Theory of Magnetic Resonance in the Heavy Rare-Earth Metals

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The spin-wave spectrum has been calculated from a Hamiltonian including anisotropic exchange, axial and hexagonal anisotropy, and magnetic fields for the various types of magnetic order which are known to occur in the heavy rare-earth metals Tb-Tm. Particular attention is paid to those spin waves which can be excited by radiation. Because of the high anisotropy, most resonances will occur in the infrared where they will be difficult to observe. However, those phases which show either ferromagnetic ordering with moments in the hexagonal planes, or spiral configurations of the moments in those planes, should display a microwave resonance when a suitably chosen magnetic field is applied in that plane. In the spiral phases an additional resonance with $\omega \propto H^2$ may also be observable.

1. INTRODUCTION

NEUTRON diffraction experiments have now made clear the nature of the magnetic order in several of the heavy rare-earth metals Gd-Tm. All these elements have the same crystal structure (hcp) but the details of the magnetic ordering vary widely from element to element and there are usually several magnetic phases in each case. Much of the experimental evidence is summarized by Elliott.[1] He was able to show that many of the variations were the natural consequence of a single Hamiltonian. The essential feature of the theory is a large axial and smaller hexagonal anisotropy arising from the crystalline electric field. The magnetic effects of this same field vary with the electronic configuration in a way which accounts for many of the observed orderings.

It is also of considerable interest to study the excited energy levels of the magnetic system. In most phases these can be described as spin waves by Fourier analyzing the deviations from the ordered state even when that state is not the ground state of the system, as is the case in some phases which do not persist down to 0°K. Some high-T phases, however, like the longitudinal spin wave found in Er, are stable because they have low free energy through a high entropy.[1–3] Such phases will be shown not to have spin-wave states.

Yosida and Miwa[2] and Kaplan[3] have already evaluated spin-wave energies in connection with their investigations of the ground state. They omitted, however, the effects of magnetic fields and the hexagonal anisotropy and used a simplified form of the axial anisotropy. By means of magnetic resonance it is possible to study the energy of certain particular spin waves in detail, and in a comprehensive theory of their behavior it is necessary to include these terms. It is also likely that the general spin-wave spectrum will eventually be determined by inelastic neutron scattering.

The stable magnetic orderings found in the metals are in general characterized by a wavelike variation in some component of $\mathbf{S}_i$ of a certain wave vector $\mathbf{k}_0$. In such cases magnetic resonance will occur with spin waves of wave vector $\mathbf{k}=\pm\mathbf{k}_0$ and possibly $\mathbf{k}=0$. In some metals the wave variations in the order are seriously distorted, especially by the hexagonal anisotropy, so that they contain a number of harmonics $n\mathbf{k}_0$. This harmonic content can be further enriched by the application of appropriate dc magnetic fields. These distortions might allow one to observe spin waves of several $n\mathbf{k}_0$ by resonance, so that by these means it would be possible to deduce the general features of the spin-wave spectrum.

The high anisotropy encountered in these metals often causes the resonances to occur in the infrared where they will be difficult to observe in metallic samples. The resonance never occurs at frequencies which are simply proportional to the applied field $\mathbf{H}$, as in normal ferromagnets.

2.1. GENERAL HAMILTONIAN

The most general Hamiltonian employed will be

$$\mathcal{H}=-\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-\sum_{i\neq j} K_{ij}S_{i\zeta}S_{j\zeta} +\sum_i\{[P_2{}^0Y_2{}^0(\mathbf{S}_i)+P_4{}^0Y_4{}^0(\mathbf{S}_i)+P_6{}^0Y_6{}^0(\mathbf{S}_i) +P_6{}^6(Y_6{}^6(\mathbf{S}_i)+Y_6{}^{-6}(\mathbf{S}_i))]+\lambda\beta\mathbf{H}\cdot\mathbf{S}_i\}, \quad (1)$$

i.e., including anisotropic exchange, a crystal field of hexagonal symmetry and a magnetic field of arbitrary direction. $\mathbf{S}_i$ is the total angular momentum of atom i (including orbital and spin), λ is the Landé factor and ζ is the direction of the hexagonal axis. The $Y_l{}^m(S)$ are the operator equivalents of the appropriate spherical harmonics[4] referred to this axis. Some discussion of the

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[1] R. J. Elliott, Phys. Rev. **124**, 346 (1961).
[2] K. Yosida and H. Miwa, J. Appl. Phys. **32S**, 8 (1961).
[3] T. A. Kaplan, Phys. Rev. **124**, 329 (1961).
[4] R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

magnitudes of the parameters J, P, etc., is given in reference 1. It was conjectured there that there should also be included in (1) interatomic interaction terms which contain more than two spin operators, arising, for example, from quadrupole-quadrupole interactions. These were thought to be responsible for continuous changes in the magnetic order (e.g., the pitch of the spiral configurations) with temperature. In view of the uncertain nature of these interactions and the algebraic complexity of the theory, they are omitted here. However, their essential effect may be included by allowing J_{ij}, K_{ij} to depend on T.

The rare-earth metals of interest have hcp structure. For the Hamiltonian (1), the fact that the lattice is not Bravais has no effect on that acoustic branch of the spin-wave spectrum which is observed in ferromagnetic resonance. The only effect would be the addition of an optical branch, probably at much higher frequencies. For this reason, we treat the lattice as though it were Bravais.

For the algebraic manipulations of this paper it is convenient to regroup the terms in (1) into sums over the various powers of $S_{i\zeta}$ present and to include $S_{i\zeta}^2$ with the anisotropic exchange by extending the sum to include $i=j$, viz.,

$$\mathcal{H}=-\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j -\sum_{i,j} K_{ij}S_{i\zeta}S_{j\zeta}+\sum_i\{K_4S_{i\zeta}^4+K_6S_{i\zeta}^6 +P_6{}^6[Y_6{}^6(\mathbf{S}_i)+Y_6{}^{-6}(\mathbf{S}_i)]+\lambda\beta\mathbf{H}\cdot\mathbf{S}_i\}. \quad (2)$$

Since this is to be Fourier analyzed, we define

$$J(q)=\textstyle\sum_j J_{ij}\cos\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j),$$
$$K(q)=\textstyle\sum_j K_{ij}\cos\mathbf{q}\cdot(\mathbf{R}_i-\mathbf{R}_j). \quad (3)$$

2.2. MAGNETIC ORDER

Experimentally it is found that in all the magnetic structures the moments in each plane perpendicular to the hexagonal axis (ζ axis) are aligned. However, this alignment varies from plane to plane along the ζ axis. There is usually a wave-like variation of the moment along this direction of some wave vector $\mathbf{k}_0$. This indicates that $J(q)$ or $J(q)+K(q)$ must be a maximum at $\mathbf{q}=\mathbf{k}_0$ depending on whether the wave-like variation is in the planar or the axial component of $\mathbf{S}$. The moment direction relative to the ζ axis is varied by the anisotropy terms in the Hamiltonian.

There are essentially three cases.

$$\text{(case } A\text{)}\quad S_{i\zeta}=S\cos\theta,\quad S_{i\xi}=S\sin\theta\cos(\mathbf{k}_0\cdot\mathbf{R}_i), \quad S_{i\eta}=S\sin\theta\sin(\mathbf{k}_0\cdot\mathbf{R}_1). \quad (4)$$

where ξ and η are mutually perpendicular directions in the hexagonal plane.

I. The moment directions lie on the surface of a cone of angle θ. This is found[5] in Er at low T.

II. By applying a field along the ξ direction greater than a certain critical value ferromagnetic alignment can be obtained in[6] Er at low T. (Putting $k_0=0$ in Eq. (4). The moments are seen to make an angle θ with the ζ axis.)

III. In the special case $\theta=0$ the system is simply ferromagnetic along ζ. This probably occurs in[7] Tm at low T.

IV. In the special case $\theta=\pi/2$ the moments lie in the plane and form a spiral. This is found in Dy,[8] Ho,[7] and probably[7] Tb at high T.

As mentioned in the introduction, the effect of the hexagonal anisotropy or a magnetic field may destroy the simple wave variations of the planar components. The pattern will still repeat after $R=2\pi/k_0$, but higher harmonics will appear in the description of $S_{i\xi}$, $S_{i\eta}$. Dy[8] shows such distortions at intermediate T and Ho,[7] large distortions at low T. These distorted spirals will be treated by perturbation theory, but only for the special case $\theta=\pi/2$.

V. In some cases the hexagonal anisotropy causes ferromagnetic alignment of the planar components, e.g.,[7] Tb and[8] Dy at low T (i.e., $\theta=\pi/2$, $k_0=0$).

$$\text{(case } B\text{)}\quad S_{1\zeta}=S\cos(\mathbf{k}_0\cdot\mathbf{R}_i),\quad S_{i\xi}=S_{i\eta}=0. \quad (5)$$

This corresponds to the high-T phase of Er.[5] It will be shown that spin waves do not describe the excited states.

(case C) All three components have a wave-like variation, corresponding to the intermediate phase of Er.[5] It will be shown that spin waves do not describe the excited states.

Thus the main results of this paper will be an evaluation of the spin-wave spectra of the five-order types of case A, and in particular of the details of magnetic resonance absorption in these cases. It is first convenient to find the various relations between the constants in (2) which hold for the various orderings.

2.3. STABILITY OF CONFIGURATIONS

In order for a configuration described by one form of A to be stable, the energy obtained by substituting (4) into (2) must be a minimum for variation against k_0 and θ.

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

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

[7] W. C. Koehler, E. O. Wollan, M. K. Wilkinson, and J. W. Cable in Rare Earth Research Developments Conference, Lake Arrowhead, California, October, 1960 (to be published), Paper III-4; W. C. Koehler, J. W. Cable, E. O. Wollan, and M. K. Wilkinson, paper presented at International Conference on Magnetism and Crystallography, Kyoto, Japan, September 25–30, 1961 (unpublished).

[8] M. K. Wilkinson, W. C. Koehler, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **32S**, 48 (1961).

$$E/N=-J(k_0)S^2\sin^2\theta-[J(0)+K(0)]S^2\cos^2\theta$$
$$+K_4S^4\cos^4\theta+K_6S^6\cos^6\theta+\lambda\beta HS\cos\theta, \quad (6)$$

since the effects of the hexagonal anisotropy and H perpendicular to ζ average out in the spiral structure. (This is not true for cases AII and AV. The conditions on the constants in these cases are described in Secs. 3.2 and 3.5, respectively.)

The pitch of the spiral is determined by $J(k_0)$ being a maximum; evaluations of J_{ij} which give the observed results are given in terms of a crude model in reference 1. The cone angle θ is given by $\partial E/\partial\theta=0$, i.e.,

$$6K_6S^6\cos^5\theta+4K_4S^4\cos^3\theta$$
$$+2[J(k_0)-J(0)-K(0)]S^2\cos\theta+\lambda\beta HS=0, \quad (7)$$

or

$$\sin\theta=0, \qquad \text{type III.} \quad (8)$$

In the absence of H, Eq. (7) gives two types of solution,

$$6K_6S^6\cos^4\theta+4K_4S^4\cos^2\theta$$
$$+2[J(k_0)-J(0)-K(0)]S^2=0, \qquad \text{type I,} \quad (9)$$

and

$$\cos\theta=0, \qquad \text{type IV.} \quad (10)$$

A small field H, in general, changes the cone angle from that given by (9) to a solution of (7). It also pulls the planar case given by (10) into a shallow cone of angle θ given by a solution of (7) of the form $\theta=\pi/2-CH$. The solution (8) is always of type III.

There are several domains of stability for the three cases described here. These domains are determined by the conditions that the second derivative of the energy must be positive for the configuration to have a relative minimum, that the energy must be an absolute minimum if there are two relative minima in a given domain, and the $\cos^2\theta$ must be real, and have value between 0 and 1. We consider the several possibilities:

(a). $$3K_6S^4+2K_4S^2+J(k_0)-J(0)-K(0)<0, \quad (11a)$$

and

$$J(k_0)-J(0)-K(0)>0. \quad (11b)$$

The stable configuration is type III if

$$K_6S^4+K_4S^2+J(k_0)-J(0)-K(0)<0, \quad (11c)$$

and type IV if

$$K_6S^4+K_4S^2+J(k_0)-J(0)-K(0)>0. \quad (11d)$$

(b). $$3K_6S^4+2K_4S^2+J(k_0)-J(0)-K(0)>0, \quad (12a)$$

and

$$J(k_0)-J(0)-K(0)<0, \quad (12b)$$

The stable configuration is type I if the further condition for the reality of $\cos^2\theta$ is satisfied.

$$K_4^2-3K_6[J(k_0)-J(0)-K(0)]>0. \quad (12c)$$

(c). (12a) and (11b) hold. If, in addition, (12c) and the following conditions hold:

$$K_6>0, \quad (13a)$$

$$K_4<0, \quad (13b)$$

$$2S^2\{-K_4^2+3K_6[J(k_0)-J(0)-K(0)]\}\cos^2\theta$$
$$-K_4[J(k_0)-J(0)-K(0)]<0, \quad (13c)$$

where

$$\cos^2\theta=-(K_4/3K_6S^2)-(1/3K_6S^2)$$
$$\times\{K_4^2-3K_6[J(k_0)-J(0)-K(0)]\}^{\frac{1}{2}}, \quad (13d)$$

then the configuration is of type I. If (12a) and (11b) hold, but not the other conditions, then the configuration is type IV.

(d). (11a) and (12b) hold. If, in addition, (12c) and the following conditions hold

$$K_6<0, \quad (14a)$$

$$K_4>0, \quad (14b)$$

$$2S^2\{-K_4^2+3K_6[J(k_0)-J(0)-K(0)]\}\cos^2\theta$$
$$-K_4[J(k_0)-J(0)-K(0)]>9K_6\{K_6S^6+K_4S^4$$
$$+[J(k_0)-J(0)-K(0)]S^2\}, \quad (14c)$$

where

$$\cos^2\theta=-(K_4/3K_6S^2)-(1/3K_6S^2)$$
$$\times\{K_4^2-3K_6[J(k_0)-J(0)-K(0)]\}^{\frac{1}{2}}, \quad (14d)$$

then the configuration is of type I. If (11a) and (12b) hold but not the other conditions, then the configuration is of type III.

3.1. THE CONE—CASE AI

This is the case for Er at low T. The hexagonal anisotropy is negligible. We consider the applied field to be along the ζ direction, the axis of the cone, so that the symmetry about the cone axis is not distorted.

Following a procedure similar to those of references 2 and 3, we define new coordinates such that the equilibrium direction of the spin at each site determines the z direction at that site.

$$\hat{x}_i=[\boldsymbol{\xi}\cos(\mathbf{k}_0\cdot\mathbf{R}_i)+\boldsymbol{\eta}\sin(\mathbf{k}_0\cdot\mathbf{R}_i)]\cos\theta-\boldsymbol{\zeta}\sin\theta, \quad (15a)$$

$$\hat{y}_i=-\boldsymbol{\xi}\sin(\mathbf{k}_0\cdot\mathbf{R}_i)+\boldsymbol{\eta}\cos(\mathbf{k}_0\cdot\mathbf{R}_i), \quad (15b)$$

$$\hat{z}_i=[\boldsymbol{\xi}\cos(\mathbf{k}_0\cdot\mathbf{R}_i)+\boldsymbol{\eta}\sin(\mathbf{k}_0\cdot\mathbf{R}_i)]\sin\theta+\boldsymbol{\zeta}\cos\theta. \quad (15c)$$

$\hat{x}_i$, $\hat{y}_i$, $\hat{z}_i$, $\boldsymbol{\xi}$, $\boldsymbol{\eta}$, and $\boldsymbol{\zeta}$ are unit vectors in the appropriate directions.

$\mathcal{H}$ can then be written in terms of the x, y, z coordinates. Keeping terms only to the second order in S_{ix}

and S_{iy},

$$-\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j=-\sum_{i\neq j} J_{ij}\{\tfrac{1}{4}\sin^2\theta[S_i^+S_j^+ + S_i^-S_j^-][1-\cos(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))]$$
$$+\tfrac{1}{2}S_i^-S_j^+[(1+\cos^2\theta)\cos(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))+\sin^2\theta-2i\cos\theta\sin(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))]$$
$$+S_{iz}S_{jz}[\sin^2\theta\cos(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))+\cos^2\theta]$$
$$+S_i^+S_{jz}[\sin\theta\cos\theta[\cos(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))-1]+\mathrm{i}\sin\theta\sin(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))]$$
$$+S_i^-S_{jz}[\sin\theta\cos\theta[\cos(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))-1]-i\sin\theta\sin(\mathbf{k}_0\cdot(\mathbf{R}_i-\mathbf{R}_j))]\}; \quad (16)$$

$$-\sum_{i,j} K_{ij}S_{i\zeta}S_{j\zeta}=-\sum_{i,j} K_{ij}\{\tfrac{1}{4}\sin^2\theta[S_i^+S_j^+ + S_i^-S_j^-]+\tfrac{1}{2}S_i^-S_j^+\sin^2\theta+S_{iz}S_{jz}\cos^2\theta-[S_i^-S_{jz}+S_i^+S_{jz}]\sin\theta\cos\theta\}; \quad (17)$$

$$K_4\sum_i S_{i\zeta}^4=K_4\sum_i\{S_{iz}^4\cos^4\theta-2S_{iz}^3(S_i^+ + S_i^-)\cos^3\theta\sin\theta+\tfrac{3}{2}S_{iz}^2[(S_i^+)^2+(S_i^-)^2+2S_i^-S_i^+]\cos^2\theta\sin^2\theta\}; \quad (18)$$

$$K_6\sum_i S_{i\zeta}^6=K_6\sum_i\{S_{iz}^6\cos^6\theta-3S_{iz}^5(S_i^+ + S_i^-)\cos^5\theta\sin\theta$$
$$+(15/4)S_{iz}^4[(S_i^+)^2+(S_i^-)^2+2S_i^-S_i^+]\cos^4\theta\sin^2\theta\}; \quad (19)$$

$$\lambda\beta H\sum_i S_{i\zeta}=\lambda\beta H\sum_i\{-\sin\theta[(S_i^+ + S_i^-)/2]+S_{iz}\cos\theta\}; \quad (20)$$

where

$$S_i^{\pm}=S_{ix}\pm iS_{iy}. \quad (21)$$

In Eqs. (17), (18), and (19) we have neglected terms arising from commutators of S_i^+, S_i^-, and S_{iz}. These terms will be of lower order in S and are neglected in keeping with the spirit of the spin-wave harmonic-oscillator approximation[9] which we now adopt. S (which is the total angular momentum) is very large in the cases of interest, so this approximation should be quite good. These terms contribute to the zero-point energy of the ground state in the spin-wave approximation, and give a contribution to the linewidth in magnetic resonance.

For the spin-wave treatment we make the usual Holstein-Primakoff approximation.[9]

$$S_i^+\approx(2S)^{\frac{1}{2}}a_i,\quad S_i^-\approx(2S)^{\frac{1}{2}}a_i^*,\quad S_{iz}=S-a_i^*a_i, \quad (22)$$

where

$$[a_i,a_j^*]=\delta_{ij}. \quad (23)$$

The Fourier transforms of a_i and a_i^* are

$$a_q=N^{-\frac{1}{2}}\textstyle\sum_i a_ie^{i\mathbf{q}\cdot\mathbf{R}_i},\quad a_q^*=N^{-\frac{1}{2}}\sum_i a_i^*e^{-i\mathbf{q}\cdot\mathbf{R}_i}, \quad (24)$$

The a_q^* and a_q are the spin-wave creation and destruction operators.

$$[a_q,a_{q'}^*]=\delta_{qq'}. \quad (25)$$

The Hamiltonian (2) becomes

$$\mathcal{H}=E_0+\textstyle\sum_q[2SA_qa_q^*a_q+SB_q(a_q^*a_{-q}^*+a_qa_{-q})], \quad (26)$$

where E_0 is given by (6), and

$$A_q=(1/2S)\{-\tfrac{1}{2}S[(1-\cos\theta)^2J(k_0+q)$$
$$+(1+\cos\theta)^2J(k_0-q)+2\sin^2\theta(J(q)+K(q))$$
$$-4\cos^2\theta(J(0)+K(0))-4\sin^2\theta J(k_0)]$$
$$+K_4S^3[-4\cos^4\theta+6\sin^2\theta\cos^2\theta]$$
$$+K_6S^5[-6\cos^6\theta+15\cos^4\theta\sin^2\theta]$$
$$-\lambda\beta H\cos\theta\}; \quad (27)$$

$$B_q=-\tfrac{1}{4}\sin^2\theta[2J(q)-J(k_0-q)$$
$$-J(k_0+q)+2K(q)-2L],$$

where

$$L=6K_4S^2\cos^2\theta+15K_6S^4\cos^4\theta. \quad (28)$$

If (7) is used, (27) reduces to

$$A_q=-\tfrac{1}{4}\{(1-\cos\theta)^2J(k_0+q)$$
$$+(1+\cos\theta)^2J(k_0-q)-4J(k_0)$$
$$+2\sin^2\theta[J(q)+K(q)-L]\}. \quad (29)$$

This reduction is strictly true only at $T=0$, since we found the expression defining θ by minimizing the energy rather than the free energy.

To find the spin-wave spectrum, it is necessary to diagonalize (26). Let α_q^* and α_q denote the transformed creation and destruction operators which diagonalize $\mathcal{H}$.

$$[\alpha_q,\alpha_{q'}^*]=\delta_{qq'}. \quad (30)$$

Let

$$\alpha_q=w_qa_q+b_qa_{-q}^*, \quad (31)$$

then

$$[\alpha_q,\mathcal{H}]=E_q\alpha_q. \quad (32)$$

This leads to the secular determinant

$$\begin{vmatrix} 2SA_q-E_q & -2SB_q \\ 2SB_q & -2SA_{-q}-E_q\end{vmatrix}=0. \quad (33)$$

Use has been made of the relation $B_q=B_{-q}$. Note that $A_q\neq A_{-q}$. (The asymmetry of A_q arises from the $S_{ix}S_{jy}$ term in the exchange energy.)

The secular determinant is solved for the spin-wave energies.

$$E_q=S(A_q-A_{-q})\pm S[(A_q+A_{-q})^2-4B_q^2]^{\frac{1}{2}}. \quad (34)$$

The spin-wave energy, E_q, must be positive for stability. If both a positive and negative energy are present for a given q (say, q_1), we attach physical meaning only to the positive value. There must be at least one positive value

[9] T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

of energy at every q for stability. Consider the case for q_1 when

$$(A_{q_1}-A_{-q_1})>[(A_{q_1}+A_{-q_1})^2-4B_{q_1}^2]^{\frac{1}{2}}.$$

There would be two positive values for E_{q_1}, but there would be two negative values for E_{-q_1}. Hence for stable spin waves

$$|A_q-A_{-q}|<[(A_q+A_{-q})^2-4B_q^2]^{\frac{1}{2}}.$$

This means that the spin-wave frequencies are

$$\hbar\omega_q=S(A_q-A_{-q})+S[(A_q+A_{-q})^2-4B_q^2]^{\frac{1}{2}}. \quad (35)$$

The values of b_q and w_q are needed to find the relative intensities of the frequencies excited in a resonance experiment. Equation (30) leads to the condition

$$w_q^2-b_q^2=1. \quad (36)$$

The secular determinant and the value of E_q give

$$b_q=\frac{A_q+A_{-q}-[(A_q+A_{-q})^2-4B_q^2]^{\frac{1}{2}}}{\{-[A_q+A_{-q}-((A_q+A_{-q})^2-4B_q^2)^{\frac{1}{2}}]^2+4B_q^2\}^{\frac{1}{2}}}; \quad (37)$$

$$w_q=\frac{2B_q}{\{-[A_q+A_{-q}-((A_q+A_{-q})^2-4B_q^2)^{\frac{1}{2}}]^2+4B_q^2\}^{\frac{1}{2}}}; \quad (38)$$

so that

$$b_q=b_{-q}, \quad w_q=w_{-q}. \quad (39)$$

The question of interest is what frequencies are excited in a resonance experiment and with what relative intensities.

Apply an rf field along the ξ direction.

$$\mathcal{H}_{\xi\mathrm{rf}}=\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i S_{i\xi}; \quad (40)$$

$$\mathcal{H}_{\xi\mathrm{rf}}=\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i [S_{ix}\cos(\mathbf{k}_0\cdot\mathbf{R}_i)\cos\theta -S_{iy}\sin(\mathbf{k}_0\cdot\mathbf{R}_i)+S_{iz}\cos(\mathbf{k}_0\cdot\mathbf{R}_i)\sin\theta]. \quad (41)$$

The terms in S_{ix} and S_{iy} give rise to resonance effects. This part of $\mathcal{H}_{\xi\mathrm{rf}}$ is obtained in terms of α^* and α by taking the Fourier transform and using the inverse of (31).

$$\alpha_q=w_qa_q+b_qa_{-q}. \quad (42)$$

Then

$$\begin{aligned}\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i [S_{ix}&\cos(\mathbf{k}_0\cdot\mathbf{R}_i)\cos\theta-S_{iy}\sin(\mathbf{k}_0\cdot\mathbf{R}_i)]\\ &=\lambda\beta H_{\mathrm{rf}}\times\tfrac{1}{4}(2SN)^{\frac{1}{2}}\{[\alpha_{k_0}^*+\alpha_{k_0}]\\ &\quad\times[(-b_{k_0}+w_{k_0})\cos\theta+(w_{k_0}+b_{k_0})]+[\alpha_{-k_0}^*+\alpha_{-k_0}]\\ &\quad\times[(-b_{k_0}+w_{k_0})\cos\theta-(w_{k_0}+b_{k_0})]\}. \quad (43)\end{aligned}$$

The absorption in the ground state arises from the terms in $\alpha_{k_0}^*$ and $\alpha_{-k_0}^*$. The frequencies $\omega(k_0)$ and $\omega(-k_0)$ are excited with relative intensities, $\mathcal{I}$,

$$\mathcal{I}(\pm k_0)\propto\lambda^2SN[(b_{k_0}^2+w_{k_0}^2)(1+\cos^2\theta) \pm 2\cos\theta+2b_{k_0}w_{k_0}\sin^2\theta]. \quad (44)$$

The frequencies are obtained from (35), (28), and (29).

$$\begin{aligned}\hbar\omega(\pm k_0)&=\pm S\cos\theta[J(2k_0)-J(0)]\\ &\quad+S\{[2J(k_0)-J(0)-J(2k_0)]\\ &\quad\times[\cos^2\theta(2J(k_0)-J(0)-J(2k_0))\\ &\quad-2\sin^2\theta[K(k_0)+L]\}^{\frac{1}{2}}. \quad (45)\end{aligned}$$

In (41) the term in S_{iz} gives rise to nonresonant absorption (z pumping).[10]

$$\begin{aligned}\lambda\beta H_{\mathrm{rf}}&\sin\theta\textstyle\sum_i S_{iz}\cos(\mathbf{k}_0\cdot\mathbf{R}_i)\\ &=-\tfrac{1}{2}\lambda\beta H_{\mathrm{rf}}\sin\theta\textstyle\sum_q[(w_q\alpha_q^*-b_q\alpha_{-q})\\ &\quad\times(w_{q+k_0}\alpha_{q+k_0}-b_{q+k_0}\alpha_{-q-k_0}^*)\\ &\quad+(w_q\alpha_q^*-b_q\alpha_{-q})(w_{q-k_0}\alpha_{q-k_0}-b_{q-k_0}\alpha_{-q+k_0}^*)]. \quad (46)\end{aligned}$$

If the rf field is applied along ζ,

$$\mathcal{H}_{\zeta\mathrm{rf}}=\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i S_{i\zeta}, \quad (47)$$

$$\mathcal{H}_{\zeta\mathrm{rf}}=\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i(-S_{ix}\sin\theta+S_{iz}\cos\theta). \quad (48)$$

The resonance arises from the term in S_{ix}.

$$\lambda\beta H_{\mathrm{rf}}\textstyle\sum_i S_{ix}\sin\theta=\tfrac{1}{2}\lambda\beta H_{\mathrm{rf}}\sin\theta(2SN)^{\frac{1}{2}} \times[w_0\alpha_0-b_0\alpha_0^*-b_0\alpha_0+w_0\alpha_0^*]. \quad (49)$$

$\omega(0)$ is excited with relative intensity

$$\mathcal{I}(0)\propto\lambda^2SN\sin^2\theta[-b_0+w_0]^2; \quad (50)$$

$$A_0=B_0=\tfrac{1}{2}\sin^2\theta[J(k_0)-J(0)-K(0)+L], \quad (51)$$

so that

$$\omega(0)=0. \quad (52)$$

When the frequency goes to zero, (50) has no meaning since it is possible to create an infinite number of spin waves of zero energy. If the hexagonal anisotropy were included, $\omega(0)$ would not be zero. This possibility is discussed in Sec. 3.4 for the case $\theta=\pi/2$.

3.2. FERROMAGNETIC ALIGNMENT CAUSED BY APPLIED FIELD GREATER THAN CRITICAL FIELD—CASE *A*II

For the cone configuration of case *A*I, if a magnetic field greater than some critical value is applied in the ξ direction, the spin configuration becomes simply ferromagnetic at an angle θ to the ζ axis.

$$\mathbf{S}_i=S(\boldsymbol{\xi}\sin\theta+\boldsymbol{\zeta}\cos\theta). \quad (53)$$

For erbium[6] this critical field is about 17 kOe at 4°K. This value is obtained from Fig. 1 of reference 6 and is the field necessary to pull the spins into ferromagnetic alignment in the $\xi-\zeta$ plane.

If $H_{\parallel}$ is the applied field along ζ and $H_{\perp}$ that along ξ, θ is given by

$$0=2S^2K(0)\cos\theta\sin\theta-4K_4S^3\cos^3\theta\sin\theta -6K_6S^5\cos^5\theta\sin\theta-\lambda\beta H_{\parallel}\sin\theta+\lambda\beta H_{\perp}\cos\theta.$$

The spin-wave spectrum may be found by the method demonstrated in Sec. 3.1 using Eqs. (16–20) with $k_0=0$

[10] E. Schlomann, J. J. Green, and U. Milano, J. Appl. Phys. **31S**, 386 (1960).

and, in addition,

$$\lambda\beta H_\perp \sum_i S_{i\xi} = \lambda\beta H_\perp \sum_i (S_{ix}\cos\theta + S_{iz}\sin\theta). \quad (54)$$

The spin-wave frequencies are

$$\hbar\omega(q) = \left\{ \left(-2S[J(q)-J(0)] - \frac{\lambda\beta H_\perp}{\sin\theta} \right) \times \left(-2S[J(q)-J(0)] - \frac{\lambda\beta H_\perp}{\sin\theta} - 2S\sin^2\theta[K(q)-L] \right) \right\}^{\frac{1}{2}}. \quad (55)$$

An rf field applied along either the ξ or ζ direction excites $\omega(0)$.

$$\hbar\omega(0) = \left\{ -\frac{\lambda\beta H_\perp}{\sin\theta} \left[-\frac{\lambda\beta H_\perp}{\sin\theta} - 2S\sin^2\theta[K(0)-L] \right] \right\}^{\frac{1}{2}}. \quad (56)$$

The maximum intensity is obtained for the rf field perpendicular to the magnetization in the $\xi-\zeta$ plane.

3.3. AXIAL FERROMAGNETISM—CASE *A*III

This is the case of ferromagnetic alignment with a static field along ζ, where the Fourier transform of the exchange energy may, however, have its maximum at some k_0 different from zero.

An rf field along ξ excites

$$\hbar\omega(k_0) = 2SK(0) - 4K_4S^3 - 6K_6S^5 - \lambda\beta H. \quad (57)$$

$\omega(k_0)$ rather than $\omega(0)$ is excited in this case because of the peculiar definition of coordinates (15), for even with $\theta=0$, x_i and y_i rotate from site to site with reciprocal pitch given by k_0.

3.4. SPIRAL IN THE PLANE—CASE *A*IV

This is the case for dysprosium at high temperatures where the helix in the plane is undistorted. This means that the hexagonal anisotropy has negligible effect in this temperature range. The results can be obtained from case AI by setting $\theta=\pi/2$.

An rf field along ξ excited $\omega(+k_0)$ and $\omega(-k_0)$ which are equal in this case.

$$\hbar\omega(\pm k_0) = 2S\{-K(k_0)[J(k_0) - \tfrac{1}{2}J(2k_0) - \tfrac{1}{2}J(0)]\}^{\frac{1}{2}}. \quad (58)$$

An rf field along ζ would excite $\omega(0)$, however, $\omega(0)=0$, so no resonance would be observed.

For this special case we wish also to study the influence of the hexagonal anisotropy energy and of a constant magnetic field in the plane of the spins on the spectrum and resonance absorptions. From Eq. (2) the Hamiltonian will be

$$\mathcal{H} = \sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{ij} K_{ij}S_{i\zeta}S_{j\zeta} + \lambda\beta H \sum_i S_{i\xi} + (P_6{}^6/2)\sum_i [(S_{i\xi}+iS_{i\eta})^6 + (S_{i\xi}-iS_{i\eta})^6]. \quad (59)$$

K_4 and K_6 do not appear with the present treatment of the problem when $\theta=90°$.

As already noted, the last two terms in (59) give rise to harmonics in the sinusoidal spin order. To study this a slightly more general form of the transformations (15) is needed. The phases $\mathbf{k}_0\cdot\mathbf{R}_i$ are replaced by unknown ϕ_i, which are to be found by an interation procedure. For instance,

$$\hat{Z}_i = \boldsymbol{\xi}\cos\phi_i + \boldsymbol{\eta}\sin\phi_i, \quad (\theta=90°). \quad (60)$$

The Holstein-Primakoff operators defined by (22) and (23) in terms of the new local spin operators are again substituted into (59). The resulting expansion in powers of the operators a_i, a_i^* is cut off at the quadratic terms, giving

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2, \quad (61)$$

where $\mathcal{H}_0$, $\mathcal{H}_1$, $\mathcal{H}_2$ are the constant, linear, and quadratic terms.

The phases ϕ_i are determined by setting $\partial\mathcal{H}_0/\partial\phi_i=0$. One has

$$\partial\mathcal{H}_0/\partial\phi_i = -2\sum_{j\neq i} J_{ij}\sin(\phi_i-\phi_j) + (\lambda\beta H/S)\sin\phi_i + 6P_6{}^6S^4\sin6\phi_i = 0. \quad (62)$$

Equation (62) turns out to be equivalent to setting $\mathcal{H}_1$ in (61) equal to zero. Equation (62) is solved by iteration:

$$\phi_i = k_0\zeta_i + \delta\phi_i^{(1)} + \delta\phi_i^{(2)} + \cdots. \quad (63a)$$

where k_0 is again the wave vector which maximizes $J(q)$.

Substituting $\phi=k_0\zeta_i$ into the last two terms of (62), (i.e., the perturbing terms) and substituting $\phi_i=k_0\zeta_i+\delta\phi_i^{(1)}$ into the first term, one finds that

$$\delta\phi_i^{(1)} = X\sin(k_0\zeta_i) + Y\sin(6k_0\zeta_i), \quad (63b)$$

$$\delta\phi_i^{(2)} = X_2\sin(2k_0\zeta_i) + X_5\sin(5k_0\zeta_i) + X_7\sin(7k_0\zeta_i) + X_{12}\sin(12k_0\zeta_i). \quad (63c)$$

Here

$$X = (\lambda\beta H/S)[2J(k_0) - J(2k_0) - J(0)]^{-1}, \quad (64a)$$

and

$$Y = 12P_6{}^6S^4[2J(k_0) - J(7k_0) - J(5k_0)]^{-1}. \quad (64b)$$

The coefficients X_2, X_5, X_7, X_{12} turn out to be rather involved expressions of second order in H and $P_6{}^6$. The quantities X, Y, which measure to first order the higher harmonic content in the spin-structure are essentially equal to the ratios of the perturbing energies to the spin-interaction energies.

$\mathcal{H}_2$ in (61) is given by

$$\mathcal{H}_2 = -S\sum_{i\neq j} J_{ij}[-2a_i^*a_i+\tfrac{1}{2}(a_i^*a_j+a_ia_j^*-a_i^*a_j^*-a_ia_j)]\cos(\phi_i-\phi_j)$$
$$-\tfrac{1}{2}S\sum_{\substack{ij\\(i\neq j)}}(J_{ij}+K_{ij})(a_i^*a_j+a_j^*a_i+a_i^*a_j^*+a_ia_j)$$
$$-2P_6{}^6S^5\sum_i[6a_i^*a_i-(15/2)(a_i^*-a_i)^2]\cos 6\phi_i-\lambda\beta H\sum_i a_i^*a_i\cos\phi_i. \quad (65)$$

The running-wave transformations (24a) are again substituted into Eq. (65). In addition the functions $\cos(\phi_i-\phi_j)$, $\cos 6\phi_i$, $\cos\phi_i$ appearing in (65) are expanded in Taylor series about $\cos[k_0(\zeta_i-\zeta_j)]$, etc., with the help of (63b). In the resulting expression all terms with vanishing momentum, (i.e., $a_q^*a_q$, a_qa_{-q}, etc.) are retained up to the second order in H or $P_6{}^6$, and all off-diagonal terms, (i.e., $a_q^*a_{q+k_0}$ etc.) up to the first order. The result is

$$\mathcal{H}_2=\textstyle\sum_q \mathcal{H}_{2q}, \quad (66a)$$

$$\begin{aligned}\mathcal{H}_{2q}\doteq{}&2SA_q{}^1a_q^*a_q+SB_q{}^1(a_q^*a_{-q}^*+a_qa_{-q})\\&+HS_q(a_{q+k_0}^*a_q+a_{q-k_0}^*a_q+\text{c.c.})\\&+HT_q(a_q^*a_{-q+k_0}^*+a_q^*a_{-q-k_0}^*+\text{c.c.})\\&+HR_q(-a_q^*a_{-q+k_0}^*+a_q^*a_{-q-k_0}^*\\&+a_{q-k_0}^*a_q-a_{q+k_0}^*a_q+\text{c.c.})\\&+P_6{}^6V_q(a_{q+6k_0}^*a_q+a_{q-6k_0}^*a_q+\text{c.c.})\\&+P_6{}^6W_q(a_q^*a_{-q+6k_0}^*+a_q^*a_{-q-6k_0}^*+\text{c.c.})\\&+P_6{}^6U_q(-a_q^*a_{-q+6k_0}^*+a_q^*a_{-q-6k_0}^*\\&+a_{q-6k_0}^*a_q-a_{q+6k_0}^*a_q+\text{c.c.}).\end{aligned} \quad (66b)$$

Here,

$$A_q{}^1=A_q+H^2A_{qH}+(P_6{}^6)^2A_{qp}, \quad (66c)$$

$$B_q{}^1=B_q+H^2B_{qH}+(P_6{}^6)^2B_{qp}. \quad (66d)$$

A_q and B_q are given by (27) and (28) with $\theta=90°$. A_{qH}, A_{qp}, B_{qH}, B_{qp}, S_q, T_q, R_q, V_q, W_q, and U_q are independent of H and $P_6{}^6$. They are rather complicated functions of $J(q+nk_0)$, $n=0$, ±1, 2, 5, or 7. R_q and U_q are odd functions of q, while all the others are even.

The general expressions (35), (37), and (38) for the frequencies and normal modes of the spin waves apply here with $\theta=90°$.

$$\hbar\omega(q)=2S\{[K(q)+J(q)-J(k_0)]\times[\tfrac{1}{2}J(k_0+q)+\tfrac{1}{2}J(k_0-q)-J(k_0)]\}^{\frac{1}{2}}, \quad (67)$$

and $\omega(0)=0$ as in (52).

Accordingly, when corrected for anisotropy and applied magnetic field $\omega(0)$ ought to lie in the microwave region, and is, thus, of special interest. An attempt to calculate the correction to $\omega(0)$ from the Hamiltonian (66) by means of conventional second-order perturbation theory led to an infinite result. This is related to the fact that the coefficients b_q and w_q given in (37) and (38) tend to infinity as $\omega(q)$ goes to zero. A semiclassical treatment which uses the equations of motion is given in the Appendix. This is similar to the method often used in antiferromagnetism, and it is shown there that the antiferromagnetic case is a rather special limit which gives qualitatively different results for some frequencies.

In this section a modified perturbation procedure due to Pryce[11] is used which is suitable for weakly interacting systems in which only the system of interest has very closely spaced energy levels. In view of (67) it will be possible to use this scheme here to attempt to find ω_0. We define energy operators

$$\mathcal{H}_{q0}=2A_q{}^1a_q^*a_q+B_q{}^1(a_q^*a_{-q}^*+a_qa_{-q}), \quad (68a)$$

$$\mathcal{H}^1=\mathcal{H}_2-\textstyle\sum_q\mathcal{H}_{q0}, \quad (68b)$$

where $\mathcal{H}_2$ is the Hamiltonian defined in Eqs. (66). Pryce's scheme then prescribes an effective Hamiltonian, $\tilde{\mathcal{H}}_0$, for the $q=0$ spin wave as follows:

$$\tilde{\mathcal{H}}_0=\mathcal{H}_{00}+\langle g|\mathcal{H}^1|g\rangle-\textstyle\sum_e(\langle g|\mathcal{H}^1|e\rangle\langle e|\mathcal{H}^1|g\rangle)(E_e)^{-1}. \quad (69)$$

Here g and e denote the ground and excited states of all spin waves except the wave $q=0$. No integration over the coordinates associated with this wave are taken when finding the matrix elements appearing in (69). $\mathcal{H}_{00}$ and $\mathcal{H}^1$ are defined in (68a) and (68b).

When Eq. (69) is applied to the Hamiltonian (66), one finds that

$$\tilde{\mathcal{H}}_0=\mathcal{H}_{00}+2S[H^2\tilde{A}_{0H}+(P_6{}^6)^2\tilde{A}_{0p}]a_0^*a_0+S[H^2\tilde{B}_{0H}+(P_6{}^6)^2\tilde{B}_{0p}][a_0^*a_0^*+a_0a_0], \quad (70a)$$

with

$$\tilde{A}_{0H}-\tilde{B}_{0H}=(X/\lambda\beta HS)(S_0-T_0+S_{k0}-T_{k0}+2R_{k0})^2, \quad (70b)$$

$$\tilde{A}_{0p}-\tilde{B}_{0p}=(Y/6P_6{}^6S^6)(V_0-W_0+V_{6k0}-W_{6k0}+2U_{6k0})^2, \quad (70c)$$

where X and Y are as defined in (64a) and (64b). Only the differences of the A's and B's as in (70b) and (70c) will be of interest. Since $A_0=B_0$ from (51), we have

$$\hbar_{\omega(0)}\sim 2s[2A_0]^{\frac{1}{2}}[H^2(A_{0H}+\tilde{A}_{0H}-B_{0H}-\tilde{B}_{0H})+(P_6{}^6)^2(A_{0p}+\tilde{A}_{0p}-B_{0p}-\tilde{B}_{0p})]^{\frac{1}{2}} \quad (71)$$

using (35), (68a), (70) and (66c,d).

However, it is found that both the expressions $(A_{0H}+\tilde{A}_{0H}-B_{0H}-\tilde{B}_{0H})$ and $(A_{0p}+\tilde{A}_{0p}-B_{0p}-\tilde{B}_{0p})$ vanish identically to first order in H and $P_6{}^6$. The second-order terms have proved too complicated to be calculated explicitly. We can see no reason why they

[11] M. H. L. Pryce, Proc. Phys. Soc. (London) **A63**, 25 (1950).

should vanish and it would seem that $\omega(0)$ will be quadratic in H and P_6^6

$$\omega_0 \sim O(H^4+H^2 P_6^{6\,2}+P_6^{6\,4})^{\frac{1}{2}}. \tag{72}$$

A time-dependent magnetic field in the plane of the spins, $\mathbf{H}_{\mathrm{rf}}$, gives to rise resonance excitation of spin waves of wave vector nk_0, where n stands for one of certain integers, as will now become apparent. If the magnetic field points in the η direction, the component of the interaction linear in spin-wave operators is, from Eqs. (15) and (22),

$$-\lambda\beta H_{\mathrm{rf}}\, i(S/2)^{\frac{1}{2}} \sum_i (a_i^*-a_i) \cos\phi_i. \tag{73}$$

From Eqs. (63) one can expand $\cos\phi_i$ as

$$\cos\phi_i = \sum_{n=0}^{13} t_n \cos(nk_0 z_i). \tag{74}$$

This expansion is substituted back into (73), the a_i^*, a_i are replaced by running-wave operators (24a), and the summation over all atoms is taken. Transformations whose form corresponds to the inverse of (31) then give for the interaction in (73)

$$-\lambda\beta H_{\mathrm{rf}} i(S/2)^{\frac{1}{2}}(N/2)^{\frac{1}{2}} \sum_n t_n(w_{nk_0}+b_{nk_0}) \times(\alpha_{nk_0}{}^*+\alpha_{-nk_0}{}^*-\alpha_{nk_0}-\alpha_{-nk_0}). \tag{75}$$

It follows that the resonance absorption intensities for the various harmonics will be proportional to

$$t_n{}^2(w_{nk_0}+b_{nk_0})^2. \tag{76}$$

From (63) one finds that to first order in H or P_6^6

$$\cos\phi_i = \cos(k_0\zeta_i) - \tfrac{1}{2}X + \tfrac{1}{2}X\cos(2k_0\zeta_i) - \tfrac{1}{2}Y\cos(5k_0\zeta_i) + \tfrac{1}{2}Y\cos(7k_0\zeta_i). \tag{77}$$

To second order, $t_9=t_{10}=t_{12}=0$, whereas $t_1, t_2\cdots, t_8, t_{11}, t_{13}$ assume rather involved forms, which in general depend on both X and Y. Since, when $\theta=90°$, A_q, given in (27), is an even function, it follows from (37) and (38) that

$$\begin{aligned}(w_q+b_q)^2 &= (A_q+B_q)^{\frac{1}{2}}(A_q-B_q)^{-\frac{1}{2}} \\ &= -2S[K(q)+J(q)-J(k_0)]/\hbar\omega(q).\end{aligned} \tag{78}$$

When $n=0$, (78) becomes

$$[w(0)+b(0)]^2 = 4SA(0)/\hbar\omega(0), \tag{79}$$

where $\omega(0)$ is presumed to be of the form (72). At a fixed frequency this leads to an intensity of absorption for this mode of the same order as that of the main resonance at wave vector k_0.

The intensity of the absorption of the second harmonic will be roughly reduced by a factor X^2 relative to that of the first harmonic, and the absorptions of the fifth and seventh harmonic by the factor Y^2. The relative intensities of the 3rd, 4th, 6th, 8th, 11th, and 13th harmonic are of fourth order in X and Y, and it appears unlikely that these can be observed.

3.5. FERROMAGNETIC ALIGNMENT IN THE PLANE CAUSED BY HEXAGONAL ANISOTROPY—CASE *AV*

This is the case for dysprosium and terbium at low temperature where with no applied field the spins are ferromagnetically aligned along the ξ axis.

The Hamiltonian is

$$\begin{aligned}\mathcal{H} = -\sum_{i\neq j} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j - \sum_{i,j} K_{ij}S_{i\zeta}S_{j\zeta} \\ \times\sum_i \{ (P_6^6/2)\sum_i [(S_{i\xi}+iS_{i\eta})^6+(S_{i\xi}-iS_{i\eta})^6] \\ +\lambda\beta H_{\shortparallel}S_{i\xi}+\lambda\beta H_{\perp}S_{i\eta}\}.\end{aligned} \tag{80}$$

We include an applied field with components along ξ and η. Such a field affects the resonance frequency in a most interesting manner, as will be shown below. If the equilibrium position of the spin points along the $+\xi$ axis with no applied field, then the net applied field must point within $\pm 30°$ of the ξ axis. Otherwise the spins will merely change their equilibrium direction to the hexagonal axis nearest H.

$$H_\perp \leqq H_{\shortparallel} \tan 30°. \tag{81}$$

To allow the equilibrium position of the spins to lie other than in the ξ direction because of the applied field, the coordinates used are

$$\hat{x}_i = -\boldsymbol{\zeta}, \tag{82a}$$

$$\hat{y}_i = -\boldsymbol{\xi}\sin\delta + \boldsymbol{\eta}\cos\delta, \tag{82b}$$

$$\hat{z}_i = \boldsymbol{\xi}\cos\delta + \boldsymbol{\eta}\sin\delta, \tag{82c}$$

where, as previously, $\hat{z}_i$ is the equilibrium direction of the ith spin.

$$\begin{aligned}\tfrac{1}{2}P_6^6 \sum_i [(S_{i\xi}+iS_{i\eta})^6+(S_{i\xi}-iS_{i\eta})^6] \\ = P_6^6 \sum_i [S_{iz}{}^6\cos 6\delta - 6S_{iz}{}^5 S_{iy}\sin 6\delta \\ -15S_{iz}{}^4 S_{iy}{}^2\cos 6\delta],\end{aligned} \tag{83}$$

keeping only terms to second order in S_{iy}.

The Hamiltonian can be written in terms of creation and destruction operators using the same technique as in case *A*I. The exchange energies can be obtained by setting $k_0=0$ and $\theta=\pi/2$ in the appropriate expressions in case *A*I.

The equilibrium energy is

$$E_0/N = -S^2J(0) + P_6^6S^6\cos 6\delta + \lambda\beta H_{\shortparallel}S\cos\delta + \lambda\beta H_\perp S\sin\delta. \tag{84}$$

The conditions that this be a minimum are

$$0 = -6P_6^6S^6\sin 6\delta - \lambda\beta H_{\shortparallel}S\sin\delta + \lambda\beta H_\perp S\cos\delta \tag{85}$$

and

$$-36P_6^6S^6\cos 6\delta - \lambda\beta H_{\shortparallel}S\cos\delta - \lambda\beta H_\perp S\sin\delta > 0. \tag{86}$$

The spin-wave frequencies are given by

$$\begin{aligned}\hbar\omega(q) = 2S\{[-J(q)+J(0)-K(q)-3P_6^6S^4\cos 6\delta \\ -(\lambda\beta H_{\shortparallel}/2S)\cos\delta - (\lambda\beta H_\perp/2S)\sin\delta] \\ \times[-J(q)+J(0)-18P_6^6S^4\cos 6\delta \\ -(\lambda\beta H_{\shortparallel}/2S)\cos\delta - (\lambda\beta H_\perp/2S)\sin\delta]\}^{\frac{1}{2}}.\end{aligned} \tag{87}$$

An rf field along the ζ direction excites $\omega(0)$.

In this ferromagnetic phase there will be considerable demagnetizing fields arising from the long-range dipole-dipole interaction. The important effects of these on the resonance frequency were first pointed out by Kittel[12] and they may be easily seen to appear in the frequency calculated here in a similar manner. Including these effects, we have

$$\begin{aligned}\hbar\omega(0)=\{[\lambda\beta H_{\parallel}\cos\delta+H_{\perp}\sin\delta)\\ +6P_6{}^6S^5\cos 6\delta+2K(0)S-(N_z-N_x)M]\\ \times[36P_6{}^6S^5\cos 6\delta+\lambda\beta(H_{\parallel}\cos\delta+H_{\perp}\sin)\\ -(N_z-N_y)M]\}^{\frac{1}{2}},\quad(88)\end{aligned}$$

where using the notation (82), N_z is the demagnetization factor of the sample along the magnetization direction in the plane, N_y along the perpendicular direction to this in the plane, and N_x along the direction of the hexagonal axis.

For $H=0$:

$$\hbar\omega(0)=2S\{[K(0)+3P_6{}^6S^4][18P_6{}^6S^4]\}^{\frac{1}{2}}.\quad(89)$$

4. PHASE WITH LONGITUDINAL SPIN VARIATIONS—CASES *B* AND *C*

In all the variations so far treated there has been one essential feature that allowed a discussion of the excited states in terms of spin waves. This was the fact that each site showed the same total-ordered magnetic moment, although its direction varied. In the high-T phase of Er the ζ component alone is ordered but this shows a sinusoidal variation in magnitude along the ζ axis. In the intermediate-T phase this component retains this property, although the ξ, η components are ordered and vary only in direction.

It is rather difficult to define spin deviations and transform the Hamiltonian in the usual way in this case. Instead, we examine the equations of motion which give an alternative method of approach in the usual case.[13] Since we shall only be concerned with the form of the result, the algebra is simplified by putting $\mathbf{H}=0$ in Eq. (2). Then

$$\begin{aligned}-\hbar\omega S^+(\mathbf{q})=[S^+(\mathbf{q}),\mathcal{H}]=-\frac{2}{\sqrt{N}}\{\sum_{i,j}[J_{ij}S_{i\zeta}S_j{}^+\\ -(J_{ij}+K_{ij})S_i{}^+S_{j\zeta}]e^{i\mathbf{q}\cdot\mathbf{R}_i}-2K_4\sum_i S_{i\zeta}{}^3S_i{}^+\\ -3K_6\sum_i S_{i\zeta}{}^5S_i{}^+\},\quad(90)\end{aligned}$$

where as before terms of order $1/S$ have been neglected in taking the commutators.

Now substituting in the equilibrium value for S_ζ,

$$S_\zeta=S\cos(\mathbf{k}_0\cdot\mathbf{R}),\quad(91)$$

gives

$$\begin{aligned}\hbar\omega S^+(\mathbf{q})\\ =S[J(\mathbf{q}+\mathbf{k}_0)-J(\mathbf{k}_0)-K(\mathbf{k}_0)\\ +\tfrac{3}{2}K_4S^2+(15/8)K_6S^4]S^+(\mathbf{q}+\mathbf{k}_0)\\ +S[J(\mathbf{q}-\mathbf{k}_0)-J(\mathbf{k}_0)-K(\mathbf{k}_0)\\ +\tfrac{3}{2}K_4S^2+(15/8)K_6S^4]S^+(\mathbf{q}-\mathbf{k}_0)\\ +(\tfrac{1}{2}K_4S^3+\tfrac{15}{16}K_6S^5)[S^+(\mathbf{q}+3\mathbf{k}_0)+S^+(\mathbf{q}-3\mathbf{k}_0)]\\ +\tfrac{3}{16}K_6S^5[S^+(\mathbf{q}+5\mathbf{k}_0)+S^+(\mathbf{q}-5\mathbf{k}_0)].\quad(92)\end{aligned}$$

In the limit $k_0\rightarrow 0$ this becomes equivalent to the spin-wave frequency derived for axial ferromagnetism [cf. Eq. (57)].

It is obvious from (92) that $S^+(\mathbf{q})$ is not a satisfactory normal coordinate of the system. Unless $n\mathbf{k}_0$ is a reciprocal lattice vector for some whole number n, (92) gives rise to an infinite set of coupled equations. Thus all the true normal coordinates obtained from their solution will contain some admixture of $S^+(0)$ and hence absorption of a resonance field will take place over the whole frequency range. In this case spin waves do not form a satisfactory description of the excited states and no resonance will be observable.

In the intermediate phase with ordered ξ and η components an equation of motion considered in the same approximation leads to a result similar in form to (92) but more complicated. The general result that $S^+(\mathbf{q})$ is not a satisfactory normal coordinate is repeated, and again spin waves do not form a correct description of the excited states and no resonance will be observable. However, there is one temperature (about 30°K) in this phase where the magnetic order repeats in an integral number (8) of lattice layers. For this temperature, there will be only five coupled equations which contain $S^+(0)$ and hence only five frequencies at which a well-defined resonance will be observed. In view of the uncertain values of the parameters, these equations have not been solved, but in general the frequencies will be of the order of the exchange and anisotropy energies; i.e., in the infrared.

5. DISCUSSION

In order to give a quantative discussion of the resonant frequencies and intensities derived in Sec. 3, it is necessary to consider some model of the exchange and anisotropy constants in Eq. (2) along with some of the experimental results. In particular, it is possible to relate various combinations of $J(q)$ to the critical field, i.e., the field necessary to change the spiral moment arrangement into a ferromagnetic one. Neglecting anisotropy, this is given by

$$[J(k_0)-J(0)]S\sin\theta=\lambda\beta H_c.\quad(93)$$

It is more convenient to define $H_c'=H_c/\sin\theta$. In Eq. (93), S is the temperature-dependent value corresponding to the ordered moment, so that it goes to zero when the spin ordering disappears.

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

[13] F. Englert, Phys. Rev. Letters **5**, 102 (1960); V. L. Ginzberg and V. M. Fain, Soviet Phys.—JETP **12**, 923 (1961).

In reference 1 it was shown that a simple model which used exchange interactions only within one layer (J_0) and between nearest (J_1) and next-nearest layers (J_2) gave a reasonable interpretation of the facts. Since in this paper the anisotropy energy is included explicitly, there is no need to make the further assumption of anisotropic exchange here. The angle in the spiral was given by

$$\cos k_0 c = \cos\alpha = -J_1/4J_2, \tag{94}$$

(c is the lattice constant along ζ), and

$$\begin{aligned}\lambda\beta H_c' &= 2S[J_1(\cos\alpha-1)+J_2(\cos 2\alpha-1)]\\ &= 4SJ_2(1-\cos\alpha)^2,\end{aligned} \tag{95}$$

using (94). It turns out that this relationship between H_c' and α is largely verified experimentally and gives some confidence in the model. Within it, various other expressions found in (45), etc., are:

$$J(2k_0)-J(0)=16SJ_2(1-\cos\alpha)^2(1+\cos\alpha)\cos\alpha, \tag{96}$$

and

$$\begin{aligned}&J(2k_0)+J(0)-2J(k_0)\\ &\quad=4SJ_2(1-\cos\alpha)^2(2\cos^2\alpha+2\cos\alpha+1).\end{aligned} \tag{97}$$

Within this model these expressions will show roughly the same variation with T as H_c', since the term $(1-\cos\alpha)^2$ shows the main variation.

The anisotropy terms appearing in (45), (56), (57), (67), and (88) determine an anisotropy field, say H_A. The actual field varies from metal to metal and can be related to the P_l^m which were calculated on a crystal-field model in reference 1. In the absence of experimental results, it does not seem worth pursuing the detailed calculations in each case. It is also convenient to define a hexagonal anisotropy field.

$$\lambda\beta H_h = P_6^6 S^5. \tag{98}$$

With these definitions, the possible resonance frequencies can be crudely written as follows.

For case I, from (45),

$$\hbar\omega \sim \lambda\beta\{H_c'\cos\theta+[H_c'(H_c'\cos^2\theta+H_A\sin^2\theta)]^{\frac{1}{2}}\}. \tag{99}$$

The intensity is proportional to (44), i.e., of normal intensity for ferromagnetic resonance. For case II, from (56),

$$\hbar\omega \sim \lambda\beta[H_\perp(H_\perp+H_A)]^{\frac{1}{2}}. \tag{100}$$

In the axial ferromagnetic case III,

$$\hbar\omega \sim \lambda\beta(H_A-H_{\parallel}). \tag{101}$$

In the planar spiral case IV the main resonant frequency is crudely like

$$\hbar\omega \sim \lambda\beta(H_c'H_A)^{\frac{1}{2}}, \tag{102}$$

with intensity like ferromagnetic resonance. Hexagonal anisotropy and an applied magnetic field in the plane have the effect of introducing harmonics in the sinusoidal spin order of the crystal, notably the zeroth, second, fifth, and seventh harmonic.

As mentioned in the Introduction, the resonance absorptions associated with these harmonics are expected to furnish independent information about the spin-wave spectrum. In addition, the relative absorption intensities, especially of the second harmonic as the transverse field is varied, might give additional information about the Fourier transform $J(q)$ of the spin interactions.

Except for the zeroth harmonic the absorptions will lie in the infrared; the associated frequencies will be given with sufficient accuracy by Eq. (67). However, the frequency $\omega(0)$ will be quite low, and is determined by H_h and $H_\perp$ using (72)

$$\hbar\omega(0) \sim \lambda\beta(H_\perp^4+H_\perp^2H_h^2+H_h^4)^{\frac{1}{2}}/H_c, \tag{103}$$

since it was found analytically that the coefficient of lower order terms $H_\perp^2$ and H_h^2 in the expression for ω_0 vanish.

Associated with an expression like (103) for ω_0 is an absorption probability of the same order of magnitude as that of the main resonance $q=k_0$. On the other hand, if the rf field is applied parallel to the c axis the corresponding absorption probability will be reduced by a factor $(H_\perp/H_c)^2$. It is expected that an extension of the analysis for the planar case to the conical case of arbitrary θ would lead to a similar qualitative result.

Estimates indicate that H_A is large, $\sim 10^5-10^6$ G, at low temperature and falls with increasing T, the slowest component, coming from $K(0)$ goes like M. In reference 1, H_h was estimated $\sim 10^3$ G in Dy, and this falls very rapidly with increasing T. H_c varies widely with T, falling to zero like M near the Néel point and also falling to zero again at T_c.

The simplest cases for observing resonances of a kind not dissimilar from the usual kind would be in the planar ferromagnet for Dy and probably Tb in their low-T phases. The frequency (88) can be brought into the microwave range if $H_\perp$ were applied in the hard direction, so that it worked against H_h (see Sec. 3.5). In the second factor of the square root in (88), for $\delta > 15°$, negative P_6^6 and λ give the term in P_6^6 opposite in sign to those in applied field. The applied field effectively cancels part of the hexagonal anisotropy field. Thus by a choice of applied field such that $\delta > 15°$ the frequency behaves roughly like

$$\hbar\omega \sim \lambda\beta[(H_h-aH)H_A]^{\frac{1}{2}}. \tag{104}$$

The frequency may be lowered to a more convenient experimental value.

The most desirable condition would be to have the applied field almost balance the hexagonal anisotropy. This could probably be done most conveniently by applying a field in the hard direction large enough to bring the spins to equilibrium in that direction ($\delta=30°$). For this angle (85) gives

$$H_\perp/H_{\parallel}=1/\sqrt{3} \tag{105}$$

while (86) demands

$$-\lambda\beta H_{\parallel}S/\sqrt{3}>-18P_6{}^6S^6. \tag{106}$$

For $\delta=30°$, (88) becomes

$$\hbar\omega(0)=2S\left\{\left[K(0)-3P_6{}^6S^4+\frac{\lambda\beta H_{\parallel}}{S}\frac{\sqrt{3}}{3}\right]\times\left[-18P_6{}^6S^4+\frac{\lambda\beta H_{\parallel}}{S}\frac{\sqrt{3}}{3}\right]\right\}^{\frac{1}{2}}. \tag{107}$$

If (106) is just satisfied, the second factor in (107) can be made quite small, and a conveniently low frequency obtained. Hence experimentally it would probably be most advantageous to use a field just strong enough to hold the spins in a hard direction.

The case of the cone spiral in Er at low T is probably most simply investigated with fields in the plane of a magnitude close to the critical field. It might also be possible to observe the resonance in the planar spiral cases, Dy and Ho at high T. Tb has such a phase at high T but over so narrow a range where M is small that the linewidth is likely to be large. As discussed below [e.g., (108)] it is possible by applying a field near the critical field to bring the resonance into the microwave region. This will be easiest at the low-T end of this phase where H_c is small and the hexagonal and applied-field distortions severe. In this temperature region it might also be possible to observe the higher harmonics at high frequencies.

Both the cone and the spiral in the plane are greatly distorted by a field with magnitude just less than the critical value. The spin-wave spectra in these circumstances are radically changed from those in the undistorted cone and spiral. In particular, the spin wave excited in a resonance experiment has low frequency. Physically, this is because such a spin wave corresponds to a ferromagnetic alignment. Its energy therefore goes to zero when the ferromagnetic phase becomes stable. This occurs at the critical field. In the presence of hexagonal anisotropy and an applied field in the plane, $H_\perp$ the resonance frequency for the cone is like (99) and that for the plane spiral like (102) with H_c' replaced by

$$H_c'\rightarrow H_c'-H_h-H_\perp, \tag{108}$$

where $H_h+H_\perp$ is an appropriate combination.

Thus, a low frequency can be obtained for $H_h+H_\perp$ slightly smaller in magnitude than H_c'.

The weak $\omega\sim0$ resonance given by (103) should also be most easily observable in the temperature regions where the spin arrangement distorts most easily, i.e., just above T_c in Dy and at all low T in Ho.

APPENDIX

There is an alternative method for treating the distorted helix in the plane (Sec. 3.4). This is to consider the equations of motion for the Fourier transforms of the spin operators. We discuss this method for a Hamiltonian similar to (59); however, for simplicity $P_6{}^6=0$ and the field is applied along the η direction. (This is done so that the limiting case of an antiferromagnet in the plane may be considered. For $k_0\zeta_i=n\pi$, the spins point alternately in the $+\xi$ and $-\xi$ directions, so that in this limiting case the field is applied perpendicular to the spins.)

$$\mathcal{H}=-\sum_{i\neq j}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j-\sum_{i,j}K_{ij}S_{i\zeta}S_{j\zeta}+\lambda\beta H\sum_i S_{i\eta}. \tag{A1}$$

Equations (63b) and (63c) for the distortions in angle hold when the applied field is along ξ. For a field along η

$$\phi_i=k_0\zeta_i+\delta\phi_i, \tag{A2a}$$

$$\delta\phi_i=x\cos(k_0\zeta_i)-x_2\sin(2k_0\zeta_i). \tag{A2b}$$

We seek the equations of motion for the components of $\mathbf{S}(q)$.

$$\mathbf{S}(q)=N^{-\frac{1}{2}}\sum{}_i(S_{ix}\boldsymbol{\varepsilon}_x+S_{iy}\boldsymbol{\varepsilon}_y+S_{iz}\boldsymbol{\varepsilon}_z)e^{i\mathbf{q}\cdot\mathbf{R}_i}. \tag{A3}$$

The Hamiltonian may be expanded correct to terms of the order H^2. Then the equations of motion may be found using the random-phase approximation as in Sec. 4. The equations obtained are

$$\hbar\omega S_x(q)=-iG(q)S_y(q)+C(q)S_y(q-k_0)+D(q)S_y(q+k_0), \tag{A4}$$

where

$$\begin{aligned}G(q)=2S\{&J(k_0)-\tfrac{1}{2}J(K_0+q)-\tfrac{1}{2}J(-k_0+q)\\&-\tfrac{1}{8}x^2[4J(k_0)-2J(2k_0)-2J(0)\\&-2J(k_0+q)-2J(-k_0+q)+2J(q)\\&+J(q+2k_0)+J(q-2k_0)]\}+\tfrac{1}{2}\lambda\beta Hx,\end{aligned} \tag{A5a}$$

$$\begin{aligned}C(q)=-\tfrac{1}{2}Sx[&J(0)-J(2k_0)-J(q)+J(q-2k_0)\\&+J(q+k_0)-J(q-k_0)]-\tfrac{1}{2}\lambda\beta H,\end{aligned} \tag{A5b}$$

$$\begin{aligned}D(q)=\tfrac{1}{2}Sx[&J(0)-J(2k_0)+J(q+2k_0)-J(q)\\&-J(q+k_0)+J(q-k_0)]+\tfrac{1}{2}\lambda\beta H,\end{aligned} \tag{A5c}$$

and

$$\hbar\omega S_y(q)=iR(q)S_x(q)+FS_x(q-k_0)-FS_x(q+k_0), \tag{A6}$$

where

$$\begin{aligned}R(q)=2S\{&J(k_0)-J(q)-K(q)\\&-\tfrac{1}{4}x^2[2J(k_0)-J(2k_0)-J(0)]\}+\tfrac{1}{2}\lambda\beta Hx,\end{aligned} \tag{A7a}$$

$$F=\tfrac{1}{2}Sx[J(0)-J(2k_0)]+\tfrac{1}{2}\lambda\beta H. \tag{A7b}$$

In general any $\mathbf{S}(q)$ is linked to $\mathbf{S}(q\pm k_0)$. We are interested in the frequency for $\mathbf{S}(0)$, $\omega(0)$, which would be zero if no applied field were present, $\mathbf{S}(0)$ is linked to $\mathbf{S}(\pm k_0)$. $\mathbf{S}(k_0)$ in turn is linked to $\mathbf{S}(2k_0)$ as well as $\mathbf{S}(0)$. We neglect the interaction with $\mathbf{S}(2k_0)$. This approximation corresponds to that of second-order perturbation theory. The secular determinant for ω then is obtained from (A4) and (A6) neglecting the terms in $\mathbf{S}(\pm 2k_0)$ that come in for $\hbar\omega\mathbf{S}(\pm k_0)$,

The secular determinant is

$$\begin{vmatrix} -\omega & -iG(0) & 0 & D(0) & 0 & C(0) \\ iR(0) & -\omega & -F & 0 & F & 0 \\ 0 & C(k_0) & -\omega & -iG(k_0) & 0 & 0 \\ F & 0 & iR(k_0) & -\omega & 0 & 0 \\ 0 & D(-k_0) & 0 & 0 & -\omega & -iG(k_0) \\ -F & 0 & 0 & 0 & iR(k_0) & -\omega \end{vmatrix} = 0. \quad \text{(A8)}$$

We express ω in a series with H as the expansion parameter:

$$\omega = \omega_0 + \omega_1 H + \omega_2 H^2 + \cdots. \quad \text{(A9)}$$

For $\omega(0)$, the frequency of interest, $\omega_0 = 0$. It is possible to expand (A8), choose the lowest order terms in H (of order H^2), and set them equal zero. (This corresponds to the usual perturbation method.) This gives a value for $H^2\omega_1^2(0)$,

$$H^2\omega_1^2(0) = R(0)\{G(0) + [1/G(k_0)] \times [D(-k_0)C(0) + D(0)C(k_0)]\}. \quad \text{(A10)}$$

Here $R(0)$ and $G(k_0)$ are of the order of H^0, C and D are of order H, and $A(0)$ is of order H^2. The expression in the curly brackets is equal to zero. Thus as noted in Sec. 3.4, $\omega(0)$ is zero to first order in H.

It is interesting to note that this result does not hold true for the case of an antiferromagnet confined to an easy plane with an applied field in the plane perpendicular to the unperturbed position of the spins.[14] For this particular case, k_0 lies at the edge of the Brillouin zone so that $+k_0$ and $-k_0$ are equivalent. This means that $\mathbf{S}(q+k_0)$ is the same as $\mathbf{S}(q-k_0)$. In this case, (A8) reduces to a 4×4 determinant with no elements connecting $q=0$ and $q=k_0$.

$$\begin{vmatrix} -\omega & -iG(0) & 0 & 0 \\ iR(0) & -\omega & 0 & 0 \\ 0 & 0 & -\omega & -iG(k_0) \\ 0 & 0 & iR(k_0) & -\omega \end{vmatrix} = 0 \quad \text{(A11)}$$

and

$$H\omega_1 = (G_0 R_0)^{\frac{1}{2}}. \quad \text{(A12)}$$

Using (A5a) and (A7a),

$$H\omega_1 = gBH\left[\frac{J(k_0) - J(0) - K(0)}{2J(k_0) - J(2k_0) - J(0)}\right]^{\frac{1}{2}}. \quad \text{(A13)}$$

[14] T. Nagamiya, K. Yosida, and R. Kubo, *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 1.

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Nuclear Magnetic Resonance in Metallic Single Crystals

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A method is presented for observing nuclear magnetic resonance absorption in metallic single crystals. Single crystals thick compared to the skin depth are used and accurate corrections are made for the distortion due to eddy currents. Experimental results for aluminum and copper single crystals with the fixed magnetic field parallel to the [001], [$\bar{1}$11], and [$\bar{1}$10] directions, respectively, demonstrate the feasibility of the method. The Knight shifts in aluminum and copper are found to be isotropic and equal to the powder values. The experimental second moments vary with orientation approximately in proportion to the theoretical second moments, but are somewhat larger, as in the powders.

INTRODUCTION

SINCE the discovery of nuclear magnetic resonance absorption (NMR) in metallic copper by Pound,[1] NMR experiments have been performed on a wide variety of metals and alloys. Because of the classical skin effect arising from eddy currents, the radio-frequency magnetic fields will penetrate a good conductor only to a depth of a few thousandths of a centimeter. For this reason, virtually all the NMR work on metals up to now has been performed on finely divided powders, thin polycrystalline foils, or thin evaporated layers. No NMR experiments have been reported on metallic single crystals. It would be highly desirable in studying many phenomena in metals, such as anisotropic Knight shifts and relaxation times or quadrupole interactions, to work with single crystals. The purpose

[1] R. V. Pound, Phys. Rev. **73**, 1112 (1948).