

## Spin-Wave Interactions in an Anisotropic Ferromagnet\*

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In the presence of anisotropic interactions the spin-wave dispersion parameter  $\mathfrak{D}(T)$  acquires an  $aT^{3/2}$  dependence in addition to the  $bT^{5/2}$  dependence due to isotropic exchange. We have used Van Vleck's anisotropic exchange, and the largest effect consistent with the magnetocrystalline anisotropy of a cubic ferromagnet is found to come from the pseudodipolar coupling. While the anisotropy appears only in the second order of perturbation theory, there is a first-order contribution to  $\mathfrak{D}(T)$  which varies as the third power of the magnetization, itself a function of temperature. Thus  $a \approx C(g\beta H_A^D/k_B T_c)^{1/2}$ , where  $C$  is the coefficient in the Bloch law,  $H_A^D$  is the pseudodipolar contribution to the anisotropy field, and  $T_c$  is the Curie temperature. The coefficient  $a$  depends upon the direction of spin-wave propagation and averages to zero over a sphere. In a first approximation, then, there is no  $T^3$  term in the magnetization. In an experiment dealing with selected propagation directions, such as spin-wave resonance or inelastic neutron scattering, since  $b \approx C/T_c$ , an effect important when  $T/T_c \lesssim (g\beta H_A^D/k_B T_c)^{1/2}$  is predicted.

### I. INTRODUCTION

SPIN-WAVE interactions are most conveniently studied by measuring the temperature dependence of the spin-wave energy.<sup>1</sup> For long wavelengths this energy varies quadratically with wave vector  $\mathbf{k}$ ; the departure at temperature  $T$  of the constant of proportionality or exchange parameter  $\mathfrak{D}(T)$  from its value at  $T=0^\circ\text{K}$  measures the average interaction of the mode under study with the thermal population of elementary excitations (magnons, phonons, conduction electrons). The interpretation of experimental results relies upon the existence of an inventory of behaviors (dependences on temperature) predicted for various models. The purpose of this paper is to call attention to an item which should be included in this "catalog" and to show that it might be very important.

The catalog of possible contributions to the temperature dependence of  $\mathfrak{D}(T)$  is reviewed in the next section, where particular attention is drawn to the occurrence of terms proportional to  $T^{3/2}$ . At sufficiently low temperatures such terms are the dominant ones, but in that region the difference between  $\mathfrak{D}(T)$  and  $\mathfrak{D}(0)$  is often immeasurably small. This situation is illustrated by consideration of the magnetocrystalline anisotropy, whose influence is usually obscured unless  $k_B T \lesssim 12g\beta H_A$ , where  $H_A$  is the effective anisotropy field,  $k_B$  is Boltzmann's constant,  $g$  the spectroscopic splitting factor and  $\beta$  the Bohr magneton. In Sec. III we discuss  $\mathfrak{D}(T)$  for a model in which there is an underlying interaction "inefficient" in producing the magnetocrystalline anisotropy; a cubic ferromagnet with anisotropic exchange as

a perturbation to the isotropic exchange.<sup>2</sup> Here a  $T^{3/2}$  term arises in the temperature dependence of  $\mathfrak{D}(T)$  which is important if  $T/T_c \lesssim (g\beta H_A^D/k_B T_c)^{1/2}$ , where  $T_c$  is the Curie temperature and  $H_A^D$  originates in the pseudodipolar part of the perturbation. We conclude in Sec. IV with the suggestion that the behavior predicted by this model may be worth looking for whenever a  $T^{3/2}$  dependence of  $\mathfrak{D}(T)$  is observed.

### II. TEMPERATURE DEPENDENCES OF THE EXCHANGE PARAMETER

In a pair of classic papers Dyson<sup>3</sup> proved that the interaction between spin waves in an ideal three-dimensional Heisenberg ferromagnet has an extremely small effect on the low-temperature thermodynamic behavior. Long-wavelength magnons pass almost undisturbed through the thermal excited spin system, and the interaction yields a correction to the Bloch law for the magnetization,  $M(0) - M(T) = CM(0)T^{3/2}$ , whose leading term is proportional to  $T^4$ . Equivalently, the exchange parameter decreases with increasing temperature as  $T^{5/2}$  because of the interaction. A clear physical interpretation of Dyson's result has been presented by Keffer and Loudon,<sup>4</sup> who explained that for an isotropic nearest-neighbor exchange coupling the spin-wave energy depends on the average angle between neighboring spins rather than on the average angle between the spins and the magnetization. The latter dependence is the interpretation of the spurious  $T^3$  term [equivalent to  $\mathfrak{D}(T) \propto T^{3/2}$ ] obtained by many authors.<sup>5,6</sup>

Mattis and Horwitz<sup>6</sup> have, on the other hand, recently shown that the excitation of spin waves shields the effects of the transverse components of the spin vectors beyond a temperature-dependent "Debye length"  $R_0(T)$  ( $\propto T^{-1/2}$ ). Thus Dyson's result must be

\* A report of this work was given at the meeting of the American Physical Society, January 1964 [Bull. Am. Phys. Soc. **9**, 112 (1964)].

<sup>1</sup> Experimental techniques include spin-wave resonance; R. E. Weber and P. E. Tannenwald, Phys. Chem. Solids **24**, 1357 (1963), inelastic neutron scattering; M. Hatherly, K. Hirakawa, R. D. Lowde, J. F. Mallett, M. W. Stringfellow, and B. H. Torrie, J. Appl. Phys. **35**, 892 (1964) and Proc. Phys. Soc. (London) (to be published), parallel pumping; R. C. LeCraw and L. R. Walker, J. Appl. Phys. **32**, 167S (1961) and discussion by C. W. Haas, Phys. Rev. **132**, 228 (1963).

<sup>2</sup> This is a well-known special case of the anisotropic exchange introduced by J. H. Van Vleck, Phys. Rev. **52**, 1178 (1937).

<sup>3</sup> F. J. Dyson, Phys. Rev. **102**, 1217, 1230 (1956).

<sup>4</sup> F. Keffer and R. Loudon, J. Appl. Phys. **32**, 2S (1961).

<sup>5</sup> See preceding references and references cited therein.

<sup>6</sup> See D. C. Mattis and L. P. Horwitz, Phys. Rev. Letters **10**, 511 (1963) and references therein.

modified if there is appreciable exchange between distant spins. For the long-range part of the exchange, a  $T^{3/2}$  dependence of the spin-wave energy obtains, because it depends on the average angle between the spins and the magnetization.

But if attention is restricted to short-range exchange, a  $T^{3/2}$  dependence of the spin-wave energy will still exist because of the magnetocrystalline anisotropy energy, which is sensitive to the average angle between the spins and the magnetization. It has been noted<sup>4,7</sup> that this is the nature of the power-law dependence of anisotropy on magnetization which may be written<sup>8</sup>

$$\frac{K_n(T)}{K_n(0)} = \left[ \frac{M(T)}{M(0)} \right]^{n(n+1)/2}, \quad (1)$$

where  $K_n$  is the anisotropy coefficient associated with spherical harmonics of degree  $n$ . The result expressed by Eq. (1) deals only with the  $\mathbf{k}$ -independent part of the spin-wave energy which originates in the anisotropic part of the Hamiltonian. Of course, it is only the  $\mathbf{k}$ -dependent part of this energy which can be regarded as modifying  $\mathfrak{D}(T)$ . A relation like Eq. (1) may be expected to be valid for this part too, and, at least for the case treated here, this is so (see the Appendix). Perhaps it is appropriate to emphasize here that the  $\mathbf{k}$ -independent part competes only with the internal magnetic field and that this competition is part of our everyday experience with magnetic materials. On the other hand, the  $\mathbf{k}$ -dependent part of the anisotropy energy is surely eclipsed by the exchange and only because its temperature dependence is expected to be more important than that of the exchange energy is it treated here.

To be complete, the temperature dependence of  $\mathfrak{D}(T)$  due to interaction of spin waves with phonons and with conduction electrons should be mentioned. These were recently discussed by Izuyama and Kubo,<sup>9</sup> who found that  $\mathfrak{D}(T)$  has a term proportional to  $T^2$  as the leading effect of the conduction electrons and one proportional to  $T^4$  as the leading effect of the phonons. If no  $T^{3/2}$  term arises from magnon-magnon interactions, then the magnon-electron interactions should dominate in metals at low temperatures.

### III. ANISOTROPIC EXCHANGE IN A CUBIC CRYSTAL

The model which we discuss is that of anisotropic exchange introduced by Van Vleck.<sup>2</sup> He showed that such anisotropic interactions as arise in a multipole expansion of the coupling between local spins could be responsible for the directional dependence of the energy,

i.e., magnetocrystalline anisotropy, in a cubic crystal. The various terms in such an expansion are named pseudodipolar, pseudoquadrupolar, and so on, with obvious reference to their form. These are treated as *perturbations* to the isotropic exchange. The range of interaction is, as for the isotropic exchange, commonly limited to nearest neighbors, and the strength of the interaction is essentially left to be determined by comparison of the calculated and experimental values of the  $K_n$ . The model suffers in a number of respects when confronted with the properties of real metals.<sup>10</sup> It nevertheless survives because it could apply to some non-metallic materials and also as a mathematical model whose properties have been subject to considerable theoretical investigation in order to enhance our understanding of the behavior of real materials.

The complete Hamiltonian is

$$H = H_{\text{EX}} + H_Z + H_D + H_Q + \dots \quad (2)$$

The exchange energy is given by

$$H_{\text{EX}} = -J \sum_{i \neq j}^{\text{n.n.}} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (3)$$

where  $2J$  is the exchange integral,  $\mathbf{S}_i$  is the spin vector attached to site  $i$ , and the sum is restricted to nearest-neighbor pairs (n.n.). The energy associated with the applied field  $H$  is

$$H_Z = -g\beta\mathbf{H} \cdot \sum_i \mathbf{S}_i. \quad (4)$$

Only the first two anisotropic terms of Van Vleck's multipole expansion have been explicitly included, the dipolar term

$$H_D = \frac{1}{2} \sum_{i \neq j} D_{ij} \{ \mathbf{S}_i \cdot \mathbf{S}_j - r_{ij}^{-2} (\mathbf{r}_{ij} \cdot \mathbf{S}_i) (\mathbf{r}_{ij} \cdot \mathbf{S}_j) \}, \quad (5)$$

and the quadrupolar term

$$H_Q = \frac{1}{2} Q \sum_{i \neq j}^{\text{n.n.}} r_{ij}^{-4} (\mathbf{r}_{ij} \cdot \mathbf{S}_i)^2 (\mathbf{r}_{ij} \cdot \mathbf{S}_j)^2. \quad (6)$$

Here  $\mathbf{r}_{ij}$  is the vector connecting sites  $i$  and  $j$ . The coefficient in the dipolar term is

$$D_{ij} = g^2\beta^2/r_{ij}^3 + D\delta(|\mathbf{r}_{ij}| - l), \quad (7)$$

where  $l$  is the nearest-neighbor distance. The  $r^{-3}$  dependence of  $D_{ij}$  is the contribution of the magnetic dipolar coupling. The anisotropic exchange between nearest-neighbor spins is measured by the pseudodipolar and pseudoquadrupolar coupling constants  $D$  and  $Q$ .

The calculation of the low-lying states of this Hamiltonian is most conveniently made by first neglecting the long-range magnetic coupling. The spin-wave energy levels, to first order in the quadrupolar and second order in the dipolar coupling constants, may be

<sup>7</sup> T. Oguchi and A. Honma, J. Phys. Soc. Japan **16**, 79 (1961).

<sup>8</sup> C. Zener, Phys. Rev. **96**, 1335 (1954); J. H. Van Vleck, J. Phys. Radium **20**, 124 (1959); H. B. Callen and E. R. Callen, Phys. Chem. Solids **16**, 310 (1960).

<sup>9</sup> T. Izuyama and R. Kubo, J. Appl. Phys. **35**, 1074 (1964).

<sup>10</sup> F. Keffer and T. Oguchi, Phys. Rev. **117**, 718 (1960); R. J. Joenk, Phys. Rev. **130**, 932 (1963).

expressed as

$$E = F(H) + VK_1(0)\Gamma_4 + \sum_{\mathbf{k}} n_{\mathbf{k}} e_{\mathbf{k}}(T), \quad (8)$$

where  $F(H)$  is a field-dependent ground-state energy,  $V$  is the sample volume,  $K_1(0)$  is the first cubic anisotropy constant with contributions proportional to  $Q$  and  $D^2/J$ ,  $n_{\mathbf{k}}$  is the number of spin waves excited of wave vector  $\mathbf{k}$ , and  $e_{\mathbf{k}}(T)$  is the energy per spin wave, in whose temperature dependence may be embodied the effects of spin-wave interactions. The anisotropy constant multiplies a function of the direction cosines of the magnetization with respect to the crystal axes;

$$\Gamma_4 = \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2 - \frac{1}{3}. \quad (9)$$

Inclusion of the magnetic dipolar coupling modifies these results in a way first discussed by Holstein and Primakoff.<sup>11</sup> In particular, the spin-wave energy becomes, at  $T=0^\circ\text{K}$ ,

$$e_{\mathbf{k}}'(0) = [e_{\mathbf{k}}(0) - g\beta N_z M(0)] \times (1 - \{g\beta 4\pi M(0) / [e_{\mathbf{k}}(0) - g\beta N_z M(0)]\} \sin^2 \theta_{\mathbf{k}})^{1/2}, \quad (10)$$

where  $N_z$  is the demagnetizing factor of the sample and  $\theta_{\mathbf{k}}$  is the angle between the direction of  $M(\mathbf{z})$  and  $\mathbf{k}$ . For  $k_B T \gg g\beta 4\pi M$ , Eq. (10) is also valid at finite temperatures with  $M(0)$  replaced by  $M(T)$ .<sup>7</sup> Hereinafter, the effect of the magnetic dipolar coupling in the radical will be disregarded. It is to be noted, on the one hand, that it has a calculable effect and, on the other hand, that it should be unimportant in the present context for  $k_B T \gg g\beta 4\pi M$ .

The quantities appearing in Eq. (8) have been evaluated by various authors,<sup>12</sup> and, including the surface demagnetizing energy, they are, at  $T=0^\circ\text{K}$ ,

$$F(H) = F_0 - g\beta NS[H - N_z M(0)] - NJZS^2, \quad (11)$$

$$VK_1(0) = \frac{1}{15} NS_4 Q S^2 (S - \frac{1}{2})^2 - \frac{15}{16} \frac{NSD^2}{ZJ}, \quad (12)$$

$$e_{\mathbf{k}}(0) = g\beta [H - N_z M(0)] + 2SJZ(1 - \gamma_{\mathbf{k}}) - \frac{V}{NS} K_1(0) [10 - 4(1 - \gamma_{\mathbf{k}})] \Gamma_4 + 2S \sum_{\mathbf{l}} E_1 \cos \mathbf{k} \cdot \mathbf{l}. \quad (13)$$

Here,  $N$  is the total number of atoms in the sample, each with spin  $S$ ,

$$S_4 = 6/5, 16/15, 3/5 \quad (14)$$

for the simple cubic (sc), body-centered cubic (bcc), and face-centered cubic (fcc) lattices, respectively,  $F_0$

<sup>11</sup> T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940). Also see C. Herring and C. Kittel, *ibid.* 81, 869 (1951).

<sup>12</sup> See Ref. 10 and references therein; also an excellent review of the entire subject of spin waves which has been prepared by F. Keffer for publication in the *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, to be published).

is just a constant,  $Z$  is the coordination number of the lattice,

$$\gamma_{\mathbf{k}} \equiv Z^{-1} \sum_{\mathbf{l}} \cos \mathbf{k} \cdot \mathbf{l}, \quad (15)$$

where the  $\mathbf{l}$ 's are the nearest-neighbor vectors and

$$E_1 = -\frac{1}{4} D [1 - 3(l_z/l)^2]. \quad (16)$$

We hasten to point out that the term involving  $\Gamma_4$  in  $e_{\mathbf{k}}$  has actually been calculated for  $\mathbf{k} \neq 0$  in the quadrupolar case only. It is the  $\mathbf{k}$ -independent part of this term which has been used in proving the application of Eq. (1) to these anisotropies ( $n=4$ ). For the pseudodipolar case we have estimated that the  $\mathbf{k}$ -dependent part will be approximately the same as in the quadrupolar case and since only the order of magnitude is significant, we have assumed that result in Eq. (13).

Because of the cubic symmetry

$$\sum_{\mathbf{l}} E_1 = 0, \quad (17)$$

so that the last term in Eq. (13) contributes only to the  $\mathbf{k}$ -dependent spin-wave energy. In the long-wavelength approximation, with  $a$  the lattice constant,

$$Z(1 - \gamma_{\mathbf{k}}) \approx k^2 a^2. \quad (18)$$

The spin-wave exchange energy becomes

$$e_{\mathbf{k}}^{\text{EX}}(0) \approx 2SJk^2 a^2 \equiv \mathfrak{D}(0)k^2. \quad (19)$$

Compared with this term, the other  $\mathbf{k}$ -dependent spin-wave energies are commonly neglected. Defining the anisotropy field

$$H_A(0) = \frac{2V}{g\beta NS} K_1(0) = \frac{2K_1(0)}{M(0)}, \quad (20)$$

the total spin-wave energy for  $ka \ll 1$  is

$$e_{\mathbf{k}}(0) \approx g\beta [H - N_z M(0) - 5H_A(0)\Gamma_4] + \mathfrak{D}(0) \left\{ 1 + \frac{g\beta H_A(0)}{ZJS} \Gamma_4 \right\} k^2 - S \sum_{\mathbf{l}} E_1(\mathbf{k} \cdot \mathbf{l})^2. \quad (21)$$

In the last term,  $\cos \mathbf{k} \cdot \mathbf{l}$  has been expanded to  $1 - \frac{1}{2}(\mathbf{k} \cdot \mathbf{l})^2$  and Eq. (17) invoked. At finite temperatures, but with constant volume,  $e_{\mathbf{k}}(T)$  is given by Eq. (21) and  $M(0)$  replaced by

$$M(T) \approx M(0) \left[ 1 - 0.306 \frac{V}{NSl^3} \left( \frac{k_B T_c}{ZJS} \right)^{3/2} \left( \frac{T}{T_c} \right)^{3/2} \right], \quad (22)$$

$\mathfrak{D}(0)$  replaced by

$$\mathfrak{D}(T) \approx \mathfrak{D}(0) \left[ 1 - 0.234 \frac{V}{NSl^3} \left( \frac{k_B T_c}{ZJS} \right)^{5/2} \left( \frac{T}{T_c} \right)^{5/2} \right], \quad (23)$$

and  $H_A(0)$  replaced by

$$H_A(T) \approx H_A(0) [M(T)/M(0)]^9 \\ \approx H_A(0) \left[ 1 - 9 \times 0.306 \frac{V}{NSJ^3} \left( \frac{k_B T_c}{ZJS} \right)^{3/2} \left( \frac{T}{T_c} \right)^{3/2} \right], \quad (24)$$

where  $T_c$  may conveniently be defined by the molecular field result

$$k_B T_c / J = \frac{2}{3} ZS(S+1). \quad (25)$$

Surely,  $g\beta H_A(0)/ZJS \ll 1$ . Still the temperature dependence of the coefficient of  $k^2$  in Eq. (21) will be dominated by that of  $H_A$  if  $k_B T \approx 12g\beta H_A(0)$ , which usually occurs for temperatures below a few degrees Kelvin. This is a rather insignificant effect.

No calculation of the temperature dependence of the last term in Eq. (21) has been given. We notice first that, if averaged over an isotropic distribution of  $\mathbf{k}$  directions, this energy vanishes. This means that, to a first approximation, it has no thermodynamic consequences. It also follows, then, that this energy can be positive or negative depending on the direction of  $\mathbf{k}$ . However, since the sign of  $D$  is unknown, we cannot predict for what directions the energy is, say, positive. On the other hand, a means for determining the sign of  $D$  is, in principle, available here. We define

$$\langle K^2 \rangle = \alpha_1^2 k_x^2 + \alpha_2^2 k_y^2 + \alpha_3^2 k_z^2, \quad (26)$$

where  $X$ ,  $Y$ , and  $Z$  are the crystal axes. Then we calculate,

$$-S \sum_1 E_1(\mathbf{k} \cdot \mathbf{l})^2 \\ = \frac{SD}{4} a^2 \times \begin{cases} 2(3\langle K^2 \rangle - k^2), & \text{sc} \\ 4(k_z^2 - \langle K^2 \rangle), & \text{bcc} \\ \frac{1}{2}(6k_z^2 - k^2 - 3\langle K^2 \rangle), & \text{fcc}, \end{cases} \quad (27)$$

which depends on both the directions of  $\mathbf{k}$  and  $\mathbf{M}$ . It is more convenient to discuss Eq. (27) as evaluated for the magnetization along an easy axis of the anisotropy ( $\langle 001 \rangle$  or  $\langle 111 \rangle$ ). Then

$$-S \sum_1 E_1(\mathbf{k} \cdot \mathbf{l})^2 = \delta (SD/4) (3 \cos^2 \theta_{\mathbf{k}} - 1) k^2 a^2 \quad (28)$$

with

|            |                       |                       |      |
|------------|-----------------------|-----------------------|------|
|            | $\langle 001 \rangle$ | $\langle 111 \rangle$ |      |
|            | 2                     | 0                     | sc   |
| $\delta =$ | 0                     | $\frac{4}{3}$         | bcc  |
|            | $\frac{1}{2}$         | 1                     | fcc. |

(29)

If we define the pseudodipolar contribution [see Eqs. (12), (20)] as

$$K_1^D(0) \equiv -\frac{1}{16} S_4 \frac{NSD^2}{VZJ} \quad \text{and} \quad H_A^D(0) = \left| \frac{2K_1^D(0)}{M(0)} \right|, \quad (30)$$

then

$$-S \sum_1 E_1(\mathbf{k} \cdot \mathbf{l})^2 = \delta \left[ \frac{1}{20|S_4|} \frac{S}{S+1} (k_B T_c) g\beta H_A^D(0) \right]^{1/2} \\ \times (3 \cos^2 \theta_{\mathbf{k}} - 1) k^2 a^2, \quad (31)$$

where the radical has the sign of  $D$  and we have used Eq. (25) to define  $T_c$ . By analogy with the  $\mathbf{k}$ -independent terms, it may be expected that the temperature dependence of this energy is as the third power of the magnetization since its angular dependence is that of the spherical harmonic of second degree. That this is indeed so is shown in the Appendix. We now multiply Eq. (31) by

$$[M(T)/M(0)]^3 \\ \approx 1 - 3 \times 0.306 \frac{V}{NSJ^3} \left( \frac{k_B T_c}{ZJS} \right)^{3/2} \left( \frac{T}{T_c} \right)^{3/2}. \quad (32)$$

Comparing the temperature dependence of this energy with that of the exchange, we find for the ratio of the  $T^{3/2}$  to the  $T^{5/2}$  terms

$$\delta (3 \cos^2 \theta_{\mathbf{k}} - 1) \frac{\mu [(S/S+1)k_B T_c g\beta H_A^D]^{1/2}}{k_B T}, \quad (33)$$

where

$$\mu = \frac{1.95Z}{(20|S_4|)^{1/2}} = \begin{cases} 2.4 & \text{sc} \\ 3.4 & \text{bcc} \\ 6.8 & \text{fcc}. \end{cases} \quad (34)$$

If the Curie temperature is a few hundred degrees and  $H_A^D$  is a few kilooersted, then this ratio can be of the order of unity for  $T \approx 50^\circ \text{K}$ ! Finally, when the ratio in Eq. (33) is much larger than unity, the  $T^{5/2}$  term is negligible and the  $\mathbf{k}$ -dependent part of the spin-wave energy may be described by  $\mathfrak{D}(T)k^2$ , where

$$\mathfrak{D}(T) \approx \mathfrak{D}(0) [M(T)/M(0)]^9 \quad (35)$$

with

$$\rho = 0.63\mu\delta (3 \cos^2 \theta - 1) \left[ \frac{2S(S+1)}{3} \frac{g\beta H_A^D(0)}{k_B T_c} \right]^{1/2}. \quad (36)$$

#### IV. CONCLUSION

The pseudodipolar coupling is "inefficient" in producing magnetocrystalline anisotropy in a cubic crystal, the second order of perturbation being required. The spin-wave energy resulting from this coupling in first order is therefore measured by the geometric mean of the pseudodipolar anisotropy and the exchange. This unusual magnitude combines with a comparatively fast temperature dependence (it varies as the third power of the magnetization) to produce the dominant variation with temperature of the curvature of the spin-

wave dispersion over a sufficient range of temperatures that it can be experimentally significant.

As an example we may discuss nickel. The expansion of the anisotropic exchange for  $S=\frac{1}{2}$  contains only the dipolar term so that the anisotropy is entirely dipolar in origin on this model [see Eq. (12)]. The lattice is fcc with the easy axis along  $\langle 111 \rangle$ . Relevant parameters are<sup>13</sup>  $M(0)=508$  G,  $K_1(0)=-7.5 \times 10^6$  ergs/cc,  $g=2.19$ ,  $JS=187k_B$ , and the coefficient in the Bloch  $T^{3/2}$  law,  $C=7.5 \times 10^{-6}$  deg $^{-3/2}$ . The radical in Eq. (36) is then  $\sim 1 \times 10^{-2}$ , and therefore

$$\rho_{Ni} \approx 0.043(3 \cos^2 \theta_k - 1) \quad (37)$$

for  $\mathbf{M}$  in an easy direction, and assuming  $D > 0$ . Finally,

$$\mathfrak{D}_{Ni}(T) \approx \mathfrak{D}_{Ni}(0) \{1 - 3.2(3 \cos^2 \theta_k - 1) \times 10^{-7} T^{3/2} - 4.1 \times 10^{-9} T^{5/2}\}, \quad (38)$$

where the coefficient of the  $T^{5/2}$  term has been calculated from Eq. (23). Notice that the  $T^{3/2}$  term is larger than the  $T^{5/2}$  term up to  $\approx 150^\circ\text{K}$  for  $\theta_k=0$ . Also, up to  $\approx 80^\circ\text{K}$  the  $\mathfrak{D}(T)$  increases with increasing temperature for  $\theta_k=\pi/2$ . For a thermodynamic property, such as the magnetization, the  $T^{5/2}$  term in  $\mathfrak{D}(T)$  leads to a  $T^4$  dependence. The  $T^{3/2}$  term has no effect however, since it averages to zero over the isotropic distribution of  $\mathbf{k}$  directions.

Experimentally, the temperature dependence of spin-wave energies has been as thoroughly investigated for nickel as for any other ferromagnet. In spin-wave resonance, a  $T^{3/2}$  term two orders of magnitude larger than that calculated here was observed.<sup>14</sup> It has been argued that the result is due to inhomogeneous magnetization and is not a measure of the "intrinsic" temperature dependence.<sup>15</sup> In inelastic neutron scattering<sup>16</sup> only a  $T^{5/2}$  term has so far been measured, the coefficient of which is an order of magnitude larger than that appearing in Eq. (28) above. The experimental situation is by no means conclusive. We have already pointed out that the anisotropic exchange model is not to be taken literally for metals. In future work we nevertheless suggest that, if a  $T^{3/2}$  dependence of the spin-wave energy is observed, it should be checked for a directional dependence such as is derived here.

#### APPENDIX

Our task is to show that the first order of the pseudodipolar coupling energy varies, due to spin-wave interactions, as  $[M(T)/M(0)]^3$ . The pseudodipolar

<sup>13</sup> B. E. Argyle, S. H. Charap and E. W. Pugh, Phys. Rev. **132**, 2051 (1963).

<sup>14</sup> T. G. Phillips and H. M. Rosenberg, Phys. Rev. Letters **11**, 198 (1963).

<sup>15</sup> P. E. Wigen, M. R. Shanabger, and C. F. Kool, Phys. Letters **7**, 109 (1963).

<sup>16</sup> R. D. Lowde (private communication).

Hamiltonian (5) may be written<sup>10</sup>

$$H_D = H^0 + H^{++} + H^{--} + H^+ + H^-, \quad (A1)$$

where

$$H^0 = \sum_{i,1} E_1 (\mathbf{S}_{Ri} \cdot \mathbf{S}_{Ri+1} - 3S_{Ri}^z S_{Ri+1}^z) \quad (A2)$$

is the only term which does not change the total  $S_z$  and, therefore, contributes a first-order energy. To go over to the spin-wave representation and obtain the correct result for the interaction due to the isotropic coupling, the Maleev transformation<sup>17</sup> is used:

$$\begin{aligned} S_{iz} &= S - b_i^* b_i, \\ S_{i+} &= 2S(1 - b_i^* b_i / 2S) b_i, \\ S_{i-} &= 2S b_i^*, \end{aligned} \quad (A3)$$

where

$$S_{i\pm} = S_{iz} \pm iS_{iy} \quad \text{and} \quad [b_i, b_j^*] = \delta_{ij}.$$

By a straightforward calculation involving introduction of the Fourier transforms  $b_k, b_k^*$ ,

$$\begin{aligned} b_i &= N^{-1/2} \sum_k b_k \exp(-i\mathbf{k} \cdot \mathbf{R}_i), \\ b_i^* &= N^{-1/2} \sum_k b_k^* \exp(i\mathbf{k} \cdot \mathbf{R}_i), \end{aligned} \quad (A4)$$

the result

$$\begin{aligned} H^0 &= E_0 + \sum_k b_k^* b_k \xi_k \\ &\quad - \sum_{k, k_1, k_2} \Gamma_{k_1, k_2} b_{k_1+k}^* b_{k_2-k} b_{k_1} b_{k_2} \end{aligned} \quad (A5)$$

is obtained. If the isotropic exchange [Eq. (3)] is included,

$$E_0 = -NJZS^2, \quad (A6)$$

$$\xi_k = 2SJZ(1 - \gamma_k) + 2S \sum_1 E_1 \cos \mathbf{k} \cdot \mathbf{l}, \quad (A7)$$

and

$$\begin{aligned} \Gamma_{k_1, k_2}^k &= (2N)^{-1} \sum_1 (J + 2E_1) \exp(i\mathbf{k} \cdot \mathbf{l}) \\ &\quad \times [1 - \exp(i\mathbf{k}_1 \cdot \mathbf{l})][1 - \exp(-i\mathbf{k}_2 \cdot \mathbf{l})] \\ &\quad + 3(2N)^{-1} \sum_1 E_1 \exp(i\mathbf{k} \cdot \mathbf{l}) \\ &\quad \times [\exp(i\mathbf{k}_1 \cdot \mathbf{l}) + \exp(-i\mathbf{k}_2 \cdot \mathbf{l})]. \end{aligned} \quad (A8)$$

Equation (A8) is the strength of the spin-wave interaction to this order in the pseudodipolar perturbation. The first term of (A8) originates in the isotropic spin coupling, vanishes if  $\mathbf{k}_1$  or  $\mathbf{k}_2$  is zero, and leads to the  $T^{5/2}$  dependence of the spin-wave energies. It is the source of Eq. (23) of our text and is now dropped from this discussion. The second term of (A8) comes from the Ising term of  $H^0$  and vanishes only if  $\mathbf{k}, \mathbf{k}_1$ , and  $\mathbf{k}_2$  vanish simultaneously. In first order there is a contribution from this term only if  $\mathbf{k} = 0$  or  $\mathbf{k} = \mathbf{k}_2 - \mathbf{k}_1$  (note that

<sup>17</sup> S. V. Maleev, Zh. Eksperim. i Teor. Fiz. **33**, 1010 (1957) [English transl.: Soviet Phys.—JETP **6**, 776 (1958)]; R. A. Tahir-Kheli and D. Ter Haar, Phys. Rev. **127**, 95 (1962). It should be emphasized that, except for  $S=\frac{1}{2}$ , this transformation is not a true representation of the spin operators. The Holstein-Primakoff representation<sup>11</sup> would serve as well. However, it does not yield, in as simple a way, the correct spin-wave interaction term resulting from isotropic exchange. See T. Oguchi, Phys. Rev. **117**, 117 (1960).

$\Gamma_{\mathbf{k}_1, \mathbf{k}_2}^{k_2-k_1} = \Gamma_{\mathbf{k}_1, \mathbf{k}_2}^0 = \Gamma_{\mathbf{k}_2, \mathbf{k}_1}^0$ . The diagonal part of  $H^0$  is therefore

$$H_{\text{diag}}^0 = E_0 + \sum_{\mathbf{k}} n_{\mathbf{k}} - 2 \sum_{\mathbf{k}_1, \mathbf{k}_2} n_{\mathbf{k}_1} n_{\mathbf{k}_2} \Gamma_{\mathbf{k}_1, \mathbf{k}_2}^0, \quad (\text{A9})$$

and the average energy associated with the excitation of a spin wave is then<sup>18</sup>

$$\bar{\xi}_{\mathbf{k}} \approx \xi_{\mathbf{k}} - 4 \sum_{\mathbf{k}'} \langle n_{\mathbf{k}'} \rangle \Gamma_{\mathbf{k}, \mathbf{k}'}^0, \quad (\text{A10})$$

where  $\langle \ \rangle$  denotes a thermal average and to this order the  $\langle n_{\mathbf{k}} \rangle$  are uncorrelated. In the long-wavelength ap-

<sup>18</sup> See, for example, M. Bloch, Phys. Rev. Letters 9, 286 (1962).

proximation for both  $\mathbf{k}$  and  $\mathbf{k}'$ , and making use of Eq. (17),

$$\bar{\xi}_{\mathbf{k}} = \xi_{\mathbf{k}} - (6/N) \sum_{\mathbf{k}'} \langle n_{\mathbf{k}'} \rangle \sum_1 E_1 \cos \mathbf{k} \cdot \mathbf{l}, \quad (\text{A11})$$

or, using Eq. (A7),

$$\bar{\xi}_{\mathbf{k}} = 2SJZ(1 - \gamma_{\mathbf{k}}) + 2S \sum_1 E_1 \cos \mathbf{k} \cdot \mathbf{l} [1 - (3/NS) \sum_{\mathbf{k}'} \langle n_{\mathbf{k}'} \rangle]. \quad (\text{A12})$$

Noting that  $[M(T)/M(0)]^n \approx [1 - (n/NS) \sum_{\mathbf{k}'} \langle n_{\mathbf{k}'} \rangle]$ , we have shown that the first-order pseudodipolar coupling energy varies as  $[M(T)/M(0)]^3$ .

## Ultrasonic Attenuation in Metals in the Fluid-Dynamic Approximation\*

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Ultrasonic attenuation in metals has been discussed from diverse points of view, and for the most part the derived attenuation formulas reduce in the first-order low-frequency limit to the viscous dissipative expression originally proposed by Mason. We undertake here a treatment for the low-frequency range which makes immediate contact with the transport-theory formalism of classical gases. "Fluid"-dynamic equations for the metal are formulated. For the electron-gas component a complete set of transport coefficients, including the several diffusion coefficients, is derived in a unified way on the simplifying assumption of a constant relaxation time. The acoustic attenuation coefficient for a longitudinal wave is deduced; the dominant term is of course the shear viscous one, but thermal and diffusion effects are also explicit.

### INTRODUCTION

MOTIVATED by the problem of ultrasonic attenuation, we develop "fluid"-dynamic equations for a metal in a manner parallel to that employed for a mixture of classical gases.<sup>1</sup> We take as a two-component model a free-electron gas coupled to an elastic ion continuum by an electromagnetic field and by collision. From the Boltzmann transport equation, fluid-dynamic equations and a complete set of transport coefficients are derived for the electron gas in a unified way; thus the various diffusion coefficients are included. For the present purpose a constant-rate relaxation term suffices for the collision integral, and we are content simply to postulate isotropic continuum equations for the ions. The equations for the two components are combined into a simplified set for the system as a whole. This, in conjunction with the equation for the relative motion of the electrons and the Maxwell equations for the associated electromagnetic field, determines the behavior of

the model. Finally, we consider the propagation of a longitudinal-plane acoustic wave and compute the attenuation coefficient.

### FORMULATION OF MACROSCOPIC EQUATIONS

The electron-distribution function  $f(\mathbf{r}, \mathbf{v}, t)$  obeys the Boltzmann transport equation

$$\mathcal{D}f \equiv \left[ \frac{\partial}{\partial t} + \mathbf{v} \cdot \frac{\partial}{\partial \mathbf{r}} + \frac{\mathbf{F}}{m_1} \cdot \frac{\partial}{\partial \mathbf{v}} \right] f = \frac{\partial f}{\partial t} \Big|_{\text{coll}}, \quad (1)$$

where symbols have their common meanings. When distinction is needed, indices 1 and 2 will be used to denote electrons and ions, respectively. Let  $\psi(\mathbf{v})$  be a particle property which is a function of velocity. According to standard transport theory,<sup>1</sup> (1) is multiplied by  $\psi(\mathbf{v})$  and integrated over velocity to obtain the equation of change for the mean value  $\langle \psi \rangle$ :

$$\frac{\partial}{\partial t} n_1 \langle \psi \rangle + \frac{\partial}{\partial \mathbf{r}} \cdot n_1 \langle \mathbf{v} \psi \rangle - n_1 \left\langle \frac{\mathbf{F}}{m_1} \cdot \frac{\partial \psi}{\partial \mathbf{v}} \right\rangle = \Delta_c \langle \psi \rangle. \quad (2)$$

Here  $n_1$  is the electron-particle density. We assume the electron and ion densities are equal.  $\Delta_c \langle \psi \rangle$  is the rate of change of  $\psi$  per unit volume due to collisions;  $\mathbf{F}$  has been

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<sup>1</sup> J. O. Hirschfelder, C. F. Curtiss, R. B. Bird, *Molecular Theory of Gases and Liquids* (John Wiley & Sons, New York, 1954), Chap. 7; S. Chapman and T. G. Cowling, *The Mathematical Theory of Non-uniform Gases* (Cambridge University Press, Cambridge, England, 1952).