Spin-Wave Theory of Magnetic Resonance in Spiral Spin Structures: Effect of an Applied Field

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The possibility of performing magnetic resonance experiments in substances with spiral spin structures is studied. Particular attention is paid to the variation of spin arrangement and spin-wave spectrum with an applied magnetic field in the plane of the spiral. Perturbation techniques give resonance frequencies for small fields and for fields near the value, H_f, required for ferromagnetic alignment. These results then allow a realistic extrapolation for the whole range of H. The effects of planar anisotropy and demagnetizing fields on the variation of spin arrangement and on the spin-wave frequencies are discussed. The actual conditions of resonance are examined for dysprosium and erbium. There is a discussion of the information that can be obtained from polycrystalline specimens with particular reference to the experimental results for $MnAu_2$.

1. INTRODUCTION

NEUTRON diffraction studies have now demonstrated the existence of spiral spin structures in a number of materials. One relatively simple class of such structures occurs in crystals with an axis of symmetry, where the moments in each layer perpendicular to this axis are aligned ferromagnetically. The direction of these moments varies, however, from layer to layer. The heavy rare-earth metals (Tb-Tm), which are hexagonal, show such structures in some of their magnetic phases, as does $MnAu_2$ which is tetragonal. Several authors[1–4] have shown that these orderings are a natural consequence of indirect exchange (via the conduction electrons) and a strong crystalline electric field. The axial variation of the latter fixes the direction of the moments relative to the symmetry axis. They often point in the plane perpendicular to this axis (Tb, Dy, Ho, and $MnAu_2$), sometimes along the axis (Tm) or at some intermediate angle (Er). There is also a smaller anisotropy of appropriate symmetry in the plane perpendicular to the axis.

It appears likely that further information about the magnetic properties could be obtained by a study of the low-lying (spin-wave) states of these systems. Inelastic neutron scattering and magnetic resonance provide convenient techniques for this purpose. Accordingly in a recent paper (referred to as I) Cooper *et al.*[5] examined the spin waves which might be observed by resonance experiments in the heavy rare earths. A realistic Hamiltonian may be written down and investigated for all the layer types of structure mentioned above. Because of the high anisotropy and spiral structure, the resonance conditions are quite different from those in usual ferromagnetic crystals.

The normal technique of ferromagnetic resonance is to vary an applied field in order to make the appropriate spin-wave frequency equal to the signal frequency. Thus it is essential to study the change in the magnetic structure and the spin-wave spectrum with magnetic field. Since the anisotropy relative to the crystal axis is so large, a field applied along the axis has little effect. A field in the plane, however, can seriously modify the spiral structure, and hence the spin-wave spectrum. Nagamiya *et al.*[6,7] have studied the modifications of structure for a spiral where the spins point entirely in the plane, and there is no planar anisotropy. For small fields they find a slight distortion of the spiral with a small net moment along **H**. At a critical field H_c there is a first-order transition to a fan-like structure with a large moment along **H**. As the field increases still further, the angle of the fan decreases continuously until complete ferromagnetic alignment is achieved in a second-order transition at $H=H_f$, which is approximately twice H_c. Typically H_c is of order 10^4 G so that all these structures will be within the range of conventional equipment.

In I these effects received only a preliminary examination. In this paper, the effect of H on the resonance frequencies will be studied by a perturbation treatment at small H and $H \sim H_f$. This allows a realistic extrapolation over the whole range of H. Comments will be made on the relationship of the spin-wave spectrum to the transitions at H_c and H_f. The effects of planar anisotropy and demagnetizing fields on the transitions and spin-wave frequencies will be discussed. The actual conditions of resonance will be examined for Dy and Er, and an attempt will be made to interpret the resonance

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data of Meyer and Asch[8,9] on $MnAu_2$. This work shows that information can be obtained from polycrystalline samples, and the theory of such situations will be discussed.

2. BASIC HAMILTONIAN

In I it was shown that in the case of a plane spiral the general Hamiltonian applicable to the heavy rare-earth metals could be reduced to a model form

$$\mathcal{H}=-\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-K_2\sum_i S_{i\zeta}^2+\lambda\beta H\sum_i S_{i\xi}, \quad (1)$$

which contained all the essential features, viz., exchange, an axial anisotropy, and a field applied in the plane. Here ζ is the symmetry axis and ξ,η are mutually perpendicular directions in the plane. The possible anisotropy of the exchange and the planar anisotropy have been dropped for ease of calculation. The effects of planar anisotropy are discussed in Sec. 6b. The axial anisotropy terms of fourth and sixth orders have also been dropped. In I it was shown that these do not enter into the spin-wave frequencies when the equilibrium spin arrangement is a planar spiral. λ is the Landé factor taken with negative sign.

The Fourier transform of the exchange energy

$$J(\mathbf{q})=\sum_i J_{ij}\cos\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j) \quad (2)$$

has its maximum at $\mathbf{q}=\mathbf{k}_0$ where $\mathbf{k}_0$ is parallel to the ζ axis. This means that when $H=0$, the spin arrangement is a spiral with an angle k_0c' between the moments of successive planes of spacing c'. For planar arrangements K_2 is negative.

μ, the equilibrium moment per atom at T, is of magnitude $-\lambda\beta S$ with

$$S=MS_0, \quad (3)$$

where M is the normalized moment and S_0 the total angular momentum (orbital plus spin) of the atom. It is convenient to define

$$\begin{aligned} a&=S^2[J(\mathbf{k}_0)-J(0)],\\ b&=S^2[J(\mathbf{k}_0)-J(2\mathbf{k}_0)],\\ c&=S^2[J(\mathbf{k}_0)-J(3\mathbf{k}_0)],\\ d&=S^2[J(\mathbf{k}_0)-J(4\mathbf{k}_0)]. \end{aligned} \quad (4)$$

It is also convenient to define a rotating coordinate system such that the Z axis is the equilibrium direction of magnetization. If θ_n is the angle of the magnetization of the nth layer with respect to the ξ axis, the unit vectors of the new system are

$$\begin{aligned} \mathbf{X}_n&=-\boldsymbol{\zeta},\\ \mathbf{Y}_n&=-\boldsymbol{\xi}\sin\theta_n+\boldsymbol{\eta}\cos\theta_n,\\ \mathbf{Z}_n&=\boldsymbol{\xi}\cos\theta_n+\boldsymbol{\eta}\sin\theta_n. \end{aligned} \quad (5)$$

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We must now proceed to make the approximations necessary to treat the various cases of interest.

3. LOW FIELDS

For $H=0$, the spin arrangement is an undistorted spiral with $\theta_n=nk_0c'$. The spin-wave spectrum and resonance frequencies for this arrangement were discussed in I. It was shown that in this case there are two frequencies that can be excited depending on the polarization of the rf field. An rf field applied in the ξ-η planes excites $\omega(\mathbf{k}_0)$.

$$\omega(\mathbf{k}_0)=[-2K_2(a+b)]^{1/2}. \quad (6)$$

An rf field along ζ excites $\omega(0)$; however $\omega(0)=0$ for $H=0$. Some discussion of the behavior of $\omega(0)$ for nonzero H was given in I. In particular, it was shown, using both the equations of motion method and a modified perturbation procedure, that the term in $\omega(0)$ linear in H vanished. It was, therefore, speculated that $\omega(0)\sim H^2$. In the present paper it is shown that this is correct and the expression for $\omega(0)$ is given.

In an experiment, the signal frequency would be held constant and dc field varied. For this reason, the variation of $\omega(\mathbf{k}_0)$ with H is of interest. It should be noted that for dysprosium, $\omega(\mathbf{k}_0)$ at $H=0$ is expected to be quite high (at least several cm^{-1}) because of the large anisotropy energy present, typical of the heavy rare earths. An analogous situation holds for the spiral-cone phase of Er as discussed in Sec. 7.

We will first discuss the value of $\omega(0)$ for nonzero H. The variation of θ_n with H can be found from the condition that $\partial E_0/\partial\theta_n=0$, where E_0 is the equilibrium energy in the molecular field approximation

$$\partial E_0/\partial\theta_n=0=2\sum_p J_{np}S^2\sin(\theta_n-\theta_p)+\mu H\sin\theta_n. \quad (7)$$

This equation can be solved by an iterative procedure described in I. To third order in H,

$$\theta_n=nk_0c'+[XH+AH^3]\sin nk_0c' +YH^2\sin^2 nk_0c'+BH^3\sin^3 nk_0c', \quad (8)$$

$$X=-\mu/(a+b), \quad (9a)$$

$$Y=\mu^2(4b-c)/4(a+b)^2c, \quad (9b)$$

$$A=[\mu^3/2(a+b)^4(b+d)c]\{(b+d)(-ac+bc) +4abd-12b^3-8ab^2\}, \quad (9c)$$

$$B=[\mu^3/3(a+b)^3(b+d)c]\{c(b+d)+6b(3b-d)\}. \quad (9d)$$

The Hamiltonian can be written in terms of the X, Y, Z components of $\mathbf{S}_i$ using (5). This gives

$$\mathcal{H}=-\sum_{i\neq j} J_{ij}[(S_{iy}S_{jy}+S_{iz}S_{jz})\cos(\theta_i-\theta_j)+S_{ix}S_{jx}] +\sum_i[-(\mu H/S)S_{iz}\cos\theta_i-K_2S_{ix}^2]. \quad (10)$$

Here the subscript i on θ labels individual spins rather than layers. The θ_i for all spins in the nth layer are equal and given by (8). Additional terms, linear in S_{iy} and in $S_{iy}S_{jz}$ would be present. These would give rise to linear spin-wave terms, but vanish to terms of second order in S_{iy} because of (7). Hence, these terms can be omitted in the noninteracting spin-wave approximation.

The equation of motion method is used to find the energy of the low-lying excited magnetic states of the system, the spin-wave states. Defining

$$S_x(\mathbf{q})=N^{-1/2}\sum_i S_{ix}e^{i\mathbf{q}\cdot\mathbf{R}_i}, \qquad (11a)$$

$$S_y(\mathbf{q})=N^{-1/2}\sum_i S_{iy}e^{i\mathbf{q}\cdot\mathbf{R}_i}, \qquad (11b)$$

the equation of motion for $S_x(\mathbf{q})$ is

$$[S_x(\mathbf{q}),\mathcal{H}]=i\hbar\dot{S}_x(\mathbf{q}). \qquad (12)$$

The random-phase approximation is used in finding the equations of motion. That is, S_{iz} is replaced by S, the thermal average, after taking the commutator. This means that spin waves are defined as deviations from the equilibrium spin arrangement at the temperature in question.

Now if $S_x(\mathbf{q})$ were an operator generating an exact excited eigenstate, then $[S_x(\mathbf{q}),\mathcal{H}]$ would be just a constant times $S_x(\mathbf{q})$, and this would immediately give the energy of the corresponding state. However, in general any $S_x(\mathbf{q})$ in itself is not such an operator. In general $[S_x(\mathbf{q}_1),\mathcal{H}]$ consists of a combination of $S_y(\mathbf{q})$'s for several $\mathbf{q}$ related to $\mathbf{q}_1$. Thus, the proper operators to generate excited states consist of a combination of $S_x(\mathbf{q})$'s and $S_y(\mathbf{q})$'s for several values of $\mathbf{q}$. The procedure for finding the $\omega(\mathbf{q})$ corresponding to the generating operators is straightforward, but to obtain results correct to a high order in H, one must solve equations in which many $\mathbf{q}$ are linked. The determinants thus become of high dimension and impossible to solve analytically.

However, to low order in H the size of the determinant which must be considered is relatively small. From (8) it appears that the angle θ_n of the nth layer contains terms in $e^{\pm irk_0nc'}$ where r is an integer, and that H^r is the lowest order which occurs in such a term. The energy was expressed in (10) in terms of θ_n. If this is now expanded in powers of H, the terms in $e^{\pm irk_0nc'}$ will mix spin waves of wave vector $\mathbf{q}\pm r\mathbf{k}_0$ to those of wave vector $\mathbf{q}$. For the present purpose $-2\leq r\leq 2$ suffices. This will give the mixing of a spin wave of given $\mathbf{q}$ with all spin waves linked to it to order H^2. The equations of motion are then of the form

$$-\hbar\omega S_x(\mathbf{q})=[S_x(\mathbf{q}),\mathcal{H}]=\sum_{-2\leq r\leq 2} iD_r(\mathbf{q})S_y(\mathbf{q}+r\mathbf{k}_0), \qquad (13)$$

$$-\hbar\omega S_y(\mathbf{q})=[S_y(\mathbf{q}),\mathcal{H}]=-\sum_{-2\leq r\leq 2} iG_r(\mathbf{q})S_x(\mathbf{q}+r\mathbf{k}_0), \qquad (14)$$

where to order H^2

$$\begin{aligned}D_0(\mathbf{q})=&S[2J(\mathbf{k}_0)-J(\mathbf{k}_0+\mathbf{q})-J(-\mathbf{k}_0+\mathbf{q})]\\&-\tfrac{1}{4}SX^2H^2[-2J(\mathbf{k}_0+\mathbf{q})-2J(-\mathbf{k}_0+\mathbf{q})\\&+2J(\mathbf{q})+J(\mathbf{q}+2\mathbf{k}_0)+J(\mathbf{q}-2\mathbf{k}_0)],\end{aligned} \qquad (15a)$$

$$\begin{aligned}D_{-1}(\mathbf{q})=D_1(-\mathbf{q})=&-\tfrac{1}{2}SHX[2J(\mathbf{k}_0)-2J(2\mathbf{k}_0)-J(\mathbf{q})\\&+J(\mathbf{q}-2\mathbf{k}_0)+J(\mathbf{q}+\mathbf{k}_0)-J(\mathbf{q}-\mathbf{k}_0)],\end{aligned} \qquad (15b)$$

$$\begin{aligned}D_{-2}(\mathbf{q})=D_2(-\mathbf{q})=&\tfrac{1}{8}SH^2X^2[-J(\mathbf{q}-3\mathbf{k}_0)-2J(\mathbf{q}-\mathbf{k}_0)\\&+2J(\mathbf{q})+2J(\mathbf{q}-2\mathbf{k}_0)-J(\mathbf{q}+\mathbf{k}_0)+J(3\mathbf{k}_0)\\&+3J(\mathbf{k}_0)-2J(0)-2J(2\mathbf{k}_0)]\\&+\tfrac{1}{2}H^2YS[2J(\mathbf{q}-\mathbf{k}_0)-J(\mathbf{q}-3\mathbf{k}_0)-J(\mathbf{q}+\mathbf{k}_0)\\&+J(3\mathbf{k}_0)-J(\mathbf{k}_0)]+\mu H^2X/4S,\end{aligned} \qquad (15c)$$

$$G_0(\mathbf{q})=2S[J(\mathbf{k}_0)-J(\mathbf{q})-K_2], \qquad (16a)$$

$$G_1(\mathbf{q})=G_{-1}(\mathbf{q})=-HXb/S, \qquad (16b)$$

$$G_2(\mathbf{q})=G_{-2}(\mathbf{q})=-(H^2X^2/2S)b. \qquad (16c)$$

It may be noted that since K_2 appears in $\mathcal{H}$ as the coefficient of $\sum_i S_{ix}^2$, (13) does not contain K_2 while (14) does. One may form the second commutator $[[S_x(\mathbf{q}),\mathcal{H}],\mathcal{H}]$ by commuting (13) with $\mathcal{H}$ and substituting (14). In some cases of interest like the rare earths, the K_2 term in $[[S_x(\mathbf{q}),\mathcal{H}],\mathcal{H}]$ is much larger than the other terms and it is possible to approximate the second commutator as

$$(\hbar\omega)^2S_x(\mathbf{q})=[[S_x(\mathbf{q}),\mathcal{H}],\mathcal{H}]\approx(-2K_2S)\sum_{-2\leq r\leq 2} D_r(\mathbf{q})S_x(\mathbf{q}+r\mathbf{k}_0). \qquad (17)$$

This yields an eigenvalue equation for ω^2. The positive and negative roots for each solution correspond simply to left- and right-handed rotation of the spin vectors in the excited state, or in the quantized formalism to creation and destruction of excitations.

It was shown in I that $\hbar\omega(0)$ had no term linear in H. It can also be shown that there is no term with $\omega^2(0)\sim H^3$. Therefore, we seek $\omega^2(0)\sim H^4$. To find $\omega^2(0)$ to order H^4, the terms necessary in the equations of motion are those for $S_x(\mathbf{q})$'s and $S_y(\mathbf{q})$'s linked to $\mathbf{q}=0$ to order H^2. These terms are just those in (13) and (14) with $-2\leq r\leq 2$. The other quantity of interest, $\omega(\mathbf{k}_0)$, is nonvanishing for $H=0$ as already discussed. The lowest order change in $\omega^2(\mathbf{k}_0)$ coming from H goes as H^2. There is an off-diagonal contribution to $\omega^2(\mathbf{k}_0)$ to this order from the $r=\pm 1$ terms in (17). The $r=\pm 2$ term which is of order H^2 connects $\mathbf{k}_0$ and $-\mathbf{k}_0$; because of the degeneracy of $\omega(\mathbf{k}_0)$ and $\omega(-\mathbf{k}_0)$ this contributes to $\omega^2(\mathbf{k}_0)$ to order H^2. Thus, both the quantities of interest, $\omega(0)$ and $\omega(\mathbf{k}_0)$, can be examined using just the $-2\leq r\leq 2$ terms retained in (13) and (14).

To find $\omega(0)$, Eq. (17) couples $S_x(0)$ to $S_x(r\mathbf{k}_0)$ for r between -2 and 2. Thus, the secular determinant is

$$|(\hbar\omega)^2\delta_{rs}-\Delta_{rs}|=0, \qquad (18)$$

where the determinant is symmetric and

$$\Delta_{rs}=[-2K_2S]D_{s-r}(r\mathbf{k}_0). \tag{19}$$

The D's have been given to order H^2 in (15). To find $\omega^2(0)$ to order H^4 requires $D_0(0)$ and $D_1(0)$ to orders H^4 and H^3, respectively. These can be easily found from the appropriate terms in $[\mathcal{H},[\mathcal{H},S_x(0)]]$ using the expression for θ_n correct to order H^3 given in (8). Since $\omega^2(0)$ is being found to order H^4, it is appropriate to consider just the terms in the secular determinant contributing to $\omega^2(0)$ to this order so that

$$\begin{aligned}
\Delta_{00}&=-\frac{K_2}{(a+b)}(\mu H)^2-\frac{K_2b(2b-c)}{c(a+b)^4}(\mu H)^4,\\
\Delta_{11}&=\Delta_{-1-1}=-2K_2(a+b)+\frac{K_2(2b+2a-c)}{2(a+b)^2}(\mu H)^2,\\
\Delta_{22}&=\Delta_{-2-2}=-2K_2c,\\
\Delta_{01}&=\Delta_{0-1}=-K_2\mu H-\frac{K_2(-c+2b)}{4(a+b)^2c}(\mu H)^3,\\
\Delta_{02}&=\Delta_{0-2}=\frac{K_2(\mu H)^2}{2(a+b)},\\
\Delta_{1-1}&=-\frac{K_2(a-b)(c-2b)}{(a+b)^2c}(\mu H)^2,\\
\Delta_{12}&=\Delta_{-1-2}=-K_2\left(\frac{3b-a-c}{a+b}\right)(\mu H),\\
\Delta_{1-2}&=\Delta_{-12}=\Delta_{2-2}=0,
\end{aligned} \tag{20}$$

and the solution is

$$\hbar\omega(0)=(\mu H)^2\left\{\frac{-K_2(c-4b)}{2(a+b)^3c}\right\}^{1/2}. \tag{21}$$

The behavior of the frequency $\omega(\mathbf{k}_0)$ to order H^2 may be obtained from the same determinant. [Although the Δ in general are required to a lower order than in (20).] In a plane spiral the states of $\pm\mathbf{k}_0$ are degenerate and $\Delta_{rr}=\Delta_{-r-r}$, $\Delta_{rs}=\Delta_{-r-s}$. It is therefore simpler to consider the symmetric and antisymmetric combinations $S_x(\mathbf{k}_0)\pm S_x(-\mathbf{k}_0)$. The positive combination will be designated $\cos k_0$ and is coupled to the $\mathbf{q}=0$ and $\cos 2k_0$ states. The negative combination is designated $\sin k_0$ and is coupled to the $\sin 2k_0$ state. Explicit calculation gives

$$\hbar^2\omega^2(\cos k_0)=-2K_2(a+b)-\frac{K_2(\mu H)^2}{2(a+b)^2}\left[-4b-4a+c+\frac{2(-b+a)(c-2b)}{c}+\frac{(-a+3b-c)^2}{(-c+a+b)}\right], \tag{22}$$

$$\hbar^2\omega^2(\sin k_0)=-2K_2(a+b)+\frac{K_2(\mu H)^2}{2(a+b)^2}\left[2b+2a-c+\frac{2(-b+a)(c-2b)}{c}-\frac{(-a+3b-c)^2}{(a+b-c)}\right]. \tag{23}$$

For the heavy rare earths, as discussed by both de Gennes[10] and Cooper,[11] one expects that the Fourier transform of the exchange field is a rapidly decreasing function of q_ζ for $q_\zeta>k_0$ so that $c\gg b>a$. Under these conditions, these frequencies may be approximately written

$$\hbar^2\omega^2(\cos k_0)\approx(-2K_2)(a+b)\left[1-\frac{(\mu H)^2(5a+b)}{4(a+b)^3}\right], \tag{24}$$

$$\hbar^2\omega(\sin k_0)\approx(-2K_2)(a+b)\left[1+\frac{(\mu H)^2(5b-7a)}{4(a+b)^3}\right]. \tag{25}$$

The frequency of the $\cos k_0$ mode decreases slowly with H while that of the $\sin k_0$ mode increases slowly.

The method using a second time derivative does not give the mixing of the $S_x(\mathbf{q})$ with the $S_y(\mathbf{q})$ to form the generating operators. This information can be obtained from (13) and (14) to zero order in H. The proper combinations are of the form $\alpha S_x(\mathbf{q})+\beta S_y(\mathbf{q})$ with $\beta/\alpha=\pm(D_0/G_0)^{1/2}$ (evaluated for $H=0$).

Next we look at the way in which these frequencies may be excited by an rf field at low values of H. First, the case when the rf field is polarized along ζ is considered. Then,

$$\mathcal{H}_{\zeta\,\mathrm{rf}}=C\sum_i S_{i\zeta}=-C\sum_i S_{ix}=C_1N^{1/2}S_x(0). \tag{26}$$

Now the creation operator for the state with energy $\hbar\omega(0)$ differs from $S_x(0)$ by terms which go to zero as H goes to zero. Thus, for small values of $\mu H/(a+b)$ (the expansion parameter) $\mathcal{H}_{\mathrm{rf}}$ corresponds to a creation operator for the spin waves of energy $\hbar\omega(0)$. Since the expression (26) is independent of H, the transition probability does not go to zero as H goes to zero. Thus, an rf field polarized along ζ excites $\omega(0)$, given by (21) with intensity typical of ferromagnetic resonance.

In I, it was shown that $\omega(\mathbf{k}_0)$ at $H=0$ is excited by an rf field in the ξ-η plane. When $H\neq 0$, as was shown above, $\omega(\mathbf{k}_0)$ splits into two frequencies, $\omega(\cos k_0)$ and $\omega(\sin k_0)$. Which of these is excited by an rf field in the plane depends on the polarization of the rf field relative to the dc field.

If the rf field is polarized along η,

$$\mathcal{H}_{\eta\,\mathrm{rf}}=C\sum_i S_{i\eta}=C\sum_i[S_{iy}\cos\theta_i+S_{iz}\sin\theta_i]. \tag{27}$$

The resonant effect comes from the S_{iy} term. To lowest

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order in $\mu H/(a+b)$, $\cos\theta_i \approx \cos\mathbf{k}_0\cdot\mathbf{R}_i$; so that

$$\mathcal{H}_{\eta\ \mathrm{reson}} \approx C\sum_i S_{iy}\cos\mathbf{k}_0\cdot\mathbf{R}_i = C_1 N^{1/2}[S_y(\mathbf{k}_0)+S_y(-\mathbf{k}_0)]. \quad (28)$$

Now $S_y(\mathbf{k}_0)+S_y(-\mathbf{k}_0)$ is equal to a combination of the creation and destruction operators for the spin-wave state with energy $\hbar\omega(\cos k_0)$ plus terms that go to zero as $\mu H/(a+b)$ goes to zero. Thus, an rf field along η (perpendicular to the dc field) excites $\omega(\cos k_0)$ given by (22). Since (28) is independent of H, the transition probability is independent of H to lowest order in H, and $\omega(\cos k_0)$ is excited with intensity typical of ferromagnetic resonance. Other frequencies which tend to $\omega(r\mathbf{k}_0)$ as H goes to zero are excited with intensities which are higher order in $\mu H/(a+b)$. For small $\mu H/(a+b)$ these will have much lower intensity.

For an rf field polarized along ξ

$$\mathcal{H}_{\xi\ \mathrm{rf}} = C\sum_i S_{i\xi} = C\sum_i(-S_{iy}\sin\theta_i + S_{iz}\cos\theta_i), \quad (29)$$

and

$$\mathcal{H}_{\xi\ \mathrm{reson}} = -C\sum_i S_{iy}\sin\theta_i \approx C_1 N^{1/2}[S_y(\mathbf{k}_0)-S_y(-\mathbf{k}_0)], \quad (30)$$

to lowest order in H. $[S_y(\mathbf{k}_0)-S_y(-\mathbf{k}_0)]$ is equal to a combination of the creation and destruction operators for the spin-wave state with energy $\hbar\omega(\sin k_0)$ plus terms that go to zero as $\mu H/(a+b)$ goes to zero. Thus, an rf field along ξ (parallel to the dc field) excites $\omega(\sin k_0)$, given by (23). As in the case of the $\cos k_0$ mode and for the same reasons, this resonance has intensity typical of ferromagnetic resonance; while other frequencies are present with intensities higher order in $\mu H/(a+b)$. For small $\mu H/(a+b)$ therefore these other frequencies have much lower intensity.

4. THE HIGH-FIELD CASE

We will next treat the case of high dc field. As has been discussed by Nagamiya *et al.*[6,7] there is a value of magnetic field

$$H_f = 2a/\mu, \quad (31)$$

such that for $H>H_f$ the equilibrium spin arrangement is ordinary ferromagnetic alignment along the dc field. For $H<H_f$, as shown by Nagamiya *et al.*, the energy is lower for a "fan" spin arrangement to be described below. That a transition occurs at $H=H_f$ can also be seen by examining the spin-wave spectrum for case AII of I, when the spin arrangement is ferromagnetic. From equation (I.55) (with $\theta=\pi/2$, so that $H_\perp=H$), it is seen that $\hbar\omega(\mathbf{k}_0)$ is zero for $H=H_f$, indicating some transition.

For $H>H_f$, the situation is just that for ordinary ferromagnetic resonance. The only frequency excited is $\hbar\omega(0)$ obtained from (I.56).

$$\hbar\omega(0) = [(-2K_2S+\mu H/S)(\mu H/S)]^{1/2}. \quad (32)$$

This frequency can be excited with usual ferromagnetic intensity by an rf field polarized in the η-ζ plane. The orbit described by the magnetization in the $\omega(0)$ mode is strongly elliptical with the major axis of the ellipse in the η-ζ plane in the η direction because of the strong anisotropy from K_2. Thus, the greatest intensity will be obtained when the rf field is polarized along η.

For H just less than H_f, the spin arrangement is a ferromagnetic fan,[6,7] given by

$$\sin(\theta_i/2) = 2\delta\sin\mathbf{k}_0\cdot\mathbf{R}_i, \quad (33)$$

$$\delta = [\mu(H_f-H)]^{1/2}/2(2a+b)^{1/2}. \quad (33b)$$

We now examine the spin waves for the fan structure and see what resonance frequencies can be excited by a uniform rf field. The Hamiltonian can be written in terms of the usual spin raising and lowering operators

$$S_i^+ = S_{ix}+iS_{iy}, \quad (34a)$$

$$S_i^- = S_{ix}-iS_{iy}. \quad (34b)$$

Omitting the terms linear in raising and lowering operators this gives

$$\mathcal{H} = -\sum_{i\neq j} J_{ij}\{\tfrac{1}{4}(S_i^+S_j^+ + S_i^-S_j^-)(1-\cos[\theta_i-\theta_j]) + \tfrac{1}{2}S_i^+S_j^-(1+\cos[\theta_i-\theta_j]) + S_{iz}S_{jz}\cos(\theta_i-\theta_j)\} - (\mu H/S)\sum_i S_{iz}\cos\theta_i - \tfrac{1}{4}K_2\sum_i[(S_i^+)^2+(S_i^-)^2+2S_i^+S_i^-]. \quad (35)$$

This can be transformed to spin-wave operators using the usual transformation

$$S_i^+ = \left(\frac{2S}{N}\right)^{1/2}\sum_q a_{\mathbf{q}}e^{-i\mathbf{q}\cdot\mathbf{R}_i}, \quad (36a)$$

$$S_i^- = \left(\frac{2S}{N}\right)^{1/2}\sum_q a_{\mathbf{q}}^* e^{i\mathbf{q}\cdot\mathbf{R}_i}, \quad (36b)$$

$$S_{iz} = S - N^{-1}\sum_{q,q'} a_{\mathbf{q}}^* a_{\mathbf{q}'} e^{i(\mathbf{q}-\mathbf{q}')\cdot\mathbf{R}_i}. \quad (36c)$$

Using the thermal average value for S is equivalent to the random-phase approximation of Sec. 3.

Comparing (36) and (11), it can be seen that

$$S_x(\mathbf{q}) = (a_{\mathbf{q}}+a_{-\mathbf{q}}^*)\frac{(2S)^{1/2}}{2}, \quad (37a)$$

$$S_y(\mathbf{q}) = (a_{\mathbf{q}}-a_{-\mathbf{q}}^*)\frac{(2S)^{1/2}}{2i}. \quad (37b)$$

Using (33), the Hamiltonian (35) may be expanded to terms of second order in δ and transformed to spin-wave operators using (36). The omission of the terms in (35) linear in S_i^+ and S_i^- is justified since these vanish to second order in δ. The transformed Hamiltonian to order δ^2 can be written as

$$\mathcal{H}=E_0+\mathcal{H}_0+\delta^2\mathcal{H}_2. \tag{38}$$

Here E_0 is the equilibrium energy. The terms quadratic in spin-wave operators and independent of δ are given by $\mathcal{H}_0$.

$$\mathcal{H}_0=-2S\sum_q[J(\mathbf{q})-J(0)]a_\mathbf{q}^*a_\mathbf{q}+\frac{\mu H}{S}\sum_q a_\mathbf{q}^*a_\mathbf{q} -\frac{K_2S}{2}\sum_q(a_\mathbf{q}a_{-\mathbf{q}}+a_\mathbf{q}^*a_{-\mathbf{q}}^*+2a_\mathbf{q}^*a_\mathbf{q}). \tag{39}$$

The term given by $\mathcal{H}_2$ is a rather complicated expression quadratic in spin-wave operators which links a given $a_\mathbf{q}$ to $a_\mathbf{q}^*$, $a_{\mathbf{q}\pm2\mathbf{k}_0}$, $a_{-\mathbf{q}}$, $a_{-\mathbf{q}\pm2\mathbf{k}_0}$. To find $\hbar\omega(\mathbf{q})$, in (38) we consider $\mathcal{H}_0$ as the unperturbed Hamiltonian and $\delta^2\mathcal{H}_2$ as the perturbation for small δ. Since $\mathcal{H}_0$ gives a nonzero value of $\hbar\omega(\mathbf{q})$ for all $\mathbf{q}$ but $\pm\mathbf{k}_0$, to find $\hbar\omega(\mathbf{q})$ [except $\omega(\pm\mathbf{k}_0)$] to order δ^2 we need consider only the terms in $\mathcal{H}$ diagonal in $|\mathbf{q}|$. The spin-wave frequency for $\mathbf{k}_0$ is a special case to be considered below. Then keeping only terms diagonal in $\mathbf{q}$, to second order in δ,

$$\mathcal{H}_{\text{diag}}=\sum_q[2SA_\mathbf{q}a_\mathbf{q}^*a_\mathbf{q}+SB_\mathbf{q}(a_\mathbf{q}^*a_{-\mathbf{q}}^*+a_\mathbf{q}a_{-\mathbf{q}})], \tag{40}$$

where

$$2SA_\mathbf{q}=\frac{\mu H}{S}-2S[J(\mathbf{q})-J(0)]-K_2S +4S\delta^2\left[2J(\mathbf{q})-J(\mathbf{q}+\mathbf{k}_0)-J(\mathbf{q}-\mathbf{k}_0)+\frac{2a}{S^2}\right], \tag{41}$$

$$2SB_\mathbf{q}=-K_2S-4S\delta^2[2J(\mathbf{q}) -J(\mathbf{q}+\mathbf{k}_0)-J(\mathbf{q}-\mathbf{k}_0)]. \tag{42}$$

The Hamiltonian of (40) is easily diagonalized using the procedure described in I, that is by transforming to new spin-wave operators $\alpha_\mathbf{q}^*$ and $\alpha_\mathbf{q}$

$$\alpha_\mathbf{q}=\omega_\mathbf{q}a_\mathbf{q}+b_\mathbf{q}a_{-\mathbf{q}}^*. \tag{43}$$

The resulting spin-wave energies are

$$\hbar\omega(\mathbf{q})=2S[(A_\mathbf{q}+B_\mathbf{q})(A_\mathbf{q}-B_\mathbf{q})]^{1/2} \tag{44}$$

correct to order δ^2. In particular, for $q=0$ this gives

$$\hbar\omega(0)=\left[\frac{\mu H}{S}\left(\frac{\mu H}{S}-2K_2S\right)-\frac{8a\delta^2}{S}(-2K_2S)\right]^{1/2}, \tag{45}$$

so that for $\mu(H_f-H)$ small compared to $(2a+b)$, $\hbar\omega(0)$ differs by only a small amount from the expression for $H>H_f$ given in (32).

The spin-wave energy for $\mathbf{q}=\pm\mathbf{k}_0$ is a special case for two reasons. First, for $\mathbf{q}=\pm\mathbf{k}_0$ the unperturbed energy, $\mathcal{H}_0$, in (38) gives $\hbar\omega(\pm\mathbf{k}_0)=0$. Second, $\delta^2\mathcal{H}_2$ connects these two degenerate states, and the problem must be properly diagonalized to remove this degeneracy. The pertinent terms in $\mathcal{H}$ (all terms linking $+\mathbf{k}_0$ and $-\mathbf{k}_0$ to order δ^2) are

$$\mathcal{H}(k_0)=2SA_{\mathbf{k}_0}[a_{\mathbf{k}_0}^*a_{\mathbf{k}_0}+a_{-\mathbf{k}_0}^*a_{-\mathbf{k}_0}] +2SB_{\mathbf{k}_0}[a_{\mathbf{k}_0}a_{-\mathbf{k}_0}+a_{\mathbf{k}_0}^*a_{-\mathbf{k}_0}^*] +\frac{2\delta^2a}{S}[-4(a_{\mathbf{k}_0}^*a_{-\mathbf{k}_0}+a_{-\mathbf{k}_0}^*a_{\mathbf{k}_0})+a_{\mathbf{k}_0}a_{\mathbf{k}_0} +a_{\mathbf{k}_0}^*a_{\mathbf{k}_0}^*+a_{-\mathbf{k}_0}a_{-\mathbf{k}_0}+a_{-\mathbf{k}_0}^*a_{-\mathbf{k}_0}^*]. \tag{46}$$

Then we seek a transformation to new creation and destruction operators, $\alpha_{\mathbf{k}_0}^*$ and $\alpha_{\mathbf{k}_0}$, which diagonalize (46).

$$\alpha_{\mathbf{k}_0}=w_{\mathbf{k}_0}a_{\mathbf{k}_0}+b_{\mathbf{k}_0}a_{-\mathbf{k}_0}^*+w_{-\mathbf{k}_0}a_{-\mathbf{k}_0}+b_{-\mathbf{k}_0}a_{\mathbf{k}_0}^*. \tag{47}$$

There are two different $\alpha_{\mathbf{k}_0}$ that diagonalize (46). One of these is the symmetric solution labeled $\cos k_0$ as in (22).

$$\alpha_{\cos k_0}=s\alpha_x(\cos k_0)+t\alpha_y(\cos k_0), \tag{48}$$

where

$$\alpha_x(\cos k_0)=(a_{\mathbf{k}_0}+a_{-\mathbf{k}_0}^*)+(a_{-\mathbf{k}_0}+a_{\mathbf{k}_0}^*) =(2/S)^{1/2}[S_x(\mathbf{k}_0)+S_x(-\mathbf{k}_0)], \tag{49a}$$

$$\alpha_y(\cos k_0)=(a_{\mathbf{k}_0}-a_{-\mathbf{k}_0}^*)+(a_{-\mathbf{k}_0}-a_{\mathbf{k}_0}^*) =i(2/S)^{1/2}[S_y(\mathbf{k}_0)+S_y(-\mathbf{k}_0)], \tag{49b}$$

with energy given by

$$\hbar\omega(\cos k_0)=[(8/S)(-K_2S)(b-a)]^{1/2}\delta. \tag{50}$$

The other solution is the antisymmetric solution labeled $\sin k_0$ as in (23)

$$\alpha_{\sin k_0}=p\alpha_x(\sin k_0)+r\alpha_y(\sin k_0), \tag{51}$$

where

$$\alpha_x(\sin k_0)=(a_{\mathbf{k}_0}+a_{-\mathbf{k}_0}^*)-(a_{-\mathbf{k}_0}+a_{\mathbf{k}_0}^*) =\left(\frac{2}{S}\right)^{1/2}[S_x(\mathbf{k}_0)-S_x(-\mathbf{k}_0)], \tag{52a}$$

$$\alpha_y(\sin k_0)=(a_{\mathbf{k}_0}-a_{-\mathbf{k}_0}^*)-(a_{-\mathbf{k}_0}-a_{\mathbf{k}_0}^*) =i\left(\frac{2}{S}\right)^{1/2}[S_y(\mathbf{k}_0)-S_y(-\mathbf{k}_0)], \tag{52b}$$

with energy given by

$$\hbar\omega(\sin k_0)=\left\{\frac{8}{S}(-K_2S)(5a+b)\right\}^{1/2}\delta. \tag{53}$$

The next order contributions to (50) and (53) are of order δ^3.

It remains to be seen which of these frequencies can be excited with a uniform rf field. If the rf field is polarized along ζ

$$\mathcal{H}_{\text{rf}\,\zeta}=C\sum_i S_{ix}=C_1N^{1/2}(a_0+a_0^*) =C_2N^{1/2}(w_0\alpha_0-b_0\alpha_0^*+w_0\alpha_0^*-b_0\alpha_0). \tag{54}$$

This excites the frequency $\omega(0)$ where the intensity $\mathcal{I}_\zeta(0)$ is given by

$$\mathcal{I}_\zeta(0)=CN(w_0-b_0)^2. \quad (55)$$

The coefficients w_0 and b_0 can be found by the same diagonalization procedure used to find the energy expression, (44). $\mathcal{I}_\zeta(0)$ for $H<H_f$ differs from that for $H>H_f$ only by terms of order δ^2, so that the intensity in the fan region is essentially the same as in the ferromagnetic region.

If the rf field is polarized along η,

$$\mathcal{H}_{\mathrm{rf}\ \eta}=C\sum_i S_{i\eta}=C\sum_i(S_{iy}\cos\theta_i+S_{iz}\sin\theta_i). \quad (56)$$

The resonant effect comes from the term in S_{iy}

$$\mathcal{H}_{\eta\ \mathrm{reson}}=C\sum_i S_{iy}\cos\theta_i, \quad (57)$$

and to second order in δ^2

$$\cos\theta_i=1-8\delta^2\sin^2\mathbf{k}_0\cdot\mathbf{R}_i. \quad (58)$$

So,

$$\mathcal{H}_{\eta\ \mathrm{reson}}=C(a_0-a_0^*)+\text{terms of order }\delta^2. \quad (59)$$

Thus a field polarized along η excites $\hbar\omega(0)$, where the intensity $\mathcal{I}_\eta(0)$ is given by

$$\mathcal{I}_\eta(0)=CN(w_0+b_0)^2, \quad (60)$$

with the same constant as in (55). It is a straightforward procedure to find b_0 and w_0 and to examine the ratio $\mathcal{I}_\eta(0)/\mathcal{I}_\zeta(0)$. This is given by

$$\mathcal{I}_\eta(0)/\mathcal{I}_\zeta(0)\approx-2K_2S^2/\mu H_f \quad \text{as} \quad \delta\to 0, \quad (61)$$

so that for large anisotropy $\mathcal{I}_\eta(0)\gg\mathcal{I}_\zeta(0)$ as expected from the ellipticity of the magnetization orbit for this mode. Thus, an rf field polarized along η (planar) will be more effective than a field polarized along ζ (perpendicular to the plane) in exciting the $\omega(0)$ resonance. The frequency $\omega(2\mathbf{k}_0)$ will also be excited by a field polarized along η but with intensity proportional to δ^4, which is probably negligibly small.

Finally, we examine the resonance excited for an rf field polarized along ξ (parallel to the dc field).

$$\mathcal{H}_{\mathrm{rf}\ \xi}=C\sum_i S_{i\xi}=C\sum_i(-S_{iy}\sin\theta_i+S_{iz}\cos\theta_i). \quad (62)$$

The resonant effect comes from the term in S_{iy}.

$$\mathcal{H}_{\xi\ \mathrm{reson}}=C\sum_i S_{iy}\sin\theta_i, \quad (63)$$

$$\begin{aligned}\sin\theta_i&=2\sin\frac{\theta_i}{2}\cos\frac{\theta_i}{2}\\&=4\delta\sin\mathbf{k}_0\cdot\mathbf{R}_i(1-4\delta^2\sin^2\mathbf{k}_0\cdot\mathbf{R}_i)^{1/2}\\&=4\delta\sin\mathbf{k}_0\cdot\mathbf{R}_i,\end{aligned} \quad (64)$$

to order δ^2.

$$\begin{aligned}\mathcal{H}_{\xi\ \mathrm{reson}}&\approx C\delta\sum_i S_{iy}\sin\mathbf{k}_0\cdot\mathbf{R}_i\\&=C_1N^{1/2}\delta[S_y(k_0)-S_y(-k_0)],\end{aligned} \quad (65)$$

and from (52)

$$\mathcal{H}_{\xi\ \mathrm{reson}}=C_2N^{1/2}\delta\alpha_y(\sin k_0). \quad (66)$$

From (51) and (52)

$$\alpha_y(\sin k_0)=(\alpha_{\sin k_0}+\alpha_{\sin k_0}^*)/2r, \quad (67)$$

so an rf field polarized along ξ excites the frequency $\omega(\sin k_0)$ given by (53) with intensity

$$\mathcal{I}_{\sin k_0}\sim N\delta^2/r^2. \quad (68)$$

The condition

$$[\alpha_{\sin k_0},\alpha_{\sin k_0}^*]=1, \quad (69)$$

gives

$$8pr=1, \quad (70)$$

and the transformation which gives the energy has

$$\frac{p}{r}=\left\{\frac{-2K_2S}{(4\delta^2/S)(5a+b)}\right\}^{1/2}. \quad (71)$$

Thus,

$$r^2=C\delta, \quad (72)$$

and

$$\mathcal{I}_{\sin k_0}\sim N\delta. \quad (73)$$

This means that the intensity of the $\omega(\sin k_0)$ resonance goes to zero with δ in the same way as the frequency goes to zero.

5. EXTRAPOLATION TO INTERMEDIATE FIELDS: BEHAVIOR AT H_c

The theory discussed in Secs. 3 and 4 deals with the circumstances for very small magnetic field, and for magnetic field in the vicinity of H_f. It is desirable to extend the discussion to intermediate fields, in particular for fields near H_c. To the present time, it has proved impossible to give an exact theory for the first-order transition at H_c. Nagamiya and co-workers[6,7] have given an approximate expression for H_c/H_f.

$$H_c/H_f=[(1+\beta_N)(2+\beta_N)]^{1/2}-(1+\beta_N), \quad (74a)$$

where

$$\beta_N=b/a. \quad (74b)$$

This expression is quite insensitive to the value of β_N. For $\beta_N=0$, $H_c/H_f=0.41$; for $\beta_N=1$, $H_c/H_f=0.45$; while the ratio tends to 0.5 as β_N goes to ∞.

To obtain the values of the resonance frequencies at fields intermediate to 0 and H_f, one can extrapolate towards $H=H_c$ from both sides using the formula for $H\sim 0$ and $H\sim H_f$. Since the frequencies are discontinuous at H_c, as discussed below, it appears that these extrapolations may be adequate right to H_c. In effect, the pertinent low-field resonance frequency expressions in Sec. 3 are expansions in $\mu H/(a+b)$ while those for high fields in Sec. 4 are expansions in $\mu H/(2a+b)$. Thus if $b\gg a$, as it is in the two-layer interaction model for the rare earths,[1,11] the expansion parameters are still small at $H=H_c$.

Since the transition at H_c is first order, the spin-wave spectrum is expected to change discontinuously. In this respect, the behavior is much different from that for the second-order transition at H_f. For the H_f transition there is a spin-wave energy that goes to zero, and the spin-wave spectrum for the fan goes over continuously into that for the ferromagnetic region. This is expected since the spin configuration changes continuously at a second-order transition. The new ground state is a state reached by small continuous deviations of spin arrangement from the old ground state, and may therefore be described by spin waves. The equality of energy for the new and old ground states at the transition implies that some spin-wave energy goes to zero. In the ferromagnetic phase the average number of spin waves with $\mathbf{q}\sim\mathbf{k}_0$ will measure the tendency of the order to distort into the fan. This number,

$$\bar{n}\sim\int k^2dk/(\exp[E(\mathbf{k})/kT]-1), \tag{75}$$

where $\mathbf{k}=\mathbf{q}-\mathbf{k}_0$ and $E(\mathbf{k})\sim H_A{}^{1/2}[\alpha k^2+(H-H_f)]^{1/2}$ at small k, remains finite and well behaved as $H\rightarrow H_f$. (H_A is the anisotropy field, $-2K_2S/|\lambda|\beta$ in the present case.) However, the derivative $\partial\bar{n}/\partial H$ diverges (near $k=0$) as is expected for a second-order transition.

For the first-order transition at H_c, on the other hand, the spin-wave spectrum is expected to be discontinuous with no spin-wave state of zero energy. Such behavior is expected, in general, for a first-order transition because the equilibrium spin configuration changes discontinuously, and the new ground state cannot be described by small spin deviations from the old ground state.

While it is impossible to give an exact description of the behavior at H_c in the present case, there is an analogous problem that can be described exactly, which illustrates these ideas. This is the situation where the Hamiltonian for the system consists of the same terms as (1) plus an additional planar anisotropy of the form, $-P\sum_i S_{i\xi}{}^2$, where P is positive and small compared to K_2. The exchange is taken as antiferromagnetic between neighboring layers only. At zero field the equilibrium spin arrangement is antiferromagnetic along ξ. As H increases, the equilibrium arrangement does not change until a field H_c^* where the spin arrangement changes discontinuously to a canted structure with the two sublattices inclined at $\pm\theta(0<\theta<\pi/2)$ to the $+\xi$ axis. As the field is increased further, θ decreases continuously until $\theta=0$ (ferromagnetic alignment) is reached at a field H_f^*.

For this antiferromagnetic model, it is possible to calculate both the equilibrium spin configuration and the spin-wave spectrum exactly for all values of H as is shown in the Appendix. The behavior at the two transitions agrees with the general discussion above.

A schematic plot of the relevant spin-wave frequencies ($\mathbf{q}=0$, $\pm\mathbf{k}_0$ and combinations) as a function

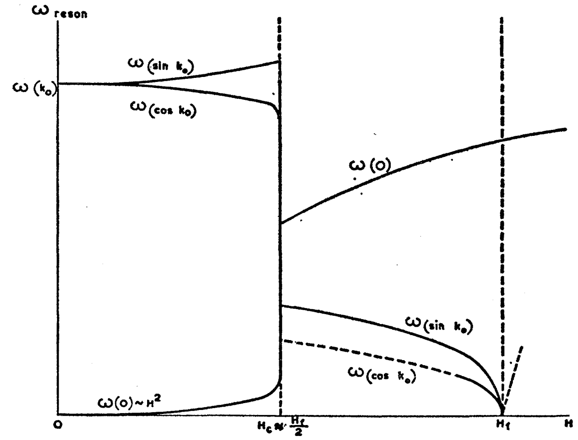

FIG. 1. Variation of resonance frequencies with applied field for the case of zero planar anisotropy. (The resonances can be excited only for values of H where the variation is indicated by a solid line.)

of H obtained from the extrapolation scheme described above is shown in Fig. 1 for the case, valid for the rare-earth spirals, where $c\gg b\gg a$ and the axial anisotropy is large. The $\mathbf{q}=0$ mode energy rises quadratically at small H and is still small at $H=H_c$ if $b\gg a$. At H_c there is a sharp discontinuity, and $\omega(0)$ goes to the value given by (45) which has only a small correction in the fan amplitude from the ferromagnetic result (32). In some cases of interest (e.g., dysprosium discussed in Sec. 7) the usual signal frequencies for ferromagnetic resonance would fall in the range of the discontinuity at H_c. In these cases, the $\omega(0)$ frequency would be observed at the critical field, H_c. Nagamiya[12] has discussed an analogous critical resonance for the antiferromagnetic resonance in $CuCl_2\cdot 2H_2O$.

The $\mathbf{q}=\mathbf{k}_0$ mode energy is large in the spiral phase. For nonzero H, $\omega(\mathbf{k}_0)$ splits into two frequencies, one increasing, the other decreasing with H, but the change is small for $H<H_c$ if $b\gg a$. There is a discontinuity at H_c from about $|\lambda|\beta(H_AB')^{1/2}$ to $|\lambda|\beta(H_AA')^{1/2}$, where H_A denotes the axial anisotropy field,

$$H_A=-2K_2S^2/\mu \tag{76a}$$

and

$$A'=a/\mu,\quad B'=b/\mu \tag{76b}$$

represent the fields corresponding to the energies a and b. The energies for both the $\sin k_0$ and the $\cos k_0$ mode fall to zero at H_f. Only the $\sin k_0$ mode is expected to be observable with reasonable intensity for $H>H_c$; and since its intensity goes as δ, it is likely to be weaker than the usual ferromagnetic resonance intensity.

In general, for the parameters pertinent to a particular material at a particular temperature, one can draw a diagram such as Fig. 1. The signal frequency then would correspond to a horizontal line at the appropriate value of ω, and resonances would be observed at fields

[12] T. Nagamiya, Progr. Theoret. Phys. (Kyoto) 11, 309 (1954).

given by its intersections with the curves. For the case where H_A is large and B' larger than A', resonance is most likely to be observed at $H=H_c$ and weakly at an H slightly less than H_f. If H_A and B' are smaller, more complex patterns may be observed.

6. EXTENSIONS OF RESONANCE CONDITIONS

Various more complicated cases than that of a plane spiral with no planar anisotropy were considered in I. The labor of extending the calculations of Secs. 3 and 4 to such cases proved prohibitive, but it is possible to make qualitative estimates of the behavior of the pertinent spin-wave frequencies based on those calculations.

A. Cone

The magnetic moments here lie along the generators of a cone of semivertical angle, ψ, so that their projections into the hexagonal plane is a spiral. This configuration occurs at low T in Er. The components are

$$\begin{aligned} S_\xi(\mathbf{R}_i)&=S\sin\psi\cos(\mathbf{k}_0\cdot\mathbf{R}_i),\\ S_\eta(\mathbf{R}_i)&=S\sin\psi\sin(\mathbf{k}_0\cdot\mathbf{R}_i),\\ S_\zeta(\mathbf{R}_i)&=S\cos\psi. \end{aligned} \tag{77}$$

The resonances for the unperturbed spiral cone have been discussed in I. For $H=0$, both the resonances corresponding to $|q|=k_0$ and that for $q=0$ can be excited depending on the polarization of the rf field. Just as for the planar spiral, $\omega(0)=0$ for $H=0$. The $+\mathbf{k}_0$ and $-\mathbf{k}_0$ frequencies, however, are split at $H=0$ for the cone. From (I.45),

$$\hbar\omega(\pm\mathbf{k}_0)=\pm\frac{(a-b)\cos\psi}{S}+\frac{1}{S}\{[a+b][\cos^2\psi(a+b)-2S^2\sin^2\psi(K_2-L)]\}^{1/2}, \tag{78}$$

(with correction of a misprint in the sign of L). Here, L depends on higher order anisotropies and is defined by (I.28). In the presence of a field along the ζ direction the cone angle changes by a small amount

$$\delta(\cos\psi)=H/H_A', \tag{79}$$

where $H_A'=2S^2(-K_2+L)/\mu$ is the axial anisotropy field which stabilizes the conical configuration. For a field along ζ, the value of $\hbar\omega(\pm\mathbf{k}_0)$ for nonzero H is obtained from (78) by simply replacing ψ by its new value. Thus, the change in (78) is small for attainable fields along ζ in the rare earths where H_A' is very large. $\omega(0)=0$ is unchanged by a field along ζ. As for the plane spiral, a field in the plane is necessary to affect the resonance frequencies.

In the presence of a field $H_\perp$ in the plane along ξ the change in cone angle ψ is small, but it is possible to break down the spiral structure. The components in the plane show a behavior similar to that discussed in Sec. 5. At a critical field $H_{\perp c}$ a fan structure is formed which closes up to give a ferromagnetic structure as $H_\perp$ is increased to a value $H_{\perp f}$. The ferromagnetic alignment is in the ξ-ζ plane at an angle to ζ slightly greater than the cone angle. The field $H_{\perp f}$ is given by

$$\mu H_{\perp f}=2a\sin\psi. \tag{80}$$

For small fields in the plane, in analogy to the results of Sec. 3, one expects the changes in $\omega(0)$ and $\omega(\pm\mathbf{k}_0)$ to be small; the expressions for the resonant frequencies are clearly expansions in $\mu H_\perp/(a+b)\sin\psi$ rather than $\mu H/(a+b)$. As $H_\perp$ passes $H_{\perp c}$ there will be a discontinuous change in the frequencies, and at $H_\perp<H_{\perp c}$ the energies will change to join continuously on to those obtained in the ferromagnetic region. From Eq. (55) of I, $\omega(\mathbf{k}_0)$ goes to zero at $H_{\perp f}$. At $H_{\perp f}$ the $\omega(0)$ frequency goes to the ordinary ferromagnetic frequency

$$\hbar\omega(0)=\left\{-\frac{\lambda\beta H_\perp}{\sin\psi}\left[-\frac{\lambda\beta H_\perp}{\sin\psi}-2S\sin^2\psi[K_2-L]\right]\right\}^{1/2}, \tag{81}$$

which is excited by an rf field in the plane perpendicular to the magnetization. Thus, the general behavior of the spin-wave resonant frequencies is again that shown in Fig. 1, and the conditions on $H_\perp$ likely to provide resonances are as for H in the planar spiral. The principal change is the splitting of the $+\mathbf{k}_0$ and $-\mathbf{k}_0$ frequencies at low dc field. The expected behavior for erbium is discussed in Sec. 7.

B. Effect of Planar Anisotropy

In addition to the applied magnetic field there is an additional force which tends to produce some distortion of the spiral arrangement of spins in the plane, namely the anisotropy within that plane which in the rare earths is hexagonal. Calculations of this effect are complicated, but the calculations carried out in I and Sec. 3 above indicate the form of the results which might be expected. As in Sec. 3 the change in the spin-wave frequencies due to this distortion alone may be expanded in powers of

$$(12H_h/G)^2, \tag{82}$$

where the hexagonal anisotropy field H_h is related to the hexagonal crystal field energy $\frac{1}{2}P_6{}^6[(S^+)^6+(S^-)^6]$ by

$$|\lambda|\beta H_h=-S^5P_6{}^6 \tag{83}$$

and

$$|\lambda|\beta G=[2J(\mathbf{k}_0)-J(7\mathbf{k}_0)-J(5\mathbf{k}_0)]S. \tag{84}$$

In the spiral structure this may be expected to cause changes in the $\mathbf{k}_0$ energy given by (6) to order $(12H_h/G)^2$. For the sharply decreasing $J(\mathbf{q})$ expected for the heavy rare-earth metals[10,11] and typical values of $\mathbf{k}_0$,

G is quite large and the change in the $\mathbf{k}_0$ frequency is likely to be small. More interesting is the change in $\omega(0)$ which in analogy with (21) may be expected to be

$$[\hbar\omega(0)]^2 \sim (12H_h)^4 H_A/G. \quad (85)$$

This gives a small nonzero $\omega(0)$ as $H \rightarrow 0$.

The addition of an external field in the presence of such an hexagonal field raises further complications. For example, the observed values of H_c and H_f will be affected since the anisotropy field may help (or hinder) the creation of a ferromagnetic arrangement as it does in Dy.[1,3,13] The effects on the transition at H_f can be seen from the results of I for the ferromagnetic phase using (I.87)

$$[\hbar\omega(\mathbf{k}_0)]^2 = |\lambda|\beta(H - 2A' + 36H_h) \times (H_A + H + 6H_h - 2A'), \quad (86)$$

which goes to zero when $H = 2A - 36H_h$, and at somewhat higher fields when H is applied in a direction other than an easy hexagonal axis. Thus, the second-order transition takes place at a new critical field $H_f' = 2A' - 36H_h$. Below this field the $\mathbf{k}_0$ energy will increase again from zero in a roughly symmetrical fashion as in Sec. 4. Also at $H > H_f'$ the $\mathbf{q}=0$ frequency is modified.

$$[\hbar\omega(0)]^2 = |\lambda|\beta(H + 36H_h)(H_A + H + 6H_h). \quad (87)$$

At $H < H_f'$ it has an extrapolation only slightly different from this while the fan structure persists.

The transition to a spiral takes place at a new critical field $H_c' = H_c - H_h$. Below this the effects of H_h are additive to (21), (24), and (25).

When H_h is large enough ($H_h > A'$), rather than just slightly perturbing the spiral structure, it may (as in dysprosium[1,3] at low T) stabilize the ferromagnetic structure so that (87) holds down to $H=0$. The hexagonal anisotropy then causes a finite spin-wave energy at $\mathbf{q}=0$ as calculated in (I.107). This energy gap in the spin wave spectrum has important consequences in the specific heat[11] and other properties. The gap may be destroyed by a field in the hard hexagonal direction. Then from (I.107)

$$[\hbar\omega(0)]^2 = |\lambda|\beta(36H_h - H)(H_A - H + 6H_h), \quad (88)$$

so that $\omega(0)$ goes to zero for $H = 36H_h$.

There is an intermediate case $A' > H_h > A'/35$ where the H_f transition is eliminated, and only a single first-order transition from spiral to ferromagnet is observed. (In this case the $\mathbf{k}_0$ spin-wave energies are greater than zero for all H.) The origin of the difference of a factor of 36 multiplying H_h in H_c' and H_f' may be seen by considering a spin of small angular deviation ϵ from the hexagonal easy axis. The energy is then $\sim P_6^6 \cos 6\epsilon$ and the anisotropy exerts a torque $\sim -d(P_6^6 \cos 6\epsilon)/d\epsilon \approx 36 P_6^6 \epsilon$. The exchange forces must overcome this torque to form a fan. At the first-order transition, however, the energy is compared and the factor 36 is absent. The resonant frequencies in the intermediate case are shown in Fig. 2.

C. Demagnetizing Field Effects

For $H > H_c$, where the crystal has a sizeable static magnetization $\mathfrak{M}$, the long range dipole-dipole interactions will give rise to large demagnetizing fields for some sample shapes. These fields will modify the static properties, in particular the transition fields H_c and H_f, as well as the resonant frequencies. If there is a large demagnetizing factor in the direction of $\mathbf{H}$ (i.e., the ξ axis), the transition at H_c cannot be sharp. The crystal will presumably form domains of fan structure where the local field is above critical, while in the rest of the crystal it remains below critical. Under these circumstances the transition will begin at $H = H_c$ but only be completed at $H = H_c + N_\xi \mathfrak{M}$ (where N is the demagnetizing factor). The resonance which occurs at the transition field will now have a width in field comparable to the demagnetizing field. It will therefore be essential to use crystals with zero demagnetizing factor in the direction of the applied field. This is the case in the usual disk geometry used in ferromagnetic resonance with the dc field parallel to the surface. Since the rf field is applied in the plane of the disk, the best intensity for $\omega(0)$ with $H \geq H_c$ and $\omega(\cos k_0)$ with $H < H_c$ is obtained with the ζ axis out of the plane. On the other hand, to observe the low-frequency $\omega(0)$ resonance at $H < H_c$, it would be necessary to have ζ in the plane. Since $\mathfrak{M}$ is continuous at H_f, there will be no similar width effect near this field; the external field at which the ferromagnetic phase begins will, however, be $H_f + N_\xi \mathfrak{M}$.

In addition, the resonant frequencies will be modified throughout the fan and ferromagnetic structures. Following Kittel[14] as in (I.88), the main resonance will become

$$[\hbar\omega(0)]^2 = \left[-2K_2 S + \frac{\mu}{S}(H - (N_\xi - N_\zeta)\mathfrak{M})\right] \times \left[\frac{\mu}{S}(H - (N_\xi - N_\eta)\mathfrak{M})\right] \quad (89)$$

instead of (32) in the ferromagnetic region. In the fan region (50) and (53) will be modified as δ becomes $[\mu(H_f + N_\xi \mathfrak{M} - H)/4(2a+b)]^{1/2}$.

D. Polycrystals

The resonance expected from polycrystalline samples of crystals with a plane spiral spin structure due to a strong axial anisotropy field are readily calculated in the case where the planar anisotropy is small. Taking

[13] D. R. Behrendt, S. Legvold, and F. H. Spedding, Phys. Rev. **109**, 1544 (1958).

[14] C. Kittel, Phys. Rev. **73**, 155 (1948).

the applied field along (0,0,1) a typical crystallite will have its axis $\boldsymbol{\zeta}$ at θ to this, say $(\sin\theta,0,\cos\theta)$. If $H \ll H_A$ the magnetization will remain in the plane perpendicular to this axis, along $\boldsymbol{\xi}=(-\cos\theta, 0, \sin\theta)$. The $\mathbf{y}$ axis, perpendicular to $\boldsymbol{\mu}$ in the plane is then (0,1,0). For the normal resonance condition, $H_{\text{rf}} \perp H$, say along $(\cos\phi, \sin\phi, 0)$.

Thus the main effect of the applied field is to produce an effective field $H \sin\theta$ in the plane, which can distort the spiral. This particular crystallite will make a transition to a fan structure when $H \sin\theta = H_c$. For a given H those crystallites with $\theta > \theta_c$ will have fan structures and those with $\theta < \theta_c$ spirals, where

$$\theta_c = \sin^{-1}(H_c/H). \tag{90}$$

The number of crystallites with axes lying between θ and $\theta + d\theta$ is proportional to $\sin\theta d\theta$. If the magnetization of each crystallite changes discontinuously by μ' at $H \sin\theta = H_c$, the magnetization observed along H coming from such crystallites in the fan phase is

$$\int_{\theta_c}^{\pi/2} \mu' \sin^2\theta d\theta = \frac{\mu'}{2}\left\{\frac{\pi}{2} - \sin^{-1}\left(\frac{H_c}{H}\right) + \frac{H_c}{H}\left[1-\left(\frac{H_c}{H}\right)^2\right]^{1/2}\right\}. \tag{91}$$

A similar expression is shown plotted by Herpin and Meriel.[15] A discontinuity occurs in $d\mu/dH$ at $H = H_c$. In the presence of planar anisotropy this discontinuity is smoothed out.

If a resonance occurs for a field H_0 applied in the plane, it will be observed at H in those crystallites with

$$\theta = \sin^{-1}(H_0/H). \tag{92}$$

Those resonances excited by $\mathbf{H}_{\text{rf}} \| \boldsymbol{\xi}$ have an intensity $\mathcal{I}(\theta)$ proportional to $\cos^2\theta$, $\mathbf{H}_{\text{rf}} \| \boldsymbol{\zeta}$ to $\sin^2\theta$, and $H_{\text{rf}} \| \eta$ to unity, when averaged over ϕ. The resonant intensity as H varies

$$\mathcal{I}(H) = \mathcal{I}(\theta) \sin\theta (d\theta/dH)$$

$$\propto \frac{H_0^2}{H^3}\left[1-\left(\frac{H_0}{H}\right)^2\right]^{1/2} \quad H_{\text{rf}} \| \xi, \tag{93a}$$

$$\propto \frac{H_0^2}{H^3}\left[1-\left(\frac{H_0}{H}\right)^2\right]^{-1/2} \quad H_{\text{rf}} \| \eta, \tag{93b}$$

$$\propto \frac{H_0^4}{H^5}\left[1-\left(\frac{H_0}{H}\right)^2\right]^{-1/2} \quad H_{\text{rf}} \| \zeta. \tag{93c}$$

The last two cases give a discontinuous rise at $H = H_0$ followed by a gradual tail. The first case gives a rise at $H = H_0$ with infinite slope and a maximum at $H = \sqrt{3} H_0/2$.

[15] A. Herpin and P. Meriel, J. Phys. Radium **22**, 337 (1961).

The $\mathbf{k}=0$ resonance is given by (93c) for $H_0 < H_c$ and by (93b) when $H_0 > H_c$ although in the latter case there is a weak component like (93c). The $\cos k_0$ resonance is like (93b) for $H_0 < H_c$ and is not observed if $H_0 > H_c$. The $\sin k_0$ resonance is like (93a). Thus, it appears that interesting information can be obtained from polycrystalline samples.

7. NUMERICAL ESTIMATES

The above theory allows a more complete discussion of the results to be expected in typical cases than was possible in I.

A. Dysprosium

The possibility of exciting the ordinary $\omega(0)$ resonance in the low-temperature ferromagnetic phase of dysprosium was discussed in I under Case AV and in Eqs. (104)–(107) of the Discussion. As discussed there, and in Sec. 6B of this paper, $\omega(0)$ might be brought to quite a low value by applying a sufficiently strong field along a hard hexagonal axis. The field necessary to do this, however, is of the order $36H_h$. Since $P_6{}^6S^6$ is about 1 or 2 cm^{-1}, this field would be of the order of fifty or a hundred thousand gauss. This can also be seen from the magnetization measurements of Behrendt *et al.*[13] The value of field necessary to align the magnetization along a hard axis at equilibrium is also $36H_h$. From Fig. 1 of Ref. 13, it can be seen that at 4.2°K with a field of 8000 G applied along a hard direction, the magnetization is not saturated, and is approaching the value with H along an easy axis quite slowly as H increases. Thus, the field necessary to bring $\hbar\omega(0)$ into the range of usual microwave frequencies is likely to be higher than that usually obtainable in a laboratory.

It is interesting to look at the possibility of doing resonance experiments in the high-temperature spiral phase of dysprosium. The three-layer model of Elliott is used to obtain the values of a, b, c and $P_6{}^6$ for a typical case at $T \sim 100$°K when $k_0c' = \pi/6$ and $M = 0.846$. The parameters so obtained are

$$a = 2.4M^2 \text{ cm}^{-1},$$
$$b = 18.4M^2 \text{ cm}^{-1},$$
$$c = 102.4M^2 \text{ cm}^{-1},$$
$$P_6{}^6S^6 = -1.56M^6 \text{ cm}^{-1},$$
$$\mu = 0.467 \times 10^{-3} M \text{ cm}^{-1}/\text{G}.$$

The value

$$K_2 = -1.48 \text{ cm}^{-1}$$

was found in Ref. 11 by fitting the difference of the parallel and perpendicular paramagnetic Curie temperatures with an axial anisotropy of the form $-K_2 S_{n\zeta}{}^2$. The point-charge crystal field model of Elliott[1] gives a value about 0.8 times this. This difference is not sufficient to affect the qualitative behavior discussed below.

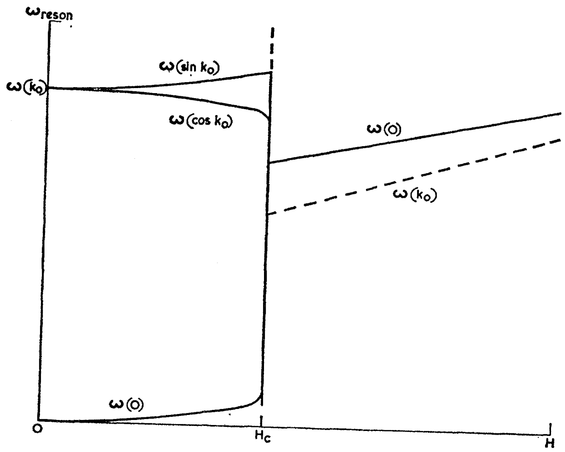

FIG. 2. Variation of resonance frequencies with applied field for the case of planar anisotropy large enough to eliminate the fan phase.

Since the hexagonal anisotropy energy drops off as M^6, its magnitude has decreased sufficiently at this temperature so that its effect is to give rise to the intermediate case discussed at the end of Sec. 6(b) and illustrated by Fig. 2. The value of H_c from exchange effects alone is 4.3×10^3 G. For a field along an easy axis, hexagonal anisotropy lowers this to $H_c'=2.9\times10^3$ G, while for a field along a hard axis the critical field $H_c''=3.4\times10^3$ G. Below H_c', the hexagonal anisotropy has negligible effect on the resonance frequencies for the sharply rising $J(\mathbf{q})$ of dysprosium. For $H<H_c'$, $\omega(0)\alpha H^2$ is given by (21). At $H=H_c'$ for field along an easy axis, $\hbar\omega(0)\approx0.01$ cm^{-1}. Thus, throughout the region $H<H_c'$, $\omega(0)$ is very low compared to the signal frequencies usual for ferromagnetic resonance experiments. It is possible that the curve of $\omega(0)$ versus H for $H<H_c'$ could be measured by radio-frequency techniques. The line has the intensity characteristic of ferromagnetic resonance, but its width may be too great. For $H>H_c'$ along an easy axis, $\omega(0)$ is given by (87), and at H_c, $\hbar\omega(0)\approx8.2$ cm^{-1}. Thus, for the usual signal frequencies between 0.1 and 1 cm^{-1}, $\omega(0)$ is probably observed as a resonance at the critical field H_c.

At $H=0$, $\hbar\omega(\mathbf{k}_0)=6.6$ cm^{-1}. The $\omega(\sin k_0)$ frequency increases by less than 1% for $H=H_c'$, while $\omega(\cos k_0)$ decreases by a similar amount. At H_c', these frequencies change discontinuously to the value 7.4 cm^{-1} obtained from (86). [The values of $\omega(0)$ and $\omega(k_0)$ at H_c'' with field along a hard hexagonal axis obtained from (I.85) and (I.87) are about 1% less than the corresponding values at H_c' for field along an easy axis.] Thus, the $\mathbf{k}_0$ frequencies are probably too high to be observed with the usual signal frequencies even as a critical resonance.

Therefore, the only resonance for dysprosium in the spiral phase observable with the usual signal frequencies is a resonance at H_c' (for dc field along an easy axis) coming from $\omega(0)$. This is excited by an rf field polarized in the ζ-η plane, probably most strongly for an rf field along η.

B. Erbium

The field for ferromagnetic alignment $H_{\perp f}$ is given in (80) in terms of a and ψ. The cone angle (27°) can be found from the neutron diffraction results[16] and the magnetization data.[17] On the other hand, $H_{\perp c}$ corresponds to the critical field[17] of 17 kOe at 4.2°K. Then if $H_{\perp f}\approx2H_{\perp c}$ as in the planar case, since ψ is known, a can be found from (80). In the two-layer model,[1] b is determined from a and the spiral angle (41°).

From a and b the splitting of the two frequencies for $\pm\mathbf{k}_0$ at $H=0$ is found to be 19.6 cm^{-1}. It is of interest to see whether the lower of these two frequencies would fall in the usual microwave range. This depends on the anisotropy term, (K_2-L). If this is evaluated on the point-charge model,[1] then the lower $\mathbf{k}_0$ frequency is about 9 cm^{-1}. The anisotropy constants obtained from the point-charge model give a value for the difference of the perpendicular and parallel paramagnetic Curie temperatures that is about half the correct value, although the cone angle is only slightly smaller than the measured value. One can obtain values of the anisotropy constants by fitting the Curie temperatures and cone angle which give values of the lower $\mathbf{k}_0$ frequency varying by 2 or 3 cm^{-1} from the value for the point-charge model. However, the values are still much greater than the usual maximum signal frequency of 1 cm^{-1}. For $H=0$, $\omega(0)$ is zero. (The planar anisotropy is negligible for erbium even at low T.) $\omega(0)$ for $H>H_{\perp f}$ is given by (81), and is about 8 cm^{-1} at $H_{\perp f}$. On the other hand, $\omega(\pm\mathbf{k}_0)$ go to zero at $H_{\perp f}$.

Thus with the exception of the splitting between the $+\mathbf{k}_0$ and $-\mathbf{k}_0$ frequencies, one expects much the same sort of behavior for the resonance frequencies in erbium as discussed for the plane spiral in Sec. 5. There is probably a resonance at $H_{\perp c}$ which is most strongly excited by an rf field along η (because this is tangent to the cone surface where it cuts the ξ-ζ plane); and also a weak resonance just below $H_{\perp f}$ excited by an rf field at the cone angle in the ξ-ζ plane (parallel the magnetization at $H_{\perp f}$).

C. $MnAu_2$

This substance is known to have a spiral structure below 360°K with moments $\mu=3.5\beta$ on the Mn atoms at $T=0$, pointing within the layers perpendicular to a tetragonal axis.[18] The static magnetic properties of polycrystalline samples have been studied by Meyer

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[16] J. W. Cable, E. O. Wellan, W. C. Koehler, and M. K. Wilkinson, Suppl. J. Appl. Phys. **32**, 49S (1961).

[17] R. W. Green, S. Legvold, and F. H. Spedding, Phys. Rev. **122**, 827 (1961).

[18] A. Herpin, P. Meriel, and J. Villain, Compt. Rend. **249**, 1334 (1959).

and Taglang.[19] From these data it is possible to derive some of the constants required in the theory to predict the resonance conditions. Between the Néel point and room temperature H_c and H_f are given in this reference, and have a ratio close to two as predicted by Nagamiya *et al.*[7] In fact, the observed $H_f'=H_f-16H_q$ is less than twice the observed $H_c'=H_c-H_q$. The quadratic anisotropy field H_q here occurs with different numerical factors obtained by analogy with the discussion in Sec. 6B. This indicates that $H_q<300$ G in this temperature region. Assuming that this is negligible, the axial anisotropy may be obtained from the observed approach to saturation in large applied fields. Using the expression given in Ref. 19

$$\mu(H)=\mu(1-b/H^2); \quad b=H_A{}^2/15, \tag{94}$$

with H_A defined as in Eq. (76a), Meyer has calculated $H_A=4.1\times10^4$ G at 170°K, 3.5×10^4 at 290°K and 3.2×10^4 at 320°K. This is roughly proportional to μ although the low T value is smaller than expected.

The value of A' [defined in Eq. (76b)] can be determined from the measured H_c, but unfortunately there is no evidence on the other Fourier transforms of the exchange. Full numerical estimates can only be made if some assumption is made, and in the absence of other information we use a three layer model similar to that in the rare earths. Here $qc=2\pi/7$ (approx.) so that the relation $c\gg b\gg a$ can again be used. This predicts $\hbar\omega(k_0)\sim4$ cm^{-1} at 320°K [from (6)] and somewhat larger at lower T. For $H<H_c$ this is little changed and $\hbar\omega(0)$ is very small ~0.05 cm^{-1} at $H=H_c$ [using (21)]. At H just greater than H_c, $\hbar\omega(\cos k_0)$ from (50) is about 2 cm^{-1} and $\hbar\omega(\sin k_0)$ a little larger. These frequencies tend to zero at H_f. At $H>H_c$

$$\hbar\omega(0)\simeq2\beta[H(H+H_A)]^{1/2}, \tag{95}$$

since the last term in (45) is negligible. This is also about 2 cm^{-1} at $H=H_c$.

Thus, for $T<320$°K the discontinuous change in $\hbar\omega(0)$ at $H=H_c$ covers the microwave range and predicts resonance at that field. A second weak resonance from the k_0 modes should occur near H_f.

At higher temperatures H_c and H_A will decrease rapidly. The effect of this may be roughly seen by expanding the scale of Fig. 1. Then the ordinate appropriate to the resonant frequency increases. Since in this substance $\omega(0)$ and $\omega(\sin k_0)$ are similar at H just greater than H_c, it appears that as T increases the $\omega(0)$ resonance will move to higher fields. Eventually as $H_A\rightarrow0$ at T_N this becomes the normal paramagnetic resonance at $\hbar\omega=2\beta H$. The k_0 resonances will move to lower fields and replace the $\omega(0)$ resonance at $H=H_c$. At still higher T this resonance will abruptly disappear to lower fields.

[19] A. J. P. Meyer and P. Taglang, J. Phys. Radium **17**, 260 (1956).

Resonance experiments have been carried out on polycrystalline samples of this substance by Meyer and Asch.[8,9] The resonance conditions of a polycrystal in Sec. 6D show that the peak occurs at the same field as it would for an applied field in the plane. Most of the results are then in agreement with the above discussion. At $T=290$ and 320°K the resonance is at H_c. By 350°K just below T_N the resonance is broader and might consist of the k_0 resonance at H_c and the $k=0$ resonance at higher fields. At $T>T_N$ normal paramagnetic resonance is observed. A serious discrepancy between theory and experiment exists, however, in the case of the single low-temperature experiment. At $T=170$°K the magnetization curves indicate $H_c\simeq10^4$ G but a strong resonance occurs at $H=6000$ G. Further low-temperature measurements are required to establish the origin of this effect.

ACKNOWLEDGMENTS

We are indebted to Dr. A. J. P. Meyer for several discussions and correspondence concerning $MnAu_2$ and to F. A. Wedgewood for assistance with some calculations.

APPENDIX

In this Appendix we summarize a study of the behavior of an antiferromagnetic model in which both a first-order and a second-order transition are present.

The Hamiltonian for the system is

$$\mathcal{H}=-\sum_{i\neq j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j+\sum_i[-(\mu H/S)S_{i\xi}-K_2S_{i\zeta}{}^2-PS_{i\xi}{}^2]. \tag{A1}$$

Here J_{ij} is taken only between spins in adjacent layers along the ζ direction. It is assumed that there is a very large exchange within each layer holding all spins in a layer parallel. Then J_{ij} is such that for $\mathbf{S}_n$ and $\mathbf{S}_{n+1}$ any spins in the nth and $(n+1)$th layers, respectively, $-2J\mathbf{S}_n\cdot\mathbf{S}_{n+1}$ gives the exchange interaction of $\mathbf{S}_n$ with the total spin of the $(n+1)$th layer, and J is negative. The anisotropy terms have K_2 negative and P positive with $P\ll|K_2|$. Also K_2 is taken much larger than any other interaction in (1) so that K_2 serves to hold the spins in equilibrium in the planes perpendicular to the ζ axis (ξ-η planes). The term in P then gives an easy axis along the ξ direction within the planes. So if H is zero, the spins are aligned parallel to the ξ direction with alternate layers antiparallel.

The behavior of the equilibrium spin configuration as H increases is as follows. For H less than a certain value $H_c{}^*$, the spin configuration is unchanged. The spins are antiparallel in alternate layers. At $H=H_c{}^*$ the spin arrangement "flops" to one where alternate layers are canted at angles $+\theta$ and $-\theta$ to H, respectively, $(0<\theta<\frac{1}{2}\pi)$. Here,

$$H_c^*=2[P(4|J|-P)]^{1/2}, \tag{A2}$$

$$\cos\theta=H/2(4|J|-P), \tag{A3}$$

(putting $\mu=1$, $S^2=1$ for convenience).

As H increases, $\cos\theta$ increases continuously (θ decreases) until $H=H_f^*$, where the spins are ferromagnetically aligned. For $H>H_f^*$, the spins remain ferromagnetically aligned.

$$H_f^*=2(4|J|-P). \tag{A4}$$

The spin-wave behavior in each of the three regions can be summarized.

Region (1). $0\leq H<H_c^*$ Antiferromagnet

For $\mathbf{q}$ along $\boldsymbol{\zeta}$ the spin-wave frequencies are given by (with lattice spacing $c'=1$ and $\hbar=1$)

$$\omega^2(q)=(-K_2)[4(2|J|+P) \pm 2(16J^2\cos^2 q+H^2)^{1/2}]. \tag{A5}$$

$\omega(q)$ and $\omega(\pi-q)$ are degenerate. The maximum of the spectrum occurs for $q=0$ or π and the $+$ sign before the square root. The minimum occurs for $q=0$ or π and $-$ sign before the square root. So that at $H=H_c^*$, the minimum of the spectrum is

$$\omega^2{}_{\min H_c^*}=4(-K_2)[2|J|+P - (4J^2+4P|J|-P^2)^{1/2}], \tag{A6}$$

but since

$$(4J^2+4P|J|-P^2)^{1/2}<(4J^2+4P|J|+P^2)^{1/2} =2|J|+P, \tag{A7}$$

this is positive. So that at the limit of $H=H_c^*$ coming from the antiferromagnetic region, all spin-wave energies are positive.

Region (2). $H_c^*<H<H_f^*$ Canted Spin Arrangement

The spin-wave frequencies are given by

$$\omega^2(q)=(-2K_2)\left\{\left[\frac{H^2}{(4|J|-P)^2}-2\right] \times[2|J|(\cos q-1)+P]+\frac{H^2}{2(4|J|-P)}\right\}. \tag{A8}$$

For $H>\sqrt{2}(4|J|-P)$, the minimum of $\omega^2(q)$ is at $q=\pi$; while for $H<\sqrt{2}(4|J|-P)$, the minimum of $\omega^2(q)$ is at $q=0$. For small P, this means that at H_c^* the minimum of the spectrum is for $q=0$, and at H_f^* the minimum is at $q=\pi$. Thus, at H_c^*

$$\omega^2{}_{\min}(q)=(-2K_2)\left(\frac{4P^2}{4|J|-P}\right). \tag{A9}$$

This is positive, so that at the limit of $H=H_c^*$ coming from the canted region all spin-wave energies are positive. At $H=H_c^*$, the spin-wave spectrum is discontinuous. It is easy to show that for small P, at $H=H_c^*$, the bottom of the spin-wave spectrum is lower and the top higher in the antiferromagnetic region as compared to the canted region.

For $H=H_f^*$, the minimum of (A8) occurs for $q=\pi$ and is equal to zero. Thus the transition at H_f^* corresponds to the spin wave frequency $\omega(\pi)$ going to zero.

Region (3). $H>H_f^*$ Ferromagnet

The spin-wave frequencies are

$$\omega^2(q)=-4K_2[2|J|(\cos q-1)+\tfrac{1}{2}H+P]. \tag{A10}$$

The minimum of this is for $q=\pi$. At $H=H_f^*$, $\omega(\pi)$ is zero. Thus, the transition at $H=H_f^*$ corresponds to $\omega(\pi)$ going to zero in both the canted and ferromagnetic region. Since the equilibrium state is the same at the $H=H_f^*$ limit for both the canted and the ferromagnetic arrangement, the spin-wave spectrum is continuous at H_f^*.