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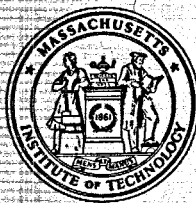
# POLARIZED NEUTRON STUDIES ON ANTIFERROMAGNETIC SINGLE CRYSTALS

R. NATHANS  
T. RISTE  
G. SHIRANE  
C. G. SHULL

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SINGLE CRYSTALS

- R. Nathans, Pennsylvania State University,  
University Park, Pennsylvania and  
Brookhaven National Laboratory, Upton, N. Y.
- T. Riste, Brookhaven National Laboratory, Upton,  
New York and Joint Establishment for Nuclear  
Energy, Kjeller, Norway
- G. Shirane, Brookhaven National Laboratory, Upton,  
New York
- C. G. Shull, Massachusetts Institute of Technology,  
Cambridge, Massachusetts

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- R. Nathans, Pennsylvania State University, University  
Park, Pennsylvania and Brookhaven National  
Laboratory, Upton, New York
- T. Riste, Brookhaven National Laboratory, Upton, New  
York and Joint Establishment for Nuclear Energy,  
Kjeller, Norway
- G. Shirane, <sup>†</sup> Brookhaven National Laboratory, Upton,  
New York
- C. G. Shull, Massachusetts Institute of Technology,  
Cambridge, Massachusetts

ABSTRACT

The theory of neutron scattering by magnetic crystals as given by Halpern and Johnson predicts changes in the polarization state of the neutron beam upon scattering which depend upon the relative orientation of the neutron polarization vector and the crystal magnetic axis. This has been investigated experimentally with a polarized beam spectrometer using single crystals of  $\text{Cr}_2\text{O}_3$  and  $\alpha\text{-Fe}_2\text{O}_3$  in which reside unique antiferromagnetic axes. Studies were made on several different reflections in both crystals for a number of different temperatures both below and above the Neel point. Our results support the theoretical predictions and indicate directions for the moments in these crystals consistent with previous work. In addition to the above, a more detailed study of the polarization changes in the (111) reflection in  $\alpha\text{-Fe}_2\text{O}_3$  at room temperature on application of a magnetic field has also been carried out. The results indicate that the principal source of the parasitic ferromagnetism in hematite is essentially independent of the orientation of the antiferromagnetic domains within the crystal.

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<sup>†</sup> Now at Westinghouse Research Laboratories, Pittsburgh, Penna.

## INTRODUCTION

The theory of neutron scattering by magnetic materials has been considered in great detail by Halpern and Johnson<sup>(1)</sup>. In this general treatment, the authors have derived expressions for both the polarization and the intensity of the scattered neutrons. In this paper we wish to deal primarily with those predictions made by Halpern and Johnson (H-J) for the changes in state of polarization of the incident neutrons arising from elastic magnetic scattering. These polarization state changes, according to H-J, will depend upon the relative orientation of the neutron polarization vector and the crystal magnetic axis.

In order to verify this predicted behavior we have selected single crystals of  $\text{Cr}_2\text{O}_3$  and  $\alpha\text{-Fe}_2\text{O}_3$ , both of which are rhombohedral and have been shown to possess unique antiferromagnetic axes with respect to their crystallographic directions<sup>(2, 3)</sup>. Since these crystals have no net magnetization we can vary the orientation between the neutron polarization vector and the magnetic spin axis without altering the directions of the magnetic fields present in the vicinity of the crystal, unlike the ferromagnetic crystal case, thereby avoiding any depolarization of the incident beam. The first part of this paper deals with the experiments on  $\text{Cr}_2\text{O}_3$  which have been used to confirm the Halpern and Johnson theory.

Since the change in the neutron polarization state depends on the angle between the polarization vector and the magnetic spin axis in the crystal, once the H-J predictions are confirmed, it is then possible to use this method for the determination of the absolute orientation of the spins within these single crystals. It should also be noted that this method offers a method of distinguishing between nuclear and magnetic scattering reflections since no change in neutron polarization state is to be expected upon coherent nuclear scattering. Finally, observations of the polarization of the reflected neutron beam can also be correlated with the behavior of the magnetic spin axis on the application of a magnetic field and such studies have been carried out on the room temperature phase of  $\alpha\text{-Fe}_2\text{O}_3$  and the results are discussed below.

The general expression for the change in the state of polarization of a neutron beam polarized along the direction  $\underline{\lambda}$  and analyzed after Bragg reflection along a direction  $\underline{\lambda}'$  as given by H-J is

$$D_{\lambda'} = \frac{(b^2 - p^2 q^2) \underline{\lambda} \cdot \underline{\lambda}' + 2bp \underline{\lambda}' \cdot \underline{q} + 2p^2 (\underline{\lambda} \cdot \underline{q}) (\underline{\lambda}' \cdot \underline{q})}{b^2 + p^2 q^2 + 2bp (\underline{q} \cdot \underline{\lambda})} \quad (1)$$

where  $b$  and  $p$  are the nuclear and magnetic structure factors for the particular (hkl) reflection, and  $\underline{q}$  is defined as

$$\underline{q} = \underline{e} (\underline{e} \cdot \underline{\kappa}) - \underline{\kappa}$$

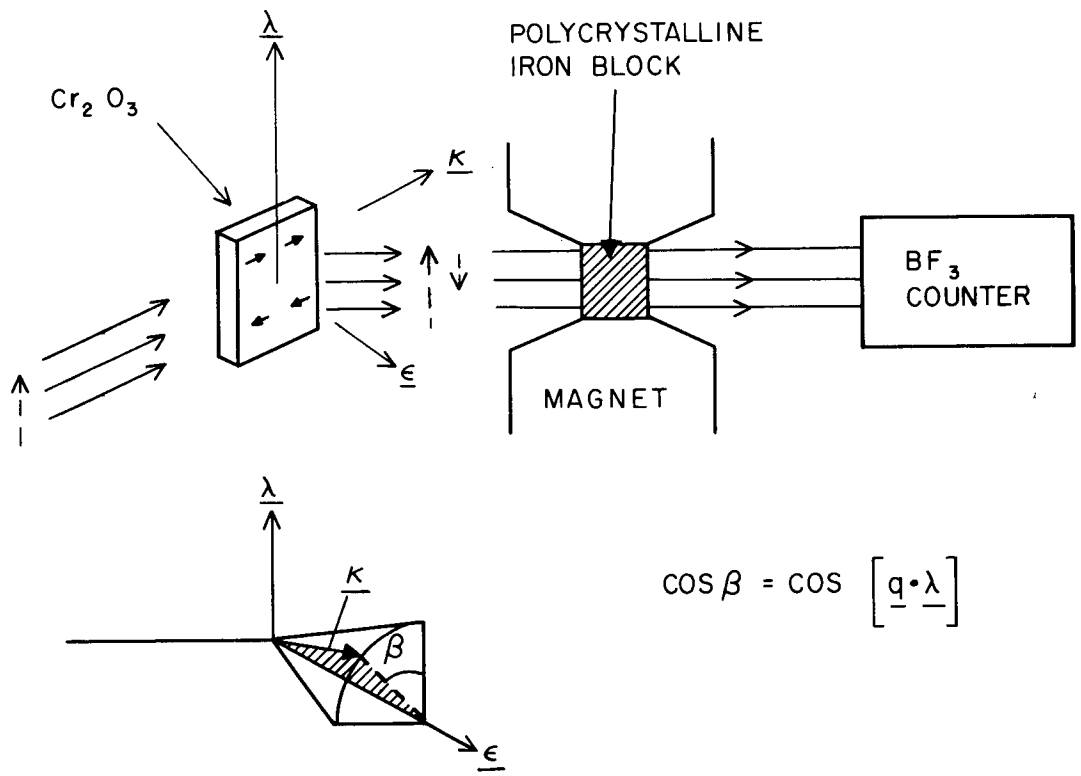


Fig. 1 - Schematic diagram of the experimental arrangement used in determining the neutron polarization after crystal scattering.

where  $\underline{e}$  is the unit scattering vector and  $\underline{\kappa}$  is a unit vector along the crystal magnetic axis. For the experimental arrangement shown in Fig. 1,  $\underline{\lambda} = \underline{\lambda}'$  and  $\underline{e} \cdot \underline{\lambda} = 0$ . Furthermore, since the crystals are antiferromagnetic the interference term  $2bp(\underline{\lambda}' \cdot \underline{q})$  goes to zero when averaged over the different atoms which contribute to the coherent reflection. The final form of the H-J equation therefore reduces to

$$D = \frac{b^2 - p^2 q^2 + 2p^2 q^2 \cos^2 \beta}{b^2 + p^2 q^2} \quad (2)$$

where  $\beta$  is the angle between  $\underline{q}$  and  $\underline{\lambda}$ . Our experiments were designed to verify Equation (2) with  $q^2$  being kept constant and with  $\beta$  varying between 0 and  $\pi/2$ . We therefore will expect that D will follow a cosine-squared dependence on  $\beta$ , going from  $D = 1$  or no change in polarization, at  $\beta = 0$  to a value of  $D = (b^2 - p^2) / (b^2 + p^2 q^2)$  at  $\beta = \pi/2$ . For a purely magnetic reflection  $D(\pi/2) = -1$  and the incident beam should undergo complete reversal in spin direction.

#### EXPERIMENTAL ARRANGEMENT

The apparatus used in these experiments consists of a standard neutron diffraction spectrometer modified to produce a beam of nearly completely polarized and monochromatic neutrons. A schematic diagram of the experimental arrangement is shown in Fig. 1. The polarized beam spectrometer has been described elsewhere<sup>(4)</sup> and it utilizes a single crystal of face-centered  $\text{Co}_{.92}\text{Fe}_{.08}$  as a polarizing monochromator. The neutrons once polarized are kept in a small controlled magnetic field throughout their entire path from the monochromator to the counter. The polarization in the scattered beam was established in most cases by measurement of the transmission through a magnetized polycrystalline block of iron. The iron block was approximately 1" thick and was magnetized in an electromagnet to a field of 7500 oersteds. On some of the later measurements on hematite, a Co-Fe crystal similar to the polarizing crystal was set to reflect neutrons scattered by the  $\alpha\text{-Fe}_2\text{O}_3$  crystal. The use of a third crystal with a high analyzing efficiency produces increased sensitivity in the polarization evaluation of the reflected beam, although the final observed intensities are smaller than that transmitted through the polycrystalline iron. The procedure was to measure the intensity for an initially polarized and then depolarized neutron beam for successive values of  $\beta$  thereby determining the shim ratio S. The resulting ratio expressed in terms of  $P_i$ , the polarization in the incident beam,  $P_x$  the analyzing efficiency of the magnetized block and D, the change in the polarization state produced on scattering is given by

$$S = 1 + P_i D P_x .$$



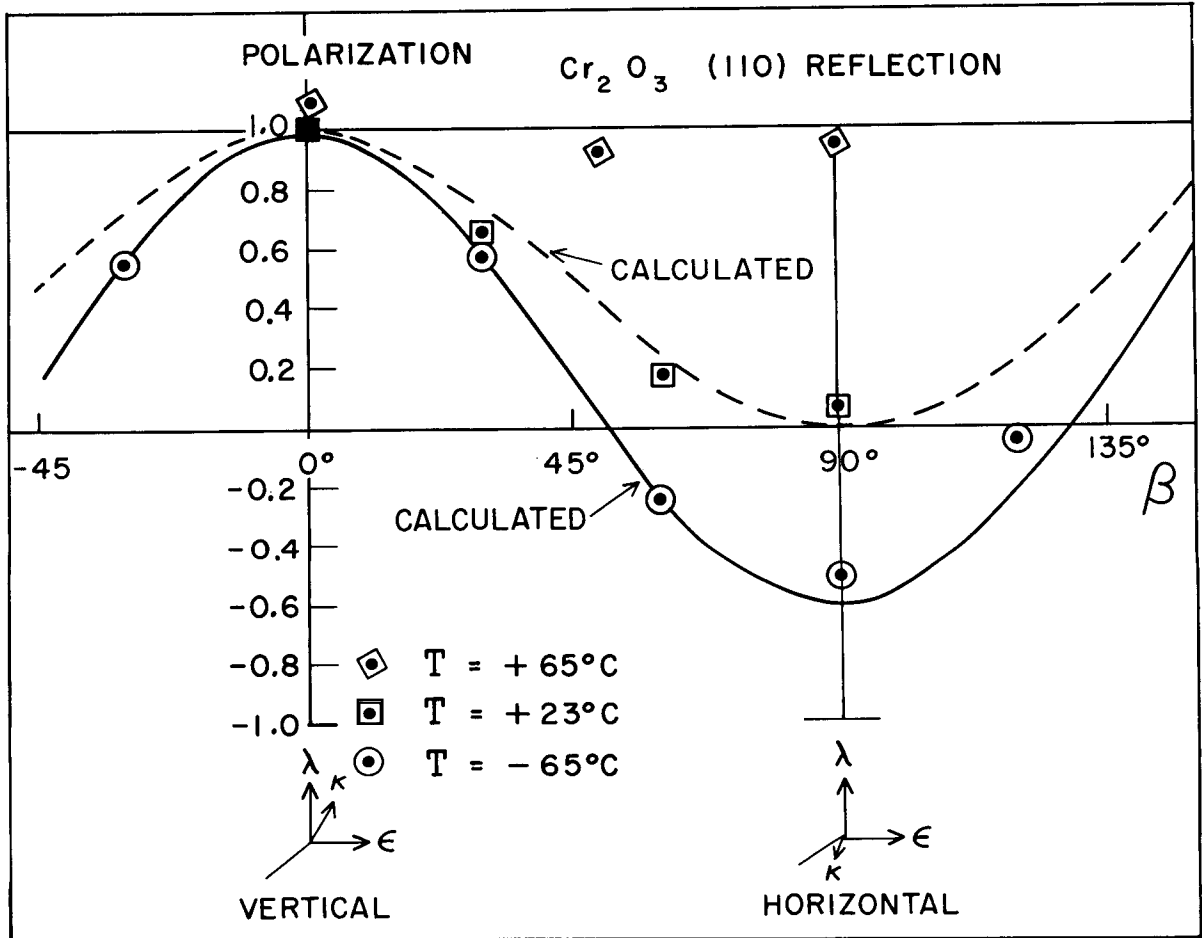


Fig. 2 - The polarization in the scattered beam from the (110) reflection of  $\text{Cr}_2\text{O}_3$  as a function of the angle  $\beta$  for different crystal temperatures.

The product of  $P_1 P_x$  was evaluated by turning to a pure nuclear reflection, in which case  $D = 1$ , and measuring the shim ratio,  $S$ .

### Cr<sub>2</sub>O<sub>3</sub>

Cr<sub>2</sub>O<sub>3</sub> is known to be antiferromagnetic (Neel temperature of 40° C) with a layer structure of chromium atoms ferromagnetically coupled<sup>(2)</sup> in layers and with adjacent layers antiparallel. Measurements on the paramagnetic susceptibility by McGuire indicate that the spins are directed along the trigonal axis of the rhombohedral unit cell. Measurements of  $D$  as a function of  $\beta$  were made on the (110) and (200) reflections. The former contains contributions from both nuclear and magnetic scattering while the latter is a purely magnetic reflection. Data on the (110) reflection were collected\* at three different temperatures. The results are shown in Fig. 2. We first observe, as predicted from equation (2) that  $D$  has a cosine-squared dependence on  $\beta$ . Secondly, at  $\beta = 0$ ,  $D = 1$  as expected, and at  $\beta = \pi/2$ ,  $D$  drops to a value which depends on the temperature. This again is expected since the ratio of magnetic to nuclear scattering decreases with increasing temperature up to the Neel point, above which there is no coherent magnetic scattering and consequently  $D$  remains at unity independent of changes in  $\beta$ . The curves corresponding to temperatures 23° C and -65° C were calculated with the use of equation (2) and neutron powder data which gave the ratio of nuclear to magnetic scattering at each of these temperatures, i. e.  $p^2 q^2 / b^2$ . The good agreement between the calculated curves and the experimental points shown in Fig. 2 supports the H-J predicted behavior for changes in the polarization of the Bragg scattered neutrons.

In the above discussion we have assumed the results of McGuire et al for the trigonal orientation of the spin axis in Cr<sub>2</sub>O<sub>3</sub><sup>(2)</sup>. The result can be confirmed independently by repeating the above measurements for one additional reflection. This follows from the fact that at  $\beta = 0$ , where  $D = 1$ , the spin axis must lie in the plane defined by  $\underline{\lambda}$  the polarization direction and  $\underline{e}$ , the scattering vector for the particular reflecting plane. In the case of the (110) reflection, the plane so defined is the (1 $\bar{1}$ 0). A similar measurement to locate  $\beta = 0$  for the (200) reflection showed the (10 $\bar{1}$ ) plane must also contain the spin direction. The intersection of the (1 $\bar{1}$ 0) and (10 $\bar{1}$ ) planes is the [111] or trigonal direction in confirmation of the previous measurements.

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\* We are indebted to Dr. T. R. McGuire of the Naval Ordnance Laboratory for supplying us with a single crystal of Cr<sub>2</sub>O<sub>3</sub>.

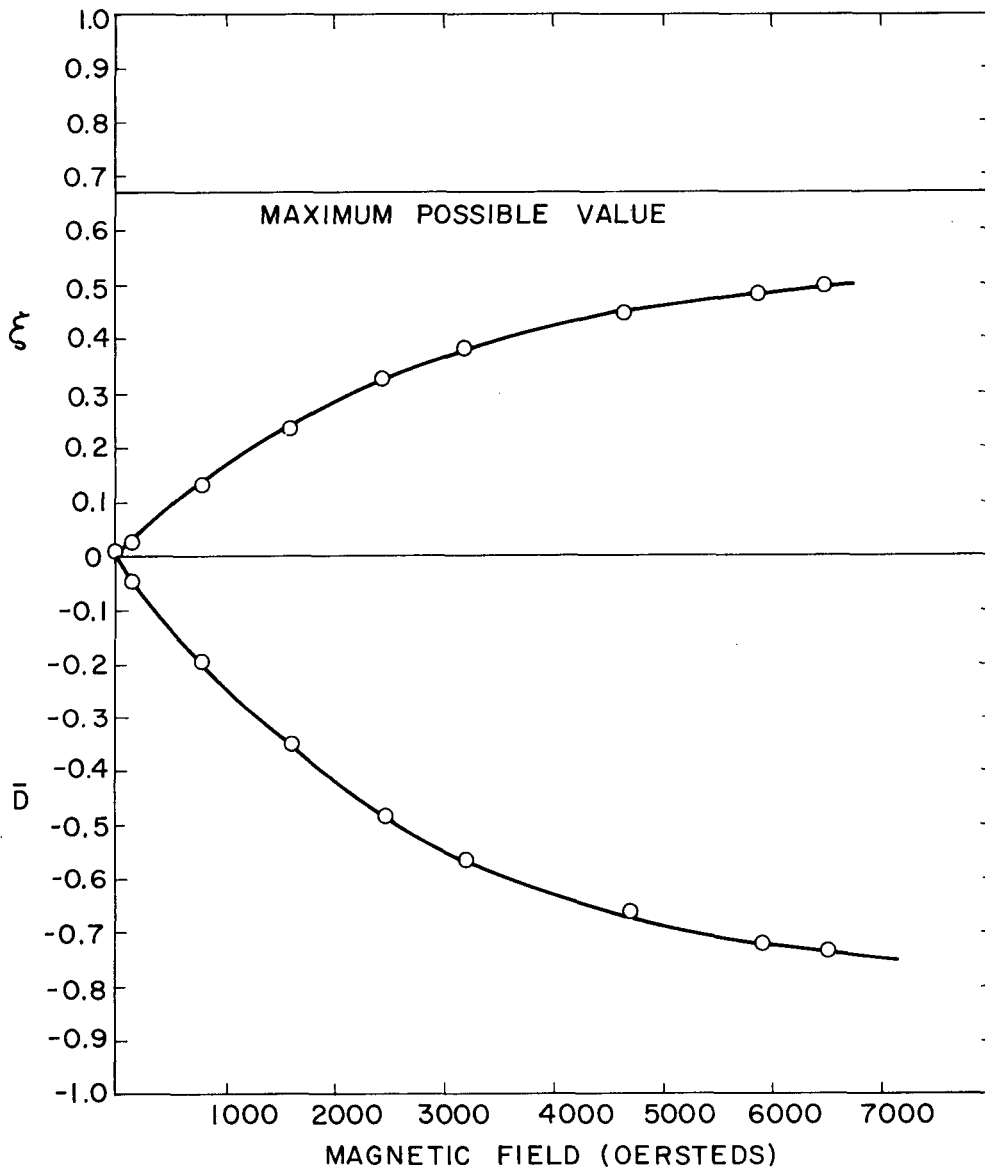


Fig. 3 - (Below) The polarization in the beam scattered from the (111) reflection of  $\alpha\text{-Fe}_2\text{O}_3$  as a function of applied magnetic field.

(Top) The excess number of spins oriented perpendicular to the direction of applied magnetic field.

$\alpha - \text{Fe}_2\text{O}_3$

The magnetic structure of hematite has been determined in the neutron diffraction studies of Shull, Strauser and Wollan<sup>(3)</sup>. This work showed that below the Neel point there exist two antiferromagnetic states. Below approximately  $-20^\circ\text{C}$  the magnetic axis lies along the trigonal axis as in  $\text{Cr}_2\text{O}_3$ . At room temperature the spins are in the (111) planes, remaining ferromagnetically coupled within the planes; adjacent planes, however, are antiferromagnetically coupled. As a further confirmation of the H-J predictions we have studied the (100) reflection of the low temperature phase of hematite. Since this reflection is purely magnetic in origin, we can expect from equation (2) that D will go from +1 to -1 as  $\beta$  goes from 0 to  $\pi/2$ . This complete reversal of the neutron polarization was found at  $\beta = \pi/2$  as predicted.

In addition to confirming the spin direction in the low temperature state of hematite as being along the trigonal axis, studies were made at room temperature of the effect of applied magnetic fields on the spin system of hematite. In this case the spins in the (111) plane can point in a number of directions. Because of the trigonal symmetry of the rhombohedral cell it is probable that the moments are directed at one of three nearest neighbors and this would lead to a set of three easy directions  $120^\circ$  apart. The value observed for D would then be the weighted average resulting from all the antiferromagnetic domains, that is

$$\bar{D} = \frac{\sum_i D_i q_i^2 N_i}{\sum_i q_i^2 N_i}$$

where  $D_i$  is the value of the polarization change,  $q_i$  the value  $q$  (as previously defined) and  $N_i$ , the fraction of spins, in each of the easy directions. In the case of the (111) reflection  $q_i = 1$  since all the spins are at right angles to the scattering vector and  $\bar{D} = \sum_i D_i N_i / \sum N_i$ . For zero magnetic field and the easy directions equally populated,  $\bar{D}$  is calculated to be zero for all angles of  $\beta$ . A study of D as a function of  $\beta$  for zero field showed this to be the case. It should be noted that this result does not exclude the possibility of a still larger number of easy directions, in fact the observed data can be shown to be consistent with any number of easy directions equal to, or greater than three.

When a magnetic field is applied within the (111) plane and parallel to the neutron polarization vector  $\lambda$ , the observed values of D for the (111) reflection are found to be changed and to become negative. This is seen in Fig. 3, where a plot of D versus applied field is shown. Increasingly large negative values for  $\bar{D}$  indicate that the effect of the applied field is to turn spins perpendicular to the applied field. We can, in fact, use the measured values of

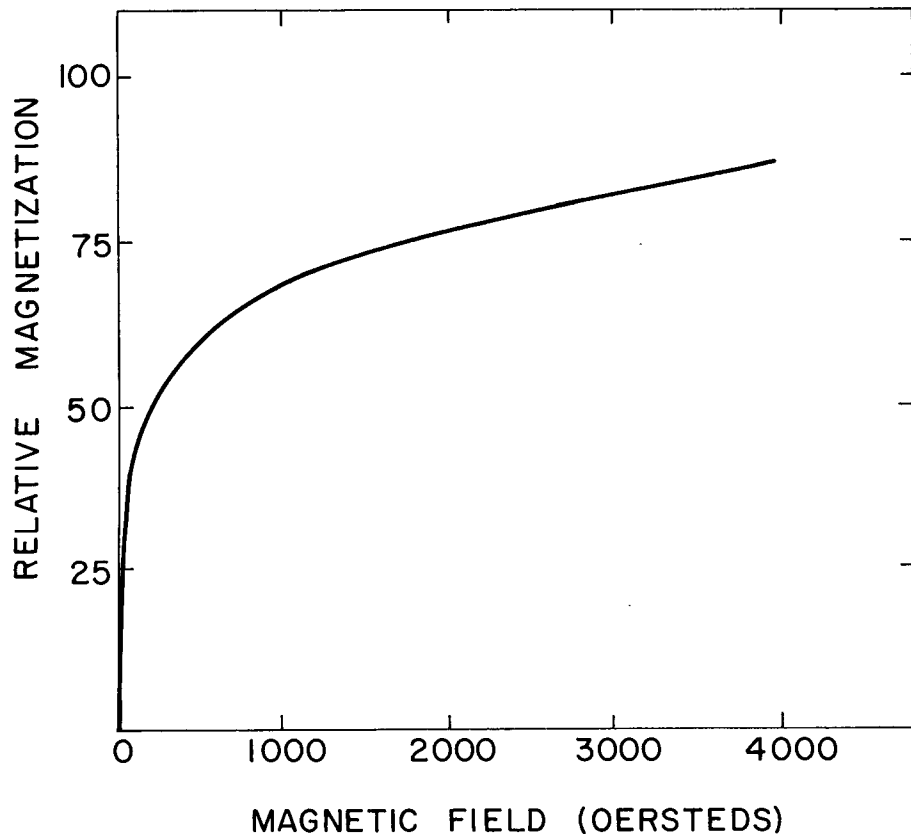


Fig. 4 - The magnetization curve of the ferromagnetic component in  $\alpha\text{-Fe}_2\text{O}_3$  after Townsend Smith<sup>(5)</sup>.

$\bar{D}$  to calculate the excess number of spins oriented perpendicular to the field over that observed at zero field. In this calculation we again assume that the spins within a given domain are directed toward one of the nearest neighbors. The upper curve in Fig. 3 gives this excess number as a function of field. We note that the resulting curve is somewhat similar in appearance to magnetization curves obtained for ferromagnetic materials with the field applied along a hard direction. Even at fields of 8400 oersteds, however, all the spins are not directed at right angles to the applied field; in that case,  $\bar{D}$  would be equal to -1. As a further point it should be noted that placing a magnetic field of equal strength along the trigonal axis produced no observable change in the direction of the spins, a result which is consistent with the previously known difficulty of rotating the magnetic spins out of the (111) plane.

It is possible to draw several conclusions from the above results on  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>. The presence of a parasitic ferromagnetic component in hematite has been the source of much experimental work<sup>(5-10)</sup>. These studies have shown that for the high temperature phase of hematite the ferromagnetism lies within the (111) sheet. The room temperature curve of magnetization versus field can be described as a weak ferromagnetism which saturates easily at low fields superimposed on a paramagnetic component. A typical magnetization curve taken from the data of Townsend Smith<sup>(5)</sup> is reproduced in Fig. 4. We also note that measurements on the magnetostrictive effects by Urquhart and Goldman<sup>(11)</sup> have suggested that the magnetostriction at room temperature is closely related to the ferromagnetic component.

The behavior of the "saturation" curve for the antiferromagnetic domains given in Fig. 3 differs significantly from that of the ferromagnetic component shown in Fig. 4. This suggests that the main source of the parasitic ferromagnetism must, for the most part, be independent of the orientation of the antiferromagnetic domains within the crystal. At low fields where the neutron data show the domains to be still essentially random, the net spins making up the ferromagnetic component have already been aligned with the field direction. One cannot rule out, however, a connection between the paramagnetic part of the magnetization curve of the ferromagnetic component and the rotation of the antiferromagnetic domains. We note that the remarks given here are consistent with theoretical considerations of Li<sup>(12)</sup>, which attribute the presence of the ferromagnetism in hematite to the uncompensated moments in the domain walls.

There remains the question of the lack of "saturation" of the antiferromagnetic domains at the higher fields applied to the crystal. In reference (11), Urquhart and Goldman have discussed the usual forces tending to line up the ferromagnetic moment with the applied field and those tending to rotate the antiferromagnetic axis perpendicular to the field. In addition, they note there is a third

force resulting from the coupling between the spins in the antiferromagnetic domains and the uncompensated spins in the domain wall. This coupling tends to keep the antiferromagnetic axis parallel to the net wall magnetization, that is, parallel to the applied field. The slow approach of the antiferromagnetic domains to saturation shown in Fig. 3 may therefore be caused by a competition between this third force and the usual force found in antiferromagnets which acts to rotate the antiferromagnetic axis perpendicular to the direction of the applied field.

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