

## Experimental CRM production in a basaltic rock; evidence for stable, intermediate palaeomagnetic directions

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### SUMMARY

Remagnetization experiments involving secondary thermoremanent and chemical remanent magnetization (TRM–CRM) have been carried out on samples from a basaltic lava flow. The rock carries a stable NRM residing in deuterically oxidized (class II–III) titanomagnetite grains, and some haematite formed by low-temperature oxidation. Heating to 525 °C in a controlled field of 52  $\mu$ T for periods ranging from 1 to 400 hr, resulted in oxidation to haematite of a parent phase close to magnetite in composition. The direction of the resulting CRM is controlled both by the remagnetizing field and the primary remanence (NRM), giving rise to stable high-blocking remanence components with intermediate directions, and precluding identification of the primary NRM component.

Control experiments carried out on chemically stable specimens gave a partial TRM (PTRM) parallel to the remagnetizing field, with blocking temperatures in agreement with single-domain theory. After removal of this secondary PTRM, the primary direction in these specimens was regained.

The results imply that caution is advisable in analysing the palaeomagnetic directions of any rock which has undergone a thermochemical event.

**Key words:** chemical remanent magnetization, palaeomagnetism, remanence deflection, rock magnetism.

### 1 INTRODUCTION

During the past two decades, the importance of secondary magnetic overprinting in palaeomagnetism has been widely recognized. In rock units with chemically stable mineralogies, secondary magnetic effects associated with burial or reheating related to tectonic events, are restricted to the acquisition of a thermo- or thermoviscous remanent magnetization (TRM–TVRM). For ambient temperatures well below the blocking temperature ( $T_B$ ) of the magnetic carriers, the primary magnetization can generally be resolved from the secondary overprint by standard palaeomagnetic demagnetization techniques.

A more complex situation arises when the reheating also results in chemical changes of the magnetic minerals. New magnetic minerals may form as a result of precipitation and subsequent grain growth from a non-magnetic matrix, or by alteration of parent magnetic phases to daughter phases with different magnetic properties. This may give rise to a chemical remanent magnetization (CRM) which often has a

magnetic stability comparable to, or greater than the original remanence, leading to difficulties in resolving the two components and determining their relative ages. Mineral growth occurs in the presence of both the Earth's magnetic field and a magnetostatic field related to the remanence in the parent magnetic phase. The possibility of exchange coupling between parent and daughter phase during chemical alteration must also be considered. The direction of a secondary CRM is therefore not self-evident.

Various authors have reported a range of apparently conflicting CRM directional behaviour for different mineralogies and experimental conditions. Porath (1968) oxidized synthetic acicular SD maghemite grains (0.4  $\mu$ m) to haematite in a field of 108  $\mu$ T by heating in air to 540° and 580 °C. The resulting CRM was parallel to the laboratory field direction  $H_{CRM}$ . In a similar set of experiments, Özdemir & Dunlop (1988) also oxidized maghemite in air at temperatures ranging from 300° to 604 °C, but used smaller grains (0.025  $\mu$ m) and a palaeomagnetically more realistic field strength of 50  $\mu$ T. This again produced a CRM parallel to the applied field  $H_{CRM}$ .

Field controlled CRM directions were also found by Hunt, Banerjee & Marvin (1986) in oceanic pillow lavas for

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magnetite-ilmenite exsolution at 300–350 °C in fields of 10–50  $\mu\text{T}$ . In contrast, Johnson & Merrill (1974) reported CRM growth along the primary remanence direction for low-temperature oxidation of SD magnetite in a field of 50  $\mu\text{T}$ . Wayman & Evans (1977) predictably also found CRM directions controlled by primary remanence when oxidizing sea-floor basalts at 300 °C in zero field.

Marshall & Cox (1971), using oceanic basalts and a field of 50  $\mu\text{T}$ , observed remanence growth both in the direction of an original NRM and in the applied field direction. They interpreted their results in terms of two separate processes, one enhancing the original NRM by increasing the saturation moment of remanence carrying grains, the other giving rise to a CRM parallel to the applied field. Their results, however, may alternatively be explained by a CRM formed in a direction intermediate between  $\mathbf{H}_{\text{CRM}}$  and  $\mathbf{J}_{\text{NRM}}$ . By the same token, the results of Johnson & Merrill (1972, 1973), who oxidized magnetite and titanomagnetite grains in an excited oxygen gas atmosphere in the 50–200 °C temperature range, are explainable both in terms of an intermediate and a field-controlled CRM.

Bailey & Hale (1981) were the first to report directionally stable CRMs intermediate between  $\mathbf{J}_{\text{NRM}}$  and  $\mathbf{H}_{\text{CRM}}$ . They oxidized sea-floor basalts containing PSD titanomaghemite at 515 °C, obtaining a magnetic phase close to magnetite in composition. For field strengths of 10 and 20  $\mu\text{T}$ , a univectorial CRM with an intermediate direction was obtained, while a field of 50  $\mu\text{T}$  was sufficient to align the CRM along the applied field  $\mathbf{H}_{\text{CRM}}$ .

Özdemir & Dunlop (1985) oxidized synthetic single-domain titanomagnetite ( $\text{Fe}_{2.4}\text{Ti}_{0.4}\text{Al}_{0.2}\text{O}_4$ ) by heating in air to temperatures between 100° and 230 °C in fields of 50 and 100  $\mu\text{T}$  respectively. They found that the CRM remained parallel to the initial remanence for low and medium degrees of oxidation ( $z \leq 0.62$ ), while the highest oxidation degrees ( $z > 0.7$ ) resulted in intermediate CRM directions.

Heider & Dunlop (1987) oxidized SD/PSD magnetite grains by heating in air at 500–520 °C. They obtained contrasting results for the single phase oxidation of magnetite to maghemite, and the multiphase oxidation of magnetite to haematite. In the first case CRM remained parallel to the initial remanence for fields as high as 1.5 mT, whereas intermediate directions were observed for fields of 50–200  $\mu\text{T}$  in the second case. They proposed a model in which exchange coupling prevails through the single phase magnetite–maghemite transition, effectively preserving the initial remanence direction. Breakdown of the crystal lattice in the magnetite–haematite transition, however, causes breakdown of exchange-coupling, and allows acquisition of CRM along the direction of a net internal field controlled jointly by  $\mathbf{J}_{\text{NRM}}$  and  $\mathbf{H}_{\text{CRM}}$ . Heider & Dunlop demonstrated a clear directional dependence on the relative strengths of  $\mathbf{J}_{\text{NRM}}$  and  $\mathbf{H}_{\text{CRM}}$  in their multiphase experiments.

In the present study we report on secondary CRM and partial TRM (PTRM) magnetizations induced in natural basaltic specimens carrying a stable NRM, by reheating to 525 °C in a field of 52  $\mu\text{T}$ . After heat treatment the specimens were subjected to standard stepwise thermal demagnetization, and the directional properties of the secondary magnetization were analysed in detail.

For comparison, a parallel set of specimens were pre-heated to 700 °C and given a total TRM by slow cooling

before being subjected to the same set of experiments. This produced samples that were apparently chemically stable, enabling us to monitor a pure PTRM/VRM type remagnetization.

Experimenting with rocks carrying a natural remanent magnetization rather than using synthetically prepared samples, prevents experimental control on factors such as magnetic grain size and mineralogy, origin and strength of primary remanence and homogeneity of the samples. We feel, however, that this is justified in order to obtain results which are more directly comparable to processes operating in nature.

## 2 CHARACTERIZATION OF STARTING MATERIAL

### 2.1 Suitability criteria

A rock suitable for CRM experiments of the type described should satisfy the following criteria.

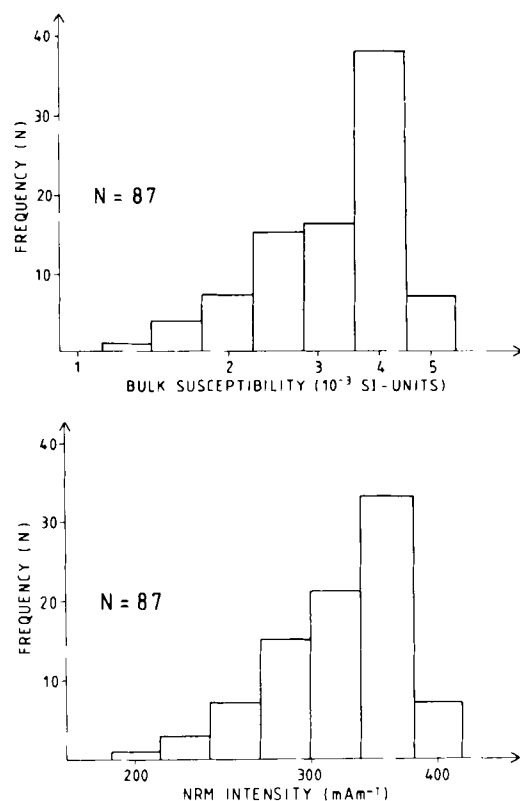
- (1) A stable, univectorial NRM.
- (2) Negligible anisotropy of magnetic susceptibility (AMS).
- (3) Uniform magnetic mineralogy.
- (4) Production of a 'daughter' magnetic mineral phase within reasonable time, and at a temperature well below the Curie temperature  $T_C$  for the 'parent' magnetic mineral. The daughter phase should preferably have a higher  $T_C$  than the parent phase.

### 2.2. Bulk magnetic properties and NRM

The rock samples used in this study were collected from an approximately 12 m thick basaltic lava flow from Crawton Bay, 30 km south of Aberdeen, Scotland. The palaeomagnetic properties of the flow have been studied by several authors (Embleton 1968; Sallomy & Piper 1973; Torsvik 1985), all reporting a reversed magnetic direction consistent with a Lower Devonian age. The presence of both normal and reversed lava flows in the same sequence, combined with a positive contact test, indicates that the observed magnetic direction is primary in origin (Torsvik 1985).

To ensure maximum homogeneity in the bulk magnetic properties and NRM directions, all specimens for the experiments were taken from a single hand sample. 32 drill cores were obtained from this sample, providing a total of 94 cylindrical specimens ( $d = 19 \text{ mm}$ ,  $h = 18 \text{ mm}$ ). Bulk susceptibility and AMS were determined on a KLY-1 low-field induction bridge, and remanence measurements were carried out on a Digico spinner magnetometer. Instrument sensitivities are of the order of  $8 \times 10^{-8} \text{ SI units}$  and  $3 \times 10^{-4} \text{ A m}^{-1}$  respectively for the specimen size used. Thermal demagnetization was performed in a Schonstedt TSD-1 furnace, with a hold time of 1 hr for each demagnetization step.

Seven of the specimens showed anomalous NRM intensities and/or directions, possibly due to spurious remanence acquired during sampling or drilling. Although the anomalous components could be removed by alternating field demagnetization to moderate field strengths ( $< 20 \text{ mT}$ ), these specimens were discarded.



**Figure 1.** Distribution of bulk susceptibility (top) and NRM intensity (bottom). Horizontal axes on logarithmic scale.

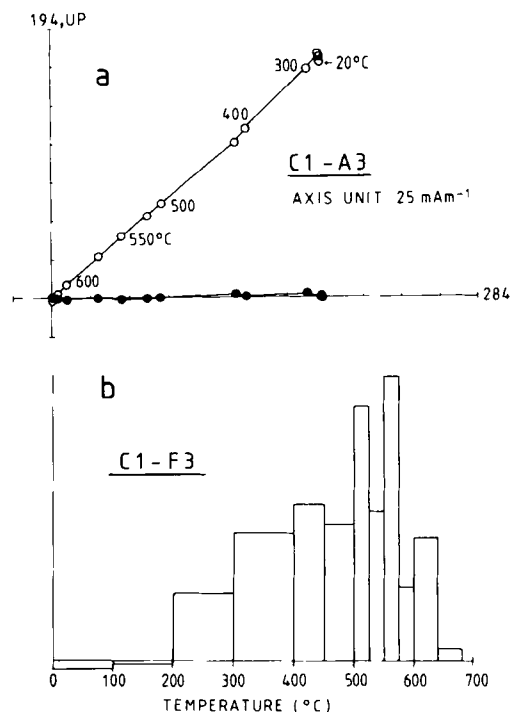
The remaining 87 specimens have NRM intensities ranging from 191 to 378  $\text{mA m}^{-1}$ , and bulk susceptibilities between 1.20 and  $4.14 \times 10^{-3}$  SI units. Both properties exhibit slightly skewed distributions when plotted on a logarithmic scale (Fig. 1), contrasting with the log-normal distributions expected for homogeneous material (Tarling 1966; Irving, Molyneux & Runcorn 1966; Latham *et al.* 1989). This can probably be explained by the topmost specimen in each drillcore having a slightly lower susceptibility and remanent intensity than the underlying specimens, attributed to surface oxidation of the rock.

NRM directions are extremely well grouped with  $D = 284$ ,  $I = -43$ ,  $k = 280$  and  $\alpha_{95} = 1^\circ$  (Fisher 1953). Alternating field (six specimens) and thermal demagnetization (six specimens) cause hardly any directional changes (Fig. 2a). Remanence unblocking takes place over the whole temperature range from 200  $^\circ\text{C}$  to 680  $^\circ\text{C}$  with a peak in unblocking just below the Curie point of magnetite (Fig. 2b). The maximum available alternating field of 100 mT is insufficient to remove the remanence, leaving on average 47 per cent of the initial remanence untouched.

### 2.3. Magnetic mineralogy

The magnetic mineralogy of the initial material as well as of the pre-heated specimens, was determined by thermomagnetic analysis, isothermal remanent magnetization (IRM) acquisition analysis and reflected light microscopy.

Microscope studies reveal titanomagnetite grains ranging in size from approximately 100  $\mu\text{m}$  down to the limit of the optical resolution ( $<1 \mu\text{m}$ ), residing in a reddish and



**Figure 2.** (a) Thermal demagnetization of specimen C1-A3 shown in optimal orthogonal vector projection. Open and closed circles denote vertical and horizontal projections respectively. (b) Unblocking temperature spectrum for specimen C1-F3.

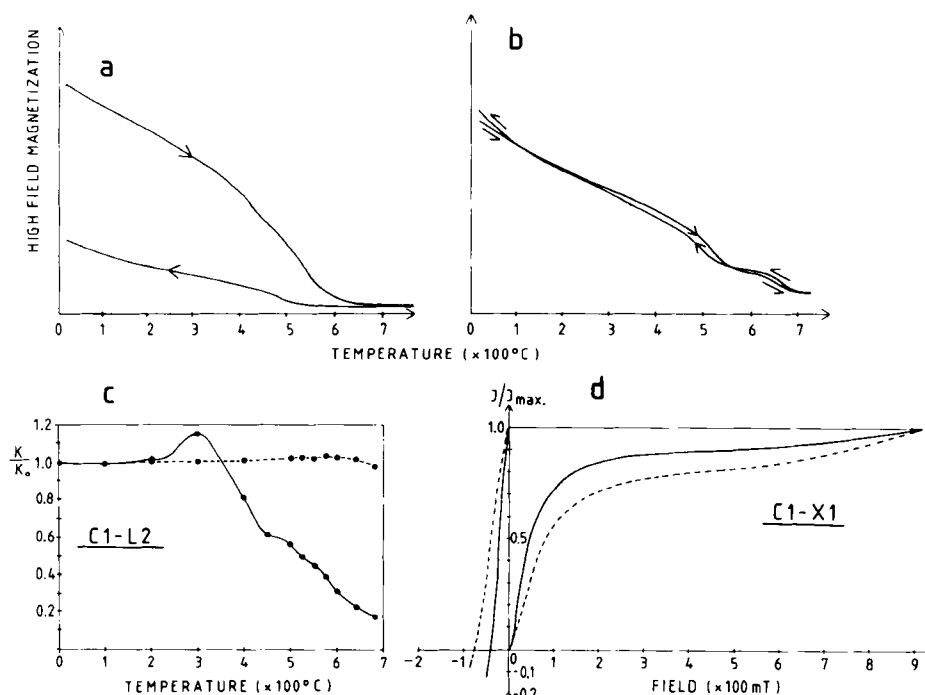
probably haematite-bearing matrix. Exsolution lamellae of ilmenite are well developed in the larger grains, indicating deuteric oxidation corresponding to oxidation class II–III (Ade Hall, Palmer & Hubbard 1971). Moderate to strong hydrothermal alteration is also evident, causing corrosion of the titanomagnetite grains, and indicating the presence of cation-deficient phases.

Thermomagnetic analysis was conducted on an automatic horizontal translation balance in a field of 800 mT, with a heating-cooling cycle to 700  $^\circ\text{C}$  of approximately 1 hr. Untreated samples show Curie points ranging from 530 $^\circ$  to 580 $^\circ\text{C}$ , occasionally with a significant decrease in  $T_C$  after heating, as shown in Fig. 3(a). Most samples also show weak reversible Curie points above 650  $^\circ\text{C}$ , indicating the presence of almost pure haematite. Upon cooling the magnetic moment of the samples has been reduced by more than 50 per cent, consistent with breakdown of part of the titanomagnetite phase to haematite. After pre-heating to 700  $^\circ\text{C}$  for 1 hr, the samples became stable against further chemical alteration, as evidenced by reversible curves (Fig. 3b) with an enhanced haematite Curie point.

Plots of bulk susceptibility variation during thermal demagnetization, indicate onset of oxidation between 300 $^\circ$  and 400  $^\circ\text{C}$  in untreated specimens, but give stable values for heat-treated specimens (Fig. 3c).

IRM-H curves (Fig. 3d) also reflect the presence of two distinct magnetic phases. After heating to 700  $^\circ\text{C}$ , the high-coercivity phase becomes more dominant, and mean IRM intensity at 920 mT is reduced from 79.9 to 24.9  $\text{A m}^{-1}$ . This is again consistent with the conversion of a titanomagnetite phase to haematite.

In summary, the samples seem to contain Ti-poor, and

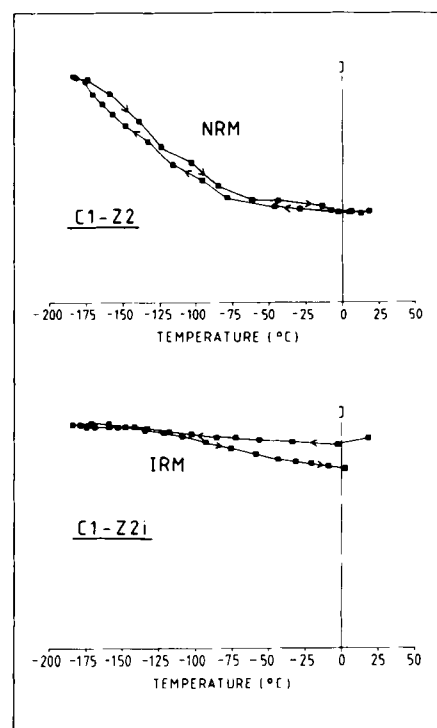


**Figure 3.** Magnetomineralogical stability before and after heating to  $700^\circ\text{C}$  for 1 hr. (a) Thermomagnetic curve before heating. (b) Thermomagnetic curve after heating. (c) Changes in bulk susceptibility during progressive thermal demagnetization before (continuous line) and after (broken line) heating. (d) IRM-H acquisition and backfield curves before (continuous line) and after (broken line) heating.

probably cation-deficient titanomagnetite and haematite formed through both high- and low-temperature oxidation of an original titanomagnetite phase. Heating causes further breakdown of the titanomagnetite phase, and formation of more secondary haematite. The extensive low-temperature oxidation indicates that the NRM is probably partly of chemical origin in addition to a TRM acquired during cooling.

#### 2.4. Domain state

Domain states were inferred from low-temperature magnetic properties and hysteresis parameters. Variations in intensities of NRM and IRM were monitored during cooling from  $20^\circ\text{C}$  down to liquid nitrogen temperature ( $-196^\circ\text{C}$ ) utilizing a low-temperature spinner magnetometer. Cooling magnetite through its isotropic point at  $-143^\circ\text{C}$  causes a sharp decrease in multidomain remanence. A slight decrease is also expected for equidimensional SD/PSD grains where the remanence is partly controlled by the magnetocrystalline anisotropy (Levi & Merrill 1978). No change is expected for acicular SD/PSD grains where shape anisotropy is the controlling factor. For low-Ti titanomagnetites ( $x < 0.15$ ) the isotropic temperature decreases with increasing Ti content (Syono 1965), but the same general behaviour as for pure magnetite is retained. Neither NRM nor IRM magnetizations show any reduction when passing through the transition (Fig. 4), indicating a purely SD/PSD origin of the remanence. A slight increase of remanent intensity on cooling is expected due to a rise in spontaneous magnetization, but the observed increase for the NRM is far too large to be explained by this factor alone. A tentative explanation is that superparamagnetic grains in the specimens cross the threshold to a stable single-domain



**Figure 4.** Changes in remanent intensity during cooling/heating cycle to  $-196^\circ\text{C}$  for NRM (top) and IRM induced at 400 mT (bottom) respectively. Cooling and heating denoted by arrows.

configuration when the critical size for this transition is lowered on cooling, and are magnetized parallel to the existing remanence.

Hysteresis properties were measured on six samples utilizing a Molspin vibrating sample magnetometer with a

peak field of 1 T. Since this field is not sufficient to saturate the haematite in the samples, some uncertainty applies to the determination of hysteresis related parameters.  $J_{rs}/J_s$  ratios were nevertheless extremely consistent, ranging from 0.19 to 0.21, while coercive force ( $H_c$ ) varied from 12.5 to 15.5 mT. The theoretical value for  $J_{rs}/J_s$  in SD grains with uniaxial anisotropy is 0.5, falling to  $<0.02$  for true MD magnetite (Dunlop 1986). The observed values are theoretically consistent with small PSD material. Experimental studies of  $J_{rs}/J_s$  versus grain size in magnetite (Day, Fuller & Schmidt 1977; Dunlop 1986) are complicated by the fact that material prepared by grinding yields consistently higher ratios than grown crystals in a lower state of stress. The ratio increases with increasing titanium content within the titanomagnetite series for grains of similar size (Day *et al.* 1977), but the behaviour of mixtures of different minerals and grain sizes (Wasilewski 1973; Day *et al.* 1977) is not well known. The observed values of  $H_c$  and  $J_{rs}/J_s$  are however, associated with 'single domain' behaviour in natural rocks as defined by the Lowrie–Fuller test (Dunlop 1983).

## 2.5 AMS

Anisotropy of magnetic susceptibility (AMS) was measured on five specimens before and after heating to 700 °C, mainly to assess the amount of remanence deflection which could be expected when magnetizing the specimens in the laboratory. The anisotropy factor  $[(k_{\max}/k_{\min}) - 1]$  varied between 1.1 and 1.4 per cent before and 0.2 and 2.1 per cent after heating. This would normally indicate a maximum remanence deflection of well below 1° (Stacey 1960; Hrouda 1982; Cogne 1987), although certain unfavourable grain size distributions may give larger deflections (Stephenson, Sadikun & Potter 1986). TRM deviation effects due to AMS are regarded as negligible for the present experiments.

## 3 EXPERIMENTAL SET-UP AND PROCEDURES

Laboratory heatings were carried out in a magnetic field environment produced by a Sperry Rand Dycome ('Dynamic Controlled Magnetic Environment') field control system. This system consists of three orthogonal sets of circular Helmholtz coils (radius 1.20 m), and a control unit enabling variation of field strength and direction at the centre of the coils. Specimens were placed in a non-inductive furnace, and heating–cooling cycles were monitored by an automatic temperature control unit. Errors in temperature determination are maximum 10 °C, and errors in estimated magnetization direction due to specimen orientation and AMS are maximum 2° (Walderhaug 1987).

The temperature was increased approximately linearly from room temperature to 525 °C during the course of 1 h. A somewhat lower heating rate was adopted close to the final temperature to prevent an initial 'overshoot' above 525 °C. Specimens were then held at this temperature for periods of 1, 3, 6, 12, 24, 48, 96, 200 and 400 hr respectively, with three chemically stable and three untreated specimens being used for each time interval. Cooling was accomplished by simply switching off the current to the furnace, causing the temperature to drop exponentially to below 100 °C in approximately 5 hr. All specimens were left in place until a

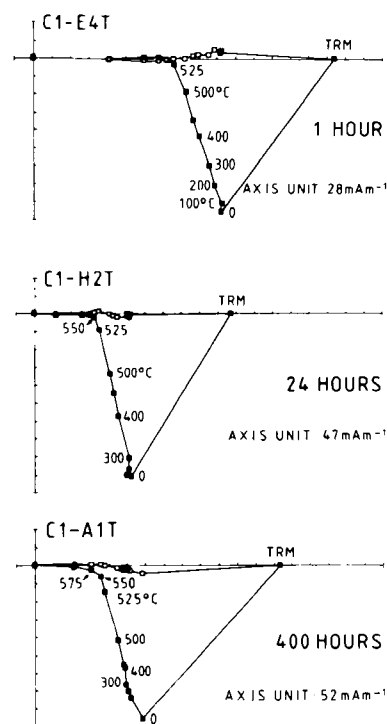
temperature of less than 50 °C had been reached. A constant field of 52  $\mu$ T along a direction of  $D = 0^\circ$ ,  $I = -17^\circ$  in the specimens' coordinate system was maintained throughout the heating–cooling cycle.

Specimens intended for use in pure TRM/VRM experiments were subjected to a similar heating–cooling cycle to 700 °C for 1 hr before commencement of the experiments described above. Heating took place in zero field, with a field of 50  $\mu$ T being applied along the specimens' cylindrical axis ( $I = -90^\circ$ ) only during cooling. The 'primary' magnetization in these specimens should therefore be a pure TRM acquired on cooling from 700 °C.

During subsequent thermal demagnetization, all specimens were held at temperature for 1 hr at each demagnetization step.

## 4 RESULTS OF TRM/VRM EXPERIMENTS

Figure 5 shows three examples of the directional behaviour upon thermal demagnetization of chemically stable specimens. The secondary PTRM/VRM is parallel to the direction



**Figure 5.** Experimental results for three pre-heated specimens shown in orthogonal vector projections. The horizontal axis is chosen along the direction of primary remanence  $J_{\text{TRM}}$ . Filled squares denote projections into the plane of magnetization defined by the primary remanence  $J_{\text{TRM}}$  and the laboratory field direction  $H_{\text{CRM}}$ . All remanence directions should ideally be confined to this plane. Open squares denote projections orthogonal to the plane of magnetization, and can be disregarded except as a measure of experimental noise. Deflections from the ideal plane appear as the angles between open squares and the horizontal axis. Points marked '0' show directions after experimental heating runs to 525 °C. A secondary PTRM parallel to  $H_{\text{CRM}}$  is removed during thermal demagnetization between 0° and 525 °C, before directions swing back to the primary remanence direction. Note increase in unblocking temperature of secondary remanence with increased heating time.

**Table 1.** Summary of experimental results for preheated specimens.  $t$  = experimental heating time;  $J_{TRM}$  = intensity of primary remanence;  $k_0$  = bulk susceptibility before heating runs to 525 °C;  $A_i$  = angle between  $J_{TRM}$  and  $H_{CRM}$ ;  $A_{525} - A_{600}$  = angular deflection from  $J_{TRM}$  after demagnetization to give temperature;  $k_1/k_0$  = Ratio of bulk susceptibilities after and before experimental heating runs.

Sample	$t$ (hours)	$J_{TRM}$ (mA/m)	$k_0$ (SI*10 <sup>-3</sup> )	$A_i$ (°)	$A_{525}$ (°)	$A_{550}$ (°)	$A_{575}$ (°)	$A_{600}$ (°)	$k_1/k_0$
C1E4T	1	460	4.76	72	3	1	1	1	1.01
C1H3T	1	447	5.99	72	1	1	1	1	1.01
C1AA3T	1	308	3.58	72	2	1	1	1	1.01
C1D2T	3	425	6.39	72	3	2	1	1	1.00
C1U3T	3	579	4.28	73	3	1	1	0	1.00
C1G1T	3	600	9.54	72	6	3	1	1	1.00
C2A3T	6	445	10.34	73	3	1	1	1	-
C1E2T	6	497	7.00	73	4	1	1	1	-
C1C2T	6	536	10.60	73	14	4	2	1	-
C2C2T	12	408	5.67	73	3	1	2	2	-
C1I1T	12	680	12.88	73	8	3	0	1	-
C1D1T	12	730	13.35	73	14	2	1	1	-
C1H2T	24	502	9.78	73	15	4	2	2	0.96
C1U1T	24	681	11.44	71	13	3	2	3	0.99
C1T3T	24	430	4.13	72	4	1	1	1	1.01
C1W2T	48	635	10.75	72	8	2	1	1	0.95
C1V4T	48	691	4.60	73	2	1	0	0	0.99
C1AA1T	48	612	9.76	72	8	3	1	1	0.95
C1D3T	96	388	3.77	73	4	2	1	1	1.00
C1B4T	96	661	4.35	73	4	2	1	1	0.99
C1L1T	96	575	10.20	73	10	3	2	1	0.95
C1O2T	200	510	7.73	73	8	3	1	1	0.95
C1E3T	200	399	3.44	74	5	2	1	1	1.00
C1N1T	200	455	4.92	73	4	2	2	2	0.99
C1D4T	400	344	4.18	73	13	3	2	2	0.98
C1A1T	400	692	12.92	74	20	9	4	3	0.88
C2A1T	400	564	12.19	73	4	4	4	3	0.91

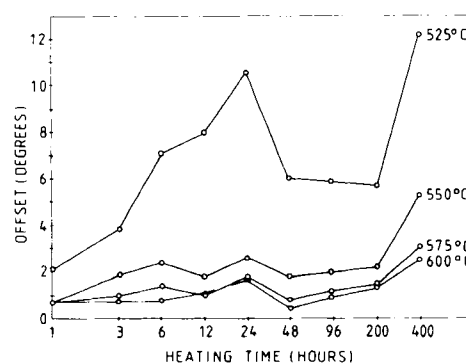
#### MEAN OF EACH TIME INTERVAL

1	405	4.78	72	2	1	1	1	1.01
3	535	6.74	72	4	2	1	1	1.00
6	493	9.31	73	7	2	1	1	-
12	606	10.63	73	8	2	1	1	-
24	538	8.45	72	11	3	2	2	0.99
48	646	8.37	72	6	2	1	1	0.96
96	541	6.11	73	6	2	1	1	0.98
200	455	5.36	73	6	2	2	1	0.98
400	533	9.76	73	12	5	3	3	0.92

of the laboratory field  $H_{CRM}$ . Unblocking of the secondary remanence is virtually complete after demagnetization to 525 °C for the shorter heating cycles, rising to 575 °C for the longer runs. After removal of the secondary component, the 'primary' remanence ( $I = -90^\circ$ ) is perfectly reproduced.

The results for a total of 27 specimens are summarized in Table 1. After demagnetization to 600 °C, all specimens are within 3° of the 'primary' field direction, and the mean directions are identical within statistical error limits ( $D = 276$ ,  $I = -89$  and  $\alpha_{95} = 1^\circ$  for 'primary' TRM;  $D = 343$ ,  $I = -89$  and  $\alpha_{95} = 1^\circ$  for 600 °C directions). The largest offsets (3°) are associated with two of the three specimens with the longest (400 hr) heating times. These two specimens also show a marked decrease in susceptibility (12 and 9 per cent respectively) during the experimental run, indicating that pre-heating to 700 °C for 1 hr was not quite sufficient to prevent further chemical change during the longest heating run. The largest directional offsets may therefore be associated with CRM formation.

Figure 6 illustrates the angular offset observed during unblocking of the secondary remanence as listed in Table 1. Theoretical curves based on single-domain theory illustrating the time-temperature dependence of blocking and unblocking of remanence, were first presented by Pullaiah *et al.* (1975). A slightly different approach by Walton (1980)



**Figure 6.** Angular offset from primary remanence direction for preheated specimens after demagnetization to 525°, 550°, 575° and 600 °C respectively. Points denote mean of the three specimens used at each heating time. Horizontal axis on logarithmic scale.

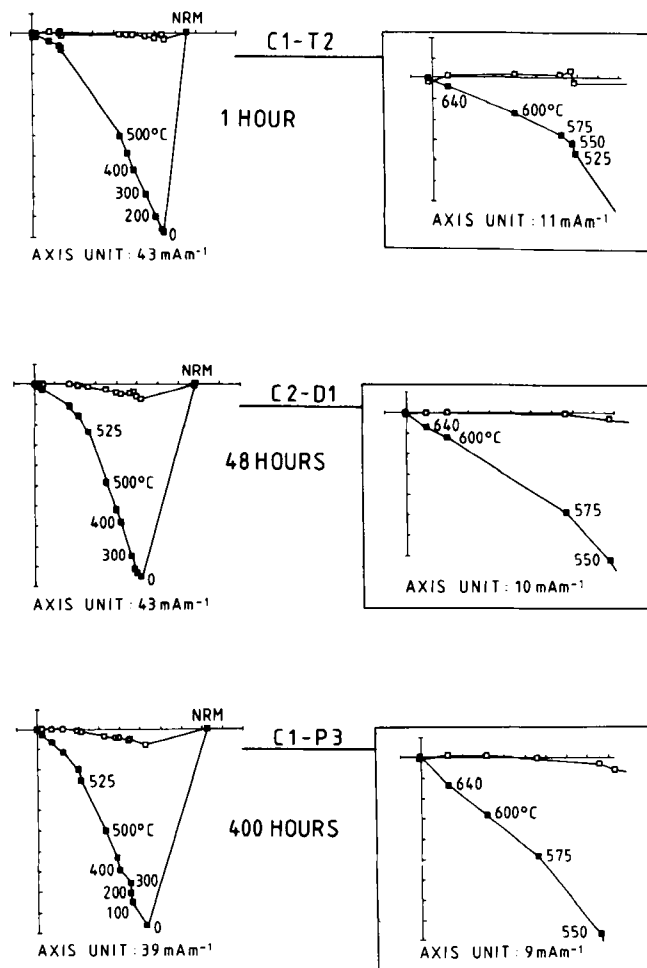
leads to a second set of curves given by Middleton & Schmidt (1982), which show a somewhat stronger time dependence. Both predict that remanence acquired in magnetite or haematite during 1 hr at 525 °C should be practically removed by demagnetization for 1 hr at the same temperature. A remanence in magnetite acquired during 400 hr at 525 °C should be removed by heating for 1 hr at

550 °C, while haematite requires approximately 560 °C according to Pullaiah *et al.*'s model, or 575 °C following Walton. Although individual specimens show some variation in behaviour, the results are in general agreement with these predictions. Discrepancies with theory on the order of 100 °C as reported by Pullaiah *et al.* (1975) are certainly not in evidence.

## 5 RESULTS OF CRM EXPERIMENTS

Figure 7 shows three examples of thermal demagnetization of untreated specimens. As can be readily seen from comparison with Fig. 5, these results are in marked contrast with those from the 'chemically stable' specimens. A secondary component parallel to the laboratory field direction is removed by demagnetization to between 525° and 575 °C, but the primary and known NRM direction cannot be retrieved. An apparently directionally stable high-temperature blocking component is observed in an intermediate position between the original NRM and the laboratory field direction  $H_{CRM}$ .

The results for all 27 specimens are summarized in Table



**Figure 7.** Experimental results for three untreated specimens shown in orthogonal vector plots. Projections and conventions as in Fig. 5. Insets show expansions of the high-blocking portions of the magnetization. Note increasing offset from  $J_{NRM}$  with increased heating time.

2 and Figs 8 and 9. Removal of the secondary PTRM/VRM, which should be complete before demagnetization to 600 °C, still leaves an angular deviation from the original NRM direction ranging between 19° and 47° for individual specimens, suggesting a fundamental difference between the remagnetization experiments involving CRM and pure TRM/VRM acquisition respectively.

Individual specimens show somewhat erratic results, but there is a general correlation ( $r = 0.73$ ) between remanence deflection and the logarithm of experimental heating time, as shown in Fig. 10(a). Reduction in susceptibility should reflect the amount of oxidation of titanomagnetite to haematite taking place in the individual specimens, and therefore the amount of CRM formation. There is, however, only a slight positive correlation ( $r = 0.58$ ) between susceptibility reduction and remanence deflection, as shown in Fig. 10(b).

## 6 DISCUSSION AND INTERPRETATION OF CRM EXPERIMENTAL RESULTS

The observed directional behaviour can conceivably be explained both by the acquisition of a directionally stable intermediate CRM, or a vector sum comprising a remnant of the original NRM plus a CRM parallel to the laboratory field. We favour a true intermediate CRM for the following reasons.

### (i) Directional stability of components

If a vector sum of two separate components is to give a directionally stable remanence direction, these two components must have almost identical blocking spectra. Although this might conceivably happen for some of the specimens, it seems highly unlikely that the majority should show the stable behaviour observed, given the individual, albeit small, differences in magnetic properties. Some specimens *do* show a slight directional change towards the NRM direction in the range between 525° and 600 °C (cf. Table 2), but this is consistent with removal of the secondary PTRM which is also observed in the chemically stable specimens (Table 1). All specimens were subjected to standard palaeomagnetic directional analysis, utilizing a least-squares algorithm developed by Torsvik (1986). Both this program and visual inspection (e.g. Fig. 7), give the impression that most of the high-blocking components easily meet any reasonable standards for palaeomagnetic stability.

### (ii) Intensity of high-blocking components

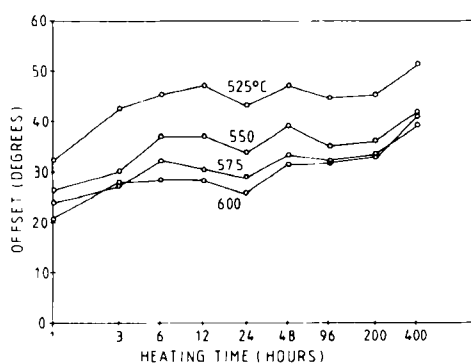
The remaining magnetic vector after demagnetization to 600 °C was resolved into components along  $J_{NRM}$  and  $H_{CRM}$  respectively, yielding an average remaining intensity of  $27.6 \pm 8.2 \text{ mA m}^{-1}$  along  $J_{NRM}$  for the 27 specimens. 12 specimens not used in the experiments were demagnetized directly to 600 °C, and yielded an average remaining intensity of  $11.1 \pm 7.1 \text{ mA m}^{-1}$ . The difference between the two groups is statistically significant at the 99 per cent confidence level, providing strong evidence that CRM must have been formed in a direction different from  $H_{CRM}$  during the experiments, and thus supporting the notion of a true intermediate CRM. Some caution is needed however, as the

**Table 2.** Summary of experimental results for untreated specimens.  $t$  = experimental heating time;  $J_{\text{NRM}}$  = intensity of primary remanence;  $k_0$  = bulk susceptibility before heating runs to 525°C;  $A_i$  = angle between  $J_{\text{NRM}}$  and  $H_{\text{CRM}}$ ;  $A_{525} - A_{600}$  = angular deflection from  $J_{\text{NRM}}$  after demagnetization to given temperature;  $k_1/k_0$  = Ratio of bulk susceptibilities after and before experimental heating runs.

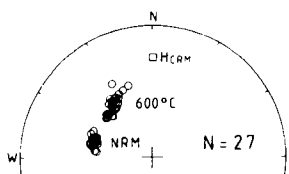
Sample	$t$ (hours)	$J_{\text{NRM}}$ (mA/m)	$k_0$ (SI*10 <sup>-3</sup> )	$A_i$ (°)	$A_{525}$ (°)	$A_{550}$ (°)	$A_{575}$ (°)	$A_{600}$ (°)	$k_1/k_0$
C1T2	1	326	34.26	65	29	26	24	23	0.56
C1Y3	1	321	29.77	67	35	26	23	19	0.50
C1C1	1	344	38.62	66	33	27	24	20	0.51
C1Z1	3	335	37.61	66	30	28	24	31	0.55
C1I2	3	371	40.88	66	49	34	31	27	0.52
C2D2	3	265	19.77	69	49	28	26	25	0.55
C2B1	6	345	38.98	68	32	30	26	21	0.39
C1S1	6	348	41.05	74	53	46	37	34	0.45
C1J1	6	356	38.59	67	51	35	34	30	0.49
C1G3	12	265	23.51	69	42	33	30	28	0.44
C1Y1	12	358	39.48	67	46	37	29	28	0.46
C1E1	12	362	35.74	68	52	41	32	29	0.46
C1P2	24	352	37.11	64	39	38	36	32	0.45
C1AB1	24	263	17.37	70	45	31	22	22	0.48
C1B2	24	298	23.63	68	46	32	28	24	0.45
C1R4	48	294	25.26	65	44	34	27	26	0.38
C1K1	48	348	36.04	73	56	46	41	37	0.40
C2D1	48	337	32.47	69	42	37	32	31	0.42
C1I3	96	240	22.12	65	39	29	28	29	0.31
C2F1	96	315	38.09	63	51	42	37	36	0.33
C1P1	96	353	37.40	61	43	34	31	30	0.41
C1R1	200	341	33.07	67	50	38	35	34	0.36
C1AB2	200	271	25.97	68	34	32	29	29	0.27
C1O1	200	350	33.45	66	52	38	36	36	0.36
C2B3	400	244	18.55	70	47	34	33	35	0.37
C1P3	400	317	32.80	68	49	45	40	42	0.30
C2A2	400	323	36.13	68	58	46	44	47	0.33

#### MEAN OF EACH TIME INTERVAL

1	330	34.22	66	32	26	24	21	0.53
3	324	32.75	67	42	30	27	28	0.54
6	350	39.54	70	45	37	32	28	0.44
12	328	32.91	68	47	37	30	28	0.45
24	304	26.04	67	43	34	29	26	0.46
48	326	31.26	69	47	39	33	31	0.40
96	303	32.54	63	44	35	32	32	0.35
200	321	30.83	67	45	36	33	33	0.33
400	295	29.16	69	51	42	39	41	0.33



**Figure 8.** Angular offset from primary remanence direction for untreated specimens after demagnetization to 525°, 550°, 575° and 600°C respectively. Points denote mean of the three specimens used at each heating time. Horizontal axis on logarithmic scale.



**Figure 9.** Primary remanence direction (NRM) and laboratory field direction ( $H_{\text{CRM}}$ ) compared to directions after demagnetization to 600°C for all untreated specimens.

difference may in part be caused by the different laboratory treatment for the two groups. Chemical changes also take place during thermal demagnetization itself, further complicating the analysis.

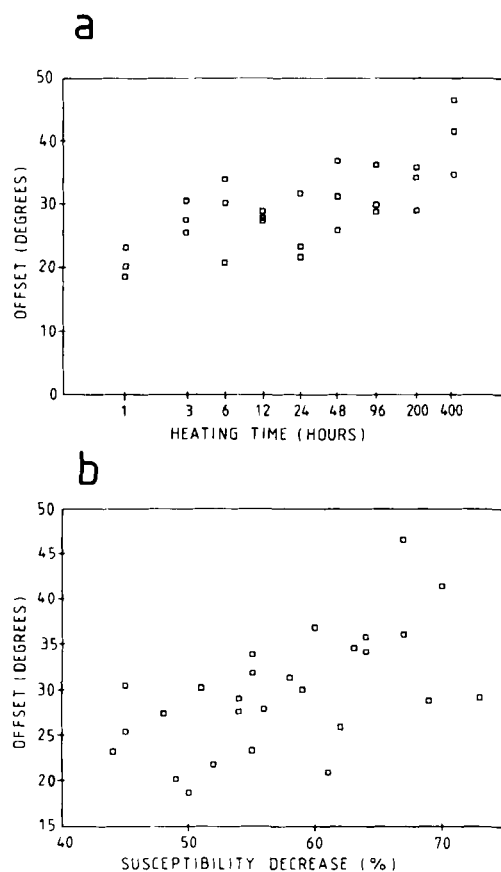
Assuming then, that the CRM direction is indeed intermediate, these results would seem to fit into the framework suggested by Heider & Dunlop (1987), where multiphase oxidation leads to intermediate direction CRM. Heider & Dunlop proposed that the CRM formed during multiphase oxidation of magnetite to haematite in their synthetic samples, was acquired parallel to a net internal field in magnetic grains given by

$$H_{\text{res}} = \beta J_r + H_{\text{CRM}} \quad (1)$$

This simple linear relationship assumes that exchange coupling between parent and daughter phases is unimportant. Their observed directions and intensities yielded a reasonably good fit to a value of 0.07 for the constant  $\beta$ , or 0.56 when corrected for unblocking of remanence and reduction of  $J_s$  on heating.

A similar calculation performed on our specimens, using NRM intensity and direction for  $J_r$ , yielded uncorrected  $\beta$  values ranging from 0.06 to 0.28 in individual specimens, or 0.44 to 2.19 after correction for unblocking of remanence up to 525°C and reversible reduction of  $J_s$  on heating (Pullaiah *et al.* 1975). These values seem unreasonably large,





**Figure 10.** Angular offset from primary remanence direction for untreated specimens after demagnetization to 600°C, as a function of experimental heating time and susceptibility decrease respectively. Heating time axis on logarithmic scale.

indicating a magnetostatic field due to initial remanence well in excess of the Lorentz interaction field, which has a value of 0.33 J in a region of uniform magnetization. One might therefore speculate that exchange coupling does after all play some role during the transition of the cubic phase to haematite.

Alternatively, the value of  $\beta$  in equation (1) may be reduced by supposing that the remanent magnetic intensity in the parts of magnetic grains undergoing oxidation is higher than the average remanent intensity ( $J_r$ ) of the grains. Oxidation to haematite would be expected to preferentially take place near grain surfaces. Both domain structure and chemical composition of the parent phase may also differ between the surface and interior of the grains, providing a possible basis for variations in remanent intensity, and thereby allowing for a purely magnetostatic origin of the observed CRM directions.

Whatever the mechanism controlling CRM formation, it seems clear that CRM formation in these large grains must be understood on the scale of each of the individual titanomagnetite grains undergoing oxidation, rather than by assuming a uniform field across the grains. Preliminary investigations of other rocks (Walderhaug, unpublished results) also suggest that the relative importance of  $J_{\text{NRM}}$  and  $H_{\text{CRM}}$  is by no means constant during the magnetite to haematite oxidation process.

The increase in remanence deflection with time presented

in Fig. 10(a) may reflect two different processes. At an early stage of each heating run, we may suppose that the net internal field  $H_{\text{res}}$  is controlled strictly by the original NRM direction and  $H_{\text{CRM}}$ . If CRM forms along this field, however, the resultant direction of remanence, and therefore  $H_{\text{res}}$ , will be deflected towards  $H_{\text{CRM}}$ . CRM formation or resetting at a later stage in each heating run may therefore give rise to directions more distant from the original NRM. The observed time dependence would also be enhanced if a larger fraction of the original NRM component survived the shorter heating runs, deflecting the remanence in short runs towards the NRM.

## 7 IMPLICATIONS FOR PALAEOMAGNETISM

The present CRM experiments have been carried out on rocks carrying an ancient remanent magnetization, and at a field strength equal to the Earth's field at high latitudes. The results should therefore indicate what would happen to a similar rock in nature during a reheating event, with one important qualification: we cannot match geological time in the laboratory, so we have to use relatively high temperatures to obtain a sufficient amount of oxidation of the parent magnetic phase.

Nevertheless, the results show that caution is needed in the interpretation of palaeomagnetic results in any rock which has been exposed to a thermochemical event. Adopting the high-blocking component in our experiments as a true representation of the geomagnetic field direction, would give an error in the determination of virtual geomagnetic pole position of the order of thousands of kilometres, which is obviously unacceptable in palaeomagnetic work.

Ideally, the intermediate directions should always be confined along great circle paths between primary remanence directions and the remagnetizing field direction. Even if individual specimens give stable directions, a group of such specimens should therefore give rise to a non-Fisherian elongated distribution. This is indeed observed for the group in Fig. 9, apparently providing a basis for discriminating between 'true' palaeofield recordings and 'false' recordings of the type described above.

In practice, however, recognizing such great circle trends in the data is not always a straightforward task. The elongation in Fig. 9 is enhanced by the different oxidation times for individual specimens, resulting in varying amounts of CRM formation. The extremely high quality of the data also makes the non-Fisherian nature of the distribution more evident than it would have been for (more normal) poorer quality data.

A smaller amount of CRM deflection, say 5°, from the direction of primary remanence, would not significantly alter the shape of the original group. Although less serious in terms of the magnitude of the error induced, such deflections would conceivably introduce a systematic bias to the data which could be extremely difficult to detect.

## 8 CONCLUSIONS

The major conclusions of this study are as follows.

- (i) A pure TRM/VRM type partial remagnetization in the

flow studied accurately preserves the direction of both the primary remanence and the magnetic overprint, and gives blocking temperatures in agreement with single-domain theory.

(ii) A partial remagnetization involving CRM formation by breakdown of a cubic parent phase to secondary haematite, gives rise to an apparently stable high-blocking component intermediate between the direction of primary remanence and the remagnetizing field.

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