

1 Regional remagnetization of Irish Carboniferous carbonates
2 dates Variscan orogenesis, not Zn-Pb mineralization

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11 **ABSTRACT**

12 Paleomagnetic methods have been used in economic geology to date
13 mineralization in sediment-hosted ore deposits and thereby help to develop ore deposit
14 models and understand the geodynamic settings in which mineralization can occur.
15 However, paleomagnetic ages are sometimes inconsistent with other geochronological
16 techniques and with geological observations. Here we test the veracity of paleomagnetic
17 ages for sediment-hosted ores through a study of the Irish Midlands ore field. We find
18 that unaltered rocks distal to mineralization that are of equivalent age to the ore host
19 sequence have comparable characteristic remanent magnetic directions to those
20 previously derived from the ores. This indicates that remagnetization of the rocks was
21 probably independent of the ore-forming process. Comparison with the apparent polar
22 wander path for Europe suggests an age of ca. 310 Ma for this event, consistent with the

23 timing of the Variscan orogeny. Fold test results support this, indicating the signal was
24 acquired after tilting and/or folding of the host rocks. Petrology and magnetic data
25 suggest that nanometric magnetite particles are the remanence carrier. Based on
26 independent geochronological and geological constraints, we conclude that
27 mineralization formed in Ireland in the early Carboniferous coincident with basin
28 development and that paleomagnetic dates were reset during the later orogenic overprint.
29 Caution is therefore warranted in the interpretation of paleomagnetic dates for ore
30 systems, and geodynamic models for mineral systems based on these may be erroneous.

31 **INTRODUCTION**

32 Sedimentary rock-hosted hydrothermal ore deposits are a major source of lead,
33 zinc, and copper. Their origins are believed to be diverse: formation synchronously with
34 their host rocks from fluids that may have vented at the seafloor (sedimentary exhalative,
35 Sedex, deposits); formation during burial and diagenesis; or synorogenic (Wilkinson,
36 2014). In many cases, due to the lack of minerals amenable to dating by radiogenic
37 isotopic methods, ore genesis and geodynamic settings of mineralization remain
38 controversial. This is because the timing of mineralization with respect to host-rock
39 deposition can only be inferred from often ambiguous textural relationships between ore
40 minerals and sedimentary structures. This uncertainty affects three important issues: (1)
41 mineral exploration models are based on an assumed age relationship between ore and
42 host rock; (2) viable sulfide precipitation mechanisms are constrained by the
43 physicochemical environment of ore formation; and (3) the geodynamic setting of well-
44 mineralized basins and the fluid-flow systems that formed them are interpreted based on
45 the assumed age of mineralization. For example, Leach et al. (2001) argued for a link

46 between carbonate-hosted Zn-Pb Mississippi Valley–type (MVT) deposits and
47 supercontinent assembly cycles involving topographically driven fluid flow ahead of the
48 advancing orogen. However, the temporal correlation between deposit genesis and major
49 collisional orogens is largely dependent on paleomagnetic ages for ore formation. If these
50 ages reflect an orogenic overprint, then this association would be expected and the
51 inferred models would be invalidated.

52 The paleomagnetic dating method involves the isolation of a characteristic
53 remanent magnetic (ChRM) orientation acquired during mineralization and comparison
54 of this with the apparent polar wander (APW) path for the terrane in question within the
55 relevant period of Earth history. Two basic assumptions apply: (1) that the measured
56 magnetic orientation was acquired during the ore-forming event; and (2) that the
57 orientation of the Earth’s magnetic field (APW path) is sufficiently well known for the
58 time period in question.

59 Here we specifically test the first assumption based on a study of the Irish
60 Midlands Zn-Pb ore field. The region provides an excellent opportunity to address the
61 veracity of paleomagnetic dating because the system has been extensively studied, yet
62 controversy remains regarding the timing of mineralization. Geological constraints and
63 recent Re-Os dating suggest that the ores are early Carboniferous (Hnatyshin et al.,
64 2015), whereas published paleomagnetic dates range from early Carboniferous to early
65 Permian (Symons et al., 2007; Pannalal et al., 2008a, 2008b); the latter have been used to
66 argue for a synorogenic or postorogenic origin for the Irish deposits.

67 **GEOLOGICAL SETTING AND SAMPLING**

68 The ore-hosting carbonate rocks are located in the central Ireland Midlands Basin,
69 which began to develop in the Mississippian during a period of dextral transtensional
70 strain across the Laurussian continental margin. This was related to oblique convergence
71 between Laurussia and Gondwana that affected much of northern Europe (e.g.,
72 McKerrow et al., 2000). The progressive Variscan collision ultimately led to the
73 amalgamation of Pangea in the late Carboniferous–early Permian (ca. 300 Ma; Stampfli
74 et al., 2013).

75 A diachronous northward marine transgression across the margin established a
76 shallow ramp environment in which a shale-limestone sequence was deposited. At some
77 stage, hydrothermal solutions flooded through the sequence and precipitated tens of
78 millions of tons of zinc and lead in the form of sulfides (Andrew, 1993; Hitzman and
79 Beaty, 1996; Wilkinson and Hitzman, 2015). The Navan deposit represents the largest
80 accumulation of ore minerals, but economically exploitable ores also formed at Tynagh,
81 Silvermines, Galmoy, and Lisheen (Fig. 1). At least 20 more subeconomic prospects have
82 been identified, making the district one of the most intensely Zn mineralized terrains on
83 Earth.

84 The most contentious issue with respect to deposit genesis is the timing (and
85 burial depth) of mineralization. Based on geological and isotopic arguments it is thought
86 that the deposits either (1) formed during deposition and early diagenesis, within ~15
87 m.y. of host rock deposition and within a few hundred meters of the paleoseafloor (e.g.,
88 Wilkinson and Hitzman, 2015; Wilkinson et al., 2005); or (2) formed during deeper
89 burial, after lithification was complete (e.g., Peace and Wallace, 2000). Paleomagnetic
90 studies have yielded remagnetizations of mostly late Carboniferous or Permian age,

91 interpreted in terms of very late epigenetic mineralization (Symons et al., 2007; Pannalal
92 et al., 2008a, 2008b).

93 In order to assess if there is a link between remagnetization and mineralization we
94 carried out a regional paleomagnetic study of host rock–equivalent age samples from 14
95 sites in the Irish Midlands over a total area of ~25,000 km², all distal to mineralization
96 (Fig. 1). Samples were mostly taken from the Waulsortian Limestone Formation; at least
97 five individually oriented samples were collected at each site.

98 **LABORATORY METHODS**

99 To determine their ChRM, samples were demagnetized using both thermal and
100 alternating-field (AF) techniques at the University of Oxford (UK). Between five and
101 nine samples from each site were subjected to AF demagnetization using a 2G SQUID
102 (superconducting quantum interference device) magnetometer with in-line AF
103 demagnetization coils. The sample natural remanent magnetizations (NRMs) were
104 demagnetized in steps of 5 mT through to 100 mT.

105 To help identify the magnetic mineral carriers and their grain size, standard
106 hysteresis measurements were made using the Princeton Measurements alternating
107 gradient magnetometer at Imperial College London (UK). Thermomagnetic curves were
108 measured in helium (He) using the Princeton Measurements vibrating sample
109 magnetometer, also at Imperial College London. Scanning electron microscopy (SEM)
110 analysis was carried out using a Cameca SX-500 microprobe located at the Natural
111 History Museum, London, in order to determine sample mineralogy and identify potential
112 magnetic carriers.

113 **RESULTS**

114 The majority of the specimens behaved similarly during AF demagnetization (Fig.
115 2). Approximately 25% appeared to contain well-defined viscous remanent
116 magnetizations acquired in Earth's current magnetic field; these were normally removed
117 by AFs of 5–10 mT. Subsequent demagnetization steps revealed the presence of a ChRM,
118 generally directed shallowly downward or upward to the south (Fig. 2). Not all samples
119 were fully AF demagnetized at the peak applied AF of 100 mT; however, directions
120 leading to the origin were clearly defined. The ChRM was successfully isolated in nine of
121 the sites. The other five sites yielded no consistent directions (sites BQ, FQ, and GB;
122 Table 1), were too weak to measure (ES), or contained strongly overlapping components
123 such that the ChRM could not be confidently isolated from overprints (RT). Site mean
124 directions in both in situ and tilt-corrected coordinates (Figs. 2D and 2E) show that tilt
125 correction causes an increase in the dispersion of the site mean directions, suggesting that
126 the characteristic magnetization was acquired after tilting and/or folding of the rocks
127 (Fig. 2F), although the increase in dispersion is not statistically significant (McFadden
128 and Jones, 1981).

129 Thermomagnetic analysis found evidence for iron oxides (magnetite) and sulfides
130 (principally pyrite that transformed irreversibly into magnetite and pyrrhotite on heating).
131 These findings are supported by SEM, which identified fine-grained iron and iron-
132 manganese oxides and pyrite (see the GSA Data
133 Repository¹). Iron oxide grains are typically 5–50 μm in diameter and mostly associated
134 with texturally late dolomite. Pyrite grains are mostly ~5–10 μm in size, occasionally
135 occurring as framboidal clusters but more often as disseminated grains in late
intergranular porosity suggestive of a diagenetic origin. With the exception of ES, the

136 samples that did not yield coherent directions contained relatively abundant pyrite and no
137 Fe oxide or dolomite. This, combined with a lack of pyrrhotite Curie temperatures on
138 thermomagnetic warming, suggests that magnetite, developed during dolomitization, is
139 the principal carrier of the ChRM.

140 Room-temperature magnetic hysteresis loops were generally wasp-waisted, which
141 is indicative of either a two-phase magnetic assemblage or the presence of thermally
142 activated, single-domain behavior, i.e., superparamagnetism (SP). Assuming magnetite as
143 a dominant carrier, the critical SP threshold size is ~30 nm (Muxworthy and Williams,
144 2008), indicating that nanometric particles, below SEM resolution, are the principal
145 source of the ChRM.

146 **DISCUSSION AND CONCLUSIONS**

147 The mean pole position (332°E; 37°S) derived in our study of unmineralized
148 rocks (Fig. 3) plots close to the 310 ± 15 Ma mean pole of the APW path for Europe of
149 Torsvik et al. (2012). This is consistent with the post-tilting and/or post-folding
character of the remanence and indicates that all the Mississippian rocks (ca. 345
152 Ma) where we could isolate the ChRM have been remagnetized. There is a clear temporal
153 association between the remagnetization and Variscan events, as documented by
154 $^{40}\text{Ar}/^{39}\text{Ar}$ ages of 314 Ma, 300 Ma (synorogenic), and 297 Ma (postorogenic) intrusions
155 in southwestern Ireland (Quinn et al., 2005). The remagnetization direction is, at 95%
156 confidence (Watson, 1983), the same as that found in mineralized and unmineralized
157 zones of the Navan deposit (Symons et al., 2002) and at Lisheen (Pannalal et al., 2008a).
158 Mean directions failed fold and conglomerate tests, indicating a secondary post-tilting
159 chemical magnetization.

160 The ChRM recorded from the deposits has been interpreted as synmineralization
161 even though it is imprinted on both ore samples and adjacent unmineralized host rocks
162 (e.g., Symons et al., 2002). Furthermore, the principal carrier, magnetite, is not known in
163 the ore assemblage; it occurs only in paragenetically early, distal iron oxide facies, such
164 as at Tynagh (e.g., Schultz, 1966). This is not surprising, given the mildly acidic
165 hydrothermal conditions, suggested by extensive host-rock dissolution and muscovite-
166 illite formation (e.g., Wilkinson et al., 2011), that are inconsistent with magnetite stability
167 (Cooke et al., 2000). We therefore argue that the same ChRM recorded in the vast
168 majority of samples is an overprint on all rocks throughout the Irish Midlands, regardless
169 of their exposure to hydrothermal ore fluids.

170 This remagnetization event also appears to have affected older rocks (Fig. 3);
171 magnetization of intermediate stability was identified in studies of the Silurian of Dingle
172 (Mac Niocaill, 2000) and of north Galway–south Mayo (Smethurst and Briden, 1988;
173 Smethurst et al., 1994). A pervasive remagnetization with a mean estimated age of $307 \pm$
174 6 Ma was also found in Devonian–Carboniferous rocks in southern Ireland (Pastor-Galán
175 et al., 2015). These also overlap those we have recorded, implying a pervasive
176 remagnetization in the late Carboniferous.

177 Given its synfolding to postfolding timing and age estimate, we attribute this
178 large-scale regional remagnetization to burial and/or fluid flow associated with the
179 Variscan orogeny, coincident with the development of the pan-European Cantabrian
180 orocline (Pastor-Galán et al., 2015). Our findings do not exclude the possibility that
181 mineralization could have been associated with such an event, but an explanation for the
182 localization of the ore deposits within such a context is lacking. Furthermore, none of the

183 published studies has demonstrated that the remagnetization and mineralization are
184 coeval. Rather, the catalogue of geological and geochemical evidence that supports an
185 early Carboniferous age for mineralization (e.g., Wilkinson and Hitzman, 2015) and the
186 recent derivation of early Chadian (346.6 ± 3.0 Ma) and Asbian (334.0 ± 6.1 Ma) Re-Os
187 dates on ore-stage pyrite (Hnatyshin et al., 2015) lead us to confidently rule out a
188 Variscan age for the ores.

189 Our results inform the wider controversial debate on the utility of paleomagnetism
190 for dating ore deposits. Globally, such dates have been used to argue for a link between
191 major episodes of Zn-Pb mineralization and the development of collisional orogens
192 during supercontinent assembly cycles (Leach et al., 2001). However, resetting of
193 magnetic signatures by orogenesis, either via fluid-related chemical reactions or burial,
194 has been known for a long time (e.g., McCabe and Elmore, 1989; Katz et al., 1998).
195 Consequently, we question the inferred link between supercontinent assembly and Zn-Pb
196 deposits (also see Kesler and Carrigan, 2002; Kesler et al., 2004) and suggest that at least
197 some of the MVT deposits reviewed by Leach et al. (2001) formed during basin
198 extension and were remagnetized later. For example, the large age discrepancies between
199 geochronological and paleomagnetic ages observed in eastern Tennessee (USA), Upper
200 Silesia (Poland), and Pine Point (Canada) can, in all cases, be explained by a post-
201 mineralization remagnetization. In the case of Pine Point, the Devonian age constraint
202 provided by Rb-Sr dating of sphalerite (Nakai et al., 1993) is consistent with sulfur
203 isotope data that cannot be explained by younger ages (Wilkinson, 2014). The Laramide
204 paleomagnetic age of 71 ± 13 Ma at Pine Point (Symons et al., 1993), as well as other

205 Laramide ages in the Western Canada Sedimentary Basin (Leach et al., 2001), can be
206 accounted for by orogenic fluid flow (Gillen et al., 1999; Kesler et al., 2004).

207 Our results from Ireland show that paleomagnetic dates here record orogenic
208 events, rationalizing their contradiction with many other lines of evidence regarding the
209 timing of mineralization in the province. Therefore, we suggest that paleomagnetic ages
210 should only be used to constrain large-scale models for fluid flow and geodynamic
211 models for mineralization where an explicit link can be made between the mechanism of
212 ore formation and remagnetization.

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335 **FIGURE CAPTIONS**

336 Figure 1. Geological map of Ireland showing the Galmoy, Lisheen, and Navan mines
337 (white circles), sample localities (black circles; codes are given in Table 1), and pre-
338 Carboniferous sample sites from previous work (white stars).

341

342 Figure 2. Three representative orthogonal projection plots. A: The data from Ballykane
343 Hill (BH4X). B: Newmarket Bypass (NB2Y). C: Kells-East (KE6X). D: Equal-area
344 projection plot for the site means. E: Same data after tilt correction. F: Results of the fold
345 test (method of McFadden and Jones, 1981).

347

348 Figure 3. Paleomagnetic pole for this study (blue) with 95% error ellipse. Data for Navan
349 (Symons et al., 2002), Lisheen, and Galmoy (Pannalal et al., 2008a, 2008b) and for
350 overprints in older rocks (Smethurst and Briden, 1988; Smethurst et al., 1994; Mac
351 Niocaill, 2000) are shown with beige and orange error ellipses, respectively. The
352 apparent polar wander path from Torsvik et al. (2012) is plotted in blue; reference poles
353 are in green. Ages are in Ma.

354

355 ¹GSA Data Repository item 2017xxx, SEM microscopy and rock magnetism, is available
online at

357 <http://www.geosociety.org/datarepository/2017/> or on request from

358 editing@geosociety.org.

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TABLE 1. SUMMARY OF SAMPLE LOCATIONS AND SITE MEAN DIRECTIONS

| Locality | Code | Age | Site | Strike/dip | N/N ₀ | R | Dec (°) | Inc (°) | K | a ₉₅ | VGP lat | VGP long |
|-----------------------|------|-----------|---------------|------------|------------------|------|------------|------------|-------|-----------------|---------|----------|
| Ballykane Hill | BH | Courceyan | 53.41N, 7.01W | | 5/7 | 4.83 | 199.6 | 19.2 | 23.7 | 16.0 | -48.3 | 331.5 |
| Barrow River | BR | Chadian | 53.02N, 7.02W | 270/10 | 5/5 | 4.75 | 191.4 | -7.4 | 16.3 | 19.6 | -39.8 | 338.1 |
| Butlersgrove Quarry | BQ | Courceyan | 52.65N, 7.03W | 284/10 | 0/7 | | | | | | | |
| Cloughjordan North | CN | Courceyan | 52.95N, 8.05W | | 5/8 | 4.98 | 183.8 | -18.0 | 241.0 | 5.0 | -46.1 | 346.6 |
| Fogharty's Quarry | FQ | Chadian | 53.05N, 8.22W | | 0/7 | | | | | | | |
| Garrylucas Beach | GB | Courceyan | 51.64N, 8.56W | 330/56 | 0/8 | | | | | | | |
| Kells East | KE | Courceyan | 52.54N, 7.25W | | 5/8 | 4.93 | 194.3 | -23.8 | 55.3 | 10.4 | -36.9 | 325.4 |
| Knockshangarry Quarry | KQ | Courceyan | 53.19N, 8.52W | | 5/5 | 4.69 | 199.1 | -2.5 | 12.8 | 22.3 | -35.7 | 327.8 |
| Newmarket Bypass | NB | Courceyan | 52.78N, 8.92W | | 5/5 | 4.86 | 200.7 | 26.5 | 28.9 | 14.5 | -20.9 | 329.6 |
| Round Tower | RT | Courceyan | 53.67N, 6.65W | 137/32 | 0/9 | | | 68 | | | | |
| Swords Roundabout | SR | Courceyan | 53.43N, 6.65W | 136/30 | 3/5 | 2.96 | 202.0 | -6.9 | 55.8 | 16.6 | -24.6 | 331.7 |
| Tory Hill | TH | Courceyan | 52.54N, 8.69W | 120/42 | 6/7 | 5.86 | 196.1 | 0.2 | 35.9 | 11.3 | -35.7 | 331.3 |
| Trim Quarry | TQ | Courceyan | 53.52N, 6.77W | 158/35 | 6/7 | 5.93 | 197.4 | -14.2 | 68.9 | 8.1 | -41.6 | 329.8 |
| Urlingford South | ES | Courceyan | 52.71N, 7.60W | | 0/8 | | | | | | | |
| Mean | | | | | 9 | 8.63 | 196.1 | -3.1 | 21.9 | 11.3 | | |

367
 368

Note: Dec—declination; Inc—inclination; N/N₀—Number of samples from which a stable direction could be determined/total number of samples; VGP—virtual geomagnetic pole.