

Variations in the magnetic properties of meteoritic cloudy zone

Claire I. O. Nichols*, James F. J. Bryson, Roberts Blukis, Julia Herrero-Albillos, Florian Kronast, Rudolf Ruffer, Aleksandr I. Chumakov, Richard J. Harrison

*Corresponding author: cion2@mit.edu

This article has been accepted for publication and undergone full peer review but has not been through the copyediting, typesetting, pagination and proofreading process, which may lead to differences between this version and the Version of Record. Please cite this article as doi: 10.1029/2019GC008798

©2020 American Geophysical Union. All rights reserved.

Variations in the magnetic properties of meteoritic cloudy zone

Claire I. O. Nichols^a, James F. J. Bryson^b, Roberts Blukis^c, Julia Herrero-Albillos^{d,e}, Florian Kronast^f, Rudolf Ruffer^g, Aleksandr I. Chumakov^g, Richard J. Harrison^b

^a Department of Earth, Atmospheric and Planetary Sciences, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, USA

^b Department of Earth Sciences, University of Cambridge, Downing Street, Cambridge, CB2 3EQ, UK

^c Helmholtz Zentrum Potsdam, Deutsches GeoForschungsZentrum GFZ, Telegrafenberg, 14473 Potsdam, Germany

^d Centro Universitario de la Defensa, Zaragoza 50090, Spain

^e Instituto de Ciencia de Materiales de Aragon, Departamento de Física de la Materia Condensada, Consejo Superior de Investigaciones Científicas, Universidad de Zaragoza, Zaragoza 50009, Spain

^f Helmholtz Zentrum Berlin, Elektronenspeicherring BESSY II, Albert-Einstein-Strasse 15, Berlin 12489, Germany

^g ESRF - The European Synchrotron, CS40220, 38043 Grenoble, Cedex 9, France

Key Words: Meteorites, Rock magnetism, Paleomagnetism

Key Point: Meteoritic cloudy zone formed at intermediate cooling rates can acquire a stable and reliable nanopaleomagnetic remanence.

Abstract

Iron and stony-iron meteorites form the Widmanstätten pattern during slow cooling. This pattern is comprised of several microstructures whose length-scale, composition and magnetic properties are dependent upon cooling rate. Here we focus on the cloudy zone: a region containing nanoscale tetrataenite islands with exceptional paleomagnetic recording properties. We present a systematic review of how cloudy zone properties vary with cooling rate and proximity to the adjacent tetrataenite rim. X-ray photoemission electron microscopy (X-PEEM) is used to compare compositional and magnetization maps of the cloudy zone in the mesosiderites (slow cooling rates), the IAB iron meteorites and the pallasites (intermediate cooling rates) and the IVA iron meteorites (fast cooling rates). The proportions of magnetic phases within the cloudy zone are also characterised using Mössbauer spectroscopy. We present the first observations of the magnetic state of the cloudy zone in the mesosiderites, showing that, for such slow cooling rates, tetrataenite islands grow larger than the multi-domain threshold, creating large-scale regions of uniform

magnetization across the cloudy zone that render it unsuitable for paleomagnetic analysis. For the most rapidly cooled IVA meteorites, the time available for Fe-Ni ordering is insufficient to allow tetrataenite formation, again leading to behaviour that is unsuitable for paleomagnetic analysis. The most reliable paleomagnetic remanence is recorded by meteorites with intermediate cooling rates ($\sim 2 - 500^\circ\text{C Myr}^{-1}$) which produces islands that are 'just right' both in size and degree of Fe-Ni order.

1 Introduction

Iron meteorites are characterised by the highly distinctive Widmanstätten pattern, a complex growth of Fe-Ni microstructures revealed by polishing and etching the metallic surface. The Widmanstätten pattern forms during slow cooling; the full range of microstructures are observed for cooling rates below $\sim 10,000^\circ\text{C Myr}^{-1}$ (Yang et al., 1997, 2010). Kamacite (α), the body centered cubic (bcc) Fe-rich endmember, nucleates and begins to grow upon cooling through $\sim 900^\circ\text{C}$. This phase makes up the bulk of the meteoritic metal and its large grain size means it is magnetically multidomain. The nucleation of kamacite causes Ni to be partitioned into the surrounding face centered cubic (fcc) taenite (γ). An 'M'-shaped Ni-diffusion profile develops, with highest Ni concentrations of ~ 50 wt% immediately adjacent to the kamacite. As Ni content drops below ~ 47 wt%, spinodal decomposition takes place forming a mixture of nanoscale $\text{Fe}_{0.5}\text{Ni}_{0.5}$ islands in an Fe-rich matrix. This microstructure is known as the cloudy zone. As Ni content continues to decrease to ~ 25 wt%, spinodal decomposition is initiated at lower temperatures. Islands are largest ($< \sim 500$ nm) in the most Ni-rich regions (Yang et al., 1997; Maurel et al., 2019). Regions with < 25 wt% Ni, below the stability of the spinodal region, undergo a martensitic transition, spontaneously transforming from fcc taenite to bcc martensite. The temperature of the martensitic transition increases with decreasing Ni content. If the transition occurs at a high temperature and the cooling rate is slow, the martensite has enough time upon further cooling to exsolve into plessite, a complex intergrowth of kamacite and taenite (Goldstein and Michael, 2006).

The cloudy zone has been shown to be an excellent paleomagnetic recorder (Uehara et al.,

2011; Bryson et al., 2014b; Einsle et al., 2018). As the islands cool below 320 °C they transform to ordered tetrataenite. Ordering occurs parallel to the {100} planes and the magnetic easy axis is aligned perpendicular to the ordered planes. The cloudy zone islands acquire a chemical transformation remanent magnetization (CTRM) which is extremely stable. Thermal overprints at relatively low temperatures ($> \sim 360$ °C) are easily observable since they can dissolve the cloudy zone entirely (Goldstein et al., 2009b), and the high intrinsic coercivity of the cloudy zone (> 1 T; Uehara et al., 2011) means an isothermal remanent magnetization (IRM) overprint is unlikely since naturally-occurring magnetic fields are rarely this strong.

The size of the islands in the cloudy zone depends on the cooling rate; the faster the cooling, the smaller the islands and this relationship is empirically well defined by the equation $d_{CZ}^{2.9} = \frac{k}{CR}$ where d_{CZ} is the diameter of the largest cloudy zone islands in nm, k is a constant, equal to 7,620,000 °C nm^{2.9} Myr⁻¹ and CR is cooling rate in °C Myr⁻¹ for a temperature range of 500 – 700 °C (Yang et al., 2010). Cooling rates are determined by measuring the diameter of tetrataenite islands within the cloudy zone and the width of the tetrataenite rim in etched samples (Yang et al., 1997, 2008, 2010; Goldstein et al., 2009a). More accurate cooling rate estimates are obtained by measuring the shape of the ‘M’-shaped Ni diffusion profile across kamacite-taenite interfaces using electron probe microanalysis (EPMA) (Goldstein et al., 2014). Island size also decreases with distance from the tetrataenite rim. Since islands form by spinodal decomposition, as opposed to nucleation and growth, islands spontaneously form at ≤ 85 % of their present day size (Maurel et al., 2019). The first islands to form are closest to the tetrataenite rim where the Ni content is highest. The Ni content of the cloudy zone affects the temperature at which it enters the spinodal region; at lower Ni contents — and therefore lower spinodal initiation temperatures — smaller islands are formed.

The cooling rate during cloudy zone formation controls the ability of this microstructure to record paleomagnetic information. Cooling rate affects island size, the degree of ordering (tetrataenite vs taenite) and packing fraction, which in turn controls the strength of interactions between islands. In this paper we will describe the optimal cloudy zone for recording paleomagnetic information, dictated by both cooling rate and distance from the tetrataenite

86 rim using X-ray photoemission electron microscopy (X-PEEM) and Mössbauer spectroscopy.
 87 We review existing data for fast and intermediate cooling rates, and present the first results
 88 characterising the magnetic properties of the cloudy zone in the slowest cooled group of iron
 89 and stony-iron meteorites; the mesosiderites. The meteorites reviewed here are summarised
 90 in Table 1. We investigate how the magnetic state of the cloudy zone changes as function of
 91 both cooling rate and proximity to the tetrataenite rim and discuss the implications for the
 92 preservation and quantification of paleomagnetic remanence.

Meteorite	Type	TT Islands (nm)	CR ($^{\circ}\text{C Myr}^{-1}$)	References
Bishop Canyon	IVA Iron	12	2500 ± 1.3	Yang et al. (2007)
Steinbach	IVA Iron	29	150	Goldstein et al. (2009b)
Chinautla	IVA Iron	32	110 ± 1.7	Goldstein et al. (2009b)
Tazewell	IAB Iron	90 – 100	20.8	Goldstein et al. (2014)
Toluca	IAB Iron	100 – 120	11.6	Goldstein et al. (2014)
Marjalahti	MG Pallasite	118 ± 3	7.6 ± 0.6	Yang et al. (2010)
Odessa	IAB Iron	120 – 300	11.6	Nichols et al. (2018)
Brenham	MG Pallasite	123 ± 3	6.2 ± 0.9	Yang et al. (2010)
Springwater	MG Pallasite	132 ± 3	5.4 ± 0.5	Yang et al. (2010)
Imilac	MG Pallasite	143 ± 4	4.3 ± 0.3	Yang et al. (2010)
Esquel	MG Pallasite	157 ± 11	3.3 ± 0.6	Yang et al. (2010)
Estherville	Mesosiderite	463 ± 32	0.2 – 0.5	Goldstein et al. (2014)

Table 1: Table summarizing the meteorites reviewed in this study including the largest tetrataenite island diameters (TT Islands) and their cooling rates at ~ 500 $^{\circ}\text{C}$ (CR). ‘MG pallasite’ refers to pallasite meteorites from the Main Group.

93 2 Sample Characterisation

94 2.1 The Mesosiderites

95 The mesosiderites are an unusual group of stony-iron meteorites, comprised of FeNi metal and a
96 range of brecciated basaltic, gabbroic, dunitic and orthopyroxene-rich clasts (Greenwood et al.,
97 2015). The composition of the silicates, combined with FeNi metal, suggests the mesosiderites
98 represent a mixture of core and crustal material. The oxygen isotope composition of the silicates
99 has been used to infer their origin on the same parent body, and their well-mixed compositions
100 and brecciated nature imply they represent a regolith breccia (Mittlefehldt, 1980). It has been
101 proposed that an impact event mixed the molten core of an impactor with the regolith breccia,
102 forming the mesosiderites.

103 An unresolved issue is the exceptionally slow cooling rates of the mesosiderites, which is
104 slower than that of any other meteorite group. Initially, the mesosiderites cooled rapidly; Fe-
105 Mg profiles in pyroxenes suggest cooling rates of $1 - 100 \text{ }^\circ\text{C yr}^{-1}$ between $1150 - 900 \text{ }^\circ\text{C}$ and
106 plagioclase overgrowth textures predict a cooling rate $\geq 0.1 \text{ }^\circ\text{C yr}^{-1}$ between $1100 - 850 \text{ }^\circ\text{C}$
107 (Delaney et al., 1980; Ruzicka et al., 1994). This initial fast cooling rate is attributed to rapid
108 thermal equilibration between hot and cold ejecta during the impact event (Scott et al., 2001).
109 At temperatures below $400 \text{ }^\circ\text{C}$, cooling rates drop to $0.2 - 0.5 \text{ }^\circ\text{C Myr}^{-1}$ based on Ni and Co
110 concentrations at kamacite-taenite interfaces (Goldstein et al., 2014; Wasson and Hoppe, 2014).
111 This is supported by the large size of the cloudy zone microstructure — tetrataenite islands
112 are $400 - 450 \text{ nm}$ in diameter (Goldstein et al., 2009a) — yielding a cooling rate estimate
113 of $0.5 \text{ }^\circ\text{C Myr}^{-1}$ (Yang et al., 1997). Fe-Mg ordering in orthopyroxene and $^{40}\text{Ar}/^{39}\text{Ar}$ ages
114 also support very slow cooling (Ganguly et al., 1994; Bogard and Garrison, 1998). These
115 extremely slow cooling rates are difficult to reconcile with the inference that the metal mixed
116 with crustal and regolith material near the parent body surface. It has been proposed that
117 after their formation, the mesosiderites were buried deep inside their parent body, which must
118 be well-insulated by a thick regolith layer (Scott et al., 2001; Greenwood et al., 2015). Samples
119 BM 65575 and BM 53764 of the Estherville mesosiderite were borrowed from the Natural

120 History Museum, London.

121 **2.2 The Pallasites**

122 The pallasites consist of an intimate mix of FeNi metal and large (cm-sized) olivine crystals
123 and were initially considered to originate from the core-mantle boundary of a differentiated
124 asteroid. It has since been shown that their variable cooling rates and paleomagnetic record
125 of an internal core dynamo firmly rule out an origin deep within a planetesimal interior (Yang
126 et al., 2010; Tarduno et al., 2012). It is now proposed that the pallasites were formed by
127 an impact between two differentiated planetesimals, resulting in the molten core of one being
128 injected into the mantle of the other. The significant difference in density between silicate
129 and metal suggests that the two phases should quickly separate; their intimate mix therefore
130 suggests initial rapid cooling following the intrusion of molten metal. Further impact events
131 and the addition of a regolith layer to the parent body could explain late, slow cooling below
132 700 °C of 2.5 – 18 °C Myr⁻¹ (Yang et al., 2010).

133 Samples of Brenham (BM 68725) and Marjalahti (BM 1920,318) were obtained from the
134 Natural History Museum, London. Previous magnetic and compositional characterisation of
135 these samples are reported by Nichols et al. (2016). Paleomagnetic results revealed evidence
136 for cessation of a dynamo on the pallasite parent body after a thermally convective dynamo
137 and prior to a compositional dynamo driven by core solidification.

138 **2.3 The IAB Iron Meteorites**

139 The IAB iron meteorites are an unusual group, since they are not thought to form via fractional
140 crystallization in a planetary core (Benedix et al., 2014). They contain silicate fragments with
141 both primitive achondritic and chondritic compositions, which supports the hypothesis that the
142 IAB parent body was partially differentiated (Benedix et al., 2000). It is likely that the IAB
143 irons formed as isolated pools of metal after an impact event disrupted the process of planetary
144 differentiation (Schulz et al., 2012). The IAB iron meteorites are rich in carbon, and exhibit
145 unusual FeNi microstructures (Buchwald, 1975; Goldstein et al., 2017; Nichols et al., 2018).

146 Samples of Toluca (TN 4389), Odessa (11538) and Tazewell (16269) were borrowed from
147 the Sedgwick Museum, University of Cambridge. These samples have previously been studied
148 using X-PEEM and results are reported in Nichols et al. (2018) and Bryson et al. (2014b).
149 Paleomagnetic results reported by Nichols et al. (2018) suggested the IAB parent body did not
150 fully differentiate and did not have a substantial metallic core capable of generating a dynamo.

151 **2.4 The IVA Iron Meteorites**

152 The IVA iron meteorites are thought to originate from a metallic core that had its overlying
153 silicate mantle removed by glancing collisions during the early solar system. Without the insu-
154 lation provided by a silicate mantle, the parent core of the IVAs is expected to have cooled far
155 quicker than most other iron meteorites. Indeed, the sizes of the low-temperature microstruc-
156 tures within these meteorites are the smallest that have been measured, with cooling rates
157 ranging from $100 - 10,000 \text{ }^\circ\text{C Myr}^{-1}$ (Yang et al., 2008). Furthermore, unlike Earth's core,
158 the rapid cooling rate of the surface of the IVA parent core is expected to result in initial solid-
159 ification at the asteroid surface, and the crystallisation front will then have advanced toward
160 the centre of the core as cooling continued. The solidification of this core therefore represents a
161 fundamentally different regime to that of our own planet and could provide crucial constraints
162 on dynamo generation (Neufeld et al., 2019).

163 A sample of the Steinbach IVA iron meteorite, sample number BM 35540 was acquired from
164 the Natural History Museum, London. Previous X-PEEM imaging and bulk paleomagnetic
165 measurements on this sample are reported by Bryson et al. (2017). Paleomagnetic results
166 revealed the first direct evidence of inward core solidification within an asteroid.

167 **3 Methods**

168 **3.1 Sample Preparation**

169 Samples were prepared in the Department of Earth Sciences, University of Cambridge, by
170 cutting $\sim 5 \times 5$ mm sections using a tile-cutting saw under running water to keep the samples

171 cool. Samples were then ground down until < 1 mm thick and polished, down to $0.25 \mu\text{m}$ -grade
172 diamond paste. They were subsequently etched for ~ 20 seconds using nital (2 % nitric acid
173 in ethanol) and examined using a reflected light microscope to check for signs of alteration or
174 shock. Samples were repolished to reveal a fresh surface prior to X-PEEM measurement. They
175 were then sputtered for ~ 18 hours using a focussed Ar-ion beam under ultra-high vacuum
176 (pressure $< 1.5 \times 10^{-5}$ mbar) while gradually decreasing the voltage from 1.2 keV to 0.4 keV
177 to remove any oxidation or surface magnetization induced by polishing. Samples were kept in
178 vacuum between sputtering and measuring (measuring pressure $< 1.0 \times 10^{-8}$ mbar).

179 **3.2 X-ray Photoemission Electron Microscopy (X-PEEM)**

180 X-PEEM was used to collect images with nanoscale resolution of the composition and magne-
181 tization of iron meteorites. X-PEEM was performed at the SPEEM UE49 beamline, BESSY II
182 (Berlin, Germany). An intense beam of monochromatic X-rays is focussed at an angle of 16° to
183 the sample surface. Secondary photoelectrons are excited by the X-rays from the top ~ 5 nm
184 of the sample surface. This technique allows compositional and magnetic imaging to be carried
185 out in the same location in quick succession by changing the energy and polarisation of the
186 X-ray beam, meaning a direct comparison can be made between composition and magnetiza-
187 tion for each region. Each image is individually assessed for light drift and charging artefacts
188 both qualitatively and quantitatively by comparing light intensity in sequential images using a
189 Student T-test; only the highest quality data are selected for further analysis. 160 images (80
190 for each polarity, or change in energy) are acquired with a 2 second exposure time per image.
191 These images are then aligned by selecting a surface feature as a reference point, and averaged
192 to improve signal to noise ratio in the final images.

193 Compositional images were acquired using linearly polarised X-rays tuned to the energy of
194 the Fe L_3 and Ni L_3 edges (~ 707 eV and ~ 852 eV, respectively). The energy of the X-ray beam
195 is fine-tuned by imaging the sample over a range of energies and precisely locating the L_3 peaks.
196 The L_3 peaks are used since they provide maximum compositional contrast when conducting
197 X-PEEM. Images were also acquired at pre-edge energies (~ 700 eV and ~ 845 eV for Fe and

198 Ni, respectively). Each on-edge image was normalised by dividing by its equivalent pre-edge
199 image. The Ni image was divided by the Fe image in order to generate a semi-quantitative map
200 of Ni/Fe ratio.

201 **3.2.1 X-ray Magnetic Circular Dichroism (XMCD)**

202 XMCD was used in combination with X-PEEM to image the magnetization state at the energy
203 of the Fe L_3 edge (Stöhr et al., 1998; Ohldag et al., 2001). XMCD intensity is a projection of
204 the magnetization onto the incident direction of the X-ray beam; negative intensities represent
205 magnetization towards the X-ray beam and are coloured red, while positive intensities (mag-
206 netization parallel to the beam) are blue. magnetization perpendicular to the beam, as well as
207 non-magnetic material appears white. Images were acquired for both left and right circularly
208 polarised X-rays, which excite electrons in opposite spin states. Since spin state directly cor-
209 relates to the direction of magnetization, this generates contrast depending on the component
210 of magnetization parallel to the X-ray beam. Images were taken along parallel sections of the
211 tetrataenite rim in order to be directly comparable. Images taken in each polarisation state
212 were subtracted from one another and divided by their sum, in order to normalise and enhance
213 magnetic contrast. Thus,

$$I = \frac{I_R - I_L}{I_R + I_L} \quad (1)$$

214 where I_R and I_L are right and left polarised X-ray intensities, respectively. I is the resulting
215 XMCD intensity (Bryson et al., 2014b).

216 **3.3 Mössbauer spectroscopy**

217 Samples of Tazewell, Esquel and Estherville were prepared by mechanical polishing on a carbide
218 paper to the desired thickness of 20 – 40 μm . Samples were further mechanically polished with
219 water based monocrystalline diamond suspension, with the final particle diameter of 0.25 μm .
220 Synchrotron Mössbauer spectroscopy was performed with the Synchrotron Mössbauer Source
221 (SMS) (Smirnov et al., 1997; Potapkin et al., 2012) at the Nuclear Resonance beamline ID18

222 (Rüffer and Chumakov, 1996) at the European Synchrotron Radiation Facility in Grenoble,
223 France . The beam from SMS is nearly 100 % polarized with the electric vector in the vertical
224 direction. Typical count rates on the sample reach 16 kHz resonant quanta. The beam can be
225 focused to a spot size of $9 \times 4 \mu\text{m}^2$ FWHM under ideal conditions (Rüffer and Chumakov, 1996).
226 For our experiment, the beam shape is approximately Gaussian with a focused beam size [h \times v]
227 of $12 \times 16.5 \mu\text{m}^2$ FWHM for the Estherville sample was achieved. The energy distribution
228 of the source follows a Lorentzian squared distribution with a linewidth of 0.27 mm s^{-1} . The
229 Doppler shift was provided by oscillating the $^{57}\text{FeBO}_3$ nuclear monochromator with a sinusoidal
230 drive and maximum velocity of 11.25 mm s^{-1} distributed over 1048 velocity channels (unfolded)
231 in the multi-channel analyzer. All spectra were acquired at room temperature ($\sim 22 \text{ }^\circ\text{C}$). The
232 storage ring was operated in $7/8 + 1$ filling mode. Samples were mounted over holes in an Al
233 holder. The holder was bolted to a large moving stage with $\pm 1 \mu\text{m}$ accuracy. Sample navigation
234 was achieved by first mapping changes in X-ray absorption as the sample was rastered across
235 the X-ray beam, enabling the XY stage positions corresponding to prominent features around
236 the sample edge to be determined. The locations of individual spectral measurements were
237 then determined by triangulation of the stage position relative to scanning electron microscope
238 (SEM) images of the samples on the holder. Using this method we estimate the uncertainty in
239 the absolute position of individual spectral measurements to be $\sim 10 - 20 \mu\text{m}$. The uncertainty
240 in the relative position of spectral measurements within a single profile is equal to the accuracy
241 of the stage movement. Due to the high intensity of synchrotron radiation, average spectrum
242 acquisition time was ~ 2800 seconds.

243 Phase fractions of kamacite, tetrataenite and antitaenite were calculated by comparing the
244 relative proportions of each phase from the Mössbauer spectra and normalising them based on
245 Fe content. Fe contents of kamacite, tetrataenite and antitaenite are assumed to be 95 at.%,
246 50 at.% and 85 at.%, respectively (Blukis et al., 2017).

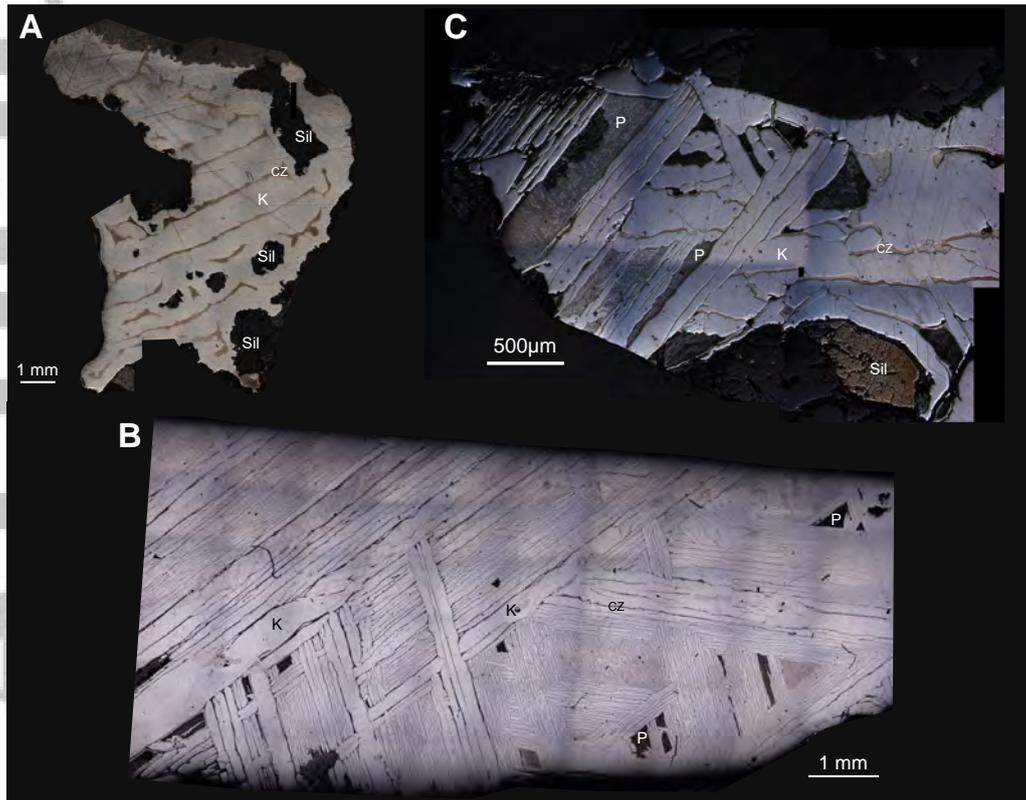


Figure 1: Polished and etched sections of (a) the Estherville mesosiderite (9.2 wt.% Ni) (b) the Brenham pallasite (10.6 wt.% Ni) and (c) the Steinbach IVA iron (9.4 wt.% Ni). Compositions are from Goldstein et al. (2014). The images highlight the clear changes in lengthscale of the Widmanstätten pattern with cooling rate; Estherville exhibits the slowest cooling rate and the coarsest features, and vice versa for Steinbach. Each image is annotated to highlight kamacite lamellae (K), the cloudy zone (CZ), plessite (P) and silicates (Sil).

247 4 Results

248 4.1 Cloudy zone behaviour as a function of cooling rate and proximity 249 to the tetrataenite rim

250 The lengthscale on which Fe-Ni microstructures form is dependent on cooling rate and Ni
251 content. The effect of cooling rate can be observed by examining polished and etched surfaces of
252 iron meteorites with similar Ni contents (Figure 1). In fast-cooled samples the Widmanstätten
253 pattern forms on a much finer scale; the kamacite lamellae are narrow (200 – 300 μm in width),
254 as are the Ni-rich regions between them. In the slowest cooled samples the Widmanstätten
255 pattern is much broader (kamacite lamellae have a width of 1 – 2 mm). These variations are

256 also observed within the individual FeNi microstructures. In this section we will review how the
257 cloudy zone microstructure varies with cooling rate, focusing on the variations in the resultant
258 magnetic behaviour.

259 The fastest cooled sample reviewed here is Bishop Canyon, a IVA iron meteorite with a
260 cooling rate of $2500\text{ }^{\circ}\text{C Myr}^{-1}$ (Yang et al., 2008). It has a tetrataenite rim with a width of
261 $80 \pm 4\text{ nm}$ and an average cloudy zone island diameter of $12 \pm 3\text{ nm}$ (Goldstein et al., 2009b).
262 A map of the magnetization direction within the kamacite, the tetrataenite rim, the cloudy zone
263 and plessite in Bishop Canyon is shown in Figure 2. All four microstructures show multidomain
264 behaviour, however domain size varies between microstructures. The largest domains ($\sim 1\text{ }\mu\text{m}$)
265 are observed in the kamacite. The tetrataenite rim is very narrow and magnetic domains
266 alternate along its length approximately once every $1 - 5\text{ }\mu\text{m}$. In the cloudy zone, narrow
267 regions of uniform magnetization (width of $100 - 200\text{ nm}$) run parallel to the tetrataenite rim.
268 There is no observable distinction between the magnetization of the islands compared to the
269 matrix in the cloudy zone; the orientation and shape of the regions of uniform magnetization
270 appear to only be controlled by the parallel boundaries between the microstructures.

271 IVA iron meteorites have a large range of cooling rates; Steinbach and Chinautla have much
272 slower cooling histories than Bishop Canyon (150 and $110\text{ }^{\circ}\text{C Myr}^{-1}$, respectively). Their
273 microstructures are therefore significantly larger; Steinbach and Chinautla have tetrataenite
274 rim widths of $195 \pm 5\text{ nm}$ and $215 \pm 25\text{ nm}$, and cloudy zone island diameters of $29 \pm 3\text{ nm}$
275 and $32 \pm 4.5\text{ nm}$, respectively (Goldstein et al., 2009b). Compositional and magnetization
276 maps of kamacite, the tetrataenite rim, the cloudy zone and plessite are shown for Steinbach in
277 Figure 3. The kamacite shows large magnetic domains ($> 1\text{ }\mu\text{m}$), consistent with multidomain
278 behaviour. The tetrataenite rim also shows multidomain behaviour, but here the domains are
279 much smaller ($200 - 300\text{ nm}$). In the cloudy zone immediately adjacent to the tetrataenite
280 rim, the magnetization pattern appears to cluster around the islands, forming small regions of
281 uniform magnetization ($< 200\text{ nm}$). Further from the tetrataenite rim in the fine cloudy zone,
282 the islands and matrix are uniformly magnetized.

283 The pallasites and IAB iron meteorites reviewed here have intermediate cooling rates ($7.6 -$

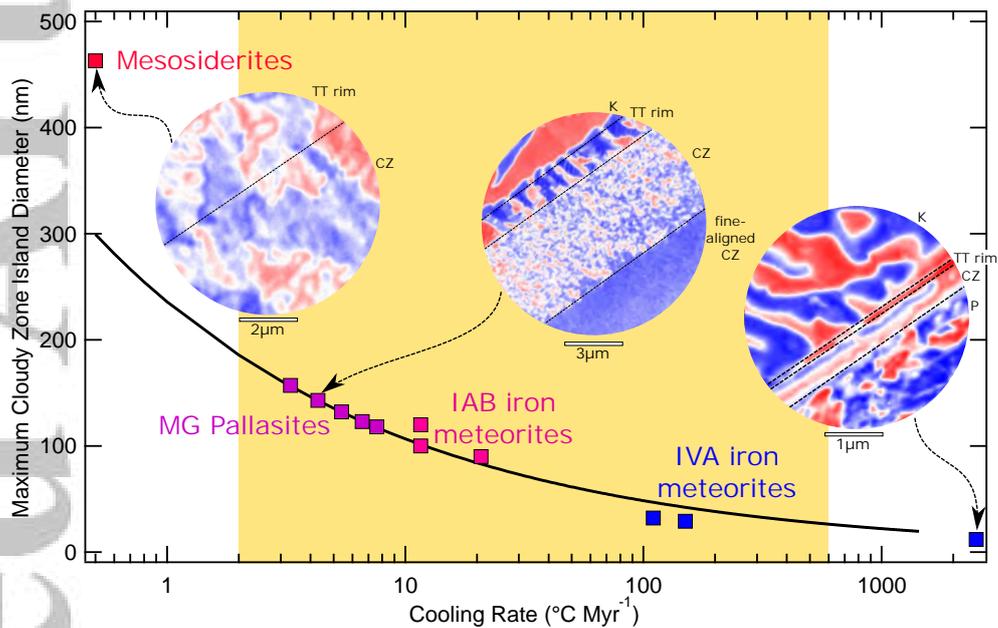


Figure 2: The graph shows the meteorites whose cloudy zone have been studied using X-PEEM. The black curve represents the empirically-derived relationship between cloudy zone island diameter and cooling rate defined by Yang et al. (2010). X-PEEM images show the magnetization in the kamacite (K), tetrataenite rim (TT rim), cloudy zone (CZ) and plessite (P) and the clear differences in the magnetic behaviour of these microstructures with cooling rate. The X-PEEM images shown are from the Estherville mesosiderite, the Marjalahti pallasite (Nichols et al., 2016) and the Bishop Canyon IVA iron meteorite (Bryson et al., 2017). The yellow region represents ideal cloudy zone for paleomagnetic study, which depends upon both cooling rate and spatial proximity to the tetrataenite rim.

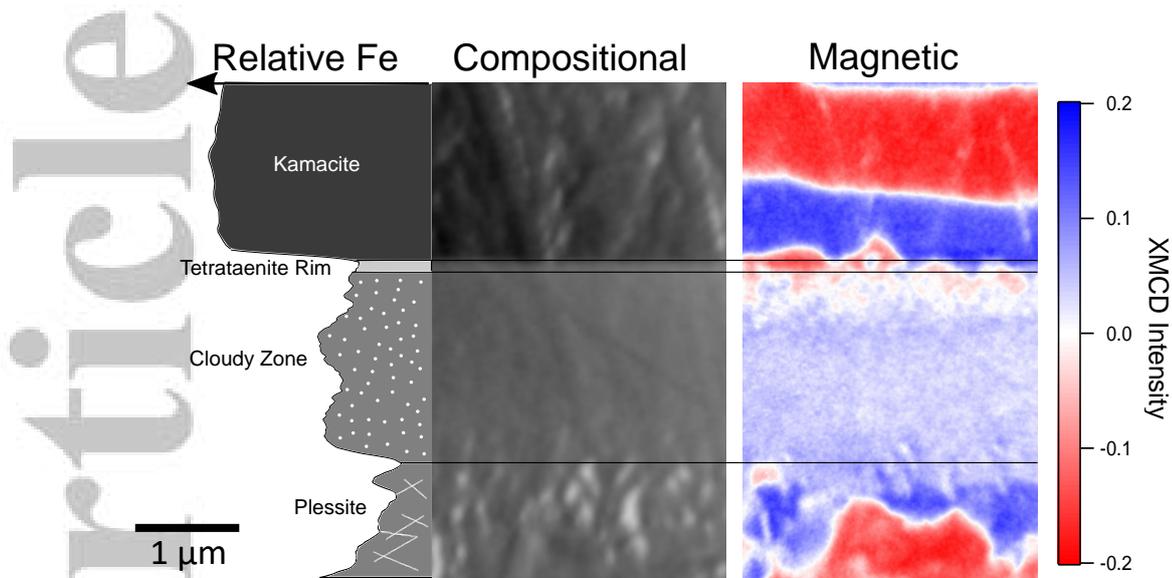


Figure 3: An example of a magnetic and compositional profile across the kamacite, tetrataenite rim, cloudy zone and plessite in the Steinbach IVA iron meteorite (Bryson et al., 2017). Note that the magnetic and compositional images do not correspond to the same region. All of the relevant microstructures were imaged in a single field of view.

284 $20.8 \text{ }^\circ\text{C Myr}^{-1}$) (Yang et al., 2010; Winfield et al., 2012; Goldstein et al., 2014). magnetization
 285 maps of the kamacite, the tetrataenite rim and the cloudy zone are shown for Imilac and
 286 Brenham in Figures 2 and 4a, respectively. The kamacite and tetrataenite rim both exhibit
 287 multidomain magnetic behaviour. Magnetic domains in the kamacite are significantly larger
 288 than those in the tetrataenite rim. The cloudy zone within ~ 4 widths of the tetrataenite
 289 rim is clustered into uniformly magnetized regions on a similar lengthscale to the size of the
 290 cloudy zone islands (90 – 160 nm) (Yang et al., 2010; Goldstein et al., 2014). In the fine
 291 cloudy zone, at distances exceeding ~ 4 tetrataenite rim widths the clustering is replaced
 292 by a region of uniform magnetization. In some cases, this transition is very sharply defined,
 293 such as in the magnetization map of Imilac (Figure 2) whereas the transition is more gradual
 294 and happens further from the tetrataenite rim in Brenham (Figure 4a). The sharpness of the
 295 boundary between the coarse and fine cloudy zone does not appear to correlate systematically
 296 with cooling rate and is also observed to vary within a single sample.

297 The mesosiderites have the slowest known cooling rates of any meteorite group. Here we
 298 present the first characterisation of the magnetic properties of the Estherville mesosiderite

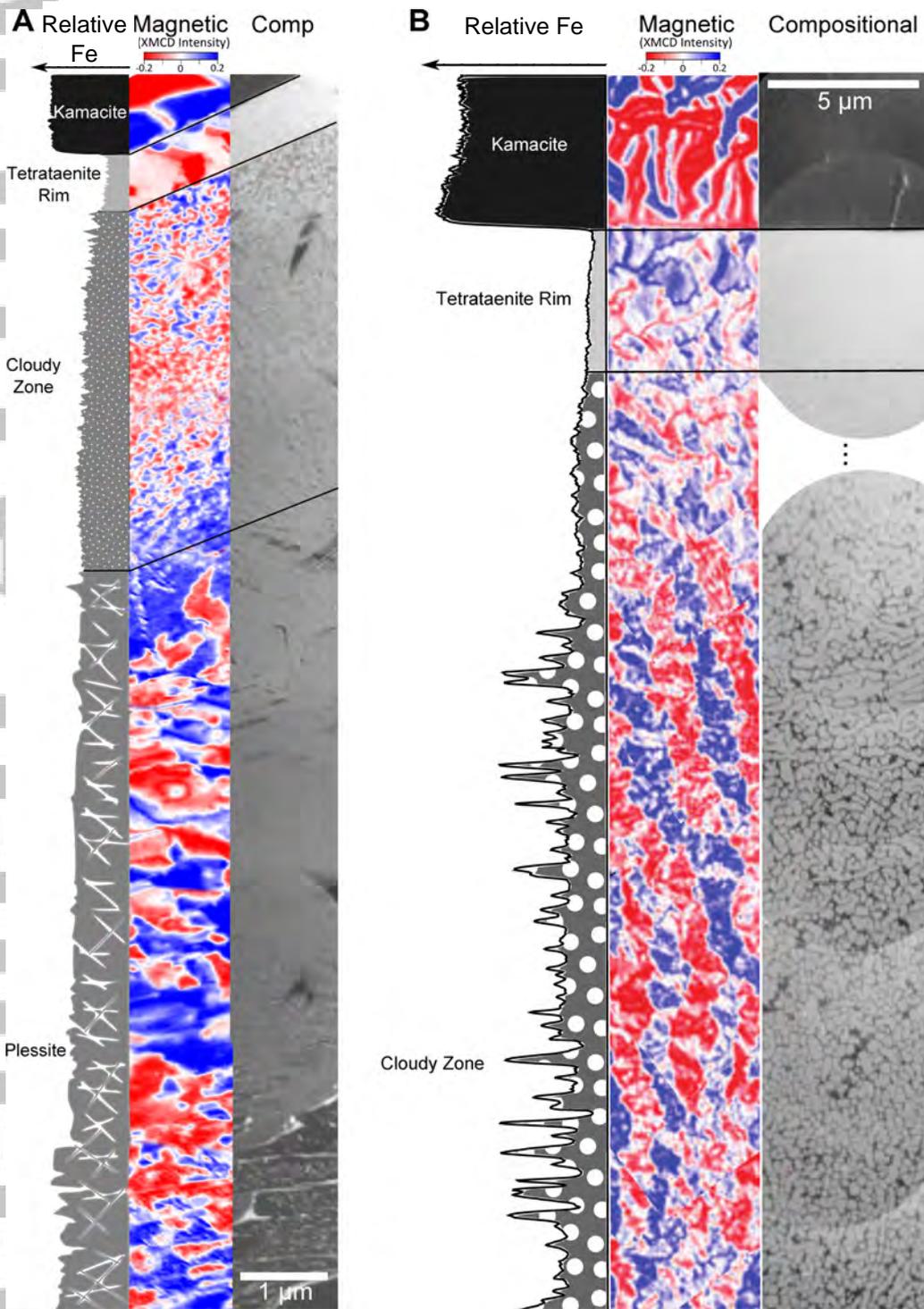


Figure 4: (a) X-PEEM images of the composition and magnetization of microstructures in the Brenham pallasite, with associated compositional profile. Brenham cooled at $\sim 6 \text{ }^\circ\text{C Myr}^{-1}$ below $\sim 500 \text{ }^\circ\text{C}$ (Yang et al., 2010). (b) X-PEEM images of the composition and magnetization of microstructures in the Estherville mesosiderite, with associated compositional profile. Estherville cooled at $\sim 0.5 \text{ }^\circ\text{C Myr}^{-1}$ below $\sim 500 \text{ }^\circ\text{C}$ (Yang et al., 1997).

299 (cooling rate $< 0.5 \text{ }^\circ\text{C Myr}^{-1}$). magnetization maps of Estherville are shown in Figures 2
300 and 4b. The microstructures here form on a much coarser scale; the tetrataenite rim width is
301 $\sim 5 \text{ } \mu\text{m}$ and the average cloudy zone island diameter is $463 \pm 32 \text{ nm}$ (Goldstein et al., 2014).
302 The kamacite has large ($1 - 2 \text{ } \mu\text{m}$ width) magnetic domains with a higher XMCD intensity than
303 the magnetization of the tetrataenite rim and the cloudy zone. The tetrataenite rim and the
304 cloudy zone both exhibit multidomain behaviour and there is no clear transition between the
305 two. Large ($\sim 2 \text{ } \mu\text{m}$ width) regions of uniform magnetization are observed as parallel stripes at
306 a high angle to the kamacite-tetrataenite-rim-interface and run directly across the tetrataenite
307 rim and cloudy zone (Figure 3).

308 4.2 Characterising the magnetic properties of the cloudy zone matrix

309 The magnetic behaviour of the cloudy zone matrix is not easily examined using X-PEEM; the
310 matrix is typically only a few nm across, and therefore cannot be spatially resolved. Matrix
311 properties are therefore evaluated using Mössbauer spectroscopy to acquire spatially resolved
312 spectral information for each magnetic phase within the cloudy zone. A previous study investi-
313 gated the behaviour of the cloudy zone in two intermediate cooled samples, Esquel and Tazewell,
314 and showed that their cloudy zones are comprised of ferromagnetic tetrataenite and paramag-
315 netic antitaenite (Blukis et al., 2017). Antitaenite has the same crystallographic structure
316 as taenite, but a different electronic structure resulting in its contrasting magnetic properties
317 (Danon et al., 1979; Rancourt, 1995; Rancourt et al., 1999; Lagarec et al., 2001). Here we
318 compare these results to new data for the slow cooled Estherville mesosiderite.

319 In all three meteorites, the percentage of the kamacite phase decreases in the cloudy zone.
320 There appears to be a systematic decrease in kamacite content with slower cooling rate, however
321 this is an artifact caused by the larger size of the focussed beam used for Mössbauer measure-
322 ments compared to the width of the cloudy zone (Blukis et al., 2017). The degree of overlap in
323 the signal between the kamacite and the cloudy zone decreases as the microstructures become
324 larger at lower cooling rates. In Esquel and Tazewell the entire kamacite signal in the cloudy
325 zone is considered to be an artifact; it is solely comprised of antitaenite and tetrataenite. This

326 is supported by a scanning precession electron diffraction (SPED) study of Tazewell in which
327 no kamacite was observed in the cloudy zone (Einsle et al., 2018). In Estherville on the other
328 hand, the width of the cloudy zone is significantly larger than the beam size. This suggests
329 there is a genuine kamacite phase component in the Estherville cloudy zone.

330 It appears that the proportions of antitaenite and tetrataenite in the cloudy zone may
331 vary with cooling rate. In Tazewell and Esquel there are approximately equal proportions of
332 each phase. In Estherville there is significantly less antitaenite in the cloudy zone and the
333 dominant phase is tetrataenite, however the ratio of tetrataenite to kamacite and antitaenite is
334 also approximately equal.

335 **4.3 Interpreting paleomagnetic signatures in the cloudy zone**

336 Paleomagnetic information is extracted from the cloudy zone by assessing the degree of bias
337 in magnetization direction in the cloudy zone (Bryson et al., 2014b; Maurel et al., 2019). The
338 cloudy zone adjacent to the tetrataenite rim in two intermediate-cooled samples, Imilac and
339 Toluca, are compared (Figure 6). Given their similarity in island size, 143 nm and 120 nm
340 respectively, Imilac and Toluca should acquire a remanence via the same mechanism in this
341 region. The cloudy zone in Imilac shows a strong bias in magnetization direction, whereas
342 Toluca shows no such bias. The bias in magnetization direction in Imilac is attributed to
343 cloudy zone formation in the presence of a strong external field (Bryson et al., 2015). The
344 random distribution of magnetization directions in the cloudy zone in Toluca is attributed to
345 formation in a null or very weak external magnetic field (Nichols et al., 2018). The behaviour of
346 the cloudy zone in this region contrasts starkly with that observed further from the tetrataenite
347 rim (e.g. Brenham pallasite in Figure 4, Steinbach IVA in Figure 3), where relatively uniform
348 magnetization is observed, irrespective of the presence or absence of a bias in cloudy zone closer
349 to the rim.

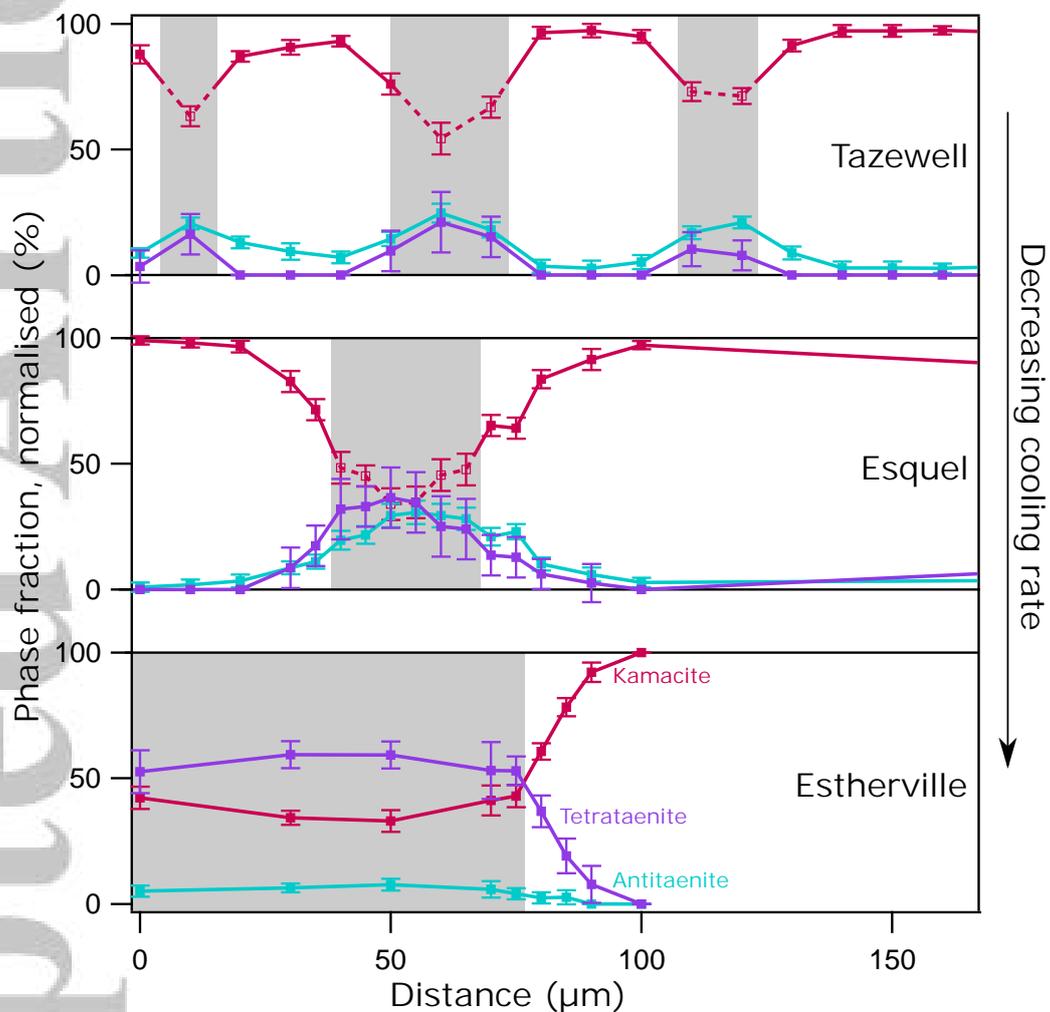


Figure 5: Normalised phase fractions of kamacite, tetrataenite and antitaenite determined from Mössbauer spectra for Tazewell, Esquel and Estherville. Profiles for Tazewell and Esquel were previously published in Blukis et al. (2017). Grey boxes highlight regions of cloudy zone. Dashed lines reflect measurement artifacts; kamacite appears to be present in the cloudy zone of Tazewell and Esquel, however this is only because the beam size is larger than the measured microstructures.

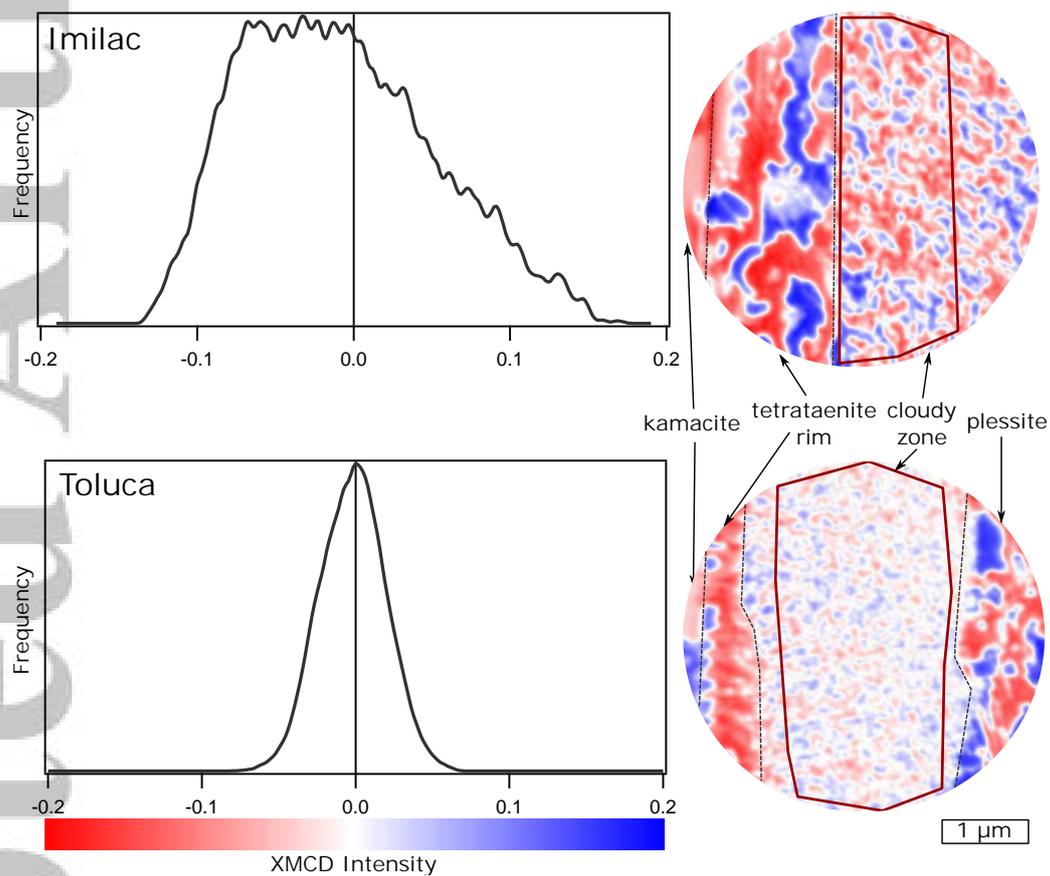


Figure 6: Histograms of the distribution of XMCD pixel intensity in the cloudy zone of Imilac and Toluca. Despite having similar cooling rates and island sizes, these cloudy zones formed under highly contrasting external magnetic fields. Imilac experienced a strong magnetic field shown by the pronounced asymmetric histogram, whereas Toluca experienced a weak or null magnetic field. The regions outlined in dark red were sampled to generate the histograms of pixel intensity.

5 Discussion

5.1 Variations in cloudy zone behaviour with cooling rate

The cloudy zone forms via spinodal decomposition in even the fastest cooled IVA iron meteorites, such as the La Grange IVA iron meteorite which cooled at $6,600\text{ }^{\circ}\text{C Myr}^{-1}$ (Yang et al., 2007, 2008). This suggests that, on meteoritic cooling timescales, spinodal decomposition is always expected to take place to some degree, although the resulting cloudy zone becomes spatially restricted at the fastest cooling rates (Figure 3). Island size decreases with increasing cooling rate (Goldstein et al., 2009a; Yang et al., 2008). We would expect, therefore, to see an increase in the coercivity of the cloudy zone with increasing cooling rate, since the coercivity of tetrataenite has been shown to increase with decreasing lengthscale (Uehara et al., 2011; Bryson et al., 2014a). It is therefore somewhat surprising that Bishop Canyon, the fastest cooled meteorite considered in this study, has a cloudy zone consisting of large regions of uniform magnetization. A study of the hysteresis properties of bulk metal from Bishop Canyon revealed low coercive force, H_c , and remanent coercive force, H_{cr} (Bryson et al., 2017). The measured hysteresis loop was closed at all applied field values, suggesting the bulk metal contains no magnetically hard material. Similar hysteresis properties have been observed in heated samples of Santa Catharina, an ungrouped IAB iron meteorite, and of ordinary chondrites (Dos Santos et al., 2015; Gattacceca et al., 2014). In both these studies, samples were heated in the laboratory to transform tetrataenite islands into taenite via Fe-Ni disordering. This Fe-Ni disordering causes a drastic change in magnetic properties, including a significant decrease in saturation remanence, M_{rs} , H_c and H_{cr} (Gattacceca et al., 2014). Disordering could be caused by shock heating during an impact event since the process only requires short-range (lattice-scale) diffusion and therefore only heating for short periods of time is required, for example complete disordering take < 100 seconds at $550\text{ }^{\circ}\text{C}$ (Goldstein et al., 2009b; Dos Santos et al., 2015). However, the Bristol IVA iron meteorite, which has not experienced significant shock (Yang et al., 2007), has been shown to contain a mixture of ordered and disordered islands using atom probe tomography (Rout et al., 2017). Since Bishop Canyon also only experienced

377 low levels of shock (< 13 GPa) (Yang et al., 2007) we argue that the cloudy zone is comprised
378 of taenite islands because cooling was too rapid for ordering to tetrataenite to occur in the
379 first place. Because of the low temperature of tetrataenite ordering (< 320 °C) kinetics are
380 significantly inhibited and ordering takes at least 10^4 years per atomic jump (Scorzelli, 1997).
381 Given the rapid cooling of Bishop Canyon (2500 ± 1.3 °C Myr⁻¹) cooling from 320 °C to room
382 temperature would take $\sim 120,000$ years allowing insufficient time for complete ordering.

383 The large uniform regions of magnetization observed in the cloudy zone of Bishop Canyon
384 are not observed in the slower cooled IVA iron meteorites, Steinbach and Chinautla. In these
385 samples, small regions of uniform magnetization are observed to cluster around islands in the
386 cloudy zone near the tetrataenite rim. Similarly their cloudy zones also have different hysteresis
387 properties suggesting the presence of tetrataenite (Bryson et al., 2017; Gattacceca et al., 2014).
388 The different magnetic properties observed in the cloudy zone of Bishop Canyon could be caused
389 by the smaller size of the islands, approaching the superparamagnetic threshold of ~ 5 nm (Neel
390 et al., 1964), or insufficient time for tetrataenite ordering. The cloudy zone in Bishop Canyon
391 shows visibly different magnetic behaviour to the fine-aligned cloudy zone in meteorites with
392 intermediate cooling rates, although the islands are comparable in size. We therefore suggest
393 that for cooling rates < 150 °C Myr⁻¹, there is sufficient time for ordering and cloudy zone
394 islands are formed of tetrataenite, while at cooling rates exceeding 2500 °C Myr⁻¹ the islands
395 do not order and remain as taenite. We do not observe superparamagnetic behaviour (in which
396 case the cloudy zone would appear unmagnetized on our observation timescales) because this
397 behaviour is likely to be suppressed by interactions with neighbouring particles (Varón et al.,
398 2013) and surface effects (Blukis et al., 2017). The drastic change in magnetic properties
399 associated with this ordering suggests that the fastest cooled meteorites are not able to record
400 paleomagnetic information; this only occurs if tetrataenite forms (Gattacceca et al., 2014; Einsle
401 et al., 2018). Clustering of magnetization around islands in the cloudy zone is observed in all
402 the samples considered here with cooling rates between $7.6 - 150$ °C Myr⁻¹. We therefore
403 assume that the magnetic properties of the cloudy zone in this range of cooling rates is similar.

404 In the slowest cooled sample considered here, the Estherville mesosiderite, the magneti-

405 zation of the cloudy zone is significantly different. This is most likely due to the large size
406 of the tetrataenite islands (463 nm) that readily permit the formation of magnetic domain
407 walls, resulting in lower coercivities (Uehara et al., 2011; Bryson et al., 2014b). The magnetic
408 behaviour is also likely to be influenced by the fundamentally different chemical composition
409 of the cloudy zone in Estherville (Figure 5). Cooling rates in the mesosiderites are so slow
410 that the cloudy zone undergoes further breakdown; the islands develop Fe-rich interiors and
411 kamacite is observed in the matrix (Yang et al., 1996). We expect the magnetic properties
412 of the cloudy zone to inherently change if the matrix becomes ferromagnetic kamacite, rather
413 than paramagnetic antitaenite which is observed in faster cooled samples (Blukis et al., 2017;
414 Einsle et al., 2018).

415 5.2 Ideal cloudy zone for paleomagnetic study

416 In this section we will define the region of cloudy zone that reliably records a paleomagnetic
417 remanence. This is controlled by both the thermal history and proximity to other microstruc-
418 tures. Based on the meteorites reviewed in this study, cloudy zone with ideal paleomagnetic
419 properties forms at cooling rates between $7.6 - 150 \text{ }^\circ\text{C Myr}^{-1}$, bounded at the upper end by
420 the rate needed to form tetrataenite and at the lower end by the need to avoid multi-domain is-
421 lands. A recent study of the Tazewell IAB iron meteorite (cooling rate $20.8 \text{ }^\circ\text{C Myr}^{-1}$) combined
422 high-resolution microscopy and micromagnetic simulations to demonstrate how paleomagnetic
423 signals are acquired by the cloudy zone (Einsle et al., 2018). Islands in the coarse and mid
424 cloudy zone form as single-vortex taenite. Upon ordering to tetrataenite at temperatures below
425 $320 \text{ }^\circ\text{C}$ the vortex transforms to a transient two-domain state, with a discrete 180° domain
426 wall separating two oppositely magnetized domains. This transformation is attributed to the
427 increase in uniaxial anisotropy induced by Fe-Ni ordering. Strong magnetostatic interaction
428 between neighbouring islands causes the domain wall to displace until it reaches the edge of
429 the island, where it denucleates permanently. The interaction-driven domain-state transition
430 from a two-domain to single-domain leaves each island uniformly magnetized either parallel
431 or antiparallel to its crystallographically defined easy axis (dictated by the crystallographic

432 orientation of Fe-Ni ordering). In the absence of an applied magnetic field, the distribution
433 of interaction fields amongst the closely packed islands is random and centred around zero.
434 Random magnetostatic interactions will lead to equal probability of domain walls displacing
435 one way or the other, resulting in a cloudy zone of single-domain islands magnetized randomly
436 along each of the six possible easy axis directions (e.g., Figure 6b). An applied field, however,
437 biases the interaction field distribution, which is no longer centered around zero, leading to a
438 cloudy zone of single-domain islands that are preferentially magnetized along whichever of the
439 six possible easy axes is closest to the applied field (e.g., Figure 6a). In this model, the presence
440 of the vortex state is the defining characteristic of cloudy zone suitable for paleomagnetic study;
441 it provides a nucleation point for the two-domain state, which in turn provides the mechanism
442 by which the magnetization state of an island can be influenced by an applied field.

443 For spherical taenite particles at 320 °C the single-domain to vortex transition size is 20 –
444 25 nm (Einsle et al., 2018). Therefore, any taenite islands that are smaller than this thresh-
445 old will adopt single-domain states rather than vortex states, with consequences for both the
446 mechanism of remanence acquisition and the strength of magnetostatic interactions between
447 the islands. A critical observation here is that typical ‘coarse cloudy zone’ behaviour is re-
448 stricted to a very narrow region immediately next to the tetrataenite rim in both the Steinbach
449 and Chinautla IVA meteorites (Figure 3 and Bryson et al. (2017)). These meteorites have
450 island diameters adjacent to the tetrataenite rim of 29 ± 3 nm and 32 ± 4.5 nm, respectively
451 (Goldstein et al., 2009b), which are both just above the threshold size for vortex behaviour.
452 This observation supports the hypothesis that the adoption of vortex states is a prerequisite for
453 generating ‘coarse cloudy zone’ behaviour of the type that is able to respond to the presence or
454 absence of a paleofield in the manner displayed in Figure 6. Islands just a short distance away
455 from the tetrataenite rim in the Steinbach and Chinautla meteorites fall below the 20 – 25 nm
456 threshold and would have adopted single-domain states.

457 We observe such variations in the magnetization state of the cloudy zone with distance
458 from the tetrataenite rim in all meteorites with cooling rates between 7.6 – 150 °C Myr⁻¹.
459 The coarse cloudy zone, immediately adjacent to the tetrataenite rim, exhibits clustering of

460 uniform magnetization around the tetrataenite islands. The fine cloudy zone, furthest from the
461 tetrataenite rim exhibits strikingly different behaviour; large patches are uniformly magnetized
462 and it is referred to as the ‘fine-aligned’ cloudy zone (Harrison et al., 2017). We propose
463 that the absence of precursor vortex states in the fine cloudy zone leads to both the increased
464 magnetostatic interactions and the lack of a mechanism to enable the magnetization state to
465 be influence by the applied field. The result is a high degree of spontaneous magnetization that
466 no longer carries useful information about the paleofield.

467 As Ni content decreases away from the tetrataenite rim, the spinodal initiation temperature
468 also decreases (Maurel et al., 2019). An important transition may occur when the spinodal
469 initiation temperature drops below 320 °C, at which point islands will form as tetrataenite
470 rather than taenite. According to current knowledge of the Fe-Ni phase diagram, this point is
471 reached when the Ni composition is bewteen $\sim 34 - 41$ wt% Ni (Maurel et al., 2019; Yang et al.,
472 1997). The uniaxial anisotropy of tetrataenite means that two-domain states or single-domain
473 states, rather than vortex states, are formed when islands grow, with the single-domain to two-
474 domain threshold size being 50 – 55 nm, roughly twice that the taenite single-domain to vortex
475 threshold (Einsle et al., 2018). Therefore, islands that grow as tetrataenite are more likely to
476 be single domain than equivalently sized taenite particles. Therefore, the limit of the reliable
477 paleomagnetic remanence acquisition is defined either by the condition $\text{Ni} > 34 - 40$ wt% and
478 island sizes $< 20 - 25$ nm, or $\text{Ni} < 34 - 40$ wt% and island sizes $< 50 - 55$ nm, whichever is
479 reached sooner.

480 Magnetostatic interactions do not have a significant influence on cloudy zone suitable for
481 paleomagnetic study. This is supported by the fact that cloudy zone which forms in the absence
482 of an external magnetic field shows a random distribution of magnetization directions (Figure
483 6).

484 6 Conclusions

485 The formation of Fe-Ni microstructures and their corresponding magnetic properties have been
486 reviewed for four groups of meteorites spanning a large range of cooling rates; the mesosiderites,
487 the IAB iron meteorites, the pallasites and the IVA iron meteorites. We identify the optimal
488 conditions that lead to the generation of cloudy zone with the best potential to acquire nanopar-
489 leomagnetic remanence:

- 490 • Islands ideally form within the taenite stability field at $T > 320$ °C and have sizes $> 20 -$
491 25 nm, such that they are able to adopt a precursor single-vortex micromagnetic state.
- 492 • Island sizes decrease with increasing distance from the tetrataenite rim, so that only
493 islands that are sufficiently close to the rim are above the $20 - 25$ nm threshold.
- 494 • Spinodal decomposition occurs at lower temperatures with increasing distance from the
495 tetrataenite rim, so that only islands that are sufficiently close to the rim will have formed
496 above 320 °C.
- 497 • Cooling rates must be slow enough ($\lesssim 150 - 2500$ °C Myr⁻¹) to allow islands to trans-
498 form to tetrataenite on cooling below 320 °C, thereby inducing a domain state transition
499 from vortex to two-domain to single-domain. This mechanism enables the cloudy zone
500 magnetization to respond to the paleofield, creating a bias that can be detected using
501 X-PEEM.
- 502 • Cooling rates must be fast enough ($\gtrsim 1$ °C Myr⁻¹) to prevent islands growing too large
503 and creating multi-domain behaviour.
- 504 • Mesosiderites (cooling rate $\lesssim 0.5 - 3.3$ °C Myr⁻¹): Cloudy zone is multidomain and does
505 not acquire a stable paleomagnetic remanence, possibly due to large island size or the
506 composition and mineralogy of the matrix.
- 507 • Pallasites and IAB iron meteorites (cooling rate $\sim 1 - 100$ °C Myr⁻¹): Cloudy zone is
508 a reliable paleomagnetic recorder (Bryson et al., 2015; Nichols et al., 2016, 2018) and

509 remanence is acquired during tetrataenite formation. Ordering occurs simultaneously
510 across the cloudy zone, therefore no time-resolved paleomagnetic signals are preserved.

511 • Intermediate cooled IVA iron meteorites (cooling rate $100 - 500 \text{ }^\circ\text{C Myr}^{-1}$): Only regions
512 of cloudy zone very close to the tetrataenite rim are large enough to form precursor vortex
513 states (Bryson et al., 2017).

514 • Fastest cooled IVA iron meteorites (cooling rate $> 150 - 2500 \text{ }^\circ\text{C Myr}^{-1}$): Cloudy zone
515 forms too quickly for the islands to order to form tetrataenite.

516 7 Acknowledgements

517 We thank Jérôme Gattacceca and an anonymous reviewer for helpful comments that improved
518 the manuscript. This work was supported by the European Research Council under the Eu-
519 ropean Union's Seventh Framework Programme (FP/2007-2013)/ ERC grant agreement num-
520 bers 320750 and 312284. We acknowledge the Helmholtz-Zentrum Berlin for the use of the
521 synchrotron radiation beam time at beamline UE49 of BESSY II. We acknowledge the Euro-
522 pean Synchrotron Radiation Facility in Grenoble, France for use of synchrotron radiation beam
523 time at the Nuclear Resonance beamline ID18. We thank the Sedgwick Museum, University
524 of Cambridge and the Natural History Museum, London for samples. All data associated with
525 this study are available on zenodo (<https://doi.org/10.5281/zenodo.3613625>).

526 References

- 527 G. K. Benedix, T. J. McCoy, K. Keil, and S. G. Love. A petrologic study of the IAB iron mete-
528 orites: Constraints on the formation of the IAB-Winonaite parent body. *Meteoritics & Plan-*
529 *etary Science*, 35:1127–1141, 2000. ISSN 10869379. doi: 10.1111/j.1945-5100.2000.tb01502.x.
- 530 G.K. Benedix, H. Haack, and T.J. McCoy. Iron and Stony-Iron Meteorites. *Trea-*
531 *tise on Geochemistry*, 1:267–285, 2014. doi: 10.1016/B978-0-08-095975-7.00109-1. URL
532 <http://linkinghub.elsevier.com/retrieve/pii/B9780080959757001091>.
- 533 Roberts Blukis, Rudolf Ruffer, Aleksandr I. Chumakov, and Richard J. Harrison. A high
534 spatial resolution synchrotron Mössbauer study of the Tazewell IIICD and Esquel pallasite
535 meteorites. *Meteoritics & Planetary Science*, 52(5):925–936, 2017. ISSN 10869379. doi:
536 10.1111/maps.12841.
- 537 Donald D. Bogard and Daniel H. Garrison. ^{39}Ar - ^{40}Ar ages and thermal history of
538 mesosiderites. *Geochimica et Cosmochimica Acta*, 62(8):1459–1468, 1998.
- 539 James F J Bryson, Claire I O Nichols, Julia Herrero-Albillos, Florian Kronast, Takeshi
540 Kasama, Hossein Alimadadi, Gerrit van der Laan, Francis Nimmo, and Richard J Har-
541 rison. Long-lived magnetism from solidification-driven convection on the pallasite parent
542 body. *Nature*, 517(7535):472–475, 2015. ISSN 0028-0836. doi: 10.1038/nature14114. URL
543 <http://dx.doi.org/10.1038/nature14114>.
- 544 James F.J. Bryson, Nathan S. Church, Takeshi Kasama, and Richard J. Harrison.
545 Nanomagnetic intergrowths in FeNi meteoritic metal. The potential for time resolved
546 records of planetesimal dynamo fields. *Earth and Planetary Science Letters*, 388:
547 237–248, feb 2014a. ISSN 0012821X. doi: 10.1016/j.epsl.2013.12.004. URL
548 <http://linkinghub.elsevier.com/retrieve/pii/S0012821X13007012>.
- 549 James F.J. Bryson, Julia Herrero-Albillos, Florian Kronast, Massimo Ghidini, Simon A.T.
550 Redfern, Gerrit van der Laan, and Richard J. Harrison. Nanopaleomagnetism of mete-
551 oritic FeNi studied using Xray photoemission electron microscopy. *Earth and Planetary*
552 *Science Letters*, 396:125–133, 2014b. ISSN 0012821X. doi: 10.1016/j.epsl.2014.04.016. URL
553 <http://www.sciencedirect.com/science/article/pii/S0012821X14002489>.
- 554 James F.J. Bryson, Benjamin P. Weiss, Richard J. Harrison, Julia Herrero-Albillos, and Florian
555 Kronast. Paleomagnetic evidence for dynamo activity driven by inward crystallisation of a
556 metallic asteroid. *Earth and Planetary Science Letters*, 472:152–163, 2017. ISSN 0012821X.
557 doi: 10.1016/j.epsl.2017.05.026. URL <http://dx.doi.org/10.1016/j.epsl.2017.05.026>.
- 558 Vagn F Buchwald. *Handbook of Iron Meteorites*. University of California Press, 1975.
- 559 J Danon, Rosa Bernstein Scorzelli, I Souza Azevedo, W Curvello, JF Albertsen, and JM Knud-
560 sen. Iron-nickel 50-50 superstructure in the Santa Catharina meteorite. *Nature*, 277(January):
561 10–11, 1979.
- 562 Jeremy S. Delaney, C E Nehru, and M Prinz. Olivine clasts from mesosiderites and howardites:
563 Clues to the nature of achondritic parent bodies. *Lunar and Planetary Science Conference*,
564 11:1073–1087, 1980.

- 565 E. Dos Santos, J. Gattacceca, P. Rochette, G. Fillion, and R. B. Scorzelli. Ki-
566 netics of tetrataenite disordering. *Journal of Magnetism and Magnetic Materi-*
567 *als*, 375:234–241, 2015. ISSN 03048853. doi: 10.1016/j.jmmm.2014.09.051. URL
568 <http://dx.doi.org/10.1016/j.jmmm.2014.09.051>.
- 569 Joshua F. Einsle, Alexander S. Eggeman, Benjamin H. Martineau, Zineb Saghi, Sean M. Collins,
570 Roberts Blukis, Paul A. J. Bagot, Paul A. Midgley, and Richard J. Harrison. Nanopaleo-
571 magnetic properties of the meteorite cloudy zone. *Proceedings of the National Academy*
572 *of Sciences*, XXX(Xx), 2018. ISSN 0027-8424. doi: 10.1073/pnas.XXXXXXXXXX. URL
573 <http://arxiv.org/abs/1803.11502>.
- 574 Jibamitra Ganguly, H Yang, and S Ghose. Thermal history of mesosiderites: Quantitative
575 constraints from compositional zoning and Fe-Mg ordering in orthopyroxenes. *Geochimica et*
576 *Cosmochimica Acta*, 58(12):2711–2723, 1994.
- 577 J. Gattacceca, C. Suavet, P. Rochette, B. P. Weiss, M. Winklhofer, M. Uehara, and Jon M.
578 Friedrich. Metal phases in ordinary chondrites: Magnetic hysteresis properties and implica-
579 tions for thermal history. *Meteoritics & Planetary Science*, 49(4):652–676, apr 2014. ISSN
580 10869379. doi: 10.1111/maps.12268. URL <http://doi.wiley.com/10.1111/maps.12268>.
- 581 J I Goldstein and J R Michael. The formation of plessite in meteoritic metal. *Meteoritics &*
582 *Planetary Science*, 41(4):553–570, 2006.
- 583 J. I. Goldstein, J. Yang, P. G. Kotula, J. R. Michael, and E. R. D. Scott.
584 Thermal histories of IVA iron meteorites from transmission electron microscopy of
585 the cloudy zone microstructure. *Meteoritics & Planetary Science*, 44(3):343–358,
586 mar 2009a. ISSN 10869379. doi: 10.1111/j.1945-5100.2009.tb00737.x. URL
587 <http://doi.wiley.com/10.1111/j.1945-5100.2009.tb00737.x>.
- 588 J. I. Goldstein, J. Yang, P. G. Kotula, J. R. Michael, and E. R. D. Scott.
589 Thermal histories of IVA iron meteorites from transmission electron microscopy of
590 the cloudy zone microstructure. *Meteoritics & Planetary Science*, 44(3):343–358,
591 mar 2009b. ISSN 10869379. doi: 10.1111/j.1945-5100.2009.tb00737.x. URL
592 <http://doi.wiley.com/10.1111/j.1945-5100.2009.tb00737.x>.
- 593 Joseph I. Goldstein, Jijin Yang, and Edward R.D. Scott. Determining cool-
594 ing rates of iron and stony-iron meteorites from measurements of Ni and
595 Co at kamacite taenite interfaces. *Geochimica et Cosmochimica Acta*, 140:
596 297–320, 2014. ISSN 00167037. doi: 10.1016/j.gca.2014.05.025. URL
597 <http://linkinghub.elsevier.com/retrieve/pii/S0016703714003548>.
- 598 Joseph I. Goldstein, Gary R. Huss, and Edward R.D. Scott. Ion microprobe analyses of
599 carbon in Fe–Ni metal in iron meteorites and mesosiderites. *Geochimica et Cosmochim-*
600 *ica Acta*, 200:367–407, 2017. ISSN 00167037. doi: 10.1016/j.gca.2016.12.027. URL
601 <http://dx.doi.org/10.1016/j.gca.2016.12.027>.
- 602 Richard C. Greenwood, Jean Alix Barrat, Edward R.D. Scott, Henning Haack, Paul C.
603 Buchanan, Ian A. Franchi, Akira Yamaguchi, Diane Johnson, Alex W.R. Bevan,
604 and Thomas H. Burbine. Geochemistry and oxygen isotope composition of main-
605 group pallasites and olivine-rich clasts in mesosiderites: Implications for the “Great

- 606 Dunitite Shortage" and HED-mesosiderite connection. *Geochimica et Cosmochimica*
607 *Acta*, 169:115–136, 2015. ISSN 00167037. doi: 10.1016/j.gca.2015.07.023. URL
608 <http://dx.doi.org/10.1016/j.gca.2015.07.023>.
- 609 R. J. Harrison, James F J Bryson, C. I. O. Nichols, and B.P. Weiss. Magnetic Mineralogy
610 of Meteoritic Metal : Paleomagnetic Evidence for Dynamo Activity on Differentiated
611 Planetesimals. In Linda T Elkins-Tanton and Benjamin P. Weiss, editors, *Planetesimals:
612 Early Differentiation and Consequences for Planets*, chapter 10, pages 204–223. Cam-
613 bridge University Press, 2017. ISBN 9781316339794. doi: 10.1017/9781316339794. URL
614 <https://www.cambridge.org/core/books/planetesimals/BAB5DC62F7B5911C96E0DA6C25419CBB>.
- 615 K. Lagarec, D. G. Rancourt, S. K. Bose, B. Sanyal, and R. A. Dunlap. Observation of a
616 composition-controlled high-moment/low-moment transition in the face centered cubic Fe-Ni
617 system: Invar effect is an expansion, not a contraction. *Journal of Magnetism and Magnetic*
618 *Materials*, 236(1-2):107–130, 2001. ISSN 03048853. doi: 10.1016/S0304-8853(01)00449-8.
- 619 Clara Maurel, Benjamin P. Weiss, and James F.J. Bryson. Meteorite cloudy zone forma-
620 tion as a quantitative indicator of paleomagnetic field intensities and cooling rates on plan-
621 etesimals. *Earth and Planetary Science Letters*, 513:166–175, 2019. ISSN 0012821X. doi:
622 10.1016/j.epsl.2019.02.027. URL <https://doi.org/10.1016/j.epsl.2019.02.027>.
- 623 David W. Mittlefehldt. The composition of mesosiderite olivine clasts and implications for the
624 origin of pallasites. *Earth and Planetary Science Letters*, 51:29–40, 1980. ISSN 0012821X.
625 doi: 10.1016/0012-821X(80)90254-X.
- 626 L. Neel, J. Pauleve, R. Pauthenet, J. Laugier, and D. Dautreppe. Magnetic Proper-
627 ties of an Iron-Nickel Single Crystal Ordered by Neutron Bombardment. *Journal of*
628 *Applied Physics*, 35(3):873, 1964. ISSN 00218979. doi: 10.1063/1.1713516. URL
629 <http://link.aip.org/link/JAPIAU/v35/i3/p873/s1&Agg=doi>.
- 630 Jerome A. Neufeld, James F.J. Bryson, and Francis Nimmo. The Top-Down Solidification of
631 Iron Asteroids Driving Dynamo Evolution. *Journal of Geophysical Research: Planets*, 124
632 (5):1331–1356, 2019. ISSN 21699100. doi: 10.1029/2018JE005900.
- 633 Claire I O Nichols, James F.J. Bryson, Julia Herrero-Albillos, Florian Kronast, Francis Nimmo,
634 and Richard J. Harrison. Pallasite Paleomagnetism: Quiescence of a Core Dynamo. *Earth*
635 *and Planetary Science Letters*, 441:103–112, 2016.
- 636 Claire I.O. Nichols, BL Getzin, Benjamin Paul Weiss, R. A. Wells, and H. H. Schmitt. The Pale-
637 oinclination and Paleointensity of the Early Lunar Dynamo. In *AGU Fall Meeting Abstracts*,
638 2018.
- 639 H. Ohldag, T. Regan, J. Stöhr, a. Scholl, F. Nolting, J. Lüning, C. Stamm, S. Anders, and
640 R. White. Spectroscopic Identification and Direct Imaging of Interfacial Magnetic Spins.
641 *Physical Review Letters*, 87(24):247201, nov 2001. ISSN 0031-9007. doi: 10.1103/Phys-
642 RevLett.87.247201. URL <http://link.aps.org/doi/10.1103/PhysRevLett.87.247201>.
- 643 Vasily Potapkin, Aleksandr I. Chumakov, Gennadii V. Smirnov, Jean Philippe Celse, Rudolf
644 Ruffer, Catherine McCammon, and Leonid Dubrovinsky. The 57 Fe Synchrotron Mössbauer

- 645 Source at the ESRF. *Journal of Synchrotron Radiation*, 19(4):559–569, 2012. ISSN 09090495.
646 doi: 10.1107/S0909049512015579.
- 647 D G Rancourt. Low-spin Fe-Ni proposed as a new mineral. *Journal of Magnetism and Magnetic*
648 *Materials*, 150(95), 1995.
- 649 D. G. Rancourt, K. Lagarec, A. Densmore, R. A. Dunlap, J. I. Goldstein, R. J. Reisener, and
650 R. B. Scorzelli. Experimental proof of the distinct electronic structure of a new meteoritic Fe
651 Ni alloy phase. *Journal of Magnetism and Magnetic Materials*, 191(3):255–260, 1999. ISSN
652 03048853. doi: 10.1016/S0304-8853(98)00366-7.
- 653 Surya S. Rout, Philipp R. Heck, Dieter Isheim, Thomas Stephan, Nestor J. Zaluzec, Dean J.
654 Miller, Andrew M. Davis, and David N. Seidman. Atom-probe tomography and transmission
655 electron microscopy of the kamacite–taenite interface in the fast-cooled Bristol IVA iron
656 meteorite. *Meteoritics and Planetary Science*, 52(12):2707–2729, 2017. ISSN 10869379. doi:
657 10.1111/maps.12988.
- 658 Rudolf Ruffer and Aleksandr I. Chumakov. Nuclear Resonance Beamline at ESRF. *Hyperfine*
659 *Interactions*, 97-98(1):589–604, 1996. ISSN 0304-3834. doi: 10.1007/BF02150199. URL
660 <http://link.springer.com/10.1007/BF02150199>.
- 661 Alex Ruzicka, William V. Boynton, and Jibamitra Ganguly. Olivine coronas, metamorphism,
662 and the thermal history of the Morristown and Emery mesosiderites. *Geochimica et Cos-*
663 *mochimica Acta*, 58(12):2725–2741, 1994. ISSN 00167037. doi: 10.1016/0016-7037(94)90140-
664 6.
- 665 T. Schulz, D. Upadhyay, C. Münker, and K. Mezger. Formation and exposure history of non-
666 magmatic iron meteorites and winonaites: Clues from Sm and W isotopes. *Geochimica et*
667 *Cosmochimica Acta*, 85:200–212, 2012. ISSN 00167037. doi: 10.1016/j.gca.2012.02.012.
- 668 RB Scorzelli. A study of phase stability in invar Fe–Ni alloys obtained by
669 non-conventional methods. *Hyperfine interactions*, 110:143–150, 1997. URL
670 <http://link.springer.com/article/10.1023/A:1012679517295>.
- 671 Edward R. D. Scott, Henning Haack, and Stanley G. Love. Formation of Mesosiderites: Frag-
672 mentation and Reaccretion of a Large Differentiated Asteroid. *Meteoritics and Planetary*
673 *Science*, 36:869–881, 2001. ISSN 10869379. doi: 10.1111/j.1945-5100.2001.tb01927.x.
- 674 G.V. Smirnov, U. van Bürc, A.I. Chumakov, A.Q.R. Baron, and R. Ruffer. Synchrotron Möss-
675 bauer Source. *Physical Review B*, 55(9):5811–5815, 1997. ISSN 01631829. doi: 10.1103/Phys-
676 RevB.55.5811.
- 677 J. Stöhr, H. A. Padmore, S. Anders, T. Stammler, and M. R. Scheinfein. Principles of x-ray
678 magnetic dichroism spectromicroscopy. *Surface review and letters*, 5(6):1297–1308, 1998.
- 679 John a Tarduno, Rory D Cottrell, Francis Nimmo, Julianna Hopkins, Julia Voronov, Austen
680 Erickson, Eric Blackman, Edward R D Scott, and Robert McKinley. Evidence for a dynamo in
681 the main group pallasite parent body. *Science*, 338(6109):939–942, nov 2012. ISSN 1095-9203.
682 doi: 10.1126/science.1223932. URL <http://www.ncbi.nlm.nih.gov/pubmed/23161997>.

- 683 Minoru Uehara, Jérôme Gattacceca, Hugues Leroux, Damien Jacob, and Cornelis Jacominus
684 van der Beek. Magnetic microstructures of metal grains in equilibrated ordinary chon-
685 drites and implications for paleomagnetism of meteorites. *Earth and Planetary Science*
686 *Letters*, 306:241–252, jun 2011. ISSN 0012821X. doi: 10.1016/j.epsl.2011.04.008. URL
687 <http://linkinghub.elsevier.com/retrieve/pii/S0012821X11002147>.
- 688 M. Varón, M. Beleggia, T. Kasama, R. J. Harrison, R. E. Dunin-Borkowski, V. F. Puentes, and
689 C. Frandsen. Dipolar magnetism in ordered and disordered low-dimensional nanoparticle
690 assemblies. *Scientific Reports*, 3:3–7, 2013. ISSN 20452322. doi: 10.1038/srep01234.
- 691 John T. Wasson and Hoppe. Co/Ni double ratios in mesosiderite metal and the unrealistically
692 low cooling rates. *77th Annual Meteoritical Society Meeting*, (5405):15–16, 2014. ISSN 1086-
693 9379. doi: 10.1016/j.molstruc.2014.05.049.
- 694 T.B. Winfield, J.I. Goldstein, and E.R.D. Scott. Cooling rate estimates for IAB and IIICD iron
695 meteorites. *43rd Lunar and Planetary Science Conference*, (1307):43–44, 2012.
- 696 C Yang, D Williams, and J Goldstein. A revision of the Fe-Ni Phase Diagram at Low Temper-
697 atures (<400 C). *Journal of Phase Equilibria*, 17(6):522–531, 1996.
- 698 C.W. Yang, D.B. Williams, and J.I. Goldstein. A new empirical cooling rate indi-
699 cator for meteorites based on the size of the cloudy zone of the metallic phases.
700 *Meteoritics & Planetary Science*, 429:423–429, 1997. ISSN 10869379. URL
701 <http://onlinelibrary.wiley.com/doi/10.1111/j.1945-5100.1997.tb01285.x/abstract>.
- 702 Jijin Yang, Joseph I Goldstein, and Edward R D Scott. Iron meteorite evi-
703 dence for early formation and catastrophic disruption of protoplanets. *Nature*, 446
704 (7138):888–91, apr 2007. ISSN 1476-4687. doi: 10.1038/nature05735. URL
705 <http://www.ncbi.nlm.nih.gov/pubmed/17443181>.
- 706 Jijin Yang, Joseph I. Goldstein, and Edward R.D. Scott. Metallographic cool-
707 ing and origin of IVA iron meteorites. *Geochimica et Cosmochimica Acta*, 70
708 (12):3197–3215, 2008. ISSN 00167037. doi: 10.1016/j.gca.2006.04.007. URL
709 <http://www.sciencedirect.com/science/article/pii/S0016703706001888%5Cnhttp://linkinghub>
- 710 Jijin Yang, Joseph I. Goldstein, and Edward R.D. Scott. Main-group pallasites: Ther-
711 mal history, relationship to IIIAB irons, and origin. *Geochimica et Cosmochimica Acta*,
712 74(15):4471–4492, aug 2010. ISSN 00167037. doi: 10.1016/j.gca.2010.04.016. URL
713 <http://linkinghub.elsevier.com/retrieve/pii/S0016703710002012>.