

## Magnetic Torque Curves for a Single Crystal of Thulium Orthoferrite ( $\text{TmFeO}_3$ )

CHINJI KURODA, TOMONAO MIYADAI, AKIRA NAEMURA, NOBUKAZU NIIZEKI, AND HISAO TAKATA

Electrical Communication Laboratory, Nippon Telegraph and Telephone Public Corporation, Musashino-shi, Tokyo, Japan

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The magnetic anisotropy of a thulium orthoferrite single crystal was investigated by means of an automatically recording torque-magnetometer in the range between 77 and 300°K. It was found that at room temperature the direction of weak magnetization,  $\sigma_0$ , associated with an antiferromagnetic canted spin arrangement is parallel to the  $c$  axis of orthorhombic structure, and that at about 100°K a magnetic transition occurs, where the direction of  $\sigma_0$  changes from the  $c$  axis to the  $a$  axis. The transition temperature obtained is the highest among those reported before on other orthoferrites. The torque curves obtained above the transition

temperature are similar to those by Sherwood *et al.* and can be explained by their model. The value of  $\sigma_0$  is equal to 0.06  $\mu_B$ /mole at room temperature. At 77°K below the transition temperature, however, inexplicable torque curves were obtained in the  $b$  plane. The curves showed a small jump at the  $c$  axis superimposed on a  $\sin 2\theta$  curve with large amplitude. The jump amplitude is almost independent of the applied field in the range of 8500 to 3000 oe. This behavior cannot be explained by Sherwood's model, but it will give information on the interaction between thulium and ferric ions.

ORTHOERRITES, which have the general formula of  $M\text{FeO}_3$  ( $M$  being Y, or one of rare earth elements) and an orthorhombic structure, have been understood as antiferromagnetic substances with a weak or parasitic ferromagnetism as the result of a canted antiferromagnetic alignment of spins.<sup>1</sup> Their magnetic properties such as spontaneous magnetization, magnetic domains, magnetic torque curves, etc., have been widely investigated by many workers.<sup>2-4</sup> The spontaneous magnetization of the parasitic ferromagnetism is highly anisotropic because of its antiferromagnetic character,<sup>3</sup> and at room temperature it is, for most orthoferrites, directed along the  $c$  axis and in some of them it is reported as changing the direction of the spontaneous magnetization,  $\sigma_0$ , from the  $c$  axis into the  $c$  plane at low temperatures.<sup>4</sup> Sherwood *et al.*<sup>3</sup> have measured magnetic torque curves for  $\text{HoFeO}_3$  and for other several ferrites, and explained their results by assuming that  $\sigma_0$  is fixed along the  $\pm c$  axis during the rotation of the magnetic field and that the paramagnetic susceptibility due to the rare earth ions is anisotropic:  $\chi_a \neq \chi_b \neq \chi_c$ .

We have also measured, by the use of an automatically recording torque magnetometer, magnetic torque curves for  $\text{TmFeO}_3$ , for which Sherwood *et al.* did not present any explicit data, and have obtained, at 77°K, somewhat different data from those of Sherwood *et al.* for  $\text{HoFeO}_3$ . The single-crystal sample used for the measurement was prepared by a flux method similar to that proposed by Remeika<sup>5</sup> and is in a form of a platelet with (001), (110), ( $\bar{1}\bar{1}0$ ) planes, referring to the orthorhombic system, as its surfaces. The lattice parameters are 5.244, 5.565, and 7.580 Å for the orthorhombic  $a$ ,  $b$ , and  $c$ -axes, respectively.

<sup>1</sup> R. M. Bozorth, Phys. Rev. Letters **1**, 362 (1958); W. C. Koehler, E. C. Wollan, and M. K. Wilkinson, Phys. Rev. **118**, 58 (1960).

<sup>2</sup> M. A. Gilleo, J. Chem. Phys. **24**, 1239 (1956); R. M. Bozorth, H. J. Williams, and D. E. Walsh, Phys. Rev. **103**, 572 (1956); H. Watanabe, J. Phys. Soc. Japan **14**, 511 (1959).

<sup>3</sup> R. C. Sherwood, J. P. Remeika, and H. J. Williams, J. Appl. Phys. **30**, 217 (1959).

<sup>4</sup> R. M. Bozorth, V. Kramer, and J. P. Remeika, Phys. Rev. Letters **1**, 3 (1958).

<sup>5</sup> J. P. Remeika, J. Am. Chem. Soc. **78**, 4259 (1956).

Our results are shown in Figs. 1 and 2. Figure 1 shows torque curves in the three principal planes,  $a$ ,  $b$ ,  $c$ , at room temperature and at liquid nitrogen temperature (77°K). As shown, the curves at room temperature are essentially the same as those of Sherwood *et al.* According to Sherwood *et al.*, the torques in the  $a$ ,  $b$ ,  $c$ , planes are given as

$$L_{a,b} = \pm \sigma_0 H \sin \theta - \frac{1}{2} (\chi_c - \chi_{a,b}) H^2 \sin 2\theta,$$

$$L_c = -\frac{1}{2} (\chi_a - \chi_b) H^2 \sin 2\alpha,$$

where  $H$  is the applied field,  $\theta$  is the angle between the direction of  $H$  and the  $c$  axis, and  $\alpha$  is the angle between  $H$  and the  $a$  axis. The dashed curves in Fig. 1 show that  $\chi_c - \chi_{a,b} \approx 0$  within the experimental errors ( $\sim 10^{-5}$  emu/g) and  $\chi_a - \chi_b$  is slightly different from zero. The discontinuity at the  $b$  or  $a$  axis, which corresponds to  $\sigma_0 H$  in the formula of Sherwood *et al.*, increases linearly with  $H$ . From this linearity with  $H$ ,  $\sigma_0$  can be obtained. At room temperature, the torque curves show that  $\sigma_0$  is directed along the  $c$  axis. As seen from the solid curves in Fig. 1, the torque curves at 77°K are remarkably

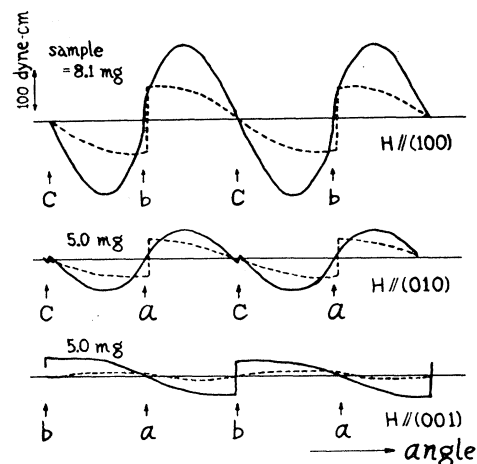


FIG. 1. Magnetic torque curves for  $\text{TmFeO}_3$  obtained in the  $a$ ,  $b$ , and  $c$  planes, respectively. Solid line, at 77°K; dashed line, at 300°K.  $H = 8500$  oe.

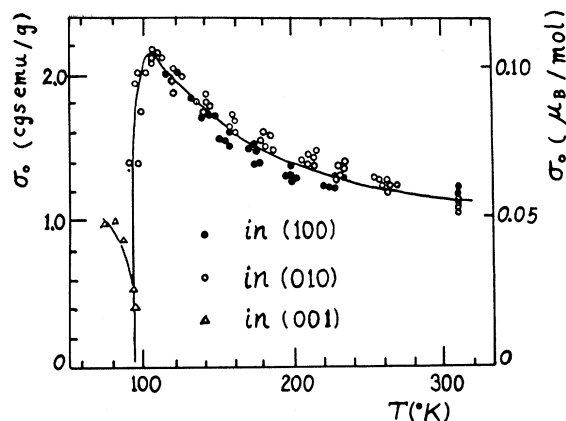


FIG. 2. Temperature variation of magnitude of  $\sigma_0$ , which was calculated from the discontinuities in the torque curves assuming  $\sigma_0 H = \Delta L$ .

different from those at room temperature and the change from the latter into the former occurs very rapidly with temperature—within several degrees. The torque curves at 77°K show that  $\sigma_0$  is directed along the  $a$  axis. The transition of the direction of  $\sigma_0$  occurs near 100°K. Figure 2 shows the temperature variation of  $\sigma_0$  and also the sharpness of the transition described above.

It is noticed that  $\chi_c - \chi_b$  and  $\chi_c - \chi_a$ , which are both positive in contrast with the data of Sherwood *et al.*, increase rapidly below the transition temperature as the temperature decreases (at 77°K they are equal to  $7 \times 10^{-4}$  and  $4 \times 10^{-4}$  emu/g, respectively) and that the discontinuity in the torque curve in the  $b$  plane occurring in the direction of the  $c$  axis, is too small compared with those for the  $c$  plane; in addition, it does not decrease linearly with decreasing  $H$ , and it seems even to increase

slightly as  $H$  is decreased. If Sherwood's model were still correct at 77°K, where the  $\sigma_0$  direction had changed for our  $\text{TmFeO}_3$ , then the torque curve in the  $b$  plane would be the one which consists of a superposition of two curves, one corresponding to the curve in the  $c$  plane, the other to the curve in the  $a$  plane which represents only a paramagnetic term with a uniaxial symmetry (i.e., containing only a coefficient of  $\sin 2\theta$ ). Furthermore, the discontinuities would increase linearly with  $H$ . But this is not the case.

Therefore, it seems to us that Sherwood's model should be, more or less, modified in order to interpret our data at 77°K. We have a qualitative explanation for the smallness and the field dependence of the discontinuity in the curve in the  $b$  plane at 77°K, if we assume that at 77°K or below the transition temperature  $\sigma_0$  can rotate "rather easily" in the  $b$  plane but that the magnetic field strength is not enough to make the direction of  $\sigma_0$  follow that of the field without any deviation of angle. On these assumptions, the uniaxial anisotropy constant of  $\sigma_0$  in the  $b$  plane may be obtained as  $3 \times 10^4$  erg/cc. However, prior to more detailed discussions on the behavior described above, it seems to be necessary to study the following problems: whether torque curves like ours are obtained near the transition temperature for  $\text{YFeO}_3$  and  $\text{LuFeO}_3$  in which  $\text{Y}^{3+}$  and  $\text{Lu}^{3+}$  are diamagnetic; whether there exists any interaction between  $\text{Tm}^{3+}$  and  $\text{Fe}^{3+}$  lattices.

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