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Single-domain-like behavior in a 3-mm natural single crystal of magnetite

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Abstract. We have observed single-domain (SD) like behavior in a 3-mm natural single crystal of magnetite following low-temperature demagnetization (LTD), which consists of zero-field cycling through the Verwey transition to erase remanence carried by pinned domain walls. We compared stepwise alternating field (AF) and thermal demagnetization curves of 1-mT total thermoremanent magnetization (TRM), 1-mT partial TRM (pTRM) acquired between the Curie point (T_C = 575°C) and 565°C, and saturation isothermal remanent magnetization (SIRM) measured with and without prior LTD. AF demagnetization curves of untreated TRM and SIRM decreased exponentially with increasing AF. SIRM was more resistant to demagnetization than was TRM, a multidomain (MD) result of the Lowrie-Fuller test. After LTD the TRM and SIRM memories had AF demagnetization curves with sigmoid shapes and initial plateaus below 8 mT in which little or no demagnetization occurred. Both features are reminiscent of SD behavior. During thermal demagnetization, untreated TRM and SIRM decreased almost linearly with increasing temperature up to ≈500°C. Such distributed unblocking temperatures T_{UB} are expected for pinned walls in MD grains. The remaining 60% of TRM and 40% of SIRM were lost over a narrow temperature interval concentrated between 560°C and T_C. The TRM memory after LTD was very stable against thermal demagnetization. There was no decrease in remanence below 550°C and very little change until 565°C, only 10°C below T_C. This very high T_{UB} fraction of TRM seems to have SD-like character. SIRM has similar behavior. In the case of pTRM, both the untreated remanence and the memory after LTD have almost entirely high T_{UB}s, identical to the range of pTRM blocking temperatures T_B, 565°C–T_C. $T_{UB} = T_B$ is a basic property of SD partial TRM. The pTRM memory fraction is also larger than that of total TRM or SIRM. These observations suggest that pTRM(T_C, 565°C) isolates an SD-like fraction of remanence similar to that constituting TRM and SIRM memory. The high AF coercivities of the TRM and SIRM memories indicate that the source of SD-like behavior may be unusually strong domain wall pinning by crystal defects formed at monoclinic twin boundaries as a result of crystal distortion from cubic to monoclinic structure in passing through the Verwey transition.

1. Introduction

It has long been hypothesized that part of the remanence of multidomain (MD) magnetites mimics that of single-domain (SD) grains in its intensity and resistance to alternating field (AF) and thermal demagnetization. SD-like regions of unusually high magnetic moment and coercivity have been attributed to deflection of spins around dislocation lines [*Verhoogen*, 1959], Barkhausen discreteness of domain wall positions [*Stacey*, 1962], regions surrounding in-phase and out-of-phase stress centers [*Kobayashi and Fuller*, 1968], domain walls themselves [*Dunlop*, 1977], surface domain structures [*Banerjee*, 1977], and metastable SD grains in which walls have failed to nucleate [*Halgedahl and Fuller*, 1983].

Experimentally, the most popular technique for isolating the SD-like part of remanence has been low-temperature demagnetization (LTD), in which a sample is cooled through the isotropic point, $T_I \approx 130$ K [*Abe et al.*, 1976], and the Verwey phase transition ($T_V \approx 115$ K [*Verwey*, 1939]), then rewarmed to room temperature, all in zero field. LTD removes a large part of MD remanence by causing domain walls to become unpinned. SD-like remanence is highlighted in the memory, the fraction of remanence remaining after LTD.

At T_I the first crystalline anisotropy constant of magnetite K_1 changes sign and momentarily becomes zero. The easy directions of magnetization switch from ⟨111⟩ (cubic) to ⟨001⟩ (monoclinic c axis). Below the Verwey transition the c axis becomes slightly elongated and inclined with respect to the other axes, producing a change in crystal symmetry from cubic to monoclinic [*Matsui et al.*, 1977], thus resulting in a twin structure.

There are two effects on domain walls. First, the vanishing of K_1 results in a considerable broadening of the walls because wall width is proportional to $K_1^{-1/2}$ [*Dunlop and Özdemir*, 1997, chapter 5]. Broad walls are less effectively pinned by localized defects [*Xu and Merrill*, 1989; *Moskowitz*, 1993] and eventually escape. Domain wall thickness also varies with temperature, with the result that unpinning is not confined to the vicinity of T_I. *Halgedahl and Jarrard* [1995] have observed large, irregular Barkhausen jumps in low-temperature cooling and reheating cycles of a magnetite crystal, particu-

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larly in what they call the "wild zone" between T_I and T_V but also over a broad temperature range above T_I.

The second effect was pointed out by *Kobayashi and Fuller* [1968]. The change in sign of K_1 causes pinning sites for domain walls to become centers of repulsion and vice versa (Kobayashi and Fuller's in-phase and out-of- phase stress centers). This effect should cause massive remobilization of walls upon cooling through T_I and raises the question of how any memory of initial remanence can be recovered during the heating half of the LTD cycle (except SD remanence pinned by shape anisotropy, which is unaffected by changes in K_1).

LTD experiments on grown synthetic magnetites [*Levi and Merrill*, 1976, 1978; *Dunlop and Argyle*, 1991; *Heider et al.*, 1992], on crushed natural magnetites [*Hartstra*, 1983; *McClelland et al.*, 1996; *Shcherbakova et al.*, 1996], and on natural single crystals of magnetite [*Ozima et al.*, 1964; *Kobayashi and Fuller*, 1968; *Levi and Merrill*, 1976, 1978] have shown that memory is a general phenomenon. MD and SD-like sources of remanence appear to coexist regardless of the origin or grain size of the magnetite. Although memory ratios (memory as a fraction of initial remanence) for thermoremanent magnetization (TRM) and saturation isothermal remanent magnetization (SIRM) are grain size dependent [*Hartstra*, 1983; *Heider et al.*, 1992; *Halgedahl and Jarrard*, 1995; *McClelland and Shcherbakov*, 1995; *Dunlop and Özdemir*, 1997, chapter 12], the stability of memory against AF and thermal demagnetization seems to be rather insensitive to grain size [*Dunlop and Argyle*, 1991; *Dunlop and Özdemir*, 1997, chapter 12].

Many factors, among them grain elongation, nonmagnetic impurities, and low-temperature oxidation [*Özdemir et al.*, 1993], can affect low-temperature memory, but crystal defects and resulting internal stress seem to play the major role [*Heider et al.*, 1992]. All natural and synthetic crystals have lattice defects of one kind or another: inclusions of a nonmagnetic phase, dislocations, voids, internal grain boundaries, and so on. Wall displacement is impeded by these defects or the stress fields they create, trapping and anchoring the wall away from its original position. The effect of defects on the magnetic properties is most apparent for large MD grains of natural magnetite. Control of coercivity and magnetic stability by internal stress is evidenced by the temperature dependence of hysteresis properties of MD magnetites at both low and high temperatures (77-300 K [*Hodych*, 1982, 1990] and 20°-580°C [*Özdemir and Dunlop*, 1997]).

The present paper describes AF and thermal demagnetization experiments on weak-field TRM, partial TRM, and SIRM and on the memories of these remanences after LTD, for millimeter- size single crystals of magnetite. The purpose of these experiments was to see whether the low-temperature memory is SD-like even in an extremely large MD crystal containing many domain walls. It is implausible that such a large crystal could fail to nucleate any walls following saturation or after heating above T_C, and so we can rule out metastable SD behavior as a source of SD-like memory. Most other suggested mechanisms for stable SD-like remanence and memory are equally unlikely. Thus we hoped to be able to clarify what actually controls memory near the upper size limit of naturally occurring MD grains.

2. Properties of the 3-mm Magnetite Crystal

Most experiments were carried out on a natural octahedral crystal of magnetite about 3 mm on edge from a chloritized ultramafic rock (Harford County, Maryland, in the Appalachian Piedmont). $^{40}Ar/^{39}Ar$ laser probe dating of a similar magnetite crystal from the same locality yielded an age of 323 ± 5 Ma [*Özdemir and York*, 1992]. This is the age of argon closure of a small biotite inclusion in the magnetite crystal and is therefore a precise estimate of the minimum age of formation of the magnetite.

X ray diffraction of a small chip of the 3-mm crystal using a Debye-Scherrer camera with Fe Kα radiation and a silicon standard gave numerous reflections characteristic of magnetite. The X ray cell edge was determined to be a = 8.39 ± 0.01 Å, in close agreement with the standard value of 8.396 Å (ASTM data file 19-629).

A companion magnetite crystal from the same locality was used for microstructural examination. This crystal was mounted in epoxy and mechanically polished for scanning electron microscopy (SEM) with energy dispersive analysis (EDA) using a Link AN-10000 X ray microanalyzer. Biotite and chlorite were the main inclusions identified.

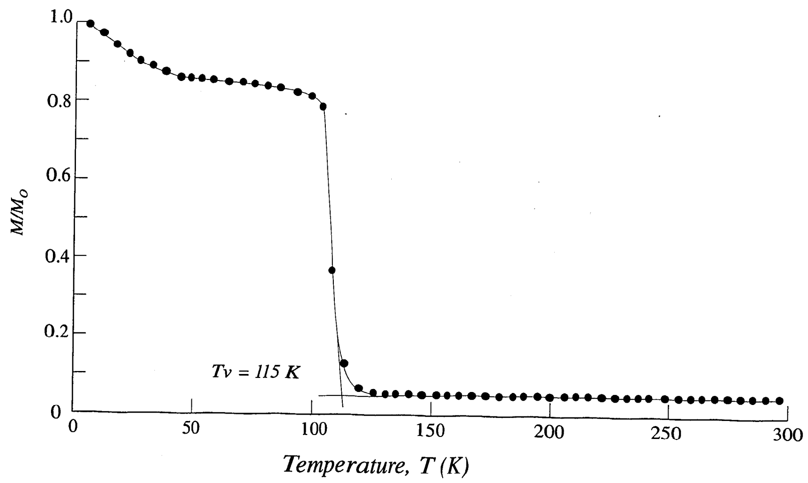

Figure 1. Normalized SIRM of a small chip of the 3-mm crystal measured during warming from 10 K to 300 K. The sharp decrease in remanence around 115 K marking the Verwey transition is diagnostic of stoichiometric magnetite.

A small chip of the crystal used for magnetic studies was measured during zero-field warming from low temperature by using a Magnetic Properties Measurement System (MPMS) with a SQUID detector. The chip was given an SIRM at 5 K, and remanence was measured at 5° intervals to 300 K. A sharp decrease in remanence was observed at the Verwey transition, T_V = 115 K (Figure 1), indicating stoichiometric magnetite [*Özdemir et al.*, 1993].

3. Room Temperature and High-Temperature Hysteresis

The magnetite crystal was sealed in an evacuated quartz capsule for the main remanence experiments described later. After these experiments the quartz capsule was opened, and the crystal was mounted on a ceramic holder. Hysteresis curves were measured at 20°C intervals from room temperature to 600°C by using a Princeton Measurements Micro VSM (vibrating sample magnetometer). A continuous jet of helium through the furnace prevented oxidation during heating. A long time constant of 500 s for each heating interval allowed the crystal to reach thermal equilibrium throughout its volume at each temperature.

The hysteresis curves have the ramp-like shape typical of large MD grains (Figure 2). Room temperature hysteresis parameters are consistent with coarse MD grains of magnetite: saturation magnetization M_s = 92.0(4) A m^2/kg, coercive force H_c = 0.46(6) mT, remanence ratio M_{rs}/M_s = 0.003, and coercivity ratio H_{cr}/H_c = 42. The susceptibility $\chi = dM/dH$ is essentially constant with respect to H over the range $0 \leq M \leq 0.58\,M_s$, above which domain rotations begin to contribute to the approach to saturation. The demagnetizing factor N, which characterizes the internal field of an MD grain, was determined as $N_{cgs} = 1/\chi = 3.59$. This is slightly lower than the value of $4\pi/3 = 4.19$ expected for a perfectly equidimensional crystal. The value estimated from $N \approx H_c/M_{rs}$ [*Néel*, 1955] is 3.53, in good agreement with the value from the slope of the hysteresis curve.

M_s for our crystal varies with temperature T as $(T_C - T)^{0.4}$ (Figure 3), as *Pauthenet* [1952] found, and disappears at the Curie temperature T_C = 585°C. An alternative determination, from the temperature dependence of initial susceptibility χ_0 measured by a Kappa bridge, is T_C = 575°C (Figure 4).

The initial susceptibility χ_0 is almost independent of temperature, increasing only 7% between 20°C and T_C. Figure 4 is a classic example of multidomain behavior. The usual Hopkinson peak in susceptibility just below T_C is absent because in the MD self-demagnetization limit, $\chi_0 = 1/N$, which is temperature independent [*Dunlop*, 1984]. For the same reason the χ values in Figure 5, determined from the slopes of hysteresis curves at various temperatures, are also virtually temperature independent. We conclude that our natural single crystal has truly MD behavior and contains domain walls whose displacements are fully responsive to self-demagnetization (apart from the small amount of pinning needed to produce a remanence).

As we mentioned earlier, for our crystal, $H_c/M_{rs} \approx N$ as determined from χ. The same proportionality between measured coercive force and saturation remanence values is maintained at high temperatures (Figure 6; normalized values of H_c and M_{rs} would be identical). This proportionality is in accord with the theoretical relation $M_{rs}(T) \approx H_c(T)/N$ for domain walls with similar pinning strengths whose displacements are limited by the internal demagnetizing field [*Stacey*, 1963; *Xu and Merrill*, 1987].

4. Methodology of Remanence Experiments

Most of the experiments on our crystal involve thermoremanent magnetization (TRM) or partial TRM (pTRM), produced by field cooling from high temperatures. To prevent chemical changes, the crystal was sealed in an evacuated quartz capsule for these experiments. Before each new remanence experiment the sample was reheated to 600°C and cooled in zero field to erase any prior remanence and to establish a standard initial state. The TRMs and partial TRMs were produced in a water-cooled noninductive furnace by cooling in a field of 1 mT from 600°C and were later thermally demagnetized in zero field in the same furnace. Extraneous fields were eliminated by placing the furnace and small Helmholtz coils inside a six-layer mu-metal shield. Fields could be nulled instantaneously during thermal demagnetization to within a few nT and the long-term drift was <10 nT. Remanence intensities and directions were measured at room temperature with a 2G superconducting magnetometer. For AF demagnetization of the remanences we used a Schonstedt demagnetizer.

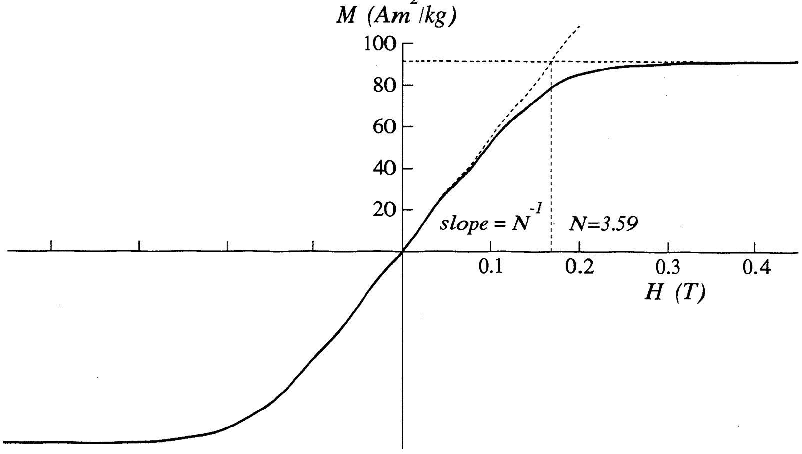

Figure 2. Saturation hysteresis measured at room temperature with a Micro VSM. Both the ramp-like form of the hysteresis loop and the value $N = 1/\chi = 3.59$ determined from the initial slope χ for demagnetizing factor N are indicators of true MD behavior.

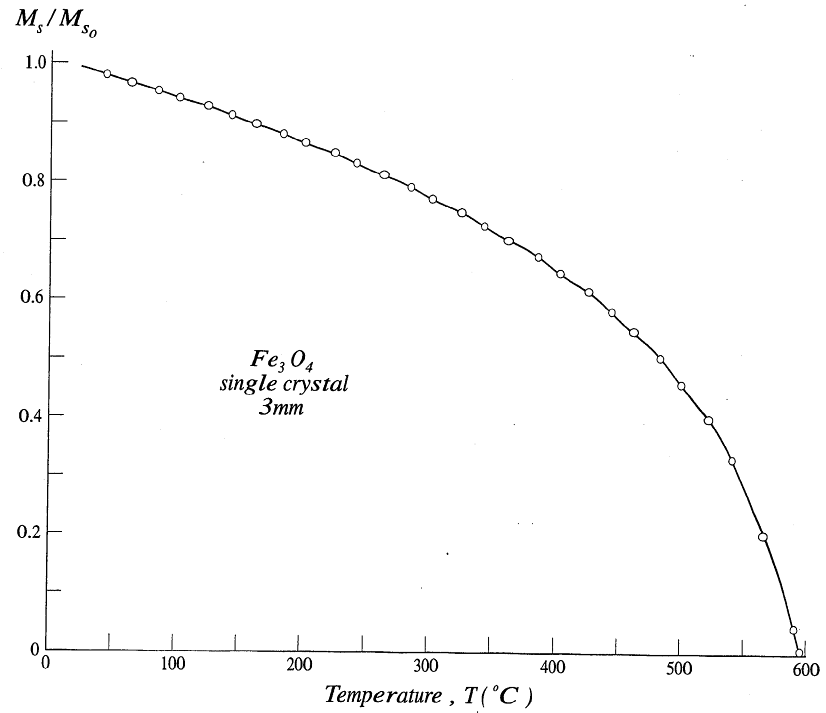

Figure 3. Strong-field thermomagnetic curve for the 3-mm magnetite crystal. Saturation magnetization $M_s \rightarrow 0$ at the Curie temperature $T_C = 585\,°C$.

AF and thermal demagnetization were also carried out for TRM and pTRM memory, the fraction of remanence remaining after low-temperature demagnetization (LTD). We carried out LTD by cooling the crystal to 77 K in a liquid N_2 dewar, allowing the temperature to equilibrate for 30 min, and warming back to room temperature, all in the zero-field environment of a six-layer mu-metal shield. As we explained in the introduction, LTD should destroy remanence due to loosely pinned walls—the walls that are most responsive to the internal demagnetizing field and impart MD response in hysteresis—leaving strongly pinned walls and possibly other sources of remanence as the carriers of low-temperature memory.

The sequence of TRM experiments was (1) thermally demagnetize, produce TRM, AF demagnetize in steps; (2) thermally demagnetize, produce TRM, LTD, AF demagnetize in steps; (3) thermally demagnetize, produce TRM, thermally clean in steps; and (4) thermally demagnetize, produce TRM, LTD, thermally clean in steps. Next, pTRM was produced by cooling the crystal in a 1-mT field from 600° to 565°C and in zero field from 565° to 20°C. This pTRM was thermally

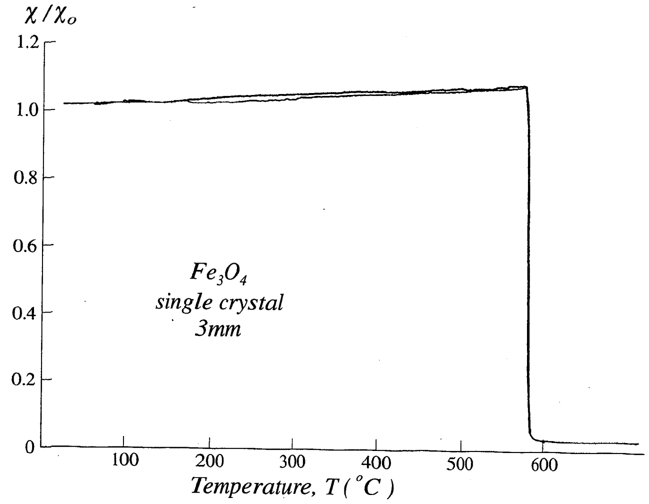

Figure 4. Temperature variation of initial susceptibility χ_0 measured continuously with a Kappa bridge during heating and cooling in an Ar atmosphere. Weak-field susceptibility is almost temperature independent with only a slight indication of a Hopkinson peak, indicating true MD behavior. The Curie temperature indicated by the sharp drop in χ_0 values is 575°C.

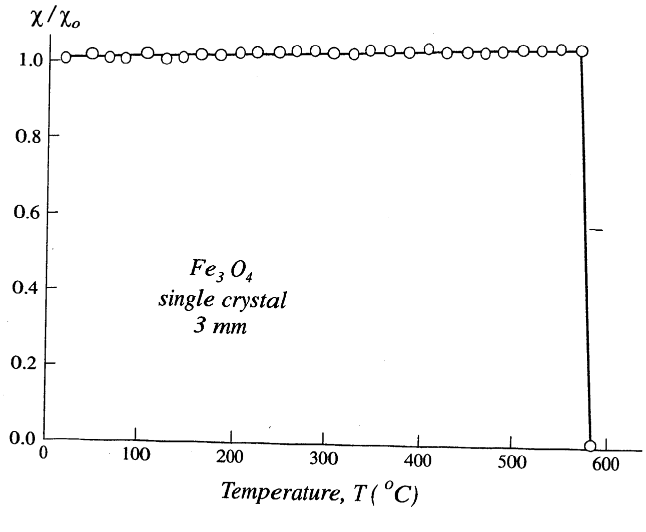

Figure 5. Temperature dependence of strong-field susceptibility determined from the slopes of hysteresis curves measured at 20°C intervals to 580°C. Strong-field susceptibility is independent of temperature, as expected from the theoretical relation $\chi = 1/N$.

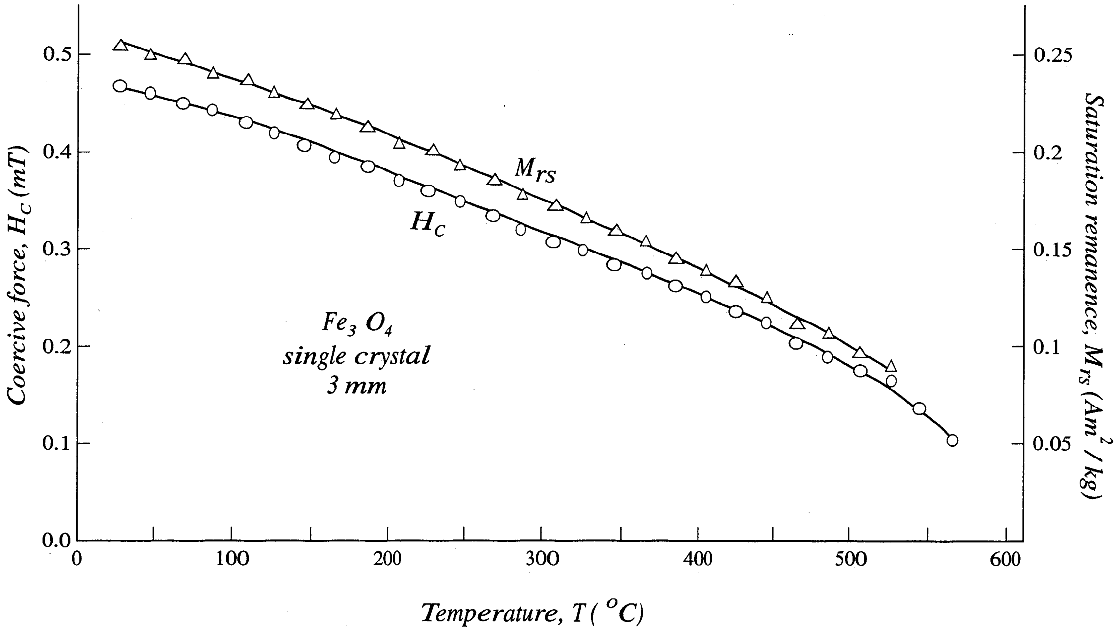

Figure 6. Thermal variations of saturation remanence M_{rs} and coercive force H_c determined from hysteresis curves measured at 20°C intervals during heating. The proportionality between $M_{rs}(T)$ and $H_c(T)$ follows the theoretical relation $M_{rs}(T) \approx H_c(T)/N$ for MD remanence carried by domain walls with similar pinning strengths.

demagnetized in steps. The pTRM was reproduced, then low-temperature demagnetized, and the pTRM memory was stepwise thermally cleaned. No AF demagnetization was carried out for pTRMs.

Finally, after all weak-field remanence experiments were complete, four identical SIRMs were produced by the 1 T field of a pulse magnetizer, with thermal demagnetization before each SIRM. The first two SIRMs were AF and thermally demagnetized in steps. The third and fourth SIRMs were treated by LTD, and the SIRM memory was then AF and thermally demagnetized.

All TRMs, pTRMs, and SIRMs were reproducible to within ±3%. There were no systematic changes as a result of repeated heatings to 600°C.

5. Results

5.1. AF Demagnetization of TRM

The AF demagnetization curve of 1-mT TRM decreases in a quasi-exponential manner with increasing field (Figure 7). This exponential form of AF decay curve is characteristic of MD behavior [*Bailey and Dunlop*, 1983]. The TRM is quite soft, with a median destructive field (MDF) of only 3.8 mT. About 80% of the initial TRM is demagnetized by an AF of 12 mT, the remaining 20% tailing off to higher coercivities. This higher-coercivity tail is also expected for MD grains and results from the combined effects of domain wall pinning and self-demagnetization [*Xu and Dunlop*, 1993].

The TRM memory after low-temperature cycling has a completely different curve (Figure 7). The exponentially decreasing low-coercivity fraction, about 75% of the original TRM, has been destroyed by LTD, leaving a remanence with single-domain (SD) like AF behavior. There is now an initial plateau region in the AF curve, extending to 5 mT or so, in which no AF demagnetization occurs, and the MDF of the memory is ≈5 times larger than that of the untreated TRM. These features are characteristic of SD or pseudo-SD (PSD) behavior.

The AF decay curves of total TRM and its memory cross each other around 25-30 mT (Figure 7). That is, the high-coercivity fraction of TRM increased as a result of low-temperature cycling. This was the only example we observed of such an increase. Possible mechanisms are considered in section 6.1.

5.2. Thermal Demagnetization of TRM

The TRM began to demagnetize as soon as it was heated above 20°C and continued to decrease at a steady rate up to ≈500°C (Figure 8). The remaining 60% of TRM demagnetized abruptly over a narrow interval, most of it between 560°C and 575°C. Previous studies have emphasized the predominantly high unblocking temperatures of TRM in magnetite grains larger than SD size [*Dunlop and Argyle*, 1991; *McClelland and Shcherbakov*, 1995]. Our observations reveal for the first time a sharp separation (also evident in Figures 9, 11, and 12) into two distinct regimes, a low-temperature region of distributed unblocking temperatures extending to ≈500°C and a high-temperature region in which the remaining TRM demagnetizes quite suddenly, mainly in the 20°C interval just below T_C. Each accounts for about one half the original TRM intensity.

The regime of distributed low unblocking temperatures must be a feature of truly MD grains, because it is much more accentuated in our results than in previous work on smaller grains. A spectrum of distributed unblocking temperatures, T_{UB}, is expected on theoretical grounds for domain walls continuously adjusting their positions and numbers as the internal demagnetizing field changes with temperature [*Dunlop and Xu*, 1994].

The very high T_{UB} fraction seems to have a more SD-like character. For one thing, this fraction is unaffected by heating to ≈560°C, by which temperature H_c has decreased about a

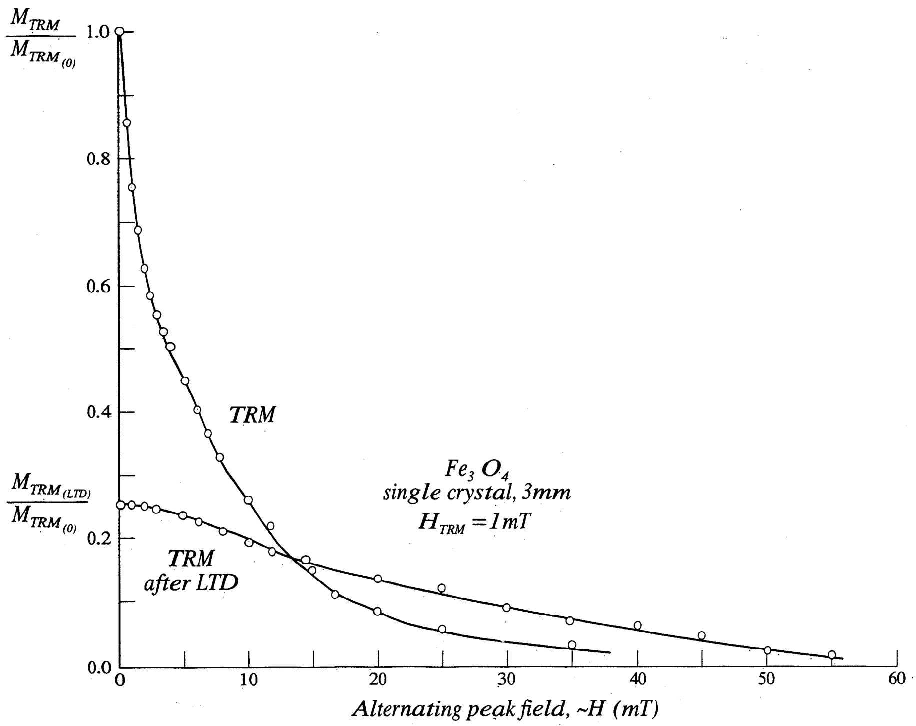

Figure 7. AF demagnetization of 1-mT total TRM with and without prior low-temperature demagnetization (LTD). Both curves are normalized to the initial value of TRM before LTD. The untreated TRM decreases in exponential fashion with increasing AF, characteristic MD behavior. The after-LTD TRM, or TRM memory, has an inflected SD-like AF decay curve with an initial plateau of no demagnetization. LTD has erased the original MD remanence and isolated the stable SD-like memory fraction of TRM.

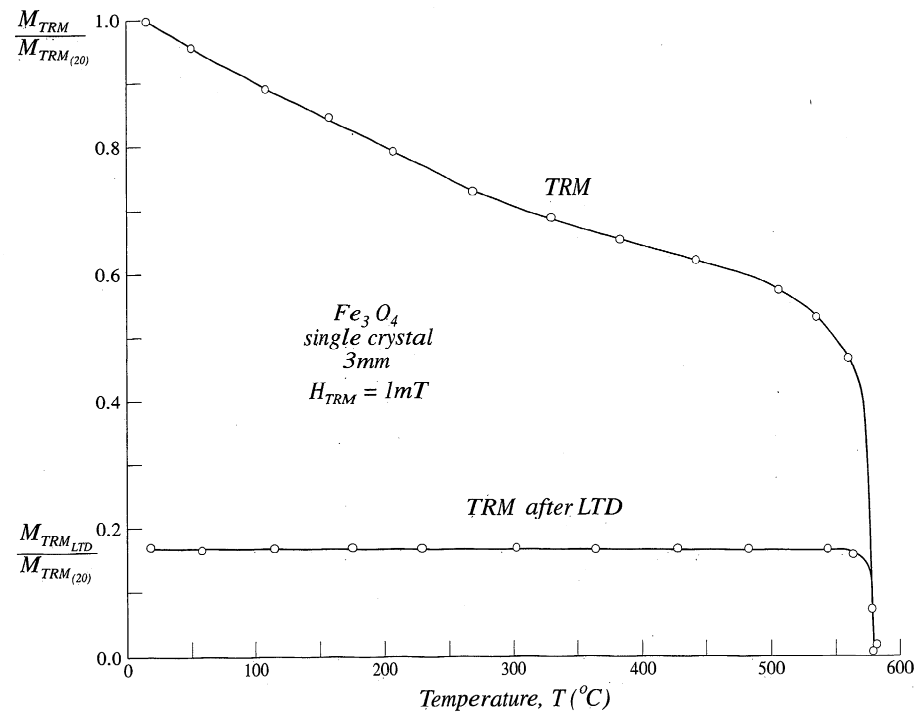

Figure 8. Stepwise thermal demagnetization of 1-mT total TRM and of the memory fraction of TRM after LTD. The original TRM has two distinct demagnetization regions: a broad range from 20°C to ≈500°C in which remanence decreases almost linearly with increasing temperature and a narrow range above 500°C of rapidly decreasing remanence. The first region, with distributed unblocking temperatures, is practically absent from the memory, which unblocks almost entirely above 565°C.

factor of 5 (Figure 6), allowing most domain walls to jump at least once. For another, the high-T_{UB} fraction is a substantial part of the TRM. Its intensity is not "screened," i.e., cancelled by the loosely pinned walls that make up the distributed T_{UB} fraction and that are evident also in high-temperature hysteresis (Figure 5). Further evidence for SD-like character will be presented in section 5.4.

After LTD the TRM became very stable against stepwise thermal demagnetization (Figure 8). There was absolutely no decrease in magnetization below 550°C and very little change until 565°C, only 10°C below T_C. The LT memory of TRM is composed entirely of the very high T_{UB} fraction of the original TRM.

Dunlop and Argyle [1991] observed a similar SD-like memory fraction of weak-field TRM, which unblocked mainly within 50°C of T_C and hardly at all below 500°C, in fine-grained (215-540 nm) magnetites. The surprising result of our study is that a 3-mm crystal of magnetite, with all the hallmarks of truly MD hysteresis behavior, also has TRM memory with practically no low unblocking temperatures. Indeed, our large crystal has an even more striking concentration of high T_{UB}s of TRM memory: exclusively above 565°C.

To check these results, we carried out similar experiments on an even larger natural single crystal of magnetite, a museum-quality 4.5-mm crystal from an unidentified locality in Ontario. The results are similar to those of the 3-mm crystal. The TRM decreased in almost perfectly linear fashion up to 350°C, then leveled out to some extent until 450°-500°C, above which the remainder of the remanence was lost quite abruptly, again mostly above 560°C (Figure 9). The only real differences from the results for the 3-mm crystal are the larger fraction of low unblocking temperatures and consequent more pronounced inflection point around 400°C separating the low- and high-temperature demagnetization regimes.

In the 4.5-mm crystal, as in the 3-mm crystal, the low-temperature memory of TRM is remarkably stable against stepwise thermal demagnetization. Absolutely no decrease (in fact, a very slight increase) occurred up to 570°C, above which the memory dropped abruptly to zero. In this crystal the TRM memory was only about 10% of the virgin TRM but was again composed entirely of the very high T_{UB} fraction of the original TRM.

The results on these two natural magnetite crystals of quite different origins are very similar, apart from the intensity of the memory fraction. The distinct regions of distributed low T_{UB}s and discrete very high T_{UB}s in the original TRM and the exclusively high T_{UB}s of the TRM memory after low-temperature cycling are features of both crystals. Although more experiments are called for, we tentatively conclude that these are general properties of MD magnetite.

5.3. AF and Thermal Demagnetization of SIRM

AF demagnetization curves of SIRM, like those of weak-field TRM, are dramatically different depending on whether or not the remanences have been treated beforehand by LTD (Figure 10). The untreated SIRM decreased quasi-exponentially with increasing AF, although not as rapidly as TRM.

About 82% of the SIRM was destroyed by LTD. This is the softest fraction, with coercivities mainly of <30 mT, and presumably resided in pinned domain walls. The SIRM memory has much higher average coercivities. Its AF demagnetization curve, like that of TRM memory, has the inflected sigmoid shape characteristic of SD remanence, with an initial plateau below 8 mT in which scarcely any demagnetization

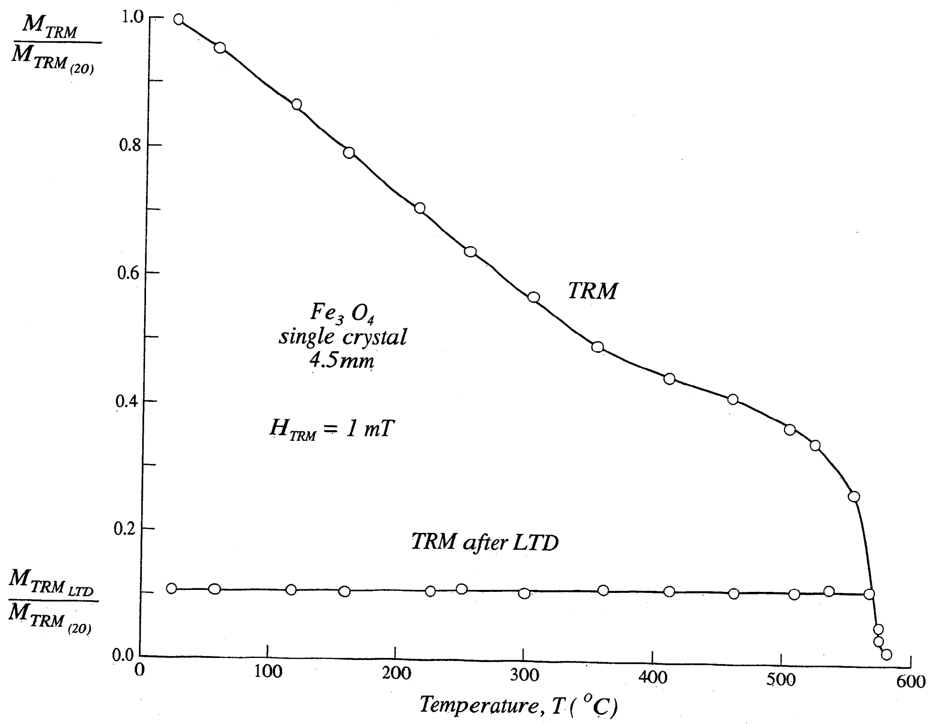

Figure 9. Thermal demagnetization curves of TRM and TRM memory as in Figure 8 but measured for a 4.5-mm octahedral crystal of magnetite. The memory is a smaller fraction of original TRM for this crystal, and there is a more marked separation between the two demagnetization regions. However, the main result is the same: LTD erases all remanence with distributed T_{UB}s, isolating an SD-like memory fraction with very high T_{UB}s.

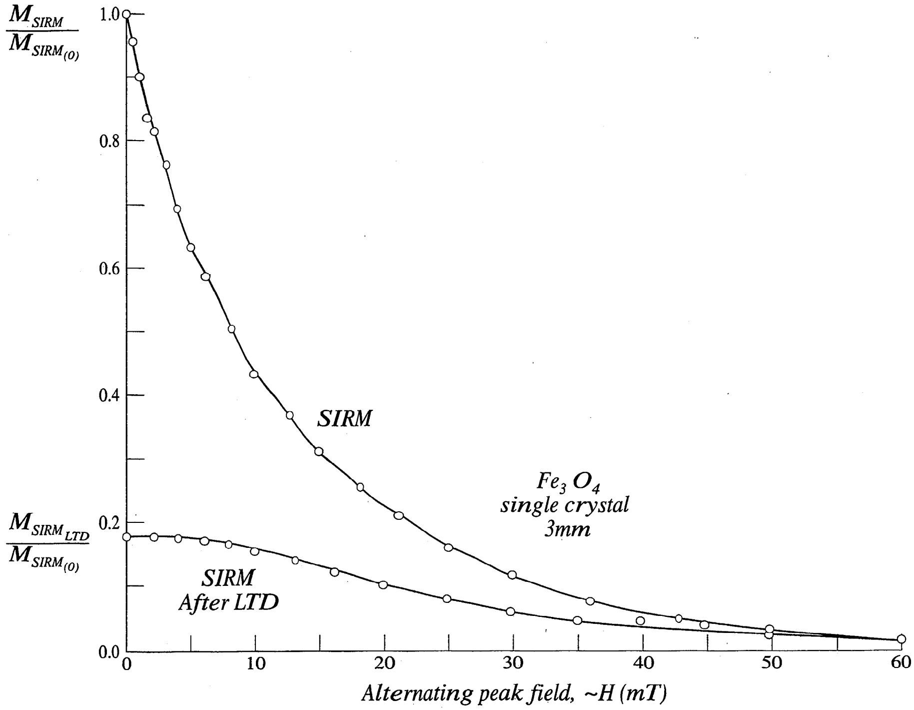

Figure 10. AF demagnetization of SIRM before and after LTD for the 3-mm crystal. As with TRM (Figure 7) the soft, exponentially decreasing MD remanence carried by loosely pinned domain walls is erased by LTD, isolating SIRM memory with an inflected SD-like AF decay curve. The initial plateau of no demagnetization extends to ≈8 mT in the case of SIRM.

occurs. One difference between the TRM and SIRM results is that the SIRM memory and total SIRM demagnetization curves merge smoothly above 40-50 mT.

Thermal demagnetization curves of SIRM with and without prior LTD have the same general features as the corresponding TRM curves. Without LTD treatment the SIRM has a broad region of almost linearly distributed unblocking temperatures extending to ≈520°C and a second narrow window of unblocking temperatures mainly above 550°C (Figure 11). The main difference is that the distributed T_{UB} remanence is about 60% of the SIRM, whereas it was <50% of the TRM. The SIRM memory after LTD, unlike TRM memory, has some low unblocking temperatures, but this fraction is <20% of the memory. Most of the SIRM memory has discrete high T_{UB}s above 550°C.

We once more checked the generality of these results by replicate experiments on the 4.5-mm crystal (Figure 12). The SIRM and TRM thermal demagnetization behaviors are practically identical, except that the low unblocking temperatures of the untreated SIRM are distributed in a perfectly linear fashion from room temperature to ≈500°C. There is no inflection around 350°-400°C. The SIRM memory, unlike that of the 3-mm crystal, has no unblocking temperatures below 500°C. Apart from these minor details the behavior of the two crystals is identical and reproduces all the features observed for TRM.

5.4. Thermal Demagnetization of Partial TRM

The thermal demagnetization results for untreated and LTD-treated TRMs (Figures 8 and 9) showed that the TRM memory has unblocking temperatures almost entirely above 565°C, implying that the memory corresponds to the part of TRM that has discrete, very high unblocking temperatures. If the TRM memory is truly SD-like, it should also be the case that its unblocking temperatures are equal to its blocking temperatures. $T_{UB} = T_B$ is a basic result for TRM blocked by SD grains [*Néel*, 1949], but in the case of MD grains a single T_B corresponds to a broad spectrum of T_{UB}s extending from room temperature (or higher in some cases [cf. *Dunlop and Xu*, 1994, Figure 13] essentially to T_C [*Bol'shakov and Shcherbakova*, 1979; *Shcherbakov*, 1981; *Shcherbakov et al.*, 1993, Appendix; *Xu and Dunlop*, 1994] because domain walls make many successive Barkhausen jumps in the course of reaching a demagnetized state.

We tested both these ideas by thermally demagnetizing a partial TRM produced between T_C and 565°C as well as the memory of the same pTRM after LTD. This partial TRM isolates the fraction of total TRM with blocking temperatures T_B higher than 565°C.

The pTRM demagnetization curve (Figure 13) is very different from the demagnetization curve of the total TRM for the same crystal (Figure 8). The part of the TRM with distributed low T_{UB}s is almost absent. Eighty percent of the partial TRM demagnetizes between 565°C and T_C, in the pTRM blocking temperature range. To a good first approximation, $T_{UB} = T_B$ for this pTRM. Notice also that the untreated pTRM demagnetizes in a fashion virtually identical to that of the LT memory of total TRM. By implication the TRM memory is carried by the same walls or regions in the crystal as the high-temperature pTRM, and these walls or regions behave in an SD-like manner.

After LTD, about 70% of the pTRM was destroyed. Thus memory constitutes only part, not all, of the SD-like high-T_B pTRM. About 10% of the pTRM memory has low unblocking temperatures with a linear distribution from 20° to ≈520°C, but the remanence erased is reversed to the remaining memory

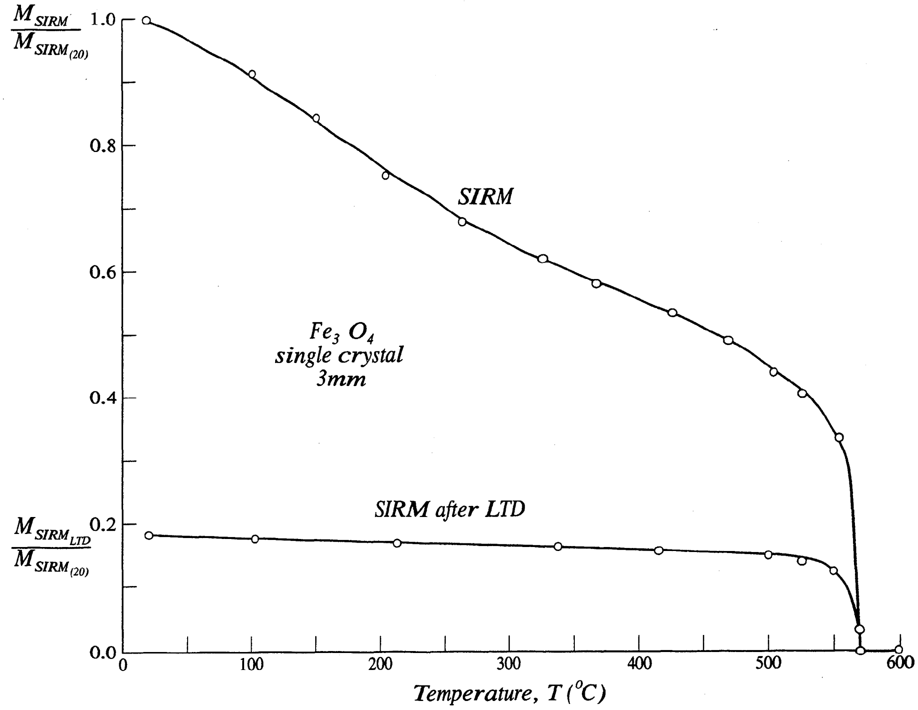

Figure 11. Stepwise thermal demagnetization of SIRM before and after LTD for the 3-mm crystal. Results are similar to those of TRM for the same crystal (Figure 8). SIRM memory after LTD lacks almost completely the prominent fraction of distributed T_{UB}s in the original SIRM and unblocks mainly above 550°C.

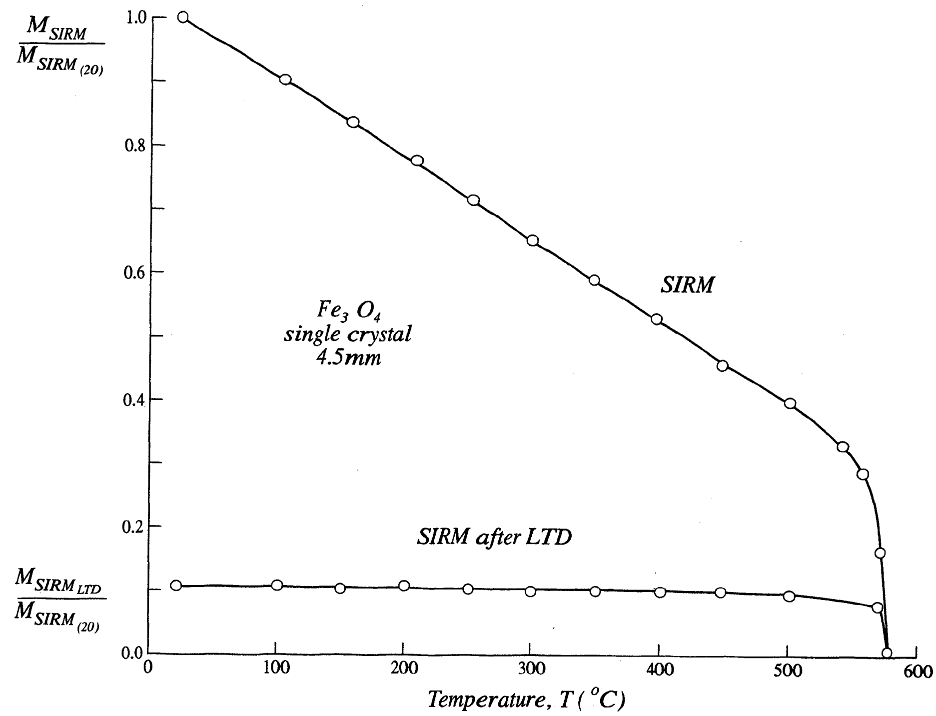

Figure 12. Thermal demagnetization of SIRM before and after LTD for the 4.5-mm crystal. Results are similar to those of Figure 11, except for the lower memory fraction and the more linear distribution of low T_{UB}s in the original SIRM. The main conclusion is the same: LTD eliminates MD remanence carried by loosely pinned walls and isolates an SD-like memory with very high T_{UB}s.

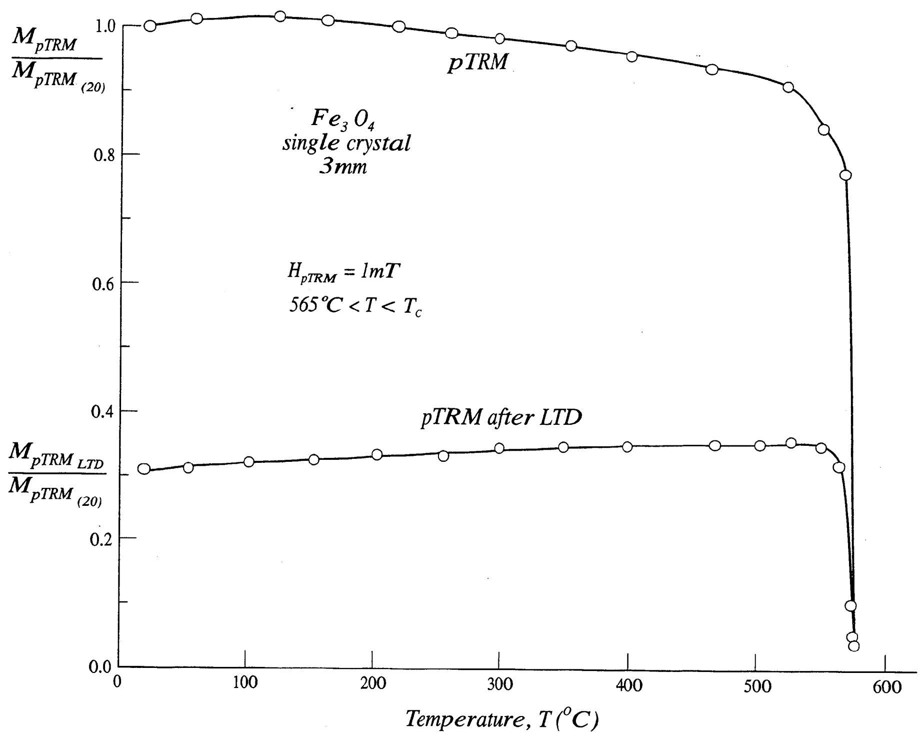

Figure 13. Thermal demagnetization of partial TRM produced by a 1-mT field applied between T_C and 565°C, both before and after LTD, for the 3-mm magnetite crystal. The low-T_{UB} part of total TRM is almost absent from pTRM(T_C, 565°C), implying a similar origin for this pTRM and for the total TRM memory after LTD. After the pTRM is low-temperature demagnetized, its intensity increases slightly with zero-field heating up to ≈550°C. Thus a small fraction of the original pTRM must have been self-reversed.

with discrete high T_{UB}s. Self-reversed fractions of pTRMs in MD magnetite have been reported by *Shcherbakov et al.* [1993] and *McClelland and Shcherbakov* [1995], but in our crystal the reversed fraction is a minor part of the pTRM.

6. Discussion

6.1. Initial Remanence Before LTD

The original TRM and SIRM have low coercivities and distributed unblocking temperatures, as would be expected for large MD grains with numerous loosely pinned walls that move easily in response to the internal demagnetizing field. Evidence of these walls is also clear in the hysteresis results. Hysteresis curves at all temperatures are ramp-like and have the same slope, $\chi = 1/N$, where N is the demagnetizing factor (about 3.6 in cgs for the 3-mm crystal) (Figures 2 and 5). Coercive force H_c and saturation remanence M_{rs} vary with temperature in similar ways (Figure 6), their ratio being the same as N determined from the main hysteresis curve. The very low remanence ratio, $M_{rs}/M_s = 0.003$, and the very high coercivity ratio, H_{cr}/H_c = 42, are hallmarks of ideal MD behavior. The initial susceptibility χ_0, like the high-field susceptibility χ, is almost constant with temperature, and the χ_0-T curve lacks any Hopkinson peak (Figure 4).

The TRM and SIRM before LTD have clear MD properties. Coercivities are mainly low, and the demagnetization curve drops exponentially with increasing AF. Such exponential or subexponential decay is an MD characteristic [*Xu and Dunlop*, 1993]. In addition, the normalized AF demagnetization curve for SIRM lies above the corresponding curve for weak-field TRM (Figure 14). This is an MD-type result of the *Lowrie and Fuller* [1971] test. After LTD the test result becomes mixed. Both TRM and SIRM curves have initial plateaus, the SIRM being somewhat harder than the TRM at low coercivities, but there is a crossover around 25-30 mT to an SD-type trend, with TRM being more resistant to demagnetization than SIRM.

This crossover is caused by the fact that the high-coercivity part of the TRM increased after low-temperature cycling (Figure 7). Instead of being demagnetized, some of the original low-coercivity TRM was transformed to a more coercive remanence with the same direction. Thus LTD is not purely a cleaning technique, although its net effect is certainly to enhance stable remanence at the expense of less coercive remanence. The increased stability could result from additional crystal defects migrating to wall pinning sites at monoclinic twin boundaries in cycling across the Verwey transition (see section 6.4).

The rationale of the *Lowrie and Fuller* [1971] test has been discussed by *Bailey and Dunlop* [1983] and *Halgedahl and Fuller* [1983]. *Xu and Dunlop* [1995] show that the test result expresses a subtle balance between the pinning of domain walls away from their equilibrium positions and screening or reduction of the remanence by soft, unpinned walls. The changeover in the test result does not actually pinpoint the SD-MD transition size, or any specific grain size, because the balance between pinning and screening changes for different levels of internal stress. However, the MD-type result is a reliable indicator of coarse grains with MD behavior.

Figure 15 compares normalized stepwise thermal demagnetization curves of 1-mT TRM and SIRM. About 40% of the TRM and ≈60% of the SIRM have broadly distributed low and intermediate unblocking temperatures. Distributed unblocking temperatures are a typical MD phenomenon resulting from

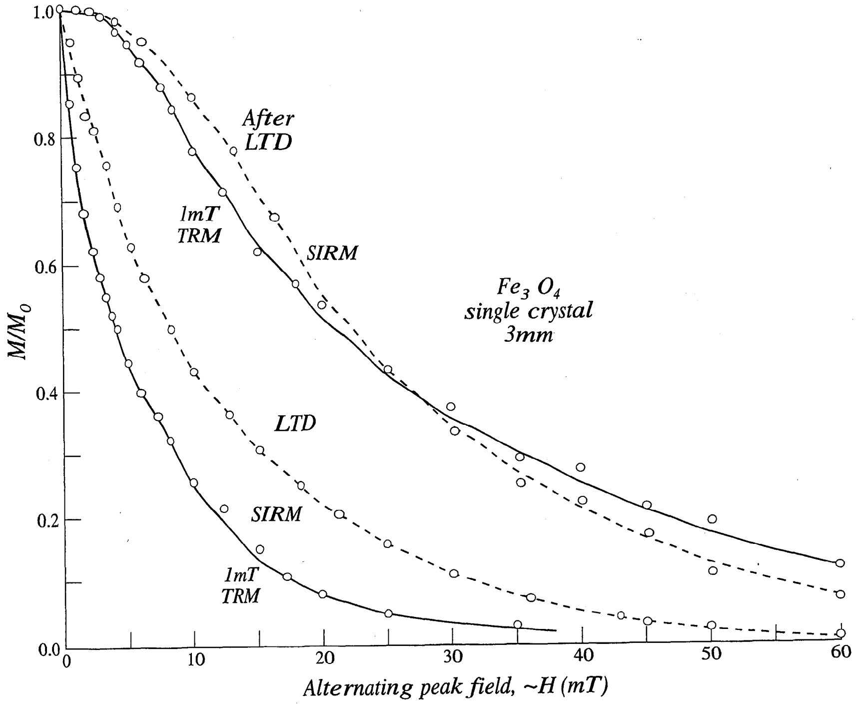

Figure 14. The AF demagnetization data of Figures 7 and 10 replotted after normalizing each curve to its own initial value of remanence, either before or after LTD. The contrast between the exponentially decaying curves of untreated TRM and SIRM and the much harder, SD-like curves of TRM and SIRM memory after LTD is brought out clearly. The result of the Lowrie-Fuller test is originally of MD type (untreated SIRM is harder than TRM) but after LTD crosses over to SD type (TRM harder than SIRM) above 25-30 mT.

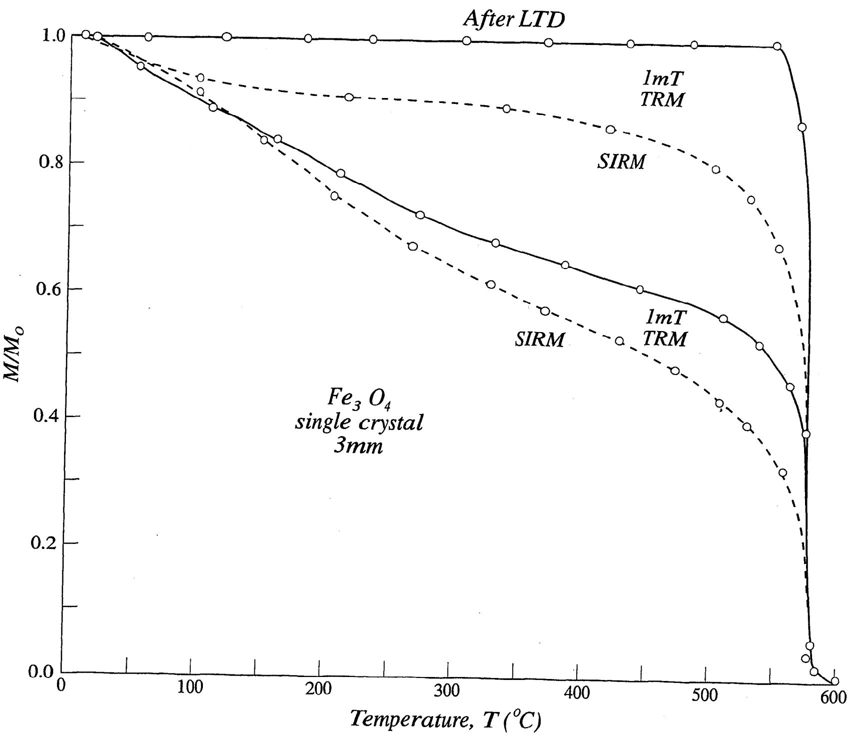

Figure 15. The thermal demagnetization data of Figures 8 and 11 replotted after normalization to initial remanence values for both original TRM and SIRM and for TRM and SIRM memories after LTD. The distributed low-T_{UB} range that is so prominent in the original TRM and SIRM data is largely or completely absent from the memories of these remanences after LTD. The SIRM memory demagnetizes mainly above 550°C, and the TRM memory demagnetizes entirely above 565°C.

successive adjustments of domain walls. The mechanism is pushing back or reequilibration of the walls by the internal demagnetizing field $\mathbf{H_d}$. The amplitude of the wall energy variations falls with heating. As the energy barriers decrease, the walls can successively jump toward the demagnetized state. This process continues to about 500°C. *McClelland et al.* [1996] recently isolated this remanence fraction in the form of pTRM (450°C, 20°C) carried by 100- to 150-μm magnetite grains and verified that unblocking continues above 450°C, in typical MD fashion. *Shcherbakova et al.* [1996] have also measured continuous distributions of T_{UB}s of pTRM (500°C, 450°C) in MD magnetite samples below, within, and above the 450°-500°C T_B range. Thus the typical MD thermal demagnetization response associated with continuous reequilibration of domain walls is well documented by other work as well as ours.

By way of contrast, above 500°C and especially above 560°C the high-T_{UB} fractions of TRM and SIRM in our crystals demagnetize sharply and have a more SD-like character. *Shcherbakova et al.*'s [1996] demagnetization curves of total TRM show a similar subdivision into separate low-temperature and high-temperature regions, separated by "shoulders" around 500°C, although the significance of these separate regimes is not remarked upon. The high-T_{UB} remanence is probably carried by hard walls, which are effectively pinned and resistant to internal field changes during heating and cooling cycles (see section 6.2).

The contrast between the thermal behavior of initial TRM or SIRM and the behavior of the TRM or SIRM memory is brought out clearly in Figure 15. The distributed T_{UB} fraction forms only 10-15% of the SIRM memory and is completely absent from the TRM memory. Notice that there is no thermal equivalent to the Lowrie-Fuller test: weak-field TRM is always more resistant to thermal cleaning than is SIRM, whatever the grain size or domain state.

The main drop in pTRM(T_C, 565°C) during zero-field heating comes above 565°C (Figure 13), in the same range as that for the TRM memory and the high-T_{UB} fraction of the total TRM (Figure 8). These remanences have the property $T_{UB} = T_B$, characteristic of SD TRM [*Néel*, 1949]. Possible mechanisms for this SD-like behavior in our very large MD crystals are considered next.

6.2. Possible Sources of SD-like Remanence in MD Crystals

Mechanisms of SD-like behavior of magnetite grains larger than SD size have been debated for 30 years. The upper limit for PSD behavior based on the grain size dependence of TRM intensity, or the related Koenigsberger ratio Q_t, is 10-20 μm [*Stacey*, 1963; *Parry*, 1965; *Dickson et al.*, 1966]. Early models appealed to regions with SD-like properties within MD crystals, so that an individual crystal could simultaneously have SD and MD responses to changing fields and temperatures. Examples are regions of deflected spins surrounding dislocations [*Verhoogen*, 1959], Barkhausen discreteness of wall positions [*Stacey*, 1962], structures at the surface terminations of domain walls [*Stacey and Banerjee*, 1974] and domain-wall moments [*Dunlop*, 1977]. In the 1980s the focus shifted to grains larger than equilibrium SD size in metastable SD states [*Halgedahl and Fuller*, 1980, 1983; *Boyd et al.*, 1984]. In this case an individual crystal would have either SD or MD behavior, but not both simultaneously.

Few of these mechanisms are viable in crystals like ours, whose volumes are orders of magnitude larger than those of 10- to 20-μm grains. Dislocation line, Barkhausen, and domain wall moments are too small to explain memories that are 10-20% of the total remanence. Surface moments associated with pinned closure and spike domains [*Özdemir et al.*, 1995] remain a possibility, but they too seem quantitatively insufficient. Metastable SD states are almost inconceivable for a millimeter size crystal like ours. Even if such a state were to exist, the observed combination of MD and SD-like behavior would not be explained. The crystal would be totally SD-like while in an SD state but would lose all SD-like properties as soon as a wall or walls nucleated.

We are forced to the conclusion that domain walls are the source of both MD and SD-like responses. Loosely pinned walls can account for hysteresis and the softer fraction of TRM or SIRM that demagnetizes at low to intermediate fields and temperatures. Strongly pinned walls must be the source of the hard, SD-like remanence that demagnetizes only at high fields and temperatures. The hardness of the SD-like remanence is mainly determined by internal stresses created by crystal defects [*Özdemir and Dunlop*, 1997]. Soft walls will respond to the internal demagnetizing field and partly screen the remanence of hard walls.

6.3. SD-like Memory After LTD

Remanence components with SD-like AF demagnetization curves following LTD have been observed previously for magnetites ranging in grain size from 0.2 μm [*Dunlop and Argyle*, 1991], just above true SD size, to 20-100 μm [*Heider et al.*, 1992; *McClelland and Shcherbakov*, 1995], in the upper PSD and lower MD range. The remarkable result of the present study is that single crystals of magnetite 3 and 4.5 mm in size also have a remanence fraction with SD-like behavior. In the earlier studies there was always the possibility of contamination by adhering finer grains, but that possibility has been eliminated in the present study.

In our crystals the memory following LTD has three SD-like properties.

1. The low T_{UB}s expected for pinned domain walls are absent. TRM and SIRM memories have almost no unblocking temperatures <550°C (Figures 8, 9, 11, 12).

2. The memory of a partial TRM with 565°C $\le T_B \le T_C$ demagnetizes sharply between 565°C and T_C (Figure 13). The blocking and unblocking temperatures are equal: $T_B = T_{UB}$.

3. AF demagnetization curves of TRM and SIRM memory have almost no low and intermediate coercivities, and there is an initial plateau of no demagnetization below 5-8 mT (Figures 7, 10). (In true SD grains the initial plateau is a result of the crystalline anisotropy threshold of ≈10-15 mT, which must be exceeded before any SD reversals can occur).

6.4. Possible Sources of SD-like Memory

Domain walls are pinned by crystal defects of many types [*Özdemir and Dunlop*, 1997] but pinning strong enough to explain SD-like coercivities of ≥30 mT after LTD is probably the result of defects and stress centers introduced when the crystal distorts from cubic to monoclinic structure in cooling through the Verwey transition. *Kobayashi and Fuller* [1968] were the first to consider the effect of stress centers on LTD and low-temperature memory. Depending on the relative orientation of the magnetoelastic and magnetocrystalline easy axes, they showed that a dislocation or dislocation pileup can produce either in-phase stress centers, which repel domain walls, or out-of-phase stress centers, which attract walls.

Such stress centers could result from twinning of the monoclinic phase below the Verwey transition. The elongation of the crystal as it deforms across the transition results in a twin structure [*Yamada et al.*, 1968; *Otsuka and Sato*, 1986;

Moloni et al., 1996]. Electron microscope observations at 77 K clearly show two kinds of boundaries between the twins, which have orthogonally oriented c axes. One type of twin boundary is perpendicular to one of the c axes, and the other makes a 45° angle with both c axes [*Chikazumi et al.*, 1971, Figure 2].

This monoclinic twin formation due to crystal distortion as the magnetite crystal cools through the cubic → monoclinic phase transition around 115 K may be the source of the defect concentrations necessary to explain recovery of remanence on warming (memory) and strong pinning. Defects will be localized along the twin boundaries where regions of the crystal elongated along different c axes meet. The magnetostriction constants λ_{100} and λ_{111} of the monoclinic phase show sharp changes below the transition temperature, probably resulting from magnetoelastic coupling with the lattice distortion [*Tsuya et al.*, 1977].

As the crystal is rewarmed through the transition and the crystal reverts to cubic symmetry with ⟨111⟩ easy axes, some domain walls will be attracted back to their former pinning sites. Note that the change of crystalline easy axes from ⟨111⟩ to ⟨001⟩ causes centers of pinning to become centers of repulsion for domain walls [*Kobayashi and Fuller*, 1968]. This occurrence, rather than wall broadening, is likely the principal cause of remanence loss in the first crossing of the Verwey transition.

6.5. SD-like Partial TRM

We separated SD-like remanence from total TRM or SIRM either as memory after LTD of the total remanence or as a high-temperature partial TRM. The two remanences are not identical—memory of total TRM has a lower intensity than pTRM(T_C, 565°C)—but the two have very similar thermal demagnetization behavior. Their unblocking temperatures T_{UB} are almost exclusively ≥565°C, which is the same as the range of blocking temperatures T_B in the case of the pTRM.

Concentrations of defects in the crystal, although not necessarily the same ones caused by low-temperature twinning and responsible for memory, are the likely cause of SD-like high-temperature partial TRM. In this way, low-temperature memory and high-temperature pTRM would have similar hard AF and thermal demagnetization behavior but not necessarily the same intensity or set of domain wall displacements.

7. Conclusions

Low-temperature demagnetization is an effective means of removing a large part of the MD remanence due to domain wall pinning and isolating the SD-like remanence. In some cases (e.g., Figure 7), part of the MD remanence is not destroyed by cycling through the Verwey transition but is transformed to memory with higher coercivity. These observations are explained by a model in which remanence is lost in cooling through the cubic → monoclinic transition at T_V, but stress centers at monoclinic twin boundaries cause repinning of walls and partial recovery of remanence (memory) at T_V in reheating.

Using low-temperature cycling and memory, we have shown that natural single crystals of magnetite 3 mm and 4.5 mm in size contain distinct SD-like and MD remanence components. The AF and thermal demagnetization characteristics of TRM, pTRM, and SIRM are dramatically different before and after LTD.

The initial remanences without low-temperature treatment have the following properties:

1. AF demagnetization curves of original TRM and SIRM decayed exponentially with increasing field, indicating MD behavior. SIRM was harder than TRM, an MD-type result of the Lowrie-Fuller test. Ramp-like hysteresis curves with a demagnetizing factor of 3.59 at all temperatures confirm truly MD behavior.
2. Thermal demagnetization of initial TRM and SIRM isolated an SD-like remanence with high unblocking temperatures, $T_{UB} > 500°C$. This remanence forms about 60% of the original TRM and about 40-50% of the original SIRM.
3. The pTRM(T_C, 565°C) has 80% of its unblocking temperatures $T_{UB} > 565°C$, in the same range as the pTRM blocking temperatures T_B. The property $T_{UB} = T_B$ is characteristic of SD behavior.

After LTD the low-temperature memory has the following properties:

1. The TRM and SIRM memories have SD-like AF decay curves with initial plateaus of no demagnetization below 5-8 mT. The two memories have quite similar decay curves; above 25 mT the TRM memory is harder than the SIRM memory, an SD-type Lowrie-Fuller test.
2. The TRM and SIRM memories have practically no unblocking temperatures below 560°C and 550°C, respectively.
3. The memory fraction of pTRM(T_C, 565°C) has mainly unblocking temperatures $T_{UB} > 565°C$, very similar to those of the memory fraction of total TRM.

Crystal defects and resulting stress centers, rather than separate SD regions in our crystal, are likely responsible for the SD-like memory. A plausible source of the defect concentrations necessary to explain the recovery of remanence in low-temperature cycling and the strong pinning of this memory is twinning of the monoclinic phase of magnetite below the Verwey transition.

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