PHYSICAL REVIEW VOLUME 85, NUMBER 2 JANUARY 15, 1952

Theory of Antiferromagnetic Resonance

F. Keffer and C. Kittel
Department of Physics, University of California, Berkeley, California
(Received October 1, 1951)

The spin resonance condition $\omega/\gamma = H_0 \pm [H_A(2H_E+H_A)]^{\frac{1}{2}}$ previously given by Kittel for a disk-shaped single-domain uniaxial or cubic antiferromagnetic crystal at 0°K with H_0 parallel to the domain axis is extended by classical calculations to cover finite temperature, ellipsoidal shape, orthorhombic symmetry, generalized two-lattice anisotropy, and arbitrary static field direction. The normal precessional modes are discussed. A quantum-mechanical derivation of the resonance equations is carried out by the method developed by Van Vleck for ferromagnetic resonance; no new features are introduced by the quantum-mechanical calculation. Several factors contributing to the line width are considered. Existing experimental data on antiferromagnetic resonance are reviewed; the data are scanty and taken in circumstances not closely related to the situation envisaged by the theory.

I. INTRODUCTION

We are concerned in this paper with the theory of electron spin resonance absorption in antiferromagnetic crystals, and we find results exhibiting very significant differences from the results with ferromagnetic and paramagnetic crystals. In an earlier[1] paper it was shown that the resonance condition in a uniaxial or cubic antiferromagnetic crystal at 0°K is given by

$$\omega/\gamma = H_0 \pm [H_A(2H_E+H_A)]^{\frac{1}{2}}, \qquad (1)$$

when the static magnetic field H_0 is parallel to the domain axis; here H_E is the Weiss exchange field, H_A is the anisotropy field, and $\gamma = ge/2mc$ is the magnetomechanical ratio; g is the spectroscopic splitting factor.

A number of the better-known antiferromagnetic crystals have Curie temperatures of the order of magnitude of 100°K, and we may for these estimate very roughly that $H_E \sim 10^6$ oersteds and $H_A \sim 10^3$ oersteds, leading to a resonance frequency $\omega \sim 5$ cm^{-1} for zero static field; to obtain resonance at a microwave frequency ~ 1 cm^{-1} it is necessary to apply a static field $H_0 \sim 5\times 10^4$ oersteds. We know also of antiferromagnetic crystals with lower Curie temperatures, and resonance experiments have been performed at Leiden[2] on a crystal ($CuCl_2\cdot 2H_2O$) with a Curie temperature ~ 5°K, so that here we should expect a zero field splitting equivalent to 10^3 to 10^4 oersteds. It seems that a splitting of this nature was observed, although the published account of the measurements is very incomplete. Our frequency estimates may be quite badly off, as there are few data from which to estimate the anisotropy field.

In Sec. II of this paper we carry out a classical calculation of the resonance frequencies for a cubic antiferromagnetic crystal at 0°K with the static field making an arbitrary angle with the axis of the antiferromagnetic domains. We consider the nature of the normal modes of motion of the spin systems, and calculate rf susceptibilities, which may be very considerably larger than in paramagnetic salts.

In Sec. III the theory is extended to arbitrary temperatures, and demagnetizing effects are considered. As we approach the Curie temperature the phenomenon comes to resemble ordinary paramagnetic resonance. The demagnetizing effect of the shape of the specimen enters in a subtle way, but is only of interest when the exchange field is not too much greater than the demagnetizing field.

In Sec. IV we consider the effect of the anisotropy energy when there is an anisotropic coupling between the antiferromagnetic sublattices. We also extend the theory to crystals of orthorhombic symmetry, of some experimental interest. Section V is concerned with line widths, and it is pointed out that polycrystalline specimens are not well-suited to experimental work because of the large orientational broadening.

In Sec. VI the resonance Eq. (1) is derived quantum-mechanically, employing the powerful and general method used by Van Vleck[3] in treating ferromagnetic resonance. It is reassuring that the quantum-mechanical calculation agrees with the classical results and introduces no new features. We may note that both the classical and quantum calculations given in this paper utilize implicitly the two sublattice model of antiferromagnetics which was employed previously by Van Vleck[4] for static calculations. This model assumes in effect that the antiferromagnetic ground state is adequately approximated by a spin function of the character $\alpha_1\beta_2\alpha_3\beta_4\alpha_5\cdots$; it is satisfying in this connection that Anderson[5] has proved that the exact ground-state eigenvalue must be close to the energy of the two sublattice approximation. For structures such as MnO, where the paramagnetic ions form a face-centered cubic lattice, there are eight sublattices. Shull, Strauser, and Wollan[6] have found by neutron diffraction that four

---

[1] C. Kittel, Phys. Rev. **82**, 565 (1951).

[2] Poulis, van den Handel, Ubbink, Poulis, and Gorter, Phys. Rev. **82**, 552 (1951).

[3] J. H. Van Vleck, Phys. Rev. **78**, 266 (1950).

[4] J. H. Van Vleck, J. Chem. Phys. **9**, 85 (1941). For a general review of antiferromagnetism the reader is referred to J. H. Van Vleck, J. phys. et radium **12**, 262 (1951).

[5] P. W. Anderson, Phys. Rev. **83**, 1260 (1951).

[6] Shull, Strauser, and Wollan, Phys. Rev. **83**, 333 (1951).

sublattices are magnetized in one direction, and four in the opposite direction. In this case in a resonance experiment we deal only with the two groups of spins, as within each group every spin sees the same exchange and anisotropy fields. In Sec. VII we review the experimental situation.

II. CLASSICAL CALCULATION FOR 0°K, CUBIC SYMMETRY

A. Static Field Parallel to Preferred Axis

This case has been briefly treated in an earlier paper.[1] The equations of motion for the two sublattices are

$$\begin{aligned}
dM_1^x/dt&=\gamma(H^z+H_0+H_A-\lambda M_2^z)M_1^y\\
&\qquad-\gamma(H^y-\lambda M_2^y)M_1^z,\\
dM_2^x/dt&=\gamma(H^z+H_0-H_A-\lambda M_1^z)M_2^y\\
&\qquad-\gamma(H^y-\lambda M_1^y)M_2^z,\\
dM_1^y/dt&=-\gamma(H^z+H_0+H_A-\lambda M_2^z)M_1^x\\
&\qquad+\gamma(H^x-\lambda M_2^x)M_1^z, \qquad (2)\\
dM_2^y/dt&=-\gamma(H^z+H_0-H_A-\lambda M_1^z)M_2^x\\
&\qquad+\gamma(H^x-\lambda M_1^x)M_2^z,\\
dM_1^z/dt&=\gamma(H^y-\lambda M_2^y)M_1^x-\gamma(H^x-\lambda M_2^x)M_1^y,\\
dM_2^z/dt&=\gamma(H^y-\lambda M_1^y)M_2^x-\gamma(H^x-\lambda M_1^x)M_2^y,
\end{aligned}$$

where H_0 is the static field, assumed in the preferred (z) direction, $\mathbf{H}$ is the rf field; $-\lambda\mathbf{M}_2$ and $-\lambda\mathbf{M}_1$ are the exchange fields acting on $\mathbf{M}_1$ and $\mathbf{M}_2$ respectively; H_A and $-H_A$ are the anisotropy fields; and $\gamma=ge/2mc$. For antiferromagnetism λ is positive. We assume small deflections from the preferred axis so that the anisotropy may be treated as an effective field directed along the axis.

To find the resonance frequency we set $\mathbf{H}=0$. Products like $M_2^yM_1^x$ can be taken as zero since the magnetization is assumed to be predominantly in the z direction.[7] Let $H_E=\lambda M_1^z=-\lambda M_2^z$ and assume M_1^x, M_2^x, M_1^y, M_2^y all proportional to $\exp(i\omega t)$.[8] The resonance frequencies are then the eigenvalues of the resonance matrix $\mathbf{A}$:

$$\mathbf{A}=\begin{vmatrix} 0 & 0 & -ia & -ib \\ 0 & 0 & ib & -id \\ ia & ib & 0 & 0 \\ -ib & id & 0 & 0 \end{vmatrix}, \tag{3}$$

where $a=\gamma(H_0+H_A+H_E)$, $b=\gamma H_E$, $d=\gamma(H_0-H_A-H_E)$. The matrix equation

$$\mathbf{AS}=\mathbf{SA}' \quad (\mathbf{A}' \text{ diagonal}) \tag{4}$$

is satisfied by

$$\mathbf{S}=\begin{vmatrix} 1 & 1 & 1 & 1 \\ -\eta^{-1} & -\eta^{-1} & -\eta & -\eta \\ i & -i & i & -i \\ -i\eta^{-1} & i\eta^{-1} & -i\eta & i\eta \end{vmatrix}, \tag{5}$$

where

$$\eta H_E=H_A+H_E+[H_A(2H_E+H_A)]^{\frac{1}{2}}, \tag{6}$$

and hence the columns of $\mathbf{S}$ are eigenvectors of $\mathbf{A}$ and represent the four normal modes N_i. The corresponding resonance frequencies (eigenvalues) are

$$\begin{aligned}
\omega_1&=-\omega_2=\gamma H_0+\gamma[H_A(2H_E+H_A)]^{\frac{1}{2}},\\
\omega_3&=-\omega_4=\gamma H_0-\gamma[H_A(2H_E+H_A)]^{\frac{1}{2}}.
\end{aligned} \tag{7}$$

The normal modes are shown in Fig. 1. N_1 and N_2 are equivalent and represent $\mathbf{M}_1$ and $\mathbf{M}_2$ in a circular clockwise precession of frequency ω_1 about $+z$, i.e., looking along the z axis one sees $\mathbf{M}_1$ and $\mathbf{M}_2$ describing unequal size circles in the same sense.

To obtain a better picture of this motion consider Fig. 1(a) with $H_0=0$. If the precessions are to be in the same sense and of magnitude ω, and with $M=|\mathbf{M}_1|=|\mathbf{M}_2|$, the following equations of motion must be satisfied:

$$\begin{aligned}
\omega M\sin\theta_1&=\gamma H_AM\sin\theta_1+\gamma M^2\lambda\sin(\theta_1-\theta_2),\\
\omega M\sin\theta_2&=-\gamma H_AM\sin\theta_2+\gamma M^2\lambda\sin(\theta_1-\theta_2).
\end{aligned}$$

Assuming θ_1 and θ_2 small, these equations may be solved for ω and for θ_1/θ_2. They yield

$$\omega=\gamma[H_A(2H_E+H_A)]^{\frac{1}{2}};\quad \theta_1/\theta_2=\eta.$$

When H_0 is added, the precessional frequency of both $\mathbf{M}_1$ and $\mathbf{M}_2$ is simply increased by γH_0. Figure 1(b) can be interpreted in an analogous manner.

For $H_0=0$ we have degeneracy, and $\omega_1=\omega_4$, $\omega_2=\omega_3$. We may combine N_1 with N_4, N_2 with N_3 into the linear combinations P_i as follows:

$$\begin{aligned}
P_1&=\eta N_1-N_4, & P_3&=\eta N_1+N_4,\\
P_2&=\eta N_2-N_3, & P_4&=\eta N_2+N_3,
\end{aligned} \tag{8}$$

giving rise to a transformation

$$\mathbf{S}'=\begin{vmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ i\epsilon & -i\epsilon & i\epsilon^{-1} & -i\epsilon^{-1} \\ -i\epsilon & i\epsilon & i\epsilon^{-1} & -i\epsilon^{-1} \end{vmatrix}, \tag{9}$$

where

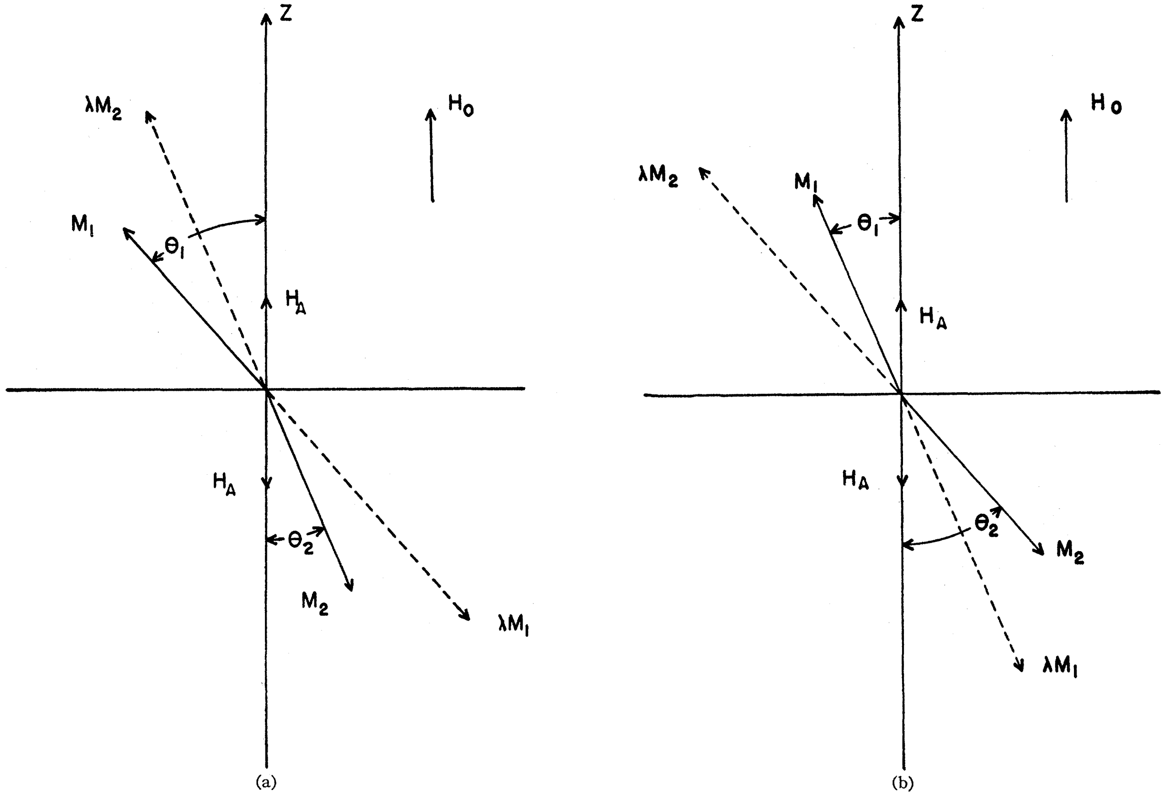
$$\epsilon=(\eta+1)/(\eta-1)=+[(2H_E+H_A)/H_A]^{\frac{1}{2}}.$$

P_1 and P_2 are now equivalent, as are P_3 and P_4. In these modes $\mathbf{M}_1$ precesses clockwise, $\mathbf{M}_2$ counterclockwise about $+z$, i.e., looking along z one sees $\mathbf{M}_1$ and $\mathbf{M}_2$ describing equal size ellipses in opposite directions, with the ratio of major to minor axis equal to ϵ. A discussion of this motion is illuminating. In mode P_1 (or P_2) the vectors $\mathbf{M}_1$ and $\mathbf{M}_2$ are colinear as they cross the yz plane and each is acted upon by only its

---

[7] Also because, for quantum-mechanical reasons, we seek transitions for which M_1^z and M_2^z are constants of the motion—other transitions giving us multiples of the fundamental frequency. For a discussion of this point see J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).

[8] They are actually proportional to $\sin\omega t$ or $\cos\omega t$. This can be taken care of, and the phase relations identified, by noting the complex amplitudes of the normal modes in, e.g., Eq. (5).



(a) (b)

FIG. 1. (a) The normal modes N_1 and N_2. $\theta_1/\theta_2=\eta$. (b) The normal modes N_3 and N_4. $\theta_2/\theta_1=\eta$.

anisotropy field, H_A. One-quarter cycle later as the vectors cross the xz plane they will be non-collinear, pointing in the same direction along x, and hence making an angle with H_E of twice that made with H_A. Each will be acted on by a field equal to $2H_E+H_A$. We compare the situation to ferromagnetic resonance in a plane surface, where the fields seen one-quarter cycle apart are B and H, and the resonance frequency is $\gamma(BH)^{\frac{1}{2}}$. Similarly we have here $\pm\gamma[H_A(2H_E+H_A)]^{\frac{1}{2}}$. The ± arises here since we deal with two magnetizations precessing in opposite directions. In mode P_3 (or P_4) we have the same argument, except the vectors are colinear as they cross the xz plane. One can, of course, form analogous combinations of the N_i using arbitrary axes x, y, resulting in a similar motion. That is, the elliptical paths cut out in the xy plane are of arbitrary orientation.

B. Susceptibilities

By solving Eq. (2) with the rf field $\mathbf{H}$ left in, it is found that, for a circularly polarized field $H^-=H^x-iH^y$, and defining $M^-=M^x-iM^y$, $M=|\mathbf{M}_1|=|\mathbf{M}_2|$, the susceptibility is

$$\chi^-=M^-/H^-=-2\gamma^2H_AM/[(\omega-\omega_1)(\omega-\omega_3)], \quad (10)$$

with ω_1 and ω_3 as given by Eq. (7). For a linearly polarized rf field

$$\chi^x=\frac{M^x}{H^x}=\frac{-2\gamma^2H_AM(\omega^2+\omega_1\omega_3)}{(\omega^2-\omega_1^2)(\omega^2-\omega_3^2)}. \quad (11)$$

These susceptibilities may be quite large compared to those of paramagnetic resonance. Assuming a relaxation frequency Δ we have for the antiferromagnetic susceptibility at $\omega=\omega_1$:

$$\chi_{AF}^-\cong-i\gamma H_AM/[(2H_AH_E)^{\frac{1}{2}}\Delta],$$

and for the paramagnetic susceptibility at $\omega=\gamma H_0$:

$$\chi_P^-\cong-i\gamma M_P/\Delta.$$

If the Δ's are the same, and with $M_P\approx\lambda^{-1}H_0$ (near the Curie point the static susceptibility is λ^{-1}), we have for the ratio of the rf susceptibilities at 0°K and just above the Curie point:

$$\chi_{AF}^-/\chi_P^-\cong(H_AH_E)^{\frac{1}{2}}/H_0; \quad (12)$$

this ratio is ~ 10 for $H_0\sim 10^4$, $H_E\sim 10^6$, $H_A\sim 10^4$ oersteds.

C. Static Field Perpendicular to Preferred Axis

Here $\mathbf{M}_1$ and $\mathbf{M}_2$ find new equilibrium positions at angles ϕ from the preferred axes. Assuming now H_0 along the x direction we introduce two new sets of

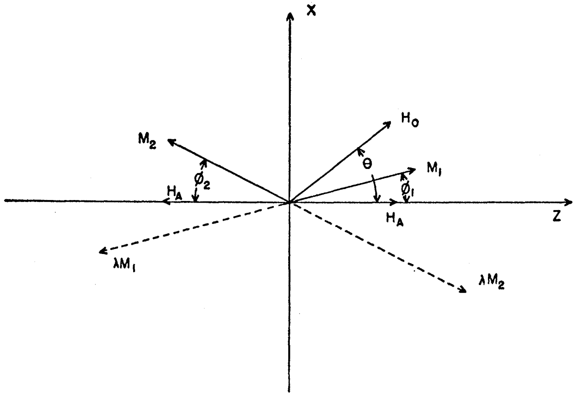


FIG. 2. H_0 at arbitrary angle.

axes: $x'y'z'$ for $\mathbf{M}_1$ (with z' in the equilibrium direction) and $x''y''z''$ for $\mathbf{M}_2$ (with z'' in the equilibrium direction of $\mathbf{M}_2$):

$$\begin{aligned} M_1^{x'} &= M_1^x \cos\phi - M_1^z \sin\phi, \\ M_1^{z'} &= M_1^x \sin\phi + M_1^z \cos\phi, \\ M_2^{x''} &= M_2^x \cos\phi + M_2^z \sin\phi, \\ M_2^{z''} &= -M_2^x \sin\phi + M_2^z \cos\phi. \end{aligned} \tag{13}$$

This transformation is applied to Eq. (2) in absence of $\mathbf{H}$. Terms like $M_2^y M_1^{x'}$ are dropped. Setting $dM_1^{z'}/dt = dM_2^{z''}/dt = 0$ gives the equilibrium condition on ϕ and hence the fundamental frequencies.[7] This gives

$$-H_0 \cos\phi + H_A \sin\phi + 2H_E \cos\phi \sin\phi = 0.$$

When $H_0 \ll H_E$, and since $H_A \ll H_E$, ϕ will be very small and hence

$$\phi \cong H_0/2H_E, \tag{14}$$

which gives for the static susceptibility

$$\chi_\perp = (M_{1\perp} + M_{2\perp})/H_0 = \lambda^{-1}, \tag{15}$$

a well-known result.

On using Eq. (14) the transformed equations of motion simplify to equations similar to Eq. (2), but in different coordinate systems. The resulting resonance matrix yields the frequencies

$$(\omega/\gamma)^2 = 2H_E H_A + H_A^2 + \tfrac{1}{2}H_0^2 \pm (H_E + H_A)(H_0^2/2H_E). \tag{16}$$

Recalling that since ϕ must be small for our approximation, H_0 must be $\ll H_E$, and using $H_A \ll H_E$:

$$\omega/\gamma \cong \pm[2H_E H_A + H_0^2]^{\frac{1}{2}}, \tag{17a}$$

and

$$\omega/\gamma \cong \pm(2H_E H_A).^{\frac{1}{2}} \tag{17b}$$

On solving the equations of motion for the rf susceptibilities one finds that, if the rf field is in the usual y direction, only the frequencies (17a) will be excited. To excite the frequencies (17b) the rf field must be in the direction of H_0.[8a]

D. Static Field at Arbitrary Angle to Preferred Axis

Let H_0 be in the xz plane at angle θ to $+z$. (See Fig. 2.) Balancing torques at equilibrium we have

$$\begin{aligned} H_0 \sin(\theta - \phi_1) &= H_E \sin(\phi_1 + \phi_2) + H_A \sin\phi_1, \\ H_0 \sin(\theta + \phi_2) &= H_E \sin(\phi_1 + \phi_2) + H_A \sin\phi_2. \end{aligned}$$

Assuming ϕ_1 and ϕ_2 small these give

$$\phi_{1,2} = \frac{H_0 \sin\theta (H_A \mp H_0 \cos\theta)}{H_A^2 + 2H_A H_E - H_0^2 \cos^2\theta}.$$

If

$$H_0^2 \ll 2H_A H_E/\cos^2\theta, \tag{18}$$

Then

$$\phi_{1,2} \cong H_0 \sin\theta (H_A \mp H_0 \cos\theta)/2H_A H_E. \tag{19}$$

It is to be noted that although Eq. (18) only requires $H_0 \ll H_E$ at $\theta \sim 90°$, it requires the more stringent $H_0 \ll (2H_A H_E)^{\frac{1}{2}}$ at $\theta < 45°$. This is because $\mathbf{M}_1$ and $\mathbf{M}_2$ attempt to become perpendicular to H_0 for minimum free energy and have weaker fields to overcome when θ is $\sim 45°$ or less. In order to remove the restriction of Eq. (18) it would be necessary to depart from the small angle approximation we have used throughout, and the equations would become much more involved.

The resonance frequency may be found by a transformation similar to Eq. (13). Let

$$y = H_0^2 \sin^2\theta / 2H_A H_E. \tag{20}$$

Thus the resonance frequencies are

$$(\omega/\gamma^2)^2 \cong 2H_E H_A + H_0^2[\cos^2\theta (1-y)^2 + \tfrac{1}{2}\sin^2\theta] \pm \tfrac{1}{2}H_0[H_0^2 \sin^4\theta + 16 H_E H_A \cos^2\theta (2+y)(1-y)^2]^{\frac{1}{2}}. \tag{21}$$

The normal modes and rf susceptibilities are quite involved. With the rf field in the usual y direction all frequencies should be observed if θ is appreciably less than 90°.

III. EXTENSION TO ARBITRARY TEMPERATURES, CUBIC SYMMETRY

A. Static Field Parallel to Preferred Axis

We assume the ordering sufficiently strong for vectors $\mathbf{M}_1$ and $\mathbf{M}_2$ to have a meaning. First the case of field along the preferred axis is worked out, then the methods of Sec. II are used to extend the result to arbitrary field direction. The equations of motion (2) are still valid providing we can assume H_A the same for both sublattices. This is adequate since near the Curie point where, as we can see from microscopic models, the anisotropy fields will be noticeably different, they will also be negligibly small. If H_0 is large enough to cause an appreciable difference in the two anisotropy fields, it will also be large enough to dominate the resonance. We assume $H_A/H_E \ll 1$ at all temperatures.

The analog of Eq. (7) when $|M_1^z| \neq |M_2^z|$ is

$$\omega/\gamma = H_0 - \tfrac{1}{2}\lambda(M_1^z + M_2^z) \pm \{H_A[H_A + \lambda(M_1^z - M_2^z)] + (\lambda/2)^2(M_1^z + M_2^z)^2\}^{\frac{1}{2}}. \tag{22}$$

[8a] We are indebted to Professor T. Nagamiya of Osaka University for calling our attention to the existence of the frequencies (17b).

This may be simplified by noting that since $M_1{}^z+M_2{}^z=\chi_{\parallel}H_0$, and on using Eq. (15),

$$\lambda(M_1{}^z+M_2{}^z)=\alpha H_0, \tag{23}$$

where

$$\alpha=\chi_{\parallel}/\chi_{\perp}; \tag{24}$$

α increases from 0 to 1 between 0°K and the Curie point. We define a temperature-dependent average H_E:

$$2H_E=\lambda(M_1{}^z-M_2{}^z). \tag{25}$$

Thus Eq. (22) becomes

$$\omega/\gamma\cong H_0(1-\alpha/2)\pm[2H_EH_A+(\alpha/2)^2H_0{}^2]^{\frac{1}{2}}. \tag{26}$$

B. Static Field at Arbitrary Angle

Figure 2 applies, except $|\mathbf{M}_1|>|\mathbf{M}_2|$. Balancing torques at equilibrium and solving for ϕ_1 and ϕ_2, assuming them small

$$\phi_{1,2}=\frac{H_0\sin\theta[H_A\mp(1-\alpha)H_0\cos\theta]}{2H_AH_E+H_A{}^2-(1-\alpha)H_0{}^2\cos^2\theta}.$$

If

$$H_0{}^2\ll 2H_AH_E/[(1-\alpha)\cos^2\theta], \tag{27}$$

then

$$\phi_{1,2}\cong H_0\sin\theta[H_A\mp(1-\alpha)H_0\cos\theta]/2H_AH_E. \tag{28}$$

Equations (27) and (28) are analogous to (18) and (19) and the remarks given below the latter apply here also.[8b] Proceeding as in the 0°K case we find the resonance frequencies by solving the characteristic equation. The result is quite complicated algebraically unless we make the assumptions

$$H_A(T)\ll H_E(T) \tag{29}$$

and

$$H_0{}^2\ll ZH_A(T)H_E(T). \tag{30}$$

Under these restrictions the resonance frequencies are

$$\begin{aligned}(\omega/\gamma)^2\cong{}&2H_AH_E+\tfrac{1}{2}H_0{}^2[1+(1+\alpha)^2\cos^2\theta]\\&\pm\tfrac{1}{2}H_0\{8H_AH_E(Z-\alpha)^2\cos^2\theta\\&+H_0{}^2[\sin^4\theta+\cos^2\theta(Z-\alpha)^2(Z\sin^2\theta+\alpha^2\cos^2\theta)]\}^{\frac{1}{2}}.\end{aligned} \tag{31}$$

This equation, in the form of H_0 as a function of ω, has been given by Nagamiya.[9] It is to be noted that his α is equal to our $(1-\alpha)$.

C. Temperature Dependence of $\alpha(=\chi_{\parallel}/\chi_{\perp})$

Van Vleck[4] has derived expressions for the susceptibility of antiferromagnetics in small fields. His results are

$$\alpha/2=(1+A)^{-1}, \tag{32}$$

where, in particular, for spin $S=1/2$

$$A=\frac{T/T_c}{1-(M/M_0)^2}$$

and for spin $S\rightarrow\infty$

$$A=\frac{T/3T_c}{1-(M/M_0)^2-2T/3T_c}.$$

In these relations T_c is the Curie temperature and M/M_0 is the ratio of the spontaneous inner magnetization of one sublattice at temperature T to that at absolute zero. This ratio rises rapidly to 1 as T goes below T_c and is plotted in books on ferromagnetism.

D. Demagnetizing Effects

We show that the central maximum of the resonance line is essentially unaffected except in materials with very low Curie points (small exchange fields). Effects on line width are considered in Sec. V.

Assuming an elliptical specimen with demagnetizing coefficients N^x, N^y, N^z, we take account of demagnetizing effects in the resonance equations by replacing $\mathbf{H}$ in Eq. (2) by

$$\mathbf{H}-[\mathbf{i}N^x(M_1{}^x+M_2{}^x)+\mathbf{j}N^y(M_1{}^y+M_2{}^y)+\mathbf{k}N^z(M_1{}^z+M_2{}^z)].$$

The general solution at arbitrary temperature and angles of H_0 is quite involved algebraically. We give two special cases.

(1) 0°K, H_0 along preferred axis.—

$$\begin{aligned}(\omega/\gamma)^2={}&H_0{}^2+H_A{}^2+H_AH_E[2+(N^y+N^x)/\lambda]\\&\pm\{4H_0{}^2H_A{}^2+4H_0{}^2H_AH_E[2+(N^y+N^x)/\lambda]\\&+H_A{}^2H_E{}^2(N^y-N^x)/\lambda\}^{\frac{1}{2}}.\end{aligned} \tag{33}$$

This reduces, for $H_0=0$, to

$$\omega/\gamma=\pm[2H_AH_E(1+N^x\lambda^{-1})]^{\frac{1}{2}} \tag{34a}$$

and

$$\pm[2H_AH_E(1+N^y\lambda^{-1})]^{\frac{1}{2}}. \tag{34b}$$

With $N^y\neq N^x$ there are four resonance frequencies. The difference between Eq. (34a) and Eq. (34b) may not be resolveable, but may serve only to broaden the lines by $\Delta\omega/\omega\sim(N^x-N^y)/2\lambda$. In MnO, e.g., where $\lambda^{-1}=\chi_{\perp}\sim10^{-4}$ we have $\Delta\omega/\omega\sim10^{-3}$ (or smaller). If the Curie point is 1°K, then $\Delta\omega/\omega\sim10^{-1}$. The ideal shape for single-crystal experiments is one with $N^x=N^y$, i.e., with cylindrical symmetry. The normal modes of Eq. (34) are the same as those given by Eq. (42) of Sec. IV.

[8b] It should be noted that $2H_AH_E/(1-\alpha)$ may well *increase* with increasing temperature. If it does, it should take larger fields H_0 as the temperature rises to cause the parallel susceptibility suddenly to jump to a high value, indicating that the magnetizations have flopped over to the hard axis (or to a perpendicular axis in case of cubic symmetry). Such an increase with temperature has been found at Leiden in $CuCl_2\cdot2H_2O$. See reference 2. This type of experiment, incidentally, offers an independent method of determining some of the quantities entering into the resonance equations, since from free energy considerations it is seen that the threshhold strength of field applied parallel to the easy axis and causing a flop is

$$H_0{}^2=2H_AH_E/(1-\alpha).$$

[9] T. Nagamiya, Prog. Theor. Phys. 6, 342 (1951).

(2) $N^x=N^y=N$, *arbitrary temperature, and arbitrary direction of* H_0.—We have $N^z=4\pi-2N$. By means of the substitutions[10]

$$H_0''=H_0[1+(3N-4\pi)\alpha/\lambda],$$
$$2H_E''=(\lambda+N)(M_1^z-M_2^z),$$

we restore the equations of motion (2) in absence of demagnetizing factors, but now in terms of the primed fields. Hence Eq. (31) now holds with H_0 replaced by H_0'' and H_E replaced by H_E''. The change is not important if $\lambda\gg N$. If $N=4\pi/3$ (spherical sample) the only change is in the size of effective H_E.

IV. MACROSCOPIC DISCUSSION OF ANISOTROPY

A. Cubic Symmetry

Since the anisotropy is of extreme importance in determining antiferromagnetic resonance, some of the possibilities are examined from a macroscopic point of view in this section. Microscopic analogs, from a quadrupolar-coupling point of view, are given in Sec. VI.

For cubic symmetry we define macroscopic anisotropy constants in the usual way. Only quartic (or higher) forms in the direction cosines of the magnetization are consistent with cubic symmetry. However, for small displacements from a preferred axis, these forms will have quadratic terms which give rise to effective anisotropy fields.

The form

$$f_A=(K_1/2)(\alpha_1^2\beta_1^2+\alpha_1^2\gamma_1^2+\beta_1^2\gamma_1^2 +\alpha_2^2\beta_2^2+\alpha_2^2\gamma_2^2+\beta_2^2\gamma_2^2),$$

where α_1, β_1, γ_1 and α_2, β_2, γ_2 are the direction cosines of $\mathbf{M}_1$ and $\mathbf{M}_2$ respectively, has a quadratic term for small displacements from the z-axis

$$f_A'\cong(K_1/2)(\alpha_1^2+\beta_1^2+\alpha_2^2+\beta_2^2). \tag{35a}$$

This is analogous to ferromagnetics. In addition, we have in antiferromagnetics the presence of forms depending upon the relative positions of $\mathbf{M}_1$ and $\mathbf{M}_2$, involving terms coupling the direction cosines. For small displacements from the z-axis these can all be reduced to

$$f_A''\cong(K_2/2)(\alpha_1^2+\beta_1^2+\alpha_2^2+\beta_2^2) \tag{35b}$$

and

$$f_A'''\cong K_3(\alpha_1\alpha_2+\beta_1\beta_2). \tag{35c}$$

We may readily identify the H_A used in Secs. II and III:

$$H_AM_1^x=(\partial/\partial\alpha_1)(f_A'+f_A'')=(K_1+K_2)M_1^x/M;$$

and we now have an additional field h_A

$$h_AM_1^x=\partial f_A'''/\partial\alpha_1=K_3M_2^x/M,$$

which adds new terms to the equations of motion (2).

The original equations of motion are restored by redefining

$$H_E'''=H_E+(K_3/M),\quad H_A'''=(K_1+K_2-K_3)/M. \tag{36}$$

The change in H_E may be neglected. Thus all the results of Secs. II and III hold with H_AH_E replaced by $\lambda(K_1+K_2-K_3)$. We have assumed anisotropy fields of the same magnitude acting on the two sublattices, and we have made use of Eq. (25).

As redefined H_A''' cannot become negative as can be seen from the following argument. For small displacements from the z-axis, and with $H_0=0$, the free energy is

$$\begin{aligned}F\cong\lambda M_1M_2[&\alpha_1\alpha_2+\beta_1\beta_2\\ &-(1-\tfrac{1}{2}\alpha_1^2-\tfrac{1}{2}\beta_1^2)(1-\tfrac{1}{2}\alpha_2^2-\tfrac{1}{2}\beta_2^2)]\\ &+[(K_1+K_2)/2](\alpha_1^2+\alpha_2^2+\beta_1^2+\beta_2^2)\\ &+K_3(\alpha_1\alpha_2+\beta_1\beta_2).\end{aligned}$$

This may be written as

$$\begin{aligned}F\cong(\lambda M_1M_2+K_3)[&\alpha_1\alpha_2+\beta_1\beta_2\\ &-(1-\tfrac{1}{2}\alpha_1^2-\tfrac{1}{2}\beta_1^2)(1-\tfrac{1}{2}\alpha_2^2-\tfrac{1}{2}\beta_2^2)]\\ &+[(K_1+K_2-K_3)/2](\alpha_1^2+\alpha_2^2+\beta_1^2+\beta_2^2),\end{aligned}$$

which immediately justifies Eq. (36). In order that F have a minimum at $\alpha_1=\alpha_2=\beta_1=\beta_2=0$, as postulated when we assume z a preferred axis, F must be a "definite" quadratic form. The condition for this is

$$\frac{\partial^2F}{\partial\alpha_1^2}\cdot\frac{\partial^2F}{\partial\alpha_2^2}>\left(\frac{\partial^2F}{\partial\alpha_1\partial\alpha_2}\right)^2$$

or

$$(\lambda M_1M_2+K_1+K_2)^2>(\lambda M_1M_2+K_3)^2,$$

so that

$$K_1+K_2>K_3. \tag{37}$$

Quantum-mechanical expressions for K_1, K_2, and K_3 are given, in terms of quadrupolar coupling constants, in Sec. VI.

B. Temperature Dependence of $H_E'''H_A'''$ ($\cong K_1+K_2-K_3$)

Here we have little to go by. As noted by Van Vleck[11] his calculations for ferromagnetic anisotropy exhibit the general trend with temperature that is observed experimentally, but do not give anywhere near as rapid a diminishment of anisotropy near the Curie point as is observed. In addition, some ferromagnetics, e.g., MnBi, exhibit an *increase* of anisotropy over a wide range of increasing temperature,[12] quite contrary to the predictions of the quadrupolar coupling model. Presumably antiferromagnetics are subject to the same vagaries.

---

[10] These substitutions can also be found by considering the free energy (less anisotropy):

$$F=-H_0(M_1^z+M_2^z)+\lambda\mathbf{M}_1\cdot\mathbf{M}_2+\tfrac{1}{2}N(M_1^x+M_2^x)^2 +\tfrac{1}{2}N(M_1^y+M_2^y)^2+\tfrac{1}{2}(4\pi-2N)(M_1^z+M_2^z)^2;$$

which may be rewritten

$$F=\tfrac{1}{2}N(\mathbf{M}_1^2+\mathbf{M}_2^2)-[H_0+\tfrac{1}{2}(3N-4\pi)(M_1^z+M_2^z)](M_1^z+M_2^z) +(\lambda+N)\mathbf{M}_1\cdot\mathbf{M}_2.$$

[11] J. H. Van Vleck, Phys. Rev. **52**, 1178 (1937).

[12] C. Guillaud, *Ferromagnétisme des alliages binaires de manganèse*, thesis, Strasbourg, 1943.

Assuming, however, that the quadrupolar model is followed, we have from Sec. VI below that the temperature dependent part of $K_1+K_2-K_3$ goes approximately as

$$[S(S+1)-3\langle(S_{1j}{}^z)^2\rangle_{\mathrm{Av}}][\langle S_{1j}{}^z\rangle_{\mathrm{Av}}]^2, \tag{38}$$

where

$$\langle S_{1j}{}^z\rangle_{\mathrm{Av}}=B_1(\theta)=\sum_{m=-S}^{S} me^{m\theta}\Big/\sum_{m=-S}^{S} e^{m\theta},$$

$$\langle(S_{1j}{}^z)^2\rangle_{\mathrm{Av}}=B_2(\theta)=\sum_{m=-S}^{S} m^2e^{m\theta}\Big/\sum_{m=-S}^{S} e^{m\theta},$$

with

$$\theta=g\beta H_0/kT.$$

$B_1(\theta)$ is the Brillouin function, and it is to be noted that

$$B_2(\theta)=[B_1(\theta)]^2+dB_1/d\theta.$$

Further details on this model as applied to ferromagnetics may be found in Van Vleck's papers.[3,11]

C. Noncubic, but Orthorhombic Symmetry

This case includes $CuCl_2\cdot 2H_2O$ which has been investigated at Leiden.[2] The anisotropy may be a quadratic function of the direction cosines without the small angle approximation. We have as the most general quadratic term, after subtracting out the rotational invariant dot product of the magnetization vectors (which acts as an exchange term)

$$f_A=(K_1'/2)(\alpha_1^2+\alpha_2^2)+(K_2'/2)(\beta_1^2+\beta_2^2)+K_3'\alpha_1\alpha_2+K_4'\beta_1\beta_2. \tag{39}$$

We choose the z axis as the preferred (direction of easiest magnetization). Hence, using arguments similar to those leading to Eq. (37) we must have

$$K_1'>K_3' \quad\text{and}\quad K_2'>K_4'. \tag{40}$$

We treat two special cases, and include demagnetizing factors.

(1) 0°K, $H_0=0$

The resonance matrix corresponding to Eq. (3) is

$$\mathbf{A}_2=\begin{vmatrix} 0 & 0 & -ia & -ib \\ 0 & 0 & ib & ia \\ ic & id & 0 & 0 \\ -id & -ic & 0 & 0 \end{vmatrix}, \tag{41}$$

where

$$a=\gamma H_E(1+N^y\lambda^{-1})+\gamma K_2'/M,$$
$$b=\gamma H_E(1+N^y\lambda^{-1})+\gamma K_4'/M,$$
$$c=\gamma H_E(1+N^x\lambda^{-1})+\gamma K_1'/M,$$
$$d=\gamma H_E(1+N^x\lambda^{-1})+\gamma K_3'/M.$$

The diagonalizing matrix becomes

$$\mathbf{S}_2=\begin{vmatrix} 1 & 1 & 1 & 1 \\ 1 & 1 & -1 & -1 \\ i\mu & -i\mu & i\nu^{-1} & -i\nu^{-1} \\ -i\mu & i\mu & i\nu^{-1} & -i\nu^{-1} \end{vmatrix}, \tag{42}$$

where

$$\mu=+[(c+d)/(a-b)]^{\frac{1}{2}},\quad \nu=+[(a+b)/(c-d)]^{\frac{1}{2}}.$$

This is closely analogous to the $\mathbf{S}'$ of Eq. (9). The eigenvalues are

$$\begin{aligned}\omega_1&=-\omega_2=+[(a-b)(c+d)]^{\frac{1}{2}},\\ \omega_3&=-\omega_4=+[(c-d)(a+b)]^{\frac{1}{2}}.\end{aligned} \tag{43}$$

The normal modes P_i are similar to those of Eq. (9) except that the elliptical paths in the xy plane are no longer of arbitrary orientation, and are of different frequencies according to whether the major axes are in the x or in the y directions.

The susceptibilities are interesting. For linearly polarized rf fields

$$\chi^x=-2\gamma M(a-b)/(\omega^2-\omega_1^2),$$
$$\chi^y=-2\gamma M(c-d)/(\omega^2-\omega_3^2).$$

Thus we have the somewhat unusual result that only the resonance frequency ω_1 should be observed with the rf field in the x direction, and only ω_3 with the rf in the y direction. Also, assuming the ω_1 and ω_3 lines to be of similar shape, the ratio of amplitudes should go as $(a-b)/(c-d)$, or as the ratio of the effective anisotropy fields for x and y displacements.

(2) 0°K, H_0 along preferred axis.

The resonance frequencies are

$$\omega^2=\gamma^2H_0^2+ac-bd\pm[(a+c-b-d)(a+b+c+d)H_0^2+(bc-ad)^2]^{\frac{1}{2}}. \tag{44}$$

The normal modes are too complicated to admit of a simple physical interpretation. All frequencies should be observed in both the x and y directions, although with quite disparate intensities as long as H_0 is small.

V. LINE WIDTHS

Powder samples should give according to Eq. (31) a fractional line width $\Delta H_0/H_0\sim 1$. With H_0 fixed and ω varied the broadening will be $\Delta\omega/\omega\sim H_0/(2H_EH_A)^{\frac{1}{2}}$. Since the microcrystals have random demagnetizing factors, there is according to Eq. (34) an additional contribution to the width of $\Delta\omega/\omega\sim 10\lambda^{-1}=10\chi_\perp$. This would vary from sample to sample, being $\sim 10^{-3}$ in MnO and becoming quite large for samples with low Curie points.

Single crystals cut cylindrically, as noted in Sec. III D, are the ideal specimens for studying antiferromagnetic resonance. The crystals should be carefully annealed in order to minimize lattice dislocations. A high density of dislocations may effectively smear out the antiferromagnetic crystal pattern, introducing many extra sets of spins in one or the other direction, and causing widely varying anisotropy and exchange forces. This problem is in some ways peculiar to antiferromagnetics, for in ferromagnetics (a) the spin pattern is not essentially changed by dislocations, and (b) the resonance frequency is not a critical function of the product of exchange and anisotropy fields.

In both powders and crystals there will also be a broadening by various relaxation mechanisms. The dipolar broadening should be small because of the Van Vleck-Gorter process of exchange narrowing[13] which is independent of sign of exchange integral and which is important when the exchange is strong.

VI. QUANTUM-MECHANICAL CALCULATION, CUBIC SYMMETRY

We follow the method used by Van Vleck[3] in treating ferromagnetic resonance. Only the case H_0 along the preferred axis will be treated; the general case follows by using transformations as in Eq. (13). As in the classical treatment, a two sublattice picture is used. We neglect dipole-dipole terms as being relatively unimportant. The anisotropy is treated as a quadrupole-quadrupole coupling. As Van Vleck points out, this is merely an artifice, representing in a simple way the complex interaction among spins, orbits, and lattice. It leaves us with unknown quadrupolar coupling constants, and the best we can do is to identify these with macroscopic counterparts.

The Hamiltonian of the system is the sum of Zeeman, exchange, and quadrupolar terms

$$\mathcal{H}=\mathcal{H}_Z+\mathcal{H}_E+\mathcal{H}_Q. \tag{45}$$

We assume spins S_{1j}^z of sublattice 1 directed opposite to H_0, so that M_1^z is in the direction of H_0. Introducing exchange coupling constants A_{jk} and quadrupolar coupling constants K_{jk} we have

$$\begin{aligned}
\mathcal{H}_Z&=H_0g\beta(\textstyle\sum_j S_{1j}^z+\sum_j S_{2j}^z);\\
\mathcal{H}_E&=\textstyle\sum_{k>j} A_{jk}\mathbf{S}_{1j}\cdot\mathbf{S}_{1k}+\sum_{k>j} A_{jk}'\mathbf{S}_{2j}\cdot\mathbf{S}_{2k}\\
&\qquad+\textstyle\sum_{k,j} A_{jk}''\mathbf{S}_{1j}\cdot\mathbf{S}_{2k};\\
\mathcal{H}_Q&=\textstyle\sum_{k>j} K_{jk}r_{jk}^{-4}(\mathbf{S}_{1j}\cdot\mathbf{r}_{jk})^2(\mathbf{S}_{1k}\cdot\mathbf{r}_{jk})^2\\
&\qquad+\textstyle\sum_{k>j} K_{jk}'r_{jk}^{-4}(\mathbf{S}_{2j}\cdot\mathbf{r}_{jk})^2(\mathbf{S}_{2k}\cdot\mathbf{r}_{jk})^2\\
&\qquad\qquad+\textstyle\sum_{k,j} K_{jk}''r_{jk}^{-4}(\mathbf{S}_{2j}\cdot\mathbf{r}_{jk})^2(\mathbf{S}_{1k}\cdot\mathbf{r}_{jk})^2.
\end{aligned}$$

Using the equations of motion and commutation relations

$$dS_{1j}^x/dt=(i/\hbar)[\mathcal{H},S_{1j}^x];\quad \text{etc.};$$
$$[S_{1j}^x,S_{1k}^y]=iS_{1j}^z\delta_{jk};\quad \text{etc.};$$

we obtain

$$\begin{aligned}
&dS_1^x/dt=\textstyle\sum_j dS_{1j}^x/dt=Z+E+Q;\quad Z=-\gamma H_0S_1^y;\\
&\hbar E=\textstyle\sum_{k,j} A_{jk}''(S_{1j}^zS_{2k}^y-S_{1j}^yS_{2k}^z);\\
&\hbar Q=\textstyle\sum_{k\neq j}K_{jk}(\alpha_{jk}S_{1j}^x+\beta_{jk}S_{1j}^y+\gamma_{jk}S_{1j}^z)^2W_{jk}\\
&\qquad+\textstyle\sum_{k,j} K_{jk}''(\alpha_{jk}S_{2j}^x+\beta_{jk}S_{2j}^y+\gamma_{jk}S_{2j}^z)^2W_{jk};
\end{aligned}\tag{46}$$

where

$$\begin{aligned}
W_{jk}&=(\alpha_{jk}S_{1k}^x+\beta_{jk}S_{1k}^y+\gamma_{jk}S_{1k}^z)(\beta_{jk}S_{1k}^z-\gamma_{jk}S_{1k}^y)\\
&\qquad+(\beta_{jk}S_{1k}^z-\gamma_{jk}S_{1k}^y)(\alpha_{jk}S_{1k}^x+\beta_{jk}S_{1k}^y+\gamma_{jk}S_{1k}^z).
\end{aligned}$$

Here α_{jk}, β_{jk}, γ_{jk} are direction cosines of $\mathbf{r}_{jk}$ relative to the field axes, since the axis of quantization must be along H_0. When H_0 is along a preferred axis we may make the field axes coincide with the crystal axes, which is a great simplification. Similar equations of motion may be obtained for S_2^x, etc.

We make the approximations that below the Curie point we may replace S_{1j}^z and S_{2k}^z by their expectation values:

$$\langle S_{1j}^z\rangle_{\text{Av}}=-2M_1^z/Ng\beta;\quad \langle S_{2k}^z\rangle_{\text{Av}}=-2M_2^z/Ng\beta; \tag{47}$$

where $N/2$ is the number of atoms in a sublattice.

Assuming that local exchange fields can be replaced by average fields taken independently over successive clusters of nearest neighbors, next nearest neighbors, etc., we may define an average exchange parameter λ

$$\lambda=(2/Ng\beta\hbar\gamma)\textstyle\sum_j A_{jk}'' \tag{48}$$

independent of k. Then we have

$$E=\gamma\lambda M_2^zS_1^y-\gamma\lambda M_1^zS_2^y. \tag{49}$$

Note that this is independent of lattice structure.

To simplify Q we replace squares by average values for each atom, assumed independent. Thus we use the relations

$$\begin{aligned}
&\langle(S_{1j}^x)^2\rangle_{\text{Av}}=\langle(S_{1j}^y)^2\rangle_{\text{Av}}=\tfrac{1}{2}[S(S+1)-\langle(S_{1j}^z)^2\rangle_{\text{Av}}];\\
&\quad S_{1j}^xS_{1j}^y+S_{1j}^yS_{1j}^x=0;\\
&\quad \tfrac{1}{2}(S_{1j}^xS_{1j}^z+S_{1j}^zS_{1j}^x)=[\langle S_{1j}^z\rangle_{\text{Av}}+\tfrac{1}{2}]S_{1j}^x;
\end{aligned}$$

with similar relations for S_2. Also we take advantage of cubic symmetry to drop terms like[14]

$$\textstyle\sum_k K_{jk}(\alpha_{jk}^2-\beta_{jk}^2),\quad \sum_k K_{jk}\alpha_{jk}\beta_{jk},\quad \sum_k K_{jk}\alpha_{jk}^3\beta_{jk},\quad \text{etc.}$$

This eventually reduces Q to

$$\begin{aligned}
\hbar Q&=[S(S+1)-3\langle(S_{1j}^z)^2\rangle_{\text{Av}}][\langle S_{1j}^z\rangle_{\text{Av}}+\tfrac{1}{2}]\\
&\qquad\times\textstyle\sum_k K_{jk}(\gamma_{jk}^4-3\beta_{jk}^2\gamma_{jk}^2)S_1^y\\
&\qquad+[S(S+1)-3\langle(S_{2j}^z)^2\rangle_{\text{Av}}][\langle S_{1j}^z\rangle_{\text{Av}}+\tfrac{1}{2}]\\
&\qquad\times\textstyle\sum_k K_{jk}''(\gamma_{jk}^4-\beta_{jk}^2\gamma_{jk}^2)S_1^y\\
&\qquad+[S(S+1)-3\langle(S_{1j}^z)^2\rangle_{\text{Av}}][\langle S_{2j}^z\rangle_{\text{Av}}+\tfrac{1}{2}]\\
&\qquad\qquad\times\textstyle\sum_k K_{jk}''(-2\beta_{jk}^2\gamma_{jk}^2)S_2^y.
\end{aligned}$$

An assumption equivalent to the classical assumption of equal anisotropy fields seen by both sublattices is that in the above equation

$$\langle(S_{1j}^z)^2\rangle_{\text{Av}}=\langle(S_{2j}^z)^2\rangle_{\text{Av}};\quad [\langle S_{1j}^z\rangle_{\text{Av}}+\tfrac{1}{2}]=-[\langle S_{2j}^z\rangle_{\text{Av}}+\tfrac{1}{2}];$$

and now defining

$$\gamma K_1/M=\hbar^{-1}q\textstyle\sum_k K_{jk}(\gamma_{jk}^4-3\beta_{jk}^2\gamma_{jk}^2); \tag{50a}$$

$$\gamma K_2/M=\hbar^{-1}q\textstyle\sum_k K_{jk}''(\gamma_{jk}^4-\beta_{jk}^2\gamma_{jk}^2); \tag{50b}$$

$$\gamma K_3/M=\hbar^{-1}q\textstyle\sum_k K_{jk}''(2\beta_{jk}^2\gamma_{jk}^2); \tag{50c}$$

---

[13] J. H. Van Vleck, Phys. Rev. 74, 1168 (1948).

[14] Strictly, these will not drop out unless we have cubic symmetry within each of the two magnetic sublattices considered. Shull's experiments (see reference 6) indicate there is no such symmetry in, e.g., MnO. However, there is, of course, symmetry within each of the eight actual sublattices of a f.c.c., and if we make the reasonable assumption that the anisotropy is associated with the super-exchange, the terms in question will go to zero.

where

$$q=-[S(S+1)-3\langle(S_{1j}^z)^2\rangle_{\mathrm{Av}}][\langle S_{1j}^z\rangle_{\mathrm{Av}}+\tfrac{1}{2}],$$

we have

$$Q=-\gamma[(K_1+K_2)/M]S_1^y-\gamma(K_3/M)S_2^y.$$

Thus Eq. (46) has become

$$dS_1^x/dt=-\gamma[H_0-\lambda M_2^z+(K_1+K_2)/M]S_1^y \\ -\gamma[\lambda M_1^z+(K_3/M)]S_2^y; \quad (51)$$

with similar equations in S_2^x, etc. Equation (51) is the same as Eq. (2) (in absence of rf field **H**) when account is taken of the anisotropy refinements of Sec. IV A. Thus we have shown that the quantum-mechanical and classical approaches lead to the same differential equations of motion, and hence to the same resonance frequencies.

It is noted that for a simple cubic lattice Eq. (50c) gives $K_3/M=0$. For more complicated lattices Eq. (50) ought not to be taken too seriously, as witness reference 14.

VII. EXPERIMENTAL SITUATION

A sudden and almost complete extinction of paramagnetic resonance absorption at microwave frequencies on cooling an antiferromagnetic below the Curie point has been noticed by several investigators. Using a single crystal of the tetragonal salt MnF_2 ($T_c \simeq 70$°K) in a field $H_0 \simeq 3000$ oersteds, Hutchison[15] has found the absorption to vanish at 64°K, independent of the orientation of H_0. The same effect has been noticed by Maxwell *et al.*,[16] using sintered Cr_2O_3 ($T_c \simeq 311$°K) in a field $H_0=3450$ oersteds.

On the other hand a continuation of strong paramagnetic absorption well below the Curie point in thin disk polycrystalline samples of the cubic compounds MnS, MnO, and MnSe has been reported from Japan.[17] In particular as MnS ($T_c \simeq 198$°K) was cooled to 78°K the resonance line at 3510 oersteds decreased quite smoothly in amplitude and increased in width. A similar result, but with the intensity more sharply reduced below the Curie point, has been found in powdered Cr_2O_3 by Maxwell *et al.*[16] The latter experiment also showed a second line below the Curie point appearing at a smaller and smaller value of H_0 (fixed ω) as the temperature decreased. It may be that these lines are caused by the presence of ferromagnetic impurities in the lattice.

The antiferromagnetic resonance discussed in this paper probably would not have been observed in any of the published experiments, because they were carried out rather far from the expected fields and frequencies. Antiferromagnetic resonance has apparently been observed at Leiden[2] in the experiment with $CuCl_2 \cdot 2H_2O$ mentioned in Sec. I. It would be of interest if experiments could be done with other crystals of very low Curie points, or with some of the above-mentioned crystals in fields $H_0 \sim 5\times10^4$ oersteds.

ACKNOWLEDGMENTS

This research has been assisted in part by the ONR. Part of it was done while one of us (F. K.) held a Whiting Fellowship. We wish to thank Mr. H. Kaplan for checking the derivations. We also are very indebted to Professor C. A. Hutchison and Dr. L. R. Maxwell for several communications regarding their experiments.

---

[15] C. A. Hutchison (private communication).

[16] Trounson, Bleil, Wangsness, and Maxwell, Phys. Rev. **79**, 542 (1950).

[17] Okamura, Torizuka, and Kojima, Phys. Rev. **82**, 285 (1951).