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Quantifying transdomain transitions in three dimensions and the limits of classical and metastable single-domain behavior using field-impressed magnetic anisotropy

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[1] Some domain imaging studies have been used as evidence for transdomain transitions and metastable single-domain states in particles larger than theoretical single-domain (SD) size. Domain imaging has certain limitations since observations are made of a two-dimensional surface, which is generally polished and may introduce stress effects, and usually a relatively small number of particles are analyzed. The present paper shows how field-impressed anisotropy of magnetic susceptibility provides independent quantitative support in three dimensions for transdomain transitions and metastable SD states. In particular, changes from positive to negative impressed anisotropy (prolate to oblate impressed ellipsoids) in a direct field (DF) of increasing strength are consistent with some small multidomain (MD) particles undergoing transitions to metastable SD states. The results suggest that DF treatment can quantify the particle size limits of metastable SD behavior. In contrast, an alternating field (AF) should not produce a metastable SD state in particles greater than theoretical SD size and experimentally produces positive impressed anisotropy in MD particles and negative values only in intrinsic SD particles. Consequently, AF treatment can be used to determine the particle size limits of theoretical classical (not metastable) SD behavior. Consistent results were obtained for particle size fractions of magnetite, titanomagnetite, pyrrhotite, and hematite. The results may also help to explain recent observations concerning the low-field variation of AC susceptibility of various minerals. Field-impressed anisotropy is a quantitative, rapid, three-dimensional technique, requiring no extra sample preparation (thus not introducing stress effects), and representative of a statistically large number of particles.

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1. Introduction

[2] The recognition that nominal multidomain (MD) particles can exhibit different numbers of domains dependent upon their magnetic history, and can undergo transdomain transitions upon application of a direct field (DF) isothermally, or upon temperature cycling in a weak field, has been a major advance in rock magnetism and paleomagnetism over the last 25 years. Much of the pioneering work has been done via domain imaging of titanomagnetite [*Halgedahl and Fuller*, 1980, 1983; *Metcalf and Fuller*, 1986; *Halgedahl*, 1991], magnetite [*Heider et al.*, 1988; *Williams and Dunlop*, 1990; *Heider and Hoffmann*, 1992; *Geiss et al.*, 1996], pyrrhotite [*Halgedahl and Fuller*, 1980], hematite [*Halgedahl*, 1995], and titanomaghemite [*Geiss et al.*, 1996]. This has led to further theoretical and experimental work on nucleation theory, LEM states [*Boyd et al.*, 1984; *Moon and Merrill*, 1985; *Merrill and Halgedahl*, 1995] and transdomain thermoremanent magnetization [*Dunlop et al.*, 1994]. A key observational feature is that particles which theoretically would be regarded as multidomain (MD) can often appear to be in a metastable SD remanent state upon cycling in a DF [e.g., *Halgedahl and Fuller*, 1980, 1983], or upon acquisition of weak field (comparable to the Earth's field) thermoremanent magnetization [*Metcalf and Fuller*, 1986; *Halgedahl*, 1991]. A number of questions have always remained regarding the domain images. First, are the two-dimensional domain images representative of the actual domain states in three-dimensional particles? Second, are stress effects, from polishing the surface of the samples in some studies, having an influence on the observed patterns [*Moskowitz et al.*, 1988]? Third, are the images of a few particles representative of the large numbers of particles in an average rock sample?

[3] One of the main purposes of the present paper is to demonstrate that measurements of field-impressed anisotropy of magnetic susceptibility can help to resolve some of these issues. Field-impressed anisotropy (FIA) has several advantages which makes it a very useful potential tool to complement domain imaging studies. It is rapid and non-

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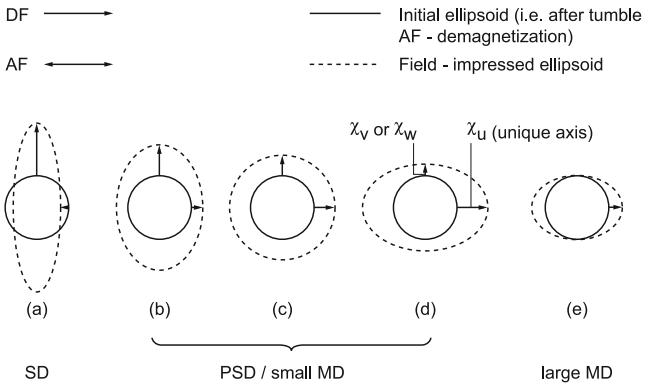

Figure 1. Schematic diagram (adapted from *Potter and Stephenson* [1990a]) illustrating previous trends of field-impressed effects for MD magnetite and SD maghemite. The field-impressed ellipsoids are ellipsoids of revolution with the unique axis (χ_u) aligned with the applied field axis. Field-impressed ellipsoids in Figures 1a and 1b are thus oblate and those in Figures 1d and 1e are prolate. AF treatment gave only oblate impressed ellipsoids for intrinsic SD particles and prolate impressed ellipsoids for all intrinsic MD particles. While DF also gave oblate impressed ellipsoids for SD particles, small MD particles exhibited a progressive change from prolate to oblate ellipsoids with increasing DF strength. Large MD particles only exhibited prolate impressed ellipsoids upon DF treatment.

destructive and requires no extra preparation of the sample. The latter advantage means that no polishing of the sample surface is required, and thus no stress-induced polishing effects are introduced. Moreover, the field-impressed ellipsoid is a quantitative measure of the changes in three dimensions, unlike two-dimensional domain images. Also, since field-impressed anisotropy is a bulk measurement, it is representative of a statistically large number of particles, whereas the practicalities of domain imaging (mainly in terms of time constraints) mean that generally only a relatively small number of particles are analyzed. In general, SD and small (submicron) MD particles exhibit the largest field-impressed effects and thus can give important information when the resolution of conventional domain imaging techniques is limited.

[4] The application of magnetic fields to paleomagnetically important minerals has been shown to alter their low-field (initial) susceptibility. *Stacey* [1961, 1963] first suggested that the application of a strong AF should cause domain alignment in MD particles and that this should result in a small anisotropy of magnetic susceptibility (AMS). Subsequently, *Bhathal and Stacey* [1969] found experimentally that such an anisotropy was indeed produced in igneous rocks and synthetic samples containing magnetite in various size ranges (from 16 to 600 μm). Further studies found effects due to an AF or DF in rocks [*Violat and Daly*, 1971; *Kapicka*, 1981], and other minerals such as hematite [*Schmidt and Fuller*, 1970; *Lecaille and Daly*, 1972; *Zapletal*, 1985; *Hejda et al.*, 1992], pyrrhotite [*Rochette*, 1988], and greigite [*Sagnotti and Winkler*, 1999]. *Potter and Stephenson* [1990a] first coined the term "field-impressed anisotropy" to describe the effects and developed a methodology for systematically quantifying the effects in three dimensions. They found that for isotropic or weakly anisotropic samples an AF or DF impressed an ellipsoid of revolution with the unique axis parallel to the applied field axis. Their results provided a rapid technique to identify the predominant domain state (SD or MD) within a sample. Intrinsic SD particles exhibited negative field-impressed anisotropy (Figure 1) with the unique axis of the impressed ellipsoid of revolution being the minimum axis (oblate impressed ellipsoid), whereas MD particles exhibited positive field-impressed anisotropy with the unique axis of the impressed ellipsoid of revolution being the maximum axis (prolate impressed ellipsoid). An aspect of their work not previously discussed in detail, and which forms the basis for the present paper, is that it provides independent support for the existence of transdomain transitions, metastable SD states, and different nonequilibrium local energy minima (LEM) states. Any observed field-impressed susceptibility anisotropy in MD material should only occur if there is a change in the domain state and/or structure (that is, a change in the number of domains present and/or a rearrangement in the orientation of the domain walls). The fact that one does observe measurable changes in anisotropy after the application of an AF or DF is strong indirect evidence for the applied fields causing changes in the domain state and/or structure.

[5] The *Potter and Stephenson* [1990a] study highlighted certain distinct differences between the application of an AF and a DF, particularly for MD particles. For all the MD magnetite samples studied (nine natural magnetite particle size fractions from <0.7 to 63 μm and one Mapico synthetic magnetite sample with particles 0.2–0.8 μm) the application of an AF resulted in positive values of field-impressed anisotropy of magnetic susceptibility [*Potter and Stephenson*, 1990a, Figure 3]. The field-impressed anisotropy was only negative after AF treatment if the particles were intrinsically SD [*Potter and Stephenson*, 1988a, 1990a, 1990b]. In contrast, the application of a DF to the MD magnetite fractions gave positive impressed anisotropy at low fields, but then reached a peak and was followed by a decrease in impressed anisotropy at higher fields up to the maximum applied field investigated (80 mT). In one case, the smallest natural magnetite sample (<0.7 μm), the impressed anisotropy actually went negative at the higher fields [*Potter and Stephenson*, 1990a, Figure 7]. The fundamental differences between AF and DF treatment will be shown in the present work to potentially enable one to quantify the particle size limits of both intrinsic (what one might term classical) SD behavior and metastable SD behavior.

[6] A previous simple model involving domain rearrangement and interactions between domains in MD particles was developed to help quantify and explain the above experimental effects [*Stephenson and Potter*, 1996]. The model predicted that at higher saturating fields almost all of the MD size fractions would exhibit a negative impressed anisotropy [*Stephenson and Potter*, 1996, Figure 5]. The model, however, did not predict the crossover from positive to negative impressed anisotropy in a systematic way with particle size, and also suggested that the 7.6–13.1 μm fraction would not go negative. There seemed to be no good reason for this when other larger particle sizes were predicted to go negative. There were also other discrep-

ancies between the modeled and experimental changes in the overall dimensions of the impressed ellipsoid. Moreover, the model assumed a constant number of domains within a MD particle when the DF was applied and then removed, whereas in practice the size of the forward domains increases and the number of domains decreases. Therefore, while the model represented a useful approach for quantifying the effects, it was not a completely accurate physical representation of what occurs in reality.

[7] There is, however, a physical mechanism that could cause a progressive change from positive to negative field-impressed anisotropy with increasing DF strength in intrinsically MD particles of a mineral such as magnetite. That mechanism is the transition to a metastable SD state of some of the particles within the sample as the DF strength increases. A uniaxial SD particle theoretically has zero susceptibility parallel to its easy magnetization direction with a maximum susceptibility perpendicular to that direction [*Stephenson et al.*, 1986; *Potter and Stephenson*, 1988b; *Potter*, 2004]. Therefore, if a significant number of particles within a sample undergo a transition to a metastable SD state upon application of a strong DF, as has been suggested from some domain studies [e.g., *Halgedahl and Fuller*, 1980, 1983], then the susceptibility parallel to the magnetization direction will decrease in comparison to the perpendicular direction, resulting in a negative field-impressed anisotropy. If this happens then one would expect more negative impressed anisotropies to occur systematically as the particle size decreases in MD magnetite particles, since the smaller particles are more likely to be capable of undergoing a transition to a metastable SD state. Hence the present paper aimed to extend the previous work of *Potter and Stephenson* [1990a] and *Stephenson and Potter* [1996] on magnetite by investigating experimentally the effect of higher applied DF strengths on the impressed anisotropy (the previous experimental work having been undertaken up to 80 mT so that the samples could be subsequently AF demagnetized). This would test whether the impressed anisotropy actually goes negative for any the other magnetite size fractions as predicted in the *Stephenson and Potter* [1996] model. It would also test whether the smaller fractions exhibit more negative impressed anisotropy in a systematic way with decreasing particle size (which was not the case in the *Stephenson and Potter* [1996] model predictions), since this would be a strong indicator that the smaller particles within the fractions had undergone a transition to a metastable SD state.

[8] This study also extends the field-impressed susceptibility anisotropy measurements to investigate the differences between the effect of an applied AF and that of a DF on discrete size fractions of other paleomagnetically important minerals: titanomagnetite (TM60 in this case), pyrrhotite, and some preliminary work on hematite. In particular, it focuses on relevant minerals and particle sizes that have been the subject of previous domain image studies involving possible transdomain transitions and metastable SD states [*Halgedahl and Fuller*, 1980, 1981, 1983; *Heider and Hoffmann*, 1992; *Halgedahl*, 1995; *Geiss et al.*, 1996]. A further motivation for this study is that it may help to explain, and support, some recent results on the low-field variation of AC susceptibility in magnetite, titanomagnetite and pyrrhotite [*Hrouda et al.*, 2006].

2. Samples and Methods

[9] Powders of magnetite (<0.7–63 μm), titanomagnetite TM60 (0.7–7.6 μm), monoclinic pyrrhotite (0.8–75 μm) and hematite (up to 76 μm) were separated further into various discrete particle size fractions. The overall ranges reflected where possible those observed in the previous domain imaging studies mentioned above. A Bahco centrifugal dust classifier was used to produce the finer fractions of magnetite, TM60 and pyrrhotite. Sieves were used to separate the larger particles of magnetite (>45 μm), as well as two hematite fractions. The pyrrhotite fractions differed very slightly in size from the magnetite and TM60 fractions, as they were separated at a different time on a different dust classifier. Small concentrations of each of the powders (about 1% by volume) were dispersed and set in resin. The resulting samples were close to being isotropic or very weakly anisotropic in terms of their intrinsic AMS. Most samples had an AMS of around 0.5–2%, where% AMS is given by $100\{(\max - \min)/\text{total}\}$.

[10] Field-impressed anisotropy of susceptibility was determined using the methodology described by *Potter and Stephenson* [1990a]. Each sample was initially tumble AF demagnetized. Three components of induced magnetization were measured when a weak field was applied along the x axis, and likewise along the y and z axes in turn, producing a total of 9 components of susceptibility as follows:

$$\begin{array}{ll} \text{Field along x} & \chi_{1x}, \chi_{1y}, \chi_{1z} \\ \text{Field along y} & \chi_{2x}, \chi_{2y}, \chi_{2z} \\ \text{Field along z} & \chi_{3x}, \chi_{3y}, \chi_{3z} \end{array} \quad (1)$$

Note that theoretically $\chi_{1y} = \chi_{2x}$, $\chi_{1z} = \chi_{3x}$ and $\chi_{2z} = \chi_{3y}$. The results were then used to define the initial ellipsoid with principal values χ_{11}, χ_{22}, χ_{33}, and the principal axes having an orientation relative to the sample axes (x, y, z) were computed from the results. A strong field (AF or DF) was then applied to each weakly anisotropic sample at some arbitrary orientation, and this changed the anisotropy of susceptibility, which could then be measured in a similar way producing a further nine components of susceptibility:

$$\begin{array}{ll} \text{Field along x} & \chi'_{1x}, \chi'_{1y}, \chi'_{1z} \\ \text{Field along y} & \chi'_{2x}, \chi'_{2y}, \chi'_{2z} \\ \text{Field along z} & \chi'_{3x}, \chi'_{3y}, \chi'_{3z} \end{array} \quad (2)$$

where $\chi'_{1y} = \chi'_{2x}$, $\chi'_{1z} = \chi'_{3x}$ and $\chi'_{2z} = \chi'_{3y}$, and a new ellipsoid with principal values χ'_{11}, χ'_{22}, χ'_{33} along with the new orientations was computed. The "difference ellipsoid", which arises from the application of the field, was then computed from the difference between the two sets of components:

$$\begin{array}{l} \left(\chi'_{1x} - \chi_{1x}\right), \left(\chi'_{1y} - \chi_{1y}\right), \left(\chi'_{1z} - \chi_{1z}\right) \\ \left(\chi'_{2x} - \chi_{2x}\right), \left(\chi'_{2y} - \chi_{2y}\right), \left(\chi'_{2z} - \chi_{2z}\right) \\ \left(\chi'_{3x} - \chi_{3x}\right), \left(\chi'_{3y} - \chi_{3y}\right), \left(\chi'_{3z} - \chi_{3z}\right) \end{array} \quad (3)$$

This "difference ellipsoid", with principal values χ_u, χ_v, χ_w (see Figure 1), specifies the changes produced by the field. Note that this procedure quantifies the changes produced by

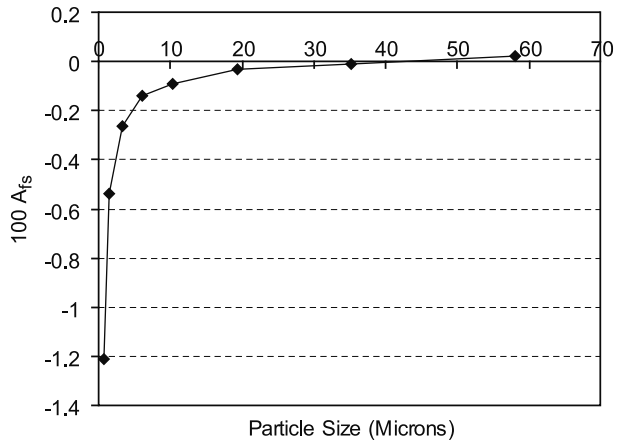

Figure 2. Values of the percentage impressed susceptibility anisotropy (100 A_{fs}) after a DF of 800 mT was applied to a series of magnetite fractions. The <0.7 μm fraction is plotted at 0.7, and the median grain size of all the other fractions is plotted. The measurement errors in 100 A_{fs} are of the same order of magnitude as the symbol size.

the field in three dimensions. The effect of an AF or DF imparted along a single axis to an isotropic or weakly anisotropic sample was found experimentally by *Potter and Stephenson* [1990a] to impress an ellipsoid of revolution (the "difference ellipsoid") with the unique axis (χ_u) aligned with the applied field axis, so that effectively $\chi_v = \chi_w$. In practice there were usually slight experimental differences between χ_v and χ_w within the measurement uncertainties so it is convenient to take an average $(\chi_v + \chi_w)/2$. Any significant differences between χ_v and χ_w (above the measurement uncertainties) might potentially give further information on domain wall orientations in MD material. (Note that for an intrinsically strongly anisotropic sample the field-impressed ellipsoid would not be expected to be an ellipsoid of revolution and some degree of triaxial behavior is likely to occur). It is useful to express the changes in terms of a field-impressed anisotropy of magnetic susceptibility parameter, A_{fs}, given by

$$A_{fs} = \{\chi_u - (\chi_v + \chi_w)/2\}/\chi'_t \quad (4)$$

where $\chi'_t = \chi'_{11} + \chi'_{22} + \chi'_{33}$ and is the sum of the principal susceptibility axes of the field-treated ellipsoid. Note that A_{fs} varies from 0 to 1 for a field-impressed prolate ellipsoid and from 0 to -0.5 for an oblate ellipsoid. It is useful to plot the percentage field-impressed anisotropy (100 A_{fs}). In practice the applied fields were given via a Digico demagnetizer for the AF case, and a Molspin pulse magnetizer for the DF case. The anisotropy of susceptibility was measured using a Digico anisotropy delineator [*Hrouda et al.*, 1983] in conjunction with a Molspin susceptibility bridge, which measures the z axis (bulk) susceptibility.

3. Results

3.1. Magnetite

[11] The percentage field-impressed susceptibility anisotropy (100 A_{fs}) produced by a strong DF of 800 mT (essentially a saturating field ten times greater than that used in the previous experiments by *Potter and Stephenson* [1990a]) applied along the z axis of seven MD magnetite fractions are shown in Figure 2. This shows that the smallest size fractions exhibit by far the most negative values of field-impressed anisotropy. Note also that there is an essentially smooth asymptotic trend of less negative field-impressed anisotropy with increasing particle size. For particle sizes above about 25 μm the impressed anisotropy essentially does not go negative within the measurement errors (which are close to the size of the symbols) compared to the tumble AF demagnetized state (zero on the 100 A_{fs} scale). Indeed for all sizes greater than 10 μm the impressed anisotropy after an 800 mT DF is very small compared to the tumble AF demagnetized state. There are, however, distinct changes in anisotropy at lower fields, and Figure 3 shows the details for some of the different fractions with increasing applied DF strength. For all fractions the impressed anisotropy starts off positive, reaches a maximum and then starts to decrease. Significantly, the point at which the impressed anisotropy goes negative is at higher applied fields as the particle size increases. (Note that fractions 7.6–13.1 μm and 13.1–25.5 μm are not shown in Figure 3 for clarity but also follow this trend.)

3.2. Titanomagnetite

[12] Figure 4 shows the results for three particle size fractions of TM60 after the application of different applied DF strengths up to 80 mT along the z axis of each sample. The maximum applied field was restricted to 80 mT as there was limited TM60 powder and the samples needed to be demagnetized for other experiments. The results exhibited a similar trend to those of magnetite in that the smallest fractions showed the most negative impressed anisotropy, and the crossover from positive to negative values of 100 A_{fs} is at higher fields as the particle size increases. However, compared to magnetite of equivalent particle size, the TM60 samples start to show negative field-impressed anisotropy at lower applied fields. This is exemplified by comparing the 0.7–2.2 μm fraction for magnetite with an equivalent fraction for TM60 in Figure 5. The magnetite sample starts to go negative at around 80–90 mT, whereas

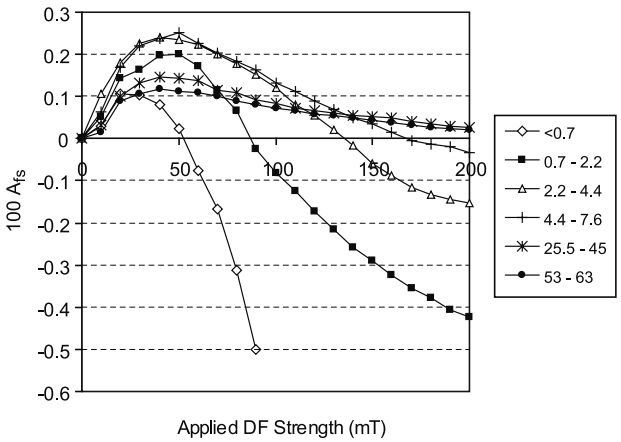

Figure 3. Values of the percentage impressed susceptibility anisotropy as a function of applied DF strength for selected magnetite fractions. The numbers in the legend refer to the particle sizes in microns.

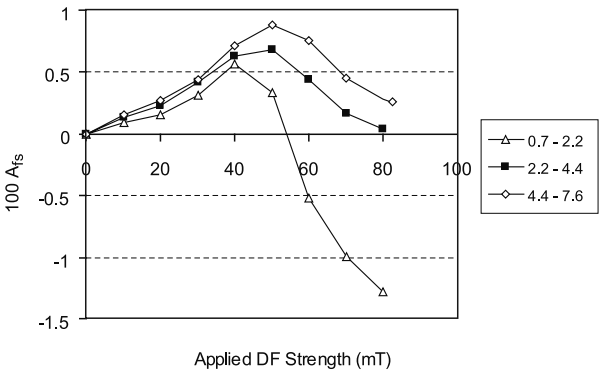

Figure 4. Percentage impressed susceptibility anisotropy as a function of applied DF strength for TM60 fractions (the numbers in the legend are the particle sizes in microns).

the TM60 sample starts to go negative at 50–60 mT. Another significant difference is that the magnitude of the impressed anisotropy (both the positive impressed anisotropy at low fields, and the negative impressed anisotropy at high fields) is larger for TM60.

[13] Figure 6 shows the field-impressed anisotropy results for TM60 due to AF treatment. The results show a similar pattern to those of magnetite [*Potter and Stephenson*, 1990a, Figure 3], with all of the fractions exhibiting a positive impressed anisotropy. However, as for the DF case, the magnitude is larger for TM60 than was the case for magnetite. A comparison of the 0.7–2.2 μm fractions for magnetite and TM60 for AF treatment bears this out (Figure 7).

3.3. Pyrrhotite

[14] Figure 8 shows the field-impressed anisotropy results for the application of a DF up to 80 mT along the z axis of eight monoclinic pyrrhotite fractions. As for the TM60 samples, the maximum applied field was restricted to 80 mT, since the pyrrhotite samples needed to be demagnetized and used for other experiments. The samples

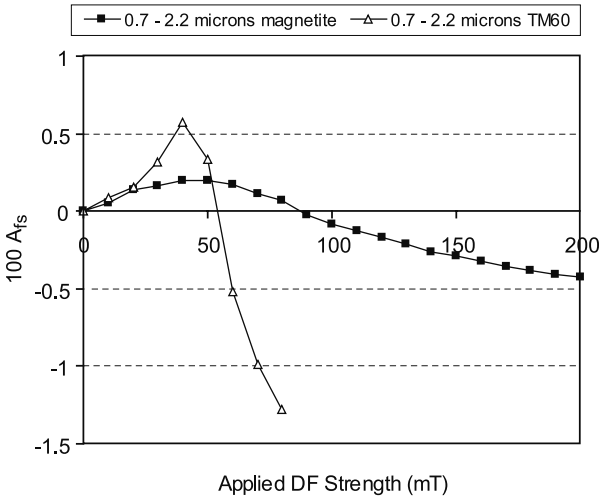

Figure 5. A comparison of the impressed susceptibility anisotropy as a function of applied DF strength for similar particle size fractions (0.7–2.2 μm) of magnetite and TM60.

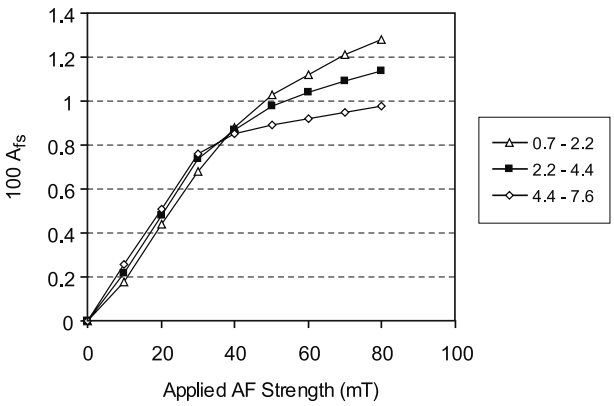

Figure 6. Percentage impressed susceptibility anisotropy as a function of peak applied AF strength for TM60 fractions (the numbers in the legend are the particle sizes in microns).

exhibit positive field-impressed anisotropy at low fields, but all the fractions (even the 63–75 μm sample) exhibit negative field-impressed anisotropy at the higher fields up to 80 mT. This is very different to the magnetite and TM60 results where only the smallest particles showed negative values. While the results up to 80 mT for fractions 0.8–22.7 μm show higher impressed anisotropy for the larger particle sizes in this range, the slope of the impressed anisotropy curves is steeper for the finer fractions at higher fields. Also the values for fractions 22.7–26.4 μm, 26.4–29 μm and 63–75 μm start to become progressively smaller with increasing particle size, and the curves for these larger particles appear closer to saturation than those for the smaller particles. These features suggest that the smaller fractions are not saturated, and that the impressed anisotropy will actually be greater for these fractions (in the same way as for magnetite and TM60) at higher fields. Pyrrhotite also exhibits more negative impressed anisotropy than magnetite or TM60.

[15] The results for AF treatment on the pyrrhotite fractions are shown in Figure 9. Significantly, none of the fractions exhibited a negative impressed anisotropy, but

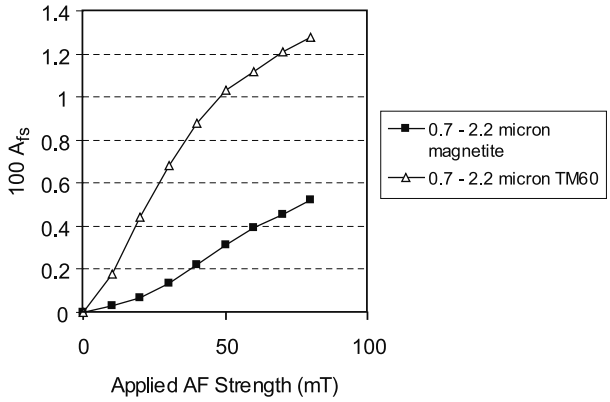

Figure 7. A comparison of the impressed susceptibility anisotropy as a function of peak applied AF strength for similar particle size fractions (0.7–2.2 μm) of magnetite and TM60.

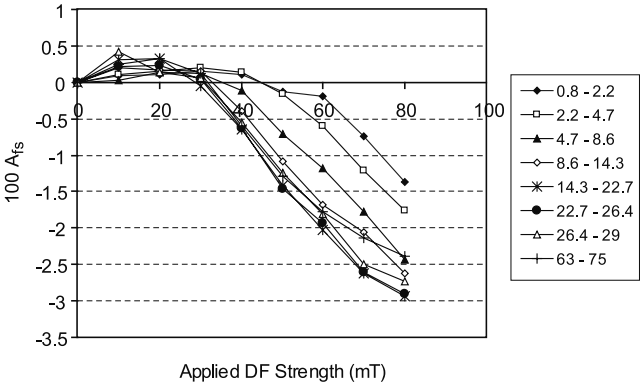

Figure 8. Percentage impressed susceptibility anisotropy as a function of applied DF strength for monoclinic pyrrhotite fractions (the numbers in the legend are the particle sizes in microns).

gave increasingly positive values with increasing AF. This is very similar to the results for MD magnetite and TM60. While the values for the finer pyrrhotite fractions are smaller than the larger fractions, again (as for the DF case) the maximum applied field of 80 mT is below the saturating field. The finer fractions show steeper curves and are further from saturation. At higher fields the maximum impressed anisotropy is likely to be higher for the finer fractions. The values of 100 A_{fs} are larger than those for magnetite, and at saturation are likely to be larger than those for TM60.

3.4. Hematite

[16] Some preliminary experiments on two available size fractions of natural hematite (specularite) were undertaken. Interestingly, the <44 μm fraction gave negative values of field-impressed anisotropy after DF and AF treatments along the z axis at 80 mT with 100 A_{fs} being -8.93 and -1.90, respectively. The value for the DF treatment is the greatest for any mineral sample studied so far. For the 44–76 μm fraction, DF and AF treatments at 80 mT produced values of 100 A_{fs} of -0.87 and 0.23, respectively. This fraction therefore behaved like many of the MD fractions of the other minerals studied, by exhibiting negative field-impressed anisotropy after a DF treatment and a positive value after AF treatment.

4. Discussion

4.1. AF Treatment and the Limits of Intrinsic (Classical) SD Behavior

[17] At the level of an individual MD particle the effect of an applied AF would be to demagnetize it and nucleate domain walls. Thus there would seem no physical mechanism whereby increasing the strength of an AF would result in a metastable SD state in any particle that is intrinsically MD. The susceptibility should not therefore decrease along the AF axis, but is more likely to increase due to alignment of domain walls and domain rearrangement. Therefore the impressed anisotropy would not be expected to go negative for any MD particle subjected to AF treatment. This is borne out by the results for all the magnetic mineralogies studied here and in previous work. The MD magnetite [*Potter and Stephenson*, 1990a, Figure 3], titanomagnetite (present study, Figure 6), pyrrhotite (present study, Figure 9), and 44–76 μm hematite particle size fractions all exhibit positive impressed anisotropy which increases (to saturation in some cases) with increasing field strength. None of these samples show a decrease in field-impressed anisotropy with increasing field strength. The only samples which exhibit a negative field-impressed anisotropy after single axis AF treatment are those which contain intrinsically SD particles such as SD γFe_20_3 [*Potter and Stephenson*, 1988a, 1990a], rocks containing predominantly SD particles [*Potter and Stephenson*, 1990b], and the <44 μm hematite fraction studied here. This suggests that AF treatment provides a means of experimentally determining the intrinsic classical (not metastable) SD/MD boundary in various minerals.

[18] Recent work on the low-field variation of AC susceptibility [*Hrouda et al.*, 2006] may also be relevant to the present study. *Hrouda et al.* [2006] made AC susceptibility measurements in 21 fields ranging from 2 to 450 A/m and found that with increasing field strength (1) for MD magnetite and titanomagnetite there were slight increases in susceptibility (though for magnetite it was difficult to tell whether the effect was real), (2) for MD pyrrhotite there were significant increases in susceptibility (as in the work of *Worm* [1991] and *De Wall and Worm* [1993]), and (3) for SD magnetite there was some suggestion that the susceptibility may have decreased. All these results have a similar pattern with AF treatment to the present results and previous work [*Potter and Stephenson*, 1988a, 1990a], namely, (1) small positive field-impressed anisotropy for MD magnetite and titanomagnetite, (2) larger positive values for MD pyrrhotite are suggested at saturation from the shape of the impressed anisotropy curves, and (3) negative values for SD magnetite particles. This suggests that *Hrouda et al.'s* [2006] small increases in susceptibility for MD magnetite, and small decreases for SD magnetite are real and not due to measurement errors. It also suggests that their results are essentially a sensitive measurement of field-impressed anisotropy at very low fields. Note that in their experiments they were measuring the AC susceptibility in different low-field strengths, whereas in the present study and our previous work the susceptibility was measured (each time using a 700 μT field in the Digico anisotropy delineator) after the sample had been subjected to an AF that was

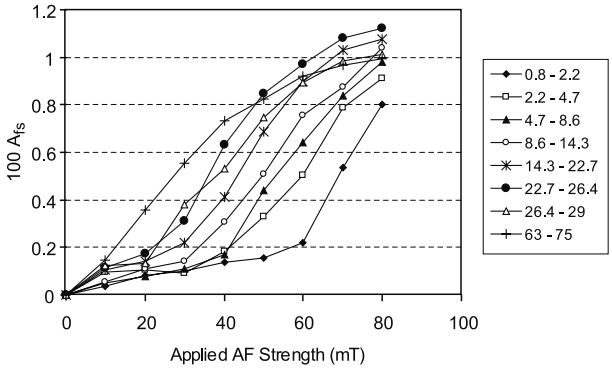

Figure 9. Percentage impressed susceptibility anisotropy as a function of peak applied AF strength for monoclinic pyrrhotite fractions (the numbers in the legend are the particle sizes in microns).

cycled from zero to its peak value and back to zero. Note also that the "low" applied AFs in the present study are higher than *Hrouda et al.'s* [2006] measuring fields. The advantage of looking at the higher field behavior in the present work is that the differences between AF and DF treatment (particularly the progressive changes from positive to negative field-impressed anisotropy upon DF treatment) are only seen at the higher fields.

4.2. DF Treatment and the Limits of Metastable SD Behavior

[19] For DF treatment of MD particles one might expect positive impressed anisotropy at low fields due to some initial domain interaction and alignment of domain walls (as for the AF case). However, with increasing applied DF strength, the flushing out of domain walls and consequent potential transitions to metastable SD states by a proportion of the particles might cause the field-impressed anisotropy to reach a peak and then decrease, since idealized uniaxial SD particles theoretically have zero susceptibility along the easy magnetization direction, and a maximum susceptibility perpendicular to this direction. For fractions containing relatively small MD particles, those that are closer to the intrinsic theoretical (classical) SD/MD boundary, the field-impressed anisotropy due to DF treatment at high fields might actually go negative, since smaller MD particles are more likely to be capable of undergoing a transition to a metastable SD state. For fractions containing relatively large MD particles, DF treatment at high fields might only result in a slight decrease in the field-impressed anisotropy (due to transitions to a metastable SD state in some particles) with increasing applied field, but the value may not necessarily go negative. The results from the present study, as well as those from previous work [*Potter and Stephenson*, 1990a], are consistent with the above scenario. The experimental results for MD magnetite and TM60 show that only the smallest fractions exhibit negative field-impressed anisotropy after DF treatment at high fields. The results for magnetite (Figure 2) show a smooth trend with particle size. Also, the impressed anisotropy goes negative at systematically higher fields as the particle size increases (Figure 3). These systematic, smooth trends with particle size were not so evident in the previous model predictions [*Stephenson and Potter*, 1996, Figures 5 and 8]. Moreover, the model predicted that all the fractions (apart from 7.6 to 13.1 μm) would exhibit negative field-impressed anisotropy at saturating fields, whereas experimentally it appears that all fractions from 25.5 to 45 μm and above do not show negative values (and in fact the 7.6–13.1 μm fraction does show a negative value). The differences between the experimental results and the model predictions are perhaps a consequence of the predictions being heavily dependent upon fitting curves to experimental results obtained up to only 80 mT. The experimental field-impressed anisotropy results, while being consistent with some particles undergoing transitions to metastable SD states, suggest that such states are much less likely to occur in particles of magnetite or TM60 above a few microns. Even above 1 μm such states would appear to be relatively rare, since for the smallest fraction (<0.7 μm) the percentage field-impressed anisotropy (about −1.2, Figure 2) is still less negative at a saturating DF than for acicular SD γFe_2O_3 particles where the value of 100 A_{fs} after a 80 mT DF was close to −2 [*Potter and Stephenson*, 1988a]. A recent model [*Potter and Stephenson*, 2006] also suggests that equidimensional SD magnetite particles should have a more negative field-impressed anisotropy than acicular particles. This suggests that only a proportion of particles in the <0.7 μm magnetite fraction (whose particles are closer to being equidimensional than acicular) may have undergone a transition to a metastable SD state. This is also consistent with the ratio of saturation remanence (M_{rs}) to saturation magnetization (M_s) for this sample, where $M_{rs}/M_s = 0.23$, which is still well below the theoretical value of 0.5 for randomly dispersed SD uniaxial particles. However, the value of 0.23 may still be higher than it otherwise would have been if none of the particles underwent a transition to a metastable SD state.

[20] *Geiss et al.* [1996] did not observe metastable SD states in domain observations of 0.5–10 μm magnetite and titanomaghemite particles in DF cycling to ± 120 mT. However, at this maximum field only the smallest MD magnetite particles are likely to have undergone a transition to a metastable SD state, according to Figures 2 and 3 of the present study. At higher DF strengths the field impressed results suggest that metastable SD behavior remains a possibility (even though it is likely to be relatively rare) within the particle size range of Geiss et al's experiments. The upper limit for metastable SD behavior in magnetite might thus be slightly larger than the SD to two-domain transition size of 0.25 μm of *Geiss et al.* [1996]. The present results showing MD type positive values of 100 A_{fs} for large magnetite particles (Figures 2 and 3) are also consistent with *Heider and Hoffmann's* [1992] view that a 70 μm magnetite particle, that appeared from domain imaging (using the magneto-optical Kerr effect) to be in a SD state upon DF treatment, may still be MD with further domains below the plane of observation.

[21] Since the critical theoretical (classical) SD/MD boundary occurs at larger particle sizes as one goes from magnetite to TM60 to pyrrhotite to hematite [*Dunlop and Özdemir*, 1997, Table 5.1], it is logical to assume that the increased particle size limits of critical metastable SD behavior may also depend upon mineralogy in a similar systematic order. In other words, the particle size range for metastable SD behavior may increase as one goes from magnetite to TM60 to pyrrhotite to hematite. If negative field-impressed anisotropy due to DF treatment of intrinsic MD particles is a reflection of metastable SD behavior, then the size range in which negative field-impressed anisotropy may occur is likely to be larger in the same sequence for the above minerals. The experimental results suggest that this is the case. Also, for an equivalent MD particle size the transition to negative field-impressed anisotropy would be expected to occur at a lower field for TM60 than magnetite if metastable SD behavior extends over a greater size range for TM60. A comparison of the magnetite and TM60 results demonstrates the latter (Figure 5). Domain imaging by *Halgedahl and Fuller* [1980, 1983] suggested that metastable SD states may exist in TM60 particles up to around 10 μm diameter. While the present results suggest that the metastable SD particle size range for TM60 is likely to be larger than that for magnetite, further field-impressed studies are needed on larger TM60 particle size fractions and

higher applied fields to help constrain the metastable SD range.

[22] The pyrrhotite fractions are all intrinsically MD according to the positive field-impressed anisotropy upon AF treatment (Figure 9). The fact that pyrrhotite shows negative field-impressed anisotropy upon DF treatment for all the size fractions from 0.8 to 75 μm (Figure 8) is consistent with the view that its metastable SD size range is larger than that of magnetite and TM60. This is consistent with some domain observations [*Halgedahl and Fuller*, 1983] of relatively large particles of pyrrhotite, including a 50 μm particle, which appeared to be in a metastable SD state after a saturation remanence. Interestingly, similar pyrrhotite fractions to the ones in the present study all gave very similar M_{rs}/M_s ratios (varying by only about 15%) as reported by *Menyeh and O'Reilly* [1991], which would also appear to be consistent with the relatively small variation in field-impressed anisotropy after DF treatment.

[23] For hematite, the negative field-impressed anisotropy for the <44 μm fraction upon AF treatment suggests that there is a reasonable proportion of intrinsic SD particles in that sample. The fact that the field-impressed anisotropy is more negative upon DF treatment may reflect some initial MD particles in the sample undergoing a transition to a metastable SD state. For the 44–76 μm hematite fraction the field-impressed anisotropy upon AF treatment was positive suggesting the particles are intrinsically MD. The negative values of field-impressed anisotropy upon DF treatment may again be associated with transitions to metastable SD states. The metastable SD size range for hematite may be relatively large compared to the other minerals studied. *Schmidt and Fuller* [1970] found that the application of a 900 mT DF in the basal plane of single hematite crystals resulted in a minimum susceptibility in the remanence direction, consistent with the present results. Their crystals were presumably a few millimeters in diameter and so metastable SD states might extend to relatively large particle sizes in hematite. *Zapletal* [1985] has also investigated field-impressed anisotropy in hematite due to a DF, and modeled the effect in terms of the preferred orientation of the magnetic moments, and assuming uniaxial anisotropy in the basal plane. More recently, *Potter and Stephenson* [2006] have quantified the effect for SD hematite in terms of the possible moment orientations within the basal plane.

5. Conclusions

[24] The following conclusions can be made from the present study:

[25] 1. Field-impressed anisotropy of magnetic susceptibility provides independent evidence for metastable SD states and transdomain transitions. The results provide complementary three-dimensional quantitative support for some previous two-dimensional domain images of these features. However, the results place constraints on the particle size limits of metastable SD behavior for different mineralogies.

[26] 2. The results for magnetite, titanomagnetite, pyrrhotite and hematite strongly suggest that field-impressed magnetic anisotropy due to DF treatment can be used to quantify the particle size limits of metastable SD behavior in these minerals, by identifying those size fractions that exhibit negative field-impressed anisotropy. This is because progressive changes from positive to negative field-impressed anisotropy (prolate to oblate impressed ellipsoids of revolution) with increasing applied DF strength are consistent with the DF treatment producing a metastable SD state in the smallest intrinsic MD particles (that is, MD in the demagnetized state).

[27] 3. Intrinsic MD particles of titanomagnetite (TM60) showed more negative field-impressed anisotropy due to DF treatment than equivalent magnetite particle sizes. The transition from positive to negative field-impressed anisotropy also occurred at lower fields for the TM60 particles. This suggests that metastable SD behavior extends over a greater particle size range for TM60 than magnetite. Intrinsically MD particles of pyrrhotite and hematite exhibit negative field-impressed anisotropy, due to DF treatment, over a much larger particle size range than magnetite or TM60. This suggests that metastable SD behavior for pyrrhotite and hematite extends over a much larger particle size range than for magnetite and TM60. This adds support to previous domain images [*Halgedahl and Fuller*, 1983] where some relatively large particles of pyrrhotite (including a 50 μm particle) preserved a metastable SD state after saturation remanence. Further impressed anisotropy results on narrow particle size fractions should help to quantify the particle size limits of metastable SD behavior for each of these minerals. It is possible that the range of metastable SD behavior may to a large extent reflect so-called pseudosingle-domain (PSD) behavior.

[28] 4. The results suggest that field-impressed magnetic anisotropy due to AF treatment can be used to quantify the limits of theoretical classical (not metastable) SD behavior. This is borne out by (1) the experimental results from all the MD material studied here and in previous studies [*Potter and Stephenson*, 1990a, 1990b] where the impressed anisotropy due to a single axis AF is positive and increases (often to saturation) with increasing field strength, and (2) the impressed anisotropy only goes negative for intrinsically SD material, as shown by the hematite results described here and previous results for SD particles [*Potter and Stephenson*, 1988a, 1990a, 1990b]. These results probably reflect the fact that single axis AF treatment should not be capable of producing a metastable SD state in an intrinsic MD particle, since the effect of the AF will be to demagnetize the particle and nucleate a domain wall or walls.

[29] 5. The present results due to AF treatment are consistent with observed changes in the low-field variation of AC susceptibility seen in magnetite, titanomagnetite and pyrrhotite by *Hrouda et al.* [2006]. Significantly, the negative field-impressed anisotropy for SD magnetite particles and positive field-impressed anisotropy for MD magnetite particles subjected to AF treatment in the present work and previous studies [*Potter and Stephenson*, 1988a, 1988b, 1990a, 1990b] are consistent with *Hrouda et al.*'s [2006] results which show a small decrease in AC susceptibility with increasing low field for SD magnetite and a small increase for MD magnetite. This suggests that their results are real and not due to measurement errors. It also suggests that their results are essentially field-impressed effects at low fields, and their equipment provides a sensitive means of detecting this.

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