

Paramagnetic Resonance Width in Iron and Nickel*

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A theory is presented to account for the unusually large linewidths found by Rodbell in the microwave resonance absorption of iron and nickel single crystals above the Curie points. The perturbing spin coupling is assumed to contain pseudodipolar (D) plus quadrupolar (Q) components. By the method of moments it is shown that the paramagnetic linewidth is proportional to $(D^2 + bQ^2)/J$, where J is the exchange integral and $b=0$ for $S=\frac{1}{2}$, and $b=1/27$ for $S=1$. The ferromagnetic anisotropy constant at 0°K is proportional to $(-D^2/J) + eQ$, where $e=0$ for $S=\frac{1}{2}$, $e\sim 2$ to 5 for $S=1$. Since D , Q , and J are probably not very temperature sensitive, measurements of paramagnetic linewidth and ferromagnetic anisotropy, together with well-known techniques for estimating J , may be used to deduce values of both D and Q . For iron and nickel it is shown that the contribution of Q to the linewidth is negligible, whereas the contribution from D is enormous, thus accounting for Rodbell's results.

I. INTRODUCTION

IN a ferromagnetic-paramagnetic resonance study of iron whisker crystals at 9 kMc/sec, Rodbell¹ has found the linewidth to rise linearly above the Curie temperature (770°C) to a value of 2000 oersteds at 850°C, the highest temperature reported. The linewidth in the ferromagnetic region was remarkably small for metals, ~ 50 oersteds or less, indicating extremely perfect crystals. At first sight it is difficult to see why the paramagnetic width should be so enormous. Preliminary measurements² on nickel crystals in the paramagnetic region indicate even larger widths.

It is the purpose of this paper to show why these ferromagnets should have such very large paramagnetic resonance linewidths and to relate these widths to the theoretical exchange, pseudodipole and quadrupole coupling constants J , D , and Q , and thereby to the measured ferromagnetic anisotropy constant K_1 .

Following Van Vleck's standard theory of ferromagnetic anisotropy³ we consider the spin Hamiltonian to be a sum of Zeeman, exchange, dipole-dipole-like, and quadrupole-quadrupole-like terms:

$$\mathcal{H} = \mathcal{H}_Z + \mathcal{H}_E + \mathcal{H}_D + \mathcal{H}_Q; \quad (1)$$

$$\mathcal{H}_Z = g\beta H_0 \sum_i S_i^z; \quad (2)$$

$$\mathcal{H}_E = - \sum_{i \neq j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j; \quad (3)$$

$$\mathcal{H}_D = \sum_{i > j} D_{ij} [\mathbf{S}_i \cdot \mathbf{S}_j - 3\mathbf{r}_{ij}^{-2} (\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})]; \quad (4)$$

$$\mathcal{H}_Q = \sum_{i > j} Q_{ij} \mathbf{r}_{ij}^{-4} (\mathbf{S}_i \cdot \mathbf{r}_{ij})^2 (\mathbf{S}_j \cdot \mathbf{r}_{ij})^2. \quad (5)$$

Here the z axis is taken along the (arbitrary) direction of the applied field H_0 . The coupling constants J_{ij} , D_{ij} , and Q_{ij} will be limited in general to nearest-neighbor J , D , and Q . It must be kept in mind that D_{ij} also includes long-range magnetic-dipole coupling; however,

the effects we consider will involve only D_{ij}^2 and hence will fall off rapidly with distance. For iron and nickel the pseudodipole portion of D is presumably very much larger than the magnetic dipole.

Throughout this paper it is assumed that D , Q , and J are temperature insensitive and in particular have roughly the same values at 0°K as in the paramagnetic region. It is probable that these parameters have only a small temperature variation in cubic crystals. According to Brenner⁴ the effect of thermal expansion can be related to measured changes of K_1 under volume strain, and he estimates K_1 of iron to increase because of thermal expansion by about 0.03% per degree. In nickel the fractional change is even smaller. Probably these changes are caused by similar small changes in D and Q . We might expect, therefore, as much as a 30% alteration of these coupling constants between 0°K and the paramagnetic region.

It is possible that some of the anisotropy arises from an interplay of spin-orbit coupling and crystalline field at the individual atomic sites. The Hamiltonian would then contain a term of the form

$$\mathcal{H}_a = \frac{1}{6} a \sum_i [(S_i^\xi)^4 + (S_i^\eta)^4 + (S_i^\zeta)^4 - \frac{1}{3} S(S+1)(3S^2 + 3S - 1)],$$

where ξ , η , and ζ are the cubic crystal axes. This term makes no contribution, however, unless $S \geq 2$, the physical reason being that only a 2^4 pole, or higher, can exhibit cubic anisotropy. (Van Vleck has pointed out that it is possible to obtain single-atom anisotropy with $S < 2$ if excited orbital levels are engaged, but that this effect is very small.) Since it is unlikely that states with S as large as 2 are important in metallic iron or nickel, it is believed that the principal contributions to the anisotropy come from pair interactions of the form of (4) or (5).

The possibility of pair interactions linear in one spin and quadratic in the other, i.e., dipole-quadrupole coupling, is ruled out by a time-reversal argument. The Hamiltonian must not change sign if all spin operators are reversed.

* R. Brenner, Phys. Rev. **107**, 1539 (1957).

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¹ D. S. Rodbell in *Growth and Perfection in Crystals* (John Wiley & Sons, Inc., New York, 1958), p. 247.

² D. S. Rodbell (private communication).

³ J. H. Van Vleck, Phys. Rev. **52**, 1178 (1937).

II. THE ANISOTROPY CONSTANT $K_1(0)$ IN TERMS OF J , D , AND Q

For a crystal with cubic symmetry, if the magnetic anisotropy energy is expanded in powers of the direction cosines $\alpha_1, \alpha_2, \alpha_3$ between the bulk magnetization and the three cubic axes, the lowest term is

$$F_1 = K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2). \quad (6)$$

Van Vleck³ shows that, in the molecular-field approximation, the Hamiltonian (1) gives rise to an anisotropy constant at 0°K of magnitude

$$K_1(0) = K_{1D}(0) + K_{1Q}(0), \quad (7)$$

with

$$K_{1D}(0) = -(3NS/16z)(D^2/J)\Omega; \quad (8)$$

$$K_{1Q}(0) = \frac{1}{3}NS^2(S - \frac{1}{2})^2Q\Omega. \quad (9)$$

Here z is the number of nearest neighbors, the factor $\frac{1}{3}$ in (9) is a correction noted in a subsequent paper by Van Vleck,⁵ and Ω is the lattice sum

$$\Omega = \sum_h [-1 + 5(\bar{\alpha}_h^2\bar{\beta}_h^2 + \bar{\beta}_h^2\bar{\gamma}_h^2 + \bar{\gamma}_h^2\bar{\alpha}_h^2)], \quad (10)$$

with $\bar{\alpha}_h, \bar{\beta}_h, \bar{\gamma}_h$ the direction cosines of the lattice vectors connecting nearest neighbors, taken with respect to the cubic axes. For simple cube, body-centered cube, and face-centered cube, respectively:

$$\Omega = -6, (16/3), 3. \quad (11)$$

A spin-wave analysis⁶ corrects K_{1D} so that Eq. (8) becomes

$$K_{1D}(0) = -(3NS/16z)\zeta(1+0.217\Delta)(D^2/J)\Omega, \quad (8')$$

$$\zeta = 2Sz\eta/(2Sz - \eta),$$

with, for sc, bcc, and fcc, respectively,

$$\eta = 1.26, 1.072, 0.974; \quad (12)$$

$$\Delta = 0, 1, 0.$$

As noted by Van Vleck, K_{1D} is intrinsically negative for bcc and fcc, whereas K_{1Q} takes the sign of Q . If K_1 is positive, as in iron, it is obvious that the contribution from Q overbalances that from D . Aside from this simple observation, however, measurements of K_1 cannot be used to separate the relative contributions from K_{1D} and K_{1Q} and hence to infer values of D and Q . Even the temperature variation of $K_1(T)$ is of no aid, since according to theory⁶ both the dipolar and quadrupolar contributions should vary as the tenth power of the magnetization. It might be possible to unscramble D and Q by comparing K_1 with K_2 , the higher-order anisotropy constant⁷; however, experimental values of

K_2 are very uncertain at present. A study of the theoretical relation between $K_2(0)$ and D, Q , and J will be reported in a future paper.

In the following sections it will be shown how measurement of the paramagnetic resonance linewidth can be combined with measurement of $K_1(0)$ to yield separate estimates of D and Q .

III. THE PARAMAGNETIC LINEWIDTH IN TERMS OF J , D , AND Q

The theoretical paramagnetic resonance linewidth to be expected from the Hamiltonian (1) will be analyzed by the method of moments, which has been used so successfully by Van Vleck⁸ to study the widths of paramagnets with small \mathcal{H}_E and no \mathcal{H}_Q . For these substances, in order to obtain moments of only the $\Delta M = \pm 1$ main line and to eliminate the small satellite lines, Van Vleck used the ingenious trick of truncating \mathcal{H}_D . Only that portion of \mathcal{H}_D was retained which commutes with \mathcal{H}_Z . The satellite $\Delta M = 0, \pm 2, \pm 3$ transitions, which are permitted in the presence of the entire \mathcal{H}_D , were thereby removed from consideration.

Anderson and Weiss⁹ and, independently, one of us¹⁰ have concluded that as \mathcal{H}_E becomes much larger than \mathcal{H}_Z , the satellite lines merge into the main line. In this situation, which includes microwave resonance in paramagnets with Curie temperatures greater than $\sim 10^\circ\text{K}$, the Hamiltonian \mathcal{H}_D must not be truncated.

A. Second Moment

The second moment of the resonance line is given by the well-known formula^{8,11}

$$\hbar^2\langle\omega^2\rangle = \langle|\mathcal{H}_E S^+|^2\rangle / \langle|S^+|^2\rangle. \quad (13)$$

Here, for convenience in calculating, we have assumed the perturbing potential to be $g\beta H^+ S^+$, with

$$S^+ = \sum_i S_i^+ = \sum_i (S_i^x + iS_i^y). \quad (14)$$

The angular brackets on the right of (13) signify a statistical average in the paramagnetic region (average of a matrix trace). Thus, sufficiently far above the Curie temperature for short-range order to be unimportant,

$$\langle|S^+|^2\rangle = N(\langle S_i^x{}^2\rangle + \langle S_i^y{}^2\rangle) = \frac{2}{3}NS(S+1). \quad (15)$$

The $\mathbf{S}_i \cdot \mathbf{S}_j$ term of \mathcal{H}_D can be incorporated into \mathcal{H}_E , and in what follows we shall understand J to include the generally very much smaller $-\frac{1}{2}D$. Then

$$[\mathcal{H}_E S^+] = g\beta H_0 \sum_i S_i^+ - 3 \sum_{i \neq j} D_{ij} F_j^i T_i^j$$

$$- \sum_{i \neq j} Q_{ij} (F_i^j T_i^j + T_i^j F_j^i) (F_j^i)^2, \quad (16)$$

⁶ J. H. Van Vleck, Phys. Rev. **78**, 266 (1950), footnote 13.

⁷ S. H. Charap and P. R. Weiss, Phys. Rev. **116**, 1372 (1959); F. Keffer and T. Oguchi, *ibid.* **117**, 718 (1960).

⁸ We are indebted to Professor T. Nagamiya for suggesting this approach. Another method of unscrambling D and Q has been suggested by S. H. Charap, Phys. Rev. **119**, 1538 (1960), who points out that these couplings lead to different temperature variations in the anisotropy of the magnetization. The effect, however, is small and probably very difficult to detect.

⁹ J. H. Van Vleck, Phys. Rev. **74**, 1168 (1948).

¹⁰ P. W. Anderson and P. R. Weiss, Revs. Modern Phys. **25**, 269 (1953).

¹¹ F. Keffer, Phys. Rev. **88**, 686 (1952).

¹² I. Waller, Z. Physik **79**, 380 (1932).

where

$$\begin{aligned} F_i^j &\equiv r_{ij}^{-1} \mathbf{S}_i \cdot \mathbf{r}_{ij} = \frac{1}{2} S_i^+ m_{ij} + \frac{1}{2} S_i^- p_{ij} + S_i^z \gamma_{ij}; \\ T_i^j &\equiv [S_i^+, F_i^j] = S_i^z p_{ij} - S_i^+ \gamma_{ij}; \\ m_{ij} &= -m_{ji} \equiv \alpha_{ij} - i\beta_{ij}; \quad p_{ij} = -p_{ji} \equiv \alpha_{ij} + i\beta_{ij}. \end{aligned}$$

Here α_{ij} , β_{ij} , and γ_{ij} are the direction cosines of \mathbf{r}_{ij} with respect to the axes of quantization. It proves convenient to call these α , β , and γ for short and to introduce the notation:

$$\begin{aligned} i^\pm &\equiv \frac{1}{2} S_i^\pm; \quad i^z \equiv S_i^z; \quad i^{zz} \equiv i^z i^z; \\ i^{\pm\pm} &\equiv i^\pm i^\pm; \quad i^{\pm z} \equiv \frac{1}{2} (i^\pm i^z + i^z i^\pm); \\ i^{00} &\equiv i^{zz} - i^+ i^- - i^- i^+ \equiv \frac{3}{2} (S_i^z)^2 - \frac{1}{2} (\mathbf{S}_i)^2; \\ (ij)^{00} &\equiv \frac{3}{2} S_i^z S_j^z - \frac{1}{2} \mathbf{S}_i \cdot \mathbf{S}_j. \end{aligned}$$

Then the following substitutions may be made in (16).

$$\begin{aligned} F_j^i T_i^j &= -[(1-3\gamma^2) i^+ j^z + p^2 i^- j^z + p\gamma (ij)^{00} - 2m\gamma i^+ j^+]; \\ \frac{1}{2} (F_i^j T_i^j + T_i^j F_i^j) &= (1-3\gamma^2) i^+ z + p^2 i^- z + p\gamma i^{00} - 2m\gamma i^{++}; \\ (F_j^i)^2 &= m^2 j^{++} + p^2 j^{--} + j^{zz} \\ &\quad - (1-\gamma^2) j^{00} + 2m\gamma j^{+z} + 2p\gamma j^{-z}; \end{aligned}$$

where in some of the terms in $F_j^i T_i^j$ the indices of summation i and j have been interchanged for convenience.

It is now relatively easy to write

$$[\mathcal{H}, S^+] \equiv U = \sum_{n=-3}^{+4} U_n, \quad (17)$$

with U_{-3} containing a net of three lowering operators:

$$U_{-3} = -2 \sum_{i \neq j} Q_{ij} p_{ij} i^+ i^- j^{--},$$

and so on up to U_4 , which contains a net of four raising operators. Then

$$\langle |[\mathcal{H}, S^+]|^2 \rangle = \sum_{n=-3}^{+4} \langle U_n U_n^* \rangle. \quad (18)$$

These terms may be evaluated with use of Eq. (15) and relations such as

$$\begin{aligned} \langle (S_i^z)^2 S_i^+ S_i^- \rangle &= (1/15) S(S+1) [2S(S+1)+1], \\ \langle (S^+)^2 (S^-)^2 \rangle &= (2/15) S(S+1) [4S(S+1)-3], \end{aligned}$$

etc. With a tedious amount of algebra Eq. (13) is finally reduced to

$$\begin{aligned} \hbar^2 \langle \omega^2 \rangle &= \frac{3}{2} N^{-1} S(S+1) \sum_{i \neq j} (1 + \gamma_{ij}^2) \\ &\quad \times \{ D_{ij}^2 + (1/675) [4S(S+1) - 3]^2 Q_{ij}^2 \}, \quad (19) \end{aligned}$$

plus Zeeman terms which need not be considered here. The dipolar portion of this expression was first obtained by Waller,¹¹ who calculated moments in the absence of \mathcal{H}_Z and therefore used the entire \mathcal{H}_D .

For a cubic crystal, with neglect of small contributions from other than the z nearest neighbors, Eq. (19) becomes

$$\hbar^2 \langle \omega^2 \rangle = 2S(S+1)z [D^2 + (1/675)\delta^2 Q^2], \quad (20)$$

with

$$\delta = (2S+3)(2S-1). \quad (21)$$

It is noted that no cross terms of the form DQ are present. The expression (20) is isotropic, that is, independent of the orientation of the magnetic field (axes of quantization) relative to the crystal axes. On the other hand, the second moment given by Van Vleck⁸ for his truncated \mathcal{H}_D is anisotropic, since truncation is with respect to \mathcal{H}_Z and differs as the direction of H_0 varies with respect to the cubic crystal axes. Van Vleck's result, when averaged over a sphere, is precisely $\frac{3}{10}$ the dipolar portion of Eq. (20). The fact that large \mathcal{H}_E , which requires use of the full \mathcal{H}_D , increases the second moment of the line is sometimes referred to as the "10/3 effect," and has been discussed in detail elsewhere.^{9,10}

B. Fourth Moment

In order to obtain a rough notion of the shape of the line, it is necessary to evaluate higher moments. The fourth moment is obtained from^{8,11}

$$\hbar^4 \langle \omega^4 \rangle = \langle |[\mathcal{H}, U]|^2 \rangle / \langle |S^+|^2 \rangle, \quad (22)$$

with U given by Eq. (17).

Let us first examine the terms in $[\mathcal{H}, U]$ contributed by \mathcal{H}_D . Of these, the terms proportional to JD are by far the largest. As in Eq. (17), we may write

$$[\mathcal{H}, U]_{JD} = \sum_{n=-1}^{+2} V_n; \quad (23a)$$

$$\langle |[\mathcal{H}, U]|^2 \rangle_{JD^2} = \sum_{n=-1}^{+2} \langle V_n V_n^* \rangle. \quad (23b)$$

The V_n are complicated expressions, but the evaluation is straightforward, if tedious. The result for the three cubic lattices is

$$\begin{aligned} \hbar^4 \langle \omega^4 \rangle_{JD^2} &= 4zJ^2 D^2 S(S+1) \left[\frac{3}{5} \delta + \frac{4}{3} S(S+1)(2z-3) \right. \\ &\quad \left. + 8S(S+1)\Delta \right], \quad (24) \end{aligned}$$

where, for sc, bcc, and fcc, respectively,

$$\Delta = 0, 0, 1. \quad (25)$$

The extra term in the fcc arises because some nearest neighbors of a given site are also neighbors of each other.

When averaged over a sphere, Van Vleck's⁸ comparative result for the truncated Hamiltonian is precisely $3/10$ the value of (24) above. Thus the ratio of the fourth moments of truncated and untruncated \mathcal{H}_D is the same as the corresponding ratio of the second

moments. Therefore, for the three cubic lattices,

$$\langle\omega^4\rangle/\langle\omega^2\rangle = 2\hbar^{-2}J^2\left[\frac{2}{3}\delta + \frac{4}{3}S(S+1)(2z-3) + 8S(S+1)\Delta\right], \quad (26)$$

for the principal term from \mathcal{H}_D , independent of truncation. We shall later argue that this equation should also hold for the principal term from \mathcal{H}_Q .

C. Exchange-Narrowed Linewidth

According to the analysis by Anderson and Weiss⁹ of paramagnetic resonance in the presence of $\mathcal{H}_E \gg \mathcal{H}_D$, the line shape is given by

$$I(\omega) = \frac{2}{\pi} \frac{(\omega_p^2/\omega_e)}{[\omega^2 + (\omega_p^2/\omega_e)^2]}. \quad (27)$$

Here ω_p is the frequency associated with the dipolar source of the linewidth, that is

$$\omega_p^2 = \langle\omega^2\rangle; \quad (28)$$

and ω_e is the rate at which the exchange modulates the dipolar perturbation. In the language of Van Vleck and Gorter,^{8,12} this modulation, which can be traced to the noncommutativity of \mathcal{H}_D and \mathcal{H}_E as exhibited by Eq. (22), causes "exchange-narrowing" of the linewidth.

The Anderson-Weiss line shape has the fourth moment

$$\begin{aligned} \langle\omega^4\rangle &= 3\omega_p^4 + \frac{1}{2}\pi\omega_p^2\omega_e^2 \\ &\approx \frac{1}{2}\pi\omega_p^2\omega_e^2. \end{aligned} \quad (29)$$

The linewidth is given by

$$\Delta\omega \approx \omega_p^2/\omega_e \approx \langle\omega^2\rangle[\frac{1}{2}\pi\langle\omega^2\rangle/\langle\omega^4\rangle]^{\frac{1}{2}}. \quad (30)$$

The arguments of Anderson and Weiss are sufficiently general to apply to \mathcal{H}_Q as well as to \mathcal{H}_D . Let us now tentatively identify ω_p^2 in the above equations with $\langle\omega^2\rangle$ of Eq. (20). Then, according to Eq. (29),

$$\langle\omega^4\rangle/\langle\omega^2\rangle \approx \frac{1}{2}\pi\omega_e^2. \quad (31)$$

The ratio of moments thus depends only on the rate ω_e at which the exchange interaction modulates the perturbation. As long as $\omega_e \gg \omega_p$, we do not expect this ratio to depend on the particular type of perturbation causing the linewidth. This ratio is in fact independent of truncation of \mathcal{H}_D , as shown by Eq. (26). Therefore, without going to the very considerable trouble of calculating $\langle\omega^4\rangle$ in the presence of \mathcal{H}_Q , we can in general relate the right side of (26) with $\frac{1}{2}\pi\omega_e^2$.

Incidentally, this argument supplies a missing link in the otherwise complete analysis given by Anderson and Weiss of the 10/3 effect of $\mathcal{H}_E \gg \mathcal{H}_Z$ on paramagnetic linewidths.

From Eq. (30) the linewidth, for the three cubic

lattices, is

$$\hbar\Delta\omega \approx \frac{z\pi^{\frac{1}{2}}S(S+1)[D^2 + (1/675)\delta^2Q^2]}{J[\frac{2}{3}\delta + \frac{4}{3}S(S+1)(2z-3) + 8S(S+1)\Delta]^{\frac{1}{2}}}; \quad (32)$$

with δ given by Eq. (21), Δ by (25).

It is seen that the contribution from a given size of Q increases rapidly with increasing S .

As mentioned above, when $\mathcal{H}_E \gg \mathcal{H}_Z$, the linewidth is isotropic. On the other hand, when $\mathcal{H}_E \ll \mathcal{H}_Z$ and the truncated \mathcal{H}_D must be used, Van Vleck finds a marked anisotropy in the main line. One way of determining the applicability of our theory to a given case is to search experimentally for linewidth anisotropy.

IV. RELATION BETWEEN PARAMAGNETIC LINEWIDTH AND FERROMAGNETIC ANISOTROPY

The results of the previous two sections are summarized by a pair of equations:

$$\hbar\Delta\omega = (a/J)[D^2(T) + bQ^2(T)]; \quad (33)$$

$$K_1(0)/N = c\{-D^2(0)/J + eQ(0)\}; \quad (34)$$

where we have emphasized the possible temperature dependence of D and Q , as discussed at the end of Sec. I. If temperature effects are neglected, then measured values of linewidth and anisotropy may be combined to yield both D and Q . We consider two cases in detail.

Case A: $S = \frac{1}{2}$

With spin $\frac{1}{2}$ there can be no quadrupolar coupling, and therefore

$$b = e = 0. \quad (35)$$

From Eqs. (8') and (32) it is easy to evaluate for sc, bcc, fcc, respectively:

$$\begin{aligned} a &= 2.66, \quad 2.95, \quad 3.07; \\ c &= -0.150, \quad 0.077, \quad 0.03, \end{aligned} \quad (36)$$

It is generally assumed that nickel, when considered from the atomic point of view, is predominantly spin $\frac{1}{2}$. If this is so, and with the fcc numbers, we expect

$$\begin{aligned} \hbar\Delta\omega &\approx (a/c)[|K_1(0)|/N] \\ &= (100)(9 \times 10^{-18} \text{ erg}), \end{aligned} \quad (37)$$

where we have inserted the standard experimental value of $K_1(0)$. The predicted paramagnetic width is thus

$$\Delta\nu \approx 2 \times 10^{11} \text{ cycles/sec.} \quad (38)$$

This extremely broad line could probably best be detected by microwave Faraday rotation. Any quadrupolar source of $K_1(0)$ would of course change the result (38). Since K_1 in nickel is negative, the presence of positive (or negative) Q would mean we have under- (or over-) estimated D and also the linewidth. (As shown below, the contribution of Q to the width is

¹² C. J. Gorter and J. H. Van Vleck, Phys. Rev. 72, 1128 (1947).

negligible.) Of course, actual measurements of $\Delta\nu$ could be used to determine the quadrupole-dipole ratio.

Preliminary measurements by Rodbell² indicate a tremendously broad paramagnetic width in nickel. Bloembergen¹³ has measured a width of ≈ 750 oe at the Curie point in a sample of electrolytic nickel foil; at higher temperatures the resonance signal became unobservably small, but there were clear indications that the linewidth rises rapidly with temperature.

Case B: $S=1$

Here, for sc, bcc, fcc:

$$\begin{aligned} a &= 4.09, & 4.62, & 4.91; \\ ab &= 0.151, & 0.171, & 0.182; \\ c &= -0.263, & 0.144, & 0.047; \\ ce &= -0.5, & 0.444, & 0.25. \end{aligned} \quad (39)$$

Let us tentatively assume that iron has predominantly spin 1, and that its positive anisotropy arises *entirely* from Q . Then, with use of the bcc numbers, we expect

$$\begin{aligned} \hbar\Delta\omega &\approx (ab/c^2e^2)[K_1(0)/N]^2J^{-1} \\ &= 0.868 \left[\frac{(6.2 \times 10^{-18})^2}{3.6 \times 10^{-14}} \right] \text{erg}. \end{aligned} \quad (40)$$

Here we have inserted the standard experimental value of $K_1(0)$, and we have evaluated J from Rodbell's¹ measurement of the exchange stiffness parameter A , using $S=1$. The predicted paramagnetic width is

$$\Delta\nu \approx 1.4 \times 10^5 \text{ cycles/sec}, \quad (41)$$

that is, less than 0.1 oe. Thus the width from Q is negligible, even in the unlikely situation that K_{1Q} is

¹³ N. Bloembergen, Phys. Rev. 78, 572 (1950).

several orders of magnitude larger than K_1 . Almost the entire width is therefore caused by D , and for bcc with $S=1$ this width is

$$\hbar\Delta\omega = (32.1)[|K_{1D}(0)|/N],$$

or

$$\Delta\nu = 3.0 \times 10^{10} \epsilon \text{ cycles/sec}. \quad (42)$$

Here the parameter ϵ is given by

$$\epsilon = |K_{1D}(0)|/K_1(0), \quad (43)$$

and is less (or greater) than unity if $|K_{1D}(0)|$ is less (or greater) than $\frac{1}{2}K_{1Q}(0)$.

For $\epsilon=1$, this width corresponds to 10.7×10^3 oe. Rodbell's measurement of 2×10^3 oe at 850°C can thus be accounted for, although the experiment must be carried to a temperature sufficiently high for the width to flatten out before a reliable estimate of ϵ can be made. Small changes of D and Q with temperature, discussed at the end of Sec. I, will of course modify these results a bit.

Experiments indicate that the paramagnetic linewidth *rises* with temperature above the Curie point in ferromagnets,^{1,2,13} but *falls* above the Néel point in antiferromagnets.¹⁴ A detailed theory of this temperature variation has not yet appeared. Short-range order effects are clearly involved, apparently in such a fashion as to decrease the local field fluctuations in ferromagnets, but to increase the fluctuations in antiferromagnets. A proper theory should fit smoothly into the analysis of this paper, which is valid in the limit $T \rightarrow \infty$.

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¹⁴ D. H. Douglass, Jr., and M. W. P. Strandberg, Physica 27, 1 (1961).