Antiferromagnetic Resonance in Systems with Dzyaloshinsky-Moriya Coupling; Orientation Dependence in αFe_2O_3

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Resonance conditions for an antiferromagnet whose sublattice magnetization vectors are canted by the Dzyaloshinsky-Moriya interaction are generalized to include an arbitrary angle θ between the applied magnetic field and the hard direction of magnetization, and the resulting normal modes are discussed. A correction term to the low-frequency resonance expressions derived by Pincus may be appreciable at higher magnetic fields and small θ. For suitable parameters, this term permits the measurement of all pertinent effective fields by resonance experiments. It is furthermore shown that demagnetization effects are negligible. Microwave resonance experiments as a function of field (to 60 kG), angle, and frequency (12 to 35 Gc/sec) on synthetic single crystals of αFe_2O_3 failed to reveal a departure from the Pincus relation, thus leading to the conclusion that $H_A/H_E < 10^{-2}$.

I. INTRODUCTION

MAGNETIZATION measurements reveal that many systems, which are known from neutron diffraction studies to be antiferromagnetic, exhibit a weak ferromagnetism, with a moment several orders of magnitude smaller than the sublattice magnetization. This has been attributed to a canting of the sublattice moments caused by one or a combination of two effects: single-ion magnetocrystalline anisotropy as is discussed by Moriya,[1] which exists in[2] $KMnF_3$ and NiF_2,[3] or a spin-spin interaction of the form $\mathbf{D}'\cdot(\mathbf{S}_1\times\mathbf{S}_2)$, which is found in αFe_2O_3, $MnCO_3$, $CoCO_3$,[4] CrF_3, and possibly FeF_3 and $CuCl_2\cdot 2H_2O$.[5,6] This latter interaction was proposed by Dzyaloshinsky[7] on the basis of a thermodynamic argument, and its physical origin has been shown by Moriya[5] to be an anisotropic superexchange proportional to the spin-orbit coupling term of the one-electron Hamiltonian. A phenomenological discussion has been given by Vonsovsky and Turov.[8] Resonance conditions for an antiferromagnet with this Dzyaloshinsky-Moriya coupling have been formulated by many authors,[9–12] but the first paper to point out the importance of the small anisotropy in the easy plane was that of Pincus.[11] Their results are extended in this paper.

Expressions derived by Pincus for αFe_2O_3, a rhombohedral crystal which above the Morin temperature at $-14°C$ has its sublattice moments lying in the [111] or "c" plane, indicate a zero-field splitting of the modes such that one exists in the 3-cm microwave region and the other in the submillimeter region. His relations for the system in an externally applied magnetic field $\mathbf{H}_0$ were given for the case of $\mathbf{H}_0$ perpendicular to [111]; i.e., $\theta=90°$. Extension of these relations to arbitrary θ by substituting $H_0 \sin\theta$ for H_0 has been made by several authors in order to compare their low-frequency resonance data with the theory. Such a procedure leads to equations which indicate that this resonance is not observable at finite fields for $\theta=0$. We shall show that an additional term becomes appreciable when θ is small, so that in fact resonance could be achieved at finite fields. An additional term also appears in the high-frequency expression, which then agrees with the prediction of Turov and Guseinov[12,13] [their low-frequency expression does not include the influence of the important (111) plane anisotropy]. In addition to deriving the resonances for arbitrary angle, we show that demagnetizing effects are small even for this case of weak ferromagnetism and the normal modes are similar to those in the hexagonal antiferromagnet $CsMnF_3$.[14]

In an attempt to observe the presence of the correction term to the low-frequency Pincus equation, room-temperature resonance experiments on synthetic single crystal αFe_2O_3 have been performed at various θ; however, the value of H_A/H_E for this system is apparently not sufficiently large to permit the term to be seen with the fields employed (see note added in proof).

II. RESONANCE CONDITIONS

We shall use the molecular field approximation and notation in a manner similar to that employed by Pincus.[11] We consider, as shown in Fig. 1, two magnetic

* National Science Foundation Predoctoral Fellow.
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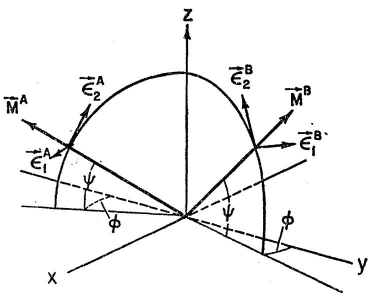

FIG. 1. Notation describing the coupled sublattice magnetizations $\mathbf{M}^A$ and $\mathbf{M}^B$ at equilibrium. The three vectors $\mathbf{M}^A$, $\boldsymbol{\varepsilon}_1{}^A$, and $\boldsymbol{\varepsilon}_2{}^A$ are mutually orthogonal as are the three vectors $\mathbf{M}^B$, $\boldsymbol{\varepsilon}_1{}^B$, and $\boldsymbol{\varepsilon}_2{}^B$, both $\boldsymbol{\varepsilon}_1{}^A$ and $\boldsymbol{\varepsilon}_1{}^B$ being parallel to the x-y plane. $\mathbf{M}^A$ and $\mathbf{M}^B$ are at an angle ψ to this plane, and their projections on the plane are at an angle φ to the y axis.

sublattices of magnetizations $\mathbf{M}^A$ and $\mathbf{M}^B$ coupled antiparallel by the usual symmetric exchange interaction expressed in pseudo-dipole form $\lambda\mathbf{M}^A\cdot\mathbf{M}^B$, confined to lie in the "c" plane by an energy characterized by the anisotropy constant K, and oriented in that plane by a small anisotropy constant K'. The parameters λ, K, and K' are taken to be positive. In addition, $\mathbf{M}^A$ and $\mathbf{M}^B$ are tipped toward each other by the antisymmetric exchange interaction $\mathbf{D}\cdot\mathbf{M}^A\times\mathbf{M}^B$ and out of the easy plane by the applied field $\mathbf{H}_0$, which is assumed to exert a much larger influence than the in-plane anisotropy so that the net moment tends to lie along $\mathbf{H}_0$. We assume there is more than twofold symmetry about the c axis, which according to Moriya[5] dictates that $\mathbf{D}$ must lie along that axis. Systems such as αFe_2O_3 and $MnCO_3$ actually have three equivalent twofold axes in the c plane, leading to a sixfold anisotropy in the resonance field as $\mathbf{H}_0$ is rotated in that plane. This symmetry has been observed in αFe_2O_3 by Tasaki and Iida,[15] and for the appropriate systems can be included in the resonance expressions by giving the c-plane anisotropy constant a $\sin 6\varphi_0$ dependence, where φ_0 is the angle between the x axis and c-plane component of $\mathbf{H}_0$.[14] We shall ignore this dependence in the following derivation.

The equations of motion for the A and B sublattices are then

$$\begin{aligned} d\mathbf{M}^A/\gamma dt= &-\lambda\mathbf{M}^A\times\mathbf{M}^B+\mathbf{M}^A\times\mathbf{H}_0 \\ &-\mathbf{M}^A\times[\mathbf{N}\cdot(\mathbf{M}^A+\mathbf{M}^B)]-\mathbf{M}^A\times(\mathbf{D}\times\mathbf{M}^B) \\ &+KM_z{}^A(\mathbf{k}\times\mathbf{M}^A)-K'M_y{}^A(\mathbf{j}\times\mathbf{M}^A), \quad (1\text{A}) \end{aligned}$$

$$\begin{aligned} d\mathbf{M}^B/\gamma dt= &-\lambda\mathbf{M}^B\times\mathbf{M}^A+\mathbf{M}^B\times\mathbf{H}_0 \\ &-\mathbf{M}^B\times[\mathbf{N}\cdot(\mathbf{M}^A+\mathbf{M}^B)]+\mathbf{M}^B\times(\mathbf{D}\times\mathbf{M}^A) \\ &+KM_z{}^B(\mathbf{k}\times\mathbf{M}^B)-K'M_y{}^B(\mathbf{j}\times\mathbf{M}^B). \quad (1\text{B}) \end{aligned}$$

The demagnetization tensor $\mathbf{N}$ is assumed to be diagonal with elements N_x, N_y, N_z that are not necessarily equal. A more convenient notation expresses the energy parameters in terms of effective magnetic fields proportional to M, the magnitude of $\mathbf{M}^A$ (which equals the magnitude of $\mathbf{M}^B$). We let $H_E=\lambda M$, $H_{DM}=DM$, $H_A=KM$, and $H_A'=K'M$. Typically, $H_E\approx10^6$ Oe, $H_{DM}\approx10^4$ Oe, $H_A\approx10^4$ Oe, and $H_A'\approx1$ Oe, but variations by a factor of 10 or more from these values should be expected for certain systems. In the following treatment we consider H_0 to be the same order of magnitude as H_{DM} and H_A; M is assumed to be 10^2 emu and N is of order 4π. Some care must be employed in preserving various orders because the expansions for the resonance conditions become somewhat complicated, and the lowest order terms eventually cancel exactly.

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By setting the total effective field experienced by $\mathbf{M}^A$ along $\boldsymbol{\varepsilon}_1{}^A$ equal to zero, we determine the equilibrium value of φ:

$$\varphi=\frac{H_0\sin\theta\cos\varphi+H_{DM}\cos2\varphi}{2H_E\cos\psi+2N_xM+H_A'} \approx\frac{H_0\sin\theta+H_{DM}}{2H_E}\left(1-\frac{N_xM}{H_E}\right). \quad (2)$$

The next order term in Eq. (2) is smaller by a factor of 10. Similarly, the equilibrium value of ψ requires the effective field along $\boldsymbol{\varepsilon}_2{}^A$ to be zero, so that

$$\psi=\frac{H_0\cos\theta\cos\psi}{H_E\cos\psi(1+\cos^2\varphi-\sin^2\varphi)+H_A+2N_zM} \approx\frac{H_0\cos\theta}{2H_E}\left(1-\frac{H_A+2N_zM}{2H_E}\right). \quad (3)$$

For the stipulated fields both φ and ψ are about 10^{-2}. The normal modes of the system will be obtained by assuming that $\mathbf{M}^A$ and $\mathbf{M}^B$ depart only slightly from these equilibrium positions, so that Eqs. (1) can be linearized. It is convenient to treat the vector operations of these equations in the Cartesian system; consequently, for the equilibrium values of the magnetizations we have the component decomposition

$$\mathbf{M}_0{}^{A,B}=M(\sin\varphi\cos\psi\mathbf{i}\mp\cos\varphi\cos\psi\mathbf{j}+\sin\psi\mathbf{k}). \quad (4\text{A,B})$$

In our notation, the upper sign is associated with the first named index. Departures of the magnetizations from $\mathbf{M}_0{}^{A,B}$ can be expressed in terms of the independent variables $M_1{}^{A,B}$ and $M_2{}^{A,B}$, the projections of $\mathbf{M}^{A,B}$ along the vectors $\boldsymbol{\varepsilon}_1{}^{A,B}$ and $\boldsymbol{\varepsilon}_2{}^{A,B}$ respectively of Fig. 1. These departures are then projected onto the $\mathbf{i}$, $\mathbf{j}$, $\mathbf{k}$ coordinates as:

$$\begin{aligned} \delta\mathbf{M}^{A,B}= &(\pm M_1{}^{A,B}\cos\varphi-M_2{}^{A,B}\sin\psi\sin\varphi)\mathbf{i} \\ &+(M_1{}^{A,B}\sin\varphi\pm M_2{}^{A,B}\sin\psi\cos\varphi)\mathbf{j} \\ &+(M_2{}^{A,B}\cos\psi)\mathbf{k}. \quad (5\text{A,B}) \end{aligned}$$

The Cartesian equations of motion are rather lengthy and will not be given here. Only terms which contain one power of $\delta M_i{}^B$ appear, since zero-order terms cancel at equilibrium, and those to higher than first order are neglected. To convert back to our $\boldsymbol{\varepsilon}_i^{A,B}$ coordinate system we use the transformations

$$\begin{aligned}\mathbf{i}&=\pm\cos\varphi\boldsymbol{\varepsilon}_1^{A,B}-\sin\varphi\sin\psi\boldsymbol{\varepsilon}_2^{A,B},\\ \mathbf{j}&=\sin\varphi\boldsymbol{\varepsilon}_1^{A,B}\pm\cos\varphi\sin\psi\boldsymbol{\varepsilon}_2^{A,B},\\ \mathbf{k}&=\cos\psi\boldsymbol{\varepsilon}_2^{A,B}.\end{aligned}\tag{6A,B}$$

Equations (1A,B) then yield the equations of motion:

$$dM_1^A/dt=\gamma(AM_2^A+BM_1^B+CM_2^B),\tag{7a}$$

$$dM_2^A/dt=\gamma(EM_1^A+FM_1^B+BM_2^B),\tag{7b}$$

$$dM_1^B/dt=\gamma(-BM_1^A+CM_2^A+AM_2^B),\tag{7c}$$

$$dM_2^B/dt=\gamma(FM_1^A-BM_2^A+EM_1^B),\tag{7d}$$

where to order 10^2 Oe the coefficients are

$$\begin{aligned}A&=(1-\tfrac{3}{2}\varphi^2-\tfrac{3}{2}\psi^2)H_E+H_A+(\psi\cos\theta+\varphi\sin\theta)H_0\\&\quad+2\varphi H_{DM}+N_zM+H_A',\\ B&=2\varphi\psi H_E-\psi H_{DM},\\ C&=(1+\tfrac{1}{2}\varphi^2-\tfrac{3}{2}\psi^2)H_E+N_zM,\\ E&=(-1+\tfrac{3}{2}\varphi^2+\tfrac{3}{2}\psi^2)H_E-(\psi\cos\theta+\varphi\sin\theta)H_0\\&\quad-2\varphi H_{DM}-N_xM-H_A',\\ F&=(1-\tfrac{3}{2}\varphi^2+\tfrac{1}{2}\psi^2)H_E+2\varphi H_{DM}+N_xM.\end{aligned}\tag{8}$$

Equations (7) can be simplified by substituting new variables defined by

$$\begin{aligned}m_1^\pm&=(M_1^A\pm M_1^B)/2,\\ m_2^\pm&=(M_2^A\pm M_2^B)/2,\end{aligned}\tag{9}$$

so that the equations of motion become

$$\begin{aligned}\dot{m}_1^\pm/\gamma&=\mp Bm_1^\mp+(A\pm C)m_2^\pm,\\ \dot{m}_2^\pm/\gamma&=\mp Bm_2^\mp+(\pm F+E)m_1^\pm.\end{aligned}\tag{10}$$

Differentiating and combining terms, we obtain

$$\begin{aligned}\ddot{m}_1^\pm/\gamma^2&=[(A\pm C)(E\pm F)-B^2]m_1^\pm\mp 2BAm_2^\mp,\\ \ddot{m}_2^\pm/\gamma^2&=[(A\pm C)(E\pm F)-B^2]m_2^\pm\mp 2BEm_1^\mp.\end{aligned}\tag{11}$$

Recognizing that $A\approx E$ to order 10^6 Oe, we can set $BE\approx BA$. Furthermore the B^2 terms in the brackets can be neglected, so that

$$\begin{aligned}\ddot{m}_{1,2}^+/\gamma^2&=-\alpha m_{1,2}^+-\eta\beta m_{2,1}^-,\\ \ddot{m}_{1,2}^-/\gamma^2&=-\beta m_{1,2}^-+\eta\beta m_{2,1}^+,\end{aligned}\tag{12}$$

where

$$\begin{aligned}\alpha&=-(A+C)(E+F),\\ \beta&=-(A-C)(E-F),\end{aligned}\tag{13}$$

$$\eta=2BA/\beta\approx H_0^2\sin\theta\cos\theta/\beta.\tag{14}$$

The equations for m_1^+ and m_2^- are thereby decoupled from the equations for m_1^- and m_2^+. It is apparent that permitting $\mathbf{H}_0$ to be at an arbitrary angle has coupled these variables in pairs, and that when $\theta=0$ or $\pi/2$ or when $H_0=0$, the $m_i^\pm$ are the normal modes. These two sets of equations (12) have identical secular equations with eigenvalues approximately given by

$$\begin{aligned}(\omega_1/\gamma)^2&=\alpha+\eta^2\beta,\\ (\omega_2/\gamma)^2&=\beta(1-\eta^2).\end{aligned}\tag{15}$$

Expanding these, we obtain

$$\begin{aligned}(\omega_1/\gamma)^2=\{&H_0\sin\theta(H_0\sin\theta+H_{DM})(1-N_xM/H_E)\\&+2H_EH_A'+H_AH_0^2(\cos^2\theta-\sin^2\theta)/2H_E\\&-H_AH_{DM}H_0\sin\theta/2H_E\\&+H_0^4\sin^2\theta\cos^2\theta/2H_EH_A\}\end{aligned}\tag{16}$$

$$\begin{aligned}\approx &H_0\sin\theta(H_0\sin\theta+H_{DM})\\&+2H_EH_A'+H_AH_0^2\cos^2\theta/2H_E,\end{aligned}\tag{17}$$

$$\begin{aligned}(\omega_2/\gamma)^2=&2H_EH_A+H_{DM}(H_0\sin\theta+H_{DM})\\&+H_0^2\cos^2\theta+2N_xMH_A.\end{aligned}\tag{18}$$

We note that terms neglected in going from Eq. (16) to (17) would give a 1% correction to H_{DM} and to γ if these parameters were measured using Eq. (17). Equations (17) and (18) reduce to the expressions derived by Pincus when $\theta=90°$, and the high-frequency relation (18) agrees with the results of Turov and Guseinov if $N_x=0$.[12,13] The last term of (17) is an addition to the Pincus relations, which *in principle* permits all of the effective fields to be determined solely from high- and low-frequency resonance experiments. This will be examined in more detail later.

III. NORMAL MODES

The fact that Eqs. (12) consist of two sets of decoupled relations initially permits us to introduce two arbitrary amplitudes, say $m_1^-=\zeta\exp(i\omega t)$ and $m_2^-=\xi\exp(i\omega t)$. Then using Eqs. (12) to determine m_1^+ and m_2^+ and reverting back to the magnetization deviation variables by the aid of Eqs. (9), we obtain

$$\begin{aligned}M_1^{A,B}&=\pm\zeta+\delta\xi,\\ M_2^{A,B}&=\pm\xi+\delta\zeta,\end{aligned}\tag{19}$$

where $\delta=1/\eta$ if $\omega=\omega_1$ and $\delta=\eta$ if $\omega=\omega_2$. Any of the equations of motion (7) relates ξ and ζ, so that by using (7a) we have

$$\frac{\xi}{\zeta}=\frac{\delta(A+C)-(B+i\omega)}{\delta(i\omega-B)-(A-C)}.\tag{20}$$

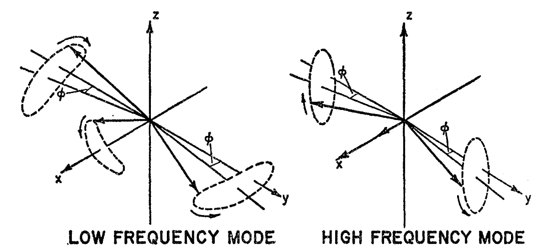

FIG. 2. Normal modes with $\mathbf{H}_0=0$. The net magnetization of the low-frequency mode precesses about the x axis, whereas for the high-frequency mode, it lies along the axis. Modes degenerate with these have magnetizations precessing in the opposite sense.

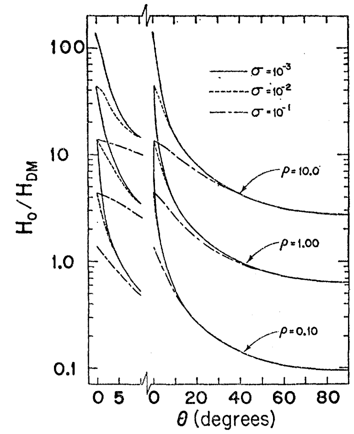

FIG. 3. Normalized theoretical curves for the dependence of the resonance field on orientation, Eq. (23). The parameters are $\rho=[(\omega/\gamma)^2-2H_EH_A']/H_{DM}^2$ and $\sigma=H_A/H_E$. Curves for $\sigma=0$ and $\sigma=10^{-3}$ would differ significantly only in the region $\theta\lesssim 0.5°$.

The modes can most easily be illustrated in the limit $H_0=0$, for which $\eta=0$. Then the amplitude ratio is

$$\begin{aligned}&\text{for}\quad \omega=\omega_1:\quad \xi/\zeta=-i(2H_E/H_A')^{1/2},\\&\text{for}\quad \omega=\omega_2:\quad \xi/\zeta=i(2H_E/H_A)^{1/2}.\end{aligned}\tag{21}$$

The magnitudes of these quantities are the ratios of major to minor axes of the ellipses traced out by $\mathbf{M}^A$ and $\mathbf{M}^B$, as shown in Fig. 2. In the indicated limit, the Dzyaloshinsky-Moriya coupling is influential only in the equilibrium canting of the magnetizations and plays no part in the first-order motion. The illustrated modes are similar to those of a hexagonal antiferromagnet such as $CsMnF_3$ which has a hard c axis with in-plane anisotropy but no $\mathbf{D}\cdot\mathbf{M}^A\times\mathbf{M}^B$ coupling.[14] However, when $H_0\neq 0$, H_{DM} *can* play an important role. For example, when $H_0H_{DM}\gg 2H_EH_A'$ and $\theta=90°$, the ratio of the axis for the low-frequency mode, Eq. (20), reduces to

$$\xi/\zeta=-i2H_E/(H_0^2+H_0H_{DM})^{1/2}.\tag{22}$$

Fink has derived the rf susceptibilities[16] and linewidths[17] for both the Dzyaloshinsky-Moriya coupled systems and those with single ion anisotropy; consequently, these properties will not be considered here. His equations for the former coupling are still valid, provided Eq. (17) is used for the resonance frequency.

IV. EXPERIMENTAL RESULTS

The high-frequency resonance of Eq. (18) has been observed in only one system, $MnCO_3$, by Fink and Shaltiel[13] and Richards,[18] who find good agreement with experiment (see note added in proof). We shall not consider this mode further.

The low-frequency mode in $MnCO_3$ has been investigated by Date[19] and Borovik-Romanov *et al.*,[20] and in αFe_2O_3 by Anderson *et al.*,[21] Kumagai *et al.*,[22] and by Tasaki and Iida.[15] Frequency dependence on field at $\theta=90°$ has been verified up to about 15 kOe, but the orientation dependence has been examined only between $\theta=90°$ and $\theta\approx 20°$ which is apparently not sufficiently close to the c axis to unambiguously reveal the presence of the last term in Eq. (17). Departures from the Pincus relation are displayed in the published figures for the smaller angles.[19,22] In attempting to see whether these deviations were significant and revealed the $\cos^2\theta$ term, we conducted resonance experiments on αFe_2O_3, giving particular attention to the small θ, large H_0 region.

For fixed frequency experiments the inverse of Eq. (17) is useful:

$$\frac{H_0}{H_{DM}}=\frac{-\sin\theta+[\sin^2\theta+2\rho(2\sin^2\theta+\sigma\cos^2\theta)]^{1/2}}{2\sin^2\theta+\sigma\cos^2\theta},\tag{23}$$

where

$$\rho=[(\omega_1/\gamma)^2-2H_EH_A']/H^2{}_{DM},$$
$$\sigma=H_A/H_E.$$

When σ is negligible this reduces to the $1/\sin\theta$ dependence of Pincus. The effect of the $\cos^2\theta$ term shown in Fig. 3, indicates that large ρ and σ favor its detection. Unfortunately this also requires a large value of H_0/H_{DM}.

Synthetic single crystal platelets of αFe_2O_3, grown and kindly furnished by Dr. A. Tasaki of the University of Tokyo, approximately 2 mm in diameter and 0.5 mm thick with the c axis perpendicular to the plane, were glued against the narrow side of a microwave guide near the end wall as shown in Fig. 4. All experiments were performed at room temperature. For the K_u band experiments, a 12-in. Varian magnet was rotated to vary θ, but for the higher frequency 8-mm experiments a transverse access Bitter solenoid was used and the waveguide itself was rotated by means of a rotating waveguide joint. The applied field H_0 was modulated at 100 cps and the reflected microwave signal was monitored by a lock-in amplifier.

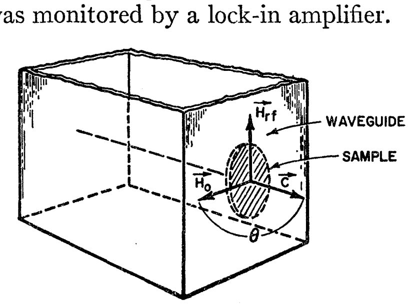

FIG. 4. Geometry of microwave resonance experiments. The direction of H_{rf} was chosen so that its coupling to the weak magnetic moment would be essentially independent of θ.

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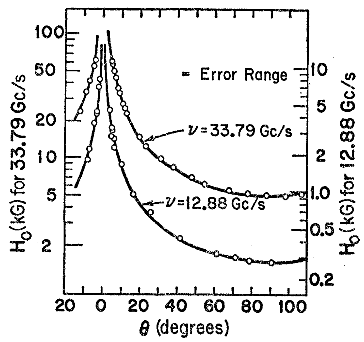

FIG. 5. Experimental results for the dependence of the resonance field on orientation for αFe_2O_3 at room temperature. The higher frequency data were obtained from a different sample than the lower. Solid curves are the respective theoretical expressions of Eq. (23) with $\sigma=0$, normalized to agree with the data at $\theta=90°$.

Measurement of H_0 versus $(\omega_1/\gamma)^2$ from 13 to 35 Gc/sec $\theta=90°$ resulted in a linear relation showing the predominant H_0H_{DM} term in Eq. (17). From this we obtain $H_{DM}=2.1\times10^4$ Oe and $2H_EH_A'=1.5\times10^7$ Oe^2, but these numbers varied by as much as 10% for the different samples investigated.

The experimental angular dependence of H_0 at 12.88 and 33.79 Gc/sec is displayed in Fig. 5. For the former the resonance was pursued up to $\theta=2°$, where the resonance became too broad to be detected. At the latter frequency, the linewidth increased from 200 Oe at $\theta=90°$ to 1700 Oe at $\theta=4.5°$; smaller values of θ could not be examined with the available fields. No evidence of the $\cos^2\theta$ term was seen at these frequencies to within the 5% experimental errors.

Since at 33 Gc/sec, $\rho\approx0.3$, the curves of Fig. 3 indicate that $\sigma=H_A/H_E$ must be less than about 10^{-2} to be undetectable with the present experimental precisions, although for small σ this is not a sensitive way to measure that ratio. Other crystals may have more favorable parameters which permit the $\cos^2\theta$ term to be detected. $MnCO_3$ offers the advantage of a small H_{DM}(3.7 kOe[19]) so that at 33 Gc/sec, $\rho=12.5$; however, H_A is known to be 2.1 kOe[13] which is a factor of 10 smaller than that suspected to exist in αFe_2O_3, and with the resulting $\sigma\approx4\times10^{-3}$, the resonance would have to be observed at $\theta<2°$ to detect the $\cos^2\theta$ term. Consequently, for αFe_2O_3 and $MnCO_3$ it will be difficult to observe this term due to line broadening, even at high fields and frequencies.

We note that a spurious indication of departure from the Pincus relation would be observed if the sample were misoriented, so that H_0 were rotated in a plane which did not contain the c axis. Such might be the case for previous measurements on $MnCO_3$,[19] although there is no indication that this affects the present experiments.

While performing the above experiments, we noticed that for some samples the resonance signal deteriorated due to the presence of H_0. While the amplitude decreased by a factor of 10 over a period of 12 min at 5 kOe, the line broadened so that the product of amplitude and linewidth at the inflection points remained constant to within 10%. This effect was independent of rf power over a 20-dB range. Annealing sufficed to prevent this except for the application of high fields (60 kOe). The cause of this effect is not yet understood.

V. CONCLUSIONS

Antiferromagnetic resonance conditions for a system with Dzyaloshinsky-Moriya coupling have been derived for $\mathbf{H}_0$ at an arbitrary direction to the hard axis and the normal modes described. An additional term to the Pincus relations is shown to be appreciable when $\mathbf{H}_0$ is nearly parallel to the axis, provided H_A/H_E is not too small. Low-frequency resonance experiments on αFe_2O_3 up to 60 kOe have failed to reveal a departure from the Pincus relations, thus indicating that H_A/H_E $<10^{-2}$. A deterioration of the resonance signal due to the presence of a magnetic field has been observed, but its origin is uncertain.

Note added in proof. We have observed antiferromagnetic and spin-flop resonances in synthetic and single-crystal αFe_2O_3 for temperatures between 4.2 and 280°K utilizing 35, 70, and 120 Gc/sec radiation and pulsed magnetic fields. The spin-flop transition has also been observed directly by differential magnetic moment measurements[23] and by Mössbauer techniques.[24] The low-temperature value of H_c is approximately 65 kG, which demonstrates that, at least below T_M, H_A is much less than 10^4 Oe estimated earlier. Extrapolation of H_A to $T=300°K$ shows that H_A is much less than 10^4 Oe so that deviations from the Pincus equations should not readily be observable in αFe_2O_3. This work will be discussed in a future publication.

[23] S. Foner, International Conference on Magnetism, 1964, Nottingham, England, Paper L3-5 (to be published).

[24] N. Blum, A. J. Freeman, and L. Grodzins, Bull. Am. Phys. Soc. 9, 465 (1964).