Effect of Dipolar Interactions on the Spin-Wave Spectrum of a Cubic Antiferromagnet*

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The effect of dipolar interactions on the spin-wave spectrum of a cubic antiferromagnet is studied. The spin-wave spectrum is found to consist of two branches whose frequencies are

$$(\hbar\omega)^2 = (g\beta)^2 [H_A + H_E a^2 k^2] [H_A + H_E (2 - a^2 k^2) - 8\pi M/3 + 8\pi M \sin^2 \partial_k]$$

and

$$(\hbar\omega)^2 = (g\beta)^2 [H_A + H_E a^2 k^2] [H_A + H_E (2 - a^2 k^2) - 8\pi M/3].$$

Here a is the lattice constant, M the magnetic moment per unit volume, k the wave vector, θ_k the angle between the magnetization and the wave vector, and H_A and H_B the anisotropy and exchange fields. The reasons for discrepancies between these formulas and those given previously by other authors are discussed.

I. INTRODUCTION

HE purpose of this short note is to call attention to an error in recent treatments1 of the effect of dipolar interactions on the spin-wave spectrum of a cubic antiferromagnet. As an introduction we briefly review some pertinent (and well-known) results for a ferromagnet. In Sec. II we then calculate the spin-wave spectrum of an antiferromagnet.

For a cubic ferromagnet it has been shown^{2,3} that the spin-wave spectrum for an ellipsoid of revolution uniformly magnetized along its axis of rotation is

$$(\hbar\omega)^2 = [Da^2k^2 + g\beta(H - N_zM)] \times [Da^2k^2 + g\beta(H - N_zM + 4\pi M \sin^2\theta_k)]. \quad (1$$

Here a is the lattice constant, M the magnetic moment per unit volume, k the wave vector, θ_k the angle between the magnetization and the wave vector, H the external field, N_z the demagnetizing factor associated with the axis of rotation, and Da^2k^2 is the exchange energy. This dispersion law is valid for $ka \ll 1$, and $kR_0 \gg 1$, where R_0 is a characteristic dimension of the sample. This latter restriction is necessary since for $kR_0 \leq 1$ plane waves are no longer proper modes of the system. The exact "magnetostatic" modes, have been investigated by Walker4 and have been shown to merge smoothly into the plane-wave solutions as kR_0 becomes large.

In an unmagnetized (multidomain) sample one finds

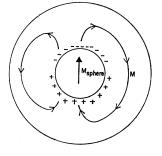
$$(\hbar\omega)^2 = \lceil Da^2k^2 \rceil \lceil Da^2k^2 + 4\pi g\beta M \sin^2\theta_k \rceil. \tag{2}$$

This relation is valid for $ka \ll 1$ and $kR_d \gg 1$, where R_d is the characteristic dimension of a domain. This result is easily demonstrated by considering a conceptual

sphere of radius R_s , such that $kR_s\gg 1$ and $R_s\ll R_d$. Since the wavelength is much less than R_s , the effect of the rest of the sample on the conceptual sphere is well approximated by an effective field. As is seen in Fig. 1, the only contribution to this effective field comes from the poles on the surface of the cavity. Since the sample is unmagnetized there are no poles on the surface of the sample. Furthermore it is assumed that the domains arrange themselves so that the effect of poles at the domain walls is negligibly small. Thus the dispersion law for an unmagnetized sample is found by using Eq. (1) and by taking $N_z = 4\pi/3$ and $H = 4\pi M/3$. Naturally, an extension of Eq. (2) valid for $kR_d \leq 1$ would require a knowledge of the exact arrangement of the domains in the sample. Such a detailed model should include the effects of lattice imperfections and impurities, since these factors affect significantly the arrangement of the domains.

If the dispersion law for an antiferromagnetic sphere consisting of a single domain were known, then formulas for (a) arbitrarily shaped samples, or for (b) the multidomain case could be derived as we have done for a ferromagnet. For this purpose we again consider a conceptual sphere whose radius R_s satisfies $kR_s\gg 1$ but with $R_s \ll R_d$. For an antiferromagnet the average magnetization is zero so that the effective field acting on the conceptual sphere is zero. This reasoning is valid both for a single-domain sample of arbitrary shape and for a multidomain sample. Hence in both cases one expects the dispersion law to be the same as for the singledomain sphere.

Fig. 1. A hypothetical arrangement of domains as viewed on a macroscopic scale. The field acting on the conceptual sphere is $+(4\pi/3)M_{\rm sphere}$ due to surface charges on the surface of the sphere caused by the magnetization of the rest of the sample. Since the sample is unmagnetized, there are no



surface poles.

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¹R. Loudon and P. Pincus, Phys. Rev. 132, 673 (1963). We shall refer to this paper as LP. Similar errors are made in F. R. Morgenthaler, Phys. Rev. Letters 11, 69 (1963); G. I. Urushadze, Zh. Eksperim. i Teor. Fiz. 39, 680 (1960) [English transl.: Soviet Phys.—JETP 12, 476 (1961)].

²A. M. Clogston, H. Suhl. L. R. Walker, and P. W. Anderson, J. Phys. Chem. Solids 1, 129 (1956).

³T. Holstein and H. Primakoff, Phys. Rev. 58, 1098 (1940).

⁴L. R. Walker, Phys. Rev. 105, 390 (1957).

II. THE SPIN-WAVE SPECTRUM OF AN ANTIFERROMAGNET

Let us first give a formal derivation of the spin-wave dispersion law using the Holstein-Primakoff transformation.3 Thus for spins on the A (up) sublattice

$$S^{z}(\mathbf{R}_{\mathbf{n}}) = S - a_{\mathbf{n}}^{\dagger} a_{\mathbf{n}}, \qquad (3a)$$

$$S^{+}(\mathbf{R}_{n}) \approx (2S)^{1/2} a_{n},$$
 (3b)

$$S^{-}(\mathbf{R}_{\mathbf{n}}) \approx (2S)^{1/2} a_{\mathbf{n}}^{\dagger}, \qquad (3c)$$

where $\mathbf{R}_{n} = (n_1 \hat{\imath} + n_2 \hat{\jmath} + n_3 \hat{k})a$. For spins on the B (down) sublattice

$$S^{z}(\mathbf{R}_{n}+\boldsymbol{\tau}) = -S + b_{n}^{\dagger}b_{n}, \qquad (4a)$$

$$S^+(\mathbf{R}_n + \boldsymbol{\tau}) \approx (2S)^{1/2} b_n^{\dagger},$$
 (4b)

$$S^{-}(\mathbf{R}_{\mathbf{n}} + \boldsymbol{\tau}) \approx (2S)^{1/2} b_{\mathbf{n}}, \qquad (4c)$$

where $\tau = \frac{1}{2}a(\hat{\imath} + \hat{\jmath} + \hat{k})$. These a and b operators obey boson commutation relations. We transform to momentum variables:

$$a_{\mathbf{n}}^{\dagger} = (2/N)^{1/2} \sum_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} \exp(-i\mathbf{k} \cdot \mathbf{R}_{\mathbf{n}}),$$
 (5a)

$$b_{\mathbf{n}}^{\dagger} = (2/N)^{1/2} \sum_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} \exp(-i\mathbf{k} \cdot \lceil \mathbf{R}_{\mathbf{n}} + \boldsymbol{\tau} \rceil), \quad (5b)$$

where N is the total number of lattice sites and k is a vector of the first Brillouin zone of a simplecubic lattice. Using these substitutions in the dipolar Hamiltonian.

$$3\mathfrak{C}_D = \frac{1}{2}g^2\beta^2 \sum_{i,j} \{\mathbf{S}_i \cdot \mathbf{S}_j - 3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})r_{ij}^{-2}\}r_{ij}^{-3}, \qquad (6)$$

one finds the quadratic part of the dipolar Hamiltonian to be

$$5C_{D} \approx \sum_{\mathbf{k}} [A(\mathbf{k}) + C(\mathbf{k})] (a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}})$$

$$+ \{ (a_{\mathbf{k}} a_{-\mathbf{k}} + b_{\mathbf{k}}^{\dagger} b_{-\mathbf{k}}^{\dagger}) B(\mathbf{k}) + \text{c.c.} \}$$

$$+ \{ E(\mathbf{k}) a_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} + \text{c.c.} \} ,$$

$$(7)$$

where c.c. means complex conjugate and

$$A(\mathbf{k}) = g^2 \beta^2 S \sum_{\mathbf{n}} \left\{ \frac{3 \cos^2 \theta - 1}{R_{\mathbf{n}^3}} \right\} (1 + \frac{1}{2} \exp(i\mathbf{k} \cdot \mathbf{R_n})), \quad (8a)$$

$$B(\mathbf{k}) = -g^2 \beta^2 S \sum_{\mathbf{n}} \frac{3}{4} \left[\frac{\sin^2 \theta e^{-2i\varphi}}{R_n^3} \right] \exp(i\mathbf{k} \cdot \mathbf{R_n}) , \qquad (8b)$$

$$C(\mathbf{k}) = -g^2 \beta^2 S \sum_{\mathbf{n}} \left\{ \frac{3 \cos^2 \theta - 1}{|\mathbf{R}_{\mathbf{n}} + \boldsymbol{\tau}|^3} \right\},$$
 (8c)

$$D(\mathbf{k}) = \frac{1}{2}g^2\beta^2 S \sum_{\mathbf{n}} \left\{ \frac{3\cos^2\theta - 1}{|\mathbf{R}_{\mathbf{n}} + \boldsymbol{\tau}|^3} \right\} \exp(i\mathbf{k} \cdot [\mathbf{R}_{\mathbf{n}} + \boldsymbol{\tau}]), \quad (8d)$$

$$E(\mathbf{k}) = -\frac{3}{2}g^{2}\beta^{2}S \sum_{n} \left\{ \frac{\sin^{2}\theta e^{-2i\varphi}}{|\mathbf{R}_{n} + \tau|^{3}} \right\} \exp(i\mathbf{k} \cdot [\mathbf{R}_{n} + \tau]) . \quad (8e) \quad (\hbar\omega)^{2} = (g\beta)^{2}[H_{A} + H_{E}a^{2}k^{2}] \\ \times [H_{A} + H_{E}(2 - a^{2}k^{2}) - 8\pi M/3 + 8\pi M \sin^{2}\theta_{k}].$$

In these equations θ and φ are the polar angles of \mathbf{R}_n

or $R_n + \tau$ relative to the direction of magnetization. We have already used the condition that $kR_0\gg 1$ to infer

$$\sum_{\mathbf{n}} \left\{ 1 - \frac{3|Z_{\mathbf{n}} - Z_{\mathbf{n}'}|^2}{|\mathbf{R}_{\mathbf{n}} - \mathbf{R}_{\mathbf{n}'}|^2} \right\} |\mathbf{R}_{\mathbf{n}} - \mathbf{R}_{\mathbf{n}'}|^{-3} \exp(i\mathbf{k} \cdot [\mathbf{R}_{\mathbf{n}} - \mathbf{R}_{\mathbf{n}'}])$$
(9)

show an insignificant dependence on $R_{n'}$ unless $R_{n'}$ is very near the surface. Cohen and Keffer⁵ have studied these dipolar wave sums and conclude that such an approximate treatment of them is justified. As for the ferromagnet, for $kR_0 \lesssim 1$ one would find spin waves are no longer appropriate for the determination of the normal modes.

Where our treatment differs from that of LP1 is in the evaluation of the lattice sums of Eq. (8). We take

$$A(\mathbf{k}) = 2\pi g\beta M \left[1 - \left(2N_z/3N_0 \right) - \cos^2 \theta_k \right], \quad (10a)$$

$$B(\mathbf{k}) = \pi g \beta M \sin^2 \theta_k e^{-2i\varphi_k}, \qquad (10b)$$

$$C(\mathbf{k}) = -\frac{4}{3}\pi g\beta M [1 - N_z/N_0], \qquad (10c)$$

$$D(\mathbf{k}) = \frac{2}{3}\pi g\beta M \lceil 1 - 3\cos^2\theta_k \rceil, \tag{10d}$$

$$E(\mathbf{k}) = 2\pi g\beta M \sin^2\theta_k e^{-2i\varphi_k}. \tag{10e}$$

Here $N_0 = 4\pi/3$ is the value of N_z for a sphere and θ_k and φ_k are the polar angles of **k** relative to the direction of magnetization. The evaluations of LP differs from those given here in that they take

$$A(\mathbf{k}) = 2\pi g\beta M \sin^2 \theta_k, \qquad (11a)$$

$$C(\mathbf{k}) = 0, \tag{11b}$$

$$D(\mathbf{k}) = 2\pi g\beta M \sin^2 \theta_k. \tag{11c}$$

Our evaluations are consistent with those of Holstein and Primakoff³ when $N_z=0$ as they have assumed. Furthermore Eqs. (10) are consistent with the formulas given by Cohen and Keffer.⁵ Note that the Hamiltonian is independent of N_z as expected since only the combination $A(\mathbf{k}) + C(\mathbf{k})$ appears. The harmonic-oscillator frequencies are readily determined from the full Hamiltonian

$$3C = g\beta H_E \sum_{\mathbf{k}} \left\{ a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} + \gamma_{\mathbf{k}} a_{\mathbf{k}}^{\dagger} b_{\mathbf{k}}^{\dagger} + \gamma_{\mathbf{k}} a_{\mathbf{k}} b_{\mathbf{k}} \right\}$$

$$+ g\beta H_A \sum_{\mathbf{k}} \left\{ a_{\mathbf{k}}^{\dagger} a_{\mathbf{k}} + b_{\mathbf{k}}^{\dagger} b_{\mathbf{k}} \right\} + 3C_D.$$

$$(12)$$

Here H_E and H_A are the exchange and anisotropy fields, respectively, and $\gamma_k = z^{-1} \sum \exp(i\mathbf{k} \cdot \mathbf{\delta})$, where z is the number of nearest neighbors and the sum is taken over all nearest-neighbor vectors 8. The two branches of the frequency spectrum are found to be

$$(\hbar\omega)^{2} = (g\beta)^{2} [H_{A} + H_{E}a^{2}k^{2}] \times [H_{A} + H_{E}(2 - a^{2}k^{2}) - 8\pi M/3],$$
 (13a)

$$\hbar\omega^2 = (g\beta)^2 \lfloor H_A + H_E a^2 k^2 \rfloor
\times \lceil H_A + H_E (2 - a^2 k^2) - 8\pi M/3 + 8\pi M \sin^2 \theta_k \rceil.$$
(13b)

⁵ M. H. Cohen and F. Keffer, Phys. Rev. 99, 1128 (1955).

These results can also be derived using the torque equations,

$$\frac{d}{dt}\mathbf{S}(\mathbf{R}) = (g\beta/\hbar)\mathbf{S}(\mathbf{R}) \times \mathbf{H}_{\text{eff}}(\mathbf{R}) , \qquad (14a)$$

$$\frac{d}{dt}\mathbf{S}(\mathbf{R}) = (g\beta/\hbar)\mathbf{S}(\mathbf{R}) \times \mathbf{H}_{eff}(\mathbf{R}) , \qquad (14a)$$

$$\frac{d}{dt}\mathbf{S}(\mathbf{R}+\tau) = (g\beta/\hbar)\mathbf{S}(\mathbf{R}+\tau) \times \mathbf{H}_{eff}(\mathbf{R}+\tau) . \qquad (14b)$$

Here $\mathbf{H}_{\text{eff}}(\mathbf{R})$ is the effective field acting on the spin at **R.** We only consider the contributions to $\mathbf{H}_{eff}(\mathbf{R})$ from dipolar fields, since LP correctly treat exchange and anisotropy fields. We suppose that only spin waves of wavelength less than k_{max} are appreciably thermally excited. We first consider the effect of spins within a distance R_s from R_s , where $k_{\text{max}}R_s \ll 1$, $R_s \gg a$. Within this sphere the spins are essentially in phase so that, due to the assumed cubic symmetry of the lattice, the spins of each sublattice create zero dipolar field at each lattice site. The dipolar fields from the remaining spins are calculated by replacing the lattice sum by an integral. The dipolar fields can then be thought of as arising from (A) a volume distribution of charge density $-\nabla \cdot \mathbf{M}(\mathbf{R})$, (B) a distribution of charge density $M(R) \cdot n$ on the surface of the sample, and (C) a similar distribution on the surface of the cavity of radius R_s . LP treat the contributions (A) and (B) correctly. Contribution (B) is zero both for the case of a single-domain and for the case of a multidomain sample. Since we take $kR_0\gg 1$, the

spatial variation of any poles on the surface of the sample is rapid in comparison to the sample dimensions. LP ignore the contribution (C), although it is nonzero. It is easy to see that this contribution to the dipolar field is

$$\delta \mathbf{H}_{\mathrm{eff}}(\mathbf{R}) = (4\pi/3) [M_A(\mathbf{R}) + M_B(\mathbf{R})], \qquad (15)$$

where, since $kR_{\epsilon}\ll 1$, we can take the spins on the surface of the conceptual sphere to be exactly in phase with those near the center of the sphere. Although the time average of this contribution is zero, the oscillating transverse components which are nonzero do modify the torque equations. When this term is added to the torque equation of LP our Eq. (13) follows.

Admittedly the difference between our results (13) and those of LP,

$$(\hbar\omega)^2 = (g\beta)^2 [H_A + 2H_A H_E + 2H_E^2 a^2 k^2],$$
 (16a)

$$(\hbar\omega)^2 = (g\beta)^2 [H_A^2 + 2(H_A + H_E a^2 k^2)]$$

$$\times (H_E + 4\pi M \sin^2 \theta_k)$$
, (16b)

are insignificant under normal conditions. Only in the rather unusual case when $4\pi M$ becomes comparable to the exchange field is the difference between Eqs. (13) and (16) important. It is interesting, however, to note that there is a criterion for the stability of the assumed antiferromagnetic array. This condition, which follows from the requirement that the frequencies in Eq. (13) be real, is

$$H_A + 2H_E > 8\pi M/3$$
. (17)

This result is not surprising since the assumed configuration is not that of lowest energy for spins interacting only via dipolar coupling.7 In contrast, the spectrum of LP [Eq. (16)] indicates no instability since the righthand side of Eq. (16) can never become negative.

⁶ Note the difference between the argument used here and the corresponding one in the Introduction. In the Introduction we chose the radius of the conceptual sphere R_s large so that $kR_s\gg 1$, thus ensuring that the motion of spins of the rest of the sample was incoherent with that of most of spins in the sphere. Then it is possible to replace the effect of the rest of the sample by an effective field. In contrast, here we take R_s to be small so that the motion of the spins in the conceptual sphere and that of the neighboring spins are coherent. In this case we must take account of the oscillating transverse components of the dipolar fields.

⁷ J. M. Luttinger and L. Tisza, Phys. Rev. 70, 954 (1946);