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Magnetic study of magnetite in the Tagish Lake meteorite

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Abstract–The saturation magnetization, saturation remanent magnetization, the coercive, and remanent coercive force were determined at room and liquid nitrogen temperatures for three pieces of the Tagish Lake meteorite. The results are compared to similar data for four other chondrites (Allende, Murray, Orgueil, and Murchison). The data suggests that the Tagish Lake meteorite is magnetically homogeneous, and is not as magnetically hard as the comparison chondrites. The magnetization measurements indicate that it contains about 10–11% multi-domain magnetite. Magnetic susceptibility measurements on all the samples from 77 K to room temperature showed a Verwey transition for all the samples which contain a significant amount of multi-domain magnetite. The coercive force data further indicate that the magnetite in Tagish Lake is multi-domain and that the grain size is small and approximately 4–9 μm.

INTRODUCTION

The general characteristics of the Tagish Lake meteorite which fell in British Columbia, Canada in January 2000 are described by Brown *et al.* (2000). The Tagish Lake meteorite is a C2 chondrite and has a significant amount of magnetite, which is finely disseminated throughout the matrix (Zolensky, 2001, pers. comm.). The Tagish Lake meteorite (19.3% iron) is compared magnetically with meteorites containing similar amounts of iron, namely the Murray (20.71% iron), the Murchison (22.13% iron), Orgueil (17.4% iron), and the Allende meteorite (23.85% iron) (see Brown *et al.*, 2000; Jarosewich, 1971; Wiik, 1956; Clarke *et al.*, 1970; von Michaelis *et al.*, 1969; Ahrens *et al.*, 1969). In addition, the low-field magnetic susceptibility was measured as a function of temperature from 77 K to room temperature for all the meteorites in order to observe and compare the Verwey transitions. Both the magnetic hysteresis loop data and the Verwey transition were used to estimate and compare the grain size of the magnetite in the Tagish Lake with that in the comparison meteorites.

EXPERIMENTAL

The three samples of the Tagish Lake meteorite were from separate pieces of the fall, but their relative spatial relationship is unknown. One sample (24-24) was divided into two parts prior to making the measurements. All three of the initial samples were quite friable and had to be handled with care.

Hysteresis loops were run on all of the Tagish Lake samples and also on the comparison meteorites. The magnetic measurements were made with both a static Faraday-type Cahn balance and a modified Princeton Applied Research vibrating sample magnetometer. Measurements were carried out as a function of temperature between room temperature and liquid nitrogen temperature. Details of the measuring techniques can be found elsewhere (Thorpe *et al.*, 1984; Alexander *et al.*, 1979).

From the hysteresis loops the saturation magnetization, J_S, the remanent saturation magnetization, J_{RS}, and the coercive force, H_C, were determined. After saturation at high fields, the applied field is reduced to zero leaving a remanent saturation magnetization, J_{RS}. If the field is subsequently increased gradually in a negative direction, the magnetization is forced to zero, at a field H_C, the coercive force. If the field is further increased in a negative direction, the magnetization is reduced to a negative value. If the negative field is removed, the value of the magnetization rises to a small value, which is less than J_{RS}. However, there is a unique negative field, which if applied and then removed, will result in the magnetization returning to zero. This field, H_R, is the remanent coercive force, and is the field at which the sample is completely demagnetized. H_R at room temperature and H'_R at liquid temperatures were determined for all of the samples.

Static magnetic susceptibility measurements were also made at a low field (2×10^4 Am^{-1}) on the Tagish Lake samples and on the comparison meteorites as a function of temperature to observe the temperature of the Verwey transition from magnetite.

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GENERAL RESULTS

The hysteresis loops for all of the Tagish Lake specimens were very similar. A hysteresis loop for one of the specimens (24-24) is shown in Fig. 1a. The loop is saturated at a field of 4×10^5 Am^{-1} which suggests that this meteorite may have a significant concentration of magnetite. The approach to saturation of the hysteresis loop is similar to that for the Orgueil meteorite (not shown) which is known to contain ~11.9% magnetite (Larson *et al.*, 1974). We assume that magnetite is the source of the saturation behaviour, because there is essentially no metallic iron present in this meteorite (Zolensky,

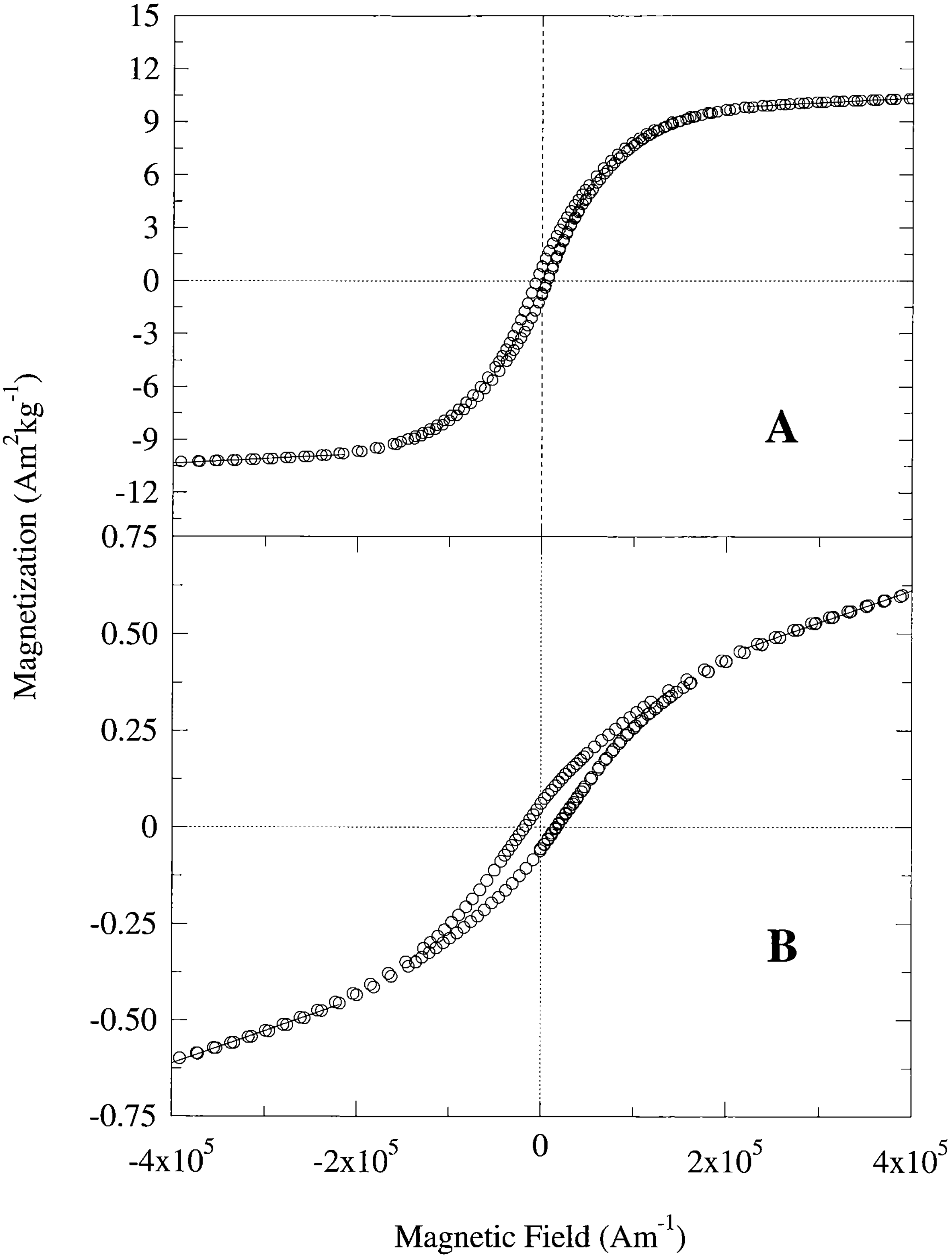

FIG. 1. The hysteresis loops determined for the Tagish Lake meteorite (A), and the Allende meteorite (B). Loop A is saturated at 4×10^5 Am^{-1}, whereas loop B is not.

2001, pers. comm.), and also because magnetite has been reported to be present in the Tagish Lake meteorite by Brown *et al.* (2000). In contrast to the hysteresis loop for the Tagish Lake meteorite, the hysteresis loop for the Allende meteorite is shown in Fig. 1b. At the same field (4×10^5 Am^{-1}) the magnetization is still increasing significantly, and suggests the presence of either small superparamagnetic grains of magnetite, some paramagnetic material, or both are responsible for the lack of saturation.

The observed saturation of the Tagish Lake hysteresis loop (Fig. 1a) in contrast to the Allende hysteresis loop suggests that there are less single and/or subdomain particles in the Tagish Lake specimen compared to the Allende meteorite. Brecher and Arrhenius (1974) in studying the hardness of the demagnetization spectra of natural and saturation remanence in several meteorites also concluded that there is a high content of single and subdomain particles in the Allende meteorite. It seems evident from the small values of J_S (Table 1) that a significant amount of the total iron in the Allende meteorite is in the form of some paramagnetic iron mineral. Thus, if the magnetite is largely in the form of subdomain particles, the contribution of superparamagnetism will be considerably smaller than the paramagnetic mineral contribution to the gradual approach to saturation.

From the hysteresis loops and other measurements described above, the magnetic parameters at room temperature are shown in Table 1, and at liquid nitrogen temperature in Table 2. The data for the four Tagish Lake specimens are comparable and indicate the magnetic homogeneity of this meteorite. In contrast, the Allende meteorite is heterogeneous (see Wasilewski, 1981).

The room-temperature values of the coercive force, H_C, for the Allende, Murray, Orgueil, and Murchison samples are ~2.5× that of the Tagish Lake meteorite (Table 1). It is well known that H_C for multi-domain magnetite is larger for samples with a small grain size, and that H_C decreases as the size increases (*e.g.*, see Fuller, 1984; also Fig. 12-4 in Dunlop and Özdemir, 1997). Although Tanaka *et al.* (1997) has cautioned that the source and preparation of magnetite can influence the value of H_C, it would seem that the relationship between H_C and grain size presented by Dunlop and Özdemir (1997) can be used for meteorites. As it is dubious that magnetite formed hydrothermally in space, we used their curve for crushed grains, rather than their curve for hydrothermal magnetite. With this in mind and using our values of H_C, we estimate the grain size of magnetite in the Tagish Lake meteorite to be about 7–9 μm. As the H_C measured for the comparison meteorites are all about the same value, we use a mean value of 1.6 and estimate a grain size of the order of 1.1 μm, which is ~6× smaller than the grain size of the Tagish Lake meteorite. From the grain size plot in Dunlop and Özdemir (1997) (their Fig. 6.7) one can estimate the magnetite grains consists of about two domains. This roughly agrees with Brecher and Arrhenius (1974) who determined the Orgueil meteorite contained 80% multi-domain magnetite grains. Most single domain magnetite grains are <0.1 μm (Dunlop and Özdemir, 1997), and thus the grain size of magnetite in the Tagish Lake meteorite is probably multi-domain with a larger grain size than the Orgueil and the other comparison meteorites.

The grain size of magnetite can also be estimated from the ratio J_{RS}/J_S using the crushed grain curve (see Fig. 12.3 in Dunlop and Özdemir, 1997). Using mean values of the ratio

TABLE 1. Measured magnetic parameters at room temperature for several specimens of the Tagish Lake meteorite.*

Sample Number and weight	Remanent coercive force H_R ($\times 10^4$ Am^{-1})	Coercive force H_C ($\times 10^4$ Am^{-1})	Saturation magnetization J_S (Am^2 kg^{-1})	Remanent saturation magnetization J_{RS} (Am^2 kg^{-1})	$R_H = H_R/H_C$	$R_J = J_{RS}/J_S$
Tagish Lake meteorite						
24-24, 38.0 mg	2.13	0.648	12.6	0.87	3.49	0.069
24-24, 14.87 mg	2.23	0.65	11.35	0.927	3.43	0.081
MM-95, 15.78 mg	2.04	0.533	8.8	0.51	3.65	0.058
MG-03, 7.9 mg	n.d.	0.638	9.0	0.53	n.d.	0.060
Allende, Murray, Orgueil, and Murchison meteorites						
Murray, USNM 6651, 70.35 mg	3.98	1.58	0.503	0.152	2.51	0.288
Murchison, USNM 5453, 37.89 mg	5.05	1.56	0.343	0.091	3.24	0.265
Murchison, USNM 5453, 60.42 mg	4.98	1.46	0.378	0.091	3.44	0.241
Allende, 41.62 mg	6.80	1.73	0.278	0.060	3.94	0.216
Orgueil, 17.32 mg	5.76	1.64	9.42	1.22	3.51	0.129

*Similar measurements on the Allende, Murray, Orgueil, and Murchison meteorites are shown for comparison.
n.d. = not detected.

TABLE 2. Measured magnetic parameters at liquid nitrogen temperature (primed symbols) for the Tagish Lake meteorite.*

Sample Number and weight	Remanent coercive force H'_R ($\times 10^4$ Am^{-1})	Coercive force H'_C ($\times 10^4$ Am^{-1})	Saturation magnetization J'_S (Am^2 kg^{-1})	Remanent saturation magnetization J'_{RS} (Am^2 kg^{-1})	$R'_H = H'_R/H'_C$	$R'_J = J'_{RS}/J'_S$
Tagish Lake meteorite						
24-24, 38.0 mg	3.29	1.77	13.8	2.10	1.95	0.152
24-24, 14.87 mg	3.13	1.7	12.9	2.15	1.84	0.166
MM-95, 15.78 mg	3.18	1.57	9.9	1.27	2.02	0.128
MG-03, 7.9 mg	n.d.	2.07	10.1	1.49	n.d.	0.147
Allende, Murray, Orgueil, and Murchison meteorites						
Murray, USNM 6651, 70.35 mg	6.9	2.43	0.493	0.214	2.83	0.434
Murchison, USNM 5453, 37.89 mg	7.22	2.30	0.332	0.141	3.14	0.425
Murchison, USNM 5453, 60.42 mg	7.91	2.23	0.364	0.144	3.54	0.396
Allende, 41.62 mg	9.71	2.13	0.276	0.087	4.35	0.283
Orgueil, 17.32 mg	6.95	3.18	10.17	2.00	2.18	0.200

*Similar measurements on the Allende, Murray, Orgueil, and Murchison meteorites are shown for comparison.
n.d. = not detected.

from Table 1 for the Tagish Lake meteorite, we estimate a grain size of about 6–7 μm, that is, somewhat less, but about the same as that estimated from the H_C values. The ratio of J_{RS}/J_S for the comparison meteorites cluster around a mean value of 0.26, if one does not include the Orgueil meteorite, which is somewhat lower. Using 0.26 for the ratio we estimate the grain size to be centered about 0.2–0.3 μm for the comparison meteorites (*i.e.*, a grain size ~4× smaller than the value estimated from the H_C data on the same meteorites). The source of this discrepancy is not clear, but it is probably related to the smaller grain size in the comparison meteorites (see Dunlop, 1973). It is interesting to note that the ratio (0.129) for the Orgueil meteorite is equivalent to a grain size of about 0.6–1.3 μm, which is comparable to the value determined from the H_C data, but smaller than the size estimated for the Tagish Lake meteorite grain size.

Assuming the data by Dunlop and Özdemir (1997) to be valid for magnetite in meteorites, their data (see their Fig. 6.7) indicates that for the above estimated grain sizes, the magnetite grains in the Tagish Lake and Orgueil meteorites contain about three to four, and two domains, respectively. A grain size of 0.2–0.3 μm for the comparison meteorites suggest that they contain less than two domains or are probably subdomain in nature. In the case of the Tagish Lake and Orgueil meteorites, the grain size is significantly larger than that of the comparison meteorites.

Dunlop and Özdemir (1997) have also plotted the ratio $R_J = J_{RS}/J_S$ as a function of $R_H = H_R/H_C$ for hydrothermal and crushed natural magnetites for a large number of samples (their Fig. 11.23). Their data fall in a relatively linear narrow band. Using the data in Table 1 we have superimposed our data on theirs (not shown). The Tagish Lake and Orgueil data fall within the band for magnetite, whereas the data for the other comparison meteorites fall well outside the band. This clearly indicates that the magnetic properties of the Tagish Lake and Orgueil meteorites are primarily dictated by the presence of multi-domain magnetite; the magnetic characteristics of the other comparison meteorites are due to other magnetic minerals, small grain size, *etc.* The larger grain size of the magnetite in the Tagish Lake meteorite, compared to the other meteorites, suggest that these meteorites are magnetically harder than the Tagish Lake meteorite.

The coercive force of the Tagish Lake meteorite increases almost 3× at liquid nitrogen temperatures (see Table 3), which is to be expected for multi-domain magnetite. In contrast, the comparison meteorites (with exception of the Orgueil) which contain more single and subdomain particles, show a smaller increase in H_C at liquid nitrogen temperature. As described by Kakol (1990), Moskowitz *et al.* (1998), and also by Schmidbauer and Keller (1996), the cubic anisotropic constant of multi-domain magnetite changes sign at a temperature of ~130 K, the isotropic point, T_i (just above the Verwey transition), and causes the coercive force to increase dramatically as the temperature decreases through T_i. Muxworthy (1999) shows the relationship between H_C and temperature, and the transition at T_i (T_k in Fig. 3 in Muxworthy, 1999). His relatively large increase in H_C at low temperatures explains the increase in H_C in the Tagish Lake meteorite at low temperatures, and is similar to that found for magnetite by Nagata *et al.* (1964). The change in H_C for both the Tagish Lake and Orgueil meteorites between room and liquid nitrogen temperatures is also in agreement with Muxworthy's work.

The saturation magnetization at 77 K is slightly higher than at room temperature as shown by the ratio J'_S/J_S in Table 3. It

TABLE 3. Ratios of the data taken at liquid nitrogen temperature (primed symbols) to the room temperature data.

Sample Number and weight	H'_R/H_R	H'_C/H_C	J'_S/J_S	J'_S/J_{RS}	R'_H/R_H	R'_J/R_J
Tagish Lake meteorite						
24-24, 38.0 mg	1.54	2.73	1.09	2.41	0.56	2.20
24-24, 14.87 mg	1.40	2.79	1.21	2.53	0.50	2.10
MM-95, 15.78 mg	1.56	2.95	1.13	2.49	0.55	2.21
MG-03, 7.9 mg	–	3.24	1.12	2.81	–	2.45
Allende, Murray, Orgueil, and Murchison meteorites						
Murray, USNM 6651, 70.35 mg	1.73	1.54	0.98	1.41	1.13	1.51
Murchison, USNM 5453, 37.89 mg	1.43	1.47	0.97	1.55	0.97	1.60
Murchison, USNM 5453, 60.42 mg	1.59	1.53	0.96	1.58	1.03	1.64
Allende, 41.62 mg	1.43	1.23	0.99	1.45	1.10	1.31
Orgueil, 17.32 mg	1.21	1.94	1.08	1.64	0.62	1.55

is well known that the saturation magnetization of magnetite increases as the temperature decreases (*e.g.*, Matsui *et al.*, 1977). The fact that the ratio is slightly >1 for the Tagish Lake and Orgueil meteorites tends to verify the presence of significant magnetite in these two meteorites.

In Fig. 2 the ratio, R_J, of the saturation remanent magnetization, J_{RS}, to the saturation magnetization, J_S, is plotted as a function of H_C (similar to a plot for carbonaceous chondrites by Sugiura, 1977). Similar to the data for the other carbonaceous chondrites, the Tagish Lake data also fall on the Sugiura plot, but at a lower ratio than the comparison meteorites. Again, this indicates that the Tagish Lake specimens are not as magnetically hard as some of the other high-iron meteorites such as the Murray, Murchison, Orgueil, or Allende meteorites. It should also be noted that the data points for all four specimens of the Tagish Lake meteorite are clustered together, whereas the data points for the Allende, Orgueil, and Murray meteorites show some spread in the data for different samples of the same meteorite. Again, this points to the greater homogeneity of the Tagish Lake meteorite.

In the Tagish Lake chondrite the value for R_H (ratio of H_R/H_C) at room temperature is ~3.6, a value not too different from the comparison meteorites (see Table 1). As discussed by Wasilewski (1973), the lower limit of the ratio is 1.0, and that for single domain particles, the Stoner–Wohlfarth model predicts a value of $R_H \approx 1.09$ (Stoner and Wohlfarth, 1948). However, if the specimen contains coarse multi-domain dispersions, a ratio closer to 4 can be expected. It appears therefore that the magnetite grains in the Tagish Lake meteorite are much larger than single domain particles, and probably in the form of coarse multi-domain particles.

VERWEY TRANSITION

If the Tagish Lake meteorite has a substantial amount of multi-domain magnetite, as the saturation magnetization measurements and other data suggests, then one should be able to observe a Verwey transition at ~120 K. At this temperature there is a conversion of the cubic lattice of Fe_3O_4 to an orthorhombic structure, which results in a magnetic susceptibility discontinuity. Figure 3 shows how the magnetic susceptibility changes with temperature for a bulk magnetite sample (Ward's research grade), and the conversion transition at ~120 K is obvious. Curve A in Fig. 4 is a similar plot for the Tagish Lake meteorite. For comparison, similar data are plotted for the Orgueil (curve B), the Murchison (curve C) and the Allende (curve D) chondrites. The change in susceptibility at the Verwey transition for the Orgueil meteorite is similar to that of the Tagish Lake meteorite. This is to be expected because they both have about the same amount of magnetite and the size of the magnetite is comparable (0.6–1.3 μm for Orgueil; 6–9 μm for Tagish Lake). However, the transition temperature is slightly different for the Orgueil and this may be due to some substitution of Ni for Fe as suggested by Banerjee and Hargraves (1971). The Murchison is reported to have from 0.75 ± 0.04% to 1.66 ± 0.8% magnetite (Watson *et al.*, 1975; Hayes *et al.*, 1986), and, as shown in Fig. 4, appears to have a small Verwey transition. In curve D in Fig. 4, there is no observable Verwey transition in the Allende chondrite. This is in agreement with Wasilewski (1981), who also reported no observable Verwey transition in the Allende chondrite. If the concentration of Fe_3O_4 is <1%, or, if the grain size of the magnetite is so small that they are single or subdomain particles (Hayes *et al.*, 1986), then no Verwey transition will be observable.

Muxworthy (1999) has measured the Verwey transition of magnetite consisting of several particle sizes, and he shows a significant decrease in the magnitude of the transition as the particle size increases from 3.0 to 108 μm. Similar to reports by Belov (1993) and Hodych (1986), Muxworthy also show a small peak at the Verwey transition for the 3.0 μm particles of Fe_3O_4, but not for the larger particles. This peak rises to ~7% above the magnetic susceptibility on the high-temperature side

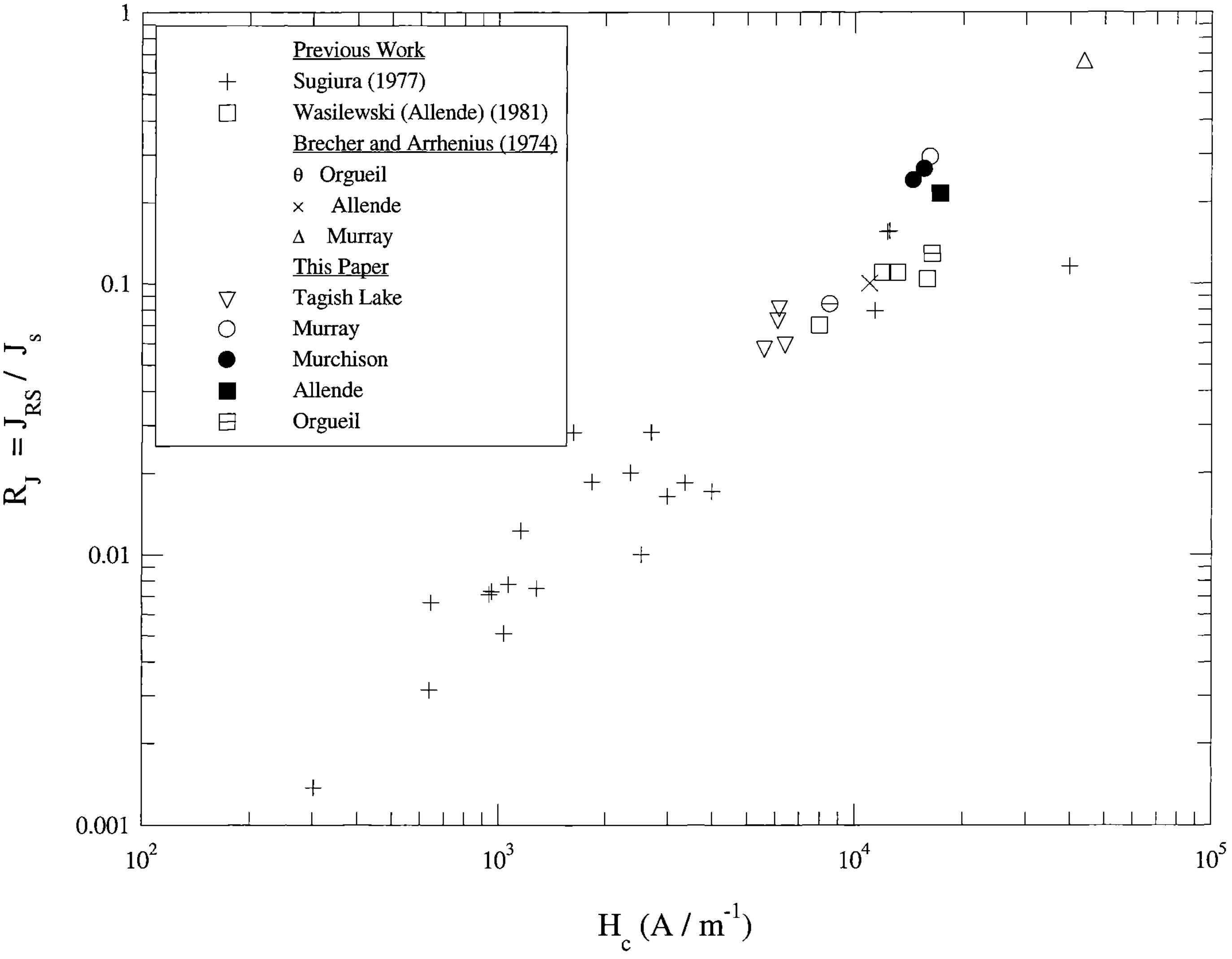

FIG. 2. The ratio, R_J, of the saturation remanent magnetization, J_{RS}, to the saturation magnetization, J_S, as a function of the coercive force, H_C (see Sugiura, 1977; Wasilewski, 1981; Brecher and Arrhenius, 1974).

of the transition. Note that the peak is not observed in Fig. 3 for bulk magnetite. However, a similar small peak can be seen for the Tagish Lake meteorite (Fig. 4a). It rises to about the same height above the high-temperature magnetic susceptibility. Likewise a similar peak can be seen for the Orgueil meteorite (curve B in Fig. 4) which also rises ~7% above the susceptibility on the high-temperature side of the transition. The source of this peak is not clear, but from Muxworthy's work it would seem to be associated with the particle size. In fact, Muxworthy suggests that this peak might be used as an indicator of particle size. Although the data does not lend itself to an accurate measurement of the peak height, its presence in the Tagish Lake meteorite would seem to indicate that the particle size of the magnetite particles of the Tagish Lake and Orgueil meteorites is >3 μm, the largest particle size measured by Muxworthy (1999). This is about the same order of magnitude as our estimates from H_C and the ratio R_J. In any event the grain size of magnetite in the Tagish Lake and Orgueil meteorites appears to be about the same as Muxworthy's magnetite sample (3 μm).

Assuming that magnetite in the Tagish Lake meteorite is stoichiometric and is the principal contributor to the saturation magnetization, one can estimate the concentration of Fe_3O_4 in the Tagish Lake meteorite. If the mean value of J_S is ~9.9 $Am^2 kg^{-1}$ (see Table 1), the estimated concentration of magnetite is ~10.7% (using 92 $Am^2 kg^{-1}$ for 100% magnetite). This value of the magnetite concentration is an upper limit, as there are probably minor amounts of other magnetic minerals contributing to the saturation magnetization. Alternatively, and assuming the magnetite in the Tagish Lake meteorite is multidomain, one may obtain an estimate of the magnetite concentration from the Verwey transition. From Fig. 3, the transition for 100% Fe_3O_4 is ~22 000 $m^3 kg^{-1}$ whereas in Fig. 4a the transition for Tagish Lake meteorite is ~2400 $m^3 kg^{-1}$.

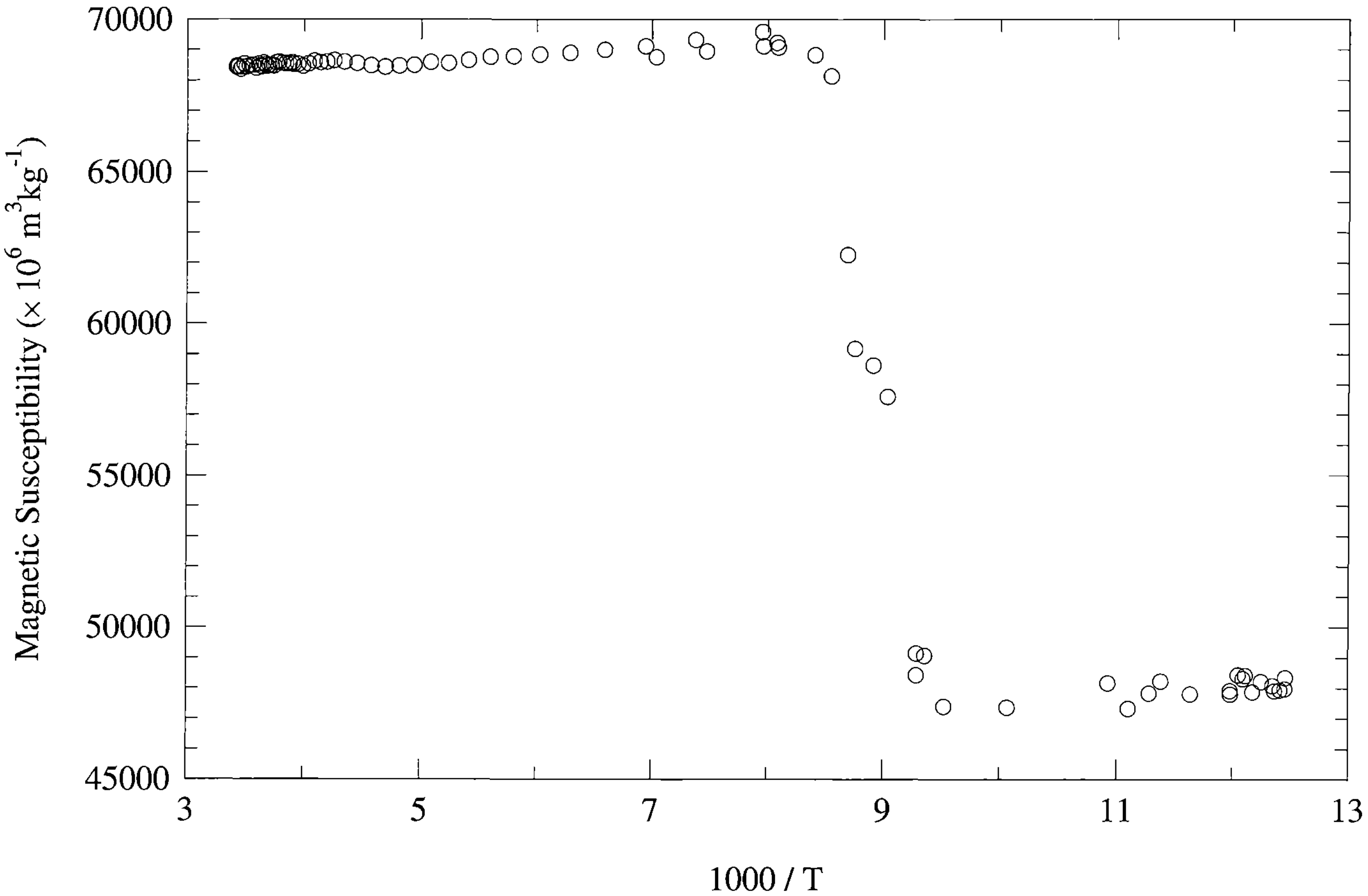

FIG. 3. The magnetic susceptibility (at a magnetic field of 2×10^4 Am^{-1}) as a function of temperature from 77 to 290 K for research grade magnetite (Ward's).

Assuming the conditions of particle size, *etc.* in Figs. 3 and 4a are not significantly different, one can estimate a concentration of 10.9% magnetite, which is about the same concentration determined from the saturation magnetization. The close correspondence between the calculations made from the saturation magnetization and the Verwey transition would seem to indicate that the broad assumptions in the calculations are justified. However, this must be considered an upper limit of the concentration, and only an approximate calculation. The low values of J_S and the small Verwey transition for the Murchison and Allende meteorites indicates that there is only a small amount of magnetite in the specimens (<2% magnetite). However, the value of J_S indicates that the observed value of H_C is probably due to magnetite, and therefore substantiates the grain size measurements described above.

DISCUSSION AND CONCLUSIONS

There is no doubt, but that magnetite is the major magnetic mineral in the Tagish Lake meteorite. The values of H_C at room temperature are relatively small and commensurate with a maximum particle size of about $(4–9) \times 10^3$ nm. This size is well above that for single domain particles, and, as attested to by the well-developed Verwey transition, the magnetite in the Tagish Lake meteorite, is multi-domain. This agrees with the discussion of the value of H_C and also of R_J which predicts that the magnetite is in the form of relatively small multi-domain particles.

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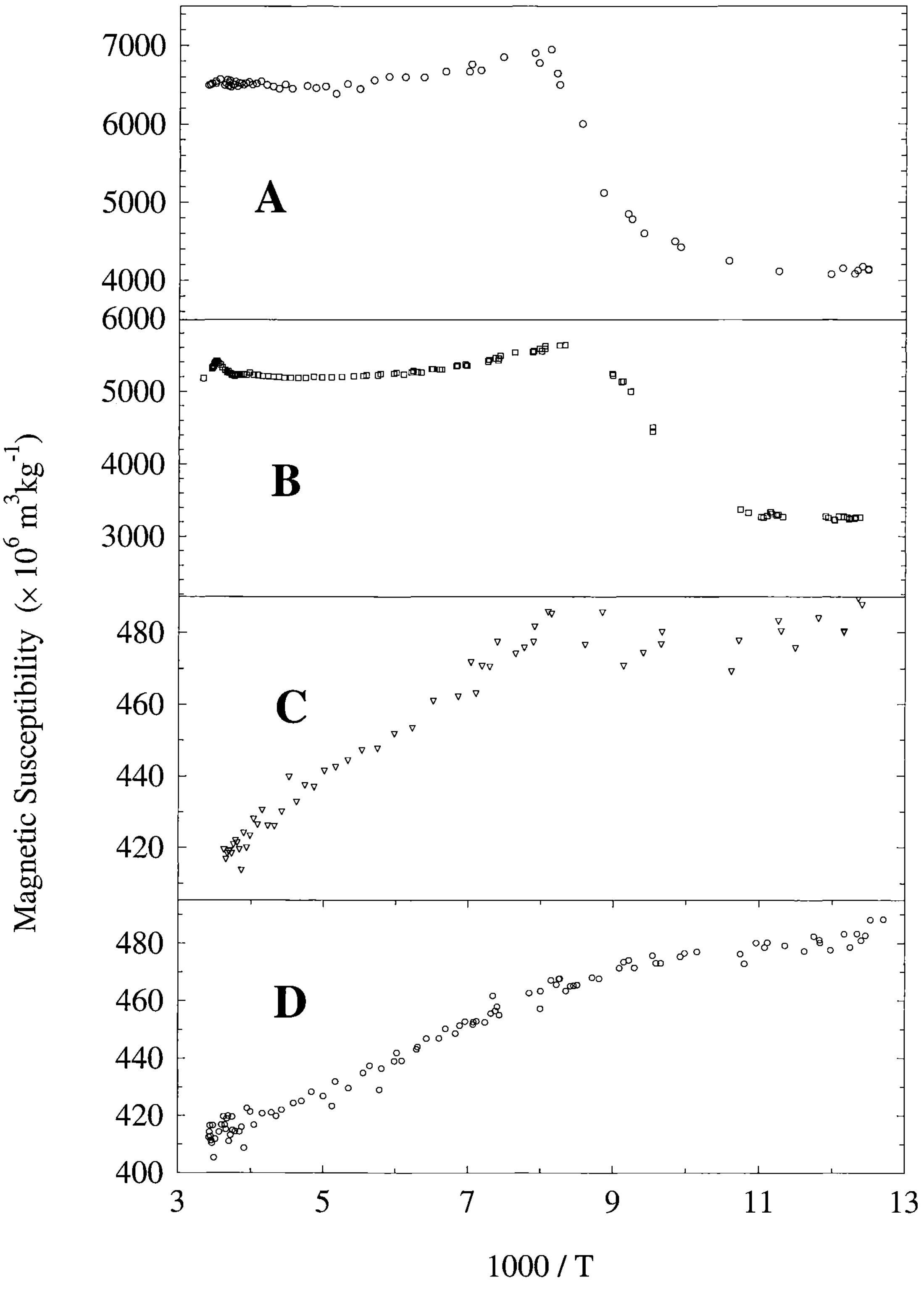

FIG. 4. The Verwey transition curves for the Tagish Lake (curve A), the Orgueil (curve B), the Murchison (curve C), and the Allende (curve D) chondrites. The prominent Verwey transition in curves A and B indicates the presence of a significant concentration of multi-domain magnetite compared to the Murchison and Allende meteorites. The Verwey transition was not observed in the Allende meteorite, probably due to the low concentration of Fe_3O_4, and subdomain particle size.

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