Partial anhysteretic remanent magnetization in magnetite

2. Reciprocity

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[1] One necessary condition for successful determination of relative paleomagnetic field intensity using anhysteretic remanent magnetization (ARM) methods is reciprocity: a partial ARM, produced by a steady field \( H \) applied over a narrow interval \((\bar{H}_2, \bar{H}_1)\) of alternating field (AF), must demagnetize over the same interval \((H_2, H_1)\). Experimentally, we find that partial ARMs of single-domain (SD) and pseudosingle-domain (PSD) grains demagnetize mainly between \( H_2 \) and \( H_1 \), whereas >50% of partial ARMs of large PSD and multidomain (MD) grains are erased below \( H_1 \), giving a low-field tail in the coercivity distribution. Natural pumices, granites, and oceanic basalts violated reciprocity, but lake sediments, gabbros, andesite, and red scoria had relatively small low-coercivity tails and are better candidates for paleointensity work. Using total ARM to simulate natural remanence, we carried out pseudo-Thellier paleointensity determinations for coarse PSD and MD grains. ARM demagnetization outweighed partial ARM acquisition at the same AF step, resulting in convex-down curves of ARM remaining versus partial ARM gained (pseudo-Arai plot). Pseudo-Arai plots predicted from experimentally determined distributions of blocking and unblocking fields agreed well with measured pseudo-Thellier results, in particular explaining convex-down MD curves.

1. Introduction

[2] Thellier’s laws of additivity, reciprocity and independance of partial thermoremanent magnetization (pTRM) are valid for single-domain (SD) grains and form the basis of Thellier-type methods of paleointensity determination [Thellier, 1938; Thellier and Thellier, 1959]. The reciprocity law states that pTRM produced by applying a field \( H \) between \( T_2 \) and \( T_1 \) (\( T_2 > T_1 \)) during cooling from the Curie temperature and zero field in all other temperature intervals is thermally demagnetized during zero-field heating over precisely the temperature interval (\( T_2, T_1 \)). In other words, blocking temperature equals unblocking temperature. Any violation of the reciprocity law will result in failure of paleointensity determination.

[1] In paper 1 [Yu et al., 2002], we test a law analogous to the Thellier additivity law, using partial anhysteretic remanent magnetization (pARM) instead of pTRM and alternating field (AF) demagnetization instead of thermal demagnetization. The present paper tests the analog of the Thellier reciprocity law: pARM produced by a steady field \( H \) applied only in the AF interval \((\bar{H}_2, \bar{H}_1)\) must be AF demagnetized over precisely the interval \((H_2, H_1)\). In other words, blocking field equals unblocking field.

2. Samples and Experiments

[4] The 10 synthetic and 18 natural magnetite- or titanomagnetite-bearing samples described in Tables 1-3 of paper 1 were again used. Magnetic grains of particular coercivities were isolated as narrowband pARMs produced over narrow intervals of AF using a Molspin demagnetizer. The standard initial state was AF demagnetized in a peak AF of 100 mT. The sequence of steps in experiments testing the reciprocity of pARM was as follows (Figure 1).

1. First, pARM\textsuperscript{m} \((\bar{H}_b, H_a)\) was produced by a steady field \( H \) applied over the AF interval from \( H_b \) to \( H_a \); from \( H_a \) to 0 mT, there was zero added field.

2. AF demagnetization was performed in either 2.5 or 5 mT steps. Double demagnetization [Tauxe et al., 1995] was carried out along orthogonal axes \((x, y, z)\), followed by \((-x, -y, -z)\), with remanence being measured after each set of three demagnetizations and then vectorially averaged.

3. Next, pARM\textsuperscript{m} \((H_b, H_a)\) was produced as in step 1 but using a higher AF interval \((H_b, H_a)\).

4. Double AF demagnetization of pARM\textsuperscript{m} \((\bar{H}_b, \bar{H}_a)\).

[5] The intervals \((H_a, H_b)\) and \((\bar{H}_a, \bar{H}_b)\) were determined from prior AF demagnetization of total ARM. They corre-
spond to 30–40% and 60–70% destruction of ARM. Thus pARM<sup>m</sup> and pARM<sup>n</sup> represent soft and hard coercivity fractions. Initial remanence in the AF demagnetized state, where significant, was vector subtracted from all pARMs. The steady field $H$ of 50–100 $\mu$T was applied along the cylindrical $(z)$ axis of the specimen.

3. Results

3.1. Synthetic Samples

[6] In AF demagnetization of samples 1–6, which contain SD or pseudosingle-domain (PSD) grains, the remanence dropped quite sharply over the pARM blocking range (dashed lines; Figures 2a and 2b), particularly for the lower coercivity pARM. Coercivity spectra $\Delta(M/M_0)/\Delta H$ (Figure 2c) show that >80% of pARM<sup>m</sup> of SD sample 1 (0.065 $\mu$m) demagnetized over the AF blocking interval 20–25 mT. The results for pARM<sup>n</sup> (50, 45 mT) are similar but more remanence unblocks below the lower blocking field of 45 mT (Figures 2b and 2d).

[7] The pARM of multidomain (MD) magnetite samples behaved quite differently (Figure 3). Although no remanence unblocking occurred above the upper blocking field (15 or 40 mT in Figures 3a and 3b), 65–80% of the pARM

![Figure 1. Schematic diagrams representing pARM acquisition: pARM<sup>m</sup> ($H<sub>b</sub>$, $H<sub>a</sub>$) and pARM<sup>n</sup> ($H<sub>d</sub>$, $H<sub>c</sub>$) were produced starting from peak AFs of $H<sub>b</sub>$ and $H<sub>d</sub>$ with the added steady field $H$ on from $H<sub>b</sub>$ to $H<sub>a</sub>$ and from $H<sub>d</sub>$ to $H<sub>c</sub>$, respectively.](image)

![Figure 2. (a–b) Results of AF demagnetization of pARM<sup>m</sup> (25 mT, 20 mT) and of pARM<sup>n</sup> (50 mT, 45 mT) for synthetic SD and PSD magnetites. (c–d) Corresponding coercivity spectra of samples 1 and 6.](image)
demagnetized below the lower blocking field (10 or 35 mT). The coercivity spectra actually peak below the blocking range (Figures 3c and 3d).

3.2. Natural Samples

Representative experimental data are shown in Figure 4 for natural samples 578 A (sediments from Lake Pepin, Minnesota [Brachfeld and Banerjee, 2000]), Km 3 (red scoria, Mount Aso, Japan [Yu, 1998]), and T 19 (Tudor Gabbro, Ontario [Yu and Dunlop, 2001]) (Figure 4). These samples have SD or PSD hysteresis properties (Table 3 and Figure 3 of paper 1). Although more of the pARM unblocked below the AF blocking range than in the synthetic SD and small PSD samples (Figure 2), the coercivity spectra peak within the blocking field interval (Figures 4c and 4d).

Samples Bu 5 and 8 (Burchell Lake Granite, Ontario [Dunlop, 1984]) and S 50 (Shelley Lake Granite, Ontario [Dunlop et al., 1984]) have MD hysteresis properties (Table 3 and Figure 3 of paper 1). Their pARM unblocking properties are also consistent with MD behavior. 70–90% of pARM and pARM unblocked below the blocking ranges of 15–20 and 20–25 mT (Figures 5a and 5b). The coercivity spectra have major low-coercivity tails and peak below the blocking field interval (Figures 5c and 5d).

4. Effect of Magnetic Prehistory on pARM

Different initial states are known to affect the thermal demagnetization of pTRM and viscous remanent magnetization of MD and coarse PSD grains [Vinogradov and Markov, 1989; Halgedahl, 1993; Shcherbakova et al., 2000; Dunlop and Özdener, 2000, 2001]. We tested whether the same is true for the AF demagnetization of pARMs. We produced pARM*(Hd, Hc) in an AF decaying from 100 mT to Hd with zero added field, from Hd to Hc with an added steady field H, and from Hc to 0 with zero added field (Figure 6). The essential difference between the two remanences is that the upper field Hd of the pARM, at which H was switched on, was approached from below in the case of pARM but from above for pARM* (compare Figures 1 and 6).

Intensities of pARM* and pARM for selected samples are listed in Table 1. Relative intensities are plotted in Figure 7. For SD and PSD samples 1 (0.065 μm), 4 (0.24 μm), 5 (0.34 μm), 6 (1.06 μm), 456 A, 578 A, T 19, and C 12 (Cordova Gabbro, Ontario [Yu and Dunlop, 2002]), the intensities of pARM* and pARM are almost identical.
However, for large PSD and MD samples 9 (16.9 μm), 10 (18.3 μm), Bu 5, Bu 8, and S 50, pARM\textsuperscript{n} is 6–14% more intense than pARM\textsuperscript{n}.

[12] A comparison of pARM\textsuperscript{*} and pARM intensities in eight different field intervals covering the entire 0–40 mT coercivity range appears in Table 2. The pARM\textsuperscript{*} > pARM in all field intervals but the difference is most accentuated for the lower fields. Despite this bias, the normalized AF demagnetization curves of pARM\textsuperscript{*} and pARM are almost indistinguishable (Figure 3).

[13] In the case of MD pTRMs, it has been observed that a thermally cooled (TC) pTRM (heated to T\textsubscript{c} and cooled to the upper blocking temperature in zero field, before switching on \(H\); the analog of pARM\textsuperscript{n\*}) has higher intensity than a thermally heated (TH) pTRM (thermally demagnetized and then heated to the upper blocking temperature in zero field, before applying \(H\); the analog of pARM\textsuperscript{n}). Our results are consistent with this pattern. According to Shcherbakova et al. [2000], the difference between TC and TH pTRM intensities quantitatively matches the TC thermal demagnetization tail extending above the upper blocking temperature to the magnetite Curie point (TH pTRMs had only minor tails). We have not observed any high-coercivity tail in AF demagnetization of either pARM\textsuperscript{n} or pARM\textsuperscript{n\*}, implying a fundamental difference between pARM and pTRM phenomena, most likely different MD magnetic microstates or domain configurations.

5. Discussion

5.1. Reciprocity of pARMs

[14] The AF demagnetization behavior of narrow-band pARMs for synthetic samples is grain-size dependent (Figures 2 and 3). The pARMs of SD and small PSD size samples demagnetize mainly over the AF range in which pARM was originally blocked. They thus obey the pARM reciprocity law: blocking and unblocking are equivalent processes. On the other hand, pARMs of MD size grains demagnetize readily below the lower blocking field. Their coercivity spectra have large low-coercivity tails and actually peak below the lower pARM blocking field (Figure 3).

[15] The lake sediments and gabbros used in this study had intermediate AF behavior, with about 50% of pARM demagnetizing over the field blocking range (Figure 4). They nevertheless have previously yielded successful paleointensity results by the pseudo-Thellier and Thellier meth-
ods, respectively [Brachfeld and Banerjee, 2000; Yu and Dunlop, 2001, 2002]. This success is rather surprising in view of their ambivalent behavior in pARM reciprocity experiments.

5.2. Implications for Paleointensity Determination

It is interesting to compare the AF demagnetization of narrowband pARM to thermal demagnetization of narrowband pTRM. Whereas MD grains always have high-unblocking temperature tails [e.g., Bolshakov and Shcherbakova, 1979], MD and SD samples alike have no high-coercivity tails during AF demagnetization of pARM (Figures 2–5). The pARM ($H_2, H_1$) produced by a steady field $H$ between $H_2$ and $H_1$ always demagnetizes totally in an AF of $H_2$. The lack of a high-coercivity tail in pARM does not necessarily imply superiority of pARM methods of paleointensity determination, however. Low-temperature tails cause large thermal demagnetization of MD and PSD pTRM below the blocking temperature range and this in turn results in non-linear paleointensity behavior [Dunlop and Özdemir, 2000, 2001]. Low-coercivity pARM tails, which are prominent in our large PSD and MD samples (Figures 3 and 5) can be expected to have the same effect in pseudo-Thellier paleointensity experiments.

![Figure 5](image-url). (a–b) The results of stepwise AF demagnetizations of Bu 5 and 8 (Burchell Lake Granite, Ontario [Dunlop, 1984]) and S 50 (Shelley Lake Granite, Ontario [Dunlop et al., 1984]). (c–d) Corresponding coercivity spectra of S 50 and Bu 5.

![Figure 6](image-url). Schematic diagrams representing pARM$^{**}$($H_d, H_c$) acquisition: pARM$^{**}$ was produced starting from a peak AF of 100 mT, with the added steady field $H$ on from $H_d$ to $H_c$. 
To test this anticipated effect, we performed simulated pseudo-Thellier paleointensity determinations [Tauxe et al., 1995] on our synthetic samples. A total ARM (simulating natural remanent magnetization) was progressively AF demagnetized and its remanence loss partially replaced in pairs of companion steps at increasing AF levels. A plot of ARM remaining versus pARM gained, the analog of the Arai plot [Nagata et al., 1963], shows an interesting grain-size dependence (Figure 8). Sample 1 (0.065 mm) comes closest to matching the ideal SD line. The MD sample (18.3 mm) has the largest deviations from this ideal line. Its strongly convex downward curve indicates that pARM lost in lower coercivity AF steps is larger than pARM gained in the same AF steps. For the PSD samples (0.24 and 1.06 mm), the downward curvature is less but still appreciable.

Similar but more pronounced downward curvature has been observed in Arai plots for simulated Thellier experiments on PSD/MD magnetites [Levi, 1977; Dunlop and Özdemir, 1997, 2001]. In practical applications, the pseudo-Thellier method therefore offers certain advantages. The sagging of the pseudo-Arai plot is less severe and would lead to a smaller error if paleointensity was estimated by fitting a line to the first few data points. More importantly, only a tail in the coercivity distribution can cause convex-down curves in the pseudo-Thellier method, whereas corresponding curvature in Thellier heating results could well be caused by chemical alteration of the magnetic minerals rather than low- or high-temperature pTRM tails.

6. Phenomenological Modeling

Traditionally, magnetic properties of isothermal processes such as AF demagnetization or ARM acquisition have been analyzed using the Preisach diagram [Preisach, 1935; Rimbert, 1959; Dunlop and West, 1969]. Although Preisach diagrams provide both qualitative and quantitative information, their interpretation can be difficult or inconclusive [Dunlop et al., 1990]. Instead we will adapt a recent phenomenological model used successfully to interpret pTRM behavior [Fabian, 2000, 2001] to analyze pARM reciprocity.

Our phenomenological model of pARMS represents the magnetic behavior of a sample by a large number of independent loops, each defined by two coercivities or blocking fields, for pARM blocking ($H_{B}$) and unblocking ($H_c$).
The number of loops in a particular area of the $H_{UB}$ vs. $H_B$ diagram is given by the density function, $c(H_{UB}, H_B)$. Two possible forms of $c$ are illustrated in Figure 9. For ideal SD grains, $c$ is confined to the diagonal, i.e., $H_B = H_{UB}$ (Figure 9a). PSD and MD grains have coercivity distributions extending up to but not beyond the AF at which steady field $H$ is switched on in pARM acquisition (Figure 9b). A distinctive feature of pARM modeling compared to pTRM modeling [Fabian, 2000, Figure 1] is that the distribution of $\chi$ (represented as shaded region) has been truncated along the diagonal, where $H_B, H_{UB}$, since there is no high-coercivity pARM tail (Figures 2–5). In pTRMs, both low-temperature ($T_{UB} \leq T_B$) and high-temperature ($T_{UB} \geq T_B$) tails are observed, where $T_B$ is the blocking (unblocking) temperature [Dunlop and Özdemir, 2001]. The distribution of $H_{UB}$ for the soft coercivity fraction, pARM, is approximately Gaussian, while the hard coercivity fraction, pARM, has a skewed $H_{UB}$ distribution (Figures 3 and 9b).

6.1. Three-Dimensional Mapping of $\chi^*$ and $\chi(H_B, H_{UB})$

In order to construct a three-dimensional (3-D) map of $\chi(H_B, H_{UB})$, experimental steps 1 and 2 (section 2) were carried out in successive stages, starting from low AFs. First, pARM ($5 \text{ mT, 0 mT}$) was produced, and then double AF demagnetization was performed to $5 \text{ mT}$ in $2.5 \text{ mT}$ steps. Next, pARM ($10 \text{ mT, 5 mT}$) was produced and AF demagnetized to $10 \text{ mT}$. Similar pARMS were generated, incrementing the upper and lower limits of $H_B$ by $5 \text{ mT}$ each time, and then AF demagnetized from $0 \text{ mT}$ to the upper $H_B$. No pARMS with $H_B \geq 40 \text{ mT}$ were simulated because pARM ($100 \text{ mT, 40 mT}$) is <2% of total ARM intensity for the MD samples. Constructing a 3-D map of $\chi^*(H_B, H_{UB})$ was carried out using exactly the same experimental sequence as for $\chi$, but with pARM*s rather than pARMS.

![Figure 9. Schematic sketches of the distribution of $H_{UB}$ versus $H_B$. (a) Ideal SD grains have identical $H_{UB}$ and $H_B$. (b) For PSD and MD grains, $H_{UB}$ is distributed up to but not beyond the diagonal line $H_{UB} = H_B$. The shaded areas depict possible $H_{UB}$ distributions.](image1)

![Figure 10. A three-dimensional view of the distribution $\chi(H_B, H_{UB})$ for an MD sample (18.3 $\mu$m magnetite).](image2)
AF demagnetization follows pARM production in the ing down to zero, e.g., 35–0 mT. Another difference is that HB between experimental procedures followed in determining incremental: each one is produced by applying the pseudo-Thellier experiment are cumulative: each is narrow AF range, e.g., 35–30 mT. The pARMs gained in because the increments of 18.3 μm MD magnetite sample.

6.2. Prediction of Pseudo-Arai Plots

[22] The experimental spectrum \( \chi(\bar{H}_B, \bar{H}_{UB}) \) for the 18.3 μm MD sample, normalized to total ARM intensity, is illustrated in Figure 10. The \((\bar{H}_B, \bar{H}_{UB})\) blocks are elongated because the increments of \( H_B \) were 5 mT but \( H_{UB} \) steps were 2.5 mT. Figure 10 demonstrates that for MD grains, the peak of \( \chi \) for each \( H_{UB} \) lies well below the matching value of \( H_B \). The distribution \( \chi^*(\bar{H}_B, \bar{H}_{UB}) \) is almost identical to \( \chi(\bar{H}_B, \bar{H}_{UB}) \), except that the intensity of each block is higher.

6.3. Additivity of pARMS

[27] Another feature of our phenomenological model is that it accounts for the additivity of pARMS, as illustrated in Figure 12. The shaded areas represent the coercivity fractions that acquire pARMS between AF limits \((H_2, H_1), (H_3, H_2)\) and \((H_3, H_1)\). The first two areas sum to give the third area, whatever the details of the coercivity distribution. In other words, the density of points \( \chi \) is immaterial to the validity of the pARM addition law.

[28] However, this picture is only partially valid in the case of MD magnetite, for which pARM < pARM* throughout the coercivity spectrum. By analogy with the law of additivity of partial TRMs, which is conventionally tested using pTRMs with a 1C initial state, our experimental test of partial ARM additivity in paper 1 used pARM* (Figure 6), not pARMS (Figure 1). As a result, the partial ARM additivity “law” in general is

\[
\text{ARM} = \sum p\text{ARM} = \sum p\text{ARM} (\text{SD and PSD, Table 1})
\]

\[
\text{ARM} = \sum p\text{ARM} > \sum p\text{ARM} (MD, 1 \text{ and } 2).
\]

Although the partial ARMs in a pseudo-Thellier experiment are pARMS rather than pARM*, the practical impact of

Figure 11. A comparison between the measured pseudo-Thellier results and the predicted pseudo-Arai plot for the 18.3 μm MD magnetite sample.

Figure 12. Graphical interpretation of the additivity of pARMS using the phenomenological model of Figure 9.
nonadditivity in the MD case is likely to be minimal because reliable paleointensities are invariably determined using SD or PSD magnetites.

7. Conclusions

1. Narrowband pARMS of SD and PSD grains were largely demagnetized over the same AF interval in which they were produced, i.e., unblocking field equals blocking field. In MD samples, coercivities were widely distributed below the blocking field range.

2. The low-coercivity tail of the pARM coercivity distribution of MD and large PSD grains causes nonlinear behavior in pseudo-Thellier paleointensity determination. The ARM loss always outweighs the pARM gained in matching AF steps, yielding a curved pseudo-Arai plot of ARM remaining versus pARM gained.

3. No high-coercivity tail has been observed during AF demagnetization of pARMS.

4. The intensity and AF demagnetization of pARM of MD grains depends on the initial state. However, pARM and pARM* have very similar normalized AF demagnetization curves.

5. The pseudo-Arai plot predicted from the experimentally determined distribution of unblocking versus blocking coercivities agreed well with the measured pseudo-Thellier simulation results, in particular providing a quantitative explanation for the convex-down form of the pseudo-Arai plots of MD samples.

6. The same model and distribution of blocking and unblocking fields explain the universal validity of the law of additivity of pARMS, observed in paper 1.

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