Effect of Antisymmetric Exchange Interaction on the Magnetization and Resonance in Antiferromagnets*

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The magnetization curves and the resonance frequencies are calculated for two types of antiferromagnets with an antisymmetric exchange interaction of the form $\mathbf{D}_{ij}\cdot\mathbf{M}_i\times\mathbf{M}_j$ and a uniaxial magnetocrystalline anisotropy term $K\sin^2\theta$. In one type of these antiferromagnets, the $\mathbf{D}$ vector is perpendicular to the easy axis. This is presumably the case in the orthoferrites. For this case, we have calculated the resonance frequency as a function of an external field applied parallel to the easy axis. In the second type of these antiferromagnets the $\mathbf{D}$ vector lies parallel to the easy axis. This is the situation in hematite below the Morin temperature. For this case, we have calculated the resonance frequency as a function of an external field applied in the plane perpendicular to the easy axis. In both cases we obtained two resonance modes, and the lower mode was found to have a frequency $W_-=0$ for a characteristic critical field, at which the antiferromagnetic axis becomes perpendicular to the easy axis.

INTRODUCTION

THE canting effect responsible for weak ferromagnetism in essentially antiferromagnetic materials is attributed primarily to two types of mechanisms, i.e., single-ion anisotropy[1] and antisymmetric exchange. The second is an exchange interaction of the form $\mathbf{D}_{ij}\cdot\mathbf{S}_i\times\mathbf{S}_j$. This term was originally suggested by Dzyaboshinsky,[2] who showed, on purely symmetrical grounds, that such a term may exist. A theoretical derivation of this term was given by Moriya[3] who showed that it was due to the effect of the spin-orbit term on the superexchange interaction. In this paper we shall deal only with antiferromagnets of the second kind, having uniaxial magnetocrystalline anisotropy of the form $K\sin^2\theta$. The $\mathbf{D}$ vector can be perpendicular to the easy axis, as is presumably the case in the orthoferrites ($R\,FeO_3$ where R is a rare-earth element), according to Treves,[4] or parallel to the easy axis, as occurs, for example, in hematite below its Morin transition temperature. (Hematite in this temperature range is in the antiferromagnetic phase, because the magnetocrystalline anisotropy energy is lower than the antisymmetric exchange energy.)

D PERPENDICULAR TO EASY DIRECTION

The Equilibrium Position

The situation where the external applied field is perpendicular to the easy axis in materials with $\mathbf{D}$ perpendicular to the easy axis had been discussed by Herrmann[5] as a special case of a Hamiltonian having a more general form of the magnetocrystalline anisotropy. We shall consider in this section the case where the external field is parallel to the easy axis, taking the X axis in the easy direction and the Y axis parallel to $\mathbf{D}$; the free energy F is given by

$$F=J\mathbf{M}_1\cdot\mathbf{M}_2-\mathbf{D}\cdot\mathbf{M}_1\times\mathbf{M}_2-(K/2M_0^2)(M_{1x}^2+M_{2x}^2)-H(M_{1x}+M_{2x}), \quad (1)$$

where $M_0=|\mathbf{M}_i|$ is the magnitude of the sublattice magnetization, J is the molecular field constant, $\mathbf{D}$ is the Dzyaloshinsky-Moriya vector, K is the uniaxial magnetocrystalline anisotropy constant, and H is the applied field in the X direction. Solving the equilibrium and stability equations obtained by considering the first and second derivatives of F, we found that there is a gradual rotation of the antiferromagnetic (AF) axis (see Fig. 1) with increasing field from a position parallel to one perpendicular to the easy direction. Thus there is no spin flop as would occur in the case of an antiferromagnet.

The equilibrium directions of the sublattice magnetization vectors $\mathbf{M}_1$, $\mathbf{M}_2$ may be defined by means of the angles α_1 and α_2, respectively, as shown in Fig. 1, or alternatively, by the canting angle Φ and the rotation angle θ of the AF axis. These sets of angles are related by $\alpha_1=\Phi-\theta$; $\alpha_2=\Phi+\theta$. The equilibrium values of Φ

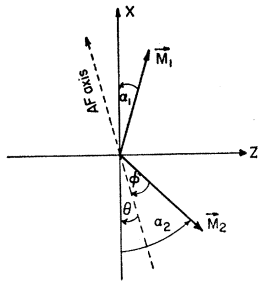

FIG. 1. Equilibrium sublattice magnetization directions.

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[1] T. Moriya, J. Phys. Chem. Solids **11**, 13 (1959).
[2] I. Dzyaloshinsky, J. Phys. Chem. Solids **4**, 241 (1958).
[3] T. Moriya, Phys. Rev. **120**, 91 (1960).
[4] D. Treves, Phys. Rev. **125**, 1843 (1962).
[5] G. F. Herrmann, J. Phys. Chem. Solids **24**, 597 (1963).

and θ are found to be

$$\sin\Phi \cong H_d H_k/(H_{||c}^2-H^2), \quad \text{for } H \leqslant H_0 \qquad (2)$$
$$\cong (H_d+H)/2H_{ex}, \quad \text{for } H_{ex} \gg H \gtrsim H_0,$$

$$\sin\theta \cong H_d H/(H_{||c}^2-H^2), \quad \text{for } H \leqslant H_0 \qquad (3)$$
$$\cong 1 \quad \text{for } H \gtrsim H_0,$$

where

$$H_{ex}=JM_0, \quad H_d=DM_0, \quad H_K=K/M_0,$$
$$H_{||c}=[(2H_{ex}+H_K)H_K]^{1/2},$$

and

$$H_0=\tfrac{1}{2}[-H_d+(H_d^2+4H_{||c}^2)^{1/2}]. \qquad (4)$$

$H_{||c}$ is the field at which spin flop would occur in the case of an antiferromagnet. The magnetization in the X direction as a function of H is $M_x=2M_0\sin\Phi \times\sin\theta$, and is shown in Fig. 2 for the case of $SmFeO_3$ at room temperature. Here we have[6] $H_{ex}=4.6\times10^6$ Oe, $H_d=6\times10^4$ Oe, and $H_K=170$ Oe. For the pure antiferromagnetic case, where $H_d=0$, we get from (3) the well-known spin flop at $H_{||c}=H_0$. As H_d increases, we see from (4) that H_0 decreases and the transition of the AF axis from parallel to perpendicular to the easy axis becomes more and more gradual. This is to be expected, as there is a corresponding increase in the ferromagnetic moment.

Resonance Frequencies

To obtain the resonance frequencies, we resort to the usual small-signal approximation. This is done by expressing the motion of the magnetization vectors in terms of their deviation from equilibrium. We follow the customary procedure[5] of transforming to two separate coordinate systems, (S_1,T_1,Y_1) and (S_2,T_2Y_2), which describe, respectively, the motion of $\mathbf{M}_1$ and $\mathbf{M}_2$. The S_1 and S_2 axes are chosen so as to coincide with the equilibrium positions of $\mathbf{M}_1$ and $\mathbf{M}_2$. Also, Y_1, Y_2, and Y are taken parallel (see Fig. 3). The transformation is formally given by

$$\begin{aligned} X_1&=S_1\cos\alpha_1+T_1\sin\alpha_1,\\ X_2&=-S_2\cos\alpha_2+T_2\sin\alpha_2,\\ Z_1&=S_1\sin\alpha_1-T_1\cos\alpha_1,\\ Z_2&=S_2\sin\alpha_2+T_2\cos\alpha_2\ . \end{aligned} \qquad (5)$$

In terms of the new variable, (1) becomes

$$\begin{aligned} F=&JM_0^2Y_1Y_2-(JM_0^2\cos2\Phi+DM_0^2\sin2\Phi)\\ &\times(S_1S_2+T_1T_2)+(JM_0^2\sin2\Phi-DM_0^2\cos2\Phi)\\ &\times(S_1T_2-S_2T_1)-\tfrac{1}{2}K(S_1^2\cos^2\alpha_1+S_2^2\cos^2\alpha_2\\ &+T_1^2\sin^2\alpha_1+T_2\sin^2\alpha_2+2S_1T_1\sin\alpha_1\cos\alpha_1-2S_2T_2\\ &\times\sin\alpha_2\cos\alpha_2)-HM_0(S_1\cos\alpha_1+T_1\sin\alpha_1-S_2\cos\alpha_2\\ &+T_2\sin\alpha_2), \end{aligned} \qquad (6)$$

where X_i, Y_i, Z_i, and S_i, Y_i, T_i are the components of unit vectors $\mathbf{R}_i$ along the sublattice magnetization vectors $\mathbf{R}_i=\mathbf{M}_i/M_0$. The dynamic equations may now be written as

$$(M_0/|\gamma|)\dot{\mathbf{R}}_i=\mathbf{R}_i\times\nabla_{\mathbf{R}_i}F\ , \quad i=1,2\ , \qquad (7)$$

where the gyromagnetic ratio $|\gamma|=1.86\times10^7\ \mathrm{Oe^{-1}\ sec^{-1}}$. In the small-signal approximation, the component of $\mathbf{M}_i$ along S_i is taken as constant, and (7) yields the set of four equations

$$\begin{aligned} \frac{M_0}{|\gamma|}\dot{T}_i&=Y_i\frac{\partial F}{\partial S_i}-S_i\frac{\partial F}{\partial Y_i}, \quad i=1,2,\\ \frac{M_0}{|\gamma|}\dot{Y}_i&=S_i\frac{\partial F}{\partial T_i}-T_i\frac{\partial F}{\partial S_i}, \quad i=1.2. \end{aligned} \qquad (8)$$

Taking the time dependence of the components of $\mathbf{M}_i$ perpendicular to S_i in the form e^{iWt}, (8) becomes

$$\begin{aligned} \omega T_1+g_1Y_1+H_{ex}Y_2&=0,\\ \omega T_2+g_2Y_2+H_{ex}Y_1&=0,\\ \omega Y_1-b_1T_1+cT_2&=0,\\ \omega Y_2-b_2T_2+cT_1&=0, \end{aligned} \qquad (9)$$

where $\omega=i(W/|\gamma|)$, and

$$\begin{aligned} c&=H_{ex}\cos2\Phi+H_d\sin2\Phi,\\ g_i&=c+H_K\cos^2\alpha_i+(-)^{i+1}H\cos\alpha_i,\\ b_i&=g_i-H_K\sin^2\alpha_i\ . \end{aligned}$$

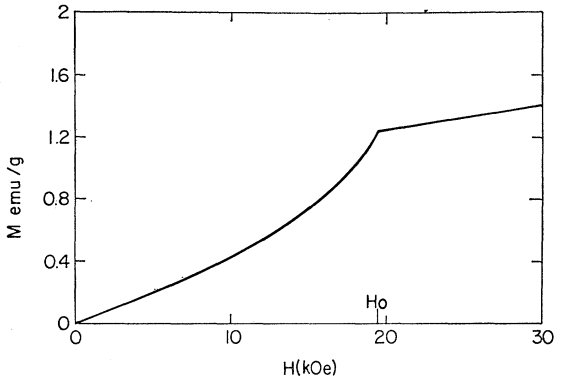

FIG. 2. Magnetization in the X (easy) direction M_X, as a function of an external field H in this direction, for the case of $SmFeO_3$ at room temperature. ($H_{ex}=4.6\times10^6$ Oe, $H_d=6\times10^4$ Oe, and $H_K=170$ Oe.)

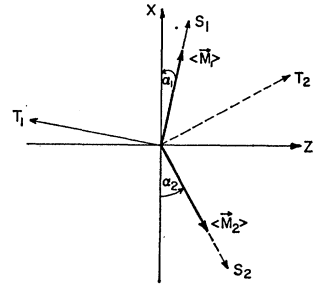

FIG. 3. Equilibrium coordinate system.

[6] G. Gorodetsky and D. Treves, Phys. Rev. 135, A97 (1964).

The condition for obtaining a nontrivial solution of this set of equations is that its determinant vanish. This requirement yields the secular equation

$$(W/\gamma)^4+A(W/\gamma)^2-B=0,$$

where

$$A\cong 2cH_{ex}-g_1b_1-g_2b_2$$

and

$$B\cong(b_1b_2-c^2)(H_{ex}^2-g_1g_2).$$

Setting $H_d=0$ gives, as a special case, the well-known antiferromagnetic modes[7] [see Fig. 4(a)]. The two modes obtained from the secular equation are

$$(W_\pm/\gamma)^2=\tfrac{1}{2}[-A\pm(A^2+4B)^{1/2}]. \quad (10)$$

This general result can be simplified for the following cases:

(1) In the range $H\ll H_d^2/H_{||c}$. Here

$$A\cong-(2H_{||c}^2+H_d^2+2H^2)$$

and

$$B\cong-(H_{||c}^4+H_{||c}^2H_d^2),$$

giving

$$\left(\frac{W_+}{\gamma}\right)^2\cong H_{||c}^2+H_d^2+2\left[1+\left(\frac{H_{||c}}{H_d}\right)^2\right]H^2,$$
$$\left(\frac{W_-}{\gamma}\right)^2\cong H_{||c}^2-2\left(\frac{H_{||c}}{H_d}\right)^2H^2 \quad (11)$$

(2) In the range $H_{ex}\gg H\geqslant H_0$. Here,

$$\alpha_1=-\left(\frac{\pi}{2}-\Phi\right),\quad \alpha_2=\frac{\pi}{2}+\Phi,\quad c\cong H_{ex}+(H_d-H)\sin\Phi,$$

$$g=g_1=g_2\cong H_{ex}+H_d\sin\Phi,$$

and

$$b=b_1=b_2\cong c+H\sin\Phi-H_K\cos^2\Phi,$$

giving

$$(W_+/\gamma)^2\cong H_d^2+H_dH,$$
$$(W_-/\gamma)^2\cong H^2+H_dH-H_{||c}^2. \quad (12)$$

It is clear from (12) that the lower mode vanishes at $H=H_0$. The resonance frequencies as a function of H, for different values of H_d, are shown in Fig. 4. [Fig. 4(e) describes the case for $SmFeO_3$ at room temperature].

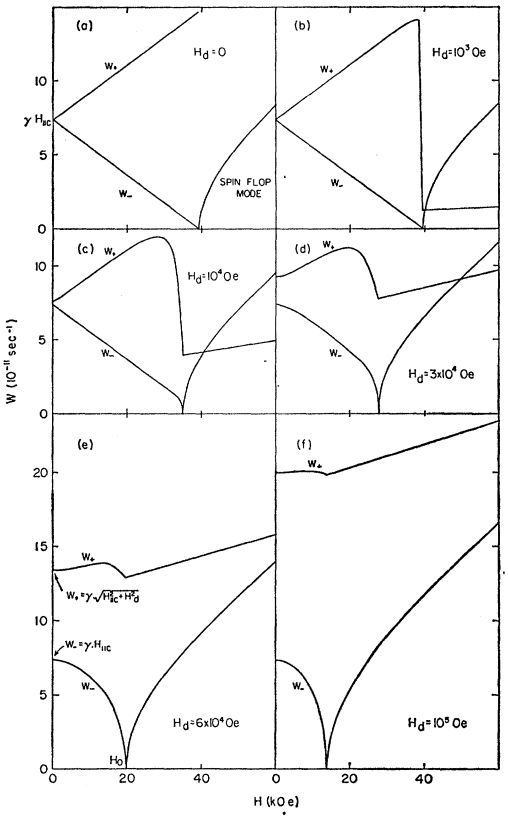

FIG. 4. Resonance frequencies $W_\pm$ as a function of H for different values of H_d: (a) $H_d=0$ Oe, (b) $H_d=10^3$ Oe, (c) $H_d=10^4$ Oe, (d) $H_d=3\times10^4$ Oe, (e) $H_d=6\times10^4$ Oe [(the case of $SmFeO_3$ at room temperature), and (f) $H_d=10^5$ Oe. All cases are for $H_{ex}=4.6\times10^6$ Oe and $H_K=170$ Oe.

D PARALLEL TO EASY DIRECTION

The Equilibrium Position

The case wherein **D** is parallel to the easy axis occurs, for example, in hematite $\alpha-Fe_2O_3$, below its Morin transition temperature ($T_M\cong263$°K). The case where the external field is parallel to the easy axis, in such a material, is well known.[8] A zero-frequency field was found at $H_{||c}=[(2H_{ex}+H_K)H_K-H_d^2]^{1/2}$, which is the critical field at which spin flop occurs. Note that for this kind of material, the expression of $H_{||c}$ is different from the corresponding critical field in the previous case. Assuming a uniaxial magnetocrystalline anisotropy $E_K=K\sin^2\theta$, one finds, in the two-sublattice approximation, with the applied field in the plane perpendicular to the easy axis, a gradual rotation of the AF axis (see Fig. 5) from parallel to perpendicular to the easy axis.[9] This rotation of the AF axis is caused by the torque due to the antisymmetric exchange interaction.

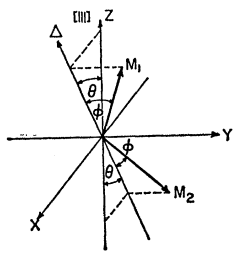

FIG. 5. Sublattice magnetization vectors.

[7] C. Kittel, Phys. Rev. **82**, 565 (1951); T. Nagamiya, Progr. Theoret. Phys. (Kyoto) **6**, 342 (1951); S. Foner, in *Magnetism*, edited by G. T. Rado and H. Suhl (Academic Press Inc., New York, 1963), Vol. I.

[8] S. Foner, in *Proceedings of the International Conference on Magnetism, Nottingham, England, 1964* (Institute of Physics and The Physical Society, London, 1965).

[9] G. Cinader and S. Shtrikman, Solid State Commun. **4**, 459 (1966).

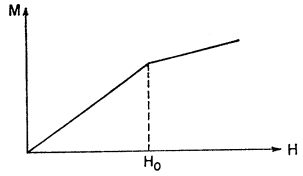

FIG. 6. Magnetization M as a function of an applied field H perpendicular to the easy axis.

The free energy for this case may be written in the form

$$F=J\mathbf{M}_1\cdot\mathbf{M}_2-\mathbf{D}\cdot\mathbf{M}_1\times\mathbf{M}_2-(K/2M_0^2)\times(M_{1z}^2+M_{2z}^2)-H(M_{1y}+M_{2y}), \quad (13)$$

where $\mathbf{D}=(0,0,D)$.

Taking Z as the crystal's easy axis (i.e., the [1,1,1] direction in hematite) and Y as the direction of the applied field, the canting angle Φ and the angle θ between the AF axis and Z are found to be

$$\sin\Phi\cong H_K H/H_{\|c}^2, \quad \text{for } H\leq H_0$$
$$\cong(H_d+H)/2H_{ex}, \quad \text{for } H_{ex}\gg H\gtrsim H_0, \quad (14)$$

$$\sin\theta\cong H_d H/H_{\|c}^2, \quad \text{for } H\leq H_0 \quad (15)$$
$$\cong 1, \quad \text{for } H\gtrsim H_0,$$

$$H_0=H_{\|c}^2/H_d. \quad (16)$$

The net magnetization $M=2M_0\sin\Phi$, as a function of H, is shown in Fig. 6.

Resonance Frequencies

As previously, we use the small-signal approximation and transform to more convenient coordinate systems. The transformed coordinate system used for each sublattice is shown in Fig. 7. Formally, the transformations are given by

$$\mathbf{X}_i=(-)^{i+1}\cos\theta\cdot\boldsymbol{\varepsilon}_x{}^i+(-)^i\sin\Phi\sin\theta\cdot\boldsymbol{\varepsilon}_y{}^i+(-)^{i+1}\cos\Phi\sin\theta\cdot\boldsymbol{\varepsilon}_z{}^i,$$
$$\mathbf{Y}_i=\cos\Phi\boldsymbol{\varepsilon}_y{}^i,+\sin\Phi\boldsymbol{\varepsilon}_z{}^i, \quad (17)$$
$$\mathbf{Z}_i=(-)^i\sin\theta\boldsymbol{\varepsilon}_x{}^i+(-)^i\sin\Phi\cos\theta\boldsymbol{\varepsilon}_y{}^i+(-)^{i+1}\cos\Phi\cos\theta\boldsymbol{\varepsilon}_z{}^i, \quad i=1,2.$$

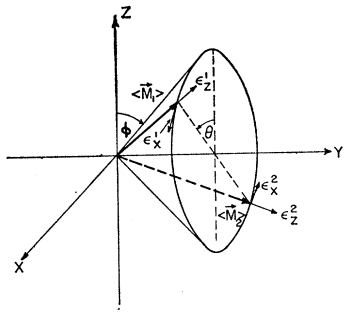

FIG. 7. Equilibrium coordinates, where X is the easy axis, $\mathbf{D}=(0,0,D)$, $\mathbf{H}=(0,H,0)$. $\boldsymbol{\varepsilon}_z{}^i\|\mathbf{M}_i$, $\boldsymbol{\varepsilon}_x{}^i$ is tangent to the base boundary of the cone, so that when $\theta=0$ we have $\boldsymbol{\varepsilon}_x{}^i\|(-)^{i+1}\mathbf{X}$. $\boldsymbol{\varepsilon}_y{}^i$ is defined so as to make $(\boldsymbol{\varepsilon}_x{}^i,\boldsymbol{\varepsilon}_y{}^i,\boldsymbol{\varepsilon}_z{}^i)$ a right-hand set. $|\boldsymbol{\varepsilon}_k{}^i|=1$; $k=x, y, z$; $i=1, 2$. θ is the canting angle between the AF axis and the easy axis Z. ϕ is the canting angle between $\mathbf{M}_1$ and the ZX plane.

By straightforward calculations, we obtain

$$(W_\pm/\gamma)^2=(B\mp A)(C\pm H_{ex})-(G\mp F)^2, \quad (18)$$

where

$$A=H_{ex}\cos2\Phi+H_d\sin2\Phi\sin\theta,$$
$$B=A+H\sin\Phi+H_K\cos2\Phi\cos^2\theta,$$
$$C=A+H\sin\Phi+H_K(\cos^2\Phi\cos^2\theta-\sin^2\theta),$$
$$G=H_d\cos\Phi\cos\theta, \text{ and } F=\tfrac{1}{2}H_K\sin\Phi\sin2\theta.$$

For $H\cong0$, (18) may be written as

$$(W_+/\gamma)^2\cong H_{\|c}^2+H^2, \quad (19a)$$
$$(W_-/\gamma)^2\cong H_{\|c}^2. \quad (19b)$$

This is the result that is obtained for the case of the antiferromagnet.[10] This equality is to be expected in this range as $E_d=-\mathbf{D}\cdot\mathbf{M}_1\times\mathbf{M}_2\cong0$ for $H\cong0$. In the range

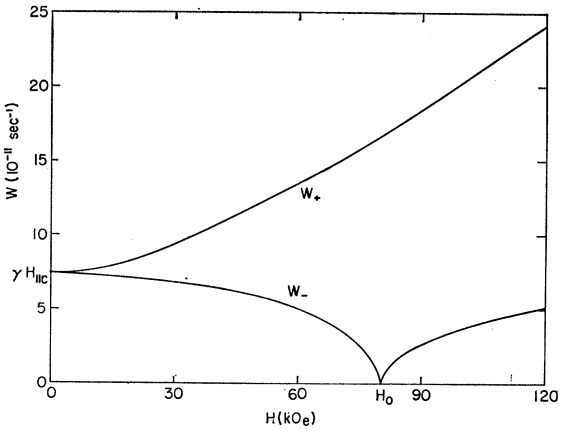

FIG. 8. Resonance frequencies $W_\pm$ as a function of H for the case of $\alpha-Fe_2O_3$ below its Morin transition temperature with $H_{ex}=10^7$ Oe, $H_d=2\times10^4$ Oe, and $H_K=100$ Oe.

$H_{ex}\gg H\gtrsim H_0$, we obtain

$$\left(\frac{W_+}{\gamma}\right)\cong H^2+H_dH, \quad (20a)$$

$$\left(\frac{W_-}{\gamma}\right)^2\cong H_d(H-H_0), \quad (20b)$$

which corresponds to Fink's results[11] above the Morin transition point, as it should. From (20b), it is clear that the lower mode W_- is equal to zero at $H=H_0$. The resonance frequencies $W_\pm$, as a function of H, are shown in Fig. 8 for $H_{ex}=10^7$ Oe, $H_d=2\times10^4$ Oe, and $H_K=100$ Oe (hematite at about 240°K). A numerical check of the first derivative of W_+ with respect to H shows a discontinuity at $H=H_0$.[12]

[10] F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).
[11] H. J. Fink, Phys. Rev. 133, A1322 (1964).
[12] I am indebted to R. Hornreich for calling my attention to the existence of this discontinuity.

CONCLUSIONS

The critical field H_0 is quite large at most temperatures. The simplest method of obtaining such high fields is by utilizing pulsed-field techniques, but, with this method, it is difficult to measure the weak ferromagnetic moment directly, in the absence of spin flop. A measurement of H_0 using a magnetic-resonance method could avoid this difficulty. Using such magnetic-resonance studies, the theoretical rotating AF-axis picture could be investigated. Also, more information about the canting mechanism, especially at low temperatures, could be obtained, and, from a knowledge of $H_0(T)$, it would be possible to find the temperature dependence of the uniaxial magnetocrystalline anisotropy constant. To calculate the resonance modes in the case where the applied field deviates slightly from the special directions considered here is somewhat more difficult as the symmetry of the equilibrium configuration is reduced. All the above calculations were performed at essentially 0°K using the molecular-field approximation.

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Orientational Order in fcc Solid Ortho-H_2. Green's-Function Treatment of the Internal-Field Approximation

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The technique of temperature-dependent Green's functions is applied to a molecular-field model of the orientational order-disorder transition of ortho-H_2 molecules on a face-centered-cubic (fcc) lattice. The chain of equations for the various Green's functions is decoupled by an approximation similar to the one used by Tahir-Kheli and ter Haar, which enables one to solve the three remaining Green's-function equations of motion. It is shown that the Green's-function approach leads to the same results as the more conventional methods.

I. MODEL

A THEORETICAL discussion of the cooperative orientational ordering of ortho-H_2 molecules on both face-centered-cubic (fcc) and hexagonal-close-packed (hcp) lattices has recently been given by Raich and James.[1,2] For the case of a rigid fcc lattice it was shown that the internal-field approximation (or molecular-field approximation) leads to a first-order phase transition between an orientationally ordered phase, stable at low temperatures, and an orientationally disordered phase, stable at high temperatures.

The purpose of this paper is to indicate how the technique of temperature-dependent Green's functions can be applied to a model of the ordering of ortho-H_2 molecules on a rigid fcc lattice, valid within the framework of the internal-field approximation. It is shown that this technique leads to the same results as the more conventional one, based on the minimization of the free energy.

Leaving off the rotational energy of the molecules, the Hamiltonian for the model to be considered is

$$H=\tfrac{1}{2}\sum_{i,j} V_{ij}(\Omega_i,\Omega_j)\,, \tag{1}$$

where $\Omega_i=(\theta_i,\phi_i)$ specifies the orientation of molecule i. If we assume that the orientational coupling of the molecules arises from quadrupole-quadrupole coupling, the potential energy of interaction of molecules i and j, V_{ij} can be written as[3]

$$V_{ij}=(20\pi/9)(70\pi)^{1/2}\Gamma_{ij}\sum_{MN} C(224;MN) \times Y_{2M}(\Omega_i)Y_{2N}(\Omega_j)Y_{4,M+N}(\Omega_{ij})^*. \tag{2}$$

[1] J. C. Raich and H. M. James, Phys. Rev. Letters **16**, 173 (1966). This paper gives a list of references to previous experimental and theoretical work on this problem.
[2] H. M. James and J. C. Raich (to be published).
[3] H. P. Gush and J. Van Kranendonk, Can. J. Phys. **40**, 1461 (1962).