

A STUDY OF IRRADIATION DAMAGE IN IRON AND Fe-Cr ALLOYS

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Abstract

Irradiation damage structures induced in pure Fe and Fe-Cr (up to 14%Cr) alloys by 2 MeV Fe⁺ ion irradiations in the temperature range 300-460°C were investigated by transmission electron microscopy.

Specimens were irradiated in bulk to doses of 1.5×10^{19} Fe⁺/m² (about 2.5 displacements per atom: dpa) and 4.5×10^{19} Fe⁺/m² (about 7 dpa).

In most cases, damage took the form of dislocation loops with diameters from 2-100nm; the loops were distributed uniformly within all the samples. At higher irradiation temperatures (400°C, 460°C), complex microstructures such as finger loops (50nm in width and 1 micron in length) and perpendicular <100> loop clusters, were observed in both pure Fe and Fe-Cr samples.

Loop sizes and densities were seen to change as a function of irradiation temperature and dose. Loop sizes were seen to increase as the increase of irradiation temperatures and doses, while loop densities only increased with increasing doses and decreased as increasing temperatures. Loops with both types of Burgers vectors (<100> and $\frac{1}{2}$ <111>) were observed in all the samples. The proportion of <100> loops was higher in Fe than that in Fe-Cr alloys at the same irradiation condition, which has can be attributed to the high mobility of $\frac{1}{2}$ <111> loops in Fe, so that a large proportion of them will escape to the (001) foil surface.

A transition in loop Burgers vectors as a consequence of increasing temperature was observed. In Fe, the proportion of <100> loops increased with increasing irradiation temperature from 40% at 300°C to 60% at 460°C. A similar trend was found in the Fe-Cr alloys, but due to the higher proportion of $\frac{1}{2}$ <111> loops in these alloys, the increase of <100> loops was not that obvious, being from 30% at 300°C to 45% at 460°C(Fe-11Cr).

The effects of irradiation dose rate on the formation of dislocation loops by 2 MeV Fe^+ ions were also investigated. These irradiations were carried out at 300°C with two different implantation dose rates: 6×10^{-4} dpa/s and 3×10^{-5} dpa/s. The implantation dose for both implantations was $0.38 \times 10^{19} \text{ Fe}^+/\text{m}^2$ (0.5 dpa). Both the average loop size and loop densities for the Fe-Cr specimens subjected to the high dose rate irradiation were higher than that in the low dose rate irradiations. Take Fe-14Cr as an example, that the loop densities in high dose rate irradiation increased about 90% compared to that in low dose rate, and the average loop size in high dose rate irradiation was 30% larger than that in low dose rate irradiation.

The 'inside-outside contrast' method was applied to determine the loop nature in all the samples. It was found that all the large loops (>5nm) are of interstitial type. Any vacancies are believed to exist in the form of small dislocation loops (<5nm) or sub-microscopic voids.

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Introduction

Energy analyses have indicated that the world energy demand will increase continuously from current power levels of 14 TW to 25-60 TW in the following 50 years [1-3]. Also growing environmental concerns on carbon dioxide emissions are pressing the industrialized nations to find replacement energy sources for fossil fuels, and nuclear fusion is a promising choice for future energy supply with the advantages of being sustainable, no greenhouse gases emissions, no long-lasting radioactive waste and the substitution of fissionable materials eliminates the possibility of serious nuclear leaking disasters [4-8].

Despite all the advantages, the structural materials of the first-wall and breeding blanket of the future commercial fusion reactors have to be carefully chosen. Since the structural materials will suffer high-flux irradiation damage from the 14MeV high energy fusion neutrons, working temperatures up to 600°C and high stresses up to 300MPa, combined with high levels of helium and hydrogen produced in transmutation reactions [1, 9-12]. As a result, the mechanical properties of the structural material will be severely damaged by radiation induced effects as hardening, swelling and embrittlement [13-18].

Moreover, another serious problem is the activation of materials caused by fusion neutrons. The unstable isotopes which are produced during neutron irradiations will cause the materials to be radioactive. As a result, 'reduced-activation' materials which eliminate the components which will cause long life radioactivity have to be used [1, 8, 19].

The ideal candidate materials should fulfill the challenging requirements, such as high thermal conductivity and heat resistance, low DBTT (Ductile-Brittle Transition Temperature), low swelling rate, high creep resistance and good corrosion resistance [6, 7, 20, 21].

In recent years, high-chromium (9-12% Cr) ferritic/martensitic steels have been considered as a potential candidate for the first-wall and breeding blanket structural materials for future fusion reactors [22, 23]. Among which, it was discovered that the low-activation ferritic/martensitic steels based on Fe-9Cr, containing small amounts of W, V and Ti, are resistant to swelling, less brittle and have better thermal properties and higher swelling resistance [5, 6, 24]. Although Fe-Cr alloys (stainless steels) became popular as early as the 1920s, it is only very recently that the radiation damage mechanisms in these complex materials begin to be studied systematically [25]. At present it is still not clear how to modify the existing steels to meet the demanding fusion reactor environment.

Model Fe-Cr alloys are now the subject of many fusion related research groups in the UK, Europe and the United States, since the fundamental aspects of radiation damage can be better understood by investigating pure materials and simple model alloys [26].

This is the approach adopted by the materials for fusion & fission group in Oxford Materials as well. The project is focused on model iron- and tungsten- based alloys and attempts to combine modelling and experimental approaches as TEM, atom probe and micromechanical testing, to give a better fundamental understanding of the radiation damage mechanism of iron- and tungsten- based alloys [27]. The work contained in this thesis is concentrated on the evolution of radiation damage of bulk Fe and Fe-Cr alloys during 2MeV Fe⁺ ion irradiation at various doses and temperatures.

Chapter 1: Literature Review

1.1 Fundamentals of Radiation Damage

1.1.1 Atomic Displacement Processes in Irradiated Metals

In order to interpret the experimental results described in the literature and this work, it is necessary to understand the fundamentals of radiation damage processes which involve the creation of point defects, their subsequent recombination and the aggregation of the point defects into visible defects clusters.

During irradiation the incident particles can lose energy through interacting with both the electrons and the atoms of the target materials. If a lattice atom receives sufficient energy to overcome a displacement barrier, then the atom will be displaced from its lattice site. The minimum energy required for this displacement is called the threshold displacement energy E_d (typically $\approx 40\text{eV}$ for Fe) [28, 29].

After the atom has been displaced from its lattice site, it can produce further displacements if its remaining energy is much greater than E_d . If its remaining kinetic energy is less than E_d , the displaced atom can only create a single interstitial-vacancy pair (Frenkel defect). The stability of the Frenkel pair depends on the separation distance between the interstitial and the vacancy. If an interstitial lies within the spontaneous recombination volume of a vacancy, they will recombine spontaneously by the attraction of their strain fields [28, 30].

In real fusion reactor conditions, the situation of an incident fast neutron is more complex as it produces numerous primary knock-on atoms. The primary knock-on atom (PKA) is defined as the lattice atom first struck and displaced by the bombarding particle, and knock-on atoms can have a wide range of energies from a few keV to MeV. These knock-on atoms lose energy through interactions with electrons and atoms of the solid. In the high energy range (a few MeV), the knock-on

atoms won't produce any permanent damage as inelastic collisions with electrons is dominant in this energy range. The few elastic collisions that happen in this range only produce well separated Frenkel pairs, as the displaced atoms may have enough energy to become sub knock-on atoms and produce more displacements [29, 30]. As the energy of the knock-on atoms decrease to a few hundred keV, the elastic collisions with lattice atoms dominate the collision events, and as the distance between elastic collisions decreases towards the interatomic separation distance, a displacement cascade is produced (see Fig.1.1.1.). The centre of the cascade may contain several hundred Frenkel defects within a volume of typical dimension of 10nm [31]. An important phenomenon occurring at this energy range is the formation of a vacancy-rich 'depleted zone' in the centre of the cascade with a shell of interstitials in the outside. It was observed by transmission electron microscopy that the depleted zone could collapse spontaneously to a vacancy loop [29, 30, 32]. To conclude, during high energy (~MeV) irradiation, the incident ions and knock-on atoms produce Frenkel defects, together with the possibility of a vacancy-rich volume only at the end of their paths.

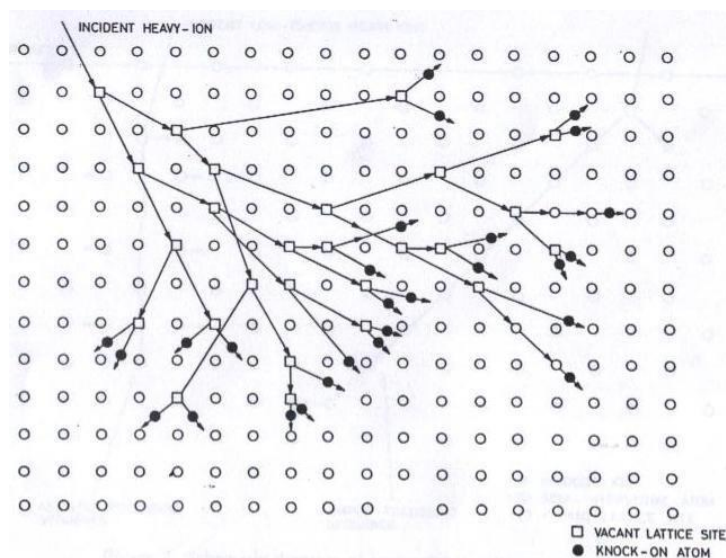


Fig.1.1.1. Displacement cascade with a vacancy-rich region in the centre [28].

During the irradiation, the majority of Frenkel defects disappear through recombination or loss to sinks such as dislocations, grain boundaries or surfaces. But some point defects meet the defects with the same character and form clusters. Because interstitials are much more mobile than vacancies, the clusters are predominantly interstitial in nature. Further interstitial migration increases the size of these interstitial clusters but it decreases the size of any vacancy clusters [32]. Since vacancy clusters are unstable during irradiation, they tend to attract mobile interstitials, while the vacancies are comparatively sessile at reactor temperature. In bcc metals, the surviving interstitials are believed to nucleate initially on {110} planes to form faulted loops with a Burger vector of $\frac{1}{2}\langle 110 \rangle$. But at an early stage in their growth, these loops unfault to remove the stacking fault, by the propagation of a shear across the fault to form a perfect loop [29, 30]. Eyre and Bullough [33] suggested that the faulted loops can shear in either $\langle 110 \rangle$ or $\langle 100 \rangle$ direction to form perfect loops with Burgers vectors of $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ respectively. As the $\langle 110 \rangle$ shear requires a higher energy than a shear in the $\langle 100 \rangle$ direction, fewer interstitial loops with a $\langle 100 \rangle$ type Burgers vector are expected to be formed. This conclusion can also explain the trend that at low temperature irradiation in Fe and Fe-Cr alloys, the majority of the loops were found to be $\frac{1}{2}\langle 111 \rangle$ loop, while $\langle 100 \rangle$ loops were only observed at elevated temperatures [34].

Voids are three dimensional vacancy clusters that are produced during high temperature irradiations at which vacancies are mobile, i.e. at temperatures between 0.2 to 0.5 T_m where T_m is the melting temperature [29]. It is believed that gas atoms are either exist in the materials before irradiation or are generated by transmutation reactions during the irradiation are necessary for the nucleation of voids, as the gas atoms can stabilise the void against collapse to vacancy loops [13, 30].

1.1.2 Basic Crystallography of Dislocation Loops

In this work, dislocation loops are the main production produced by the injected ions during irradiation. Therefore, in order to understand the profound effects of dislocation loops on the irradiation induced mechanical property changes, it is essential to learn the crystallography of dislocation loops in metals.

Pure Edge Loops and Non-edge Loops

An interstitial pure edge loop is formed when a platelet of atoms is inserted between two layers of regularly spaced crystal planes. If a platelet of atoms is removed from a plane, and the crystal is allowed to collapse, a vacancy pure edge loop will be formed (see **Fig.1.1.2.1**). Pure edge loops lie in the plane normal to their Burgers vector.

Glide of pure edge loops is controlled by the normal glide restrictions for an edge dislocation, and the motion will not change the projected area. **Fig.1.1.2.2a** shows the glide cylinder of a pure edge loop. The glide cylinder is defined by lines parallel to the Burgers vector and going through all points on the dislocation loop. The structure of a pure edge loop is considered steady, since it can only change its size by absorbing interstitial atoms or vacancies. A non-edge loop is produced when half of a pure edge loop glides on its cylinder while the other half remains stable (see **Fig.1.1.2.2b**). This non-edge loop has the same glide cylinder as the original pure-edge loop, but if it has screw parts it can glide on any plane **[29, 35, 36]**.

Shear Loops

A shear loop is formed by the sheared region within the crystal, when a dislocation runs from one crystal interface to the next. The boundary region of the dislocation is composed of displacements with pure edge and pure screw character. For a shear loop, its Burgers vector is in the plane of the loop and forms varying angles with the line dislocation (see **Fig.1.1.2.3**). A shear loop is unstable, and it can shrink or grow conservatively.

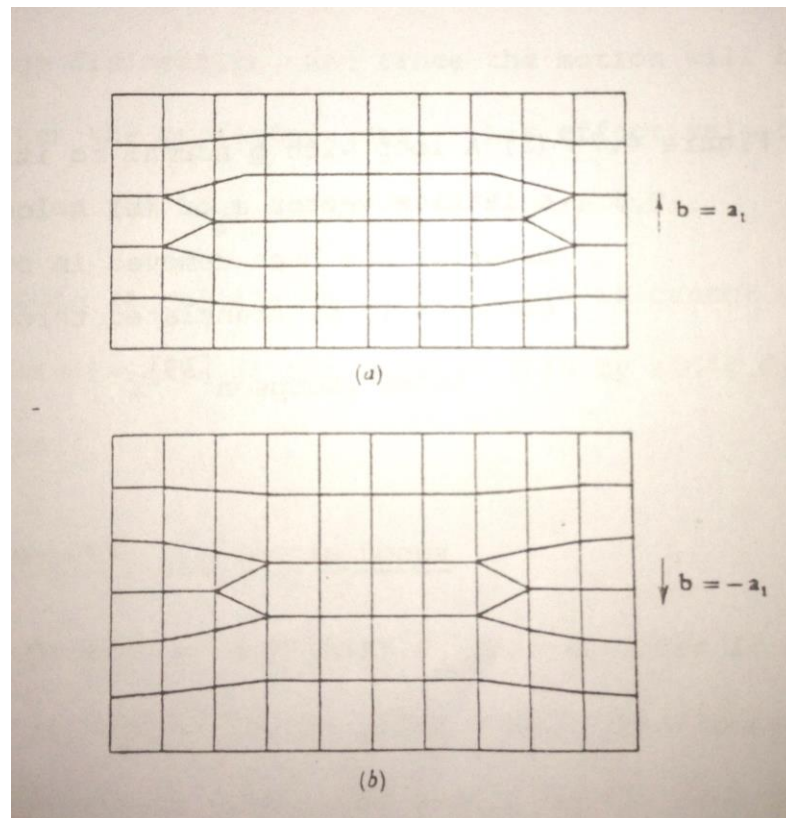


Fig.1.1.2.1. Burgers vector of an interstitial pure edge loop (a), a vacancy loop (b) [35].

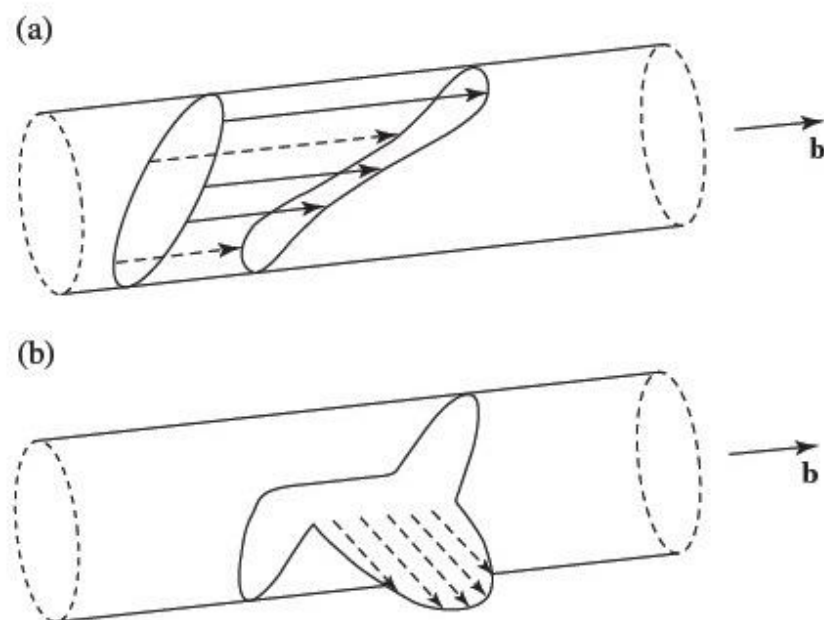


Fig.1.1.2.2. The dislocation loop shown in (a) can glide so that its area projected normal to the Burgers vector does not change. (b) This shows that a screw segment can leave the surface of this cylinder without changing the projected area [35].

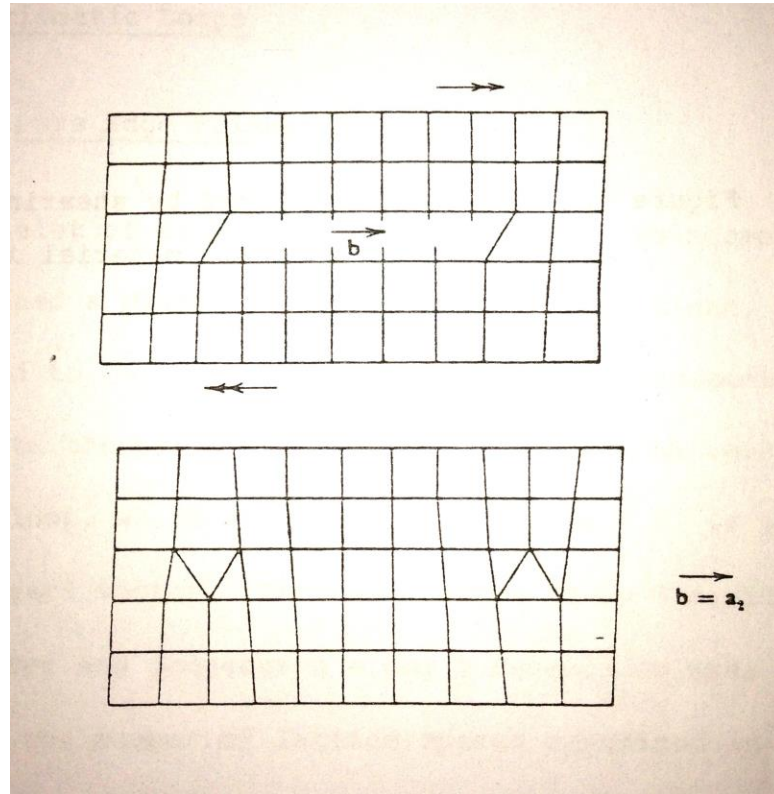


Fig.1.1.2.3. Formation of a shear loop[35].

1.2 Approaches to Understand Radiation Damage Mechanisms

Since no current facilities can produce neutrons with an energy as high as 14MeV, in the foreseeable future the radiation damage inside the fusion reactor will be simulated by other means such as fission neutrons, protons and heavy-ions [37-40]. After the irradiation experiments, the microstructure evolution can be studied by the experimental methods such as Transmission Electron Microscopy(TEM), Atom Probe Field Ion Microscopy(APFIM) [41-44], Small Angle Neutron Scattering(SANS) [31, 45-48], and Positron Annihilation Spectroscopy(PAS) [48-52].

1.2.1 Simulations of Fusion Neutron Damage

In fast neutron irradiation, the production of cascades is the major primary damage mechanism, and typically over 95% of the displacements occur in cascades produced by the displaced atoms with the energy of 5keV to 200keV [28]. Therefore in fusion neutron irradiation, knock-on atoms with the energy less than 200keV are the major producers of the radiation damage.

In radiation damage studies, heavy-ions are not used to simulate the fusion neutron directly, but to simulate the primary knock-on atoms (PKAs) created by fast-neutrons, since these injected heavy-ions have similar energies to the PKAs. During the irradiation, the incident atoms with energies of a few MeV penetrate into the target, and produce dense collision cascades near the end of their paths when the energy falls to $\leq 200\text{keV}$ [8, 28].

Apart from the displacement cascades, the fast neutrons will also produce nuclear transmutations of the type (n, p) and (n, α), which generate hydrogen and helium gas [53-56]. These gas atoms along with the vacancies produced through the cascade events can determine the subsequent void growth dynamics. Therefore, in order to simulate the real lattice environment within the fusion reactor, simultaneous

implantations of He^+ and H^+ during the heavy-ion bombardment are necessary [57-59].

Advantages of the use of heavy ions and gas ions are that the energy, dose, and dose rate of the ion beam and the irradiation temperature can be controlled allowing cascade collapse to be studied systematically. Since dose rates of charged particles can be much higher than of reactor fast neutrons, large doses can be built up in convenient times [8, 60-62]. Moreover, ion irradiations do not cause activation and transmutation effects, which allows safe handling in experiments.

However, the disadvantages are that the ion range is much smaller than that of neutrons so that bulk effects may not be simulated realistically, also the dose rate of ion irradiation is much higher than neutron irradiation so any dose rate effects should also be taken into consideration [8, 63].

1.2.2 The Advantages of TEM in Radiation Damage Study

TEM (Transmission Electron Microscopy) has played a central role in characterizing radiation damage microstructures and deducing atomistic mechanisms. Nowadays TEM can form images at magnification of the order of 10^5 or higher with a resolution of 1nm or even better. By using TEM, researchers can actually 'see' the defects clusters and other complex microstructures. With skilful analysis, TEM can be used to reveal the Burgers vectors, nature, size and densities of the irradiation induced damages [64].

Moreover, combined with in-situ irradiation facilities, TEM can allow the accumulation of radiation damage to be observed directly, and TEMs with analytical equipment such as Energy-dispersive X-ray spectroscopy (EDX) [65-70], electron energy loss spectroscopy (EELS) [71-76], can also give information on local chemical changes on a very fine scale as well.

1.3 Radiation Damage Evolution in Fe and Fe-Cr alloys

Fe and Fe-Cr alloys are the basis of many complex ferritic/martensitic steels which are considered as the promising candidate structural materials for fission and fusion reactors. As described in the 'Introduction', pure materials and simple model alloys are often used to understand the fundamental aspects of radiation damage. Since the 1960s, numerous experiments have been done on the understanding of damage development in Fe, Fe-Cr alloys and steels. In this chapter we review the previous achievements, with an emphasis on TEM studies in Fe and Fe-Cr alloys.

1.3.1 Radiation Damage in Pure Fe

Cascade phenomena have been studied extensively during the 1960s, since these displacement -cascades contain the majority of point defects produced by irradiation, which play a crucial role in the understanding of irradiation induced microstructure changes. Scientists found that in many pure materials, i.e. Cu, Cu₃Au, Co etc., vacancy loops are produced directly in cascades by a process of 'cascade collapse' [40, 61, 64]. However in iron, visible damage is only seen at high doses when cascade overlap becomes significant ($>10^{16}$ ions m⁻²) [77-79].

For instance, in 1978, Jenkins et al. [79] studied low-dose thin foil experiments in pure Fe. No visible damage was observed at doses lower than 5×10^{16} ions m⁻², during self-ion irradiation with the energies of 40-240 keV at RT. However they also found that in very heavy or molecular ion irradiations, vacancy loops can be produced in individual cascades.

In 1987, Robertson et al. [9] studied radiation damage in pure Fe by low dose fission neutron irradiation at PLUTO/Harwell. They found that at 353K, below a dose of 4×10^{23} neutrons m⁻², no visible damage was observed. At doses above 4×10^{23} neutrons m⁻², small loops appeared near line dislocations and grain boundaries, and as the dose increased, visible loops were present in the matrix.

This phenomenon implied that invisible vacancy clusters are produced at cascade sites, but only aggregate to form visible loops under overlap doses.

In the 1980s, by using in-situ irradiation facilities in radiation damage studies, many heavy-ion experiments could be observed in real time. Robertson et al. [77, 78] observed that visible loops only appeared at overlap doses ($>10^{16}$ ions m^{-2}) at RT and 30K, during their self-ion implantation experiments in pure iron. In 2008, in-situ studies by Yao et al. [26] at RT and 300°C also confirmed that individual cascades do not collapse in iron.

Also in the 1960s, contradictory results were produced on the type of loop Burgers vector in ion/neutron irradiated pure iron. For instance, Eyre and Bartlett [80] made an analysis of the loops in the neutron-irradiated iron at 60°C, and annealed up to 500°C. Both interstitial and vacancy loops were present. The loops in the annealed foil had Burgers vector of $\frac{1}{2}\langle 111 \rangle$. At nearly the same time, Masters [81] found that after a 550°C, self-ion irradiation at 150keV, only interstitial loops with Burgers vector of type $\langle 100 \rangle$ were present. Both observations were confirmed by other authors, some found that only $\frac{1}{2}\langle 111 \rangle$ loops were observed [4, 45], others found that most loops, if not all, have $\langle 100 \rangle$ Burgers vector [27, 29], while other results showed that during irradiation, both loop types appeared, and the relative amounts of the loops depend on the alloy composition, radiation temperatures [82-87].

The detailed irradiation conditions used in the experiments mentioned above are listed in **Table 1.3.1.1**. Based on these observations, it seems that both interstitial and vacancy type of $\frac{1}{2}\langle 111 \rangle$ have been observed in iron irradiated at ambient temperature, and interstitial loops of $\langle 100 \rangle$ type have been found at elevated temperatures. One explanation for the apparently contradictory observations is that $\langle 100 \rangle$ loops only appear at 500-600°C during heavy-ion irradiation in Fe, as the formation of $\langle 100 \rangle$ interstitial loops require higher energy [29].

In 1965, Eyre and Bullough [33] suggested that the interstitial loops were first aggregated on {110} planes to form faulted loops with $\frac{1}{2}\langle 110 \rangle$ Burgers vectors. Because of the high stacking-fault energy contained in the loops, these loops will remove the stacking fault, at an early stage, by a shear in $\langle 100 \rangle$ or $\frac{1}{2}\langle 111 \rangle$ direction. As the shear in $\langle 111 \rangle$ direction needs higher energy than a shear in $\langle 100 \rangle$ direction, so at lower temperatures, fewer interstitial loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vector are expected to be formed.

It is only recently that Dudarev et al. [88] suggested that due to spin fluctuations in Fe, an elastic instability occurs around the alpha-gamma phase transition temperature, and that this instability causes a dramatic reduction of the shear stiffness constant, which has a strong effect on the stabilities of $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ prismatic edge loops. This theory predicts that at the temperatures below 350°C, loops of $\mathbf{b}=\frac{1}{2}\langle 111 \rangle$ are more stable, while at higher temperature the loops with Burgers vector of $\langle 100 \rangle$ become relatively more stable.

Materials	Year	Radiation Conditions	Results	Ref
α -Fe	1964	Fission neutron; Dose $\sim 2 \times 10^{20}$ ions/cm ⁻² at 60°C	$\mathbf{b} = \frac{1}{2}\langle 111 \rangle$; both interstitial and vacancy loops were present	[80]
α -Fe	1965	150keV; protons/Fe+; Doses $\sim 1.7 \times 10^{16}$ ions/cm ² ; Dose rate $\sim 4 \times 10^{14}$ ions/cm ² sec; 550°C	Interstitial loops, $\mathbf{b} = \langle 100 \rangle$	[81]
α -Fe	1966	Fission neutron; E > 1.45 MeV; irradiated at 30-90°C; post irradiation heating at 260~600°C	Interstitial loops, $\mathbf{b} = \frac{1}{2}\langle 111 \rangle$	[4]
α -Fe	1973	Electron irradiation; 1 MeV; 550°C	$\mathbf{b} = \langle 100 \rangle$ Burgers vector	[27]
α -Fe	1965	low-dose fission neutron (E > 1MeV)	No visible damage when doses < 4×10^{23} neutrons m ⁻² at 353K; smaller loops had $\mathbf{b} =$ $\frac{1}{2}\langle 111 \rangle$; larger loops had $\mathbf{b} =$ $\langle 100 \rangle$; interstitial in nature	[33]
α -Fe	1978	40-240keV; Fe+; Doses < 5×10^{16} ions m ⁻² ; RT	Both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were found in approximately equal numbers.	[79]
α -Fe	2000	neutron irradiation to a fluence of 0.06 dpa; 280°C	About 80 % have Burgers vectors $\mathbf{b} = \langle 100 \rangle$, and the remainder have $\mathbf{b} = \frac{1}{2}\langle 111 \rangle$.	[29]
α -Fe	2006	neutron irradiated at 50 – 70°C; 0.001 – 0.79 dpa	Interstitial loops, $\mathbf{b} = \frac{1}{2}\langle 111 \rangle$	[45]

Table.1.3.1.1. Irradiation conditions used in the experiments mentioned in this section.

1.3.2 Radiation Damage in Fe-Cr alloys

Ferritic alloys, i.e. Fe-based alloys with body-centred-cubic structure, are the basis of many complex steels which are considered as the potential first-wall and breeding blanket structural materials for future fusion reactors [89-91]. This kind of material is well known to be highly resistant to swelling and damage accumulation in the radiation environment, and high chromium contents are known for good resistance against corrosion [8, 14, 23, 92]. It was known that the Cr content of steels and Fe-Cr alloys affects both fracture behaviour and void swelling. The alloys containing 9wt% Cr have the lowest DBTT, are less brittle and are resistant to erosion corrosion in the temperature range 300-500°C [8, 14, 93, 94]. Chromium is commonly added to steels to increase corrosion and oxidation resistance, also it is widely known that a relatively high amount of interstitial Cr atoms can increase hardenability and high-temperature strength [95-97].

Before the 1980s, the majority of TEM studies on radiation damage effects were focused on Fe. Only mechanical property studies and some limited TEM investigations were conducted in Fe-Cr alloys and ferritic steels. Some of the irradiation conditions and results of irradiation experiments in Fe-Cr alloys which are mentioned in this section are listed in **Table.1.3.2.1.**

Compared with pure materials such as Fe, Cu, Ni, etc., cascade collapse in Fe-Cr alloys has not been studied extensively. However recent in-situ thin foil irradiation experiments carried out in Argonne by Yao et al. [26] showed that the early stage of damage development in Fe-Cr alloys was very similar to that of Fe. Visible loops were observed only at overlap doses. The threshold doses for Fe and Fe-Cr alloys depended on materials, foil orientation and irradiation temperature, but all fell in the range of 10^{16} - 10^{17} ions m^{-2} .

Several researchers have observed that the addition of Cr atoms decreases the size of the loops, and increases the loop number density. For instance, Matijasevic et al. [25] studied Fe-Cr model alloys with different Cr concentrations (0, 2.5, 5, 9 and 12 wt%Cr). The materials were irradiated with neutrons at 300°C in the Belgian Reactor 2 (BR2) reactor to three different doses (0.06, 0.6 and 1.5 dpa). Both tensile tests and TEM observations were used to investigate the radiation hardening mechanisms. They found that the size of loops increased with dose, but decreased with Cr concentration, also the density of loops increased with dose up to 0.6 dpa. For the mechanical properties, they found that hardening is much higher in all Fe-Cr alloys that they tested than that in pure Fe. A minimum hardening rate was found around 9%Cr.

Porollo et al. [98] observed similar effects of Cr effects on loop size and densities. They found that at 400°C, in neutron irradiated Fe-Cr alloys, a mixed population of loops with $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ Burgers vectors were observed, and the addition of Cr atoms results in a decrease of loop size and increase of loop number density. Moreover, it was discovered that the presence of Cr atoms affects the loop Burgers vectors and swelling. Gelles et al. [99, 100] observed that the fraction of $\frac{1}{2}\langle 111 \rangle$ loops increased with increasing Cr content, from $0.5 \times 10^{14} \text{ n.m}^{-2}$ at 9%Cr to $0.8 \times 10^{14} \text{ n.m}^{-2}$ at 18%Cr. This observation can also explain the change in swelling rate according to different Cr content in Fe-Cr alloys. In their experiments, Fe-3%Cr has the least void-swelling, due to the large fraction of $\langle 100 \rangle$ loops. A peak swelling of 7.4% was found in a specimen of Fe-9%Cr irradiated at 425°C to 200dpa. It was thought that $\langle 100 \rangle$ loops are sessile in ferritic materials, therefore the mobility of the dislocation networks is reduced, which may cause the lower swelling rate.

This assumption was later confirmed by Katoh et al. [22] at the Fast Flux Test Facility Materials Open Test Assembly (FFTF/MOTA). They did neutron irradiation at 698 K to 140 dpa, and found similar results on swelling as a function to Cr content.

Irradiation damage in Fe-Cr has also been studied by in-situ heavy-ion irradiations by Jenkins et al. [101], Yao et al. [26] and Hernandez-Mayoral et al. [102] Compared with pure Fe, they founds that the damage were in a much finer scale, and loop loss and 'hopping' motion were reduced in Fe-Cr alloys. These experiments are discussed in depth in 1.4.

Materials	Year	Radiation Conditions	Results	Ref
Fe-Cr	1982	Doses : $\sim 10^{26} \text{ n m}^{-2}$; 400-450°C;	Proportion of $a/2\langle 111 \rangle$ loops increased with increasing Cr content.	[99]
Fe-9Cr/ Japanese Ferrite Martensitic Steels	1986	40keV, Cu^+ , Fe^+ ; Doses: 8×10^{16} - 8×10^{17} ions m^{-2} ; RT	Dislocation loops: 1-3nm; 90% vacancy, 10% interstitial; B-vector not determined.	[103]
Fe-10% Cr and Fe-10% Cr-1%Ni alloys	1988	Electron; 1 MeV ; Flux: $1.0 \times 10^{22} \text{ e/m}^2 \text{ s}$ $\sim 5.0 \times 10^{23} \text{ e/m}^2 \text{ s}$.	The growth of interstitial loops was slower in the presence of Cr; Nucleation rate had a very strong dependence on irradiation temperature;	[104]
Fe-Cr (3 to 18% Cr)	1995	Doses : $\sim 10^{26} \text{ n m}^{-2}$; 400-450°C;	Proportion of $a/2\langle 111 \rangle$ loops increased with increasing Cr content.	[100]
Fe-Cr alloys and Fe-Cr-C alloys	1995	high-dose neutron irradiation;	The void swelling was minimised for low chromium content (i.e Fe-3Cr), and the peak swelling of 0.7% was found in Fe-9Cr.	[22]
Fe-Cr (0, 2, 6, 12 and 18 at. % Cr).	1998	Neutron (BR-10 fast reactor); 400°C; Doses: 5.5-7.1dpa.	Most of the dislocation loops in Fe, Fe-2Cr and Fe-6Cr had $\mathbf{b} = a\langle 100 \rangle$; the loops were interstitial in nature	[24]
Fe-Cr alloys	1998	400°C in the BR-10 fast reactor; Doses: 5.5-7.1dpa.	Agrees well with former experiments, that void swelling was minimised for low chromium content.	[24]
Fe-9Cr	2004	Electron; 1000kV; Temperatures: 110~ 680 K	Transformation of the Burgers vector of dislocation loops was observed.	[105]
Fe-Cr alloys	2007	Neutron irradiation in Belgian Reactor 2 (BR2)	Both $a\langle 100 \rangle$ and $a/2\langle 111 \rangle$ dislocation loops were observed.	[106]
Fe-Cr alloys (0, 2.5, 5, 9,12 wt%Cr)	2008	Neutron irradiation at 300°C to three different doses (0.06, 0.6 and 1.5 dpa).	Loop size increases with dose, but decreases with Cr concentration	[25]

Table.1.3.2.1. Irradiation conditions used in the experiments mentioned in this section.

1.3.3 Irradiation Dose Rate Effects

In fusion reactors, the blanket structural materials are exposed to 14MeV neutrons at a higher dose rate than in fission reactors (10^{-12} ~ 10^{-11} dpa/s in fission reactors compared with 10^{-7} dpa/s in future fusion reactors) [107, 108]. Irradiation experiments using lower energy fission neutrons can examine bulk effects not explored by other irradiation types [109-111]. Heavy-ion experiments are conducted at high dose rates (10^{-4} ~ 10^{-5} dpa/s) so that cumulative dose levels can be achieved in very short times [112-115]. Both types of irradiation are used to simulate the radiation environment within fusion reactors. Therefore it is important to evaluate the effects of dose rate on micro-structural evolution and to provide data for modelling of dose rate effects to conduct assessments of irradiation performance in a fusion environment [116]. In the 1980s the increasing concern in using fast reactor irradiations for studies of light-water reactor (LWR) materials drove the need for understanding dose-rate effects, which were focused on the swelling and creep rates of austenitic stainless steels [117-122].

It is known that dose rate has a strong effect on micro-structural evolution and resultant macroscopic property changes. For example, in 1979, Lewthwaite et al. [123] studied creep behaviour in several different casts of austenitic stainless steels under different damage rates; the results showed that the creep rates decrease as dose rate increases. In 1993, Garner et al. [124], proposed that the irradiation temperature and the rate of atomic displacement would be the most important environmental variables for swelling in the Pressurized Water Reactors (PWRs). In 2007, Okita et al. [125] studied the dose rate effects on 4.0 MeV nickel ion irradiated Fe-15Cr-16Ni. After irradiation, they observed that the swelling rate increased with decreasing dpa rate, and this phenomenon was observed in four different temperatures from 300°C to 600°C, with three different dose rates (1×10^{-4} ,

4×10^{-4} and 1×10^{-3} dpa/s) at each temperature (300,400,500,600°C). This increase in swelling is believed to be due to the earlier un-faulting of larger Frank loops produced at lower density and the subsequent evolution of the dislocation network. In 2010, Hernandez-Mayoral et al. [81] studied flux and dose effects at the BR2 (Mol, Belgium). TEM analysis showed that both the defect densities and sizes increased with dose (see Table.1.3.3.1). It was also found that at the same dose level (dpa), irradiation at higher flux generated a higher defect density.

Material	Dose (dpa)	Average size (nm) (total number measured loops)	Maximum size (nm)	Density ($\times 10^{20}$ dislocation loops/m ⁻³)
Fe	0.026	—	—	3.4 ± 0.4
	0.051	4.9 ± 0.3 (251)	15	8.6 ± 0.8
	0.10	7.1 ± 0.2 (663)	21	12.8 ± 0.8
	0.19	10.2 ± 0.3 (1721)	49	39.1 ± 0.7

Table.1.3.3.1. Summary of quantitative results obtained from TEM images[81].

These observations have drawn attention to rates of defect recombination, and defect interaction which are believed to play an important role in the explanation of dose rate effects. Still, only limited studies have been made of dose-rate effects in structural materials, and these have concentrated mostly on swelling and creep where quite strong effects have been found. No systematic TEM studies have been conducted to investigate micro-structural changes in response to irradiation dose rate.

1.3.4 Summary

1. In both Fe and Fe-Cr alloys, visible loops were observed only at overlap doses for self-ion irradiation. However, irradiation with heavier ions can produce spontaneous cascade collapse.
2. Both $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ loops were seen in pure Fe, and as the irradiation temperature increases, many researchers observed that the proportion of $\langle 100 \rangle$ loops increases, which is consistent with the theoretical framework developed by Dudarev et al.[88] based on the anisotropic elasticity theory, that $\frac{1}{2}\langle 111 \rangle$ loops are

stable at temperature below 350°C, while at higher temperatures $\langle 100 \rangle$ loops become increasingly stable.

3. In Fe-Cr alloys, it was found that the damage structures were in a finer scale compared with Fe which was irradiated at similar conditions. The addition of Cr atoms results in a decrease of loop size and increase of loop number density. Moreover, the fraction of $\frac{1}{2}\langle 111 \rangle$ loops increases with increasing Cr content.

4. It was reported in many cases that 9%Cr alloys have the greatest swelling among other Fe-Cr alloys after irradiation. One explanation is that the swelling is suppressed by preferential formation of sessile $\langle 100 \rangle$ loops at low Cr content.

5. Irradiation dose rate is observed to have profound effects on swelling and creep rate of Fe-Cr alloys, it was discovered that at high dose rate irradiations, voids were formed with higher density and smaller size, which is attributed to the defect recombination, and defect interaction during the irradiation.

1.4 Dynamic Observations of Heavy-ion Damage in Fe & Fe-Cr alloys

In **1.3**, numerous ex-situ irradiation experiments have been described which were designed to understand the fundamental aspects of radiation damage in ferritic materials by investigating pure Fe and simple model Fe-Cr alloys.

One of the difficulties in radiation damage study is that quantitative comparisons between different experiments are difficult to draw, since the experiments were done under different doses, dose rates and irradiation temperatures and by using different kinds of ions and ion energies. Moreover, sometimes even when two experiments were carried out under similar conditions, the materials purity may also lead to different results. Considering that the irradiation damage studies are particularly time and expense consuming, the trends on radiation damage study are hard to conclude and sometimes contradictory results were published.

In the 1980s with the aid of in-situ TEM irradiation facilities, studies have been made on the irradiation damage in Fe and Fe-Cr alloys. Normally, these in-situ TEM experiments use much lower energy ions (eg.150keV) compared with 2MeV in the bulk experiments. However, the damage mechanism in thin-foils is still a good representation of the bulk, since as described in **1.1**, higher energy ions (>1MeV) initially lose most of their energy through electronic excitation and cause displacement damages when their energy falls below 200keV.

A series of in-situ thin foil ion irradiation experiments in pure Fe and Fe-Cr alloys were carried out by the Oxford Materials Group: Yao et al. **[26, 34]**, Hernandez-Mayoral et al. **[102]**, and Jenkins et al. **[101]**. In these experiments, they revisited earlier thin-foil ion irradiation experiments in pure Fe and Fe-Cr binary alloys **[61, 126-131]**. With the aid of the Argonne NEM-Tandem Facility, the evolution of radiation damage microstructures over a larger dose range, and under certain temperatures could be followed dynamically, which enabled them to draw conclusions as to trends on Cr effects, radiation temperature effects, and dose effects.

Although in-situ experiments can be a good representation (as described previously) of the bulk, parallel ex-situ experiments were of particular importance as well.

Since one of the most important limitations for in-situ thin foil experiments is that during irradiation the foil surface acts as a strong sink for mobile point-defects so that the damage development may be different compared with that in the bulk materials used in ex-situ irradiation experiments **[64]**.

The materials investigated in these in-situ experiments included ultra-high purity Fe and Fe-Cr (5, 8, 11 wt%) polycrystalline alloys and Fe-Cr (9,18wt%) single crystal alloys **[26]**. Thin foils were prepared by electro-polishing and irradiated with 100-150 keV Fe⁺ and Xe⁺ ion at room temperature, 300°C and 500°C in the Argonne NEM-

Tandem Facility [101]. The microscope was operated at 200keV. Detailed experimental conditions can be found in [26, 101].

1.4.1 Damage Evolution at RT and 300°C

At Doses Less Than 2×10^{18} ions m^{-2} (1dpa)

At RT, Yao et al. [26] found that for all the materials and irradiation conditions, small dislocation loops with diameter of 1-5nm first appeared at doses of about 10^{16} to 10^{17} ions m^{-2} . These threshold doses are found to be 2-3 orders of magnitude higher than in several other metals irradiated under similar conditions. At 300°C and RT, it was also observed that loops in all the materials appeared suddenly at doses less than about 2×10^{18} ions m^{-2} (~1dpa) and then did not grow. Since substantial cascade overlap occurs at doses greater than about 10^{16} ions m^{-2} , Jenkins et al. [101] concluded that in Fe and Fe-Cr alloys, loops are not formed directly by the collapse of individual cascades, which is consistent with the mechanism suggested by Robertson et al. [132], that visible loops are only seen at higher doses when cascade overlap happens.

At RT and 300°C, Yao et al. [26] found that both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were present, and at lower doses, $\langle 100 \rangle$ loops were in a majority (50-60% in Fe-Cr, over 80% in Fe). Compared with Fe-Cr alloys, a higher proportion of $\langle 100 \rangle$ loops was likely to be observed in Fe. Dynamic observations have shown that more loops are lost during irradiation of Fe than of Fe-Cr alloys by the possible mechanism of gliding to the specimen surface. The influence of surface image forces may influence the morphology of loops as well. In UHP Fe, it was found that the number of loops retained in the specimen was strongly dependent on the foil orientation: loop densities were lowest in $\{111\}$ foils and highest in $\{110\}$ foils. In Fe-Cr alloys, due to the pinning effect of Cr atoms, less loop loss was observed.

In all the materials at both 300°C and RT, during and after the ion irradiation, loop motion which was termed ‘hopping’ was observed by Yao et al. [26]. The hopping took the form of one-dimensional jumps from one position to another over distances of more than 10nm within 1/30 s (the video frame rate). The direction of the hopping was always parallel to the projection of a $\langle 111 \rangle$ direction, which is consistent with the Burgers vector analysis that all the hopping loops have $\frac{1}{2}\langle 111 \rangle$ Burgers vector. No loop hopping in $\langle 100 \rangle$ directions was observed. It was also observed that both loop loss and hopping were reduced in Fe-Cr alloys, which again suggests that Cr atoms have a pinning effect to reduce the loop mobility.

During the in-situ experiments, it was not possible to determine the nature of the loops. Yao et al. [26] attempted to determine the loop nature by ex-situ experiments carried out on specimens irradiated with 30 keV Ga⁺ ions at RT. The results indicated that at least some loops in thin areas of the foil were found to be vacancy type by first black-white contrast analysis. But it was difficult to give a quantitative result.

At Doses Greater Than 2×10^{18} ions m⁻² (1dpa)

At doses greater than 2×10^{18} ions m⁻² (1dpa), Hernandez-Mayoral et al. [102] found that complex microstructures developed in the thick regions of foils. At RT, in UHP Fe, loop strings consisting of several small $\frac{1}{2}\langle 111 \rangle$ loops were observed. At doses above about 5×10^{18} ions m⁻², loop strings started to coalesce and formed loops of size greater than 10nm (**see Fig.1.4.1.1**).

As the irradiation progressed, loops continued to grow by mechanisms which might involve both climb and glide. Loop shrinkage was also observed, which may be evidence of the existence of vacancy clusters.

At 300°C, in UHP Fe (6.5dpa), larger finger-shaped loops were observed which were developed by the growth and coalescence of smaller loops (**see Fig.1.4.1.2**).

It was found that all the larger loops had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, and they were all interstitial in nature. In Fe8Cr, the damage development was similar to UHP Fe, while the finger shaped loops were less developed and not well aligned as in UHP Fe.

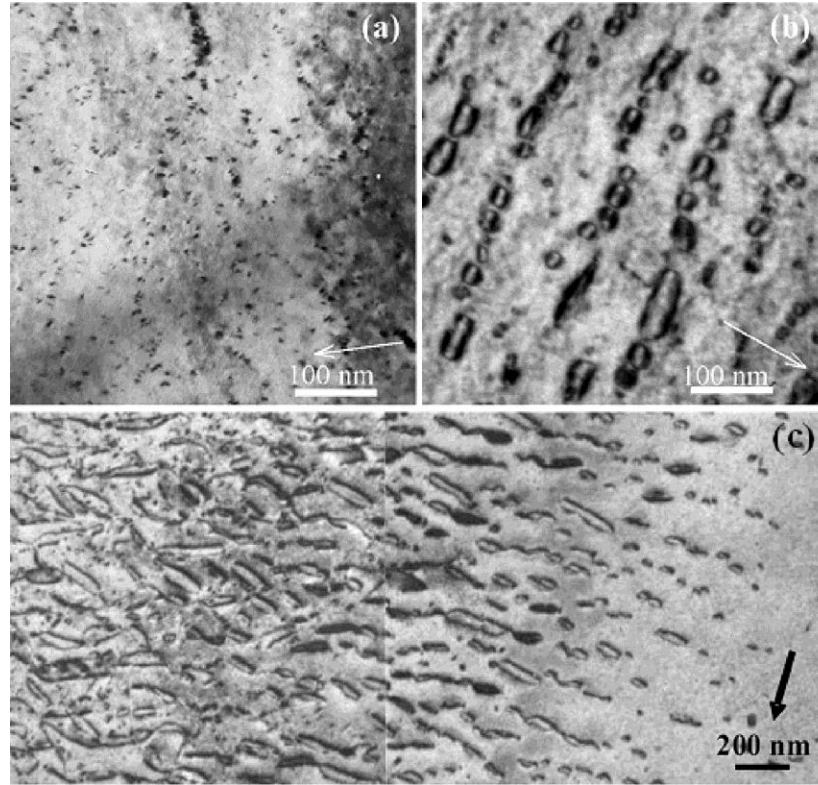


Fig.1.4.1.1. Damage development in UHP Fe irradiated with 150 keV Fe^+ ions at 300°C seen in an ex situ experiment. BFK images taken with $\mathbf{g} = \bar{1}\bar{1}0$ at doses (a) $2 \times 10^{18} \text{ ions m}^{-2}$ (~1.3 dpa); (b) $10^{19} \text{ ions m}^{-2}$ (~6.5 dpa); (c) $2 \times 10^{19} \text{ ions m}^{-2}$ (~13 dpa). Images (a) and (c) were obtained with the foil oriented near (111) and image (b) was taken near the [001] pole [102].

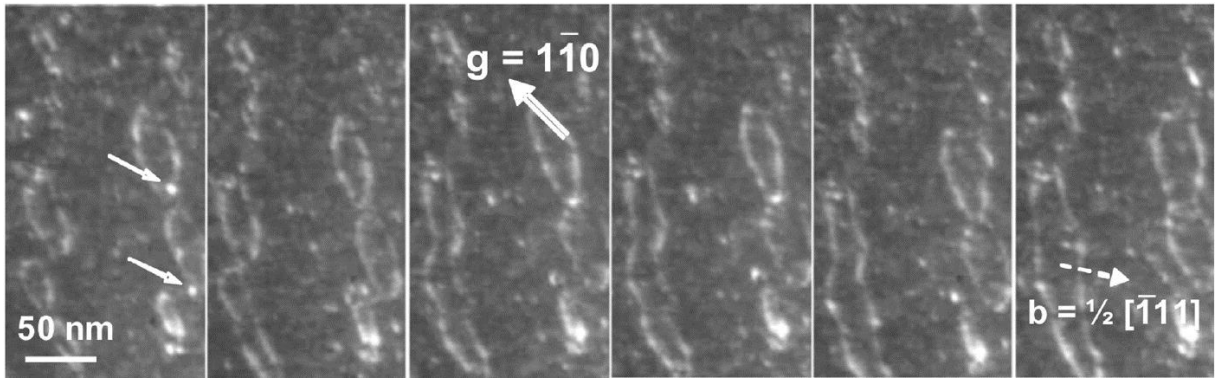


Fig.1.4.1.2. A series of frames grabbed from a video recording, showing loop growth and coalescence in UHP Fe irradiated with 150 keV Fe^+ ions at 300°C over a dose range from about $10^{19} \text{ ions m}^{-2}$ (~6.5 dpa) to about $1.6 \times 10^{19} \text{ ions m}^{-2}$ (~10 dpa). Weak-beam micrographs taken with $g = 1\bar{1}0$. The projection of the Burgers vector of the loops is shown [102].

1.4.2 Damage Development at 400°C to 500°C

Yao et al. [34] carried out further in-situ thin foil irradiations at 400°C to 500°C. They found that at 400°C, the damage structures at lower doses were similar to that seen in 300°C experiments, but loops were on a small scale of less than 5nm and most of the loops were very mobile. As the irradiation progressed, loop coalescences were observed. Small strings of $\frac{1}{2}\langle 111 \rangle$ loops were seen to coalesce into a larger $\frac{1}{2}\langle 111 \rangle$ loop. Both $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ loops were observed. The number density of $\langle 100 \rangle$ loops was larger, but they were much smaller than $\frac{1}{2}\langle 111 \rangle$ loops on average, which indicated that most of the point defects were still contained in $\frac{1}{2}\langle 111 \rangle$ loops. At 450°C, except for the development of larger-scale microstructures, other phenomena were quite similar to that at 400°C. In all the materials, the majority of the loops had Burgers vector of type $\langle 100 \rangle$ with rectilinear shape; a few chains of $\frac{1}{2}\langle 111 \rangle$ loops were also observed. At 465°C, mobile small $\frac{1}{2}\langle 111 \rangle$ loops were seen to be subsumed by a big $\langle 100 \rangle$ loop, and caused the $\langle 100 \rangle$ loop to grow, which indicated that all the loops were of the same nature. Further nature analysis proved that they were interstitial loops.

At 500°C, damage evolution in UHP Fe was found to be quite different compared with lower temperatures. Only $\langle 100 \rangle$ loops were observed at this temperature. At a dose of 10^{19} ions m^{-2} (6.5dpa), loops formed fairly regular arrays and reached the sizes of several hundred nanometres (see Fig.1.4.2.1). Inside-outside analysis showed that these big $\langle 100 \rangle$ loops were interstitial in nature.

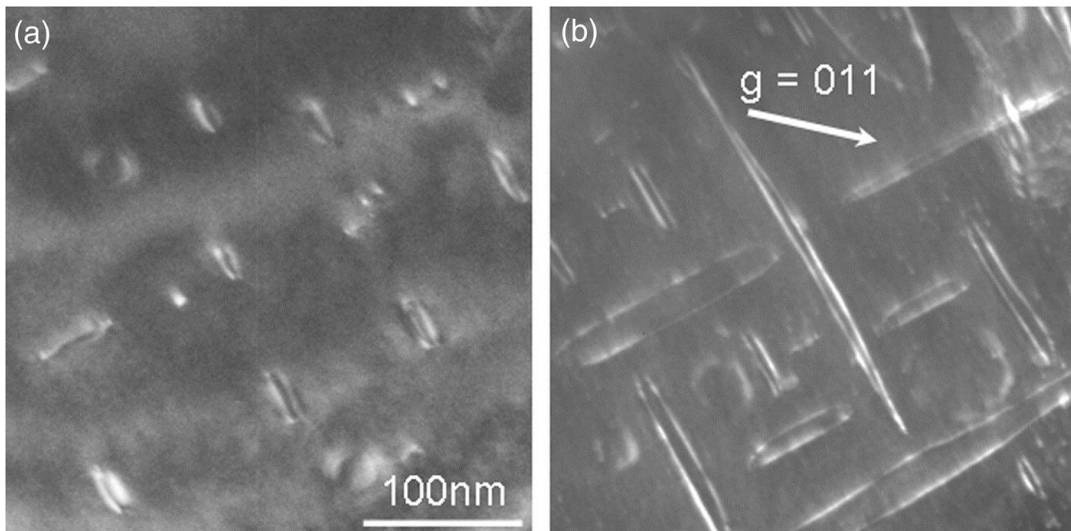


Fig.1.4.2.1. Formation of $\langle 100 \rangle$ loops in an $[001]$ foil at 500°C: (a) at a dose of 3×10^{18} ions m^{-2} ; (b) the same area at a dose of 1×10^{19} ions m^{-2} [34].

For Fe8Cr specimens irradiated at 500°C, the majority of the loops were found to be $\langle 100 \rangle$ loops, only a few loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vector were observed. The damage structures in Fe8Cr were on a much finer scale with a much higher loop number densities. Small voids were identified at 13dpa.

The transition of loop Burgers vector from $\frac{1}{2}\langle 111 \rangle$ at lower temperature to $\langle 100 \rangle$ at higher temperature which was found in the in-situ experiments, provided further evidence that $\langle 100 \rangle$ loops become increasingly dominant in Fe and Fe-Cr alloys with increasing irradiation temperature. Parallel modelling work by Dudarev et al. [88, 133] has also indicated that at temperatures $T < 350^\circ\text{C}$, prismatic pure-edge $\frac{1}{2}\langle 111 \rangle$ loops are unconditionally stable, whereas at temperatures $T > 550^\circ\text{C}$, square $\langle 100 \rangle$ pure-edge loops were unconditionally stable. Their research showed that at the

temperatures between 350°C <math>T < 550^\circ\text{C}</math>, $\frac{1}{2}\langle 111 \rangle$ loops are thermodynamically unstable and could lower their energy by change shape or transforming to $\langle 100 \rangle$ loops. Although the mechanism of loop transition is still not clear, the dynamic observations made by Yao et al. [26] indicated that small mobile $\frac{1}{2}\langle 111 \rangle$ loops could be subsumed by sessile $\langle 100 \rangle$ loops in a very short time.

1.4.3 Summary

- 1) In all materials, small (2-4nm) dislocation loops first appeared at doses between 10^{16} to 10^{17} ion m^{-2} . Loops with both Burgers vectors were observed. Evidence showed that in pure Fe, the majority of the loops were of $\langle 100 \rangle$ type [26, 101].
- 2) Loop hopping was seen to have been introduced by ion and electron beam irradiation. Some hopping loops were found to have $\frac{1}{2}\langle 111 \rangle$ Burgers vector, while no hopping $\langle 100 \rangle$ loops were seen [26, 101].
- 3) Loss of $\frac{1}{2}\langle 111 \rangle$ loops to the surface was seen frequently during the ion irradiation and under the electron beam. In Fe, it was found that the number of loops retained was strongly dependent on the foil orientation. In Fe-Cr, far less loop loss was seen, which suggest that the main effect of Cr atoms is reducing loop mobility [26, 101].
- 4) At higher doses, complex microstructures were observed in thicker area of the foils. At 300°C, in UHP Fe, the damage took the form of prismatic $\frac{1}{2}\langle 111 \rangle$ interstitial loops, and large finger loops were developed by the coalescence of smaller loops. At 500°C, in UHP Fe, larger edge loops with $\langle 100 \rangle$ Burgers vectors were observed. In Fe-8Cr, similar damage structures were developed, but with smaller loops and at a finer scale [102].
- 5) A transition of microstructure of pure Fe between irradiation temperatures of 300 to 500°C was observed. At lower temperature, it was observed that the microstructure was dominated by interstitial loops of $\frac{1}{2}\langle 111 \rangle$ types. At 450°C,

interstitial loops with $\langle 100 \rangle$ Burgers vectors dominated in most areas, while $\frac{1}{2}\langle 111 \rangle$ loops still present. At 500°C, no visible $\frac{1}{2}\langle 111 \rangle$ loops were seen, and the specimen was dominated by larger edge-on $\langle 100 \rangle$ loops. An explanation of the relative stabilities of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops as the function of irradiation temperature was given and it was believed that $\frac{1}{2}\langle 111 \rangle$ loops are thermodynamically unstable and could lower their energy by change shape or transforming to $\langle 100 \rangle$ loops at elevated temperatures [34, 101].

1.5 Comparison between Thin-foil and Bulk irradiations in Fe and Fe-Cr alloys

Advantages of in-situ TEM experiments are that this technique can not only provide direct observation of cascade damage evolution, but also allow the irradiation temperature and dose effects to be studied in a continuous and systematic way using the same sample with the same irradiation conditions. However disadvantages of this technique are that, since the sample has to be very thin (less than 200nm), the foil surfaces can act as strong sinks for mobile point defects, and the surface image forces may affect the microstructure evolution of radiation damage.

In some cases, in-situ thin-foil irradiations produce similar microstructural evolution as in the bulk samples, but in other cases, thin-foil irradiation can lead to very different microstructural changes. Therefore the similarity of damage evolution observed from in-situ irradiations between that from the bulk samples is one of the greatest concerns.

Damage Microstructures

The thin-foil and bulk experiments for self-ion irradiated Fe and Fe-Cr alloys share many similarities. In low dose bulk experiments, when cascades were well separated, it was observed that these isolated cascades couldn't collapse to vacancy loops in Fe

under self-ion irradiations. Visible loops were only observed at cascade overlap doses. This observation is consistent with that seen in thin-foil experiments in both Fe and Fe-Cr alloys.

In bulk experiments loop sizes increased with both dose and temperatures, which were similar to those found in the thin-foil experiments.

For thin-foil experiments in Fe, at doses around 10^{18} ions m^{-2} , loop strings composed by small $\frac{1}{2}\langle 111 \rangle$ loops were seen. As the dose increased, at doses around 10^{19} ions m^{-2} , large finger-shape loops with the size of hundreds of nm were produced by the coalescence of big interstitial loops. At irradiation temperatures above 450°C , regular arrays of large $\langle 100 \rangle$ loops were seen. However in bulk experiments, none of the structures described above were seen.

Cr Effects

In thin-foil experiments, observations showed that both loop hopping and loss were restricted in Fe-Cr alloys compared with that in pure Fe. Also it was found that the loop densities in Fe and Fe-Cr alloys were similar at 300°C , and both the average and maximum loop sizes decreased with increasing Cr concentration.

In bulk experiments, although it is impossible to observe loop hopping, loop sizes are smaller at high Cr alloys, which is consistent with thin foil experiments. Evidence also indicated that in high Cr alloys, loop densities were higher than that in low Cr alloys, which is different to what is found in thin-foil experiments. In thin-foil experiments, loop densities appeared to have no obvious dependence with Cr content. This is believed to be attributed to the frequent loop loss in thin-foil experiments.

Loop Burgers Vectors and Nature

$\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ loops were observed in both bulk and thin-foil experiments. In thin-foil experiments in Fe, a loop Burgers vectors transaction from 300°C to 500°C was observed, with $\frac{1}{2}\langle 111 \rangle$ loops dominating at lower temperatures and increasingly

$\langle 100 \rangle$ loops appearing at higher temperatures. In bulk experiments, since isolated experiments were carried out under various conditions, even for the experiments that were claimed to be done under similar conditions, different materials purities, temperature control, and irradiation facilities can also have strong effects on the results. Thus comparison between different bulk experiments is always problematic. In the observation of Burgers vector transaction in Fe, no systemic experiments were done in bulk experiments. But many isolated bulk experiments in Fe did find that at higher temperatures, more $\langle 100 \rangle$ loops were observed.

In thin-foil experiments the majority of the loops, both large loops ($>5\text{nm}$) and small loops ($\sim 2\text{nm}$) were found to be interstitial loops. Vacancy loops were only seen at very low doses (cascades overlap doses) in thin-foil experiments. Moreover, in Fe-8Cr samples, small voids with the size of 2nm were observed. In bulk experiments, although many vacancy loops were seen under neutron irradiation and very heavy ion irradiations (e.g. tungsten ion), only interstitial loops were found under self-ion irradiations, and no voids were seen in self-ion irradiations.

1.6 Aims of This Work

As described in the literature review, self-ion irradiated bulk Fe and Fe-Cr alloys have been studied by many researchers since the 1970s. But unlike the in-situ thin-foil experiments where continuous data can be achieved during only one continuous irradiation with increasing temperatures or doses, in bulk irradiations, every data point for a certain temperature or dose has to be done separately, which requires a vast investment in time and resources. Therefore, the majority of the published researches are based on few kinds of materials irradiated under a very limited range of conditions. Thus in bulk experiments, it is very hard to discern trends for such topics as Cr effects, temperature effects and dose effects.

Taking advantage of the advanced TEM resources, and over 20 years experiences in radiation damage research in Oxford Materials, along with the convenient irradiation facility in Surrey University, the experiments designed in this work intend to develop a comprehensive and systemic understanding of the radiation induced damage in bulk Fe and Fe-Cr alloys with a wide range of Cr content(0% Cr, 5%Cr, 8%Cr, 11%Cr), under a range of elevated irradiation temperatures (300°C,400°C,460°C), and different irradiation doses (2.5dpa, 7dpa) with a roughly constant irradiation dose rate. These comprehensive experiments in bulk samples will also be a good comparison with the thin-foil experiments, and will address the question as to how much of the knowledge from thin-foil experiments can be transferred to bulk samples.

Another part of the experiments in this work is designed to understand dose rate effects. Irradiation dose rate is believed to have profound effects on damage microstructure evolution in Fe and Fe-Cr alloys. In radiation damage studies, self-ion implantations are most commonly used to understand the damage evolution under nuclear fission and fusion environment (**see 1.2.1**). In order to build up the dose in short time, the irradiation dose rate in self-ion implantation are usually much higher than those in real fission and fusion reactors. Therefore, to apply the observations gained by self-ion implantations, it is essential to understand the role of dose rate. As described in the literature review, though many studies aiming to understand the dose rate effects in void swelling were done since the 1980s, no work has been done on the dose rate effects in interstitial clusters, which is particularly important in self-ion irradiated Fe and Fe-Cr alloys, since the majority (if not all) of the visible loops in self-ion irradiation are believed to be interstitial loops.

Chapter 2: Experimental Methods

In this chapter the experimental methods which have been used in this project are described in detail. The compositions of the materials used in this work are presented in **2.1**. The heavy-ion irradiation facility, the irradiation conditions applied in this work and the SRIM calculations of damage profiles are presented in **2.2** and **2.3** respectively. In **2.4**, the techniques used to prepared ferritic specimens for transmission electron microscopy (TEM) are described. The TEM techniques employed are described in **chapter 3**.

2.1 Materials

This project involved two different kinds of polycrystalline Fe and Fe-Cr materials with different purities.

Ultra-High Purity (UHP) materials

UHP polycrystalline Fe and Fe-Cr materials were produced by EFDA under contract EFDA-06-1901 [134]. **Table 2.1.1.** shows their impurity content in the delivered state.

<i>Alloy</i>	<i>C wt ppm</i>	<i>S wt ppm</i>	<i>O wt ppm</i>	<i>N wt ppm</i>	<i>P wt ppm</i>	<i>Cr</i>
Fe	3 (4)	2 (2)	5 (4)	2 (1)	<5	<2 ppm
Fe 5%Cr	3 (4)	3 (3)	4 (6)	3 (2)	<5	5.40 wt%
Fe 10%Cr	4 (4)	6 (4)	3 (4)	3 (3)	<5	10.10 wt%
Fe 14%Cr	4 (5)	6 (7)	4 (4)	5 (5)	<10	14.25 wt%

Table 2.1.1. Chemical analysis of the alloys in the as-delivered final metallurgical condition (in brackets) [134].

High Purity (HP) materials

HP polycrystalline Fe (5N purity) contained 0.01 wt% impurities including 0.013wt%C. HP polycrystalline Fe-Cr alloys (5, 8, 11 wt.% Cr) were produced by Metal Crystals and Oxides Ltd., Cambridge, from starting materials of purity 99.99+%. All HP materials were supplied as cold-rolled foil of thickness 0.1 ~ 0.3 mm.

2.2 Irradiation Experiments

All irradiations which were carried out in this work were performed at Surrey Ion Beam Centre using Fe^+ ions. The facility consists of a 2MV Tandem accelerator and a 200kV High Current implanter, which are capable of providing a wide energy range from 2keV to 4MeV with over 70 ion species available. Samples of size up to 40cm x 40cm can be implanted with wafer end stations configurable to 200mm at temperatures from -253°C to 500°C [135].

Before irradiation, samples in the form of 3mm discs (not pre-thinned TEM samples) were put into a custom-built sample holder with a capacity of 46 TEM discs, and the holder was then mounted flat to the implantation plate against the heating element in the target chamber (**see Fig.2.1.2**). The temperature of the sample holder was measured by an embedded thermocouple inside one of the sample holders (**see Fig.2.1.1**). The heating element behind the implantation plate is capable in principle of heating up to 1000 °C, although due to heat losses the maximum temperature measured on the sample holder is around 500°C.



Fig.2.2.1. *Implantation plate with thermocouple mounted within the sample holder.*



Fig.2.2.2. *TEM specimen holder with capacity of holding 46 discs, mounted on the hot stage in the target chamber.*

Irradiation Conditions

Two series of irradiation experiments were carried out in this project in order to study irradiation damage under different implantation conditions.

Series 1: Low/high dose self-ion irradiation

The parameters to be investigated in this series of experiments were irradiation temperature, irradiation dose, and composition of Cr. The irradiations were carried out at three different temperatures (300°C, 400°C, 460°C) with two different implantation doses (low-dose $\approx 1.5 \times 10^{19} \text{ Fe}^+/\text{m}^2$ and high-dose $\approx 4.5 \times 10^{19} \text{ Fe}^+/\text{m}^2$) at each temperature. The vacuum during the implantation was better than $5 \times 10^{-4} \text{ Pa}$. In order to produce a uniform damage profile (**see Fig.2.3.2**), Fe^+ ions were implanted with energies of 2MeV, 1MeV and 0.5MeV.

Series 2: Low/high dose-rate self-ion irradiation

The parameter to be investigated was irradiation dose-rate. These irradiations were carried out at 300°C with two different average implantation dose-rates: $6 \times 10^{-4} \text{ dpa/s}$ and $3 \times 10^{-5} \text{ dpa/s}$ (see the definition for 'dpa' in **2.3**). The implantation dose for both implantations was $0.38 \times 10^{19} \text{ Fe}^+/\text{m}^2$. Ion beam and environmental conditions for the implantation are shown in **Table 2.2.1**. The vacuum during the implantation was below $5 \times 10^{-4} \text{ Pa}$. In order to produce a uniform damage profile (**see Fig.2.2.3**), Fe^+ ions were implanted with energies of 2MeV, 1MeV and 0.5MeV. During the three energies, the beam current was carefully maintained by adjusting conditions at the source(coarse control),and by changing the size of the beam raster area (a fine control to within $0.1 \mu \text{ A/m}^2$).During the irradiation, the ion beam was focused to a diameter of 5mm and spread over the implantation area by scanning the beam. The scanning frame rate of the beam over the samples was 4kHz, which gives a displacement damage of each frame of $1.5 \times 10^{-7} \text{ dpa}$ for the high dose rate irradiation and $7.5 \times 10^{-9} \text{ dpa}$ for low dose-rate irradiation [**116**]. Both of the localised flux doses

were below the required dose for cascade overlap in Fe based alloys (about 0.01dpa) [26]. Because of the scanning beam, the localised flux dose-rate is higher than the average dose-rate given at the beginning of the paragraph. The calculation of localised flux rate can be seen in **chapter 9**.

<i>Irradiation temperature (°C)</i>	<i>Dose rate (dpa/s)</i>	<i>Energy (MeV)</i>	<i>Dose (ions/m²)</i>	<i>Dose rate (ions/m²/s)</i>	<i>Beam current (μ A/m²)</i>
300	6×10^{-4}	0.5	5×10^{17}	1.87×10^{15}	299
		1	7.5×10^{17}	2.16×10^{15}	345
		2	2.5×10^{18}	2.28×10^{15}	366
	3×10^{-5}	0.5	5×10^{17}	9.35×10^{13}	15
		1	7.5×10^{17}	1.08×10^{13}	17.3
		2	2.5×10^{18}	1.14×10^{14}	18.3

Table.2.2.1. Irradiation conditions used at Surrey Ion Beam Centre, UK [116].

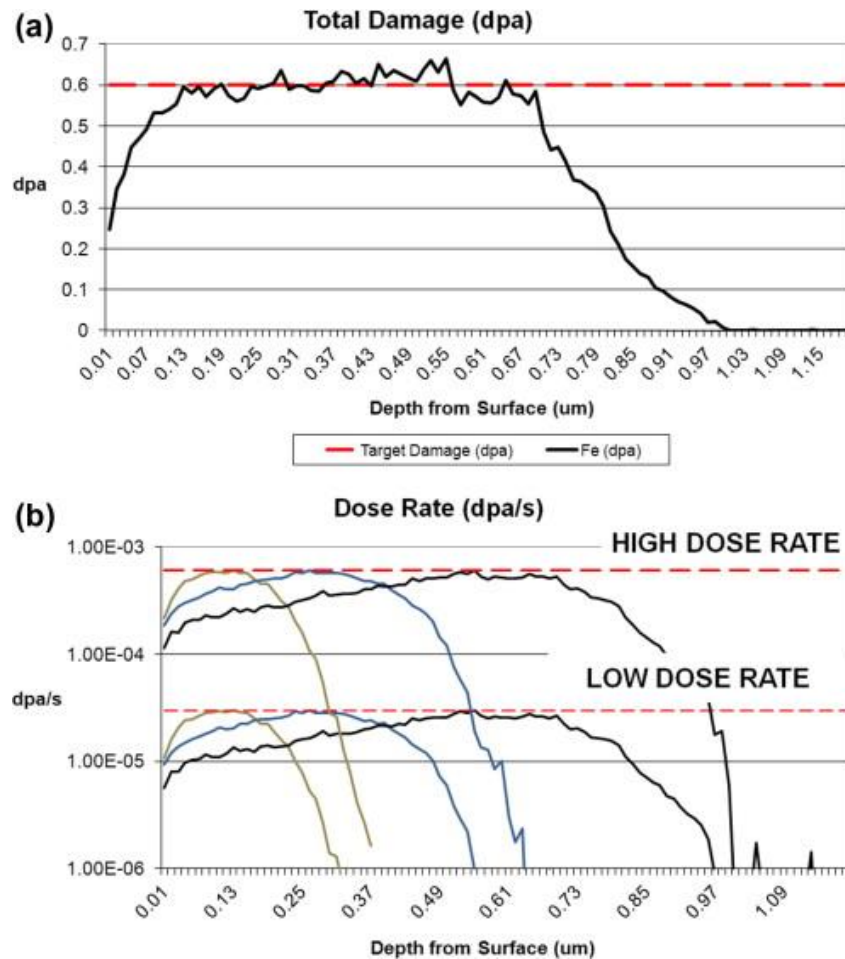


Fig.2.2.3. STRIM calculations showing displacement damage with implantation depth: (a) displacement damage level for all implantations and dose rates, and (b) damage rates for high dose rate (6×10^{-4} dpa/s) and low dose rate (5×10^{-5} dpa/s) for each implantation energy [116].

2.3 SRIM Calculations

The Stopping and Range of Ions in Matter (SRIM) programme [136] was used to simulate the radiation damage produced by Fe⁺ ions into iron and Fe-Cr alloys. A recommended displacement energy of 40eV was applied in all the simulations [137].

Fig.2.3.1 is the SRIM calculation for 2MeV Fe⁺ ion irradiation of an Fe target, which shows how the damage dose within the sample varied according to the sample depth.

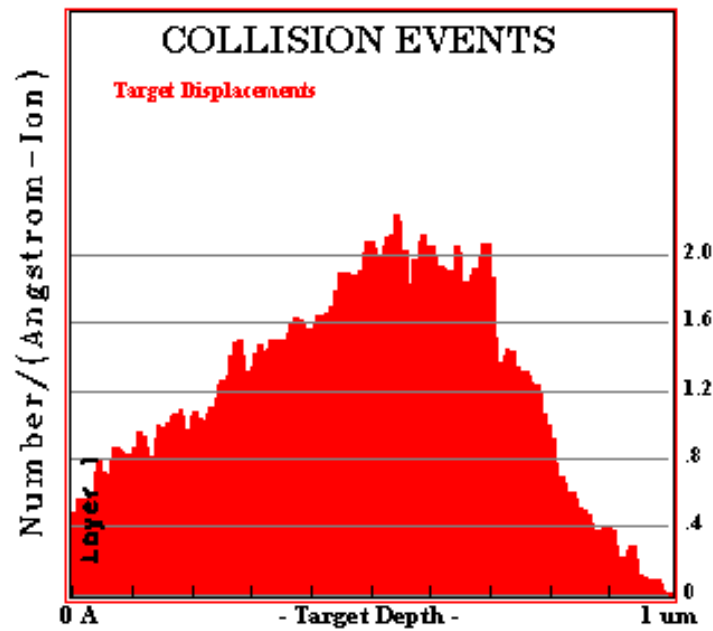


Fig.2.3.1. Fe target displacements after a single energy implantation of 1000 collision events.

In radiation damage studies, in order to compare the radiation conditions prevailing in different accelerators and reactors using different particles, the implantation dose is usually measured in displacements per atom (dpa), that is the number of times on average which each atom is displaced from its original lattice site.

With the aid of **Fig.2.3.1**, the **dpa** value of a collision event can be calculated using the equation below:

$$dpa = \frac{N_{displacements}}{N} \Phi$$

For an implantation of $1.5 \times 10^{19} \text{ Fe}^+/\text{m}^2$, Φ is $1.5 \times 10^{19} \text{ Fe}^+/\text{m}^2$. Fe and Fe-Cr alloys has a bcc lattice structure with a lattice constant of $a=0.2863\text{nm}$, and with $Z=2$ atoms per unit cell. So the density of Fe atoms per m^3 is $N=ZV/a^3 \approx 8.88 \times 10^{28} \text{ atoms} \cdot \text{m}^{-3}$.

Referring to **Fig.2.3.1**, $N_{\text{displacements}}$ at the peak damage layer is $\sim 2 \text{ \AA}^{-1} \cdot \text{ion}^{-1}$.

Therefore the peak damage level for this implantation is about 3.4 dpa.

In **Fig.2.3.1**, it can be seen that the damage peak of about 3.4 dpa is buried under the surface of the irradiated sample at a depth of 500nm to 600nm. This would make the sample preparation very difficult as in order to view the microstructure at the peak damage layer, a controlled amount of material would have to be removed from the surface. In an attempt to achieve this, a controlled electro-polishing method was developed, which will be described in **2.4.2**. However it was found that the reproducibility of this method was not sufficient in single-energy implants to guarantee success. This polishing method is estimated to remove material at a rate of $100 \pm 50 \text{ nm/s}$, so there would be a significant possibility that the 100nm-wide peak damage layer could be missed during polishing.

In practice this problem was overcome by employing a modified irradiation method, whereby triple-energy irradiation with energies of 2MeV, 1 MeV and 0.5MeV were carried out successively, to provide a much flatter damage profile and a broader peak damage layer of 600nm, as shown in **Fig.2.3.2**.

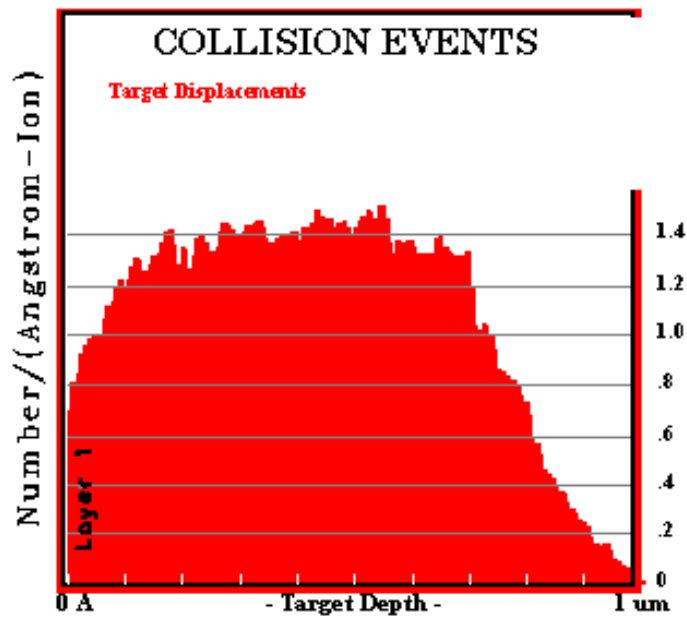


Fig.2.3.2. Fe target displacements after a triple energy implantation of 1000 collision events.

Since all the experiments in this project were designed to access the peak damage layer, the **dpa** value cited in this thesis all refer to the maximum dose given by SRIM. Using the same equation, all the peak dpa values of implantation experiments in this project were found as follows:

	<i>Implantation dose(Fe^+/m^2)</i>	<i>dpa</i>
Low dose implantation	1.5×10^{19}	2.55
High dose implantation	4.5×10^{19}	5.10
Dose rate implantation	0.38×10^{19}	0.65

2.4 Specimen Preparation

Specimen preparation is the most crucial part of the diffraction contrast experiments carried out in this project because the operation of the microscope and the quality of micrographs are strongly dependent on the quality of the TEM specimens. A good ferrite TEM specimen should be thin (around 100 nm), with a small hole in the middle of the disc. Moreover, there should be an edge area with flat surface around the small hole. Since ferritic specimens oxidise quickly in air, it is recommended that the prepared specimen be introduced to the microscope immediately after thinning and later stored in water-free ethanol if needed for further analysis.

2.4.1. Pre-irradiation preparation

This process is a combination of mechanical polishing, heat treatment and electro-polishing. The aim is to produce thin 3mm discs with a deformation-free flat surface and low dislocation density in the matrix.

Mechanical polishing

The received materials were first mechanical thinned by a series of silicon-carbide papers to around 100 μ m (grit sizes: 20 μ m, 15 μ m, 8 μ m). For magnetic materials such as Fe and Fe-Cr alloys, the final specimen must be of minimum mass since the magnetic field of the specimen will strongly disturb the electron beam. For Fe specimen the thickness must be below 100 μ m, while for Fe-Cr specimen the thickness should be below 120 μ m. After mechanical polishing, the materials were punched into standard 3mm TEM discs using a 'SPI Disc Punch'.

Annealing

These 3mm discs were then moved into a vacuum furnace with a vacuum of about 10^{-6} mbar. In order to produce a dislocation free microstructure with large grain size ($\sim 50\mu$ m), specimens were annealed at 800°C for 8h, followed by slow cooling for 20

hours. The final microstructure had a grain size of $\sim 50\mu\text{m}$ with very few dislocations (see Fig.2.4.1.1.).

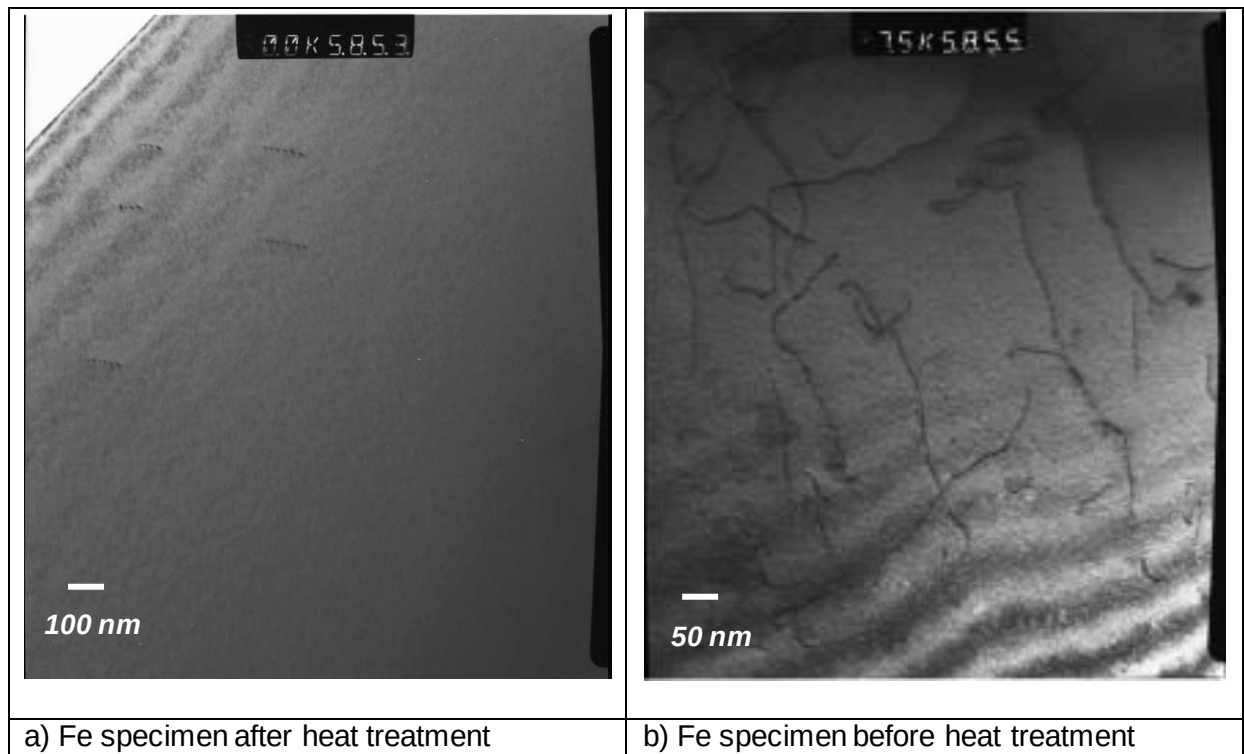


Fig.2.4.1.1. The effects of heat treatment.

Pre-irradiation electro-polishing (beaker-bath polishing method)

After heat treatment the discs were electro-polished for a few seconds in a bath. This process removes any surface deformation caused by mechanical polishing and oxidation layer on the surface. The electro-polishing conditions are described in detail in 2.4.2.

2.4.2. Post-irradiation preparation

As described in 2.2, the aim of this project was to view the radiation damage at the peak damage layer. This was achieved by a well-controlled electro-polishing removal of the top surface (about 300nm), followed by jet electro-polishing to achieve electron transparency [138].

Top surface removal (beaker-bath polishing method)

As explained in 2.2, the irradiation dose within an implanted sample varies according to the sample depth, so before the preparation of electron-transparent TEM foils a top surface layer of 400nm with a margin of allowable error of $\pm 200\text{nm}$ needs to be removed. This was done by using an accurately controlled, bath electro-polishing technique.

Fig.2.4.2.1. shows the electro-polishing set-up. The specimen was held by platinum-tipped tweezers (platinum was chosen due to its compatibility with the electrolyte) and connected to the anode with the irradiated surface facing downwards.

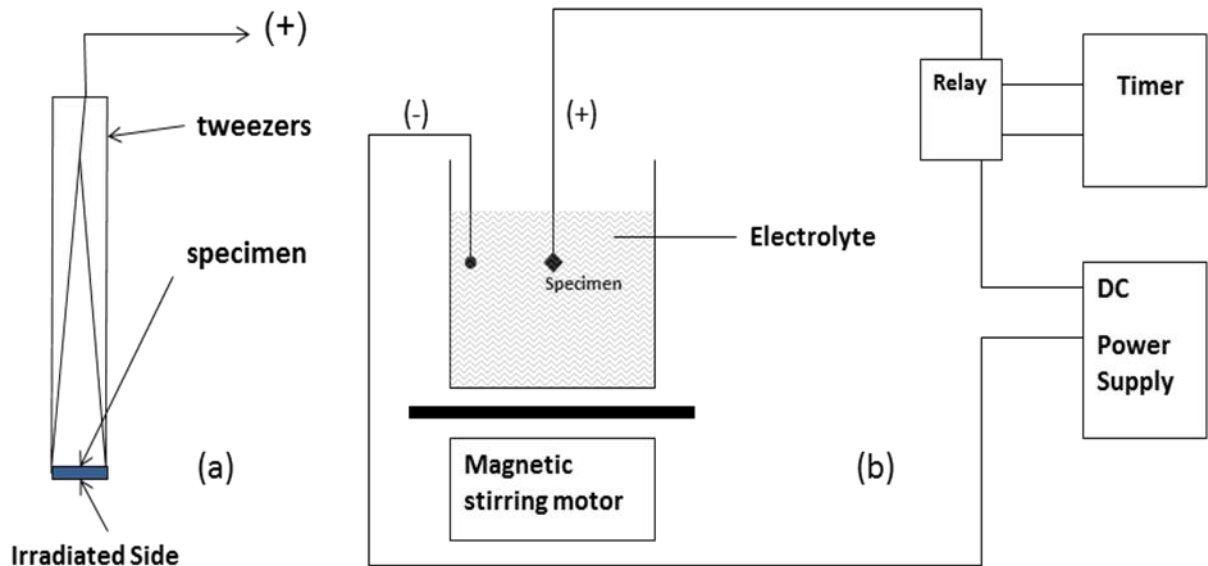


Fig.2.4.2.1. (a) tweezers and specimen; (b) diagram for bath electro-polishing technique.

The electrolyte used for all the ferrite materials in this project comprised 5% HClO_4 and 95% methanol. Before polishing, the electrolyte was cooled to $-30\text{ }^\circ\text{C}$ by addition of liquid nitrogen. The polishing time was controlled by a relay timer with an accuracy of 0.01s. Acceptable electro-polishing conditions should provide a uniform polishing rate, and a clean surface without pitting or etching. The electro-polishing conditions are described in detail in a following section below. It was found that for Fe and Fe-Cr alloys used in this project, good polishing could be achieved with 30V at a temperature of -30°C and a current of about 2A [138].

Beaker-bath polishing rate

In order to estimate the electro-polishing rate, both micro-indentation and FIB-indentation methods were applied by a former Dphil student in Oxford Materials Department (Sen Xu, graduated in 2009). Based on his measurements using the same electrolyte and polishing conditions as described above, the typical polishing rate for Fe-Cr alloys is 100 ± 50 nm per second. The polishing condition may vary slightly according to different Cr contents. Detailed explanation can be found in Sen's thesis [8, 138].

Jet electro-polishing

The final step of sample preparation was jet electro-polishing. This was achieved by a TenuPol 5 Jet -electropolisher using an electrolyte containing 5% HClO_4 and 95%methanol.The TenuPol jet–electropolisher is widely used in TEM specimen preparation. During polishing the jets normally squirt electrolyte onto both sides of the sample, and form an electron transparent area in the middle of the sample. However, for the irradiated specimen used in this project single side polishing was required, since the peak damage region was on one side of the disc. This was achieved by masking the irradiated side of the disc using Lacomite varnish [138].

The steps are illustrated in **Fig.2.4.2.2.**

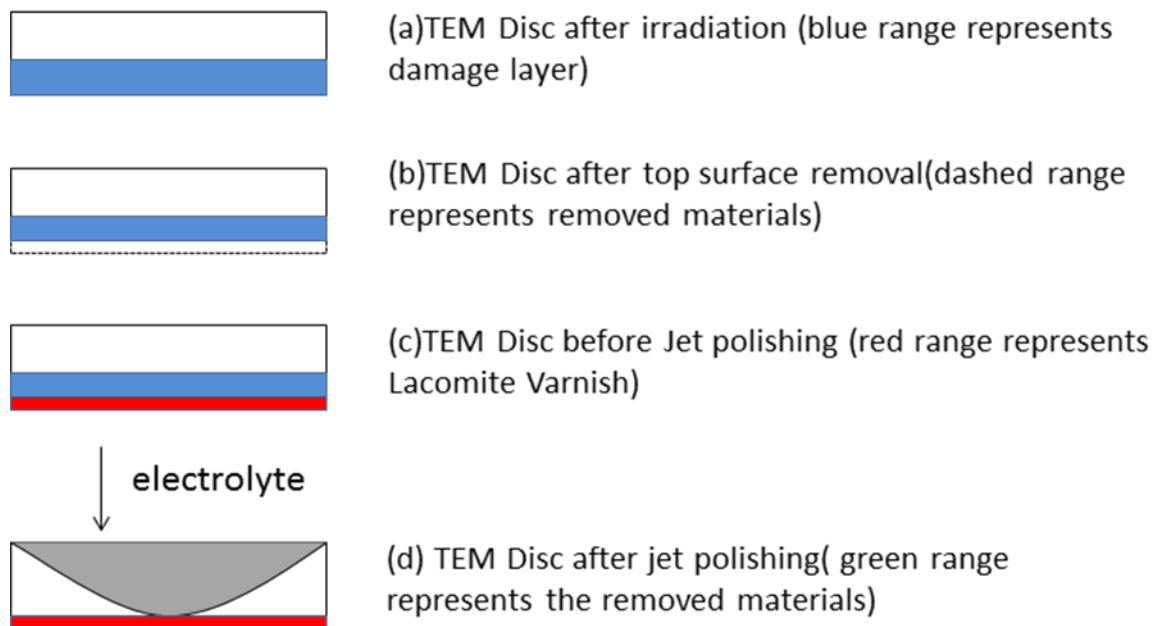


Fig.2.4.2.2. Jet electro-polishing treatment of implantation damaged TEM discs.

Electro-polishing conditions

Fig.2.4.2.3. shows schematically the effect of the current density on the quality of electro-polished materials. In the electro-polishing process, the current is determined by the voltage, temperature and electrolyte concentration. Too high or too low a current will severely damage the sample surface. Insufficient current causes etching along grain boundaries or at surfaces, while excessive current causes large numbers of small holes (diameter: 4nm-10nm) on the sample [139]. In the jet-polishing process, the polishing temperature and electrolyte are chosen, and an appropriate voltage found. In ferrite materials, the window of suitable polishing conditions is small. This region may vary from 2V to 0.2V, depending on the composition of the material, according to the hand-drawn current-voltage curves. Moreover, the purity of the materials also tends to have a strong impact on the voltage needed for good polishing.

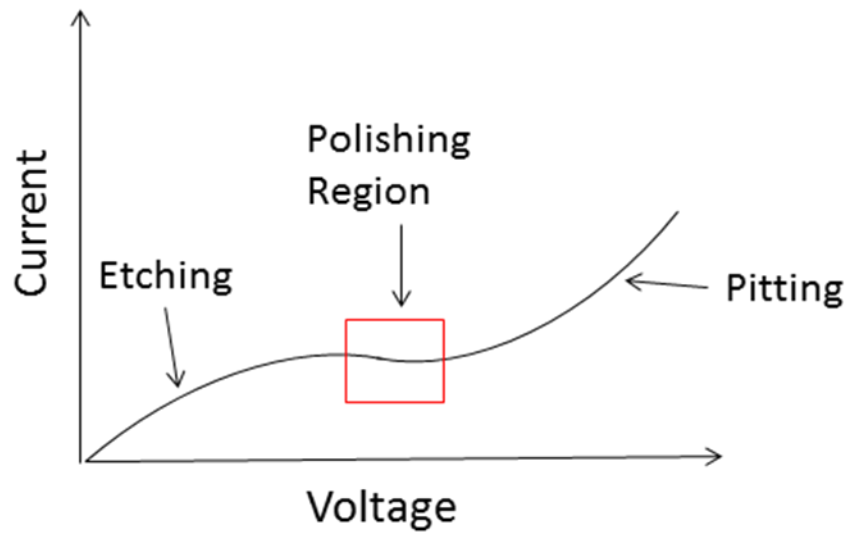


Fig.2.4.2.3. Voltage-Current curve for electro-polishing [139].

The specific Jet-polishing conditions for the materials used in this project are given in **Table 2.4.2.1.**

Materials	Electrolyte	Temp., °C	Volts
HP Fe	5% HClO_4 , 95%methanol	-40	21.5
HP Fe-5Cr, Fe-8Cr,Fe-11Cr	5% HClO_4 , 95%methanol	-40	20.5
UHP Fe, Fe-5Cr	5% HClO_4 , 95%methanol	-45	21
UHP Fe-10Cr	5% HClO_4 , 95%methanol	-45	21.3
UHP Fe-14Cr	5% HClO_4 , 95%methanol	-45	20.5
HC Fe	5% HClO_4 , 95%methanol	-45	21
HC Fe-5Cr, Fe-12Cr	5% HClO_4 , 95%methanol	-45	21.5

Table 2.4.2.1. Electrolytes and conditions used for jet-polishing (HC stands for high carbon).

2.4.3. Post-polishing cleaning and specimen storage

Because of the use of Lacomite varnish and the strong causticity of HClO_4 , post-polishing cleaning is essential. After electro-polishing, specimens were washed immediately in cold methanol (-30°C) and then moved first to Lacomite remover for 20 minutes, and then to methanol for 15 minutes. Due to oxidation of Fe and Fe-Cr alloys, it is suggested that specimens should be introduced into the TEM immediately after cleaning. Specimens which needed to be stored were kept in water-free ethanol where they could remain in good condition for up to two weeks.

Chapter 3: TEM Techniques and Data Analysis

In this chapter, the transmission electron microscopy techniques applied in this work to collect and analyse the radiation damage structure in Fe and Fe-Cr alloys will be described in detail. The theoretical basis of electron diffractions will not be presented.

In radiation damage studies, the dislocation loops are measured by determining the distribution of damage image widths, loop number density, loop Burgers vector, loop nature and loop habit plane. In this work, the loop habit plane is taken as the upward drawn normal of the dislocation loop, and the loop number densities are counted by the loops appeared within a certain volume. In order to make a comparable analysis between the micrographs taken from different samples or different areas within a sample, some standard conditions have to be applied. In diffraction contrast experiments, the reciprocal lattice vector of the operating reflection is $\mathbf{g}$, $\mathbf{Z}$ is defined as the upward drawn image plane normal, and $\mathbf{w}_g$ is the Bragg deviation parameter which is equal to $\mathbf{s}\epsilon_g$ ($\mathbf{s}$ is the deviation of the incident electron beam from the Bragg angle, ϵ_g is the extinction length of the operating reflection).

Throughout this work all quantitative comparisons are made between micrographs taken under the same grain orientations and diffraction conditions.

Bright-field kinematical conditions are often used when imaging small loops. In practice, the image is firstly formed under a two-beam condition with a small positive deviation parameter ($\mathbf{s}_g > 0$), then the foil is tilted away from its Bragg condition to avoid the dynamical contrast effects. The contrast showed in such conditions is believed to be more confined to the real physical size of the loops. If different diffraction conditions are applied in this work, they will be defined before being used. Detailed contrast mechanisms and experimental techniques can be seen in [64].

In 3.1, the method to determine the foil thickness which is essential for the loop number densities measurement is described. In 3.2 and 3.3, the methods employed

to determine the loop number density and image size distributions are described. In **3.4**, **3.5** and **3.6**, the method for the determination of loop Burgers vector, loop habit plane and loop nature are described.

3.1 Foil Thickness Measurement

For the accurate measurement of loop number densities, the data of foil thickness is essential. In the experiments described in this work, convergent beam electron diffraction (CBED) patterns are used to interpret the thickness of a selected area in a foil. In practice, a two-beam condition is firstly set up using one strongly excited **hkl** reflection. Then if the beam is focused on the selected area and the microscope switched to diffraction mode, the CBED disks will contain parallel fringes, as shown in **Fig.3.1.1**.

In order to calculate the thickness, an equation derived from the two beam dynamical theory of electron diffraction is needed.

$$\left(\frac{S_i}{n_i}\right)^2 = \frac{-1}{\xi_g^2} \left(\frac{1}{n_i}\right)^2 + \frac{1}{t^2}, \quad [140]$$

where t is the foil thickness, ξ_g is the extinction distance of the excited reflection **g**, n_i is an integer. S_i expresses the deviation of the i -th minimum from the exact Bragg position and is defined by the equation given below.

$$S_i = \frac{\lambda}{d^2} \left(\frac{L_i}{L_0}\right),$$

where λ is the wavelength of the incident beam and d is the spacing of the reflection planes.

The data needed in advance are the spacing of the reflection planes (d) and the wavelength of the incident beam (λ). The distances to be measured are L_0 which is the distance from the centre of the diffracted beam profile to the centre of the

transmitted beam, and L_1, L_2, L_3 from the centre of the diffracted beam profile to each of the successive minima (**see Fig.3.1.1**).

The next step is to plot $(S_i/n_i)^2$ vs. $(1/n_i)^2$ for different values of n_i (i.e. $n_i=1, n_i=2 \dots$) to find out which value of n_i results in a straight line. Then the foil thickness is given by the intercept of the straight line with the $(S_i/n_i)^2$ axis. It is said that this method of thickness determination has an accuracy of better than $\pm 2\%$ [8, 140, 141].

Table 3.1.1-3.1.2 shows the data needed beforehand and the obtained values after the application of the equation. It was found that in this case $n_1=4$ produced the linear plot shown in **Fig.3.1.2**. The value of the intercept of the line with the vertical axis is 0.0003 nm^{-2} . So the foil has a thickness of $58 \pm 20 \text{ nm}$, where the error comes from the uncertainty in the measurements of fringe separations.

i	L_i/L_0	λ	d	S_i
1	0.119	0.00251nm	0.101nm	0.0293
2	0.239	0.00251nm	0.101nm	0.0588
3	0.354	0.00251nm	0.101nm	0.0871

Table.3.1.1. CBED Data for thickness determination.

		$i=1$	$i=2$	$i=3$
$n_1=1$	$(S_i/n_i)^2$ vs. $(1/n_i)^2$	$8.58*10^{-4}$ vs.1.00	$8.64*10^{-4}$ vs.0.25	$8.43*10^{-4}$ vs.0.111
$n_1=2$	$(S_i/n_i)^2$ vs. $(1/n_i)^2$	$2.15*10^{-4}$ vs.0.25	$3.84*10^{-4}$ vs.0.111	$4.74*10^{-4}$ vs. $6.25*10^{-2}$
$n_1=3$	$(S_i/n_i)^2$ vs. $(1/n_i)^2$	$9.54*10^{-5}$ vs. 0.111	$2.16*10^{-4}$ vs. $6.25*10^{-2}$	$3.03*10^{-4}$ vs. $4*10^{-2}$
$n_1=4$	$(S_i/n_i)^2$ vs. $(1/n_i)^2$	$5.37*10^{-5}$ vs. $6.25*10^{-2}$	$1.38*10^{-4}$ vs. $4*10^{-2}$	$2.11*10^{-4}$ vs. $2.78*10^{-2}$
$n_1=5$	$(S_i/n_i)^2$ vs. $(1/n_i)^2$	$3.43*10^{-5}$ Vs.0.04	$9.60*10^{-5}$ Vs. $2.78*10^{-2}$	$1.55*10^{-4}$ Vs.0.02

Table.3.1.2. CBED Data for thickness determination.

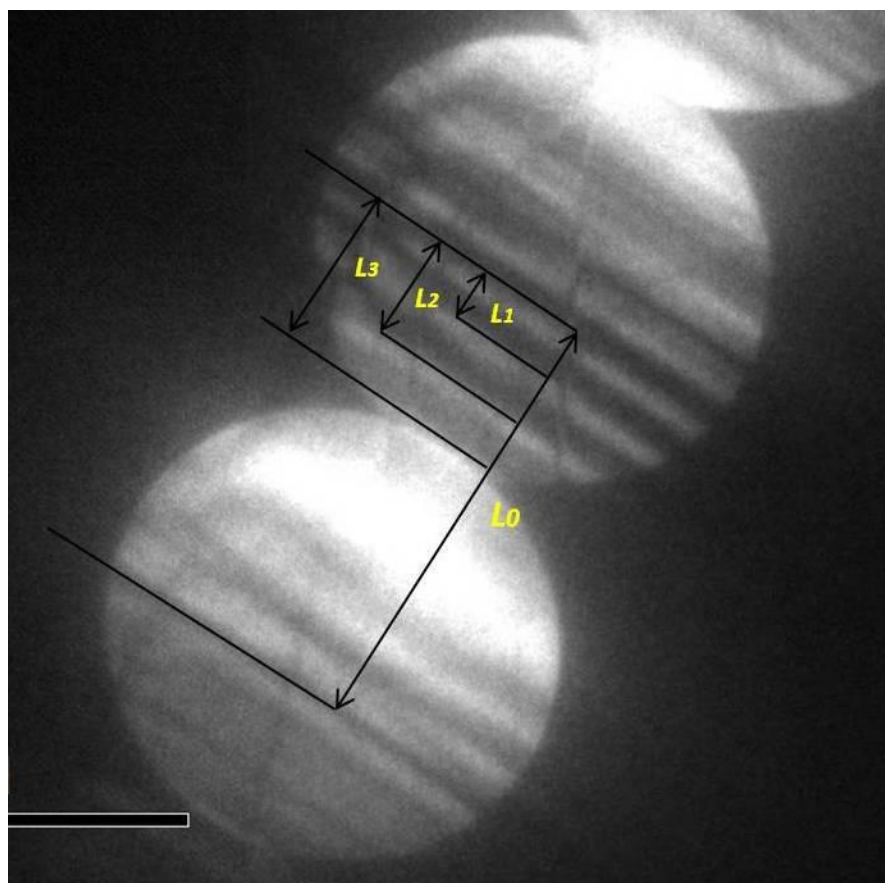


Fig.3.1.1. Thickness fringes in a CBED pattern from pure Fe taken under two-beam conditions with (1-10) strongly excited.

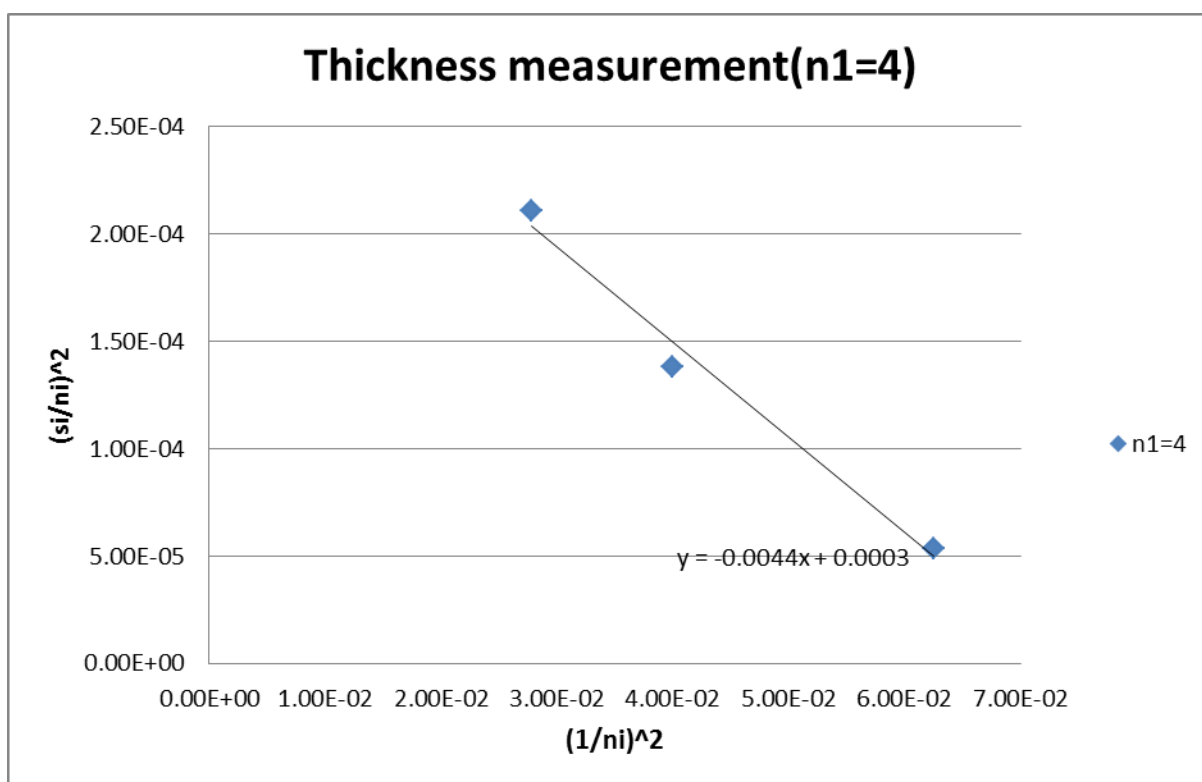


Fig.3.1.2. Graph used to determine the foil thickness.

3.2 Determination of Loop Density

The loop density is defined as the number of visible loops within a given volume. The majority of the loop number density measurements presented in this work are carried out from micrographs recorded in $g=-110$ near the $[001]$ zone axis. All the micrographs were taken under bright-field kinematical conditions. When interpreting TEM micrographs, it is important to be aware that the TEM presents us with 2D images of 3D samples, thus these micrographs have no depth sensitivity and they are only projection images which cannot represent the real size and position of the features inside the sample [141].

In counting loop number densities, all the data were collected from at least two selected areas of one or two samples containing about 800 loops. Since the loop number density is a measurement of the number densities within a volume, a good knowledge of the foil thickness is essential. As described in 3.1, in this work the foil thickness was measured by the convergent beam method or by counting thickness fringes from the foil edge. For the samples described in this work, the thickness varied from 120nm to 150nm.

3.3 Determination of Loop Size Distribution

For loop size measurements, most of the measurements were also carried out from micrographs recorded in $g=-110$ near the $[001]$ zone axis. For each analysis, at least 300 loops were measured. When measuring loop image size, it is essential to bear in mind that the loops appear in the micrographs are projection images, and their image size varies with loop morphology, loop depth, foil thickness and deviation parameter. The only way to accurately measure the real size of an individual loop is by doing a series of tilting experiments such as those described by Jenkins et al., where images of the same defect were taken under a systematic variation of $|s_g|$ [38, 140].

But due to the large number of loops and difficulties on sample tilting, it is impossible to carry out such measurements for all the loops in the foils in this study. In practice, loop image size distributions are generally obtained by measuring image sizes on a single micrograph with a given $\mathbf{g}$ vector near a certain pole, and the parameters are kept the same through comparisons between different samples or different areas in the same sample. In 2006, simulations by Zhou et al. [142] also showed that size distributions measured in a single micrograph are not an accurate representation of the true defect size distribution, since the image size is about equally likely to be smaller or larger compared to the real defect size. However, Zhou also claimed that the average image size of the population is likely to be a reasonably good approximation to the average true loop size of the population.

3.4 Determination of Loop Burgers Vectors

TEM micrographs are 2D images of 3D samples, when interpreting samples with high loop densities, as most of the cases in this work, these projection images cause great trouble with loop Burgers vector analysis and nature characterisations. This is because these analyses require not only large-angle tilting (10° - 30°), which leads to transformations in the projected position and shape of the loops, but also require comparisons of the same features taken from different projected positions.

Sometimes, because of the high loop density and movements of the loop projected images, it was impossible to complete a full analysis on certain samples. The experiments were made particularly difficult by problems inherent in tilting ferromagnetic specimens which perturbs the electron beam.

The difficulties due to image shifts at different projections can be alleviated by modern computer technology which allows images taken at different projection positions to be switched very quickly using picture viewing software on a computer

screen, thus revealing the trends of movements of the projected images. In this way, even samples with very high loop densities can be studied with confidence.

In bcc Fe and other ferritic alloys dislocation have two types of Burgers vector:

$\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$. The Burgers vectors **b** is determined by using the standard **$\mathbf{g} \cdot \mathbf{b} = 0$** invisibility criterion. The standard procedure of Burgers vector determination is as follows:

- 1) Find one zone axis and determine one **g** vector.
- 2) Record the bright field and dark field images of a selected area which contains the targeted dislocations.
- 3) Tilt the specimen to a different **g**, whilst keeping the selected area in the same position within the screen, then record the bright and dark field images.
- 4) Repeat process ③, if necessary.

In some cases, in order to get the accurate Burgers vectors, contrast experiments at more than one zone axis is required. The detailed theory of this technique can be found in [64].

Fig.3.4.1 is an example of a typical contrast experiment carried out in an Fe specimen irradiated at 300°C to a dose of 2.5dpa. The five micrographs were taken near the [001] and [011] foil orientation under bright-field kinematical conditions using the five different diffraction vectors available. A **$|\mathbf{g} \cdot \mathbf{b}|$** table was given for these diffraction conditions. Since **$\mathbf{g} \cdot \mathbf{b} = 0$** will appear as very vague or even zero image contrast, the type of Burgers vectors can be determined by observing the appearance of loops in each micrograph. In **Fig.3.4.1**, the dotted circle marks the loops which have **$\mathbf{g} \cdot \mathbf{b} = 0$** .

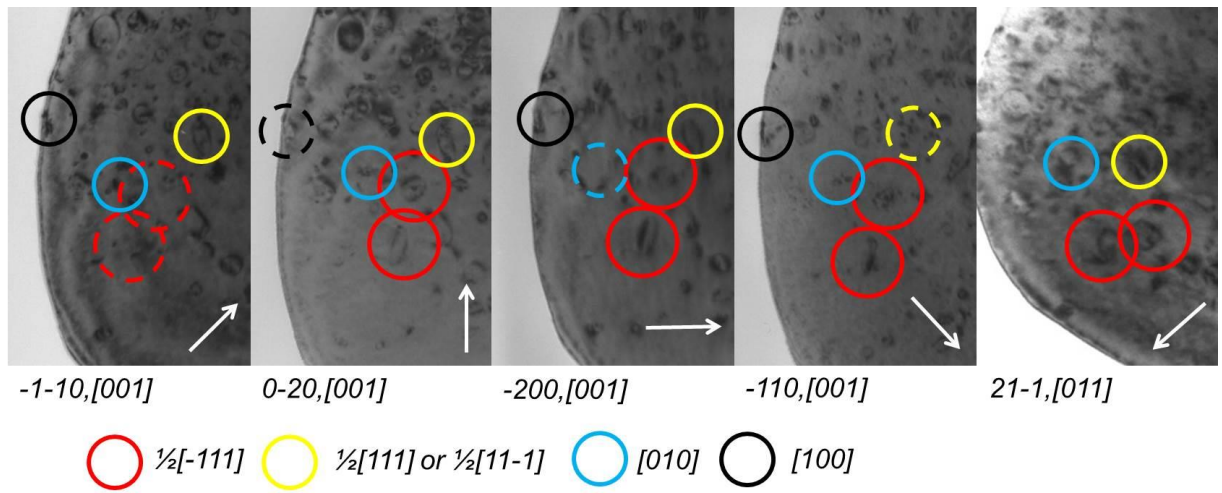


Fig.3.4.1. *g.b* contrast experiments of an Fe specimen irradiated at 300°C.

g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(110)$	1	0	0	1	1	1	0
$\pm(020)$	1	1	1	1	0	2	0
$\pm(200)$	1	1	1	1	2	0	0
$\pm(-110)$	0	1	1	0	1	1	0
$\pm(21-1)$	1	1	0	2	2	1	1

Table.3.4.1. *g.b* table for $[001]$, $[011]$ zone axes.

3.5 Determination of Loop Nature

In radiation damage studies, the loop nature (interstitial or vacancy) is one of the most important data to understand the complex defect mechanisms such as the formation of the loops, interaction between migrating point-defects with the loops etc. Also it is probably one of the most difficult pieces of information to get. There are normally several methods to determine the nature of dislocation loops, and each has its advantages and limitations. Detailed introduction of the methods can be found in the book 'Characterization of Radiation Damage By Transmission Electron Microscopy' [64]. In this work, we use the inside-outside technique to study the loop nature. This method is considered as the most reliable method of determining the nature of a loop, and it is widely applied in the bigger loops which show well-defined inside-outside contrast. This method is difficult to use for loops smaller than about 10 nm in diameter, and loops smaller than 5 nm in diameter cannot be analysed.

3.5.1. Pure Edge Loops

To illustrate the principles of the inside-outside technique, we first consider a pure edge loop as an example. A definition of pure edge loop is quoted below [143]:

'Pure edge loops (prismatic dislocation loop) are closed dislocations which, in an unstressed crystal, lie in the plane normal to their Burgers vector $\mathbf{b}$. As a shear stress is applied, a prismatic loop elongates in the glide cylinder defined by $\mathbf{b}$ and the dislocation's line sense ξ . Once ξ is parallel to $\mathbf{b}$, the dislocation can cross slip and escape the glide cylinder.'

Fig.3.5.1.1. shows the definition of loop Burgers vector. The direction of loop normal $\mathbf{n}$ and the electron beam direction $\mathbf{z}$ are defined as shown and should be kept consistent throughout the experiments. The loop Burgers vectors are defined by the Finish-Start/Right-hand (FS/RH) convention.

For an edge vacancy loop (see image a), $\mathbf{b}$ is antiparallel to the upward-drawn loop normal $\mathbf{n}$, and therefore: $\mathbf{b} \cdot \mathbf{n} < 0$, $\mathbf{b} \cdot \mathbf{z} > 0$.

For an edge interstitial loop (see image b), $\mathbf{b}$ is parallel to the upward-drawn loop normal $\mathbf{n}$, and therefore: $\mathbf{b} \cdot \mathbf{n} > 0$, $\mathbf{b} \cdot \mathbf{z} < 0$.

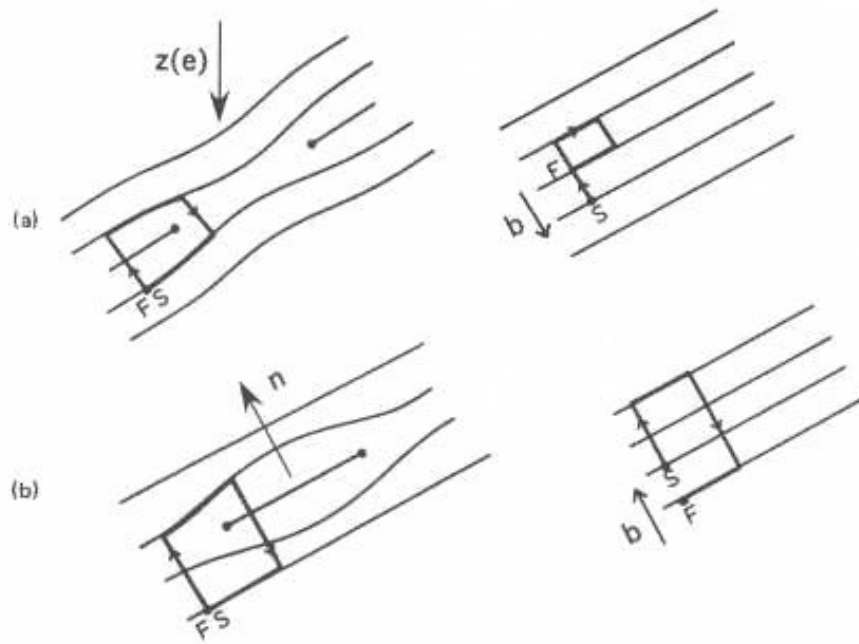


Fig.3.5.1.1. Definition of Burgers vectors [64].

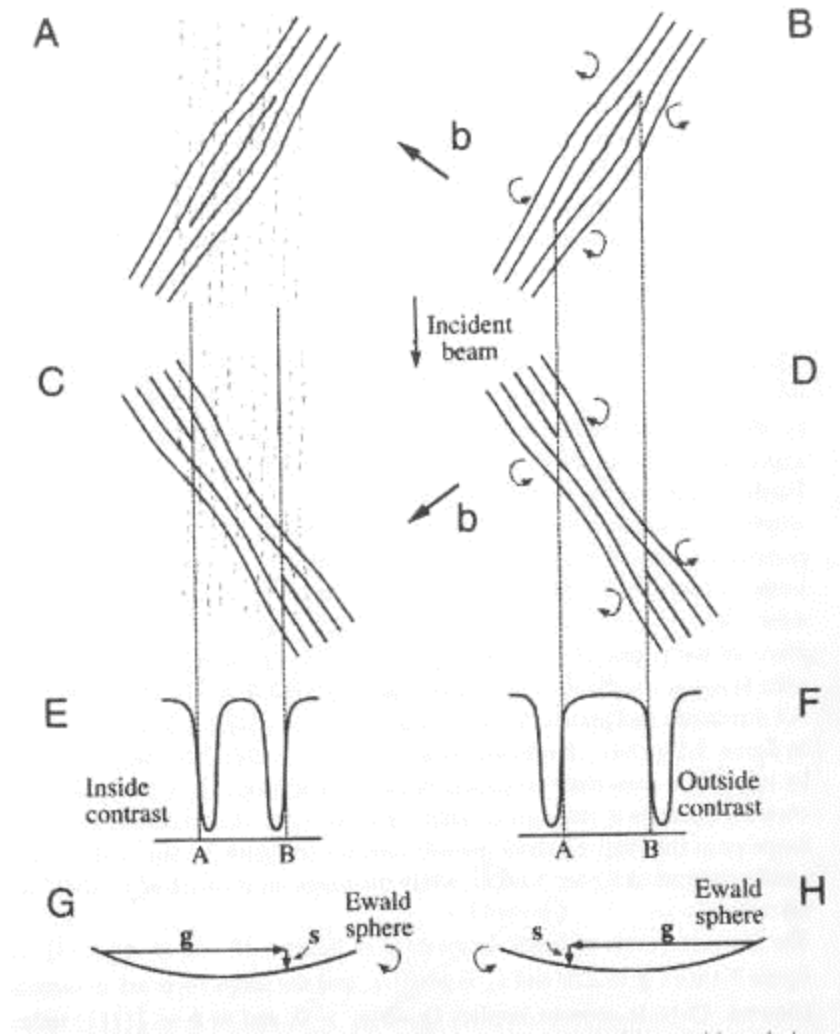


Fig.3.5.1.2. The principle of the inside-outside technique for loop nature determination [64].

Fig.3.5.1.2. illustrates the physical principle of the inside-outside technique. As can be seen from the image, inclined interstitial(A,B) and vacancy(C,D) edge loops are imaged under either bright-field kinematical or dark-field weak-beam conditions with $S_g > 0$ using the diffraction vector $\mathbf{g}$ shown in the image. The curved arrows show the sense of lattice rotations caused by the loop strain fields that try to bring the reflecting planes back towards the Bragg condition. These regions diffract strongly, therefore the peak of the image lies either inside or outside the projection of the loop, thus generating inside or outside contrast. Formally, inside contrast arises when $(\mathbf{g} \cdot \mathbf{b})S_g < 0$ and outside when $(\mathbf{g} \cdot \mathbf{b})S_g > 0$.

So, in order to determine the nature of a pure edge loop, the following steps were followed:

- a) Find the loop Burgers vector by using the ' $\mathbf{g} \cdot \mathbf{b} = 0$ ' invisibility criterion' that described in **3.4**.
- b) Find the sign of $\mathbf{b}$ by determining the diffraction vector in which the loop shows inside or outside contrast. This step is achieved through imaging the loops using both positive and negative $\mathbf{g}$, whilst keeping the S_g positive and approximately constant.
- c) Find the loop Burgers vector by using the formulas: $\mathbf{n} \cdot \mathbf{b} > 0$ (for interstitial loop), and $\mathbf{n} \cdot \mathbf{b} < 0$ (for vacancy loop).

In practice, a large amount of loops were found to be non-edge type, thus the use of the technique is not straightforward, and will be fully outlined in the following section.

3.5.2. Non-edge Loops

The application of inside-outside technique to non-edge loops was first introduced by Maher and Eyre [144]. They found that in certain 'safe' orientations, non-edge loops will show the same inside-outside contrast as that of a pure edge loop of the same Burgers vector, while for the unsafe orientations, the loops will show reverse contrast behaviour. Detailed principles for the definition of the 'safe' orientation can be found in [30, 64]. In practice, if the angle between the loop Burgers vector and the beam direction is less than $(90^\circ - \Phi)$, the loop is considered 'safe'. Φ is the maximum angle between the loop nucleation plane and the Burgers vector.

Take an Fe sample irradiated at 300°C to the dose of 2.5dpa as an example. In **Fig.3.5.2.1**, a nature analysis using inside-outside technique for a non-edge loop is shown. From **Fig.3.4.1**, it can be seen that the circled loop in **Fig.3.5.2.1** had Burgers vector of $\pm \frac{1}{2} [\bar{1}11]$. The loop was first imaged near [001] zone axis, using

the diffraction vector $\mathbf{g}=200$, then the specimen was tilted to $[013]$, where the images of $\pm\mathbf{g}$ were taken. In Fe, if we consider the Eyre-Bullough mechanism (see 1.2), which is the loops are nucleated on $\{110\}$ planes, then the maximum angle between an $\frac{1}{2}\langle 111 \rangle$ loop and its possible nucleation planes is 35° , which means that the foil orientations that make an angle less than 55° with an $\frac{1}{2}\langle 111 \rangle$ Burgers vector are 'safe' for the loop. In Fig.3.5.2.1, in an $[013]$ foil, the angle between the beam direction and the $\frac{1}{2}[\bar{1}11]$ Burgers vector is 43° , therefore such a non-edge loop will show the same image contrast as a pure edge loop with the same Burgers vector. Using the method for a pure-edge loop, we can see the loop shows outside contrast in $\mathbf{g}=-200$, and inside contrast in $\mathbf{g}=200$, therefore $\mathbf{b} \cdot [\bar{2}00] > 0$, so $\mathbf{b} = +\frac{1}{2}[\bar{1}11]$, $\mathbf{b} \cdot \mathbf{n} > 0$, and the loop is interstitial in nature.

3.6 Determination of Loop Habit Plane

There is one assumption in the previous determination of the loop nature, which is the maximum angle between the loop Burgers vector $\mathbf{b}$ and the real loop normal $\mathbf{n}$. This assumption can be double checked by finding the habit plane of the loop. In practice, it is very difficult to make an accurate measurement of the inclination of a loop. But the sense of inclination can be determined by finding the sense of the rotation of the projected loop image which indicates the changes in loop normal during a large angle tilt. There are normally two ways to do so:

- a) Estimating the loop normal by observing the change in the projected direction of the loop major axis.
- b) Tilting the loop to an edge-on configuration and evaluate the $\mathbf{n}$ from the loop trace.

After taking the inside-outside images near $[013]$, the specimen was further tilted to $[011]$ zone axis, and **Fig.3.5.2.2**, illustrates the change of the loop major axes from $[001]$ towards $[011]$.

In **Fig.3.5.2.1**, as the sample was tilted from $[001]$ to $[011]$, the main axis of the circled loop was turning anticlockwise, which is consistent with a circular loop lying within the $(\bar{1}11)$ plane. The loop is therefore of pure-edge type or close to pure-edge. Thus the analysis in **3.5** is reasonable, and the loop is interstitial in nature.

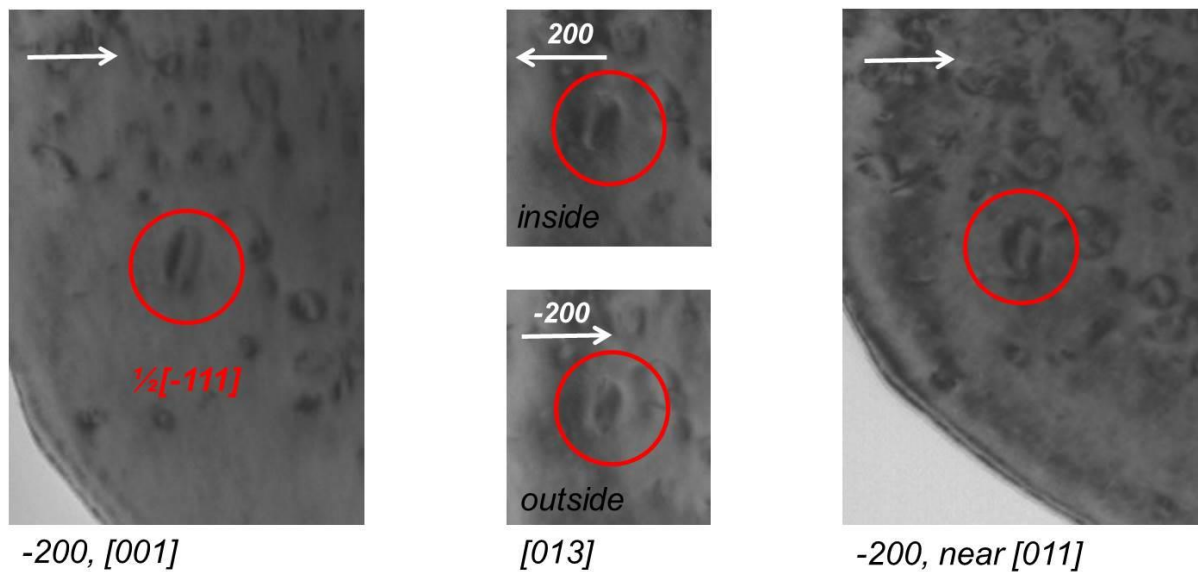


Fig.3.5.2.1. Nature analysis of an Fe sample using inside-outside technique, tilting along $g=200$ from $[001]$ to $[011]$.

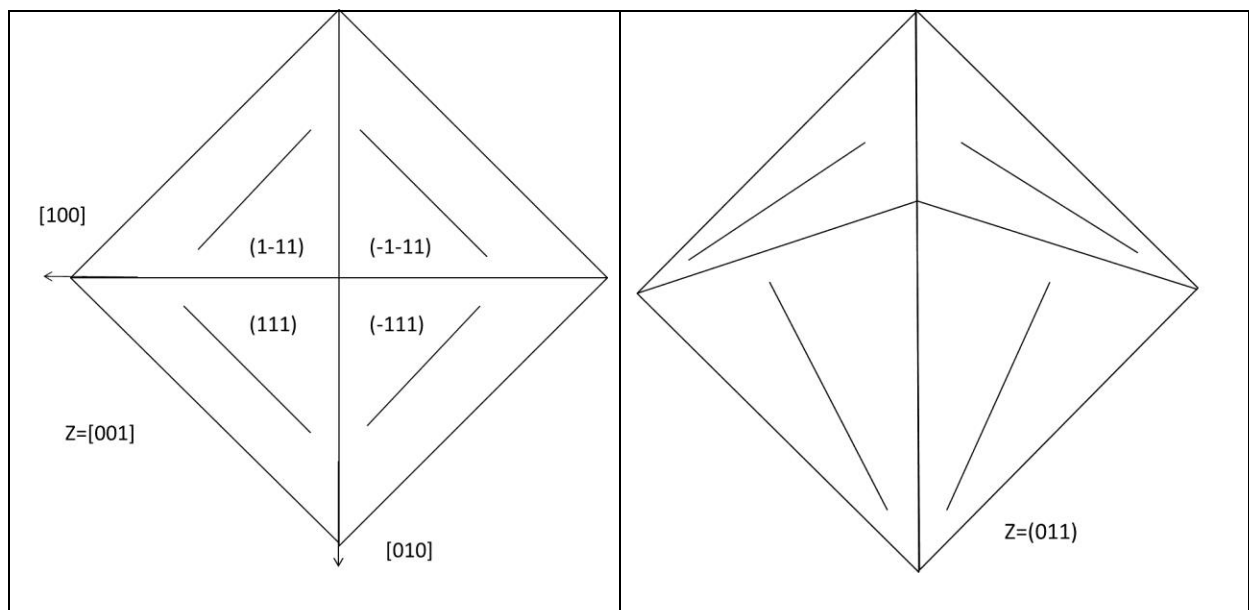


Fig.3.5.2.2. Demonstration of tilting experiments from $[001]$ to $[011]$.

Chapter 4: Radiation Damage in Fe

In this chapter, the characterisation of heavy-ion irradiated iron at elevated temperature (300 -460°C) and doses to 7dpa will be presented. The dose-rate applied in the Fe samples and the following Fe-5Cr, Fe-8Cr, Fe-11Cr samples were not constant, since it was impossible to well-control the dose-rate over such a long period irradiation, but the rates were very close to the 'high dose-rate' that has been described in 2.2. Damage produced by self-ion implantation in Fe consists of a high density of dislocation loops with sizes of several nanometres to a hundred nanometres. With increasing irradiation temperature, the loops interacted with each other to form large finger shaped loops or loop clusters of size several hundred nanometres. The number densities, sizes and Burgers vectors of the loops were analysed and compared between samples under different irradiation conditions. Nature analyses of big loops (larger than 5nm) were performed, and all the loops were found to be interstitial in nature. Through all the experiments, the defocusing method was applied to look for the existence of voids. However no voids were found in any experiments.

4.1 Irradiation Damage Microstructures

Fig.4.1.1 shows four micrographs illustrating the self-ion damage in Fe specimens irradiated from 300°C to 460°C at two implantation doses (2.5, 7dpa). The micrographs were taken in a similarly thick (80-120nm) area of the specimen. At 300°C (**Fig.4.1.1.a**), the damage microstructures took the form of small dislocation loops with size of around 2-4nm. At 400°C (**Fig.4.1.1.b.d**), dislocation loops were much bigger than those at 300°C, and more complex features such as finger-shaped loops, and loop clusters of several hundred nanometres were formed. In particular, in the specimen irradiated at 400°C to a dose of 2.5dpa (**Fig.4.1.1.b**), perpendicular

loop clusters appeared homogeneously within the foil. At 460°C (**Fig.4.1.1.c**), the damage structure consisted of isolated dislocation loops, and no large loop clusters or finger-shaped loops were observed. More micrographs and descriptions of the damage features are presented in the following paragraphs.

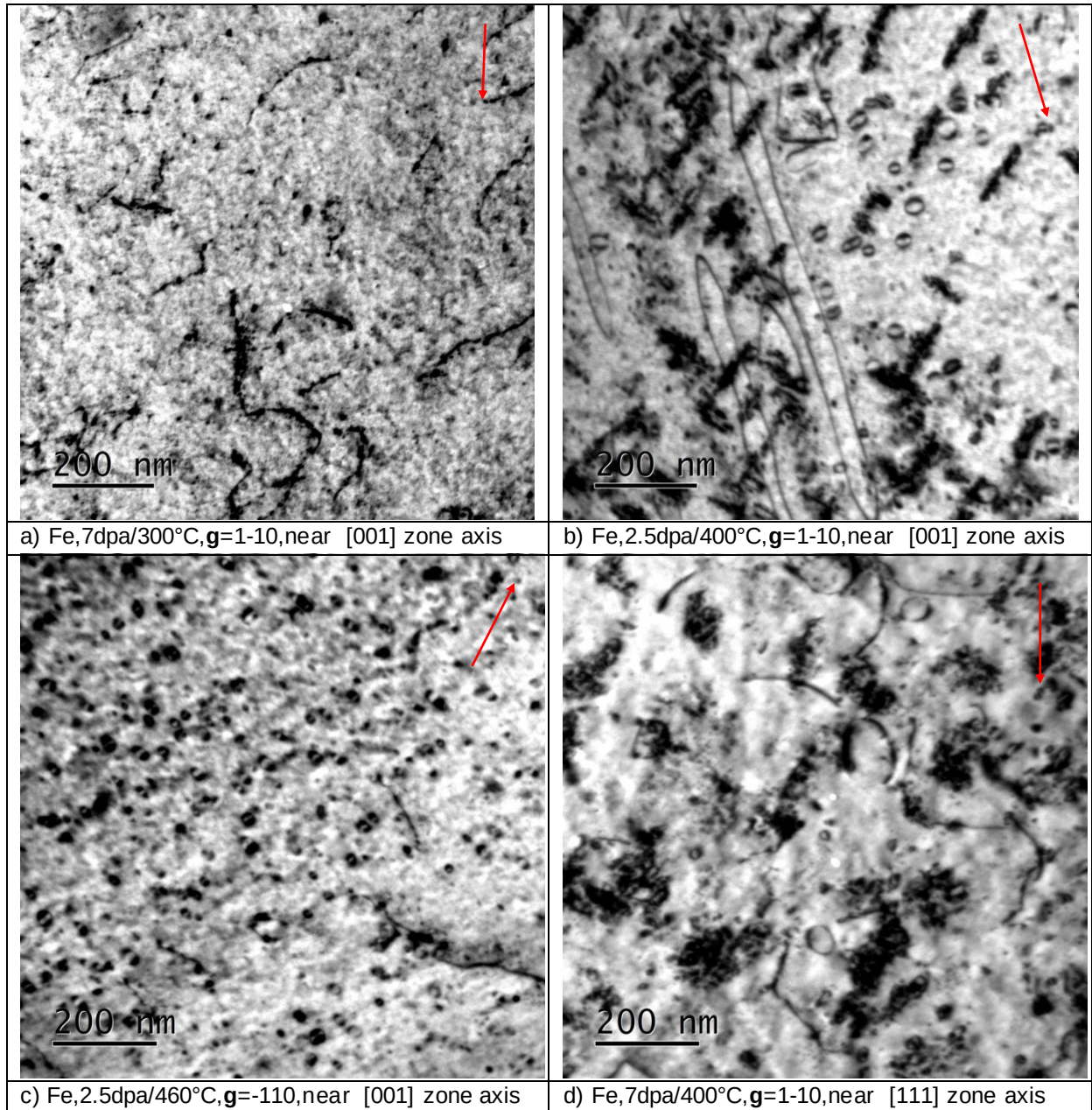


Fig.4.1.1. Micrographs of pure Fe samples irradiated at different temperatures and doses.

Fe specimen irradiated at 300°C to a dose of 7dpa

Fig.4.1.2. shows micrographs of an Fe specimen irradiated at 300°C to a dose of 7dpa. All the damage appeared to be small dislocation loops with sizes 2-10nm. The line dislocations seen in this sample were probably caused by sample preparation as they were inhomogeneous and not seen in the other samples irradiated at the same condition. Moreover, in **Fig.4.1.2**, the majority of the loops appeared near the line dislocations, and they were restricted to a narrow channel which had a projected width of 50nm on either side of the dislocation line. Burgers vector analysis also indicated that clusters of loops had the same Burgers vector. In **Fig.4.1.3**, it can be seen that the line dislocations had either $\frac{1}{2}[111]$ or $[100]$ type Burgers vectors, while 70% of the loops in the green circle had Burgers vector of $[010]$ and the rest had $[100]$. The nature of the loops could not be determined as the loops were too small.

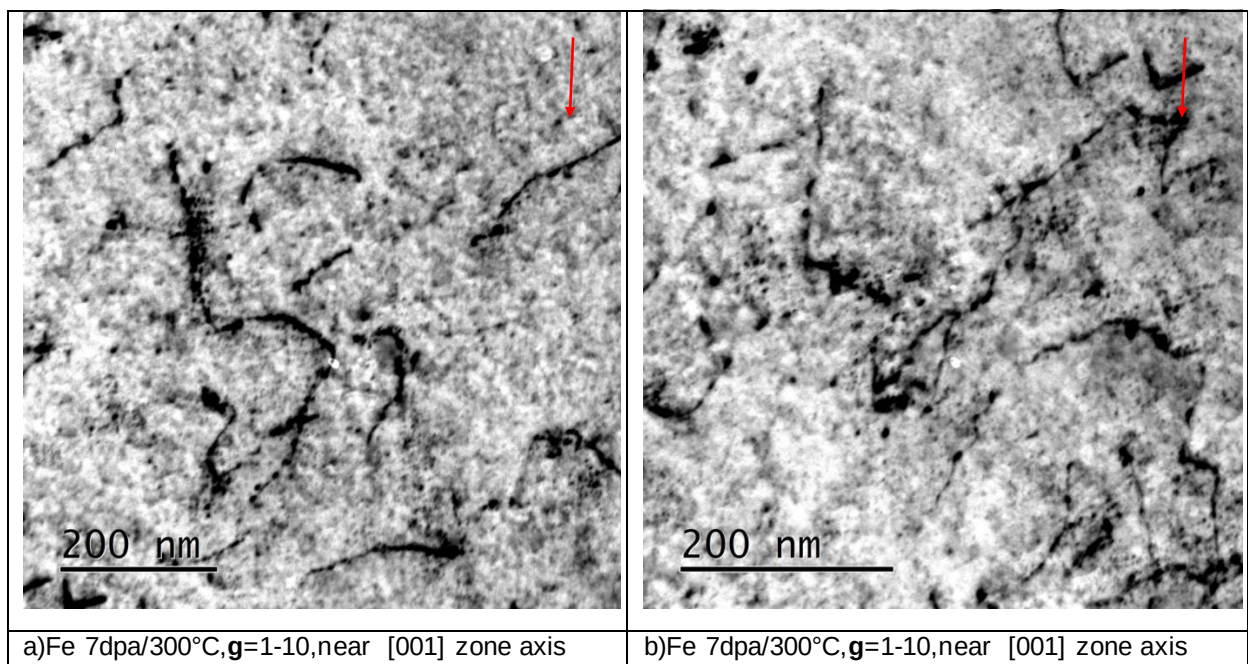


Fig.4.1.2. TEM micrographs of Fe samples irradiated at 300°C to a dose of 7dpa.

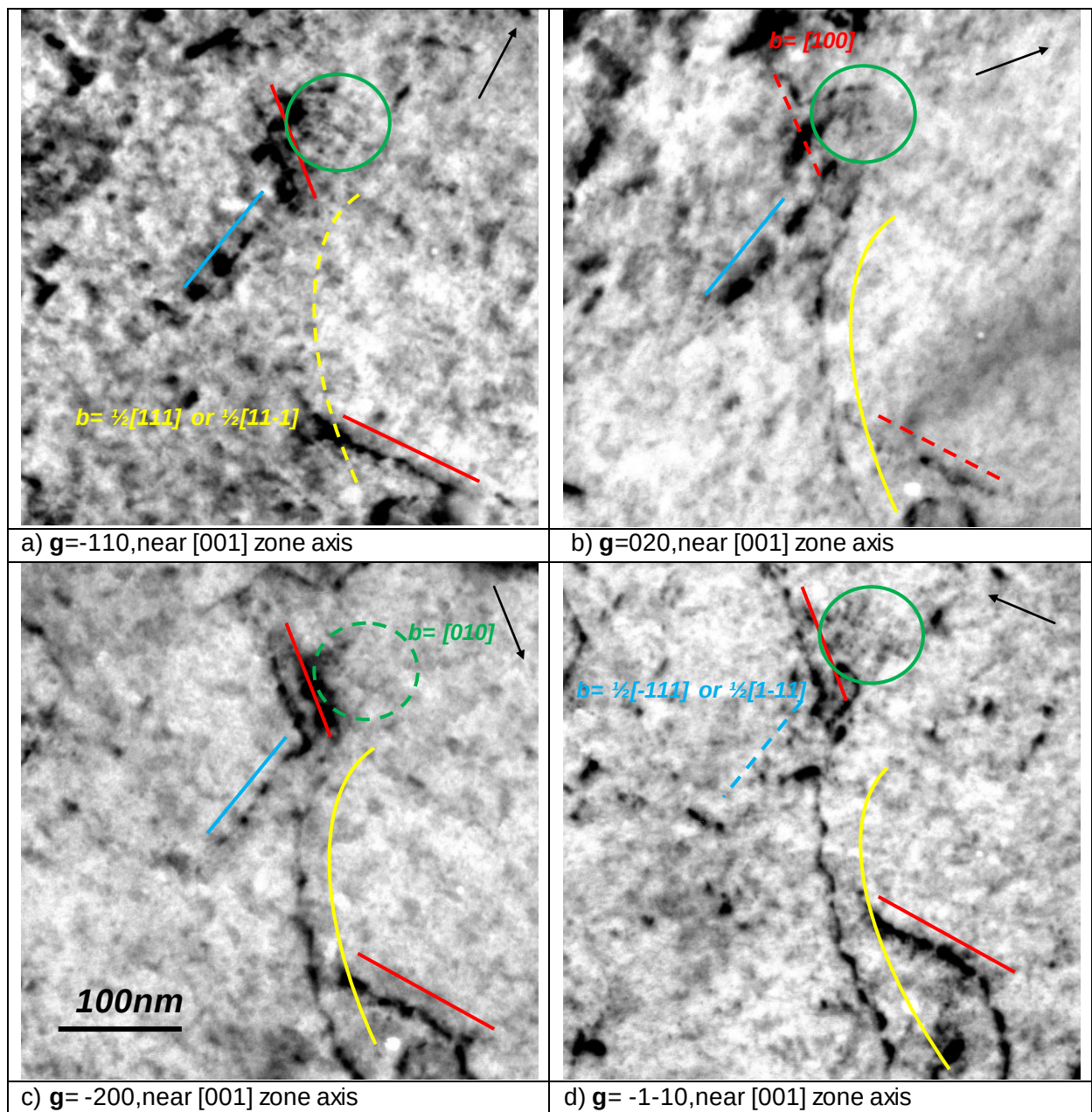


Fig.4.1.3. TEM micrographs of $g.b$ analysis for line dislocations and the associated loops in Fe samples irradiated at 300°C to a dose of 7dpa.

Fe specimen irradiated at 400°C to a dose of 2.5 dpa

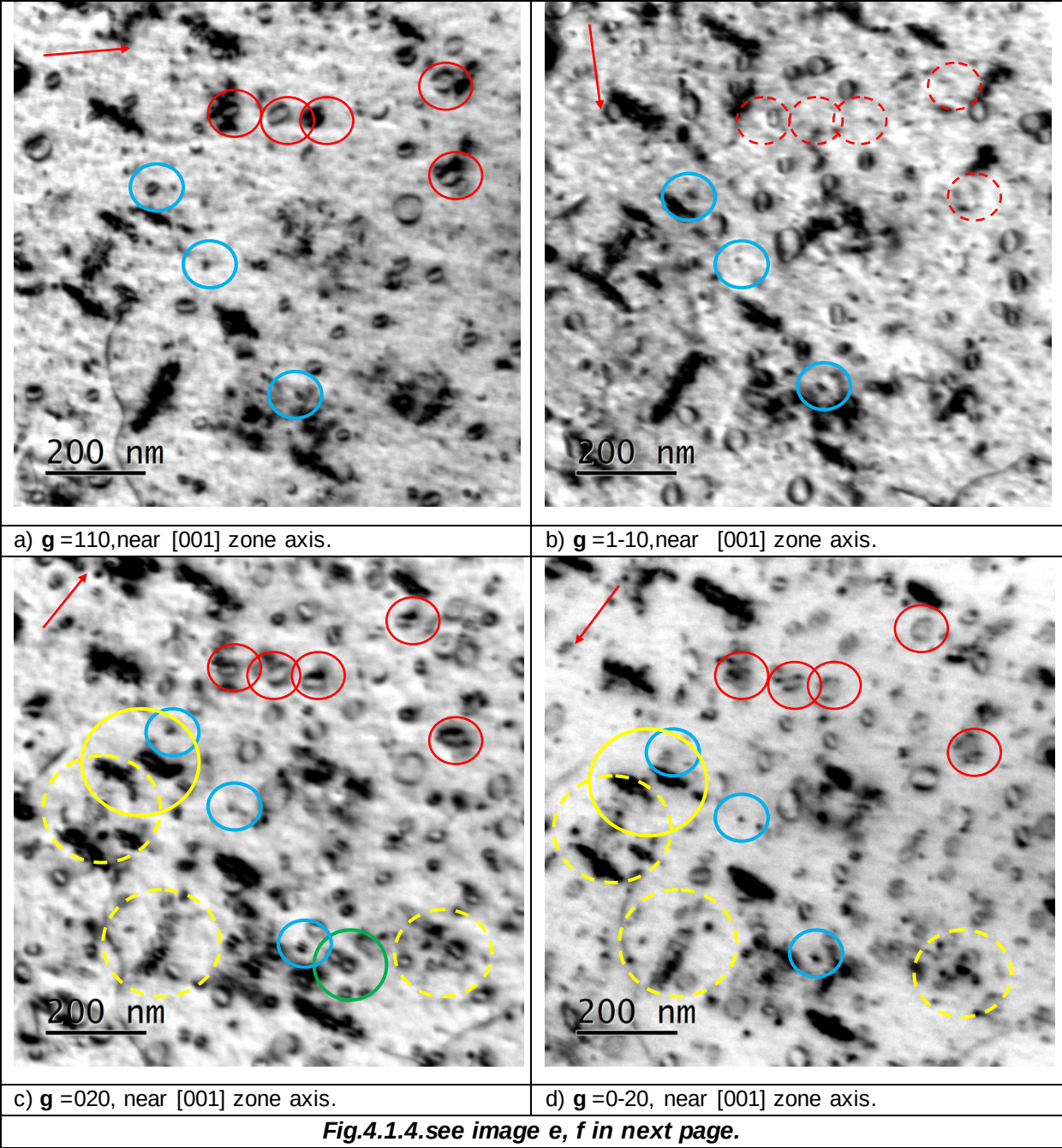
Fig.4.1.4. - Fig.4.1.6, show micrographs of an Fe specimen irradiated at 400°C to a dose of 2.5 dpa. At this temperature, the damage developed into more complex structures such as inhomogeneous finger-shaped loops of size several hundred nanometres and perpendicular loop clusters aligned roughly along the $\langle 100 \rangle$ directions, along with a large number of dislocation loops which were much bigger (50nm vs. 10nm) than those seen in the 300°C implantation. This is the first time that perpendicular loop clusters have been observed in heavy-ion irradiation experiments in Fe. Micrographs were taken to reveal the detailed structure and dislocation interactions of this feature.

Fig.4.1.4.-Fig.4.1.5 show micrographs of the same area imaged under several different diffraction conditions ($g = 110$, $\bar{1}\bar{1}0$, ± 020 and ± 200) near a $[001]$ zone, and again under a diffraction condition $g = \bar{1}\bar{1}0$ after tilting near to three different zone axes. Following the projection images in **Fig.4.1.4**, it can be seen that the perpendicular clusters were composed of several separate loops with a size of about 5nm and numerous smaller loops which were visible as black dots. The larger loops were very close to each other, but there was no evidence of coalescence (**see yellow circles**).

In **Fig.4.1.5**, a comparison of projection images of the clusters from $[001]$, $[11\bar{3}]$ to $[111]$ is presented. It can be seen that from the $[001]$ to the $[111]$ projection direction, the clusters gradually became broader, which indicates that the habit plane of these clusters is closer to the (111) plane than to the (001) plane. **g.b** analyses showed that loops of both $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ type were present. $\langle 100 \rangle$ loops are visible as orthogonal lines or pairs of parallel lines in **Fig.4.1.4.c.d.e.f**. These loops show weak contrast or even disappear in $g = (020)$ or (200) consistent with this observation. All

the larger loops (10-20nm) had Burgers vector of $\frac{1}{2}\langle 111 \rangle$ type (see red circles), while all the $\langle 100 \rangle$ loops were of size less than 5nm (see blue circles).

In **Fig.4.1.4.c**, a loop string formed by two circled $[010]$ loops (in green) is observed along the $\langle 100 \rangle$ direction, which might be the starting stage of the formation of a loop cluster.



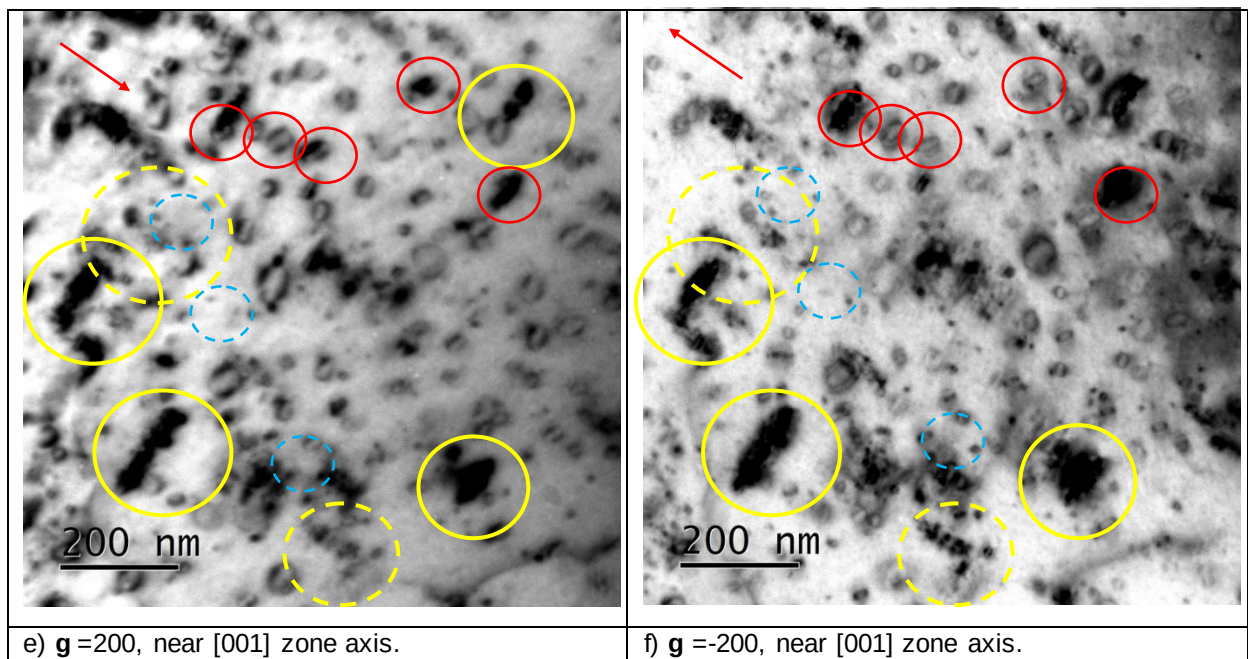


Fig.4.1.4. TEM micrographs of perpendicular loop clusters at different diffraction conditions. Fe sample irradiated at 400°C to a dose of 2.5 dpa.

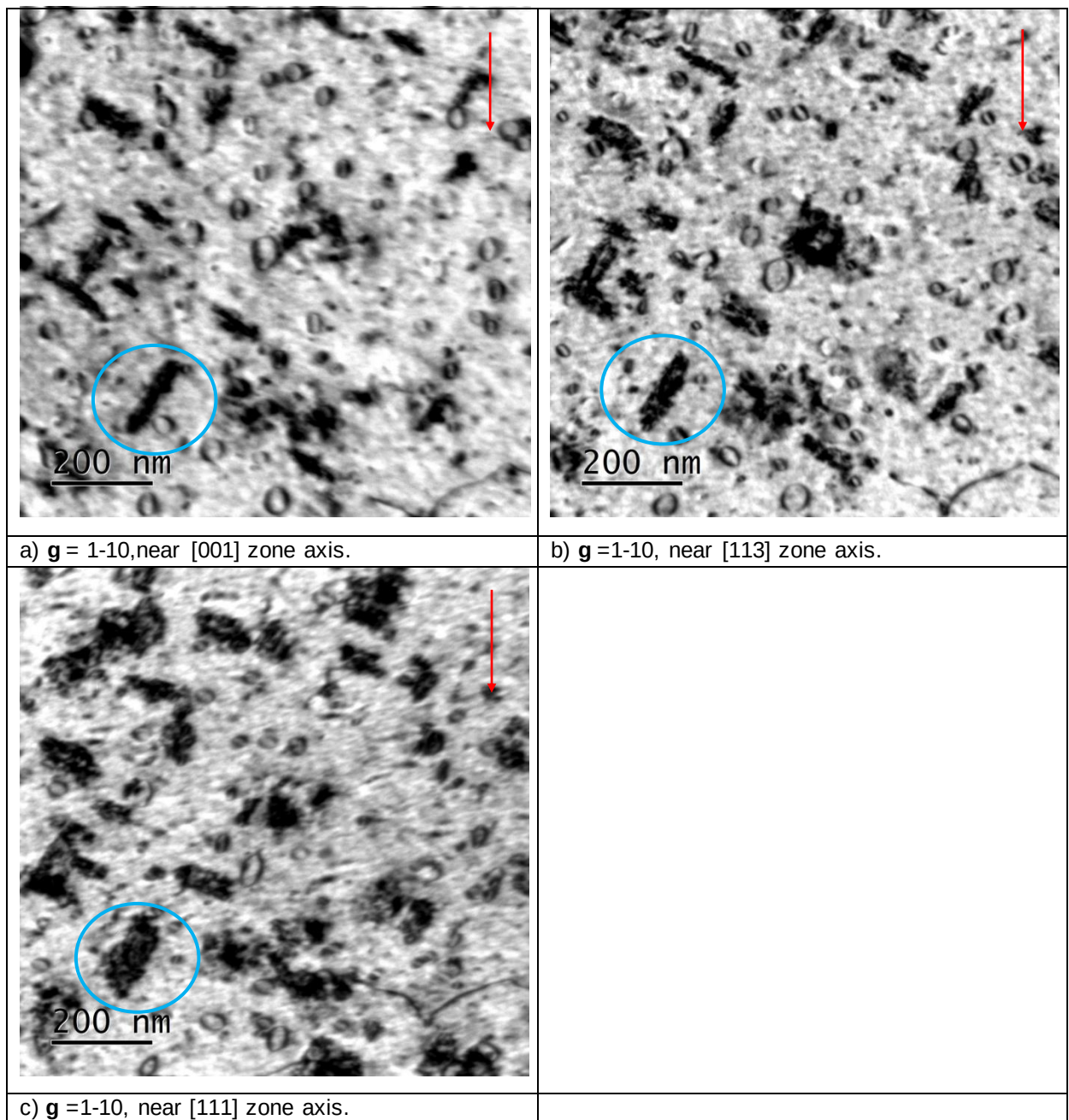
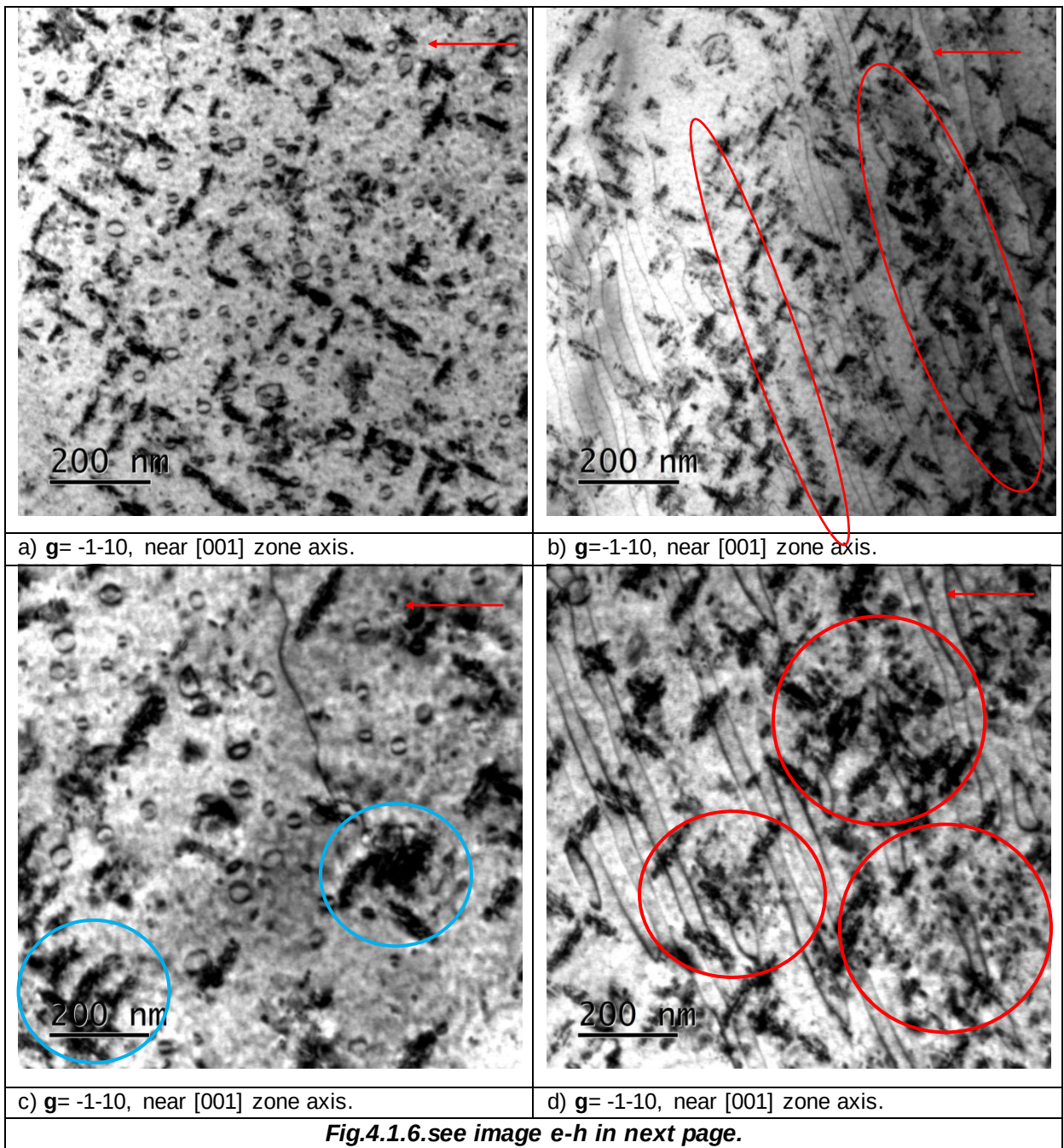


Fig.4.1.5. TEM micrographs of perpendicular loop clusters at different zone axes. Fe sample irradiated at 400°C to a dose of 2.5 dpa.

In **Fig.4.1.6**, complex damage interactions are observed. The interactions can be summarized as the following types:

1. Loop-loop interactions: As described in the previous paragraph, that $\langle 100 \rangle$ loops with same Burgers vectors could form perpendicular loop clusters of the same Burgers vectors (**see Fig.4.1.4.**).
2. Loop-cluster interactions: Small $\langle 100 \rangle$ loops visible as black dots are observed assembling around the perpendicular loop clusters, which indicates that perpendicular loop clusters can absorb nearby $\langle 100 \rangle$ loops to grow along the original $\langle 100 \rangle$ direction (**see Fig.4.1.6.g.h.**).
3. Cluster-cluster interactions: In **Fig.4.1.6.(see blue circles)**, perpendicular loop clusters can interact with other clusters to form irregular shaped loop clusters.
4. Cluster-finger-shaped loop interactions: In **Fig.4.1.6.b.d.f**, it was observed that when loop clusters and finger shape loops interacted, the perpendicular loop clusters lost their regular arrays and became accumulations of irregular black dots, while the finger-shaped loops were broken into line dislocations and gradually absorbed into the irregular loop accumulations.



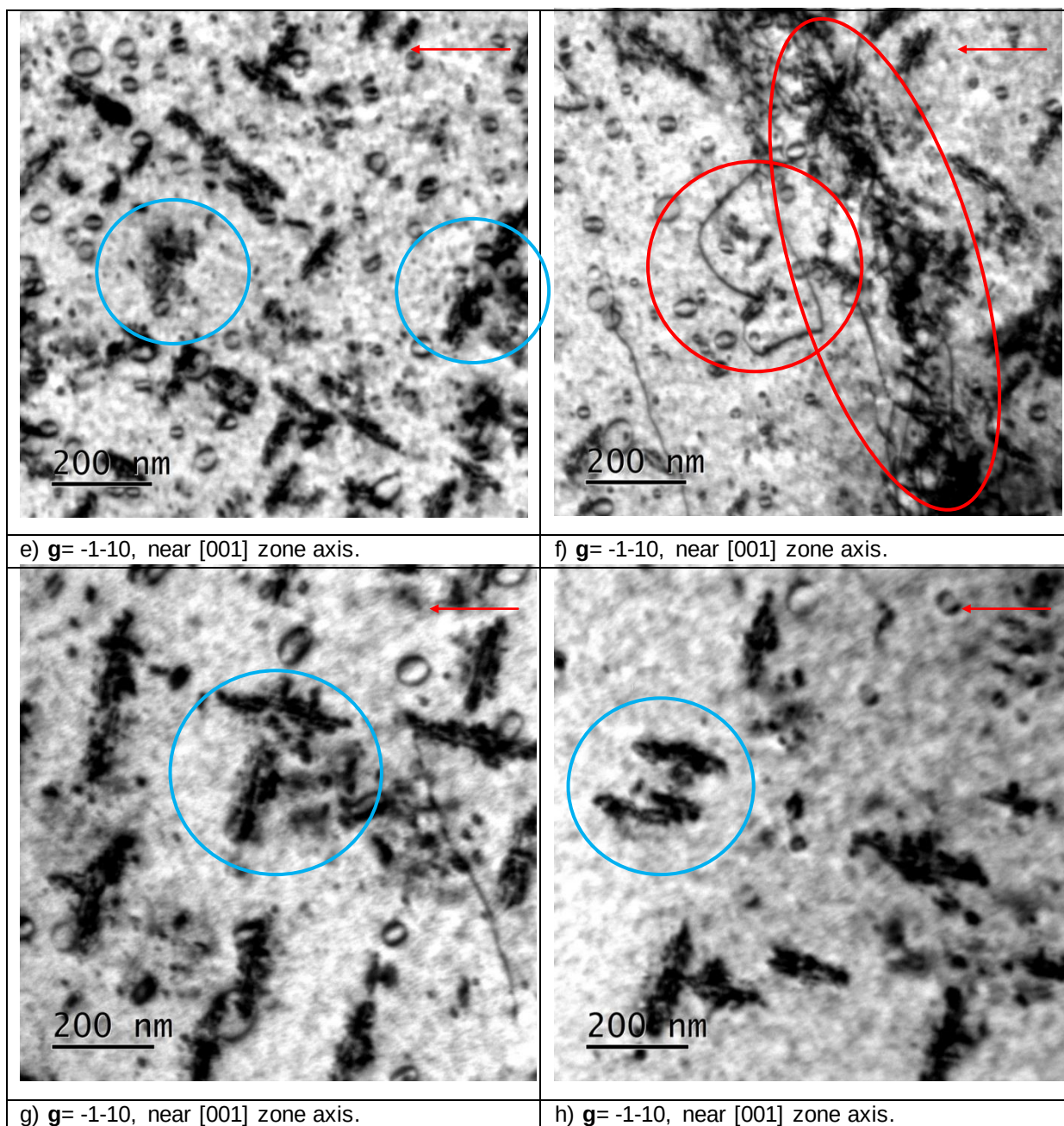


Fig.4.1.6. TEM micrographs of dislocation interactions in Fe sample irradiated at 400°C to a dose of 2.5 dpa.

Fe specimen irradiated at 400°C to a dose of 7dpa

Fig.4.1.7. shows TEM micrographs of an Fe specimen irradiated at 400°C to a dose of 7dpa. At this irradiation condition, the microstructure of the foil was dominated by large irregular loop clusters with a size of several hundred nanometres and line dislocations. It was interesting that at this dose, some of the clusters still had perpendicular features (**see Fig.4.1.7.a**) similar to those observed in the 2.5dpa irradiation. Moreover, some of the line dislocations still maintained the curve features (**see Fig.4.1.7.b**) which looked like the finger-shaped loops seen in the 2.5 dpa irradiation. Compared with the microstructure at 2.5dpa, the population of $\frac{1}{2}\langle 111 \rangle$ loops decreased.

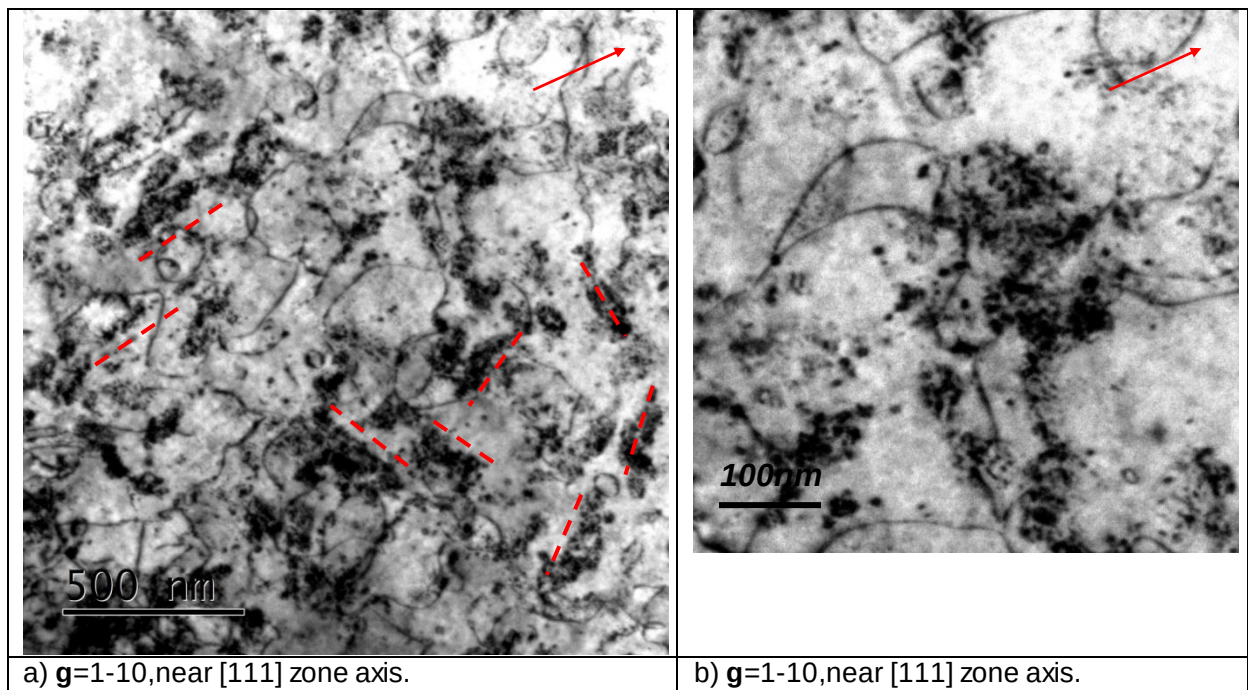
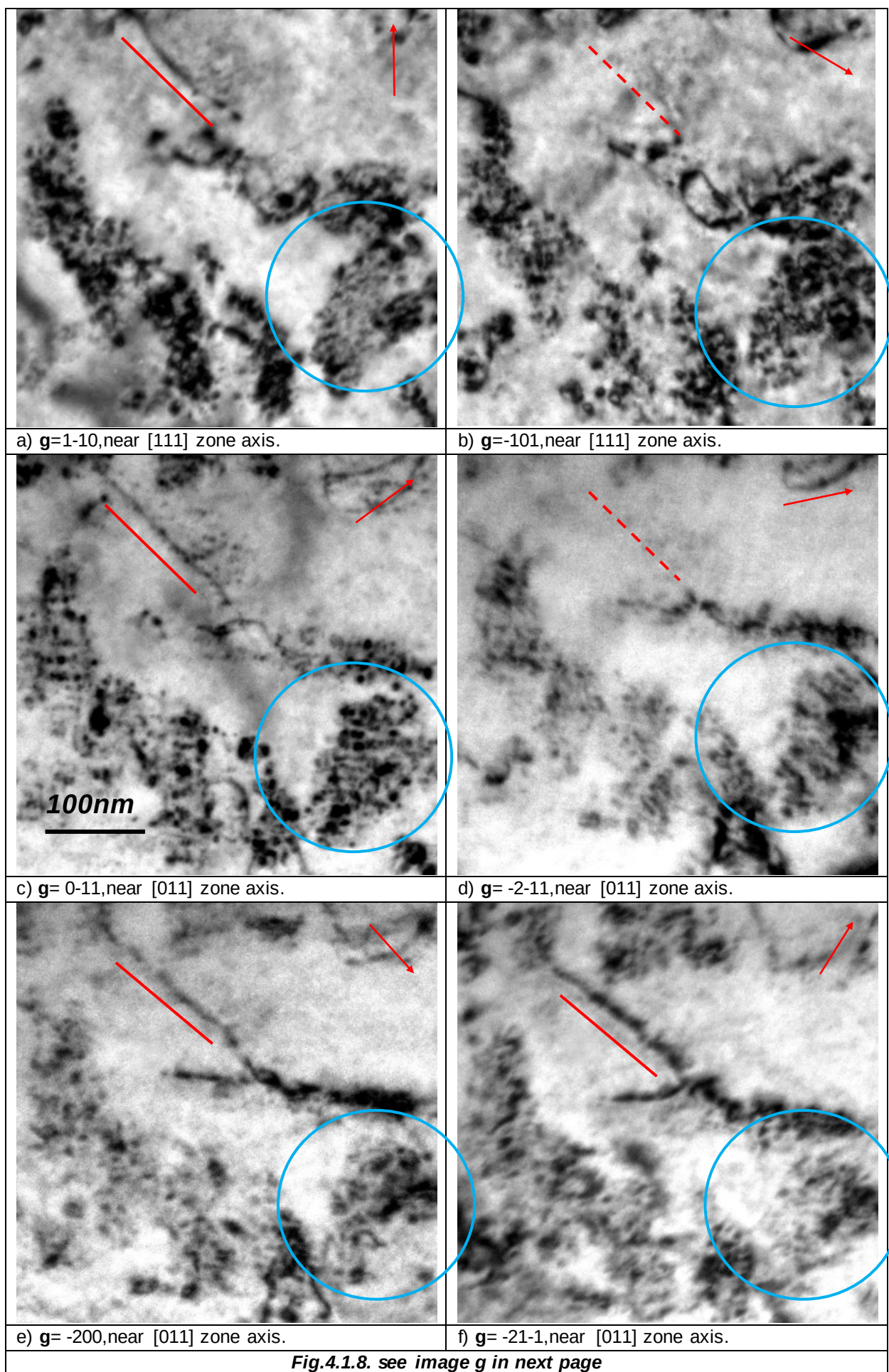


Fig.4.1.7. TEM micrographs of Fe samples irradiated at 400°C to a dose of 7dpa.

Fig.4.1.8. shows TEM micrographs of loop clusters under different diffraction conditions. The loop clusters in 7dpa irradiation were again formed by numerous small loops. However, **g.b** analysis showed that the clusters in the 7dpa irradiation contained loops of both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors, unlike the clusters formed in the 2.5dpa irradiation that were composed only of $\langle 100 \rangle$ loops. Taking the

large irregular cluster inside the blue circle as an example, and following its contrast changes in **Fig.4.1.8**, it can be seen that although the cluster as whole was visible under all the diffraction conditions, groups of loops within the cluster showed weak contrast or even disappeared in some of the diffraction conditions. This is the evidence that the clusters were formed of loops with both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors. The analysis showed that about 70% of the loops inside the clusters had $\langle 100 \rangle$ Burgers vectors, while 30% loops had $\frac{1}{2}\langle 111 \rangle$ Burgers vectors. In **Fig.4.1.8**, a line dislocation (**see the red line**) is also analysed. It was found that the line dislocation showed contrast extinction in $\mathbf{g} = \overline{1}01, \overline{2}\overline{1}1$, while it was visible in all the other diffraction conditions. Thus the line dislocation has a Burgers vector of $\frac{1}{2}[1-11]$, which might indicate that the finger loops seen at 2.5 dpa also have $\frac{1}{2}\langle 111 \rangle$ Burgers vectors, since finger loops broke into curved dislocation lines as the implantation dose increased from 2.5dpa to 7 dpa.



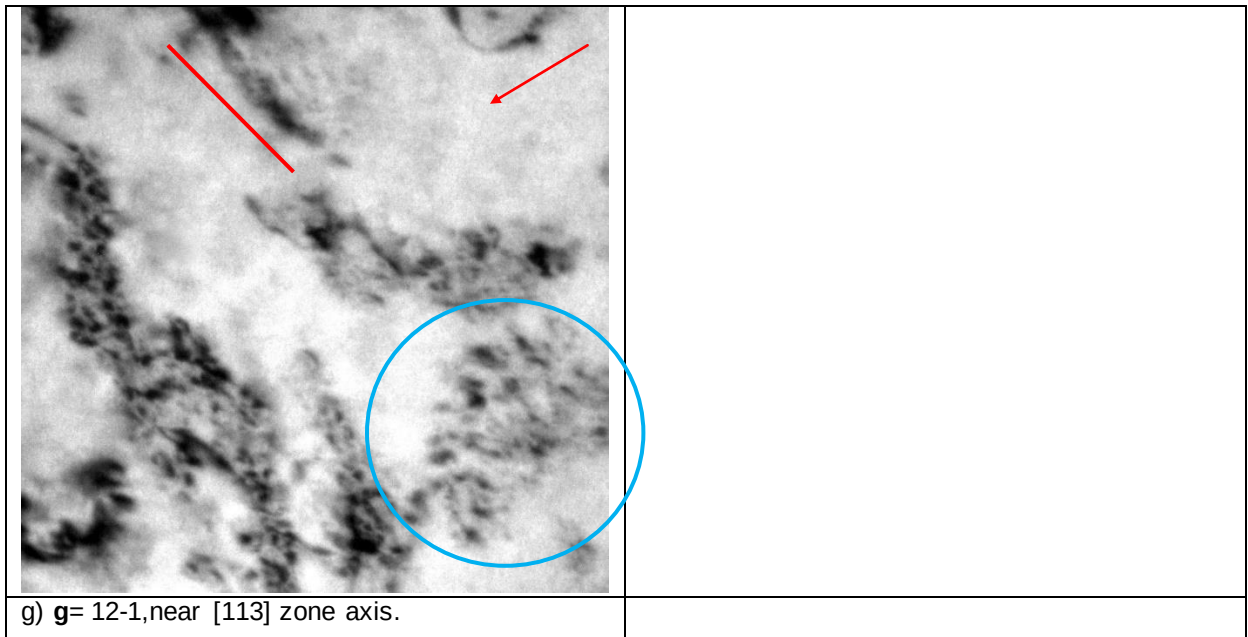


Fig.4.1.8. TEM micrographs of irregular loop clusters at different diffraction conditions in an Fe sample irradiated at 400°C to a dose of 7dpa.

Fe irradiated at 460°C to a dose of 2.5 dpa

Fig.4.1.9. shows TEM micrographs of Fe samples irradiated at 460°C to a dose of 2.5 dpa. At this temperature, unlike the microstructures observed at 2.5/7 dpa in the 400°C implantation, no loop clusters or finger-shaped loops were observed. Loops with an average size of 13 nm were homogeneously distributed within the foil. The average size of the loops was larger than those at 2.5/7 dpa in the 400°C implantation. Loop strings were observed in many cases (**see the red lines**).

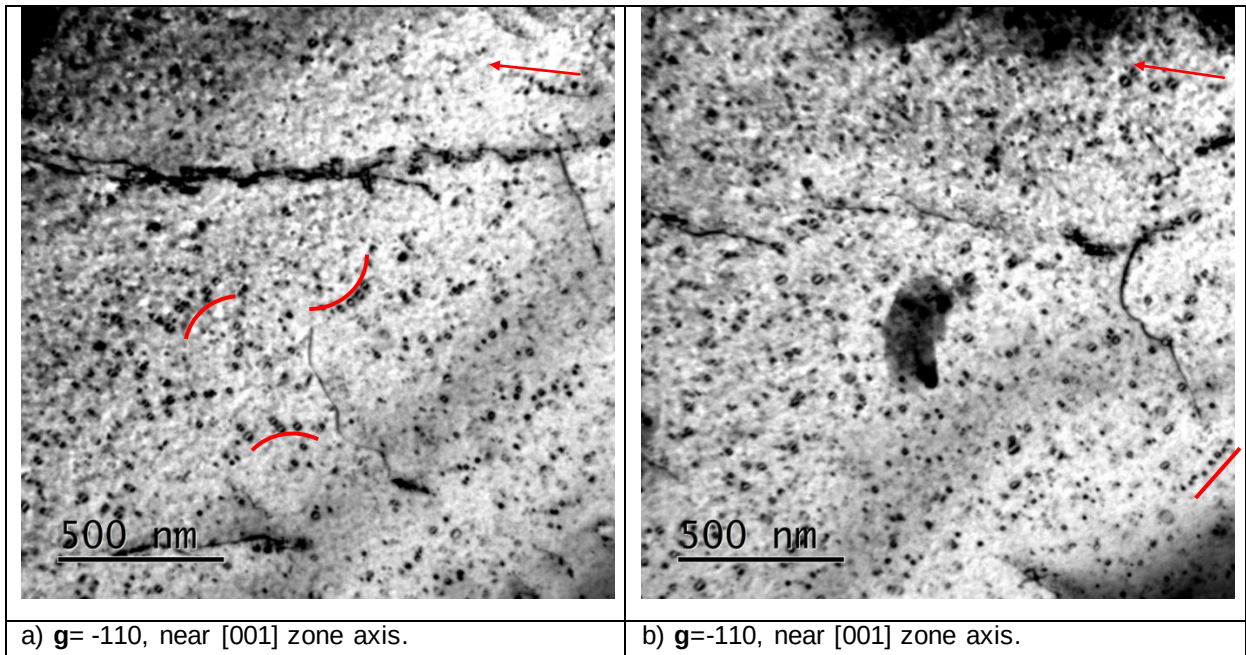


Fig.4.1.9. TEM micrographs of Fe samples irradiated at 460°C to a dose of 2.5 dpa.

4.2 Loop Image Size Distributions

Fig.4.2.1 shows the loop image size distributions measured for Fe samples irradiated at temperatures from 300°C to 460°C at two different doses (2.5-7dpa). The histograms in the figure represent the percentage of the visible loop population within different loop image size ranges, based on the measurement of 200-400 loops at the same diffraction conditions ($g=\bar{1}10$ near [001] zone axis). Before discussing these loop size distributions, two facts are worthy of notice: (1) In the foil irradiated at 400°C/2.5dpa, the perpendicular loop clusters were not taken into account, since it was impossible to measure the sizes of the loops inside the clusters. Moreover the finger-shaped loops with a size of several microns were also not included, since they were too big and the distribution was non-uniform; (2) In the foil irradiated at 460°C/2.5dpa, the broken finger-like loops were not taken into consideration for the same reason.

The effects of irradiation temperature on the loop size distributions are shown at both 2.5dpa and 7dpa. An increase in temperature from 300°C to 400°C and from 400°C to 460°C caused a shift to larger loop sizes in the distributions.

Comparing the data from both 2.5 and 7dpa irradiation at 400°C, an increase in dose did not cause a shift to larger loop size, but on the contrary to smaller loop size. The reason for this is apparent in the microstructures shown in **Fig.4.1.1**. At the higher dose the size of the loop clusters increased remarkably, but due to the absence of big $\frac{1}{2}\langle 111 \rangle$ loops, which were believed to be absorbed by the loop clusters, the average loop size decreased.

Fig.4.2.2.-Fig.4.2.3 represents the average loop sizes and maximum loop size variation under different irradiation conditions. At 7dpa, the average loop size increased from 6nm at 300°C to 9nm at 400°C, while the maximum loop size

increased from 16nm at 300°C to 82nm at 400°C. It is clear that below 400°C, both average loop size and maximum loop size increased with both irradiation temperature and dose.

At 2.5dpa, both the average and maximum loop sizes decreased as the temperature increased from 400°C to 460°C. However, as the larger number of small loops contained in the perpendicular clusters were not taken into consideration, this conclusion is still debatable. Looking again at **Fig.4.1.1.b.c**, and considering the large number of small loops inside the clusters at 400°C, it is reasonable to believe that the average loop size also increased with increasing temperature at 2.5dpa. However the absence of complex structures at 460°C also indicates that the recombination rate has increased remarkably around this temperature.

Looking at the data in 400°C implantations, the average loop size decreased and the maximum loop size increased as the implantation dose increased. Since the small loops inside the clusters in the 2.5dpa implantation were not taken into account, it is reasonable to conclude that the average loop sizes were similar at each dose.

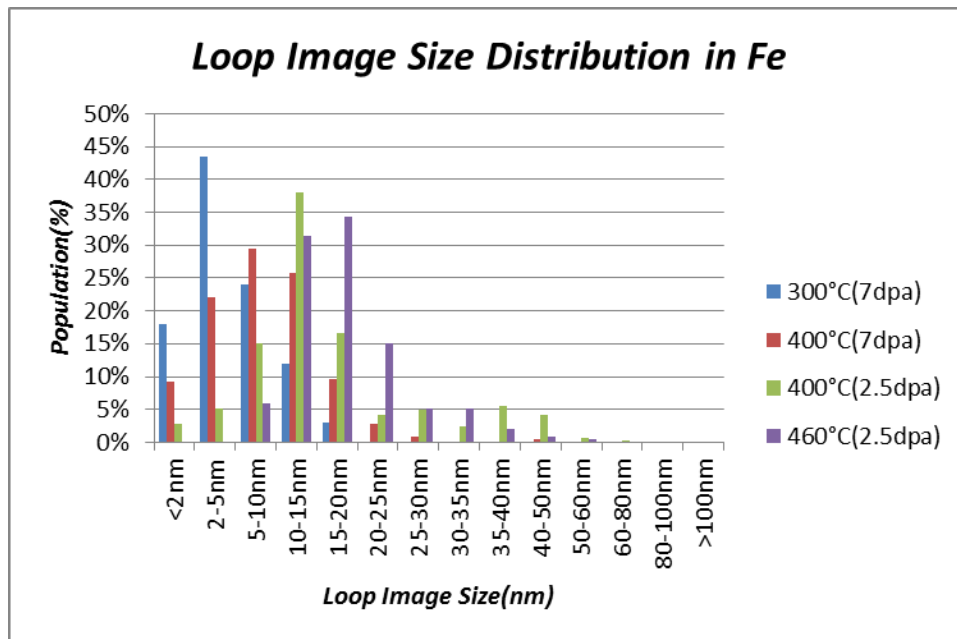


Fig.4.2.1. Loop size distributions in Fe irradiated at different temperatures and doses.

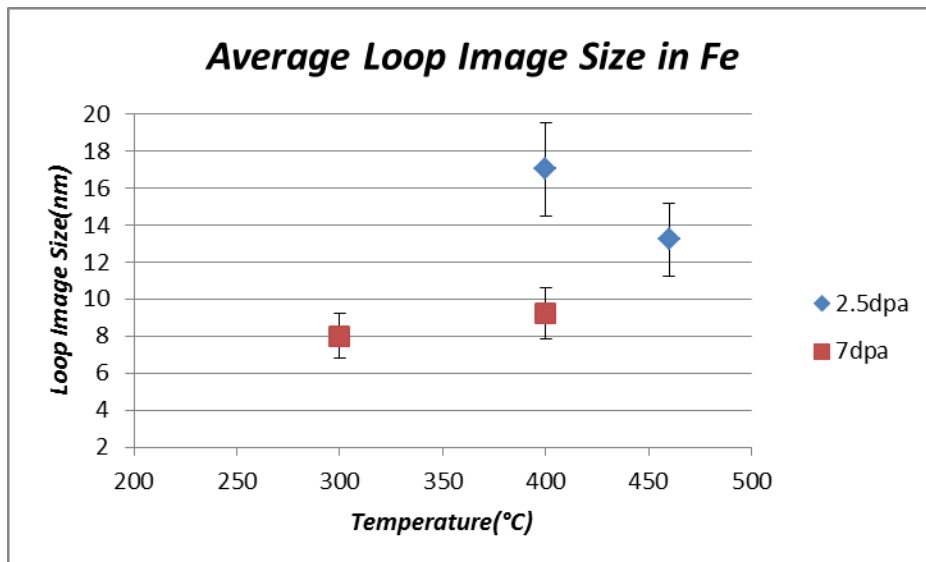


Fig.4.2.2. Average loop sizes for Fe at different irradiation temperatures and doses.

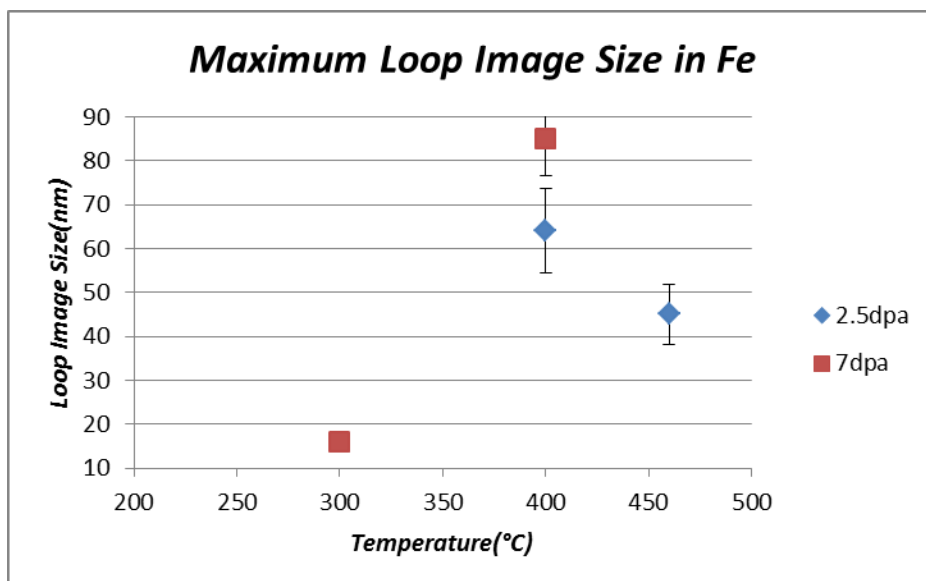


Fig.4.2.3. Maximum loop sizes for Fe at different irradiation temperatures and doses

4.3 Loop Number Densities

Accurate measurement of volume number densities requires knowledge of foil thickness. In this work, foil thicknesses were measured by the convergent beam method, and for the micrographs presented in this chapter varied from 100-130nm.

Fig.4.3.1 shows the loop number densities at different irradiation conditions. The number density at 300°C was the highest, and at irradiation temperatures $\geq 400^{\circ}\text{C}$ the loop number densities were similar and showed no obvious dependence on dose.

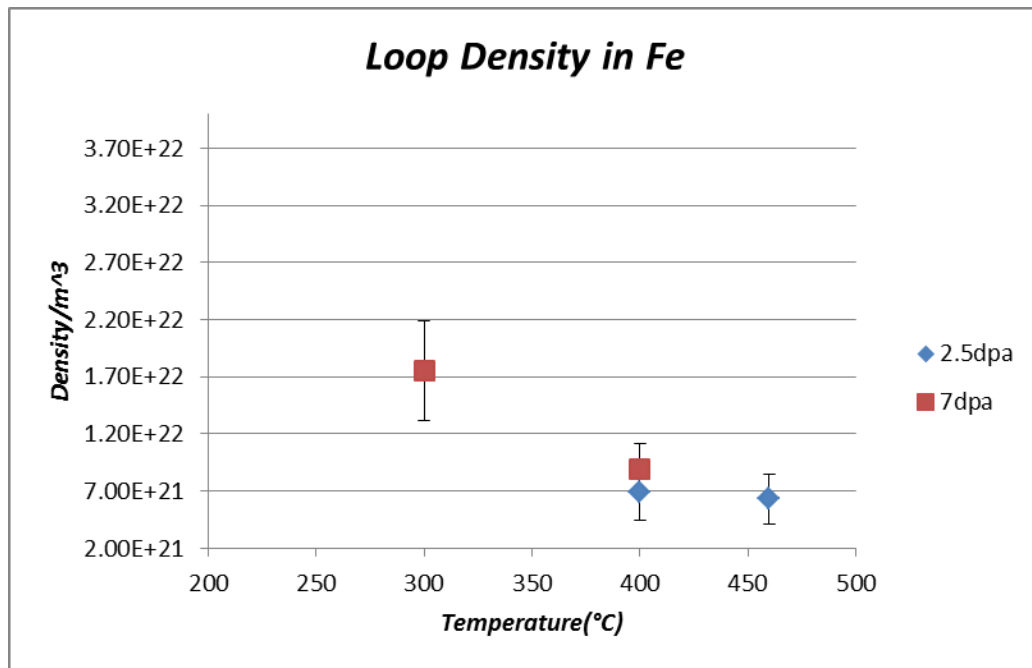


Fig.4.3.1. Dislocation loop densities as a function of irradiation temperatures and dose.

4.4 Loop Burgers Vectors

The results of **g.b** analysis in this chapter are based on the loops whose Burgers vectors could be accurately identified in a selected area. Most of the **g.b** experiments were performed near the [001] foil orientation under kinematical bright-field diffraction conditions, using the four available diffraction vectors near the pole, as this is the most effective way to determine the type of Burgers vector. However a limitation is that at this orientation loops with Burgers vector [001] are invisible in all four diffraction conditions. Also, in some cases, loops with size less than about 3nm could not be characterised due to weak image contrast or poor surface conditions. Thus the $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratios reported in this work are more representative of larger loops. Normally the conclusions in this chapter are based on the analysis of ~100 loops whose Burger vectors could be accurately identified.

4.4.1 Characterisation of 300°C/7dpa specimens

Fig.4.4.1.1 shows a series of micrographs taken near the [001] zone axis in a Fe specimen irradiated at 300°C to a dose of 7dpa. Images were taken using all four reflections available at this zone axis. With the aid of **Table 4.4.1.1**, one can distinguish $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops immediately by comparing the images obtained from diffraction vectors **g**=200 and 020. All loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vectors stay in contrast in both **g**=200 and 020 diffraction vectors, while the two types of $\langle 100 \rangle$ loops (i.e. **b** = [100] and [010]) are out of contrast in one of these two **g** vectors.

In **Fig.4.4.1.1**, it can be seen that the loops circled in blue are invisible either in (020) or in (200) diffraction vectors, while these loops stay in contrast in (110) and $(\bar{1}10)$ diffraction vectors. Thus these circled loops in blue have $\langle 100 \rangle$ Burgers vectors. Of 98 loops analysed, 45 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, 32 had Burgers vectors of type $\langle 100 \rangle$ and it was not possible to identify the Burgers vectors of 21 loops. Thus in Fe irradiated at 300°C to a dose of 7dpa, about 58% were $\frac{1}{2}\langle 111 \rangle$ loops and 42% loops were $\langle 100 \rangle$ loops.

g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(110)$	1	0	0	1	1	1	0
$\pm(020)$	1	1	1	1	0	2	0
$\pm(200)$	1	1	1	1	2	0	0
$\pm(-110)$	0	1	1	0	1	1	0

Table.4.4.1.1. $g.b$ table for $[001]$ zone axis.

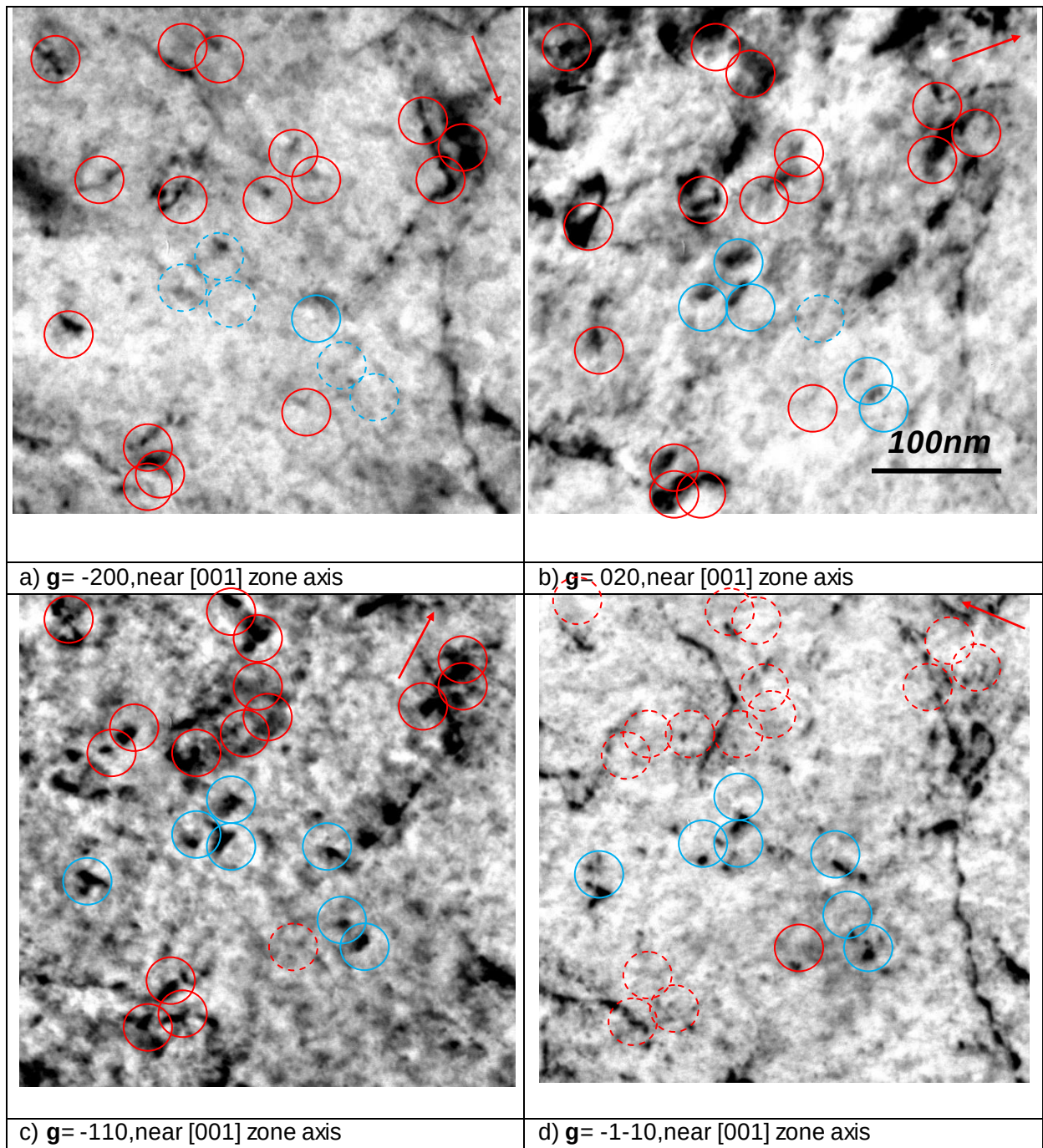


Fig.4.4.1.1. $g.b$ contrast experiments near $[001]$ zone axis, for Fe specimen irradiated at $300\text{ }^{\circ}\text{C}$ to a dose of 7dpa; $b=\frac{1}{2}\langle 111 \rangle$ (red circles) and $b=\langle 100 \rangle$ (blue circles).

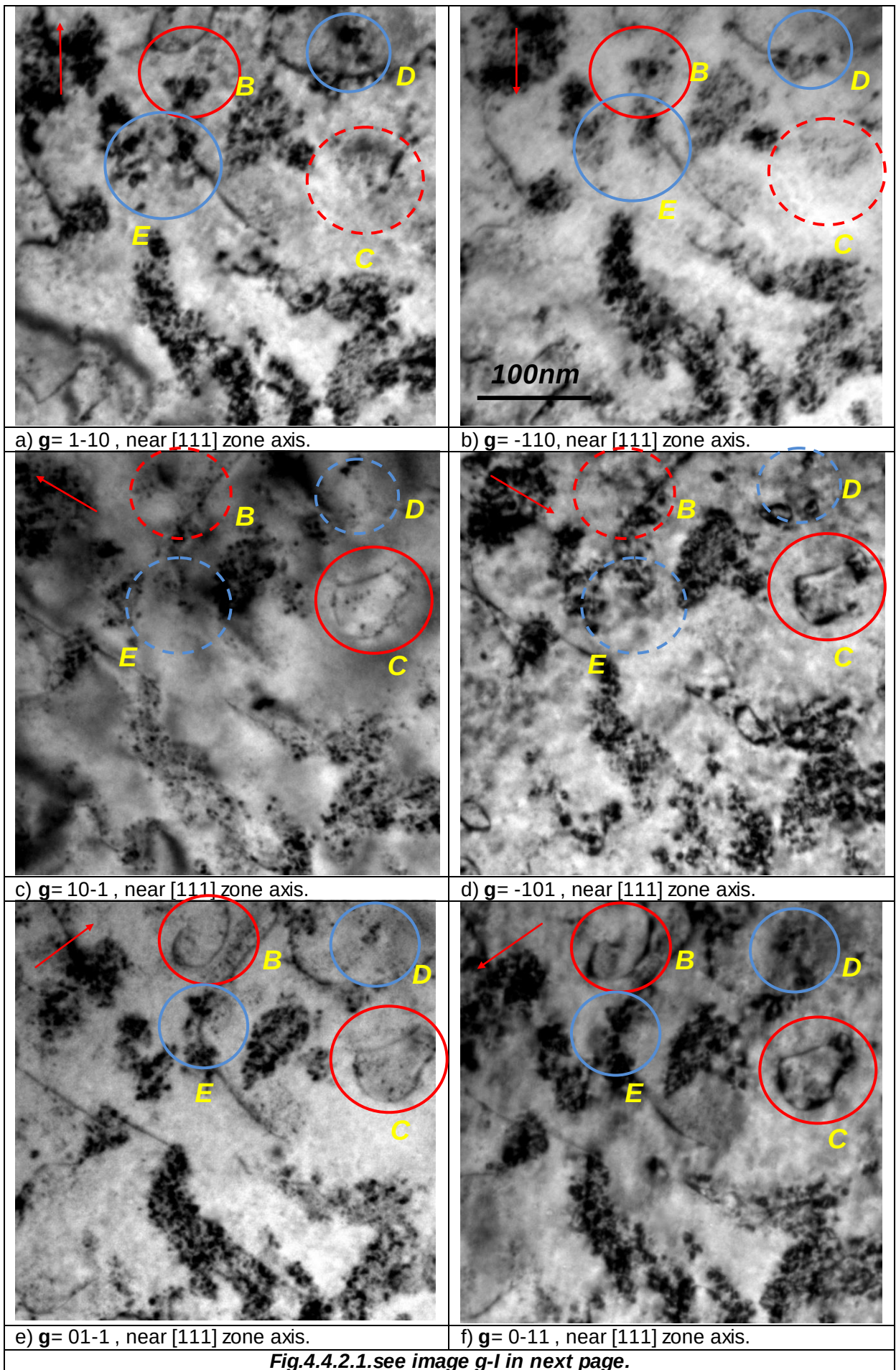
4.4.2 Characterisation of 400°C/7dpa specimens

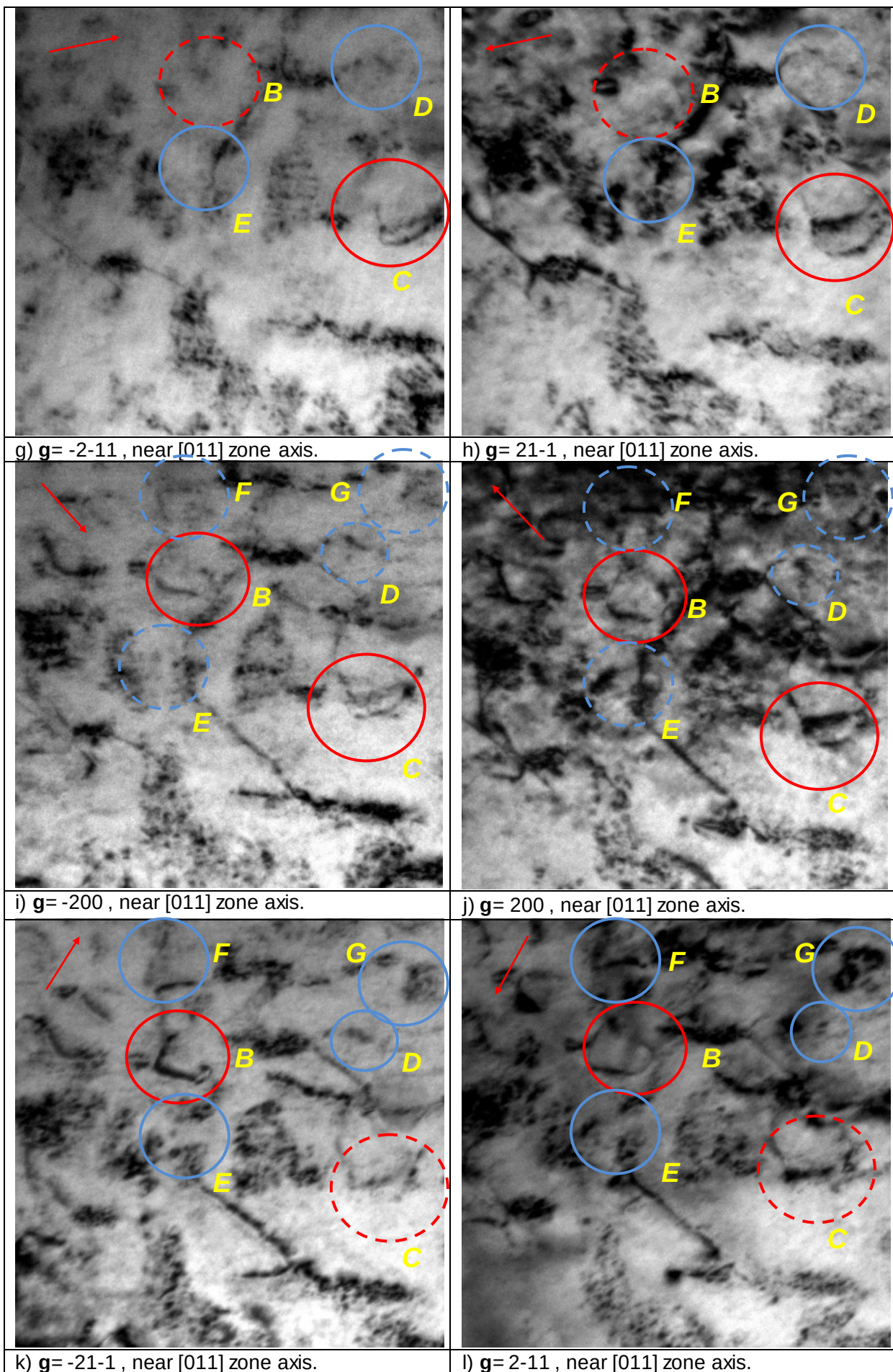
Fig.4.4.2.1 shows the **g.b** contrast experiments near [111], [011], and [113] zone axes, for an Fe specimen irradiated at 400°C to a dose of 7dpa. Both +**g** and –**g** images were obtained. Probably this was one of the most difficult **g.b** analyses throughout this work, not only because the favoured [001] orientation was unavailable, but also because the presence of large loop clusters, which contained numerous loops of both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ type, made the image contrast very ambiguous (**see 4.1**). Thus, in this case sometimes experience are needed to draw a reliable conclusion.

For example, in **Fig.4.4.2.1**, loop B was invisible in $\mathbf{g}=\pm(10\bar{1})$, but showed contrast in $\mathbf{g}=\pm(\bar{1}10)$ and $\pm(01\bar{1})$ near the [111] pole. Then the specimen was tilted to the [011] pole, where further four reflections were reached. It can be seen that loop B disappeared again in $\mathbf{g}=\pm(21\bar{1})$. With the aid of **Table.4.4.2.1**, the conclusion can be drawn that loop B has Burgers vector $\frac{1}{2}[\bar{1}\bar{1}1]$. Similar methods can be applied to loops E, F, D and G, which showed contrast in all the reflections except $\mathbf{g}=\pm(10\bar{1})$ and $\pm(200)$, so they have Burgers vector [010]. Of 135 loops analysed, 40 loops could not be characterised, 45 loops had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, and the rest had Burgers vectors of type $\langle 100 \rangle$. It can be seen that in Fe irradiated at 400°C to a dose of 7 dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were ~47% and 53% respectively.

[111]	[011]	[113]					
g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(-110)$	0	1	1	0	1	1	0
$\pm(10-1)$	0	1	0	1	1	0	1
$\pm(01-1)$	0	0	1	1	0	1	1
$\pm(2-11)$	1	1	2	0	2	1	1
$\pm(200)$	1	1	1	1	2	0	0
$\pm(21-1)$	1	1	0	2	2	1	1
$\pm(12-1)$	1	0	1	2	1	2	1

Table.4.4.2.1. *g.b* table for [111], [011] and [113] zone axes.





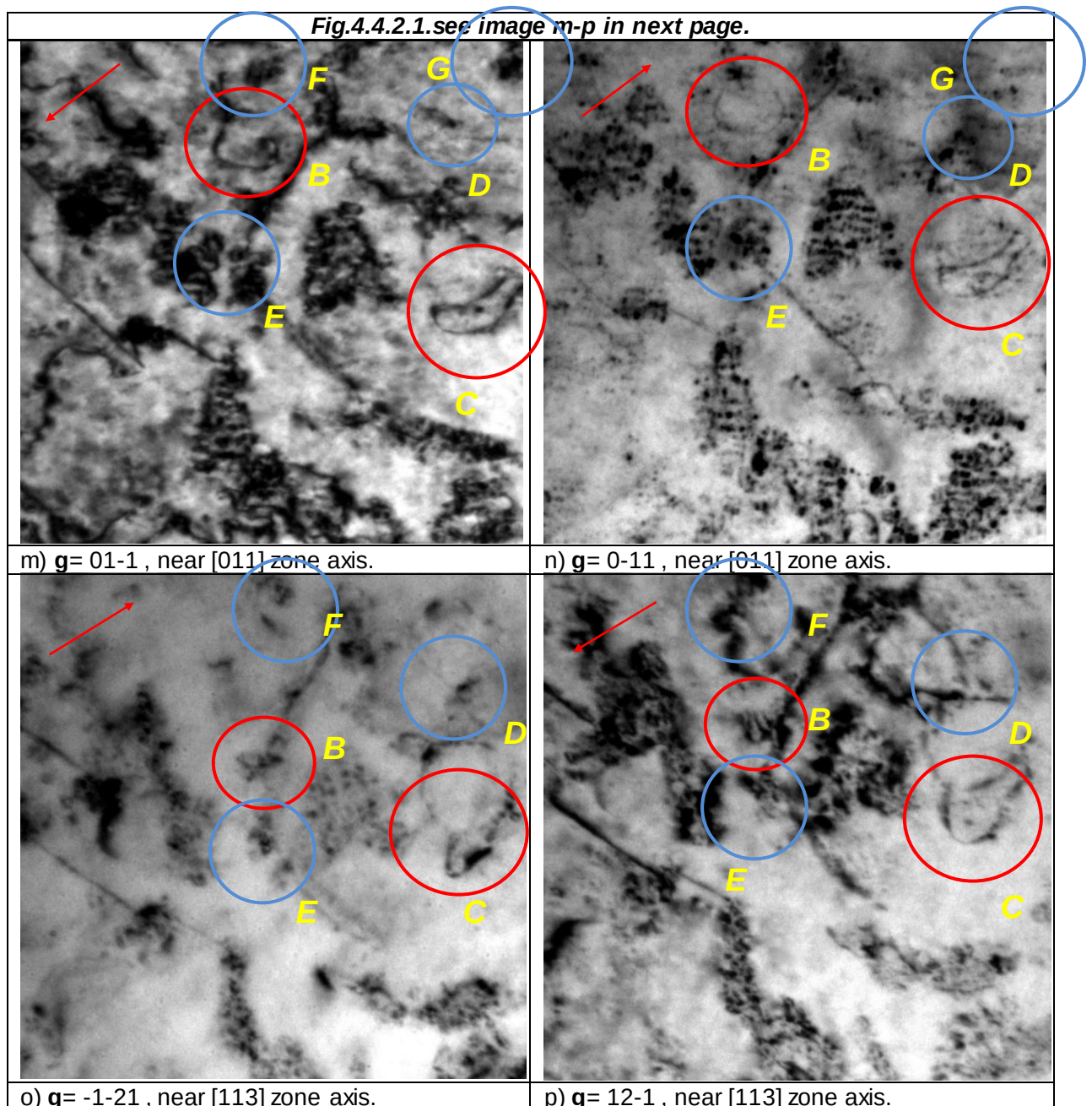
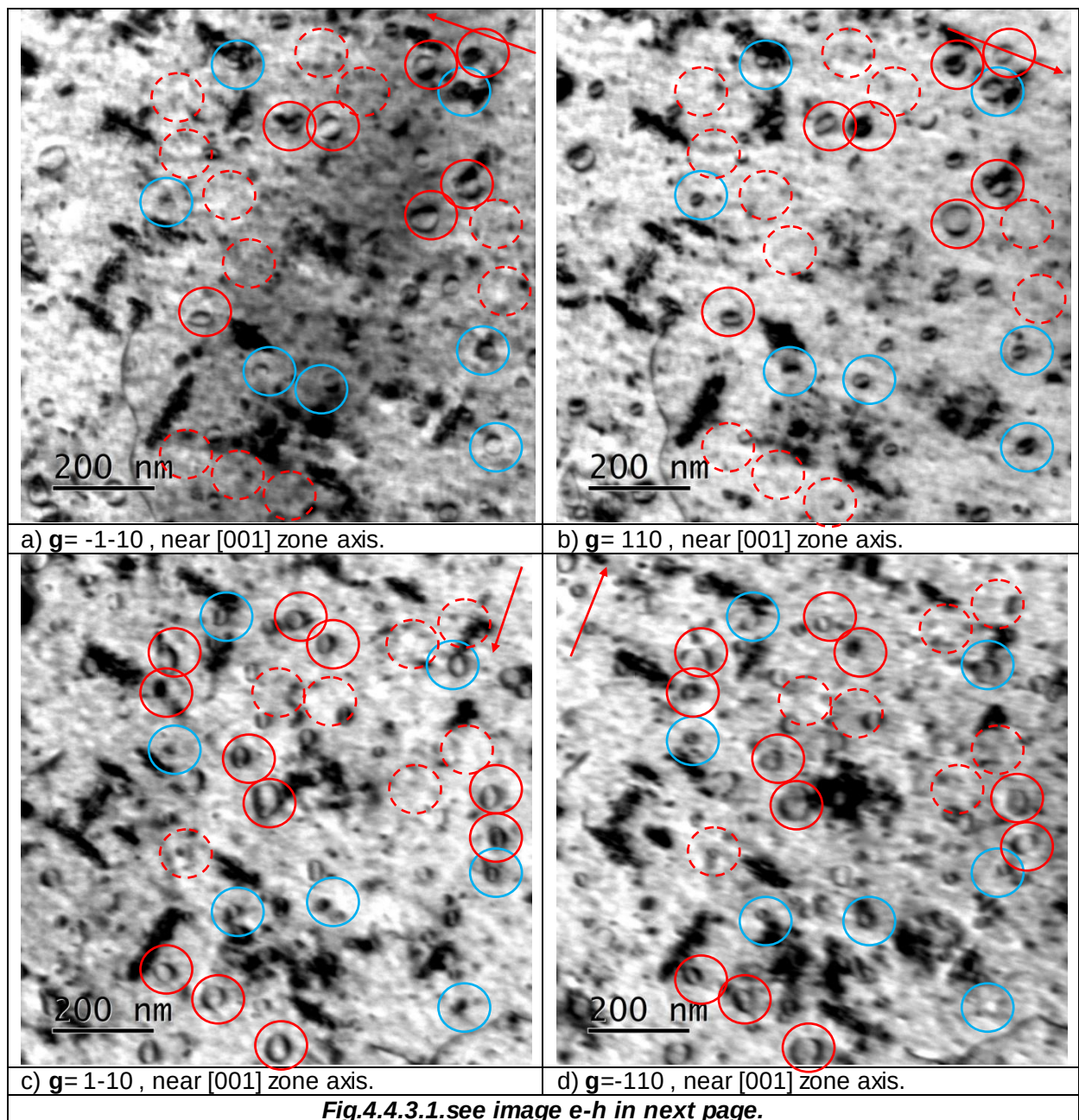


Fig.4.4.2.1. $g.b$ contrast experiments near $[111]$, $[011]$ and $[113]$ zone axes, for Fe specimen irradiated at 400°C to a dose of 7dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

4.4.3 Characterisation of $400^\circ\text{C}/2.5\text{dpa}$ specimens

Fig.4.4.3.1 shows eight micrographs from a same area of a Fe specimen irradiated at 400°C to 2.5dpa. The micrographs were taken with four different diffraction vectors close to the $[001]$ zone axis. As described in 4.4.1, at this orientation just two images obtained in $g=200$ and $g=020$ are sufficient to distinguish between loops with $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ Burgers vectors. The loops ringed in red are of $\frac{1}{2}\langle 111 \rangle$ type, while the

loops ringed in blue are of $\langle 100 \rangle$ type. Overall, of 138 loops counted, 61 had Burgers vector of type $\frac{1}{2} \langle 111 \rangle$ and 59 had Burgers vectors of type $\langle 100 \rangle$ whilst the Burgers vectors of 18 loops could not be identified. It can be seen that in Fe specimen irradiated at 400°C to a dose of 2.5dpa, the proportions of $\frac{1}{2} \langle 111 \rangle$ and $\langle 100 \rangle$ loops were ~51% and 49% respectively.



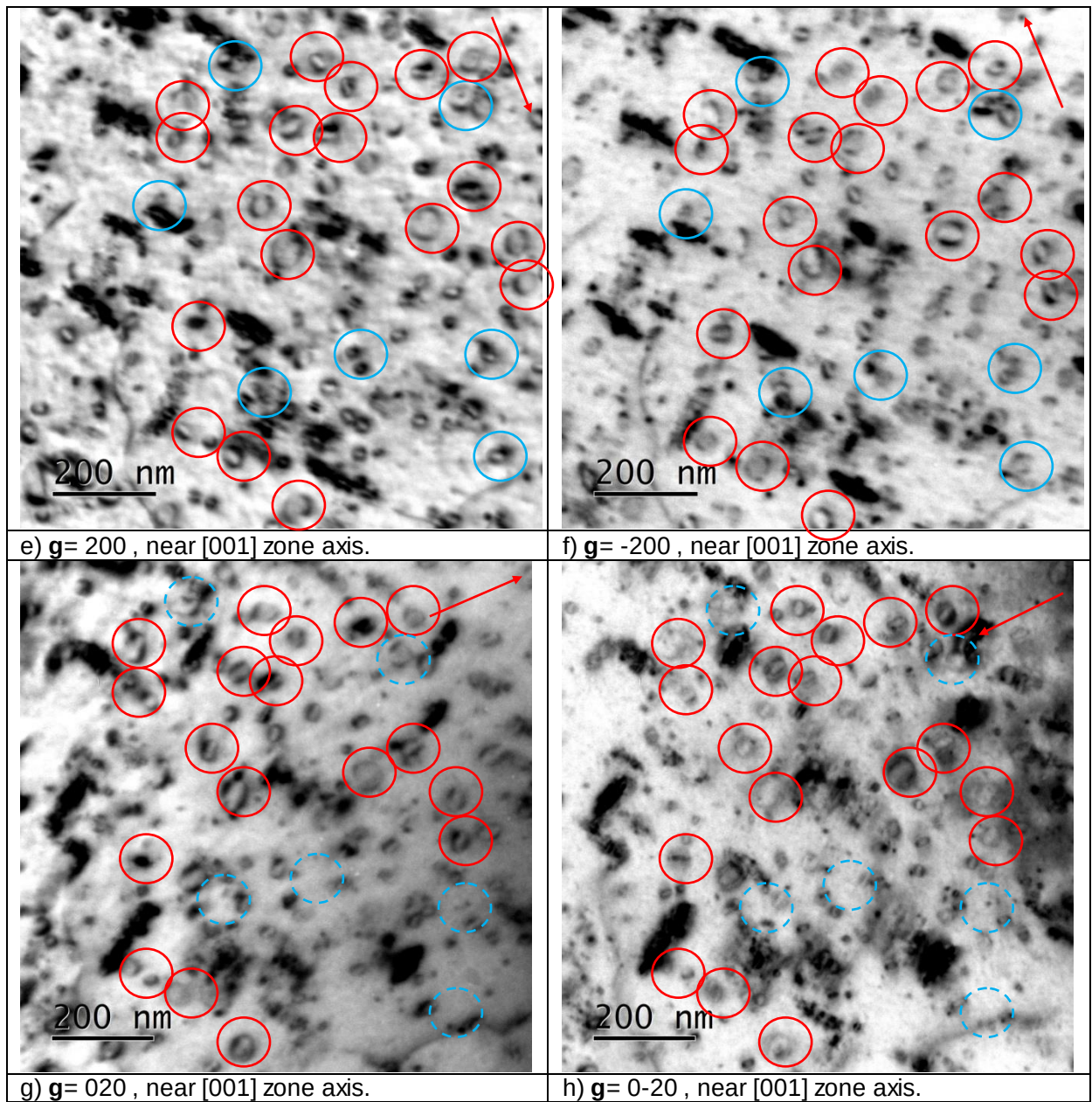
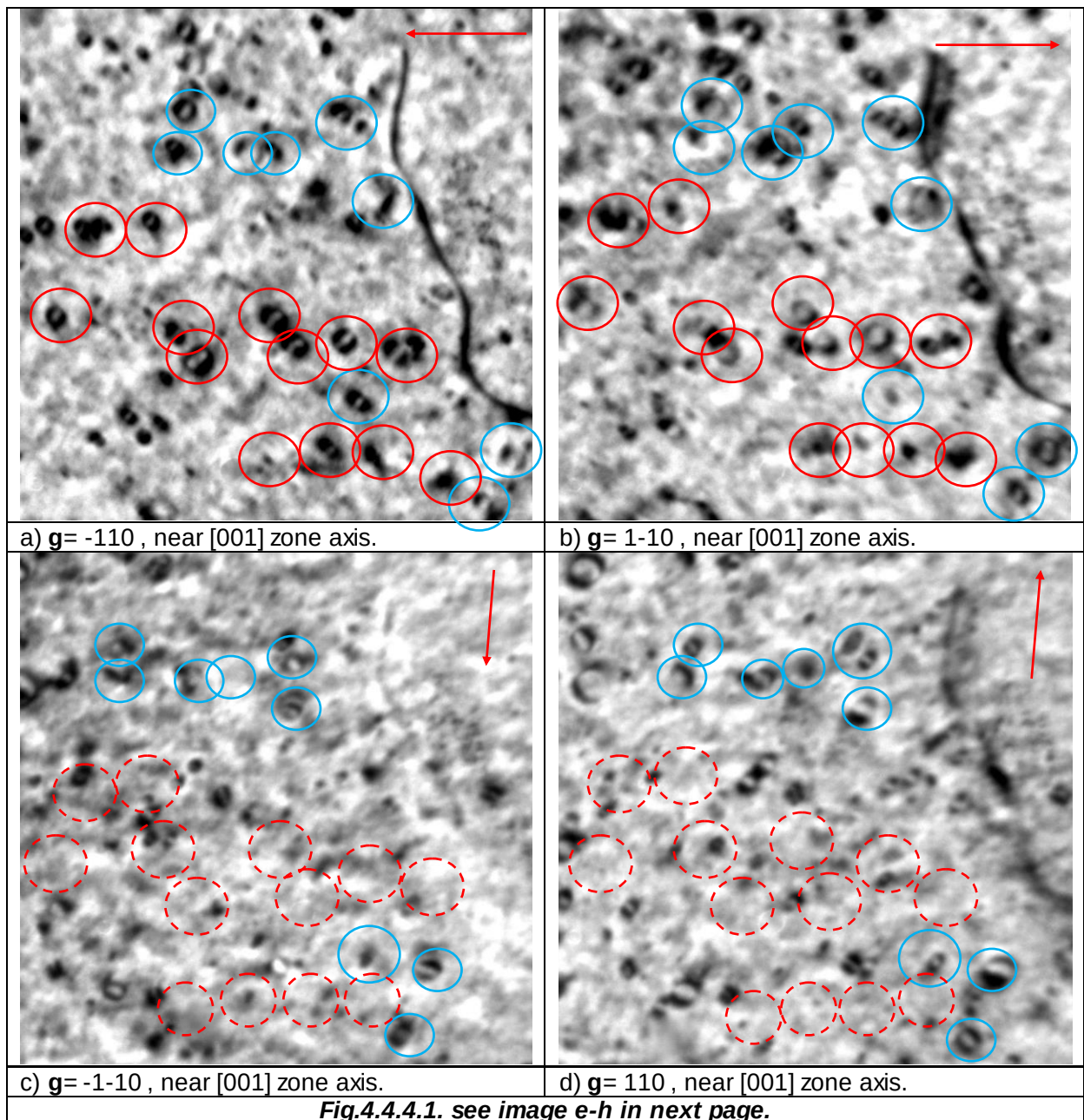


Fig.4.4.3.1. ***g.b** contrast experiments near $[001]$ zone axis, for an Fe specimen irradiated at 400 °C to a dose of 2.5 dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).*

4.4.4 Characterisation of 460°C/2.5dpa specimens

Fig.4.4.4.1 shows the contrast experiments near the [001] zone axis, with both +**g** and –**g** micrographs. Analysis proceeded as above. Of 103 loops which were analysed it was found that 33 loops were of $\frac{1}{2}\langle 111 \rangle$ type, and 62 were of $\langle 100 \rangle$ type, whilst it was not possible to identify the Burgers vectors of 8 loops. It can be seen that in Fe irradiated at 460°C to a dose of 2.5dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were ~35% and 65% respectively.



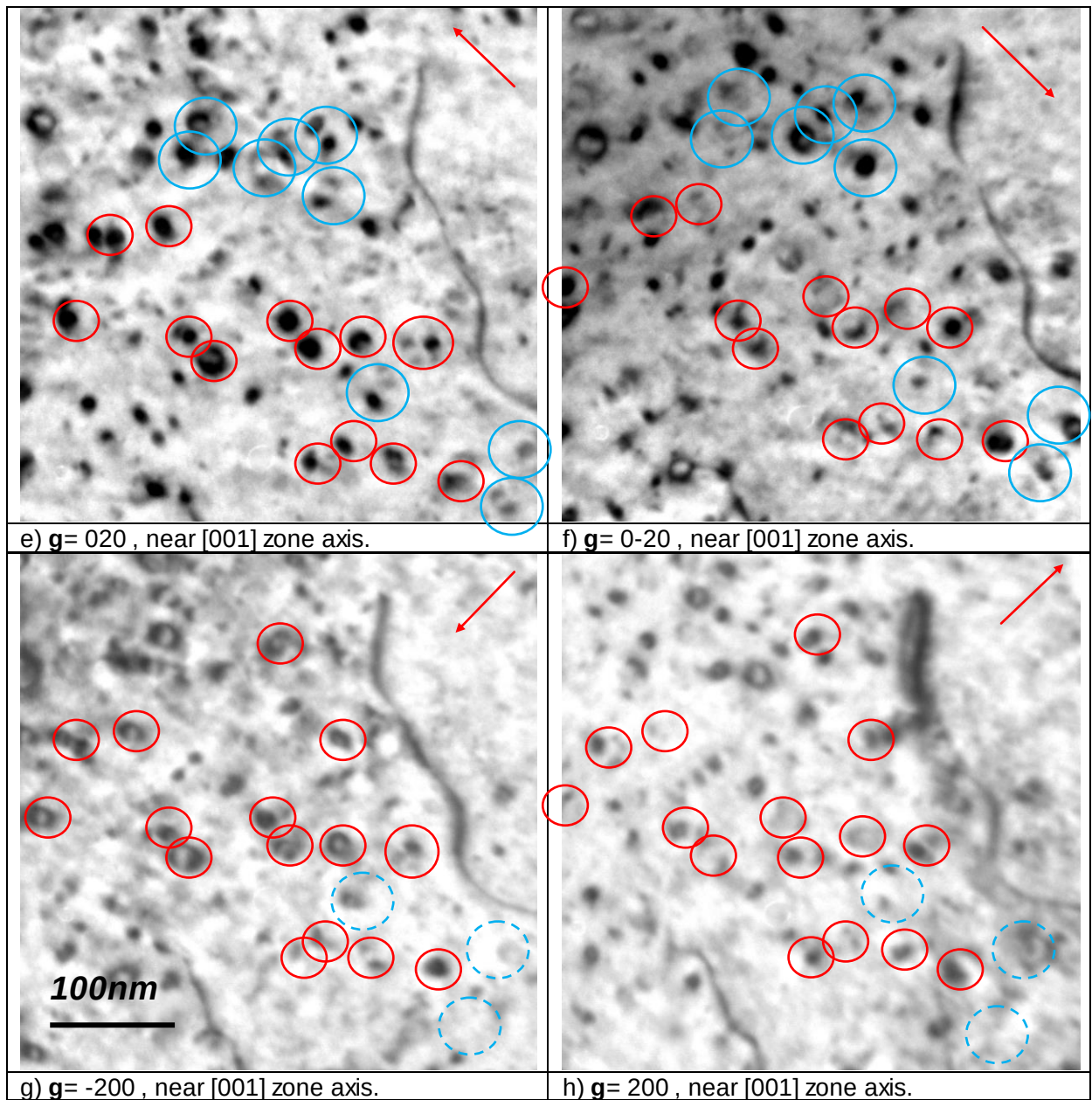


Fig.4.4.4.1. $g.b$ contrast experiments near $[001]$ zone axis, for Fe specimen

irradiated at 460 °C to a dose of 2.5 dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

4.5 Loop Nature Determination

The loop nature determinations in this work were carried out by the inside-outside technique. The full description of the method can be found in chapter 3.4. In this chapter, three successful attempts were made to determine the nature of the loops produced in Fe at different irradiation conditions.

4.5.1 Characterisation of 400°C/7dpa specimens

Fig.4.5.1.1 shows the inside-outside contrast experiments for an Fe specimen irradiated at 400°C to 7dpa. Loop B shows outside contrast in **image (b, d)**, and loop C shows outside contrast in **image (a, c)**. With the aid of the **g.b** analyses in **Fig.4.4.2.1**, the Burgers vector of B is $\pm \frac{1}{2} [1\bar{1}1]$, while the Burgers vector of C is $\pm \frac{1}{2} [11\bar{1}]$.

In this case, the [111] foil orientation is “safe” for both loops B and C. Therefore even if loops B and C are non-edge loops, they will show the same image contrast as a pure edge loop with the same Burgers vectors. Detailed discussion on safe orientations can be found in **chapter 3.4**.

The condition for the generation of outside contrast is $(\mathbf{g} \cdot \mathbf{b})s_g > 0$. s_g is positive, since the micrographs were taken under bright-field kinematical conditions.

For loop B: $\mathbf{b} \cdot (\mathbf{0}\bar{1}1) > 0$;

So the Burgers vector of loop B is $+\frac{1}{2} [1\bar{1}1]$. B is interstitial in nature.

For loop C: $\mathbf{b} \cdot (\mathbf{0}1\bar{1}) > 0$;

So the Burgers vector of loop C is $+\frac{1}{2} [11\bar{1}]$. C is also interstitial in nature.

In addition, a tilting experiment was done to determine the loop habit plane. In this case, the specimen was tilted along $\mathbf{g} = \mathbf{0}1\bar{1}$ from [111] to [011]. Images (a, b) were taken close to the [111] orientation, and images (c, d) were taken close to the [011]

orientation. It can be seen that loop B turns from an ellipse shape to a round shape as the specimen was tilted, which shows that its habit plane is between the $(1\bar{1}1)$ and (011) plane. The projection image indicates that its habit plane is closer to $(1\bar{1}1)$ than (011) . The projection image of loop C shrinks as the specimen was tilted from $[111]$ to $[011]$, which indicates that it lies on or very close to the $(11\bar{1})$ plane. Both of these loops are therefore close to edge. Of 6 other loops analysed at this irradiation condition, all were interstitial.

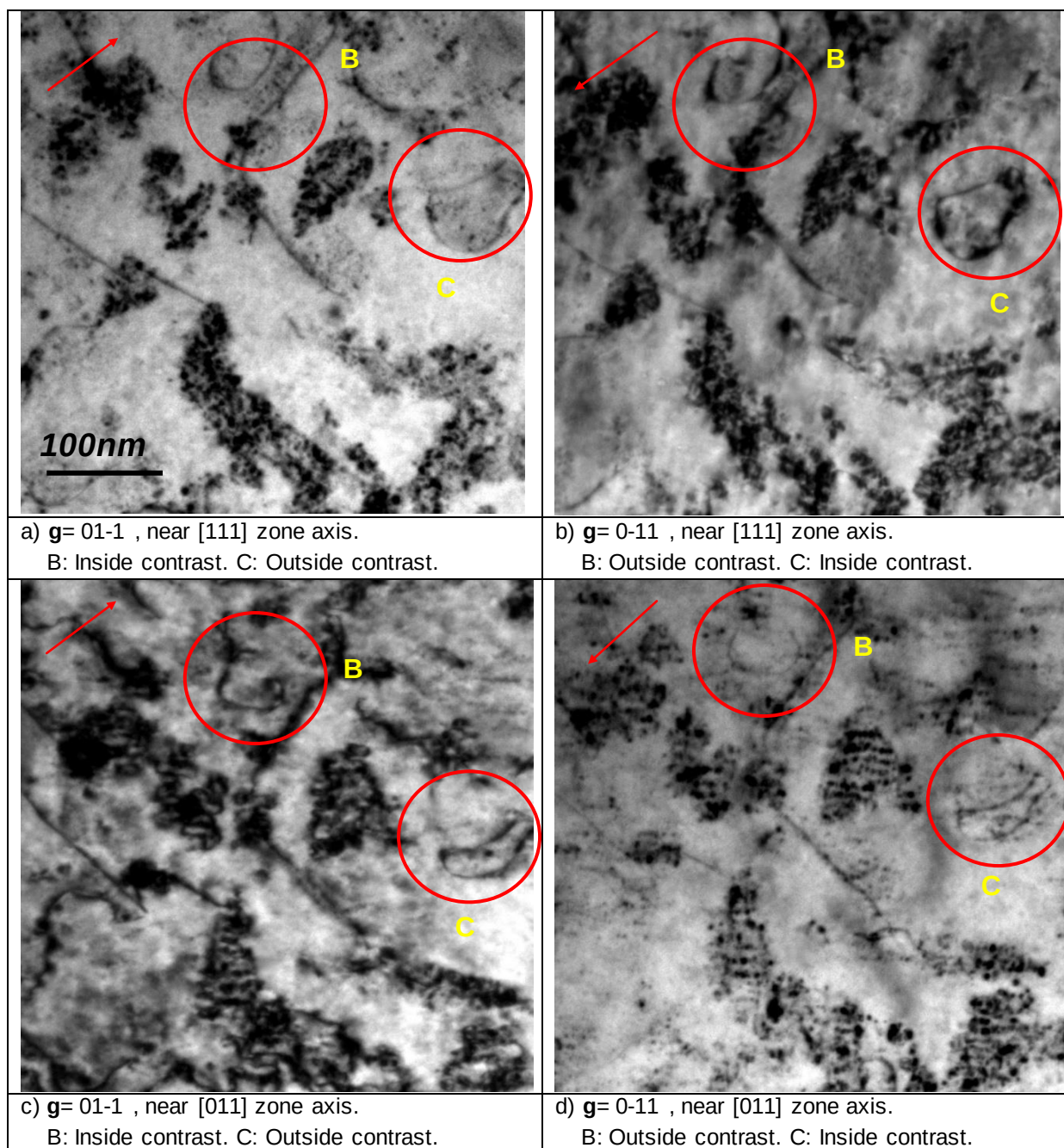


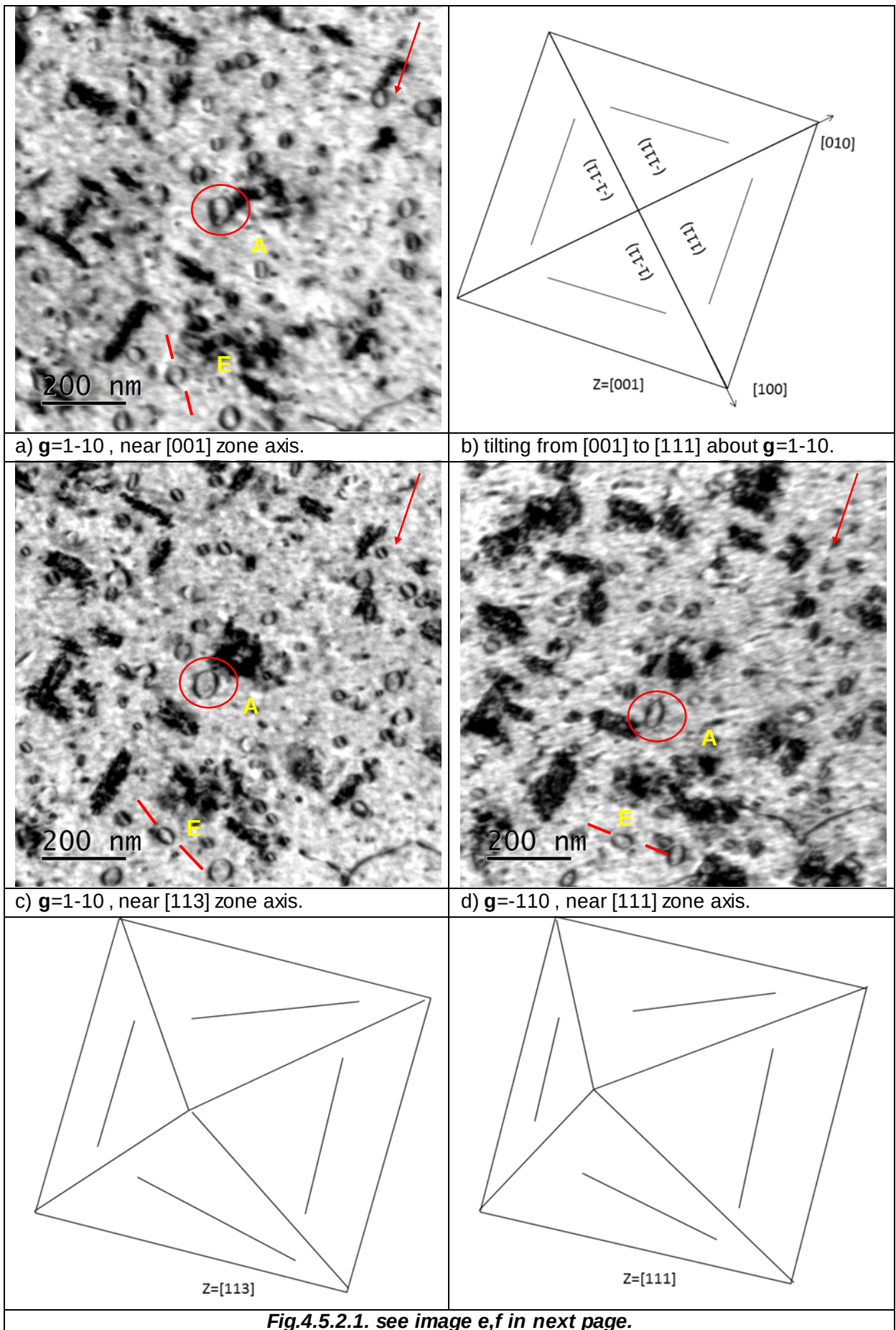
Fig.4.5.1.1. Inside-outside contrast experiments for Fe specimen irradiated at 400°C to 7dpa.

4.5.2 Characterisation of 400°C/2.5dpa specimens

Fig.4.5.2.1. shows the TEM micrographs for the determination of loop nature and habit planes in an Fe specimen irradiated at 400°C to 2.5dpa. Burgers vector analysis in this specimen has been presented in **chapter 4.4.3, Fig.4.4.3.1.** The circled loops in **Fig.4.5.2.1** are visible in $g=020$, 200 and $\bar{1}10$, whilst invisible in $g=110$, which shows that they have Burgers vector of type $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$.

Images (a, b, c, d) show a tilting experiment along $g=\bar{1}10$ from $[001]$ to $[113]$ then to $[111]$. Comparing the image projections, it is found that the major axis of loop A turns clockwise from the image taken in $[001]$ to the images taken in $[113]$ and $[111]$. This is consistent with a circle on the $(1\bar{1}1)$ plane. Thus loop A has Burgers vector $\pm \frac{1}{2}[1\bar{1}1]$. The major axis of loop E turns anti-clockwise in the same tilting sequence, which is consistent with a circle on the $(\bar{1}11)$ plane. So loop E has Burgers vector of $\pm \frac{1}{2}[\bar{1}11]$.

In **images (e, f)**, it can be seen that loop A shows outside contrast in $g=200$ and inside contrast in $g=-200$, while loop E shows inside contrast in $g=\bar{2}00$ and outside contrast in $g=200$. The condition for the generation of outside contrast is $(g.b)s_g > 0$. s_g is positive, since all the micrographs were taken under bright-field kinematical conditions. So both loop A and E are both interstitial in nature. Using the same method, 10 further loops were analysed, and all were interstitial in nature.



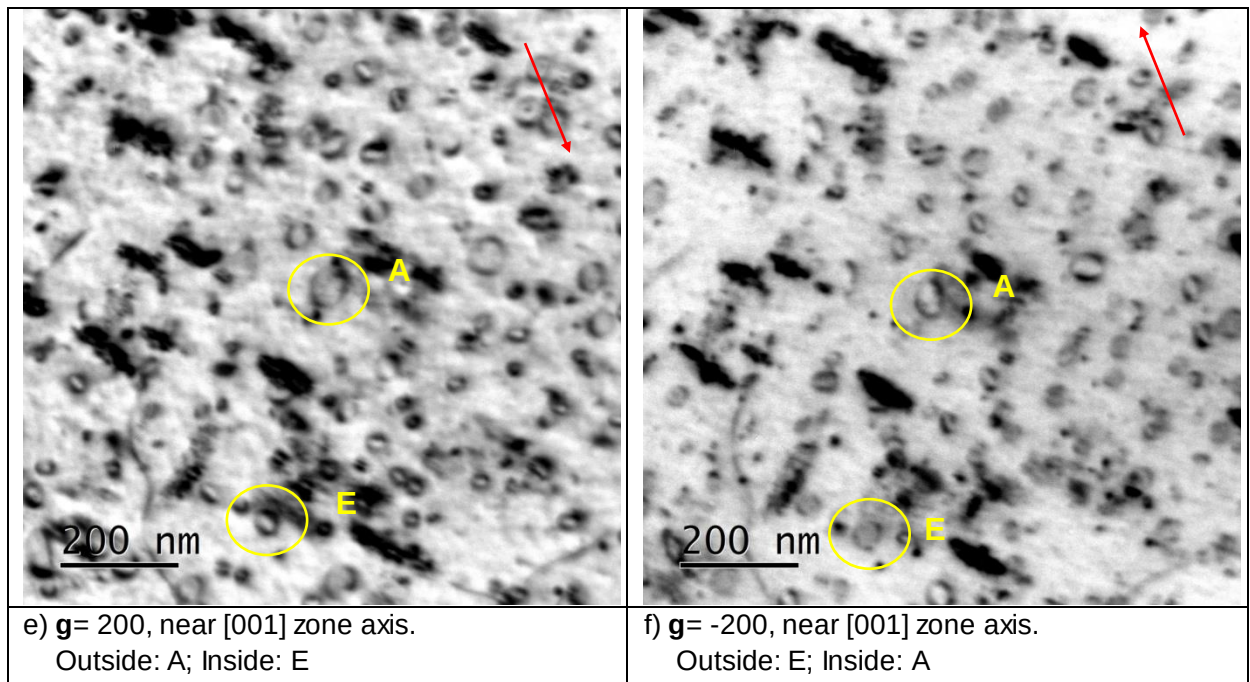
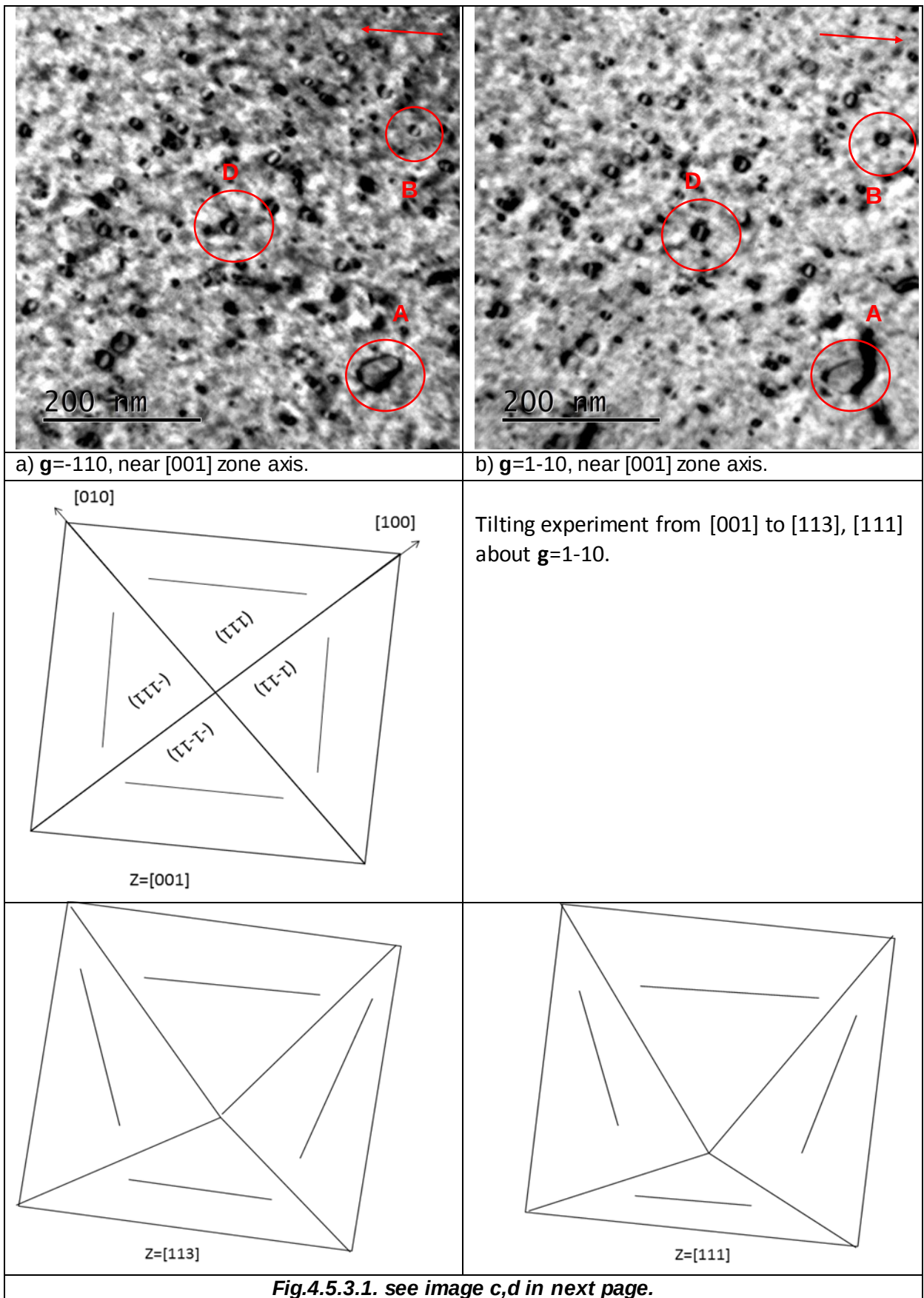


Fig.4.5.2.1. Micrographs for nature and habit plane determination.

4.5.3 Characterisation on 460°C/2.5dpa specimens

Fig.4.5.3.1. shows the TEM micrographs for the determination of loop nature and habit planes in an Fe specimen irradiated at 460°C to 2.5dpa. The specimen was first imaged at the $[001]$ orientation which was also its original orientation during irradiation, then the specimen was tilted towards the $[113]$ and $[111]$ directions. Micrographs with both $+g$ and $-g$ were taken. From the **g.b** analysis in 4.4.4, it can be seen that loops A, B, and D have Burgers vector of $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. From the tilting experiments in **Fig.4.5.3.1**, it can be concluded that the major axis of loop A turned clock-wise when the sample was tilted from $[001]$ towards $[111]$, which is consistent with a circle in the $(1\bar{1}1)$ plane, so loop A has Burgers vector $\frac{1}{2}[1\bar{1}1]$. Since it shows outside contrast in $g=1\bar{1}0$, using the same method described above, it is clear that loop A is interstitial in nature. Loops D and B turn anti-clock wise, which is consistent with a circle in the $(\bar{1}11)$ plane, so these loops have Burgers vector $\frac{1}{2}[\bar{1}11]$. These loops show outside contrast in $g=\bar{1}10$, and so are also of interstitial nature.



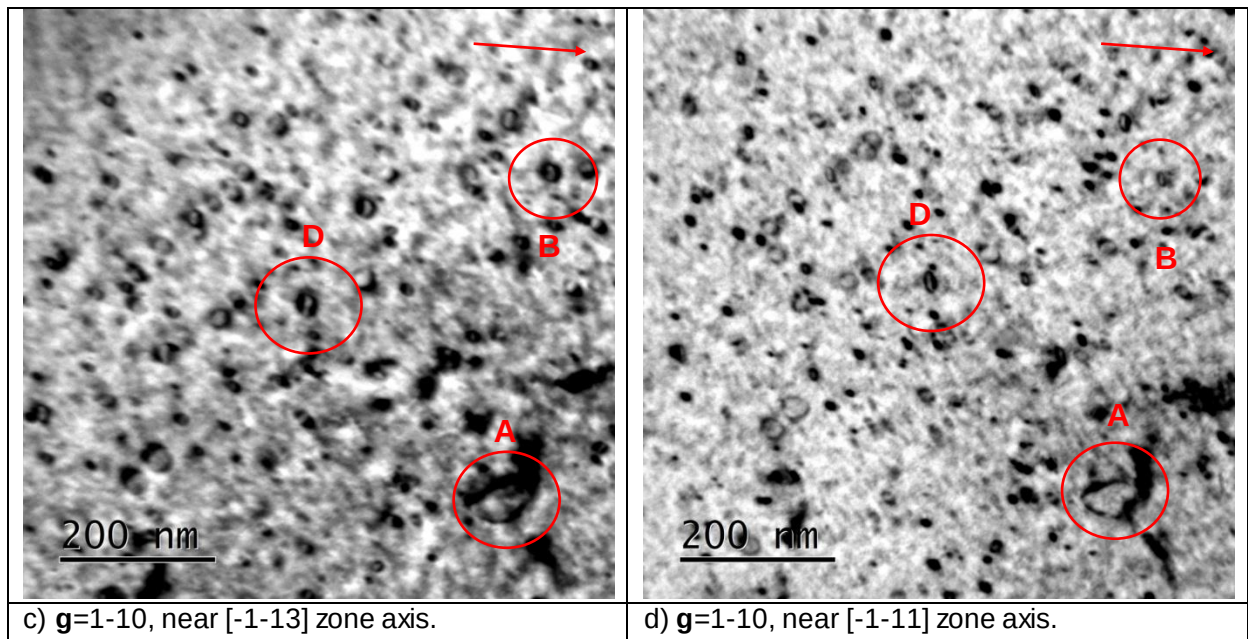


Fig.4.5.3.1. Micrographs for nature and habit plane determination.

4.6 Summary of Results

Microstructures

1. At 300°C (7dpa) the damage microstructures consist of a high density of small dislocation loops with a size of around 2-4nm. At 400°C (2.5dpa,7dpa), dislocation loops were much bigger than those found at 300°C, and more complex features such as finger shape loops, and large loop clusters of several hundred nanometres were formed. At 460°C (2.5dpa), the damage structure returned to be dislocation loops with a size of around 10-20nms, and no complex features were observed apart from some loops strings.
2. At 400°C (2.5dpa), perpendicular loop clusters were observed. Burgers vector analysis showed that these perpendicular loop clusters were formed by $\langle 100 \rangle$ loops. Evidence suggested that these loop clusters formed first from a string of $\langle 100 \rangle$ loops with a size of 5-10nm, then the loop strings grew by absorbing small $\langle 100 \rangle$ loops, which appeared as black dots. Apart from the $\langle 100 \rangle$ loop clusters, most of the large loops (20-30nm) were of $\frac{1}{2}\langle 111 \rangle$ type, which could

form strings and coalesce to form finger-shaped loops with a size of several microns.

3. At 400°C, the changes in microstructure with dose can be explained by the coalescence of $\langle 100 \rangle$ loop clusters and $\frac{1}{2}\langle 111 \rangle$ finger-shaped loops to form large irregular shaped loop clusters with a size of several hundred nanometres.

Loop Sizes:

4. Apart from exceptions caused by clustering of loops, trends were found that as the temperature and irradiation dose increased both the average and maximum loop sizes increased.

Loop Number Density:

5. The number density in Fe peaked at 300°C, and decreased at higher temperatures because of the development of complex microstructures.
6. There was no significant effect of irradiation dose on the loop number densities.

Loop Burgers Vectors:

7. As the temperature increased, the proportion of $\langle 100 \rangle$ loops increased. The implantation dose did not have an effect on loop Burgers vector distributions.

Loop Nature:

8. All loops analysed were of interstitial nature.

Chapter 5: Radiation Damage in Fe5Cr

In this chapter the microstructures of heavy-ion irradiated Fe-5Cr model alloys are presented. The materials were irradiated under the same conditions as pure Fe (**chapter 4**). TEM micrographs were taken to study the irradiation-induced microstructure changes. The observed grains were all oriented close to a [001] zone, and the contrast experiments were performed to gain information on Burgers vector analysis, loop size distributions, defect densities and loop natures as described in **chapter 4**. Results for each irradiation temperature and dose will be described separately and then all results will be combined and discussed in **chapter 8**.

5.1 Irradiation Damage Microstructures

Representative TEM micrographs of the microstructure of Fe5Cr alloys irradiated at different temperatures and doses are shown in **Fig.5.1.1**. The micrographs were taken in a moderately thick area of about 100nm. It can be seen that as the irradiation temperature and dose increase, the damage microstructures developed from isolated small loops to complex structures such as loop clusters, strings and finger-shaped loops. These trends were similar to those in pure Fe, where complex structures developed under some conditions. The average and maximum loop sizes in Fe5Cr alloy were smaller than in pure Fe, while the loop densities in Fe5Cr were much higher than in Fe.

Fig.5.1.2 shows the microstructure of an Fe5Cr foil irradiated at 300°C to a dose of 7dpa. At this temperature, the damage took the form of a high density of dislocation loops with size from 2-20nm. Compared with the Fe foil irradiated at the same temperature and dose, the loop density was significantly higher. In **Fig.5.1.2.b**, a few loop strings of 4-10 small loops can be observed. The strings are aligned along or very close to the [110] direction perpendicular to the diffraction vector $\mathbf{g}=-110$.

Fig.5.1.3. shows how the microstructure developed at 400°C at a dose of 2.5dpa in an area of thickness about 100nm. At this temperature, the loop sizes were much bigger than at 300°C. Loop clusters which were seemed to be perpendicular to each other appeared as well, similar to the clusters found in Fe at the same irradiation condition.

Fig.5.1.4. shows the microstructure of a Fe5Cr foil irradiated at 400°C to a dose of 7dpa. Finger-like loops of a size of ~ a hundred nanometres are observed uniformly distributed within the foil. **g.b** analysis showed that these large loops have $\frac{1}{2}\langle 111 \rangle$ Burgers vectors. TEM micrographs of an Fe5Cr foil irradiated at 460°C to a dose of 7dpa are illustrated in **Fig.5.1.5.** At this temperature, irregular shaped loop clusters of size ~hundreds of nanometres appeared. No loop strings, perpendicular loop clusters or finger-like loops were observed.

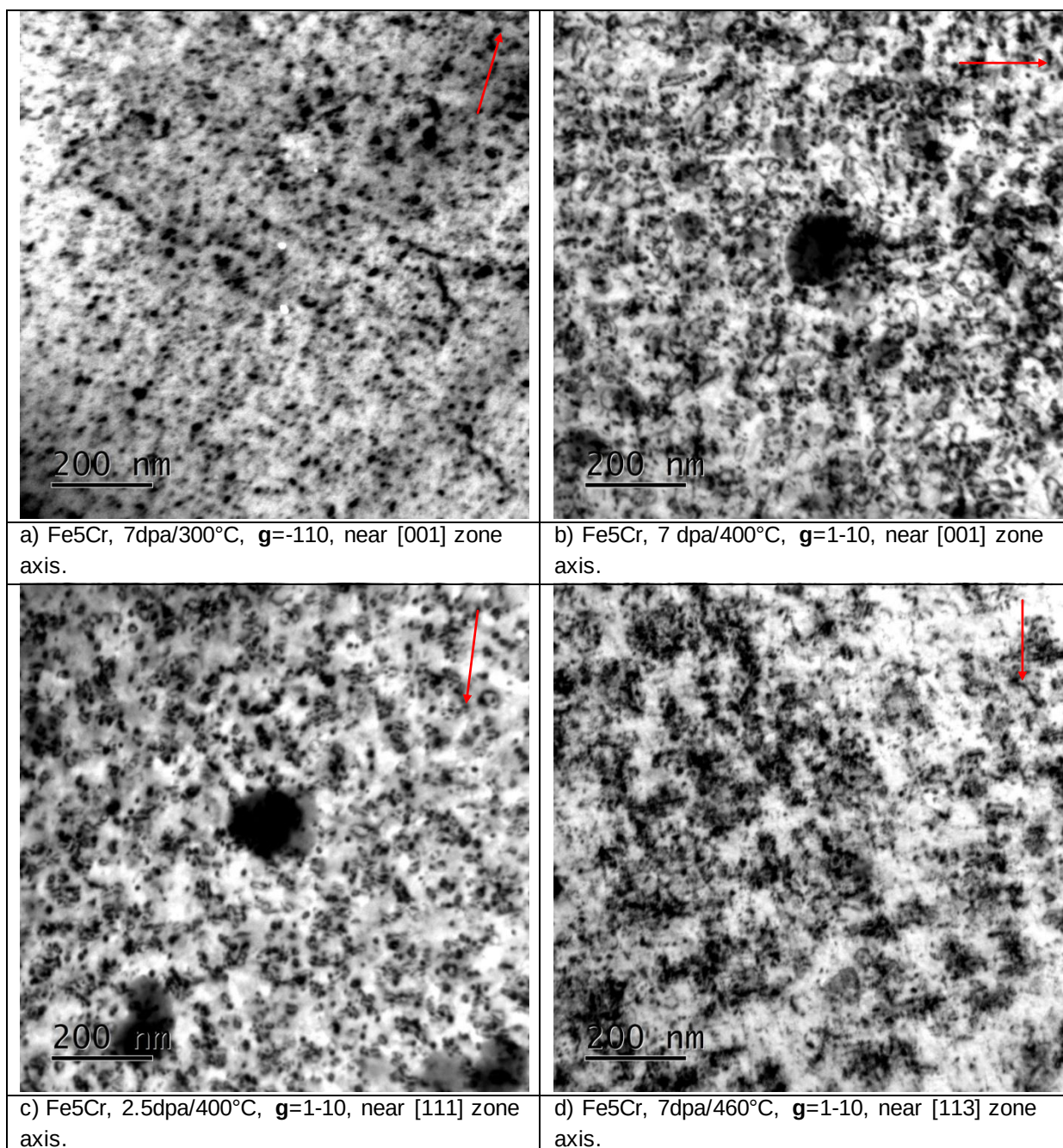


Fig.5.1.1. Micrographs of Fe5Cr samples irradiated at different temperatures and doses.

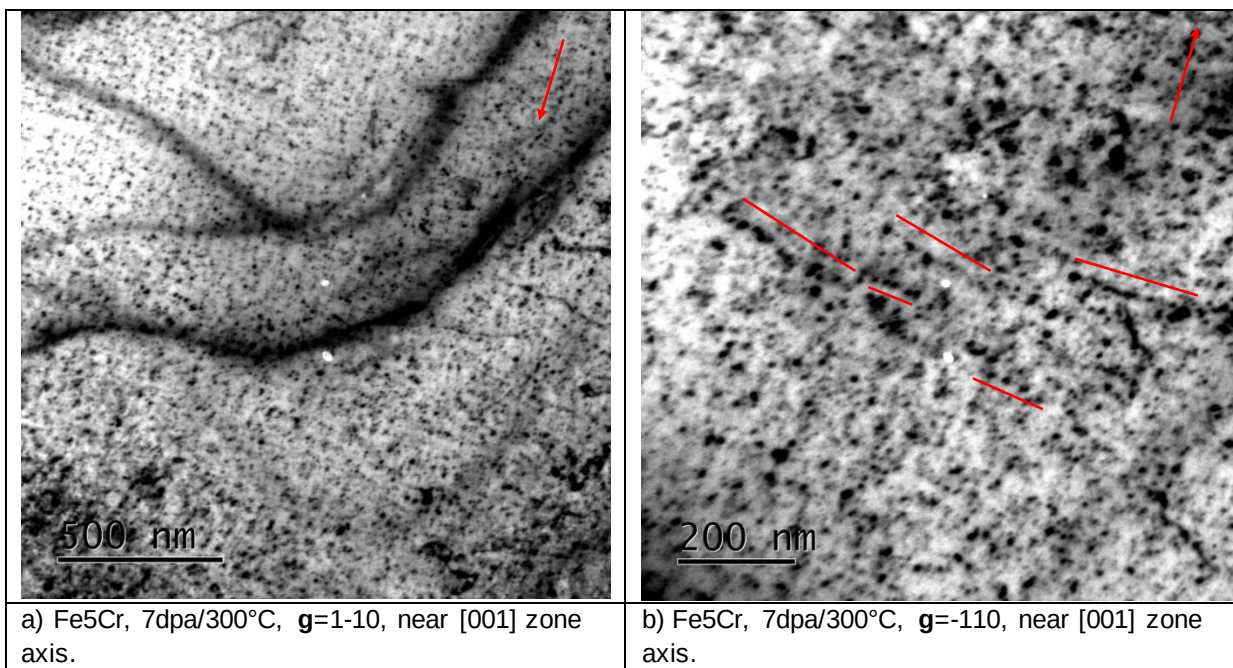


Fig.5.1.2. TEM micrographs of Fe5Cr samples irradiated at 300°C to a dose of 7dpa.

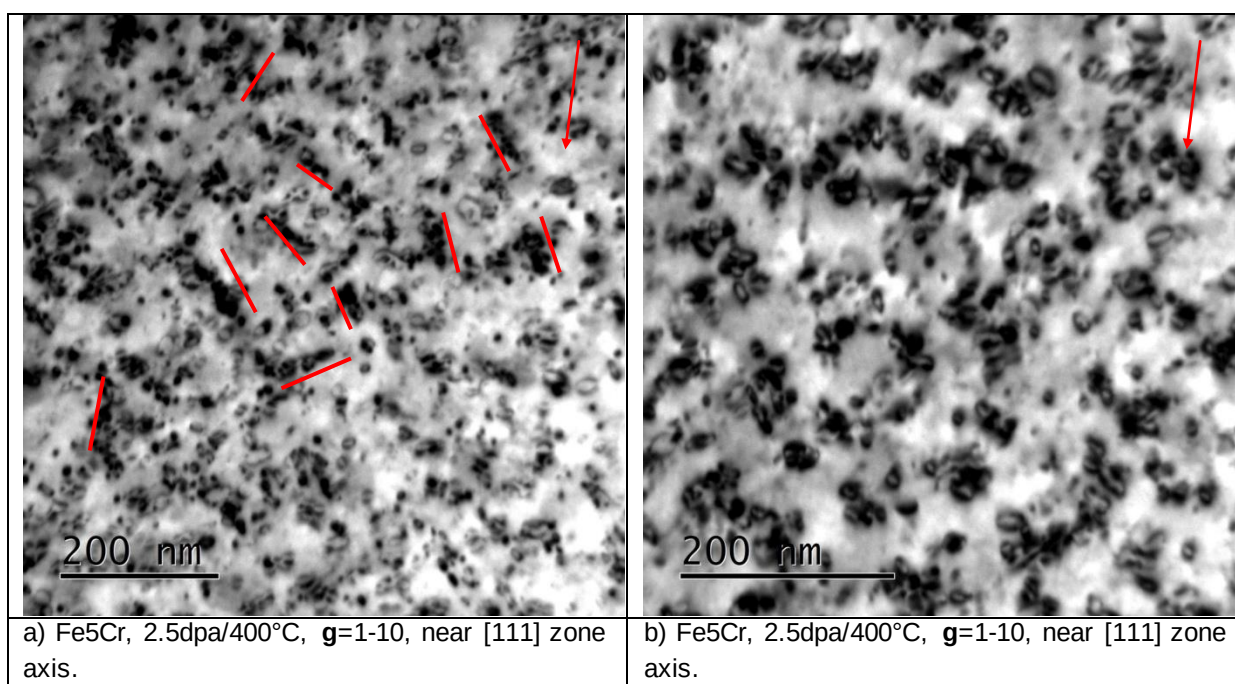


Fig.5.1.3. TEM micrographs of Fe5Cr samples irradiated at 400°C to a dose of 2.5dpa.

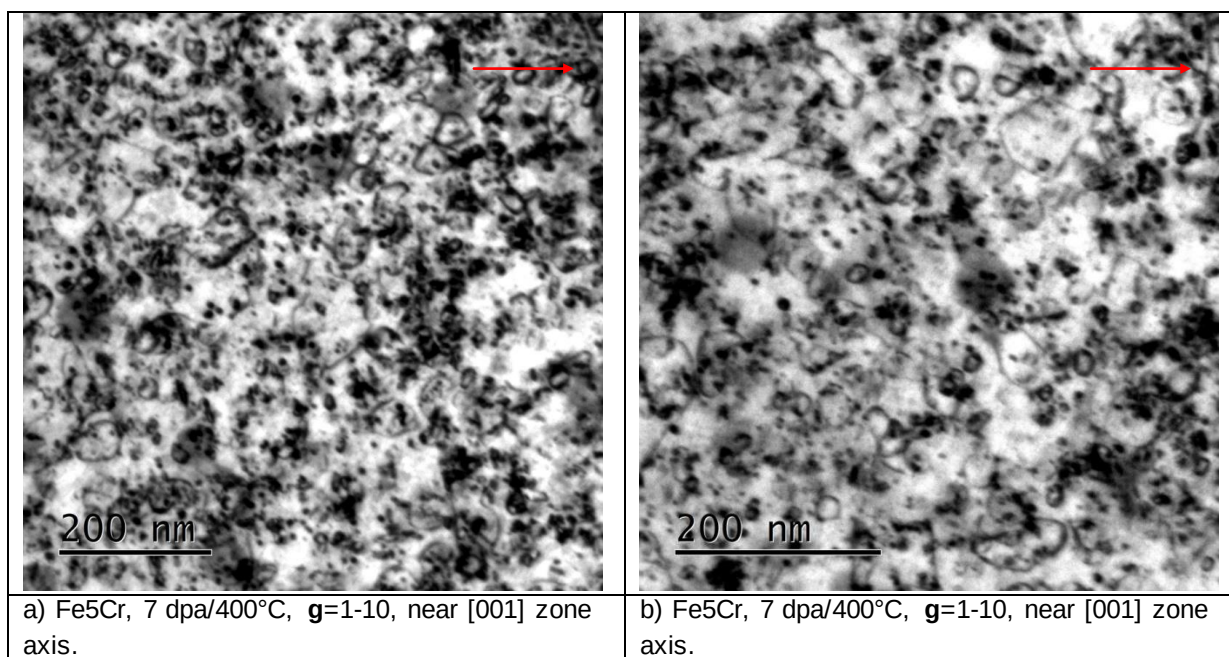


Fig.5.1.4. TEM micrographs of Fe5Cr samples irradiated at 400°C to a dose of 7dpa.

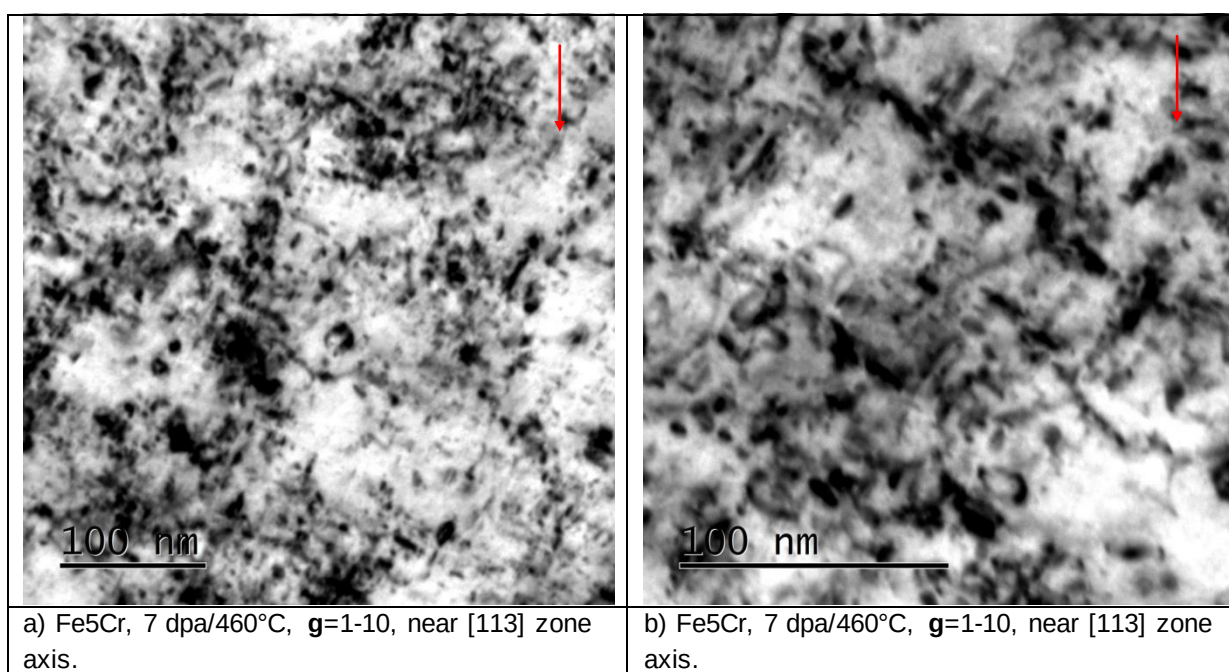


Fig.5.1.5. TEM micrographs of Fe5Cr samples irradiated at 460°C to a dose of 7dpa.

5.2 Loop Image Size Distributions

The sizes of the dislocation loops in the Fe₅Cr specimens were measured by the same method as in pure Fe. In **Fig.5.1.1**, micrographs of Fe₅Cr foils irradiated at different temperatures and doses are presented. In 300°C/7dpa and 400°C/2.5dpa samples, the damage mainly took the form of isolated dislocation loops, while in 400°C/7dpa and 460°C/7dpa samples, large loop clusters and dislocation networks appeared uniformly within the sample. The emergence of clusters and networks explains the unexpected conclusions drawn from the following data in this paragraph. In **Fig.5.2.1**, loop size distributions for 7 dpa implantations at 300°C, 400°C and 460°C, show that loop sizes increase from 300°C to 400°C but decrease from 400°C to 460°C. The average loop sizes shown in **Fig.5.2.2**, indicate the same trend.. However, the maximum loop sizes shown in **Fig.5.2.3** increased by a factor of 4 between 300°C and 400°C, and by a further factor of 1.1 times from 400°C to 460°C. In **Fig.5.2.1**, comparisons between two 400°C implantations at 2.5dpa and 7dpa, show that the increase in implantation dose caused a shift to larger loop sizes. Similar conclusion can be seen in the average loop size distribution and maximum loop size distribution in **Fig.5.2.2** and **Fig.5.2.3** respectively.

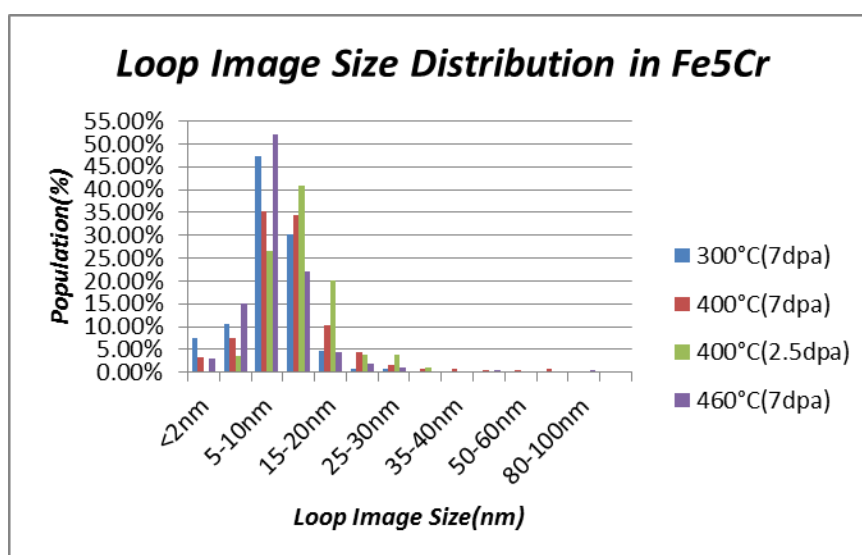


Fig.5.2.1. Loop image size distribution in Fe5Cr specimen irradiated at different temperatures and doses.

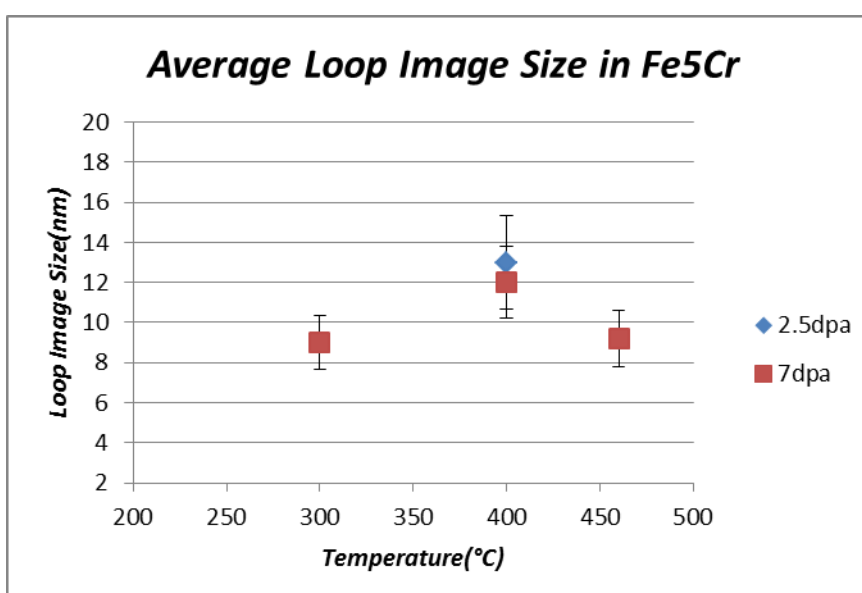


Fig.5.2.2. Average loop sizes in Fe5Cr at different irradiation conditions.

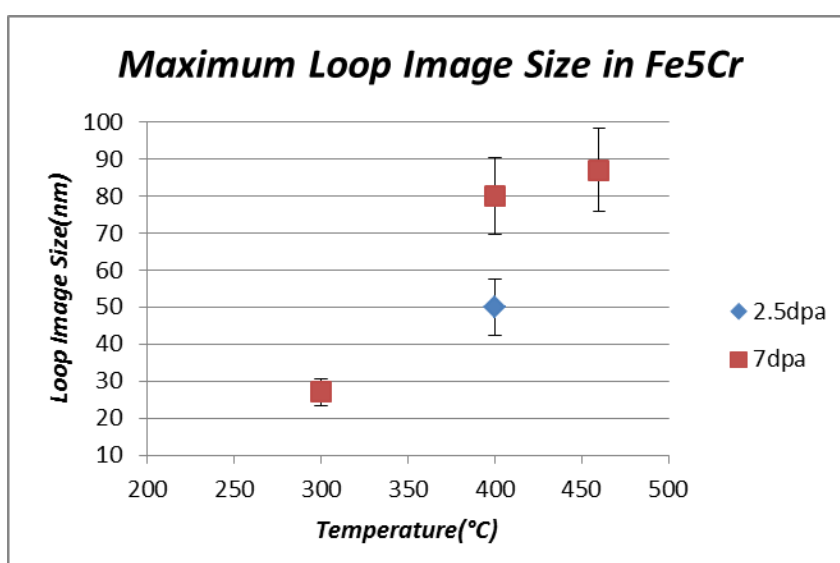


Fig.5.2.3. Maximum loop sizes in Fe5Cr at different irradiation conditions.

5.3 Loop Number Densities

Fig.5.3.1 shows the loop number densities in Fe5Cr based on 800-1000 counts in selected areas of the micrographs which were taken under the same diffraction conditions. Number densities increased with increasing implantation temperature, unlike the trends in Fe samples. At 400°C, it can be concluded that the densities increased with increasing implantation dose.

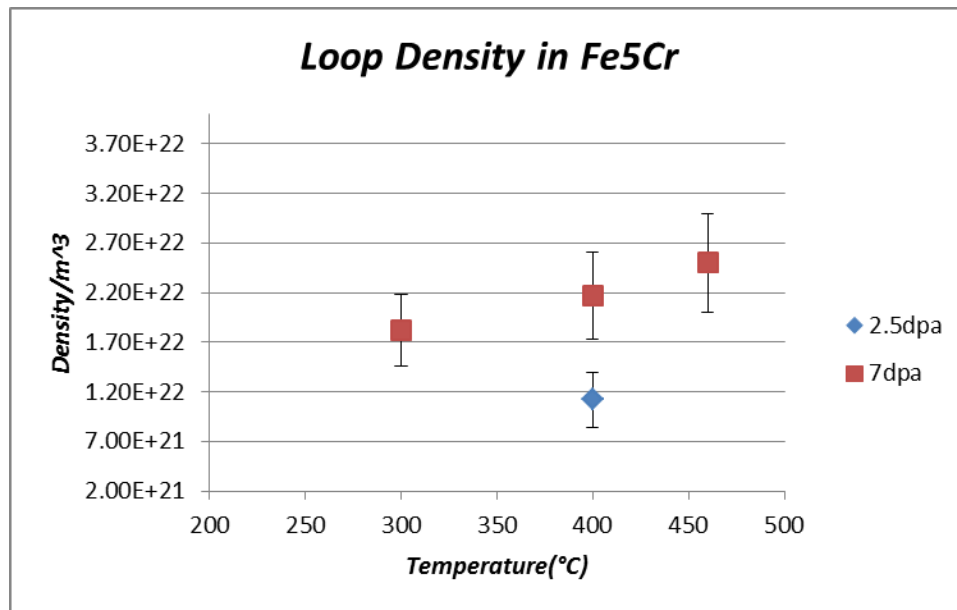


Fig.5.3.1. Loop number densities in Fe5Cr.

5.4 Loop Burgers Vectors

The Burgers vectors analyses for Fe5Cr irradiated were carried out by the same diffraction contrast method described in **chapter 3**. All conclusions are based on analyses of about 100 loops in a selected area.

5.4.1 Characterisation of 300°C/7dpa specimens

Fig.5.4.1.1 shows the Burgers vector analysis of an Fe5Cr specimen irradiated at 300°C to a dose of 7dpa. The micrographs were taken close to [001] foil orientation, with both +**g** and –**g** micrographs. The loops circled in red have $\frac{1}{2}\langle 111 \rangle$ type Burgers vector, while the loops circled in blue have $\langle 100 \rangle$ type Burgers vectors. Of the 108 loops counted, about 70 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, 30 had Burgers vectors of type $\langle 100 \rangle$ and it was not possible to identify 8 loops. In an Fe5Cr specimen irradiated at 300°C to a dose of 7 dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops are therefore ~70% and 30% respectively.

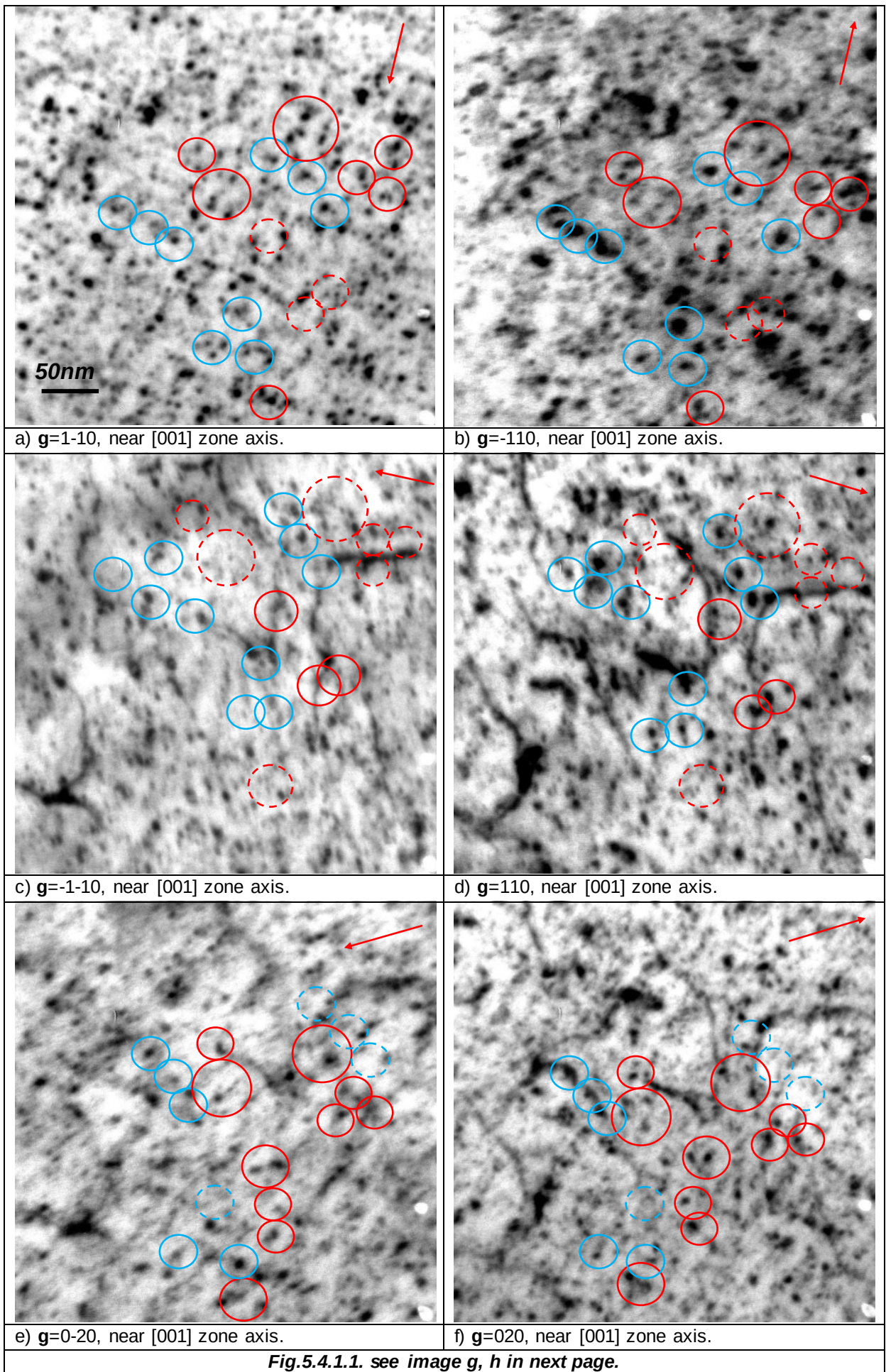


Fig.5.4.1.1. see image g, h in next page.

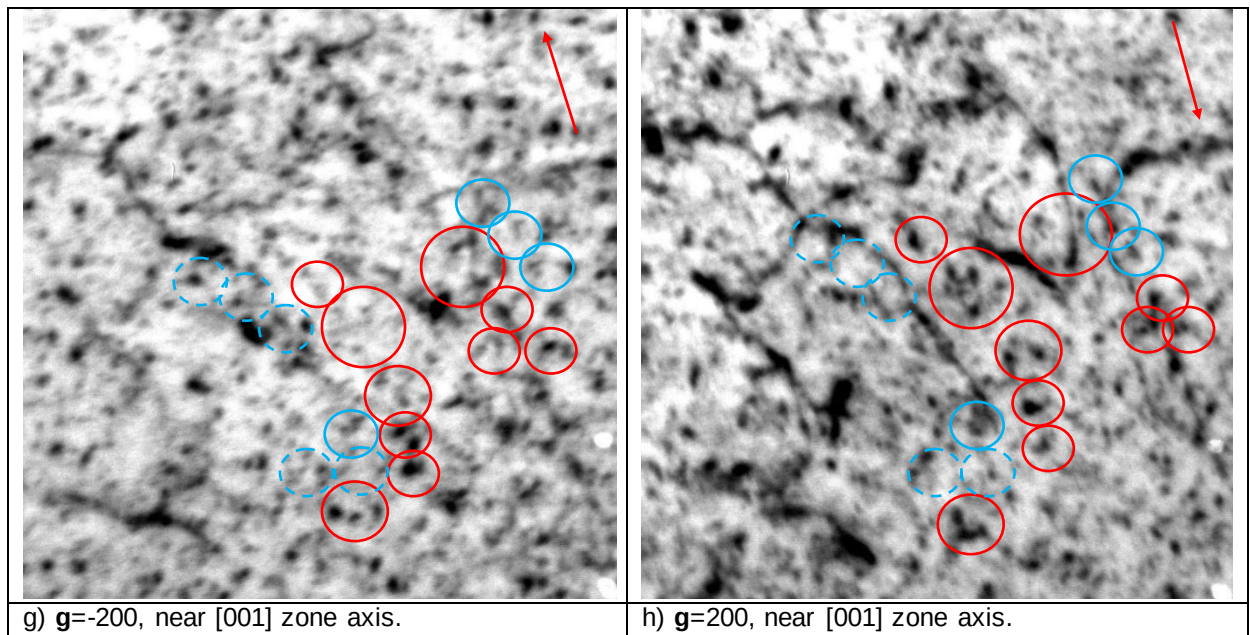


Fig.5.4.1.1. Burgers vector analysis of Fe5Cr irradiated at 300°C to a dose of 7dpa; $b=\frac{1}{2}\langle 111 \rangle$ (red circles) and $b=\langle 100 \rangle$ (blue circles).

5.4.2 Characterisation of 400°C/7dpa specimens

Fig.5.4.2.1 shows a series of images taken near the $[001]$ foil normal orientation in an Fe specimen irradiated at 400°C to a dose of 7dpa. Loops were then analysed as above. The loops circled in red have Burgers vectors of $\frac{1}{2}\langle 111 \rangle$ type, while the loops circled in blue have Burgers vectors of $\langle 100 \rangle$ type. Of the 89 loops appearing in a selected area, 46 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$ and 31 had Burgers vectors of type $\langle 100 \rangle$, and 12 loops could not be identified. In an Fe5Cr specimen irradiated at 400°C to a dose of 7dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were therefore ~60% and 40% respectively.

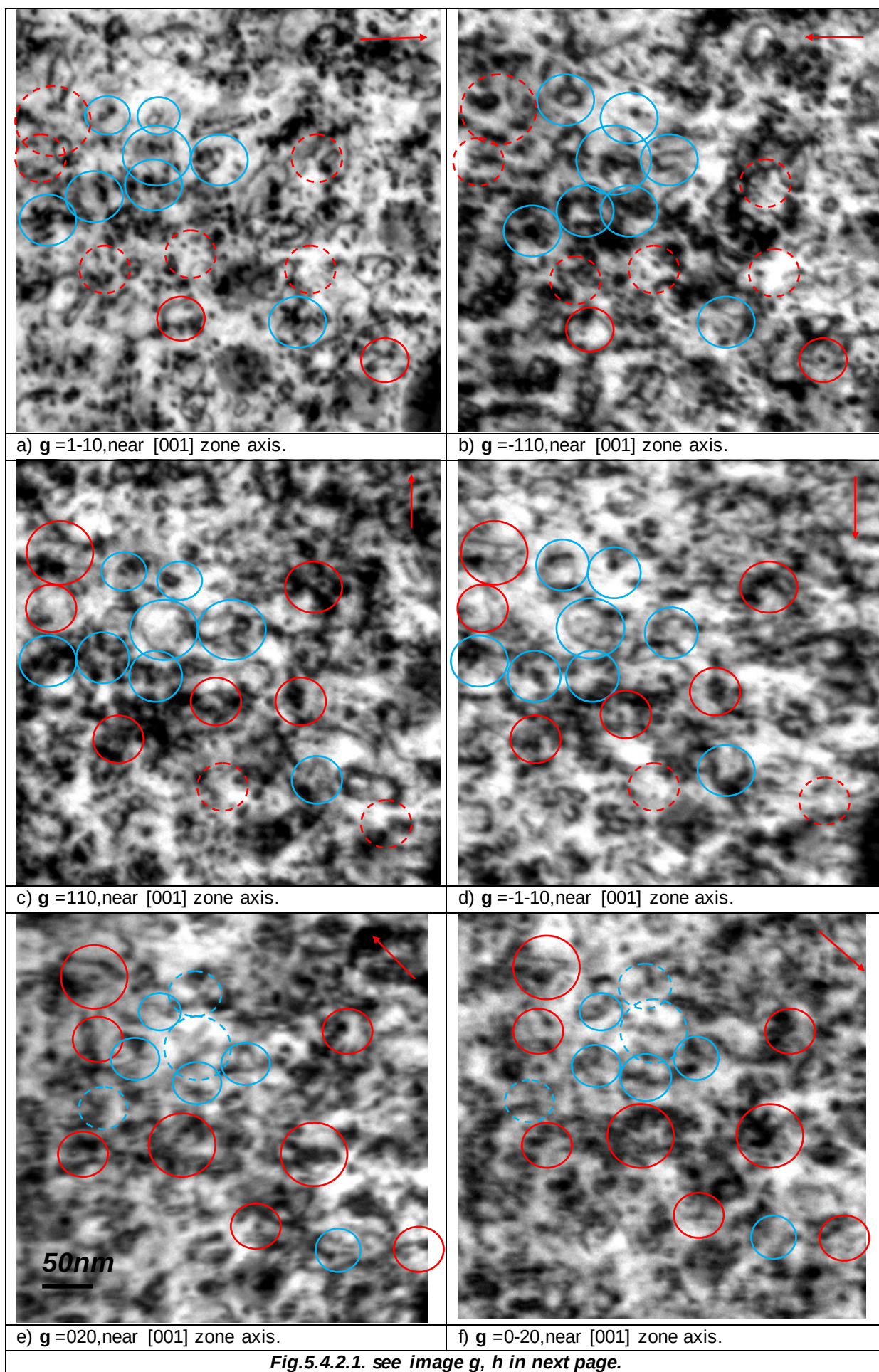


Fig.5.4.2.1. see image g, h in next page.

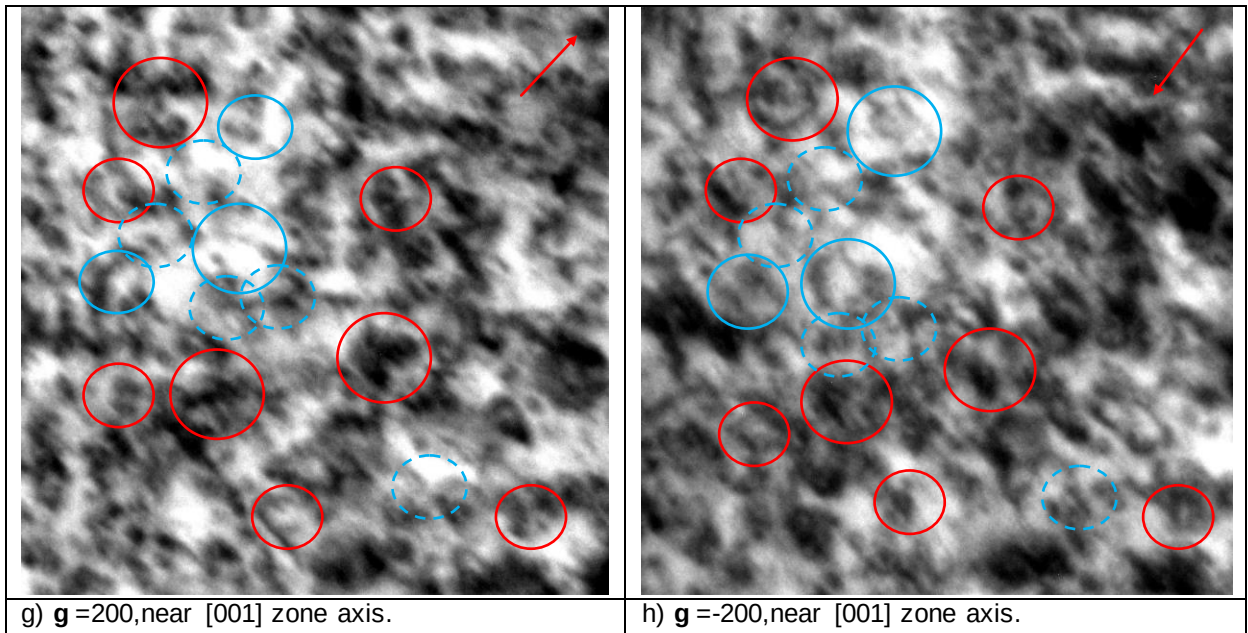
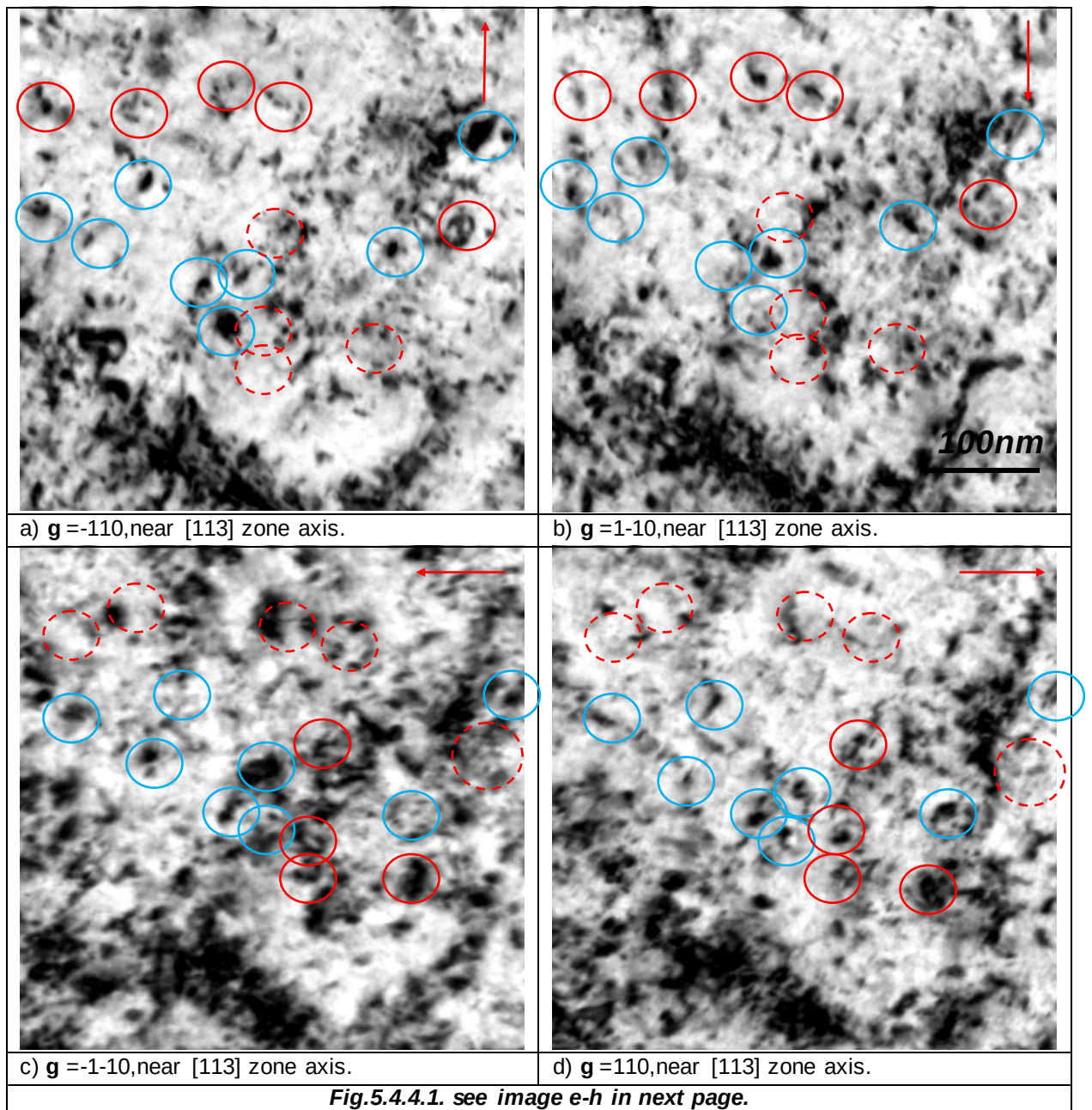


Fig.5.4.2.1. Burgers vector analysis of Fe5Cr irradiated at 400°C to a dose of 7dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

5.4.4 Characterisation of 460°C/7dpa specimens

Fig.5.4.4.1 shows a series of images taken near the [001] foil normal orientation in a Fe5Cr specimen irradiated at 460°C to a dose of 7dpa. Using the method described above, the Burgers vector of the loops were found. Of the 124 loops visible in the area, 55 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, 51 had Burgers vector of type $\langle 100 \rangle$, and 18 loops were too small for their Burgers vectors to be determined.

In an Fe5Cr specimen irradiated at 460°C to a dose of 7dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were therefore ~52% and 48% respectively.



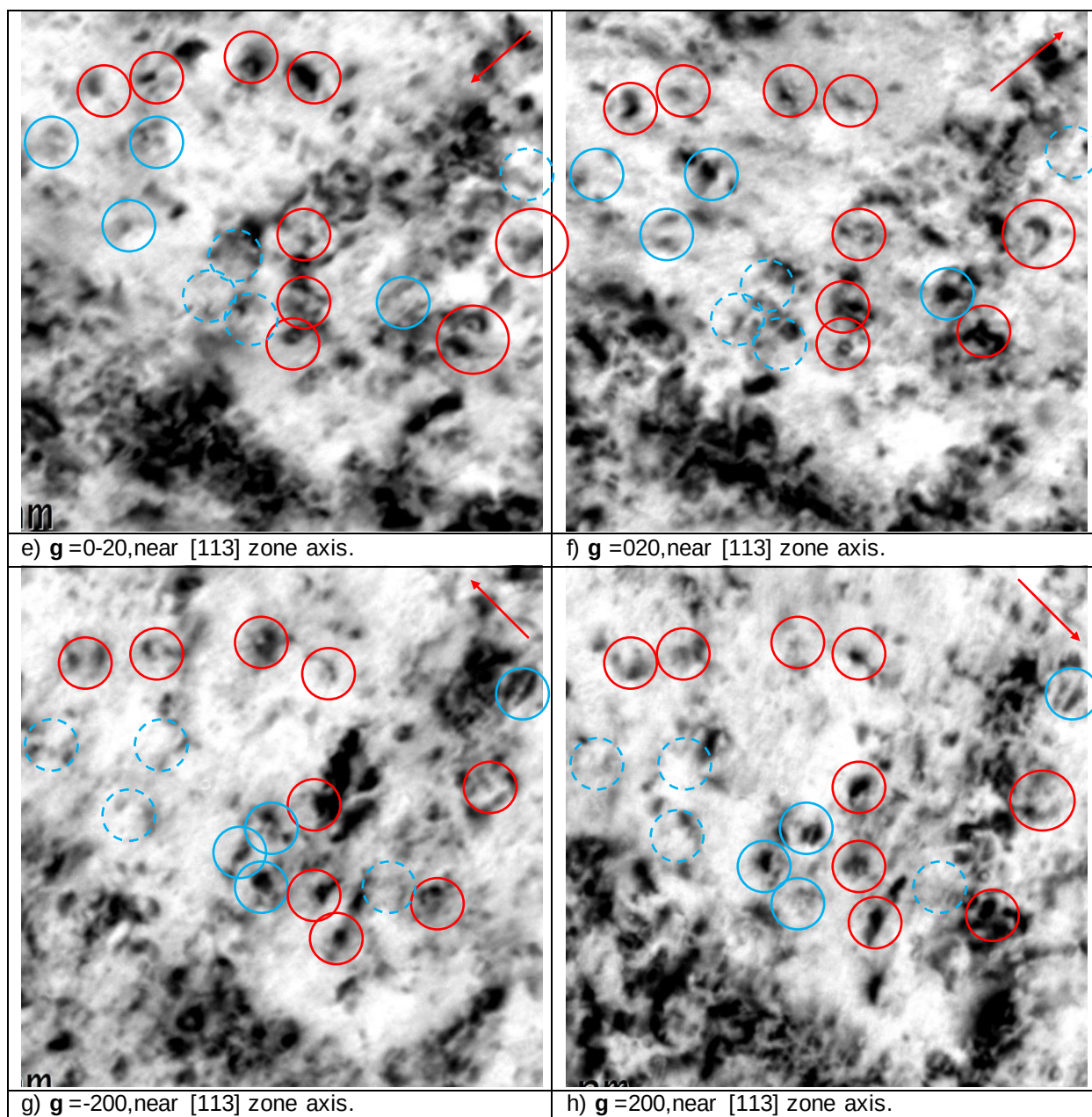


Fig.5.4.4.1. Burgers vector analysis of Fe5Cr irradiated at 460°C to a dose of 7dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

5.5 Loop Nature Determination

In Fe5Cr alloys, most of the loops were too small to show clear inside-outside contrast, and only a few loops in 400°C/7 dpa specimens were suitable for the nature determination. The analysis is presented in the following paragraph.

5.5.1 Characterisation of 400°C/7 dpa specimens

Fig.5.5.1.1 shows the inside-outside method for loop nature determination in a Fe5Cr specimen irradiated at 400°C to 7 dpa. From the loop **g.b** analysis in **5.4.2**, it can be concluded that the circled loops have Burgers vector of type $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. As the specimen was tilted from the [001] zone axis to the [111] zone axis, the major axis of the loops turned anti-clockwise, which is consistent with a circle in the $(\bar{1}11)$ plane. Thus these two loops have Burgers vector of type $\frac{1}{2}[\bar{1}11]$.

Moreover, in **images (a,b)**, the loops show outside contrast in **g**=-110, and inside contrast in **g**= $1\bar{1}0$. The condition for the generation of outside contrast is $(\mathbf{g.b})s_g > 0$. s_g is positive, since all the micrographs were taken under bright-field kinematical conditions. So both loops are interstitial in nature. Using the same method, 6 further loops were successfully analysed, and were all found to be interstitial in nature.

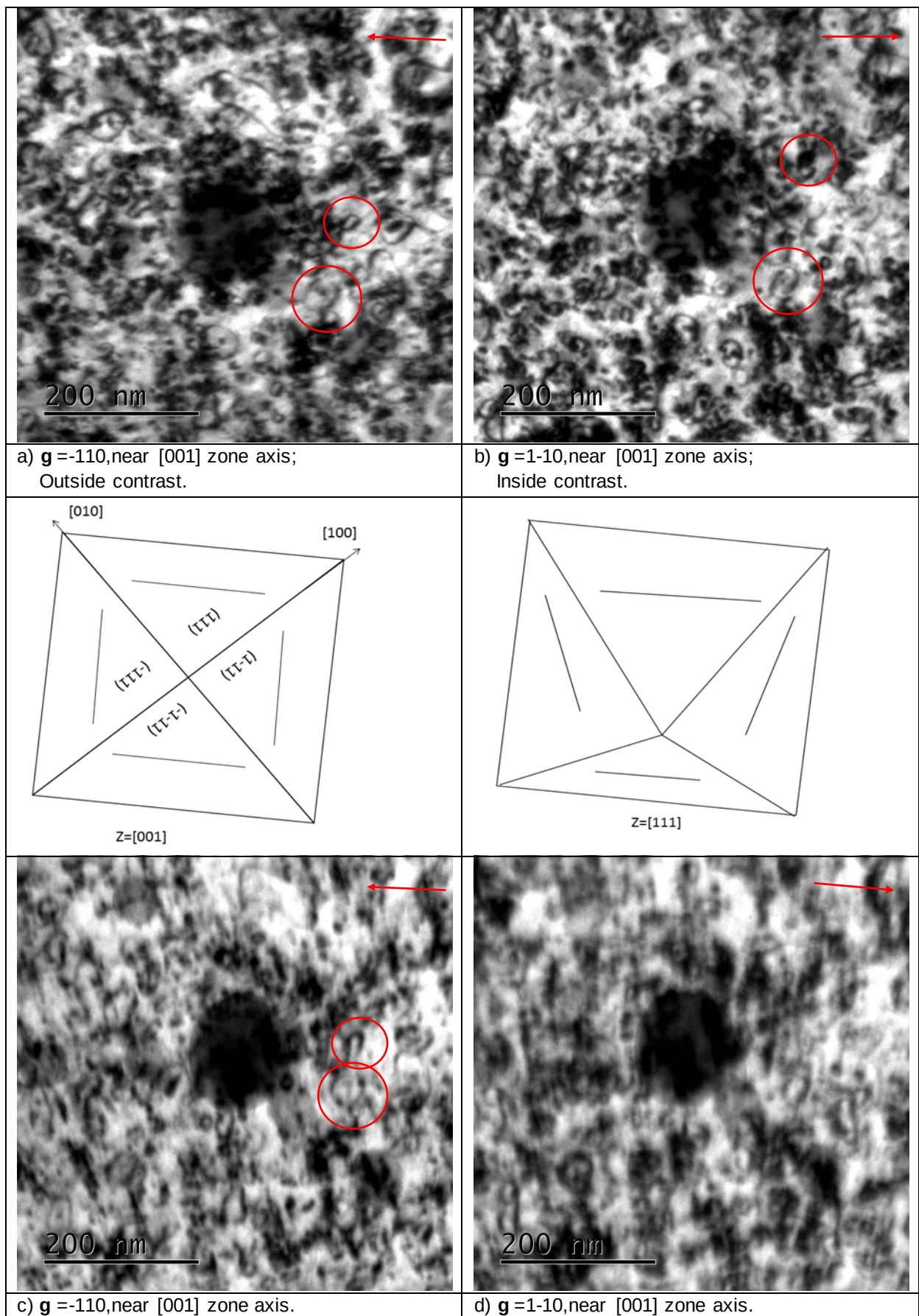


Fig.5.5.1.1. Inside-outside contrast experiments for Fe5Cr specimen irradiated at 400°C to 7dpa.

5.6 Summary of Results

Microstructures:

1. At 300°C (7dpa), radiation damage in Fe5Cr took the form of a high density of dislocation loops with some loop strings. At 400°C (2.5dpa,7dpa), more complex features such as small finger-shaped loops with a size of 30-50nms and loop clusters which were similar to the perpendicular loop clusters in Fe appeared. At 460°C (7dpa), larger areas of loop clusters which were formed by numerous small black dots were observed.

Loop Sizes:

2. Trends found in Fe5Cr were similar to those in Fe: as the temperature and irradiation dose increased both the average and maximum loop size increased.

Loop number densities:

3. The loop densities in Fe5Cr alloys increased as the irradiation temperature increased from 300°C to 460°C.
4. The irradiation dose had a strong effect on loop density in Fe5Cr, unlike pure Fe. A higher irradiation dose caused a higher loop density.

Loop Burgers vectors:

5. There was a trend in Fe5Cr that as the temperature increased, the proportion of $\langle 100 \rangle$ loops increased slightly.

Loop Nature:

6. All loops in Fe5Cr which were analysed were found to be interstitial in nature.

Chapter 6: Radiation Damage in Fe8Cr

In this chapter the microstructure of heavy-ion irradiated Fe-8Cr alloy will be presented. The irradiation conditions were the same as in pure Fe and Fe-5Cr. Again the observed grains were all oriented close to a [001] zone, and the TEM micrograph images were taken with different diffraction vectors to gain information on Burgers vector analysis, loop size distributions, defect densities and nature. All results will be discussed in **chapter 8**.

6.1 Irradiation Damage Microstructures

A series of micrographs of Fe8Cr specimens irradiated from 300°C to 460°C is shown in **Fig.6.1.1** to **Fig.6.1.3**. The damage structures in Fe8Cr irradiated at 300°C were similar to those found in Fe5Cr and Fe under the same condition. The damage at this temperature took the form of small dislocation loops, and no complex microstructures were observed. Compared with pure Fe specimens, Fe8Cr specimens which were irradiated at 400°C and 460°C contained a higher density of much smaller loops.

Fig.6.1.2. shows the damage structure in Fe8Cr irradiated at 400°C to a dose of 7dpa. At this temperature, finger-like loops and irregular shaped dislocation loops were observed. Loop clusters also seemed to form and interact with smaller loops. In **Fig.6.1.2.a** (see blue circles), steeply inclined $\langle 100 \rangle$ loops are imaged.

Fig.6.1.3. shows the damage structure in Fe8Cr irradiated at 460°C to a dose of 7dpa. At this temperature, perpendicular loop chains lying along $\langle 110 \rangle$ directions are observed.

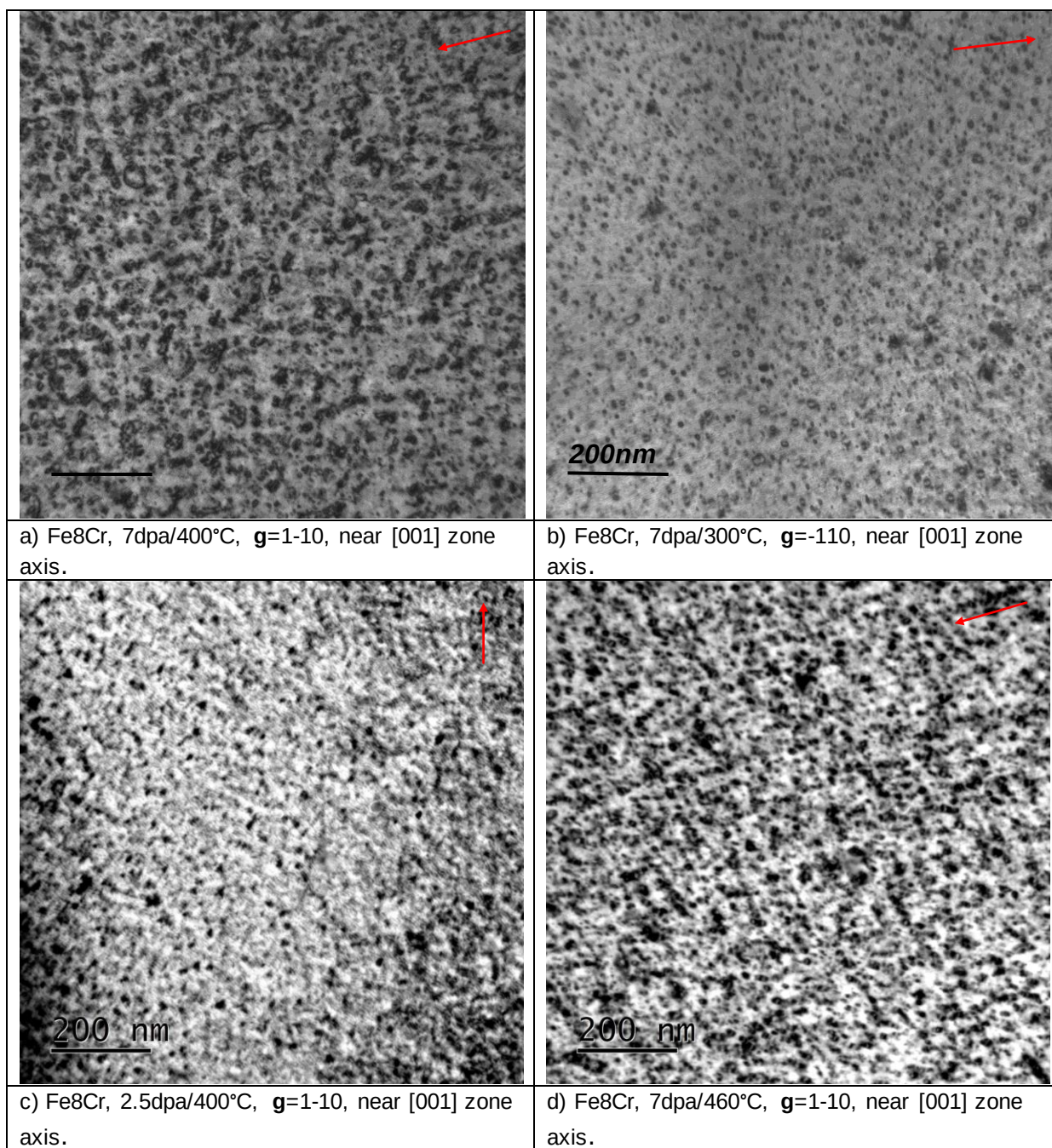


Fig.6.1.1. TEM micrographs of Fe8Cr specimen irradiated at different temperatures and doses.

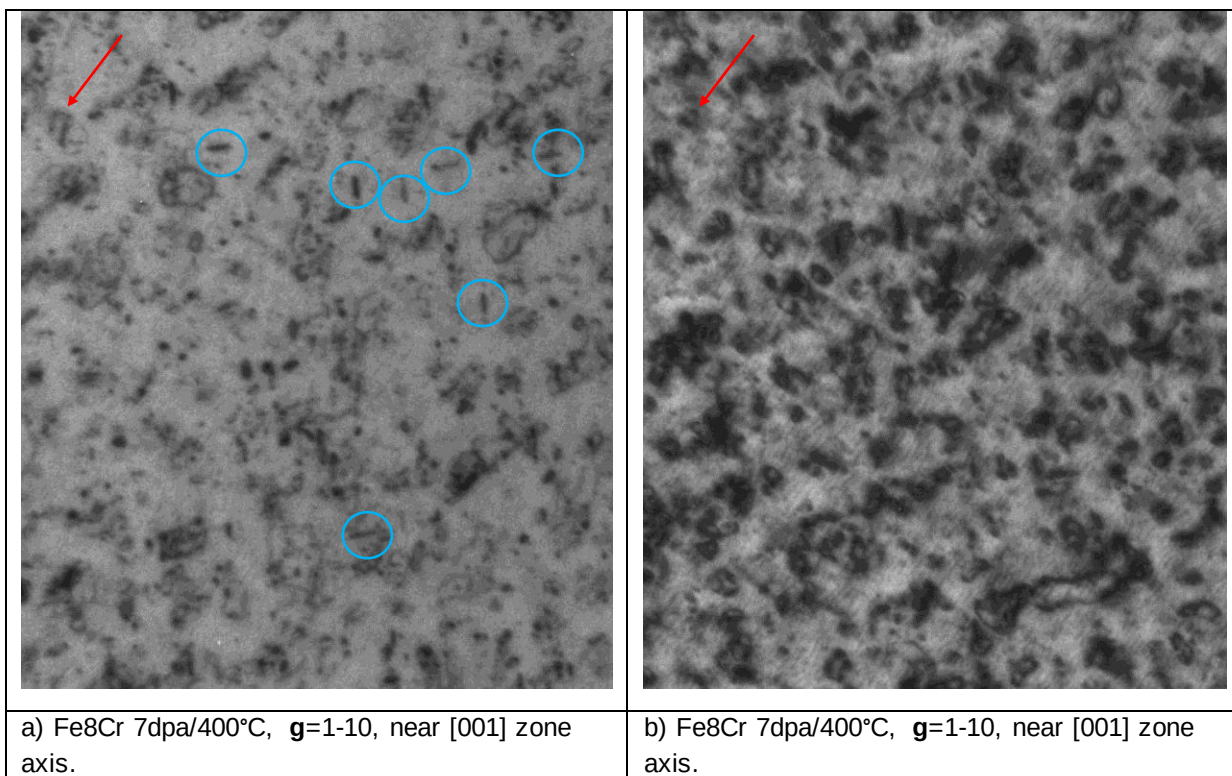


Fig.6.1.2. TEM micrographs of Fe8Cr specimen irradiated at 400°C to a dose of 7dpa.

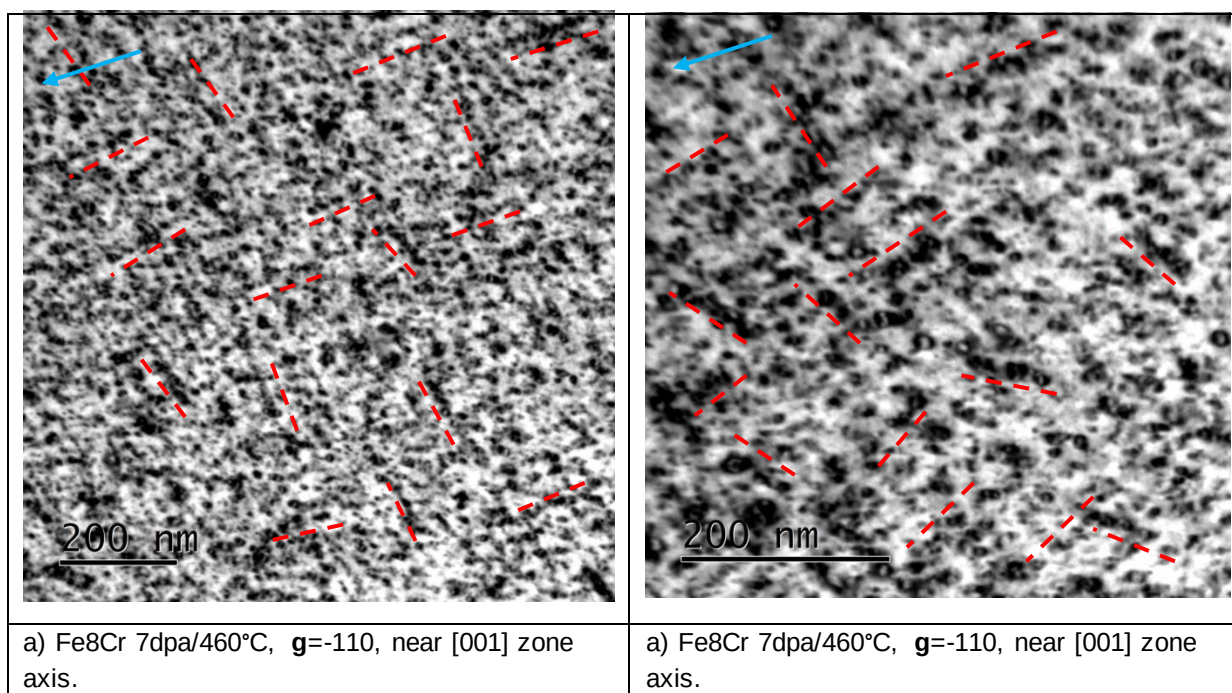


Fig.6.1.3. TEM micrographs of Fe8Cr specimen irradiated at 460°C to a dose of 7dpa.

6.2 Loop Image Size Distributions

Unlike the microstructures of Fe and Fe5Cr which showed complex features, the microstructures in Fe8Cr were composed mainly of dislocation loops and a few loop strings in some cases (see **Fig.6.1.1**). Thus the image size and density measurements shown in the following paragraph are more representative compared with the data in Fe and Fe5Cr.

Fig.6.2.1 shows the loop image size distributions in Fe8Cr. A comparison of the loop image size distributions in 300°C/7dpa, 400°C/7dpa and 460°C/7dpa specimens shows that as the temperature increased, the loop size distribution shifted to larger sizes. An increase in dose also caused larger loop sizes. These trends are consistent with those found in Fe5Cr and Fe, although the emergence of complex features in Fe and Fe5Cr caused a contrary trend at certain irradiation conditions.

Fig.6.2.2. and **Fig.6.2.3** shows the average and maximum loop sizes in Fe8Cr. Both the average and maximum loop sizes increase with irradiation temperature and dose consistent with the loop image size distributions in **Fig.6.2.1**.

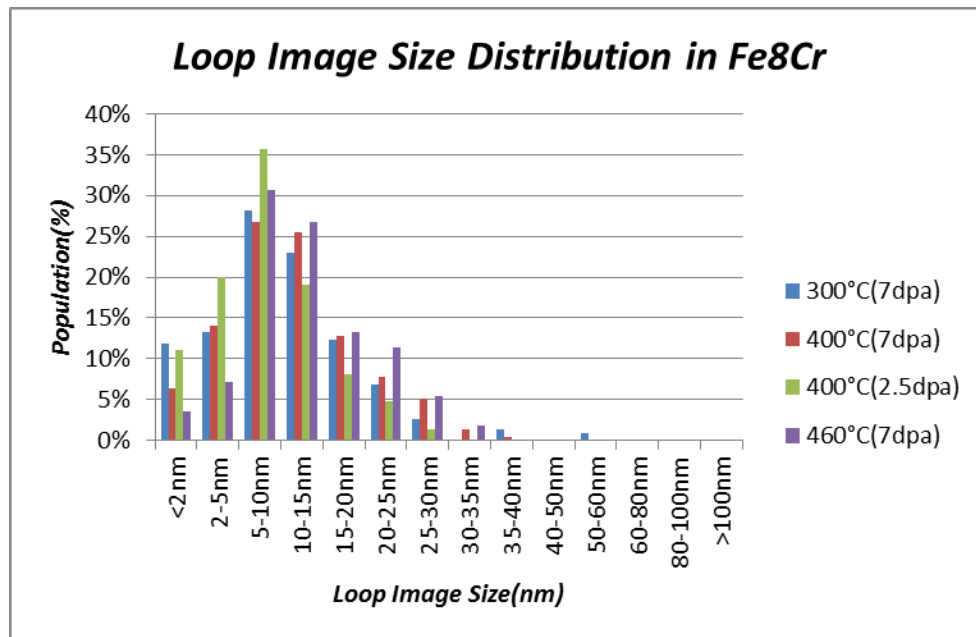


Fig.6.2.1. Loop size distribution of Fe8Cr alloy at different irradiation conditions.

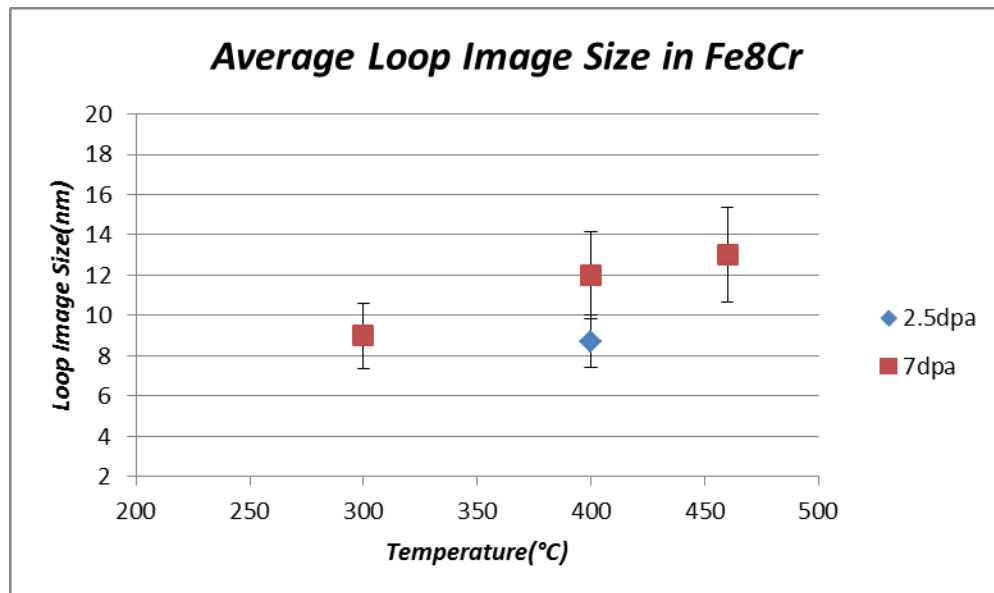


Fig.6.2.2. Average loop image size in Fe8Cr at different irradiation conditions.

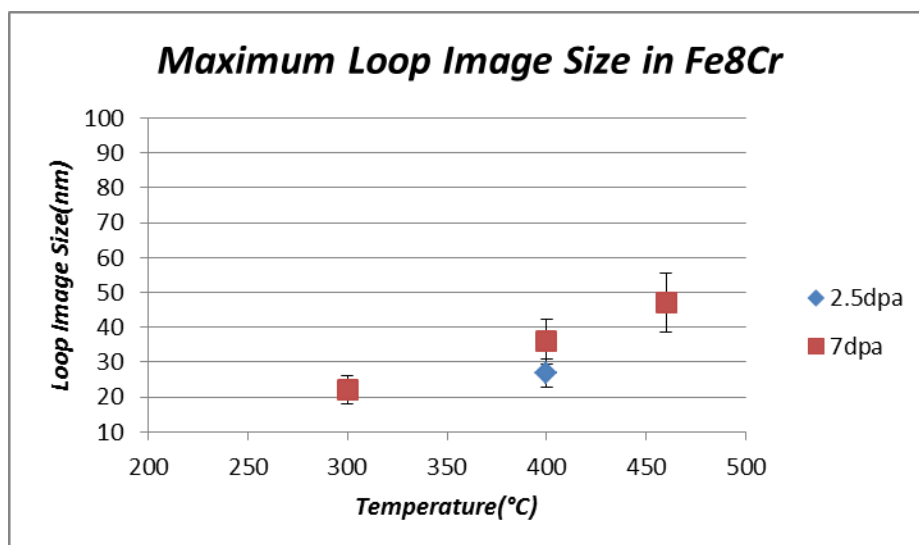


Fig.6.2.3. Maximum loop image size in Fe8Cr at different irradiation conditions.

6.3 Loop Number Densities

The measurements of loop number densities in Fe8Cr were based on at least 500 counts using micrographs taken at the same diffraction conditions. In specimens irradiated to 7dpa, the irradiation temperature had little effect on loop densities. However, at 400°C, an increase in implantation dose did cause an increase in loop density. These trends are consistent with those observed in Fe and Fe5Cr.

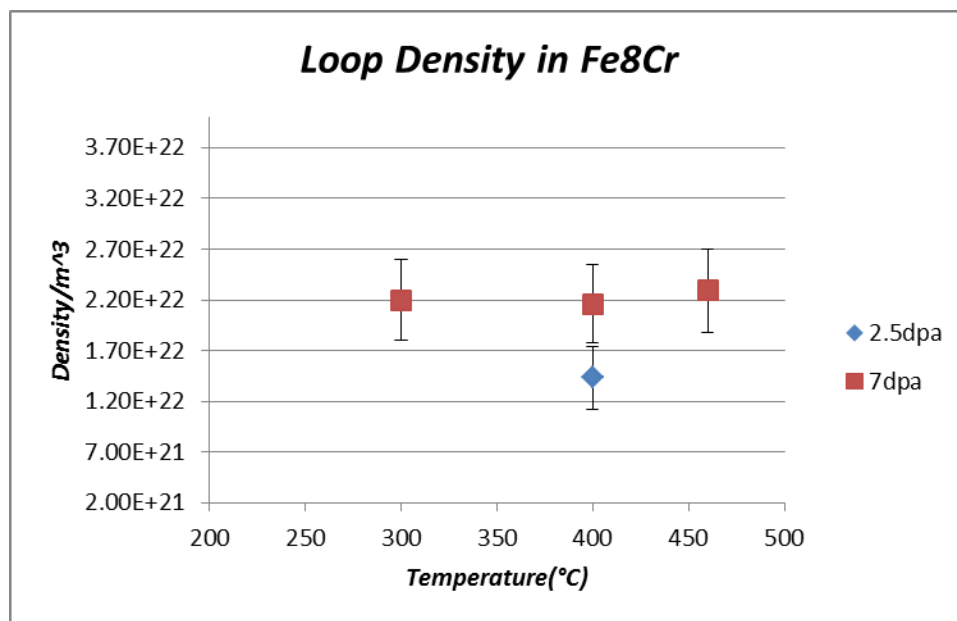


Fig.6.3.1. Loop number densities in Fe8Cr at different irradiation conditions.

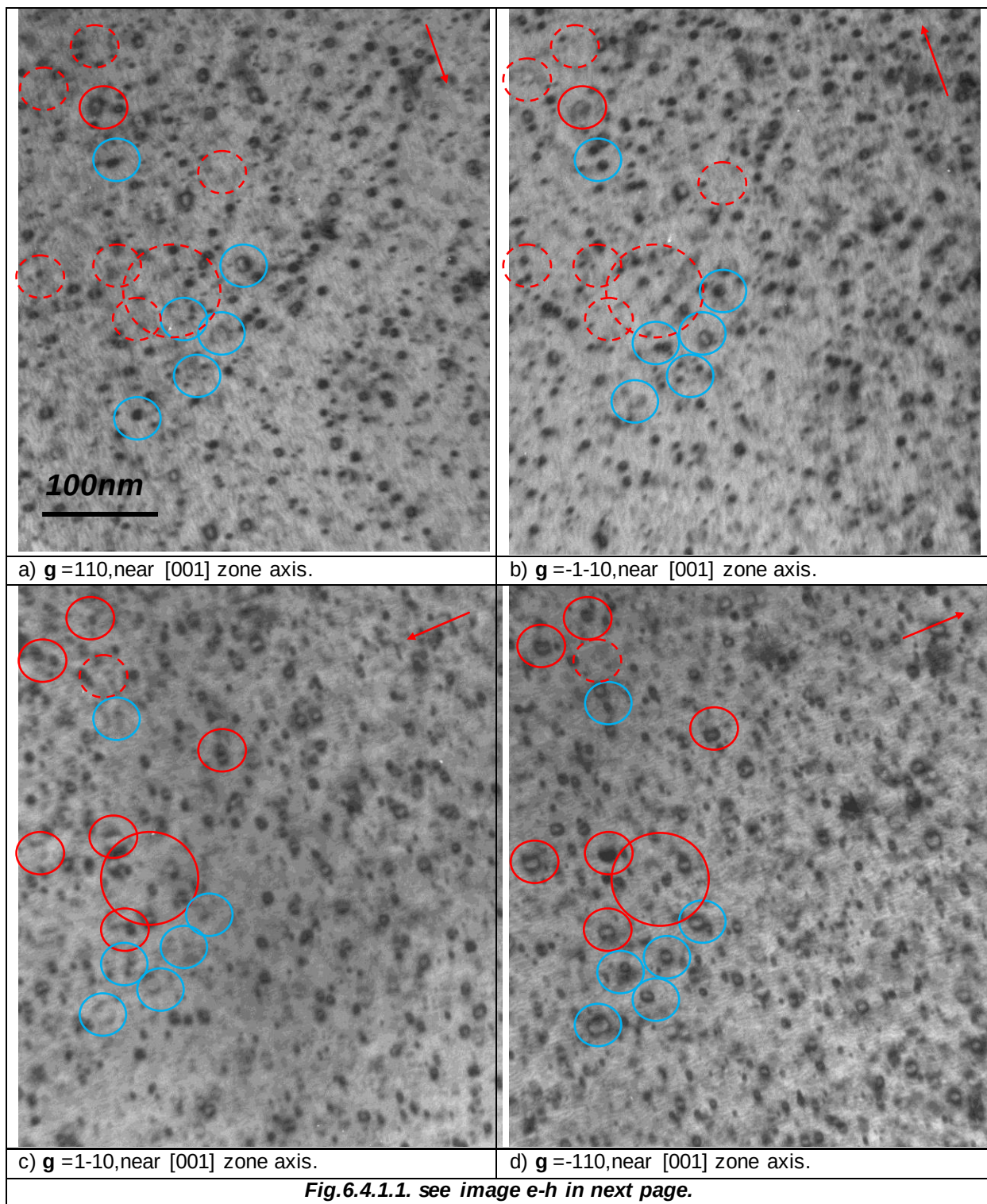
6.4 Loop Burgers Vectors

The Burgers vectors of dislocation loops in Fe8Cr were characterised by the diffraction contrast method described in previous chapters.

6.4.1 Characterisation on 300°C/7dpa specimens

Fig.6.4.1.1 shows the **g.b** analysis carried out in Fe8Cr irradiated at 300°C to 7dpa.

The red circles mark loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vectors and $\langle 100 \rangle$ loops are marked with blue circles. Of the 137 loops analysed, it was found that 78 loops had $\frac{1}{2}\langle 111 \rangle$ type Burgers vectors and 34 had $\langle 100 \rangle$ type Burgers vectors, whilst 25 loops could not be identified. In Fe8Cr irradiated at 300°C to a dose of 7dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were therefore ~70% and 30% respectively.



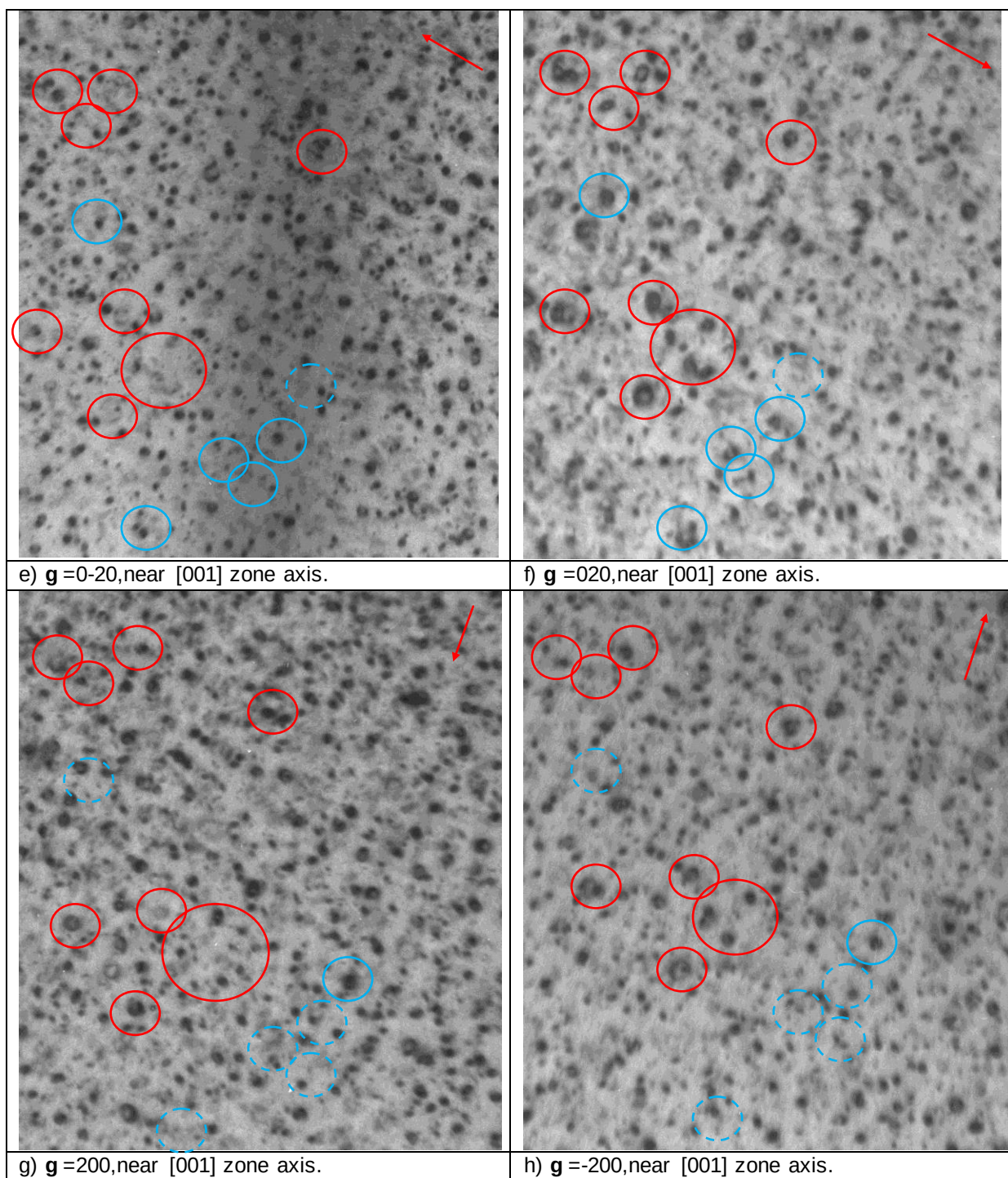
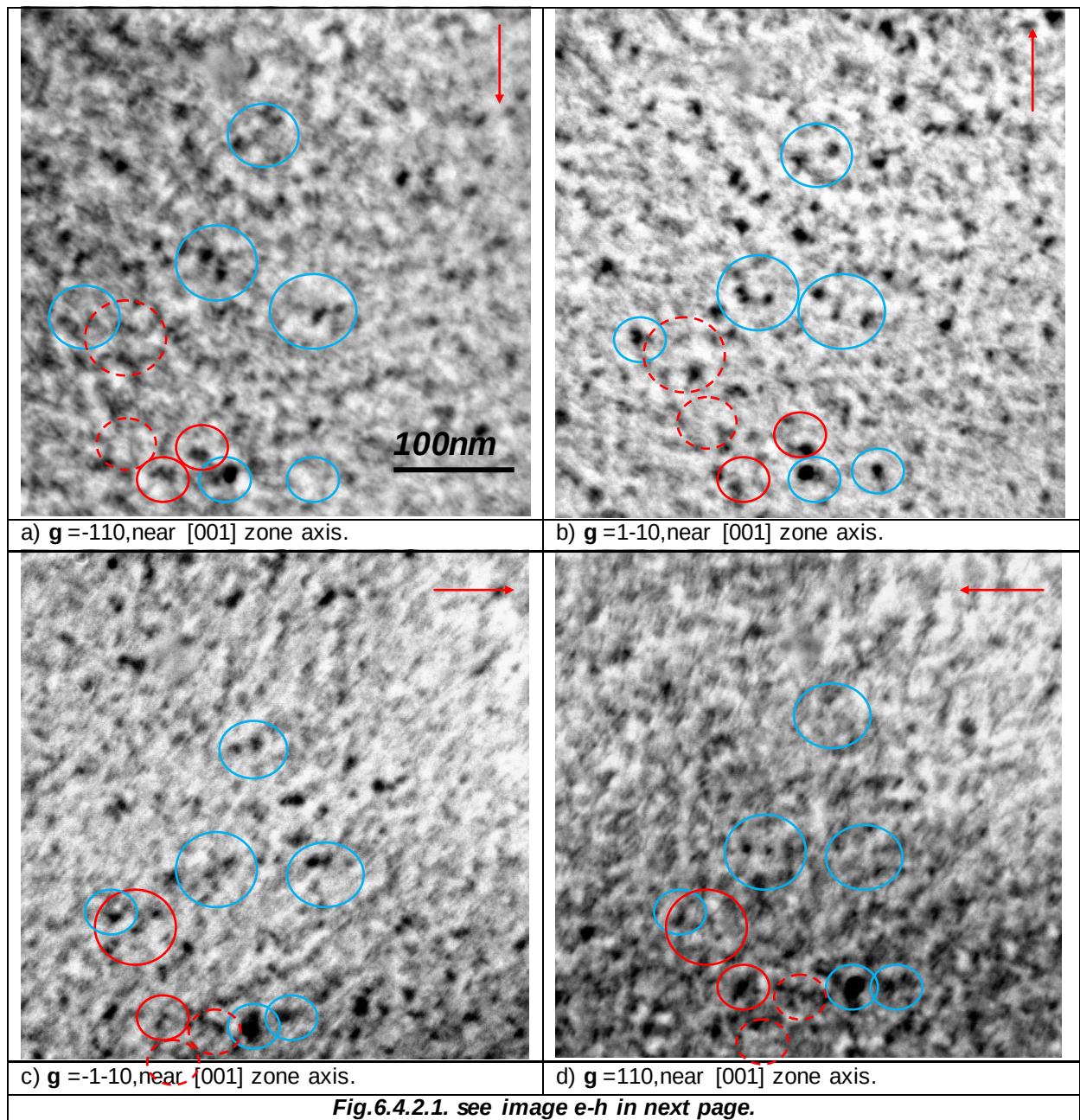


Fig.6.4.1.1. Contrast experiments for $g.b$ analysis of Fe8Cr irradiated at 300°C to 7dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

6.4.2 Characterisation of 400°C/2.5dpa specimens

Fig.6.4.2.1 shows a series of images taken near the [001] zone axis in an Fe8Cr specimen irradiated at 400°C to a dose of 2.5dpa. Of the 103 loops appearing in the selected area, 53 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, 23 had Burgers vector of type $\langle 100 \rangle$, and 17 loops could not be identified. In Fe8Cr irradiated at 400°C to a dose of 2.5dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were therefore ~ 62% and 38% respectively.



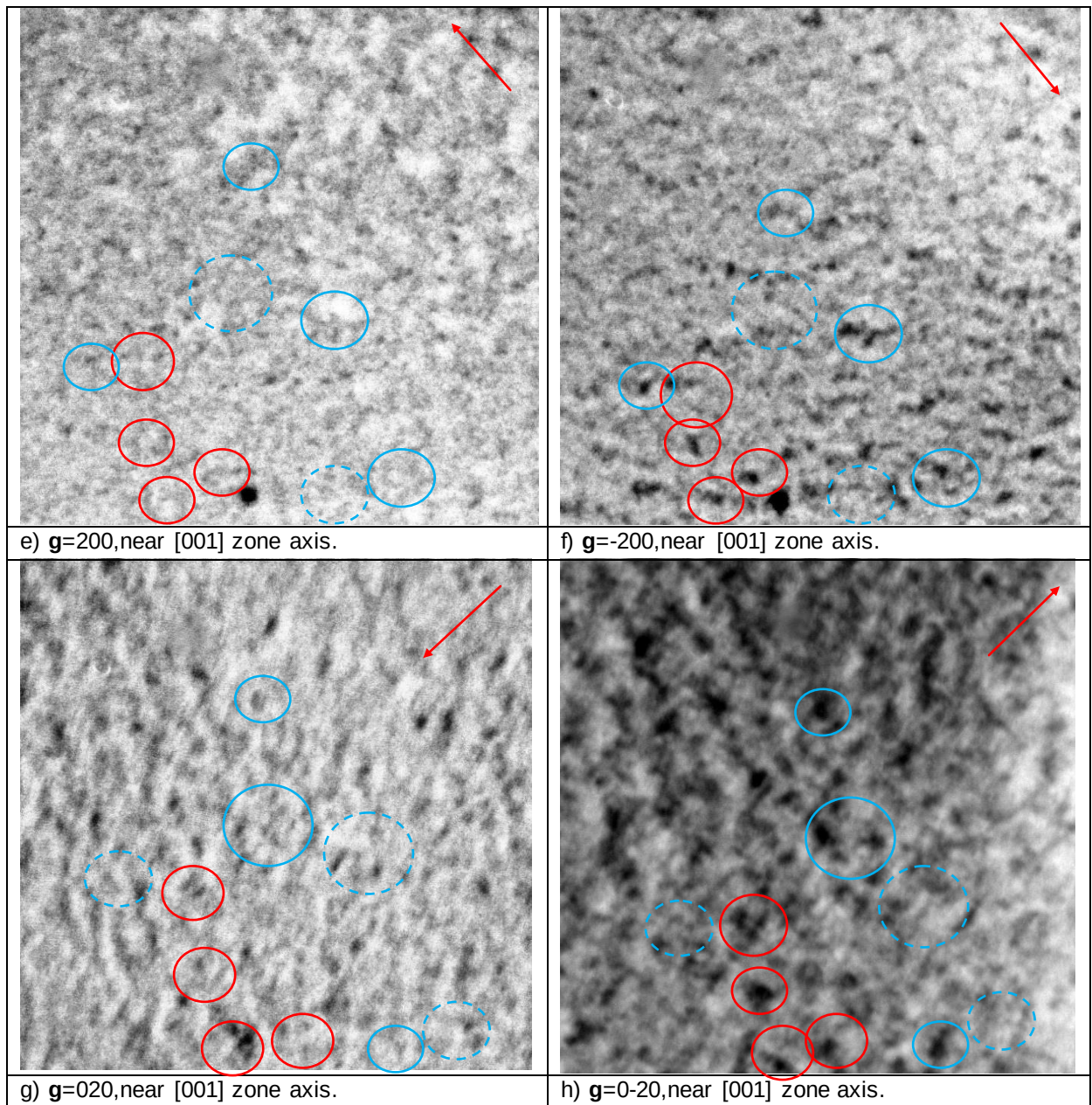
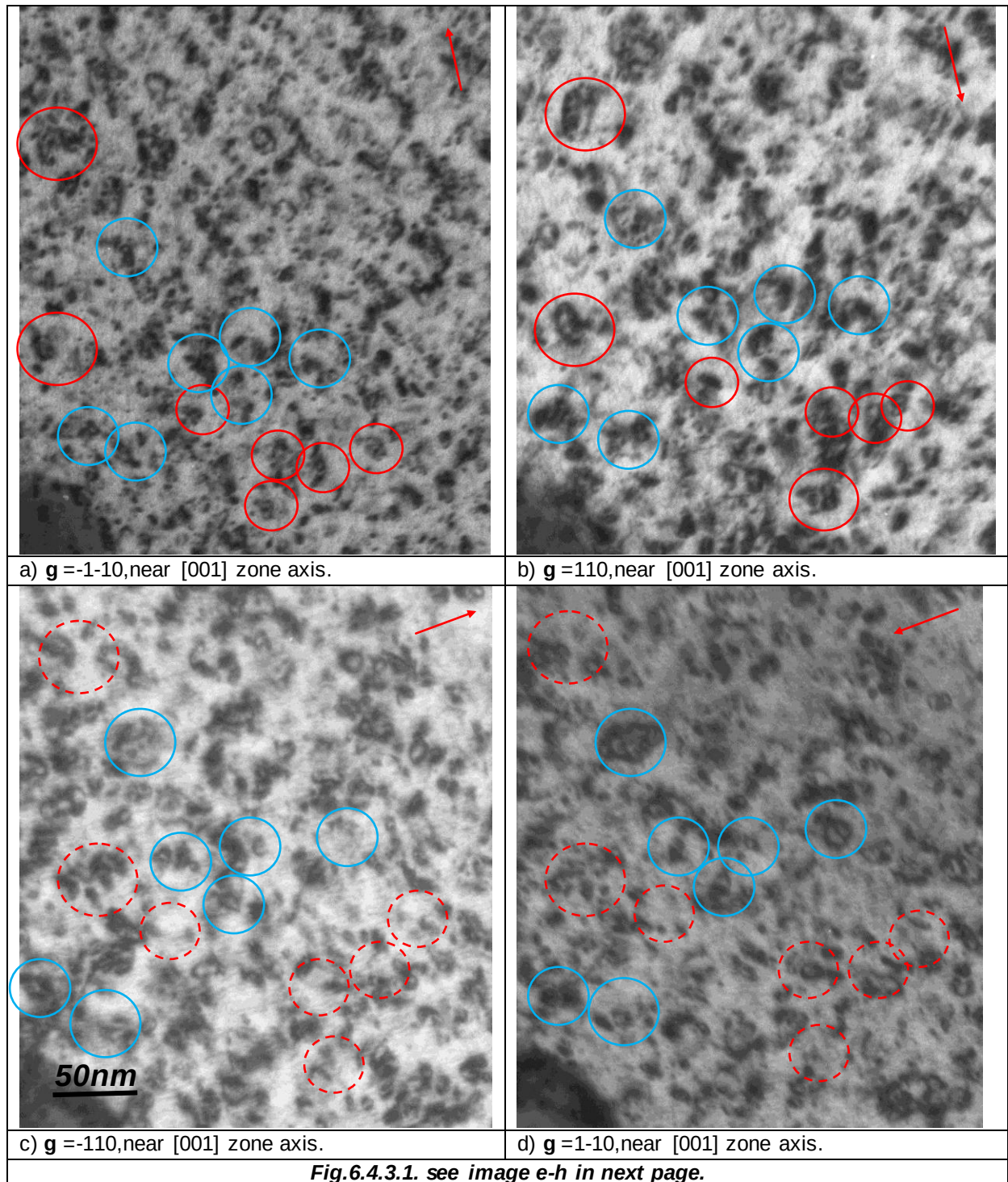


Fig.6.4.2.1. Contrast experiments for $g.b$ analysis of Fe8Cr irradiated at 400°C to 2.5 dpa; $b=\frac{1}{2}\langle 111 \rangle$ (red circles) and $b=\langle 100 \rangle$ (blue circles).

6.4.3 Characterisation of 400°C/7dpa specimens

Fig.6.4.3.1. shows the Burgers vector analyses for Fe8Cr specimen irradiated at 400°C to a dose of 7dpa. Of the 112 loops analysed in the selected area, 50 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$ and 41 had Burgers vectors of type $\langle 100 \rangle$, and 21 could not be analysed. For Fe8Cr irradiated at this condition, ~55% of the loops were therefore of $\frac{1}{2}\langle 111 \rangle$ type, while 45% of the loops were of $\langle 100 \rangle$ type.



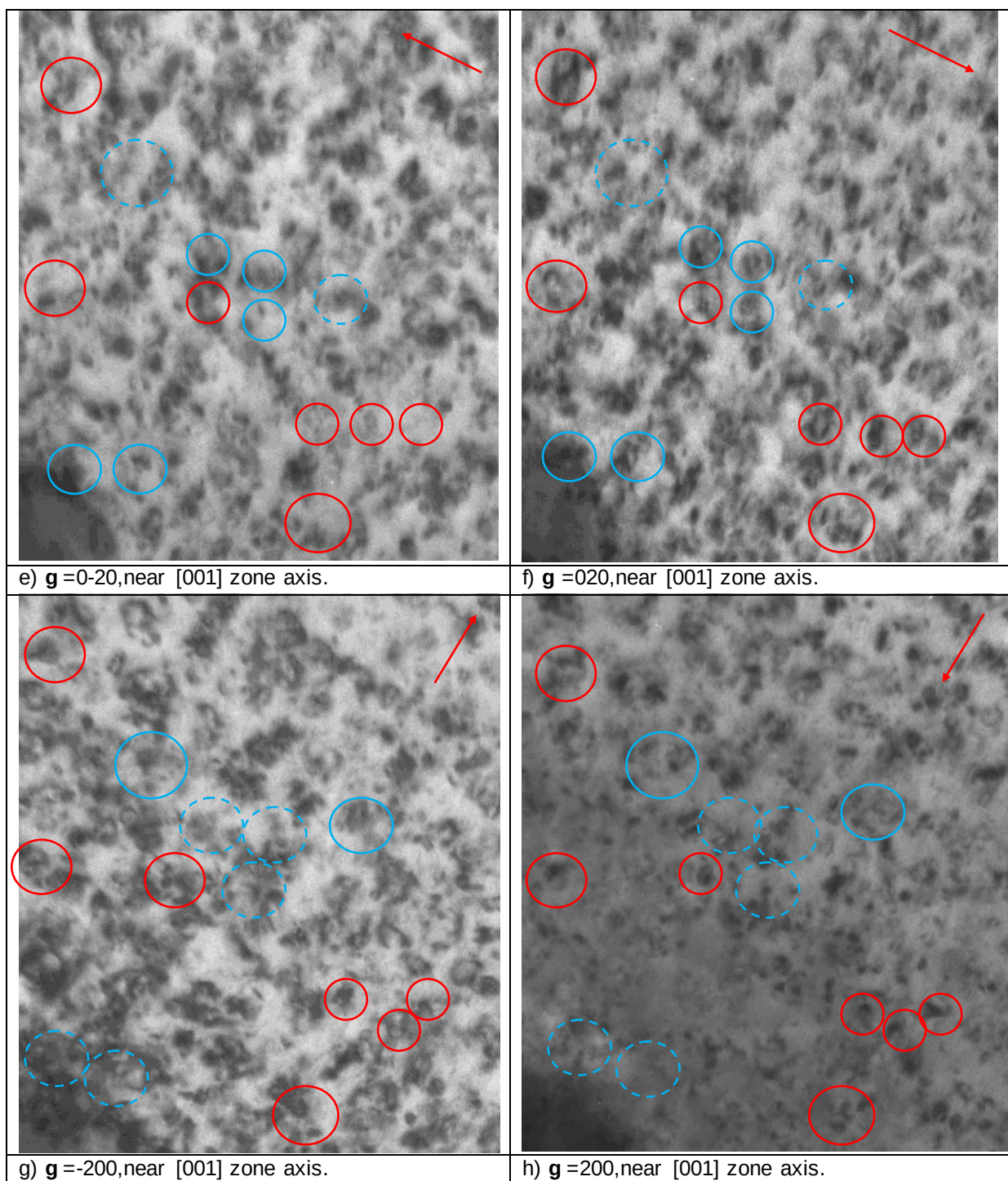
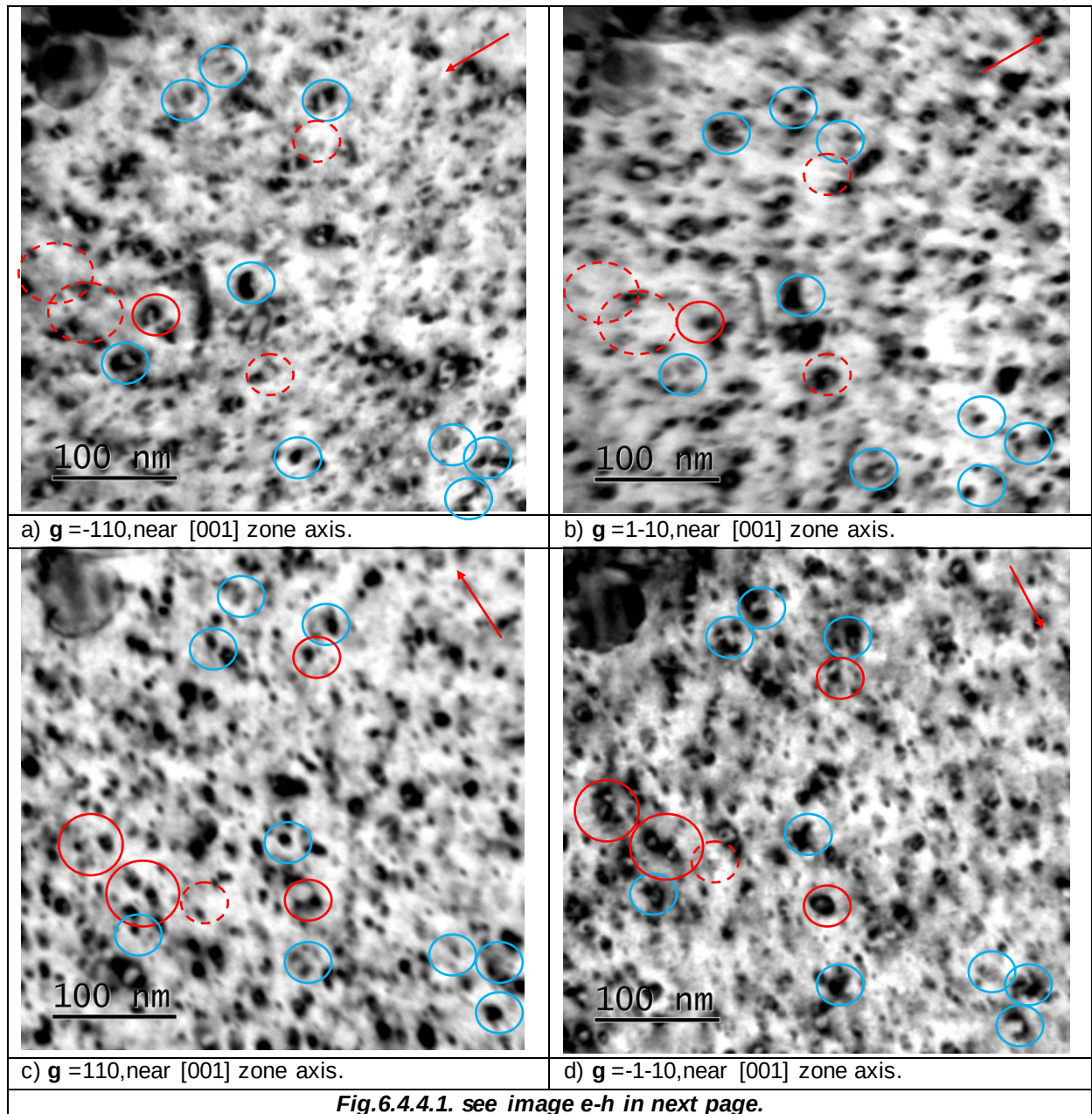


Fig.6.4.3.1. Contrast experiments for $g.b$ analysis of Fe8Cr irradiated at 400°C to 2.5dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

6.4.4 Characterisation of 460°C/7dpa specimens

Fig.6.4.4.1 shows the Burgers vector analyses for an Fe8Cr specimen irradiated at 460°C to 7dpa. Of the 153 loops in the selected area, 68 loops had Burgers vector of $\frac{1}{2}\langle 111 \rangle$ type, 68 loops had $\langle 100 \rangle$ type Burgers vector, and 17 loops were too small to be identified. The proportions are therefore about 50% for $\frac{1}{2}\langle 111 \rangle$ type and 50% for $\langle 100 \rangle$ type.



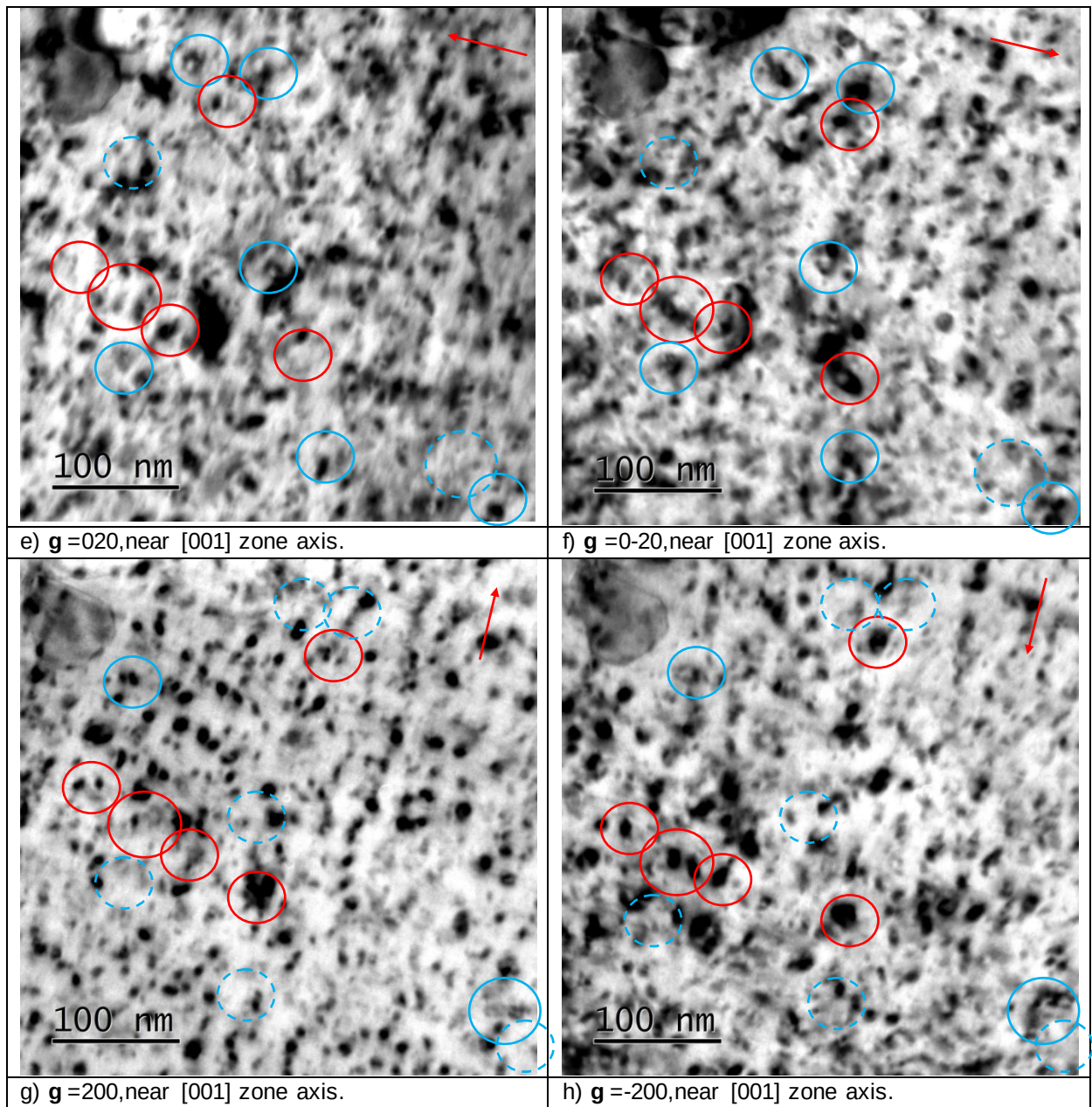


Fig.6.4.4.1. Contrast experiments for $g.b$ analysis of Fe8Cr specimen irradiated at 400°C to 2.5dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).

6.5 Loop Nature Determination

Nature analysis in Fe₈Cr was using the same inside-outside method as described in the previous chapters.

6.5.1 Characterisation on 300 °C/7dpa specimens

Fig.6.5.1.1 shows the nature determination and habit-plane analysis in a Fe₈Cr specimen irradiated at 300 °C to a dose of 7dpa. The **g.b** analysis described in **6.4.1** shows that these loops have Burgers vector of type $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. In **Fig.6.5.1.1**, as the specimen is tilted from the [001] to the [113] zone axis, the marked loops turn clockwise, which is consistent with a circle on the (1-11) plane. Thus these loops have Burgers vector of type $\frac{1}{2}[\bar{1}11]$. In **image (a,c)**, these loops all show outside contrast in **g**=1-10. Therefore, they are interstitial in nature. Using the same method, it was found that all 10 loops analysed were interstitial in nature.

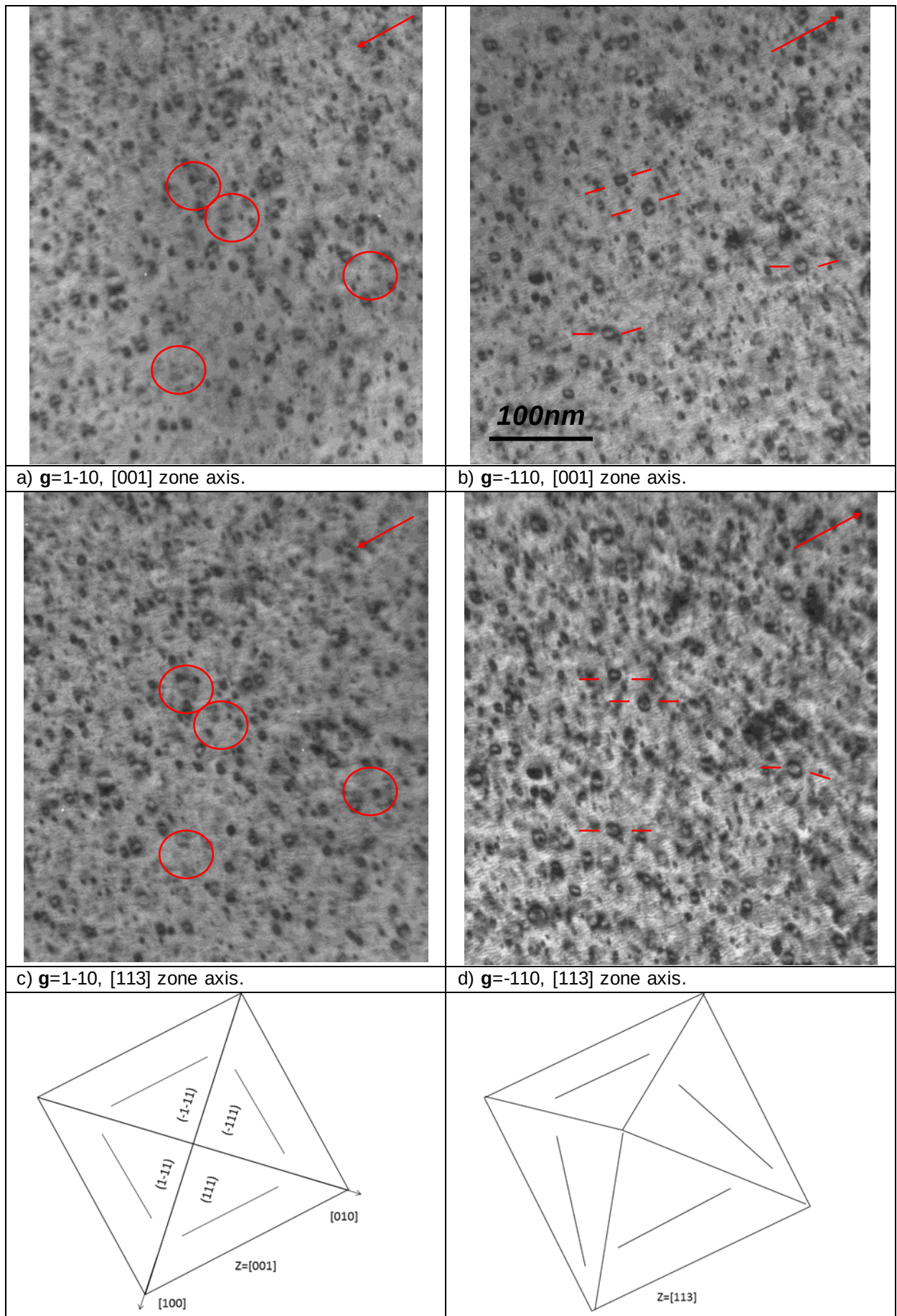


Fig.6.5.1.1. Nature determination and habit-plane analysis for Fe8Cr alloy irradiated at 300 °C to 7dpa.

6.5.2 Characterisation of 400°C/7dpa specimens

Fig.6.5.2.1. shows the loop nature and loop habit plane determination for a Fe8Cr specimen irradiated at 400°C to a dose of 7dpa. The two marked loops have Burgers vector of type $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$, and their major axes turned anti-clockwise as the sample was tilted from the [001] zone axis to the [111] zone axis. Therefore they have Burgers vector of type $\frac{1}{2}[\bar{1}11]$.

Both loops show outside contrast in $\mathbf{g}=-110$, and show inside contrast in $\mathbf{g}=1-10$. So they are both interstitial in nature. All five loops analysed in the specimen were interstitial.

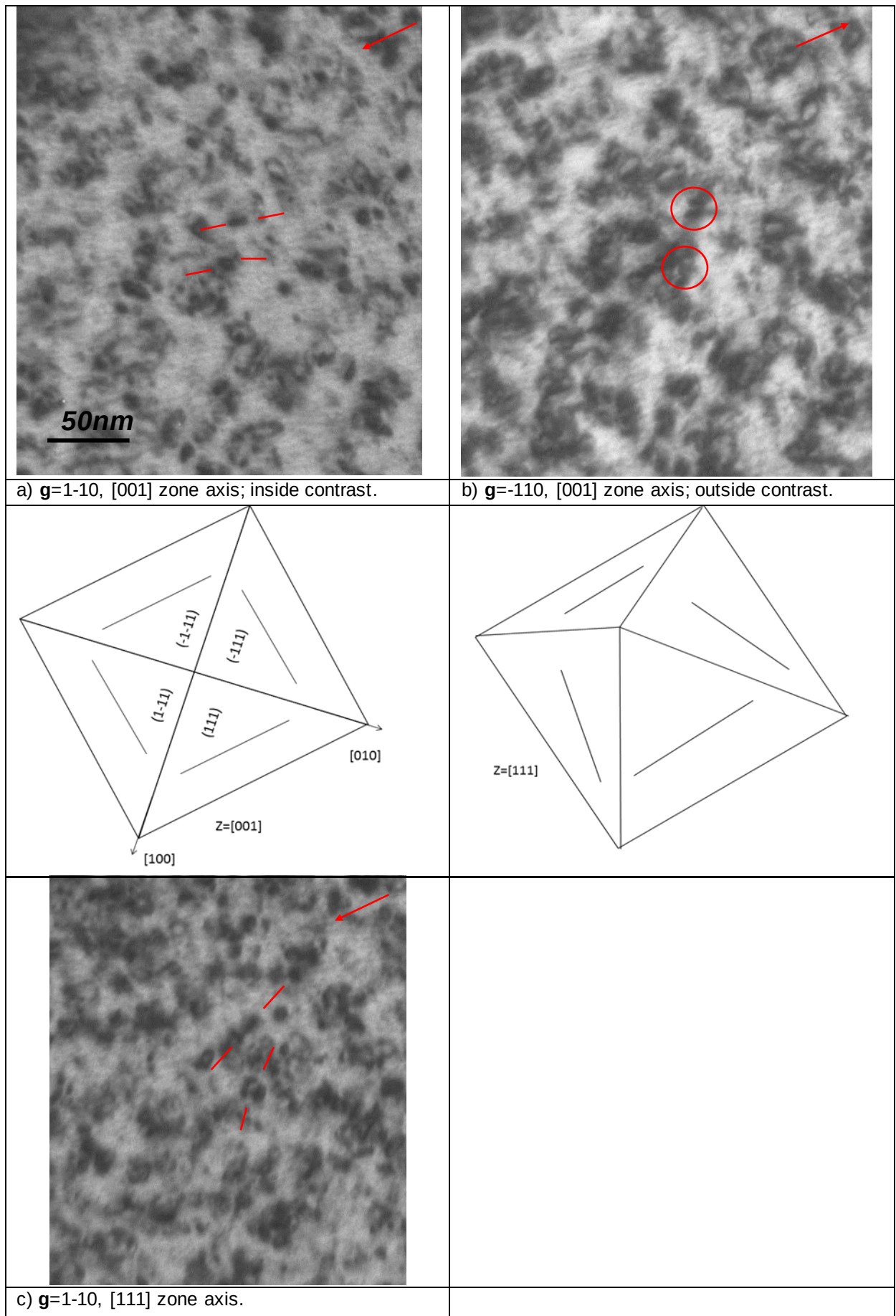


Fig.6.5.2.1. Nature and loop habit plane determination in Fe8Cr specimen irradiated at 400°C/7dpa.

6.6 Summary of Results

Microstructures:

1. At all temperatures, the radiation damage in Fe8Cr took the form of dislocation loops. No finger-shaped loops or larger irregular loop clusters were observed. At 400°C (7dpa), loop interactions were observed. At 460°C (7dpa), loop strings which had a regular perpendicular shape appeared.

Loop Sizes:

2. In Fe8Cr, both the average and maximum loop sizes increased with irradiation temperature and dose.

Loop number densities:

3. The densities in Fe8Cr alloys at all temperatures were very similar at the same dose (7dpa).
4. A higher irradiation dose caused a higher loop density in Fe8Cr at 400°C.

Loop Burgers vectors:

5. The trend of temperature effects was not so clear in Fe-Cr as that in Fe, but comparing the data at 300°C to that at 460°C, the proportion of $\langle 100 \rangle$ loops did increase about 20%.
6. Comparing the data from Fe and Fe5Cr alloys, it can be concluded that as the Cr content increased, the proportion of $\frac{1}{2}\langle 111 \rangle$ loops increased.

Loop Nature:

7. All loops which were analysed were found to be interstitial in nature.

Chapter 7: Radiation Damage in Fe11Cr

In this chapter, the characterisation of heavy-ion irradiated Fe-11Cr at elevated temperature and doses is presented.

7.1 Irradiation Damage Microstructures

In **Fig.7.1.1**, it can be seen that the damage structures in Fe11Cr specimens irradiated at 300°C and 400°C were similar to those found in Fe8Cr irradiated at the same temperature. At 300°C, the damage structure contained a high density of small dislocation loops, while at 400°C, larger loops, loop clusters and finger-like loops appeared. At 460°C, the microstructures in Fe11Cr were very similar to those found in pure Fe specimen irradiated at 400°C at the same 7dpa dose: large irregular shaped loop clusters and broken finger-shaped loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vectors were observed.

In **Fig.7.1.2**, a few curved line dislocations are seen in a Fe11Cr foil irradiated at 400°C to the dose of 2.5dpa. It is possible that these line dislocations were produced by irradiation. They were only found in Fe11Cr samples. The dislocation lines have $\frac{1}{2}\langle 111 \rangle$ Burgers vectors. The line dislocations were observed moving toward the centre of the sample. Their twisted shapes indicate pinning by dislocation loops.

In **Fig.7.1.3**, four representative micrographs of Fe11Cr alloy irradiated at 460°C to a dose of 7dpa are presented. It can be seen that the radiation induced microstructure in Fe11Cr consists of small loops which appear as black dots, larger dislocation loops which normally had $\frac{1}{2}\langle 111 \rangle$ Burgers vectors, loops clusters and broken finger-like loops. These microstructures were very similar to those found in Fe samples irradiated at 400°C to the same dose.

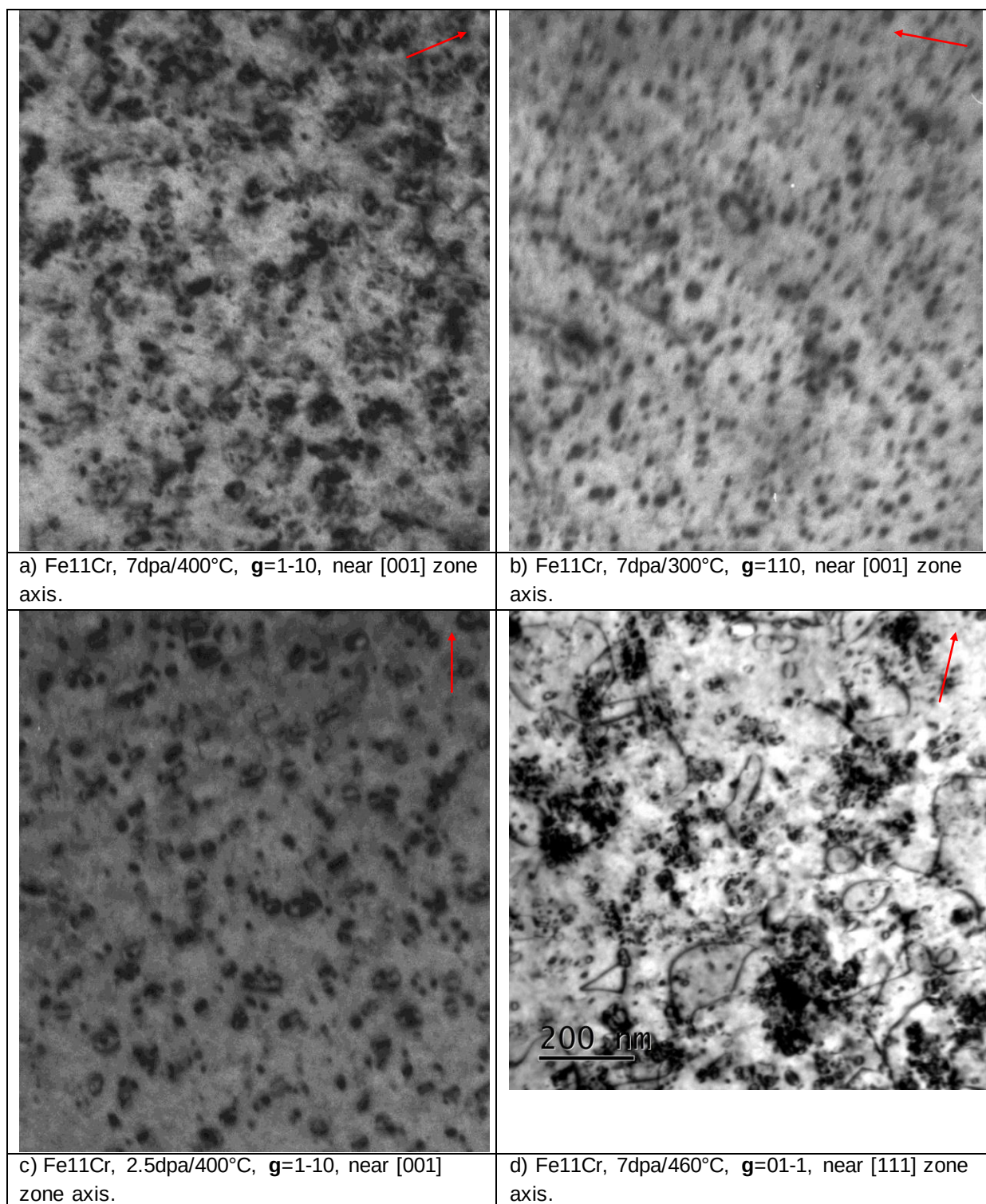


Fig.7.1.1. TEM micrographs of Fe11Cr samples irradiated at different temperatures and doses.

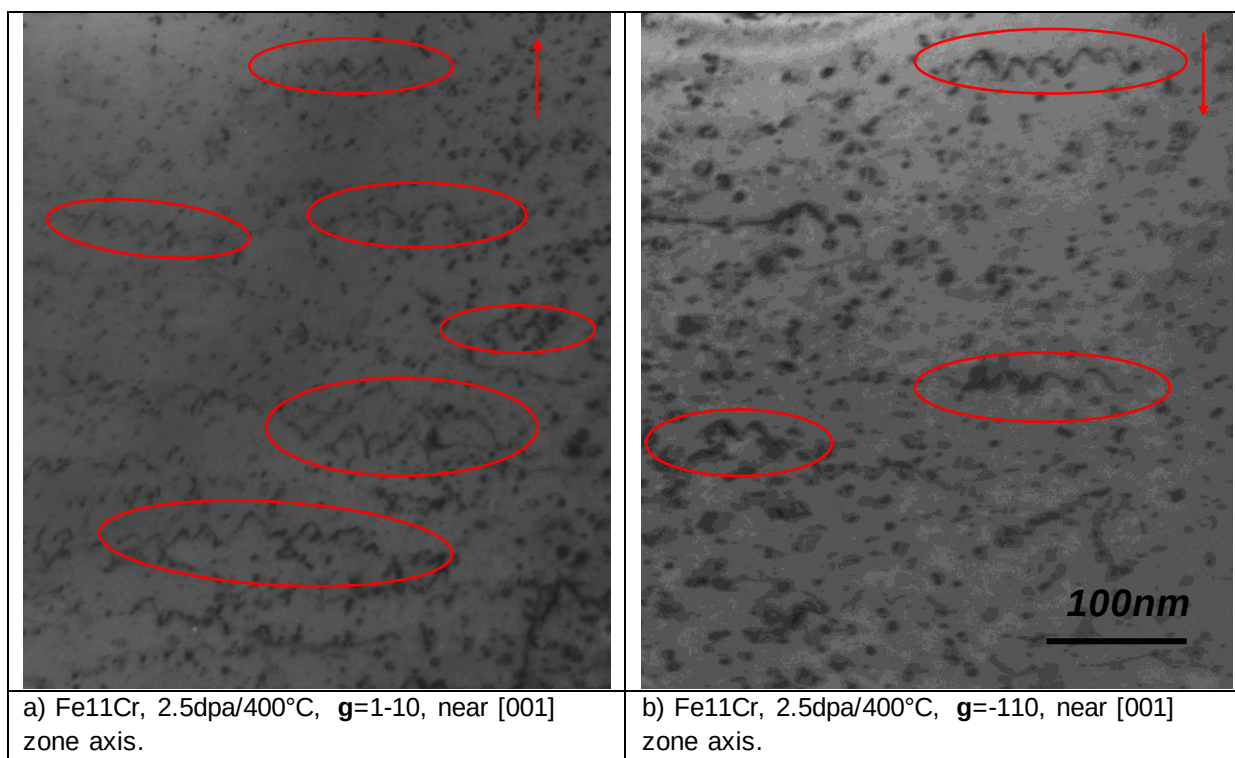


Fig.7.1.2. TEM micrographs of Fe11Cr samples irradiated at 400°C to a dose of 2.5dpa.

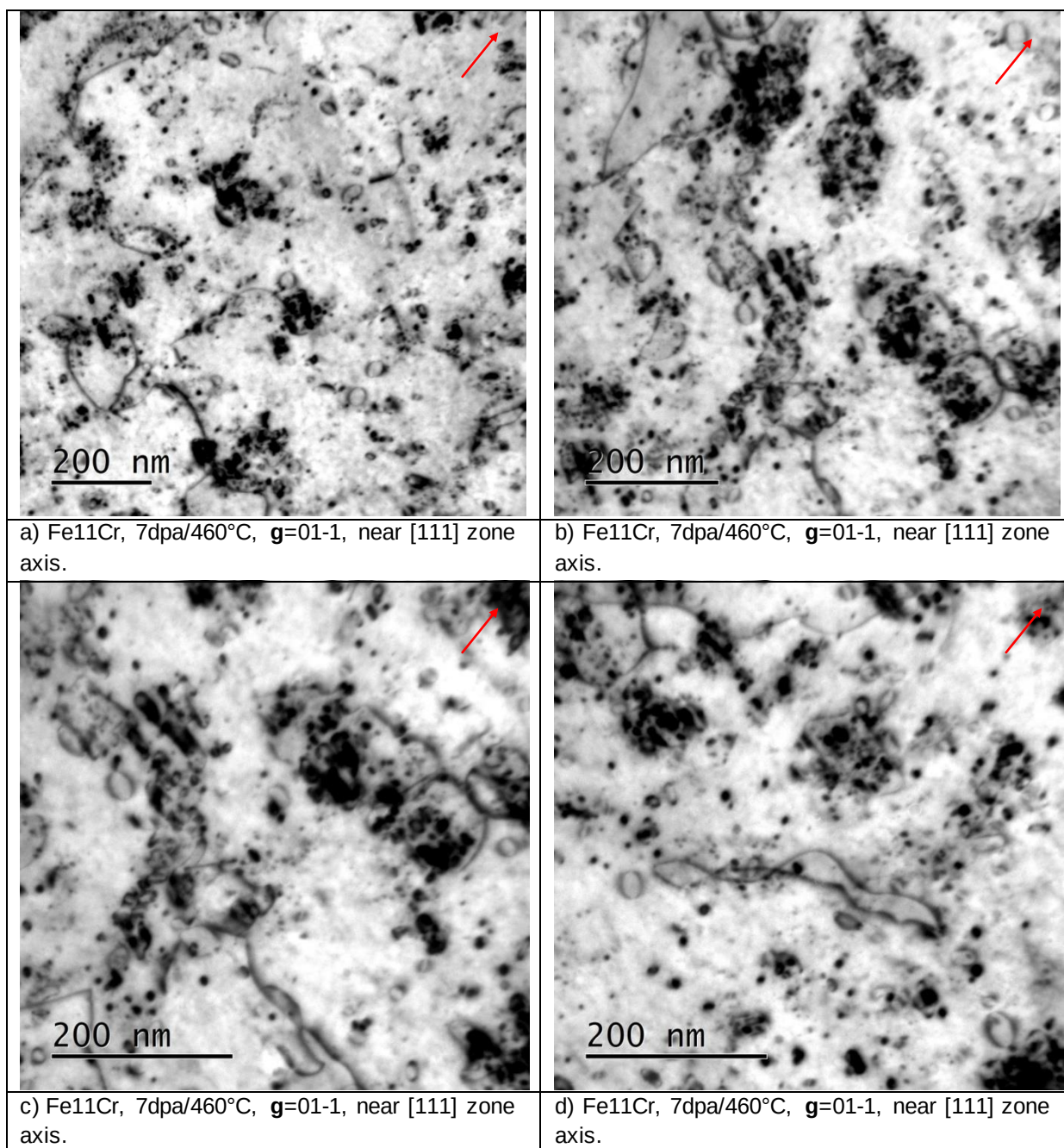


Fig.7.1.3. TEM micrographs of Fe11Cr samples irradiated at 460°C to a dose of 7 dpa.

7.2 Loop Image Size Distributions

The damage structure in heavy-ion irradiated Fe11Cr took the form of isolated dislocation loops, except in the 460°C/7dpa specimen when large loop clusters and broken finger-like loops appeared which were very similar to those observed in Fe at 400°C/7dpa (see **Fig.7.1.1**). The emergence of finger-like loops explains the great increase in average and maximum loop size at 460°C in Fe11Cr.

Fig.7.2.1 shows the loop image size distributions in Fe11Cr from 300°C to 460°C at 2.5dpa and 7dpa. **Fig.7.2.2** and **Fig.7.2.3** show the average and maximum loop size in Fe11Cr respectively.

The trends contained in the results can be summarised as follows:

- a. From 300°C to 460°C, at a given irradiation dose, the loop image sizes increased with irradiation temperature.
- b. At a given implantation temperature, the loop image size increased with dose.

Both trends are consistent with those found in Fe, Fe5Cr and Fe8Cr.

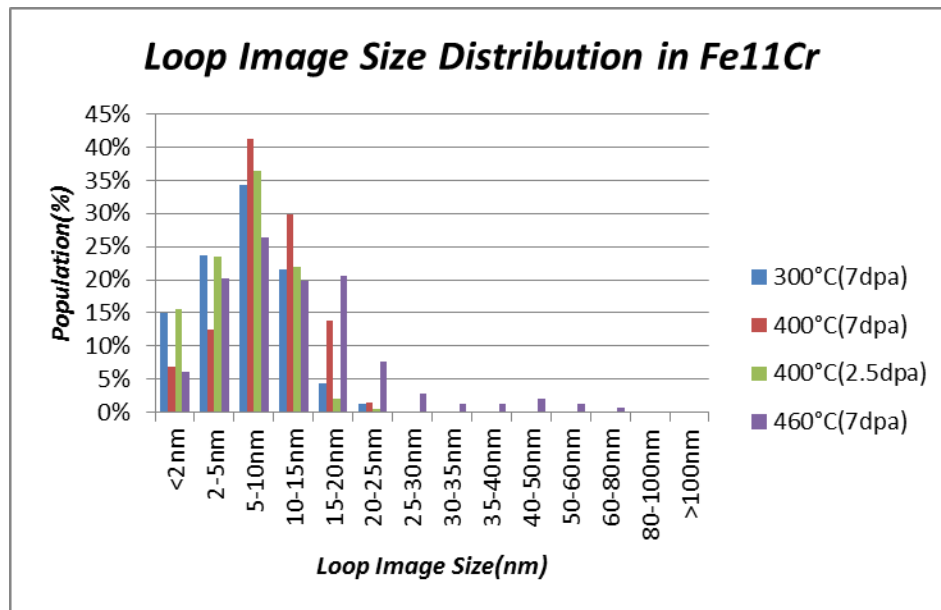


Fig.7.2.1. Loop image size distributions in Fe11Cr.

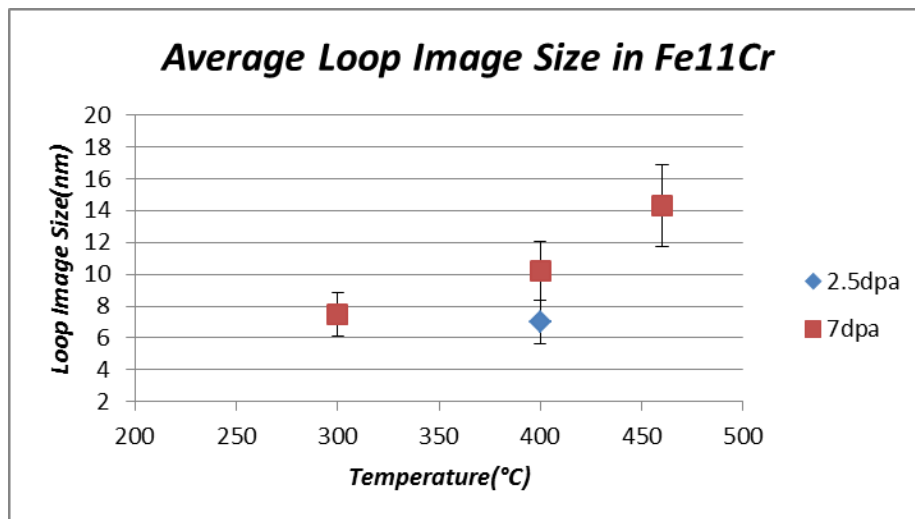


Fig.7.2.2. Average loop image size in Fe11Cr.

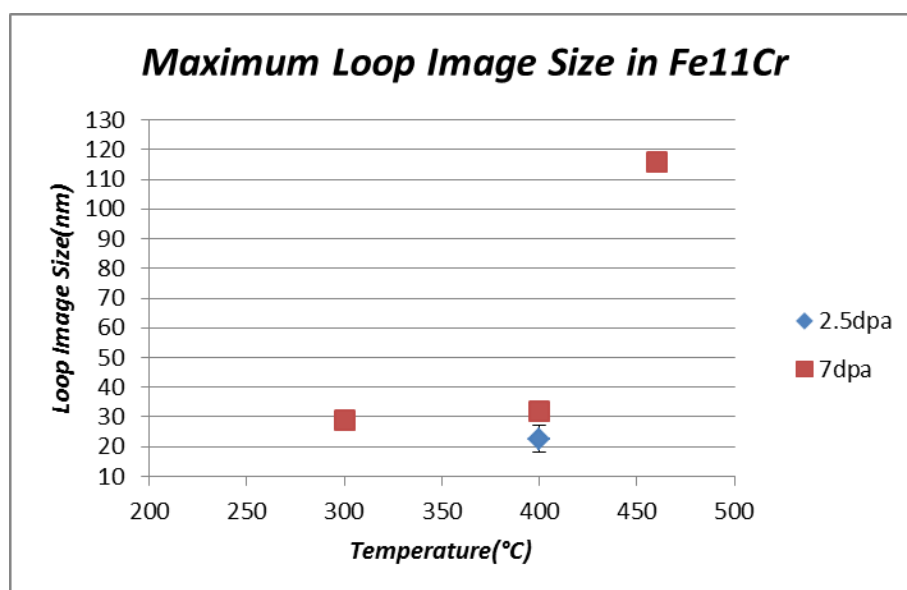


Fig.7.2.3. Maximum loop image size in Fe11Cr.

7.3 Loop Number Densities

Fig.7.3.1 shows the loop densities in Fe11Cr at different irradiation conditions. The noticeable decrease at 460°C is caused by the formation of finger-like loops and larger loop clusters. Moreover, it is seen that the loop densities at 300°C and 400°C at 7dpa were similar, which is consistent with the trend found in Fe8Cr. At 400°C, the loop density increased with implantation dose.

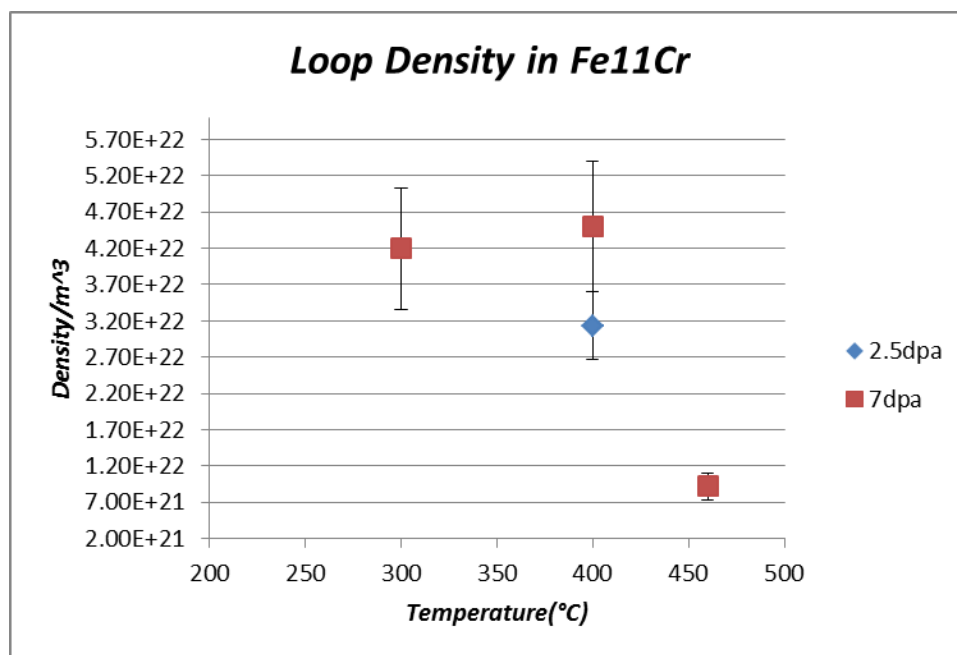


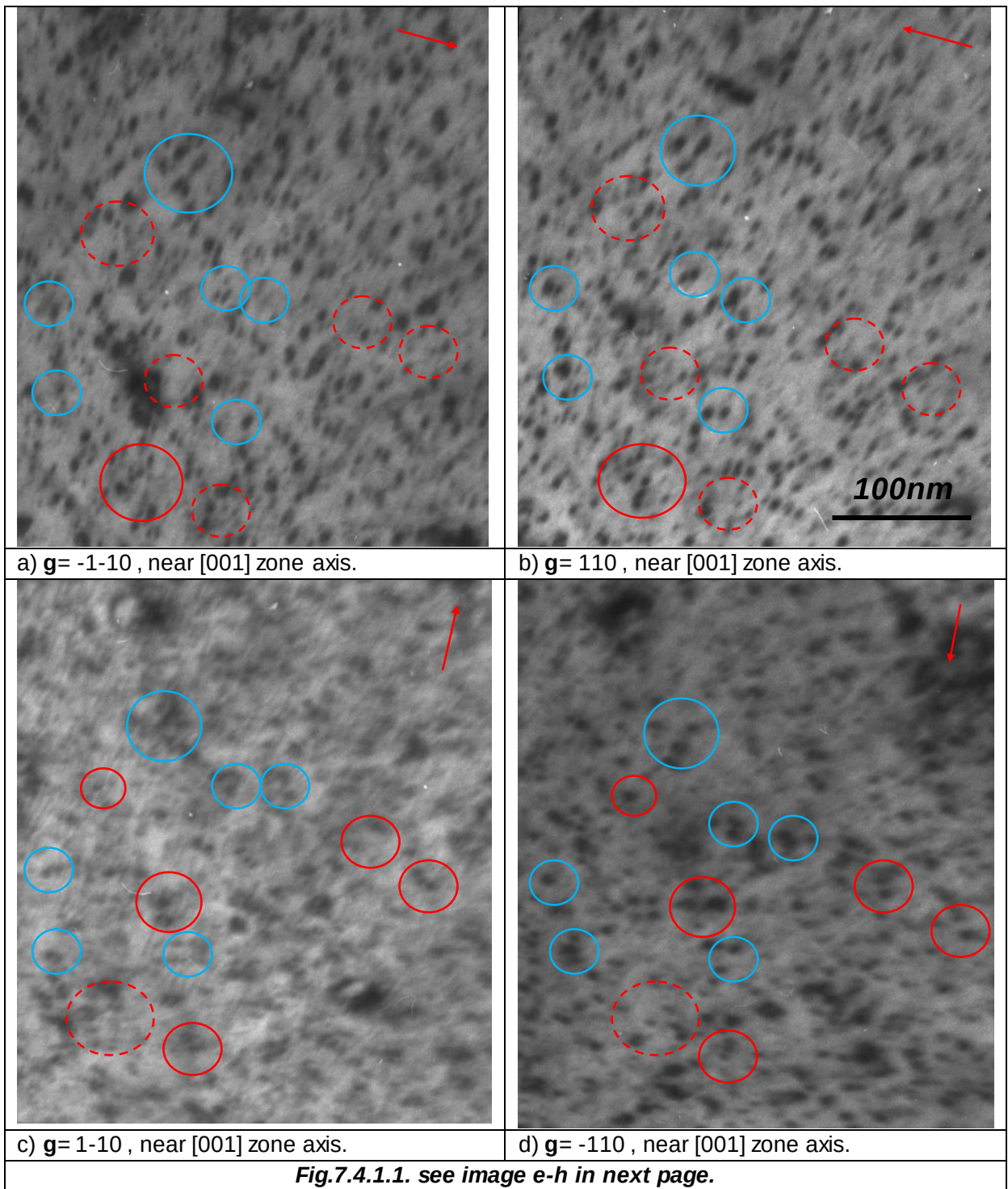
Fig.7.3.1. Loop densities in Fe11Cr at different irradiation conditions.

7.4 Loop Burgers Vectors

The Burgers vectors analyses for Fe11Cr irradiated were made using the same diffraction contrast method described in **chapter 3**.

7.4.1 Characterisation on 300°C/7dpa specimens

Fig.7.4.1.1 shows the contrast experiments for a Fe11Cr specimen irradiated at 300°C/7dpa near a [001] zone axis. Of 111 loops, 70 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, 23 had Burgers vectors of type $\langle 100 \rangle$ and 18 loops could not be identified. In Fe11Cr irradiated at 300°C/7dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ loops and $\langle 100 \rangle$ loops were therefore ~ 75% and 25% respectively.



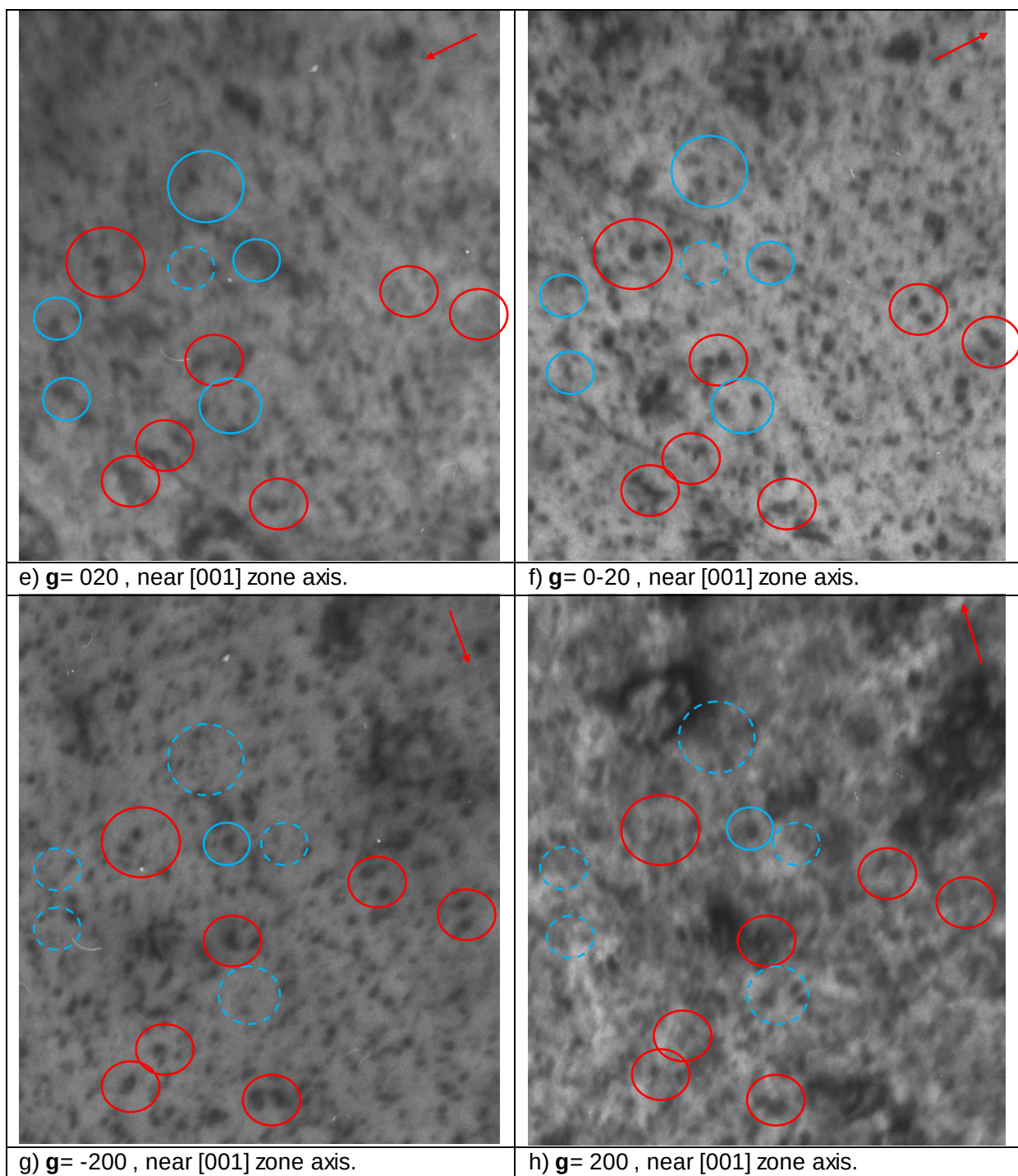
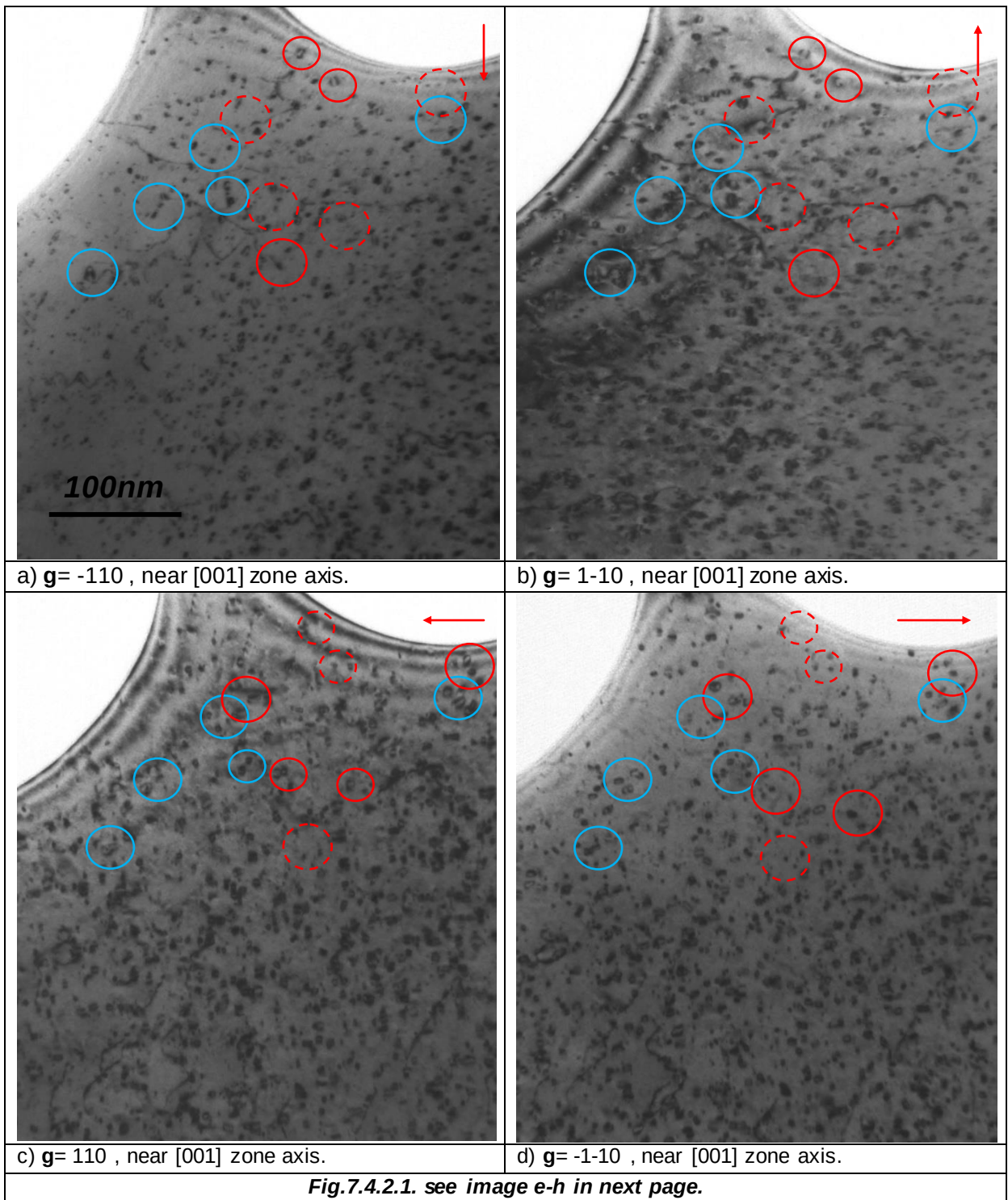


Fig.7.4.1.1. ***g.b** contrast experiments near $[001]$ zone axis, for Fe11Cr specimen irradiated at $300\text{ }^{\circ}\text{C}$ to a dose of 7dpa; $b=\frac{1}{2}\langle 111 \rangle$ (red circles) and $b=\langle 100 \rangle$ (blue circles).*

7.4.2 Characterisation of 400°C/2.5dpa specimens

Fig.7.4.2.1 shows a series of micrograph taken near the [001] zone axis in a Fe11Cr specimen irradiated at 400°C to a dose of 2.5dpa. Of 160 loops analysed, 107 had Burgers vectors of type $\frac{1}{2}\langle 111 \rangle$, 42 had Burgers vectors of type $\langle 100 \rangle$ and 11 loops could not be analysed. In Fe11Cr irradiated at 400°C to a dose of 2.5dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ loops and $\langle 100 \rangle$ loops were therefore ~72% and 28% respectively.



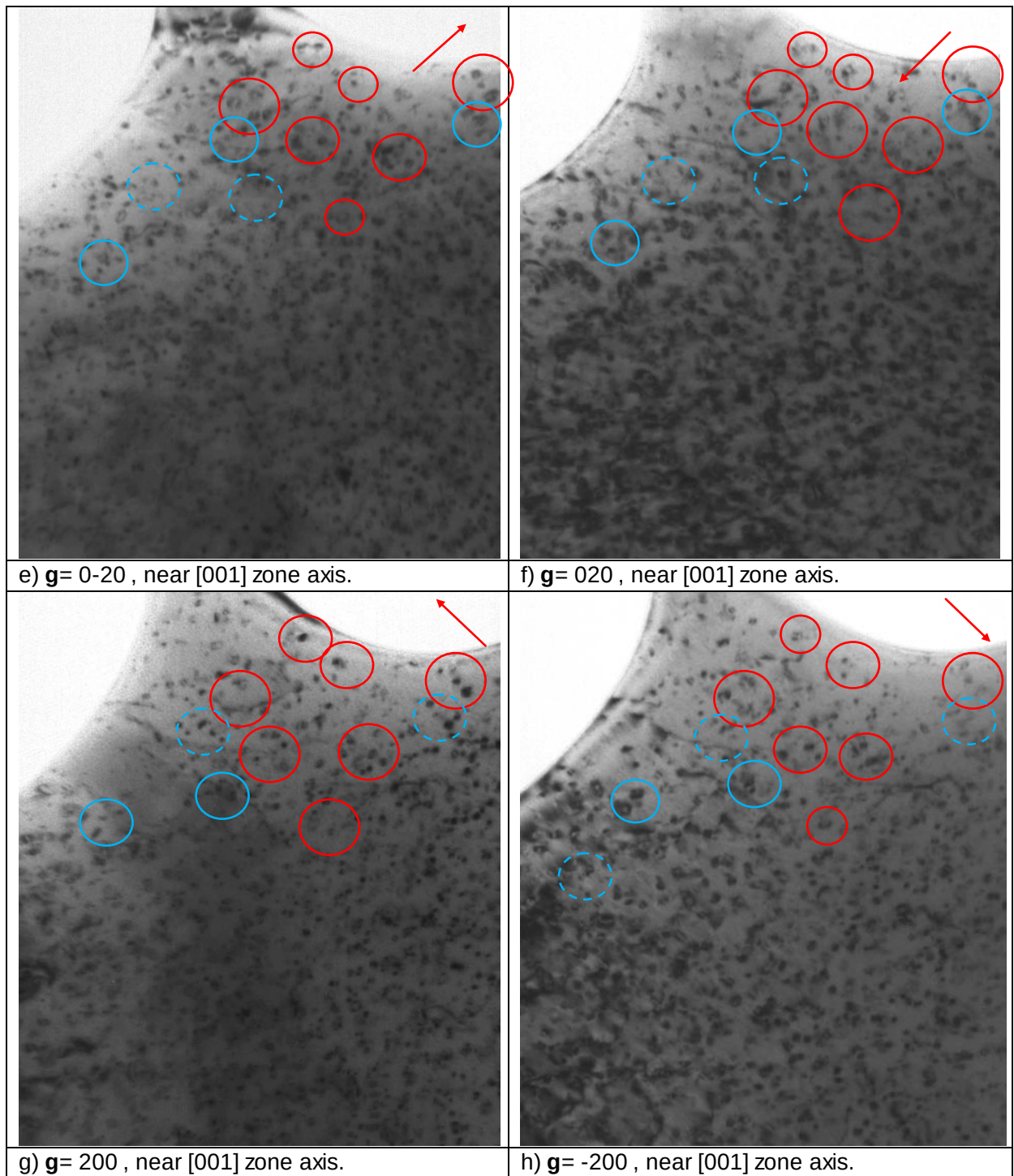
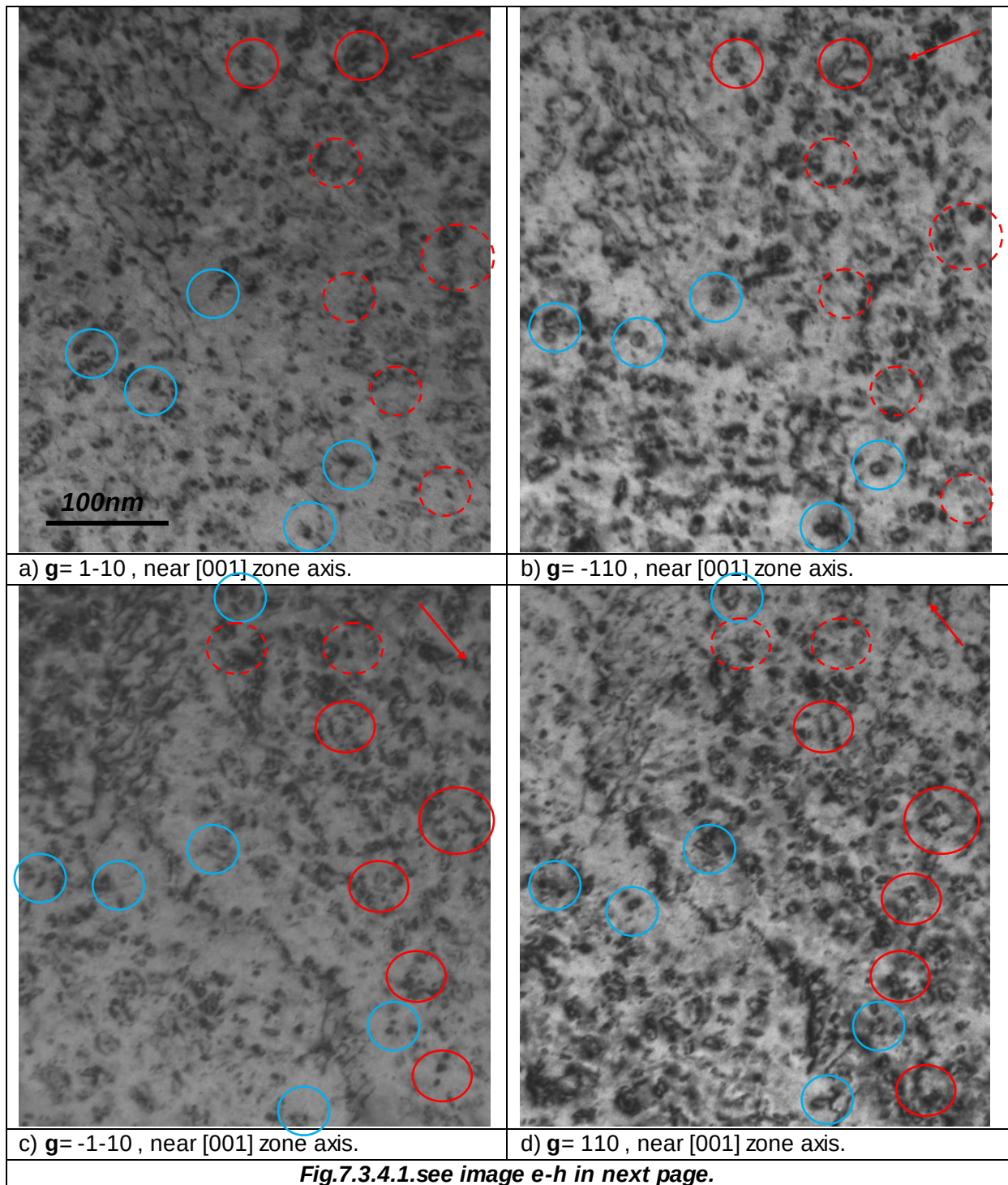


Fig.7.4.2.1. *$g.b$ contrast experiments near $[001]$ zone axis, for Fe11Cr specimen irradiated at $400\text{ }^{\circ}\text{C}$ to a dose of 2.5dpa ; $b=\frac{1}{2}\langle 111 \rangle$ (red circles) and $b=\langle 100 \rangle$ (blue circles).*

7.4.3 Characterisation on $400^{\circ}\text{C}/7\text{dpa}$ specimens

Fig.7.4.3.1 shows a series of micrograph taken near the $[001]$ zone axis in a Fe11Cr specimen irradiated at 400°C to a dose of 7dpa . Of 80 loops analysed, 45 loops had Burgers vectors of type $\frac{1}{2} \langle 111 \rangle$, 19 had Burgers vectors of type $\langle 100 \rangle$ and 16

loops could not be analysed. In Fe11Cr irradiated at 400°C to a dose of 7dpa, the proportions of $\frac{1}{2}$ $\langle 111 \rangle$ loops and $\langle 100 \rangle$ loops were therefore ~70% and 30% respectively.



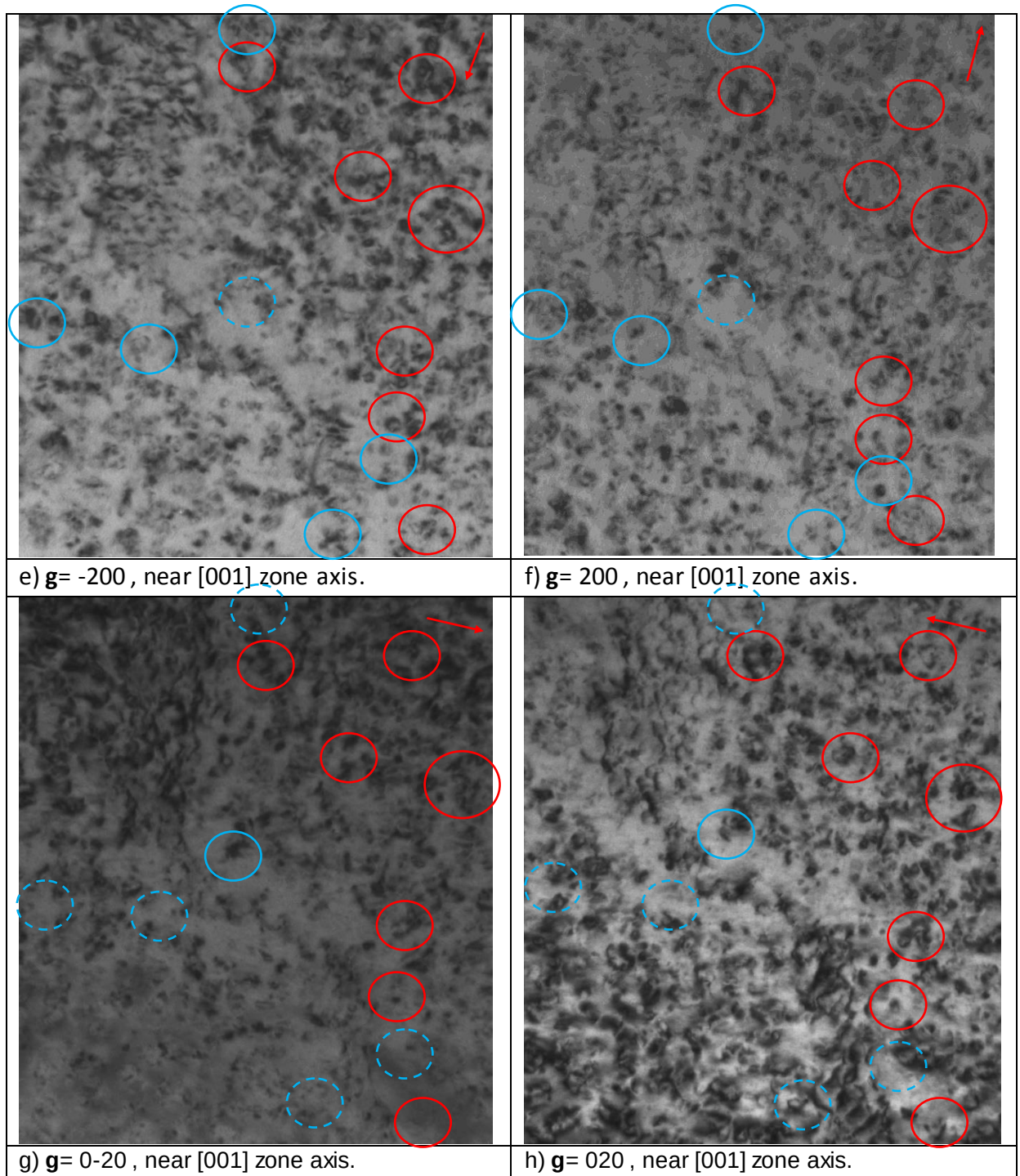


Fig.7.4.3.1. ***g.b** contrast experiments near $[001]$ zone axis, for Fe11Cr specimen irradiated at 400 °C to a dose of 7dpa; $b = \frac{1}{2}\langle 111 \rangle$ (red circles) and $b = \langle 100 \rangle$ (blue circles).*

7.4.4 Characterisation of 460°C/7dpa specimens

Fig.7.4.4.1 shows the Burgers vector analysis for an Fe11Cr specimen irradiated at 460°C to the dose of 7dpa. In this case the favoured [001] orientation was unavailable, and in order to obtain a complete **g.b** analysis the specimen was tilted from [111] to the [011] and [131] zone axes. Seven diffraction vectors in total were reached with both +**g** and –**g** images. With the aid of **Table.7.4.4.1**, the Burgers vector of the given specimen can be analysed as follows:

The loops circled in red were invisible in **g**=-110 and 2-11, whilst visible in all the other diffraction vectors, so their Burgers vectors is $\frac{1}{2}[1\bar{1}\bar{1}]$. The loops circled in blue were invisible in **g**=-110 and 200, and show clear contrast in all the other five diffraction vectors, which shows that their Burgers vector is [001]. Similarly the loop circled in yellow has Burgers vector $\frac{1}{2}[1\bar{1}1]$. Of the 100 loops analysed, 51 loops had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, 41 had Burgers vector of $\langle 100 \rangle$, and 8 loops were either too small or it was impossible for the author to track the contrast changes. In Fe11Cr irradiated at 460°C to a dose of 7dpa, the proportions of $\frac{1}{2}\langle 111 \rangle$ loops and $\langle 100 \rangle$ loops were ~55% and 45% loops respectively.

[111]	[011]	[131]					
<i>g/b</i>	<i>a/2[111]</i>	<i>a/2[-111]</i>	<i>a/2[1-11]</i>	<i>a/2[11-1]</i>	<i>a[100]</i>	<i>a[010]</i>	<i>a[001]</i>
$\pm(-110)$	0	1	1	0	1	1	0
$\pm(10-1)$	0	1	0	1	1	0	1
$\pm(01-1)$	0	0	1	1	0	1	1
$\pm(2-11)$	1	1	2	0	2	1	1
$\pm(200)$	1	1	1	1	2	0	0
$\pm(21-1)$	1	1	0	2	2	1	1
$\pm(1-12)$	1	0	2	1	1	1	2

Table.7.4.4.1. **g.b** table for [111], [011] and [131] zone axes.

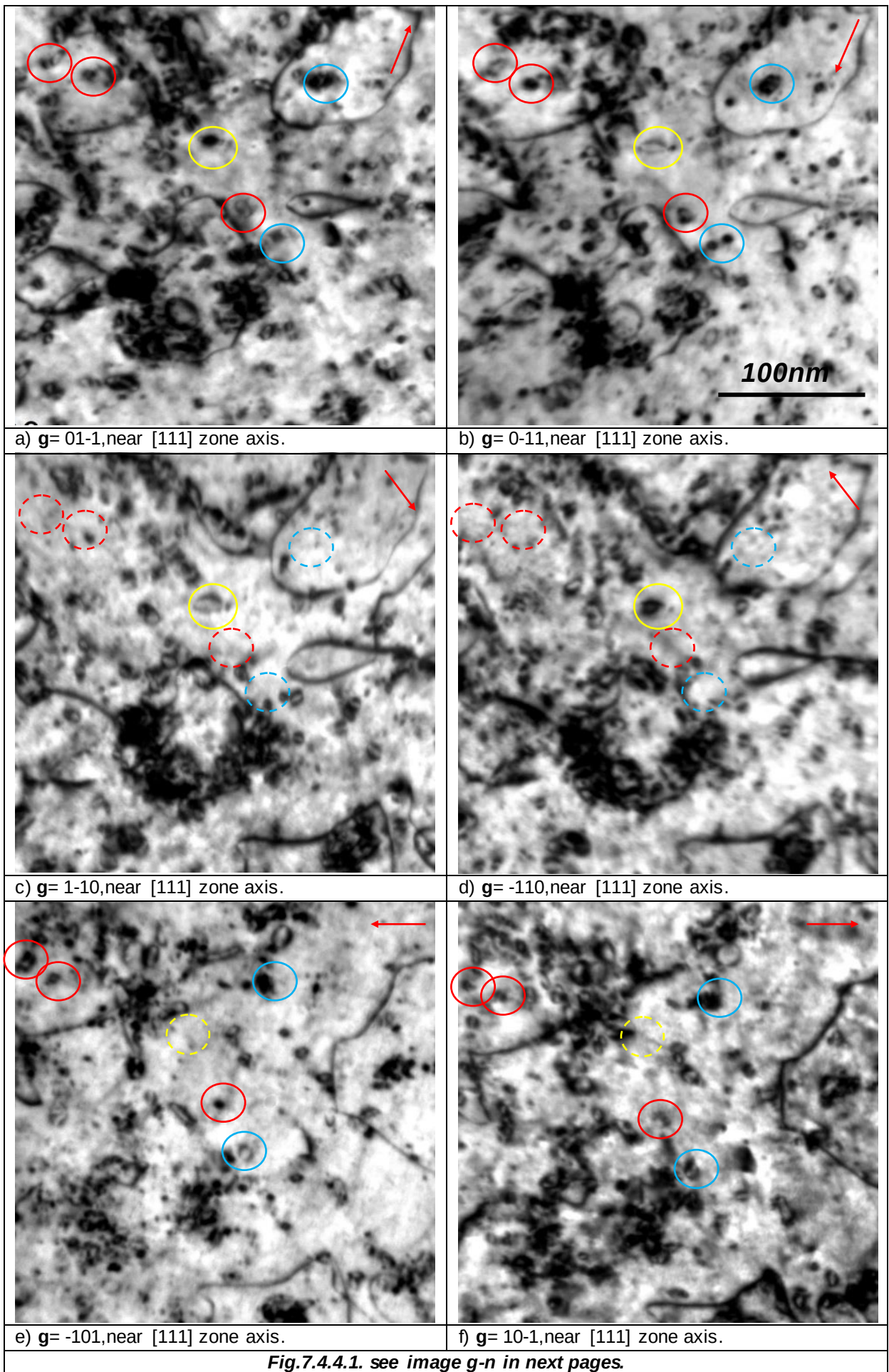
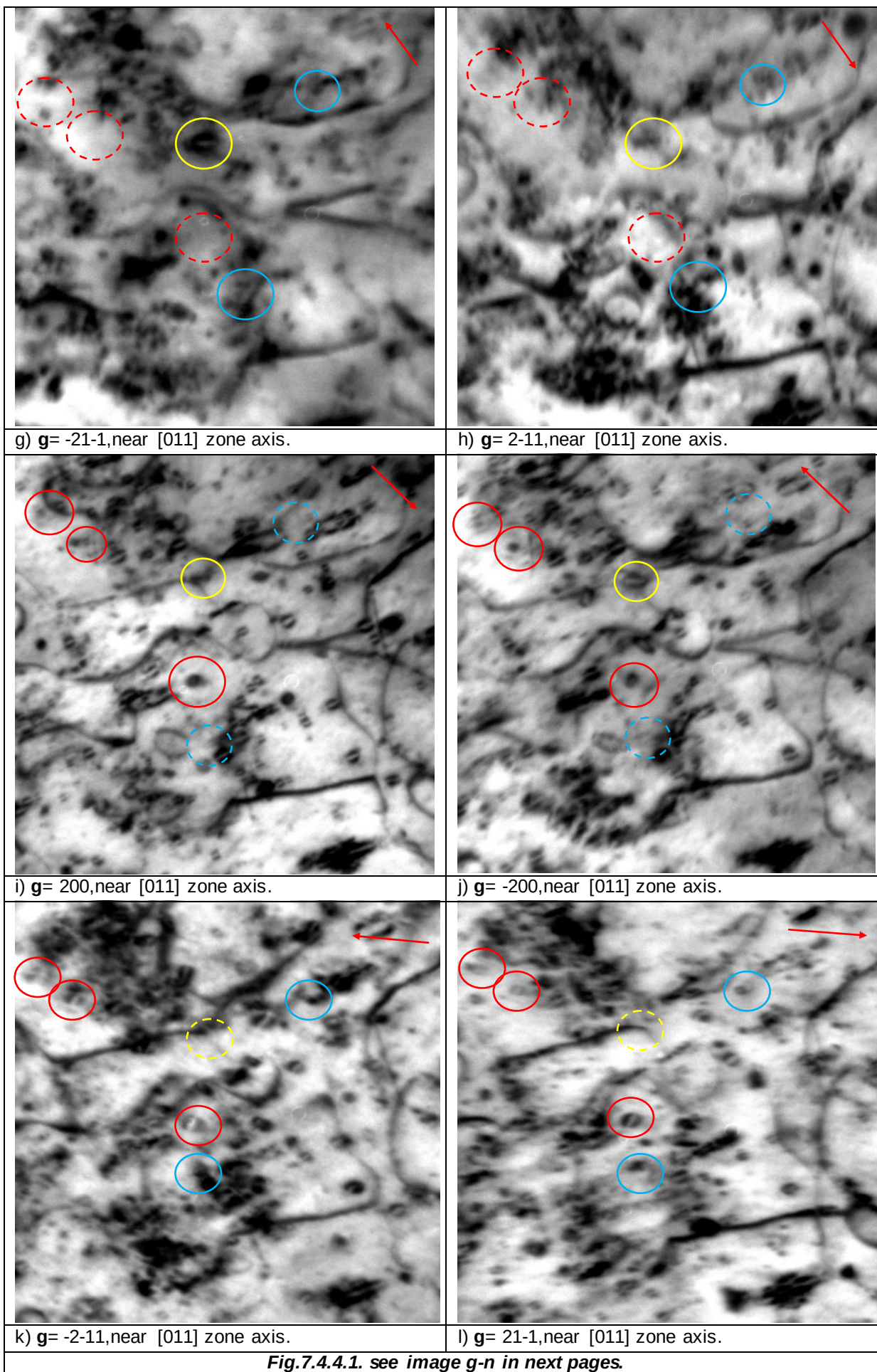
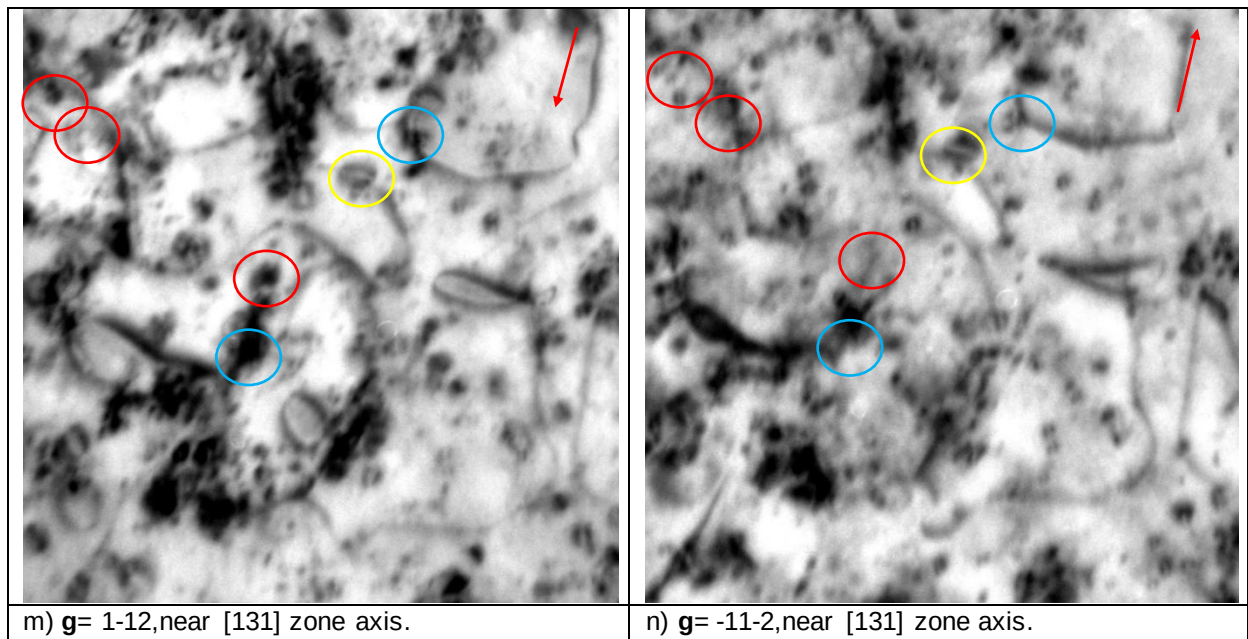


Fig.7.4.4.1. see image g-n in next pages.





m) $g= 1-12$, near $[131]$ zone axis.

n) $g= -11-2$, near $[131]$ zone axis.

Fig.7.4.4.1. Loop $g.b$ analyses in Fe11Cr at $z=[111],[011]$ and $[131]$, irradiated at 460°C to 7dpa. (red circle: $b=\frac{1}{2}[11-1]$, yellow circle: $b=\frac{1}{2}[1-11]$, blue circle: $b=[001]$)

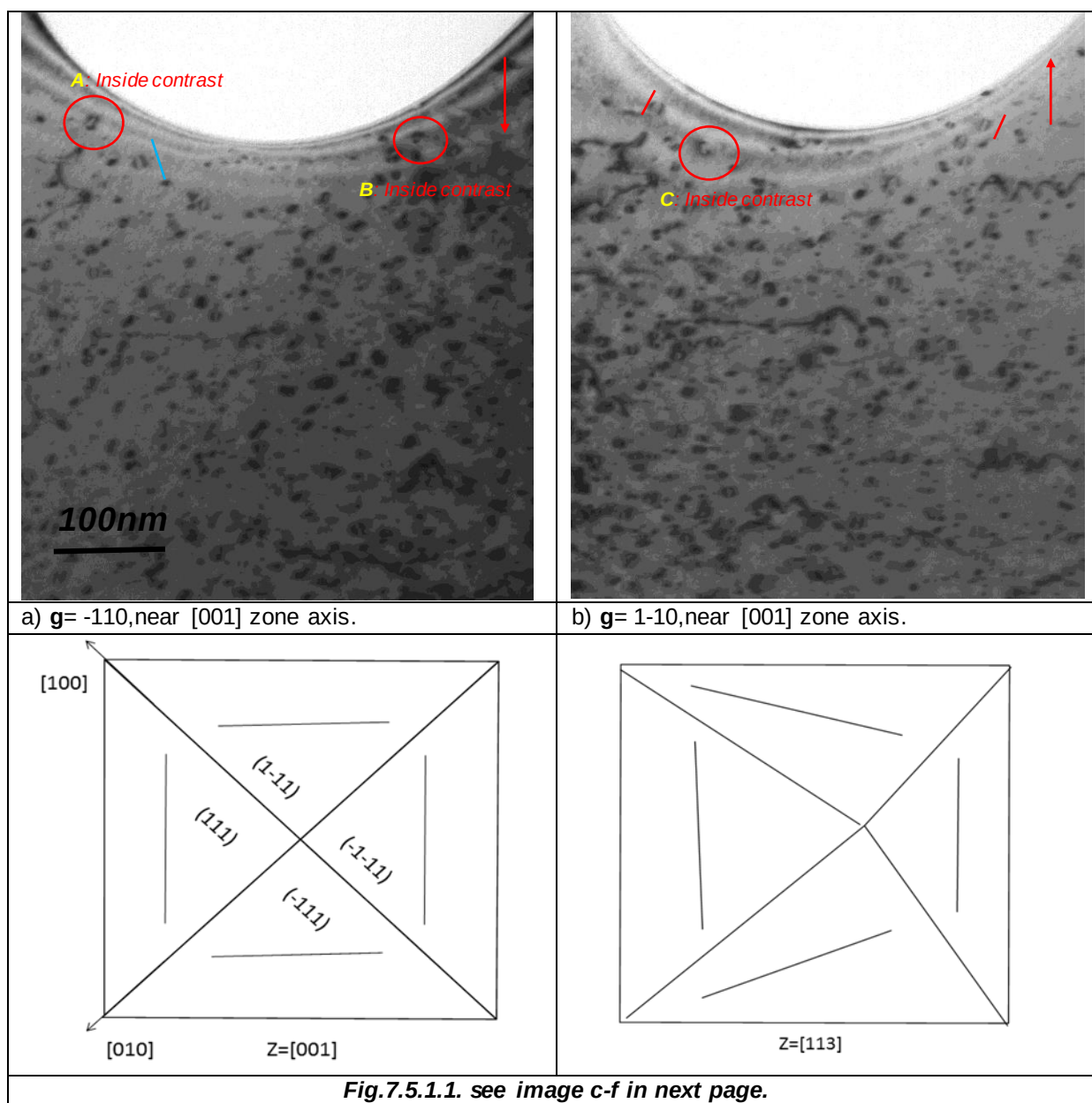
7.5 Loop Nature Determination

The loop nature determinations in Fe11Cr specimens were made using the same inside-outside method with the analyses of loop habit plane.

7.5.1 Characterisation of $400^{\circ}\text{C}/2.5\text{dpa}$ specimens

Fig.7.5.1.1. shows the nature determination and habit plane analysis in a Fe11Cr specimen irradiated at $400^{\circ}\text{C}/2.5\text{dpa}$. In **7.4.2**, Burgers vector analysis showed that loops A,B and C all had Burgers vector of type $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. The tilting experiments in **Fig.7.5.1.1**, shows that the long axes of loops A and B turn clockwise as the specimen is tilted from the $[001]$ pole towards the $[113]$ and $[111]$ poles. This is consistent with a circle in the $(1\bar{1}1)$ plane: thus loops A and B have Burgers vector $\frac{1}{2}[1\bar{1}1]$. In **image (a,b)**, loops A and B show outside contrast in $g=1-10$, and so are interstitial in nature. Similarly, it can be seen that the long axis of loop C turns anti-clockwise during the tilting experiments, which indicates that its habit plane is $(\bar{1}11)$, and so loop C has Burgers vector $\frac{1}{2}[\bar{1}11]$. Since loop C shows outside contrast in

$g=-110$, it is also interstitial in nature. All 7 loops analysed in the specimen were found to be interstitial.



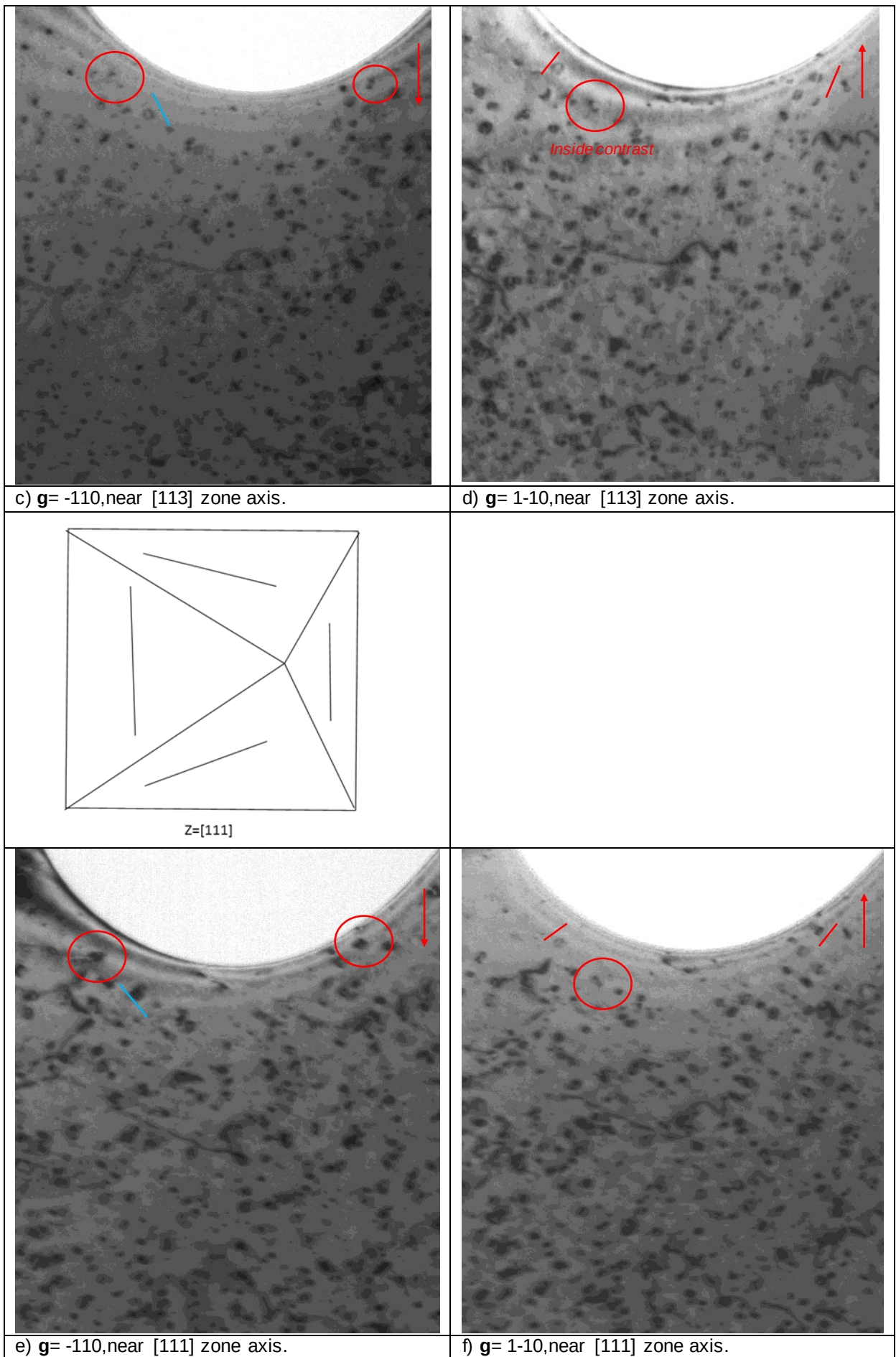


Fig.7.5.1.1. Loop nature determination and habit plane analysis.

7.5.2 Characterisation of 400°C/7dpa specimens

Fig.7.5.2.1. shows the loop nature and habit plane determination for a Fe11Cr specimen irradiated at 400°C/7dpa. The circled loops have Burgers vector of either $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. From the images, it can be seen that loops A, B and C all turn anti-clockwise as the sample is tilted from the [001] to the [111] zone axis. This indicates that these loops are on the $(\bar{1}11)$ plane, and so they have Burgers vector $\frac{1}{2}[\bar{1}11]$. These loops show outside contrast in $\mathbf{g}=-110$. Using the method described previously, it can be concluded that they are all interstitial in nature. Loop D can also easily be shown to be interstitial in nature. All 6 loops analysed in this irradiation condition were interstitial.

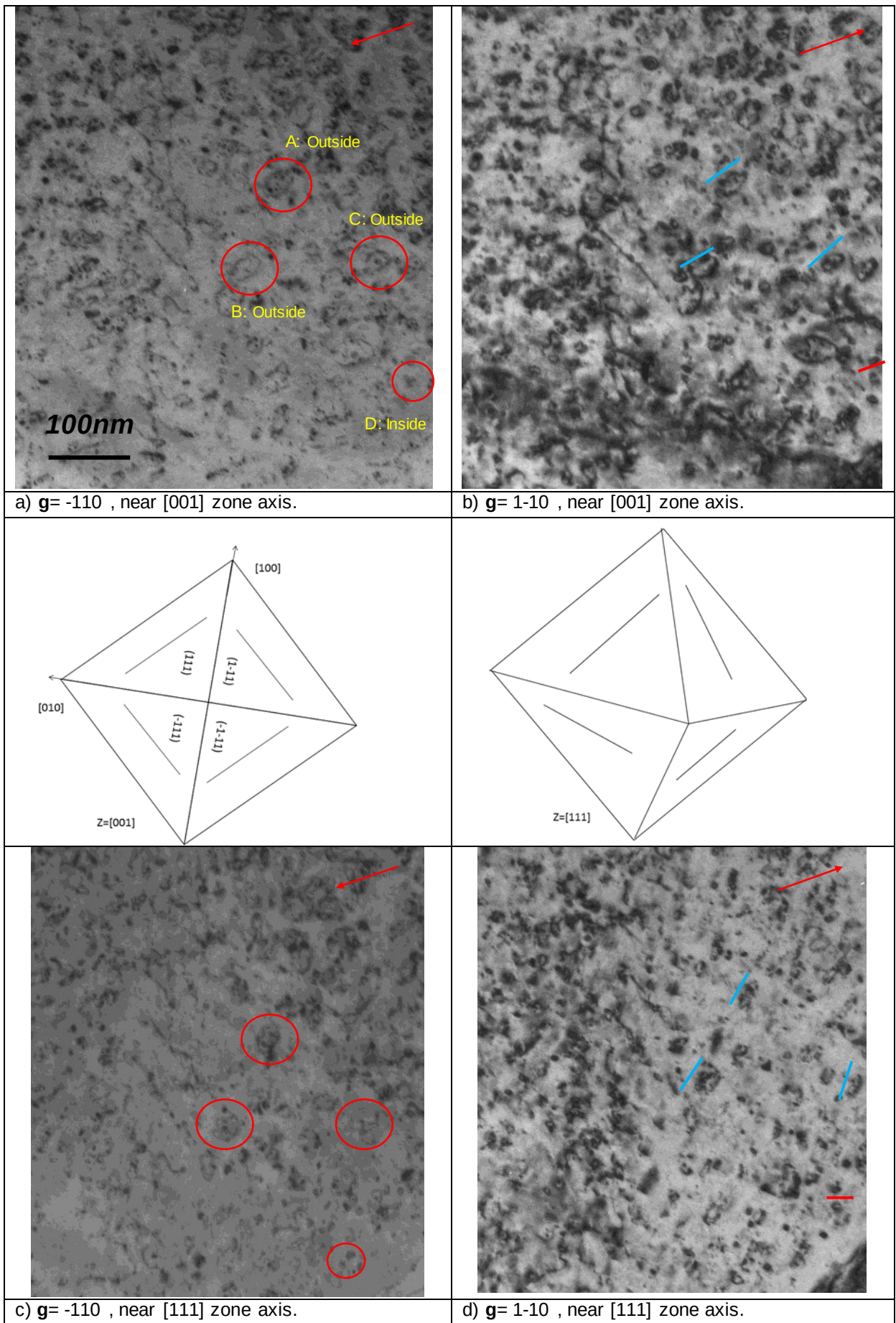


Fig.7.5.2.1. Loop nature and habit plane analyses in Fe11Cr specimen irradiated at 400°C/7dpa.

7.5.3 Characterisation of 460°C/7dpa specimens

Fig.7.5.3.1 shows the nature and habit plane determination for a specimen irradiated at 460°C/7dpa. The contrast analysis in **7.4.4** allows the Burgers vectors of the circled loops in **Fig.7.5.3.1** to be identified unambiguously, since the analysis was carried out in three different zone axes with seven reflections. Loops A and B have Burgers vectors $\frac{1}{2}[11\bar{1}]$. Loops C, D and E have Burgers vectors $\frac{1}{2}[\bar{1}11]$. Loops A and B show outside contrast in $\mathbf{g}=10\bar{1}$, while loops C, D and E show outside contrast in $\mathbf{g}=\bar{1}01$. Thus all of the loops are interstitial in nature. It proved impossible to determine the habit planes of these loops by a tilting experiment, probably because they were too small.

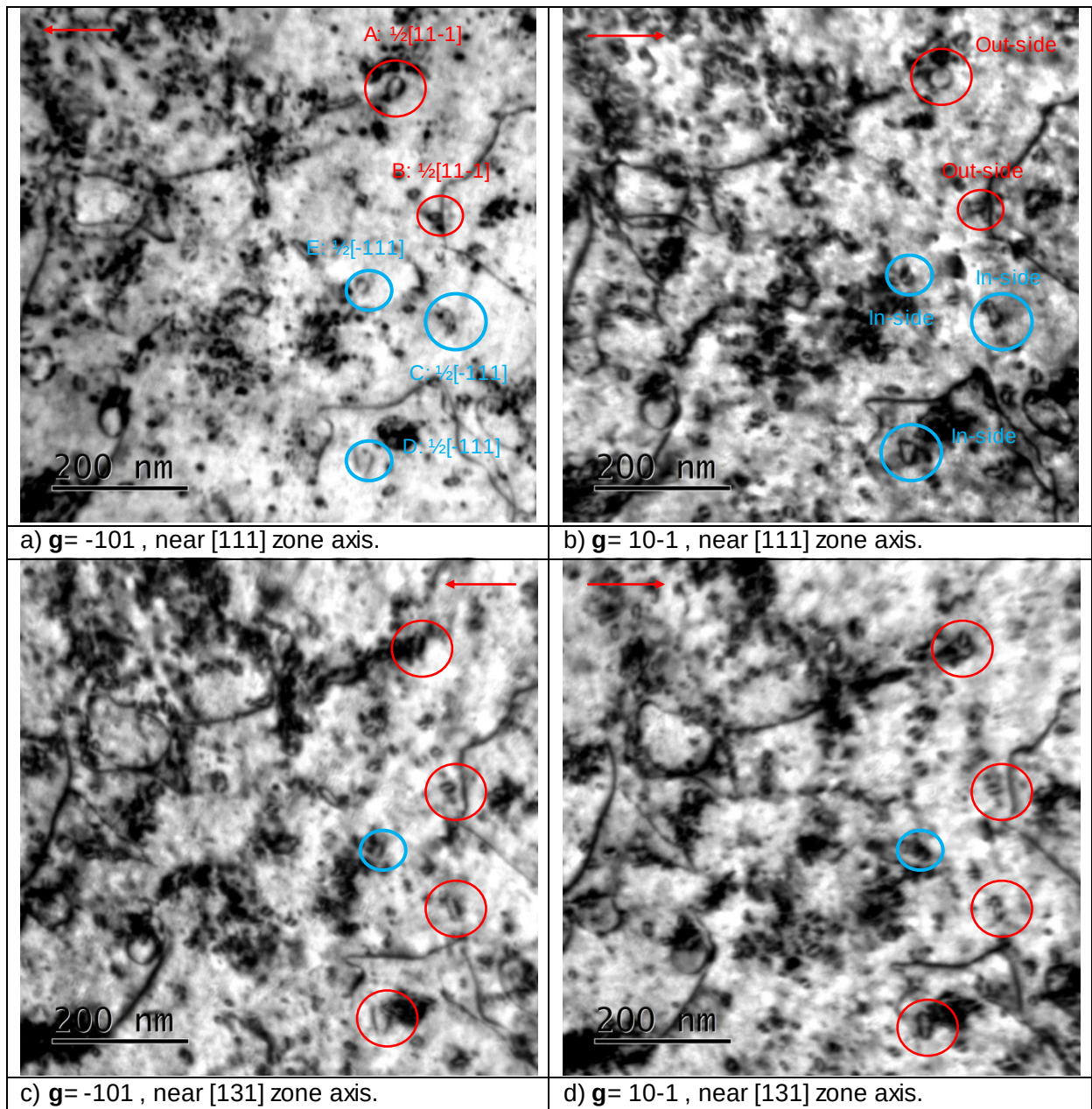


Fig.7.5.3.1. Loop nature and habit plane analyses in Fe11Cr specimen irradiated at 460°C/7dpa.

7.6 Summary of Results

Microstructures:

1. The microstructure development in Fe11Cr at 300°C and 400°C was very similar to that found in the Fe8Cr specimen. The damage microstructure took the form of small dislocation loops, and at 400°C (7dpa), dislocation interactions were observed. At 460°C, complex microstructures such as large loop clusters and broken finger-shaped loops appeared, very similar to the features seen in Fe irradiated at 400°C to 7dpa.

Loop Sizes:

2. In Fe11Cr, the average and maximum loop size stayed stable at 300°C and 400°C, while there was a large increase from 400°C to 460°C.

Loop number density:

3. The loop density in Fe11Cr alloys increased slightly from 300°C to 400°C. The large drop at 460°C was caused by the onset of complex microstructures.
4. At 400°C a higher loop density was observed at 7dpa than at 2.5dpa.

Loop Burgers vectors:

5. Little trend on the temperature effects on loop Burgers vector was seen.
6. The trend for an increasing proportion of $\frac{1}{2}\langle 111 \rangle$ with increasing Cr content continued.

Loop Nature:

7. All loops which were analysed were found to be interstitial in nature.

Chapter 8: Discussion and Conclusions

Due to the resolution limit of TEM and the foil quality, the observations made in this work relate only to clusters of size larger than ~ 2 nm, which comprise of the order of 100 point defects. Clusters of size less than 2 nm are almost certainly present, perhaps even in large numbers, but they cannot be observed directly by TEM.

8.1 Loop Nature

Throughout this work, the loop nature was determined using the inside-outside contrast method which has been fully described in **3.5**. Although this method is the most reliable way to determine the nature of loops, its limitation is that it can only be used for loops which show clear inside and outside contrast. That means the loops had to be relatively large, normally with a diameter of at least 5 nm. In this work, it was found that all loops which were analysed were interstitial in nature. The sizes of the examined loops ranged from 5-100 nm. So the question left is: where are the vacancies?

There are several possibilities. First, vacancy loops may exist of size smaller than 5nm. Evidence for this can be seen in the work of Yao et al. **[26, 102]**, who studied the nature of loops in Fe-11Cr produced by low-dose 30 keV Ga⁺ FIB irradiation. The loops they observed were of size about 2-5 nm, and B-W contrast method was applied to study their nature. They found that at least some of the loops were vacancy in nature, and were probably produced by a cascade collapse mechanism. However the defect yield was low, so most cascades did not collapse to loops. Another possibility is that the vacancies exist in the form of micro-voids which are invisible under TEM. Experimental results supporting this possibility were obtained by Zinkle and Singh's work **[145]**. These authors found using positron annihilation methods that a high density of small three-dimensional vacancy clusters was

produced in Fe-Cr by 70°C neutron irradiation. The diameters of these vacancy clusters ranged from 0.3-1.0nm, and thus they were too small to be detectable by TEM. Moreover, in the in-situ experiments of Jenkins et al. [101] in Fe-8Cr alloys irradiated with 150 keV Fe⁺ ions at 500°C, small voids with the diameter of about 2 nm were found. Furthermore, in this in-situ work small $\frac{1}{2}\langle 111 \rangle$ loops were seen to coalesce into large interstitial loops at 300°C, and small mobile $\frac{1}{2}\langle 111 \rangle$ loops were seen to be subsumed into large interstitial $\langle 100 \rangle$ loops at higher temperature (>450°C) [34]. Both of these observations suggest that large numbers of small loops were of interstitial nature as well, although it cannot be excluded that some of the visible small loops may have vacancy nature.

Another question is: after the formation of micro-vacancy clusters, why do they not grow to clusters of visible sizes? One possible explanation is related to the mobility of point defects. As described in 1.1, interstitials are much more mobile than vacancies. After the formation of initial clusters, the large number of migrating interstitials will increase the size of interstitial clusters but decrease the size of vacancy clusters.

8.2 The Formation of Complex Damage Microstructures

8.2.1 The Development of Perpendicular Loop Clusters

In Fe specimens irradiated at 400°C to a dose of 2.5 dpa, perpendicular loop clusters were observed, distributing homogeneously within the sample. As described in chapter 4, the clusters consisted of planar accumulations of $\mathbf{b} = \langle 100 \rangle$ loops (see Fig.4.1.5, Fig.4.1.6.), formed in the {100} or {010} planes, and each containing numerous small loops of size 2-10nm. The clusters aligned roughly along $\langle 100 \rangle$ directions. The loops were pinned together and there was no evidence of coalescence. This formation of loop clusters seems similar to the mechanism described in loop raft formation [102, 146-149].

A loop raft is an alignment of loops which is believed to result from loop–loop elastic interaction. In order to achieve an overall low energy configuration, loops on neighbouring planes adjust their positions and orientations and form loop rafts. Loop rafts were first observed in graphite in 1963, and have since been seen in many other materials [150-155].

In 2005, Wen et al. [149] discovered in kinetic Monte Carlo simulations that raft formation can be achieved by prismatic glide of one-dimensional (1D) interstitial clusters, combined with strain-field-induced Burgers vector rotations. Wen et al. described their model as follows [149]:

“Mutual elastic interactions between clusters were found to affect their distribution and motion drastically. Because of mutual interaction, two clusters that are oriented along non-parallel crystallographic directions will either coalesce forming a larger one or rotate and pin one another at a short distance and move jointly in the same direction. Once two clusters are pinned together, they have less chance to change their orientation, and therefore their motion becomes almost pure one-dimensional. As this process proceeds, some additional clusters may be trapped into this pinned structure by changing their Burgers vectors.”

Wen et al.’s model appears to be a good explanation of the observed loop cluster formation in our work, although in their work it was impossible to view loop interactions in real time. Evidence can be listed as follows:

- a) The loop clusters were mainly formed by loops of the same Burgers vector;
- b) The loops within the clusters interacted with each other by pinning, rather than by coalescence;
- c) Small loops visible as black dots were observed to accumulate around and inside existing clusters.

The nature of the loops within the clusters cannot be determined. Their mobility and nature determinations of larger loops suggest however that they are more likely to be interstitial than vacancy.

Besides the presence of $\langle 100 \rangle$ loop clusters, several other observations in this work can also be explained by Wen et al.'s simulations:

- a) In Fe samples irradiated at 460°C to a dose of 2.5 dpa, loop strings containing up to 10 loops with $\mathbf{b} = \frac{1}{2}\langle 111 \rangle$ Burgers vector were seen, with these loop strings aligned roughly along $\langle 110 \rangle$ directions. The formation of the strings again seemed to involve some sort of cooperative interactions.
- b) In Fe samples irradiated at 300°C to a dose of 7dpa, the majority of the loops were restricted to a narrow channel with a projected width of 50nm on either side of line dislocations. This observation is consistent with Wen et al.'s simulations. In their study, they found that small clusters are often trapped near the cores of line dislocations. In order to be absorbed by the line dislocations, the small clusters change their Burgers vectors to become parallel to that of the line dislocation [149].

Loop strings and loop rafts were also observed in Fe5Cr, Fe8Cr and Fe11Cr at elevated temperatures, but in a less regular shape.

8.2.2 The Development of Finger Loops

In Fe specimens irradiated at 400°C to a dose of 2.5dpa, finger-shaped loops with length up to one micron and width of about 50 nm were seen, distributed inhomogeneously within the sample. Neighbouring finger-shaped loops aligned parallel to each other and close to $\langle 110 \rangle$ direction. **g.b** analysis carried out in Fe specimens irradiated at 400°C to a dose of 7 dpa proved that the finger loops had $\frac{1}{2}\langle 111 \rangle$ Burgers vectors.

Parallel in-situ TEM studies on Fe and Fe-Cr alloys by Hernandez-Mayoral et al. [26, 34, 101, 102], showed that finger loops are formed by the coalescence of overlapping large interstitial loops. These authors studied heavy-ion irradiations of Fe and Fe-Cr alloys using in-situ TEM methods on the Argonne MEM-Tandem Facility. In their experiments, at doses above 10^{18} ions m^{-2} loop strings along or close to $\langle 110 \rangle$ directions were observed. The strings consisted of interstitial loops with $\frac{1}{2}\langle 111 \rangle$ Burgers vectors. As the irradiation progressed, loops strings were seen to evolve into resolvable loops. As discussed in the previous paragraph, this process can be explained by Wen et al.'s model [149]. In their kinetic Monte Carlo simulations, loop strings of five or more act as the nuclei for large interstitial loops, formed by climb and coalescence. Overlapping large interstitial loops coalesced further to form finger loops of length several hundred nanometres and width several tens of nanometres. Although the detailed mechanism of coalescence is still unclear, it is believed that this process also involves both climb and glide. A possible description of the coalescence mechanism from the work of Hernandez-Mayoral et al. is quoted below [102]:

“Attempts to model loop coalescence by Novokshanov (private communication) by dislocation dynamic simulations are in an early stage. The modelling done so far is only for edge loops. This has shown that when hexagonal edge loops overlap in the z (foil normal) direction they can coalesce by the glide of segments on their joint slip plane. This leads to a stepped habit plane. This may be relevant in understanding how large finger loops can form without intersecting the foil surface; they may not lie entirely on a single habit plane, but would be stepped. The smaller constituent loops may initially have large edge components, but the average habit plane is close to the foil plane. Further simulations better reflecting the geometry of our experiments are in progress.”

In our work it was not possible to view the formation of finger loops directly. However the observations of small loop strings and the coalescence of smaller finger loops

indicates that the mechanism of finger loop formation is probably the same in bulk and thin foil irradiations.

Besides the coalescence mechanism, there are two other phenomena which attract the attention but remain unclear:

- a) Unlike the perpendicular $\langle 100 \rangle$ loop clusters, the $\frac{1}{2}\langle 111 \rangle$ finger loops were distributed inhomogeneously within the sample. What drives the formation of finger loops in these specific areas?
- b) The finger loops are aligned along or close to the $[1-10]$ direction perpendicular to the diffraction vector $\mathbf{g}=\mathbf{110}$, and the majority of $\frac{1}{2}\langle 111 \rangle$ loops which appeared in the proximity had Burgers vectors $\mathbf{b} = \frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$, which indicates that perhaps only the other two $\frac{1}{2}\langle 111 \rangle$ variants developed into finger loops. If that was the case, then again what was the driving force?

One possible explanation is that local stresses in those specific areas were different, which encouraged the formation of finger loops along the $[1-10]$ direction.

8.2.3 The Development of Damage Structure at Elevated Doses

In Fe samples irradiated at 400°C to a dose of 2.5 dpa, complex microstructures which have been described in detail in previous paragraphs were seen (**see chapter 4**). As the implantation dose was increased to 7dpa, strong interactions between $\frac{1}{2}\langle 111 \rangle$ finger-loops and $\langle 100 \rangle$ loop clusters were observed. During the interaction, $\frac{1}{2}\langle 111 \rangle$ finger-loops were broken into twisted line dislocations, twining around the $\langle 100 \rangle$ loop clusters which lost their regular pattern. It can be seen from **Fig.4.1.7**, that after the interaction, the microstructure developed into irregular shaped clusters of several hundred nanometres in size, both in width and length. Inside the clusters were numerous loops seen as black dots with size of around 2-5 nm. Further Burgers vector analysis indicated that both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops were present inside the clusters, and for the selected cluster shown in **Fig.4.1.8**, about 70% of the loops had

$\langle 100 \rangle$ Burgers vector. The mechanism of this complicated loop interactions is not clear.

8.3 Effects of Temperature on Damage Microstructure

As described in the literature review (**chapter 1.3.1**), numerous determinations of the types of loop Burgers vectors in ion/neutron irradiated iron have been made [77, 80, 81, 156, 157]. A large number of the results even seem contradictory. For example, some researchers discovered only $\langle 100 \rangle$ loops in their samples, while others found both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops. Moreover, the relative proportions of the loops were sometimes quite different [132, 158, 159]. However a clear pattern seems to appear that $\langle 100 \rangle$ loops predominate in Fe at high irradiation temperatures ($>400^\circ\text{C}$).

The in-situ experiments carried out by Yao et al. [34] provide further evidence that $\langle 100 \rangle$ loops become increasingly dominant in Fe with increasing temperature. In their experiments, Yao et al. found that at 400°C , about 80% of loops had $\langle 100 \rangle$ Burgers vector, while their mean size was only about one-third of that of the $\frac{1}{2}\langle 111 \rangle$ loops. At 450°C , a majority of the loops were of $\langle 100 \rangle$ type, and very few large $\frac{1}{2}\langle 111 \rangle$ loops were still present in the sample. At 492°C , no $\frac{1}{2}\langle 111 \rangle$ loops were found during the irradiation.

The ex-situ bulk irradiations of Fe carried out in this work show a consistent transition in loop Burgers vectors from 300°C to 500°C , with $\frac{1}{2}\langle 111 \rangle$ loops dominating at lower temperatures, and $\langle 100 \rangle$ loops dominating at higher temperatures. The same trend was found in Fe-Cr alloys (Fe-5Cr, Fe-8Cr, Fe-11Cr). However the proportion of $\frac{1}{2}\langle 111 \rangle$ loops in the alloys was higher at every irradiation temperature compared with that in Fe.

In parallel with Yao et al.'s experiments, Dudarev et al. [88, 160] did theoretical calculations of loop self-energies as a function of irradiation temperatures. Their

calculations showed that in α -Fe, a dramatic reduction of the shear stiffness constant C' occurred near the α - γ phase transition temperature ($T_c = 912^\circ\text{C}$). This reduction in C' has a strong effect on the anisotropic elastic self-energies of dislocations and the relative stabilities of $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$ dislocation loops. Dudarev et al.'s theory predicts that $\langle 100 \rangle$ loops will become relatively more stable in pure Fe with increasing temperature, which is consistent with the observations in both in-situ and ex-situ experiments.

Besides its effects on loop Burgers vectors, the irradiation temperature has profound effects on loop sizes and densities. It can be seen from **Fig.8.3.1.-8.3.2**, that both the average and maximum loop sizes increase with increasing irradiation temperature. The loop density in Fe decreases with increasing temperature. In Fe-Cr alloys, loop densities were fairly similar through all the implantation temperatures, which may be ascribed to the pinning effect of Cr atoms.

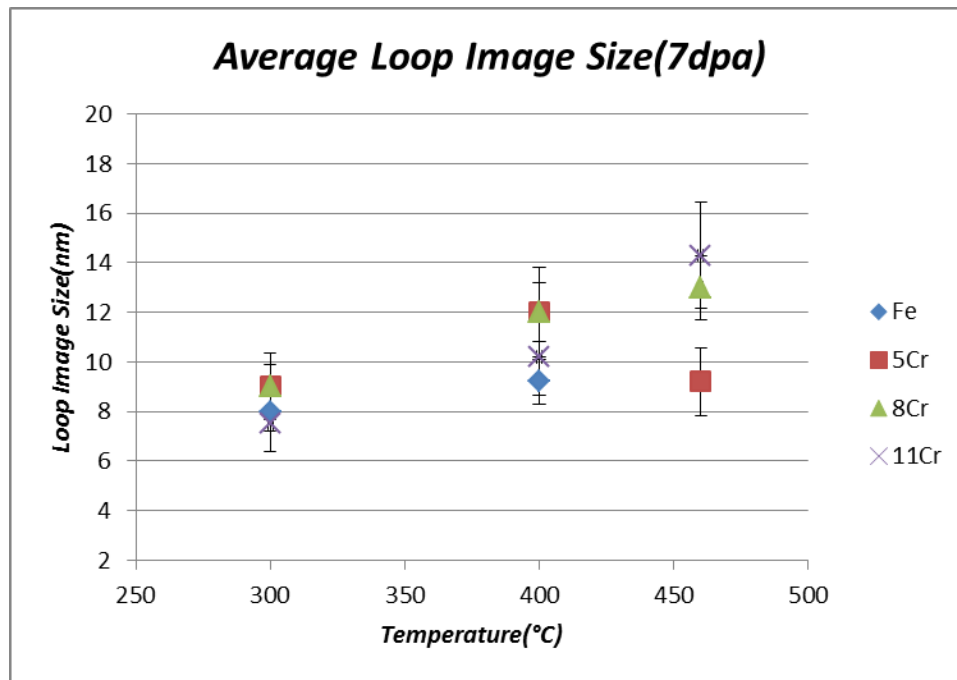


Fig.8.3.1. Average loop image size distribution for Fe and Fe-Cr alloys at different temperatures.

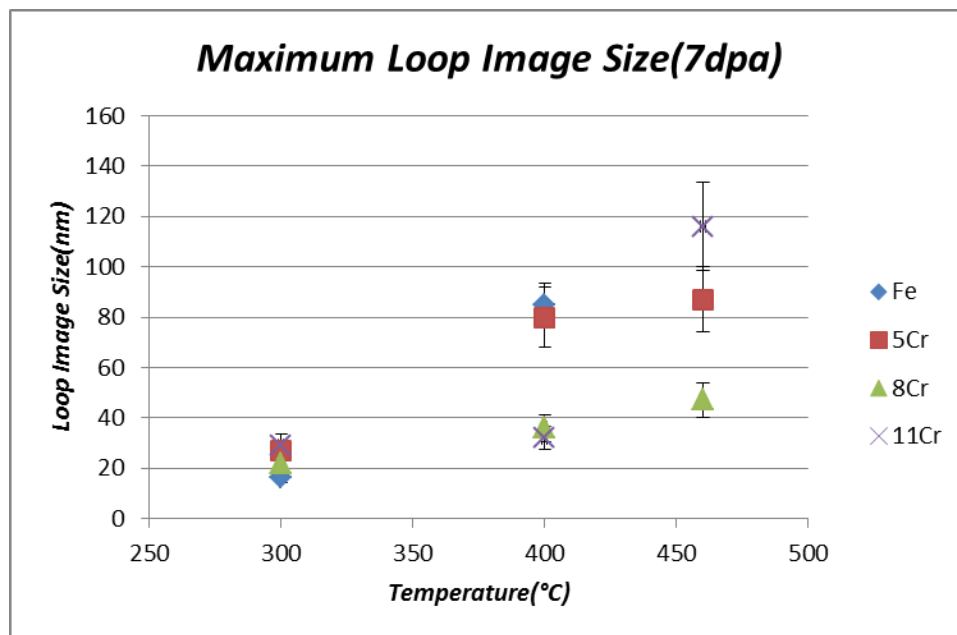


Fig.8.3.2. Maximum loop image size distribution for Fe and Fe-Cr alloys at different temperatures.

8.4 Effects of Cr on Damage Microstructure

8.4.1 Effects on Loop Burgers Vectors

Dislocation loops with both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors were observed both in Fe and Fe-Cr alloys (Fe-5Cr, Fe-8Cr, Fe-11Cr). At 400°C, the proportion of $\frac{1}{2}\langle 111 \rangle$ loops in the pure Fe foil was of the order of 45%, while in the Fe-Cr alloys the proportion was about 50%-70%. This result is consistent with the published works on ion-irradiated and neutron-irradiated materials [14, 81, 161-163]. For example, Gelles et al. [99] found that both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops appeared in their experiments, and a reduction of chromium to the Fe-3Cr range resulted predominantly in $\langle 100 \rangle$ dislocation structures. Porollo et al. [24] also found that dislocation loops with $\mathbf{b}=\langle 100 \rangle$ were dominant in alloys with lower Cr content (Fe, Fe-2%Cr and Fe-6%Cr).

The results presented in this thesis are also consistent with the general trend found in the in-situ experiments, in that the proportion of $\frac{1}{2}\langle 111 \rangle$ loops in an Fe sample was lower than that in Fe-Cr alloys. However, in bulk irradiations, the absolute proportion of $\frac{1}{2}\langle 111 \rangle$ loops was higher than in the in-situ work.

Two possible explanations were proposed by Yao et al. [26] to explain their observations. First, they suggested that the different proportions of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops in the in-situ experiments on Fe and Fe-Cr alloys were caused by the glide of $\frac{1}{2}\langle 111 \rangle$ loops to the foil surface. In an (001) foil, both [100] and [010] loops would be retained in the foil, as their Burgers vector has no component towards the foil surface. However the glide cylinders of all four $\frac{1}{2}\langle 111 \rangle$ variants intersect the foil surface, and thus a large fraction of such loops may be lost during the irradiation due to the surface image force. The dynamic observations confirmed that highly mobile $\frac{1}{2}\langle 111 \rangle$ loops were frequently lost to the surface, especially during ion irradiation.

However in bulk irradiation samples, the loss of $\frac{1}{2}\langle 111 \rangle$ loops to the surface is believed to be less than in thin foils. In thin-foil implantations, the influence of the surface image forces is expected to be severe during implantation, but in bulk samples this effect only arises after thin-foil preparation, when the microstructure is effectively pinned. Little or no loop loss was expected in the TEM in bulk-irradiated foils, although loop loss immediately on sample preparation may occur. Loop loss under the electrons irradiation from the TEM is also possible as well.

The second possible explanation is that $\langle 100 \rangle$ loops are energetically more favoured in Fe compared with Fe-Cr alloys. As yet no simulations have been reported to support this assumption.

8.4.2 Effects of %Cr on Loop Sizes &Densities

Fig.8.4.2.1.-Fig.8.4.2.3.show the distributions of number densities, average sizes, and maximum sizes of loops in Fe and Fe-Cr alloys at 400°C at a dose of 2.5 dpa. Both loop sizes and densities are strongly affected by the Cr content. This is unlike the in-situ work of Yao et al. [26, 102] where little dependence of loop densities on Cr content was found.

The trends that we observed are as follows:

- a) The average and maximum loop sizes saw a clear decrease as the Cr content increased from 0% to 11%.
- b) The loop densities in Fe, Fe5Cr and Fe8Cr were quite similar, while the loop density in Fe11Cr saw an apparent increase compared with the other materials irradiated at the same conditions.

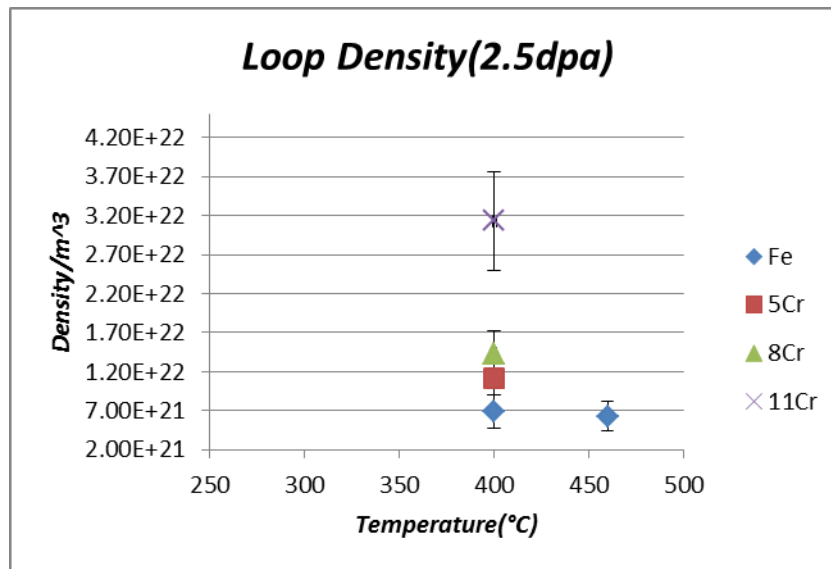


Fig.8.4.2.1. Loop density distributions of Fe and Fe-Cr alloys at 400 °C to a dose of 2.5 dpa

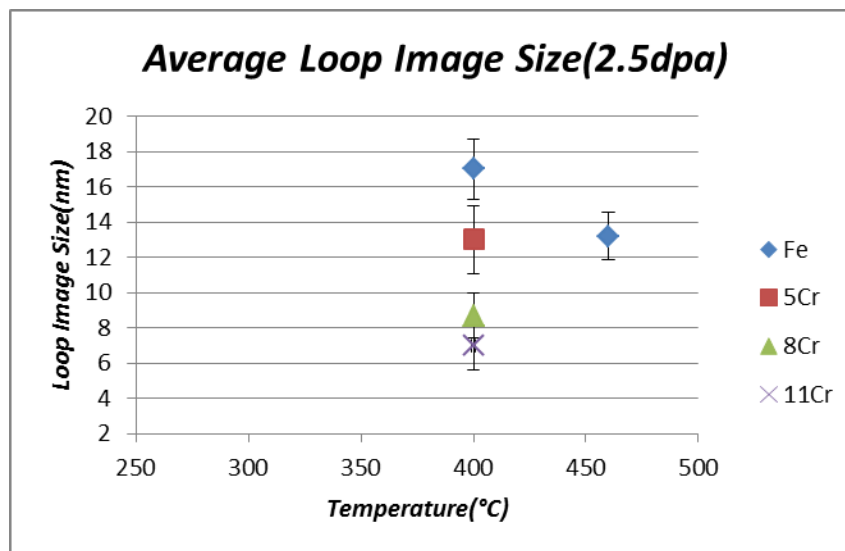


Fig.8.4.2.2. Average loop size distributions of Fe and Fe-Cr alloys at 400 °C to a dose of 2.5 dpa.

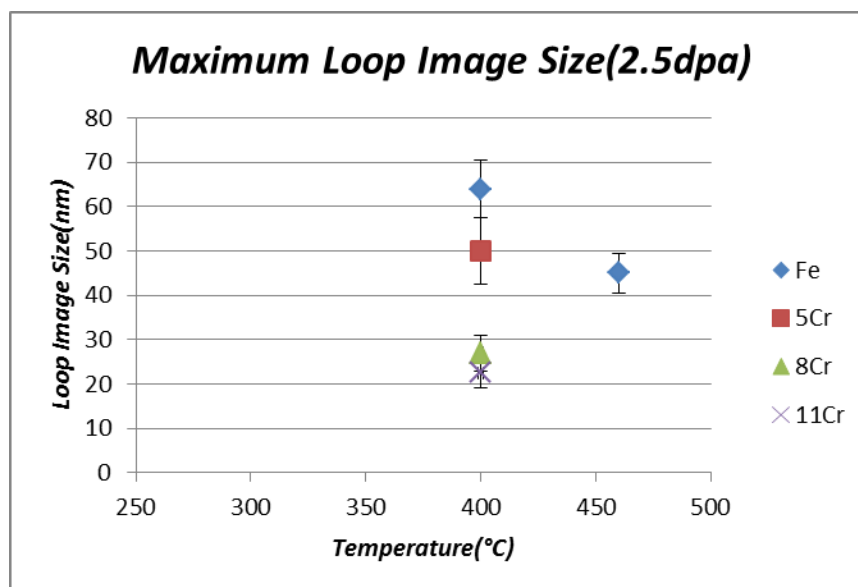


Fig.8.4.2.3. Maximum loop size distributions of Fe and Fe-Cr alloys at 400 °C to a dose of 2.5 dpa.

The different results between thin foil and bulk irradiated samples are believed to be due to loop loss during irradiation in thin foil samples. In the thin foil experiments, loop densities were found to depend strongly on the foil orientation in Fe, but not in Fe-11Cr alloys. In pure Fe, Yao et al. [26] found that at the same irradiation conditions, loop number densities were higher in (110) foils than (001) and (111) foils. This observation is easy to understand. In a (110) foil, the glide cylinders of two $\frac{1}{2} \langle 111 \rangle$ variants as well loops with [001] Burgers vector lie in the foil plane, and thus these loops are not able to glide to the surface. However in an (001) foil, all loops with $\frac{1}{2} \langle 111 \rangle$ Burgers vector are able to glide to the surface, as their Burgers vectors have components towards the foil surface. A high proportion of loops may also be lost to the surface in a (111) foil. In the Fe-Cr alloys loops were less mobile, so fewer were lost and so the effect of foil orientation is less. The effect of foil orientation has not been investigated in bulk-irradiated samples. As discussed above, loop loss to the surface is less likely in bulk-irradiated samples, but may account to some extent for the difference in loop populations in Fe and the Fe-Cr alloys.

8.5 Conclusions

- 1) The evolution of radiation damage under heavy-ion irradiation in bulk samples of pure Fe and Fe-5Cr, Fe-8Cr and Fe-11Cr alloys has been investigated for two irradiation doses (2.5dpa, 7dpa), and at three irradiation temperatures (300°C, 400°C, 460°C).
- 2) Loops with both $\langle 100 \rangle$ and $\frac{1}{2} \langle 111 \rangle$ Burgers vectors were identified in all the samples through all the irradiation conditions. In Fe, the proportion of $\langle 100 \rangle$ loops was higher than in the Fe-Cr alloys at the same irradiation condition. This result may be due in part to the high mobility of $\frac{1}{2} \langle 111 \rangle$ loops in Fe, in that a

proportion of them will be lost to the foil surface on thin-foil preparation. In Fe-Cr samples, especially high Cr alloys as Fe-11Cr, the pinning effects of Cr atoms lead to less loop loss, and so both the total loop density and the proportion of $\frac{1}{2}\langle 111 \rangle$ loops are higher than in Fe.

- 3) Cr effects on loop sizes have been observed, with a decrease in both the average loop size and maximum loop size with increasing Cr content. Also it was found that in Fe-11Cr, the loop density was higher than that in Fe and low Cr alloys under the same irradiation conditions. In Fe-Cr alloys with a high Cr content, the Cr atoms restrict the mobility of interstitial clusters: thus such clusters can only interact with very nearby clusters which lead to a higher density of visible loops with smaller sizes.
- 4) Inside-outside methods were used to determine the nature of the loops. All the loops which were analysed in this work were interstitial in nature. Since this technique can only be applied to loops larger than about 5nm, it can only be concluded that at least the majority of the large loops in our samples were interstitial in nature. Small vacancy loops (less than 5 nm) and small voids have been identified by other researchers, which indicates that maybe the vacancy clusters exist in these forms.
- 5) In Fe irradiated at 400°C to the dose of 2.5 dpa, complex microstructures as loop strings, perpendicular $\langle 100 \rangle$ loop clusters, and $\frac{1}{2}\langle 111 \rangle$ finger loops were observed. The formation of these structures probably involves cooperative alignment and interaction, coalescence of smaller loops. The exact mechanism by which these microstructures formed is still unclear.
- 6) Similar complex features were also observed in Fe-5Cr, Fe-8Cr and Fe-11Cr. But the finger loops and perpendicular $\langle 100 \rangle$ loop clusters were less developed and not as regularly aligned as in Fe.

- 7) At the highest dose of 7 dpa, interaction between $\frac{1}{2}\langle 111 \rangle$ finger loops and perpendicular $\langle 100 \rangle$ loop clusters was observed. Loop clusters as large as 1 micron in width and length appeared during and after the interaction. The loop clusters were composed of numerous small loops with both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors.
- 8) A transition in loop Burgers vectors with increasing irradiation temperature has been observed. At 300°C the proportions of both types of loops were roughly the same. Taken that the $\frac{1}{2}\langle 111 \rangle$ loops were much bigger, the majority of the interstitials were contained in $\frac{1}{2}\langle 111 \rangle$ loops. As the temperature was increased, the proportion of $\langle 100 \rangle$ loops increased. An explanation of these observations has been given in terms of recent theoretical calculations on the relative stabilities of loops as a function of irradiation temperature.

Chapter 9: Radiation Damage at Different Irradiation Dose Rate

Up to now, only limited studies have been made of dose-rate effects in structural materials, and these researches have concentrated mostly on swelling and creep where quite strong effects have been found [164-167]. Unfortunately, no systematic TEM studies have been conducted to investigate micro-structural changes in response to irradiation dose rate.

In this work, the author has designed a series of irradiations to study dose rate effects in UHP-Fe5Cr, 10Cr, 14Cr alloys. Irradiations were carried out at 300°C with two different average implantation dose rates (6×10^{-4} dpa/s and 3×10^{-5} dpa/s). The implantation dose for both implantations was $0.38 \times 10^{19} \text{ Fe}^+/\text{m}^2$ (0.6 dpa). Detailed experimental conditions can be found in 2.2.

In 9.1, the experimental conditions for the micrographs are presented while the damage structures produced in the different materials by iron ions are considered in 9.2. Quantitative analyses of damage structures are presented in 9.3. Mechanisms which might account for the present observations are discussed in 9.4.

9.1 Experimental Conditions

Comparisons of damage structures were made from different areas within a single specimen or from one specimen to another at different irradiation doses. Care was taken to maintain consistent imaging conditions (foil orientation, diffraction vector and deviation parameters) during the experiments. Only grains with an orientation close to [001] or [011] were considered, because they contain variants of expected Burgers vector types which lie in the plane of the examined foil, and so will not slip out of the foil through the influence of surface image forces.

9.2 Irradiation Damage Microstructures

In this section, damage microstructures produced by high- and low-dose rate irradiations of UHP-Fe5Cr, Fe10Cr and Fe14Cr are presented separately, while quantitative results on loop densities, Burgers vectors and loop nature are presented in the following section.

9.2.1 UHP-Fe5Cr at High/Low Dose Rate

The dependence of the damage structure in UHP-Fe5Cr on the implantation dose rate can be qualitatively evaluated from the micrographs presented in **Fig.9.2.1.1(a)-(d)**. The main points to note are:

- (1) Based on observations of non-irradiated foils, where dislocations seemed to be introduced in some foils during sample preparation, the dislocation lines seen in (a) were probably not introduced by irradiation. They were caused by sample preparation in this particular specimen, while in micrograph (b) which was taken from a different specimen, there were few dislocation lines.
- (2) The dislocation loop density in HDR specimens was clearly greater than that in LDR specimens, considering that these examined areas were of similar thickness.
- (3) Most loops produced under both irradiation conditions (LDR and HDR) were of a similar size (which will be illustrated quantitatively in **9.3.2**)
- (4) However, large loops with a size of tens of nanometres (**see red circles in (a), (b)**) only appeared in LDR samples, while all other loops were less than or around 5 nm. Qualitative analysis will be described in **9.4**.

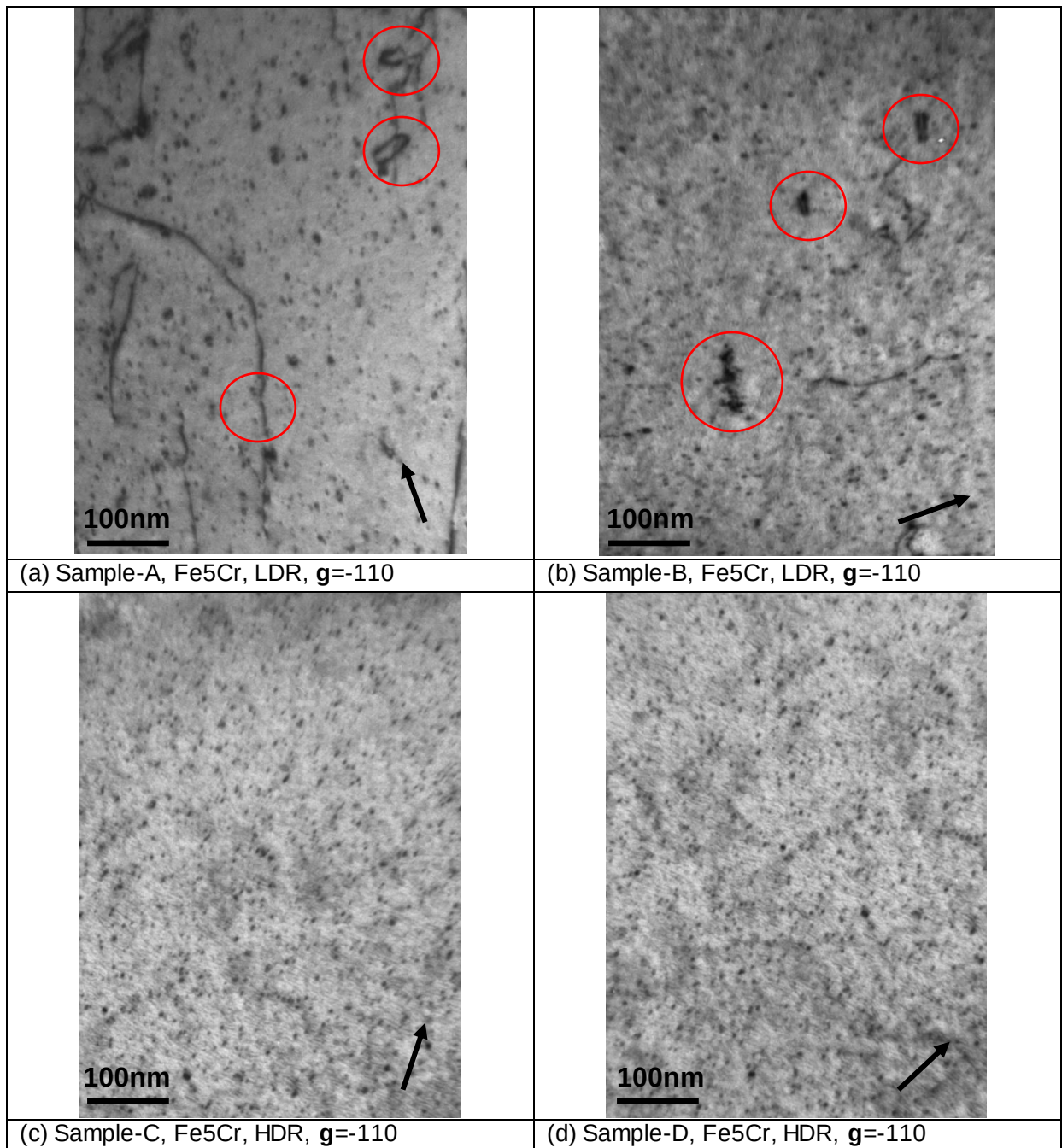


Fig.9.2.1.1. Micrographs of Fe5Cr showing: (a),(b) specimens irradiated at low dose rate at 300°C to 0.6dpa; (c),(d) specimens irradiated at high dose rate at the same irradiation temperature and dose. All the micrographs were taken near an [001] zone axis with $g=-110$.

9.2.2 UHP-Fe10Cr at High/Low Dose Rate

The damage structures in UHP-Fe10Cr took the form of small dislocation loops of size less than 5nm (**see Figure 9.2.2.2**). There were similarities and differences to Fe5Cr. Different from what was found in Fe5Cr, in Fe10Cr no large loops were seen in LDR specimens. The same trend in loop density which was found in Fe5Cr samples was seen: since all samples were confirmed to be of similar thickness by the convergent beam method, the loop density in HDR samples was higher than that in LDR samples. Unlike what was found in Fe5Cr samples, the average loop size was clearly larger in HDR samples than in LDR samples. These trends will be qualitatively described in **9.3**.

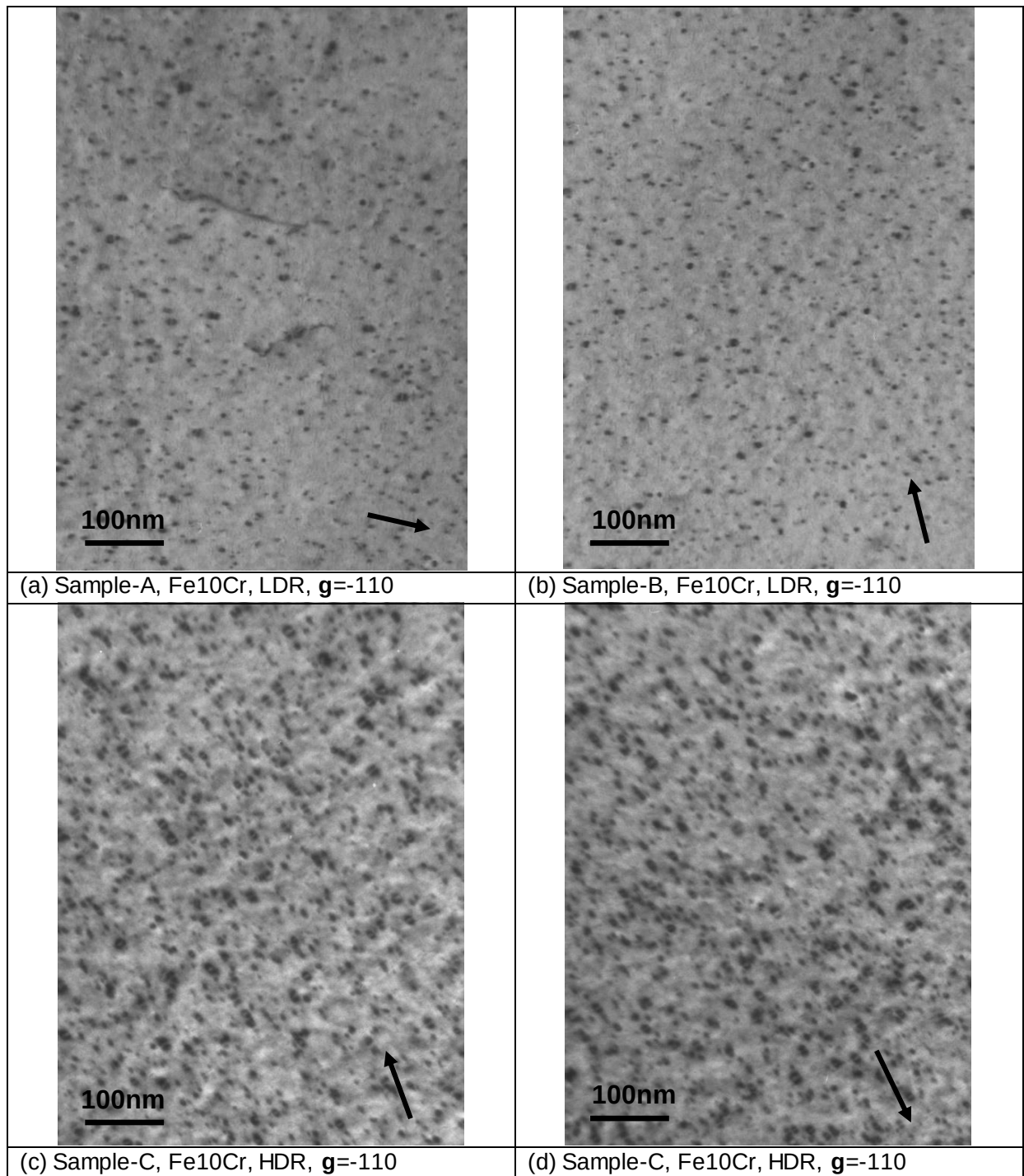


Fig.9.2.2.1. Micrographs of Fe10Cr showing: (a),(b) specimens irradiated at low dose rate at 300°C to 0.6dpa; (c),(d) specimens irradiated at high dose rate at the same irradiation conditions. All the micrographs were taken near an [001] zone axis with $g=-110$.

9.2.3 UHP-Fe14Cr at High/Low Dose Rate

The trends found in UHP-Fe14Cr samples were the same as those in UHP-Fe10Cr samples (see Fig.9.2.3.2):

(1) In HDR samples, the loop density was clearly higher than that in LDR samples.

(2) In HDR samples, the average loop size was larger than that in LDR samples.

Moreover, compared to the loops in Fe10Cr samples, loop sizes in Fe14Cr samples were somewhat larger and apparently there were more edge-on loops (see red circles) seen as pairs of straight lines under a diffraction condition $\mathbf{g}=-110$ near the $[001]$ zone axis. Later $\mathbf{g}\cdot\mathbf{b}$ analysis in 9.3.4 discovered that those edge-on loops had $\frac{1}{2}\langle 111 \rangle$ Burgers vectors.

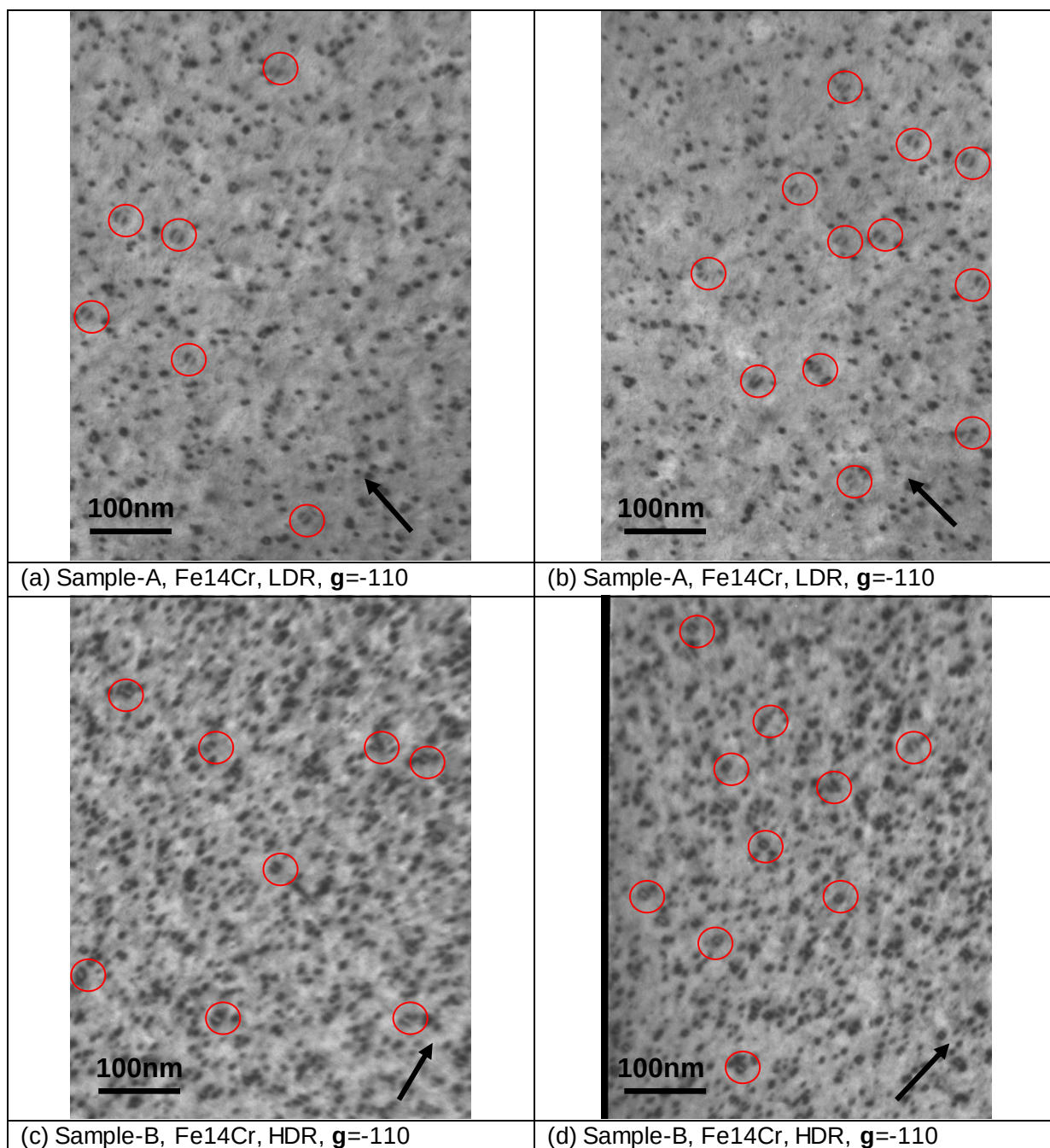


Fig.9.2.3.1. Micrographs of Fe10Cr showing: (a),(b) specimen irradiated at low dose rate at 300°C to 0.6dpa; (c),(d) specimen irradiated at high dose rate at the same irradiation temperature and dose. All the micrographs were taken near an [001] zone axis with $g=-110$.

9.3 Irradiation Damage Characterizations

In this section, quantitative analyses of loop densities, sizes, Burger-vectors and nature are presented. Number density and loop image size measurements presented in this section were carried out from micrographs recorded in $\mathbf{g}=-110$ near the $[001]$ zone axis. All the micrographs were taken under bright-field kinematical conditions. For the samples described in this chapter, the thickness varied from 130nm to 150nm. For loop size measurements at least 300 loops were measured in at least two different areas. The results of all the $\mathbf{g.b}$ analyses in this section are based on the loops whose Burgers vectors could be accurately identified in selected areas. Normally the number of loops analysed in this section in order to reach a reliable conclusion was around 200. In some of the cases in this work it was difficult to identify the Burgers vectors of loops smaller than about 3nm, due to weak image contrast or poor surface conditions. This means that the $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratios presented in this work are more representative of larger loops ($>3\text{nm}$). In addition to this, account has to be taken of loops which remain out-of contrast throughout all the imaging conditions used. In the most common foil orientation $\mathbf{n} = [001]$ used in this work, loops with $\mathbf{b} = [001]$ are out-of-contrast ($\mathbf{g.b} = 0$) in all four diffraction vectors which are available near this pole. See **Table 8.3.1**.

g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(110)$	1	0	0	1	1	1	0
$\pm(020)$	1	1	1	1	0	2	0
$\pm(200)$	1	1	1	1	2	0	0
$\pm(-110)$	0	1	1	0	1	1	0

Table.9.3.1. $\mathbf{g.b}$ table for $n = [001]$.

9.3.1 Loop Number Densities

Fig.9.3.1.1 shows the loop number densities at the two different irradiation dose rates. It can be seen that a 20x increase in dose rate caused about a 2x increase in loop density in Fe5Cr and Fe14Cr, but only a 1.4x increase in Fe10Cr. At the lower dose rate, the loop densities of the three given alloys were similar, whilst at the higher dose rate except the loop densities in Fe5Cr and Fe14Cr were similar but significantly higher than in Fe10Cr.

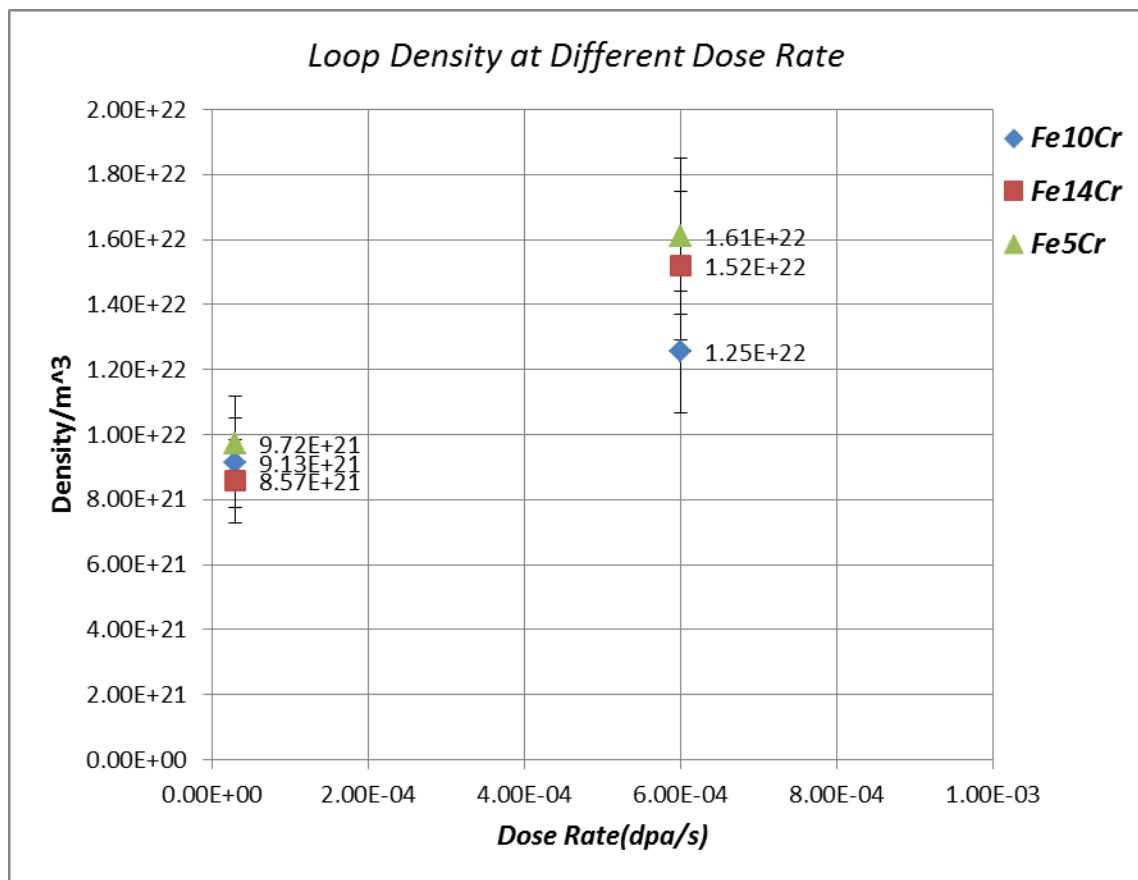


Fig.9.3.1.1. Loop number densities in UHP-Fe-Cr alloys (5Cr, 10Cr, 14Cr) at LDR and HDR.

9.3.2 Loop Image Size Distributions

Fig.9.3.2.1 shows the loop image size distributions measured for Fe5Cr samples at HDR and LDR at 300°C. The histograms presented in **Fig.9.3.2.1** represent the percentage of the visible loop population within different loop image size ranges based on the measurement of 300-400 loops at the same diffraction conditions ($g=-110$ near $[001]$ zone axis). Due to the resolution limitation, loops smaller than 2nm are usually invisible. Loops which were smaller than 3nm were put into the '<3nm' bin. There is no trend that a 20x increase in irradiation dose rate causes a significant change in loop sizes. The loops in LDR samples have a wider distribution and few loops with extremely large size (>20nm) were noted.

Fig.9.3.2.2 and **Fig.9.3.2.3** both show a clear shift to larger loop sizes at larger dose rate in Fe10Cr and Fe14Cr samples respectively. In LDR samples (Fe10Cr, Fe14Cr) the majority of the loops (>50% for Fe10Cr, >60% for Fe14Cr) fall in the range of 5-10nm, while in HDR samples the majority of the loops (>60% for Fe10Cr, >50% for Fe14Cr) have the size of 10-15nm.

The changes in average loop sizes with dose rate in Fe5Cr, Fe10Cr and Fe14Cr are shown graphically in **Fig.9.3.2.4**. It is clear that loop sizes increase with dose rate in Fe10Cr and Fe14Cr. Fe10Cr samples have the largest increase (about 60%) in loop size, while in Fe5Cr there is no obvious change in average loop size.

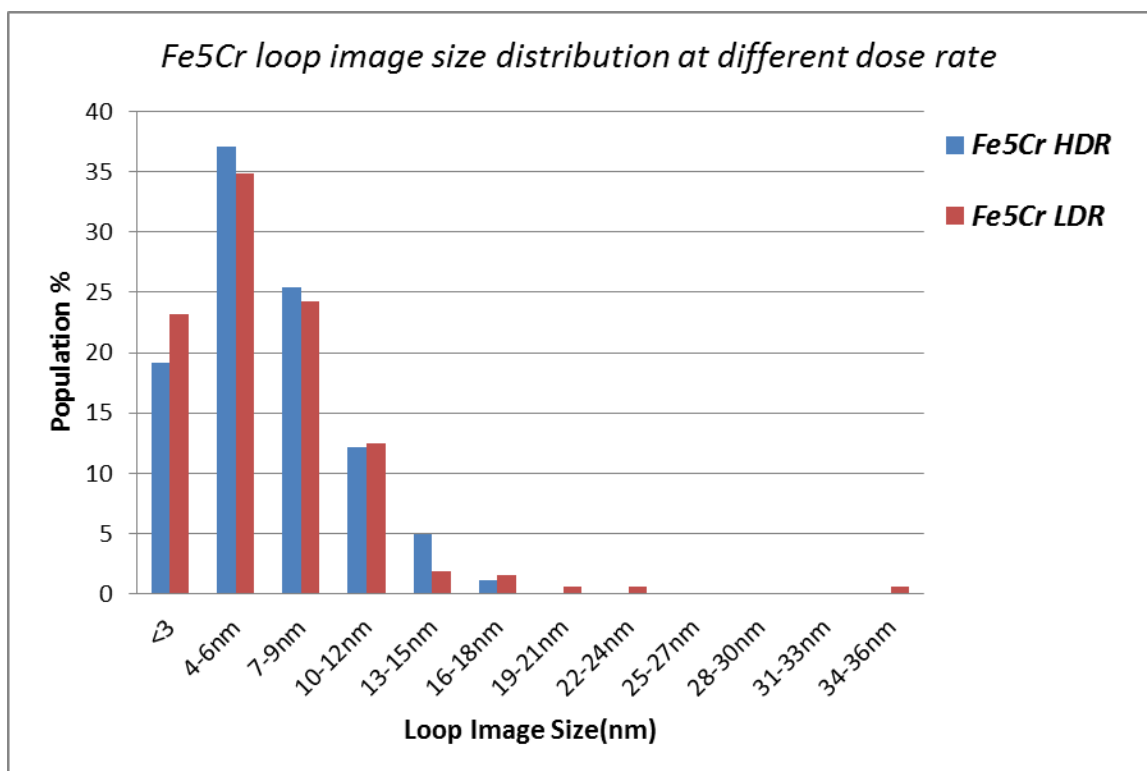


Fig.9.3.2.1. Loop image size distributions in UHP-Fe5Cr at LDR and HDR.

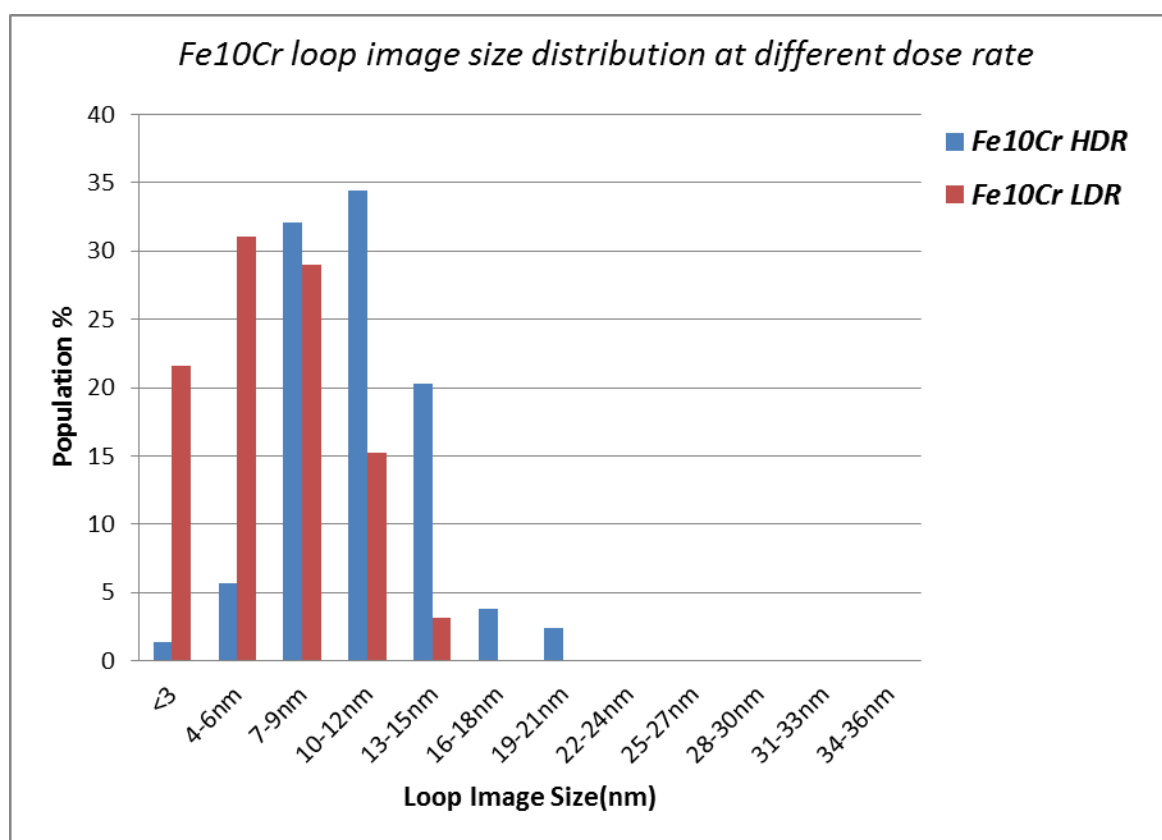


Fig.9.3.2.2. Loop image size distributions in UHP-Fe10Cr at LDR and HDR.

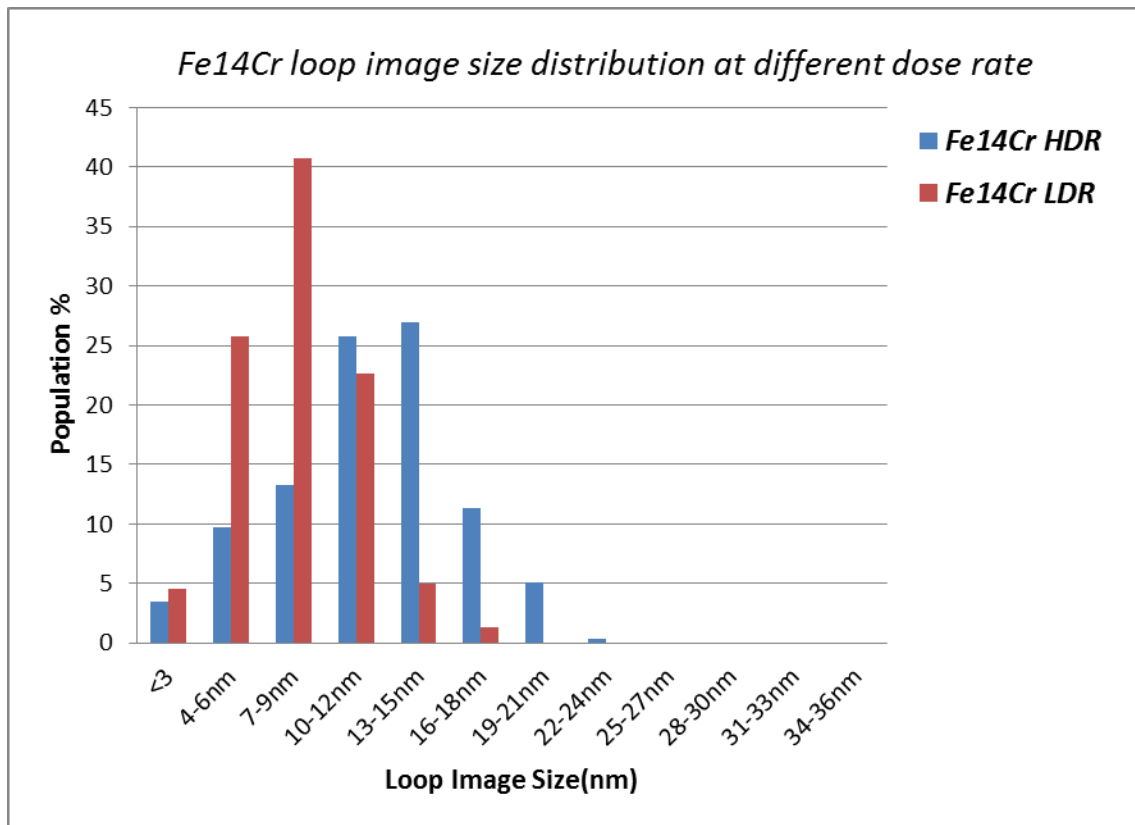


Fig.9.3.2.3. Loop image size distributions in UHP-Fe14Cr at LDR and HDR.

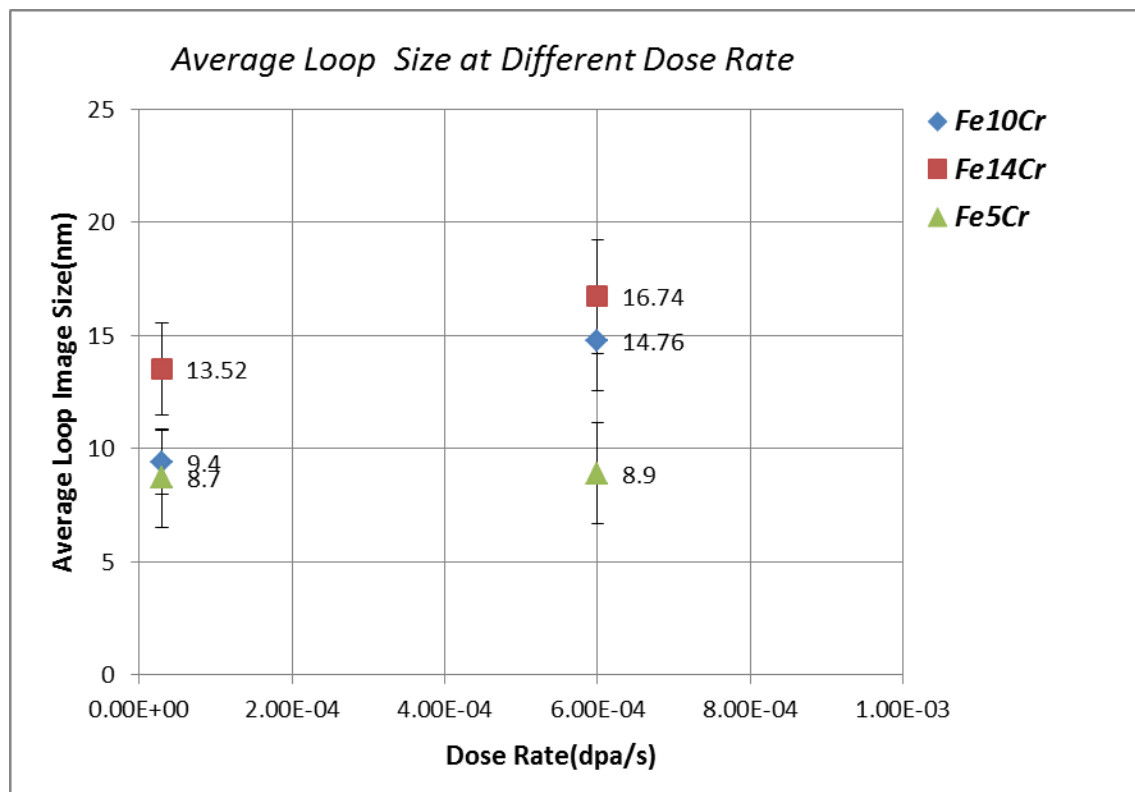


Fig.9.3.2.4. Average loop sizes for Fe5Cr, Fe10Cr and Fe14Cr at different dose rates.

9.3.3 Loop Burgers Vectors

In bcc Fe and FeCr alloys, there are two possible types of Burgers vector: $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$. The Burgers vectors of the loops were determined by use of the standard $\mathbf{g} \cdot \mathbf{b} = 0$ invisibility criterion which was described in detail in 3.3.

UHP-Fe5Cr

Table 9.3.1. shows the $|\mathbf{g} \cdot \mathbf{b}|$ table for [001] foil orientation. With the aid of four images taken in the four reflections available at this zone axis, a fairly complete $\mathbf{g} \cdot \mathbf{b}$ analysis could be done. All the loops of type $\frac{1}{2}\langle 111 \rangle$ stay in contrast in both $\mathbf{g} = 200$ and 020 diffraction conditions, while two variants of $\langle 100 \rangle$ loops are out of contrast in one of the $\langle 100 \rangle$ $\mathbf{g}$ -vectors. Loops with $\mathbf{b} = [001]$ would remain out of contrast in all the reflections. In practice, two images taken in $\mathbf{g} = 200$ and 020 are sufficient to distinguish between loops of type $\langle 100 \rangle$ and $\frac{1}{2}\langle 111 \rangle$.

Fig.9.3.3.1. shows a series of images taken near the [001] zone axis in a Fe5Cr specimen irradiated at low dose rate at 300°C to a dose of 0.6 dpa. The images from the same area in four reflections are presented. Both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loops are present. Some loops are ringed for easier identification. The loops which are visible in images (a), (c), (d), while are out of contrast in (b) have $\mathbf{b} = \frac{1}{2}[-111]$ or $\frac{1}{2}[1-11]$. Those only visible in (b), (c), (d), have $\mathbf{b} = \frac{1}{2}[111]$ or $\frac{1}{2}[11-1]$. Those visible only in (a), (b), (d) have $\mathbf{b} = [100]$. Those visible only in (a), (b), (c) have $\mathbf{b} = [010]$.

Of the 184 loops visible in this area, 40 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$ and 103 had Burgers vector of type $\langle 100 \rangle$ whilst 41 loops of the loops were too small for the Burgers vector to be identified.

Similar Burgers vector analysis was carried out on Fe5Cr samples irradiated at high dose rate at the same temperature and dose. Of the 209 loops counted, 58 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, 113 had Burgers vector of type $\langle 100 \rangle$ and it was not possible to identify 38 loops. The results for the $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratio on Fe5Cr

samples are presented in **Fig.9.3.3.2**. It is seen that in Fe5Cr the majority (over 60%) had Burgers vector of type $\langle 100 \rangle$ at both dose rates and there was no obvious change in the $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratio.

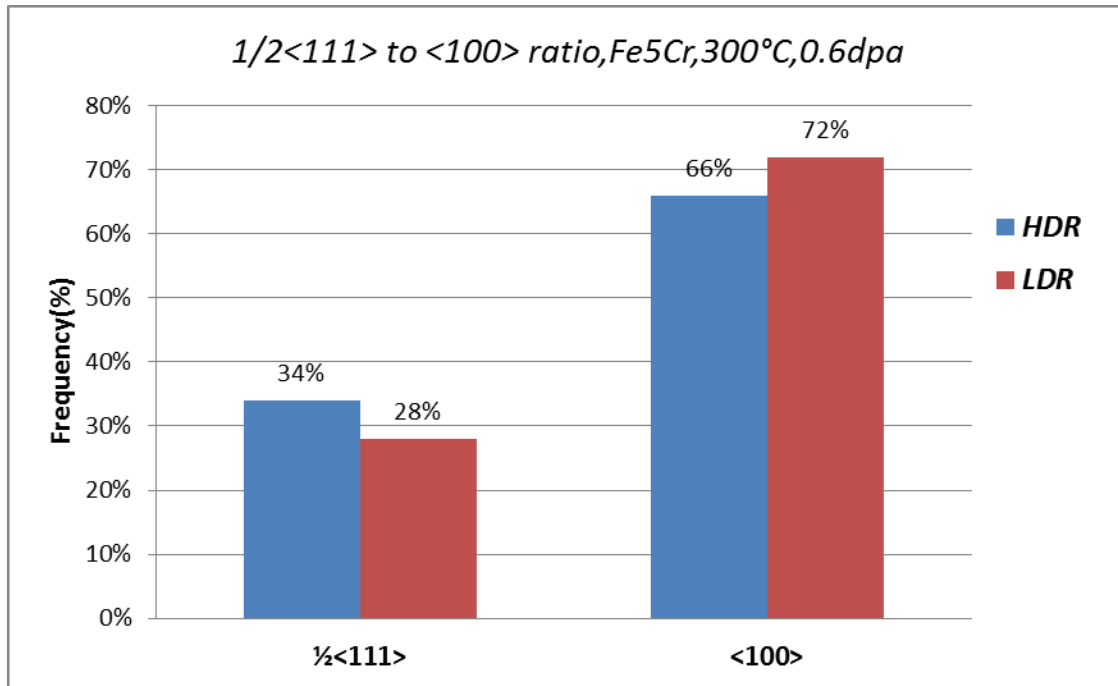


Fig.9.3.3.2. Fraction of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loop types in Fe5Cr. 184 loops counted in the selected area.

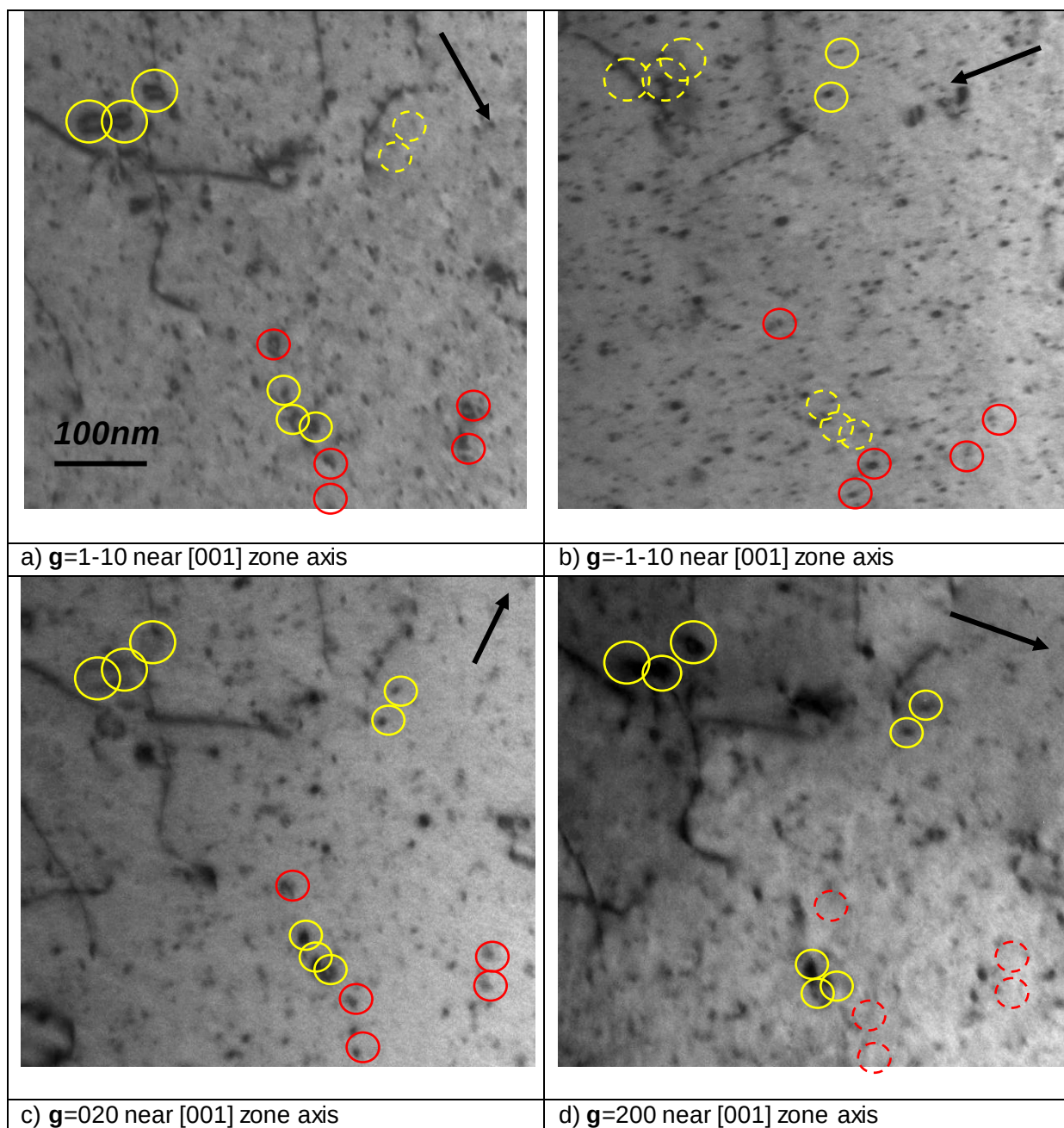


Fig.9.3.3.1. Loop morphology in Fe5Cr at $z= [001]$, irradiated at 300°C to 0.6dpa, LDR. (yellow circle: $b=\frac{1}{2}\langle 111 \rangle$, red circle: $b=\langle 100 \rangle$, dotted circle: invisible).

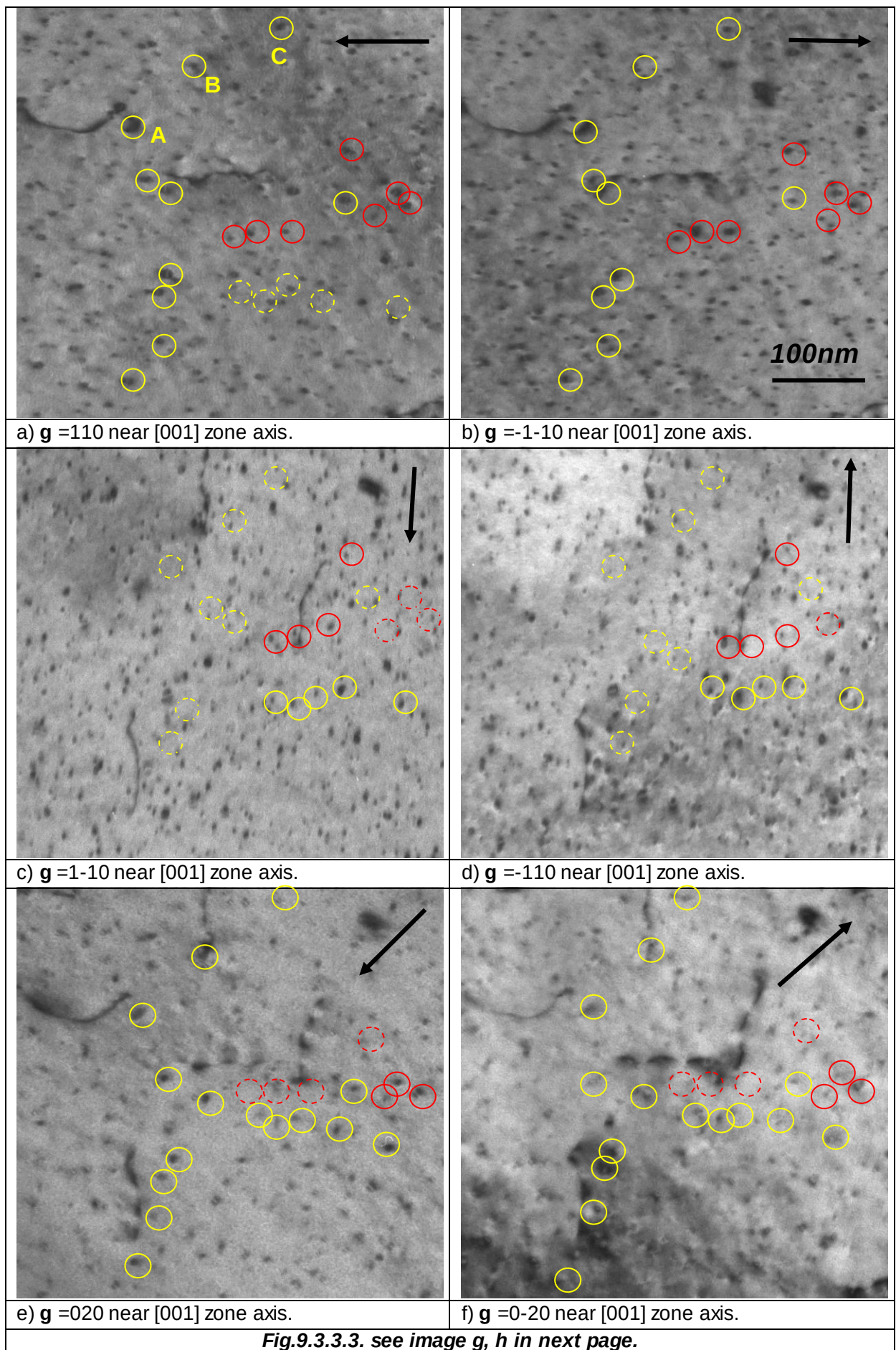
UHP-Fe10Cr

Fig.9.3.3.3. shows a series of images taken near the [001] foil normal orientation in a Fe10Cr specimen irradiated at low dose rate at 300°C to a dose of 0.6 dpa.

Kinematical bright-field (KBF) micrographs of a same area were taken using the four diffraction vectors available at this pole ($\mathbf{g} = 110, 1\text{-}10, 200, 020$), and both $+\mathbf{g}$ and $-\mathbf{g}$ micrographs were obtained. Loops were then analysed as above.

Of the 123 loops appearing in this area, 46 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$ and 56 had Burgers vectors of type $\langle 100 \rangle$ and 21 were too small for their Burgers vectors to be determined. Similar Burgers vector analysis was carried out for Fe10Cr samples irradiated at high dose rate at the same temperature and dose. Of the 201 loops counted, 72 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, 95 had Burgers vector of type $\langle 100 \rangle$ and 34 loops were too small for their Burgers vectors to be determined. The results for the $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratio for Fe10Cr samples are presented in

Fig.9.3.3.4. It is seen that in Fe10Cr, the proportions of $\frac{1}{2}\langle 111 \rangle$ loops were 43% and 45% for high dose rate samples and low dose rate samples respectively.



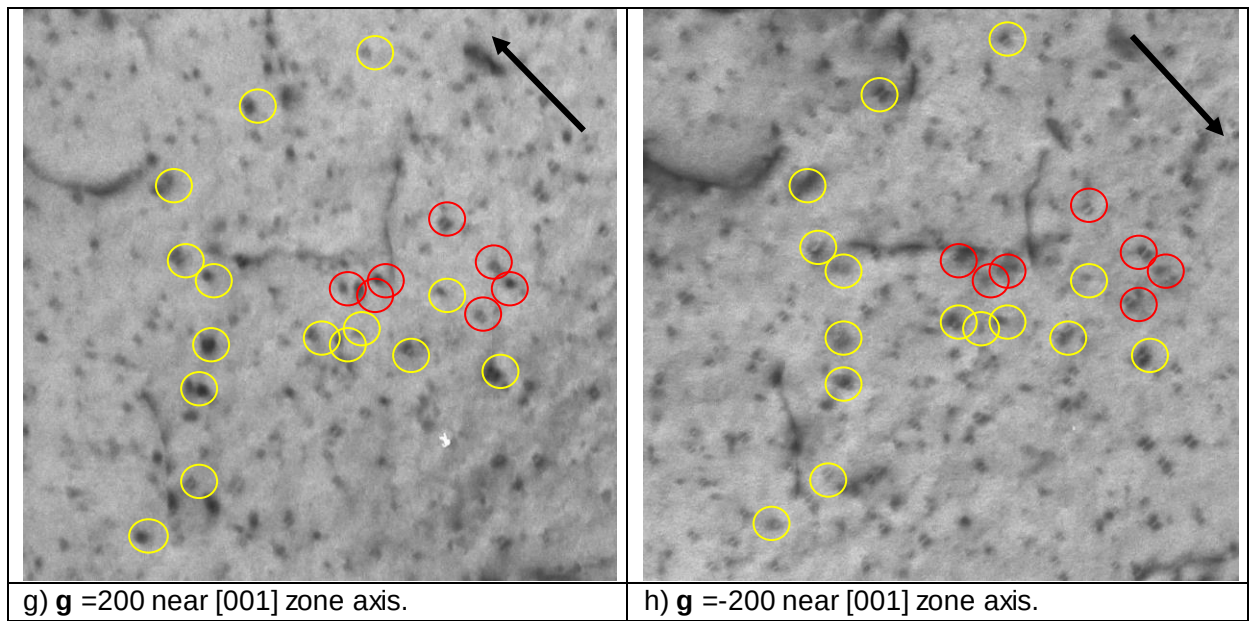


Fig.9.3.3.3. Loop morphology in Fe10Cr at $z = [001]$, irradiated at 300°C to 0.6dpa . $b = \frac{1}{2}\langle 111 \rangle$ (yellow circle) and $b = \langle 100 \rangle$ (red circle).

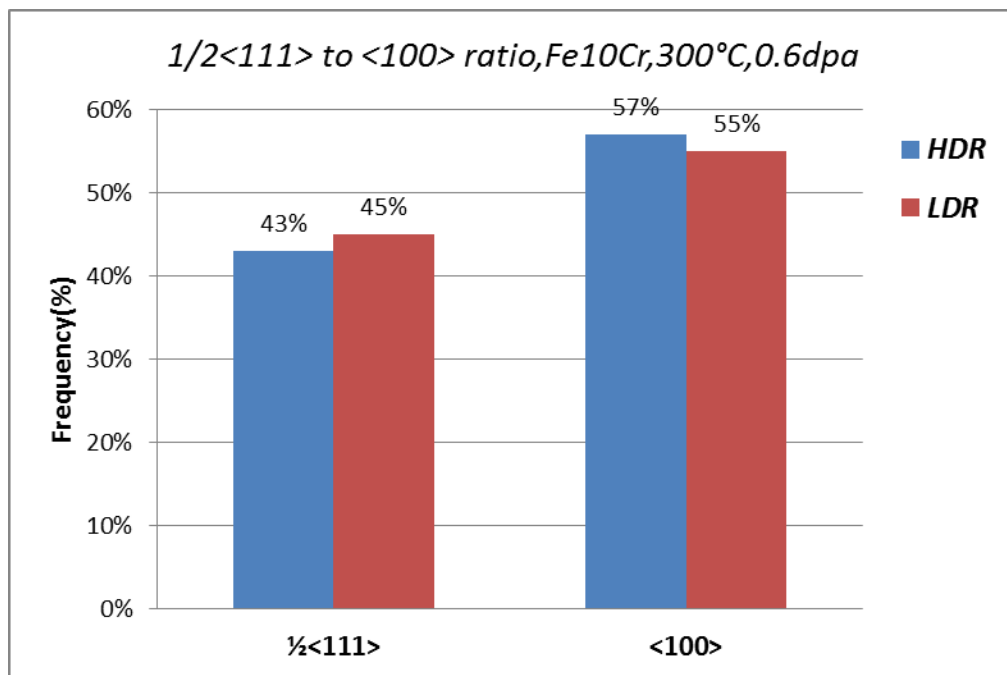


Fig.9.3.3.4. Fraction of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loop types in Fe10Cr. 123 loops counted in the selected area.

UHP-Fe14Cr

Fig.9.3.3.5. shows a series of images taken near the [001] foil normal orientation in a Fe14Cr specimen irradiated at low dose rate at 300°C to a dose of 0.6 dpa.

Both +**g** and –**g** Kinematical bright-field (KBF) micrographs of a same area were taken using the four diffraction vectors available at this pole (**g**= 110, 1-10, 200, 020).

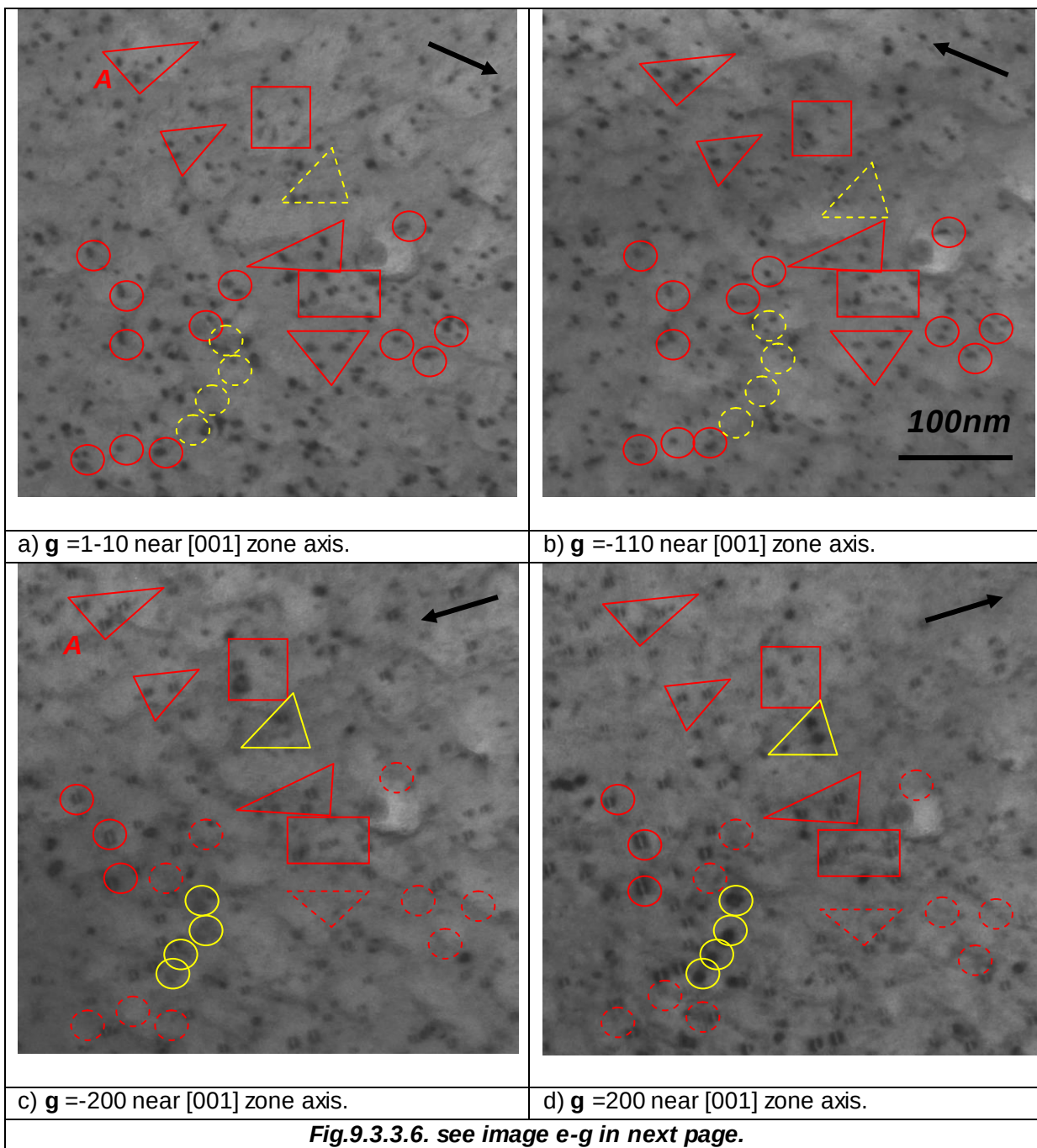
Using the method described above, the Burgers vector of the loops were found.

Loops inside the red triangle **A** show contrast in **g**= 110, 1-10, 200, and disappear in **g**= 020, which indicates that these loops have Burgers vector of **b**= ± [100]. Moreover, these [100] loops show characteristic 'parallel double line' contrast in **g**=± 200 micrographs in **images (c) and (d)**, which further demonstrate their pure edge character (these loops lie on edge on (100) planes).

Of the 100 loops visible in this area, 39 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$ and 43 had Burgers vector of type $\langle 100 \rangle$ whilst 18 loops were too small for their Burgers vectors to be determined. Similar Burgers vector analysis was carried out for Fe14Cr samples irradiated at high dose rate at the same temperature and dose. Of the 126 loops counted, 50 had Burgers vector of type $\frac{1}{2}\langle 111 \rangle$, 53 had Burgers vector of type $\langle 100 \rangle$ and 23 loops were too small for their Burgers vectors to be determined.

The results for $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ ratio on Fe5Cr samples are presented in

Fig.9.3.3.4. It is seen that in Fe14Cr, the proportions of $\frac{1}{2}\langle 111 \rangle$ loops were 43% and 45% for high dose rate samples and low dose rate samples respectively.



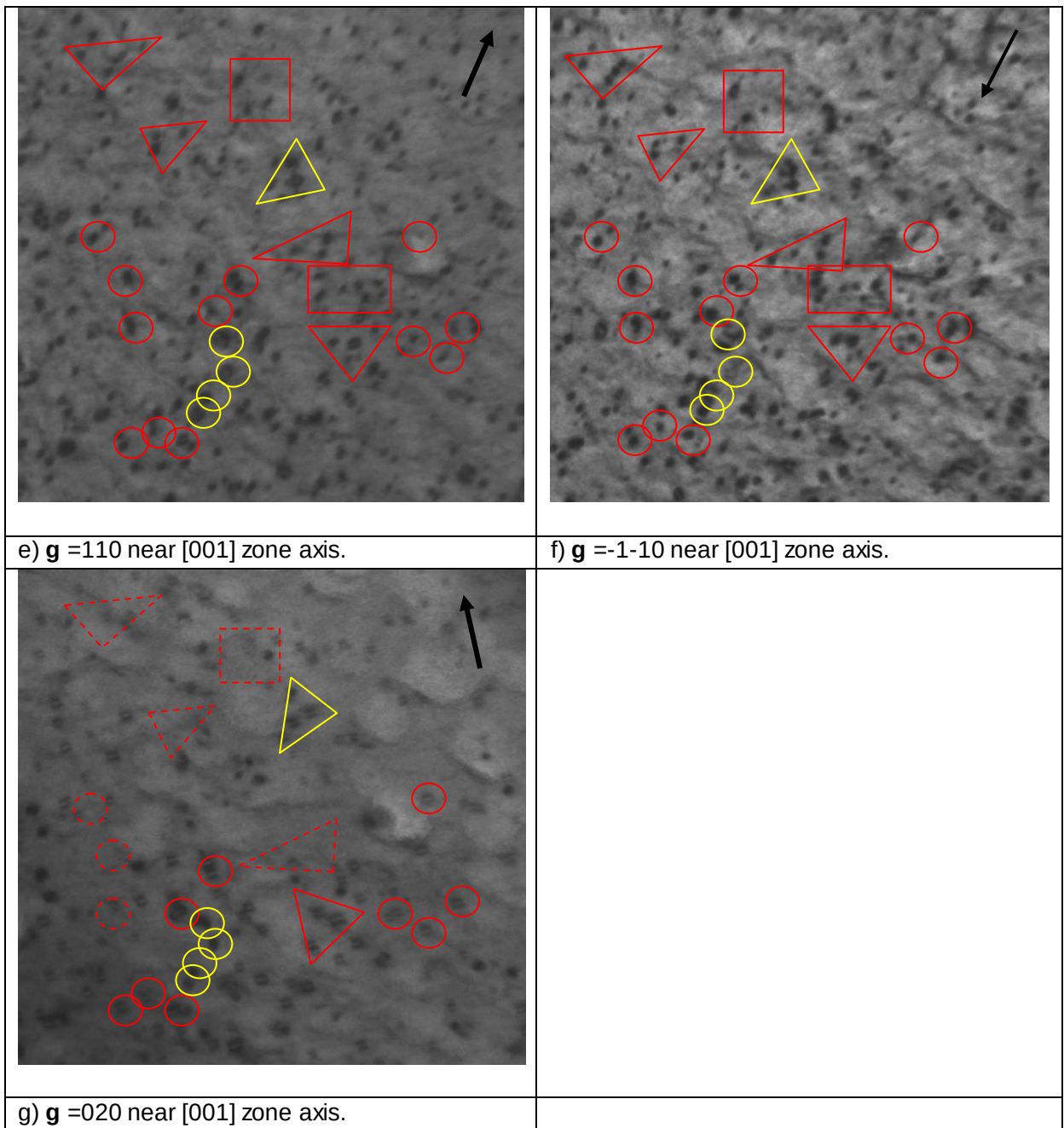


Fig.9.3.3.6. Fraction of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loop types in Fe10Cr. 100 loops counted in the selected area.

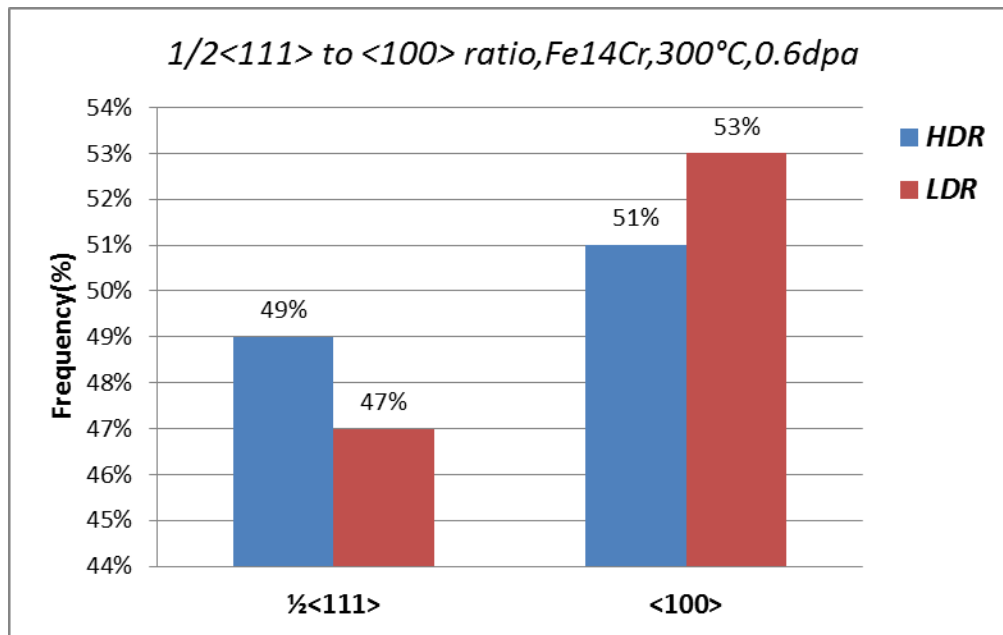


Fig.9.3.3.6. Fraction of $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ loop types in Fe10Cr. 100 loops counted in the selected area.

9.3.4 Loop Nature

Determination of the nature of the dislocation loops is one of the most difficult analyses in diffraction contrast experiments. The inside-outside technique used in this work is potentially the most reliable way to determine the nature of the loops. But it is always difficult to get all the information required to process a reliable nature analysis, in particular the loop habit plane. In this section, two successful attempts were made to determine the nature of the loops produced in the low dose rate irradiation in Fe5Cr and Fe14Cr alloys. Loops formed in Fe10Cr samples were too small to show clear inside-outside contrast.

UHP-Fe5Cr

Due to the small size (<5nm) of the majority of loops presented in the samples, only a few loops were big enough to show clear inside and outside contrast. Three loops were tested, and all of them were interstitial in nature.

Fig.9.3.4.1- Fig.9.3.4.3, shows an example of the inside-outside technique carried out on the LDR Fe5Cr sample. A description of the inside-outside technique can be found in **chapter 3**.

Due to the difficulties in tilting ferromagnetic specimens, it was not always possible to carry out a full analysis of loop Burgers vectors. But in this case, the author succeeded in tilting a specimen between the [001] and [111] poles. Bright-field kinematical micrographs were taken in all the reflections available at these orientations with both $+\mathbf{g}$ and $-\mathbf{g}$. The $|\mathbf{g} \cdot \mathbf{b}|$ table used for this sample is shown in **Table.9.3.4.1**, and confirms that a complete analysis of Burgers vector is possible.

1. Loop Burgers Vector

In this case, the foil normal was near the [001] pole. In **Fig.9.3.4.1**, the ringed loop is visible in images (a), (b) and (c) with $\mathbf{g} = 0-20$, $1-10$, and 200 respectively taken close to the [001] pole, which shows that its Burgers vector is either $\frac{1}{2}[\bar{1}11]$ or $\frac{1}{2}[1\bar{1}1]$. Then the sample was tilted towards the [111] pole, where images in two additional reflections $\mathbf{g} = 01-1$ and -101 were taken. Since it is found that the circled loop was also out of contrast in $\mathbf{g} = 01-1$, its Burgers vector is $\mathbf{b} = \pm \frac{1}{2}[\bar{1}11]$.

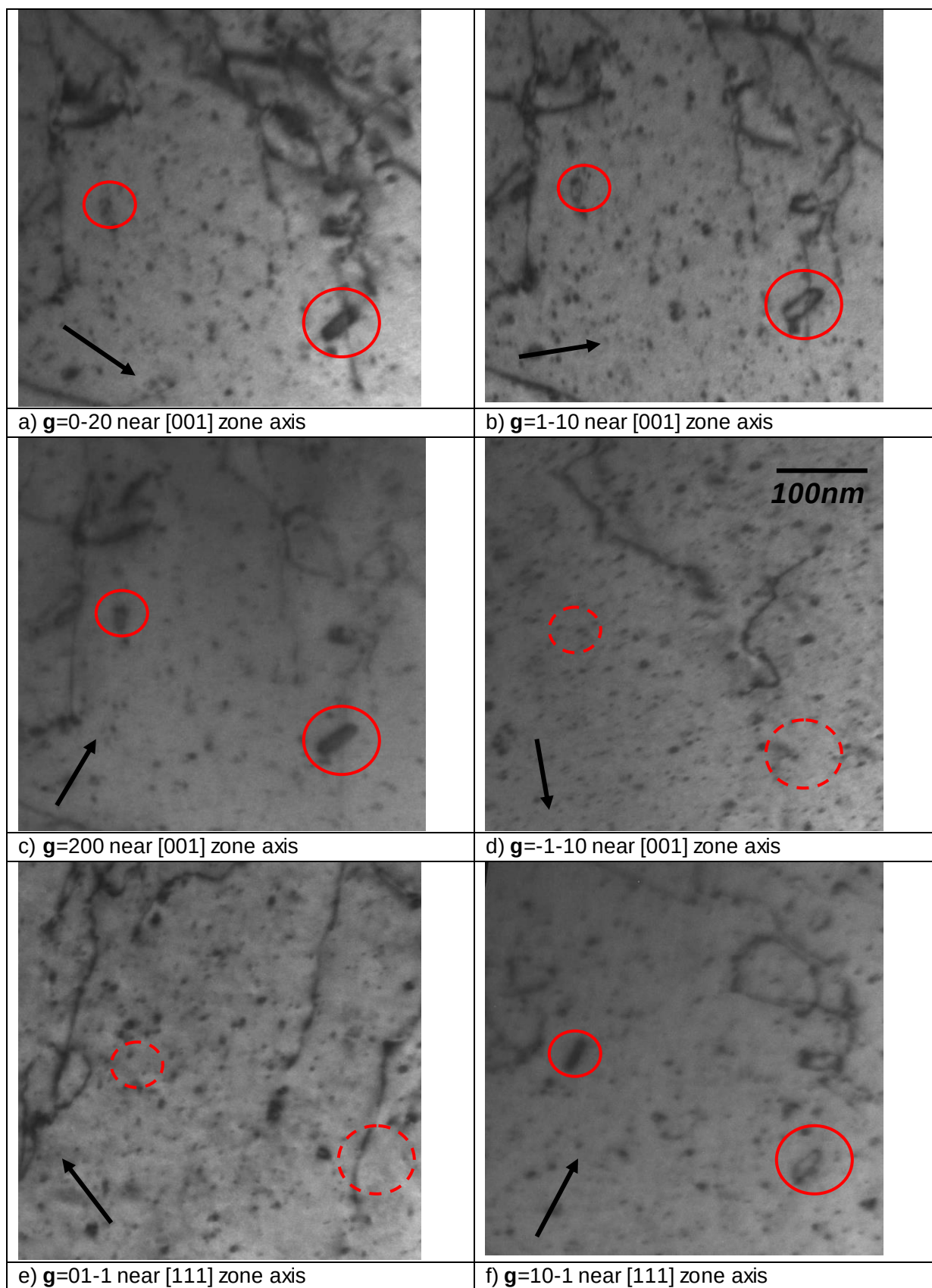


Fig.9.3.4.1. Loop Burgers analysis in Fe5Cr at $z = [001]$, irradiated at 300°C to 0.6dpa.

g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(110)$	1	0	0	1	1	1	0
$\pm(020)$	1	1	1	1	0	2	0
$\pm(200)$	1	1	1	1	2	0	0
$\pm(-110)$	0	1	1	0	1	1	0
$\pm(10-1)$	0	1	0	1	1	0	1
$\pm(01-1)$	0	0	1	1	0	1	1

Table.9.3.4.1. $|g.b|$ table for reflections near $[001]$ and $[111]$ poles.

2. Loop inside/outside contrast

The inside/outside contrast method for the determination of loop nature is most easily applied to pure edge loops. For non-edge loops, this method can most easily be applied to loops in certain ‘safe’ orientations where they will show the same inside/outside contrast behaviour as pure edge loops. As described in **3.4**, if the angle between the beam direction and the Burgers vector is less than $(90^\circ - \emptyset)$, where $\emptyset$ is the maximum angle between the loop nucleation plane and the Burgers vector, the loop is ‘safe’ in that orientation. In bcc crystals, for loops with an $\frac{1}{2}\langle 111 \rangle$ Burgers vector, if the angle between the foil orientation and Burgers vector is less than 55° , this foil orientation is considered ‘safe’ for that loop. Detailed discussion on safe orientations can be found in **3.4**. In this case, the angle between the $[001]$ foil orientation and the $\frac{1}{2}[\bar{1}11]$ Burgers vector is very close to 55° , which complicates the analysis. However, if we look at **Fig.9.3.4.2**, the loop appears edge-on in the micrographs taken near $[001]$ foil orientation, under $g=0-20$ and -110 , which is consistent with a loop with $b=\pm \frac{1}{2}[\bar{1}11]$ lying on its possible nucleation plane (-110) . So the inside-outside technique is applicable in this case.

In **Fig.9.3.4.2**, the electron beam direction is close to $[001]$. The loop shows outside contrast in **(b)**, **(c)**, and shows inside contrast in **(a)**, **(d)**. The condition for the generation of outside contrast is **(g.b)** $s_g > 0$. s_g is positive, since the micrographs were taken under bright-field kinematical conditions. In this case:

$$\mathbf{b} \cdot (020) > 0 \text{ and } \mathbf{b} \cdot (-110) > 0$$

So the Burgers vector of the loop is $+\frac{1}{2}[\bar{1}11]$, therefore, have interstitial nature.

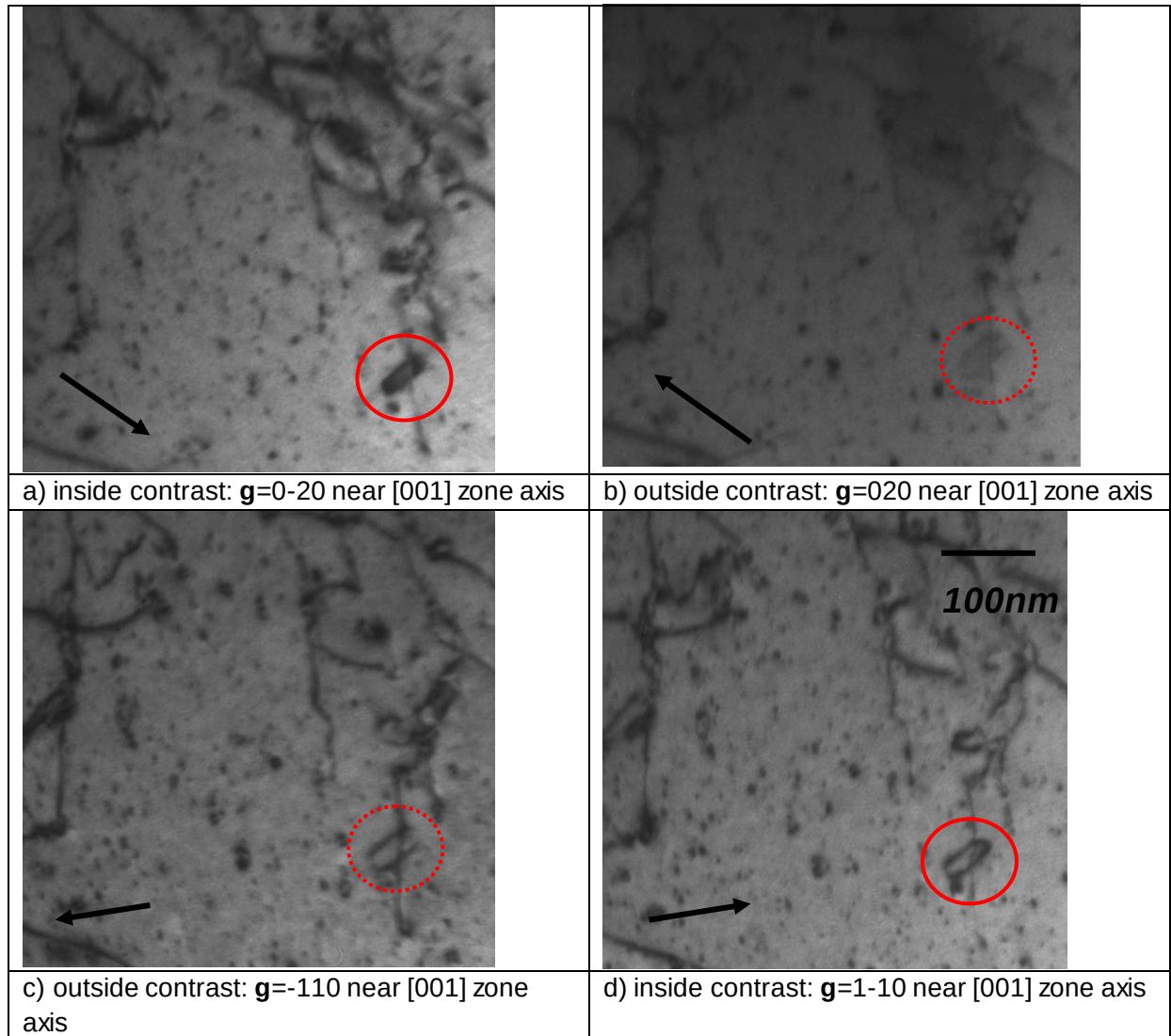


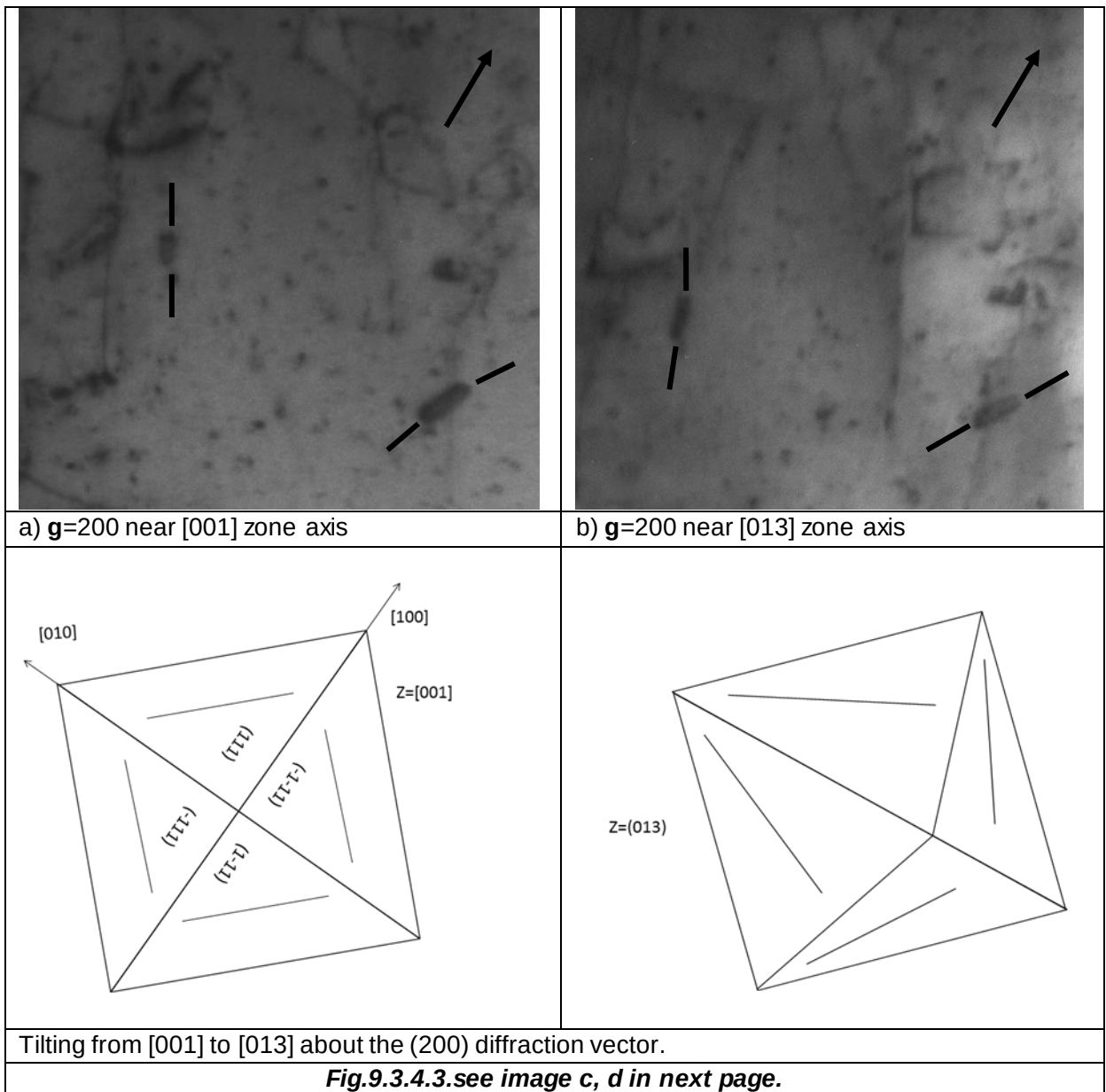
Fig.9.3.4.2. Determination of the nature of loops using the inside/outside contrast method.

3. Loop habit-plane

In the previous inside/outside nature analysis, the only assumption is that the maximum angle between **b** and **n** is 35° (detailed explanation can be found in **3.4**).

By finding the loop habit plane, the loop normal **n** can be determined, thus the assumption can be removed.

In this case, the specimen was tilted along two axes (**g**=200 and **g**=-110) from [001] orientation to [013] and [113] respectively. The beam direction in (a) is close to [001], in (b) it is close to [013]. Tilting is from [001] to [013] about the (200) diffraction vector. The major axis of the loop turns anti-clockwise which is consistent with a loop on a (-111) plane. In **image (c) and (d)**, tilting is about (-110) diffraction vector from [001] to [113]. The major axis rotates anti-clockwise which is again consistent with a circle on a (-111) plane. Therefore the loop is pure-edge, or close to pure-edge, and so the assumption in the inside/outside analysis is correct. The loops have interstitial nature.



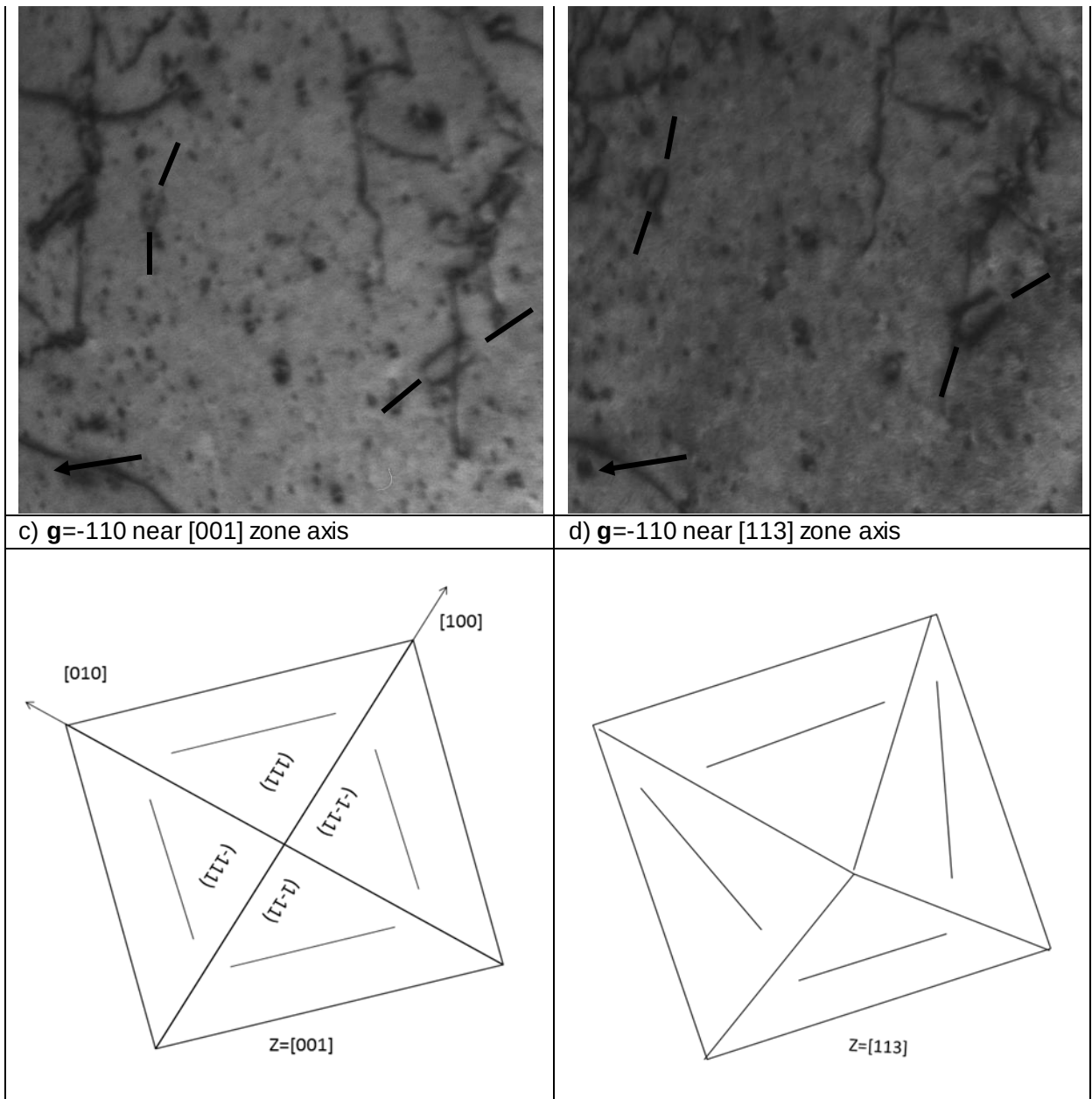


Fig.9.3.4.3. Tilting experiments to determine the loop habit-plane.

UHP-Fe14Cr

Eight loops were analysed, and all of them were interstitial in nature. **Fig.9.3.4.4-**
Fig.9.3.4.5 is an example of the inside-outside technique carried out on the LDR Fe14Cr samples. In this case, bright-field kinematical micrographs for both $+\mathbf{g}$ and $-\mathbf{g}$ were taken around the [001] and [012] foil orientations, using the six reflections available. The $|\mathbf{g}\cdot\mathbf{b}|$ table used for this sample is shown in **Table.9.3.4.1**.

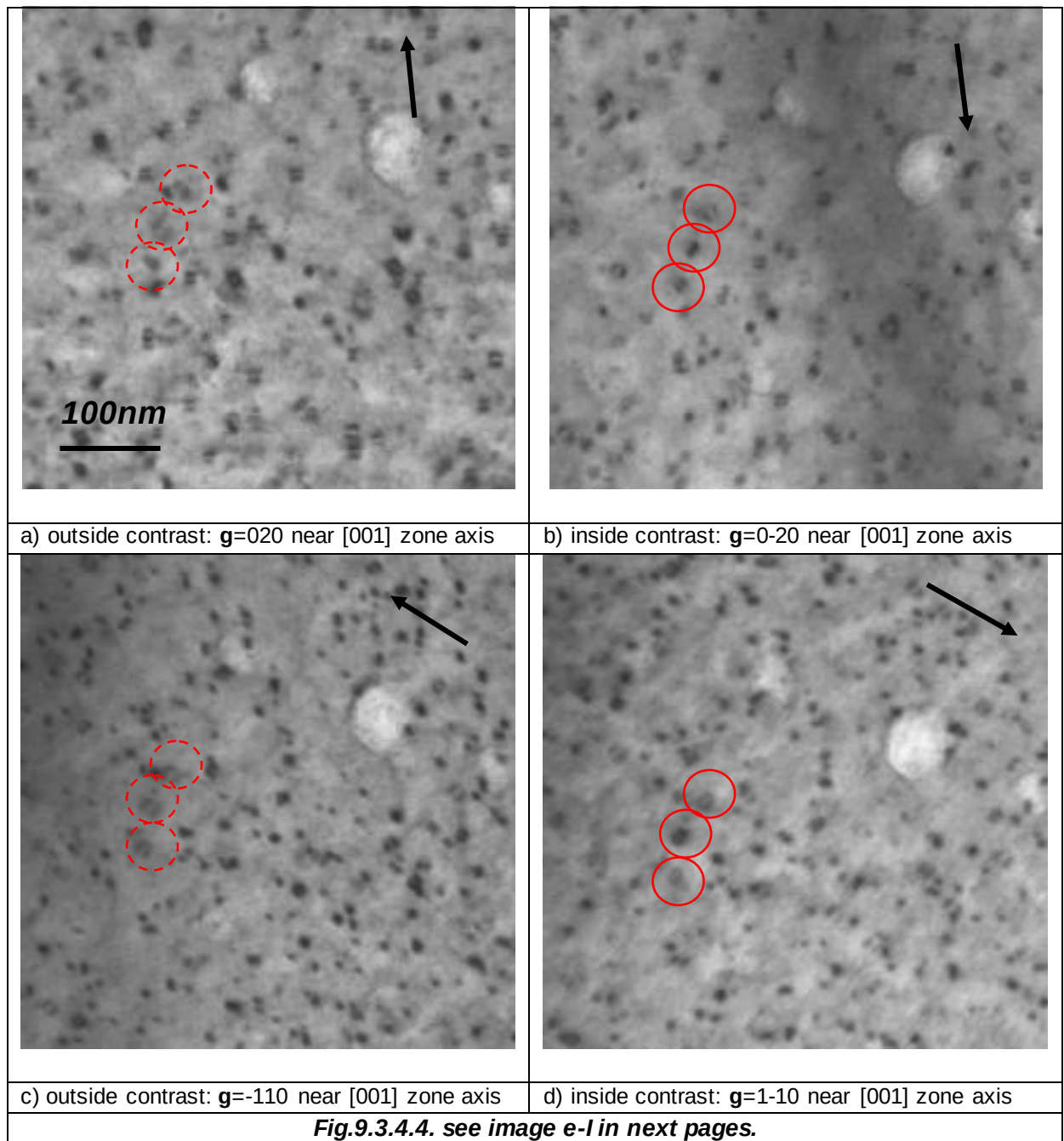
1. Loop Burgers Vector & Inside/Outside Analysis

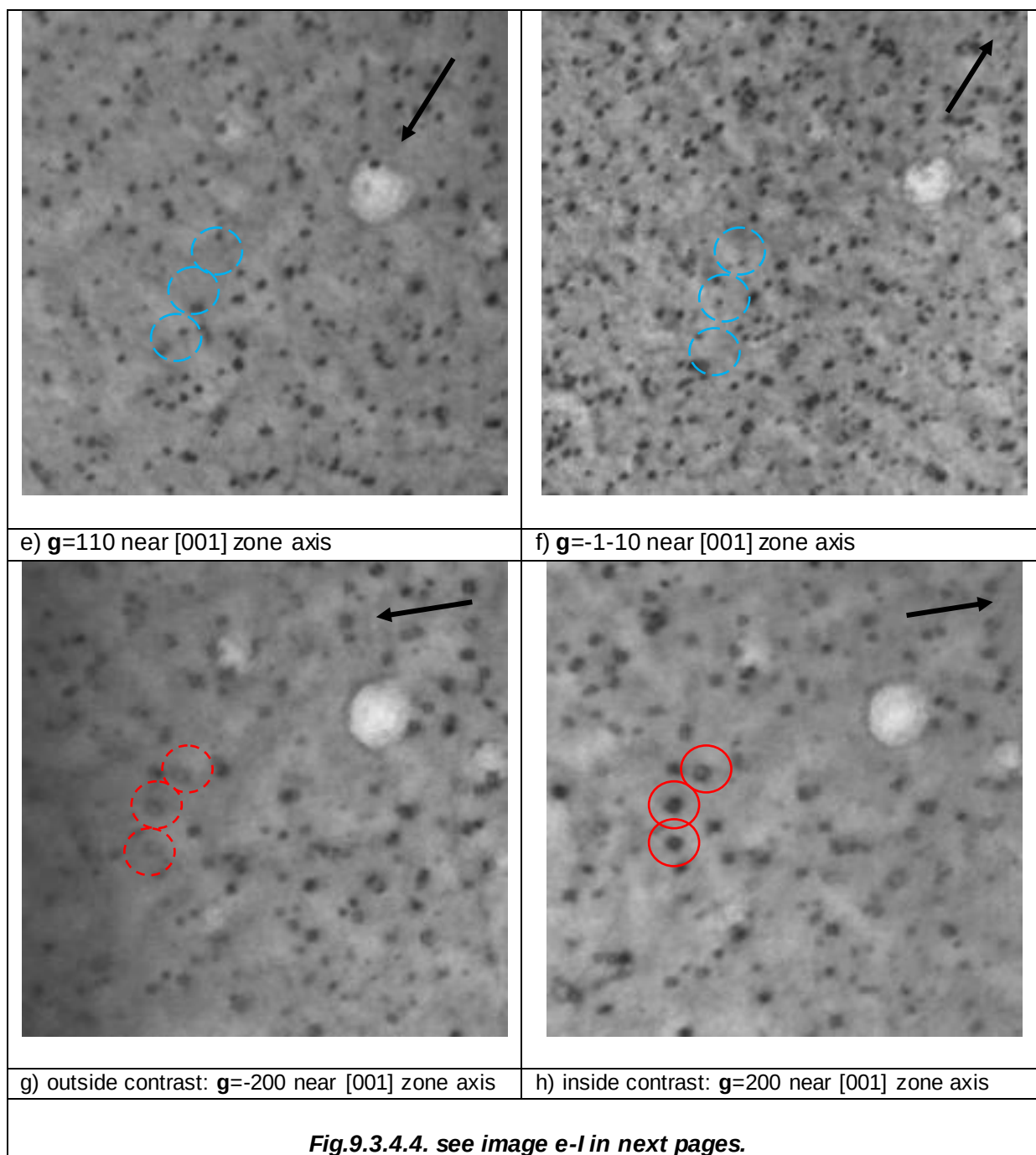
Images **(a)-(h)** were taken near the [001] pole. The ringed loops are visible in images taken in $\pm\mathbf{g}=020, 200, 1-10$, and invisible in images taken in $\pm\mathbf{g}=110$, which indicates that they have Burgers vector either $\pm\frac{1}{2} [\bar{1}11]$ or $\pm\frac{1}{2} [1\bar{1}1]$. Then the sample was tilted towards the [012] pole, where two additional reflections were taken. The loops were visible in $\pm\mathbf{g}=1-21$, while invisible in $\pm\mathbf{g}=12-1$, which shows that the Burgers vector of the circled loops is $\pm\frac{1}{2} [\bar{1}11]$.

The inside/outside contrast analysis is also shown in **Fig.9.3.4.4**. As described in **3.4**, the inside-outside method is strictly suitable to pure edge loops and loops in safe orientations. In this case, the angles between either [001] or [012] foil orientations and the $\frac{1}{2} [\bar{1}11]$ Burgers vector is less than 55° . Thus these orientations are safe. In images **(a)-(h)**, the electron beam direction is close to [001]. The loops show outside contrast in **(a), (c)** and **(g)**, and inside contrast in **(b), (d)** and **(h)**. The condition for the generation of outside contrast is $(\mathbf{g}\cdot\mathbf{b})s_g > 0$. s_g is positive, since the micrographs were taken under bright-field kinematical conditions. So the Burgers vector of the loops is $+\frac{1}{2} [\bar{1}11]$. The loops have interstitial nature. The same can be concluded from images **(i)-(j)** which were taken around [012] pole.

g/b	$a/2[111]$	$a/2[-111]$	$a/2[1-11]$	$a/2[11-1]$	$a[100]$	$a[010]$	$a[001]$
$\pm(110)$	1	0	0	1	1	1	0
$\pm(020)$	1	1	1	1	0	2	0
$\pm(200)$	1	1	1	1	2	0	0
$\pm(-110)$	0	1	1	0	1	1	0
$\pm(12-1)$	1	0	1	2	1	2	1
$\pm(1-21)$	0	1	2	1	1	2	1

Table.9.3.4.2. $|g \cdot b|$ table for reflections near $[001]$ and $[012]$ poles.





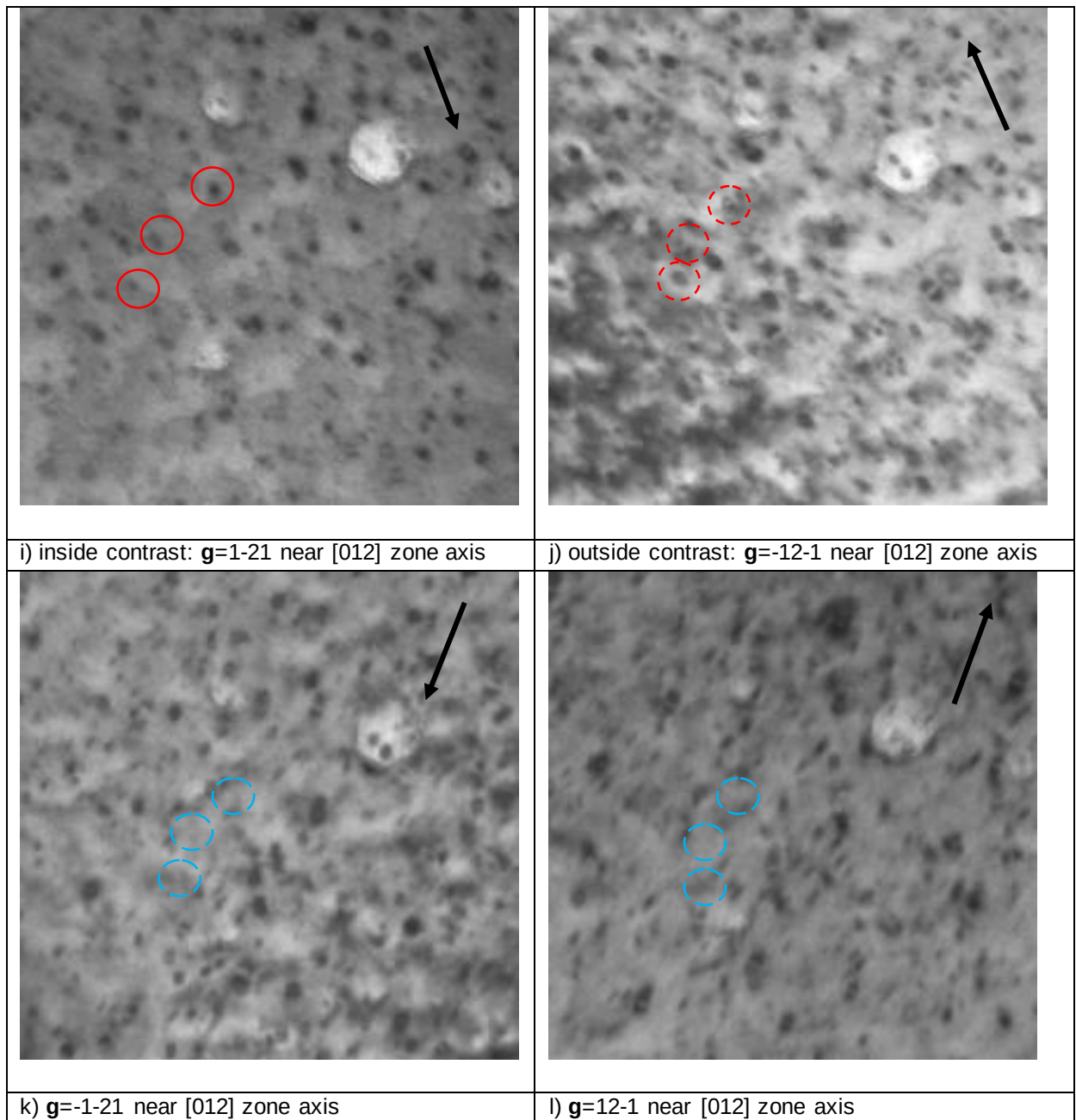
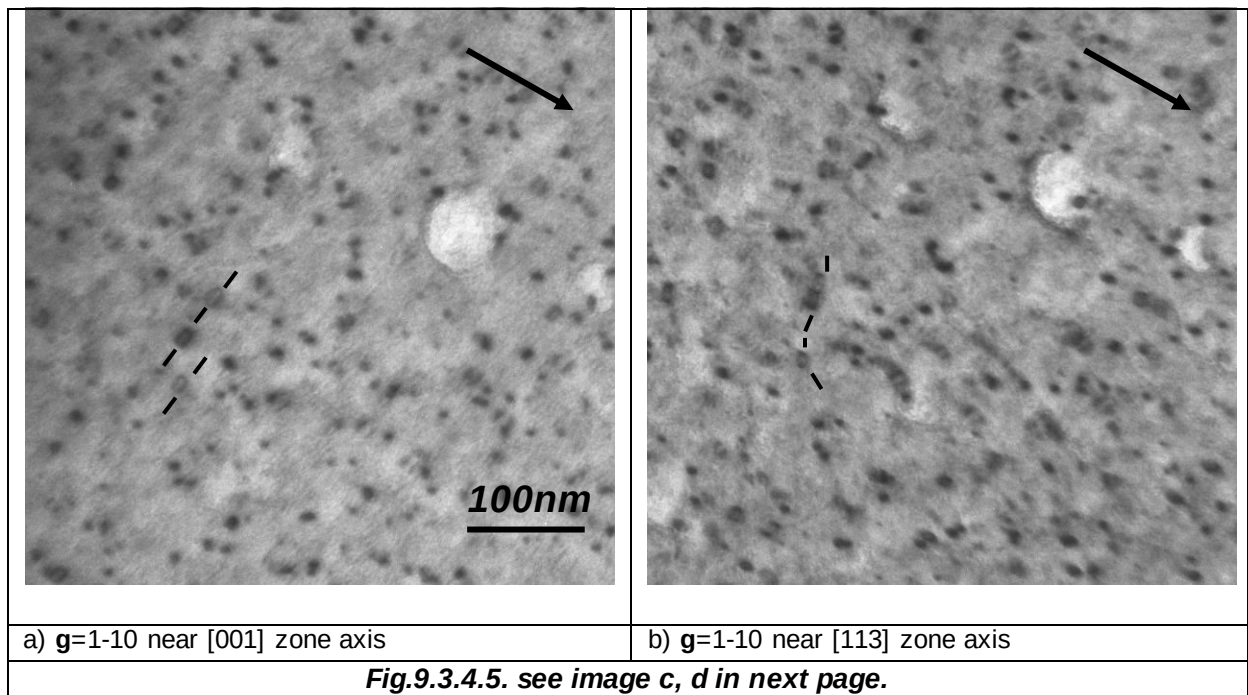


Fig.9.3.4.4. Loop Burgers analysis and inside/outside analysis in Fe14Cr at $z= [001]$, $[012]$ and $[113]$ irradiated at 300°C to 0.6dpa.

2. Loop habit-plane

Loop habit-plane analysis was carried out in order to double check the loop nature which was concluded in the previous inside/outside analysis. In the following images, the specimen was tilted along two axes ($g=0-20$ and $1-10$) from $[001]$ pole to $[113]$ and $[102]$ poles respectively. In **image (a)-(b)**, tilting is about the $(1-10)$ diffraction vector from $[001]$ to $[113]$. The major axis of the loops rotate anti-clockwise which is consistent with a circle on a (-111) plan. In image **(c)-(d)**, tilting is about the (020) diffraction vector from $[001]$ to $[102]$ pole. The major axis of the loops rotate anti-clockwise which is again consistent with a circle on a (-111) plane. Therefore the analysis from the inside/outside analysis is correct, the loops have interstitial nature.



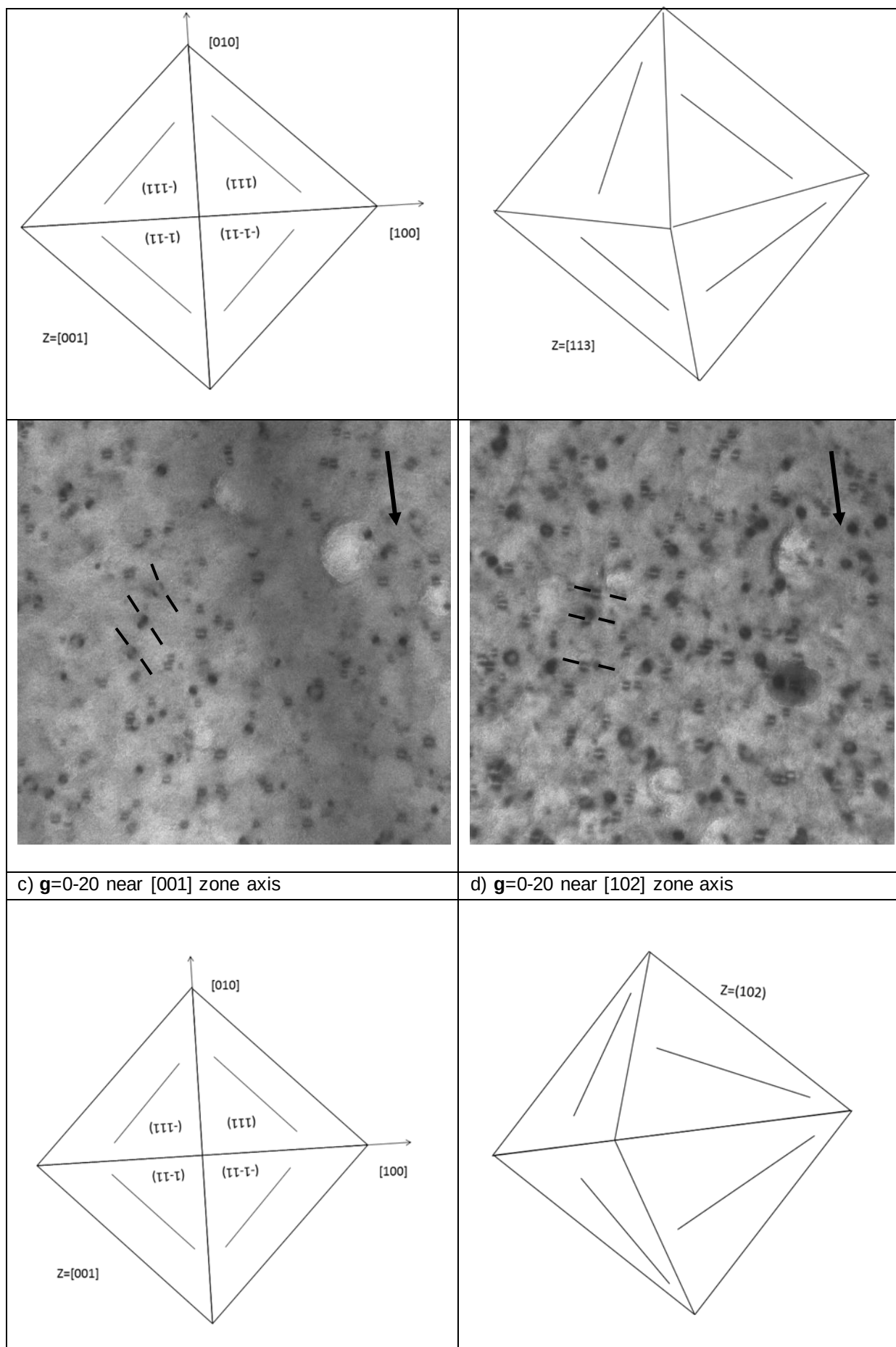


Fig.9.3.4.5. Tilting experiments to determine the loop habit-plane.

9.4 Discussion

In this section the experimental results of heavy-ion induced dose-rate effects are discussed in relation to results from the literature.

9.4.1. Dose-rate Effects

In this work, the implantations were done using a scanning beam with a scanning frame rate of 4kHz, and the beam currents for high dose-rate and low dose-rate implantations were maintained constant during each flux. The dose-rate given in 2.2, is the average dose-rate, which is calculated by the total implantation dose and time, while in reality the instantaneous dose-rate is higher.

In order to interpret the dose-rate effects, it is essential to know the instantaneous dose-rate for both high dose-rate and low dose-rate irradiations. Also it is very useful to simplify the beam scanning process to an equivalent un-scanning circumstance, where we have an ion beam focused on an area with the diameter of 5mm with the instantaneous irradiation dose-rate of G , the beam is switched on-off for 4000 times in a second, with the duration for each implantation pulse T_1 . The beam currents of both high dose-rate and low dose-rate irradiations are maintained constant within each pulse.

The instantaneous dose-rate G , and the implantation pulse duration T_1 can be calculated as follows:

$$T_1 = B / (4000A) = 7.85 \times 10^{-6} \text{ s};$$

$$T_2 = (A - B) / (4000A) = 1/4000 - T_1 = 2.42 \times 10^{-4} \text{ s};$$

$$G_H = D_H / T_1 = 0.019 \text{ dpa/s}; \quad G_L = D_L / T_1 = 0.001 \text{ dpa/s};$$

B is the beam area (5mm diameter);

A is the total area scanned (5cm x 5cm);

T_2 is the gap duration time between two irradiation pluses;

D_H is the displacement of each frame for high dose-rate (1.5×10^{-7} dpa);

D_L is the displacement of each frame for low dose-rate (7.5×10^{-9} dpa).

Since the implantation in our experiments is composed by a series of pulses (each lasting for T_1) with a gap of T_2 in between. We now consider the dose-rate effects at two stages.

Stage 1: The interactions of the clusters produced by a single pulse within T_1 ;

Stage 2: The interactions of the clusters produced by two pulses with a gap of T_2 .

Recent molecular dynamics simulation carried out by Calder et al. [168], suggested that relatively large clusters of self-interstitial atoms can form at the very early stage of cascade development, while large vacancy clusters may subsequently formed by the end of the thermal spike phase. They first defined a zone called 'spaghetti', which is created by a supersonic shock front expanding from the primary recoil event. They suggested that larger interstitial clusters can form on the periphery of the spaghetti, if a hypersonic-recoil creates damage with a supersonic shock ahead of the main supersonic front. When the two supersonic fronts meet, the main one injects atoms into the low density core of the other sub-supersonic shocks, these atoms become interstitial atoms during the rapid recovery of the surrounding crystal. The corresponding number of the vacancies is then formed in the spaghetti core as the crystal cools. **Fig.9.4.1.1.** is the schematic representation of their model of the cascade development over time (the whole process takes about 5-10ps).

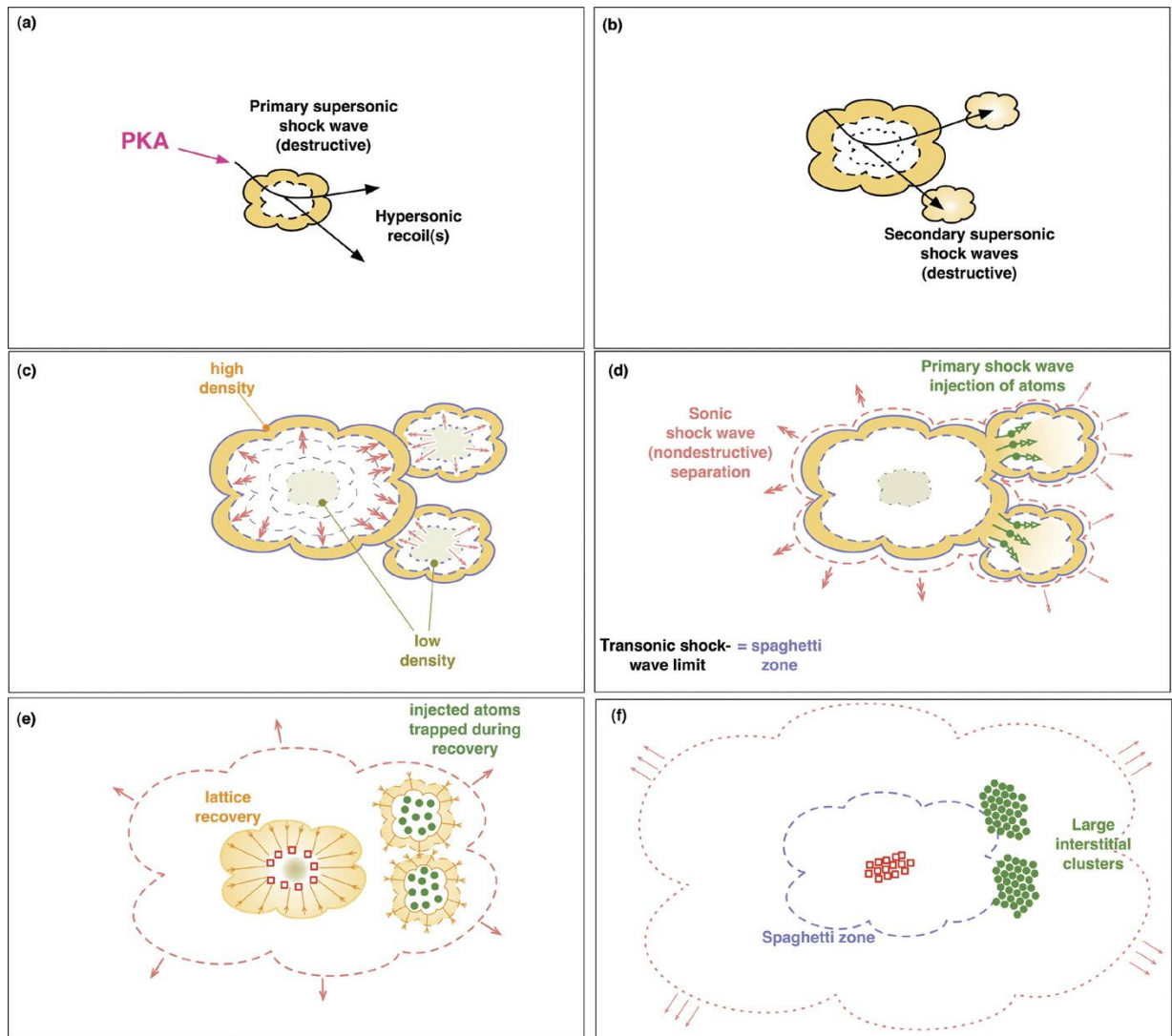


Fig.9.4.1.1. Schematic representation of cascade development over time and interstitial cluster formation [168].

Their explanation of **Fig.9.4.1.1** is quoted below [168]:

Most of the supersonic shock front does not encounter damage formed by a hypersonic event and the atoms displaced by the front outside the spaghetti zone simply return to their lattice sites by elastic relaxation after the time of about 2 ps. However, when an energetic impact event produces one or more hypersonic recoils (two are shown in **image a**) beyond the main front (**see image b**), the main and secondary supersonic shock waves with high atomic density at their fronts and low density disorder behind expand and meet (**image c**). It appears from the evidence of the visualisation techniques that the main front then injects atoms into the core of the secondary region(s) (**image d**). The number of atoms there exceeds the number of

*lattice sites, and the excess atoms become trapped by the rapid recovery of the surrounding crystal (**image e**) and form an interstitial cluster.*

The interstitial atoms formed in this way are matched by vacancies formed during solidification of the hot disordered core of the main zone, which is a relatively slow process. The whole process can take about 10-20 ps.

In our dose-rate experiments, the experimental evidence shows that at the higher dose- rate irradiation both the loop densities and the loop sizes are greater than in the low dose-rate irradiation. Applying Calder et al.'s interstitial cluster formation model to the dose-rate experiments, at **stage 1**, we can estimate that at higher dose-rate irradiations, the density of the cascades in a given volume within very short time period (a few ps), is much greater than that in low dose-rate irradiations. Therefore this leads to a higher density of collision events between hypersonic shock waves and sub-supersonic shock waves. Thus in the high dose-rate irradiation, the density of interstitial clusters is expected to be higher.

At **stage 2**, we then consider the interactions of the supersonic and hypersonic waves created by two pulses. The duration between two pulses is 2.42×10^{-4} s is long compared with the collision event (about 10-20ps), therefore long before the starting of the following pulse, the collision events in the previous pulse have already cooled down. So there will not be interaction between the supersonic and hypersonic waves created by two pulses.

However interactions between the interstitial clusters created by different pulses may occur. As described at **stage 1**, in the high dose-rate irradiation, there will be more clusters produced during each pulse. Therefore the distance between nearby clusters will be closer compared with that in low dose-rate irradiation. And previous clusters will also act as sinks for the new clusters and self-interstitials. Therefore it is easier for the interstitial clusters to grow into visible size at high dose-rate irradiation.

Moreover, in high dose-rate irradiations, since the concentrations of mobile interstitial clusters and defects within the sample are greater, the mobile $\frac{1}{2}\langle 111 \rangle$ interstitial loops have a higher chance (compared with sessile $\langle 100 \rangle$ loops) to absorb mobile interstitials by hopping along their glide cylinders. The consequence is that the loop size increase during high dose-rate implantation will be more evident for the materials containing more $\frac{1}{2}\langle 111 \rangle$ loops. The relatively lower loop size increase in the Fe-5Cr alloy compared with higher-Cr alloys can be attributed to the lower proportion of $\frac{1}{2}\langle 111 \rangle$ loops. In Fe-5Cr, 30% of the loops were $\frac{1}{2}\langle 111 \rangle$ type, whereas in Fe-10Cr/Fe-14Cr, about 50% of the loops were $\frac{1}{2}\langle 111 \rangle$ type.

In the previous arguments, we only talked about the formation and interactions between interstitial clusters, while vacancy clusters were believed to be formed within the core of the spaghetti zone according to Calder et al.'s model. Therefore although the experimental observations and the model are quite consistent, the assumption is that the loops observed in the micrograph are interstitial in nature, or at least a great proportion of them are interstitial loops. In this work, inside-outside technique was used to test the nature of the loops. Of the loops we tested (about 5-10 in each sample), all were interstitial in nature. However this method can only be apply to larger loops which show clear inside-outside contrast, so we can only conclude that at least most loops larger than 5nm are interstitial in nature.

We now consider the nature of the large number of small loops ($<5\text{nm}$) and the possible existence of vacancy clusters. The literature survey (**see 1.3, 1.4**) showed although vacancy loops were seen during neutron irradiation in Fe based alloys, in self-ion irradiated iron based alloys only interstitial loops were found. And in the dynamic observation of self-ion irradiated iron based alloys, Yao et al. found that cascades only collapsed at overlap doses, when at least some of the very small loops they observed were of vacancy nature. They also observed that in one case in

their Fe-8Cr alloy, vacancies in the form of micro-voids were present. In their dynamic observations, large loops proved to be interstitial in nature; also small mobile loops were seen to be absorbed by large interstitial loops, and resulted in the growth of the larger loop. This is evidence that some of the small loops in self-ion irradiated iron based alloys are interstitial. Therefore it is reasonable to believe that most of the loops seen in the micrographs are interstitial in nature, and so our argument on dose-rate effects is valid.

The description given in this section gives an assumption to explain the observed phenomenon on loop density and size changes, which attributes to the dose-rate effects. However, there might be other factor that could have the same effects on the observed phenomenon, like the temperature effects.

During the experiments, the temperature was measured by the thermocouple buried inside the sample holder. This design can accurately measure the temperature of the bulk sample, while during irradiation the localized temperature along the path of the injected ions and its knock-ons can be higher than the matrix, but this temperature alert within the sample cannot be measured by the present method. During irradiation, the 2 MeV ions produces dense defects only in the end of their path when energy falls to a few hundred keV, the rest of the energy lost to the inelastic collision with electrons, therefore will increase the localized temperature. Although the heat will quickly transfer to the matrix within the cooling phase period of cluster formation, this short period of localized temperature increase might still increase the migration and formation of interstitial clusters, especially in high dose rate irradiations, where the density and frequency of injected ions within each pulse are about 100 times higher, compared with that in low dose rate experiments. Further insight into this assumption requires modelling work on the localized temperature increase during irradiation.

9.4.2. Cr Effects

We compare here the results of this chapter with those reported in **chapters 4-8**.

- 1) First, a higher proportion of $\langle 100 \rangle$ loops was found in the work reported in this chapter compared with the equivalent results of **chapters 4-8** (300°C, 7dpa), where we found that only ~30% of the loops were $\langle 100 \rangle$ type in all alloys. One possible explanation may be attributed to the pinning effects of the impurity atoms contained in the less-pure materials used for the experiments described in **chapter 4-8**. These materials were made from an alloy power with a starting purity of 99.99%, but lower purity after casting. The materials for dose-rate effects were UHP alloys with composition listed in **Table.2.1**. Since all the results were taken from foils with $\{001\}$ orientation, a larger proportion of $\frac{1}{2}\langle 111 \rangle$ loop may glide to the foil surface in these purer alloys, while $\langle 100 \rangle$ loops can be well preserved within the foil because their Burgers vectors have no component towards the foil surface.
- 2) Second, in this chapter the fraction of $\frac{1}{2}\langle 111 \rangle$ loops was found to increase with increasing Cr content, from 30% in Fe5Cr to nearly 50% in Fe10Cr and Fe14Cr. This trend is consistent with that found in **chapters 4-8**.
- 3) Third, in the 300°C (0.6 dpa) implantation described in this chapter, the loop number densities all fall in the range from $8 \times 10^{21}/\text{m}^3$ to $10 \times 10^{21}/\text{m}^3$, and little effect of Cr was found. This is consistent with the limited data for 300°C irradiations reported in **chapters 4-8**, where little effect of Cr was apparent on either loop number densities or loop average sizes. A clear increase in loop densities as a function of Cr content was however observed in the 400°C, 2.5 dpa implantations. One possible explanation is due to the increased loop mobility at higher temperatures in the lower-Cr alloys. In such alloys loops can glide out of the foil or merge with other loops. Due to the pinning effects of Cr atoms, in high

Cr alloys more loops are preserved within the foil, and the coalescence of loops is reduced.

- 4) Fourth, average loop sizes in Fe-14Cr alloys were significantly larger than in Fe-10Cr and Fe-5Cr alloys. This result was unexpected. In **chapter 4-8**, in the 300°C implantation to a dose of 7 dpa, the average loop sizes in different Fe-Cr alloys were roughly the same. One explanation can be attributed to the interstitial cluster diffusivity. It has been shown that with increasing Cr concentration, the cluster diffusivity first decreases and then increases. The minimum diffusivity is found at about 10 at% Cr [169]. If interstitial clusters have higher diffusivity in Fe-14Cr, it is reasonable to expect larger sizes of interstitial loops.

9.5 Conclusions

1. The radiation damage microstructure observed in this work are large quantity of dislocation loops with both $\frac{1}{2}\langle 111 \rangle$ and $\langle 100 \rangle$ Burgers vectors.
2. Nature determination using inside-outside methods indicated that all the loops tested were interstitial loops. No voids were seen.
3. Dose rate effects were observed in self-ion irradiated Fe-Cr alloys, that in high dose rate experiment both the loop densities and sizes were greater. Same trend were observed in all the irradiated materials (Fe-5Cr, Fe-8Cr, Fe-14Cr). A possible explanation using the recent molecular simulation on the formation of interstitial clusters was given.
4. Conclusions on Cr effects are fully described in **9.4.2**.

Chapter 10: Future Work

In this work a relatively comprehensive research has been done on the self-ion irradiated Fe and Fe-Cr alloys with a wide range of Cr content at various temperatures and doses. Conclusions on Cr effects, temperature effects, dose effects have been drawn. Complex microstructures as loop strings, finger shape loops and perpendicular $\langle 100 \rangle$ loop clusters were observed the first time in bulk Fe irradiated at 300°C and 400°C. Transition of loop Burgers vectors from $\frac{1}{2}\langle 111 \rangle$ to $\langle 100 \rangle$ type as elevated irradiation temperatures was observed as well, which is consistent with that in thin-foil experiments.

Firstly, as an extension of this work, it is essential to carry out irradiations at higher temperatures ($>500^\circ\text{C}$). Due to the limitation of the implantation facility, the highest temperatures achieved in this work was 460°C, thin-foil irradiation indicated that the damage evolution at 500°C may be different as both vacancy and interstitial defects become highly mobile at high temperature.

Secondly, in order to simulate the real fusion reactor environment with the production of transmutation gases by energetic neutrons, the next step of the research could be dual beam implantation. The JANNUS facility in France should have the capability of dual-beam or even triple-beam irradiations, as it is designed for irradiation damage studies.

Also dose rate effects on the evolution of interstitial clusters in Fe-Cr alloys have been studied at 300°C as well. The observation showed that both the loop densities and sizes were increased at high dose rate irradiation.

Thirdly, as no vacancy loops were observed in self-ion irradiated Fe and Fe-Cr alloys. It is interesting to find where the vacancies are going during ion irradiation. One possibility is that in self-ion irradiated Fe and Fe-Cr alloys, vacancies exist in the form

of micro-voids as which has been seen in Fe-8Cr thin-foil experiments. Therefore, higher voltage TEM (>200kV) could be used to look for the micro-voids.

Fourthly, the research can be extended to more complex model alloys or realistic materials such as Ferritic/Martensitic steels. Therefore, analytical microscopy techniques such as EELS, along with the diffraction contrast technique can be very useful in characterising realistic steels.

For dose rate experiments, it is necessary to see how dose rate affect the microstructure evolution in pure Fe, as only Fe-Cr alloys were analysed in this work. Moreover, the dose rate experiments could also extend to higher irradiation temperatures, and it will also be very interesting to observe the dose rate effects using in-situ TEM.

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