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Experimental study of the origin and properties of the defect moment in single domain haematite

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Summary. The properties of the 'defect moment' of two fine-particle synthetic haematite powders were studied at low and room temperatures, in weak and moderate fields with respect to the saturation field of this mineral, and as a function of annealing. Two components were separated, (a) a component of low coercivity due to lattice defects, which disappears at the Morin transition, (b) a component with higher coercivities, due to non-magnetic impurities, which persists below the Morin transition and is unaffected by annealing.

1 Introduction

There have been many studies in the last 20 years on the magnetic properties of haematite, bearing witness to the lively interest in this mineral among physicists and geophysicists. Haematite often has magnetic properties which are variable and even contradictory, depending on whether the mineral is present in the form of a single crystal or as a powder. Therefore the interpretation of the results has been difficult and these difficulties have still not been completely overcome. Nevertheless, the development of palaeomagnetic studies of sedimentary rocks containing haematite necessitates a fundamental understanding of the behaviour of this mineral.

Haematite has a crystallographic structure of the corundum type; it crystallizes in the rhombohedal system. Magnetically, various studies have established the following characteristic properties, (a) an antiferromagnetic structure (Endo 1937), (b) a characteristic transition in the susceptibility (Morin 1950) in the neighbourhood of -15°C (Morin temperature, T_M), (c) a Néel point at 675°C (T_N) (which is the same as the Curie point).

Neutron diffraction studies (Shull, Strauser & Wollan 1951), have explained these observations; above the Morin transition, the spins are in the basal plane of the crystal, when the temperature goes below this transition point, the spins reorient themselves along the ternary axes.

Between -20 and $+675^{\circ}$C, in the absence of an external field, one can measure a weak ferromagnetism in the basal plane, superimposed on the antiferromagnetic structure (Townsend-Smith 1916; Chevallier & Mathieu 1943). Since the Curie point coincides with T_N and since the saturation remanence (σ_{rs}) also undergoes a transition at the Morin

temperature, it appears that this ferromagnetism is intimately connected with the antiferromagnetism. The theoretical explanation which seems to explain this phenomenon best was given by Dzyaloshinsky (1958), who proposed the existence of a spin-canting or imperfect antiparallelism of the elementary moments in the basal plane, giving rise to a resultant moment perpendicular to the antiferromagnetic axes. This moment is called the 'fundamental moment' (it is nothing more than the anisotropic moment of Néel 1953). Below the Morin transition, the spins lie precisely along the ternary axes in one sense or the other and haematite then becomes a perfect antiferromagnet, without any fundamental moment.

Although the theory just presented predicts that no magnetization should persist below T_M, experiment often shows the existence of a moment at low temperatures. This magnetization is isotropic and its intensity varies according to the shape of grains, method of preparation and the nature (powder or single crystals) of the haematites being studied. It corresponds to a 'defect moment' which can be reduced by annealing, changed by neutron irradiation (Gallon 1968) and can increase by quenching (Smith & Fuller 1967). The origin of this moment could be either, (a) in magnetic impurities; epitaxial layers of magnetite or maghemite (Néel 1949) or (b) in the moment of antiferromagnetic domain walls, which appear as result of lattice defects (Li 1956).

Before the publication of Dzyaloshinsky's theory, these hypotheses had been put forward as a means of explaining the remanent moment of haematite. In our present state of understanding, they could all (whether chemical or structural defects are involved) be the cause of the defect moment, but up until now it has not been possible definitively to choose one over the others.

As far as the relative stability of these two components is concerned, the results, which are often different depending on the material studied (natural or synthetic single crystals or synthetic powders), have led to the following conclusions:

(a) Haigh (1957) and Dunlop (1971), working with synthetic haematite powders, both at ordinary temperatures and below the Morin transition, were able to obtain separately the magnetization curves of the two components (Haigh 1957) or to follow the relative stability of these two components in the course of heatings which appeared progressively to destroy the defect moment (Dunlop 1971). These authors concluded that, (1) the fundamental moment is a high coercivity moment (H_{Cr} of the order of several thousands of Oe)*, (2) the defect moment is much softer.

(b) Smith & Fuller (1967) studying a natural haematite made up of an aggregate of single crystals, arrived at a completely different conclusion, namely the defect moment remaining at low temperatures is a hard moment (with coercivities approximately equal to 3000 Oe), with the result that the fundamental moment must have very low coercivities, perhaps as small as 100 Oe.

With the object of testing experimentally the validity of Dzyaloshinsky's theory in the region that is the subject of this study, Lecaille & Daly (1972) studied the basal plane anisotropy in fine-grained haematite containing single domain grains; they demonstrated the multiaxial character of the basal plane anisotropy, which reconciles the experimental facts with theory.

Our study has been carried out with the object of defining more precisely the properties and the origin of the defect moment carried by fine-grained haematite, adding to the experiments just described results obtained in a region still very little studied, namely, magnetizations acquired in fields which are weak or moderate with respect to the saturation field of haematite. We have studied the variation of induced magnetization, isothermal

*H_{Cr} = remanent coercive force.

remanent magnetization (IRM) and thermoremanent magnetization (TRM), as a function of temperature (between ≈ -80 and $+20^{\circ}$C); it is well known that heatings at high temperatures – from 900°C onwards – considerably modify the properties of haematite, very probably as a result of a decrease in the amount of impurities and the relaxation or annealing out of internal stress (Pozzi 1972). In particular, the magnetic properties at low temperature are changed by annealing. For this reason, we have studied the influence of annealing temperature on induced and remanent magnetizations. Likewise we have measured the hysteresis curve in a field of maximum intensity ≈ 20 kOe (or 2 Tesla) as a function of annealing, with a view to obtaining information about characteristics of the hysteresis curve and, as a result, further information about the origin and evolution of the defect moment.

2 Experimental conditions

2.1 APPARATUS

The measurements were carried out with a symmetrical astatic magnetometer (Pozzi 1967). Cylindrical samples, placed in a Dewar vessel located in the front of the magnetometer, were cooled in steps to $\approx -80^{\circ}$C by nitrogen gas, then reheated to ambient temperature. Two translations of the sample in front of the central magnet, one vertical, the other a rotation, allowed us to measure the remanent magnetization along the axis of the sample and perpendicular to it. The Dewar vessel and magnetometer were both located in zero field.

2.2 PREPARATION OF THE SAMPLES

The study was carried out on two synthetic haematites of different commercial origin and characteristics,

Mapico 110-2: cubical grains between 0.2–1.3 μm, obtained by oxidizing magnetite (Fe_3O_4);

Mapico R297: spherical grains of size 0.3–0.8 μm, obtained by decomposition of $FeSO_4, 7H_2O$.

Each powder served to make three samples; one sample contained unheated powder, a second contained a moderately annealed powder (820°C) designated MA and a third strongly annealed sample (990°C) designated SA. In order to obtain identical annealing conditions, in each case the two powders were heated at the same time, in air, the annealing temperature being maintained constant for 2 hr. Both samples were then allowed to cool slowly for 24 hr to room temperature.

3 Experimental results

3.1 AMBIENT TEMPERATURE MEASUREMENTS

The initial susceptibility of all six samples was measured in a field of 0.84 Oe (or 0.84×79.6 A/m). The values obtained are given in Table 1 (third column).

Hysteresis curves measured on the same samples in fields with a maximum intensity of 20 kOe (or 2 T) are given in Fig. 1 (R 297 – below, 110-2 – above). These curves were drawn after subtracting out the antiferromagnetic component of magnetization which – according to Néel & Pauthenet (1952) and Creer (1967) – is $20.4\pi \times 10^{-9}$ SI units over the range -196 to 680°.

These curves demonstrate the existence of an important ferromagnetism with coercivities which increase as a function of annealing, thus providing evidence for a 'hardening' of the ferromagnetic moment with annealing temperature, in accord with the findings of Dunlop (1971).

Table 1. Initial susceptibility and characteristic hysteresis parameters (ambient temperature measurements) for the three samples of each haematite (unheated powder, (MA) powder and (SA) powder).

Haematite	$T°C$	χ_i (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	H_C (Oe or 10^{-1} mT)	σ_{rs} (10^{-3} emu/g or 10^{-3} Am^2/kg)	σ_s (10^{-3} emu/g or 10^{-3} Am^2/kg)	σ_{rs}/σ_s	Remarks
	–	56.3	2000	154	250	0.62	unheated powder
R 297	820	50.7	2400	150	235	0.68	
	990	40.0	5200	194	240	0.81	
	–	217.0	650	200	410	0.49	unheated powder
110-2	820	60.5	2000	140	220	0.65	
	990	40.5	4800	200	235	0.85	

Notes:

$T°C$ (annealing temperature), χ_i (initial susceptibility), H_C (coercive force), σ_{rs} (saturation remanence), σ_s (saturation magnetization).

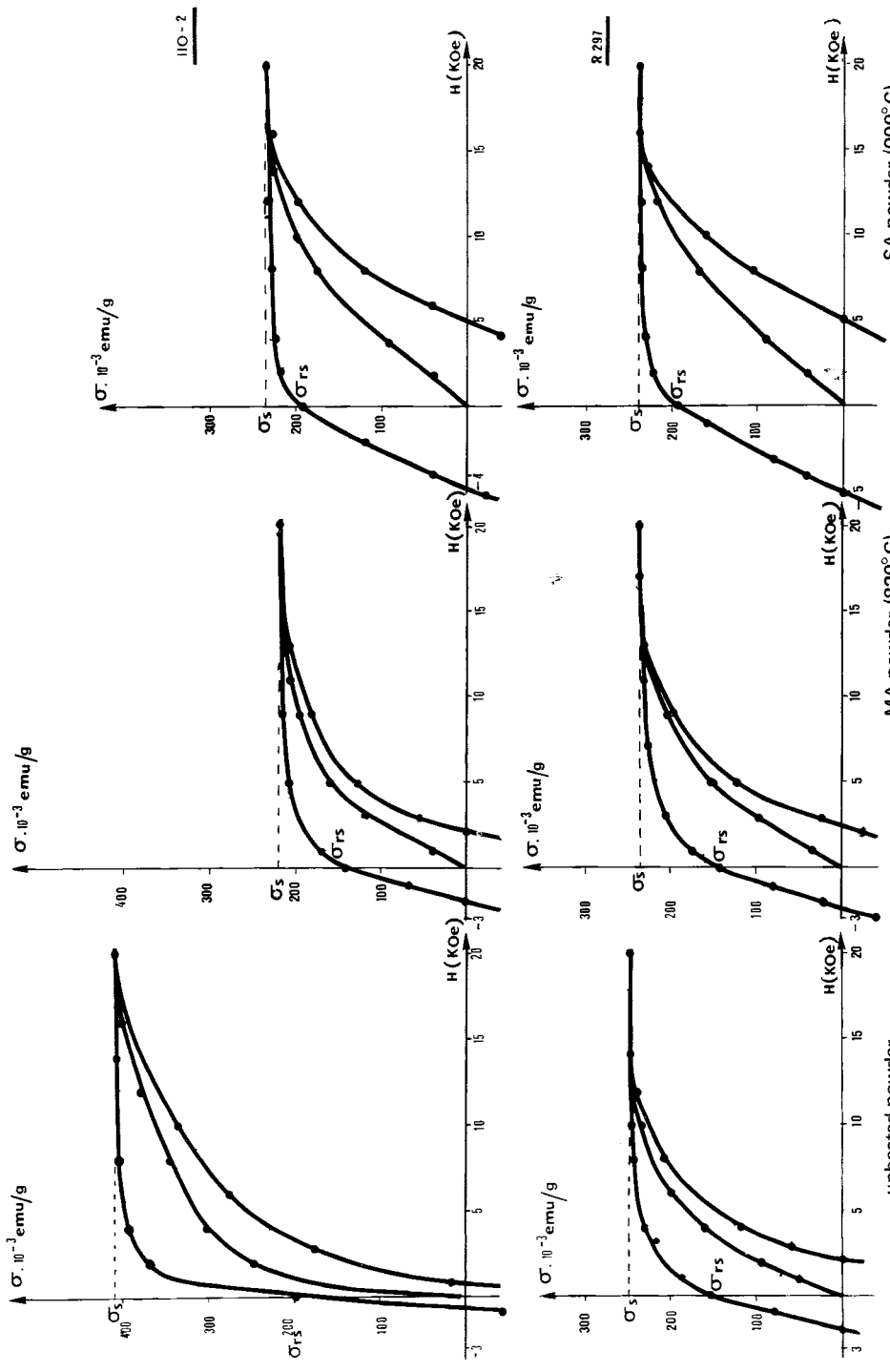

Figure 1. Hysteresis curves measured on the three samples of each haematite (unheated powder, 820°C annealed powder (MA),and 990°C annealed powder (SA)); R 297 – below, 110-2 – above.

Table 1 also contains the characteristic hysteresis parameters (H_C, σ_{rs}, σ_s), as well as the ratio σ_{rs}/σ_s which, according to Wohlfarth & Tonge (1957), is indicative of the type of anisotropy (uniaxial or multiaxial) in the basal plane. In fact, for:

(a) a field H sufficiently intense to align all the spins in the basal plane, without rotating spins out of this plane, these authors predict the ratios

$\sigma_{rs}/\sigma_s = 0.5$ for uniaxial anisotropy (magnetoelastic)
$\sigma_{rs}/\sigma_s = 0.750$ for multiaxial anisotropy (magnetocrystalline),

(b) a field H sufficiently strong to pull the spins out of the basal plane and into alignment with the field

$\sigma_{rs}/\sigma_s = 0.637$ for uniaxial anisotropy (magnetoelastic)
$\sigma_{rs}/\sigma_s = 0.955$ for multiaxial anisotropy (magnetocrystalline).

Since the field used in these measurements was 20 kOe (or 2 T), the measured magnetization ratios are probably closer to the second case.

3.1.1 Effect of annealing at 820°C

The values given in Table 1 allow the following observations:

Sample R 297. (a) There is practically no variation in σ_s and σ_{rs}, (b) the other two parameters (H_C and χ_i) also change very little.

Thus, overall, heating to 820°C has not had any important effect. If a defect moment exists, it has not been changed.

Sample 110-2. σ_s decreases to almost the half of its original value, demonstrating the disappearance of the defect magnetization.

By comparison of measurements before and after heating (refer to the values of H_C, χ_i, σ_{rs}), it appears that this sample contains a highly magnetic impurity, i.e. a defect moment of chemical origin which is soft and characterized by a spontaneous magnetization and a susceptibility much stronger than that of the fundamental moment in haematite. It could be due to maghemite whose anisotropy, if due to shape, is uniaxial ($\sigma_{rs}/\sigma_s = 0.5$). Theoretically, this impurity should be pseudo-single domain, since this value in reality is < 0.5.

3.1.2 Effect of annealing at 990°C

Since the anneal at this temperature does not merely follow the previous anneal at 820°C but, rather, is a first anneal of previously unheated powders at this high temperature, we will compare lines 1 and 3 in Table 1 for each of the samples.

Sample R 297. For this sample, which was practically unaffected by a 820°C anneal, the constancy of σ_s demonstrates that we are not seeing here a variation in a defect moment of chemical origin. On the other hand, the increase in σ_{rs}/σ_s, as well as the decrease in χ_i, indicates that a defect moment with uniaxial anisotropy, whose origin is perhaps in lattice defects, has disappeared; as a result, the underlying multiaxial crystalline anisotropy becomes more prominent and thus the haematite behaves as if the number of anisotropy axes had increased.

Sample 110-2. The same reasoning applied to the 820°C anneal remains valid for the 990°C anneal.

Changes between 820 and 990°C. On the other hand, if one concentrates on the changes between 820 and 990°C (lines 2 and 3 in Table 1 for the case of both haematites), the situation is quite different. Both haematites behave in the same way. There is (a) a decrease

in χ_i, (b) the coercive force H_C doubles, (c) σ_{rs}/σ_s increases, (d) σ_s remains essentially constant.

One can draw the following conclusions from these facts (a) a defect moment whose origin must lie in lattice defects has decreased (χ_i decreases and H_C doubles), (b) this defect moment cannot be of magnetic origin because σ_s remains essentially constant and (c) it must have uniaxial anisotropy (the 990°C annealed sample has a larger value of σ_{rs}/σ_s).

As a result of its origin, this defect moment cannot be completely separated from the fundamental moment; the two moments appeared to be initimately linked. As a result, one can suppose that the large increase in H_C is a result of a variation in the fundamental moment itself, resulting from a change in crystal structure. Thus, as a result of measurements made at ambient temperature, it appears that

(1) if a defect magnetization of chemical origin is present, it can be removed by heating to moderate temperatures,

(2) a defect magnetization whose origin is related to lattice defects (present in both cases studied here) can be separated into two components, (a) a component which decreases as the annealing temperature increases and (b) a component which survives heatings (constancy of σ_s for R 297 (20–820–990°C) and for 110-2 (820–990°C)).

In order to achieve a more precise interpretation of the results obtained, measurements at low temperatures were obviously necessary.

3.2 LOW-TEMPERATURE MEASUREMENTS

3.2.1 Weak-field susceptibility

The susceptibilities χ_i of the heated powders, given in the preceding section, as well as susceptibilities χ_{lt} (at ≈ -80°C), both measured in a field of 0.84 Oe (or 0.84 × 79.6 A/m), are given in Table 2. In the same table we give the values of the initial susceptibility due to the defect moment ($\chi_{d(i)}$, the effective total contribution of a weak ferromagnetism, i.e. canted + defect moment), calculated by subtracting from χ_i the fundamental antiferromagnetic susceptibility at room temperature, which is $20.4\pi \times 10^{-9}$ SI units or 20×10^{-6} emu/g. For micronsized grains, Lecaille & Daly (1972) have shown that the anisotropy of susceptibility is very slight; later, Lecaille (1977, private communication) has shown for the same haematites that the fundamental susceptibility is very little different from 20×10^{-6} emu/g.

In Table 2 (column 6) is also given the low-temperature susceptibility due to the defect moment ($\chi_{d(lt)}$), calculated after having subtracted out from χ_{lt} the value of the fundamental antiferromagnetic susceptibility at low temperatures. At these temperatures, the antiferromagnetic direction is along the ternary axis and is no longer canted; the susceptibility is thus zero along this axis but remains at 20×10^{-6} emu/g along two perpendicular directions in the basal plane. If one supposes that the ensemble of all crystalline easy axes of the grains represents a uniform distribution, the fundamental susceptibility at low temperature has the value 2/3 of $20.4\pi \times 10^{-9}$ SI units (or 20×10^{-6} emu/g) or 13.3 in these units. Thus $\chi_{d(lt)} = \chi_{lt} - 13.3$. In the seventh column of Table 2, we give the difference $\chi_{d(i)} - \chi_{d(lt)}$ which represents the susceptibility due to that part of the weak ferromagnetic moment which disappears below the Morin transition, i.e. the spin canted part.

In the preceding section, we remarked that after the 820°C anneal, just as after the 990°C anneal, the values of initial susceptibility indicate the existence of a defect moment (if one is in agreement with the calculated values for the susceptibility of the fundamental moment).

Table 2. Susceptibilities at low temperature for the two annealed powders (MA and SA) of each haematite.

Haematite	T°C	χ_i (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	$\chi_{d(i)}$ (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	χ_{lt} (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	$\chi_{d(lt)}$ (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	$\chi_{d(i)} - \chi_{d(lt)}$ (10^{-6} emu/g or $4\pi \times 10^{-9}$ SI)	$\chi_{d(lt)}/\chi_{d(i)}$
R 297	820	50.7	30.7	23.8	10.5	20.2	0.34
	990	40.0	20.0	20.8	7.5	12.5	0.38
110-2	820	60.5	40.5	27.6	14.3	26.2	0.35
	990	40.5	20.5	21.6	8.3	12.2	0.40

Notes:

T°C (annealing temperature), χ_i (initial susceptibility), $\chi_{d(i)}$ (initial susceptibility due to the defect moment), χ_{lt} (overall low-temperature susceptibility), $\chi_{d(lt)}$ (low-temperature susceptibility due to the defect moment).

Table 3. Numerical values of the characteristic parameters obtained by measuring the evolution of the remanent magnetization during one cooling–heating cycle.

Haematite	Remanent magnetization	h (Oe or 10^{-1} mT)	σ_{ri} (10^{-4} emu/g or 10^{-4} Am^2/kg)		σ_{lt} (10^{-4} emu/g or 10^{-4} Am^2/kg)		σ_{rl} (10^{-4} emu/g or 10^{-4} Am^2/kg)		σ_{lt}/σ_{rl}	
			MA	SA	MA	SA	MA	SA	MA	SA
R 297	IRM	100	1.57	1.71	0.16	0.15	0.31	0.24	0.52	0.63
		400	10.23	15.45	1.43	1.24	3.27	2.47	0.44	0.50
		1000	45.30	55.43	4.08	3.88	11.30	9.42	0.36	0.41
	TRM	0.42	18.8	263.0	1.32	10.52	9.78	129.0	0.13	0.08
		0.84	36.1	492.0	2.53	9.84	18.80	226.0	0.13	0.04
110-2	IRM	100	1.51	1.10	0.14	0.18	0.33	0.19	0.42	1.00
		400	15.7	12.7	0.63	0.69	2.20	0.69	0.29	1.00
		1000	75.9	53.8	1.52	2.15	13.3	2.98	0.11	0.72
	TRM	0.42	37.3	328.5	1.12	13.12	15.29	154.0	0.07	0.09
		0.84	76.8	570.0	5.38	28.5	33.8	273.5	0.16	0.10

Notes:

h (field in which the initial remanent magnetization was produced), σ_{ri} (initial remanent magnetization), σ_{lt} (low-temperature remanent magnetization), σ_{rl} (memorized remanent magnetization after the first cooling–heating cycle).

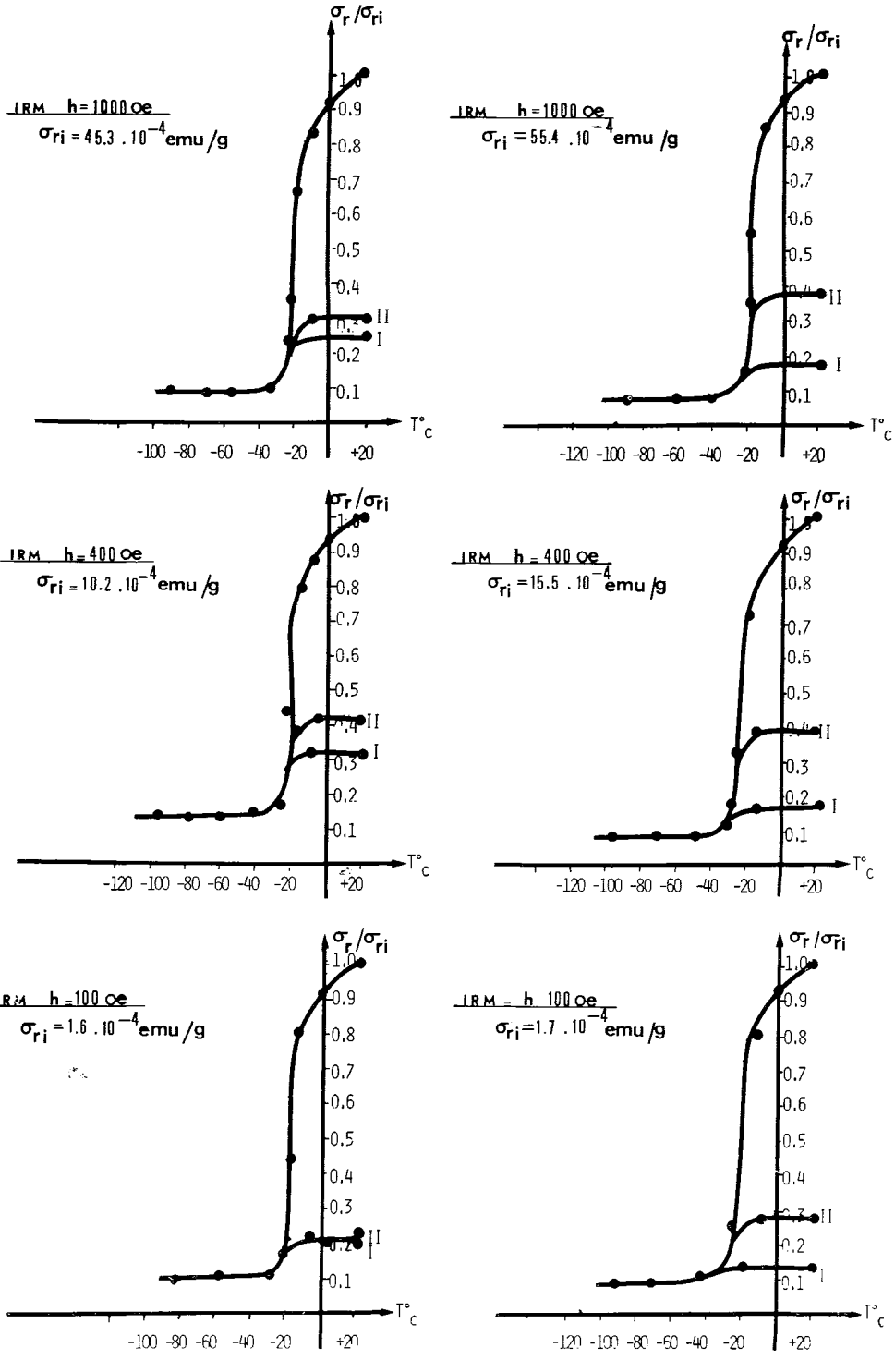

Figure 2. Variation of the IRM as a function of temperature, for haematite R297.

According to the values in Table 2, it would appear that this defect moment is composed of two parts, (a) a component which disappears below T_M and which, surprisingly, varies as a function of annealing (see the values of $\chi_{d(i)} - \chi_{d(lt)}$; (b) a component which does not undergo the Morin transition and remains unaffected by annealing (see the values of $\chi_{d(lt)}/\chi_{d(i)}$ which are essentially the same for the two haematites).

These conclusions will now be tested by studying the evolution with temperature of the various types of remanent magnetization currently used in palaeomagnetism.

3.2.2 Isothermal remanent magnetization (IRM) and thermoremanent magnetization (TRM) in weak fields; variation of the ferromagnetic moment in the course of a temperature cycle

The evolution of the remanent intensity for the two annealed samples of each haematite was measured in the region of the Morin transition, for (a) IRMs acquired at ordinary temperatures in fields of 100, 400 and 1000 Oe (or 10, 40 and 100 mT) and (b) total TRMs acquired by cooling in fields of 0.42 and 0.84 Oe (or 0.042 and 0.084 mT).

The form of the curves was the same for the two powders of different origin, thus we shall present only the results for R 297, the numerical results being contained in Table 3. The variation of remanent magnetization σ_r, normalized to the initial remanent magnetization σ_{ri} before cooling, as a function of temperature, is given in Fig. 2 (IRM) and Fig. 3 (TRM). Each curve is drawn after measurements on two successive cooling–heating cycles; with a view to facilitating the observation of the effects of annealing, the results of the same experiment, carried out on the two specimens annealed at different temperatures (MA and SA), are compared: to the left, specimen MA, to the right, specimen SA.

One observes well-known phenomena characteristic of haematite at low temperatures, after several successive thermal cycles; decrease of the magnetization after reheating to ambient temperature, with a memory of the initial direction but a decreased intensity (Morin 1950; Haigh 1957), cyclic variation of the intensity of magnetization recovered – between two values both weaker than the initial value – (zig-zag effect) in the course of successive thermal cycles (the intensities recovered after an odd number of thermal cycles being weaker than intensities recovered after an even number of cycles (Nagata, Yama-Ai & Akimoto 1961)).

Furthermore, one notices,

(1) a characteristic property which is independent of the nature of the haematite studied, of the type of magnetization and of annealing; namely, at low temperatures there remains a remanent magnetization (σ_{lt});

In the case of IRM:

(2) there is a different variation of the initial remanent magnetization σ_{ri} depending on previous annealing history, for both haematites,

(3) likewise for each value of h (the field in which the initial remanent magnetization was produced), the values of σ_{lt} are very similar, independent of annealing,

(4) σ_{rI} decreases as a result of annealing, both for R 297 and 110-2,

(5) the contribution of σ_{lt} to the values of σ_{rI} is more important for weak inducing fields (see the ratio σ_{lt}/σ_{rI}).

In the case of TRM, we observe:

(6) a considerable increase in σ_{ri} as result of annealing, for both haematites studied,

(7) a very pronounced increase in σ_{rI} between moderate- and the high-temperature anneals,

(8) σ_{lt} is very small compared to σ_{rI}.

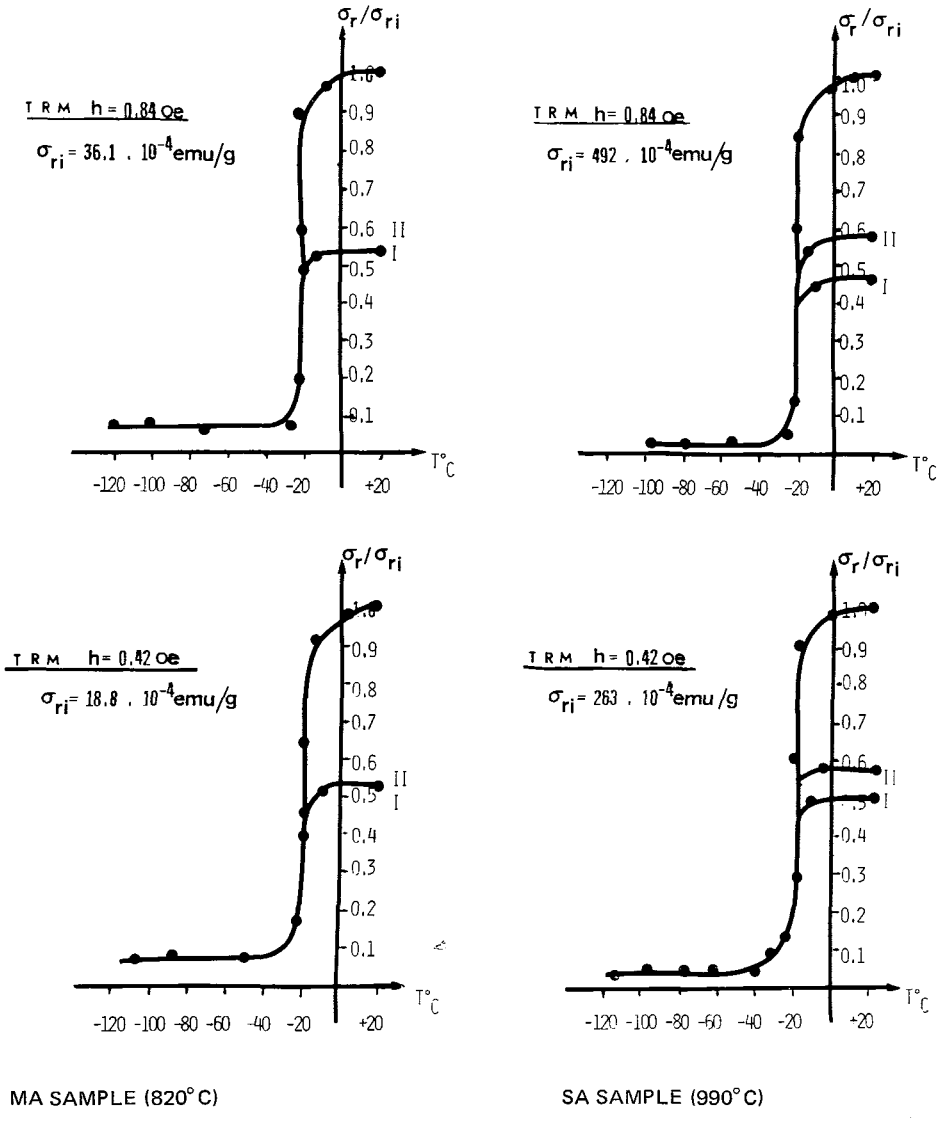

Figure 3. Variation of the TRM as a function of temperature, for haematite R297.

The analysis of these observations must be made in the light of results obtained in the previous section, namely the origin of the defect moment in haematite R 297 is not the same as that of the defect moment in haematite 110-2. We have concluded that for 110-2 the defect moment is due in large part to an impurity (probably maghemite) which disappears in the course of annealing, whereas the defect moment of haematite R 297 is due principally to defects in the crystal lattice. These conclusions explain very well, in the case of the IRM, the variation in initial remanent magnetization σ_{ri} as result of annealing.

Also well explained for R 297 is the fact that σ_{ri} increases as a result of annealing. If the defect magnetization is due to internal stress, which makes the orientation of the elementary moments more or less difficult, this variation is to be expected since the constraints within the crystal relax as the annealing temperature increases.

Also explained for 110-2 is the fact that σ_{ri} decreases with annealing. This observation is to be expected if the defect magnetization is due to maghemite which disappears during annealing.

In the case of TRM, the explanation of the variation of σ_{ri} with annealing is less easy to see; we know that the grains involved in the acquisition of this remanence must normally exhibit a range of blocking temperatures and thus a range of relaxation times. The large increase in σ_{ri} between anneals at moderate and high temperatures ($\approx$ 14-fold for R 297 and $\approx$ 8-fold for 110-2), leads one to believe that the effect of such a heating is expressed by an increase in relaxation times of the grains. A number of hypotheses could be suggested but the weak-field experiments with which we have been concerned do not allow us to choose among the hypotheses.

The fact that a magnetization remains at low temperature, independent of the nature of the haematite, the type of magnetization or of annealing, as well as the fact that the values of σ_{lt} are practically the same in the case of each IRM for both anneals, confirms the conclusions of a previous section (3.2.1) that there remains below T_M a particular sort of defect moment which is, *so far as its intensity is concerned,* unaffected by annealing. As far as the behaviour of this magnetization during the second part of a thermal cycle is concerned (from -80 to $+20^\circ$C) as well as its contribution to the overall memory (σ_{rI}), let us recall that Iwata, Iwata & Yamamoto (1962) and Smith & Fuller (1967), as a result of strong field studies, concluded that σ_{rI} is a very stable magnetization carried by high-coercivity grains. As σ_{rI} is closely linked to σ_{lt}, they concluded that that part of the defect moment which does not undergo the Morin transition is a 'hard' moment and that the fundamental moment which disappears below -20°C is a 'soft' moment. Iwata *et al.* (1962) did not state any precise conclusion with regard to the magnetic behaviour of these two moments, but did suggest that the magnetization due to the defect moment remaining at low temperatures controls the memory recovered after reheating to ordinary temperature, rather in the manner of a nucleus which remembers the original orientation of the fundamental moment.

For the interpretation of our measured results (4) and (5), we are going to recall the other conclusion of Section 3.2.1; the component of the defect moment which varies with annealing disappears below the Morin transition. One can suppose that the magnetization σ_{rI} is composed of three components;

$$\sigma_{rI} = \sigma_{lt} + \sigma_f + \sigma'$$

where σ_{lt} is the defect magnetization which is unaffected by annealing and which remains at low temperatures, σ_f is the magnetization of the fundamental moment which is remembered, σ' is the defect magnetization which is memorized, this last component varies with annealing and disappears in crossing the Morin transition.

The decrease in the values of σ_{rI} with annealing, as well as the more important contribution of σ_{lt} to the values of σ_{rI} in the case of weak field IRM, could indicate two conclusions;

(a) σ_{lt} is probably a medium-coercivity magnetization,

(b) σ_{rI} is probably a high-coercivity magnetization (on this point, our results agree with those of Smith & Fuller), the coercivities of σ_{rI} are higher than those of the initial remanence.

If these last conclusions can be confirmed, one can make the following hypothesis,

(a) σ' is a magnetization carried by grains with low coercive forces which, as a result, carry a small fraction of the magnetic memory; since the memory decreases with increasing annealing temperature, the variation of σ_{rI} with annealing is also explainable,

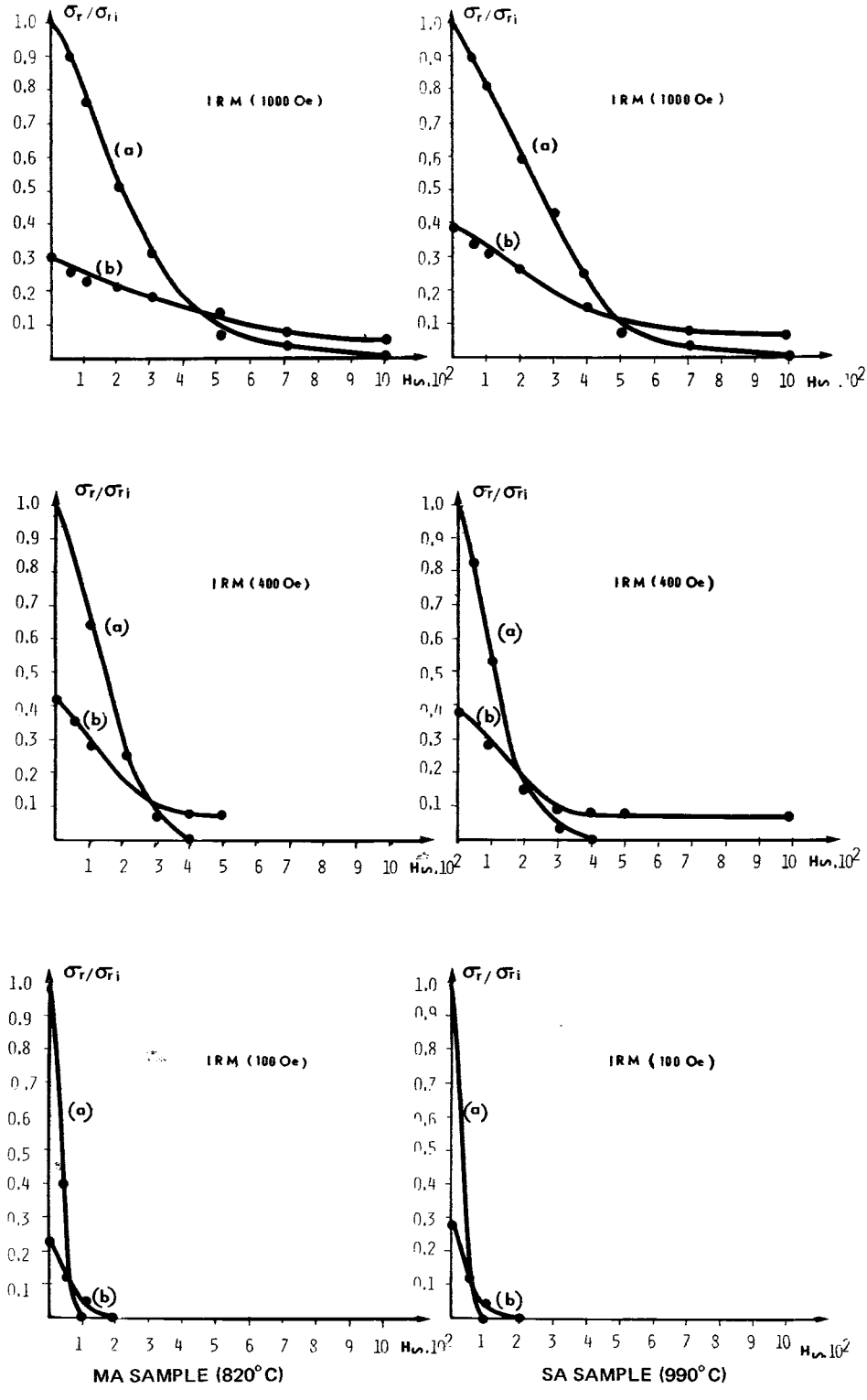

Figure 4. Alternating-field demagnetization curves of: (a) the initial IRMs and of (b) the memorized IRMs after a second cooling below the Morin transition, for haematite R297.

(b) σ_f is due to the fraction of the fundamental magnetization with the highest coercivities. Thus, overall, σ_{rI} would appear as a magnetization with coercivities higher than those of the initial remanence.

For haematite 110-2, although the variation of σ_{rI} with annealing is the same as that of R 297, the values obtained after annealing at 990°C are rather curious: it would seem that the fundamental moment has not kept a magnetic memory and that σ_{rI} contains only a single component, namely the defect moment remaining at low temperatures.

The experiments we have made do not permit us to give an explanation of the results (7) and (8) concerning the contribution of σ_{lt} to σ_{rI} and the variation of σ_{rI} as a function of annealing in the case of TRM. Further experiments on a pure haematite which has no defect magnetization would be necessary.

In order to verify the above hypothesis about the coercivities of the magnetic memory, we have studied the behaviour during alternating field demagnetization of the IRM memory.

3.2.3 Resistance to alternating-field demagnetization of the memory component of IRM

Alternating-field demagnetization curves of IRMs measured at room temperature after a second cooling below the Morin transition are shown in Fig. 4(b), together with those of the initial IRMs (a) (normalized with respect to the initial IRM).

Although initial IRMs are all completely destroyed by alternating fields of the same order as the field which produced the IRM, the fraction of the magnetization which survives a second thermal cycle is more resistant; fields larger than those which created the initial IRM are necessary completely to demagnetize the IRM memory. The annealing at 990°C does not seem to produce any detectable difference in these values, although, according to the results obtained in Section 3.1.1, the coercive force of the sample has practically doubled.

4 Conclusions

The results of the experiments carried out on synthetic powders of fine-grained haematites, in fields which are weak and moderate with respect to the saturation field, at low temperatures and at room temperature, as well as at different annealing temperatures, lead to the following observations: in the case of IRM;

(1) The defect moment of haematite appears in different forms which may coexist; (a) a magnetization with low coercivities which decreases with increasing annealing temperature and disappears at the Morin transition, its origin is likely related to lattice defects, (b) a magnetization with somewhat higher coercivities which is unaffected by heating and remains also at low temperatures. This magnetization cannot be attributed to lattice defects since it does not vary with annealing; nor can it be attributed to magnetic impurities (σ_s remains constant, unaffected by annealing). It could be due to non-magnetic impurities but the mechanism by which these impurities affect the magnetization remains to be explained.

(2) Heating haematite powders carrying this annealing independent defect magnetization to a higher temperature scarcely changes the saturation magnetization but it does increase the coercivity and the number of anisotropy axes as well as decreasing the susceptibility. The magnetocrystalline anisotropy becomes more and more important with respect to the magnetoelastic anisotropy ($\sigma_{rs}/\sigma_s > 0.8$), an observation which agrees with the conclusions of Dunlop (1971). It is possible that these changes can be attributed to the defect magnetization, but this is difficult to demonstrate.

(3) The remanent magnetization which is remembered after cooling and heating through the Morin transition is more resistant to alternating fields than the initial magnetization. However, this resistance does not increase with the annealing temperature, unlike the coercive force of the sample. This later observation could be explained by the fact that the resistance of an IRM to alternating-field demagnetization depends only on the strength of the field applied to the sample and not on the overall coercivity of the sample.

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