

## EARTH SCIENCES

# Intrinsic paleointensity bias and the long-term history of the geodynamo

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Many geodynamo models predict an inverse relationship between geomagnetic reversal frequency and field strength. However, most of the absolute paleointensity data, obtained predominantly by the Thellier method from bulk volcanic rocks, fail to confirm this relationship. Although low paleointensities are commonly observed during periods of high reversal rate (notably, in the late Jurassic), higher than present-day intensity values are rare during periods of no or few reversals (superchrons). We have identified a fundamental mechanism that results in a pervasive and previously unrecognized low-field bias that affects most paleointensity data in the global database. Our results provide an explanation for the discordance between the experimental data and numerical models, and lend additional support to an inverse relationship between the reversal rate and field strength as a fundamental property of the geodynamo. We demonstrate that the accuracy of future paleointensity analyses can be improved by integration of the Thellier protocol with low-temperature demagnetizations.

## INTRODUCTION

The history of Earth's magnetic field, including geomagnetic polarity reversals, secular variation, and field strength variation, is crucial for understanding the geodynamo and planetary evolution. Although the geodynamo could have started as early as during the Hadean (1), there has been controversy regarding whether long-term variations of geomagnetic polarity reversals, secular variation, and field strength are intrinsically related or whether they have varied independently throughout geological history. In particular, although a large class of geodynamo models consistently predicts an inverse relationship between geomagnetic reversal frequency and paleointensity (2, 3), to date, most of the paleointensity data—dominated by results from bulk volcanic rocks—fail to confirm this relationship (4). Low paleointensity values (40 to 60% of the present-day intensity) are universally recorded during periods of high reversal rate (notably, a portion of the late Jurassic) (5–7). However, high values that markedly exceed the present-day intensity are rare during periods with no or few reversals (most notably, the Cretaceous Normal Polarity Superchron) (4, 8). This discrepancy suggests either that the numerical models are not accurately describing geodynamo behavior or that there is a pervasive and unrecognized bias in the experimental data that accounts for the absence of high paleointensity values.

The vast majority of absolute paleointensity data have been obtained using the double-heating Thellier method, in which the natural remanent magnetization (NRM) of a sample is gradually replaced with a thermal remanent magnetization (TRM) induced in a known magnetic field in a series of paired heating steps (9, 10). The data points corresponding to each temperature step are evaluated using an NRM-remaining versus a partial TRM (pTRM)-gained plot [Arai plot (11)], and the paleointensity is calculated from the slope of a linear segment, corresponding to the unblocking temperature range of the primary remanent magnetization. A fundamental requirement of the Thellier method is that the paleointensity signal is carried by single-domain (SD) magnetic grains that obey the Thellier laws of reciprocity, inde-

pendence, and additivity (12). For these ideal grains, the Arai plot is perfectly linear and yields an accurate estimate of the paleofield strength (13). However, rocks used in paleointensity analyses practically always contain larger magnetic grains in the pseudo-SD (PSD) and/or multidomain (MD) states for which the Thellier laws do not hold (14). Thellier experiments on these rocks typically result in nonlinear Arai plots (15, 16). However, notwithstanding this limitation, most published syntheses of geodynamo and Earth's interior processes treat paleointensity data from these nonideal carriers as representative of the true paleofield strength [for example, see the study of Biggin *et al.* (17)].

Here, we scrutinize the fidelity of rocks containing nonideal magnetic grains as paleointensity recorders by presenting a systematic paleointensity and rock magnetic investigation of synthetic magnetite-bearing samples with a broad range of grain sizes that represent the SD, PSD, and MD magnetic domain states. Paleointensity experiments were conducted using two methods: the double-heating Thellier method, as modified by Coe (10) and Coe *et al.* (18) (hereafter referred to as the conventional Thellier method), and the low-temperature demagnetization Thellier (LTD-Thellier) method (see Materials and Methods) (19).

## RESULTS

### Rock magnetism

Rock magnetic and paleointensity experiments were conducted on 24 synthetic samples grouped into eight series of three samples each. Samples of the M0.35, M0.75, M1.5, M5, M12, M30, and M250 series contained nearly stoichiometric magnetite powders with grain size modes of 0.35 (acicular), 0.75, 1.5, 5, 12, 30, and 250  $\mu\text{m}$ , respectively (see Materials and Methods). Samples of the MIX series contained a mixture of the  $\sim 250\text{-}\mu\text{m}$  magnetite (70%) with the acicular magnetite (30%).

Temperature dependences of low-field magnetic susceptibility [ $\kappa(T)$ ] measured for each series reveal the presence of a single magnetic phase with a Curie temperature of  $\sim 585^\circ$  to  $590^\circ\text{C}$ , which is consistent with nearly pure magnetite (fig. S1). A characteristic peak observed at  $\sim 153^\circ\text{C}$  ( $\sim 120\text{ K}$ ), associated with the Verwey transition (20), indicates that the magnetite is nearly stoichiometric (fig. S1). All  $\kappa(T)$  curves are reversible, which confirms the thermal stability of our samples. In addition, after completion of paleointensity experiments,  $\kappa(T)$  curves

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were measured from one of the used samples. All measured curves are similar to those obtained from fresh specimens, which indicates that no magneto-mineralogical alteration occurred during the Thellier experiments.

Magnetic hysteresis loops measured for each series have normal shapes (fig. S2, A to C). The M0.35 samples plot close to the SD region of the Day plot (fig. S2D) (21). Although acicular magnetite grains in these samples are larger than the SD threshold for equidimensional magnetite [0.05 to 0.06  $\mu\text{m}$  (22)], their nearly SD state is defined by the dominant role of shape anisotropy in the elongate particles (23). The M0.75, M1.5, M5, M12, M30, and MIX series have PSD characteristics, plotting along SD-MD mixing lines (24) toward the MD region of the Day plot with increasing grain size (fig. S2D). The M250 series has typical MD characteristics (fig. S2D and Table 1). No anisotropy of magnetic hysteresis properties was observed.

In addition, one specimen from each series was placed in a ceramic nonmagnetic boat and heated together with the samples used for paleointensity determinations during conventional Thellier and LTD-Thellier experiments. Magnetic hysteresis parameters for these specimens after completion of paleointensity experiments were indistinguishable from those measured initially, which further attests to the thermal stability of our samples.

No anisotropy of TRM was observed for any series. The TRMs imparted in three orthogonal directions were indistinguishable within experimental errors.

### Paleointensity experiments

Before paleointensity experiments were conducted, we modeled the NRM in each sample with a full TRM imparted by cooling from 700°C (in nitrogen) in a 50- $\mu\text{T}$  magnetic field ( $H_{\text{NRM}}$ ). All measured samples yielded interpretable data in both conventional Thellier and LTD-Thellier experiments. For all samples, the full TRM imparted after the final in-field heating step (595°C) was within 3% of the initially imparted TRM, which further attests to their thermal stability. Paleointensity values were calculated by fitting both high-temperature (typically, 525° to 585°C) and low-temperature (100° to 450°C) segments on the Arai plot (Table 1 and tables S1 and S2). Unless stated otherwise, paleointensity values cited below were obtained by fitting high-temperature Arai segments.

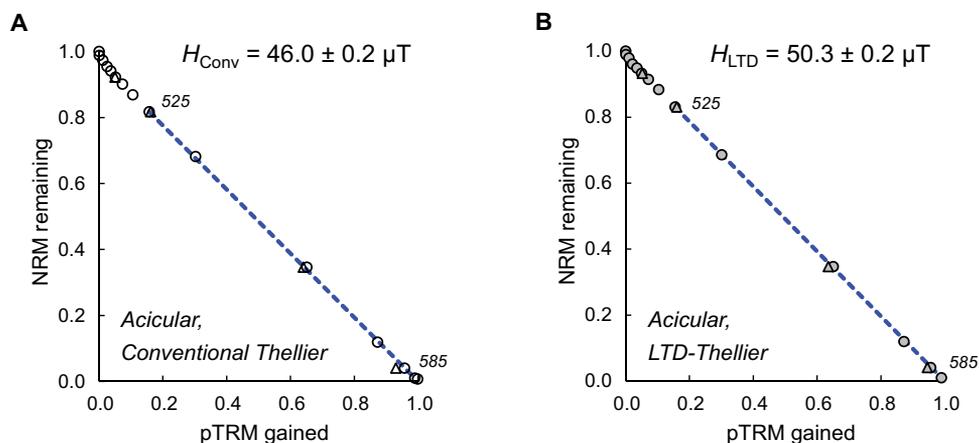
Both conventional Thellier and LTD-Thellier experiments on the M0.35 samples resulted in almost ideal, linear Arai plots (Fig. 1), consistent with their nearly SD magnetic state. Slight deviation from linearity at low temperatures most likely reflects the presence of a small amount of equidimensional magnetite in PSD grains. The series-mean paleointensity values are statistically indistinguishable from one another and from  $H_{\text{NRM}}$  (Table 1).

For all other sample series, the conventional Thellier experiments resulted in concave-up (double-slope) Arai plots, where curvature increased with magnetic grain size (Fig. 2). Mean paleointensity values underestimated  $H_{\text{NRM}}$  by ~8 to 58% (Table 1). The MIX samples yielded Arai plots similar to those obtained from the M1.5 and M0.75 samples (Fig. 3), with the mean paleointensity underestimating  $H_{\text{NRM}}$  by ~10%. For all series, mean paleointensity values calculated from low-temperature Arai segments significantly overestimated  $H_{\text{NRM}}$  by 16 to 280%, with a larger within-series scatter (Table 1).

In LTD-Thellier paleointensity experiments yielded less curved Arai plots compared to their conventional Thellier counterparts for all sample series except for M250. However, the linearization effect strongly depended on magnetic grain size. The M0.75 and M1.5

**Table 1. Summary of paleointensity results obtained using the conventional Thellier and LTD-Thellier methods.**  $M_{rs}/M_s$  is the ratio of saturation remanence to saturation magnetization. "HT fit" and "LT fit" denote the paleofield values ( $H$ ) calculated by fitting high- and low-temperature segments of the Arai plot, respectively (see text).  $\Delta H$  is the standard deviation.

| Sample     | $M_{rs}/M_s$ | Conventional Thellier              |                                    | LTD-Thellier                       |                                    |
|------------|--------------|------------------------------------|------------------------------------|------------------------------------|------------------------------------|
|            |              | HT fit                             | LT fit                             | HT fit                             | LT fit                             |
|            |              | $H \pm \Delta H$ ( $\mu\text{T}$ ) |
| M0.35-1    | 0.375        | 48.8 $\pm$ 0.3                     | 61.1 $\pm$ 2.2                     | 49.3 $\pm$ 0.3                     | 55.2 $\pm$ 2.4                     |
| M0.35-2    | 0.384        | 49.0 $\pm$ 0.3                     | 56.1 $\pm$ 3.5                     | 49.7 $\pm$ 0.2                     | 53.0 $\pm$ 1.9                     |
| M0.35-3    | 0.367        | 49.7 $\pm$ 0.1                     | 56.7 $\pm$ 5.3                     | 50.1 $\pm$ 0.1                     | 50.2 $\pm$ 0.1                     |
| SD mean    | 0.375        | 49.2 $\pm$ 0.5                     | 58.0 $\pm$ 2.7                     | 49.7 $\pm$ 0.4                     | 52.8 $\pm$ 2.5                     |
| M0.75-1    | 0.192        | 46.0 $\pm$ 0.2                     | 87.5 $\pm$ 3.3                     | 50.3 $\pm$ 0.2                     | 57.9 $\pm$ 2.9                     |
| M0.75-2    | 0.193        | 45.0 $\pm$ 0.2                     | 91.2 $\pm$ 4.8                     | 49.9 $\pm$ 0.2                     | 51.5 $\pm$ 3.6                     |
| M0.75-3    | 0.182        | 46.4 $\pm$ 1.3                     | 70.2 $\pm$ 10.4                    | 49.5 $\pm$ 0.6                     | 59.4 $\pm$ 5.8                     |
| M0.75 mean | 0.189        | 45.8 $\pm$ 0.7                     | 83.0 $\pm$ 11.2                    | 49.9 $\pm$ 0.4                     | 56.3 $\pm$ 4.2                     |
| M1.5-1     | 0.151        | 44.0 $\pm$ 0.1                     | 107.3 $\pm$ 11.1                   | 49.1 $\pm$ 0.1                     | 54.6 $\pm$ 3.8                     |
| M1.5-2     | 0.149        | 44.1 $\pm$ 0.4                     | 99.3 $\pm$ 6.8                     | 49.5 $\pm$ 0.6                     | 52.8 $\pm$ 5.2                     |
| M1.5-3     | 0.139        | 43.4 $\pm$ 0.9                     | 105.7 $\pm$ 11.5                   | 48.6 $\pm$ 0.4                     | 61.1 $\pm$ 3.6                     |
| M1.5 mean  | 0.146        | 43.8 $\pm$ 0.4                     | 104.1 $\pm$ 4.2                    | 49.1 $\pm$ 0.5                     | 56.2 $\pm$ 4.4                     |
| M5-1       | 0.105        | 40.5 $\pm$ 0.8                     | 117.1 $\pm$ 7.5                    | 48.3 $\pm$ 0.3                     | 62.9 $\pm$ 2.3                     |
| M5-2       | 0.113        | 39.8 $\pm$ 1.1                     | 121.7 $\pm$ 8.2                    | 47.0 $\pm$ 0.5                     | 75.9 $\pm$ 8.0                     |
| M5-3       | 0.110        | 38.9 $\pm$ 1.3                     | 143.0 $\pm$ 11.5                   | 47.8 $\pm$ 0.4                     | 68.9 $\pm$ 6.1                     |
| M5 mean    | 0.109        | 39.7 $\pm$ 0.8                     | 127.3 $\pm$ 13.8                   | 47.7 $\pm$ 0.7                     | 69.2 $\pm$ 6.5                     |
| M12-1      | 0.074        | 35.2 $\pm$ 1.5                     | 145.2 $\pm$ 14.2                   | 45.2 $\pm$ 0.8                     | 80.9 $\pm$ 12.7                    |
| M12-2      | 0.076        | 35.8 $\pm$ 0.6                     | 104.7 $\pm$ 11.1                   | 44.5 $\pm$ 0.9                     | 82.2 $\pm$ 6.0                     |
| M12-3      | 0.077        | 36.6 $\pm$ 1.7                     | 137.7 $\pm$ 13.6                   | 45.0 $\pm$ 0.4                     | 73.8 $\pm$ 3.0                     |
| M12 mean   | 0.076        | 35.8 $\pm$ 0.7                     | 129.2 $\pm$ 21.6                   | 44.9 $\pm$ 0.4                     | 79.0 $\pm$ 4.5                     |
| M30-1      | 0.048        | 28.2 $\pm$ 1.2                     | 116.3 $\pm$ 12.2                   | 34.3 $\pm$ 1.6                     | 136.3 $\pm$ 5.3                    |
| M30-2      | 0.046        | 28.5 $\pm$ 2.0                     | 139.3 $\pm$ 8.2                    | 33.7 $\pm$ 1.8                     | 122.5 $\pm$ 3.6                    |
| M30-3      | 0.043        | 29.4 $\pm$ 1.9                     | 132.3 $\pm$ 7.6                    | 35.0 $\pm$ 1.6                     | 110.4 $\pm$ 3.0                    |
| M30 mean   | 0.046        | 28.7 $\pm$ 0.6                     | 129.3 $\pm$ 11.8                   | 34.3 $\pm$ 0.7                     | 123.1 $\pm$ 13.0                   |
| M250-1     | 0.024        | 20.7 $\pm$ 1.6                     | 184.7 $\pm$ 15.6                   | 21.9 $\pm$ 1.3                     | 157.5 $\pm$ 12.6                   |
| M250-2     | 0.025        | 21.0 $\pm$ 1.8                     | 180.7 $\pm$ 15.4                   | 22.1 $\pm$ 1.4                     | 159.5 $\pm$ 10.6                   |
| M250-3     | 0.024        | 21.2 $\pm$ 2.2                     | 206.9 $\pm$ 8.8                    | 22.8 $\pm$ 2.0                     | 173.4 $\pm$ 6.4                    |
| M250 mean  | 0.024        | 21.0 $\pm$ 0.3                     | 190.7 $\pm$ 14.1                   | 22.3 $\pm$ 0.5                     | 163.5 $\pm$ 8.7                    |
| MIX-1      | 0.131        | 43.1 $\pm$ 1.4                     | 104.4 $\pm$ 6.2                    | 47.8 $\pm$ 0.6                     | 72.7 $\pm$ 3.9                     |
| MIX-2      | 0.125        | 44.7 $\pm$ 1.0                     | 92.6 $\pm$ 6.2                     | 48.3 $\pm$ 0.4                     | 66.2 $\pm$ 1.9                     |
| MIX-3      | 0.135        | 45.3 $\pm$ 0.9                     | 87.6 $\pm$ 5.6                     | 48.7 $\pm$ 0.3                     | 103.3 $\pm$ 5.1                    |
| MIX mean   | 0.130        | 44.4 $\pm$ 1.1                     | 94.8 $\pm$ 8.6                     | 48.3 $\pm$ 0.4                     | 80.7 $\pm$ 19.8                    |



**Fig. 1. Examples of paleointensity determinations from an M0.35 sample containing acicular (SD) magnetite.** Data are obtained using the conventional Thellier (A) and LTD-Thellier (B) methods. NRM remaining versus pTRM gained (circles) is shown. Triangles are pTRM checks. The blue dashed line is the best-fit line for a high-temperature Arai segment used to calculate paleointensity estimates  $H_{Conv}$  (A) and  $H_{LTD}$  (B) (the numbers in italics indicate the temperature range, in degrees Celsius). The best-fit lines for low-temperature Arai segments are not shown for clarity.

samples yielded nearly linear Arai plots (Fig. 2, A and B) and mean paleointensity values that accurately reproduce  $H_{NRM}$  (Table 1). The M5 samples resulted in slightly curved Arai plots (Fig. 2C), with mean paleointensity within 5% of  $H_{NRM}$ . We note that Xu and Dunlop (16) did not observe improved linearity of LTD-Thellier Arai plots for a sample containing  $\sim 6\text{-}\mu\text{m}$  magnetite grains. Although the reason for this discrepancy remains unclear, we speculate that it may reflect inefficient LTD in the previous study due to sample oxidation. The M12 samples gave rise to double-sloped Arai plots (Fig. 2D), with mean paleointensity within  $\sim 10\%$  of  $H_{NRM}$ . Similarly, the MIX series yielded a mean paleointensity that underestimates  $H_{NRM}$  by 3 to 4% (Fig. 3 and Table 1).

The linearization effect of the LTD-Thellier method was much less expressed for the M30 series (Fig. 2E), which produced a mean paleointensity value that significantly (by 31%) underestimates  $H_{NRM}$ , albeit to a lesser extent than the conventional Thellier method (Table 1). For the M250 series, the LTD-Thellier method did not result in significant improvement of the Arai plot linearity compared with conventional Thellier data (Fig. 2F). The corresponding mean paleointensity underestimated the true field by  $\sim 55\%$  (Table 1). These results are consistent with behavior observed for MD magnetite in previous studies (13, 16).

For all measured samples, the low-temperature segments of LTD-Thellier Arai plots are, on average, shallower than their conventional counterparts (Figs. 2 and 3). Nevertheless, corresponding mean paleointensity values overestimated  $H_{NRM}$  by 6 to 226% (Table 1).

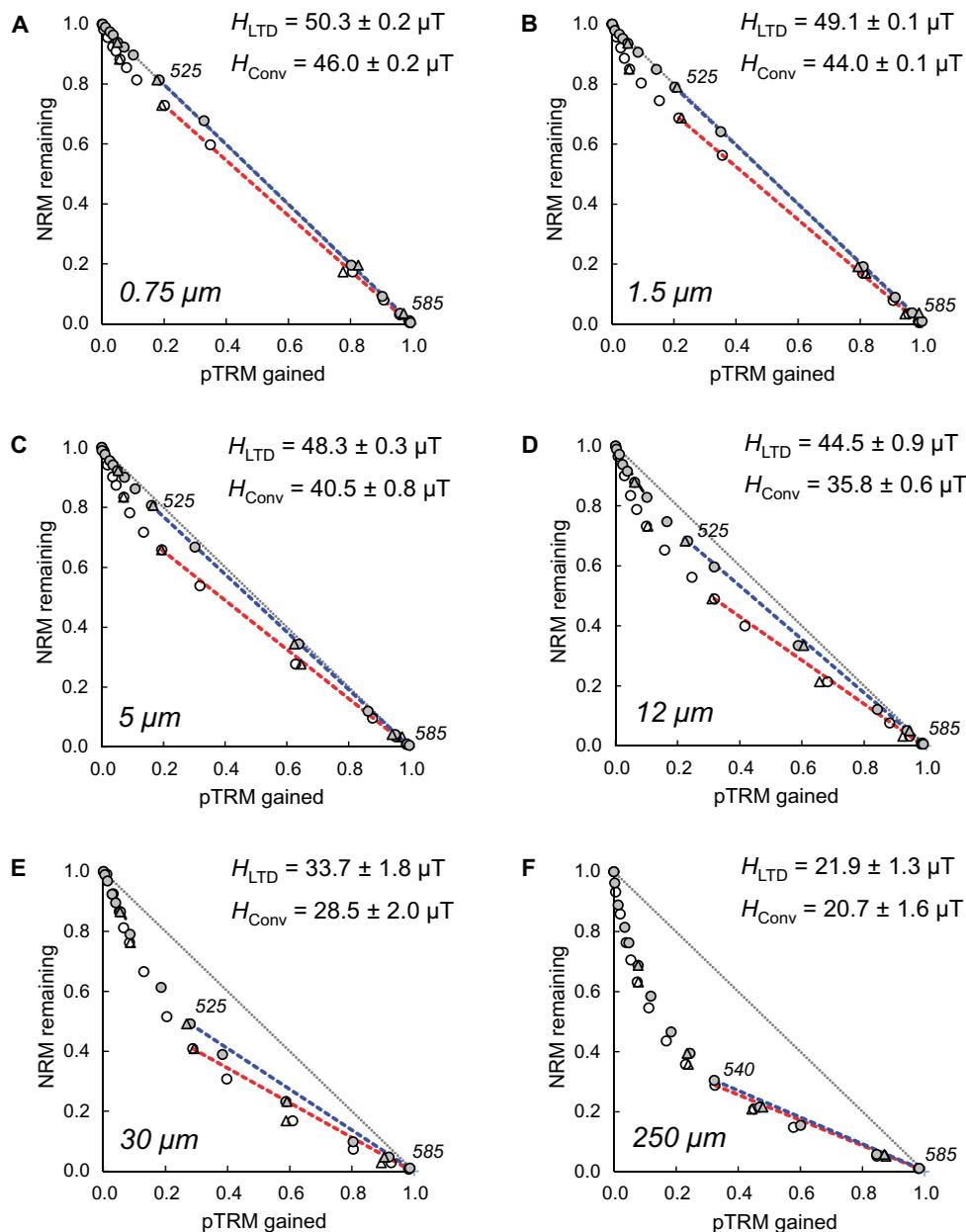
## DISCUSSION

Most absolute paleointensity data have been obtained using variants of the conventional Thellier method from rocks containing PSD magnetic grains, with saturation remanence to saturation magnetization ( $M_{rs}/M_s$ ) ratios as low as 0.1 [for example, see the study of Zhu *et al.* (8)]. Our results demonstrate that these nonideal Thellier experiments may underestimate the true paleofield strength by as much as 25% (Table 1 and Fig. 4). We note that, although curvature of Arai plots and attendant bias may be reduced when the directions of NRM and laboratory magnetic field  $H_{lab}$  are not parallel, non-linear Arai plots have been observed even in perpendicular Thellier

experiments [for example, see the study of Xu and Dunlop (16)]. Because intrinsic low-field bias is fully controlled by magnetic domain state, it should affect all nonideal data independent of the magnetic mineralogy of studied rocks.

Pervasive bias due to the presence of nonideal carriers of paleointensity signal identified by our analyses represents a fundamental physical mechanism that contributes to the dominance of low virtual dipole moment values in the global paleointensity database. In some cases, this bias can be compounded by several additional mechanisms, such as experimental alteration of basalts (25) and basaltic glass (26), low-temperature oxidation, and hydrothermal alteration (27). These factors are likely to account for the deficit of paleointensity values exceeding the present-day value ( $\sim 8 \times 10^{22} \text{ Am}^2$ ) observed from bulk rock data during the Cretaceous Normal Polarity and the Kiaman Reversed Polarity Superchrons [for example, see the studies of Zhu *et al.* (8) and Garcia *et al.* (28)]. Therefore, our results provide a feasible explanation for the apparent discordance between experimental data based on whole rock analyses and numerical models, and lend additional credibility to an inverse relationship between geomagnetic reversal rate and field strength. This interpretation is further corroborated by high-fidelity paleointensity data from single silicate crystals that contain SD magnetic inclusions (29), which consistently indicate high field strengths ( $12.5 \times 10^{22}$  to  $12.7 \times 10^{22} \text{ Am}^2$ ) during superchrons (30–32) and low field strengths ( $4.1 \times 10^{22}$  to  $5.2 \times 10^{22} \text{ Am}^2$ ) during periods of high reversal frequency, such as the late Jurassic (5).

An important corollary of our investigation is that statistical treatments of the global paleomagnetic database from bulk rock samples may not truly represent the long-term behavior of the field strength. Consequently, our data call into question the veracity of some long-term features of the field, such as the Mesozoic Dipole Low (MDL) (33). The MDL was originally proposed on the basis of Thellier analyses of bulk rocks, which suggested geomagnetic field strengths much lower than those of the present day. However, our results support a contrasting view that the apparent low field strength during the MDL may reflect a low-field bias due to rock magnetic effects, and, instead, that the real “MDL” is reduced to an interval of high reversal frequency in the late Jurassic at  $\sim 150$  to 170 million years ago (5, 34).

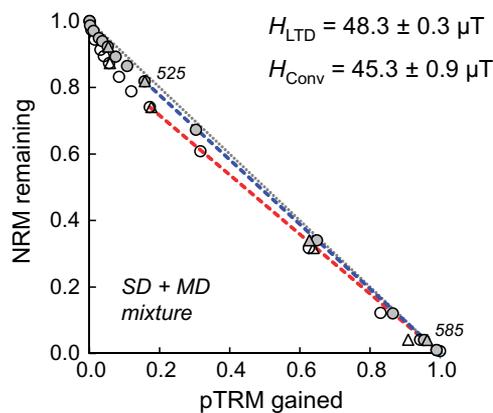


**Fig. 2. Examples of Thellier paleointensity determinations.** Data from series M0.75 (A), M1.5 (B), M5 (C), M12 (D), M30 (E), and M250 (F). NRM remaining versus pTRM gained (circles) is shown. Open and closed symbols represent data measured using the conventional Thellier and LTD-Thellier methods, respectively. Triangles are pTRM checks. Red and blue dashed lines are the best-fit lines for high-temperature Arai segments (the numbers in italics indicate the corresponding temperature range, in degrees Celsius) used to calculate paleointensity estimates  $H_{\text{Conv}}$  and  $H_{\text{LTD}}$  for the conventional Thellier and LTD-Thellier methods, respectively. The best-fit lines for low-temperature Arai segments are not shown for clarity. The negative unit slope line (dotted line) represents the ideal paleointensity behavior expected for an ideal SD carrier (see text).

Similarly, with pervasive fundamental low-field paleointensity bias, some other inferences about long-term geodynamo features, such as the “Proterozoic Dipole Low” (35) or a consistently low long-term global average axial dipole magnetic moment [for example, see the study of Wang *et al.* (36)], require additional scrutiny and may need to be revised. A thorough analysis of the original paleointensity publications is required to determine the extent to which the data are affected by the low-field bias.

Our experimental analyses also demonstrate that, in addition to low-field bias, nonideal Thellier experiments substantially overesti-

mate the true paleofield strength if the low-temperature segment of a double-slope Arai plot is used for best-line fitting, even for PSD samples containing relatively small (0.5 to 1.5  $\mu\text{m}$ ) grains (Table 1). Thus, our study corroborates conclusions of previous studies [for example, see the study of Shcherbakov and Shcherbakova (15)] that these paleointensity values should not be accepted as a faithful paleofield estimate. Nevertheless, data affected by such a high-field bias [for example, see the study of Thomas and Piper (37)] have been included in the global paleointensity database; noncritical acceptance of these

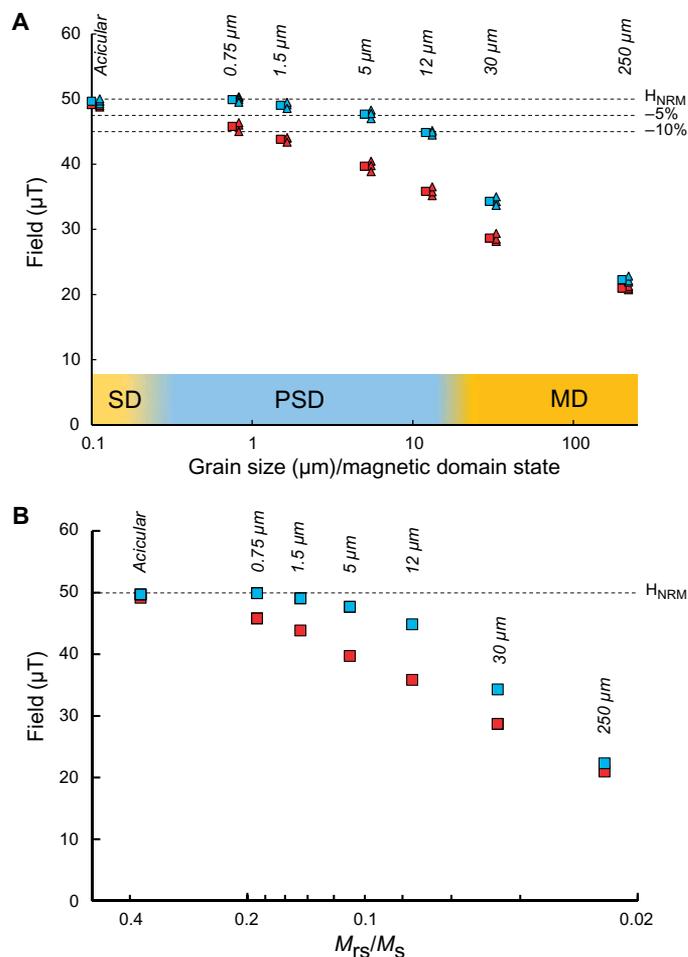


**Fig. 3. An example of Thellier paleointensity determinations from the MIX series.** NRM remaining versus pTRM gained. Open and closed symbols indicate the data measured using the conventional Thellier and LTD-Thellier methods, respectively. Triangles are pTRM checks. Red and blue dashed lines are the best-fit lines for high-temperature Arai segments (the numbers in italics indicate the corresponding temperature range, in degrees Celsius) used to calculate paleointensity estimates  $H_{Conv}$  and  $H_{LTD}$  for the conventional Thellier and LTD-Thellier methods, respectively. The best-fit lines for low-temperature Arai segments are not shown for clarity. The negative unit slope line (dotted line) represents the ideal paleointensity behavior expected for an ideal SD carrier (see text).

data can lead to artifacts in reconstructing geodynamo history and in estimating the age of inner core (38).

Although it is virtually impossible to correct existing data for bias due to nonideal magnetic carriers and other factors, all possible efforts should be made to obtain more accurate determinations in new paleointensity experiments. Our investigation contributes to these efforts by demonstrating that integration of the conventional Thellier approach with LTD-Thellier represents an efficient tool to improve the accuracy of paleointensity determinations by eliminating or significantly reducing intrinsic bias in samples containing PSD magnetite (<10- $\mu\text{m}$  grain size). Furthermore, our data from the MIX series indicate that the LTD-Thellier method may also produce more accurate results from rocks containing two distinct populations of magnetite grains (for example, large magnetite phenocrysts and submicrometer magnetite inclusions in silicate crystals). Although the LTD-Thellier method is only effective for magnetite-bearing rocks, nearly stoichiometric PSD magnetite [for example, formed by subsolidus reactions during initial cooling (39)] represents the most common remanence carrier in both extrusive and intrusive igneous rocks used for paleointensity investigations. The LTD behavior of natural samples may differ to some extent from that of our synthetic samples, due to the pinning of domain walls at exsolution phase boundaries and/or crystalline lattice defects; however, the intrinsic paleointensity bias is unlikely to be significantly affected by these mechanisms (40). All studies using the LTD-Thellier method, albeit limited in number, have resulted in higher success rates and increased quality of paleointensity results (41–43).

Complete understanding of the physical mechanisms responsible for the efficiency of the LTD-Thellier approach awaits development of a comprehensive theory of both thermal remanence and LTD in PSD and MD magnetite. The LTD-Thellier method was originally proposed on the basis of the notion that LTD preferentially removes less stable remanence components carried by MD magnetite and, thus, should magnify the paleointensity signal carried by SD grains and/or



**Fig. 4. Summary of paleointensity results.** Red and light blue symbols represent data obtained using the conventional Thellier and LTD-Thellier methods, respectively. (A) Grain size/magnetic domain state dependence. Triangles, data for individual samples (tables S1 and S2); squares, mean paleointensity values (Table 1). The 0.1- $\mu\text{m}$  grain size used for the M0.35 (acicular magnetite) sample is arbitrary. Individual data points are shifted with respect to the mean for clarity. Horizontal dashed lines correspond to the field of initial NRM acquisition ( $H_{NRM} = 50 \mu\text{T}$ ) and to the 5 and 10% underestimate levels of  $H_{NRM}$ . (B) Mean paleointensity values versus the mean  $M_{rs}/M_s$  (saturation remanence to saturation magnetization) ratios (Table 1).

SD-like structures in larger grains [for example, see the study of Schmidt (19)]. Although the fraction of remanence removed by LTD in our experiments was increasingly larger for samples containing larger size grains, reaching a maximum for MD samples, the linearization effect of the LTD-Thellier approach was most strongly expressed for our PSD samples and is practically absent for MD samples (Table 1). We suggest that this dichotomy may reflect the inherent difference between PSD and MD magnetite in the relative effect of crystallographic twinning (44) or thermal activation of magnetic vortices (45) in monoclinic magnetite on the LTD process.

## MATERIALS AND METHODS

### Sample preparation

For our synthetic samples, we used six different magnetite powders characterized by log-normal grain size distributions with modes of 0.35, 0.75,

1.5, 5, 12, and 30  $\mu\text{m}$ . The 0.35- $\mu\text{m}$  powder contained acicular grains with aspect ratios ranging from  $\sim 1.6$  to  $1:12$ , whereas magnetite grains in the five other powders have an irregular, nearly equidimensional shape (46). An additional powder, prepared by crushing natural magnetite crystals, had a grain size ranging from 100 to 400  $\mu\text{m}$  with a mode of  $\sim 250$   $\mu\text{m}$ . To obtain nearly stoichiometric magnetite, we reduced the powders in a  $\text{CO}_2/\text{CO}$  (9:1) atmosphere at  $400^\circ\text{C}$  for 4 hours. Temperature dependence of low-field magnetic susceptibility measured from the reduced powders upon cycling to  $700^\circ\text{C}$  in argon was reversible and revealed the presence of a single magnetic phase with Curie temperatures of  $\sim 585^\circ$  to  $590^\circ\text{C}$ , which is consistent with nearly pure magnetite.

To prepare samples for paleointensity experiments, we thoroughly mixed the reduced magnetite powders with dry  $\text{CaF}_2$  powder and added a minute amount of distilled water to the mixtures to produce a dense paste. Next, 1-cm<sup>3</sup> cubes were made out of the paste, which was air-dried for 24 hours; after which, the drying process was completed by gradually heating the samples to  $400^\circ\text{C}$  (in nitrogen) for 5 hours. Three samples were prepared from each magnetite powder (referred to as the M0.35, M0.75, M1.5, M5, M12, and M30 series). An additional set of three samples (the M250 series) was prepared using the crushed natural magnetite powder. Finally, three samples (the MIX series) containing a mixture of  $\sim 250$ - $\mu\text{m}$  magnetite (70%) with acicular magnetite (30%) were prepared. The same procedure was used to create additional samples for rock magnetic experiments.

### Rock magnetism

Magnetic hysteresis parameters (coercivity,  $H_c$ ; coercivity of remanence,  $H_{cr}$ ; saturation remanence,  $M_{rs}$ ; and saturation magnetization,  $M_s$ ) were measured at room temperature using an alternating gradient magnetometer (AGM) (model 2900, Princeton Measurement Corporation). The maximum applied field of 0.5 T was used for both magnetic hysteresis loop and backfield demagnetization curve measurements. To test for magnetic anisotropy, we rotated samples ( $45^\circ$  increments) on parallel and perpendicular AGM stages, and measured magnetic hysteresis parameters after each rotation.

Temperature dependences of low-field magnetic susceptibility,  $\kappa(T)$ , were measured upon cycling the samples from room temperature to  $700^\circ\text{C}$  (in argon) using an AGICO MFK-1FA magnetic susceptibility meter equipped with a high-temperature furnace and cryostat. The  $\kappa(T)$  curves were also measured upon heating from  $\sim 192^\circ\text{C}$  ( $\sim 180$  K) to room temperature before and after the high-temperature thermomagnetic runs. The field applied in all  $\kappa(T)$  experiments was 200 A/m.

To test for anisotropy of TRM, we compared the magnitudes of TRMs imparted in a cubic sample from each series in three mutually orthogonal directions. TRMs were imparted by cooling samples from  $700^\circ\text{C}$  (in nitrogen) in a 50- $\mu\text{T}$  field using a thermal specimen demagnetizer (model TD-48SC, ASC Scientific).

### Thellier paleointensity experiments

Conventional Thellier experiments followed the Thellier protocol modified by Coe (10) and Coe *et al.* (18). Zero-field and in-field heatings were done in a thermal specimen demagnetizer (model TD-48SC, ASC Scientific) in an inert (99.9% nitrogen) atmosphere. For the in-field steps, the laboratory magnetic field ( $H_{\text{lab}} = 50$   $\mu\text{T}$ ) was applied parallel to the NRM direction. Samples were always placed at exactly the same position and orientation in the thermal demagnetizer. The temperature steps at  $100^\circ$ ,  $200^\circ$ ,  $300^\circ$ ,  $350^\circ$ ,  $400^\circ$ ,  $450^\circ$ ,  $500^\circ$ ,  $525^\circ$ ,  $540^\circ$ ,  $555^\circ$ ,  $565^\circ$ ,  $575^\circ$ ,  $585^\circ$ , and  $595^\circ\text{C}$  were used. To monitor for exper-

imental alteration, we performed four pTRM checks at  $400^\circ$ ,  $525^\circ$ ,  $555^\circ$ , and  $575^\circ\text{C}$ . The pTRM checks were judged to be successful if they fell within 5% of the original pTRM value. In both conventional Thellier and LTD-Thellier experiments, the magnetic remanence was measured using a 2G Enterprises 760-R superconducting rock magnetometer located in a magnetically shielded laboratory. The resulting paleointensity data were processed using the ThellierTool-4.22 software (47).

LTD-Thellier experiments followed the same protocol as described above except that samples were subjected to two successive LTD steps after each heating. The LTD was conducted by immersing samples in liquid nitrogen in a magnetic field-free environment. The samples warmed to room temperature at a natural rate (no forced heating was applied). The number of LTD treatments needed for effective demagnetization of MD-like remanences was determined experimentally by applying several LTDs to a laboratory TRM induced in a 50- $\mu\text{T}$  field. For all sample series, the difference in remanence measured after the second and third LTD did not exceed 1%. The remanence directions before and after LTDs were indistinguishable.

### SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at <http://advances.sciencemag.org/cgi/content/full/3/2/e1602306/DC1>

Supplementary Results

fig. S1. Typical dependences of low-field magnetic susceptibility versus temperature.

fig. S2. Magnetic hysteresis properties.

table S1. Paleointensity results obtained using the conventional Thellier method by fitting the high- and low-temperature segments of the Arai plots.

table S2. Paleointensity results obtained using the LTD-Thellier method by fitting the high- and low-temperature segments of the Arai plots.

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## Intrinsic paleointensity bias and the long-term history of the geodynamo

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