GEOPHYSICAL RESEARCH LETTERS, VOL. 27, NO. 9, PAGES 1311-1314, MAY 1, 2000

Effect of Grain Size and Domain State on Thermal Demagnetization Tails

David J. Dunlop and Özden Özdemir

Geophysics, Physics Department and Erindale College, University of Toronto, Toronto, Canada

Abstract. Thermal demagnetization of viscous remanence (VRM) and partial thermoremanence (pTRM) of 20- and 135-µm natural magnetites reveals a broad spectrum $f(T_{UB})$ of unblocking temperatures T_{UB}, both > and < T_B, the blocking temperature. In contrast, 0.04-µm single-domain (SD) grains demagnetize sharply at $T_{UB} \approx T_B$. High- and low-T_{UB} tails of $f(T_{UB})$ were wider for 135-µm multidomain (MD) grains than for 20-µm PSD grains. 10-mT alternating-field demagnetization rendered the VRM of the 20-µm magnetite more SD-like in subsequent thermal cleaning, selectively erasing both low- and high-T_{UB} tails of $f(T_{UB})$. Primary and secondary remanences are then cleanly separated, and non-linear Thellier paleointensity determination becomes linear. Our results agree well with (T_B, T_{UB}) data for 10^5-year thermoviscous overprints in the Milton Monzonite over the range of log(time) common to the two studies. Because $(T_{UB})_{av} \approx T_B$ for PSD and MD as well as SD grains, the Pullaiah et al. paleothermometry method will work if $(T_{UB})_{av}$ values are used instead of $(T_{UB})_{max}$ values.

Introduction

Viscous remanent magnetization (VRM) acquired at constant temperature T over a long time t and partial thermoremanent magnetization (partial TRM or pTRM) acquired during cooling through a range of T commonly overprint natural remanent magnetization (NRM) in rocks. Both are acquired at a blocking temperature T_B (or a range of T_B's). Time is also involved because rotation of spins in a single-domain (SD) grain or motion of a domain wall (DW) in a multidomain (MD) grain is thermally activated. The relaxation time is

$$1/\tau = C \exp(-E_b / kT) \qquad (1)$$

[Néel, 1949], where $C \approx 10^{10}\ s^{-1}$ and k is Boltzmann's constant. Longer t means a higher probablility of surmounting the energy barrier E_b and therefore a larger VRM or pTRM.

In paleomagnetic studies, the goal is to isolate the primary NRM by erasing VRM and pTRM overprints. Thermal demagnetization should give a clean separation for SD grains. The unblocking temperature T_{UB} is equal to T_B if t is similar for magnetization and demagnetization. For MD grains, anomalously high T_{UB}'s are found, forming a tail extending to the Curie point T_C [Bol'shakov and Shcherbakova, 1979]. Separating primary and secondary NRM's is then difficult.

In paleothermometry, the thermoviscous overprint is the target and the goal is to determine T_B in nature on a geological time scale from measurement of T_{UB} on a laboratory time scale [Pullaiah et al., 1975]. This is only simple for SD grains, with their 1-1 relation between T_B and T_{UB}. For MD grains, a single T_B gives a spectrum $f(T_{UB})$, including anomalously high T_{UB}'s.

Paper number 1999GL008461.
0094-8276/00/1999GL008461$05.00

In determining paleointensity of the geomagnetic field, NRM of TRM origin is replaced, in steps, by laboratory pTRM's. The ratio of ancient / laboratory field is given by the NRM / pTRM ratio [Thellier and Thellier, 1959]. The NRM-pTRM graph is linear only for SD grains with $T_{UB} = T_B$. As we will show later, for MD grains anomalously *low* T_{UB}'s cause non-linearity and complicate the Thellier method.

Earlier [Dunlop and Özdemir, 1993] we thermally demagnetized laboratory VRM's and pTRM's of SD magnetites and confirmed the predictions of Néel [1949]. The present experiments are on larger grains of magnetite, obtained by crushing natural crystals, separating fractions with mean grain sizes of 20 µm and 135 µm, and annealing to relieve internal stress. 135-µm grains have truly MD behavior (DW's fully responsive to internal demagnetizing fields). 20-µm magnetites are in the pseudo-single-domain (PSD) range and have a blend of MD and SD-like responses. We examine how grain size and magnetic domain state affect thermal demagnetization and test methods of reducing high- and low-T_{UB} tails, to make the thermal response SD-like and paleomagnetically more useful.

Experiments

Stepwise thermal demagnetization of VRM's produced in 3.5 hr exposures to a field of 0.1 mT at fixed temperature T_B gave very different results for SD, PSD and MD magnetites (Figure 1). VRM was produced after AF demagnetizing the SD and 20-µm samples. The initial state for the 135-µm data shown was thermally demagnetized but an AF demagnetized state gave similar results. In each case, 40-50% of the VRM was erased by zero-field heating to T_B, but the spread of T_{UB}'s about T_B covered a range of ±35°C for the SD grains, about ±150°C for the 20-µm grains, and practically the entire range from T_0 to T_C = 580°C for the 135-µm MD grains. 5% and 95% limits were used to arrive at these ranges, but the broadening of the T_{UB} spectrum is obvious to the eye. It is clear that high-T_{UB} tails, which have been much discussed in the literature [Worm et al., 1988; Halgedahl, 1993; McClelland and Shcherbakov, 1995; McClelland et al., 1996], are not the only hallmark of MD behavior. There is a matching tail of low T_{UB}'s in both the 20-µm and 135-µm data. The symmetry of the low-T_{UB} and high-T_{UB} tails, with a mean T_{UB} very close to T_B, must be physically significant. We give an interpretation later.

For SD grains, VRM and pTRM are analogs. We calculate from eqn. (1) that an SD pTRM acquired between 370°C and 350°C should activate the same range of τ as a VRM acquired over 3.5 hr at 350°C. Figure 2 shows that the same equivalence holds for much larger grains, in this case 135 µm. VRM's produced at 350°C have thermal demagnetization curves indistinguishable from those of a pTRM produced from 370-350°C. Initial states were AF demagnetized. The mechanism of VRM or pTRM production in these large MD grains is quite different from SD rotation, but the VRM / pTRM equivalence still holds.

We next investigated the effect of alternating-field (AF) demagnetization on VRM's acquired at 350°C (135 µm) or 357°C (20

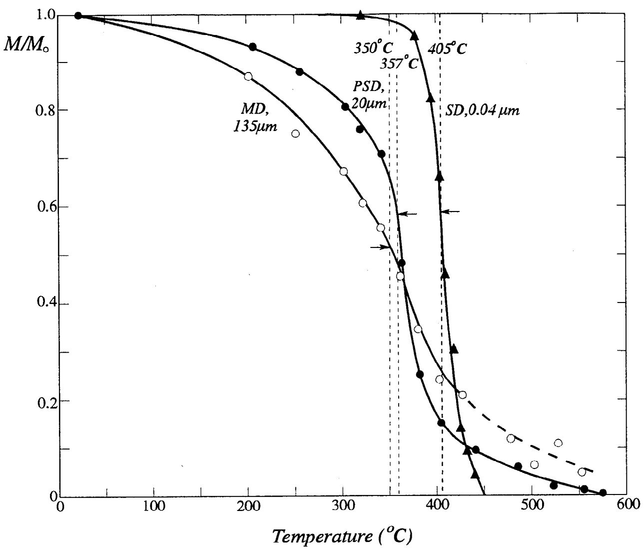

Figure 1. Stepwise thermal demagnetization of VRM's produced over 3.5 hr at $T_B = 350°C$ (135 μm magnetites), 357°C (20 μm), and 405°C (0.04 μm). Unblocking temperatures T_{UB} are concentrated near T_B for SD grains but broadly distributed for PSD and MD grains, with both low-T and high-T tails. The remanence when $T_{UB} = T_B$ (arrows) is 50-60% in all cases. Thus the mean of the T_{UB} distribution is $\approx T_B$, whatever the grain size or domain state.

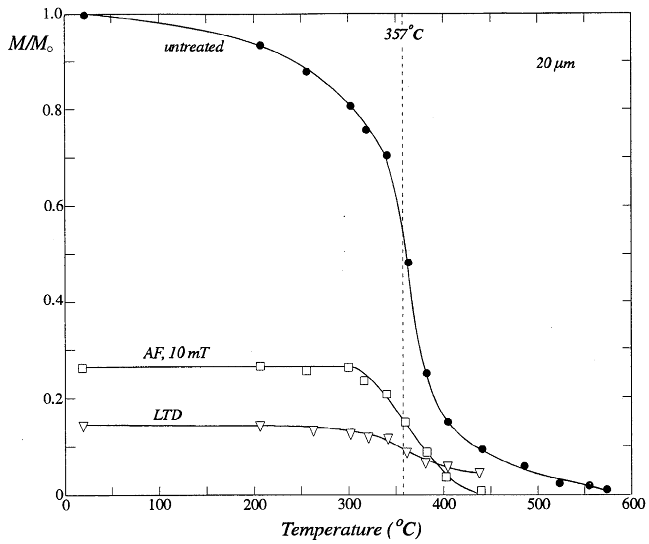

Figure 3. The effect of pre-treatments on the thermal demagnetization of VRM acquired at 357°C by 20 μm magnetite grains. 10-mT AF cleaning greatly reduces both low-T_{UB} and high-T_{UB} tails in subsequent thermal demagnetization. Prior LTD suppresses only the low-T_{UB} tail.

μm). VRM is very soft, with a median demagnetizing field of ≈5 mT in both cases. About 30% of the VRM remains after a peak AF of 10 mT.

The residual VRM of the 20-μm sample after 10-mT AF demagnetization was now thermally demagnetized in steps. Following full AF cleaning, VRM was again induced at 357°C and subjected to low-temperature demagnetization (LTD), zero-field cycling to 77 K and back to room temperature T_0. The residual after LTD, ≈17% of the initial VRM, was then thermally demagnetized.

Both LTD and AF cleaning selectively remove low-stability remanence, e.g., that of loosely pinned DW's. The effect on the T_{UB} spectrum is seen in Figure 3. AF pre-treatment resulted in no thermal demagnetization below ≈300°C or above 425°C: low-T_{UB} and high-T_{UB} tails essentially disappeared. Demagnetization now occurs within ±60°C around $T_B = 357°C$. LTD removed most of the low T_{UB}'s but not all the high T_{UB}'s. There is a clear association between the tails of the T_{UB} spectrum and low-stability remanence removed by AF cleaning or LTD.

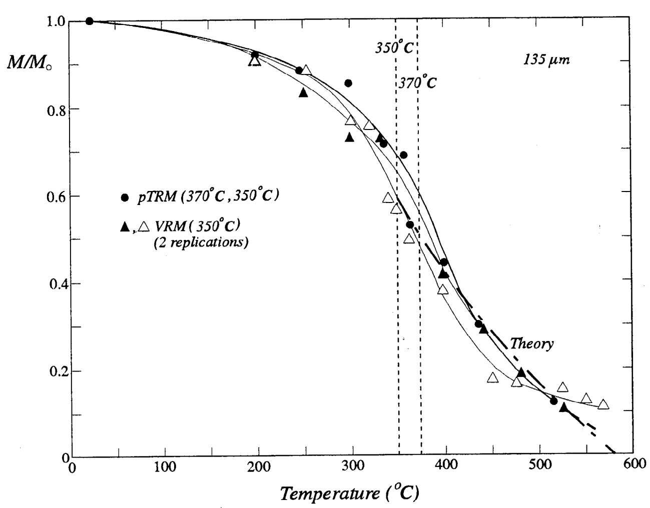

Figure 2. Stepwise thermal demagnetization data for 135 μm magnetite grains, demonstrating the equivalence of VRM and narrow-band pTRM with the same T_B range. The high-T tail is as predicted by MD pTRM theory [Xu and Dunlop, 1994].

Paleothermometry

The average T_{UB} of a VRM or pTRM is quite close to its average T_B, not only for SD grains, but for PSD and MD grains as well (Figures 1, 2). Paleothermometry (see Introduction) would work satisfactorily for grains of any size or domain state if average values of T_B and T_{UB} were used. In practice, overlapping T_{UB} spectra of the primary NRM and the VRM/pTRM overprint make $(T_{UB})_{av}$ of the overprint hard to determine. $(T_{UB})_{max}$ of the overprint, above which the primary demagnetizes univectorially in a vector plot, is easy to pinpoint and is generally used.

Figure 4 compares (T_B, 3.5 hr) and ($(T_{UB})_{max}$, 100 s) for VRM's reduced to the 10% level by rapid heating to $(T_{UB})_{max}$. For 0.04 μm SD grains, lines joining such pairs of points on a t−T graph parallel the Pullaiah et al. [1975] SD remagnetization contours. For the 20-μm and 135-μm grains, the lines joining T_B and T_{UB} pairs are increasingly oblique to the Pullaiah et al. contours, because PSD and MD $(T_{UB})_{max}$ values are "anomalously" high compared to SD values. If $(T_{UB})_{av}$ values were plotted instead, the PSD and MD lines would parallel the contours.

In the Milton Monzonite, Dunlop et al. [1997] found three groups of samples with thermal overprints carried by SD magnetite (A in Figure 4), MD magnetite (C), and intermediate sizes or mixtures of SD and MD (B). Interestingly, the lines joining the Milton A, B, and C ($(T_{UB})_{max}$, t_{lab}) points to their common (T_B, t_{nature}) point roughly parallel the present SD, PSD, and MD lines over the range of log t common to the two studies. The interpretation of the Milton thermal data is thus confirmed by the present control study, in which grain size and domain state are known. Finding reasonable $(T_{UB})_{av}$ values from the B and C thermal data is a subject for future study.

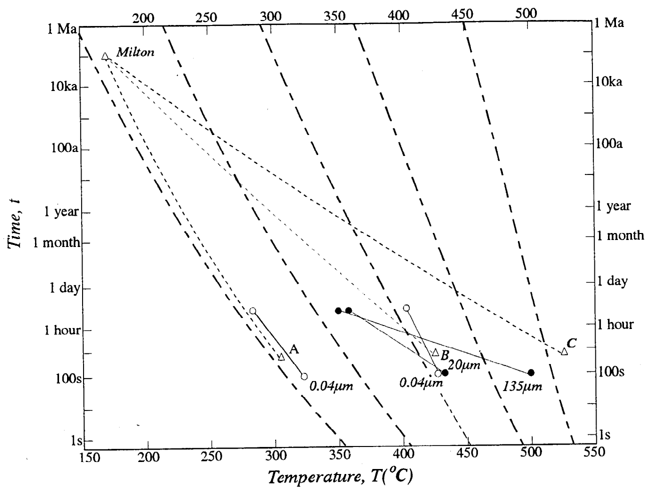

Figure 4. A comparison of corresponding (t, T) values in remanence blocking and unblocking. The 20 μm and 135 μm data pairs of this paper intersect the Pullaiah et al. [1975] t–T contours for SD magnetite (dot-dash lines), but more or less parallel the PSD and MD data pairs for the Milton Monzonite (B, C, dashed lines). If $(T_{UB})_{av}$ values were used instead of $(T_{UB})_{max}$ values, the 20 μm and 135 μm lines would mimic SD behavior and parallel the Pullaiah et al. lines (see text)

Discussion

We have examined how grain size and domain state affect thermal demagnetization of VRM and pTRM with narrow ranges of T_B, and how these remanences might be pre-treated in the case of PSD and MD grains to render them more useful paleomagnetically. Our results confirm that VRM and pTRM are analogous for MD grains (Figure 2), just as they are for SD grains. This has often been assumed, but not directly demonstrated.

We have verified that thermal demagnetization tails extending much above T_B are a feature of grains larger than SD size. These tails increase in size and extent as grain size increases (Figure 1). In large grains, a clean separation of primary and secondary NRM's by thermal cleaning is difficult and paleothermometry methods predicated on SD behavior predict increasingly deviant paleotemperatures (Figure 4). A partial remedy is LTD prior to thermal cleaning or, preferably, 10-mT AF pre-cleaning, which efficiently removes remanence with anomalously high T_{UB}'s (Figure 4).

Our two new discoveries are (1) that the spectrum of anomalous T_{UB}'s extends below as well as above T_B, and (2) that the T_{UB} spectrum is symmetric, with a mean value close to T_B. Previous studies have not reported this behavior but it is of considerable practical importance. In a Thellier paleointensity experiment on PSD or MD grains, NRM demagnetization in low-T steps will greatly outweigh pTRM acquisition in companion steps because of anomalously low T_{UB}'s in thermal demagnetization. This effect persists until the NRM is about half-demagnetized and is the cause of "sagging" NRM–pTRM plots observed for PSD and MD grains [Levi, 1977; Dunlop, 1998].

The second effect, the symmetry of the T_{UB} spectrum, is potentially of great interest in paleothermometry. If we can devise a reliable method of determining $(T_{UB})_{av}$ of a thermoviscous overprint in the presence of primary NRM, the Pullaiah contours will, to a good approximation, apply to PSD and MD as well as SD grains. This would be a satisfying solution to a vexing problem.

These two effects have not been observed previously. Bol'shakov and Shcherbakova [1979] and other Russian workers used pTRM's acquired over broad T_B ranges extending down to T_0. The low-T_{UB} thermal demagnetization tail then occupies part of the range of T_B's and is not obvious. Worm et al. [1988] studied pTRM's produced over a narrower T_B range (400-350°C), but their first heating step was 380°C, and they normalized the demagnetization data to an assumed full pTRM at 350°C. Halgedahl [1993] continuously demagnetized VRM's produced at 225°C, starting from 225°C. Only thermal demagnetization beginning from room temperature will reveal low T_{UB}'s.

What causes the continuous distribution of T_{UB}'s from T_0 to T_C, with a mean around T_B (Figures 1, 2)? Why are the low- and high-T_{UB} tails of $f(T_{UB})$ selectively erased by AF cleaning or LTD (Figure 3)? The mechanism of thermal demagnetization is either (de)nucleation of domains [Halgedahl, 1991], causing discontinuous changes in internal demagnetizing field H_d that move all DW's, or small jumps, one DW at a time, driven by the continuously changing balance between $H_d(T)$ ($\propto M_s(T)$) and wall pinning ($\propto H_c(T)$) [Xu and Dunlop, 1994].

Theoretically, $H_c(T)$ should vary as $K_1(T)/M_s(T)$ (K_1 is magnetocrystalline anisotropy) if nucleation controls H_c [Goodenough, 1954] but as $\lambda(T)/M_s(T)$ (λ is magnetostriction) if pinning of DW's by defects governs H_c. The latter variation is almost always observed in heating [Heider et al., 1987; Özdemir and Dunlop, 1997]. Observations of domains in magnetite crystals at elevated T have shown on the one hand, successive nucleations at a sharp corner in heating to ≈120°C with few changes at higher T [Heider et al., 1988], but on the other, domain widths in a variety of crystals that varied very little until T ≥450°C [Ambatiello et al., 1999].

This somewhat conflicting evidence suggests that domain nucleation in magnetite is confined to two ranges, just above T_0 (where K_1 decreases rapidly with T) and between ≈450°C and T_C. If so, low-T and high-T tails in thermal demagnetization data may result from domain (de)nucleation, while more continuous thermal demagnetization at intermediate T would be due to readjustments of DW positions with few changes in the number of walls and domains. Such a picture does not explain why pre-treatment by partial AF cleaning or LTD, which should affect the least strongly pinned walls, preferentially removes VRM or pTRM with the lowest and the highest T_{UB}'s. Nucleation or denucleation events cause profound changes in domain widths and demagnetizing fields, and should move strongly as well as weakly pinned walls. Our understanding of the causes of broad and symmetric T_{UB} distributions in MD grains remains incomplete.

Many authors have stressed the importance of magnetic prehistory in MD grains, e.g., the initial state and how it was achieved. Halgedahl [1993] found that a thermally demagnetized initial state resulted in VRM's with pronounced high-T_{UB} tails whereas an AF demagnetized initial state suppressed most of the tail. Our results have the same trend but there is only a modest broadening of the T_{UB} spectrum (to low as well as high T_{UB}'s) for a thermally cleaned initial state. We also investigated the effect of a thermally cooled (TC) initial state, in which the sample is heated to T_C and cooled to T_B in zero field. Shcherbakova et al. [2000] report that this state results in a higher intensity pTRM with a larger high-T_{UB} tail than does a thermally demagnetized (or TH, for thermally heated) initial state. In our 135 μm sample, VRM intensities for the AF, TH, and TC states were 6.05, 5.57, and 9.72 mA/m, respectively, but the T_{UB} spectrum for the TC state was only slightly broader than that of the TH state. We will report details of this work elsewhere.

Conclusions

1. Thermal demagnetization of SD VRM or pTRM is sharp, with $T_{UB} = T_B$. For PSD and MD VRM or pTRM, a single T_B gives a spectrum $f(T_{UB})$.
2. The width of $f(T_{UB})$ increases as grain size increases.
3. As well as a high-T tail, complicating separation of primary and secondary NRM, $f(T_{UB})$ has a low-T tail, causing non-linear Thellier paleointensity behavior.
4. $(T_{UB})_{av} \approx T_B$ for PSD and MD as well as SD grains. The Pullaiah et al. [1975] paleothermometry method will work if $(T_{UB})_{av}$ values are used.

Acknowledgments. Our measurements were made at the Institute for Rock Magnetism, which is funded by the Keck Foundation, NSF, and University of Minnesota. We thank Mike Jackson and Jim Marvin for their help and Sue Halgedahl, Phil Schmidt and Wyn Williams for careful reviews. This research was supported by NSERC Canada through grant A7709 to DJD.

References

Ambatiello, A., K. Fabian, and V. Hoffmann, Magnetic domain structure of multidomain magnetite as a function of temperature: observation by Kerr microscopy, *Phys. Earth Planet. Inter., 112*, 55-80, 1999.

Bol'shakov, A.S., and V.V. Shcherbakova, A thermomagnetic criterion for determining the domain structure of ferromagnetics, *Izv. Phys. Solid Earth, 15*, 111-117, 1979.

Dunlop, D.J., Thermoremanent magnetization of non-uniformly magnetized grains, *J. Geophys. Res., 103*, 30561-30574, 1998.

Dunlop, D.J., and Ö. Özdemir, Thermal demagnetization of VRM and pTRM of single domain magnetite: no evidence for anomalously high unblocking temperatures, *Geophys. Res. Lett., 20*, 1939-1942, 1993.

Dunlop, D.J., Ö. Özdemir, and P.W. Schmidt, Paleomagnetism and paleothermometry of the Sydney Basin, 2. Origin of anomalously high unblocking temperatures, *J. Geophys. Res., 102*, 27285-27295, 1997.

Goodenough, J.B., A theory of domain creation and coercive force in polycrystalline ferromagnetics, *Phys. Rev., 95*, 917-932, 1954.

Halgedahl, S.L., Magnetic domain patterns observed on synthetic Ti-rich titanomagnetite as a function of temperature and in states of thermoremanent magnetization, *J. Geophys. Res., 96*, 3943-3972, 1991.

Halgedahl, S.L., Experiments to investigate the origin of anomalously elevated unblocking temperatures, *J. Geophys. Res., 98*, 22443-22460, 1993.

Heider, F., D.J. Dunlop, and N. Sugiura, Magnetic properties of hydrothermally recrystallized magnetite crystals, *Science, 236*, 1287-1290, 1987.

Heider, F., S.L. Halgedahl, and D.J. Dunlop, Temperature dependence of magnetic domains in magnetite crystals, *Geophys. Res. Lett., 15*, 499-502, 1988.

Levi, S., The effect of magnetite particle size on paleointensity determinations of the geomagnetic field, *Phys. Earth Planet. Inter., 13*, 245-259, 1977.

McClelland, E., and V.P. Shcherbakov, Metastability of domain state in multidomain magnetite: Consequences for remanence acquisition, *J. Geophys. Res., 100*, 3841-3857, 1995.

McClelland, E., A.R. Muxworthy, and R.M. Thomas, Magnetic properties of the stable fraction of remanence in large multidomain (MD) magnetite grains: single-domain or MD? *Geophys. Res. Lett., 23*, 2831-2834, 1996.

Néel, L., Théorie du traînage magnétique des ferromagnétiques en grains fins avec applications aux terres cuites, *Ann. Géophys., 5*, 99-136, 1949.

Özdemir, Ö., and D.J. Dunlop, Effect of crystal defects and internal stress on the domain structure and magnetic properties of magnetite, *J. Geophys. Res., 102*, 20211-20224, 1997.

Pullaiah, G., E. Irving, K.L. Buchan, and D.J. Dunlop, Magnetization changes caused by burial and uplift, *Earth Planet. Sci. Lett., 28*, 133-143, 1975.

Shcherbakova, V.V., Shcherbakov, V.P., and Heider, F., Properties of partial thermoremanent magnetization in PSD and MD magnetite grains, *J. Geophys. Res., 104*, 767-781, 2000.

Thellier, E., and O. Thellier, Sur l'intensité du champ magnétique terrestre dans le passé historique et géologique, *Ann. Géophys., 15*, 285-376, 1959.

Worm, H-U., M. Jackson, P. Kelso, and S.K. Banerjee, Thermal demagnetization of partial thermoremanent magnetization, *J. Geophys. Res., 93*, 12196-12204, 1988.

Xu, S., and D.J. Dunlop, Theory of partial thermoremanent magnetization in multidomain grains, 2. Effect of microcoercivity distribution and comparison with experiment, *J. Geophys. Res., 99*, 9025-9033, 1994.

David J. Dunlop and Özden Özdemir, Geophysics, Physics Department and Erindale College, University of Toronto, 3359 Mississauga Road North, Mississauga, Ontario, Canada L5L 1C6. (dunlop@physics.utoronto.ca)

(Received November 26, 1999; revised February 14, 2000; accepted March 8, 2000.)