Fig. 1) with a g value of 3.3 ± 0.3. The large uncertainty is due to the initial width and weakness of the line which is compounded when it splits in a field. To a first approximation we would expect a magnon sideband to have a g value equal to $g_{\text{exc}} - g_{\text{mag}}$ or 2.7. Between 4 and 140°K, beyond which line 5 is too broad to observe, the separation between line 5 and line 1 falls somewhat faster than the sublattice magnetization measured in neutron scattering.[30] In fact, probably coincidentally, it falls like a Brillouin function for spin $\frac{3}{2}$. This difference may arise from the effect of exciton-magnon coupling on the energy renormalization. Zone-boundary magnon energies are not yet known for Cr_2O_3, but we can extrapolate Samuelsen's neutron data[31] out to the zone boundary, and find an energy ≈ 250 cm^{-1}. An exciton-magnon density of states peak (line 5) 227 cm^{-1} from the $k=0$ exciton energy (line 1) is then reasonable. Wickersheim[3] has previously assigned line 5 as a vibronic, but with the additional information that we now have on its properties it appears that his assignment was incorrect.

In summary, this section has presented some new data for lines 1–5, of which the circular polarizations of the magnetically split components of lines 1 and 4 and the temperature shifts of the separations of lines 1 and 4, and 2 and 3, confirm very well the reassignments made in this work. Unfortunately, the temperature-dependent integrated intensities, perhaps the most interesting data, remain unexplained and should be a good topic for future study.

ACKNOWLEDGMENTS

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Direct Measurements of Anisotropy Energy and Anisotropic Magnetization in Gallium Iron Oxide

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Direct measurements are reported of the dependence of the anisotropy torque on the orientation of an applied magnetic field in the *ac* and *bc* crystallographic planes of ferrimagnetic $Fe_xGa_{2-x}O_3$ for $x=1.11$. Temperatures between 4.2 and 294°K and applied fields up to 110 kOe were used. Anisotropic spontaneous magnetizations, as well as isotropic susceptibilities and field-dependent anisotropy energies, were determined by comparing the experimental data with a new thermodynamic analysis of the magnetic properties of spontaneously magnetized orthorhombic crystals. The larger of the values obtained at 4.2°K for the zero-field anisotropy energy (3.22×10^6 erg/cm^3 and 5.65×10^6 erg/cm^3 in the *ac* and *bc* planes, respectively) is the largest value of this quantity measured so far in any insulator. The contributions of classical dipolar interactions to the zero-field anisotropy energy at 0°K are calculated to be 1.22×10^6 and 4.19×10^6 erg/cm^3 in the *ac* and *bc* planes, respectively. The remainder of the anisotropy energy is shown to be probably due to one-ion anisotropy or antisymmetric exchange or both. The largest values obtained for the fractional anisotropy of the spontaneous magnetization (0.24 and 0.55 in the *ac* and *bc* planes, respectively, at 294°K) are, to our knowledge, the largest values of this quantity measured so far in any material. It is found, moreover, that the anisotropic part of the spontaneous magnetization does not tend to zero as the temperature approaches 0°K. Direct measurements of the anisotropic magnetization are reported for 276 and 294°K, and the results are found to agree satisfactorily with those deduced from the torque measurements.

I. INTRODUCTION

GALLIUM iron oxide, $Fe_xGa_{2-x}O_3$ (where $0.7 < x < 1.4$), was first synthesized by Remeika,[1,1a] who found it to be both ferromagnetic and piezoelectric. In 1964 Rado[2] reported that it is the first ferromagnetic material known to exhibit the magnetoelectric effect. Other authors have studied its spontaneous moment and susceptibility,[3,4] its crystallographic structure,[5,6] its

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ferrimagnetic order,[7,8] and its resonance properties.[9,10] The magnetocrystalline anisotropy of this material has been deduced from magnetization data by Pinto[4] and by Nowlin and Jones,[3] from small-angle torque measurements by Schelleng[11] and from ferrimagnetic resonance measurements by Dweck.[10] This anisotropy was found to be orthorhombic in nature and unusually large in magnitude. The crystal *c* axis is the magnetically easy or preferred direction, the *a* axis is the intermediate direction, and the *b* axis is the hard direction.

One purpose of this paper is to report direct torque measurements of the anisotropy constants of $Fe_xGa_{2-x}O_3$, made possible by the very high sustained fields at the Naval Research Laboratory High-Field Magnet Facility. Because the anisotropy energy of this material, except near the Curie point, is greater than its Zeeman energy in the presence of fields normally available in laboratories, such direct torque measurements have not been made previously. In conjunction with this study, the magnetic-dipole contribution to the anisotropy has been calculated, and some observations on other possible sources of anisotropy in this material have been made.

Another purpose of this paper is to show that $Fe_xGa_{2-x}O_3$ exhibits an unusually large anisotropic magnetization, and that there is evidence that this magnetization persists even at temperatures as low as 4.2°K. The concept of anisotropic magnetization (i.e., the dependence of magnetization on crystallographic directions, resulting in a field-dependent anisotropy in ferromagnetics and ferrimagnetics) was first proposed by Callen and Callen.[12] According to their theory, however, the anisotropic magnetization vanishes at 0°K. Various mechanisms which do lead to a nonvanishing anisotropic spontaneous magnetization at 0°K were recently discussed by Rado.[13]

After presenting a thermodynamic analysis of the torque measurements (Sec. II), we describe the specimen (Sec. III) and apparatus (Sec. IV) used in the experiments. Next we report the results of the torque measurements and discuss their accuracy (Sec. V). This is followed by a calculation of the contribution to the anisotropy energy arising from dipole-dipole interactions (Sec. VI) and by a discussion of possible contributions of other mechanisms (Sec. VII). Finally, we report a direct measurement of anisotropic magnetization (Sec. VIII).

II. ANALYSIS OF TORQUE MEASUREMENTS

The torque method for the experimental determination of magnetocrystalline anisotropy in a ferromagnet or ferrimagnet requires the knowledge of a suitable functional relation $L(\mathbf{H})$ between the magnitude L of the torque density and the magnitude, as well as direction, of the magnetic field $\mathbf{H}$. While the variables L and $\mathbf{H}$ are directly measurable, the various anisotropy constants are unknown parameters which can be determined by comparing $L(\mathbf{H})$ with appropriate experiments. In deriving the required relation $L(\mathbf{H})$ for the case of an orthorhombic crystal, we find it both necessary and sufficient to allow (a) the magnitude M of the total magnetization $\mathbf{M}$ to be anisotropic so that the anisotropy constants may be field-dependent, (b) the susceptibility χ above technical saturation to be nonzero but isotropic so that only the spontaneous part of M may be anisotropic, and (c) the anisotropy energy density to be a sufficiently large fraction of $\mathbf{M}\cdot\mathbf{H}$ so that $\mathbf{M}$ may not be parallel to $\mathbf{H}$. It should be noted that while the generalization (a) was already used in Aubert's[14] work on crystals of cubic symmetry, a simultaneous consideration of (a), (b), and (c) has not yet been published for any symmetry. Nevertheless, our relation $L(\mathbf{H})$ still lacks full generality because it neglects (d) all demagnetizing effects, (e) a possible anisotropy of χ, and (f) all anisotropy constants of order higher than 2. The assumption (d) reflects the fact that in gallium iron oxide the ratio of $2\pi M^2$ to the first-order anisotropy constant is small compared to unity at any temperature T. Assumptions (e) and (f), on the other hand, can only be justified by noting that the relation $L(\mathbf{H})$ derived below does suffice to represent our torque data on gallium iron oxide.

To define the various anisotropy constants, we expand the Helmholtz free-energy density F of the magnetic sample in powers of the direction cosines α_a, α_b, α_c of $\mathbf{M}$ with respect to the principal axes a, b, and c of the orthorhombic crystal. The anisotropy constants are the nonvanishing coefficients contained in this expansion. Thus the coefficients of $\alpha_a^k\alpha_b^l\alpha_c^m$ (where k, l, m are zero or positive integers) for odd values of $n=k+l+m$ must vanish because F is invariant under time reversal. The coefficients for even values of n, on the other hand, form the components of a time-invariant polar tensor of rank n. As may be verified by appropriate tables,[15] the number of independent components of this tensor in the case of orthorhombic symmetry is three for $n=2$ and six for $n=4$. Because of the identity $\alpha_a^2+\alpha_b^2+\alpha_c^2=1$, however, one of the α's may be eliminated so

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[15] R. R. Birss, in *Symmetry and Magnetism* (North-Holland Publishing Co., Amsterdam, 1964). See Tables VII and IV.

that the number of independent tensor components is reduced to two for $n=2$ and three for $n=4$. Hence we obtain

$$F=K_0+K_{1a}\alpha_a^2+K_{1b}\alpha_b^2+K_{2a}\alpha_a^4+K_{2b}\alpha_b^4+K_{2ab}\alpha_a^2\alpha_b^2, \qquad (1)$$

where K_0 is the isotropic part of F, K_{1a} and K_{1b} are the first-order "intrinsic" anisotropy constants, and K_{2a}, K_{2b}, and K_{2ab} are the second-order "intrinsic" anisotropy constants. The K's, as well as F, are specified by the independent variables T and $\mathbf{M}$. If we describe the direction of $\mathbf{M}$ by the polar coordinates θ and φ, where $\alpha_a=\sin\theta\cos\varphi$, $\alpha_b=\sin\theta\sin\varphi$, and $\alpha_c=\cos\theta$, then the independent variables of F become T, M, θ, and φ. Next, we introduce the thermodynamic potential

$$\tilde{F}(T,H_M,\theta,\varphi)=F(T,M,\theta,\varphi)-MH_M, \qquad (2)$$

where H_M denotes $(\mathbf{H}\cdot\mathbf{M})/M$. The quantity $\tilde{F}$ may be regarded either as the total free-energy density of the magnetic sample and the "H reservoir" or as that particular Legendre transform of F in which H_M rather than M is one of the independent variables. It is $\tilde{F}$ rather than F which will be used for deriving $L(\mathbf{H})$.

The considerations which follow will be restricted to the case where $\mathbf{H}$ is in the ac plane ($\varphi=0$) so that, by virtue of the crystalline symmetry, $\mathbf{M}$ is also in this plane. It is easily shown, however, that the corresponding results for the bc plane ($\varphi=\pi/2$) can be obtained from those for the ac plane by merely replacing a by b throughout the subscripts. Proceeding with the case of the ac plane, we write Eq. (1) in the form

$$F(T,M,\theta)=K_0(T,M)+K_{1a}(T,M)\sin^2\theta+K_{2a}(T,M)\sin^4\theta, \qquad (3)$$

and then justify the analogous expansions

$$M(T,H_M,\theta)=M_0(T,H_M)+M_{1a}(T,H_M)\sin^2\theta+M_{2a}(T,H_M)\sin^4\theta, \qquad (4)$$

$$\tilde{F}(T,H_M,\theta)=K_0'(T,H_M)+K_{1a}'(T,H_M)\sin^2\theta+K_{2a}'(T,H_M)\sin^4\theta \qquad (5)$$

by using symmetry arguments similar to those underlying Eq. (1). Here M_0, M_{1a}, and M_{2a} denote, respectively, the isotropic, first-order anisotropic, and second-order anisotropic parts of M. The coefficients K_0', K_{1a}', and K_{2a}', which are analogous to K_0, K_{1a}, and K_{2a}, may be referred to as "extrinsic" anisotropy constants. Their values are given by the equations

$$K_0'(T,H_M)=K_0(T,M)-M_0(T,H_M)H_M, \qquad (6)$$

$$K_{pa}'(T,H_M)=K_{pa}(T,M)-M_{pa}(T,H_M)H_M, \qquad (7)$$

where p denotes the order (1 or 2) of the pertinent anisotropic quantity. Equations (6) and (7) are derived by a comparison of Eq. (5) with the equation obtained by substituting Eqs. (3) and (4) into Eq. (2). Next we take the partial derivative of Eqs. (6) and (7) with respect to H_M and simplify the result by using the thermodynamic relations

$$M_0=-\partial K_0'/\partial H_M \quad \text{and} \quad M_{pa}=-\partial K_{pa}'/\partial H_M, \qquad (8)$$

which follow from $M=-\partial\tilde{F}/\partial H_M$ and Eq. (5). The expressions

$$\partial K_0(T,M)/\partial H_M=H_M\partial M_0(T,H_M)/\partial H_M, \qquad (9)$$

$$\partial K_{pa}(T,M)/\partial H_M=H_M\partial M_{pa}(T,H_M)/\partial H_M \qquad (10)$$

obtained in this way will now be used to modify Eqs. (6) and (7). We assume that M_0 is given by the relation

$$M_0(T,H_M)=M_{00}(T)+\chi(T)H_M, \qquad (11)$$

where $M_{00}(T)$ is the spontaneous part of the isotropic magnetization. Substitution of Eq. (11) into Eq. (9) yields

$$K_0(T,M)=K_{00}(T)+\tfrac{1}{2}\chi(T)H_M^2, \qquad (12)$$

where $K_{00}(T)$ is a constant of integration, and use of Eqs. (11) and (12) in Eq. (6) gives

$$K_0'(T,H_M)=K_{00}(T)-M_{00}(T)H_M-\tfrac{1}{2}\chi(T)H_M^2. \qquad (13a)$$

Since χ is assumed to be isotropic, the anisotropic magnetizations M_{pa} are spontaneous rather than induced so that the $\partial M_{pa}/\partial H_M$ must vanish. According to Eq. (10), therefore, the K_{pa} are independent of H_M and dependent on T only. The two equations implied by Eq. (7) may thus be written in the form

$$K_{1a}'(T,H_M)=K_{1a}(T)-M_{1a}(T)H_M, \qquad (13b)$$

$$K_{2a}'(T,H_M)=K_{2a}(T)-M_{2a}(T)H_M. \qquad (13c)$$

It should be noted that Eqs. (13) are preferable to Eqs. (6) and (7) for two reasons. Firstly, Eqs. (13) contain only two independent variables (T and H_M), and secondly, Eqs. (13) display the functional dependencies on H_M, explicitly. Thus we substitute Eqs. (13) into Eq. (5) and use the resulting expression

$$\tilde{F}(T,H_M,\theta)=K_{00}(T)-M_{00}(T)H_M-\tfrac{1}{2}\chi(T)H_M^2+[K_{1a}(T)-M_{1a}(T)H_M]\sin^2\theta+[K_{2a}(T)-M_{2a}(T)H_M]\sin^4\theta \qquad (14)$$

as the basis for our calculation of $L(\mathbf{H})$. We note that the various free-energy densities contained in Eq. (14) describe the intrinsic anisotropy and the interaction of $\mathbf{H}$ with the induced magnetization (χH_M), as well as with the components (M_{00}, M_{1a}, and M_{2a}), of the spontaneous magnetization. The term $-\frac{1}{2}\chi(T)H_M^2$ is just what one expects in view of the fact that the infinitesimal work of magnetization is $-MdH_M$, rather than H_MdM, whenever the total free-energy density (which is $\tilde{F}$ in our case) does contain the interaction ($-MH_M$) between $\mathbf{M}$ and $\mathbf{H}$. To simplify the notation, the independent variable T will henceforth be dropped from Eq. (14) and its consequences.

The equilibrium value of θ must minimize $\tilde{F}$ for a fixed value of $\mathbf{H}$. Thus θ must satisfy the equilibrium condition

$$(\partial\tilde{F}/\partial\theta)_{H,\psi}=0, \qquad (15)$$

which we do not write down explicitly. Here ψ is the angle between **H** and the c axis so that H_M is $H\cos(\psi-\theta)$. In principle, we could now determine the equilibrium value of θ from Eq. (15), substitute this value into Eq. (14), and then calculate L from the equation

$$-L=\partial\tilde{F}/\partial\psi. \qquad (16)$$

For computing purposes, however, it is convenient to proceed differently. We write Eq. (16) in the form

$$-L=(\partial\tilde{F}/\partial\psi)_\theta+(\partial\tilde{F}/\partial\theta)_\psi(\partial\theta/\partial\psi), \qquad (17)$$

where the second term vanishes because of Eq. (15). Use of Eq. (14) in Eq. (17) then yields

$$\begin{aligned}-L=\tfrac{1}{2}\chi H^2\sin 2(\psi-\theta)&+M_{00}H\sin(\psi-\theta)\\ &+M_{1a}H\sin(\psi-\theta)\sin^2\theta\\ &+M_{2a}H\sin(\psi-\theta)\sin^4\theta, \qquad (18)\end{aligned}$$

where the terms on the right-hand side represent the torque densities exerted by **H** on the induced magnetization and on the three components of the spontaneous magnetization. Next we substitute the explicit form of Eq. (15) into Eq. (18) to obtain

$$\begin{aligned}-L=[K_{1a}-M_{1a}H\cos(\psi-\theta)]\sin 2\theta&\\ +2[K_{2a}-M_{2a}H\cos(\psi-\theta)]\sin^2\theta\sin 2\theta&, \qquad (19)\end{aligned}$$

which can be written in the alternative form

$$-L=(K_{1a}'+K_{2a}')\sin 2\theta-\tfrac{1}{2}K_{1a}'\sin 4\theta. \qquad (20)$$

Finally, we eliminate θ from Eqs. (18) and (19) in order to calculate the desired functional dependence of L on H and ψ. If we make the somewhat drastic assumptions

$$K_{2a}=\chi=M_{1a}=M_{2a}=0, \qquad (21)$$

then we obtain the exact result

$$-\psi=\tfrac{1}{2}\sin^{-1}(L/K_{1a})+\sin^{-1}(L/M_{00}H). \qquad (22)$$

Equation (22) does not, however, agree with our torque data on gallium iron oxide at any temperature. If, on the other hand, we drop the assumptions of Eq. (21), then the solution of Eqs. (18) and (19) can no longer be obtained in closed form. Accordingly, we developed an iterative procedure for deriving numerical solutions with the help of an electronic computer. It is these solutions (together with their analog for the bc plane) that were used for plotting the theoretical curves.

III. SPECIMEN

The crystal used in this study was grown by a flux method.[16] It is the same crystal sample as that used by Pinto[4] in measuring the spontaneous moment of this material between 78 and 300°K. In the course of the present study the moment was remeasured at 78, 276, and 294°K using the same vibrating sample magnetometer as that used by Pinto. Agreement is excellent at 78°K and although the present magnetization values at the higher temperatures (48 emu/cm^3 at 276°K and 19 emu/cm^3 at 294°K) are somewhat higher than Pinto's, the differences are within the experimental error of the measurements. In addition, the moment was measured at 4.2°K and found to be equal to 185 emu/cm^3.

The nominal composition parameter, derived from the proportions of raw material used, is $x=1.08$. A systematic comparison of the Curie point of this specimen and the Curie points of chemically analyzed specimens, as measured by Nowlin and Jones,[3] indicates that a value of $x=1.11$ is more appropriate. The above mentioned value of the spontaneous magnetization of the sample at 4.2°K is in good agreement with that predicted by Nowlin and Jones for a sample with $x=1.11$. This value for the composition parameter will be used throughout the present paper.

The unit-cell dimensions of a powder sample made from crystals grown with the sample used in this study were measured by x-ray techniques and found to be $a=8.76$, $b=9.41$, $c=5.09$ Å. These dimensions and the composition parameter $x=1.11$ lead to a crystal density of 5.44 g/cm^3. The weight of the sample was 8.73 mg.

Since the specimen did not have the spherical shape preferred for anisotropy studies, the results should, in principle, be corrected for the effect of demagnetization fields due to external shape. The sample was roughly in the form of a parallelepiped whose longest dimension was along the crystallographic c axis. If the actual shape of the sample is approximated by a prolate spheroid of the same length and average width then the shape-dependent contribution to the anisotropy energy is about 0.02×10^6 erg/cm^3 at 4.2°K. This contribution is negligible compared to the measured magnetocrystalline anisotropy.

IV. APPARATUS

The high applied fields used in this study were supplied by one of the Bitter-type magnets of the High-Field Magnet Facility of the Naval Research Laboratory. The magnet used had a 2½-in. bore and provided an axial field of up to 110 kOe. Because of the axial nature of this field, the usual frictionless type of torque magnetometer could not be used. Instead, an apparatus, shown in Fig. 1, involving four sapphire jewel bearings (J) was designed. This apparatus consists of two light pulleys (U and L) connected by a loop of cotton thread (T). The sample (B) is attached to the lower pulley so that the axis of rotation of the pulley is perpendicular to the crystal plane in which the anisotropy is to be measured. A small scale (A), viewed by a microscope (R), is attached to the upper pulley to

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[16] Crystal prepared by F. Molea and obtained through the kindness of R. V. Jones.

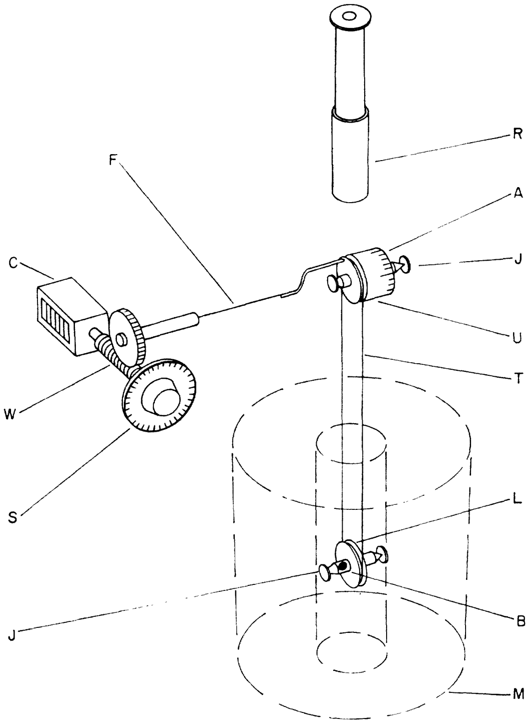

FIG. 1. Schematic drawing of the torque anisotropy measuring apparatus. The symbols are explained in the text.

indicate the angular position of the sample in the field. A calibrated phosphor bronze torsion fiber (F) is also attached to the upper pulley parallel to its axis. This fiber was used both to apply a restoring torque to the sample and to adjust its angular position. The opposite end of this fiber can be twisted by means of a worm gear arrangement (W). The fiber used in this experiment was made of 0.01-in.-diameter phosphor bronze wire and had a torsion constant of 1.54×10^{3} dyn cm/rad. A counter (C) and scale (S) is provided to determine this twist, and therefore the torque, to within 1/2000 of a turn. The whole apparatus is placed in a vacuum chamber, the lower pulley being at the bottom of a long tube. This tube extends into a double Dewar which is placed in the magnet (M).

To obtain the range of temperature desired for this study, liquefied helium, nitrogen, and "Freons" (fluorocarbons) 14, 13, 22, and 114 were used. The sample temperature was brought to that of the cryogenic liquid by introducing helium gas into the vacuum chamber. With the field turned off, the sample was then rotated through 180°. At each 10°, as noted on the scale on the upper pulley, the reading of the torsion-fiber counter and scale was noted. This process was then repeated with the field on. The difference between the torsion-fiber scale readings with the field off and with the field on represented the torque exerted on the crystal by the field at that crystal position. As a check, the field-off readings were again made, and the results averaged. To further minimize the errors inherent in the apparatus due to bearing friction, the measurements were repeated several times. From these data, curves of torque L versus field angle ψ were plotted at each field H and temperature T.

The presence of friction in the jewel bearings will introduce a random error into the determination of the measured torque. Consequently, the data exhibit a certain amount of scatter, most evident at the highest temperatures where the torques are smallest. This lead to an error of $\pm 0.05\times 10^{6}$ erg/cm^{3} in all values of anisotropy constants and ± 1.0 emu/cm^{3} in the determination of the anisotropic magnetization. Of more importance was a consistent error introduced by the stretching of the loop of cotton thread connecting the two pulleys. Because of this, the measured position of a point on a torque curve will be shifted away from the c axis by an angle proportional to its torque value, although the torque value itself will not be affected. This error will cause a distortion of the shape of the torque curves, but will not affect the magnitude of the torque maximum. The properties of the thread are such that this error is important only at the lowest temperatures where the torques are greatest.

V. RESULTS OF TORQUE MEASUREMENTS

Torque curves in both the ac and bc planes of $\mathrm{Fe}_x\mathrm{Ga}_{2-x}\mathrm{O}_3$ ($x\approx 1.11$) were taken at 4.2, 78, 145, 233, 276, and 294°K and at fields of 8, 16, 24, 32, 40, 59, and 90 kOe, and sometimes 4, 20, 75, and 110 kOe. The values of the observed torque maxima are shown graphically in Figs. 2 and 3 for those cases in which the measured maxima represent analytical maxima. It can be shown that in the absence of anisotropic magnetization the magnitudes of these torque maxima at a given temperature will not depend on the applied field. Yet at every temperature these magnitudes are seen to increase with increasing field, as shown by the slope of the lines connecting the points corresponding to a given temperature. This indicates that some form of anisotropic magnetization is present at every temperature, even at the low value of 4.2°K.

The L versus ψ data were then fitted to theoretical curves based on Eqs. (18) and (19) in which, for the present purpose, only first-order terms in anisotropy energy and anisotropic magnetization were retained. All the curves corresponding to a given temperature and crystallographic plane were required to have the same value of K_1 and M_1, and the curves corresponding to both planes at a given temperature were required to have the same values of M_{00} and χ. As a first approximation, Pinto's values were used for M_{00}. The $H=0$ intercepts of the constant temperature curves in Figs. 2 and 3 were used for K_1, the slopes of these curves were used for M_1, and χ was taken to be zero. Sets of theoretical curves of L versus ψ were generated by means of

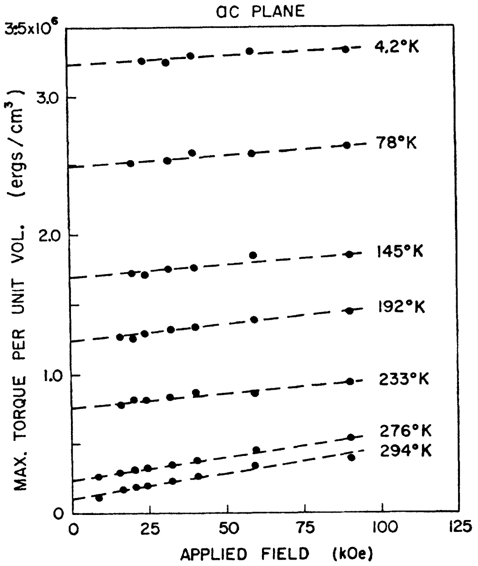

FIG. 2. Maximum measured anisotropy torque in the *ac* plane of $\mathrm{Fe}_x\mathrm{Ga}_{2-x}\mathrm{O}_3$ ($x=1.11$) as a function of applied field and temperature. Each point represent the maximum torque on a single curve of torque versus applied-field angle (ψ) at the temperature and applied-field magnitude (H) indicated. Only those points are included for which the observed torque maximum represents a true analytical maximum.

a computer and compared to the data. Where necessary, the original parameters were varied to improve the agreement between the measured and calculated curves. This process proved successful for all temperatures except 4.2 and 78°K, with only a few small changes in the approximate parameters needed to give curves that agreed with the data within the experimental error quoted above. An example of such fit is shown in Fig. 4. Except at the two highest temperatures, 276 and 294°K, the fit did not require invoking any appreciable susceptibility. The susceptibility needed at these two temperatures is in reasonable agreement with that derived by Pinto[4] from magnetization measurements.

It can be shown that had L in Figs. 2 and 3 been plotted as a function of H_M, the component of the field along the direction of the moment, the assumptions initially made for K_1 and M_1 would have been exactly valid. Unfortunately, the calculation of H_M itself requires a previous knowledge of the final parameters, so that such a plot could not at first be made. Using the values found from the fitting process, however, L could be plotted versus H_M and the parameters K_1 and M_1 could then be redetermined. These parameters were only slightly different from those assumed in the calculation of H_M and were therefore self-consistent. Theoretical curves calculated using these new parameters also fell within the experimental error of the data. Table I lists the basic parameters K_{1a}, K_{1b}, M_{00}, M_{1a}, M_{1b}, and χ found in this study. Except at 4.2 and 78°K these parameters were derived by the fitting process described above.

The experimental L versus ψ curves at 4.2 and 78°K could not be fitted using only the first-order terms in anisotropy energy and anisotropic magnetization. This was partially due to the angular distortion (error in ψ) introduced into the torque curves by stretching of the loop of string connecting the sample pulley and the scale pulley (see Sec. IV). This distortion is proportional to the torque being measured and inversely proportional to the Young's modulus of the string. The highest torques are found at the lowest temperatures so the

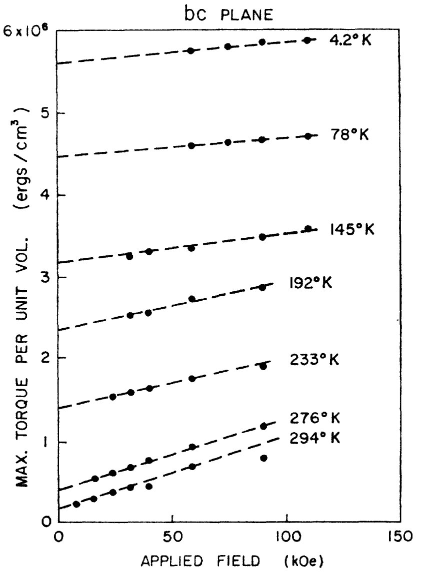

FIG. 3. Maximum measured anisotropy torque in the *bc* plane of $\mathrm{Fe}_x\mathrm{Ga}_{2-x}\mathrm{O}_3$ ($x=1.11$) as a function of applied field and temperature. Each point represents the maximum torque on a single curve of torque versus applied field angle (ψ) at the temperature and applied field magnitude (H) indicated. Only those points are included for which the observed torque maximum represents a true analytical maximum.

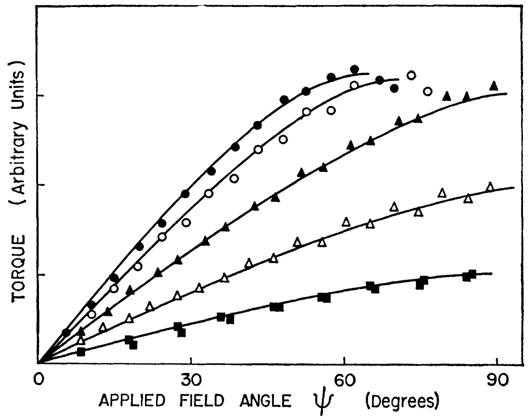

FIG. 4. Example of fit of theoretical curves to experimental points of torque versus applied field angle ψ. The measurements were made in the *bc* plane of $\mathrm{Fe}_x\mathrm{Ga}_{2-x}\mathrm{O}_3$ ($x=1.11$) at 145°K in various applied fields H (■, 8; △, 16; ▲, 32; ○, 59; and ●, 90 kOe).

TABLE I. Anisotropy energy and anisotropic magnetization of $Fe_xGa_{2-x}O_3$ (where $x=1.11$), as measured by torque methods. The values of the parameters K_{1a}, K_{1b}, M_{1a}, and M_{1b} given for 4.2 and 78°K were found from the magnitudes of the maximum torques and may contain higher-order contributions (see text). At all other temperatures the values of all the parameters were found by fitting the entire torque curve.

T (°K)	M_{00} (emu/cm^3)	χ	K_{1a} (10^6 erg/cm^3)	$-M_{1a}$ (emu/cm^3)	K_{1b} (10^6 erg/cm^3)	$-M_{1b}$ (emu/cm^3)
4.2	187	≈ 0	3.22	1.1	5.65	1.7
78	167	≈ 0	2.50	1.5	4.48	1.4
145	138	≈ 0	1.64	1.6	3.16	3.4
192	112	≈ 0	1.27	1.8	2.40	5.1
233	84	≈ 0	0.77	1.8	1.45	5.1
276	42	0.0010	0.26	3.0	0.44	8.1
294	13	0.0015	0.13	3.1	0.19	7.2

distortions there will be greatest. These are somewhat compensated for by the fact that the Young's modulus also increases with decreasing temperature. During the experiments, there is a temperature gradient along the string from room temperature to the sample temperature, and as this gradient, in practice, is neither measurable nor reproducible, an accurate correction cannot be made. It is expected, however, that a greater part of the string is near room temperature, so that an estimate of the maximum error can be made by using the room-temperature value of the Young's modulus. From this it is found that at 78°K most of the difference between the theoretical curves and the data arises from string stretching.

It should be reemphasized that while dependent on the torque, this error will only affect the determination of the angle ψ at which the external field is being applied. Thus, while it will change the apparent position of the torque maximum, it will not affect its magnitude. The arguments for the presence of anisotropic magnetization at these temperatures given in the first paragraph of this section will not be affected by the presence of this error.

At 4.2°K the stretch of the string can account for at most half of the above mentioned distortion of the torque curves. Furthermore, in the *ac* plane the observed distortion is in the opposite direction from that which would be caused by this source. This indicates the presence of higher-order anisotropy terms, most probably second-order terms of the form included in Eq. (1). The appearance of these terms at 4.2°K is not unreasonable because the temperature dependence of first- and second-order terms will undoubtedly be different. The value of the maximum torque depends, of course, on all orders of anisotropy, but as the effect of the string stretch error cannot be accurately determined, K_{1a} and K_{1b} represent only "effective" anisotropy constants at this particular temperature.

The values of the basic bulk parameters given in Table I for 4.2 and 78°K were derived merely from the maximum values of the torque as measured at the various applied fields. The values of K_1 derived above are plotted as a function of temperature in Fig. 5. The dashed line in this graph represents the results obtained by Pinto who applied a relatively incomplete model to his magnetization measurements and used only low fields. Our results, on the other hand, involved direct torque measurements of K_1 at high field, and are based on a much more complete analysis. Dweck[10] measured the anisotropy fields $2K_{1a}/M$ and $2K_{1b}/M$ for this material using ferrimagnetic resonance but did not take into consideration the possibility of a field dependence of K_{1a}', K_{1b}', and M. His results are given in Table II along with our values of the anisotropy field calculated for those values of H_M that equal Dweck's applied fields. The agreement is reasonable considering that the experiments were done on different crystals whose nominal composition parameter (determined by the ingredients of the melt) was $x=1.1$. It should be noted that our value $K_{1b}=5.65\times10^6$ erg/cm^3 at 4.2°K is the largest anisotropy constant measured so far in any insulator.

Finally, the deduced values of M_{1a} and M_{1b} are plotted in Fig. 6. All values except those at 4.2 and 78°K result from the fitting of the entire experimental torque curves to theoretical curves. The values of M_{1a}

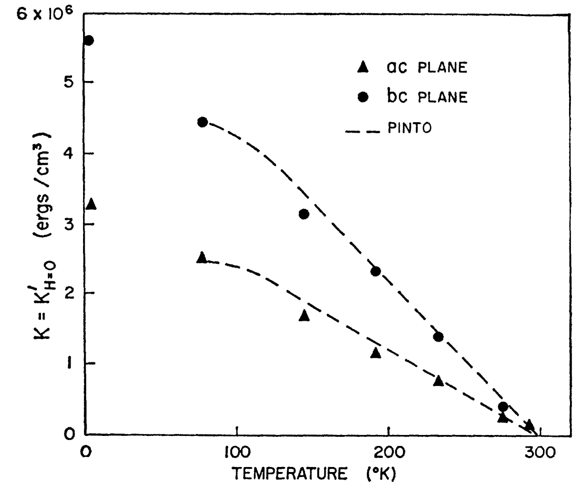

FIG. 5. Curves of K_{1a} (triangles) and K_{1b} (circles) versus temperature for $Fe_xGa_{2-x}O_3$ ($x=1.11$). The two points at 4.2°K probably include some admixture of higher-order anisotropy terms (see text). The dashed lines represent Pinto's[4] results obtained from magnetization measurements at low fields.

TABLE II. Comparison of anisotropy fields of $Fe_xGa_{2-x}O_3$ deduced from the torque measurements of the present paper ($x=1.11$) and from the resonance measurements of Dweck ($x\approx1.1$). Each K_{1a}' and K_{1b}' is listed for that value of H_M which equals the applied field H_{appl} used by Dweck to obtain his H_a and H_b.

T (°K)	H_{appl} (kOe)	$2K_{1a}'/M_{00}$ (kOe)	H_a (kOe)
4.2	40.5	34.9	31.0
78	38.5	30.8	28.5
145	36.0	25.2	25.0
192	33.0	23.2	21.5
233	30.0	18.8	15.5
276	26.0	12.7	8.5
		$2K_{1b}'/M_{00}$ (kOe)	H_b (kOe)
4.2	72.0	61.3	58.0
78	68.0	54.8	53.0
145	62.0	48.8	46.0
192	50.0	47.1	40.0
233	41.0	38.9	32.0

and M_{1b} at 4.2 and 78°K are derived from the field dependence of the maxima of these torque curves. The error shown is the previously mentioned effect on these values (via scatter of the points on the torque curves) of friction in the bearings of the apparatus. In both the *ac* and *bc* planes there are indications that the anisotropic magnetization does not go to zero as the temperature approaches 0°K. Because of the large experimental error, however, strong evidence for a nonvanishing anisotropic magnetization at 4.2°K can be claimed for the *bc* plane only. The largest anisotropy of spontaneous magnetization observed here ($M_{1b}/M_{00}=0.55$ at 294°K) is to our knowledge the largest yet found in and material.

VI. CALCULATION OF DIPOLAR ANISOTROPY

Classical dipole interactions between the ions of a noncubic magnetic material make a contribution to its first-order anisotropy. If the position and ordering of these ions are known, this contribution can be directly calculated. Structure studies[5,6,8] have shown that $Fe_xGa_{2-x}O_3$ has four inequivalent types of cation sites. Although nominally called the Fe_{I}, Fe_{II}, Ga_{I}, and Ga_{II} sites, they are all occupied to varying degrees by Fe^{+++} ions. It should be noted here that there is some nonuniformity in the literature as to the labelling of these sites, and that the labelling first introduced by Bertaut *et al.*[6,8] will be used in this paper. Each unit cell contains four crystallographically equivalent sites of each type. An exact treatment of the effect of an arbitrarily directed field on this material would therefore require consideration of at least 16 magnetic sublattices.

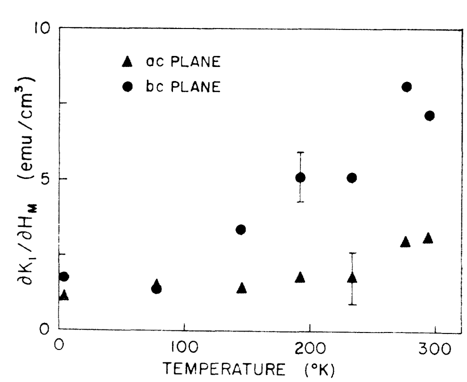

FIG. 6. Temperature dependence of the anisotropic magnetization components $M_{1a}=-\partial K_{1a}'/\partial H_M$ (triangles) and $M_{1b}=-\partial K_{1b}'/\partial H_M$ (circles) in $Fe_xGa_{2-x}O_3$ ($x=1.11$). The experimental errors shown apply to all points.

It is known[1,17] that in the absence of an external field the net magnetization is along the *c* axis. As to the directions of the individual moments, Bertaut *et al.*[8] found by neutron diffraction that any departure from ferrimagnetic ordering[7,6] (i.e., any canting of the spins away from the *c* axis) is nonexistent or small. This result is also supported by the absence, at 4.2°K, of any significant susceptibility along the *c* axis for fields above technical saturation. Measurements at this laboratory show that the susceptibility (which is dimensionless in the Gaussian system when referred to unit volume) is less than 0.2×10^{-5} along the *c* axis, while it is approximately 21×10^{-5} along the *a* axis. If the susceptibility at this low temperature is considered to be due mainly to rotations of the ionic moments toward the applied field, this would indicate an "average" canting of the moments from the *c* axis of no more than 5 deg.

In calculating the dipolar anisotropy it will be assumed that exchange interactions maintain the moments of the ions within each type of site parallel to each other so that $Fe_xGa_{2-x}O_3$ can be described as a ferrimagnet composed of four sublattices, each having a sublattice moment and a sublattice anisotropy. It will further be assumed that intersublattice exchange is strong enough, even in the presence of a field, to maintain the original coaxial orientations of the sublattices relative to each other. The dipolar contribution (K_{1a}^{dip}) to the anisotropy energy in the *ac* plane of the entire ferrimagnet will then be the difference between its dipolar energy when the net moment is oriented along the *a* (or *x*) direction and when it is oriented along the *c* (or *z*) direction. Thus one obtains

$$K_{1a}^{\mathrm{dip}}=\tfrac{1}{2}(4\mu^2/V)\sum_{i,j} s_{ij}\nu_i\nu_j\Phi_{ij}^{a}, \tag{23}$$

$$\Phi_{ij}^{a}=-3\sum_{|\mathbf{r}|<R}\frac{x^2-z^2}{|\mathbf{r}|^5}. \tag{24}$$

Here *i* and *j* each run over the four types of lattice sites, taking on the value 1 for Fe_{I}, 2 for Fe_{II}, 3 for Ga_{I}, and 4 for Ga_{II}. The factor 4 represents the four equivalent sites per unit cell, μ is the magnetic moment of each ion, V is the volume of the unit cell, the factor $\frac{1}{2}$ is

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[17] E. A. Wood, Acta Cryst. 13, 682 (1960).

inserted to prevent double counting, ν_i is the fractional occupation of the sublattice i, and s_{ij} takes the value $+1$ or -1 depending upon whether sublattices i and j are parallel or antiparallel to each other. In Eq. (24) the sum is taken over all the ions in sublattice j that fall within a radius R of a given central ion in sublattice i. The radius R taken is large enough to include all ions that effectively contribute to the anisotropy. The vector $\mathbf{r}$, with components x, y, z, connects the central ion of sublattice i with a given ion in sublattice j. An analogous set of equations can be written for K_{1b}^{dip} by replacing a by b and x by y in Eqs. (23) and (24).

A computer was used to calculate the sums Φ_{ij} for both the *ac* and *bc* planes of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$. The positions of the sites in the unit cell were taken from the x-ray studies of Abrahams *et al.*[5] on a crystal which they describe as $x>1.0$. The unit-cell dimensions, however, were taken from x-ray measurements on a powdered sample grown in the same lot as the sample used in this work. The sums Φ_{ij} were carried out over all those ions of sublattice j which are located in 20 successive spherical shells of 5 Å thickness about a central ion of sublattice i. At least 4600 ions were considered for each pair of i, j values. The results in the first shell of each sum were checked by hand calculations. As expected, the value of the sums stayed reasonably constant beyond the first few shells from the central ion, although some oscillation about an average was observed. These average values are given in Table III.

The ferrimagnetic ordering of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ on the sites 1, 2, 3, and 4 was first determined by Mössbauer measurements[7,8] to be $+-+-$. The sublattice occupations for a sample with $x=1.15$ were found by neutron diffraction studies[8] to be $\nu_1=0.73$, $\nu_2=0.87$, $\nu_3=0.62$, and $\nu_4=0.08$. For use in estimating the dipolar anisotropy of the specimen used in this work, these occupations must be corrected to apply to a crystal with $x=1.11$. For such a crystal the number of ferric ions per unit cell is

$$4(\nu_1+\nu_2+\nu_3+\nu_4)=8x=8.88\,,$$

where the factor 4 represents the number of equivalent sites per unit cell and the factor 8 gives the number of formula units per unit cell.

The spontaneous magnetization at 4.2°K for this crystal has been found to be 33.9 emu/gm. Using this value for 0°K, we have

$$\nu_1-\nu_2+\nu_3-\nu_4=2M_{00}V/(8\mu)=0.417\,.$$

Thus the sum of the two "spin-up" site occupations $(\nu_1+\nu_3)$ is 1.319 and the sum of the two "spin-down" occupations $(\nu_2+\nu_4)$ is 0.901. The corresponding numbers for Bertaut's[8] crystal $(x=1.15)$ are 1.350 and 0.950, respectively.

There are insufficient data to predict exactly the division of the occupations between the two sites in each group. If it is assumed that these occupations change proportionally as x goes from 1.15 to 1.11, however, individual sublattice occupations of 0.715, 0.826, 0.606, and 0.076 are derived for $x=1.11$. Using these results and Eq. (23), the values for the dipolar contribution to the anisotropy of such crystals are found to be $K_{1a}^{\text{dip}}=1.22\times10^6$ erg/cm^3 (or 0.281 cm^{-1} per ion), $K_{1b}^{\text{dip}}=4.19\times10^6$ erg/cm^3 (or 0.964 cm^{-1} per ion).

TABLE III. Intra- and intersublattice dipolar sums Φ_{ij} (see text) in units of 10^{24} cm^{-3} for $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ using the unit-cell dimensions for $x=1.11$. In each pair the upper number is calculated in the *ac* plane and the lower number is calculated in the *bc* plane.

Sublattice	Fe$_{\text{I}}$	Fe$_{\text{II}}$	Ga$_{\text{I}}$	Ga$_{\text{II}}$
Fe$_{\text{I}}$	0.003 0.014	0.000 −0.042	−0.011 0.100	0.005 −0.033
Fe$_{\text{II}}$		0.099 0.150	−0.018 −0.090	−0.058 −0.027
Ga$_{\text{I}}$			0.121 0.171	−0.044 −0.086
Ga$_{\text{II}}$				0.096 0.148

The dipolar sums given in this section were calculated some time ago and are referred to by Dweck[10] in analyzing the results of his ferrimagnetic resonance measurements. As the present paper was being prepared, Levine, Nowlin, and Jones[18] very kindly sent us a preprint of a paper that also included dipolar calculations for $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$. Their results give $K_{1a}^{\text{dip}}=1.55\times10^6$ erg/cm^3 and $K_{1b}^{\text{dip}}=5.24\times10^6$ erg/cm^3. In making their calculations they estimated their occupation numbers by linear interpolation between certain assumed conditions[18] at the two ends of the range $0.7<x<1.4$ in which the composition parameter x of this material can be varied. However, even when we use their occupation numbers with our dipolar sums, agreement between the two calculations is not achieved. Levine *et al.* concluded that dipolar processes are the sole source of anisotropy in the *bc* plane of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$, and that in the *ac* plane some sort of anisotropic exchange also contributes. As will be shown in Sec. VII, our conclusions are somewhat different.

VII. OTHER SOURCES OF ANISOTROPY

The bulk magnetic moment of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ is the sum of several sublattice moments, each with a different temperature dependence. Because of this, the fact that the anisotropy K_1 seems to vary linearly[4] with M_{00}^2 over a rather large temperature range has little bearing on the determination of the sources of anisotropy in this material. Other experimental facts, however, do permit the following comments.

The measured values of the total orthorhombic anisotropy of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ at 4.2°K were found to be

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[18] B. F. Levine, C. H. Nowlin, and R. V. Jones, Phys. Rev. **174**, 571 (1968).

$K_a = 3.22\times10^6$ erg/cm^3 and $K_b = 5.65\times10^6$ erg/cm^3. As noted earlier, these values were found from the values of the torque maximum extrapolated to $H_M = 0$, and they include both first- and second-order terms. The contribution to the first-order anisotropy at $T = 0$°K due to classical magnetic dipolar processes has been found to be $K_{1a}^{\text{dip}} = 1.22\times10^6$ erg/cm^3 and $K_{1b}^{\text{dip}} = 4.19\times10^6$ erg/cm^3. If it is assumed that the conditions at 4.2°K will be little different from those at 0°K, this leaves a residual anisotropy of $K_a^{\text{res}} = 2.00\times10^6$ erg/cm^3 and $K_b^{\text{res}} = 1.46\times10^6$ erg/cm^3. It is this residual anisotropy which must now be explained.

An estimate of the anisotropy due to the symmetric[19] part of the anisotropic exchange (such as pseudodipolar exchange) can be made by using the expression $(2-g)^2J/g^2$, where g may be taken here to represent the "average" g factor of the ions at the various sites and J the "average" isotropic exchange energy of these ions. In a complicated system such as $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ these average quantities can only be estimated. By using the paramagnetic Curie temperature of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ which Bertaut[8] *et al.* find to be 1180°K, one estimates $J \approx 1.63\times10^{-13}$ erg. Dweck,[10] from ferrimagnetic resonance studies of this crystal, found the value $g = 1.96$, which would give an anisotropic exchange of 6.52×10^{-17} erg and hence a corresponding anisotropy of 0.14×10^6 erg/cm^3. Dweck's value of g is a bulk "effective" g for the crystal as a whole, and can be quite different from the ionic g values involved. A better estimate may be achieved by using the value $g = 2.00\pm0.01$ found by Folen[20] in his paramagnetic resonance studies of β-Ga_2O_3:Fe^{3+}, a crystal whose structure is significantly more distorted than that of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$. Thus the g factor of β-Ga_2O_3:Fe^{3+} should be more anomalous than that of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$. Nevertheless, Folen's value of g would give, as an upper limit, an anisotropic exchange of only 0.41×10^{-17} erg and hence a corresponding anisotropy of 0.01×10^6 erg/cm^3. Thus, while anisotropy due to symmetric exchange may be present, its contribution is probably negligible.

The antisymmetric (Dzyaloshinskii-Moriya)[21–23] part of the anisotropic exchange can also lead to a bulk anisotropy. An estimate of this contribution to the anisotropy can be made by using the expression[21] $(2-g)J/g$. Dweck's value of g gives an antisymmetric exchange of 3.26×10^{-15} erg and hence a corresponding anisotropy of 6.93×10^6 erg/cm^3, while Folen's value of g gives, as an upper limit, an antisymmetric exchange of 0.82×10^{-15} erg and hence an anisotropy of 1.73×10^6 erg/cm^3. The canting angle β produced by antisymmetric exchange is given by the formula[21] $\tan 2\beta \sim (2-g)/g$. Folen's g value gives an angle β of at most 1°, which is well within the limit of 5° found in Sec. VI. Thus, antisymmetric exchange may be a major contributor to the anisotropy in $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$.

The remaining source of orthorhombic anisotropy is single-ion crystal-field processes. In an orthorhombic ferromagnet the anisotropy due to single-ion processes K^{si} can be found by using the lengthy expressions[13] whose leading terms are

$$K_{1a}^{si} = -N(D-E)S(S-\tfrac{1}{2}), \tag{25a}$$

$$K_{1b}^{si} = -N(D+E)S(S-\tfrac{1}{2}), \tag{25b}$$

where N is the number of magnetic ions per unit volume, S is the ionic spin, D is the quadratic axial crystal-field parameter, and E is the quadratic rhombic crystal-field parameter. Both D and E may include overlap and covalency effects.[24] Gallium iron oxide is, of course, a ferrimagnet and each of its four sites will have its own D and E, but Eq. (25) can be used to estimate the average D and E that would be needed to give single-ion anisotropies equal to the residual anisotropies mentioned above. If S is taken to be $\frac{5}{2}$, we find that D is equal to -1.81×10^{-17} erg or -0.090 cm^{-1} and E is equal to 0.29×10^{-17} erg or 0.014 cm^{-1}. These values can be compared to those describing the ferric ion in the axially distorted cubic sites[25] of Al_2O_3, where $D = 0.1719$ cm^{-1}, or to those corresponding to the ferric ion in one of the octahedral sites[26] in kyanite (Al_2SiO_5), where D has been found to be 1.3 cm^{-1} and E to be 0.057 cm^{-1}. Therefore, the values of D and E needed to explain the residual anisotropy of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$ as being due to crystal-field processes are quite reasonable, especially as the sites in this material are probably even more distorted then those in the examples given above.

Thus, we conclude that while dipolar interactions do contribute considerably to the anisotropy of $\text{Fe}_x\text{Ga}_{2-x}\text{O}_3$, antisymmetric exchange and one-ion anisotropy are both capable of making significant contributions as well. The paucity of information presently available prohibits us from identifying the residual anisotropy in more detail.

Except near 0°K, the anisotropic magnetization found in this material can most probably be explained, following Callen and Callen,[12] by a statistical treatment of the same mechanisms that give rise to the anisotropy. In principle, the exact contributions of these mechanisms can be calculated but in practice such calculations cannot be made without a knowledge of the exchange energies and crystal-field parameters of the ferric ions on the various sites. Near 0°K, on the other hand, the statistical contributions must vanish, so that, according

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[19] J. Kanamori, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, p. 127.

[20] V. J. Folen (to be published).

[21] T. Moriya, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I, p. 85.

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[23] T. Moriya, Phys. Rev. **120**, 91 (1960).

[24] J. Kondo, Progr. Theoret. Phys. (Kyoto) **23**, 160 (1960); R. R. Sharma, T. P. Das, and R. Orbach, Phys. Rev. **155**, 338 (1967).

[25] G. S. Bogle and H. F. Symmons, Proc. Phys. Soc. (London) **73**, 531 (1959).

[26] G. P. Troup and D. R. Hutton, Brit. J. Appl. Phys. **15**, 1493 (1964).

to Rado,[13] the anisotropic magnetization possibly arises from the anisotropy of the g factor or from some "admixture"-type mechanism.

VIII. DIRECT MEASUREMENT OF ANISOTROPIC MAGNETIZATION

Considering the large magnitude of the anisotropic magnetization indicated by the torque data, it was thought that a direct measurement of this phenomenon would be of interest. A vibrating sample magnetometer[27] was used for this study. Such an instrument measures that component of the moment of the sample that is directed along an externally applied field. If this field is applied along the easy axis of the material, then the moment measured corresponds to "technical saturation" provided the field is high enough to sweep out any domain structure. If the field is applied along one of the other major axes, then the component of the moment in the field direction will not correspond to "technical saturation" until the magnitude of the field reaches the "anisotropy field" of the material. While theoretically a sharp change in the slope of the M versus H curve is predicted at this point, in practice a more rounded change in slope is expected. If no anisotropic magnetization is present, the moments measured at this field and above will be the same as the corresponding moments measured in the easy direction in this same field range. If anisotropic magnetization is present, the moments measured in the two directions in this field range will be different. As the applied field is increased above the anisotropy field, this difference will be maintained, though both moments may increase slightly, due to the finite susceptibility of the material.

The sample of $Fe_xGa_{2-x}O_3$ was cut in half and one of the halves mounted in the magnetometer so that measurements could be made along either the c or the a crystal axis. The field available to the vibrating sample magnetometer was limited to 18 kOe, so that meaningful measurements could only be made in the ac plane and at the two highest temperatures, namely, room temperature and 276°K. Fortunately, these are the temperatures where the expected anisotropic moment would be the greatest. Figure 7 shows the curves measured at 276°K. The value of M_{1a} found was -4.4 emu/cm³. This is to be compared to the value of 3.0 emu/cm³ found from the torque measuremenst. The agreement is not unreasonable considering the large error of both experiments. The values of spontaneosu magnetization ($M_{00}=47.8$ emu/cm³) and susceptibility ($\chi=9.7\times10^{-4}$) deduced from Fig. 7 are in good agreement with those found above. At room temperature the anisotropic moment was found to be 4.7 emu/cm³, which is again comparable with the value (3.1 emu/cm³) found from the torque-curve analysis. Although these direct measurements were rather crude (because of the smallness of the sample) and limited to two temperatures, they do substantiate the presence of a large anisotropic spontaneous magnetization in $Fe_xGa_{2-x}O_3$. It should be noted, moreover, that the parallelism of the two curves of Fig. 7 at high fields is consistent with the assumption made in analyzing the torque curves (see Sec. II) that the anisotropic magnetization is spontaneous rather than induced.

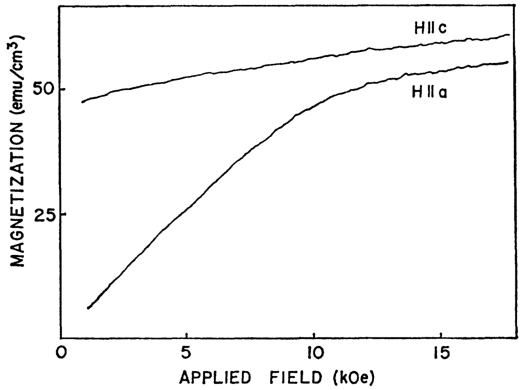

FIG. 7. Direct measurement of magnetization at 276°K in the ac plane of $Fe_xGa_{2-x}O_3$ ($x=1.11$). The two curves represent, respectively, the component of the magnetization along the applied field when the latter is directed along the a and c axes of the crystal. The anisotropy of the spontaneous magnetization is shown by the separation of these curves in the (high-field) region where they are parallel.

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[27] S. Foner, Rev. Sci. Instr. **30**, 548 (1959). The instrument used in the present work was built by A. Pinto.