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Low temperature magnetic transition of chromite in ordinary chondrites

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[1] We present low temperature magnetic data (thermal demagnetization, hysteresis) obtained on thirty three ordinary chondrite meteorites. A magnetic transition is observed in the 40–80 K range (average 66 K), and is interpreted as a ferrimagnetic to paramagnetic phase transition. We present quantitative analyses of changes in magnetization across the magnetic phase transition and chemical elemental analyses to argue that chromite, a common accessory mineral in meteorites, is the mineral undergoing the phase transition, and not troilite or tetrataenite as proposed in previous studies. Moreover, we observe a correlation between the measured Curie temperature and chemical composition of chromites in ordinary chondrites. Low temperature magnetic measurement are thus a sensitive indicator of chromite composition in these meteorites, and as such a possible proxy to the understanding of their thermal metamorphism. **Citation:** Gattacceca, J., P. Rochette, F. Lacroix, P.-E. Mathé, and B. Zanda (2011), Low temperature magnetic transition of chromite in ordinary chondrites, *Geophys. Res. Lett.*, 38, L10203, doi:10.1029/2011GL047173.

1. Introduction

[2] Knowledge of the magnetic field of our solar system in the past opens a window into the activity of the young Sun and differentiation processes of asteroids and planets [e.g., Weiss *et al.*, 2010]. This knowledge may be acquired from paleomagnetic and mineral magnetic studies on meteorites. Magnetic characterization of meteorites is also a sensitive tool to better constrain their mineralogy [e.g., Rochette *et al.*, 2005], petrogenesis [e.g., Rochette *et al.*, 2009], and/or petrofabrics [e.g., Gattacceca *et al.*, 2008]. In most meteorites, including ordinary chondrites (OC), the main ferromagnetic minerals at room temperature are FeNi alloys, in the form of kamacite, taenite, and to a lesser extent tetrataenite and awaruite. However, at low temperature (LT), other minerals can become ferromagnetic (*sensu lato*) and contribute to the magnetic properties and remnant magnetization of meteorites.

[3] Recently, through LT magnetic measurements, Kohout *et al.* [2007] identified the contribution of daubreelite (FeCr₂S₄) in an enstatite chondrite. They observed a Curie temperature at 170 K and a second magnetic transition at 60 K. Previously, LT magnetic transition attributed to chromite was observed in the Martian meteorite ALH84001 near 90 K [Weiss *et al.*, 2004], and in the Apollo lunar basalt 12063 near 100 K [Banerjee, 1972]. It has been proposed recently that troilite

(FeS, antiferromagnetic at room temperature), an almost ubiquitous mineral in meteorites with typical modal abundances of several %, becomes a weak ferromagnet below 70 K [Kohout *et al.*, 2007, 2010].

[4] In this paper, we further investigate the LT magnetic properties of OC, and discuss the origin of a magnetic transition observed around 65 K. We show that chromite, not troilite, is responsible for this transition, and discuss the implications of this observation.

2. Material and Methods

[5] LT magnetic measurements were performed primarily at CEREGE (Aix-en-Provence, France) with a Princeton[®] Micromag Vibrating Sample Magnetometer equipped with a LT cryostat. The maximum applied field is 1 T, the moment sensitivity is $\sim 5 \cdot 10^{-9}$ Am², and the working temperature range is 10–400 K. Additional measurements were performed with a MPMS from Quantum Design[®], both at IPGP (Paris, France) and at IRM (Minneapolis, USA). This instrument has a moment sensitivity of 10^{-11} Am², and can operate in the 1.9–400 K temperature range. The VSM offers more flexibility for field control and allows rapid measurement of hysteresis cycles. The MPMS offers better temperature control. Results obtained with the two instruments are almost identical (Figure 1b).

[6] Measurements were performed on 33 OC samples in the 15–175 mg range. Most samples originate from the collection of the Museum of Natural History (Paris, France). Only meteorite falls were chosen to avoid oxy-hydroxides that are formed during terrestrial weathering of FeNi alloys. Samples were selected to cover the different groups (H, L, and LL), and the different petrologic types (from 3 to 6) as defined by Van Schmus and Wood [1967]. Samples and their magnetic properties are listed in Table 1.

[7] In order to observe possible magnetic transitions, we measured temperature dependence of the saturation isothermal remanent magnetization (SIRM) acquired at LT (typically 10 K) on heating. Most measurements were performed with the VSM, with a temperature sweep rate of 5 K/min. LT SIRM was acquired after field cooling (FC) in 1 T or after zero field cooling (ZFC). In the latter case SIRM was imparted in a 2.5 T field. Hysteresis loops were measured at room temperature for all samples and at variable LT for six selected samples.

3. Results of Low Temperature Magnetic Measurements

[8] In all studied OC samples, both unequilibrated (petrographic type 3) and equilibrated (petrographic types 4, 5 and 6), the LT SIRM is partially unblocked at LT, as evidenced by a non monotonic decrease of SIRM during

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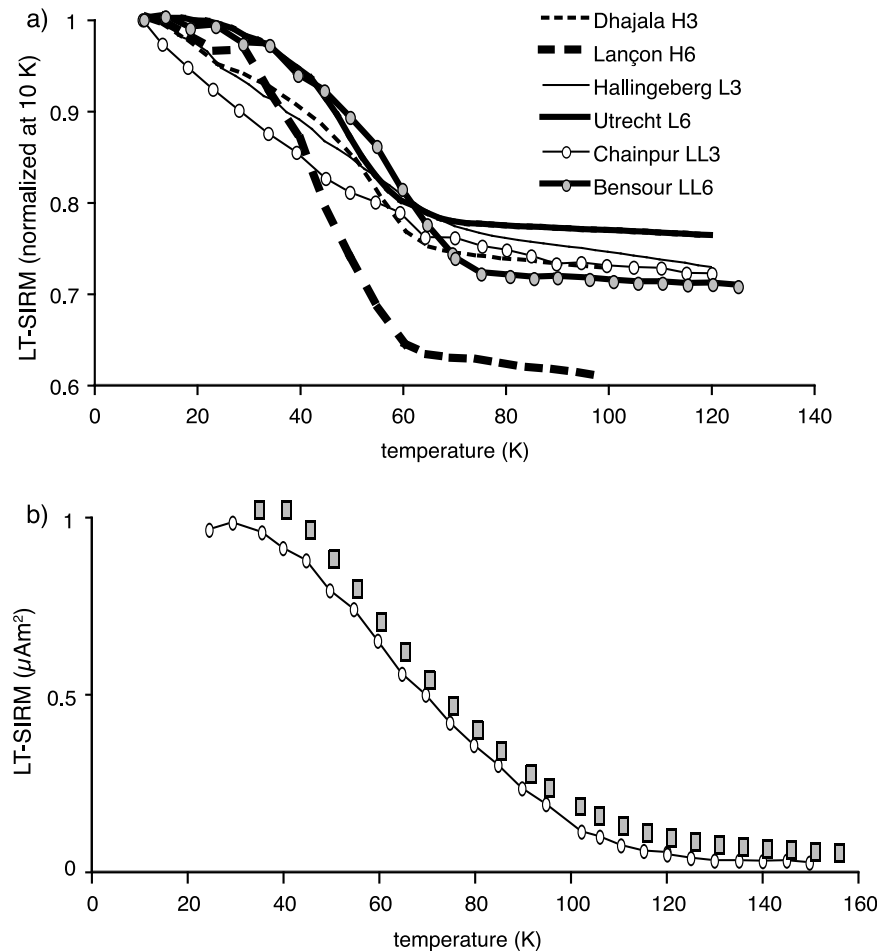


Figure 1. (a) Thermal evolution of FC LT SIRM for a representative set of ordinary chondrite samples. There is no significant evolution of the SIRM in the 120 K to 300 K temperature range, so the temperature scale is cut at 120 K. (b) Thermal evolution of ZFC LT SIRM for a natural terrestrial chromite. The same sample was measured with a VSM (circles) and a MPMS (boxes).

heating (Figure 1). During subsequent cooling in zero field, there is no recovery of the remanent magnetization, which shows that the decay of SIRM is not attributable to a magnetic phase transition within a ferromagnetic state, and corresponds to standard thermal demagnetization. FC and ZFC experiments give the same results (Figure S1 of the auxiliary material) indicative of a low coercivity (soft) ferrimagnet.¹ The maximum unblocking temperatures were evaluated using the two-tangent method of Grommé *et al.* [1969]. Results are summarized in Table 1. These temperatures range from 40 to 80 K, with 90% of data clustered in the 55–80 K range, and a mean value of 66 ± 9 K (all mean values in this article are given $\pm$ s.d.). The mean amplitude of this magnetic transition is 88 ± 24 mAm²/kg ($n = 32$), excluding Semarkona (LL3.00 chondrite) which is a clear outlier. The residual LT SIRM above the transition is due to the FeNi alloys present in the meteorites: kamacite, taenite, tetrataenite, and martensite [Uehara *et al.*, 2011].

[9] In the same temperature range in which the LT SIRM is unblocked, hysteresis properties undergo significant changes, with a decrease of saturation magnetization (M_S), saturation remanent magnetization (M_{RS}) and coercivity B_C

(Figure 2). Hysteresis loops saturate below 1 T (Figure S2). Although the M_S data are noisy due to the correction of high-field susceptibility (computed over the 0.9–1 T field interval), the decrease of M_S with increasing temperature indicates that a ferromagnetic mineral is going through its Curie temperature (T_C). The mean variation of M_S across T_C is 168 ± 53 mAm²/kg ($n = 5$), the variation of M_{RS} being 100 ± 16 mAm²/kg ($n = 6$), i.e., undistinguishable of the variation of the LT SIRM across the magnetic transition computed on the same six samples (95 ± 27 mAm²/kg). The M_{RS}/M_S of the phase undergoing a magnetic disordering across the transition can thus be estimated to be 0.67.

[10] The absence of variation of the SIRM acquired just above the transition temperature upon cooling down to 10 K and warming up to the transition temperature (Figure S1) is an additional proof that this transition corresponds to a Curie temperature, and not to another type of crystallographic transition.

4. Discussion

4.1. Chromite

[11] Chromite (FeCr_2O_4) is an ubiquitous accessory mineral in meteorites, in particular in OC. Cr_2O_3 concentrations in OC are very constant, with a mean value of

¹Auxiliary materials are available in the HTML. doi:10.1029/2011GL047173.

Table 1. Studied Samples and Their Magnetic Properties^a

Sample	Group	Mass (mg)	Low Temperature T _C	LT-SIRM at 20 K (mAm ² /kg)	LT-SIRM Lost Across T _C (mAm ² /kg)	M _{RS} at 300 K (mAm ² /kg)	M _S at 300 K (Am ² /kg)
Dhajala	H3	121	66	306	66	278	33.9
Forest Vale	H4	30	52	115	59	54.6	36.9
Ochansk	H4	28	57	493	86	367	46.4
Sainte Marguerite*	H4	176	40	84	84	0.0178	0.11
Agen	H5	93	57	499	83	442	28.5
Pultusk	H5	67	63	431	100	303	42.7
Kernouvé	H6	69	42	426	71	320	42.9
Lançon	H6	71	58	194	65	243	41.6
Tieschitz	H/L3	20	75	528	95	359	14.1
Hallingeberg	L3	27	70	463	85	341	21.7
Khohar	L3	57	75	542	38	364	20.7
Mező-Madaras	L3	59	80	343	76	221	14.9
Kendleton	L4	24	67	348	117	222	10.7
Saratov	L4	45	66	485	115	377	20.1
Homestead	L5	72	66	445	86	346	11.7
Tadjera	L5	138	65	115	52	55.4	14.2
Utrecht	L6	76	60	562	114	472	16.7
Bishunpur	L/LL3	62	78	502	80	381	15.3
Bjurböle	L/LL4	28	59	534	61		
Cynthiana	L/LL4	21	67	560	80		
Semarkona	LL3	26	63	1380	326	567	5.08
Chainpur	LL3	94	75	425	89	195	5.48
Parnallee	LL3	70	75	318	62	225	6.32
Soko-Banja	LL4	73	62	518	105	403	4.09
Olivenza	LL5	27	76	677	80	572	2.5
Tuxtuac	LL5	45	71	83	76	8.64	0.43
Douar Mghila	LL6	51	70	485	118	357	2.5
Jelica	LL6	90	70	176	91	84.2	0.97
Kilabo	LL6	38	69	114	94	20.7	1.97
Kilabo	LL6	16	70	578	140	423	1.6
Oued el Hadjar	LL6	52	63	583	151	412	2.49
Bensour	LL6	29	70	280	74	215	1.32
Benguerir	LL6	59	64	604	114	431	4.43

^aT_C is computed from the LT SIRM data.^bMeasurement on a single separated chondrule.

0.52 ± 0.05 wt% (n = 175 [Jarosewich, 1990]). Chromite being the only significant Cr-bearing phase, an average mass content of 0.77 wt % can be computed for chromite in OC. Several LT phase transitions are known in chromite as evidenced by LT measurement of the heat capacity of chromite that show three anomalies at 37 K, 69 K and 124 K, the Curie point being the second one [Klemme *et al.*, 2000]. Below ordering temperature it shows a ferrimagnetic behavior like most cubic spinels and therefore can carry a significant spontaneous remanent magnetization [e.g., Shirane *et al.*, 1964]. One should thus expect to detect chromite T_C on LT measurements of either saturation remanence, low field susceptibility or saturation magnetization. We present in Figures 1b and 2 the results of LT magnetic measurements on natural terrestrial chromite-bearing ores (from the Tiebaghi mine in New-Caledonia) obtained with the same experimental procedure than the measurements on meteorites. These measurements show a T_C of the studied terrestrial chromites in the 100–120 K range. Pure chromite is reported to have a T_C of 70 [Robbins *et al.*, 1971; Klemme *et al.*, 2000] to 80 K [Shirane *et al.*, 1964], and a saturation magnetization M_S of 16 Am²/kg [Robbins *et al.*, 1971]. Natural chromite shows a number of solid solutions with various substitutions affecting Fe²⁺ and Cr³⁺. In equilibrated OC, the range of substitution is quite narrow [Johnson and Printz, 1991; Wlotzka, 2005]. Typical composition for equilibrated L/LL and LL chondrites (as cation per total of

3 cations after Johnson and Printz [1991]) is Cr 1.56, Fe 1.00, Al 0.21, Mg 0.10, Ti 0.06, V as well as Mn 0.02, the rest being Si, Zn, etc. (each <0.01).

[12] Predicting precisely the T_C of such a composition is impossible because each substitution cannot be accounted independently. Qualitatively we can estimate the tendencies based on linear interpolation from T_C of end members: MnCr₂O₄ (45 K), Mg or ZnCr₂O₄ (15 K), FeAl₂O₄ (13 K), and FeV₂O₄ (109 K) [Klemme *et al.*, 2000; Masrour *et al.*, 2010; Schieber, 1967]. V³⁺ and Mn²⁺ substitutions (by the same amount) have a very small net effect (ΔT_C of +29 K and −35 K respectively). Al and Mg substitutions of 0.10 and 0.21 reduces T_C from 75 (assumed T_C for FeCr₂O₄) to ≈15 K (T_C of MgCr₂O₄ and FeAl₂O₄) by 10 and 10.5%, respectively, i.e., results in a predicted T_C of 63 K. Ti⁴⁺ substitution, as it implies a correlative addition of divalent cation can have contrasted effects depending on the added cation. Banerjee [1972] reports a T_C of 100 K (i.e., ΔT_C of 25 K) for a 0.14 substitution of Ti and Fe (Fe_{1.14}Cr_{1.7}Ti_{0.14}O₄). The substitution of 0.14 Cr³⁺ by Fe³⁺ (Fe_{1.14}Cr_{1.86}O₄) results in a T_C of 93 K [Robbins *et al.*, 1971]. Fe³⁺ substitution is usual in terrestrial rocks, but should be minimal for equilibrated OC due to low oxygen fugacity (indicated by the presence of Fe⁰). Therefore the observed mean T_C of 66 K is compatible with the composition of OC chromites. The mineral that loses its ferrimagnetism at 66 K in ordinary chondrites has a M_{RS}/M_S of 0.67 (§3). Using this M_{RS}/M_S

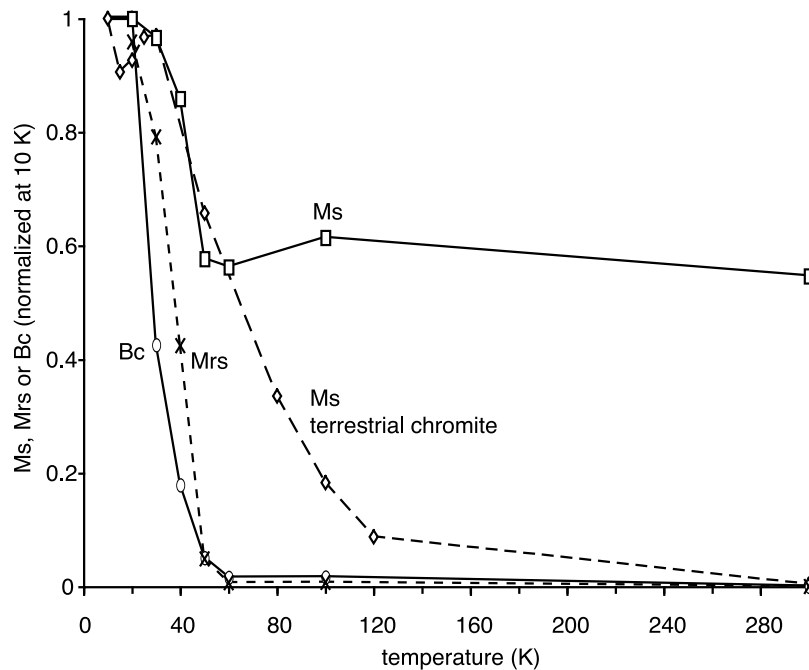


Figure 2. Evolution of hysteresis properties as a function of temperature for a separated chondrule from Ste-Marguerite OC. This sample was selected because of its very low metal content which allows better observation of the magnetic signal of chromite in particular for saturation magnetization. A curve for a terrestrial chromite-bearing ore is given for comparison (broken line).

value, a chromite content in OC of 0.77 wt %, and M_S value for pure chromite ($16 \text{ Am}^2/\text{kg}$), we can predict a LT M_{RS} of $83 \text{ mAm}^2/\text{kg}$ due to chromite in OC. This is in perfect agreement with our measurements ($88 \pm 24 \text{ mAm}^2/\text{kg}$). Using the M_S variation across T_C revealed by hysteresis measurements ($168 \pm 53 \text{ mAm}^2/\text{kg}$, see §3), we can compute an average M_S of $22 \pm 7 \text{ Am}^2/\text{kg}$ for chromite in OC, again in agreement with pure chromite data. However, one assumption for these quantitative analyses is that the chromite magnetic grain size is uniform between samples because M_{RS} is a grain size (magnetic domains) dependent magnetic property.

4.2. Troilite and FeNi Minerals

[13] Previous observation of a magnetic transition near 70 K (resulting in a decrease of remanence and induced magnetization upon heating) in a troilite extract of Bruderheim (L6) has been attributed to an intrinsic property of troilite [Kohout *et al.*, 2007]. Recent measurements on a troilite extract from the Cape York iron meteorite [Kohout *et al.*, 2010] evidence the same transition at 70 K. The SIRM of troilite below 70 K was estimated by Kohout *et al.* [2007] to be about $0.46 \text{ Am}^2/\text{kg}$. Troilite, being a major constituent of ordinary chondrites, could thus be proposed to explain our data instead of chromite. However, it is antiferromagnetic at room-temperature, and no LT phase transition of any kind has ever been evidenced in this major phase [e.g., Coey and Rouxboisson, 1979]. The absence of transition at 70 K during the zero field cooling of room temperature SIRM (Figure S1) also strongly argues against the existence of a Morin type transition advocated by Kohout *et al.* [2007]. Moreover, using the LT SIRM value proposed by Kohout *et al.* [2007] to account for our observed LT M_{RS} data would require 21 wt. % troilite in OC, a value incompatible

with that obtained from chemical analysis of OC: $5.45 \pm 1.15 \text{ wt. \%}$ ($n = 175$ [Jarosewich, 1990]). To provide a final test of our conclusion, we powdered two samples that had been already measured (Kilabo, 37.5 mg, and Agen), and leached them in concentrated HCl at 80°C for 4 hours, a treatment that should have dissolved most troilite present [e.g., Torigoye and Shima, 1991]. Remeasurement of LT SIRM heating curves reveals that the 70 K transition is unchanged (Figure S3). Therefore, we conclude that the troilite extract from Bruderheim that was used by Kohout *et al.* [2007, 2010] was contaminated by chromite, and that troilite remains a pure antiferromagnet at any temperature below its Néel point. Such a contamination is likely if the troilite fraction was obtained by a combination of magnetic extraction and heavy liquid extraction, leaving the dense and paramagnetic or antiferromagnetic minerals together (i.e., troilite + chromite). Moreover, chromite is usually found in contact with troilite or in inclusion within troilite [e.g., Ramdohr, 1967; Rubin, 2003]. Therefore hand-picking of troilite grains under a binocular microscope cannot prevent significant chromite contamination. From the $0.46 \text{ Am}^2/\text{kg}$ transition obtained by Kohout *et al.* [2007] on “troilite” and the $11.2 \text{ Am}^2/\text{kg}$ LT SIRM of chromite (see above), we estimate that a contamination of the troilite extract by chromite of only about 4 wt. % is enough to account for their low temperature observations.

[14] Our data show that there is no correlation between M_S or M_{RS} measured at room temperature, and the amplitude of the LT transition in OC (Table 1). M_S being proportional to the bulk amount of FeNi metallic minerals, and M_{RS} being indicative of the tetrataenite amount, this observation rules out the hypothesis that the LT magnetic transition in OC is due to kamacite, taenite, or tetrataenite. Nagata and Carleton [1989] had measured the LT properties

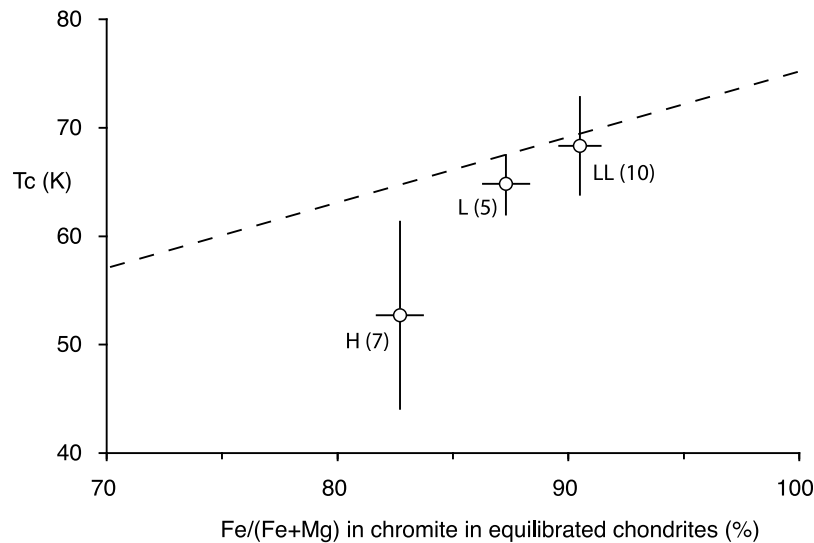


Figure 3. Correlation between average chromite Curie point (from Table 1, with indication of the number of data) and ratio Fe/(Fe+Mg) after Wlotzka [2005] on equilibrated H, L and LL chondrites. The broken line shows the expected correlation for a pure $\text{Fe}_{(1-x)}\text{Mg}_x\text{Cr}_2\text{O}_4$ solid solution, using a linear interpolation from end members.

of “non metallic matrix” of the Saint-Séverin LL6 ordinary chondrite, and observed an increase in coercivity and M_{RS} below 70 K, which they tentatively interpreted as a magnetic transition in tetrataenite. Based on close similarity with our LL6 data, their observation is attributable to chromite which is present in the non magnetic fraction of the matrix.

4.3. Implications for Ordinary Chondrites

[15] Having attributed unambiguously the LT magnetic transition in OC to chromite, we can discuss the insights given by these magnetic measurements into OC mineralogy.

[16] It is noticeable that within a given chondrite group, the amplitude of the LT magnetic transition (and hence, to the first order, the total amount of chromite in the samples) is restricted to a rather narrow range (38–151 mAm^2/kg for the whole dataset, excluding the anomalous sample Semarkona), and is much less variable than the room temperature M_{S} and M_{RS} that are controlled by the amount of FeNi metal grains. This reflects the smaller grain size of chromite and its homogeneous distribution at the scale of the studied samples (a few tens of mg), with respect to the large FeNi metal grains whose distribution is heterogeneous at this scale, especially in metal-poor LL chondrite [e.g., Gattacceca et al., 2003].

[17] We observe a slight increase of the transition amplitude from H to L and LL chondrites with median values of 77, 85, and 93 (all in $10^{-2} \text{ Am}^2/\text{kg}$) respectively. However, the Cr_2O_3 amount is essentially the same in all three groups: 0.50 wt. % for H and L, 0.52 wt. % for LL [Jarosewich, 1990]. We also observe a slight increase of the transition temperature from H, to L, and LL chondrites. The mean transition temperature for equilibrated chondrites are 53, 65, and 68 K for H, L, and LL chondrites respectively (Figure 3). This evolution reflects an increase in Fe content and decrease in Mg content from H to LL [Wlotzka, 2005]. These chemical differences are probably also responsible for the observed differences in the transition amplitude.

[18] It can also be observed in Figure 1a that the transition is sharper for equilibrated OC than for unequilibrated ones.

This reflects the fact that the composition of chromites in unequilibrated OC covers a wider composition spectrum than in equilibrated ones [Johnson and Printz, 1991; Krot et al., 1993], and therefore result in a spread of unblocking temperatures. The median transition temperature for unequilibrated OC (75 K, $n = 8$) is significantly higher than for equilibrated ones (64 K, $n = 22$). This is also attributable to the spread of unblocking temperatures, because the two-tangent method used to determine the transition temperature is biased towards the maximum blocking temperature.

5. Conclusions

[19] We observe a LT magnetic transition in all thirty-three ordinary chondrites we studied, with a Curie temperature in the 40–80 K range (average 66 K). Comparison with existing LT magnetic data, and bulk chemical analyses points unambiguously towards chromite (with varying degree of substitution by Al, Mg, ...) as the mineral responsible for this transition. Previous studies attributing such transition to troilite or tetrataenite should be reappraised.

[20] Although the T_{C} of chromite is way too low to make it a potential carrier of remanent magnetization in any present day environment in the Solar system (a 75 K temperature will be achieved only by a body located farther than the orbit of Saturn [e.g., Spencer et al., 1989]), the ferromagnetic properties of chromites in chondrites may have had some implications for the magnetic aggregation of chromite grains in the solar nebula, as already proposed by Krot et al. [1993]. Magnetic clumping is a major process accounting for dust aggregation the Solar nebula [e.g., Dominik and Nübold, 2002].

[21] On the other hand, LT magnetic properties give some insights into OC mineralogy. Firstly, it appears that chromites in H, L and LL chondrites, have distinct chemical composition that can be detected using T_{C} value. Secondly, magnetic measurements are sensitive to the larger spread in chromite compositions of unequilibrated OC versus equilibrated ones. As a sensitive indicator of the chemical composition spectrum

of chromite population, and in view of the high sensitivity of chromite to thermal metamorphism in chondrites [Johnson and Printz, 1991], we surmise that further studies may be able to establish a robust link between LT magnetic properties and incipient thermal metamorphism in ordinary chondrites.

[22] The identification of ferrimagnetic chromite at LT in chondrites also offers a new potential technique for separating chromite grains in these meteorites through magnetic sorting at LT in liquid nitrogen, taking advantage of the increase of its magnetic susceptibility at this temperature.

[23] **Acknowledgments.** We thank the two anonymous reviewers for their helpful comments. This work was funded by the Agence Nationale de la Recherche (project ANR-09-BLAN-0042). The MPMS XL-5 EverCool at IPGP was financed through the Région Ile-de-France SESAME program, CNRS-INSU equipment grant, BQR-IPGP and ANR-06-JCJC-0144 research grants. IPGP contribution 3153.

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