*Geophys. J. Int.* (2009) **177,** 949–957 doi: 10.1111/j.1365-246X.2009.04152.x



# Continuous and stepwise thermal demagnetization: are they equivalent?

David J. Dunlop

*Geophysics, Physics Department, University of Toronto, Toronto,* ON M5S 1A7*, Canada. E-mail: dunlop@physics.utoronto.ca*

Accepted 2009 February 10. Received 2009 February 10; in original form 2008 December 11

## SUMMARY

Continuous thermal demagnetization, in which measurements of magnetization are made at high temperature $T$ during heating, is considerably faster than the conventional palaeomagnetic method of stepwise demagnetization, in which measurements are made at room temperature $T_0$ in a series of cooling-reheating cycles. In the case of single-domain (SD) grains, the two methods give equivalent results after the continuous measurements are converted to equivalent room-temperature values by correcting for the reversible decrease of spontaneous magnetization $M_S$ between $T_0$ and $T$. To test for equivalence of the two methods in larger pseudo-SD and multidomain grains, three different samples containing magnetite of different grain sizes and origins were heated in zero magnetic field and measurements taken either continuously at $T$ during heating or at $T_0$ after a set of cooling steps from $T$. Two samples contained 100–125 μm (mean 110 μm) and 125–150 μm (mean 135 μm) sieve fractions from a crushed natural crystal of magnetite, while the third sample is a natural diabase core sample containing coarse magnetite in the dark minerals and groundmass and fine magnetite in the plagioclase. Two different vibrating-sample magnetometers were used for continuous demagnetization and a mini-furnace and SQUID magnetometer were used for stepwise measurements. Both total thermoremanent magnetization (TRM) and partial TRMs with non-overlapping blocking temperatures ($T_C$, $T_I$) and ($T_I$, $T_0$) were demagnetized. The Thellier laws of partial TRMs are approximately although not exactly obeyed. In $M_S(T)$-corrected continuous data, there is little overlap of unblocking temperatures: pTRM ($T_C$, $T_I$) demagnetizes almost entirely above $T_I$ and pTRM ($T_I$, $T_0$) almost entirely below $T_I$, demonstrating reciprocity and independence. The stepwise measurements decrease more rapidly in intensity with increasing $T$ than the corrected continuous results, some of which increase slightly with heating up to $T \approx T_I$. Some additional decay of magnetization must occur during cooling from $T$, where the continuous measurement is made, to $T_0$, where the stepwise result is measured. There are no high-temperature measurements of subsidiary cooling-heating cycles to confirm this deduction, but continuously recorded heating-cooling cycles below room temperature in inverse thermal demagnetization (or low-temperature demagnetization) show both reversible and irreversible features, depending on the remanence tested. The most important conclusion of this study is that $M_S(T)$-corrected continuous demagnetization results do not exactly reproduce measured stepwise demagnetization results except for very fine grains, of SD size or close to it. Continuous thermal demagnetization cannot be used in general as a time-saving alternative to stepwise demagnetization if exact equivalence is required.

**Key words:** Archaeomagnetism; Palaeointensity; Rock and mineral magnetism.

## 1 INTRODUCTION

In most palaeomagnetic and palaeointensity studies, thermal demagnetization is carried out in stepwise fashion. A sample is heated in zero field in steps of increasing temperature $T$ and cooled in zero field between steps to room temperature $T_0$, where the residual magnetization vector $\boldsymbol{M}_0(T)$ is measured. More detailed thermal demagnetization results can be obtained quite rapidly by measuring $\boldsymbol{M}(T)$ continuously with a high-temperature vector magnetometer. However, $\boldsymbol{M}(T)$ includes the reversible change in spontaneous magnetization $M_S(T)$ and thus decreases more rapidly than $\boldsymbol{M}_0(T)$. The desired irreversible decrease in magnetization with heating can be obtained by dividing $\boldsymbol{M}(T)$ by normalized saturation magnetization $M_S(T)$

$$\boldsymbol{M}_0(T) = \boldsymbol{M}(T)/\boldsymbol{M}_S(T) \quad (1)$$

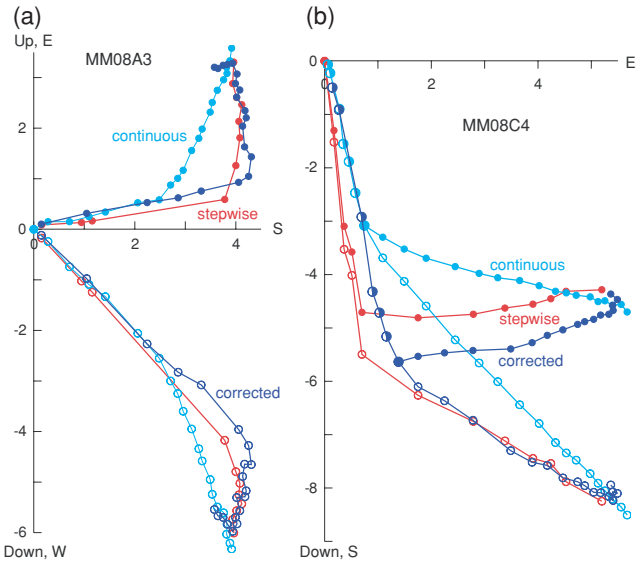


**Figure 1.** Results of stepwise and continuous thermal demagnetization of a bivectorial remanence in two samples of the Milton Monzonite, displayed on orthogonal vector diagrams (data: Schmidt & Clark 1985). Open and filled points are vertical- and horizontal-plane projections, respectively. Agreement is poor between the stepwise and untreated continuous data for the lower-$T$ partial TRM but becomes quite close when the continuous data are corrected by dividing each result by $M_S(T)$.

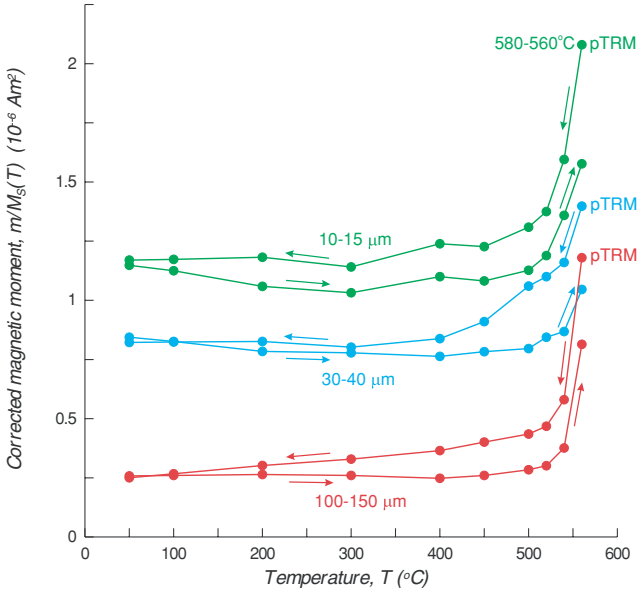


**Figure 2.** Zero-field cycling of pTRM (580 °C, 560 °C) from 560 °C → 20 °C → 560 °C for magnetite samples of various grain sizes (data: McClelland & Sugiura 1987). Moments have been corrected to equivalent room-temperature values by dividing each result by $M_S(T)$. Each pTRM decreases sharply on cooling below 560 °C but recovers part of its intensity in heating back to 560 °C.

using the measured high-field thermomagnetic curve to approximate $M_S(T)$.

Schmidt & Clark (1985) tested the equivalence of stepwise and continuous methods of thermal demagnetization in resolving multivectorial magnetizations in samples of the Milton Monzonite (SE Australia). Some of their results are redrawn in Fig. 1, with the originally separated stepwise, continuous and corrected (according to eq. 1) data combined in a single vector plot (Zijderveld 1967) for comparison. In stepwise heating, sharp junctions between low- and high-temperature data trajectories demonstrate non-overlapping unblocking temperature spectra of the two remanence components. In the continuous demagnetization trajectories, the junctions are rounded and occur much closer to the origin because of the more rapid decrease of $\boldsymbol{M}(T)$ compared to $\boldsymbol{M}_0(T)$. Linear fits to the continuous demagnetization trajectories give the correct direction for the high-temperature component, but the direction of the low-temperature component is seriously in error because of apparent overlap of the unblocking temperatures of the components.

The corrected data show that the apparent overlap of components is an artefact of making observations of magnetization that are systematically biased by $M_S(T)$. When the reduction in $\boldsymbol{M}$ due to $M_S(T)$ is corrected for, the agreement between the stepwise and corrected continuous trajectories becomes quite close. In particular, both show sharp elbows at comparable temperatures (around 460–480 °C).

Schmidt & Clark (1985) believed that their samples had multidomain character but more recent work on the Milton Monzonite (Dunlop *et al*. 1997) indicates that at most sites remanence carriers have the characteristics of single-domain (SD) or small pseudo-single-domain (PSD) grains. This view is supported by the demagnetization results of Fig. 1. More than one-third of the total remanence of sample A3 and about one-half that of sample C4 demagnetize below 480 °C. The two components are partial thermoremanent magnetizations or pTRMs produced by the same field, the samples being reoriented during cooling. Unblocking temperatures are fairly uniformly distributed down almost to $T_0$. Such an abundance of distributed low and intermediate unblocking temperatures is characteristic of SD not multidomain behaviour.

The results of Schmidt & Clark (1985) have important practical implications. Each continuous demagnetization run took about one hour at a heating rate of ≈10 °C min$^{-1}$. Although measurements of $\boldsymbol{M}$ (made with a flux-gate sensor) were only recorded at intervals of 10–50 °C, modern high-temperature magnetometers are capable of continuous tracking of $\boldsymbol{M}$ with changing $T$ (Wack & Matzka 2007; Dunlop 2008). Very detailed continuous demagnetization data can be obtained in a fraction of the time required for repeated heating-cooling-reheating cycles to a limited set of temperatures (in the case of Schmidt and Clark's experiments, 12 temperatures) in stepwise demagnetization. Measuring $M_S(T)$ with a vibrating-sample magnetometer (VSM) using chips from palaeomagnetic cores, is equally rapid. If the procedure of eq. (1) can reliably 'deconvolve' $\boldsymbol{M}(T)$ data and transform them into the corresponding $\boldsymbol{M}_0(T)$ results, a considerable saving of time and improvement in the detail of data coverage are possible.

The purpose of this paper is to test the equivalence of continuous and stepwise heating procedures in grains much larger than SD size. Although the author is not aware of previous work aimed specifically at testing this question, results by McClelland & Sugiura (1987) suggest that there could be problems. Most of their data concern changes in pTRMs during cooling after the acquisition field is suppressed but they show also continuous thermal demagnetization results for some of the pTRMs when they are subsequently reheated from $T_0$. Three of the most interesting sets of cooling + heating results are redrawn on a common scale in Fig. 2. These are renormalized $M$ values of the 580–560 °C pTRMs for three grain-size fractions of magnetite: 10–15, 30–40 and 100–150 μm. The first is near the upper limit for PSD behaviour and the other two are truly multidomain. $M$ was measured with a VSM.

The pTRM intensities for all three samples decrease by similar amounts during zero-field cooling, most strongly just below the lowest blocking temperature of 560 °C, where the field was turned off, but to some extent right down to $T_0$. The decreases are claimed not to be due to thermoviscous decay. The cause is not entirely clear but the phenomenon is well substantiated (Sugiura 1989; Muxworthy 2000).

The behaviour during zero-field heating is remarkable (Fig. 2). Instead of further decreases in remanence, the pTRM intensity rises. The heating curves tend to retrace the cooling curves, and more than one-half of the remanence lost (supposedly irreversibly) during cooling is regained by heating back to 560 °C. The 540–400 °C pTRMs (not shown) behave as expected and decrease steadily to near zero on heating. The 580–540 °C pTRMs have intermediate behaviour, decreasing very gradually with heating and remaining far from a demagnetized state at 560 °C. In one case, the 100–150 μm fraction, the intensity increases spectacularly above 500 °C to a value higher than the room-temperature pTRM.

These heating results seem to defy the physical principle that magnetization cannot increase without a field to cause that increase. Magnetization in a single-component system should always remain constant or decrease in zero field. Apparently the higher-temperature pTRMs are a dual-component system, that is, they are sums of remanences with two different sources. McClelland & Sugiura (1987) propose regions within the 'magnetite' with different Curie temperatures and therefore different $M_S(T)$ functions at high temperatures. Whatever the precise mechanism, the existence of dual sources of remanence, each with its own temperature behaviour, seems likely to compromise or destroy the equivalence of corrected continuous and stepwise thermal demagnetization data. Cooling to $T_0$ in stepwise treatment evidently causes decreases in pTRM intensity that are only partly recovered in reheating to $T$ (Fig. 2).

McClelland & Sugiura (1987) did not carry out any stepwise experiments. In this study, stepwise and continuous thermal demagnetization results for similar samples are compared and their equivalence (or non-equivalence) is assessed.

## 2 SAMPLES AND EXPERIMENTAL METHODS

This study investigates three different samples in three separate sets of experiments. Two of the samples contain annealed magnetite grains obtained as sieve fractions after crushing a large magnetite crystal. The purity of the magnetite was verified by a measured Curie point of 580 °C and a sharp Verwey transition around 115 K. The sieve fractions were 100–125 μm (nominal mean size 110 μm) and 125–150 μm (nominal mean size 135 μm). These magnetites were heated in vacuum for 2–3 hr at 650–700 °C, both to anneal out internal stress and to achieve a reproducible initial state for thermal work.

The 135 μm magnetite was dispersed in $CaF_2$ (≈1 per cent by weight) and then packed tightly with quartz wool in a 7 mm OD quartz capsule. The sample was ≈4 mm in diameter and ≈8 mm long. The capsule was sealed off after ≈1 day's pump-down to a hard vacuum. The multidomain-size magnetite grains in this sample presumably contain large numbers of easily mobilized domain walls. The sample was used in stepwise thermal demagnetization experiments.

The 110 μm magnetite was dispersed in a small amount (≈4 mm × 3 mm × 2 mm) of refractory cement on the ceramic sample holder of a high-temperature VSM (Princeton Measurements, MicroVSM). This sample of multidomain-size magnetite was used in continuous thermal demagnetization experiments. Measurements on the 110 and 135 μm magnetite samples were carried out during visits to the Institute for Rock Magnetism, University of Minnesota.

The third sample is a rock minicore, ≈10 mm in diameter × 10 mm long, drilled from a Matachewan Diabase palaeomagnetic core (site TK49, Dunlop *et al.* 2005). TK49 samples contain inclusions of SD magnetite in plagioclase phenocrysts, coarse individual magnetite crystals, and multidomain-size magnetites associated with dark ferromagnesian minerals. All are Ti-free, based on ≈575 °C Curie points in Kappabridge AC susceptibility measurements and high-field thermomagnetic curves, both done in Ar. In a palaeointensity simulation experiment (Dunlop 2008), about two-thirds of the TRM was estimated to have SD sources and one-third had multidomain sources. The sample thus has overall PSD character. This sample was used in continuous thermal demagnetization experiments at the Université de Montpellier II (France). It was initially stabilized by repeatedly being given a 50-μT TRM and thermally demagnetized.

Various pTRMs were produced in the vacuum-sealed 135 μm sample using a small non-inductively wound resistance furnace just large enough to accommodate the sample and a Pt-Rh thermocouple. The furnace was enclosed in a quartz water jacket on which a single-layer solenoid had been wound. Fields could be applied along the sample axis with this solenoid or perpendicular to the sample using miniature Helmholtz coils. The coils and furnace were located inside a six-layer cylindrical μ-metal shield, which reduced the ambient field to ≤3 nT during stepwise thermal demagnetization. Magnetization measurements were made with a 2G superconducting magnetometer.

The 110 μm sample was not vacuum sealed but was heated in a stream of flowing He to inhibit oxidation. A 200-μT field, produced by the electromagnet in the MicroVSM, was used to produce TRM and reciprocal pairs of pTRMs ($T_C$, $T_I$) and ($T_I$, $T_0$), with $T_I = 560$, 540 and 500 °C. These temperatures were chosen to match those used by McClelland & Sugiura (1987). 560 °C is the equipartition temperature for the 110 μm sample, that is, equal amounts of pTRM are produced above and below this temperature. Before starting continuous thermal demagnetization runs, the field was zeroed by initially AF demagnetizing the sample and adjusting the magnet current until the sample had zero induced moment. Slight adjustments of the null field were required above 550 °C. Measured pTRMs were recorded at 50 °C intervals up to 400 °C, at 25 °C intervals from 400 to 500 °C, and 10 °C intervals above 500 °C. Each recorded magnetic moment is the average of 10 successive VSM readings.

The TK49 minicore was heated in a vibrating thermomagnetometer of Russian design with enhanced electronics (Poidras *et al.* 2009). Heating was in Ar at a rate of ≈10 °C min$^{-1}$. TRM and pTRMs, reciprocal pairs with $T_I$ = 345C, 470, 515–520 and 550 °C, were produced by a 50-μT field. Magnetization was measured and recorded 'continuously' (at ≈8 s or 1.3 °C intervals) during cooling and zero-field reheating.

Thermomagnetic curves of all samples were measured using the MicroVSM with a 0.5 T applied field. $M_S(T)$ results are compared in Fig. 3 with the Bochirol & Pauthenet (1951) curve for magnetite. There are minor variations between samples, for example, $T_C$ of the 110 μm sample is about 5 °C lower than the other $T_C$'s, but the curves all have the same general aspect.

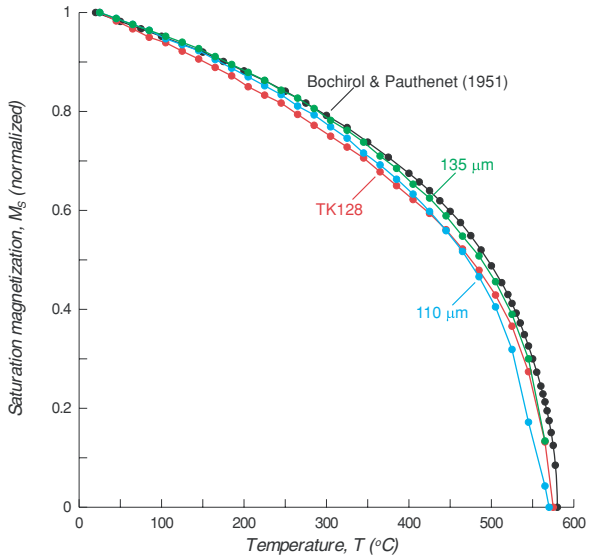


**Figure 3.** Magnetization measured in a 0.5-T field as a function of temperature $T$ for sister samples of those used in pTRM experiments. The results, all resembling the standard $M_S(T)$ curve of magnetite (Bochirol & Pauthenet 1951), were used to convert continuous thermal demagnetization data to equivalent room-temperature values.

## 3 THERMAL DEMAGNETIZATION RESULTS

### 3.1 Results for the 110 μm sample

Continuous thermal demagnetization data for the 110 μm sample appear in Fig. 4(a). Dividing each measured value by $M_S$ at the same $T$ (from Fig. 3) gives the results of Fig. 4(b), which trace the irreversible change of TRM or pTRM with $T$. The before-heating values of pTRM ($T_C$, 560 °C) and pTRM (560 °C, $T_0$) are equal: 560 °C is the equipartition temperature for pTRM acquisition. When heated to 560 °C, TRM drops to approximately one-half its room-temperature value (Fig. 4b); the decrease is much greater in Fig. 4(a) because of the rapid drop in $M_S$ above 500 °C. The equipartition would be perfect if pTRM (560 °C, $T_0$) demagnetized entirely below 560 °C and pTRM ($T_C$, 560 °C) entirely above 560 °C. This is close to being the case in Fig. 4(b). Thus Thellier's (1938) laws of pTRMs are approximately if not exactly obeyed.

Dividing by $M_S(T)$ has a great magnifying effect, particularly above 500 °C where $M_S$ drops below one-half its room-temperature value and ultimately to zero at $T_C$. Minor inflections in the measured data become spikes in the corrected data, for example, for the TRM and the ($T_C$, 500 °C) and ($T_C$, 560 °C) pTRMs. These features cannot be taken at face value; they represent amplified noise. However, certain robust trends are clear in Fig. 4(b). While pTRMs produced between $T_I$ and $T_0$ always decrease monotonically with heating, as does the total TRM, pTRMs produced between $T_C$ and $T_I$ gradually increase with heating. This is true even for pTRM ($T_C$, 540 °C), excluding the 300 °C point, and is unmistakable for the ($T_C$, 500 °C) and ($T_C$, 560 °C) pTRMs. The increases begin in some cases at quite low temperatures and tend to level out above 450 °C, apart from spikes. The huge increases above 500 °C reported by McClelland & Sugiura (1987), in some cases more than double the room-temperature pTRM value (Fig. 2, 100–150 μm), are not seen in the present results.

### 3.2 Results for the TK49 minicore

Fig. 5(a) illustrates continuous thermal demagnetization data for the TK49 minicore as well as the cooling curve with field $H$ applied from $T_C$ to $T_0$ (TRM acquisition + reversible induced magnetization). Measurement noise is more obvious than in Fig. 4(a) because magnetization is recorded quasi-continuously by the thermomagnetometer. The demagnetization data were fitted with a spline and digitized at a set of fixed $T$ before being divided by $M_S(T)$. The renormalized curves (Fig. 5b) are smooth without any high-$T$ artefacts. The ($T_C$, 550 °C) and (550 °C, $T_0$) pTRMs are nearly equal at $T_0$; pTRM equipartition occurs at a temperature slightly higher than 550 °C. TRM drops to a little over one-half its room-temperature

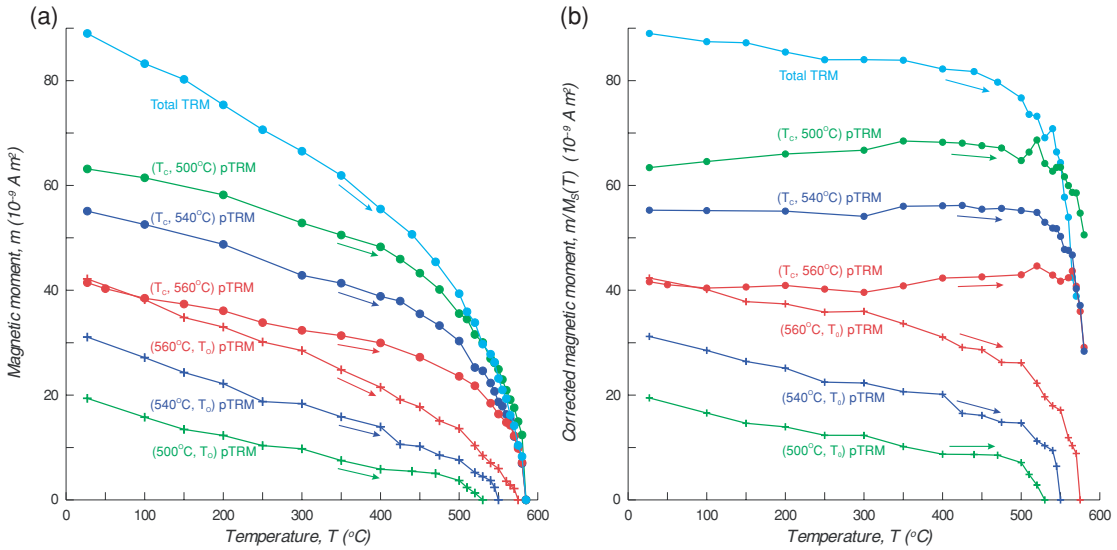


**Figure 4.** (a) Raw and (b) $M_S(T)$-corrected continuous thermal demagnetization data for TRM and various pTRMs of the 110 μm magnetite sample. Measurements are averages of 10 successive VSM readings made during brief pauses at each $T$. After correction, the TRM and lower-$T$ pTRMs decrease monotonically with heating but the higher-$T$ pTRMs are constant or increase slightly up to approximately their respective minimum blocking temperatures $T_I$. Spikes in the highest-$T$ data are artefacts generated by dividing noisy data by small values of $M_S(T)$.

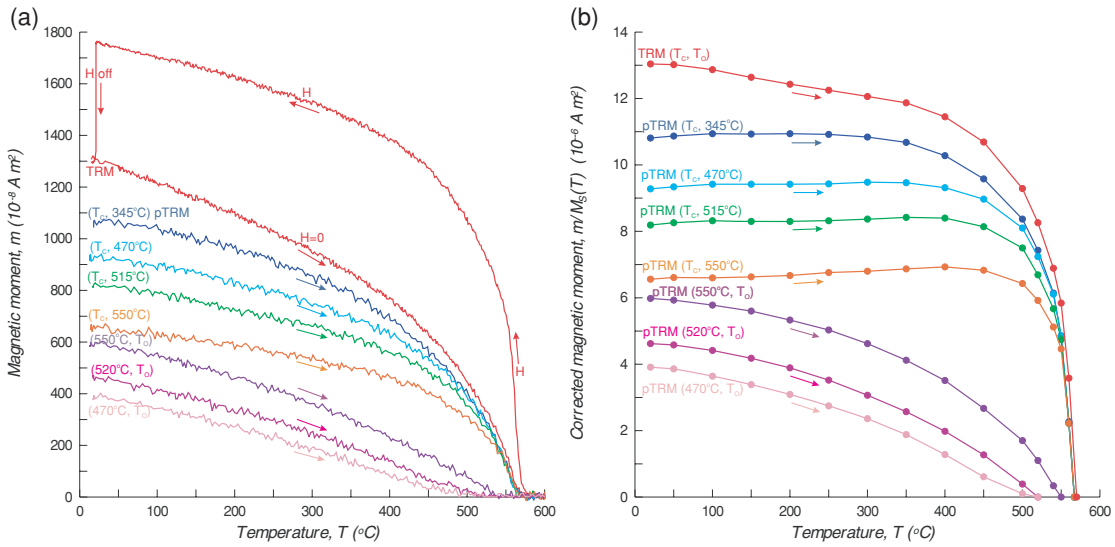


**Figure 5.** (a) Raw and (b) $M_S(T)$ corrected continuous thermal demagnetization data for TRM and various pTRMs of the TK49 minicore. Measurements were made on the fly at ≈1.3 °C intervals in continuous heating in a thermomagnetometer. After smoothing, digitization and correction of the raw data, the TRM and lower-$T$ pTRMs decrease monotonically with heating but the higher-$T$ pTRMs are constant or increase slightly up to approximately 100 °C below their respective minimum blocking temperatures $T_I$.

intensity at 550 °C. The demagnetization of each pTRM begins/ends approximately at the temperature $T_I$ where $H$ was turned off/on during cooling. TK49, like the 110 μm sample, comes close to obeying the Thellier laws in spite of a substantial population of multidomain grains.

A striking result in Fig. 5(b) is that all the high-$T$ pTRMs increase in intensity in zero-field heating, starting immediately above $T_0$ and continuing to increase up to 250 °C for pTRM ($T_C$, 345 °C), to 350 °C for pTRM ($T_C$, 470 °C), to 400 °C for pTRM ($T_C$, 515 °C) and to ≈450 °C for pTRM ($T_C$, 550 °C). This progressive growth in the uppermost temperature of increasing pTRM is compatible with the results for the 110 μm sample, although the changeover from increasing to decreasing moment is more difficult to judge in Fig. 4(b). As a rough guideline, pTRM intensity increases until ≈100 °C below the temperature $T_I$ at which $H$ was switched off during pTRM production.

These results contrast in a number of ways with those of McClelland & Sugiura (1987). First, the increase in pTRM intensity is confined to temperatures much below those involved in pTRM production. Second, the increase occurs more or less uniformly with heating, starting at $T_0$. Third, all the pTRMs ($T_C$, $T$) of both samples exhibit these increases. McClelland and Sugiura, on the other hand, observed increases in intensity during heating only for their highest-$T$ pTRM, ($T_C$, 560 °C). All other pTRMs decreased monotonically on heating, except for the 100–150 μm sample where the intensity decrease changed to an increase for the highest few temperature steps. In addition, the nature of the intensity increases was entirely different: minor or non-existent at lower $T$ with quasi-exponential increases just below 560 °C (Fig. 2).

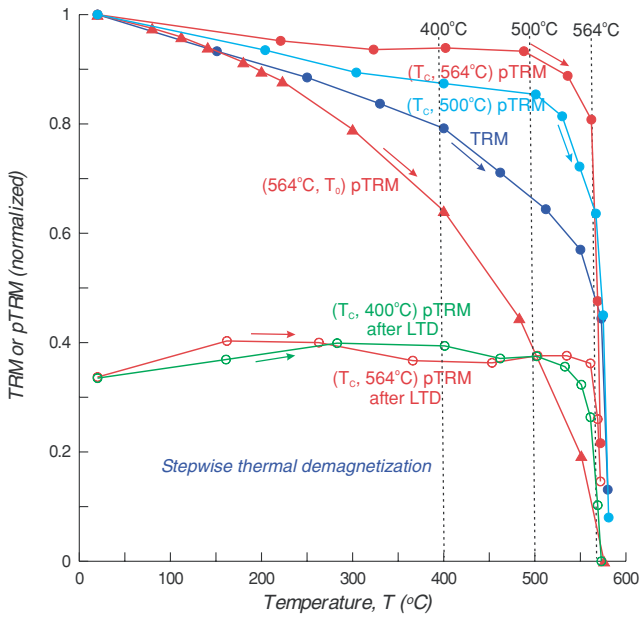


**Figure 6.** Stepwise thermal demagnetization of TRM and selected pTRMs of the 135 μm magnetite sample. All measurements were made by a SQUID magnetometer after cooling the sample to room temperature. In contrast to the replica room-temperature results represented by corrected continuous thermal data, these truly room-temperature measurements decrease monotonically with heating for all pTRMs. However, after LTD of the (580 °C, 564 °C) and (580 °C, 400 °C) pTRMs, room-temperature remanence increases with heating up to approximately 564 and 400 °C, respectively.

### 3.3 Results for the 135 μm sample

The intent of correcting continuous thermal demagnetization data (Figs 4b and 5b) is to isolate irreversible changes in pTRM intensity by removing the masking effect of $M_S(T)$ variations. The same information is given directly (although much more laboriously) by stepwise thermal demagnetization. Representative stepwise data for the 135 μm sample appear in Fig. 6. TRM and a full set of reciprocal pTRMs ($T_C$, $T_I$) and ($T_I$, $T_0$) were thermally demagnetized, as were the 'memories' of TRM and pTRMs ($T_C$, 564 °C) and ($T_C$, 400 °C) after low-temperature demagnetization (LTD). LTD consists of zero-field cycling to below the magnetite Verwey transition and back to $T_0$. It was carried out using a small liquid $N_2$ dewar lowered into three nested μ-metal shields. Because the Earth's field

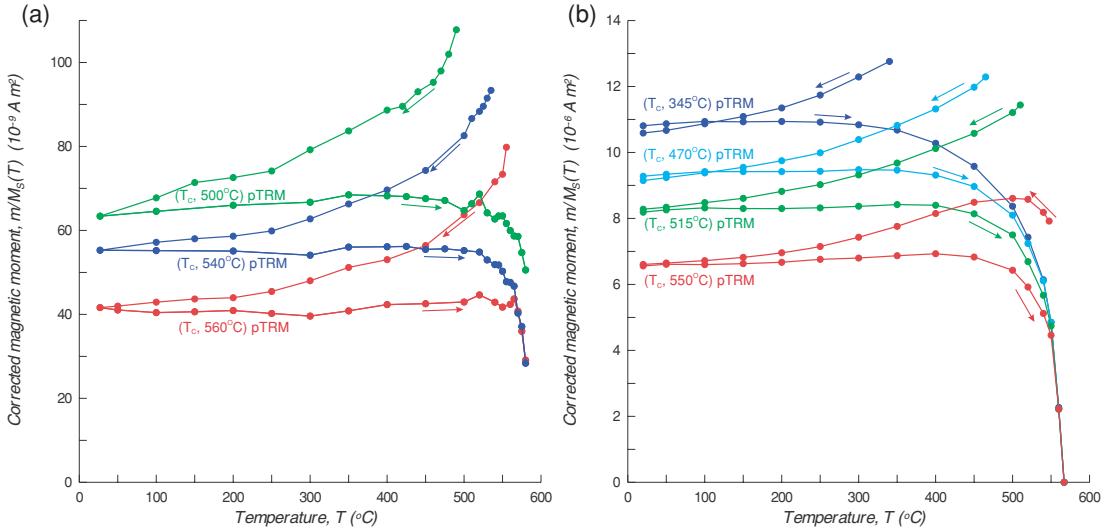


**Figure 7.** Zero-field cycling of various pTRMs in the style of Fig. 2 for (a) the 110 μm magnetite samples and (b) the TK49 minicore. Corrected to room-temperature values, pTRM intensities decrease with cooling, although not as markedly as in Fig. 2. However, on heating, the curves are almost flat and recovery of pTRM intensity is minimal, in contrast to the Fig. 2 results.

in the laboratory is mainly vertical, the zero-field environment during LTD was not as low as that during thermal demagnetization in a six-layer horizontal shield, but it was below 20 nT.

TRM and pTRM (564 °C, $T_0$) decrease monotonically with heating (Fig. 6), as do pTRMs (500 °C, $T_0$), (400 °C, $T_0$) and (300 °C, $T_0$) (not shown). The renormalized continuous thermal demagnetization curves for TRM and similar pTRMs had the same behaviour (Figs 4b and 5b). What is new is that all the high-temperature pTRMs—($T_C$, 564 °C) and ($T_C$, 500 °C), as well as ($T_C$, 400 °C) and ($T_C$, 300 °C) (not shown)—also decrease monotonically as $T$ rises, with a tendency to reach a plateau at intermediate $T$. The corresponding corrected continuous demagnetization data unmistakably increased with heating for the first 250–450 °C. In stepwise demagnetization, only the LTD memories of pTRMs ($T_C$, 564 °C) and ($T_C$, 400 °C) increase with heating.

Although it is not obvious from the normalized data in Fig. 6, 564 °C is the pTRM equipartition temperature for the 135 μm sample: pTRM ($T_C$, 564 °C) = pTRM (564 °C, $T_0$). The normalized TRM drops to almost exactly half when heated to 564 °C. In other respects, the Thellier laws are not as closely obeyed as they were in Figs 4(b) and 5(b). Although pTRM (564 °C, $T_0$) demagnetizes to within 5 per cent at 564 °C, pTRMs (500 °C, $T_0$), (400 °C, $T_0$) and (300 °C, $T_0$) have substantial demagnetization tails extending above 500, 400 and 300 °C, respectively. The high-temperature pTRMs ($T_C$, 564 °C), ($T_C$, 500 °C), ($T_C$, 400 °C) and ($T_C$, 300 °C) all demagnetize appreciably below their minimum blocking temperatures to levels ranging from 82 to 92 per cent. These are not gross violations of Thellier's laws but they are hallmarks of multidomain behaviour.

## 4 DISCUSSION

### 4.1 Partial recovery of pTRM in zero-field heating

Fig. 7 shows acquisition as well as demagnetization curves for the high-temperature pTRMs ($T_C$, 500 °C), ($T_C$, 540 °C) and ($T_C$, 560 °C) of the 110 μm sample and pTRMs ($T_C$, 345 °C), ($T_C$, 470 °C), ($T_C$, 515 °C) and ($T_C$, 550 °C) of the TK49 minicore. There are marked differences between these results and those of McClelland & Sugiura (1987), shown in Fig. 2.

Instead of demagnetizing in zero-field heating, the ($T_C$, 560 °C) pTRMs of McClelland and Sugiura's samples increase with increasing $T$, especially from 540 to 560 °C. For their 100–150 μm sample, which is comparable in grain size to two of our samples, the moment increases by a factor 3.5 between room temperature and 560 °C. Cooling and heating curves are subparallel. This is particularly noticeable for the 10–15 μm sample whose heating curve reproduces the ups and downs of the cooling curve between 200 and 540 °C. Decreases of pTRM in zero-field cooling are thus not all permanent but are partly recovered in zero-field heating. At 560 °C, where the two curves diverge the most, one-half to two-thirds of the pTRM is restored. The ($T_C$, 560 °C) pTRM must be carried by two phases having oppositely directed magnetizations with very different temperature dependences. The real loss of pTRM with cooling to $T_0$ tends to be overshadowed by reversible changes in the moments of the two phases, the balance swinging one way as $T$ decreases and back again as $T$ increases.

The results for TK49 and the 110 μm sample are entirely different from those of McClelland & Sugiura (1987, Fig. 7). The pTRMs do decrease in zero-field cooling, although less markedly than the ($T_C$, 560 °C) pTRMs in Fig. 2, but the zero-field heating curves are flat. There is little recovery of moment on heating, and the small increases that do occur are confined to lower $T$ ranges. In stepwise demagnetization there is no recovery at all (Fig. 6), but the after-LTD memories of two of the pTRMs do increase with null-field heating up to the minimum blocking temperature $T_1$. Two magnetite-like phases with distinct thermal properties were invoked by McClelland & Sugiura (1987) to explain their results. None of the present samples had a second Curie temperature, even TK49 with its three different types of magnetite (Section 2). Nevertheless two phases with opposed magnetizations seem to be indicated by the stepwise heating results following LTD.

The large decreases in moment of our pTRMs during zero-field cooling followed by only minor recovery in zero-field heating could be an reflection of irreversible domain state changes. Grains in SD

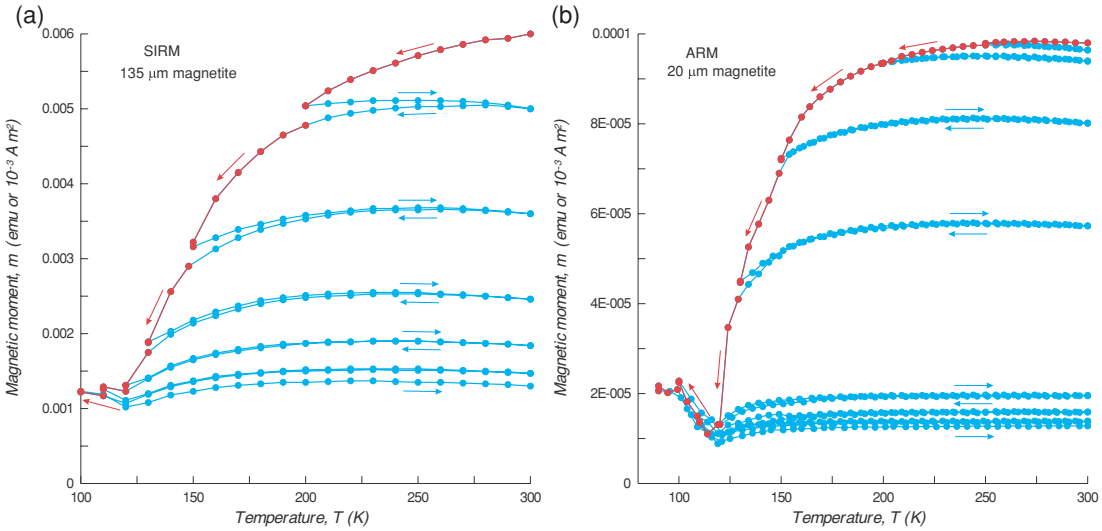


**Figure 8.** Stepwise and continuous thermal demagnetization results during zero-field low-temperature cycles. The process amounts to inverse thermal demagnetization in that (a) SIRM and (b) ARM are erased during cooling and partially recover upon heating. The main cooling curve, in red, is the analogue of the heating curves in Figs 2, 4(b), 5(b) and 7. The subsidiary heating-cooling cycles to room temperature, in blue, continuously track the process of stepwise demagnetization. This process is almost reversible in heating and cooling of ARM but somewhat irreversible for SIRM.

states near $T_C$ would have large remanence drops if they nucleated a wall or vortex during cooling, while in zero-field reheating the remanence would recover very little because any SD moments would be oriented at random. An SD → 2-domain transformation is theoretically difficult in all but ultrafine grains ($\leq$0.1 μm for magnetite) because the energy barrier between states rapidly grows larger than available thermal $kT$ energy as $T$ drops below $T_C$ (Dunlop *et al.* 1994; Muxworthy *et al.* 2003). Nevertheless, experimental nucleation of domains has been observed at much lower temperatures in a 30-μm magnetite crystal (Heider *et al.* 1988).

Could these unusual properties—pTRMs that decrease rather than staying constant during cooling below the lowest blocking temperature and then recover to varying extents on heating—be artefacts? The results shown in Figs 2, 4(b), 5(b) and 7 are not primary data but measured moments $m$ that have been divided at each temperature by $M_S(T)$. If the $M_S(T)$ values are systematically low, $m/M_S(T)$ will be systematically high, particularly above 500 °C where $M_S$ changes rapidly.

Temperature lag of the sample interior could lead to such a systematic error during heating. During cooling, on the other hand, the surface cools ahead of the sample interior and $M_S(T)$ is systematically high. The small moment increases in our samples in zero-field heating could possibly be explained in this way. However, it seems unlikely that a lag in $T$ could account for the large increases in McClelland and Sugiura's ($T_C$, 560 °C) pTRMs during zero-field heating, particularly since the changes during cooling—which cannot be explained by thermal lag—are larger still.

### 4.2 Equivalence of stepwise and continuous thermal demagnetization

We turn now to the practical question of whether continuous thermal demagnetization results, after correction, reproduce stepwise demagnetization results. For SD or small PSD magnetites, Schmidt & Clark's (1985) results (Fig. 1) suggest they do. For multidomain magnetites, all pTRMs decay monotonically in stepwise heating (Fig. 6), while in continuous thermal demagnetization, correcting measurements to equivalent room-temperature moments, high-$T$ pTRMs remain constant or increase slightly up to the minimum blocking temperature (Figs 4b, 5b and 7). The renormalized continuous demagnetization results do not reproduce the stepwise cleaning results.

We can gain some insight from the results of Figs 2 and 7. In each sample there is some irreversible loss in pTRM intensity as a result of cooling in zero field from the minimum blocking temperature $T_I$ where the field that produced the pTRM was switched off. The permanent loss is the difference between the cooling and reheating curves at any chosen $T$. In stepwise cleaning, we reheat the sample to some intermediate $T$ (not to $T_C$ as in these experiments) and cool back to $T_0$, where the moment is remeasured. There must be a further irreversible pTRM decrease in this cooling step because the remeasured moment is less than the original pTRM. Each partial cooling curve must therefore slope downward to the left (measurements reduced to $T_0$), just like the initial cooling curve.

Note that the initial decrease in pTRM intensity in cooling to $T_0$, anomalous though it appears when compared to Néel's (1949) SD blocking theory, would not itself destroy the equivalence between stepwise and thermal demagnetization. The culprit is the further irreversible decrease that apparently occurs when the pTRM is heated to an intermediate $T$ and cooled back to $T_0$. To be sure of this, we need to compare stepwise and continuous methods in a single experiment, with measurements taken throughout repeated stepwise heating-cooling cycles.

### 4.3 Analogue experiments using inverse thermal demagnetization

Dunlop (2008) reported continuously tracked measurements in Thellier cycles (zero-field heating, in-field cooling) to increasing temperatures. Similar data for zero-field cycles are lacking but Dunlop (2003) made an analogous comparison of stepwise and continuous low-temperature demagnetization (LTD) or inverse thermal demagnetization. Some examples are shown in Fig. 8. Remanence is erased by zero-field cooling from $T_0$ to $T$ and partially regained in zero-field heating from $T$ back to $T_0$. Results have not been corrected to $T_0$ values because $M_S(T)$ is almost constant below room temperature.

Programmable SQUID systems make it possible to automate these rather intricate experiments and to delineate the main cooling curve (red) and subsidiary heating-cooling cycles (blue) in great detail. The measurements in Fig. 8 were made with a Quantum Design MPMS-2 magnetometer and cryostat at the Institute for Rock Magnetism. Most high-$T$ VSMs are programmed to take measurements at fixed intervals in unidirectional heating from $T_0$ to $T_C$, and the nested heating-cooling steps needed for a direct comparison of stepwise and continuous thermal demagnetization require direct operator control. Less laborious is the variable-field translation balance (VFTB; Petersen (2009)), which can be programmed to do nested heating-cooling steps at high temperatures.

There is some irreversible decrease in saturation isothermal remanence (SIRM) as a result of the heating-cooling cycles involved in stepwise LTD although the major losses occur in the main cooling curve, the envelope of the stepwise cycles (Fig. 8a). Losses in subsidiary cycles are largest for the 200 and 150-K steps but also appreciable for the 130 and 120-K steps. In contrast, anhysteretic remanence (ARM) behaves essentially reversibly in $T \rightarrow T_0 \rightarrow T$ cycles (Fig. 8b). The blue heating and cooling curves overlap almost perfectly, except within $\approx$10 K of $T$, and the red curve is an exact replica of measurements made during a single cooling run from 300 to 90 K.

ARM is a closer analogue of TRM than SIRM but before inferring too much from these analogue thermal curves, actual pTRMs need to be low-$T$ cycled. The samples in Fig. 8 do have grain sizes similar to the samples used in thermal demagnetization experiments. Note that the continuous and stepwise inverse thermal demagnetization results are not equivalent. Over most ranges of $T$, SIRM and ARM demagnetize more rapidly when monitored continuously than after stepwise reheating to $T_0$ (compare the red points at the beginning of each subsidiary heating-cooling cycle to the blue points at $T_0$). This is opposite to the trend for thermal demagnetization, where room-temperature-equivalent magnetization drops more rapidly in stepwise than in continuous measurements (Figs 4b, 5b and 6).

## 5 CONCLUSIONS

Three different magnetites, two of truly multidomain size (110 and 135 μm) and the third (TK49) a mixture of SD and multidomain grains, come close to obeying the Thellier (1938) laws of pTRM additivity, reciprocity and independence. In demagnetization, TRM drops to about half its room-temperature intensity when heated to the equipartition temperature $T_{eq}$ for which pTRM ($T_C$, $T_{eq}$) = pTRM ($T_{eq}$, $T_0$), implying additivity. There is relatively little permanent loss of moment in demagnetization up to the minimum blocking temperature $T_I$ of pTRMs ($T_C$, $T_I$), particularly in (corrected) continuous data, while pTRMs ($T_I$, $T_0$) demagnetize almost entirely over their blocking range $T_0 \rightarrow T_I$. These observations imply that blocking and unblocking temperatures are reciprocal and that the two non-overlapping pTRMs are approximately independent.

In previously reported results (McClelland & Sugiura 1987) some high-$T$ pTRMs (corrected to room-temperature values) increased strongly during continuous thermal demagnetization. This is quite unlike SD behaviour. In the present experiments all high-$T$ pTRMs grew during zero-field heating but only by small amounts (Figs 4b) and 5b). Stepwise measurements show no intensity increase, suggesting that it might be an artefact of using sample surface temperatures in the correction procedure. Heating curves in general are very different from initial zero-field cooling curves, rather than mirroring them as in McClelland and Sugiura's results.

Stepwise and corrected continuous demagnetization data disagree significantly. Sample moment drops more rapidly with heating in stepwise than in continuous demagnetization. Continuous demagnetization is quick, because no repeated cooling cycles to room temperature are needed, and it yields detailed results. Unfortunately, for multidomain grains the corrected continuous data do not reproduce the stepwise data. This is the diametric opposite of the good agreement observed for SD grains.

Continuous tracking of pTRMs, not only during heating to $T_C$ but also throughout subsidiary cooling-heating steps, needs to be carried out as an integral experiment in order to understand the physical reason for the disagreement between continuous (main heating curve) and stepwise (subsidiary cycles) results. There must be a further decrease of moment during the cooling steps but whether this is a permanent loss or is partially recovered in the reheating half of the cycle is unknown at this point.

Some insight is given by analogue experiments in which remanence is demagnetized both continuously and in steps back to room temperature in the course of cooling below $T_0$ rather than heating above $T_0$. This inverse thermal demagnetization (usually called LTD) gives reversible cycles for ARM but significantly irreversible cycles for SIRM. However, pTRMs have so far not been tested in this way.

## ACKNOWLEDGMENTS

The TK49 experiments were done at the Université de Montpellier II, France. I thank Michel Prévot for inviting me and providing research facilities and Thierry Poidras for his experimental help. The other experiments were done at the Institute for Rock Magnetism, which is funded by NSF's Earth Sciences Division, the Keck Foundation, and the University of Minnesota. Thanks to Mike Jackson, Jim Marvin and Peat Sølheid for their help during my visits to the IRM. The ingenious mini-furnace used for pTRM production and stepwise demagnetization was designed by Özden Özdemir. Comments by the reviewers, Phil. Schmidt and John Shaw, improved the paper. This research is part of a program supported by the Natural Sciences and Engineering Research Council of Canada (grant A7709).

## REFERENCES

Bochirol, L. & Pauthenet, R., 1951. Aimantation spontanée des ferrites, *J. Phys.,* **12,** 249–251.

Dunlop, D.J., 2003. Stepwise and continuous low-temperature demagnetization, *Geophys. Res. Lett.,* **30**(11), 1582, doi:10.1029/2003GL017268.

Dunlop, D.J., 2008. Paleointensity determination using continuous thermal measurements by a high-temperature vibrating thermomagnetometer, *Geochem. Geophys. Geosyst.,* **9,** Q09Y03. doi:10.1029/2008GC002054.

Dunlop, D.J., Newell, A. J. & Enkin, R. J., 1994. Transdomain thermoremanent magnetization, *J. geophys. Res.,* **99,** 17 741–17 755.

Dunlop, D.J., Özdemir, Ö. & Schmidt, P.W., 1997. Paleomagnetism and paleothermometry of the Sydney Basin. 2. Origin of anomalously high unblocking temperatures, *J. geophys. Res.,* **102,** 27 285–27 295.

Dunlop, D.J., Zhang, B. & Özdemir, Ö., 2005. Linear and nonlinear Thellier paleointensity behavior of natural minerals, *J. geophys. Res.,* **110,** B01103. doi:10.1029/2004JB003095.

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

McClelland, E. & Sugiura, N., 1987. A kinematic model of TRM acquisition in multidomain magnetite, *Phys. Earth Planet.Inter.,* **46,** 9–23.

Muxworthy, A.R., 2000. Cooling behaviour of partial thermoremanences induced in multidomain magnetite, *Earth planet. Sci. Lett.,* **184,** 169–179.

Muxworthy, A.R., Dunlop, D.J. & Williams, W. 2003, High-temperature magnetic stability of small magnetite particles, *J. geophys. Res.,* **108(**B5), 2281. doi: 10.1029/2002JB002195.

Néel, L., 1949. Théorie du traînage magnétique des ferromagnétiques en grains fins avec applications aux terres cuites, *Ann. Géophys.,* **5,** 99–135.

Petersen, N., 2009. Petersen Instruments website: http://www.vftb.com/vftb_em.shtml

Poidras, T., Camps, P. & Nicol, P., 2009. Controlled atmosphere vibrating thermo-magnetometer ($C_{at}$VTM): a new device to optimize the absolute paleointensity determinations, *Earth planet. Space,* **61,** 101–110.

Schmidt, P.W. & Clark, D.A., 1985. Step-wise and continuous thermal demagnetization and theories of thermoremanence, *Geophys. J.R. Astron. Soc.,* **83,** 731–751.

Sugiura, N., 1989. Measurement of magnetization at high temperatures and the origin of thermoremanent magnetization: a review, *J. Geomag. Geoelectr.,* **41,** 3–17.

Thellier, E., 1938. Sur l'aimantation des terres cuites et ses applications géophysiques, *Ann. Inst. Phys. Globe Univ. Paris,* **16,** 157–302.

Wack, M. & Matzka, J., 2007. A new type of three-component spinner magnetometer to measure the remanence of rocks at elevated temperatures, *Earth planet. Space,* **59,** 853–862.

Zijderveld, J.D.A., 1967. A.C. demagnetization of rocks: analysis of results, in *Methods in Palaeomagnetism*, pp. 254–286, eds Collinson, D.W., Creer, K.M. & Runcorn, S.K., Elsevier, Amsterdam.