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## **To cite this version:**

J. Gattacceca, C. Suavet, P. Rochette, B. Weiss, M. Winklhofer, et al.. Metal phases in ordinary chondrites: Magnetic hysteresis properties and implications for thermal history. Meteoritics and Planetary Science, 2014, 49 (4), pp.652-676. 10.1111/maps.12268 hal-03289394

# **HAL Id: hal-03289394 <https://hal.science/hal-03289394v1>**

Submitted on 18 Jul 2021

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Meteoritics & Planetary Science 49, Nr 4, 652–676 (2014) doi: 10.1111/maps.12268

## Metal phases in ordinary chondrites: Magnetic hysteresis properties and implications for thermal history

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(Received 21 June 2013; revision accepted 19 December 2013)

Abstract–Magnetic properties are sensitive proxies to characterize FeNi metal phases in meteorites. We present a data set of magnetic hysteresis properties of 91 ordinary chondrite falls. We show that hysteresis properties are distinctive of individual meteorites while homogeneous among meteorite subsamples. Except for the most primitive chondrites, these properties can be explained by a mixture of multidomain kamacite that dominates the induced magnetism and tetrataenite (both in the cloudy zone as single-domain grains, and as larger multidomain grains in plessite and in the rim of zoned taenite) dominates the remanent magnetism, in agreement with previous microscopic magnetic observations. The bulk metal contents derived from magnetic measurements are in agreement with those estimated previously from chemical analyses. We evidence a decreasing metal content with increasing petrologic type in ordinary chondrites, compatible with oxidation of metal during thermal metamorphism. Types 5 and 6 ordinary chondrites have higher tetrataenite content than type 4 chondrites. This is compatible with lower cooling rates in the 650–450 °C interval for higher petrographic types (consistent with an onion-shell model), but is more likely the result of the oxidation of ordinary chondrites with increasing metamorphism. In equilibrated chondrites, shock-related transient heating events above approximately 500 °C result in the disordering of tetrataenite and associated drastic change in magnetic properties. As a good indicator of the amount of tetrataenite, hysteresis properties are a very sensitive proxy of the thermal history of ordinary chondrites, revealing low cooling rates during thermal metamorphism and high cooling rates (e.g., following shock reheating or excavation after thermal metamorphism). Our data strengthen the view that the poor magnetic recording properties of multidomain kamacite and the secondary origin of tetrataenite make equilibrated ordinary chondrites challenging targets for paleomagnetic study.

#### INTRODUCTION

Knowledge of the magnetic mineralogy and magnetic properties of extraterrestrial materials is crucial for interpreting the paleomagnetism of meteorites (see Weiss et al. [2010] for a review), for interpreting the magnetic anomalies of planetary surfaces (e.g., Dunlop and Kletetschka 2001; Wasilewski et al. 2002; Arkani-Hamed 2005; Rochette et al. 2005 for Mars; Rochette et al. 2010; Wieczorek et al. 2012 for the Moon). It also sheds light on the redox conditions prevailing during the formation of solid

matter in the solar system (e.g., Rochette et al. 2008), and on its evolution through time. Intrinsic magnetic properties, in particular susceptibility, can be used as a rapid classification tool (Rochette et al. 2003, 2008, 2009), allowing scanning of large meteorite collections for preliminary classification (Folco et al. 2006) or in search of anomalous samples (e.g., Kohout et al. 2010), and with potential application to the characterization of asteroids (Kohout et al. 2008). The anisotropy of magnetic susceptibility has been used to study the compaction history of chondrites (Sneyd et al. 1988; Gattacceca et al. 2005).

Most comprehensive studies about the magnetic properties of meteorites were focused on magnetic susceptibility (e.g., Rochette et al. 2003, 2008, 2009), magnetic anisotropy (Gattacceca et al. 2005, 2008), and thermomagnetic behavior (e.g., Lovering and Parry 1962; Herndon et al. 1976; Nagata and Funaki 1982; Momose and Nagai 1983). In contrast, little is known about the magnetic hysteresis properties of meteorites, particularly for metal-bearing stony meteorites. Hysteresis properties are sensitive proxies of grain size (e.g., Peters and Dekkers 2003), the presence of defects in the crystal structure (e.g., Ozdemir and Dunlop 1997), and the bulk abundances of magnetic minerals. In particular, hysteresis properties are very sensitive to the microstructures of metal grains and may thus be a good proxy of the thermal history of meteorites (e.g., Leroux et al. 2000).

Hysteresis properties of 22 meteorites, including 14 ordinary chondrites, were measured by Sugiura (1977), but most of these are Antarctic finds whose magnetic properties are likely to be affected by terrestrial weathering (Uehara et al. 2012). The only significant data set of hysteresis properties for meteorite falls is the report by Terho et al. (1992) that provides hysteresis parameters for 26 ordinary chondrite falls. However, no detailed interpretation is given in this report and a subsequent publication (Pesonen et al. 1993).

In this study, we present magnetic hysteresis properties of 91 ordinary chondrite falls. Data for 10 HEDs (howardites, eucrites, diogenites) achondrites are also provided for comparison. This data set, based mainly on new measurements, is the basis for a discussion on the magnetic mineralogy of ordinary chondrites, its relation with the thermal and shock history of meteorites, and the implications for paleomagnetism.

## SAMPLES AND METHODS

## Samples

Only meteorite falls were studied to avoid biases due to terrestrial weathering and the associated

formation of ferric phases (in particular, maghemite) in meteorites finds (Kohout et al. 2004; Bland et al. 2006). Weathering has been shown to drastically modify the hysteresis properties of ordinary chondrites even at its incipient stages (Uehara et al. 2012). In this study, we increase the hysteresis properties database significantly with new measurements on 109 ordinary chondrite samples representing 70 different meteorites. Sample selection was made to span the three ordinary chondrite classes (H, L, and LL), as well as a few chondrites from the intermediate classes H/L and L/LL (Kallemeyn et al. 1989). In each class, we selected samples spanning the whole spectrum of thermal metamorphism, from type 3 to type 6 (as defined by Van Schmus and Wood 1967). For the L class, we also selected samples spanning all six levels of shock, from stages S1 to S6 (as defined by Stöffler et al. 1991). For all three classes, meteorites of metamorphic type 3 are overrepresented in our database with respect to the average population, but types 4 to 6 are represented approximately correctly relative to one another. For L chondrites, heavily shocked meteorites of shock stage S4 and above are also overrepresented. For comparison with meteorites containing FeNi minerals but with completely different petrogenetic and thermal histories, 19 HED samples were also studied. Studied samples range from 20 mg to 3 g in mass. Most samples are from the collection of the Museum National d'Histoire Naturelle (MNHN, Paris, France).

## Magnetic Measurements

Readers not familiar with the measurement and meaning of rock magnetic parameters may refer to Collinson (1983) and Dunlop and Ozdemir (1997) for in-depth explanations. Magnetic hysteresis properties were measured with a Princeton Micromag vibrating sample magnetometer (VSM) with a noise level of about 1 nAm<sup>2</sup> and a maximum applied field of 1 T. Hysteresis loops allow the determination of coercivity  $(B_C)$ , saturation magnetization  $(M<sub>S</sub>)$ , saturation remanent magnetization  $(M_{RS})$ , high-field susceptibility  $(\chi_{HF},$ including both diamagnetic and paramagnetic contributions). Coercivity of remanence  $(B_{CR})$  was evaluated through DC back-field demagnetization of the saturation remanence. Saturation remanent magnetization was also measured with a 2G DC SQUID magnetometer model 755R (noise level  $10^{-11}$  Am<sup>2</sup>) after saturation in a 1 T field using a pulse magnetizer from Magnetic Measurements. The  $M_{RS}$ value measured this way is more accurate than that obtained with the VSM (whose measurements are more sensitive to the geometry of the sample) and was used to recalculate the  $M_{\rm S}$  through the  $M_{\rm RS}/M_{\rm S}$  ratio

provided by the VSM. First-order reversal curves (FORC) were acquired using the same VSM using a field sampling interval of 10 mT. FORC data were processed with a smoothing factor of three using UNIFORC software (Egli et al. 2010). Mass normalized low-field magnetic susceptibility  $\gamma$  of the same samples was measured in an AC field of 200 A  $m^{-1}$  (peak field) and frequency 976 Hz with a MFK1 apparatus from Agico (sensitivity  $5 \times 10^{-13}$  m<sup>3</sup>). Magnetic susceptibility was measured along three orthogonal directions and averaged to account for the variability introduced by the often strong magnetic anisotropy of FeNi-bearing meteorites (Gattacceca et al. 2005). The ferromagnetic susceptibility was then computed as  $\chi_f = \chi - \chi_{HF}$ . All magnetic measurements were performed at CEREGE (Aix-en-Provence, France). Previous hysteresis data (Nagata and Sugiura 1976; Sugiura 1977; Nagata et al. 1986; Terho et al. 1992) were included in our database only for meteorites that were not measured in our study, amounting to 21 meteorites.

Because grain shape can affect some magnetic properties (like apparent magnetic susceptibility), X-ray microtomography was used to evaluate the shape of metal grains in some ordinary chondrites. Data were collected at resolutions of  $13-19 \mu m/voxel$  with monochromatic X-rays using the 13-BMD beamline located at the Advanced Photon Source (APS) of Argonne National Laboratory. These resolutions are known to be adequate for observing the morphology and size distributions of metal and sulfide grains in ordinary and other chondrites (Friedrich et al. 2008). Details of the data collection can be found in Friedrich et al. (2008, 2013). Three-dimensional examination of ordinary chondrite metal grain shapes was accomplished digitally isolating them in images and computing best fitting ellipsoids following Friedrich et al. (2008). Metal grain shapes were inferred from major, intermediate, and short ellipsoid axes using the Zingg (1935) parameters and nomenclature.

## Anisotropy Effects

Anisotropy of magnetic susceptibility in ordinary chondrites is strong, ubiquitous, and controlled mostly by the preferential alignment of metallic grains (Gattacceca et al. 2005). Remanence anisotropy can also be strong in ordinary chondrites because of the crystalline anisotropy of tetrataenite (e.g., Gattacceca et al. 2003). Therefore, measurement of hysteresis properties along a single direction may result in a significant departure from the anisotropy-averaged value.

Measurements on eight 0.4 g cubic samples of the Bensour LL6 chondrite reveal an average anisotropy

degree (ratio of maximum to minimum value) of  $1.28 \pm 0.08$  for  $M_{\text{RS}}$ . In such a meteorite, measurement of  $M_{RS}$  along a single direction may result in a maximum departure of  $\pm 12\%$  from the anisotropyaveraged value. Measurements on 0.5 g cubic samples of Pultusk H4 chondrite give an anisotropy degree of 1.18 for  $M_{RS}$ , indicating a maximum departure of  $\pm 8\%$ from the average value.  $B_{CR}$  values for the same Pultusk samples differ by up to 20% from the average value. Therefore, the effects of magnetic anisotropy can significantly bias  $M_{RS}$  and  $B_{CR}$  (up to approximately 10 and approximately 20%, respectively) if hysteresis is measured along a single random axis. This is especially true for LL chondrites that have stronger remanence anisotropy than L and H chondrites because of the higher relative abundance of tetrataenite. However, measurements of several randomly oriented samples help to minimize this bias, an approach that has been used for most LL chondrites studied in this study with an average of three samples (and up to 15 samples) measured per meteorite.

## Approach to Saturation

In most previous studies that provided hysteresis measurements on FeNi-bearing meteorites (or lunar rocks), saturation magnetization was computed after subtracting the contribution of paramagnetism and diamagnetism estimated by a linear fit of the reversible part of the hysteresis loop (generally in the 0.8–1 T or 0.9–1 T interval to take into account the nonsaturation of FeNi multidomain grains below approximately 0.7 T). However, in the reversible part of a hysteresis loop, the magnetization  $M$  does not increase strictly linearly with applied field  $B$ , but is better expressed as  $\widehat{M} = M_{\rm S} + \chi_{\rm HF} B + \alpha B^{\beta}$  (e.g., Fabian 2006).  $\alpha$  and  $\beta$ are negative parameters that are mineral dependent. In most cases, the term  $\alpha B^{\beta}$  is negligible, and  $\chi$ <sub>HF</sub> and  $M_{\rm S}$ are well approximated by a linear fit of the reversible part of the hysteresis loops. However, as shown below, this approximation does not hold for ordinary chondrites and leads to underestimation of  $M<sub>S</sub>$  and overestimation of  $\gamma$ <sub>HF</sub>. Instead, we used the approach to saturation method to estimate  $M<sub>S</sub>$  and  $\chi$ <sub>HF</sub>.  $\alpha$  was determined for each meteorite, so as to minimize the residual of the linear regression of  $M - \alpha B^{\beta}$  where we assumed  $\beta = -2$ , the theoretical and observed value for defect free minerals (Fabian 2006).  $M<sub>S</sub>$  and  $\chi$ <sub>HF</sub> are the intercept and slope of the regression, respectively. The regressions were computed on the  $|B| > 0.8$  T portions for all four branches of the hysteresis cycle. This algorithm was tested on a hysteresis loop measured at room temperature on a sample made of 76 wt% of gadolinium oxide (70 mg) of  $Gd_2O_3$  from Aldrich with a theoretical paramagnetic susceptibility of  $1.85 \times 10^{-6}$  m<sup>3</sup> kg<sup>-1</sup>; see Sagnotti et al. 2003) and 24 wt% metallic iron (10  $\mu$ m synthetic powder in multidomain state from Merck), with a theoretical  $M<sub>S</sub> = 218$  Am<sup>2</sup> kg<sup>-1</sup>, a proportion roughly similar to that found in H ordinary chondrites. The algorithm gave a reliable value for  $M<sub>S</sub>$  with a departure from the theoretical value of only 0.6%, whereas the classic linear fit underestimates the theoretical value by 17%.  $\chi$ <sub>HF</sub> is overestimated by a factor of 2.2 when using the linear fit, and is underestimated by a factor of 3.7 when using approach to saturation. In conclusion, the approach to saturation provides a more reliable estimate for  $M<sub>S</sub>$  (and thus the mass content of metallic iron), but fails to provide a reliable estimate of  $\chi$ <sub>HF</sub> because the magnetization of metallic iron is too strong and masks the signal from paramagnetic minerals.

Most hysteresis loops measured in this study were processed using approach to saturation. We find that a linear fit in the 0.9–1 T interval on average underestimates  $M<sub>S</sub>$  by 19% (SD 7%,  $n = 68$ ). For the few loops that were noisy at high fields (above approximately 0.7 T) and could not be processed using approach to saturation, we used a linear fit in the 0.9– 1 T and corrected the  $M<sub>S</sub>$  value using this 19% underestimate. It is noteworthy that approach to saturation is applicable only on the reversible part of the hysteresis loops, and is not strictly applicable to samples that are not saturated in a field of 1 T, the maximum field used for our measurements. A few tetrataenite-dominated chondrites (as identified by their higher  $B_{CR}$ , see Hysteresis Properties in Different Ordinary Chondrite Classes section) are clearly not saturated in 1 T (e.g., Guidder in Fig. 1A), leading to underestimation of  $M<sub>S</sub>$  when using both a linear fit in the 0.9–1 T range or the approach to saturation.

## INTRODUCTION TO FERROMAGNETIC MINERALS IN ORDINARY CHONDRITES

Before presenting the results of our hysteresis measurements, we give a brief overview of the magnetic minerals that are found in ordinary chondrites and their magnetic properties. With the exception of magnetite and cohenite that are present in some of the most unequilibrated chondrites like Semarkona (Hutchison et al. 1987; Krot et al. 1997), the only significant ferromagnetic minerals at room temperature are metallic FeNi alloys. These alloys are found mostly in the form of kamacite ( $\alpha$  bcc, with  $\sim$ 5 wt% Ni), taenite ( $\gamma$  fcc, with  $10-50$  wt% Ni, but paramagnetic at room temperature for Ni < 30 wt%), tetrataenite ( $\gamma$ " ordered tetragonal, approximately 50 wt% Ni), martensite (metastable distorted bcc), and rare awaruite  $(\gamma)$  ordered  $FeNi<sub>3</sub>$ ). These minerals can exist as a single phase, like kamacite that forms homogeneous grains up to several 100 μm, or can form complex microstructures such as plessite and zoned taenite (e.g., Reisener and Goldstein 2003; Yang et al. 1997a; Uehara et al. 2011). Plessite is a fine-grained  $(\leq 10 \mu m)$  intergrowth of kamacite and tetrataenite formed during decomposition of martensite below 500–300 °C (depending on the local Ni content) and down to temperatures below 200 °C (Reisener and Goldstein 2003; Goldstein and Michael 2006). Plessite can also be formed by reheating martensite in the kamacite + taenite stability field, i.e., between  $400^{\circ}$ C and 700 °C depending on the local Ni content.

We note that the terminology used in metallography and cosmochemistry studies is not always sufficient for a detailed understanding of magnetic properties. For instance, "taenite" grains are usually compositionally zoned in equilibrated ordinary chondrites, and they actually contain not only taenite (in relatively small amounts) but also a tetrataenite rim, a cloudy zone, and a martensite and/or plessite core (Scott 1973; Reisener and Goldstein 2003). The cloudy zone is composed of tetrataenite islands in a kamacite/ martensite matrix, although the nature of the matrix is still debated (Bryson et al. 2014). As a result, even though the average Ni content of such "taenite" is about 35 wt% Ni in ordinary chondrites (Afiattalab and Wasson 1980; Smith et al. 1993), this value likely represents an average value of fine tetrataenite grains with approximately 50 wt% Ni and paramagnetic taenite with less than 30 wt% Ni, as already discussed by, e.g., Nagata et al. (1986). Another example of this is the Santa Catharina Ni-rich ataxite, in which regions composed of pure cloudy zone described as tetrataenite islands of approximately 20 nm in a taenite matrix also have a bulk composition of 35 wt% Ni (Danon et al. 1979a; Zhang et al. 1990). Another issue is that tetrataenite is often reported in chondrites based on microprobe analyses only. But chemical analyses alone cannot distinguish between tetrataenite (formed by slow cooling) and taenite (formed by fast cooling) with the same composition. For instance, an approximately 100 lm euhedral "tetrataenite" grain described in an impact melt rock clast in the Jelica LL6 chondrite (Rubin 1994) is likely to be taenite in view of its position in a quenched impact melt rock. Similarly, the "tetrataenite" grains described in Semarkona (LL3.00) and Krymka (LL3.2) based on elemental analyses only (Rambaldi and Wasson 1984; Reisener and Goldstein 1999) are unlikely to be ordered in view of the low metamorphism temperature suffered by these meteorites, but are more likely nickel-rich taenite.

Examination of the FeNi phase diagram (e.g., Reuter et al. 1989; Yang et al. 1997a) shows that any taenite in slowly cooled meteorites should have a Ni



Fig. 1. Representative hysteresis loops: Guidder (LL5), Ausson (L5), Pultusk (H4), Juvinas (eucrite). The data are displayed before correction for high-field susceptibility.

content below 30 wt% Ni ( $\gamma_1$  phase), otherwise it would have decomposed by entering the spinodal region on cooling below 400–300 °C. As a consequence, any taenite in slowly cooled meteorites should be paramagnetic at room temperature (Crangle and Hallam 1963). This is confirmed by Mössbauer spectroscopy of the metallic phases of some ordinary chondrites (e.g., Nagata et al. 1986). However, the presence of room temperature ferromagnetic taenite (i.e., with  $Ni > 30$  wt%) has also been detected by Mössbauer spectroscopy in two other ordinary chondrites (Scorzelli et al. 1994; Kong and Ebihara 1996), sometimes in very significant amounts, suggesting nonequilibrium processes like fast postshock cooling or unusual cooling histories with initial slow cooling down to approximately 400°C, allowing formation of Ni-rich taenite followed by quenching that prevented decomposition into tetrataenite. Indeed, because of the slow solid state diffusion of Ni in FeNi (see, e.g., Hopfe and Goldstein 2001), thermodynamic equilibrium is not easily reached at low temperatures.

The formation of tetrataenite also requires a low cooling rate. Tetrataenite has been found in iron meteorites with metallographically determined cooling rates in the 650–400 °C range of up to several

thousands of  $^{\circ}$ C Myr<sup>-1</sup>, but is absent in shock reheated iron meteorites (Scott 1973; Goldstein et al. 2009). Petrographic observations also show the absence of taenite with Ni concentrations inside the tetrataenite range in slowly cooled equilibrated ordinary chondrites, except in the reheated zone close to the fusion crust (Clarke and Scott 1980). However, the exact cooling rate limit that allows formation of tetrataenite is not known as experiments cannot access this low cooling rate range, and numerical simulations have not been performed for the low-temperature range of the FeNi diagram (Mishin et al. 2005).

Combination of bulk and microprobe chemical analyses indicates that kamacite makes up to 88, 76, and 0–53 wt% of FeNi metal in H, L, and LL chondrites, respectively (Afiattalab and Wasson 1980). The tetrataenite, kamacite, and paramagnetic taenite contents in equilibrated ordinary chondrites were estimated by Mössbauer spectroscopy on a limited number of ordinary chondrite falls (Danon et al. 1979b; Nagata et al. 1986, 1991). The average kamacite, tetrataenite, and paramagnetic taenite contents computed from these measurements are reported in Table 1. There is a fair agreement between the kamacite content based on chemical analyses and Mössbauer

	$wt\%$ metal Technique Chemical analyses	$wt\%$ Ni in metal Chemical analysis	Fraction of kamacite in metal $(wt\%)$		Fraction of tetrataenite in metal $(wt\%)$	Fraction of $M_s$	Fraction of $M_{RS}$ carried
			Chemical analysis	Mössbauer	Mössbauer	carried by kamacite %	by kamacite %
References	a	a	h.	$\mathbf{c}$	$\mathbf{c}$	d	d
H	$18.20 \pm 1.44$ (N = 25) 10 (N = 28) 88			$65 (N = 1)$	$18(N = 1)$	-89	48
H/L	$13.79(N = 1)$	$12(N = 1)$					
L	$8.45 \pm 0.81$ (N = 53) 15 (N = 54) 76			$80 \pm 6$ (N = 2) 12 (N = 2)		-83	22
L/LL	$6.57 \pm 1.16$ $(N = 2)$ 16 $(N = 2)$						
LL	$2.94 \pm 1.51$ (N = 14)	$30 (N = 14)$ 53		$35 \pm 16$ (N = 6) 49 (N = 6)		56	3

Table 1. FeNi mineralogy in ordinary chondrites.

a Computed from Jarosewich (1990).

<sup>b</sup>Afiattalab and Wasson (1980).

<sup>c</sup>Computed using data on meteorite falls from Danon et al. (1979b); Nagata et al. (1986, 1991) (Mössbauer spectroscopy). For L chondrites, data for Shaw meteorite are not included based on its peculiar shock history (Danon et al. 1979b). For LL chondrites, Tuxtuac is an outlier and has not been included.

dSee text (see Contribution of Kamacite and Tetrataenite section).





a<br>Afiattalab and Wasson (1980).

 $<sup>b</sup>$ Smith et al. (1993).</sup>

<sup>c</sup>Crangle and Hallam (1963).

<sup>d</sup>Nagata et al. (1991).

<sup>e</sup>Néel et al. (1964) on synthetic tetrataenite.<br><sup>f</sup>Eunaki et al. (1986) on multidomain tetrat

Funaki et al. (1986) on multidomain tetrataenite.

g Nagata et al. (1986) on single-domain tetrataenite using Santa Catharina meteorite that contains only single-domain tetrataenite as ferromagnetic mineral (Danon et al. 1979a; Zhang et al. 1990).

<sup>h</sup>Rochette et al. (2009) on multidomain iron spheres.

i Nagata et al. (1975) on a EH3 chondrite with only multidomain kamacite as ferromagnetic mineral.

Butler and Banerjee (1975).

k This study.

Taenite is paramagnetic at room temperature for  $Ni < 30$  wt%.

spectroscopy. The situation for the most unequilibrated ordinary chondrites is somewhat different because their peak metamorphic temperatures below 400 °C prevented the formation of tetrataenite and preserved martensite, which would otherwise have decomposed into plessite (like in equilibrated chondrites).

The intrinsic magnetic properties of the FeNi minerals found in ordinary chondrites are summarized in Table 2. Because  $B_{\rm C}$ ,  $B_{\rm CR}$ , and  $M_{\rm RS}$  values depend closely on the magnetic domain state and hence the microstructure in which the minerals are found, these values are not given in Table 2. For instance, tetrataenite grains within plessite have lower coercivity than the

smaller tetrataenite grains in the cloudy zone (Uehara et al. 2011), and measured  $B_{CR}$  values for tetrataenite in meteorites range from 77 mT (Funaki et al. 1986) to approximately 1000 mT (Uehara et al. 2011) on large grains (approximately  $500 \mu m$ ) extracted from Toluca iron meteorite and on the cloudy zone of Ausson L5 and Agen H5 chondrites, respectively. Similarly, measured  $B<sub>C</sub>$ values for kamacite range from 1 mT (Nagata et al. 1975) to 600 mT (Lappe et al. 2011) for a EH3 chondrite and on a synthetic dusty olivine, respectively. As a general rule for ordinary chondrites, tetrataenite stands out with a much higher coercivity and saturation remanence (Funaki et al. 1986; Nagata et al. 1986, 1991; Uehara

et al. 2011) than kamacite and martensite that are characterized by low coercivity and low saturation remanence (Nagata et al. 1975; Wasilewski 1981) consistent with their multidomain state. Indeed, the magnetic single-domain size for kamacite, which is controlled by shape anisotropy for nearly all grain elongations, is restricted to a very narrow range: approximately 20–50 nm and approximately 17–22 nm for grains with axial ratios of 0.5 and 0.8, respectively (Butler and Banerjee 1975). By comparison, the critical size below which tetrataenite becomes superparamagnetic is 7 nm at room temperature (Nagata et al. 1991). The single-domain grain-size range for tetrataenite is presently unknown, but can be constrained on the basis of its grain-size independent (intrinsic) magnetic material parameters. According to Néel et al. (1964) and Paulevé et al. (1968), the magnetocrystalline anisotropy constant K is 1.0–1.3 10<sup>6</sup> J m<sup>-3</sup> and the saturation magnetization  $M<sub>S</sub>$  is 1.3  $\times$  10<sup>6</sup> A m<sup>-1</sup>. With these values, we obtain 1.5 to 2 T for the microcoercivity  $\mu_0H_k = 2 K/M_s$  and 0.94– 1.22 for the dimensionless magnetic hardness parameter  $Q = K/K_d$ , where  $K_d = \frac{1}{2}\mu_0 M_s^2$  is the scale for the stray field energy. According to Rave et al. (1998), the singledomain state in equidimensional particles with  $Q = 0.94$ -1.22 is stable to an edge length of at least  $L_{\text{min}}$  = 20 to 23  $L_{\text{ex}}$ , where  $L_{\text{ex}}$  is the exchange length with  $L_{\text{ex}} = \sqrt{A/K_d}$ , and A is the exchange constant, which for the tetragonal  $(L1_0)$  Fe<sub>0.5</sub>Ni<sub>0.5</sub> alloy is not available, but can be approximated by that of disordered Fe<sub>0.5</sub>Ni<sub>0.5</sub>  $A = 15.5 \times 10^{-11}$  J m<sup>-1</sup> (e.g., Hennemann and Siegel 1976). Thus, with  $L_{ex} = 3.8$  nm, the singledomain state is the energetically most stable magnetization configuration in particles with grain sizes up to at least  $L_{\text{min}} = 77{\text -}89$  nm. Depending on the magnetization history, the single-domain state may exist as a metastable state well beyond  $L_{\text{min}}$ . This is in agreement with micromagnetic simulations of the cloudy zone showing that tetrataenite islands up to approximately 100 nm in diameter have a single-domain behavior (Bryson et al. 2014).

#### RESULTS AND DISCUSSION

#### Spatial Homogeneity of Hysteresis Properties

We tested the consistency of hysteresis properties in a given meteorite by measuring multiple fragments for some meteorites. The data indicate that  $B_{CR}$ ,  $B_C$ , and  $M_{\rm RS}/M_{\rm S}$  have well-defined values for a given meteorite even for small samples down to a few tens mg (Fig. 2). This remains true even for LL chondrites that have low metal concentration and high magnetic anisotropy (see Anisotropy Effects section). For example, we measured the hysteresis properties of 16 samples of the Bensour



Fig. 2. Hysteresis properties for multiple samples of selected meteorites.

meteorite and these properties are constant for masses as low as 20 mg. However, because of the possible presence of a discrete number of large metallic grains, for small samples (a few tens of mg),  $M_{RS}$ ,  $M_S$ , and  $\chi$ can show strong departures from the mean value, especially in metal-poor LL chondrites (see, e.g., Gattacceca et al. 2003). Like bulk susceptibility (Rochette et al. 2003) or anisotropy of magnetic susceptibility (Gattacceca et al. 2005), hysteresis properties are representative intrinsic properties of a given ordinary chondrite at the scale of a few tens of mg. However, in LL chondrites, reliable estimates of  $M_{\rm RS}$  and  $M_{\rm S}$  require masses above 100 mg.

The consistency of hysteresis properties within a given meteorite enables merging results from multiple samples of the same meteorite. Merged values were computed in the following way:  $M_{RS}$  and  $M_S$  were mass-weighted,  $B<sub>C</sub>$  was M<sub>S</sub>-weighted (giving similar results to the approach of Nagata and Funaki [1982] for computing  $B_C$  for a mixture of two ferromagnetic materials), and  $B_{CR}$  were  $M_{RS}$ -weighted. This ensures that the mean values are equal to what would have been measured had the different samples been combined for a single measurement. The results, averaged at the meteorite level, are given in Table 3.

## Hysteresis Properties in Different Ordinary Chondrite Classes

The hysteresis properties of H and L chondrites are broadly similar, with low  $M_{RS}/M_S$  and high  $B_{CR}/B_C$ (Table 4, and representative cycles provided in Fig. 1). Together with the curvature observed on the hysteresis cycles up to 0.7 T (due to the high demagnetizing field of multidomain kamacite), these values show that soft multidomain FeNi metal (in the form of mostly kamacite and with a possible small contribution from taenite with  $>30$  wt% Ni in some unusual cases)











Table 3. Continued. Hysteresis properties of ordinary chondrites and HED achondrites. Table 3. Continued. Hysteresis properties of ordinary chondrites and HED achondrites. "Shock stage as defined by Stöffler et al. (1991).<br>"Uden (LL7) has been merged with LL6 for the mean in Table 3. Gursum (H4/5) results have been merged with H5 in Table 3, also based on the proposed H5 reclassification (Sc et al. 1990). °Uden (LL7) has been merged with LL6 for the mean in Table 3. Gursum (H4/5) results have been merged with H5 in Table 3, also based on the proposed H5 reclassification (Schultz et al. 1990).<br>et al. 1990).<br>"Terho et al. (19

eTerho et al. (1992).

fNagata and Sugiura (1976).

gNagata et al. (1986).

hKrol and Lang (1996).

See text for definition of magnetic parameters.



Table 4. Mean hysteresis properties by meteorite class, metamorphic type, and shock stage. Table 4. Mean hysteresis properties by meteorite class, metamorphic type, and shock stage.



Fig. 3. Hysteresis properties for ordinary chondrites and HED. Each circle corresponds to one meteorite.

dominates the hysteresis properties. Conversely, LL chondrites stand out with higher  $M_{RS}/M_S$  and lower  $B_{CR}/B_C$ , with almost no overlap with H and L chondrites (Fig. 3). Along with the "wasp-waisted" shape of their hysteresis loops, this is attributable to the higher tetrataenite contribution to hysteresis properties in these meteorites, as previously observed (Nagata and Funaki 1982; Nagata 1988; Pesonen et al. 1993; Gattacceca et al. 2003). In L chondrites, the existence of samples with  $M_{RS}/M_S$  0.02 and  $B_{CR}/B_C$  of 20–40 that are outside of the theoretical zone for multidomain grains reflects the significant contribution of tetrataenite

to the hysteresis properties (Fig. 3). This is also visible in the "wasp-waistedness" of their hysteresis loops, although this is less pronounced than in LL chondrites (Fig. 1). L/LL and H/L chondrites have hysteresis properties broadly similar to L and H chondrites.

As expected, H, L, and LL chondrites have distinct ranges for  $M<sub>S</sub>$  (Fig. 4a). This stems from their different bulk metal abundances that is the basis of the definition of these groups and which also clearly controls their magnetic susceptibility measurements (Rochette et al. 2003). The mean  $M<sub>S</sub>$  values for H, L (Table 4) correspond to a minimum metal content of  $18.0 \pm 3.1$ and  $8.0 \pm 2.7$ , respectively, based on the simplistic assumption that all the metal is in the form of kamacite with  $M<sub>S</sub> = 224$  Am<sup>2</sup> kg<sup>-1</sup>. These values are lower limits because tetrataenite has a lower  $M<sub>S</sub>$  than kamacite, and paramagnetic taenite does not contribute to  $M<sub>S</sub>$ . They compare well with the Fe + Ni + Co abundances obtained from chemical analyses of the metallic fraction of meteorites (computed from the data of Jarosewich [1990] by selecting falls only, excluding meteorites described as weathered by the author, using updated classifications for some meteorites, and discarding values more than 3 SD away from the mean value for H and L chondrites):  $18.2 \pm 1.4$  wt%  $(n = 25)$  for H and  $8.5 \pm 0.8$  wt%  $(n = 53)$  for L (Table 1).

Although they have distinct ranges for  $M<sub>S</sub>$ , it is noteworthy that H, L, and LL chondrites have similar ranges for  $M_{RS}$  (Fig. 4b and 5), as already noticed on a more limited  $M_{RS}$  database by Rochette et al. (2003). The observation in Rochette et al. (2003) that H chondrites have lower  $M_{RS}$  scatter than L and LL



Fig. 4. Frequency histograms of  $M_S$  and  $M_{RS}$  for LL, L, H chondrites, and HED achondrites. The percentages are computed within each group. The number of data for each class is indicated between parentheses. The  $M_{RS}$  histogram includes data from Rochette et al. (2003).



Fig. 5.  $M_{RS}$  versus  $M_S$  for H, L, LL, and HED meteorites. The dotted lines are lines of constant  $M_{RS}/M_S$ .

is also confirmed, as well as the possible existence of two clusters for H chondrites with a gap of  $M_{RS}$ values in the  $0.38-0.44 \text{ Am}^2 \text{ kg}^{-1}$  interval in our database, these two groups being undistinguishable in terms of petrographic type or shock stage. The smaller scatter observed for H chondrites is due at least in part to their higher metal concentration that enables more accurate bulk measurements compared with L and LL chondrites for samples of similar size. The absence of correlation between  $M_{RS}$  and  $M_S$  within each class (Fig. 5) confirms the strong decoupling between induced and remanent magnetic properties in ordinary chondrites.

It is noteworthy that HED achondrites do not follow the same trend (Fig. 5, Table 3). For an equivalent  $M<sub>S</sub>$ , they have much lower  $M<sub>RS</sub>$ , in agreement with the usual absence of taenite and tetrataenite (e.g., Mayne et al. 2009). Moreover, HEDs show a reasonably linear relationship between  $M_{RS}$  and  $M<sub>S</sub>$  (Fig. 5), indicating that the same mineral controls the remanent and the induced magnetic properties. This mineral, multidomain kamacite, varies only in concentration. This was already indicated by the good correlation between  $M_{RS}$  and magnetic susceptibility in HED meteorites (Rochette et al. 2009) and lunar rocks (Rochette et al. 2010).

 $\chi$ <sub>HF</sub> average values for H, L, LL, and HED are the following:  $5.3 \pm 2.0$ ,  $2.8 \pm 0.9$ ,  $1.4 \pm 0.5$ , and  $0.4 \pm 0.6$  $10^{-6}$  m<sup>3</sup> kg<sup>-1</sup>. This apparent trend with chondrite class is clearly an artifact of incomplete removal of the ferromagnetic slope, as the paramagnetic contribution

should increase from H to LL chondrites in view of the increasing Fe content of the silicates. Only the HED values provide a significant estimate of the true paramagnetic contribution that is in good agreement with the theoretical value of  $0.35 \times 10^{-6}$  m<sup>3</sup> kg<sup>-1</sup> that can be computed from the elemental composition of these meteorites (see discussion in Gattacceca et al. 2008).

#### First-Order Reversal Curve Properties

We measured the first-order reversal curve (FORC) properties of three chondrites: Guidder (LL5), Chergach (H5), and Tamdakht (H5). Despite the strong differences between hysteresis loops for H and LL chondrites (Fig. 1), their FORC diagrams share the same distinct features (Fig. 6). This strongly suggests the presence of the same magnetic phases with different mixing ratios. Three distinct features are apparent, which are most pronounced in Guidder: (1) a highcoercivity ridge running parallel to the  $B<sub>C</sub>$  axis with some small offset toward positive bias field; (2) a lowcoercivity ( $B<sub>C</sub> < 100$  mT) contribution, which is also offset toward positive bias field by a few tens of mT; and (3) a diagonally running positive ridge with a negative trough attached to its right, both extending from the low-coercivity maximum at an angle of about  $45^{\circ}$  from the  $B_C$  axis down to negative bias field. This distinct set of FORC features has already been observed by Acton et al.  $(2007)$  in chondrules of the Bjurböle L/LL4 chondrite. Acton et al. (2007) ascribed the diagonal ridge to interactions between a very lowcoercivity ("nearly superparamagnetic") component and a magnetically hard component. We also interpret the diagonal ridge in terms of interactions between hard and soft components, but suggest that the coercivity of the magnetically soft component (kamacite) needs not be low in absolute terms, but just distinctly lower than that of the magnetically hard phase (tetrataenite) in order for both components to have a mutual biasing effect. The simplest way to demonstrate the presence of interactions is by way of feature (2). Here, the shift of the low-coercivity distribution toward positive bias field represents "earlier" downswitching of the soft component along the descending major hysteresis branch (at a reversal field that is smaller than the switching field of the soft component when isolated), and delayed upswitching along the FORC, i.e., the soft component is destabilized by the presence of the stray fields from the hard component. Premature downswitching and delayed upswitching reflect demagnetizing interactions, which are also reflected by the fact that IRM acquisition requires higher fields than demagnetization of the saturation remanence does



Fig. 6. First-order reversal curve diagrams. To better bring out the high-coercivity part of Chergach and Tamdakht, which are dominated by viscous contributions, their FORC diagrams were truncated at the low-coercivity side.

(Cisowski-Wohlfarth R values are 0.25 for Guidder and 0.15 for the Tamdakht sample). The magnetic interactions between kamacite and tetrataenite had already been observed at microscopic scale in H ordinary chondrites scale by magneto-optical imaging (Uehara et al. 2011).

#### Hysteresis and Susceptibility

Ferromagnetic susceptibility and  $M<sub>S</sub>$  are well correlated for all three classes of ordinary chondrites (Fig. 7). The  $M_S/\chi_f$  ratios are  $1.83 \times 10^5$ ,  $2.54 \times 10^5$ , and  $3.77 \times 10^5$  A m in H, L, and LL chondrites, respectively (Table 4). This illustrates again the dichotomy between H and L chondrites, and LL chondrites. In H and L chondrites, the induced magnetic properties are most likely controlled by multidomain kamacite, although some influence of tetrataenite is already visible in the L group. However, the  $M_S/\chi$  value in H and L chondrites (overall mean value is  $2.32 \times 10^5$ ) is significantly lower than the theoretical  $M_S/\chi$  =  $4.12 \times 10^5$  A m obtained using  $M<sub>S</sub> = 224$  Am<sup>2</sup> kg<sup>-1</sup> for kamacite (Crangle and Hallam 1963) and  $\chi = 5.44 \times 10^{-4} \text{ m}^3 \text{ kg}^{-1}$  determined for multidomain iron spheres (Rochette et al. 2009). The discrepancy is even stronger if one takes into account that part of the metal is in the form of tetrataenite that has likely a higher  $M_s/\gamma$  than kamacite (see discussion in Rochette et al. [2003] about the magnetic susceptibility of tetrataenite).

This deviation can be attributed to three factors that can increase apparent susceptibility: the presence of superparamagnetic grains and/or the presence of



Fig. 7.  $\chi_f$  versus  $M_S$  for the three classes or ordinary chondrites. The two properties are measured on the same sample. The dotted line is the theoretical line for multidomain kamacite (see text).

elongated metallic grains, and/or magnetic interactions. Indeed, superparamagnetic metal grains can have a magnetic susceptibility up to 10 times that of multidomain grains (see, e.g., Van de Moortele et al. 2007). We tested for the presence of superparamagnetic FeNi grains (i.e., with a size below about 20 nm for kamacite [Dunlop and Özdemir 1997] and  $7 \text{ nm}$  for tetrataenite [Nagata et al. 1991]), by measuring the frequency dependence of low-field susceptibility of

Table 5. FeNi grain shapes in ordinary chondrites.

Meteorite		Median aspect ratio	Mean aspect ratio	SD	$\boldsymbol{n}$
Baszkówka	L5	1.87	1.99	0.55	2873
Estacado	H6	1.94	2.06	0.58	9999
Kernouve	H6	1.78	1.92	0.56	8254
Kyushu	L6	1.94	2.07	0.61	8680
Middlesbrough	L6	1.90	2.02	0.58	6340
MIL99301	LL6	1.91	2.03	0.57	2921
Moorleah	L6	1.93	2.06	0.59	2653
Saratov	L4	2.00	2.12	0.62	2361
Tennasilm	L4	1.93	2.09	0.63	4014
Utrecht	L6	1.90	2.02	0.56	2445
10) Mean $(n)$ $=$		1.91	2.04	0.05	

 $n =$  number of analyzed grains.

several dozens of ordinary chondrite falls (detailed results not shown here). The measured variations are almost within detection limit (Table 4) and exclude a significant contribution of superparamagnetic FeNi grains to the magnetic susceptibility of ordinary chondrites. Regarding the shape of the grains, using a mean susceptibility  $K = (N + 1/3)/N(1 - N)$  for an ellipsoid of revolution where  $N$  is the demagnetization factor that depends on the aspect ratio of the ellipsoid, the misfit between the measured and theoretical  $M_{\rm s}/\chi$ for H and L chondrites requires N values of 0.08 or 0.75, indicating ellipsoid aspect ratio of 3 to 4 for prolate grains, and about 0.2 for oblate grains (e.g., Carmichael 1989). Furthermore, our XR microtomography found that all samples have a remarkably similar distribution of grain shape, with aspect ratio (ratio of maximum over minimum dimension) clustered around a mean value of 2.0 (Table 5). In all studied meteorites, about 50% of the grains are classified as equant, 30% prolate, 15% oblate, and 7% triaxial using Zingg criteria (Zingg 1935). Such a shape results in an approximately 18% increase in magnetic susceptibility with respect to spherical grains and is not enough to account for the observed discrepancy. Magnetic interactions are evidenced by the FORC properties (see First-Order Reversal Curve Properties section) and result in a higher magnetic susceptibility. Finally, metal grains with silicate or troilite inclusions, which are common in ordinary chondrites, have the same magnetic susceptibility as solid metal grains with the same shape, but lower  $M_s$ , again leading to a decrease in the  $M_S/\chi$  ratio.

Because apparent magnetic susceptibility is increased by the combination of these effects, it cannot be used directly to estimate quantitatively the amount of metal in ordinary chondrites.  $M<sub>S</sub>$  is a better proxy of the bulk metal content. However, the good correlation between the two properties (Fig. 7) justifies the use of magnetic susceptibility for classification of ordinary chondrites (Rochette et al. 2003), all the more because susceptibility can be measured on much larger samples than hysteresis (up to several dozen grams), which allow averaging heterogeneities in composition.

## Contribution of Kamacite and Tetrataenite

Combining the results from Mössbauer spectroscopy and chemical analyses (Table 1), the magnetic properties of the individual metallic phases (Table 2), and our measurements of meteorites, we obtain a picture of the contribution of each magnetic phase to the bulk magnetic properties of ordinary chondrites. Relative to the meteorite total mass, kamacite amounts to 16, 6.8, and 1.0  $wt\%$  in H, L, and LL chondrites, respectively (Table 1). This estimate was computed by combining the total abundance of metal determined by chemical analyses (Jarosewich 1990) and the kamacite fraction in the metal provided by Mössbauer for L and LL chondrites, and from chemical analysis for H chondrites (because only one H chondrite fall has been studied by Mössbauer spectroscopy [Danon et al. 1979b] and this measurement yielded an unrealistically high tetrataenite fraction of 18%). This translates into a contribution of kamacite of 89, 84, and  $56\%$  to the  $M<sub>S</sub>$  of H, L, and LL chondrites, respectively. A reasonable estimate for  $M_{RS}$  of kamacite in ordinary chondrites can be derived from the hysteresis properties of H chondrites with the lowest coercivity that are dominated by multidomain kamacite and give  $M_{\text{RS}}/M_{\text{S}} = 5 \times 10^{-3}$ . This value is close to the value of  $4 \times 10^{-3}$  measured for a synthetic sample made of dispersed  $10 \mu m$  pure iron grains by Bezaeva et al. (2010). Using a  $M_{\text{RS}}/M_{\text{S}}$  of  $5 \times 10^{-3}$ , we can estimate the contribution of kamacite to  $M_{RS}$  to 48, 22, and 3.0% in H, L, and LL chondrites, respectively. These numbers, which do not follow the trend for  $M_s$ , confirm the decoupling between induced and remanent properties in ordinary chondrites.

## Hysteresis Properties, Metamorphism, Shock, and Irradiation

We have seen that the broad picture is the same for all ordinary chondrites, with induced magnetic properties (susceptibility and  $M<sub>S</sub>$ ) controlled by the amount of kamacite (except in some metal-poor LL chondrites), and the remanent properties increasingly influenced by tetrataenite from H to L to LL chondrites. However, it is noteworthy that in detail, different meteorites within the same class have significantly distinct hysteresis properties (Fig. 2). For

instance, coercivity of remanence varies from 9 to 453 mT in L chondrites, and from 5 to 335 mT in H chondrites. Therefore, hysteresis properties may reveal the different histories of these meteorites. Table 4 provides the average hysteresis properties as a function of class, petrographic type, and shock stage.

The scope of this study is not to discuss individual meteorites in detail, but rather to derive general mechanisms that can affect the magnetic properties. We identify three events that may affect the hysteresis properties of meteorites: thermal metamorphism, shock metamorphism, and irradiation. Thermal metamorphism affects the grain size of FeNi metal (Afiattalab and Wasson 1980; Easton 1983), modifies their mineralogy (e.g., Reisener and Goldstein 2003), and may anneal their defects. Shock metamorphism also affects the mineralogy (Taylor and Heymann 1971; Scorzelli et al. 1987; Ming et al. 1995; Bennett and McSween 1996; Smith and Goldstein 1997), the grain size (Friedrich et al. 2008) and the shape distribution (Gattacceca et al. 2005; Friedrich et al. 2008) of metal grains. Shock may also result in silicate darkening in which micron-sized metal blebs form in silicates (e.g., Bennett and McSween 1996; Van de Moortèle et al. 2007), and may lead to thermal annealing in some cases (Rubin 2004). Irradiation by protons has been shown to modify the magnetic properties of kamacite (Butler and Cox 1974), and irradiation by neutrons leads to ordering of taenite into tetrataenite (Paulevé et al. 1962). In natural settings, irradiation can occur at the surface of the parent body for regolith breccia samples, or while the meteorite is being transferred to the Earth under the form of a meter-sized meteoroid.

Because of the small size of the studied samples, it is not possible to provide a detailed discussion of the variation in the mass normalized magnetic properties like  $M<sub>S</sub>$ ,  $M<sub>RS</sub>$ , or  $\chi$ . These properties can be biased by nugget effects, especially in LL chondrites. However, it is noteworthy that in agreement with the strong decrease in magnetic susceptibility with increasing petrographic type observed in LL chondrites (Rochette et al. 2003), a similar trend is visible, although not as clearly, for  $M_S$ :  $M_S = 6.3 \pm 2.3$  ( $N = 8$ ) for LL3 and LL4, and  $M_s = 3.6 \pm 1.9$  ( $N = 17$ ) for LL5 and LL6 (Fig. 8a). This can be accounted for only in part by a higher proportion of tetrataenite versus kamacite and taenite in LL5 and LL6, and requires a higher bulk metal content in LL3 and LL4. Similarly, we observe a higher  $M<sub>S</sub>$ , and hence a higher bulk metal content, in L3 and L4 ( $M<sub>S</sub> = 22.1$ ,  $N = 7$ ) compared with L5 and L6 chondrites ( $M<sub>S</sub> = 16.6$ ,  $N = 22$ ), and in H3 and H4 ( $M<sub>S</sub> = 44.8$ ,  $N = 8$ ) compared with H5 and H6 chondrites ( $M<sub>S</sub> = 38.6$ ,  $N = 16$ ). This is in agreement with the decrease in the modal abundance of metal in ordinary chondrites as a result of oxidation during metamorphism (McSween and Labotka 1993).

Mass independent magnetic properties (e.g.,  $B_C$ )  $B_{CR}$ ,  $M_{RS}/M_S$ , and  $M_S/\chi$ ) can be interpreted with more confidence.  $B_{CR}$  and  $B_C$  increase with petrologic type in all classes (Fig. 8c, except for LL6 chondrites that are discussed below), which indicates that increasing thermal metamorphism is accompanied by increasing formation of tetrataenite. Among ordinary chondrites, this effect is most visible in LL chondrites that have the lowest amount of kamacite, which otherwise dilutes the tetrataenite signal. The increasing amount of tetrataenite with increasing metamorphism temperature is compatible with a lower cooling rate, allowing more diffusion of Ni in taenite during cooling above 400 °C, and eventually formation of tetrataenite by spinodal decomposition of this taenite below 400 °C. This overall inverse correlation between metamorphic grade and cooling rate would be compatible with an onion-shell model for the ordinary chondrite parent bodies (e.g., Kleine et al. 2009), at least while they cool in the subsolidus temperature range. However, recent metallographic cooling rate determination shows that this onion-shell model is likely not valid (Scott et al. 2013). A more plausible explanation is that metamorphism is accompanied by oxidation of metal in ordinary chondrites, as documented by the evolution with metamorphism of the iron content of silicates, of the relative abundances of silicates, and of the composition of metal (McSween and Labotka 1993; Rubin 2005; Dunn et al. 2010). The increasing Ni concentration in metal with increasing metamorphism as a result of oxidation will increase the tetrataenite abundance, as revealed by our measurements.

Notable exceptions are the LL6 chondrites that have significantly lower  $B_{CR}$  and  $M_{RS}$  than LL5 chondrites (Figs. 8b and 8c). This may be due to the formation of larger tetrataenite rims in these meteorites, because such rims have a multidomain behavior with lower  $B_{CR}$  than the smaller tetrataenite grains in the cloudy zone of taenite (Uehara et al. 2011). This process may not be visible in H6 and L6 chondrites because of the lower Ni content in the metal compared with LL. A good correlation between the width of the tetrataenite rim and the metallographic cooling rate has been found for IVA irons meteorites (Goldstein et al. 2009). Another explanation of the lower  $B_{CR}$  and  $M_{RS}$  in LL6 chondrites may be the increasing size of the tetrataenite particles in the cloudy zone. Again, tetrataenite grain size in the cloudy zone is correlated with the cooling rate at approximately 350–450 °C (Yang et al. 1997b). All together, this may indicate lower cooling rates in LL6 than in LL5 chondrites.  $M_S/\chi$  variations are small



Fig. 8. Average hysteresis properties  $(\pm 1$  SD) as a function of petrologic type for the three classes of ordinary chondrites. For L5 and L6 chondrites, our database purposely contains a large number of meteorites with shock stage >S3 (see Samples section). To avoid a bias, these heavily shocked meteorites are not included in the average values displayed here.

except for LL chondrites where this ratio increases with petrographic type, especially in LL6 chondrites (Fig. 8d). At the moment, we have no satisfactory explanation of this observation. Because remanent and induced properties are uncorrelated in ordinary chondrites, variations of  $B_{CR}/B_C$  and  $M_{RS}/M_S$  with thermal metamorphism, although significant, are also difficult to interpret.

In our database, only L chondrites display the full range of shock stages (S1–S6). The highly shocked L chondrites show significantly lower  $B_{\rm C}$ ,  $B_{\rm CR}$ ,  $M_{\rm RS}$ , and  $M_{\rm RS}/M_{\rm S}$  compared with less shocked L chondrites (S1– S3). S4 meteorites have intermediate properties (Table 4, Fig. 9). This indicates a lower tetrataenite content in highly shocked L chondrites (S4–S6). This lower content can be explained in part by disordering by the shock wave, an effect that has been documented experimentally (Scorzelli et al. 1987). In particular, the

thermal excursion associated with the impact and the ensuing fast cooling result in disordering of the initial tetrataenite and decomposition into disordered Ni-rich ferromagnetic taenite ( $\gamma_2$  phase). This is well in agreement with the postshock temperature increase above 600 °C estimated for S5 and S6 shock stages (Stöffler et al. 1991). In view of the typical equilibrium temperatures of approximately 165 K encountered in the asteroid belt (e.g., Spencer et al. 1989), such temperature increases lead to postshock temperature above approximately 500 °C, which is enough to disorder tetrataenite in a matter of minutes, although the knowledge of the disordering kinetics of tetrataenite as a function of temperature is still fragmentary (Wasilewski 1988). In S4 chondrites, it is likely that localized temperature excursions, as indicated by the presence of melt pockets and melt veins, result in localized disordering of tetrataenite. The effect of shock



Fig. 9. Average hysteresis properties  $(\pm 1 \text{ SD})$  as a function of shock stage for L chondrites.

heating on the metallography has long been recognized (Taylor and Heymann 1971), in particular with the disappearance of cloudy taenite upon heating above approximately 500 °C (Scott 1973), the decomposition of martensite into plessite upon heating above 500 °C (e.g., Reisener and Goldstein 2003), and the formation of new martensite during rapid cooling from above approximately 700 °C (e.g., Yang et al. 1997a). But the magnetic signature of this transient heating event has never been documented.

Finally, we observe no correlation between the magnetic properties of the studied meteorites and the cosmic ray exposure age (Graf and Marti 1994, 1995) for H and LL chondrites. Irradiation does not produce a detectable influence on the magnetic properties of ordinary chondrites.

These general trends do not explain the unusual results obtained for a limited number of outliers. These deserve individual discussions that are beyond the scope of this study. As an example, Ste. Marguerite and Forest Vale, both unshocked H4, have distinctly lower  $M_{\rm RS}/M_{\rm S}$  than other H chondrites. This can be related to the distinctly high metallographic cooling rates  $(>1000 \degree C/Myr)$  measured for these two meteorites and attributed to excavation by impact (Scott et al. 2010). The fast and unusual cooling of Forest Vale has also been inferred from the study of Cr spinel and chromite (Wlotzka 2005) and recently confirmed to be as high as  $10^4$  °C/Myr (Ganguly et al. 2013). This fast cooling resulted in a limited or null amount of tetrataenite in these meteorites. This exemplifies the potential for magnetic measurements to detect samples with unusual cooling history.

It must be emphasized, however, that the interpretation of hysteresis properties is not unique. For instance, low  $B_{CR}$  values in L chondrites are characteristic of both L3 (in which no tetrataenite formed) and heavily shocked equilibrated L chondrites (in which tetrataenite has been disordered into taenite). Moreover, some individual results remain enigmatic. For instance, the equilibrated L chondrite Mount Tazerzait (L5, S1) and Pê (L6, S1) have magnetic properties at the two extremes of the range covered by equilibrated L chondrites with  $B_{CR} = 16$  and 453 mT, respectively, a discrepancy that remains unexplained.

#### Correlation with Microscopic Observations

It has been shown recently that the remanence carrier with the highest  $M_{RS}$  in ordinary chondrites is the cloudy zone of zoned taenite grains (i.e., submicron tetrataenite grains embedded in a kamacite/martensite matrix) (Uehara et al. 2011). The  $M_{RS}$  of larger tetrataenite grains, as found in the tetrataenite rim of zoned taenite or in zoneless plessite, is much lower, and that of kamacite even lower.

IRM acquisition curves for bulk samples of LL chondrites show three distinct components with coercivity peaking at <10 mT, approximately 60 mT, and approximately 650 mT, and extending up to 1.2 T (Fig. 10a). This was already observed in separated chondrules from Bjurböle L/LL4 chondrite (Acton et al. 2007). The same is observable in L chondrites (Fig. 10b). In H chondrites, the high-coercivity peak is faint but still visible (Fig. 10c). It is noteworthy that the two highcoercivity peaks correspond closely to the coercivity of remanence observed at microscopic scale for large tetrataenite grains and fine-grained tetrataenite in the cloudy zone (Uehara et al. 2012). The low-coercivity peak can be attributed to multidomain kamacite. It is also noteworthy that highly shocked chondrites lack the two high-coercivity peaks (Fig. 10d), which indicates that the original tetrataenite has been disordered. The drastic changes in magnetic properties associated with reheating events are demonstrated by the disappearance of the two high-coercivity peaks upon heating to 600 °C in controlled fugacity atmosphere for 20 min (Fig. 10c). This is additional proof that tetrataenite, with different domain states, is responsible for the two high-coercivity peaks of the coercivity spectrum.



Fig. 10. IRM acquisition curves and their derivative for a selection of ordinary chondrites. For Pê (L6, S1), the gray circles (respectively, open circles and broken line) are the results after the sample was heated at 450 °C (respectively, 600 °C) for 20 min.

The size of tetrataenite grains in the cloudy zone of ordinary chondrites ranges from approximately 10 nm to approximately 100 nm (Yang et al. 1997a; Uehara et al. 2011) and is related to the cooling rate of the meteorite in the 350–450 °C range (Yang et al. 1997b). This size range corresponds almost exactly to the size

range in which tetrataenite is in a single-domain magnetic state  $(7 \text{ nm to} > 80-90 \text{ nm})$ , see Introduction to Ferromagnetic Minerals in Ordinary Chondrites section). This single-domain behavior accounts for the high coercivity and high  $M_{RS}$  of the cloudy zone, as opposed to the low coercivity and low remanence of kamacite grains that are in multidomain state. The single-domain state of tetrataenite in the cloudy zone is also in good agreement with the high  $B_{CR}$  values (up to 1 T) measured at microscopic scale in this structure (Uehara et al. 2011) and with micromagnetic simulations (Bryson et al. 2014).

## IMPLICATIONS FOR PALEOMAGNETISM

The understanding of the magnetic properties of the different FeNi minerals contained in ordinary chondrites has implications for their potential for paleomagnetic studies. In equilibrated ordinary chondrites, the possible carriers of a paleomagnetic record are kamacite, tetrataenite, and possibly ferromagnetic taenite. The situation is the same for the least primitive unequilibrated chondrites that have been heated above approximately 400 °C (allowing formation of tetrataenite), which correspond more or less to a petrologic type above 3.5 (Huss et al. 2006). Although kamacite has a simple behavior upon cooling, with no low-temperature martensitic transition, it is present in ordinary chondrites as large grains that have a strong multidomain behavior with typical  $B_{CR}/$  $B_C \sim 45$  and  $M_{RS}/M_S \sim 0.005$ , making them poor paleomagnetic recorders of dubious reliability (e.g., Tikoo et al. 2012). Tetrataenite can be found in a number of microstructures: in the cloudy zone of zoned taenite grains where it is found mostly as single-domain grains; in zoned plessite grains (in H and L equilibrated chondrites only, see Reisener and Goldstein 2003) as multidomain grains, or as massive multidomain grains in LL chondrites (Clarke and Scott 1980). Because of its high coercivity, single-domain tetrataenite in the cloudy zone may appear as an ideal paleomagnetic recorder. However, tetrataenite is a secondary mineral formed by ordering of a Ni-rich taenite precursor below the critical ordering temperature of 320 °C (Paulevé et al. 1962). Although it has been proposed that it may record the magnetization of its precursor taenite grain (e.g., Wasilewski 1988; Nagata and Funaki 1982), closer examination of the FeNi phase diagram shows that the precursor taenite grains become ferromagnetic only when cooling below 400 °C for the tetrataenite rim, and below 350 °C (maximum value) for the single-domain tetrataenite in the cloudy zone (Yang et al. 1997a; Uehara et al. 2011). As a consequence, the precursor taenite grains (with Ni content approximately 50 wt% Ni and a Curie temperature of approximately 500 °C) can only record a magnetization when the temperature goes below 400 °C (for the rim) or below 350 °C (for the cloudy zone). This magnetization is either a thermochemical remanent magnetization or a partial thermoremanent magnetization acquired while cooling between 400 °C and 320 °C for the rim, and 350 °C and 320 °C for the cloudy zone.

It is not clear whether tetrataenite grains can inherit any remanent magnetization from their parent taenite grain. In any case, the single-domain tetrataenite in the cloudy zone can only record magnetic fields that are present after the meteorite has cooled below 350 °C (if it inherits the remanent magnetization of the precursor taenite) or 320  $\rm{^{\circ}C}$  (if it does not). Therefore, slow cooling following thermal metamorphism down to 400 °C represents a time gap during which no paleomagnetic record can be preserved in equilibrated ordinary chondrites. The shortest time gap is obtained for type 4 chondrites (heated to about 700 °C; Huss et al. 2006) that have cooled relatively fast. For meteorites such as Bath, a H4 with a cooling rate of 80  $\rm{°C}$  Myr<sup>-1</sup> (Willis and Goldstein 1981), this time gap is only about 4 Myr. In view of the old age of the peak metamorphism temperature in ordinary chondrites, type 4 ordinary chondrites may well have cooled below 400–320 °C around 10 Ma after the formation of the solar system, but for type 6 chondrites, such cooling may require more than 100 Ma (Kleine et al. 2009; Henke et al. 2012).

For the most primitive ordinary chondrites (below petrographic type 3.5), the potential carriers of paleomagnetic information are kamacite and martensite. Magnetite and cohenite that have been reported in a few very primitive ordinary chondrites (Hutchison et al. 1987; Krot et al. 1997) are not discussed here. As in equilibrated chondrites, multidomain kamacite is a very poor paleomagnetic recorder, and martensite loses any potential paleomagnetic record of its precursor taenite when the martensitic transformation occurs (Momose et al. 1984) (although for most of the range of Niabundances over martensite forms, the parent taenite will have a Curie point below room temperature). Only single-domain kamacite present in dusty olivine grains appears to be a potential source of paleomagnetic information in unequilibrated ordinary chondrites (Uehara and Nakamura 2006; Lappe et al. 2011), although accessible only through magnetic microscopy.

#### **CONCLUSION**

We have shown that hysteresis properties measured on samples of a few tens of mg are distinctive intrinsic properties of individual ordinary chondrites, and we

provide a large data set of hysteresis properties of ordinary chondrite falls. The metal contents derived from magnetic measurements are in agreement with those estimated previously from chemical analyses. We evidence a decreasing bulk metal content with increasing petrologic type in ordinary chondrites, in agreement with progressive oxidation of metal with increasing thermal metamorphism. The bulk magnetic properties of ordinary chondrites can be approximated with a mixture of multidomain kamacite and tetrataenite. Kamacite dominates the induced magnetic properties in all ordinary chondrites, and two distinct populations of tetrataenite grains dominate the remanence properties. This picture derived from bulk magnetic properties is in very good agreement with microscopic observations, with a dichotomy between tetrataenite grains in the magnetic single-domain size range (7 nm to at least 80–90 nm) that are found in the cloudy zone of taenite grains, and larger  $(> \mu m)$ multidomain tetrataenite grains are found in plessite or in the rim of zoned taenite grains.

The presence of tetrataenite requires initial thermal metamorphism above 400 °C followed by slow cooling, with cooling rate of a few thousands  $\rm{^{\circ}C}$  Myr<sup>-1</sup> or lower in the subsolidus temperature range, which explains its absence in fast-cooled FeNi-bearing achondrites (e.g., HED and lunar meteorites) in which multidomain kamacite controls both the induced and the remanent magnetic properties. The bulk amount of tetrataenite is inversely correlated with the cooling rate above 400 °C (slower cooling allowing more diffusion of Ni), and the size of the tetrataenite precipitates in the cloudy zone and the thickness of tetrataenite rims are inversely correlated with the cooling rate. Therefore, the presence of tetrataenite, its distribution between different microstructures, and its grain-size distribution are controlled by the thermal history of the meteorites.

Highly equilibrated chondrites (types 5 and 6) have a higher tetrataenite content than type 4 chondrites. This may suggest lower cooling rates in the  $650-450$  °C interval, which would be supportive of an onion-shell model at least while cooling through this temperature interval, but a more likely origin for this observation is the increasing Ni content in metal with increasing metamorphism as a result of oxidation. In equilibrated chondrites, shock-related transient heating events above approximately 500 °C result in the disordering of tetrataenite into Ni-rich taenite, and an associated drastic change in magnetic properties (decrease in saturation remanence, coercivity, and coercivity of remanence). This effect is maximal for chondrites with shock stage S5 and S6, but is already visible starting from shock stage S4. As a good indicator of the amount of tetrataenite, hysteresis properties (in

particular,  $B_{CR}$ ,  $M_{RS}$ , and the coercivity spectrum derived from IRM acquisition) are very sensitive proxies of the thermal history of ordinary chondrites. Both low cooling rates (during thermal metamorphism) and high cooling rates (following shock or thermal metamorphism) can be identified. Our data set strengthens the view that because of the poor paleomagnetic behavior of multidomain kamacite and the secondary nature of tetrataenite, equilibrated ordinary chondrites are challenging targets for paleomagnetic study.

Acknowledgments—The research leading to these results has received funding from Agence Nationale de la Recherche (project ANR-09-BLAN-0042), and People Programme (Marie Curie Actions) of the European Union's Seventh Framework Programme (FP7/2077- 2013) under REA grant agreement N°298355. C.S. was supported by the European Commission through the Marie Curie Actions–RTNs ORIGINS (Project ID: 35519). C.S. and B.P.W. thank the Brown-Massachusetts Institute of Technology (MIT) NASA Lunar Science Institute, the NASA Origins Program, and Thomas F. Peterson, Jr. We thank the Muséum d'Histoire Naturelle de Paris for the loan of meteorites. Philippe Thomas (Meteoritica) kindly provided Chergach and Tamdakht samples. Rainer Bartoschewitz kindly provided Braunschweig samples. We acknowledge Francois Demory (CEREGE) for his help in the laboratory. This study benefited from constructive insights provided by J. I. Goldstein, G. Kletetschka, and associate editor E. R. D. Scott.

Editorial Handling—Dr. Edward Scott

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