Stepwise and continuous low-temperature demagnetization

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[1] Magnetites with sizes from 1 μm to 135 μm were cooled in zero field and their magnetizations $M(T)$ measured continuously. $M(T)$ changed reversibly in cooling from T_0 = 300 K to 200 K, and in subsidiary warming-cooling cycles $T_i \rightarrow T_0 \rightarrow T_i$ for any T_i. Changes in $M(T)$ in cooling from 200 K to 130 K were largely irreversible due to decreasing magnetocrystalline anisotropy which promotes wall unpinning and domain nucleation. Low-temperature demagnetization (LTD) is almost complete by 130 K in 20–135 μm magnetites but in 1–14 μm magnetites further LTD occurs on cooling to 120 K as magnetocrystalline easy axes change and domains reorganize at the Verwey transition. The observed irreversible changes are the basis of stepwise LTD as a method of paleomagnetic "cleaning." Decrements ΔM in remanence due to cooling are most accurately measured at T_0, requiring a set of warming-cooling cycles $T_i \rightarrow T_0 \rightarrow T_i$. A less accurate method, continuous LTD, measures decrements $M(T_i) - M(T_{i-1})$ from the main cooling curve below 200 K, without intermediate warming-cooling cycles; this requires remanence measurements at $T_i < T_0$. Stepwise or continuous LTD curves $M(T_i)$ discriminate among remanence types and grain sizes. The signal of finer (PSD) grains is enhanced compared to coarser (MD) grains. Analogous to the *Lowrie and Fuller* [1971] test, the inverse thermoremanence (ITRM) of 1–14 μm grains is harder to stepwise LTD than saturation remanence (SIRM), while anhysteretic remanence (ARM) is harder than either; for 20–135 μm multidomain grains, ITRM is softer than SIRM. *INDEX TERMS:* 1525 Geomagnetism and Paleomagnetism: Paleomagnetism applied to tectonics (regional, global); 1527 Geomagnetism and Paleomagnetism: Paleomagnetism applied to geologic processes; 1540 Geomagnetism and Paleomagnetism: Rock and mineral magnetism; 1594 Geomagnetism and Paleomagnetism: Instruments and techniques. **Citation:** Dunlop, D. J., Stepwise and continuous low-temperature demagnetization, *Geophys. Res. Lett.*, *30*(11), 1582, doi:10.1029/2003GL017268, 2003.

1. Introduction

[2] Thermal and alternating field (AF) demagnetization are used by paleomagnetists to selectively erase or "clean" the natural remanent magnetization (NRM) of rocks. Both techniques are carried out in stepwise fashion. Increasing temperatures or AFs mobilize successively higher blocking temperature or coercivity fractions of the NRM.

[3] Low-temperature demagnetization (LTD) is zero-field cycling between room temperature T_0 and the Verwey transition T_V = 120 K. Domains and domain walls in magnetite reorganize as a result of changes in magnetocrystalline anisotropy, most notably at the isotropic point T_K = 130 K, where K_1 changes sign, and at T_V as the crystal symmetry changes from cubic to monoclinic. LTD is an effective cleaning technique [*Ozima et al.*, 1964; *Merrill*, 1970; *Dunlop and Argyle*, 1991; *Heider et al.*, 1992] but has rarely been applied in paleomagnetism [*Dunlop et al.*, 1997; *Warnock et al.*, 2000], and then only as a single step to 77 K.

[4] Magnetization changes continuously during cooling, not just at T_K and T_V [*Özdemir et al.*, 2002], but how much of this is reversible remains unclear. The present paper reports reversible and irreversible changes when various remanences are rewarmed to T_0 after cooling is interrupted at $T > T_K$. The permanent losses due to zero-field cycling between T_0 and a set of $T_i > T_K$ demonstrate the feasibility of stepwise LTD as a cleaning technique.

2. Samples and Experiments

[5] Nine natural magnetites were prepared by crushing large single crystals. Fractions with mean grain sizes of 0.6, 1, 3, 6, 9, 14 and 20 μm were separated using a centrifuge and 110 and 135 μm fractions by sieving. Samples were vacuum annealed for several hours at 700°C to relieve internal stress.

[6] Samples were cooled to $T \geq 30$ K and axial magnetization was measured in a Quantum Design MPMS2 magnetometer. Three remanence types were used: anhysteretic remanence (ARM) produced at T_0 by a 0.1 mT steady field superimposed on an AF decaying from 100 mT to 0; saturation isothermal remanence (SIRM) in 1 T; and inverse thermoremanence (ITRM) produced by warming from 30 K to T_0 = 300 K in 0.2 mT. Stepwise LTD was performed by cycling from T_0 to a set of T_i (250, 200, 150, 130, 120, 110, 100 and 90 K) and back to T_0 in zero field. Magnetization was measured at 5 K intervals, although only the results at T_0 are needed for a practical stepwise LTD procedure.

3. Low-Temperature Cycling Results

[7] Continuous measurements during low-temperature cycling of ARM give curves with a wide variety of shapes (Figures 1 and 2). In some cases, the magnetization first increases with zero-field cooling from T_0 to ≈200 K, passes through a broad peak, then decreases with further cooling (Figure 1). At the other extreme, magnetization may decrease steeply in initial cooling and then more gradually below ≈200 K (Figure 2). The curves have the same shape for each cycle and nest within the envelope curve tracing LTD in a single cycle to 90 K.

[8] The cooling curve from T_0 to each temperature T_i where cooling is suspended and warming begins contains both reversible and irreversible changes in magnetization M.

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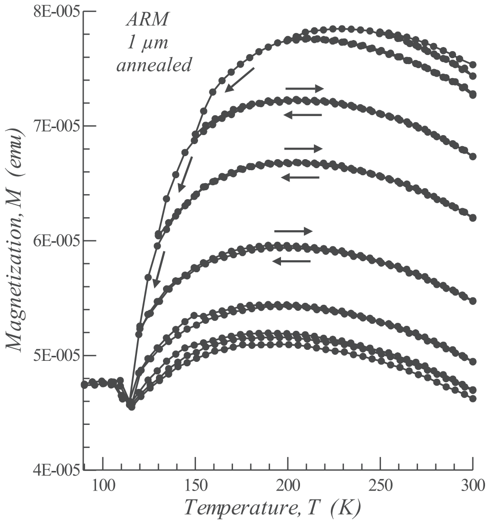

Figure 1. Zero-field cycling of ARM of 1 μm magnetite. Intermediate warming-cooling cycles are reversible and nest within the main cooling curve. Irreversible decreases in ARM measured at 300 K (right) constitute stepwise LTD. Approximately equal decreases measured along the main curve (left) below ≈200 K constitute continuous LTD. Changes in M along the main curve between 300 and 200 K are mainly reversible.

However, all changes in M in the intermediate warming-cooling cycle from $T_i \rightarrow T_0 \rightarrow T_i$ are entirely reversible: the $T_0 \rightarrow T_i$ cooling curve exactly retraces the $T_i \rightarrow T_0$ warming curve. All irreversible changes ΔM occur along the main envelope cooling curve between T_i and T_{i+1}. The permanent decrements ΔM that define the stepwise LTD curve $M(T_i)$ are measured at T_0. Unexpectedly, approximately the same decrements ΔM are measured directly from the main cooling curve as $M(T_i) - M(T_{i+1})$ when 90 K $\leq T_i \leq$ 200 K. That is, below 200 K, changes in M along the envelope cooling curve are almost entirely irreversible.

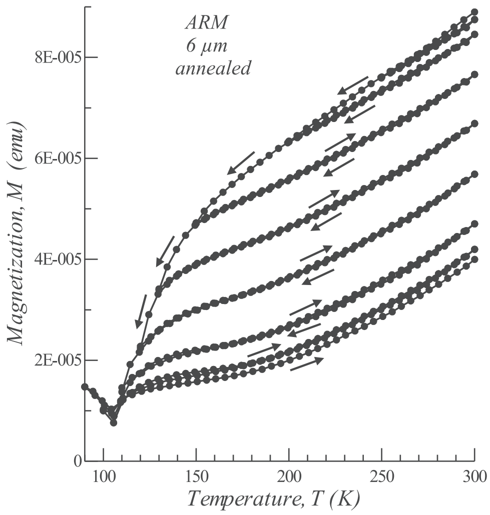

Figure 2. Zero-field cycling of ARM of 6 μm magnetite. Reversible changes in M from 300 to 200 K are very different from those in Figure 1, but the pattern of irreversible changes is identical.

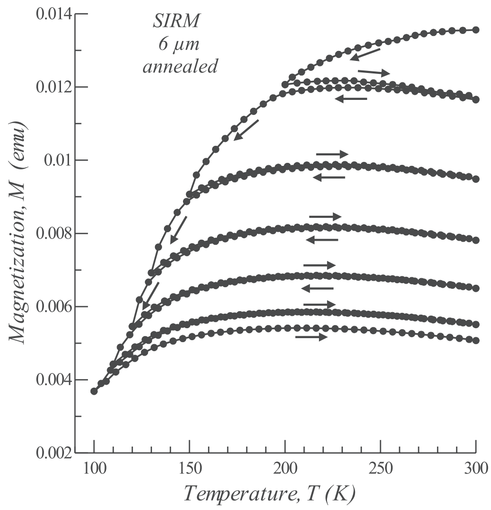

Figure 3. Zero-field cycling of SIRM of 6 μm magnetite. Unlike ARM, all samples have the same pattern of changes in SIRM, including considerable irreversible loss in M in cooling to 200 K and slight further irreversibility in intermediate warming-cooling cycles.

[9] Conversely, in the 200–300 K range, the point-by-point slopes dM/dT of the envelope and nested curves reflect mainly reversible domain rotations or displacements of domain walls. These are quite variable in the case of ARM. In the 6 μm magnetite, walls are driven strongly towards a demagnetized state in cooling from T_0 to 250 and 200 K but return almost to their original states when the sample is rewarmed to T_0; there is <10% permanent loss ΔM (Figure 2). Walls in the 1 μm magnetite are driven in the opposite direction, away from the demagnetized state, over the 300–200 K range, but the pattern of irreversible magnetization change ΔM is identical (Figure 1).

[10] Low-temperature cycling curves for ITRM, measured as the first (zero-field) of two cycles to T_i in an inverse Thellier paleointensity experiment [*Dunlop and Yu*, 2003], have similar shapes to the ARM curves. However, SIRM curves for all samples have the shape illustrated in Figure 3, regardless of the varied shapes of the ARM and ITRM cycling curves. SIRM involves different wall motions than ARM and ITRM, including considerable irreversible remanence loss in the 200–300 K range. There is also a consistent irreversibility in the second SIRM cooling from $T_0 \rightarrow T_i$. The data fall slightly below the $T_i \rightarrow T_0$ warming curve, showing a small further loss ΔM.

4. Stepwise LTD Results

[11] Like AF or thermal demagnetization curves, stepwise LTD curves of ITRMs (Figure 4) plot the fraction of initial remanence remaining after a complete demagnetization step, here an $H = 0$ cycle from T_0 to T_i and back to T_0, as a function of T_i. The LTD spectrum of ITRM peaks at lower T as grain size decreases, although 1, 3, 6 and 9 μm magnetites have quite similar curves. The largest fraction of ITRM is demagnetized between 130 and 120 K for the smaller grain sizes and between 150 and 130 K for the 20 and 135 μm magnetites (Figure 4). A judicious choice of demagnetizing temperature greatly enhances the remanence carried by the finer grains. The 130 K step, for example,

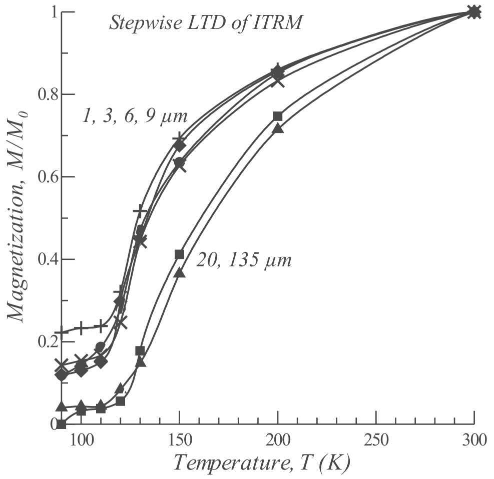

Figure 4. Results of stepwise LTD of ITRM for selected samples. All measurements were made at room temperature in intermediate warming-cooling cycles from a set of low temperatures *T*. There is a clear separation between harder LTD curves for PSD-size (1, 3, 6, 9 μm) and softer curves for MD-size (20, 135 μm) magnetites.

reduces the initial ITRM of the 20 and 135 μm magnetites to <20% but leaves ≈50% of the ITRM of the 1, 3, 6, and 9 μm grains untouched. The grain-size dependence of LTD stability for ARM and SIRM is similar but less pronounced.

[12] For 1, 3, 6 and 9 μm magnetites, ITRM is ''harder'' than SIRM, i.e., it must be cooled to a lower *T* to reach the same level of LTD, but ITRM is ''softer'' to LTD than weak-field ARM (Figure 5). For 20, 110 and 135 μm magnetites, ITRM is softer to stepwise LTD than SIRM (Figure 5); ARM (not shown) is harder in early LTD steps, but ultimately (130 K and lower) marginally softer than either. Thus stepwise LTD selectively erases certain types of remanence. Equally important, stepwise LTD is diagnostic of grain size because the relative stabilities of ITRM and SIRM are opposite below and above the magnetite PSD threshold (10–20 μm).

[13] ITRM is ''reciprocal'' to LTD in the same way that ARM is reciprocal to AF and thermal demagnetization, respectively. The hardness trends for AF demagnetization of ARM and SIRM in SD/PSD and MD magnetites are identical to the analogous ITRM/SIRM trends (Figure 5). ARM is more resistant to AF demagnetization than SIRM for SD and PSD magnetites, while the opposite is true for MD magnetites [*Dunlop and West*, 1969; *Lowrie and Fuller*, 1971; *Johnson et al.*, 1975; *Bailey and Dunlop*, 1983]. The results of Figure 5 thus establish a ''Lowrie-Fuller test'' for stepwise LTD.

5. Discussion

[14] The idea of stepwise LTD to a series of *T* between 180 and 135 K was proposed by *Halgedahl and Jarrard* [1995], who were the first to demonstrate irreversible Barkhausen jumps in magnetization during cooling in this range. *Hodych et al.* [1998] interrupted cooling at 135 K and found that MD and large PSD size magnetites had already lost more than half their initial SIRM before reaching T_K. Five-step LTD results were reported for ARM in 11 μm magnetite [*Muxworthy et al.*, 2003].

[15] *Halgedahl and Jarrard* [1995] suggested that measurements of *M* during low-temperature cooling (continuous LTD) might provide an equivalent paleomagnetic treatment. The results of Figures 1–3 support this provided data are taken below 200 K. Changes in *M* along the main cooling curve are largely irreversible in this range, whereas 300–200 K changes are almost reversible. Intermediate cycles from $T_i \rightarrow T_0 \rightarrow T_i$ are reversible for $T_i < 200$ K, and so ΔM measured after rewarming from T_i to T_0 in stepwise LTD is very nearly equal to $M(T_i) - M(T_{i-1})$ measured along the envelope cooling curve in continuous LTD.

[16] The shapes of the main cooling curves for SIRM in the present study (Figure 3) resemble those of *Hodych et al.* [1998]. They showed that the shapes matched the variation $H_C(T)$ of bulk coercive force over the 300–130 K range and hypothesized that internal stress controls H_C, the variation being due to magnetostriction $\lambda_S(T)$. Since our magnetites are annealed, stress is likely not the sole cause of H_C. In any case $\lambda_S(T)$ varies much less between 300 and 120 K than magnetocrystalline anisotropy K_1.

[17] *Özdemir* [2000] showed that $H_C(T)$ in large MD magnetites follows $\lambda_S(T)/M_S(T)$ between 300 and 170 K but is closer to $K_1(T)/M_S(T)$ between 170 and 130 K. Thus in the 300–200 K range where the present samples show mainly reversible changes in SIRM, ARM and ITRM, wall segments move (by large amounts in some cases: Figure 2) without breaking free of their pinning points, probably dislocations. Pinning is determined by $\lambda_S(T)/M_S(T)$, which dictates $H_C(T)$. Below ≈200–180 K, where most irreversible changes ΔM occur (Figures 1–5) K_1 plummets and walls broaden, promoting their unpinning from dislocations. Nucleation of new walls is favored by strongly decreasing $K_1(T)$ and probably occurs in this range, as it does during heating from T_0 to ≈400 K where $K_1(T)$ also decreases strongly [*Heider et al.*, 1988].

[18] The efficacy of stepwise LTD in cooling from ≈200 K to T_K = 130 K is well accounted for by the strong

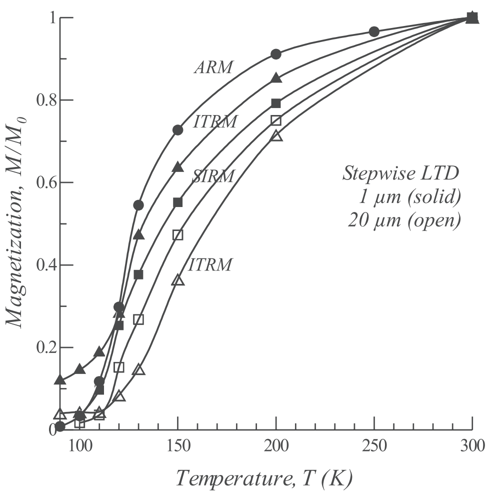

Figure 5. Comparative stepwise LTD curves for ARM, ITRM and SIRM of PSD-size (1 μm) and MD-size (20 μm) magnetites. The remanence of the finer fraction of magnetite in any real sample will be enhanced by stepwise LTD. The results also establish a ''Lowrie-Fuller test'' for LTD: ITRM is harder than SIRM for PSD grains but softer than SIRM for MD grains.

decrease and ultimate vanishing of K_1 over this range. Stepwise LTD is essentially complete by 130 K for MD (20–135 μm) magnetites, but for the PSD magnetites (1–14 μm) further demagnetization ΔM occurs between 130 K and T_V = 120 K (Figures 4 and 5). Since $K_1(T)$ increases over this range, further nucleation or unpinning of walls seems unlikely. More likely the reordering of crystalline easy axes at T_V causes a reorganization of the domains, expressed in this final ΔM. Cycling to $T < T_V$ (110, 100, 90 K) causes little or no further demagnetization.

[19] The relative stepwise LTD efficiencies of various grain sizes and types of remanence shown in Figure 5 are in addition to the well known contrasts in memory when LTD is carried out in a single step across T_V and back to T_0. The LTD memories are only 5–20% for ITRM (Figure 4) but are much larger for SIRM and ARM in the finer grains. They have been subtracted from the ARM and SIRM data in Figure 5, in order to more clearly compare the shapes of the LTD demagnetization spectra. If not removed from the data, their effect is to further separate the stepwise LTD curves for ARM relative to SIRM and for fine relative to coarse grains, thereby actually enhancing the resolving power of the stepwise LTD method and its usefulness in separating NRM components of different origins.

6. Conclusions

[20] 1. Magnetization $M(T)$ changes reversibly when magnetite is cooled from T_0 to ≈200 K, and in subsidiary warming-cooling cycles $T_i \rightarrow T_0 \rightarrow T_i$ for 90 K $\leq T_i \leq T_0$. Walls move large distances but they remain pinned, and no new walls nucleate.

[21] 2. Changes $M(T)$ in cooling from ≈200 K to T_K = 130 K are largely irreversible and result from the precipitous decrease in K_1, promoting wall unpinning and nucleation of new domains. LTD is almost complete by 130 K in 20–135 μm magnetites.

[22] 3. In 1–14 μm PSD magnetites, further LTD occurs on cooling to T_V = 120 K as easy axes change and domains reorganize.

[23] 4. Irreversible changes 2 and 3 are the basis of stepwise LTD. Decrements ΔM in initial remanence are most accurately measured at T_0 in a set of warming-cooling cycles $T_i \rightarrow T_0 \rightarrow T_i$.

[24] 5. An approximate method (continuous LTD) is to measure decrements $M(T_i) - M(T_{i-1})$ from the main cooling curve below 200 K. This may be sufficiently accurate for paleomagnetic applications, but it requires remanence measurements at $T_i < T_0$.

[25] 6. Stepwise or continuous LTD curves $M(T_i)$ discriminate among remanence types and grain sizes. Signals of finer (PSD) grains are enhanced at the expense of coarser (MD) grains.

[26] 7. Stepwise LTD provides an analog to the *Lowrie and Fuller* [1971] test. For 1–14 μm PSD grains ITRM is harder to demagnetize than SIRM (and ARM is harder than either), whereas for 20–135 μm MD grains, ITRM is softer than SIRM. ITRM is the natural standard of comparison because it is the reciprocal remanence to LTD.

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