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A large gyromagnetic effect in greigite

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SUMMARY

Rotational and anhysteretic remanent magnetization (RRM and ARM) acquisition curves of two sediment samples containing greigite have been measured up to peak fields of 80 mT at rotation frequencies between 5 and 95 revolutions per second (rps). At 95 rps the ARM (70 μT direct field, applied antiparallel to the RRM) increased almost linearly with peak field but the RRM increased approximately exponentially. The effective field (B_g), defined in this case as $70 \times$ RRM/ARM, was about 1100 μT for the two samples after the application of an alternating field (AF) of 80 mT peak. B_g is approximately 10 times higher than previously observed for magnetite of size 1 μm. Although greigite is the dominant ferrimagnetic mineral present in these samples, other studies (Snowball 1997a) have shown that low concentrations of detrital multidomain magnetite are also present, so that the high value of B_g must be regarded as a lower limit.

Unlike a magnetite sample that was used for comparison, both greigite samples had a negative RRM at all rotation frequencies below 50 rps but, like magnetite, there was an increase in RRM when it changed from negative to positive at 50 rps. In addition, unlike magnetite, the ARM was not constant but approximately halved as the RRM became strong and positive above 50 rps (ARM antiparallel to RRM). Thus there appeared to be an interaction between the ARM and the RRM. Further investigation of this interaction by applying weak direct fields parallel and then antiparallel to the RRM (80 mT peak AF, 95 rps rotation rate) showed that the ARM was linear with direct field but was indeed always smaller when the direct field producing it was antiparallel to the RRM.

At present it is not clear why gyromagnetic remanences are so strong in greigite. Such high values of B_g have never been observed before except in the very special case of a self-reversing lithium chromium ferrite near its moment compensation temperature. These high values of B_g might enable RRM to be used as an indicator for greigite if they turn out to be unique to this mineral.

Key words: demagnetization, greigite, gyroremanence, magnetite, remanent magnetization, rock magnetism.

INTRODUCTION

The mineral greigite is often found in marine and freshwater sediments and contributes to the natural remanent magnetization (NRM) since it may acquire a strong chemical remanent magnetization (CRM). The paper by Snowball (1997a) contains a list of references to work carried out on the magnetic properties of rocks containing this mineral. In particular, gyromagnetic remanences have been identified and studied by Snowball (1997a,b) and Hu *et al.* (1998) in sediments from Swedish lakes and the Tibetan Plateau, respectively. It is interesting to note that Krs *et al.* (1990, 1991) observed that during static AF demagnetization of greigite-bearing samples from Bohemia, strong components of anhysteretic remanent magnetization (ARM) were produced in fields as low as 40–60 mT. The results of this paper suggest that these components might have been a gyroremanent magnetization (GRM) rather than ARM. GRM refers to a remanence of gyromagnetic origin in general, but the term rotational remanent magnetization (RRM) (which is a GRM) is retained for historical reasons to denote a gyroremanence produced when specimen rotation is involved.

GRM PRODUCED BY STATIC AF DEMAGNETIZATION

To examine the strength of the gyromagnetic effect in greigite and to compare it with that observed for magnetite, two representative sediment samples from Sweden were used. The first was from Bara Mosse (labelled BAM8) and the second was from Bjorkerods Mosse (labelled BMR7). Both samples were taken from sections of fresh sediments (from horizons known to contain high concentrations of greigite) and were impregnated with low-viscosity epoxy resin. Cubes of side 1 cm were cut from the solidified resin. Comparison of NRM measurements with those of standard palaeomagnetic samples (not impregnated) indicated that the NRM was not affected by the resin impregnation.

Stationary AF demagnetization of the NRM showed that, after the initial decrease, the intensity started to increase above about 80 mT (Fig. 1a), and the direction of magnetization changed in a systematic manner (Fig. 1b). This indicated that another remanence was being acquired (see Snowball 1997a,b) for results on similar samples).

In Fig. 1(a), the NRM remaining is the remanence observed after stationary AF demagnetization along the x-, y- and z- (down) axes (laboratory coordinates) in that order. In Fig. 1(b), as the intensity starts to increase above 80 mT (the last three points on each curve), a constant final direction in the x, y-plane (i.e. zero inclination) is approached. This is a characteristic of GRM produced by the above technique (Stephenson 1993), that is, it appears in a plane at 90° to the last AF axis used. The direction within that plane is determined by the intrinsic anisotropy of the sample. It is anisotropy that, in a static AF, is responsible for producing a predominant sense of switching of the moments of single-domain particles that leads to a GRM. Hu *et al.* (1998) have also found GRM with the above characteristics in greigite-bearing samples from the Tibetan plateau.

RRM AND ARM ACQUIRED AT LOW ROTATION RATES

Gyroremanences can also be produced in isotropic (or anisotropic) samples by, for instance, continuously rotating the sample in a slowly decreasing alternating field. At high rates of rotation the switching of the moments is all in the same direction (Stephenson 1988a) and so a large gyroremanence is acquired (RRM). At low rates, there is probably a mixture of positive and negative switching depending on the rotation rate (divided by the AF frequency) and on the coercivity distribution. This means that the results at low rates are more difficult to interpret (see Stephenson 1980). The reversal of sign at 50 rps is due to an overall change in the sense of the switching of the moments in the AF.

Snowball (1997a,b) measured RRM acquisition of greigite samples at a rotation rate of 5 rps in alternating fields up to 110 mT peak and found a very marked effect, the slope of the RRM versus field curve increasing with peak fields especially above about 40 mT. The results he obtained were very similar in shape to those shown in Figs 6 and 7 described below. He also measured the static longitudinal ARM in a weak field, $b = 100$ μT (parallel to the AF), and found that it had about the same magnitude as the RRM, that is, the effective field B_g ('gyrofield') producing RRM was also about -100 μT, where B_g is defined as (Potter & Stephenson 1986)

$$B_g = b \times \mathrm{RRM}/\mathrm{ARM}\,, \qquad (1)$$

where the ARM is the rotational ARM produced under the same AF conditions as the RRM (use of a static ARM as above is only an approximation to this). Snowball's estimate of B_g at low rotation rates is much higher than the values measured by Potter & Stephenson (1986) at low rotation rates for different magnetite size fractions. The RRM at this low rotation rate was antiparallel to the rotation vector, hence the negative B_g. Since more consistent results for magnetite (and rocks) are obtained at high speeds of rotation (Stephenson 1980; Potter & Stephenson 1986), it is better to make comparisons between samples at high rates of rotation and to use the ARM acquired under the same conditions in the calculation of B_g (see next section). Since B_g depends on AF strength as well as rotation rate, Potter & Stephenson (1986) have chosen 80 mT peak AF (fairly high but easily attainable) and 95 rps (lowest rotation rate for a large RRM at 50 Hz AF) as 'standard conditions' for

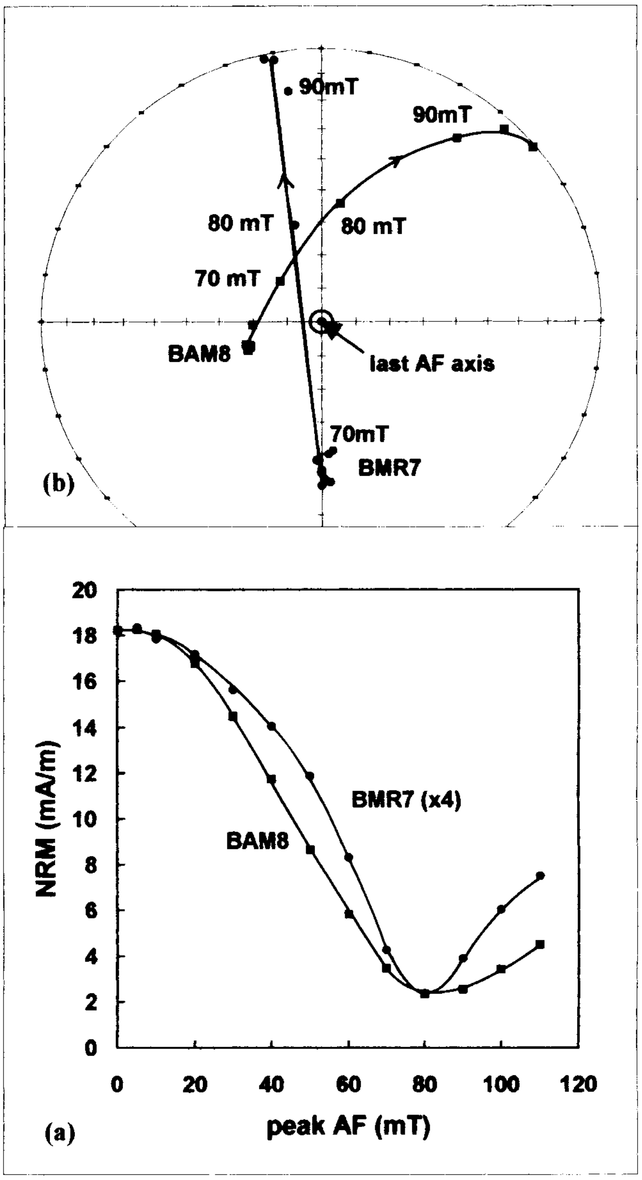

Figure 1. (a) The variation of intensity with peak AF for BAM8 and BMR7 during stationary AF demagnetization of the NRM. After demagnetizing along each axis in turn the resultant remanence was that remaining after the last demagnetization. Note that for BMR7, the NRM has been multiplied by 4 before plotting. (b) The corresponding direction changes during AF demagnetization for the two samples shown on a conventional stereogram. The outer great circle represents the x, y-plane. The last demagnetization for each AF was along the z-axis as shown.

comparative purposes. Provided rotational ARM is linear with direct field, any value of field is acceptable. (For these samples, ARM was linear with direct field up to at least 200 μT—see Fig. 5).

RRM AND ARM ACQUIRED AT HIGH ROTATION RATES

Magnetite sample (2.2–4.4 μm)

To see whether large B_g values would be obtained at high rotation rates, specialized equipment designed for this purpose (Stephenson & Molyneux 1987) was used. To provide a direct comparison with the greigite samples and to check the calibration, a fractionated magnetite sample of particle size in the range 2.2–4.4 μm (100 mg dispersed in resin) was measured to compare its results with those obtained many years previously (Potter & Stephenson 1986). In the present test, the *z*- (downwards) component of RRM and ARM (in 70 μT) was measured after rotation about a vertical axis at various speeds between 5 and 95 rps in the presence of an AF (aligned horizontally), which was slowly reduced from a peak value of 80 mT. Each rotation rate was used three times. These were (i) clockwise, (ii) anticlockwise and (iii) anticlockwise with a 70μT field applied downwards along the rotation axis. In (i) and (ii) the vertical ambient field component was cancelled. Thus in (i) and (ii) RRMs of opposite sign were produced along the rotation axis while in (iii) an RRM was produced together with a rotational ARM. The only difference between (i) and (ii) was the sense of rotation and so half the difference of the magnetizations produced in (i) and (ii) gave the RRM. The only difference between (ii) and (iii) was the addition of a 70 μT downward-acting field and so the difference between (iii) and (ii) gave the rotational ARM (Stephenson & Molyneux 1987). Fig. 2 shows the dependence of RRM and rotational ARM (in 70 μT) on rotation frequency. At 95 rps B_g was 28 μT, in agreement with values obtained previously for this sample (2.2–4.4 μm). [Potter & Stephenson (1986) found empirically that for magnetite particles of size *d* covering the range 0.2–90 μm, $B_g(\mu T) = 100/d(\mu m)$.] In Fig. 2, note the general shape of the graph including the negative RRM at very low rotation rates, the small positive RRM around 25 rps, the negative RRM just below 50 rps (AF frequency 50 Hz) and in particular that the ARM is constant, independent of both the rate of rotation and the RRM that was simultaneously acquired.

Greigite samples

In contrast, Figs 3 and 4 show the result for the two greigite samples. Below 50 rps, the RRM is always negative, unlike the magnetite sample. Above 50 rps, like magnetite, a strong positive RRM is produced. The ARM, however, unlike magnetite, does not remain constant but approximately halves when the strong RRM is acquired. Note that at high rotation rates the RRM is positive, that is, parallel to the rotation vector, i.e. upwards during rotations (ii) and (iii). In (iii), however, the 70 μT field producing the ARM is downwards and so the ARM is acquired antiparallel to the RRM. Thus, for these greigite samples, ARM acquired when the direct field is antiparallel to the RRM is smaller (and B_g therefore greater) than when the direct field is parallel to the RRM (as at low rotation speeds).

Using the three-spin method above (95 rps, 80 mT peak AF, 70 μT direct field antiparallel to the RRM) gives values for B_g of (1050 ± 30) μT for BAM8 and (1070 ± 40) μT for BMR7. These are very large values and are some 10 times larger than the 100 μT obtained for 1 μm magnetite (Potter & Stephenson 1986). Thus to produce an ARM equal to the RRM under the same AF and rotation conditions, a bias field some 20 times stronger than the Earth's field must be applied. These values of B_g are approximately equal to the previous highest value measured for B_g defined in the above way. This is all the more

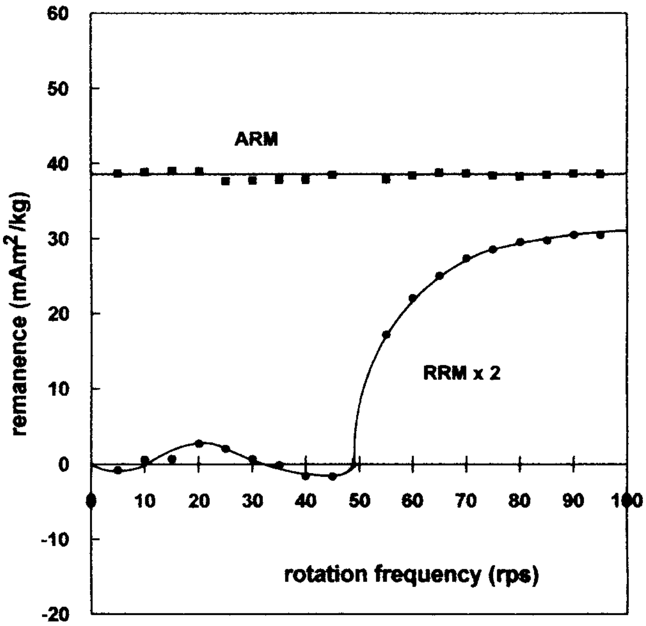

Figure 2. The variation of RRM and ARM (in 70 μT direct field) with rotation speed for the 2.2–4.4 μm magnetite sample at 80 mT peak field. Note that the RRM has been multiplied by 2 before plotting.

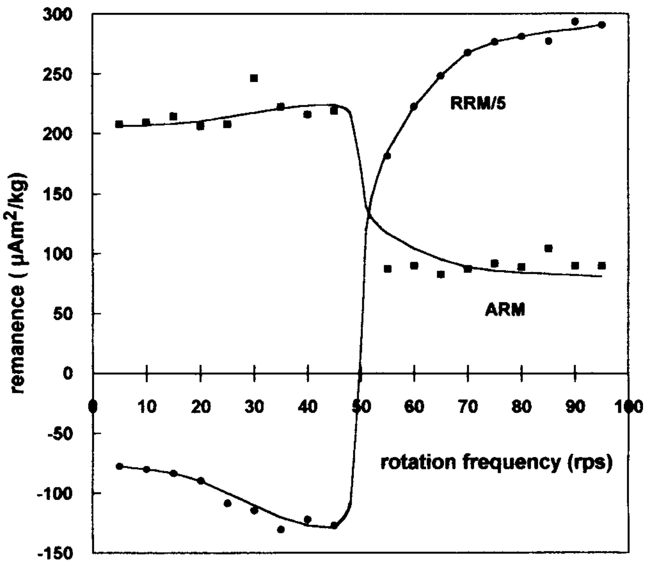

Figure 3. The variation of RRM and ARM (in 70 μT direct field) with rotation speed for BAM8 at 80 mT peak field. The line through the ARM results is obtained from eq. (2). Note that the RRM has been divided by 5 before plotting.

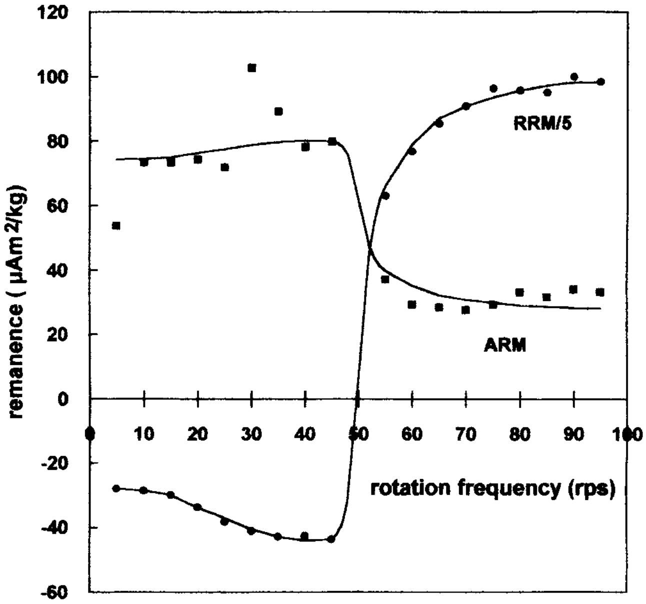

Figure 4. The variation of RRM and ARM (in 70 µT direct field) with rotation speed for BMR7 at 80 mT peak field. The line through the ARM results is obtained from eq. (2). Note that the RRM has been divided by 5 before plotting.

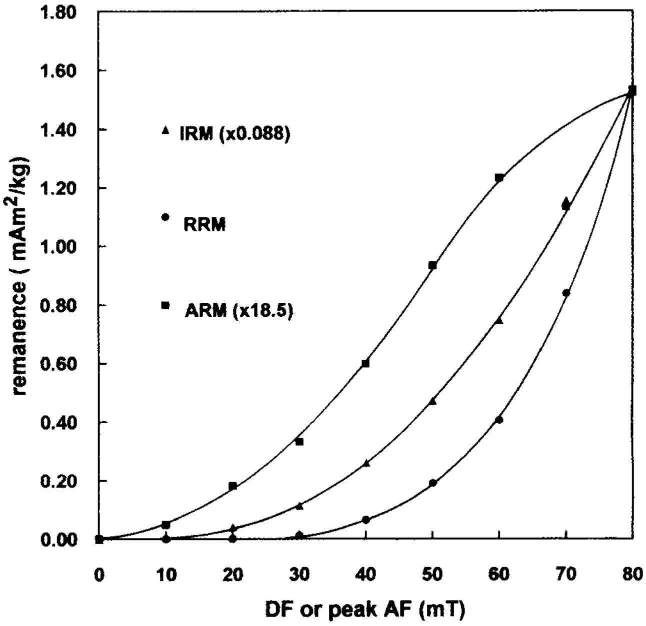

Figure 6. The variation of RRM and ARM (in 70 µT direct field) with peak field for BAM8 at 95 rps. Note that the ARM has been multiplied by 18.5 before plotting. The acquisition of IRM in a direct field (DF) of the same magnitude as the peak AF and along the same direction as the RRM and ARM is also shown for comparison. The IRM results have been multiplied by 0.088 before plotting.

remarkable because the previous record was held by a synthetic ferrite under very special conditions, namely a lithium chromium ferrite ($Li_{0.5}Cr_{1.25}Fe_{1.25}O_4$) with a compensation temperature, T_m (about 30 °C), at which the opposed moments of the two sublattices, which exhibit different temperature dependences of their magnetization, exactly balance. Because the angular momenta associated with the moments do not compensate at this temperature there is angular momentum but very little moment, so the gyromagnetic effect is very big

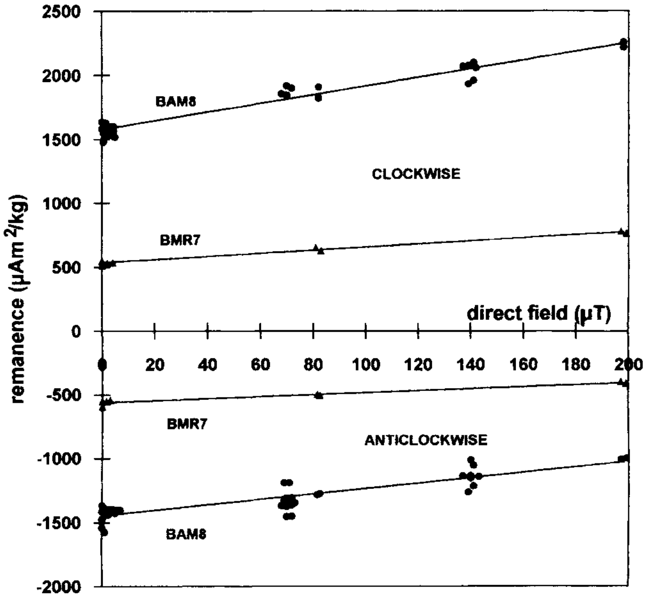

Figure 5. The dependence of remanence for BAM8 and BMR7 on weak direct field (up to 198 µT) after being spun about a vertical axis clockwise and anticlockwise at 95 rps in an increasing and then a slowly decreasing horizontal AF of maximum peak value 80 mT. The weak direct field was always applied downwards, that is, along a direction specified by the vector describing clockwise rotation. At zero direct field an RRM is acquired whose sign is governed by the sense of rotation. The ARM acquired per unit direct field (slope of the graphs) is greater when the direct field is parallel to the rotation vector (clockwise). Because there are a large number of points for some fields, they have been spread slightly along the abscissa to show them more clearly.

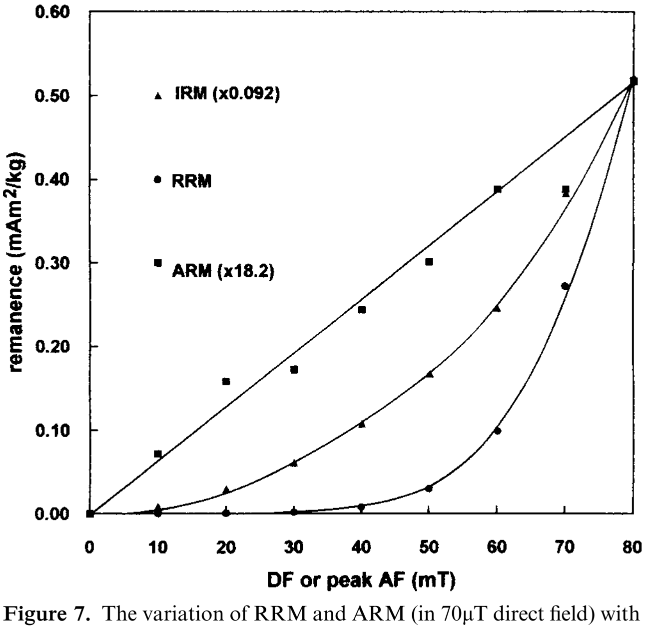

Figure 7. The variation of RRM and ARM (in 70µT direct field) with peak field for BMR7 at 95 rps. Note that the ARM has been multiplied by 18.2 before plotting. The acquisition of IRM in a DF of the same magnitude as the peak AF and along the same direction as the RRM and ARM is also shown for comparison. The IRM results have been multiplied by 0.092 before plotting.

(Stephenson 1988b). The maximum B_g measured near T_m was 1030 μT (but if the temperature had been closer to T_m then presumably B_g would have been higher).

INTERACTION BETWEEN ARM AND RRM

Because there is no obvious physical reason why rotational ARM should have any dependence on rotation rate (and this is borne out by the result for magnetite in Fig. 2), it seems reasonable to conclude that the change in ARM observed at 50 rps is a consequence of the large change in RRM that occurs at this rotation rate and that is unavoidably simultaneously acquired with the ARM. It appears, therefore, that there is an interaction between the RRM and the ARM.

To investigate this interaction further and to show that it is unconnected with rotation rate, both samples were spun clockwise and anticlockwise at 95 rps with direct fields of 0, 82 and 198 μT applied either parallel or antiparallel to the rotation vector. BAM8 was also measured at 70 and 140 μT. Fig. 5 shows the result. It can be seen that for both samples the ARM acquisition is linear with field to within the measurement uncertainties (which can be estimated from the scatter of the points) and is more easily acquired when the sample is rotated clockwise, that is, the rotation vector is downwards and parallel to the applied direct field.

The slopes of the lines in units of (μA m^2 kg^{-1}) μT^{-1} are

BAM8: 3.38 ± 0.15 (clockwise) and 2.14 ± 0.19 (anticlockwise) ,

BMR7: 1.20 ± 0.07 (clockwise) and 0.78 ± 0.07 (anticlockwise) .

When the direct field is antiparallel to the RRM (anticlockwise rotation), the slope is significantly smaller (ARM less easily acquired) than when the direct field is parallel to the RRM, just like the results of Figs 3 and 4. There is thus qualitative agreement between these two sets of results [one set with the rotation rate varied, the other set with the direct field varied and sense of (constant) rotation changed].

In the absence of any theoretical model to explain these results, it seems reasonable to attempt to quantify the interaction empirically. A tentative relationship between ARM and RRM is the following:

$$\mathrm{ARM} = (A + k\mathrm{RRM})b\,, \qquad (2)$$

where $b = \mu_0 h$ and is the weak field applied to produce the ARM. This equation allows the slope of ARM versus direct field to depend on the RRM via an interaction constant k. If the RRM is positive (defined positive in eq. 2 when parallel to the weak direct field) then the slope is larger than when the RRM is negative (antiparallel to the direct field). From the four slopes, the values of A in units of (μA m^2 kg^{-1}) μT^{-1} and k in units of (μT)$^{-1}$ for the two samples are

BAM8: $A = 2.73 \pm 0.31$ (clockwise) and $k = 410 \pm 170$ (anticlockwise) ,

BMR7: $A = 1.00 \pm 0.17$ (clockwise) and $k = 380 \pm 280$ (anticlockwise) .

From Fig. 5 it is possible to calculate values of B_g from the four slopes by dividing the magnitude of the RRM by the slope. This yields the direct field required to produce an ARM equal to the RRM (i.e. B_g). The results in μT are

BAM8: 466 ± 31 (clockwise) and 679 ± 79 (anticlockwise) ,

BMR7: 445 ± 27 (clockwise) and 713 ± 73 (anticlockwise) .

The highest values (anticlockwise) are lower than the values quoted previously from the results of Figs 3 and 4 (1050 and 1070 μT). The reason is that while the RRM for this new set of measurements has about the same value, the ARM is almost 50 per cent higher than before (and thus B_g is lower) when it is antiparallel to the RRM. The reason for this is unknown but since there was a gap of 3 yr between the data acquisition for Figs 3 and 4 and Fig. 5, it is possible that chemical change within the sample may be responsible. In spite of this quantitative difference there is qualitative agreement regarding the nature of the interaction in that in both sets of results the ARM is always smaller when it is acquired antiparallel to the RRM.

Eq. (2) was fitted by a least-squares method to the data of Figs 3 and 4. The line drawn through the ARM points is the result of this. The values of A in units of (μA m^2 kg^{-1}) μT^{-1} and k in units of (μT)$^{-1}$ are as follows:

BAM8: $A = 2.57 \pm 0.14$ (clockwise) and $k = 970 \pm 30$ (anticlockwise) ,

BMR7: $A = 0.91 \pm 0.09$ (clockwise) and $k = 1040 \pm 60$ (anticlockwise) .

Thus while eq. (2) is a good fit to the data, the constant k (the interaction constant) is, for these earlier results, higher than for the results of Fig. 5, indicating that in the latter case k may have decreased with time due to chemical change. (It should be noted that if the same equation were to be applied to the magnetite result of Fig. 2, the decrease of ARM above 50 rps would be about 1 Am^2 kg^{-1}, a value that is just too small to be detected.)

COMPARISON OF THE ACQUISITION OF RRM, ARM AND IRM IN THE SAME STRENGTH OF FIELD

Figs 6 and 7 show the variation of RRM and ARM with peak AF at 95 rps for the two samples. The RRM increases rapidly with increasing AF in a similar way to the results obtained by Snowball at 5 rps and has a higher coercivity in agreement with the observations of Mahon & Stephenson (1997).

Another way of appreciating the large gyromagnetic effect in greigite is to compare the RRM acquired in a given peak alternating field with the isothermal remanent magnetization (IRM) acquired in the same direct field. Figs 6 and 7 also show the result for the acquisition of IRM for the two samples. The IRM is far from saturation at 80 mT for both samples, showing that there is a considerable amount of high-coercivity material present. The acquisition curve is less steep than the acquisition of RRM, showing that the RRM is dominated by the highest-coercivity material, as expected (see Mahon & Stephenson 1997). The RRM at 80 mT peak AF for BAM8 and BMR7 is 8.8 and 9.2 per cent of the IRM, respectively. To appreciate the significance of this result consider the following explanation of AF demagnetization.

When an AF of 80 mT peak is applied to a static isotropic sample it acquires alternate positive and negative IRMs in each successive half-cycle of the field. Thus the amount of remanence that is being moved around by the field in successive half-cycles is the IRM acquired in one half-cycle of field. Neglecting viscous effects, this is essentially the same as the IRM acquired in a direct field of 80 mT. As the field slowly decreases, these IRMs of alternate sign gradually reduce and their resultant (that is, the sum of any positive and negative remanences left in the sample) ideally becomes zero after AF demagnetization is finished.

In the case of the acquisition of RRM in a peak field of 80 mT with sample rotation, the AF is similarly moving a remanence approximately equal to the IRM in 80 mT but the gyromagnetic effect operates preferentially on some of this remanence (mainly that carried by the higher-coercivity particles) in such a way that after the AF has reduced to zero, a remanence (i.e. an RRM) is left in the sample at right angles to the AF (i.e. along the rotation axis). If there were no gyromagnetic effect this resultant remanence would be zero.

Thus the IRM in 80 mT is a measure of the maximum amount of remanence that is available at room temperature at that field. In the case of the two greigite samples the gyromagnetic effect is apparently able to deflect about 9 per cent of this remanence out of the plane in which the field acts [presumably by switching the moments of those particles carrying the high-coercivity part of the IRM—see Mahon & Stephenson (1997)]. If the results are extrapolated to 100 mT, this percentage rises to something like 15 per cent and may be even higher at stronger fields. For comparison, an examination of previous results indicates that at 80 mT for 0.7 μm magnetite the same figure is 0.65 per cent and for the 2.2–4.4 μm magnetite sample used here it is 0.17 per cent.

CONCLUSIONS

Greigite exhibits a very large gyromagnetic effect as measured by the parameter B_g. This is about 1 mT and is about an order of magnitude higher than that typically observed for 1 μm magnetite or magnetic tape particles, and about 30 times as large as that of the 2.2–4.4 μm magnetite sample described here. These large values must be regarded as a lower limit for greigite because it is likely that the samples used may well have magnetite as a contaminant, which would depress the value of B_g. In the 2.2–4.4 μm magnetite sample, remeasured here for comparison, the rotational ARM is independent of rotation rate and is not influenced by the RRM that is simultaneously acquired, but in the greigite samples there seems to be an interaction between the ARM and the RRM. This is perhaps not so surprising in view of the observation that the gyromagnetic effect is capable of deflecting some 9 per cent of the maximum available remanence to 90° away from the AF axis.

To understand this behaviour would involve a detailed study of the fundamental physics of GRM acquisition in greigite. Meanwhile, it may be possible to use these effects to identify greigite, although it is possible that some other minerals may also turn out to exhibit large effects. To obtain the maximum likelihood of detecting greigite, the three-spin method of Stephenson & Molyneux should be used under ‘standard conditions’, that is, 95 rps, 80 mT peak AF and a known direct field for the rotational ARM of about 70 μT (although higher fields up to 200 μT could be used on the basis of the linearity of ARM with applied direct field of these samples). A condition previously used for all samples investigated to date but not explicitly specified (because of its unimportance for magnetite) is that the direct field should be aligned antiparallel to the rotation vector (and thus antiparallel to the RRM at this rotation speed). This gives the largest value for B_g for greigite as defined by eq. (1) because of the interaction effect.

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