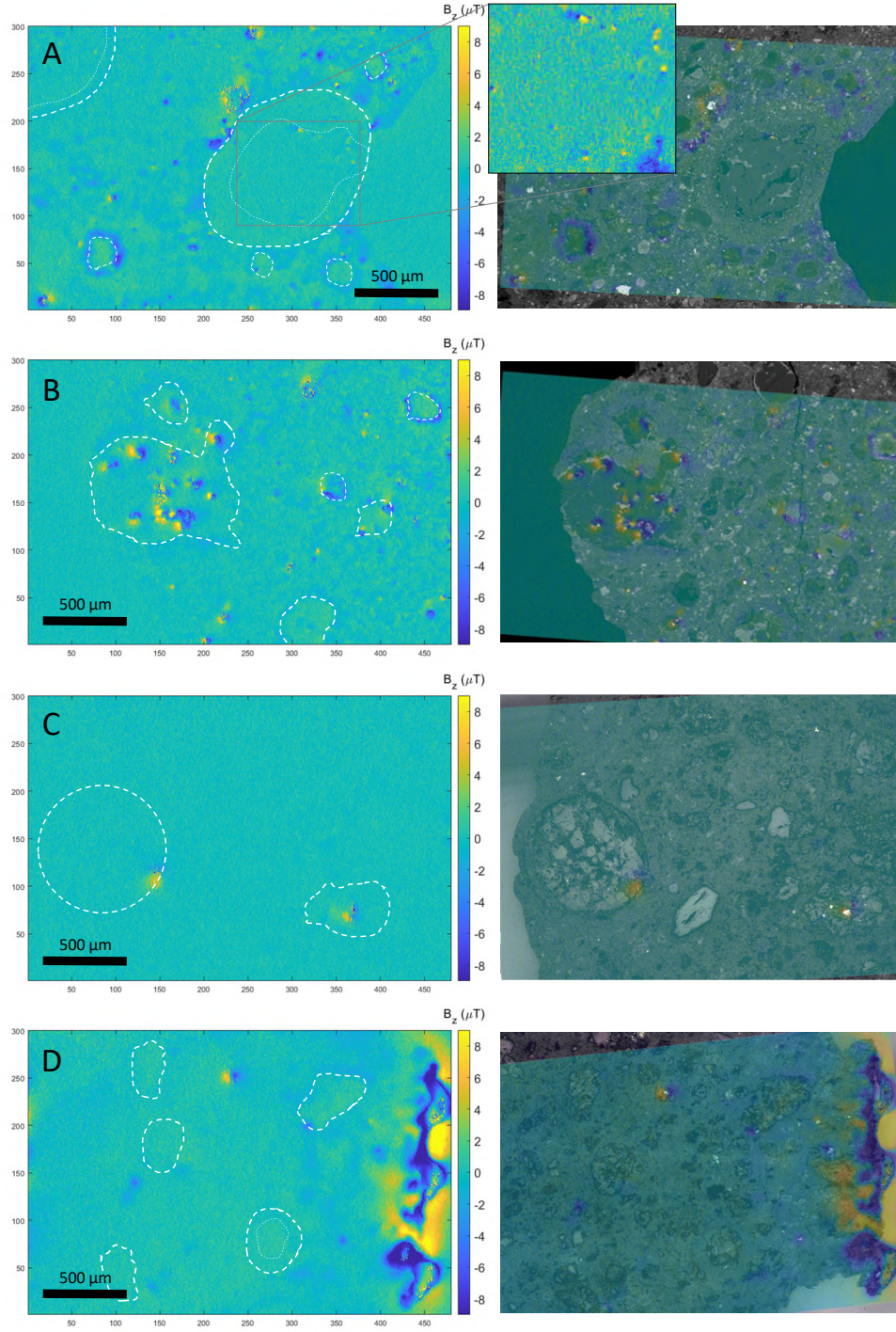


Figure 4.9: QDM maps of the NRM of different regions in MCY 05231, along with the same maps overlain on top of BSE and optical images. A. Location 6, showing a chondrule surrounded by an FGR, with small metal inclusions. The inset shows a magnified view of the inclusions, with exaggerated colorbar (-3 to $3 \mu\text{T}$). The inclusions are magnetised in a different direction than the matrix grains to the left of the chondrule, with the exception of one inclusion to the right of the inset, which corresponds to pyrrhotite. (*Continues on the next page*)

Figure 4.9: (*Continued*) B. Location 7, showing a chondrule with several unidirectional magnetite inclusions, magnetised in a direction orthogonal to the smaller chondrules to the right. C. QDM map measured at 30 μm lift-off showing two chondrules magnetised in different directions. The signal in the larger chondrule is carried by magnetite, and in the smaller chondrule it is carried by metal with minor magnetite. D. QDM map of the fusion crust, measured at 30 μm lift off. The background signal is slightly stronger within ~ 1 mm from the fusion crust, but the signal to the right of the map seems unaffected by the heating. The first two maps are overlain on BSE images, and the last two maps are overlain on optical (reflected light) images. The dashed lines trace the location of chondrules. Where two outlines surround the same shape, they indicate the FGR and the main body of the chondrule.

confidence level. The metal is magnetised in an approximately orthogonal direction ($D = 244.8^\circ$, $I = -9.6^\circ$, $\alpha_{95} = 19.1^\circ$), which does not overlap the direction indicated by magnetite and pyrrhotite (F value of 10.36, above the critical value of 3.44). The summary data for the stereonets (sample size, mean vector directions, α_{95} , k, mean length) are shown in Table C.6.

The Watson randomness test (Watson 1956) suggests that each individual phase, and the matrix as a whole are non-random to 95% confidence (Table 4.1). Furthermore, although the weak signals in the background of the matrix could not be fitted to dipoles, their N-S orientation suggests that they would plot similarly to the pyrrhotite and magnetite on the stereonets.

By contrast, the chondrules are randomly oriented both irrespectively of phase, and within each phase (Fig. 4.11A, Table 4.1). The stereonets in Fig. 4.11 only have one data point per chondrule, but in one instance, dipoles could be fitted to several signals within one chondrule. The signals are mostly unidirectional, as shown in Fig. 4.12, and they are oriented approximately 90° away from the other signals on the same map (Fig. 4.9B). The inclusions are a very fine intergrowth of Fe-S-O-bearing phases. The presence of magnetite is inferred based on EDS (phases dominated by FeO are present, but they always appear to contain 5-10% of S and Ni, likely due to the finely intergrown textures; Fig. 4.12A-E), the strong magnetic signals of the inclusions (9-12 μT , stronger than most signals produced by pyrrhotite), and the unidirectionality

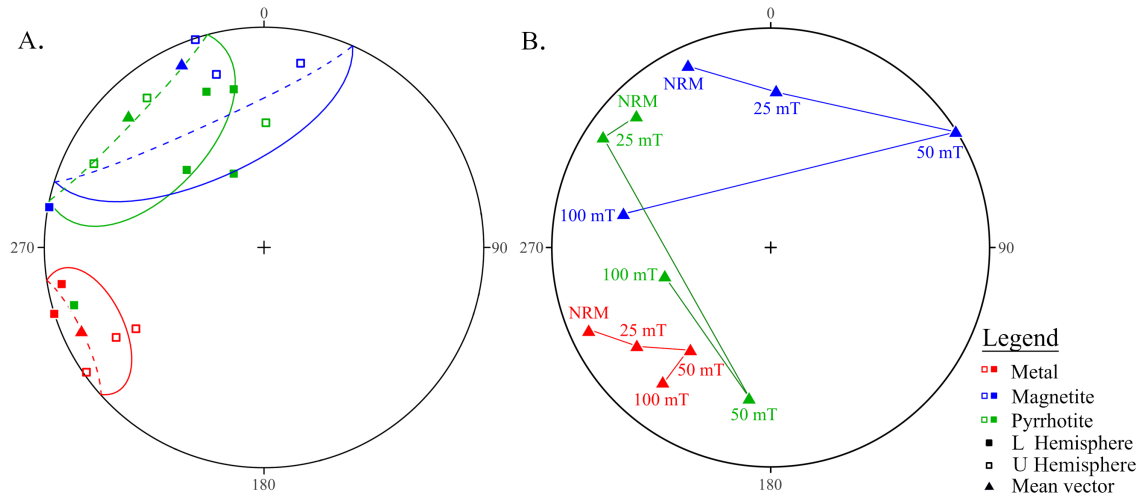


Figure 4.10: A. NRM and B. AF demagnetisation behaviour for the fitted dipoles in the matrix. The colours correspond to the magnetic phase that makes up the majority of the grain: pyrrhotite (green), magnetite (blue), and metal (red). The mean vectors for each phase are shown by triangles, surrounded by the $\alpha 95$ confidence circle when it could be calculated. For the demagnetisation behaviour (B), only the mean vectors for each AF step are shown. The data corresponding to each step are shown in Appendix C. Solid/hollow symbols correspond to the lower/upper hemisphere, respectively.

of the signals (Fig. 4.9B, 4.12). Unidirectionality within a chondrule would not be expected from any mineral apart from metal, or a secondary phase that inherits a signal from metal. However, the presence of metal is ruled out since it would be straightforward to identify based on high brightness in both optical and electron imagery. It is therefore suggested that these inclusions were initially composed of uniformly magnetised metal that has now been altered to a fine magnetite-sulfide intergrowth which inherited its magnetisation from the original metal (Sections 4.4.1, 4.4.2).

Another chondrule with several similarly oriented metal inclusions is shown in the inset of Fig. 4.9A. Because these inclusions produce very faint magnetic signals, only the top inclusion could be fitted. Nevertheless, the similar magnetic orientations of the inclusions could indicate that this chondrule is also magnetised.

Furthermore, visual inspection of Figure 4.9C reveals that the two signals on the map (produced by two chondrules) are oriented differently from each other. These

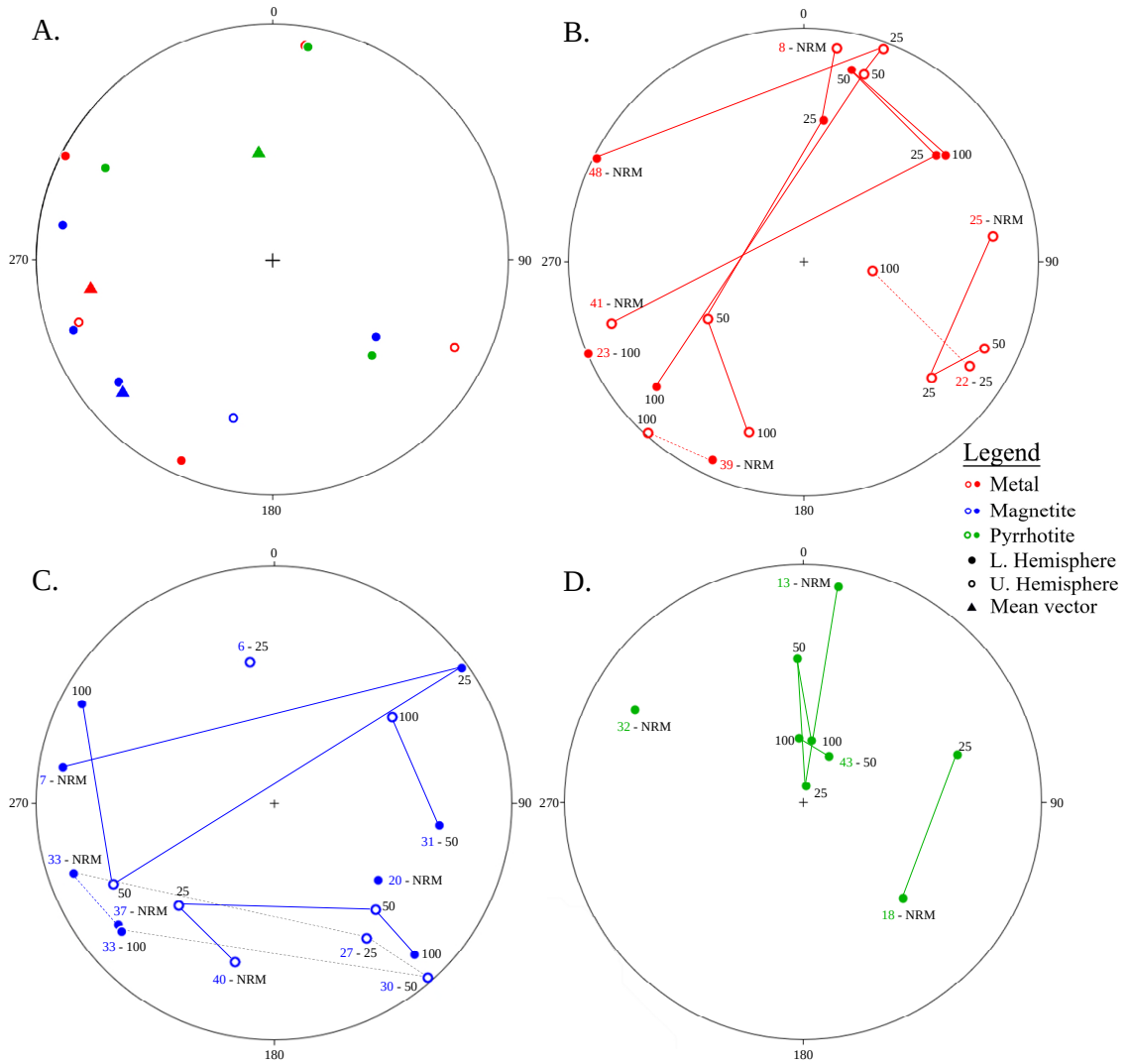


Figure 4.11: Equal-area stereonet showing the NRM (A), demagnetisation of metal (B), magnetite (C) and pyrrhotite (D) in each individual chondrule. The numbers in red, blue and green are the datapoint numbers (Appendix C), and the numbers in black are the AF intensities at each point (25, 50 or 100 mT). Dashed lines indicate one or two missing steps, when the signals corresponding to those steps could not be fitted to dipoles. The grey dashed lines in stereonet C link different signals from the same chondrule. Solid/hollow symbols correspond to the lower/upper hemisphere, respectively.

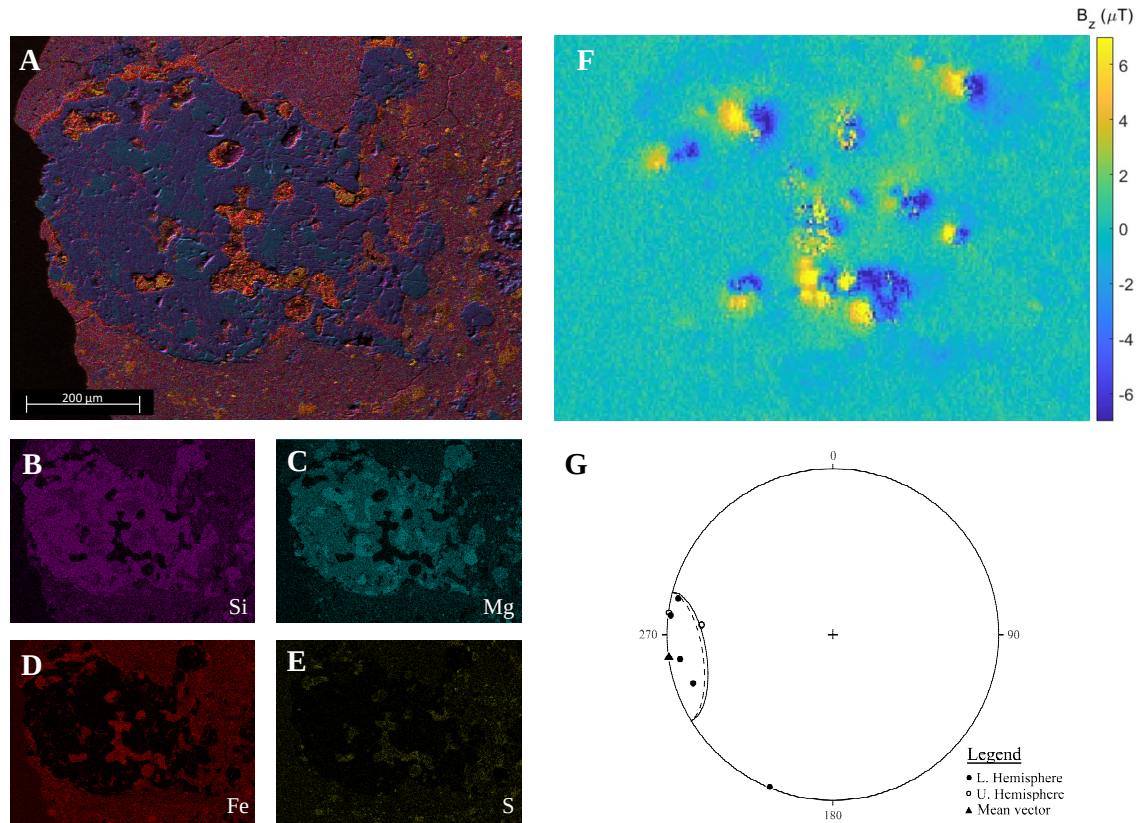


Figure 4.12: Chondrule showing unidirectional magnetic signals. A. EDS map; B. Si elemental map; C. Mg elemental map; D. Fe elemental map; E. S elemental map; F. QDM map of the NRM; G. Directions of the magnetic signals in the chondrule, shown on an equal-area stereonet.

observations highlight the fact that the matrix and chondrules behave differently from a magnetic perspective, which is discussed in Section 4.4.2.

4.3.3.2 AF demagnetisation

The mean vectors for the different phases at each AF step are plotted in Fig. 4.10B (matrix) and Fig. 4.11B-D (chondrules, separated by phase). In addition, a separate stereonet for each AF step is plotted in Fig. C.1.

The signals in each matrix phase generally become more scattered with each AF step, with the exception of magnetite in the 50 mT step, which is more clustered than the NRM or 25 mT step (Table C.6). Starting with the 25 mT step, all phases fail the Watson test for randomness (indicating that they are random) with the exception

of the metal, which is non-random at 25 mT but becomes random at 50 mT. The randomness test can only be applied to datasets with sample sizes greater than or equal to 5, so a few phases could not be tested (magnetite in the matrix and pyrrhotite in chondrules; Table 4.1).

The mean vectors for magnetite and pyrrhotite in the matrix remain statistically identical in the 25 mT step ($F = 1.17$, where the critical value is 3.56), but become distinct in the 50 mT step.

In the chondrules, the dipoles remain random throughout the demagnetisation sequence (Table 4.1). Fig. 4.11B-D shows the demagnetisation of each chondrule point, separated by phase. Only one point per chondrule was plotted. All stereonet show that most of the chondrules retain their magnetisation until 50 mT, when they randomise. Furthermore, many of the 25 mT and 50 mT points are close to each other in Fig. 4.11B, indicating a coercivity above 50 mT. There are a few exceptions of lower-coercivity grains that randomise by 25 mT (points 41 in Fig. 4.11B, and 7 and 33 in Fig. 4.11C). Incomplete sequences and missing steps are due to signals which did not pass the fitting criteria at certain AF steps. For point 33 in Fig. 4.11C, the 25 mT and 50 mT signals did not pass the fitting criteria, so different points in the same chondrule were used instead. This is the same chondrule as in Fig. 4.12, where the carrier is likely to be magnetite, so the lower coercivity is expected.

4.3.3.3 ARM

A 200 μ T ARM at 150 mT AF field was applied to the sample in order to understand how the magnetisation is acquired by different phases. The ARM remagnetised most grains, as shown in Fig. 4.13 and in the maps in Fig. 4.8. The mean direction of the magnetisation is 257.6° declination and -5.8° inclination ($\alpha_{95} = 9.6^\circ$), which is consistent with the direction that the ARM was applied in (270° declination and 0° inclination), accounting for potential error when manually orienting the sample prior to ARM acquisition. The background field carried by small grains in the matrix is

			NRM	25 mT	50 mT	100 mT
All data		R	18.89	5.29	3.36	8.69
		N	40	34	32	34
		R ₀	10.2	9.41	9.13	9.41
		R>R ₀	Y	N	N	N
Chondrules		R	4.1	4.04	1.62	2.82
		N	13	9	10	11
		R ₀	5.75	4.76	5.03	5.29
		R>R ₀	N	N	N	N
Matrix		R	11.58	4.71	3.47	6.35
		N	18	17	15	15
		R ₀	6.79	6.6	6.6	6.19
		R>R ₀	Y	N	N	Y
Chondrule	Magnetite	R	3.15	1.87	2.18	0.53
		N	5	4	4	4
		R ₀	3.50	n/a	n/a	n/a
		R>R ₀	N	n/a	n/a	n/a
	Metal	R	3.15	1.87	1.25	0.53
		N	5	5	5	5
		R ₀	3.5	3.5	3.5	3.5
		R>R ₀	N	N	N	N
	Pyrrhotite	R	1.49	1.78	1.90	2.00
		N	3	2	2	2
		R ₀	n/a	n/a	n/a	n/a
		R>R ₀	n/a	n/a	n/a	n/a
Matrix	Magnetite	R	3.33	3.16	3.44	2.00
		N	4	4	4	4
		R ₀	n/a	n/a	n/a	n/a
		R>R ₀	n/a	n/a	n/a	n/a
	Metal	R	4.76	4.30	1.82	2.60
		N	5	5	6	5
		R ₀	3.50	3.5	3.85	3.5
		R>R ₀	Y	Y	N	N
	Pyrrhotite	R	5.72	2.24	2.21	2.37
		N	8	7	6	6
		R ₀	4.48	4.18	3.85	3.85
		R>R ₀	Y	N	N	N

Table 4.1: Results of the Watson randomness test (Watson 1956) applied to the fitted dipoles in MCY 05231. The values for R (resultant vector), N (sample size), and R₀ (critical value based on N) are given in each case. The test result (whether R exceeds R₀) is 1 if the test succeeds (the null hypothesis of randomness is rejected and directions are statistically non-random), and 0 otherwise. The test cannot be performed on sample sizes below 5, which is indicated as n/a in the table. The results correspond to 95% confidence.

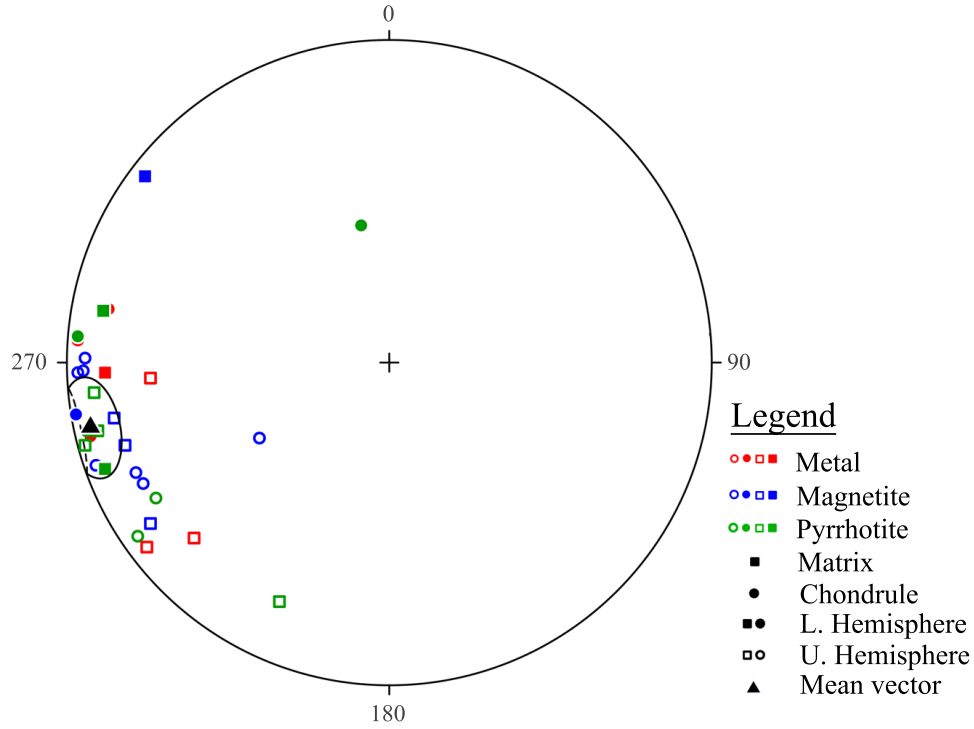


Figure 4.13: All fitted dipoles after the sample was magnetised by a $200\ \mu\text{T}$ ARM at 150 mT AF field applied in the y-direction of the map.

stronger in the ARM than it was in the NRM ($\pm 2.5\ \mu\text{T}$ in the ARM vs. $\pm 1.5\ \mu\text{T}$ in the NRM).

4.4 Discussion

In this section, I discuss the likely magnetite formation pathways in this sample (Section 4.4.1) and I interpret the magnetic record of the sample (Section 4.4.2). After ruling out atmospheric heating (Section 4.4.3) and the spatial resolution of the QDM (Section 4.4.4) impacting the results, I propose a magnetic history for CM chondrites (Section 4.4.5) and discuss implications for bulk palaeomagnetic studies (Section 4.4.6).

4.4.1 Magnetite formation

4.4.1.1 Pseudomorphic replacement of metal

When magnetite is seen in association with metal, either mantling metal grains (e.g., Fig. 4.5A) or in close proximity to metal (e.g., the magnetite inclusion in Fig. 4.6C), it is likely that the magnetite formed through pseudomorphic replacement of metal, a well-documented process observed by several previous studies (Palmer & Lauretta 2011, Sridhar et al. 2021). These grains contain trace Ni (2-3 wt.%) as observed by Palmer & Lauretta (2011) in magnetite formed through replacement of metal, although I note that the magnetite rims are often thin (up to 5 μm), so beam overlap with nearby metal (containing 5-7% Ni) is likely and could account for the apparent Ni content in magnetite. Although metal grains with magnetite rims are present in the matrix, their large sizes make them too noisy to be included in the QDM analysis, so they are not included in Fig. 4.10. However, the two magnetite outlines (Fig. 4.5B, C) are included, because the absence of metal prevented noise from affecting the signal. Based on the presence of Ni, it seems likely that the first magnetite outline (Fig. 4.5B) formed via replacement of metal, as the metal could have been plucked out during thin section preparation.

The other instance of magnetite occurrence is in association with sulfides (Fig. 4.6E, 4.12). Despite the potential influence of the sulfides, magnetic unidirectionality in a chondrule would be expected from chondrules containing metal that was magnetised in the solar nebula (Borlina et al. 2021). Metal can be readily identified in both optical and electron images based on its high brightness, but no metal was found associated with the intergrowth. However, the unidirectionality of the signal makes it very likely that the mineral assemblage in this chondrule also formed from metal, and that magnetite inherited the magnetisation. In addition, the size (30-150 μm) and morphology (irregular, droplet-like) of the magnetite-sulfide inclusions are also consistent with formation from metal. I therefore propose that this assemblage also formed through replacement of metal.

4.4.1.2 Pseudomorphic replacement of pyrrhotite

When intergrown with pentlandite and pyrrhotite, the magnetic portion is dominated by magnetite (as pentlandite is non-magnetic, and pyrrhotite forms a minor part of the grain; e.g., Fig. 4.5E). Therefore, magnetite is expected to dominate the magnetic susceptibility. Previous studies have identified such intergrowths as altered PPIs, where magnetite forms through pseudomorphic replacement of pyrrhotite (Singerling & Brearley 2020).

4.4.1.3 Precipitation from solution

Previous work on MCY 05231 suggests that the majority of magnetite in this meteorite is in single-vortex state, based on FORC diagrams (Sridhar et al. 2021). This corresponds to submicron sizes, making it difficult to identify these grains in microscopy and QDM maps, as the pixel size on the QDM maps is $5.2\ \mu\text{m}$. However, since there is a strong background signal in the matrix, and it becomes strongly magnetised after the application of the ARM (Fig. 4.8), the presence of submicron magnetic grains can be inferred. Furthermore, the background demagnetises by 25 mT, which is more consistent with magnetite than with pyrrhotite. This is because the coercivity of pyrrhotite (70-85 mT; e.g., Roberts et al. 2006) is higher than the coercivity of magnetite (up to 50 mT, more commonly 25-30 mT; e.g., Hodych et al. 1998). I therefore suggest that the background signal is caused by submicron magnetite grains. They could be formed through pseudomorphic replacement of submicron metal grains, which have previously been found in CO chondrites (Davidson et al. 2019). However, in CM chondrites, examples of pseudomorphic replacement are at least a few microns in size (Palmer & Lauretta 2011). It is more likely that the submicron magnetite instead forms through precipitation from Fe-rich solution, which has been documented to result in very small (100-200 nm) matrix grains in CI and C2 chondrites (e.g., Bryson et al. 2020b, Render et al. 2022). The magnetic orientation of this signal (roughly N-S; Fig. 4.8) makes it likely that the submicron

magnetite was magnetised by the same field that magnetised the pyrrhotite, which argues against inheritance of the remanence from an extant phase, consistently with magnetite formed through precipitation. This could be verified with higher resolution microscopy.

4.4.2 Magnetic record

Fig. 4.10 and Table 4.1 show that all magnetic phases are magnetised randomly in the chondrules, and that there are two uniform magnetisations in the matrix (one carried by magnetite and pyrrhotite, and one carried by metal). In this section, I discuss possible interpretations of these results.

4.4.2.1 Chondrules

Chondrules have previously been found to carry pre-accretional TRMs in their metal inclusions (e.g., Fu et al. 2014, Borlina et al. 2021, Fig. 4.1). When chondrules are incorporated into a parent body, it is expected that the gravitational collapse would lead to the random chondrule orientations (Borlina et al. 2021). Although each chondrule remains uniformly magnetised when considered in isolation, an assemblage of chondrules would display random directions. This is consistent with the results presented here, which show random directions among chondrules (Fig. 4.10), carried by metal and/or magnetite formed through pseudomorphic replacement of metal. Where more than one signal could be fitted to the same chondrule (in Fig. 4.12), the signals were mostly unidirectional within the chondrule, which is also consistent with previous chondrule studies. Furthermore, the demagnetisation sequences for each chondrule (Fig. 4.11) suggest that many of the chondrules with only one fitted dipole are also magnetised, since they retain a signal to 50 mT or higher, although this interpretation is limited by the coarse demagnetisation steps. Taken together, these observations suggest that the chondrules are individually magnetised, and that they were accreted in random directions during the formation of the parent body, in agreement with Fu et al. (2014) and Borlina et al. (2021).

4.4.2.2 Matrix

In the matrix, the two magnetic minerals that likely formed through alteration (magnetite and pyrrhotite) are magnetised in the same direction in both the NRM and the first AF step (25 mT), after which they become scattered (pyrrhotite at 50 mT, and magnetite at 100 mT; Fig. 4.10, C.1). This suggests that they were both magnetised by the field that was present while they were forming, acquiring a CRM, as previously suggested by Cournède et al. (2015). The metal in the matrix is also magnetised, in a different direction than the alteration products (Fig. 4.10). Possible interpretations of this signal are discussed in Section 4.4.2.3C.

4.4.2.3 Bulk magnetic record

A. Pyrrhotite

Considering the chondrules and matrix together, the only phase that may be magnetised in the same direction in both components is pyrrhotite. Although pyrrhotite is rarely seen in chondrules, the 2-3 accepted dipole fits of chondrule pyrrhotite plot closely to the matrix pyrrhotite in the NRM (Fig. 4.10). This is expected since pyrrhotite likely forms through alteration of troilite, and since troilite is non-magnetic, there is no signal to inherit from an extant grain and pyrrhotite would record the field present during alteration.

In the two examples where magnetite forms through alteration of PPIs in the matrix, it likely formed through alteration of primary pyrrhotite (Singerling & Brearley 2018). The primary pyrrhotite that Singerling & Brearley (2018) identified is Fe-rich and stoichiometrically very close to troilite (48 - 49.5 atomic%, close to the ideal 50 atomic% Fe in FeS). In pyrrhotite, magnetism generally increases with decreasing Fe content, as less Fe leads to more lattice vacancies (e.g., Halder 2017). The high Fe content of primary pyrrhotite makes it likely to be non-magnetic, which suggests that alteration of primary pyrrhotite to magnetite would lead to magnetite acquiring a CRM as it forms, rather than inheriting a pre-existing signal. If primary pyrrhotite

can be magnetised in the solar nebula, the consistency of the magnetic directions in the PPI grains with the other (secondary) pyrrhotite and magnetite grains would suggest that magnetite cannot inherit a magnetisation from primary pyrrhotite.

B. Magnetite

Magnetite behaves differently between chondrules (random orientations) and matrix (unidirectional). Two possibilities emerge to explain the difference: the field which magnetised the matrix magnetite was not active when the magnetic grains in the chondrules acquired their signal, or the field was active but the magnetite in the chondrules did not record it.

Metal is one of the first phases to oxidise during aqueous alteration, as seen from the abundance of magnetite in the least altered CO chondrites (e.g., DOM 08006, Davidson et al. 2019), and from laboratory alteration experiments showing that magnetite forms early (Suttle et al., 2022). If the PPIs in the matrix are primary (Singerling & Brearley 2018), then pyrrhotite alteration to magnetite could happen at the same time as metal alteration, or slightly later (Suttle et al. 2021). However, if pyrrhotite alteration happened later, the nebular field it experienced would be weaker (Bai & Goodman 2009), implying a stronger field during metal alteration. Therefore, the magnetisation of the matrix (including PPIs) implies that a field was present while the metal in the chondrules was altered.

The second possible explanation is that the magnetite in the chondrules was unable to record the field. There is strong evidence that metal in chondrules is magnetised (Fu et al. 2014, Borlina et al. 2021), and since both the metal and magnetite are randomly oriented in the chondrules, it is likely that magnetite is able to inherit the magnetisation of the metal, which may prevent it from becoming magnetised by the local field.

C. Metal

Finally, the metal in the matrix is well clustered in a different direction than magnetite and pyrrhotite (Fig. 4.10). It appears to remain stable during demag-

netisation – although some metal grains randomise, leading to more scatter with increased AF field (Fig. C.1), the mean vectors for the metal stay in the same region (Fig. 4.10). The grains are scattered across the sample, ruling out reheating due to atmospheric passage, or a uniformly magnetised clast. The apparent high coercivity of these grains, together with the lack of spatial correlation with the reheated region next to the fusion crust suggests that the metal magnetisation is primary (i.e., not overprinted).

The magnetisation direction of the matrix metal differs from the direction of pyrrhotite and magnetite, which requires that the field which magnetised the matrix metal did not affect pyrrhotite, magnetite, or the chondrules. Two possibilities emerge: impact heating (Bland et al. 2014) prior to alteration, or an accretional detrital remanent magnetisation (ADRM; Fu & Weiss 2012). The possibilities are discussed below.

Impact heating Studies on impact heating and compaction have found that the matrix of a chondrite can be heated up to 1000 K due to low-velocity impacts, while chondrules act as heat sinks due to their much lower porosity (Bland et al. 2014, Muxworthy et al. 2017). Impacts are very common in the early solar system and they could plausibly create temperatures above or close to the high Curie temperature of kamacite (760°C) in the matrix of a chondrite, particularly close to a chondrule (Bland et al. 2014). Chondrules and their inclusions would be unaffected by the initial heating event, but they may thermally re-equilibrate with the matrix shortly after the impact (see below). To explain the difference in magnetic directions between metal and the minerals formed through alteration (magnetite and pyrrhotite), the impact heating would have to take place before the onset of aqueous alteration on the CM parent body, and the field would have to change direction relative to the parent body between the impact and the onset of alteration.

The shock stage of MCY 05231 has not yet been studied. The flattened chondrules could be evidence for shock (as observed in Murchison; Hanna et al. 2015), but

irregularly-shaped chondrules are common and they can also be a primary feature (Hanna & Ketcham 2018). Constraining shock stage was not an objective of this study, so even though no other shock indicators were immediately obvious (e.g., rock fabrics, veins, shocked olivines; Hanna et al. 2015), their presence cannot be ruled out. It is also possible that the alteration grade of MCY 05321 (more advanced than Murchison) and the early timing of the impact (pre-alteration) could have erased a rock fabric. On the other hand, thermal evolution modelling based on WIS 91600 was performed by Bryson et al. (2020b), and found that, despite not being heated by the initial shockwave, chondrules thermally equilibrate with the matrix within 2 seconds from the impact, such that the final temperature reached by the chondrules is only 35°C less than the peak matrix temperature. CM chondrites contain a slightly higher proportion of chondrules vs. matrix than WIS 91600 (15% in WIS 91600, Birjukov & Ulyanov 1996, vs. 20% in CM chondrites, on average; Weisberg et al. 2006), which increases the difference in temperature between chondrules and matrix. However, if the matrix is heated to 760°C (to remagnetise all/most of the metal), it is likely that the chondrules would be heated to a high enough temperature that some proportion would be remagnetised, producing a directional bias that is not observed.

Accretional Detrital Remanent Magnetisation Fu & Weiss (2012) hypothesised that ferromagnetic grains in the solar nebula can align with the nebular field, and that this alignment could be preserved during planetesimal formation. Assuming CM chondrite formation at 5 AU based on the likely location of the snow line (Cieza et al. 2016), and a background nebular field value of 10^{-5} T (Turner & Sano 2008), the size range of the grains expected to align with the field is 30 μm up to 6 mm (aspect ratio = 2) or 4 cm (aspect ratio = 1.1) in a turbulent nebula, and higher in a quiescent nebula. The metal grains whose moments were measured here are at the lower end of this range (30-60 μm), so ADRM is a plausible explanation for the metal magnetisation in the matrix.

However, the chondrules did not align with the field, and they were instead accreted on to the CM parent body in random orientations (Section 4.4.2.1). This could be due to their larger size, combined with smaller specific magnetic moments of chondrules, since they contain ferromagnetic inclusions within mafic minerals instead of being pure ferromagnetic particles like the metal grains. The time needed for a particle to align with the local field is proportional to the (negative) specific magnetic moment, and inversely proportional to particle radius, such that small, strongly magnetic particles would align with the local field much faster than large, weakly magnetic chondrules (Fu & Weiss 2012). The difference in the specific magnetic moment of a metal grain vs a chondrule could be up to 6 orders of magnitude ($10^{-5} \text{ Am}^2\text{kg}^{-1}$ for the chondrules in Allende measured by Carporzen et al. (2011), and $10 \text{ Am}^2\text{kg}^{-1}$ for pure Fe, Wang et al. 2010), so metal grains are much more likely to align with the nebular field. The size range above is calibrated for magnetic chondrules, indicating that the chondrules should nonetheless align with the field, so perhaps the reason for the random chondrule orientations is that the alignment of chondrules is more difficult to maintain during accretional collisions with other particles — in this case, the metal grains may have time to realign during accretion, but the chondrules might not. The total magnetic moment of the strongest chondrule measured here is $9.6 \times 10^{-11} \text{ Am}^2$ (point 18 in Table C.1), which is weaker than the most magnetic chondrules measured by both Fu et al. (2014) and Borlina et al. (2021). Their chondrules are randomly oriented, indicating that they were also unable to align and/or remain aligned with the nebular field during accretion, so it is not surprising that the chondrules measured here are not aligned either.

4.4.3 Atmospheric heating

The potential influence of atmospheric heating on the magnetic record is considered here. A fragment of fusion crust is present in the bottom right of the sample (Fig. 4.7), giving a very strong magnetic signal (Fig. 4.9 D). Since the fusion crust is continuous,

it could not be fitted to a dipole, so it is not possible to draw firm conclusions on the direction of the magnetisation. Nevertheless, a magnetic signal ~ 1.5 mm away from the fusion crust on the same map in Fig. 4.9D appears to be magnetised differently than the fusion crust. The signal is carried by metal with a Curie temperature of 760°C , which rules out heating above this temperature. The background signal is slightly elevated within 1 mm from the fusion crust, which could be due to reheating. For this reason, no dipoles within 1 mm of the crust were fitted and included in the analysis. Furthermore, the stereonets in Fig. 4.10 were replotted after removing all datapoints within 3 mm of the fusion crust on the entire thin section, by extrapolating the exposed fusion crust. This led to more scatter in the results since there are fewer datapoints, but no trends have changed.

I therefore conclude that heating from atmospheric passage was limited to 1 mm from the fusion crust, in agreement with previous studies (Weiss et al. 2000, 2002), so it is unlikely to have affected the data used in this study.

4.4.4 Limitations of the QDM

The spatial resolution of the QDM maps in the present setup is $5.2\text{ }\mu\text{m}$ (Section 4.2.3). This size exceeds the typical single domain (SD) and vortex (SV) size ranges of the magnetic carriers in MCY 05231 (Table 1.1) and as a result, the signals included in the net moment inversion analysis are fields generated by large grains or clusters of smaller grains (Section 4.3.1), not individual SD and SV grains. At the same time, Fig. 4.10 and 4.11 show that the magnetic signals included in the analysis often retain their magnetisation to 50 mT during AF demagnetisation, giving confidence that the signals are carried by high-coercivity grains. This is likely due to the presence of clusters of SD/SV grains in the measured signal, which is consistent with previous work showing that clusters of SD magnetite in zircons produce stable magnetic signals that can be measured using a QDM (Weiss et al. 2018, Tang et al. 2019, Fu et al. 2021b). Previous work also found that the magnetic signals from clusters of SD/SV

grains are sometimes faint and non-dipolar, which is consistent with the faint signals produced by the submicron magnetite grains in the matrix of MCY 05231 (Section 4.4.1). The high coercivity of the analysed signals (Fig. 4.10 and 4.11), combined with the similarity between the measured background signal (e.g., Fig. 4.8) and previously observed signals from SD magnetite suggest that the discussion in this chapter is unlikely to be negatively impacted by the spatial resolution of the QDM.

4.4.5 Magnetic history

The data presented here are consistent with three stages of magnetisation for the CM chondrites:

1. Chondrule magnetisation in the solar nebula
2. Metal magnetisation, either in the solar nebula as an ADRM, or through impact heating on the parent body
3. Formation of magnetite and pyrrhotite through aqueous alteration

These steps are detailed below and sketched in Fig. 4.14.

1. Chondrule magnetisation (Fig. 4.14.1) In line with previous studies, I suggest that the chondrules were magnetised in the solar nebula and that they accreted in random directions, such that their magnetisation directions appear random. The magnetic carrier is primary metal.

2. Isolated metal grain magnetisation (Fig. 4.14.2) The two possibilities for metal magnetisation are presented in Section 4.4.2.2. On the basis that impact heating would be expected to create a directional bias in the chondrules, it is more likely that the metal in the matrix preserves an ADRM (Fu & Weiss 2012). Although uncertainties remain as to why the chondrules did not also align with the nebula

field, it could be due to accretion on to the CM parent body disturbing the chondrule alignment more strongly than it disturbs the metal alignment. Regardless of the mechanism of magnetic acquisition, by the end of this stage, the rock contains randomly oriented chondrules, and unidirectional metal in the matrix.

3. Aqueous alteration (Fig. 4.14.3) Aqueous alteration leads to the formation of secondary pyrrhotite and magnetite. Magnetite likely forms via three mechanisms: pseudomorphic replacement of metal (likely inheriting the metal remanence), precipitation from Fe-rich solution (forming small matrix magnetite, which is expected to acquire a CRM during formation), and pseudomorphic replacement of primary pyrrhotite (expected to acquire a CRM during formation).

Of the three magnetite formation mechanisms above, magnetite formed through precipitation carries the most uncertainty. While the presence of submicron magnetite was identified based on the background signal on the QDM maps (Section 4.4.1) and FORC diagrams (Sridhar et al. 2021), there is a possibility that these small magnetite grains may also have formed through pseudomorphic replacement of finer metal grains. Fine metal grains were found in pristine CO chondrites (Davidson et al. 2019), so they could plausibly have been present in the unaltered precursor to CM chondrites. This is presently unconstrained due to the altered nature of CM chondrites, where the only metal grains left unaltered in the matrix are the coarse grains (20-50 μm range). Therefore, submicron magnetite being formed through pseudomorphic metal replacement is a possibility. However, I suggest that it is unlikely considering that the background signal is oriented approximately N-S on the maps (Fig. 4.8), which is consistent with the pyrrhotite and the coarse magnetite directions, but very different from the metal direction (Fig. 4.10). If submicron metal were present in the sample, it would be expected to carry the same signal as the larger metal grains, considering the likely primary origin of the metal magnetisation. The alignment of the submicron matrix magnetite with pyrrhotite instead of metal then suggests that it did not inherit

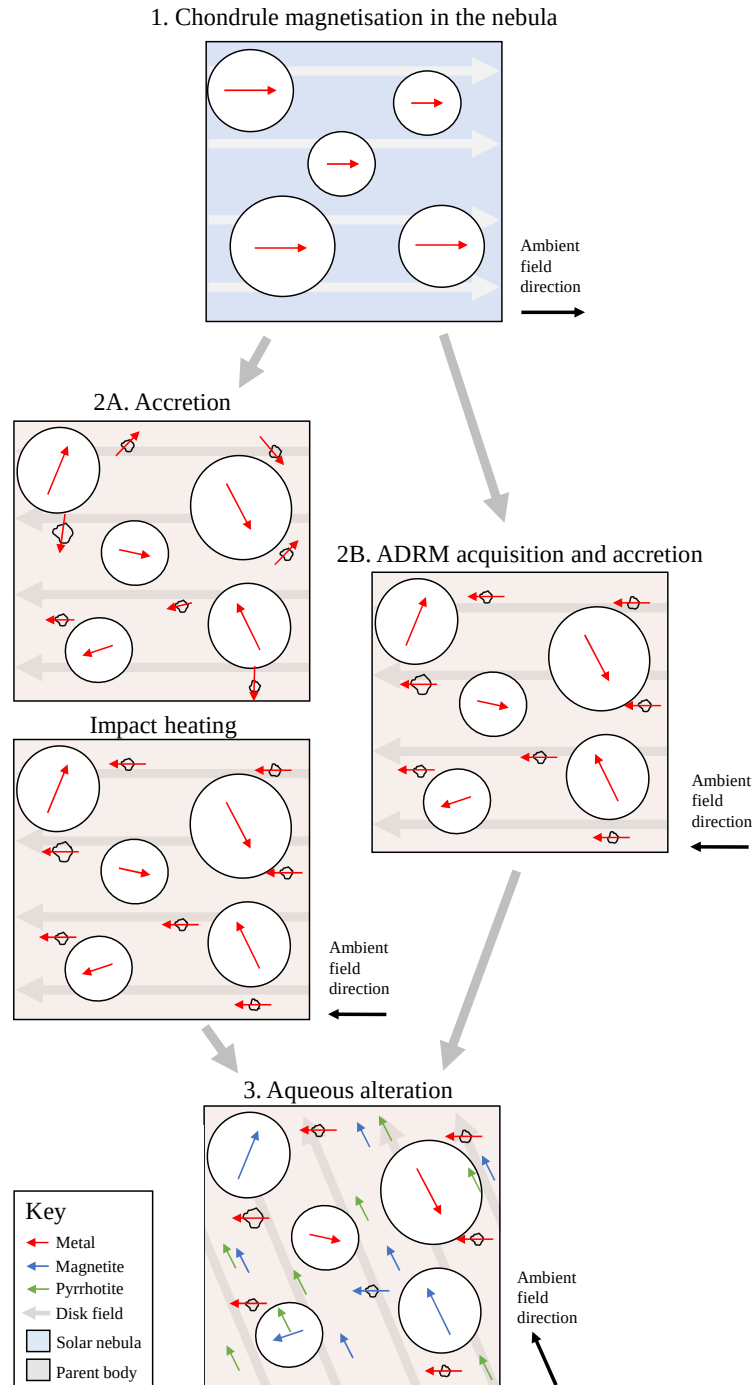


Figure 4.14: Schematic showing the proposed magnetisation sequence for MCY 05231. 1. Chondrules form in the solar nebula and their magnetic constituents are magnetised uniformly. When they are incorporated into a parent body, the uniform magnetisation becomes random on cm-scale (Borlina et al. 2022). 2A. Grains of metal in the matrix are not initially magnetised, and become magnetised by impact heating that raises the temperature of the matrix, without affecting the chondrules as much (Bland et al. 2014). 2B. Alternatively, the metal grains acquire an ADRM in the solar nebula (Fu & Weiss 2012), which is preserved during accretion. (*Continues on the next page*)

Figure 4.14: (*Continued*) At the end of stage 2, the matrix is carrying a unidirectional matrix signal, and a random chondrule signal. 3. Aqueous alteration leads to the formation of pyrrhotite and magnetite. The pyrrhotite forms from a non-magnetic precursor (troilite), so it acquires a CRM while it forms. The magnetite that forms through pseudomorphic replacement of metal inherits the signal from the metal (predominantly in the chondrules), whereas magnetite which forms through precipitation acquires a CRM (in the matrix). This leads to randomly magnetised chondrules and a uniformly magnetised matrix.

the magnetisation of the metal, which favours a precipitative origin that would allow it to record a CRM as it forms.

Since metal is more common in chondrules, pseudomorphic replacement of metal dominates the chondrule signal, whereas other formation mechanisms (replacement of pyrrhotite and precipitation) dominate the matrix signal. Examples of pseudomorphic replacement of metal to form magnetite have been observed in the matrix, and where they could be fitted and included on stereonet (in the outline examples detailed in Section 4.3.1 and 4.4.1), they plot in the same direction as the pyrrhotite, differently from the metal cluster. It is possible that pseudomorphic replacement of metal does not always inherit the remanence (the shape and size of the grains may play a role that requires further study), or alternatively, I note that the origin of these grains is uncertain, making them non-representative examples of pseudomorphic replacement of metal. Nevertheless, considering the unidirectionality of metal in the matrix, it is predicted that some magnetite grains will align with the metal rather than the pyrrhotite.

4.4.6 Implications for bulk magnetic studies of the solar nebula

The bulk magnetic signal of MCY 05231 is influenced by the bulk signal of each constituent phase: pyrrhotite (uniformly magnetised in the entire sample), magnetite (uniformly magnetised in the matrix, but random in chondrules), and metal (uniformly magnetised in the matrix, in a different direction than pyrrhotite and mag-

netite; random in chondrules).

Since all pyrrhotite in the sample is unidirectional, pyrrhotite is expected to dominate the bulk signal. At the same time, only some magnetite is uniformly magnetised, so the bulk signal due to magnetite will be a combination of matrix magnetite (most of which is unidirectional, with potential limited exceptions for the magnetite that forms via replacement of metal in the matrix) and the randomly magnetised magnetite in chondrules. The primary signal from matrix metal could further complicate bulk studies, although this metal forms a very small part of CM chondrites (1% in the least altered CM chondrite studied by Rubin et al. (2007), so expected to be $\ll 1\%$ in the moderately altered MCY 05231, and almost absent from highly altered CMs), so its contribution to the remanence is likely very small.

Only a portion of magnetite being uniformly magnetised is consistent with the bulk magnetic study published by Cournède et al. (2015), who found that the remanence of the bulk meteorite is dominated by pyrrhotite, despite the susceptibility being dominated by magnetite. In some meteorites (Banten and Nogoya), their thermoremanent curves show that some signal is still present after the demagnetisation of pyrrhotite at 200-300°C. They interpret this as due to alteration during laboratory heating, but it would also be consistent with only a portion of the magnetite in the sample carrying a HT signal.

Inheritance of magnetisation from metal to magnetite could have significant implications for studies that have found zero-field recordings (e.g., Gattacceca et al. 2016, Borlina et al. 2022). This is because the lack of a field recorded by CO and CV chondrites could be due to magnetite inheriting the (random) pre-accretionary remanence of metal, and thus being unable to record the field present during alteration, even if a field were present (Borlina et al. 2022). This applies only if all or most of the magnetite in those meteorites formed through pseudomorphic replacement of metal, which is not necessarily the case – if different alteration pathways form magnetite in CO and CV meteorites as they do in CMs, then a zero-field recording in magnetite

grains that are able to record a field may still indicate that no field was present. However, it may be difficult to resolve the signal of such magnetite grains from the overall signal, especially as pseudomorphic magnetite tends to be coarser, and it may dominate the bulk signal as a result.

Furthermore, palaeointensity methods which involve applying a laboratory field to a sample will magnetise the entire magnetic assemblage of the sample, whether or not all minerals carried an NRM originally (Bryson et al. 2023). If a mineral was not originally magnetised, palaeointensities obtained using these methods could be underestimated. If only a portion of a mineral is magnetised, for example because a mineral formed via different reactions and/or at different times, the palaeointensity could still be underestimated by an amount which depends on the portion of unmagnetised material (Bryson et al. 2023). This could affect palaeointensity estimates for CM chondrites (Cournède et al. 2015, Bryson et al. 2023), CV chondrites (Gattaccecchia et al. 2016) and CO chondrites (Borlina et al. 2022), with implications for the strength and evolution of the solar nebula field.

4.5 Conclusions

- Using optical and electron microscopy, I found that the magnetite in MCY 05231 likely formed via three mechanisms. The majority of magnetite formed through pseudomorphic replacement of metal, especially in chondrules. Magnetite also likely formed through pseudomorphic replacement of pyrrhotite in PPIs, which were found exclusively in the matrix. Based on a weakly magnetic background signal that demagnetises by 25 mT in the matrix, and on FORC diagrams (Sridhar et al. 2021), I also inferred the presence of submicron magnetite grains, likely formed through precipitation from Fe-rich solution in the matrix. Therefore, pseudomorphic magnetite dominates the chondrule signal, while magnetite formed via different pathways dominate the matrix signal.

- Quantitative moment analysis of the QDM magnetic signals suggests that the signals produced by metal and magnetite in chondrules are randomly oriented, although uniformly magnetised within each chondrule. By contrast, the matrix carries two uniform magnetisations – pyrrhotite and magnetite are uniformly magnetised in a direction approximately orthogonal to the direction of the matrix metal. In addition, submicron grains of pyrrhotite and magnetite in the matrix produce a weak background signal oriented similarly to the coarser pyrrhotite and magnetite in the matrix.
- Based on the random orientations of the magnetic signals carried by the chondrules, together with the uniform magnetisation within at least one chondrule, I suggest that the chondrules carry a preaccretionary remanence. Since the signals are carried by metal and magnetite formed via replacement of metal, it is likely that magnetite that forms via pseudomorphic replacement of metal inherits the magnetisation of the metal, regardless of the field present during the alteration.
- The uniform magnetisation of the pyrrhotite and magnetite present in the matrix suggests that they both recorded the field that was active during their formation through aqueous alteration, likely as a CRM. Both the magnetite grains formed through replacement of primary pyrrhotite and those formed through precipitation from Fe-rich solution become magnetised by the local field. Due to its high Fe content, it is unlikely that primary pyrrhotite was magnetic, so there is no magnetisation to inherit and the newly formed magnetite becomes magnetised by the local field. In the case of magnetite formed through precipitation, there is no extant grain from which a remanence could be inherited, so magnetite records the field present when it grows through its blocking volume, as hypothesised for magnetite forming as framboids and plaquettes (Bryson et al. 2023).

- Inheritance of magnetic remanence from metal to magnetite could affect previous palaeointensity measurements in chondrites, as a low palaeointensity could be caused by magnetite not being able to record a field (as it instead inherits the random, pre-accretionary remanence of metal), rather than no field being present. This could lead to the palaeointensity being underestimated by an extent which depends on the amount of magnetite that forms via replacement of metal as a fraction of total magnetite in the sample (Bryson et al. 2023). Therefore, samples where magnetite forms via different pathways (e.g., samples rich in framboids and plaquettes such as WIS 91600 and Winchcombe; Bryson et al. 2020b, 2023) would be less affected than samples where all or most magnetite forms via pseudomorphic replacement of metal (e.g., CO chondrites; Borlina et al. 2022). Future work could be directed at understanding the conditions under which magnetite can inherit a remanence from metal, as well as further investigating reactions that form magnetite and their effects on the overall mineral assemblage – this could aid early identification of samples which can be used reliably for palaeomagnetism, if the same conditions that form precipitative magnetite also form other minerals or textures that are more readily identifiable.

Chapter 5

Summary & Future work

5.1 Summary

In this thesis, I presented a suite of magnetic and geochemical data to advance our understanding of secondary processes affecting planetesimals in the early solar system. I focused primarily on two types of processes (impacts and aqueous alteration) by studying stony-iron and chondritic meteorites. I also discussed the credibility of palaeomagnetic studies of meteorites and suggested directions for future work.

In **Chapter 2**, I used bulk palaeomagnetic techniques combined with electron microscopy to study the mesosiderite Estherville. The palaeomagnetic measurements (bulk NRM measurements, AF demagnetisation, IRM acquisition and ARM fidelity experiments) revealed that Estherville does not record a bulk high-coercivity (HC) magnetic signal, and that it does not have the recording fidelity to record a field lower than 150 μm . Some high-coercivity material (coercivity range 200 - 600 mT) was detected in Estherville, and based on microscopy and the palaeomagnetic results, I suggested that the high-coercivity grains are nanometric taenite inclusions in a pyroxene crystal. I also used the palaeomagnetic results to place constraints on the formation of mesosiderites. Mesosiderites are able to record strong fields based on their fidelity, and they are likely to have experienced such fields due to their slow cooling rate and inferred burial at depth in the parent body. The fact that they did not experience them could be due to several reasons: parent body properties

(the parent body could not generate a strong field), mesosiderite location in the parent body (not close enough to the core to experience a strong field), or due to the mesosiderite-forming impact. If the impact was destructive, it could have scrambled the core and shut off convection and dynamo generation. Furthermore, the impact likely happened during a period of magnetic quiescence in the solar system, when no palaeointensity could be recovered from any meteorite due to a proposed gap between thermal and compositional convection. If the remanence acquisition period was during this gap, that could also explain why mesosiderites did not record a field.

Since mesosiderites are known to be abundant in tetrataenite, I also used different methods to identify it. I found that electron microscopy methods are not very effective, because of the compositional overlap with taenite and the small tetragonal distortion occurring during ordering. At the same time, magnetic methods can readily identify single-domain tetrataenite because of its high coercivity.

Finally, since low-fidelity samples are not well suited to analysis using traditional palaeomagnetic techniques, I also used EBSD to study the crystallography of the cloudy zone islands, to test the hypothesis that cloudy zone formation in a field causes a bias in the c-axis orientation of tetrataenite in the cloudy zone. I found that all cloudy zone islands have the same crystallographic orientation, and suggested that this is due to the slow cooling rate of mesosiderites (much slower than the iron meteorite where this phenomenon was observed) which causes all cloudy zones islands to be connected, making it energetically difficult to align with a field.

In **Chapter 3**, I investigated the formation of Main Group pallasites. I tested the hypothesis that two distinct metal textures (matrix metal and metal pockets trapped in olivines) contain metal of different origins, by measuring their respective compositions using LA-ICP-MS on the pallasite Seymchan. I found no systematic compositional difference between the matrix and pocket metal in Seymchan, or among different pockets, which suggests that all metal in Seymchan has the same origin. I also measured a IIIAB iron meteorite (Henbury) and found compositional similarities

to Seymchan, lending further evidence to the hypothesised genetic link between Main Group pallasites and IIIAB irons. To reconcile chemical and textural observations from this study and previous work, I suggested the following formation model for pallasites:

1. A differentiated body (likely the IIIAB parent body) impacts the pallasite parent body and inserts core metal into the pallasite mantle;
2. The metal and olivines equilibrate texturally, trapping metal in pockets within the olivines;
3. A second impact fractures the olivines, leading to unequilibrated secondary features;
4. Burial following the impact leads to variable degrees of annealing in different pallasites;
5. One or more recent impacts create loose pallasites, some of which reach the Earth as meteorites.

I also discussed the possibility of primary metal being mixed in with the impactor's metal, and suggested that this is unlikely. Finally, I discussed the timescales involved in this model and proposed a mechanism that allows textural equilibration of the pockets to occur before gravitational separation of olivines and metal.

In **Chapter 4**, I turned my attention to aqueous alteration and its effects on the magnetic mineralogy of CM chondrites. I used QDM magnetic mapping to spatially resolve the magnetic signals of the CM chondrite MCY 05231, as well as microscopy to identify the source of each signal. I found two uniform magnetisations within the matrix, one carried by pyrrhotite and magnetite, and another carried by primary metal. I hypothesised that the magnetisation carried by pyrrhotite and magnetite was acquired when these minerals formed during aqueous alteration, while the metal could carry a primary magnetisation (potentially an Accretional Detrital Remanent

Magnetisation) or an early TRM due to impact heating. At the same time, the chondrules are randomly magnetised. Based on unidirectionality of signals in each chondrule, as well as chondrule magnetisations often being blocked up to 50 mT, I suggest that each chondrule is uniformly magnetised. However, the chondrules appear random due to being incorporated in the CM parent body in random orientations.

I used these results combined with petrological observations to suggest that the magnetite in MCY 05231 forms via three different pathways: pseudomorphic replacement of metal, pseudomorphic replacement of primary pyrrhotite, and precipitation from Fe-rich solution. I found that magnetite formed through replacement of metal is common in the chondrules, and since it is not aligned with the pyrrhotite in the matrix (instead being randomly oriented like the chondrule metal), it is likely to have inherited its magnetisation from the metal. On the other hand, magnetite formed from primary pyrrhotite and magnetite precipitated from solution are uniformly magnetised in the matrix, suggesting that they were magnetised during alteration instead of inheriting a remanence from a precursor. I used these results as evidence for the previously proposed possibility that pseudomorphic magnetite may inherit the remanence of the metal in CM chondrites (Borlina et al. 2022, Bryson et al. 2023), confirming that the different pathways of magnetite formation can influence magnetic record. Finally, I discussed implications for bulk magnetic studies of the solar nebula magnetic field.

5.2 Future work

5.2.1 Stony-iron meteorites

In Chapter 2, I suggested that there are high-coercivity taenite inclusions trapped in pyroxene in the silicate portion of mesosiderites. Future work could study these inclusions using *in situ* magnetic imaging (QDM or SQUID microscopy) to determine if they are uniformly magnetised, since the silicate part of mesosiderites may carry a pre-impact remanence. The nature of this remanence and whether it is partially or

completely overprinted could then be used to learn about the metal-silicate mixing impact in mesosiderites. Alternatively, if the remanence is not affected by the impact, the inclusions may preserve evidence of early, thermally-generated dynamo on the mesosiderite parent body, which could be used to place further constraints on its physical properties (e.g., parent body radius, volatile content).

In Chapter 3, I presented new chemical data on the metal in pallasites and reconciled these data with textural observations from previous studies. In agreement with previous work, I found both texturally equilibrated and unequilibrated features in Seymchan. The unequilibrated features may be partially re-equilibrated by annealing in deeper meteorites (Walte & Golabek 2022), but it is currently unclear how this correlates with cooling rate, due to the diversity of textures and grain sizes found in pallasites. An opportunity for future research is to analyse the extent of chemical equilibration between metal and olivines, and the connection between textural equilibrium, chemical equilibrium and cooling rate. Chemical zonation in pallasite olivines is a well documented feature in some elements (Leitch et al. 1979, Tomiyama & Huss 2005, Boesenberg et al. 2012), and it is generally attributed to diffusion during cooling. The presence of zonation suggests that the olivines are not in chemical equilibrium with the metal (Tomiyama & Huss 2005), but there is evidence for partial re-equilibration after the metal-silicate mixing event (McKibbin et al. 2013, Donohue et al. 2018), affecting some elements more than others (McKibbin et al. 2013). The degree of rounding in olivines has been found to influence the length scales of the concentration profiles, with more rounding leading to wider profiles (Tomiyama & Huss 2005). It would be informative to extend these studies to pallasites with different cooling rates and olivine textures (angular and fragmental), to fully understand the origin of different types of pallasites.

Furthermore, while the metal pockets have been shown to be in textural equilibrium with the olivines (e.g., Walte et al. 2020), the extent of chemical equilibrium remains an open question. It would be valuable to investigate the chemical relation-

ship between the metal and olivines adjacent to secondary, non-texturally-equilibrated features such as fractures and veinlets, and to compare the results to pallasites with slower cooling rates, in which the same features were annealed to various extents. The relationship between olivine and metal next to non-texturally-equilibrated secondary features could act as a baseline for the amount of diffusion that would take place in faster-cooled pallasites such as Seymchan, where little to no textural equilibration had time to occur. This could be used to determine the extent to which the slower-cooled, more annealed versions of the same features (and, ultimately, the metal pockets in full textural equilibrium) equilibrated chemically, which would allow a full evaluation of the relationship between textural and chemical equilibrium over a range of different cooling rates.

Given the complex nature of pallasites, a joint chemical and textural study is required to fully explain their formation, and understanding the chemical relationship between olivines and metal could be a key step to achieve this.

5.2.2 Chondrites

One of the main findings in Chapter 4 was that magnetite formed through different mechanisms records different directions of magnetisation. The presence of submicron precipitative magnetite was inferred based on QDM maps and previous FORC studies, but this interpretation is limited by the resolution of the microscopy methods used here. Higher-resolution microscopy such as TEM would be able to provide more certainty on the origin of these grains, and help us fully understand their origin. While framboids and plaquettes (both forms of precipitative magnetite) have been studied increasingly more in recent years, small precipitative grains isolated in the matrix (as identified by Render et al. 2022, for example) are insufficiently understood, and they could represent a significant portion of the magnetite population in CM chondrites. Furthermore, the chemical conditions that facilitate magnetite precipitation could also lead to the formation of other minerals or features. If they are more readily identifiable

than submicron magnetite, they could be used as proxies for precipitative magnetite and aid early identification of samples that can be used reliably for palaeomagnetism.

Secondly, I also provided evidence for inheritance of remanence between metal and magnetite forming through pseudomorphic replacement of metal. Micromagnetic modelling could be used to understand the conditions that facilitate inheritance from metal to magnetite. A preliminary micromagnetic modelling study was performed by Kirby (2023), who found that inheritance is influenced by the size and aspect ratio of the metal, with elongated grains being able to inherit the metal signal over a wider size range than round grains. Future work could extend these simulations to larger sizes and more diverse shapes, to obtain a complete picture of the requirements of magnetic inheritance from metal to magnetite.

Finally, the investigation into aqueous alteration and impacts in this thesis could be complemented by studying meteorites which show evidence of both processes. The CM chondrite groups contains meteorites with ample evidence for impact heating (e.g., Quirico et al. 2018), both before and after alteration. The interplay between these processes is a topic of intense research (e.g., Lee et al. 2016, King et al. 2019), and palaeomagnetic investigation could help to place constraints on the complicated parent body reprocessing experienced by these meteorites. For example, a meteorite heated after alteration (e.g., Jbilet Winselwan; King et al. 2019) may preserve a partial thermal overprint which could help constrain the peak metamorphic temperature achieved through impact heating. Coupled with modelling, this approach could provide information on the strength of the impact, and help to reconstruct the characteristics of the meteorite before the impact.

Appendix A

Pallasite supplementary data

A.1 Sulfide composition

The composition of troilite (FeS) found in veinlets and next to kamacite is given in Table A.1. The troilite in veinlets is highly altered, as evidenced by the rim of oxides surrounding the troilite (Fig. A.1), which may have caused deviations from the ideal FeS composition.

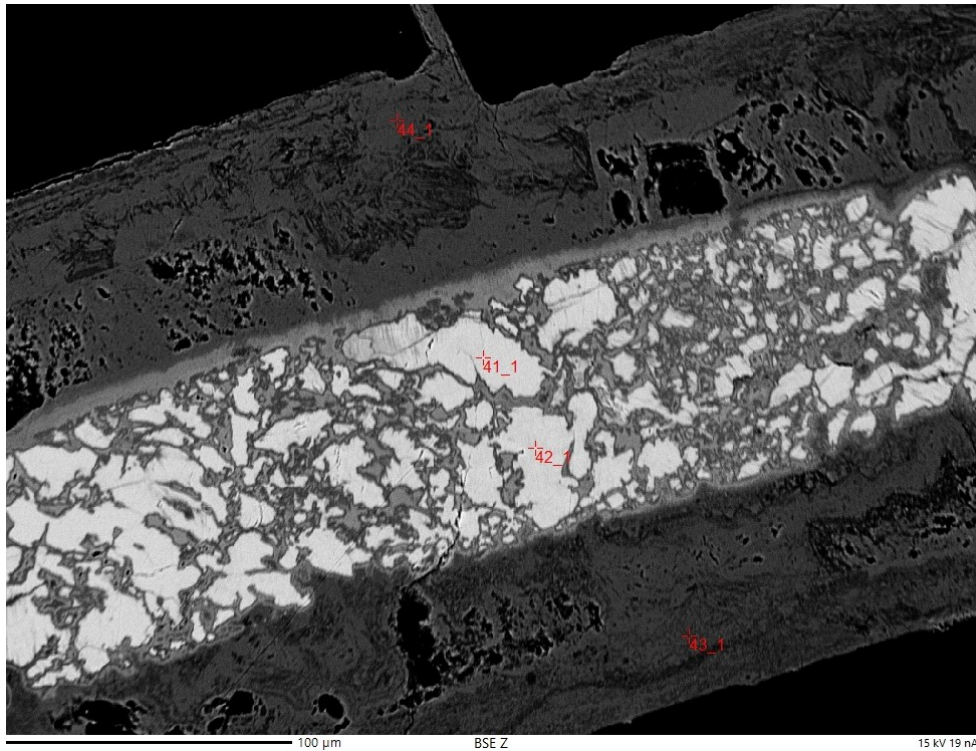


Figure A.1: BSE image of a highly altered veinlet in Seymchan, composed of troilite surrounded by a rim of oxides.

	Mean wt%	σ	Mean at%	σ	n
Fe	63.12	1.83	49.68	1.30	13
S	36.30	0.28	49.77	0.44	13
Cr	0.11	0.02	0.09	0.01	13
Co	0.16	0.12	0.12	0.09	13
Ni	0.64	1.31	0.48	0.99	8
Total	100.33		100.14		

Table A.1: EPMA data of sulfide composition, measured in veinlets or in association with kamacite. The composition is consistent with troilite (FeS). n represents the sample size, and it is lower for the Ni measurements because some measurements were below detection limit (approx. 650 ppm), and thus excluded from the averaging. The total Ni content of the sulfides may therefore be overestimated. σ = standard deviation; wt% = weight %; at% = atomic %.

A.2 Individual pocket compositions

Phase	Pocket number		Fe (%)	Ni (%)	Co (%)	Cu (ppm)	Ga (ppm)	Ge (ppm)	W (ppm)	Os (ppm)	Ir (ppm)	Au (ppm)
Kamacite	1	Mean	92.89	6.76	0.63	83.35	27.73	79.30	0.42	0.84	0.80	1.66
		σ	0.35	0.15	0.01	7.73	2.83	3.32	0.28	0.37	0.20	0.19
	2	Mean	92.19	7.01	0.59	81.48	26.23	79.20	0.32	0.58	0.64	1.59
		σ	0.20	0.13	0.02	4.64	2.47	3.05	0.11	0.31	0.21	0.24
	3	Mean	92.22	6.93	0.58	77.08	25.07	74.29	0.64	0.95	0.69	1.56
		σ	1.00	0.31	0.02	6.29	1.75	2.98	0.47	0.42	0.36	0.21
	4	Mean	92.16	6.86	0.54	69.48	26.60	75.14	0.51	0.67	0.68	1.71
		σ	0.43	0.42	0.02	6.65	2.37	4.27	0.06	0.16	0.24	0.18
	5	Mean	92.55	7.11	0.57	85.39	27.64	77.73	0.66	1.13	0.48	2.01
		σ	0.34	0.11	0.02	7.48	1.87	4.17	0.25	0.30	0.14	0.24
	6	Mean	92.79	6.71	0.52	75.19	27.07	78.30	0.54	0.70	0.62	2.18
		σ	0.36	0.31	0.02	5.39	2.72	3.12	0.16	0.28	0.18	0.22
	7	Mean	92.30	7.22	0.57	82.17	28.05	77.82	0.79	0.43	0.62	2.06
		σ	0.15	0.07	0.02	7.00	3.13	1.72	-	-	0.17	0.09
	8	Mean	92.15	6.97	0.52	73.75	25.86	82.23	0.45	0.86	0.64	1.52
		σ	0.33	0.18	0.02	5.67	2.68	4.51	0.18	0.42	0.19	0.13
	9	Mean	92.07	7.01	0.57	85.66	27.75	81.83	0.47	0.95	0.59	1.73
		σ	0.21	0.08	0.02	7.36	1.60	2.60	0.11	0.13	0.19	0.14
	10	Mean	92.08	7.01	0.60	91.58	24.59	79.50	0.55	0.33	0.50	1.58
		σ	0.17	0.16	0.005	4.05	0.96	4.27	-	0.13	0.12	0.09
	11	Mean	92.18	6.96	0.54	73.69	27.51	80.84	0.53	0.42	0.31	1.76
		σ	0.34	0.17	0.01	4.56	2.80	2.82	0.09	-	0.10	0.18
Taenite	2	Mean	83.55	15.90	0.37	255.1	27.47	60.03	0.45	1.15	0.82	2.61
		σ	1.35	1.58	0.01	29.32	2.81	3.25	0.22	0.86	0.40	0.41
	4	Mean	83.87	15.69	0.37	259	25.45	59.22	0.62	0.62	0.74	2.52
		σ	1.37	1.27	0.02	42.36	2.41	3.27	0.19	0.15	0.38	0.49
	5	Mean	82.86	17.17	0.36	317.8	29.23	62.54	0.47	0.76	0.52	3.36
		σ	2.63	2.54	0.02	61.12	4.00	2.97	0.28	0.09	0.15	0.66
	6	Mean	85.60	14.34	0.38	208.4	26.03	58.34	0.57	1.31	0.88	2.77
		σ	1.42	1.52	0.02	14.49	2.74	3.20	0.11	0.53	0.24	0.22
	7	Mean	82.27	17.63	0.34	289.2	28.03	61.88	0.63	0.97	0.64	3.31
		σ	2.08	2.00	0.02	44.27	2.64	5.30	0.09	0.29	0.18	0.50
	8	Mean	84.48	14.89	0.38	239.2	25.78	64.95	0.57	0.98	0.70	2.35
		σ	1.98	2.23	0.02	40.55	3.63	9.00	0.21	0.39	0.29	0.86
	9	Mean	85.43	14.35	0.38	243.4	25.07	56.54	0.50	0.62	0.64	2.24
		σ	0.74	0.75	0.01	15.12	2.32	1.67	0.21	0.54	0.22	0.19
	11	Mean	83.59	16.04	0.36	272.8	25.02	62.41	0.42	0.68	0.35	2.53
		σ	2.60	2.82	0.03	70.64	3.19	5.91	0.12	0.36	0.15	0.49

Table A.2: Mean and standard deviations for the concentrations of Fe, Ni, Co (measured using EMPA) and Cu, Ga, Ge, W, Os, Ir, Au (measured using LA-ICP-MS) in each pocket, separated into kamacite and taenite. For the pockets where no σ is given for W and Os, the mean is based on only one measurement due to the other measurements being below detection limit.

Appendix B

Statistical methods

B.1 Cluster analysis criteria

B.1.1 Silhouette statistics

Silhouette statistics (Rousseeuw 1987) are a method of evaluation of cluster analysis configurations, comparing how similar a datapoint is to other points within its own cluster (cohesion), and how different it is from the points in the neighboring cluster (separation).

Cohesion is measured as the mean distance between a point i and all other points in the same cluster, and it is given by $a(i)$ in Equation B.1:

$$a(i) = \frac{1}{|C_I| - 1} \times \sum_{j \in C_I} d(i, j) \quad (\text{B.1})$$

where C_I is the number of datapoints in the cluster I , and $d(i, j)$ is the distance between datapoints i and j in cluster I . i must be different to j , hence normalising by $C_I - 1$.

Separation is given by $b(i)$, the mean distance between i (in cluster I) and all points in the neighboring cluster, J (Equation B.2). The minimum value of this distance is taken to indicate the neighboring cluster.

$$b(i) = \min\left(\frac{1}{|C_J|} \times \sum_{j \in C_J} d(i, j)\right) \quad (\text{B.2})$$

where C_J is the number of datapoints in the cluster J , and $d(i, j)$ is the distance between point i in cluster I and point j in cluster J .

The silhouette value $s(i)$ can then be computed as:

$$s(i) = \frac{a(i) - b(i)}{\max(a(i), b(i))} \quad (\text{B.3})$$

where $a(i)$ and $b(i)$ are defined in Equations B.1 and B.2.

The optimal number of clusters is the number that corresponds to the highest arithmetic mean of all silhouette values in a clustering configuration.

B.1.2 Gap statistics

Gap statistics (Tibshirani et al. 2001) are a different method to evaluate cluster analysis results. They compare the within-cluster variance W_k of the data to the variance that would be expected in reference data that does not contain any clustering. W_k is given by:

$$W_k = \sum_{k=1}^K \frac{1}{2n_k} D_k \quad (\text{B.4})$$

where K is the total number of clusters, n_k is the number of points in cluster C_k , and D_k is the intra-cluster distance between points in a given cluster. D_k is defined as:

$$D_k = \sum_{x_i \in C_k} \sum_{x_j \in C_k} \|x_i - x_j\|^2 \quad (\text{B.5})$$

where x_i and x_j are both points in the same cluster C_k .

The gap value Gap_n is calculated by subtracting $\log W_k$ from the variance of the reference datasets. The reference datasets are obtained by sampling uniformly from the space occupied by the real data, using B Monte Carlo simulations and averaging them, thus obtaining the estimate of the reference variance, $E_n^*\{\log W_k\}$. This is summarised by the gap statistic formula:

$$Gap_n(k) = E_n^*\{\log W_k\} - \log W_k \quad (\text{B.6})$$

In this study, $B = 100$ Monte Carlo simulations were used. The error associated with averaging them is:

$$s'_{k+1} = \sqrt{1 + \frac{1}{B}} sd(k) \quad (\text{B.7})$$

where $sd(k)$ is the standard deviation of the Monte Carlo simulations, and $\sqrt{1 + \frac{1}{B}}$ accounts for simulation error.

The optimal number of clusters is the smallest k such that $Gap(k) \geq Gap(k + 1) - s'_{k+1}$, representing the greatest change between the real variance and the reference variance (within error) between k and $k + 1$ clusters. This method allows the evaluation of different clustering configurations for $k > 1$, and unlike silhouette statistics, it can also determine that there is no clustering ($k = 1$).

Appendix C

Dipole fitting data

C.1 NRM

Data-point	M (Am ²)	Inc (°)	Dec (°)	D	Component (C/M)	Phase	Reason for rejection (if applicable)
1	7.9E-12	49.10	315.30	0.77	M	Pyr	
2	5.8E-11	16.37	329.00	0.57	?	?	
3	1.7E-12	-15.16	295.93	0.75	M	Pyr (mostly)	
4	3.2E-12	1.21	341.85	0.77	M	Mgt	
5	1.1E-12	17.13	270.53	0.46	M	Pyr	D < 0.5
6	3.8E-12	-22.60	256.95	0.40	C	Mgt	D < 0.5
7	2.2E-12	11.20	279.50	0.60	C	Mgt	
8	1.3E-11	-8.90	8.95	0.73	C	Metal + mgt	
9	3.5E-12	-35.39	52.20	0.53	M	All	Needs quadrupole fit
10	6.0E-13	-5.28	169.28	0.39	C	N/A	Needs quadrupole fit
11	2.1E-12	-31.64	237.22	0.85	M	Metal + mgt	
12	9.7E-12	19.81	344.70	0.52	M	Mgt	
13	4.3E-12	9.51	9.43	0.53	C	Pyr	
14	3.8E-12	17.78	207.58	0.39	M	Pyr	D < 0.5
16	2.8E-11	9.09	147.06	0.66	C	Metal + mgt	Noise
17	2.2E-12	-1.02	280.46	0.69	M	Mgt (mostly)	

18	9.6E-11	41.08	133.98	0.70	C	Pyr (mostly)	D < 0.5
19	3.2E-12	15.73	11.52	0.66	M	Mgt (mostly)	
20	1.5E-12	44.25	126.69	0.64	C	Mgt + metal	
21	1.0E-12	1.66	295.67	0.52	M	N/A	
22	4.9E-12	-27.48	112.51	0.65	C	Metal	
23	4.2E-12	13.86	222.52	0.28	C	Metal + mgt	
24	1.1E-13	-15.31	321.82	0.52	M	Pyr (mostly)	
25	2.3E-13	-15.72	115.69	0.62	C	Metal	
26	1.2E-11	25.74	340.02	0.54	M	Pyr	
27	2.5E-12	0.00	202.09	0.63	C	Mgt (mostly)	
28	3.0E-11	1.94	276.56	0.76	C	Mgt (mostly)	Needs quadrupole fit
29	3.7E-12	7.44	260.60	0.52	C	Mgt (mostly)	
30	8.8E-12	-0.42	277.18	0.68	C	Mgt (mostly)	
31	2.2E-12	18.89	336.26	0.44	?	Mgt	
32	1.1E-11	20.86	298.92	0.69	C	Pyr	Noise
33	7.6E-11	11.90	250.28	0.51	C	Mgt (mostly)	
34	1.6E-12	22.83	342.46	0.39	M	Pyr	
35	1.4E-12	-43.14	1.11	0.66	M	Pyr	
36	2.4E-12	7.53	259.64	0.75	M	Metal + mgt	Noise
37	1.5E-11	18.12	231.57	0.70	C	Mgt (mostly)	
38	1.5E-11	34.60	158.18	0.56	M	Metal/all	
39	1.4E-11	7.56	204.31	0.76	C	Metal + mgt	
40	9.2E-12	-31.59	193.59	0.83	C	Mgt	Strong background
41	4.7E-12	-15.42	252.03	0.65	C	Metal	
42	2.4E-12	60.48	337.83	0.73	M	Pyr	
43	1.4E-11	-40.04	-0.92	0.39	C	Pyr	
44	9.0E-12	0.30	252.39	0.83	M	Metal	
45	1.8E-11	28.20	349.15	0.72	M	Pyr	
46	1.6E-12	10.75	252.93	0.62	M	Pyr	

48	1.6E-12	2.36	296.72	0.41	C	Metal	D < 0.5
49	4.3E-12	-1.50	234.74	0.67	M	Metal	
50	8.7E-12	-22.32	238.52	0.71	M	Metal	
51	1.6E-12	-72.19	154.16	0.51	C	Pyr	
52	7.0E-13	-22.11	264.61	0.62	C	Metal	
53	7.5E-12	5.30	273.56	0.69	C	Metal	

Table C.1: Dipole fits for the NRM. N/A denotes a signal generated by a grain below the surface of the thin section. C = chondrule, M = matrix, Mgt = magnetite, Pyr = pyrrhotite, D = dipolarity parameter.

C.2 25 mT

Data-point	M (Am ²)	Inc (°)	Dec (°)	D	Component (C/M)	Phase	Reason for rejection (if applicable)
1	1.4E-12	32.73	336.19	0.33	M	Pyr	Needs quadrupole fit
2	3.4E-11	-18.49	293.69	0.52	?	?	
3	9.6E-13	-38.24	323.23	0.61	M	Pyr (mostly)	
4	9.9E-13	13.20	19.34	0.62	M	Mgt	
5	6.6E-13	5.03	271.52	0.58	M	Pyr	
6	7.6E-13	-39.72	350.47	0.68	C	Mgt	
7	9.4E-13	3.20	54.32	0.62	C	Mgt	
8	2.3E-12	39.09	8.23	0.79	C	Metal + mgt	
9	9.9E-13	20.13	44.71	0.59	M	All	
10	2.6E-13	14.92	115.63	0.36	C	N/A	D < 0.5
11	1.7E-12	-62.74	256.00	0.86	M	Metal + mgt	
12	3.4E-12	65.00	19.51	0.76	M	Mgt	
13	2.2E-12	84.56	13.03	0.71	C	Pyr	
14	9.6E-13	13.58	241.78	0.64	M	Pyr	
16	1.1E-11	6.30	133.40	0.85	C	Metal + mgt	
17	1.3E-12	7.53	305.55	0.75	M	Mgt (mostly)	
18	2.4E-11	32.84	73.54	0.63	C	Pyr (mostly)	
19	2.5E-12	18.18	27.37	0.69	M	Mgt (mostly)	
20	5.2E-13	64.57	17.47	0.19	C	Mgt + metal	Needs quadrupole fit
21	2.9E-12	-38.19	296.53	0.61	M	N/A	Needs quadrupole fit
22	6.7E-12	-17.31	122.37	0.71	C	Metal	Needs quadrupole fit
23	1.5E-12	-8.54	121.74	0.24	C	Metal + mgt	

24	7.4E-14	-35.09	317.29	0.56	M	Pyr (mostly)	Poor fit
25	1.6E-13	-27.09	132.17	0.66	C	Metal	
26	4.2E-12	53.31	307.49	0.58	M	Pyr	
27	1.6E-12	-31.41	145.64	0.58	C	Mgt (mostly)	
28	1.3E-11	-31.38	253.00	0.79	C	Mgt (mostly)	
29	2.4E-12	-14.20	279.59	0.59	C	Mgt (mostly)	Too faint
30	2.0E-12	-6.43	259.33	0.57	C	Mgt (mostly)	
31	2.0E-13	47.71	218.30	0.31	?	Mgt	
32	1.1E-12	22.01	88.95	0.35	C	Pyr	
33	4.5E-12	-32.58	206.81	0.48	C	Mgt (mostly)	
34	3.4E-12	19.12	238.16	0.68	M	Pyr	Too faint
35	7.1E-13	-27.14	57.48	0.41	M	Pyr	
36	1.1E-12	-8.91	182.84	0.72	M	Metal + mgt	
37	1.1E-12	-6.37	50.17	0.47	C	Mgt (mostly)	
38	1.1E-11	29.94	121.07	0.65	M	Metal/all	
39	6.9E-12	-18.64	183.29	0.53	C	Metal + mgt	Needs quadrupole fit
40	5.5E-12	-40.54	222.38	0.76	C	Mgt	
41	3.2E-12	28.34	51.35	0.57	C	Metal	
42	1.6E-12	29.95	139.19	0.58	M	Pyr	
43	4.1E-12	-6.65	15.33	0.41	C	Pyr	
44	6.2E-12	-15.27	245.72	0.79	M	Metal	Too faint Needs quadrupole fit
45	9.4E-12	23.36	-0.16	0.63	M	Pyr	
46	4.1E-12	3.78	323.58	0.53	M	Pyr	
48	3.2E-12	-3.58	20.58	0.60	C	Metal	
49	3.5E-12	-16.18	241.87	0.73	M	Metal	
50	6.0E-12	-13.83	246.45	0.68	M	Metal	Too faint Needs quadrupole fit
51	2.2E-12	-17.59	185.69	0.43	C	Pyr	
52	3.1E-13	-0.79	240.60	0.48	C	Metal	
53	1.8E-12	-5.88	255.65	0.68	C	Metal	

Table C.2: Dipole fits for the 25 mT AF step. N/A denotes a signal generated by a grain below the surface of the thin section. C = chondrule, M = matrix, Mgt = magnetite, Pyr = pyrrhotite, D = dipolarity parameter.

C.3 50 mT

Data-point	M (Am ²)	Inc (°)	Dec (°)	D	Component (C/M)	Phase	Reason for rejection (if applicable)
1	3.4E-13	-36.99	92.78	0.29	M	Pyr	D < 0.5
2	1.9E-11	-33.46	222.96	0.57	?	?	
3	7.3E-13	0.52	129.31	0.24	M	Pyr (mostly)	D < 0.5
4	3.3E-13	23.39	73.13	0.39	M	Mgt	
5	7.6E-13	-39.24	345.46	0.68	M	Pyr	D < 0.5
6	6.3E-13	-33.03	348.08	0.43	C	Mgt	
7	2.6E-12	-25.29	245.92	0.68	C	Mgt	
8	5.5E-13	-50.77	238.76	0.64	C	Metal + mgt	
9	2.5E-12	17.28	2.00	0.36	M	All	Needs quadrupole fit
10	2.8E-13	53.83	129.44	0.19	C	N/A	D < 0.5
11	1.0E-12	-43.45	27.22	0.68	M	Metal + mgt	
12	2.5E-12	-45.13	33.70	0.68	M	Mgt	Can't fit
13	2.0E-12	39.83	358.02	0.75	C	Pyr	
14	2.7E-12	-7.69	245.95	0.73	M	Pyr	
16	1.4E-11	12.10	247.46	0.86	C2	Metal + mgt	
17	1.7E-12	23.71	58.54	0.69	M	Mgt (mostly)	D < 0.5
18	0.0E+00	0.00	0.20	0.00	C	Pyr (mostly)	
19	3.3E-12	-5.45	50.73	0.85	M	Mgt (mostly)	
20	3.3E-13	-8.07	220.06	0.23	C	Mgt + metal	D < 0.5
21	3.6E-12	-23.50	22.26	0.54	M	N/A	
22	3.0E-12	-12.75	275.09	0.59	C2	Metal	D < 0.5
23	8.8E-12	0.55	246.57	0.69	C	Metal + mgt	
24	8.7E-14	-12.50	147.44	0.72	M	Pyr (mostly)	
25	1.5E-13	-19.56	82.34	0.61	C	Metal	

26	5.3E-12	29.77	209.00	0.53	M	Pyr	Needs quadrupole fit
27	1.8E-12	-14.41	172.13	0.48	C2	Mgt (mostly)	Needs quadrupole fit
28	3.5E-12	-79.92	354.11	0.72	C2	Mgt (mostly)	Poor fit
29	2.2E-12	1.93	147.16	0.40	C2	Mgt (mostly)	$D < 0.5$
30	1.4E-12	-1.95	141.56	0.60	C2	Mgt (mostly)	
31	4.4E-13	25.45	87.86	0.52	?	Mgt	
32	9.8E-13	5.29	178.62	0.28	C	Pyr	Splits into several dipoles
33	2.2E-11	-6.58	172.96	0.46	C	Mgt (mostly)	
34	1.9E-11	-5.11	254.84	0.91	M	Pyr	$D < 0.5$
35	7.8E-13	-8.40	96.19	0.68	M	Pyr	
36	4.3E-12	-5.39	163.31	0.69	M	Metal + mgt	
37	1.5E-12	-22.60	117.89	0.37	C	Mgt (mostly)	Too faint
38	6.5E-11	8.15	234.54	0.85	M	Metal/all	
39	1.6E-11	54.46	280.92	0.40	C	Metal + mgt	Too faint
40	4.0E-12	-37.64	131.25	0.67	C	Mgt	
41	4.4E-12	16.96	14.30	0.78	C	Metal	
42	1.3E-12	-1.30	163.55	0.52	M	Pyr	
43	1.7E-12	72.48	30.62	0.78	C	Pyr	
44	5.1E-12	-44.60	211.01	0.82	M	Metal	
45	6.4E-13	38.59	270.84	0.26	M	Pyr	Poor fit
46	4.7E-12	10.14	253.92	0.40	M	Pyr	Needs quadrupole fit
48	9.0E-12	-17.39	18.08	0.81	C	Metal	
49	1.8E-12	31.33	46.36	0.51	M	Metal	
50	7.3E-12	-20.38	258.34	0.73	M	Metal	
51	6.4E-14	-25.77	197.55	0.44	C2	Pyr	$D < 0.5$
52	3.0E-13	11.10	256.70	0.57	C2	Metal	
53	4.0E-13	41.27	179.02	0.43	C2	Metal	Too faint

Table C.3: Dipole fits for the 50 mT AF step. N/A denotes a signal generated by a grain below the surface of the thin section. C = chondrule, M = matrix, Mgt = magnetite, Pyr = pyrrhotite, D = dipolarity parameter.

C.4 100 mT

Data-point	M (Am ²)	Inc (°)	Dec (°)	D	Component (C/M)	Phase	Reason for rejection (if applicable)
1	3.6E-13	-15.75	187.79	0.16	M	Pyr	Needs quadrupole fit
2	1.3E-11	-13.60	181.23	0.61	?	?	
3	1.2E-12	-14.04	148.11	0.64	M	Pyr (mostly)	
4	1.1E-12	-13.62	119.44	0.51	M	Mgt	
5	9.2E-13	-47.54	309.30	0.73	M	Pyr	Poor fit
6	3.8E-12	-9.54	337.37	0.38	C	Mgt	
7	1.5E-12	10.53	297.41	0.52	C	Mgt	
8	1.5E-12	-24.45	197.54	0.67	C	Metal + mgt	
9	8.2E-13	-35.57	303.46	0.41	M	All	D < 0.5
10	3.5E-13	38.87	268.04	0.62	C	N/A	
11	2.4E-12	-42.65	250.78	0.85	M	Metal + mgt	
12	2.7E-12	-11.08	267.49	0.55	M	Mgt	
13	1.6E-12	68.87	8.30	0.64	C	Pyr	
14	1.5E-12	2.35	241.57	0.61	M	Pyr	
16	5.9E-12	1.55	302.46	0.76	C2	Metal + mgt	
17	8.2E-13	-6.81	316.49	0.72	M	Mgt (mostly)	
18	3.7E-12	-24.36	144.06	0.40	C	Pyr (mostly)	Splits into several dipoles
19	3.8E-12	-31.52	275.19	0.53	M	Mgt (mostly)	
20	3.2E-13	-49.04	300.18	0.22	C	Mgt + metal	Needs quadrupole fit
21	1.4E-12	49.55	225.84	0.38	M	N/A	Needs quadrupole fit
22	4.1E-12	-65.70	98.08	0.75	C2	Metal	
23	1.3E-11	-38.19	179.95	0.66	C	Metal + mgt	

24	8.3E-14	4.05	317.59	0.64	M	Pyr (mostly)	D < 0.5
25	2.9E-13	14.96	105.16	0.49	C	Metal	
26	1.6E-12	-30.02	262.47	0.50	M	Pyr	
27	1.8E-12	1.72	120.42	0.70	C2	Mgt (mostly)	Poor fit
28	2.4E-11	8.01	211.35	0.70	C2	Mgt (mostly)	
29	4.3E-13	-31.06	321.70	0.27	C2	Mgt (mostly)	
30	3.6E-12	-20.22	268.60	0.62	C2	Mgt (mostly)	D < 0.5
31	5.9E-13	-38.65	54.30	0.60	?	Mgt	
32	5.6E-13	-39.47	211.68	0.28	C	Pyr	
33	2.2E-11	17.36	229.68	0.66	C	Mgt (mostly)	Splits into several dipoles Needs quadrupole fit
34	2.7E-12	-22.90	102.47	0.56	M	Pyr	
35	5.3E-13	-39.16	234.07	0.45	M	Pyr	
36	1.2E-12	-42.31	185.86	0.66	M	Metal + mgt	Too faint
37	2.3E-12	42.15	291.89	0.43	C	Mgt (mostly)	
38	3.4E-11	-7.87	97.84	0.69	M	Metal/all	
39	4.9E-12	-1.37	221.87	0.52	C	Metal + mgt	Poor fit
40	3.9E-12	13.90	137.29	0.66	C	Mgt	
41	3.5E-12	25.17	53.22	0.53	C	Metal	
42	2.4E-12	-79.06	249.08	0.81	M	Pyr	Poor fit
43	1.7E-12	68.24	-3.10	0.78	C	Pyr	
44	1.9E-11	5.03	247.99	0.60	M	Metal	
45	7.9E-13	-17.12	234.80	0.23	M	Pyr	Splits into several dipoles Needs quadrupole fit
46	6.9E-12	-18.54	307.25	0.38	M	Pyr	
48	4.5E-12	18.90	229.16	0.71	C	Metal	
49	3.4E-13	-4.59	289.63	0.26	M	Metal	Signal cut off
50	5.2E-12	27.35	241.57	0.51	M	Metal	
51	1.6E-13	58.10	10.91	0.59	C2	Pyr	
52	1.3E-12	-7.97	233.44	0.66	C2	Metal	

53	9.3E-13	28.68	128.71	0.56	C2	Metal	
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Table C.4: Dipole fits for the 100 mT AF step. N/A denotes a signal generated by a grain below the surface of the thin section. C = chondrule, M = matrix, Mgt = magnetite, Pyr = pyrrhotite, D = dipolarity parameter.

C.5 200 μ T ARM

Data-point	M (Am ²)	Inc (°)	Dec (°)	D	Component (C/M)	Phase	Reason for rejection (if applicable)
1	7.7E-12	-60.09	238.69	0.52	M	Pyr	Signal partially cut off
2	2.0E-10	-7.96	236.22	0.51	?	?	
3	2.5E-12	-1.00	253.67	0.53	M	Pyr (mostly)	Needs quadrupole fit
4	7.3E-12	-15.54	252.38	0.72	M	Mgt	
5	2.8E-12	-4.48	175.12	0.34	M	Pyr	D < 0.5
6	2.9E-12	-51.37	239.43	0.56	C	Mgt	
7	8.3E-12	-15.58	246.35	0.86	C	Mgt	
8	1.5E-12	-18.99	174.78	0.81	C	Metal + mgt	Poor fit
9	2.0E-12	-13.87	261.44	0.44	M	All	D < 0.5
10	8.2E-13	3.18	87.66	0.59	C	N/A	
11	3.4E-12	-19.58	227.92	0.72	M	Metal + mgt	
12	8.0E-12	-11.87	235.90	0.80	M	Mgt	
13	1.4E-12	54.42	349.02	0.56	C	Pyr	
14	4.9E-12	6.59	249.28	0.70	M	Pyr	
16	1.8E-11	5.46	256.06	0.74	C2	Metal + mgt	
17	2.6E-12	5.93	307.20	0.61	M	Mgt (mostly)	
18	7.1E-11	3.80	274.63	0.66	C	Pyr (mostly)	
19	2.1E-12	-11.08	259.73	0.44	M	Mgt (mostly)	Needs quadrupole fit
20	3.0E-13	37.10	206.43	0.23	C	Mgt + metal	Needs quadrupole fit
21	4.7E-12	-26.39	128.09	0.46	M	N/A	Needs quadrupole fit
22	1.3E-11	12.54	280.77	0.73	C2	Metal	
23	2.5E-11	-3.95	273.88	0.77	C	Metal + mgt	

24	3.7E-13	-19.62	204.59	0.53	M	Pyr (mostly)	
25	2.3E-13	15.96	82.72	0.44	C	Metal	D < 0.5
26	7.5E-12	48.10	273.08	0.36	M	Pyr	D < 0.5
27	9.5E-12	-4.02	250.42	0.65	C2	Mgt (mostly)	
28	7.2E-11	-4.09	267.99	0.72	C2	Mgt (mostly)	
29	6.2E-12	-7.80	268.74	0.44	C2	Mgt (mostly)	D < 0.5
30	8.1E-12	-6.02	268.24	0.66	C2	Mgt (mostly)	
31	3.2E-12	-14.40	258.30	0.57	?	Mgt	
32	1.8E-11	-4.39	257.17	0.78	C	Pyr	
33	9.6E-10	-6.88	270.64	0.60	C	Mgt (mostly)	
34	4.6E-12	10.90	280.03	0.57	M	Pyr	
35	5.0E-12	-8.93	263.96	0.71	M	Pyr	
36	5.0E-12	-6.03	232.56	0.58	M	Metal + mgt	
37	5.3E-11	2.20	260.41	0.87	C	Mgt (mostly)	
38	1.0E-11	67.31	242.14	0.58	M	Metal/all	Too faint
39	5.6E-12	-6.58	263.98	0.29	C	Metal + mgt	Too faint
40	8.4E-12	-26.06	273.92	0.49	C	Mgt	D < 0.5
41	6.0E-12	5.93	237.14	0.33	C	Metal	D < 0.5
42	4.0E-12	-3.05	254.72	0.62	M	Pyr	
43	1.4E-12	-5.91	235.34	0.54	C	Pyr	
44	1.0E-11	13.20	267.77	0.73	M	Metal	
45	1.1E-11	3.15	247.51	0.38	M	Pyr	D < 0.5
46	4.2E-12	-8.04	256.74	0.74	M	Pyr	Can't fit
48	1.0E-11	-7.17	21.64	0.37	C	Metal	D < 0.5
49	3.8E-11	-16.73	276.39	0.37	M	Metal	Signal cut off
50	7.9E-12	-27.15	265.90	0.64	M	Metal	
51	3.1E-13	-17.55	232.34	0.72	C2	Pyr	
52	2.9E-12	-16.35	234.06	0.74	C2	Metal	
53	5.4E-11	-9.73	240.43	0.48	C2	Metal	D < 0.5

Table C.5: Dipole fits for the 200 μ T ARM. N/A denotes a signal generated by a grain below the surface of the thin section. C = chondrule, M = matrix, Mgt = magnetite, Pyr = pyrrhotite, D = dipolarity parameter.

C.6 Stereonets

Dataset	Constituent	Phase	N	Dec (°)	Inc(°)	α_{95}	k	R/N
NRM	Matrix	Pyrrhotite	8	320.5	15.3	38.1	3.1	0.7148
		Magnetite	4	335.6	10.6	49	4.5	0.8327
		Metal	5	244.8	-9.6	19.1	16.9	0.9528
	Chondrule	Pyrrhotite	3	352.8	52.4	—	1.3	0.496
		Magnetite	5	228.3	16.3	—	2.2	0.6308
		Metal	5	260.8	-23.2	—	1.1	0.2636
25 mT	Matrix	Pyrrhotite	7	314.5	-11.1	—	1.3	0.3194
		Magnetite	4	2	29.9	57	3.6	0.7902
		Metal	5	233.1	-25.1	35.2	5.7	0.8591
	Chondrule	Pyrrhotite	2	68.2	59.9	—	—	0.8886
		Magnetite	4	83.4	-69.5	—	1.4	0.4684
		Metal	5	66.4	5.8	—	1.9	0.5738
50 mT	Matrix	Pyrrhotite	6	188.5	-34.2	—	1.3	0.3674
		Magnetite	4	55.2	-0.1	44.1	5.3	0.859
		Metal	6	218	-40.1	—	1.2	0.3025
	Chondrule	Pyrrhotite	2	7	57	—	—	0.9501
		Magnetite	4	146	-14.1	—	1.8	0.5778
		Metal	5	0.4	-41.9	—	1.2	0.3316
100 mT	Matrix	Pyrrhotite	6	253.6	-48	—	1.4	0.3946
		Magnetite	4	279.4	-32.3	—	1.5	0.4998
		Metal	5	218	-21.2	—	1.7	0.52
	Chondrule	Pyrrhotite	2	2.5	68.7	—	—	0.9993
		Magnetite	4	216.1	10.6	—	0.9	0.1328
		Metal	5	196.7	-18.2	—	1.3	0.3841
ARM	All	All	30	257.6	-5.8	9.6	8.5	0.8866

Table C.6: Summary statistics for the stereonet, showing the mean vector (declination and inclination) of each phase (pyrrhotite, magnetite, metal) in each meteorite constituent (matrix and chondrules), as well as the α_{95} , k and mean length (R/N) of the vector.

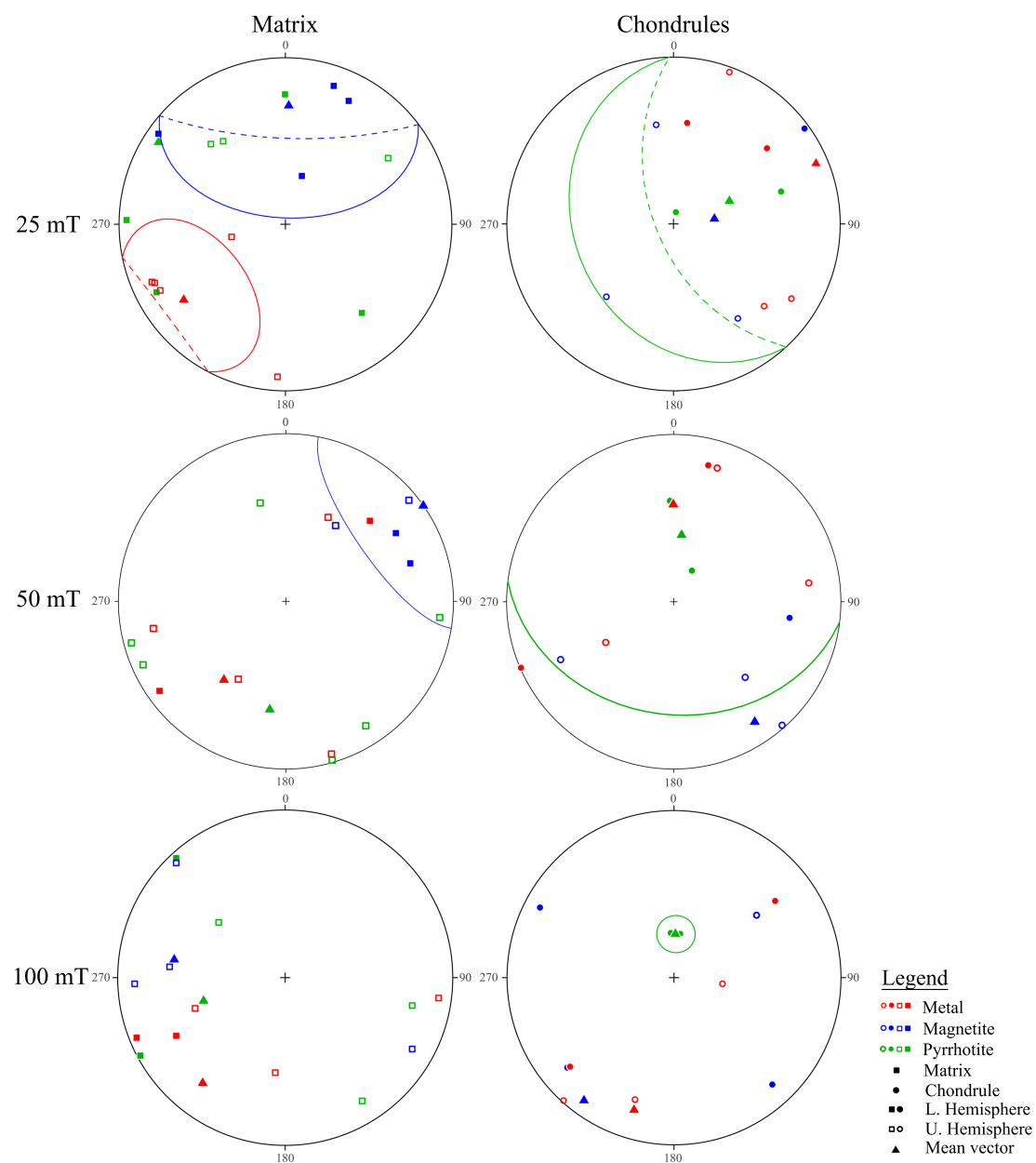


Figure C.1: Equal area lower hemisphere stereonet showing the fitted dipoles in the matrix (left column) and chondrules (right column) for the 25 mT, 50 mT and 100 mT AF steps. The triangles indicate the positions of the mean vectors, sometimes accompanied by $\alpha 95$ confidence ellipses for the more clustered datasets.

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