First-order symmetry of weak-field partial thermoremanence in multi-domain ferromagnetic grains. 1. Experimental evidence and physical implications

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Abstract

We present experimental evidence that partial thermal demagnetisation and remagnetisation treatments produce effects to the measured thermoremanent magnetisation (TRM) of an assemblage of multidomain (MD) ferromagnetic grains that are symmetrical to first order. Just as for assemblages of single domain (SD) grains, measured TRM is affected by a partial thermomagnetic treatment to an extent dependent only on the difference of the magnetic field vectors between that used to impart the TRM and that used in the partial treatment and is insensitive to the absolute field intensity. The only difference between MD and SD TRM is that the effect of a partial treatment in the former partly comprises a non-reciprocal component which cannot easily be erased. The results suggest a new paradigm for MD TRM whereby a full TRM is regarded as a background state rather than a string of partial TRMs and thermal demagnetisation treatments may be considered as overprinting the TRM rather than erasing it. The symmetrical behaviour can be explained through a simple kinematic model incorporating a temperature-dependent domain structure. This model, although greatly oversimplified, also exhibits numerous other observed traits of MD TRM behaviour including the additivity of pTRMs, the exponentially decreasing progressive effects of iterated treatments, and the concave-up Arai plots produced by a Thellier palaeointensity experiment. We argue that the conventional domain wall-pinning process is only dominant in highly stressed grains and may be responsible for a preference for MD grains to be demagnetised and consequently for a loss of symmetry which is barely discernable in this study.

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1. Introduction

Obtaining a complete model of MD TRM remains a primary research frontier in the discipline of rock magnetism. The motivation for this work is provided not only by our purely physical interest; there are also good practical reasons to develop our understanding in this field. TRM is the primary sources of stable magnetisation that igneous rocks acquire in nature. These generally contain ferromagnetic grains which are larger than the SD threshold and, consequently, a great deal of palaeomagnetic information is derived from MD (sensu lato) grains carrying TRM. This is especially true for the practise of geomagnetic palaeointensity determination which requires the remanence to be a pure TRM and for
which, MD grains are proven to behave non-ideally. A sound grasp of TRM behaviour in MD grains is therefore a prerequisite for obtaining reliable time-series of full-vector geomagnetic variation over geological time.

In 1955, Néel proposed a model for TRM acquisition in multi-domain (MD) grains which was based on the process of domain wall-pinning by imperfections in the crystal lattice [1]. This predicts MD-like behaviour in certain respects [2,3] and remains our primary working model. Nevertheless, there are also many other observed aspects of MD TRM behaviour which Néel’s *hysteretic* model cannot account for; these have led to the development of alternative physical models of MD TRM based on temperature-dependent domain structures [4,5]. A significant problem in the development of a complete model of MD TRM and a testament to its intriguing and complex nature is the fact that studies are still now revealing previously unknown properties of its behaviour (e.g. [6–8]). Considerable progress has therefore recently been made in producing non-physical models which are capable of at least encapsulating the various traits of MD TRM into self-consistent frameworks [9,10]. More recently still, a model based on non-equilibrium statistical physics has been presented and proved consistent with numerous aspects of MD TRM behaviour [11,12].

The present study is concerned with reporting and explaining an unexpected and hitherto unnoticed characteristic of TRM behaviour in assemblages of MD grains. We will firstly provide experimental evidence that, with respect to partial demagnetisation and remagnetisation treatments, MD grains behave in a far more similar fashion to SD grains than was previously believed. Following this, we will discuss the generality of the observations as well as their implications for MD TRM theory. The sister-paper to this study [13] deals entirely with absolute palaeointensity determinations, providing further tests of this study’s findings, and dealing in depth with their implications with respect to this branch of palaeomagnetism.

2. Samples, methods, and terminology employed

Three rock samples referred to as MAG (a magnetite ore), DIO (a diorite), and BAS (a basalt) were used in the experiments. Repeated heating–cooling cycles were performed in air between 700 °C and $T_r$, in order to make them resistant to heating-induced alteration.

Table 1 provides a summary of their magnetic properties and Fig. 1 shows the recorded variations of low-field susceptibility with temperature.

From these it is apparent that samples MAG and DIO contain a predominance of MD magnetite with dominant magnetocrystalline anisotropy. The BAS sample was rather more complex with multiple phases evident in the susceptibility–temperature curve and little support for the isotropic point of magnetite from LT memory. This sample contained titanomagnetite oxidised at high temperature to various degrees with the dominant phases close to pure magnetite. While the measured pTRM tail for this sample is consistent with MD grains carrying the TRM, its measured hysteresis parameters are appropriate for a pseudo-single domain assemblage. This probably indicates that a significant proportion of single domain grains coexist alongside MD grains in the BAS sample. Nevertheless, as will become apparent in the experimental results to follow, BAS does contain a sufficient MD component to allow it to be useful for this study.

The high petrographic variability between the samples implies a similar degree of heterogeneity in the properties (grain size, stress regime, quantity and properties of impurities, etc.) of the contained ferromagnetic grains; the rock magnetic measurements are consistent with this hypothesis. This itself suggests that the consistent results produced by the samples in the experiments described below are likely to be applicable to a very wide range of naturally-occurring (titanomagnetite). Nonetheless, we shall return to the issue of the generality of the observations later.

The instrument employed was a single axis vibrating thermo-magnetometer (VTM) exclusive to the Palaeomagnetic Laboratory at Université Montpellier II. It is capable of continuous measurement of the magnetic moments of 1 cm cylindrical samples heated and cooled in argon at a rate of 10 °C/min between 700 and 100 °C and in a magnetic field between −100 and +100 μT applied along one axis of the sample.

The experiments carried out for this study involved a large number of thermal treatments (demagnetisations and remagnetisations). The protocol pertaining to these treatments was kept constant in order to isolate, as far as
possible, their first-order effects and to make the treat-
ments symmetrical with respect to the applied field and
the heating/cooling rates. Each treatment entailed the
following procedure:

1. Heating from 100 °C to $T_i$ in magnetic field $H_i$ at a
rate of 10 °C/min
2. Hold at temperature $T_i$ for 10 min in magnetic field $H_i$
3. Cooling from $T_i$ to 100 °C in applied magnetic field
$H_i$ at a rate of 10 °C/min
4. Hold at 100 °C for 10 min in magnetic vacuum
5. 50 measurements of the remanence were made in a
magnetic vacuum at 100 °C over a period of ap-
proximately 3 min and the average of these was taken.

A baseline measurement temperature of 100 °C was
employed in order to allow the experiments to be
carried out at a far faster rate than if $T_i$ was used. The
amount of TRM blocking below 100 °C was observed
to be negligible so this will have had no discernable
effect on the results. A 3 min measurement period was
used to ensure that the sample’s remanence had fully
stabilised and that viscous effects were not present in
the results.

Each treatment will be referenced in this study by two
figures given in brackets; the first corresponds to $T_i$ and
the second to $H_i$. For example, treatment (400 °C; 50 μT)
would refer to a thermal remagnetisation carried out in a
field of 50 μT between 400 and 100 °C. Numerous full

Fig. 1. Magnetic susceptibility versus temperature for the three samples used in this study.
TRM treatments (700 °C; 50 μT) were made during the course of all experiments to check that the measured TRM was consistent and that alteration was not affecting the samples.

We note here that the type of treatment used as standard in this study has not received explicit attention by previous studies aiming to elucidate the properties of pTRM in MD grains. The resulting pTRM is not, strictly speaking, a pTRM$_h$ (also known as pTRM* [6,9]) since these are imparted with step 1 above carried out in zero field. However, it made sense to use this ‘new’ type of treatment because firstly, it is commonly the type already used in practical palaeointensity experiments, and secondly, because it implies a symmetry between thermal demagnetisation (when both heating and cooling takes place in an identical applied field of zero) and thermal remagnetisation treatments. This symmetry in experimental protocol may well be essential to the producing the symmetrical effects which the rest of this study is concerned with and which, we argue, simplify our understanding of the pTRM acquisition process.

Fig. 2. Roquet and Arai plots showing the experimental results of a Coe modified Thellier experiment performed on the three samples. Circles (squares) on the Roquet plots indicate the remanence measured after demagnetisation (remagnetisation) treatments while crosses indicate the demagnetisation treatments after reflection to allow comparison with the remagnetisation results. Note the nonlinear temperature scales.
3. Symmetry of thermal demagnetisation and remagnetisation curves

Our first experiment was simply a Coe-modified Thellier palaeointensity experiment [14,15] performed on the samples after they had first been given a full TRM in the laboratory through treatment ($T_c; H_0$). The experiment consisted of double-heating stages to fixed temperatures; the first of these was performed in zero field ($T_i; 0$) and the second in a field $H_a$ which was equal in intensity and direction to $H_0 (T_i; H_a)$. The temperature was incrementally increased and the double-treatment repeated. After each double-treatment, we recorded the amount of TRM remaining after the demagnetisation treatment and calculated the amount of pTRM acquired by the remagnetisation treatment from the vector difference of the remanence measured after the two treatments.

Fig. 2 displays the results as Roquet and Arai plots which are entirely in keeping with many other studies

![Graphs showing Roquet and Arai plots](image)

Fig. 3. Roquet and Arai plots showing the experimental results of isolated demagnetisation and remagnetisation experiments performed on the three samples. Circles (squares) on the Roquet plots indicate the remanence measured after demagnetisation (remagnetisation) treatments. Triangles indicate the remanence during the second remagnetisation experiment where the samples were stepwise overprinted using a field equal but antiparallel to that used to impart the initial full TRM (see text). Crosses (asterisks) indicate the demagnetisation (overprint remagnetisation) treatments after transformation to allow comparison with the remagnetisation results. Note the nonlinear temperature scales.
Eq. (1) is striking because it has no sensitivity to absolute field values. Therefore it does not discriminate between a demagnetisation and a remagnetisation treatment but ascribes changes in measured TRM as due only to differences in the applied field during the full \((T_i, T_r)\) and the partial \((T_i, T_r)\) treatments.

To test the generality of this relationship, further experiments were performed. To begin, we repeated the experiment already discussed using different absolute field values whilst retaining their relative difference of \(\Delta H\). In the new case, the full TRM was acquired in a field of \(1/2 H_0\) while the subsequent partial treatments were entirely undertaken using an equal and opposite field \((-1/2 H_0)\).

The Roquet plots in Fig. 3 show, to excellent first-order agreement, that Eq. (1) is true in this case also, specifically,

\[
\Delta \text{TRM}(T_i, T_r, 0) = -\Delta \text{TRM}(T_i, T_r, -H_0) \\
= \Delta \text{TRM}(T_i, T_r, -1/2H_0 -1/2H_0)
\]

for all values of \(T_i\).

If we assume a linear dependence of \(\Delta \text{TRM}\) on \(\Delta H\),

\[
\Delta \text{TRM}(T_i, T_r, \Delta H) = a(T_i, T_r) \cdot \Delta H
\]

then Eq. (3) may be generalised to,

\[
(1/\Delta H_1) \Delta \text{TRM}(T_i, T_r, \Delta H_1) = (1/\Delta H_2) \Delta \text{TRM}(T_i, T_r, \Delta H_2)
\]

In the specific case of \(H_0=0 \mu T\), this corresponds to a law of linearity of pTRM:

\[
p\text{TRM}(T_i, T_r, H_a) = a(T_i, T_r) \cdot H_a
\]

We tested this law for our samples by repeatedly fully demagnetising them and then imparting a pTRM using

\[
\Delta \text{TRM}(T_i, T_r, H_a), (T_i; 0), (T_{i+1}; 0), \ldots, (T_c; 0), (T_i; H_a), (T_{i+1}; H_a), \ldots, (T_c; H_a)
\]

which produced isolated demagnetisation and remagnetisation curves.

Fig. 3 shows the actual measurements which are in sharp contrast to the results of the Coe-modified Thellier experiment. The demagnetisation and remagnetisation curves shown on the Roquet plot are now first-order symmetrical and the corresponding Arai plot is linear instead of concave-up. These observations demonstrate, for the case of isolated processes, the equality:

\[
\Delta \text{TRM}(T_i, T_r, \Delta H) = -\Delta \text{TRM}(T_i, T_r, -\Delta H) \tag{1}
\]

where

\[
\Delta H = H_a - H_0 \tag{2}
\]

Both terms in Eq. (1) may be read as “the change in the measured TRM resulting from a treatment carried out in the temperature interval \((T_i, T_r)\) in an applied field \((H_a)\), which is \(\Delta H\) greater than the field \((H_0)\) used to produce the initial state of the sample”.

Fig. 4. Plots of \(\Delta \text{TRM}\) after a partial thermomagnetic treatment with field \(H_a\) applied between the temperatures 500 °C and \(T_0\) (100 °C) after the sample was first cooled from \(T_0\) to \(T_0\) in field \(H_0\). The cross on each plot indicates the maximum deviation from symmetrical behaviour (~4%) observed in independent measurements of a different sample (see Generality of the symmetry observations section in the text).
various values of $H_a$ and measuring the magnitude of the acquired pTRM. The straight lines shown in Fig. 4 demonstrate its first-order validity for low values of $H_a$.

If the general relationships (4) and (5) are also valid then the slope of the straight line should not vary when $H_a \neq 0$ $\mu$T. Fig. 4 also shows experimental results confirming that, to first order, this is the case for the three samples studied here.

The experimental results and relationships presented thus-far are astonishing in the sense that they are precisely what we would expect for samples containing only SD grains. Whilst it should be remembered that these samples (particularly BAS) are likely to contain at least some SD grains, it has already been shown by the results of the palaeointensity experiment (Fig. 2) that all three samples violate Thellier’s laws of reciprocity and independence for SD grains [17] to a significant degree. Consequently, it is beyond doubt that the MD grains within the samples are also responsible for the observed behaviour. The MD behaviour of the samples with respect to TRM acquisition will be more explicitly demonstrated in the next section where the symmetry relationship is extended to include pTRM tails.

4. Symmetry of non-reciprocal effects

Two of the most widely recognised features of TRM behaviour in MD grains are the concave-up Arai plots produced by palaeointensity experiments and the so-called ‘tail’ of pTRM first reported by Shashkanov and Metallova [18] and subject to much scrutiny since. What has not been widely recognised, or at least communicated, is that the processes responsible for these two phenomena are qualitatively symmetrical.

Consider the isolation of a high-temperature tail of a pTRM, the experimental process is: $(T_c; 0), (T_r, H_a), (T_i, 0)$.

The first steps in a simulated Coe-modified Thellier palaeointensity experiment are: $(T_c; H_a), (T_r, 0), (T_i, H_a)$ which, with respect to the field applied during each treatment, is symmetrical i.e. where the applied field was zero in the first experiment, it is now switched on and vice versa. Qualitatively at least, the results are also symmetrical. Just as the final treatment in the first experiment fails to fully reverse the effects of the second treatment and re-establish the absolute zero state (AZS), so the final treatment in the second experiment fails to fully reverse the prior partial demagnetisation treatment and re-establish the initial full TRM state.

Both the $(T_i, H_a)$ treatment in the first experiment and the $(T_i, 0)$ treatment in the second can be thought of as imparting a remanence which is partly non-reciprocal. It is this non-reciprocal component which is responsible for assemblages of MD grains violating Thellier’s laws of independence and reciprocity of pTRM. The effects of a single treatment can therefore be decomposed which, using the same nomenclature as with Eq. (1) above, leads to:

$$
\Delta \text{TRM}(T_i, T_r, H_1) = \text{RECIP}(T_i, T_r, H_1) + \text{NONRECIP}(T_i, T_r, H_1)
$$

To investigate the magnitude and longevity of the non-reciprocal components, three experiments were performed which involved the treatments given below.

Experiment 1 $(T_c; 0), (T_r, H_a), (T_i; 0), (T_{i+1}; 0), (T_{i+2}; 0), (T_{i+3}; 0)$ etc.

Experiment 2 $(T_c; H_a), (T_r; 0), (T_i; H_a), (T_{i+1}; H_a), (T_{i+2}; H_a), (T_{i+3}; H_a)$ etc.

Experiment 3 $(T_c; 1/2H_a), (T_r; -1/2H_a), (T_i; 1/2H_a), (T_{i+1}; 1/2H_a), (T_{i+2}; 1/2H_a), (T_{i+3}; 1/2H_a)$ etc.

These experiments were designed to firstly isolate the non-reciprocal component and subsequently, to progressively remove it. The results are given in Fig. 5 and demonstrate that, experimental error notwithstanding, the non-reciprocal components of the effects of all three treatments are first-order symmetrical in terms of both their magnitude and their resilience to removal by subsequent treatments made to higher temperatures.

This observation may be combined with relationships (5) and (7),

$$
(1/\Delta H_1)\text{RECIP}(T_i, T_r, H_1) = (1/\Delta H_2)\text{RECIP}(T_i, T_r, H_2)
$$

$$
(1/\Delta H_1)\text{NONRECIP}(T_i, T_r, H_1) = (1/\Delta H_2)\text{NONRECIP}(T_i, T_r, H_2)
$$

These relationships would be perfectly valid for assemblages of SD grains providing that the non-reciprocal component was taken to be zero. This implies that to first-order, and considering only the experiments performed by this study, it is only the non-zero non-reciprocal component that separates TRM behaviour in MD grains from that in SD grains.

5. Comparison with previous studies

Symmetrical behaviour of partial TRM in MD grains was demonstrated in a different context by McClelland and Sugiura [4] who observed symmetrical cooling curves of normalised remanence imparted by pTRM.
(T_{c}, T_{i}) and pTRM (T_{i}, T_{r}) over the temperature range \( T_{i} - T_{r} \). They developed a kinematic model to describe this domain reconfiguration process; we will, like them, appeal to temperature-dependent domain structure as the explanation for the symmetrical behaviour observed here.

Shcherbakov and Shcherbakova [19] constructed Arai plots from isolated measurements which were linear, similar to those produced by this study and plotted in Fig. 3. However, their Arai plots were not derived in the same manner as those in this study and simply confirmed the relationship:

\[
pTRM_b(T_i, T_r) = pTRM_a(T_i, T_i) - t[pTRM_a(T_i, T_i)]
\]

where \( pTRM_b(T_i, T_r) \) is a pTRM formed by cooling in-field in the temperature interval \( (T_i, T_r) \) after first heating from \( T_i \) to \( T_r \) in zero field; \( pTRM_a(T_i, T_i) \) is a pTRM formed by cooling in-field in the temperature interval
(\(T_i, T_t\)) after first cooling from \(T_c\) in zero field; finally \(t\) [\(\text{pTRM}_a (T_i, T_t)\)] is the high-temperature ‘tail’ of this pTRM remaining after thermal demagnetisation between \(T_i\) and \(T_t\).

They later observed that the amount of remanence removed by thermal demagnetisation of a full TRM across the interval \((T_i, T_t)\) was given by the following relationship:

\[
\text{DEMAG}(T_i, T_t) = \text{pTRM}_a(T_i, T_t) - r[\text{pTRM}_a(T_i, T_t)] + \Delta \text{pTRM}_a(T_c, T_t) \tag{11}
\]

Where \(\Delta \text{pTRM}_a (T_c, T_t)\) is the ‘low-temperature tail’ of \(\text{pTRM}_a (T_c, T_t)\) which is removed by the treatment DEMAG \((T_c, T_t)\).

Our linear Arai plots in Fig. 3 indicate the relationship

\[
\text{pTRM}_c(T_i, T_t) = \text{DEMAG}(T_i, T_t) \tag{12}
\]

where \(\text{pTRM}_c\) refers to the unique type of pTRM used in this study (where the field was applied both during heating to and cooling from \(T_i\)). This type of pTRM has been observed to be larger than a conventional \(\text{pTRM}_a\) (V. Shcherbakov, personal communication, 2005) and, through substitution of Eqs. (10) and (11) would satisfy the relationship:

\[
\text{pTRM}_c(T_i, T_t) = \text{pTRM}_b(T_i, T_t) + \Delta \text{pTRM}_a(T_c, T_t) \tag{13}
\]

Physically, this is not an unappealing result although it remains to be explicitly tested.

The approach taken by numerous studies [6,19] of considering a full TRM as a string of pTRMs with distinct blocking temperatures and high and low temperature ‘tails’ defining the unblocking temperatures is not well-suited to dealing with the results of this study. Applying this approach to the results shown in Fig. 5, experiment 1 would be discussed in terms of the imparting and subsequent removal of a high temperature pTRM tail. Similarly, experiment 2 would correspond to the removal of a low temperature pTRM tail and the subsequent replacement of this lost remanence with progressively larger pTRMs. Going on to develop equations to describe the observed symmetrical behaviour in these terms leads to extremely complex relationships which are unhelpful in forming an intuitive understanding of the behaviour.

Consequently, we suggest that in the future, the different approach used so far in this study be considered alongside the more conventional paradigm for MD TRM. The new paradigm states that a full TRM is regarded as a ‘background state’ in a similar manner to the AZS. Subsequent treatments (with a peak temperature below \(T_c\)), be they demagnetisations or remagnetisations made in any field, may then be described in terms of their effects on the total remanence of the sample (\(\Delta \text{TRM}\)) with no special consideration given to ‘blocking’ or ‘unblocking’ of pTRMs. One way in which this radically alters our perception of pTRM behaviour is that partial thermal demagnetisation treatments need no longer be regarded as ‘removing’ remanence but may just as easily be thought of as ‘overprinting’ it with a new remanence of zero magnitude (comprising its own reciprocal and non-reciprocal components).

We stress that we are not claiming this new approach to be ‘superior’ to that taken by others; in fact they are exactly equivalent. However, for the results presented here, it is more helpful in forming an understanding. There remain however, other examples of MD TRM behaviour (the symmetrical high and low temperature tails of a narrow-band pTRM [7] for example) in which the former approach provides a more intuitive basis for understanding.

6. A kinematic model of MD TRM incorporating the symmetrical behaviour

The observed first-order symmetry between isolated thermal demagnetisation and remagnetisation curves are not consistent with the 2-domain hysteretic model of MD TRM [20] which predicts the partial demagnetisation process to be more effective than the remagnetisation process in MD grains.

There is strong evidence that the domain structure of MD grains does not become fixed at high temperature as the hysteretic model predicts but rather is a function both of temperature and applied field. Observations by [21–24] suggest a picture where domain wall position and even the number of domains can change significantly well below \(T_c\) and that the entire grain reconfigures its magnetisation in response to these changes.

Models of TRM in MD grains with temperature-dependent domain structure were developed by [4,5] and achieved significant successes in describing certain aspects of MD TRM behaviour. The essence of the models is that the energy landscape of an MD grain is made up of four components, namely: the magnetostatic interaction with the external field, the magnetostatic interaction with the internal demagnetising field, the exchange and the anisotropic energies. These have different temperature dependencies and therefore any
change in temperature alters the overall magnetic structure of the grain. For a very detailed discussion, see [5]. The kinematic model of MD TRM developed by Shcherbakov et al. [5], has the capacity to explain the symmetry observations reported here as well as a host of other observed behaviour as we will now demonstrate.

We consider a large grain or assemblage of grains divided into \( N \) cells of uniform and uniaxial magnetisation. The number of cells which are magnetised in a net positive direction is \( n^+ \) and the number magnetised negatively is \( n^- \). The normalised magnetisation, \( m = M (T)/M_s(T) \) is given by,

\[
m = \frac{n^+-n^-}{N} \tag{14}
\]

Shcherbakov et al. [5] defined the rate (with temperature, not time) with which cells with magnetisation anti-parallel to the field switch to being aligned parallel as \( c^- \) and the rate at which cells makes the reverse transition is made as \( c^+ \). From this, they derived the following kinematic equation to govern thermoremanence in an MD grain:

\[
\frac{dm}{dT} = (c^+-c^-)-(c^++c^-)m \tag{15}
\]

We consider each rate as a product of a temperature-dependent and a temperature-independent function, the latter of which determines the bias placed on a cell by the magnetostatic interaction energy. This second function is the temperature-independent probability of the transition taking place,

\[
c^+ = f(T)P^+ \quad c^- = f(T)P^- \tag{16}
\]

The probabilities will be governed by the depth of the potential wells which the cells are currently trapped in and we assume that the anisotropy energy is uniform throughout the grain. The energy of a particular cell (with magnetisation \( m_i \)) is therefore only unequally influenced by its interaction with the applied external field, \( H \), and the internal demagnetising field, \( H_d \). The energies associated with these interactions are,

\[
E_h = -\mu_0 H \cdot M_s(T) \cdot m_i \quad \text{(external field interaction)} \quad (17)
\]

\[
E_d = -\mu_0/2H_d \cdot M_s(T) \cdot m_i \quad \text{(internal field interaction)} \quad (18)
\]

where \( \mu_0 \) is the permeability constant and \( N \) is the demagnetising factor. The value of \( m_i \) is limited to a single positive or negative value and the \( M_s(T) \) terms are incorporated into \( f(T) \). Consequently, it is the values of \( H \) and \( m \) that impart the bias to the rates of transition which may now be written,

\[
c^+ = f(T)(1/2 + g(H)-h(m)) \tag{19a}
\]

\[
c^- = f(T)(1/2-g(H) + h(m)) \tag{19b}
\]

where \( g \) and \( h \) are functions of \( H \) and \( m \) respectively. Subbing these into Eq. (15) produces,

\[
\frac{dm}{dT} = f(T)[2g(H)-2h(m)-m] \tag{20}
\]

For small values of \( H \) and \( m \), we assume that \( g(H) \) and \( h(m) \) are linear functions of \( H \) and \( m \) like the energies whose influence they represent. Defining \( g(H) \) as linear is a necessary and sufficient condition for producing linearity of TRM and pTRM. The kinematic equation may now be written,

\[
\frac{dm}{dT} = f(T)(uH-vm) \tag{21}
\]

where \( u \) and \( v \) are constants. It is easy to see that Eq. (21) represents a highly intuitive relationship when considering a grain as a whole (or even an assemblage of grains). It states that the grain attempts to obtain the equilibrium state whereby \( uH-vm=0 \), but that it may become trapped far from this state if the thermal energy is insufficient to allow it to reconfigure fully. Since \( dm/dt \) depends only on the degree of disequilibrium between the grain’s magnetisation and the applied field and not on the absolute value of \( H \), it will exhibit the same symmetric behaviour as reported experimentally by this study.

It is worth noting that Eq. (21) bears a strong similarity to the kinematic Eq. (21) produced by Fabian [9]. Fabian derived his relationship from first principles associated with magnetic phase theory and it is reassuring that this more general approach produces a qualitatively identical result to our own. Our \( f(T) \) is equivalent to \( \lambda(T) \) in his model which he described as a function ‘summarising the frictional effects of structural metastability.’ We will go further than he and attempt to quantitatively approximate this function in just a moment.

Despite the hugely oversimplified nature of the discrete model from which the kinematic equation was derived, it also has other impressive predictive properties for MD TRM behaviour which we will now demonstrate.
that each region is dominated by uniaxial magnetocrystalline anisotropy. This is invalid of course for magnetite which has cubic magnetocrystalline as well as magnetostrictive anisotropy; however, it will not affect the qualitative results. Since the coercivity is proportional to the magnetocrystalline constant $k$, and inversely proportional to $M_s$, and since $k$ itself is proportional to $M_s$ raised to the power of 3 we have:

$$f(T) = A' \exp(-b' \cdot M_s^2 / T)$$

Assuming that $M_s \propto (T_c - T)^{0.5}$:

$$f(T) = A' \exp(-b' \cdot (T_c - T) / T)$$

Our derived function does not have an indefinite integral with respect to $T$ and consequently we are required to approximate its value; we do this using the mid-point method and 100 rectangles.

Our master equations are therefore:

$$|H - m(T_1)| = |H - m(T_0)| \exp\left[- \int_{T_1}^{T_0} A \cdot \exp(b(T_c - T) \cdot T^{-1}) dT \right]$$

(cooling)

$$|H - m(T_1)| = |H - m(T_0)| \exp\left[- \int_{T_0}^{T_1} A' \cdot \exp(b(T_c - T) \cdot T^{-1}) dT \right]$$

(heating)

where $A' < A$, and $m(T_c)$ is fixed at zero.

We are free to choose the values of the constants in order to make the behaviour of the model as realistic as possible. So long as the values allow $m(T_1)$ to saturate close to the value of $H$ by room temperature after cooling from $T_c$, all the behavioural aspects of the model described below are fully demonstrated. Each simulated treatment produces two possible values for $m(T_1)$ but, given $H$ and $m(T_0)$, the most logical of these is always obvious.

Fig. 6 presents the results of a host of experiments simulated using the model with the parameters $A=50$, $A'=8$, and $b=10$. The unblocking temperature spectrum is not terribly realistic being somewhat sigmoidal with a sharp transition at high temperatures rather than a gradual progression becoming most extreme just below.
the Curie temperature. At low temperatures, the model also does not behave very realistically because the very small pTRMs that are acquired, say over the temperature range $T_c/2$ to $T_c$, have high temperature tails of greater than 50% of the pTRM itself which have not been observed in experiments.

These relatively minor problems are products of our choice of $f(T)$ which is clearly oversimplified. However, this choice does not affect the qualitative behaviour of the model which matches experimental observations so well. Fig. 6a and b demonstrate the successful simulation of the symmetry of reciprocal and non-reciprocal effects of treatments with constant absolute values of $\Delta H$ (Fig. 6a and b). Fig. 6c and d show examples of behaviour which Shcherbakov et al. [5] already demonstrated that the model exhibits, namely: the additivity of pTRM, and the symmetry of the decrease in $m$ below $T_i$ subsequent to the imparting of a pTRM($T_c, T_i$) with the increase of $m$ during the imparting of a pTRM($T_i, T_r$).

Inequalities between blocking and unblocking temperature arise naturally from the model. Dunlop and Özdemir [7] reported quasi-symmetrical low and high-temperature tails of narrowband, intermediate-temperature pTRMs. This behaviour is exhibited by the model providing a suitable range of temperatures is chosen for the pTRM (Fig. 6e). At lower or higher temperatures, higher or lower temperature tails (respectively) dominate but both are retained to some degree.

This simple model is also able to predict other experimentally observed aspects of MD TRM behaviour which are more complex and were not considered when the model was conceived. The Coe-modified Thellier experiment, undertaken with equal $H$ used to impart the full TRM and the pTRMs, is demonstrated to naturally produce a concave-up Arai plot in agreement with experiments (Fig. 6f). The sister-article of this study [13] will show that the model also produces results in excellent qualitative agreement with experiments for other types of protocols and with different vectors of the field used to impart the pTRM relative to that used for the full TRM.

Two recent studies [8,12] have demonstrated the progressive effects of iterated treatments of the magnetisation of MD samples. Fig. 6g shows that, in agreement with experimental observations [8], the model predicts that iterated demagnetisation treatments progressively remove remanence at an exponentially-decreasing rate. Fig. 6h shows that the model also successfully simulates the effects of iteratively producing and isolating a tail of a pTRM; the extent of the tail increases at an exponentially decreasing rate as was shown experimentally [12].

A major limitation of the model is that it implicitly assumes that the grain obtains its global energy minimum (GEM) when its net magnetisation is equal to the applied field. This is plainly untrue since many possible domain configurations with different internal energies could produce an equal net magnetisation. A corollary of this is that the model is sensitive to thermomagnetic pre-history only to the extent that it affects the value of $m(T)$. Consequently a modelled pTRM$_a$ ($T_1, T_2$) is indistinguishable from a pTRM$_b$ ($T_1, T_2$). I.E. the model wrongly predicts that the propensity of a demagnetised grain to acquire a pTRM is unchanged by cycling its temperature from $T_i$ to $T_r$ and back to $T_i$ in zero applied field. We point out however, that in agreement with experiment, the type of pTRM outlined in this study (field on during both heating from to $T_i$ to $T_r$ and cooling back to $T_i$) produces a larger effect on the simulated magnetisation than a pTRM$_b$.

A further limitation of the model used here is its lack of a probabilistic component. It has been shown that an MD grain may have entirely different domain configurations after repeated identical, thermal treatments [23]. Fabian [11] posited that a successful model of MD TRM must be based on a theory of non-equilibrium statistical physics and developed such a theory. His theory has the potential to explain and model all observed aspects of MD TRM behaviour including the symmetry reported by this study. However, at present it is difficult to constrain since the transition matrices it employs are required to be extremely complex and are beyond the capabilities of our current micromagnetic models to obtain.

7. Generality of the symmetry observations

Experimental evidence suggests that the symmetry of partial thermoremanence in MD grains is not perfect. Thus far, in describing the symmetry as first-order, we have conceded this but have not yet dealt with the deviations.

Both the empirical relationships found by this study and the kinematic model predict that the measured remanence of a sample already containing a full TRM imparted in field $H$, and subsequently heated to a temperature $< T_c$ and cooled in the same field, $H$, should remain unchanged. This was tested using our BAS sample and found to be the case to within a 2% margin.

For the other samples, careful examination of Fig. 4 demonstrates that $\Delta$TRM is not in fact precisely zero when $\Delta H = 0$ and $H \neq 0$. Further experimental data was provided by V.P. Shcherbakov (personal communication, April, 2005) using the stabilised igneous rock sample referred to as 12b in previous studies [6,19]. He observed that the measured remanence decreased by
between 1% and 4% upon heating to various temperatures between 300 and 550 °C. We plot the maximum value of these reductions (obtained when \( T = 400 \) °C) as crosses on Fig. 4 for comparison with our data. For our samples BAS and DIO, this maximum value of 4% does not constitute a noticeable deviation from the straight lines used to infer that the symmetry relationship is first-order correct. In the case of sample MAG, it does fall significantly away from the trend although in this case the offset is strongly exaggerated by the small scale of the \( y \)-axis in this figure resulting from the very hard blocking temperature spectrum of this particular sample. Overall, it appears that the observations made independently by V.P. Shcherbakov are not inconsistent with the data presented here. Consequently, the symmetry observations that form the core of the present study constitute only a first-order phenomenon for MD grains within natural rock samples.

Far more difficult to reconcile with this study are the results shown in Fig. 2 and Table 2 of a study by McClelland et al. [25]. Performing the same experiment as described above, they observed maximum reductions in measured remanence ranging in value from 11% to 131% of the full TRM (the latter implying the acquisition of a self-reversed pTRM). These results are certainly not consistent with the notion of symmetry of partial theromremanence as even a first-order phenomenon. However, McClelland et al. conclude that their samples (crushed and sieved magnetite) ‘is likely to have a much greater proportion of crystal defects and imperfections than … uncrushed natural material.’ The difference in the stress regimes of the samples used by them and ourselves could be the explanation for the contradictory results. There is some evidence that the high stress regime (or some other factor) anomalously affected other results produced in that particular study. Shcherbakov et al. [6] demonstrated, using both natural and synthetic samples, that the ratio of pTRM\(_b\) \((T_1, T_2)\) to pTRM\(_a\) \((T_1, T_2)\) is less than 1 for MD samples and that it decreases as the MD content increases. Table 1 of [25] indicates that their samples were rather haphazard in this respect; two of them produced ratios close to 1, another produced a ratio greater than 1 and no relationship with mean grain size was apparent.

On the basis of the above argument, we tentatively suggest that symmetry is likely to dominate the behaviour of MD grains that are not subject to internal stress beyond that likely to occur commonly in nature. Of course this requires explicitly testing as soon as possible. By the same token, we acknowledge that second-order asymmetry in terms of a slight tendency for MD grains to adopt lower net magnetisation, is probably a general feature of MD TRM behaviour. The source of this behaviour will be discussed in the next section.

8. Discussion

It is an extremely engaging fact that so many of the diverse aspects of MD TRM behaviour can be at once described by the simple kinematic relationship given in Eq. (21) which was itself derived from the hugely simplified discrete model developed in Shcherbakov et al. [5]. This fact suggests that MD TRM is, for the samples considered, predominantly controlled by a simple governing process. The key element of this process is that changes in temperature, even at low absolute temperatures, produce spontaneous changes in the magnetisation distribution of the grain which may take the form either of domain wall motion or nucleation. This domain reorganisation allows the grain to progress towards an equilibrium state whereby the magnetostatic energy resulting from the interaction of its magnetisation with both the external applied field and the internal demagnetising field is minimised.

The model is grossly oversimplified in that it is spatially homogenous — it assumes that positively and negatively magnetised cells are randomly distributed across the grain, and that conversions between the states may also take place with equal probability anywhere in the grain. In reality, the energy cost of producing a domain wall to separate regions of different magnetisations is very high. Consequently, identical cells will be clustered together (in domains) and the conversions will only take place close to extant domain walls or at nucleation sites. The model exhibits realistic behaviour regardless of this serious shortcoming probably because the simplifications it employs are true in a statistical sense. In this case, the temperature-independent probability of the magnetisation flipping in any one cell should be regarded as the probability of this event occurring providing that an extant domain wall can accommodate it.

The symmetric behaviour observed by this study appears in the model as a natural consequence of the linear dependence of the temperature-independent probability of cell-magnetisation reversal on both external and internal field intensity. However, the model does not incorporate the second-order asymmetric behaviour discussed in the previous section. An obvious explanation for the asymmetric behaviour is that, upon cooling from \( T_c \) in an applied field \( H \), the grain becomes trapped in a state of magnetisation that has a higher value of \( m \) than that of the equilibrium state (for \( H \)) at lower temperatures. That is, the domain walls become “pinned” by rising energy barriers. If the grain is then heated again
to some intermediate temperature, and even if \( H \) is kept constant, thermal fluctuations will allow it to reconfigure towards its equilibrium state of lower magnetisation. Internal stresses contribute to the microcoercivity distribution making it possible for a grain to become trapped further from its equilibrium state subsequent to cooling from \( T_c \) in an applied field. By this reasoning, we would expect highly-stressed grains to display asymmetric behaviour to a greater extent than low-stress grains. This is consistent with the assumedly highly-stressed nature of the samples used by McClelland et al. which behaved in a highly asymmetric manner.

The kinematic model in its present simplified form cannot incorporate the behaviour just described. During cooling from \( T_c \) in \( H \), the grain quickly adopts its global energy minimum (GEM) state and maintains it all the way down to \( T_r \). Since the value of \( m \) begins at zero (at \( T_c \)) and cannot overshoot the optimum value of \( H \), a local energy minimum (LEM) cannot result in greater magnetisation than that produced by the GEM and therefore the magnetisation cannot decrease upon further heating in field. Both this problem and the more general one of a GEM not being defined solely by the value of \( m \) could be overcome with a more sophisticated kinematic model based on the rate of change of magnetostatic energy with temperature and incorporating the different energy terms directly.

9. Conclusions

The experimental results produced by this study were derived using just three natural samples (although of diverse origins). The generality of the conclusions listed below are therefore uncertain. However, our results do not appear to contradict any of those produced by previous studies which used natural or low-stress synthetic samples. The sister-article to this study [13] makes predictions of the results of various types of Thellier experiments using models incorporating symmetric behaviour. In qualitative terms, these results are highly-consistent with their published experimental counterparts produced using numerous different samples to those in this study. This increases our confidence in the generality of the findings reported here.

1. To first-order and in the weak-field regime, the effects of isolated demagnetisation and remagnetisation treatments (the latter with the field kept on throughout) on the measured TRM of natural samples containing MD grains are symmetrical. Just as for SD grains, the effects are linearly-related only to the difference in applied field vectors between that used to produce the background state (full TRM or AZS) and that used in the partial treatment and are insensitive to the absolute values of the field.

2. All partial treatments produce effects that consist of a reciprocal and a non-reciprocal (high temperature tail) component. In practical terms, a demagnetisation treatment may be thought of as producing a ‘zero-field pTRM’ which overprints the TRM.

3. The observed symmetry is inconsistent with models of MD TRM based on the wall-pinning mechanism alone (e.g. [1,20]). However, a simple kinematic relationship based on a discrete model incorporating temperature-dependent domain structure [5] is proposed. This has the capacity to explain these results as well as a host of other observed traits of MD TRM (including additivity of pTRM, symmetrical unblocking tails of narrow-band pTRM, and exponentially-decreasing progressive effects of iterative treatments).

4. The first-order symmetry may result from the linear dependence of the magnetostatic energy, at the sub-grain level, on both the intensity of the external field and the net magnetisation of the entire grain.

5. Second-order asymmetrical behaviour, whereby a grain containing a TRM preferentially reduces it magnetisation upon heating, is barely discernable in the samples used here but may, nonetheless, be a general feature of MD TRM. This could well be caused by thermal fluctuations unpinning walls which were previously trapped beyond the GEM position for the given temperature and applied field intensity. Since increased internal stress increases the probability of this wall-pinning process occurring, we would expect high-stress samples to exhibit a greater degree of asymmetry.

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