# Concerning the Theory of Ferromagnetic Resonance Absorption

J. H. VAN VLECK Harvard University, Cambridge, Massachusetts (Received January 5, 1950)

The first part of this paper gives a simple quantum-mechanical derivation of Kittel's formula for the resonance frequency of ferromagnetic materials. The interactions between the elementary dipoles are handled directly, rather than through the ad hoc introduction of macroscopic demagnetization factors, as is usually done. In Section 3, Kittel's corrections for the effect of anisotropy on frequency are derived from the microscopic standpoint with a model having quadrupolar coupling between atoms. Section 4 discusses qualitatively the effect of exchange narrowing on the line-width. In Section 5 it is shown that Kittel's relation g-2=2-g' connecting the spectroscopic splitting factor g and the gyromagnetic ratio g' is a general consequence of first-order perturbation theory. Throughout the paper it is stressed that ferromagnetic bodies may have important short-range forces of dipolar structure which arise from anisotropic exchange rather than from true magnetic coupling. It is shown that in the first approximation, inclusion of these anomalous forces does not modify Kittel's results.

## 1. INTRODUCTION

N interesting development in microwave spectro-A scopy is the discovery of sharp resonance lines in ferromagnetic bodies in the presence of a constant magnetic field perpendicular to the oscillating or "measuring" field. At first thought, one would expect the resonance to occur at the Larmor frequency  $gHe/4\pi mc$ , provided, of course, that one includes the Landé factor g in defining this frequency. Actually, the resonance frequency depends on the shape of the specimen and involves the demagnetizing corrections. Kittel<sup>1</sup> shows that if the constant field is applied along the z axis, and the measuring field along the x direction, as we suppose throughout the paper, then the resonance frequency is

$$\nu = \frac{ge}{4\pi mc} \{ [H + (N_y - N_z)M] [H + (N_x - N_z)M] \}^{\frac{1}{2}}, \quad (1)$$

where M is the z component of the intensity of magnetization, and where  $N_x$ ,  $N_y$ ,  $N_z$  are respectively the demagnetizing corrections for fields applied in the x, y, and z directions. The case most commonly investigated is that in which the specimen is much thinner in the xthan in the y and z directions. Then (1) reduces to the simple and by now well-known formula

$$\nu = (ge/4\pi mc)(BH)^{\frac{1}{2}}$$

In evaluating the shape of the specimen, allowance must be made for any diminution in the effective size which results from limitation of penetration by the skin effect. The direction perpendicular to the wall of a wave guide is thus usually to be classed as short even though the material may extend indefinitely in this direction.

Kittel derived his fundamental formula, our Eq. (1), by a very ingenious method based on classical theory. Although his type of argument is such that one expects it to hold also in quantum theory, nevertheless, it is desirable to have a quantum-mechanical derivation of (1). The quantum eigenvalues for the entire specimen regarded as a single unit have been obtained by Luttinger and Kittel,<sup>2</sup> and by Richardson.<sup>3</sup> Their resulting derivation of (1), however, still utilizes the classical macroscopic theory of demagnetizing corrections. Polder<sup>4</sup> has given a general formal proof of the equivalence of the classical and quantum-mechanical results, so that there is no loss of rigor in using classical demagnetizing fields. Any approach of this character is, however, a little devious. In Section 2 we shall give a simple quantum-mechanical proof of (1) in which the demagnetizing corrections follow as a direct consequence of the inclusion of the interaction between the elementary dipoles.

## 2. DERIVATION OF KITTEL'S FORMULA

For our purpose any terms can be dropped from the Hamiltonian function which do not depend on spin orientation. The Hamiltonian matrix can hence be taken to be

$$\mathcal{K} = Hg\beta\sum_{j} S_{zj} + \sum_{k>j} A_{jk} \mathbf{S}_{j} \cdot \mathbf{S}_{k} + \sum_{k>j} D_{jk} [\mathbf{S}_{j} \cdot \mathbf{S}_{k} - 3r_{jk}^{-2} (\mathbf{r}_{jk} \cdot \mathbf{S}_{j}) (\mathbf{r}_{jk} \cdot \mathbf{S}_{k})],$$
(2)

where  $\beta$  is the Bohr magneton  $he/4\pi mc$ , and  $\mathbf{S}_{i}$  is the spin moment vector of atom *j*, measured in multiples of the quantum unit  $h/2\pi$ . The first right-hand member of (2) is the Zeeman energy associated with the constant magnetic field applied along the z axis. It is unnecessary for us to include explicitly the energy associated with the alternating magnetic field directed along the x axis, inasmuch as the method of moments which we use evaluates

<sup>&</sup>lt;sup>1</sup> C. Kittel, Phys. Rev. **71**, 270 (1947); **73**, 155 (1948). <sup>2</sup> J. M. Luttinger and C. Kittel, Helv. Phys. Acta **21**, 480 (1948). <sup>3</sup> J. M. Richardson, Phys. Rev. **75**, 1630 (1949). <sup>4</sup> D. Polder, Phil. Mag. **11**, 99 (1949).

the resonance frequencies directly without requiring examination of the work performed by the alternating field. The second member of (2) is the spin-dependent part of the exchange energy, and, except for a constant factor, the coefficient  $A_{jk}$  is identical with the exchange integral. The third member of (2) embodies the ordinary dipolar interaction, i.e., the mutual magnetic energy of the spins, and in addition any other coupling which has a dipolelike structure as regards dependence on spin alignment. With conventional dipolar coupling, the value of the coefficient  $D_{jk}$  is

$$D_{jk} = g^2 \beta^2 / r_{jk}^3.$$
 (3)

Actually, however, the interplay between spin-orbit interaction and orbital valence may give an energy of dipolar type as regards dependence on spin alignment, an effect sometimes called anisotropic exchange coupling. In other words, in the language of nuclear physicists, there may be "tensor" forces not of purely magnetic origin. This non-magnetic, pseudo-dipolar coupling (which is closely related to what is called rho-type tripling in the spectra of diatomic molecules) was discussed in considerable detail in a paper<sup>5</sup> of the author on ferromagnetic anisotropy, and so will not be elaborated here. Suffice it to say that such coupling is of comparatively short range. Thus the deviations of  $D_{jk}$  from the value (3) may be expected to be important only when atoms j and k are adjacent, or nearly so. In a previous article,<sup>6</sup> henceforth referred to as *l.c.*, in which we treated the width of paramagnetic (not ferromagnetic) resonance lines, we assumed that the coefficient of the term of dipolar structure had the normal, purely magnetic value (3). This supposition was quite warranted in the case of nuclear paramagnetism, but may not be in that of electronic paramagnetism if the magnetic ions are closely spaced. In the latter event, the mean square line-breadth may be higher than that calculated on the basis of purely magnetic coupling, and correspondingly there will be an increase in the amount of "exchange narrowing" (a term explained in l.c.) which is needed to explain the sharpness of paramagnetic resonance lines.

The first part of the term of (2) involving  $D_{jk}$  merely introduces a coupling of the same structure, viz. a scalar product, as that involved in the exchange member of (2), and hence may be omitted if the coefficient  $A_{jk}$  is redefined. This we henceforth tacitly suppose done.

The equations of motion

$$d\mathbf{S}_j/dt = (2\pi i/h) [ \Im(\mathbf{S}_j - \mathbf{S}_j \Im) ]$$
<sup>(4)</sup>

along with the commutation relations

$$S_{x_j}S_{y_k} - S_{y_k}S_{x_j} = i\delta_k{}^j S_{z_j} \quad \text{etc.}$$
<sup>(5)</sup>

give immediately

$$(h/2\pi)S_x = -Hg\beta S_y + 3\sum_{k \neq j} D_{jk}(\alpha_{jk}S_{xj} + \beta_{jk}S_{yj} + \gamma_{jk}S_{zj})(-\beta_{jk}S_{zk} + \gamma_{jk}S_{yk})$$
(6a)

$$(h/2\pi)\hat{S}_y = Hg\beta S_x + 3\sum_{k\neq j} D_{jk}(\alpha_{jk}S_{xj} + \beta_{jk}S_{yj} + \gamma_{jk}S_{zj})(\alpha_{jk}S_{zk} - \gamma_{jk}S_{xk})$$
(6b)

$$(h/2\pi)\dot{S}_{z} = 3\sum_{k \neq j} D_{jk}(\alpha_{jk}S_{xj} + \beta_{jk}S_{yj} + \gamma_{jk}S_{zj})(-\alpha_{jk}S_{yk} + \beta_{jk}S_{xk}).$$
(6c)

Here S denotes the total spin vector  $\sum_{j} S_{j}$ , and  $\alpha_{jk}$ ,  $\beta_{jk}$ ,  $\gamma_{jk}$  are the direction cosines of  $r_{jk}$ . We throughout suppose the wave-length long compared to the effective extension of the sample in the direction of propagation, so that the important vector for the absorption is the sum  $S_x = \sum_j S_{xj}$  of the x components of the individual atomic spins.

We now proceed to make certain approximations which are usually well warranted, and whose physical meaning is clarified to a considerable extent by the calculation itself. We omit from (6) all terms involving products  $S_{x_i}S_{x_k}$ .  $S_{x_i}S_{y_k}, S_{y_i}S_{y_k}$ . This omission can be justified in either of two counts. In the first place, in ferromagnetic media, the material will be magnetized practically to saturation in the z direction, and so the matrix elements of  $S_{xj}$  or  $S_{uj}$  will be small compared with those of  $S_{zj}$ . Also, quite irrespective of magnitude, the effect of the omitted terms is to introduce satellite lines falling in a different frequency region than that in which we are interested.<sup>7</sup> These satellites correspond to harmonics of the frequency (1), and will be faint compared with the fundamental.

In the remaining terms, involving  $S_{x_j}S_{z_k}$  or  $S_{y_j}S_{z_k}$ , we replace  $S_{z_k}$  (or  $S_{z_j}$ ) by its mean value<sup>8</sup>  $-M/g\beta N$ . Here M is the macroscopic intensity of magnetization, and N the number of atoms per unit volume. The effect of including a correction for the difference  $[S_{z_j} - (-M/g\beta N)]$  would be to introduce oscillations which are of importance for the width of the line, but not its central frequency.

With the approximations explained in the two preceding paragraphs, Eqs. (6) become

$$(h/2\pi)\hat{S}_x = -Hg\beta S_y - 3(M/g\beta N)\sum_{k\neq j} D_{jk} [-\alpha_{jk}\beta_{jk}S_{xj} + \beta_{jk}\gamma_{jk}(M/g\beta N) + (\gamma_{jk}^2 - \beta_{jk}^2)S_{yj}],$$
(7a)

$$(h/2\pi)\hat{S}_{y} = Hg\beta S_{x} - 3(M/g\beta N)\sum_{k \neq j} D_{jk} [\alpha_{jk}\gamma_{jk}(-M/g\beta N) + \alpha_{jk}\beta_{jk}S_{yj} + (\alpha_{jk}^{2} - \gamma_{jk}^{2})S_{xj}],$$
(7b)

$$(h/2\pi)\dot{S}_{z} = 3(M/g\beta N)\sum_{k\neq j} D_{jk}(\alpha_{jk}\gamma_{jk}S_{kj} - \beta_{jk}\gamma_{jk}S_{kj}).$$
(7c)

<sup>5</sup> J. H. Van Vleck, Phys. Rev. 52, 1178 (1937). <sup>6</sup> J. H. Van Vleck, Phys. Rev. 74, 1168 (1948).

<sup>9</sup> For further details on the satellites see reference 6, p. 1170. <sup>8</sup> The minus sign appears here because the ratio of magnetic moment to angular momentum is negative for electron spin. The state of lowest energy is consequently that in which the spin angular momentum is antiparallel to the field.

Now if there are planes of symmetry at each atom, sums involving products of different direction cosines will vanish. Thus we have

$$\sum_{k} \alpha_{jk} \beta_{jk} D_{jk} = 0, \quad \text{etc.} \tag{8}$$

(If there are not such symmetry planes, sums such as (8) will alternate in sign as one goes from one lattice point to another, and when the summation over j is performed, the effect is substantially the same as though (8) were satisfied, at least as far as the short-range forces are concerned.)

The classical demagnetization factor  $N_x$  in the x direction has the value<sup>9</sup>

$$N_{x} = N^{-1} \sum_{k} \left[ 1 - 3\alpha_{jk}^{2} \right] r_{jk}^{-3} + (4\pi/3), \qquad (9)$$

where N is the number of atoms per unit volume. This formula presupposes that the sum in (9) is independent of j, or in other words that the concept of a uniform demagnetization factor for the specimen is valid. A similar assumption is also necessary to make the longrange forces satisfy (8). Only an ellipsoidally cut body fulfills this criterion, and the Kittel formula (1) applies rigorously only for this shape.

If the coefficients  $D_{jk}$  have the "normal" or classical values (3), the non-vanishing sums in (7) can be evaluated by means of (9). Even if there are anomalous short-range forces which add a correction term to (3)for closely spaced atoms, nevertheless these corrections will disappear if the atoms are spaced with cubic symmetry, for then the anomalous contributions will cancel out of differences such as  $\sum_{k} (\gamma_{jk}^2 - \beta_{jk}^2) D_{jk}$ which are involved in (7). Fortunately, the common ferromagnetic materials, e.g., iron or nickel, have cubic lattices.<sup>10</sup> It is not necessary that the body be cut in a cube, and it is, in fact, the deviations from cubic or spherical symmetry which make sums of type in (9)different from zero, and the demagnetization factors different from  $4\pi/3$ . Boundary effects, which are evidenced by the usual demagnetizating fields, are not influenced by anomalous short-range forces of dipolar structure, since these forces fall off much more rapidly than those caused by true magnetic coupling, and so give rise only to sums which converge much more rapidly than the inverse cube type entering in (9). If we use (3), (8), and (9), the double sums in (7)

reduce to single ones over j which can immediately be simplified by using the relations  $S_{y} = \sum_{j} S_{yj}$  etc. Thus Eqs. (7) become

$$(h/2\pi)\dot{S}_{x} = -g\beta HS_{y} + (N_{z} - N_{y})g\beta MS_{y}, \qquad (10a)$$
  
$$(h/2\pi)\dot{S}_{y} = g\beta HS_{x} - (N_{z} - N_{x})g\beta MS_{x}, \quad \dot{S}_{z} = 0. \quad (10b, c)$$

These are precisely Kittel's equations. To solve them, we set

$$S_x = S_{x_0} \exp(2\pi i\nu t), \quad S_y = S_{y_0} \exp(-2\pi i\nu t).$$

Then (10a) and (10b) yield a pair of simultaneous homogeneous linear equations in the amplitudes  $S_{x_0}, S_{y_0}$ . For a non-trivial solution, the determinant of the coefficients must vanish, and so one obtains the standard expression (1) for the resonance frequency.

#### 3. EFFECT OF ANISOTROPY ON THE **RESONANCE FREQUENCY**

Kittel has considered how the resonance frequency is influenced by the addition of an anisotropy energy macroscopically of the form

$$E = K_1(\lambda_z^2 \mu_z^2 + \mu_z^2 \nu_z^2 + \lambda_z^2 \nu_z^2), \qquad (11)$$

where  $\lambda_z$ ,  $\mu_z$ ,  $\nu_z$  are the direction cosines of the magnetization relative to the principal cubic axes. Kittel handles this anisotropic term by showing that its effect can be included by proper alteration of the demagnetization factors to be used in (1).

We will derive Kittel's anisotropic corrections in a different fashion. He does not investigate the microscopic origin of the energy of anisotropy. We will instead start with an explicit microscopic potential. As our potential is somewhat special in character, our calculation is less general than Kittel's. Also it is more complicated. Nevertheless, despite the power and simplicity of the macroscopic method, it is perhaps worth while to give our microscopic derivation of Kittel's results since it starts more basically and since it reveals somewhat more clearly the approximations inherent in obtaining the final formulae.

The simplest model of anisotropy is that in which there is a coupling between spins of the quadrupolar type

$$\sum_{k>j} K_{jk} r_{jk}^{-4} (\mathbf{S}_j \cdot \mathbf{r}_{jk})^2 (\mathbf{S}_k \cdot \mathbf{r}_{jk})^2.$$
(12)

The addition of (12) to the Hamiltonian (2) adds the following member to the right side of Eq. (6a):

$$-\sum_{k\neq j} K_{jk} (\alpha_{jk} S_{xj} + \beta_{jk} S_{yj} + \gamma_{jk} S_{zj})^{2} [(\alpha_{jk} S_{xk} + \beta_{jk} S_{yk} + \gamma_{jk} S_{zk}) (-\beta_{jk} S_{zk} + \gamma_{jk} S_{yk}) + (-\beta_{jk} S_{zk} + \gamma_{jk} S_{yk}) (\alpha_{jk} S_{xk} + \beta_{jk} S_{yk} + \gamma_{jk} S_{zk})]. \quad (13a)$$

There are corresponding additional members (13b), (13c), which we shall not write down, for the remaining equations of motion (6b), (6c).

We now proceed to simplify the expressions (13) as follows. We replace each term which involves the square

<sup>&</sup>lt;sup>9</sup>See, for instance, J. H. Van Vleck, J. Chem. Phys. 5, 326 and 327 (1937). <sup>10</sup> In non-cubic crystals such as cobalt, the effect of anomalous short-range forces does not drop out because of symmetry con-siderations. Then these forces presumably have an important influence on the resonance frequency. They also, however, give rise to anisotropy. Presumably their contribution to the frequency is indirectly included, at least approximately, in any macroscopic theory giving the correction to the resonance frequency caused by the phenomenological energy of anisotropy in the non-cubic case.

of a spin component by its average value, and assume that the averages can be taken over the different atoms independently. We suppose that the quantization is dominantly with respect to the direction of the applied magnetic field, and that any distortion caused by demagnetization corrections or anisotropy is subordinate. The averages are then symmetrical around the z axis and we have

$$\langle S_{x_j}^2 \rangle_{\mathsf{AV}} = \langle S_{y_j}^2 \rangle_{\mathsf{AV}} = \frac{1}{2} [S(S+1) - \langle S_{z_j}^2 \rangle_{\mathsf{AV}}].$$
<sup>(14)</sup>

We can take

$$S_{x_j}S_{y_j} + S_{y_j}S_{x_j} = 0 \tag{15}$$

since this type of product is small in value and since also it averages to zero if there is symmetry about the z axis. Unless the spin quantum number of an individual atom is very large, it is an inadequate approximation to replace  $\frac{1}{2}(S_{x_j}S_{z_j}+S_{z_j}S_{x_j})$  by  $(-M/Ng\beta)S_{x_j}$ , one's first guess. Instead it is necessary to use the better approximation

$$\frac{1}{2}(S_{xj}S_{zj}+S_{zj}S_{xj})=\left[\langle S_{zj}\rangle_{kv}+\frac{1}{2}\right]S_{xj}=\left[(-M/Ng\beta)+\frac{1}{2}\right]S_{xj}.$$
(16)

The justification of (16) is that if one is close to saturation the important switches in the magnetic quantum number of an atom are those between state  $S_z = -S$  of alignment of magnetic moment parallel (i.e. spin angular momentum<sup>8</sup> antiparallel) to the field, and the next most parallel state  $S_{2j} = -S + 1$ . In other words, the effect is more or less<sup>11</sup> neglected of transitions to states having  $S_{z_j} = -S+2$ , -S+3, etc. An analogous approximation is surreptitiously made, for instance, in Moller's extension<sup>12</sup> to the case  $S > \frac{1}{2}$  of Bloch's spin wave treatment of magnetism at low temperatures. Since it is necessary to suppose that one is close to saturation, the assumptions which we are making in connection with the anisotropic corrections are probably more drastic than those we employed in Section 2 to obtain Kittel's formula for the isotropic case.

When we use (14), (15), and (16) (along with a relation identical with (16) except that x is replaced by y), the expression (13a) becomes

$$2[\frac{1}{2}S(S+1) - \frac{3}{2}\langle S_{z_{j}}^{2}\rangle_{k_{v}}][(-M/g\beta N) + \frac{1}{2}][S_{v}\sum_{k}K_{jk}(\gamma_{jk}^{4} - 3\beta_{jk}^{2}\gamma_{jk}^{2}) + 3S_{x}\sum_{k}K_{jk}(\alpha_{jk}^{3}\beta_{jk} + \beta_{jk}^{3}\alpha_{jk})] + 2[\frac{1}{2}S(S+1) - \frac{3}{2}\langle S_{z_{j}}^{2}\rangle_{k_{v}}]^{2}\sum_{k}K_{jk}\gamma_{jk}^{3}\beta_{jk}.$$
 (17a)

In writing (17a), we have utilized simplifications afforded by the fact that sums of the form  $\sum_k K_{ik} (\alpha_{ik}^2 - \beta_{ik}^2)$ or  $\sum_{k} K_{jk} \alpha_{jk} \beta_{jk}$  must vanish for a cubic crystal, since a quadratic dependence on direction cosines is incompatible with cubic symmetry. Also we have assumed the atoms similarly situated, so that the sums over k are independent of j.

The direction cosines  $\alpha_{jk}$ ,  $\beta_{jk}$ ,  $\gamma_{jk}$  which we have used so far are those of  $\mathbf{r}_{jk}$  relative to a set of axes x, y, z such that the applied field H is along z and the oscillating field along x. We must now express these direction cosines in terms of those  $\lambda_{jk}$ ,  $\mu_{jk}$ ,  $\nu_{jk}$  of  $\mathbf{r}_{jk}$  relative to the principal cubic axes X, Y, Z, together with the direction cosines connecting the x, y, z and X, Y, Z systems. We shall use the notation  $\lambda_x$ ,  $\mu_x$ ,  $\nu_x$  for the direction cosines connecting the x axis with the X, Y, Z ones, with an analogous significance for  $\lambda_y$ ,  $\mu_y$ ,  $\nu_y$  etc. Now because of the equivalence of the principal cubic axes, there are relations of the form

$$\sum_{k} K_{jk}(\gamma_{jk}^{4} - 3\beta_{jk}^{2}\gamma_{jk}^{2}) = \frac{1}{3}\sum_{k} K_{jk}[\lambda_{z}^{4} + \mu_{z}^{4} + \nu_{z}^{4} - 3\lambda_{z}^{2}\lambda_{y}^{2} - 3\mu_{y}^{2}\mu_{z}^{2} - 3\nu_{y}^{2}\nu_{z}^{2}][1 - 5(\lambda_{jk}^{2}\mu_{jk}^{2} + \mu_{jk}^{2}\nu_{jk}^{2} + \nu_{jk}^{2}\lambda_{jk}^{2}],$$

$$\sum_{k} K_{jk}\beta_{jk}\gamma_{jk}^{3} = \frac{1}{3}\sum_{k} K_{jk}[\lambda_{y}\lambda_{z}^{3} + \mu_{y}\mu_{z}^{3} + \nu_{y}\nu_{z}^{3}][1 - 5(\lambda_{jk}^{2}\mu_{jk}^{2} + \mu_{jk}^{2}\nu_{jk}^{2} + \nu_{jk}^{2}\lambda_{jk}^{2})].$$

If we use these relations and introduce the abbreviations

$$\begin{split} f(a, b) &= \lambda_a^4 + \mu_a^4 + \nu_a^4 - 3(\lambda_b^2 \lambda_a^2 + \mu_b^2 \mu_a^2 + \nu_b^2 \nu_a^2), \quad g(a, b) = \lambda_b \lambda_a^3 + \mu_b \mu_a^3 + \nu_b \nu_a^3, \quad (a, b = x, y, z). \\ Q &= \frac{2}{3} \sum_k K_{jk} \Big[ 1 - 5(\lambda_{jk}^2 \mu_{jk}^2 + \mu_{jk}^2 \nu_{jk}^2 + \nu_{jk}^2 \lambda_{jk}^2) \Big] \Big[ \frac{1}{2} S(S+1) - \frac{3}{2} \langle S_{zj}^2 \rangle_{kv} \Big] \Big[ (-M/Ng\beta) + \frac{1}{2} \Big], \end{split}$$
(18)

$$w = \left[\frac{1}{2}S(S+1) - \frac{3}{2}\langle S_{z_j}^2 \rangle_{\text{AV}}\right] / \left[(-M/g\beta N) + \frac{1}{2}\right],\tag{19}$$

the expression (17a) becomes

$$Qf(z, y)S_y + 3Q[g(x, y) + g(y, x)]S_x + Qg(z, y)w.$$
 (20a)

The corresponding expressions added to the y and z equations of motion are

$$-Qf(z, x)S_{x} - 3Q[g(x, y) + g(y, x)]S_{y} - Qg(z, x)w, Qg(z, x)S_{y} - Qg(z, y)S_{x}.$$
(20b, c)

When (20a), (20b), (20c) are added to the right sides of Eqs. (10a), (10b), (10c) respectively, it is seen that the

<sup>&</sup>lt;sup>11</sup> The relation (16) is rigorously correct if the spins are aligned completely antiparallel (see reference 8) to the field, so that the expectation value of  $Sz_i$  is -S. If, instead,  $\langle Sz_j \rangle_{AV} - (-S)$  is not identically zero, but small compared with S, use of the familiar relation  $(Sx_j \pm iSy_j)(M_S; M_S \mp 1) = [S(S+1) - M_S(M_S \mp 1)]^{\frac{1}{2}}$  shows that the matrix amplitudes connecting a state of given  $Sz_i$  with  $Sz_i + 1$ will be considerably larger than those connecting it with  $S_{z_i}-1$ . Hence (16) is a somewhat better approximation for the case of incomplete alignment than might appear at first sight. <sup>12</sup> C. Moller, Zeits. f. Physik 82, 559 (1933).

equations of motion are of the type form

$$\dot{S}_x = aS_x + bS_y + c_1, \quad \dot{S}_y = cS_x - aS_y + c_2, \quad \dot{S}_z = w^{-1}[-c_1S_x - c_2S_y].$$
 (21a, b, c)

The presence of the terms  $c_1$ ,  $c_2$  in (21a), (21b) is caused by the fact that when one of the principal cubic axes does not coincide with the direction of the applied field, the equilibrium position for the direction of magnetization is not the same as that of the applied field. These  $c_1$ ,  $c_2$  terms can be effectively eliminated by shifting the location of the z axis. To first powers in  $c_1$ ,  $c_2$ , the direction cosines of the angles which the new z axis makes with the original x and y axes are respectively

$$\frac{(-ac_1-bc_2)}{(-M/Ng\beta)(a^2+bc)} \quad \text{and} \quad \frac{(-cc_1+ac_2)}{(-M/Ng\beta)(a^2+bc)}$$

If second powers of  $c_1$  and  $c_2$  are disregarded, this rotation makes the equations of motion in the new, i.e. slightly rotated coordinate system the same as in the old except that all terms in  $c_1$ ,  $c_2$  are entirely suppressed.

In virtue of the preceding paragraph we can set  $c_1 = c_2 = 0$ . The roots of the secular equation are  $\pm i(bc+a^2)^{\frac{1}{2}}$ . The resonance frequency is therefore

$$\nu = g\beta h^{-1} \{ [H + (N_y - N_z)M - Qg^{-1}\beta^{-1}f(z,y)] [H + (N_x - N_z)M - Qg^{-1}\beta^{-1}f(z,x)] + 9g^{-2}\beta^{-2}Q^2 [g(x,y) + g(y,x)]^2 \}^{\frac{1}{2}}.$$
(22)

The constant Q involved in (22) is intimately connected with the proportionality factor  $K_1$  appearing in the energy of anisotropy (11). In an earlier paper the writer calculated the amount of magnetic anisotropy to be expected from the present model. This calculation showed that if there is quadrupolar coupling of our type (12), and if Q and w are defined as in (18) and (19), then the relation connecting  $K_1$  and Q is<sup>13</sup>

$$Q = -2K_1/Nw. \tag{23}$$

If the atomic dipoles are nearly all parallel to the applied field, then  $\langle S_z^2 \rangle_{AV}$  is very nearly equal to  $S^2$ , where S is the spin quantum number of an atom. Also then M is practically the same as  $-Ng\beta S$ . Under these circumstances (23) reduces to

$$Q = -2K_1 g\beta/M.$$
(24)

The formula (22) for the resonance frequency, as it stands, is rather cumbersome, because we have assumed that the constant magnetic field is arbitrarily oriented relative to the principal cubic axes. Our calculation is consequently somewhat more general than Kittel's. He assumed that both the applied and measuring magnetic fields are in the 001 plane, so that in our notation  $\lambda_y = \mu_y = \nu_z = \nu_x = 0$ . With this specialization (22) becomes

$$\nu = g\beta h^{-1} \{ [H + (N_y - N_z)M + 2K_1M^{-1}(\frac{3}{4} + \frac{1}{4}\cos4\theta)] [H + (N_x - N_z)M + 2K_1M^{-1}\cos4\theta] \}^{\frac{1}{2}}.$$
 (25)

where  $\theta$  is the angle between the constant magnetic field and the 100 axis. Equation (25) agrees exactly with the corresponding formula of Kittel, after correction of the latter for a minor algebraic error<sup>14</sup> which, incidentally, was detected by comparison of his calculation and ours. For the case that the x and z axes are in the 011 plane, Eq. (22) reduces to an expression given by L. R. Bickford.<sup>\*</sup>

#### 4. EFFECT OF DIPOLAR INTERACTION ON THE WIDTH OF THE ABSORPTION LINES

We now turn to the subject of the width of the microwave resonance lines in ferromagnetic media. Four possible mechanisms of line-broadening are (a) interatomic forces of dipolar structure (b) spin-lattice interaction (c) damping caused by the eddy currents involved in the skin effect and (d) dependence of the anisotropy corrections in Eq. (22) on the orientation of the magnetic field relative to the principal cubic axes. We will be concerned primarily with (a). As a matter of fact, (d) is not a true broadening at all, but gives an

apparent line-width in powders because the specimen is composed of differently oriented microcrystals whose resonance frequencies consequently do not quite coincide. Thus (d) is inoperative in a single crystal. Also, even in a powder, it is negligible in a substance such as supermalloy whose anisotropy is very small. The importance of (b) is a mooted question. Akhieser<sup>15</sup> actually obtains line-widths due to (b) which are comparable with, or even greater than, the observed widths, but his calculation has been criticized by Polder<sup>4</sup> on the ground that the spin-waves important for resonance are not as short as supposed by Akhieser. Consequently (b) may be less important than Akhieser's results would imply.

If perturbations by eddy currents are neglected, ordinary, i.e., isotropic exchange coupling does not

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<sup>&</sup>lt;sup>13</sup> J. H. Van Vleck, Phys. Rev. 52, 1195 (1937), especially Eqs. (38), (42), and (43). The quantity  $B_2$  entering in Eq. (42) is the same as  $\langle S_{2j}^2 \rangle_{Av}$  in the present model. The following error is to be noted in Eq. (38); it should contain a factor  $\frac{1}{3}$  multiplying  $\Omega_S$ .

<sup>&</sup>lt;sup>14</sup> See reference 8 of C. Kittel, Phys. Rev. 76, 743 (1949).

<sup>\*</sup> L. R. Bickford, Phys. Rev. (to be published).

<sup>&</sup>lt;sup>15</sup> A. Akhieser, J. Phys. U.S.S.R. 10, 217 (1946).

broaden the resonance line, since this type of interaction commutes with any component of the total magnetic moment, and so does not spoil the latter's constancy. The simplest kind of non-isotropic potential is that of dipolar structure. Quadrupoles and other higher order poles would be more complicated to treat and would presumably not give any materially different results. It is considerably more difficult to discuss dipolar broadening, even qualitatively, in ferromagnetic than in paramagnetic media. In loc. cit.,6 the writer studied breadths in the latter at some length. Even then, it was not possible to obtain an explicit formula for the line-shape. The best that could be done was to calculate the second and fourth moments of the deviation of the frequency from its mean value, and deduce qualitative conclusions from these computations. In the ferromagnetic case, however, the application of the method of moments strikes a snag if the field is applied in an arbitrary direction. The essential feature of the method which we employed was the use of the invariance of the diagonal sum, but with the exclusion of certain parts of the Hamiltonian, viz. the portion non-diagonal in the total magnetic quantum number M, because this portion definitely had the effect of introducing satellite lines<sup>7</sup> rather than of broadening the main line. In ferromagnetic materials, however, the dipolar interaction cannot be regarded as a small perturbation. The gist of our calculation in Section 2 was, in fact, to show how the demagnetization corrections caused by dipolar coupling influence the central resonance frequency. The component of magnetic moment in the direction of the applied field ceases to be a constant of the motion unless two of the demagnetizing factors are equal, and the field is applied along the direction of the third. Unless these conditions are satisfied our previous method cannot be used. We are therefore able to give a formula for the second moment only for the special case of axial symmetry. For this case, the calculation can be made by the same general method as that used in *l.c.*, but with two important changes in detail, as follows:

(a) It is no longer allowable to take

$$\langle S_{z_j}^2 \rangle_{\mathsf{Av}} = \langle S_{x_j}^2 \rangle_{\mathsf{Av}} = \langle S_{y_j}^2 \rangle_{\mathsf{Av}} = \frac{1}{3}S(S+1), \quad \langle S_{z_j} \rangle_{\mathsf{Av}} = 0 \quad (26)$$

since the magnetization cannot be supposed very weak in ferromagnetic bodies.

(b) The coupling of dipolar structure need not be supposed to have a true magnetic origin, or in other words, the coefficient  $D_{jk}$  in (2) need not necessarily have the value (3).

The formula for the mean square deviation of the frequency from the Kittel value (1) (with  $N_x = N_y$ ) turns out to be

$$\langle \Delta \nu^2 \rangle_{\mathsf{A}\nu} = h^{-2} [\langle S_{zj}^2 \rangle_{\mathsf{A}\nu} - \langle S_{zj} \rangle_{\mathsf{A}\nu}^2] \sum_k D_{jk}^2.$$
(27)

As is to be expected, this expression reduces to Eq. (10) of reference 6 if one can use (26) and (3), as one can for paramagnetic media with purely dipolar coupling.

The corresponding formula for the mean fourth power deviation would be tedious and unrepaying to calculate. Suffice it to note that  $\langle \Delta \nu^4 \rangle_{AV}$  would surely contain terms of the order  $A^2D^2/h^4$ , which will be very large compared to  $3[\langle\Delta\nu^2\rangle_{AV}]^2 \sim D^4/h^4$  inasmuch as the exchange coupling coefficient A will be very large in ferromagnetic media. As a result, the lines will have a far smaller half-breadth than one would calculate from (27) under the supposition of a Gaussian shape, for with a given second moment, enhancement of the fourth moment makes the line more sharply peaked. This is the phenomenon of "exchange narrowing" which we explained more fully in *l.c.* Much of the contribution to (27) comes, in fact, from transitions in which the energy change is of the order of magnitude of the exchange integral. Such transitions give rise to absorption lines of vastly higher frequency than the Kittel resonance frequency in which we are interested. The only reason that the expression (27) still remains of the order  $D^2/h^2$  rather than  $A^2/h^2$  is that these transitions are down by a factor of the order  $A^2/D^2$  in intensity as compared with those having nearly the resonance frequency. Lines having frequencies of the order A/hclearly should be regarded as satellites whose contributions to (27) are spurious for our purposes. Exchange narrowing is essentially an expression of the fact that the square root of the expression (27) furnishes a gross overestimate of the effective line-width because these spurious contributions have not been striken out.

Despite the inutility of (27) for quantitative purposes, certain qualitative conclusions can be drawn from (27) as follows:

(I) Exchange narrowing should in general be even more pronounced in ferromagnetic than in paramagnetic media, all other things being equal. The basis for this statement is that the second moment is of roughly the same order of magnitude in both types of media, but ferromagnetic materials will presumably have larger exchange integrals and hence larger fourth moments. There is thus no difficulty in understanding why ferromagnetic substances have much sharper resonance lines than one would calculate from the root mean square deviation of the dipolar field.

(II) Part of the line-breadth may be caused by short-range pseudo-dipolar forces rather than those of true magnetic origin. In another paper<sup>5</sup> the writer stressed the fact that in ferromagnetic materials the coupling coefficients in the potential of dipolar structure usually has a value materially larger than (3). Consequently one must not be surprised if sometimes lines are, despite (a), more diffuse in ferromagnetic than in paramagnetic substances. This situation could arise if the ferromagnetic material has such anomalously large coefficients  $D_{jk}$  as to more than offset the tendency towards more narrowing caused by larger exchange integrals. In *l.c.* we did not mention the possibility of anomalous dipolar coefficients in paramagnetic bodies. If the magnetic density is high, such anomalies are possible, but in general presumably less common than in ferromagnetics. Since effects (I) and (II) work in opposite directions, there is obviously considerable leeway in explaining line-width phenomena.

(III) There should be no discontinuity in the linewidth at the Curie point. This is one point on which the theory makes a definite prediction, and is in accord with experiment. Theoretically, as the Curie point is approached from below, the mean values involved in (27) approach the limits (26), and so (27) passes continuously into the corresponding formula of loc. cit. for the second moment for the purely paramagnetic case. The same is also true of the higher moments. Experimentally Bloembergen finds that in nickel and supermalloy, the two materials so far studied in this regard, the line-width does not change at the Curie point. This fact, however, does not distinguish between the broadening mechanisms (a), (b), and (c), for all three involve no abrupt changes at the Curie temperature.

(IV) The line-breadth caused by dipolar effects should decrease as the temperature is lowered from the Curie point to the absolute zero, for the expressions  $\langle S_{z_j}^2 \rangle_{Av}$  and  $\langle S_{z_j} \rangle_{Av}^2$  involved in (27) approach equality as the magnetization approaches its maximum value  $Ng\beta S$ . The dipolar line-breadth should hence vanish at the absolute zero. Existing experimental data do not extend to very low temperatures, but are of such a character as to make it highly unlikely that the linewidth vanishes at T=0. In supermalloy, for instance, Bloembergen<sup>16</sup> finds little, if any change in line-width between 300 and 600°K even though the magnetization changes by a factor changes by a factor over 2 in this interval. At 300°, the magnetization is almost as high as at T=0, and one would hence expect little further contraction in dipolar broadening to set in below room temperatures. In nickel Bloembergen finds that the line-width actually decreases when the anisotropy increases. These various experimental facts indicate pretty clearly that dipolar coupling between similarly situated atoms, such as is embodied in Eq. (27), is not the main cause of broadening in ferromagnetic materials. Nor can spin-lattice interaction be the chief mechanism of line-broadening at room temperatures and lower, for any broadening of such origin should vary drastically with temperature, and disappear completely at very low temperatures. This difficulty would not arise if the line-width were caused by the interaction of the spins with the eddy currents involved in the skin effect, but unfortunately a calculation by Kittel and Herring<sup>17</sup> shows that this process fails to give the observed width by a factor 10 or so. It is thus at present something of a mystery what is the actual mechanism of line-broadening in ferromagnetic media. It is just possible that theoretically a finite breadth of dipolar origin should persist even at T=0 because of crystalline imperfections, or in other words, because

the crystal is composed effectively of somewhat disjointedly connected microcrystals, each with slightly different resonance frequencies. It is conceivable that large enough breadths at low temperatures might be obtained if this fashion if there are large short-range pseudo-dipolar forces due to anisotropic exchange.

In this connection one can try generalizing (27) by abandoning the restriction that all atoms are similarly situated, so that  $\sum_{k} D_{jk}$  is no longer independent of j. By a calculation of the same general type as that in *l.c.*, but whose details we omit, it is found that the mean resonance frequency and mean square deviation are then given by

$$\langle \nu \rangle_{\mathsf{Av}} = (He/4\pi mc) + \mathfrak{N}^{-1}h^{-1}\sum_{j,k} D_{jk}\langle S_{zj} \rangle_{\mathsf{Av}}$$

$$\langle \Delta \nu^2 \rangle_{\mathsf{Av}} = \mathfrak{N}^{-1}h^{-2}\sum_{j,k} D_{jk}^2 [\langle S_{zj} \rangle_{\mathsf{Av}} - (\langle S_{zj} \rangle_{\mathsf{Av}})^2]$$

$$+ h^{-2} [\mathfrak{N}^{-1}\sum_{j} (\sum_{k} D_{jk})^2]$$

$$- \mathfrak{N}^{-2} (\sum_{j,k} D_{jk})^2] (\langle S_{zj} \rangle_{\mathsf{Av}})^2,$$

$$(29)$$

where  $\mathfrak{N}$  is the total number of atoms in the crystal. This formula is unfortunately rather hard to interpret. If  $\sum_{k} D_{jk}$  is not independent of j, the expression (29) does not vanish at T=0, and so gives the impression that the resonance line can remain diffuse even at T=0. However, the non-vanishing of (29) at T=0 is caused by transitions involving changes in the total spin  $\mathfrak{S}$  of the crystal from  $\Re S$  to  $\Re S - 1$ . Most of these transitions are associated with energy changes of the order of magnitude of the exchange integral A, and so their contributions to (29) are irrelevant since they represent weak satellites of completely different frequency than the resonance line. Some of these transitions, however, may be more intense than the rest, and involve relatively small changes in exchange energy, so that they should be classed as wings of the central resonance line rather than as satellites. In fact, the lowest Bloch spin waves for  $S_z = -(\Re S - 1)$  have almost the same exchange energy as that of the sole state  $S_z = -\Re S$ . It is thus conceivable that with dissimilarly situated atoms, some breadth persists even at T=0. When, however, the atoms are similarly situated, the resonance line at T=0 will consist of but a single component, as evidenced by the behavior of (27). Physically, this is because it is immaterial energetically which atom has  $S_{z_j} = -(S-1)$  in a state  $S_{z_j} = -(\Re S-1)$  when  $\sum_k D_{jk}$ is independent of j. Under this condition the dipolar interaction does not break down the selection rule  $\Delta \mathfrak{S} = 0$  for the matrix elements of  $S_x$  connecting  $S_z = -\Re S$  with  $S_z = -(\Re S - 1)$  and the absorption arising from the deepest state  $S_z = -\Re S$  consists of but a single line.

## 5. THE PROBLEM OF THE g-FACTOR IN FERROMAGNETIC RESONANCE

Doubtless we have given the impression that Kittel's formula (1) fits the observed resonance frequencies very well. Actually it does so only if the g-factor be given anomalously large values. For instance, in nickel the values of g yielded by experiments on ferromagnetic

 <sup>&</sup>lt;sup>16</sup> N. Bloembergen, Phys. Rev. (to be published).
 <sup>17</sup> C. Kittel and C. Herring, Phys. Rev. 77, 725 (1950).

resonance range from 2.19 to 2.42. On the other hand, the gyromagnetic ratio found by Barnett for nickel is 1.93. The discrepancies for other materials are set forth in detail in an interesting paper by Kittel.<sup>18</sup>

As Polder,4,19 Kittel,18 and the writer20 have independently pointed out, there is no reason why the g-factors yielded by microwave and magnetomechanical experiments need be the same. Conceptually, they relate to different quantities. The absorption measurements determine the factor g in the formula  $E = E_0$  $+Mg\beta H$  for the Zeeman energy states. Kittel aptly calls this kind of g the spectroscopic splitting factor. We shall also follow Kittel in using the letter g' for the anomaly (as compared with the classical orbital value -e/2mc) in the ratio of magnetic moment to angular momentum, which is the ratio which enters in gyromagnetic experiments.

Although it is quite possible in principle to understand why g and g' should be different, the theory strikes a snag as soon as one tries to explain quantitatively the discrepancy between g and g'. One can show quite generally that in the first approximation, the deviations of g and g' from 2 should be equal and opposite, so that

$$g - 2 = 2 - g'.$$
 (30)

This relation has been discovered independently by Kittel and the writer. Kittel has already published his derivation.<sup>18</sup> Ours is rather more general, as it is not based on a particular model, and so it is perhaps not superfluous if we give it here.

*Proof of (30).* Let us fictitiously imagine that there are fields  $H_0$  and  $H_s$  acting solely on the orbital and spin magnetic moments respectively. The actual physical case has, of course,  $H_0 = H_s$ . Let us first consider a one-atom or one-electron system in which the orbital angular momentum is quenched by the crystalline field, i.e., consists solely of non-diagonal elements, and in which spin-orbit coupling is disregarded. It is not necessary to specify anything about the crystalline field. The model can, for instance, be the usual one of a single atom with one or more bound electrons employed in the theory of magnetism by Schlapp, Penney, and others, or it can be the itinerant electron model of conduction band theory, used in magnetic problems by Mott, Brooks,<sup>21</sup> et al. If we disregard squares of  $H_o$ , the energy

levels are of the form

$$E = E_0 + 2M_S \beta H_S, \qquad (31)$$

where  $M_s$  is the spin equatorial quantum number. Now introduce as simultaneous perturbations the interaction of the orbital magnetic moment with the field  $H_o$ , and the spin-orbit coupling. The portions of the Hamiltonian function corresponding to these two perturbations we denote respectively by  $V_1$  and  $V_2$ . Both of these perturbations involve only non-diagonal elements, and so the energy is affected only in the second approximation

$$\Delta E = \sum_{l} |V_1 + V_2|_{jl^2} / h \nu_{jl}$$

In microwave spectra we are concerned with the linear Zeeman effect, and for this the only relevant part of  $|V_1+V_2|^2$  is the cross term  $V_1V_2+V_2V_1$ . Now  $V_1$  is independent of  $M_s$ , and so the only matrix elements of  $V_2$  which can contribute to the cross term are those which connect states j and l having the same values of  $M_s$ . These elements arise from the part  $AL_zS_z$  of  $A\mathbf{L}\cdot\mathbf{S}$  and are proportional to  $M_{S}$ . Therefore

$$\Delta E = \text{const.} + aH_0M_s + O(H_0^2). \tag{32}$$

The constant term we can assume independent of  $M_s$ , as we suppose the lowest spin multiplet is not decomposed in the absence of a magnetic field. Because of the Kramers degeneracy, this will always be the case if the spin is  $\frac{1}{2}$ . For spins of 1 or  $\frac{3}{2}$ , the multiplet will not be decomposed if there is cubic symmetry. Apart from squares of the magnetic fields, the total energy, which is the sum of (31) and (32), is of the form

$$E = E_O + \beta M_S (2H_S + aH_O). \tag{33}$$

The spectroscopic splitting factor is obtained by setting  $H_0 = H_s$ , and is hence

$$g=2+a.$$
 (34)

In terms of the partition function  $Z = \sum_{j} \exp(-E_{j}/kT)$ , the anomaly in the gyromagnetic ratio is

$$g' = \frac{\left[\partial \log Z / \partial H_o + \partial \log Z / \partial H_s\right]_{H_s = H_o}}{\left[\partial \log Z / \partial H_o + \frac{1}{2} \partial \log Z / \partial H_s\right]_{H_s = H_o}}$$

inasmuch as the derivatives  $-\partial E_i/\partial H_0$  and  $-\partial E_i/\partial H_s$ are respectively the orbital and spin magnetic moments of the state of energy  $E_j$ . Furthermore we have  $a\partial E_i/\partial H_s = 2\partial E_i/\partial H_o$ , and we may suppose a to be a small quantity, for otherwise the perturbation calculation has no basis. Hence

$$g' = (2+a)/(1+a) = 2-a.$$
 (35)

Equation (30) follows immediately from (34) and (35).

The preceding calculation assumed that the individual atoms or itinerant electrons could be regarded as isolated magnetic units. As long as the Zeeman effect is linear, inclusion of exchange or dipolar coupling between these units does not, however, spoil the validity of (30). Such coupling does not influence the relative

 <sup>&</sup>lt;sup>18</sup> C. Kittel, Phys. Rev. **76**, 743 (1949).
 <sup>19</sup> D. Polder, Phys. Rev. **73**, 1116 (1948).
 <sup>20</sup> J. H. Van Vleck, Physica **15**, 197 (1949).
 <sup>21</sup> H. Brooks, Phys. Rev. **58**, 909 (1940). As Kittel notes, Brooks purports to calculate g', but really computes g. However, because of a counterbalancing error, the numerical result which Brooks gives for g' (g in his notation), is correct, as one suspects since he obtains g' < 2. The compensating sign change arises because he does not allow for inversion of the spin-orbit parameter for  $d^{4}$ as compared with d. He implies that use of negative frequency denominators for transitions to unfilled states of lower energy incorporates the effect of this inversion, but actually this is not the case, for the sign behavior of the frequency denominators is the same for d and  $d^9$ 

contributions of the orbital and spin moments, and so does not change g'. Exchange interaction commutes with all spatial components of the total magnetic moment, and so does not influence g. The gist of Kittel's calculation, of ours of Section 1, is that the spectroscopic splitting factor is unaltered by dipolar interaction provided one takes for the effective field that given by the radical in Eq. (1).

Inadequacy of (30). Unfortunately, the approximate equality of g-2 and 2-g' predicted by (30) is not confirmed by experiment. The table in Kittel's paper shows that g-2 always exceeds 2-g', and is often twice as large. It is improbable that the discrepancies are caused by the fact that (30) includes only the first-order effect of spin-orbit interaction. The higher order perturbations, in the first place, are presumably small. What is more, they act in the wrong direction. If, for instance, one went to the extreme of assuming the crystalline field small compared with the spin-orbit coupling, a model based on the configuration d,<sup>9</sup> the, one most appropriate to nickel, would have g'=6/5 a value far too low.

How, then, can one explain the fact that the observed values of g are too high to fit (30)? Kittel reviews a number of suggestions made by various authors, all of which he dismisses as rather unlikely. We should like to add to this list one more remote possibility. In all the derivations of Kittel's fundamental formula, our Eq. (1), the effect of the dipolar and pseudo-dipolar forces on the frequency is included only in the first approximation. This approximation is doubtless adequate if the dipolar coupling is only of true magnetic origin. However, it is just conceivable that higher order corrections might alter the central frequency appreciably if there are large short-range forces of dipolar structure caused by anisotropic exchange. Our calculation shows that in the first approximation, such forces are without influence on the resonance frequency if the atoms are cubically spaced, but there is no assurance that this is true in higher orders. If these pseudodipolar effects are large, one would expect that there should be a large anisotropy, since forces of dipolar structure lead to anisotropy in the second approximation,<sup>5</sup> as has been shown by van Peijpe<sup>22</sup> and by the writer. On the other hand, the mean resonance frequency for a powder composed of cubic crystals is

influenced by these forces only in the third approximation (apart, of course, from the demagnetization corrections contained in Kittel's formula). The experimental data are not complete enough as yet to reveal whether there is a correlation between the amount of deviation from the formula g-2=2-g' and the amount of anisotropy. Supermalloy is a material with almost no anisotropy. It is unfortunate that no gyromagnetic measurements are available for supermalloy, so as to test whether formula (30) holds much more exactly than usual for this substance. One's guess is that it does not, for such a behavior would be out of line with that for other materials.

It should be pointed out that in Section 3 we derived the expression (25) for the effect of anisotropy on the resonance frequency only for a model in which the anisotropy is caused by the quadrupolar coupling (12). Actually, however, anisotropy can arise from the second-order effect of dipolar interaction, and this is the only mechanism operative if the atomic spin is  $\frac{1}{2}$ . The reason that we employed the dipolar rather than quadrupolar model in Section 3 is that the latter yields anisotropy in the first rather than second approximation, and so is easier to handle. The great merit of Kittel's macroscopic derivation of (25) is that it does not require any particular model, and so the relation between anisotropy and frequency given in (25) is presumably valid for dipolar as well as quadrupolar coupling. Yet the macroscopic approach by means of effective demagnetization factors is to a certain extent phenomonological. It would be interesting to treat the higher approximations of the dipolar model from the microscopic standpoint, and see whether it gives (25) in the second order, and especially whether it yields a significant shift in the resonance frequency in still higher orders if there are large anisotropic exchange forces. It looks as though we are between Scylla and Charybdis; presumably the higher order corrections yield either effectively isotropic coupling which commutes with the magnetic moment and so does not influence the frequency, or else an anisotropy already included in Kittel's phenomonological approach.

All told, the explanation of the anomalously high spectroscopic *g*-factors is at present an unsolved problem.

In writing the present paper, the writer has benefited greatly from stimulating discussions with Dr. C. Kittel and Dr. N. Bloembergen.

<sup>&</sup>lt;sup>22</sup> W. F. van Peijpe, Physica 5, 465 (1937).