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Temperature Dependence of the Resonance Frequency of Ferrimagnets and Antiferromagnets*

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A spin-wave calculation of the temperature dependence of the resonance frequency of a two-sublattice ferrimagnet leads to a modification of the macroscopic result at low temperatures which may be conveniently represented by a temperature-dependent Weiss field parameter

$$\lambda(T)=\lambda_W\left(\frac{\gamma_2 M_1(0)+\gamma_1 M_2(0)}{\gamma_2 M_1(T)+\gamma_1 M_2(T)}\right)^{1/2}.$$

The analysis for the antiferromagnet is consistent with the Kanamori-Tachiki semiphenomenological spin-wave theory. The complete Nagamiya-Keffer-Kittel formula is recovered provided that $\chi_\perp$ takes the value predicted by linear spin-wave theory rather than molecular field theory.

1. INTRODUCTION

The resonance frequency of a two-sublattice ferrimagnet in an applied field H and anisotropy fields $H_A^{(1)}$ and $H_A^{(2)}$ on the sublattices 1 and 2, respectively, is [1]

$$2\omega=\pm[\gamma_1(H+H_A^{(1)})+\gamma_2(H-H_A^{(2)})+\lambda(\gamma_1 M_2-M_1\gamma_2)] +\{\lambda^2(\gamma_1 M_2-M_1\gamma_2)^2+2\lambda[\gamma_1(H+H_A^{(1)})-\gamma_2(H-H_A^{(2)})] \times(\gamma_1 M_2+\gamma_2 M_1)+[\gamma_1(H+H_A^{(1)})-\gamma_2(H-H_A^{(2)})]^2\}^{1/2}. \quad (1.1)$$

Here $\gamma_1=g_1\mu_B$, $\gamma_2=g_2\mu_B$, and λ is the Weiss field parameter. M_1 and M_2 are the sublattice magnetizations. Antiferromagnetic coupling is assumed. Equation (1.1) was derived in the molecular field approximation which one might expect to be valid at high temperatures when spin correlations are unimportant. The subject of this paper is a spin-wave analysis of a two-sublattice ferrimagnet or an antiferromagnet in which the temperature dependence of the resonance frequency arises from spin-wave interactions. In a high-temperature approximation, the spin-wave theory yields (1.1) exactly, if appropriate substitutions of effective fields for microscopic parameters are made. At low temperatures, however, the spin-wave results differ from (1.1), but in such a manner that the form of (1.1) is preserved provided that the following substitution is made:

$$\lambda\to\lambda(T)=\lambda\left(\frac{\gamma_2 M_1(0)+\gamma_1 M_2(0)}{\gamma_2 M_1(T)+\gamma_1 M_2(T)}\right)^{1/2}. \quad (1.2)$$

Thus the use of a temperature-dependent Weiss field parameter presents the spin-wave results as a simple modification of the macroscopic theory at low temperatures.

A special case of the ferrimagnet is the antiferromagnet, which has been the subject of investigation by several authors. The macroscopic theory is due to Nagamiya[2] and Keffer and Kittel[3] (NKK). Keffer and Kittel obtained for the resonance frequency of an antiferromagnet, in the presence of an axial crystal field and applied field H, the expression

$$\frac{\omega}{\gamma}=\pm H\left(1-\frac{1}{2}\frac{x_\parallel}{x_\perp}\right)+\left[\frac{2K(T)}{x_\perp}+\left(\frac{1}{2}H\frac{x_\parallel}{x_\perp}\right)^2\right]^{1/2}, \quad (1.3)$$

where $x_\perp$ takes its molecular field value which is temperature independent, $K(T)$ is the uniaxial anisotropy constant, and $\chi_\parallel$, $\chi_\perp$ are the parallel and perpendicular susceptibilities, respectively. On the other hand, the Kanamori-Tachiki[4] phenomenological spin-wave theory suggests that, in zero applied field, the substitution of $\chi_\perp(T)$ for $\chi_\perp$ – where the temperature dependence of $\chi_\perp(T)$ is given by spin-wave theory – properly accounts for spin correlations omitted by (1.3). The Kanamori-Tachiki result is in good agreement with experiment.

Previously, Oguchi and Honma[5] had calculated the temperature dependence of the resonance frequency in an applied field by explicitly treating spin-wave interactions. They diagonalized the bilinear spin-wave Hamiltonian and applied the transformation to terms of fourth order in spin-wave operators (such terms arise from expansion of the Holstein-Primakoff[6] substitutions for Bose operators). This essentially perturbative treatment led to an ambiguity concerning the nature the spin-wave interaction terms – more specifically, spin-wave interactions arising from anisotropy and exchange were not separated correctly. Consequently, low-temperature corrections to the macroscopic results (1.1) were attributed to a change in the power of the anisotropy constant rather than the substitution $\chi_\perp\to\chi_\perp(T)$. Antiferromagnetic resonance will be treated as a special case of ferrimagnetic resonance in this paper.

Contrary to the Oguchi-Honma technique, the spin-wave interaction terms are subjected to a temperature-dependent Hartree-Fock approximation, so that the appropriate substitutions of measurable macroscopic parameters are quite transparent. The complete Keffer-Kittel result, including the term in H^2 [Eq. (1.3)] which did not appear in the Oguchi-Honma analysis, is recovered provided that $\chi_\perp(T)$ is substituted for $\chi_\perp$.

In Sec. 2 the ferrimagnetic system is examined by a microscopic-equations-of-motion method. The spin-wave energies so obtained reduce, in the limit of zero external field and anisotropy, to those of Nakamura and Bloch.[7] In Secs. 3 and 4 the special cases of antiferromagnetic and ferrimagnetic resonance, respectively, are discussed.

2. SPIN-WAVE ENERGIES

Let the system be characterized by the Hamiltonian

$$\mathcal{H}=\gamma_l H\sum_l (S_l^z)+\gamma_m H\sum_m (S_m^z)-B\sum_l (S_l^z)^2 - B\sum_m (S_m^z)^2-2\sum_{(l,m)} J_{lm}\vec{S}_l\cdot\vec{S}_m \quad , \tag{2.1}$$

where $\gamma_l=g_l\mu_B$ and $\gamma_m=g_m\mu_B\cdot\vec{S}_l$, $\vec{S}_m$ are spins on the sublattices l and m, respectively, and are not necessarily equal; J_{lm} is negative, resulting in antiferromagnetic coupling, and (l, m) denotes a sum over pairs. B is a uniaxial anisotropy parameter and the applied field H is in the negative Z direction.

Spin-wave operators $\vec{S}_q^{(l)}$, $\vec{S}_q^{(m)}$ are defined

$$\vec{S}_l=(1/\sqrt{N})\sum_{\vec{q}}\vec{S}_{\vec{q}}^{(l)}e^{i\vec{q}\cdot\vec{R}_l} \quad , \tag{2.2a}$$

$$\vec{S}_m=(1/\sqrt{N})\sum_{\vec{q}}\vec{S}_{\vec{q}}^{(m)}e^{i\vec{q}\cdot\vec{R}_m} \quad , \tag{2.2b}$$

where N is the number of atoms per unit volume per sublattice. The spin-wave operators obey the commutation relations

$$[S_{\vec{q}}^z, S_{\vec{q}'}^{\pm}]=\pm(1/\sqrt{N})S_{\vec{q}+\vec{q}'}^{\pm} \quad , \tag{2.3a}$$

$$[S_{\vec{q}}^+, S_{\vec{q}'}^-]=(2/\sqrt{N})S_{\vec{q}+\vec{q}'}^z \quad . \tag{2.3b}$$

From (2.1) and (2.2), the Hamiltonian becomes

$$\mathcal{H}=-\sum_{\vec{q}} J(\vec{q})[\tfrac{1}{2}(S_{\vec{q}}^{(l)+}S_{-\vec{q}}^{(m)-}+S_{\vec{q}}^{(l)-}S_{-\vec{q}}^{(m)+})+S_{\vec{q}}^{(m)z}S_{\vec{q}}^{(l)z}] - B\sum_{\vec{q}} S_{\vec{q}}^{(l)z}S_{-\vec{q}}^{(l)z}-B\sum_{\vec{q}} S_{\vec{q}}^{(m)z}S_{-\vec{q}}^{(m)z} +\gamma_l H(\sqrt{N})S_o^{(l)z}+\gamma_m H(\sqrt{N})S_o^{(m)z} \quad , \tag{2.4}$$

where $$J(\vec{q})=\sum_j J_{ij}e^{i\vec{q}\cdot\vec{R}_{ij}} \quad . \tag{2.5}$$

Then the equations of motion for $S_{\vec{q}'}^{(l)z}_{(m)}$ are

$$[S_{\vec{q}_1}^{(l)+},\mathcal{H}]=-(2/\sqrt{N})\sum_{\vec{q}} J(\vec{q})(S_{\vec{q}_1-\vec{q}}^{(l)z}S_{\vec{q}}^{(m)+}-S_{\vec{q}}^{(m)z}S_{\vec{q}_1-\vec{q}}^{(l)+}) +(B/\sqrt{N})\sum_{\vec{q}}(2S_{\vec{q}_1-\vec{q}}^{(l)+}S_{\vec{q}}^{(l)z}+S_{\vec{q}_1}^{(l)+}/\sqrt{N})-\gamma_l HS_{\vec{q}_1}^{(l)+} , \tag{2.6a}$$

$$[S_{\vec{q}_1}^{(m)+},\mathcal{H}]=(2/\sqrt{N})\sum_{\vec{q}} J(\vec{q})(S_{\vec{q}_1-\vec{q}}^{(m)z}S_{\vec{q}}^{(l)+}-S_{\vec{q}}^{(l)z}S_{\vec{q}_1-\vec{q}}^{(m)+}) +(B/\sqrt{N})\sum_{\vec{q}}(2S_{\vec{q}_1-\vec{q}}^{(m)+}S_{\vec{q}}^{(m)z}+S_{\vec{q}_1}^{(m)+}/\sqrt{N})-\gamma_m HS_{\vec{q}_1}^{(m)+} . \tag{2.6b}$$

The following redefinitions of $S_{\vec{q}}^{(l)\pm}_{(m)}$ are appropriate:

$$S_{\vec{q}}^{(l)\pm}\to S_{\vec{q}}^{\pm}, \quad S_{\vec{q}}^{(l)z}\to S_{\vec{q}}^z , \quad S_{\vec{q}}^{(m)\pm}\to T_{\vec{q}}^{\mp} , \quad S_{\vec{q}}^{(m)z}\to T_{\vec{q}}^z \ . \tag{2.7}$$

Complete ground-state alignment and spin magnitude are defined by

$$S_{\vec{q}}^{(z)}|0\rangle=-S(\sqrt{N})\delta_{\vec{q},0}|0\rangle; \quad T_{\vec{q}}^z|0\rangle=T(\sqrt{N})\delta_{\vec{q},0}|0\rangle , \tag{2.8}$$

where $|0\rangle$ is a hypothetical fully aligned ground state.

$S_{\vec{q}}^z$, $T_{\vec{q}}^z$ may be expressed as normal ordered products of spin-wave annihilation and creation operators by an expansion due to Wortis[8]:

$$S_{\vec{q}}^z=-S(\sqrt{N})\delta_{\vec{q},0}+(1/2S\sqrt{N})\sum_{\vec{q}'} S_{\vec{q}+\vec{q}'}^+S_{-\vec{q}'}^-+\cdots , \tag{2.9a}$$

$$T_{\vec{q}}^z=T(\sqrt{N})\delta_{\vec{q},0}-(1/2T\sqrt{N})\sum_{\vec{q}'} T_{\vec{q}+\vec{q}'}^+T_{-\vec{q}'}^-+\cdots . \tag{2.9b}$$

Substitution of (2.9) into (2.6) allows the equations of motion to be decoupled in a symmetrical fashion.[9] We shall use slightly different expansions from (2.9), however. Thus,

$$S_{\vec{q}}^z=-S(\sqrt{N})\delta_{\vec{q},0}+(1/\sqrt{N})\sum_{\vec{q}'} a_{\vec{q}+\vec{q}'}^+a_{\vec{q}'}^-+\ \ldots , \tag{2.10a}$$

$$T_{\vec{q}}^z=T(\sqrt{N})\delta_{\vec{q},0}-(1/\sqrt{N})\sum_{\vec{q}'} b_{\vec{q}+\vec{q}'}^+b_{\vec{q}'}^-+\ \ldots , \tag{2.10b}$$

which imply

$$S_{\vec{q}}^+=(2T)^{1/2}a_{\vec{q}}^+, \quad S_{\vec{q}}^-=(2S)^{1/2}a_{-\vec{q}}^+ , \tag{2.11a}$$

$$T_{\vec{q}}^+=(2T)^{1/2}b_{\vec{q}}^+, \quad T_{\vec{q}}^-=(2S)^{1/2}b_{-\vec{q}}^- , \tag{2.11b}$$

so that $a_{\vec{q}}^{\pm}$, $b_{\vec{q}}^{\pm}$ obey boson commutation relations. Equations (2.10) and (2.11) may be identified with the expansion used by Brout[10] for just the Heisenberg Hamiltonian.

Equations (2.10) and (2.11) are substituted into (2.6) and the equations of motion linearized in the following manner. Only terms of one and three operators are retained in the equations of motion. All fluctuation parts of the three-operator terms are neglected and these terms are then decoupled symmetrically. It may be seen from (2.6) that the relevant nonzero expectation values are $\langle a_{\vec{q}}^+a_{\vec{q}}^-\rangle$, $\langle b_{\vec{q}}^+b_{\vec{q}}^-\rangle$, $\langle a_{\vec{q}}^+b_{\vec{q}}^+\rangle$, and $\langle a_{\vec{q}}^-b_{\vec{q}}^-\rangle$. Therefore, the linearized equations of motion may be written

$$[a_{\vec{q}_1}^+,\mathcal{H}]=-\Delta A_{\vec{q}_1}a_{\vec{q}_1}^+-A_{\vec{q}_1}^{(3)}a_{\vec{q}_1}^+-\Delta B_{\vec{q}_1}b_{\vec{q}_1}^--B_{\vec{q}}^{(3)}b_{\vec{q}_1}^- , \tag{2.12a}$$

$$[b_{\vec{q}_1}^-,\mathcal{H}]=-\Delta A_{\vec{q}_1}b_{\vec{q}_1}^-+A_{\vec{q}_1}^{(3)}b_{\vec{q}_1}^--\Delta B_{\vec{q}_1}a_{\vec{q}_1}^++B_{\vec{q}_1}^{(3)}a_{\vec{q}_1}^+ \quad , \tag{2.12b}$$

where $\Delta A_{\vec{q}} = \frac{1}{2}[A_{\vec{q}}^{(1)} - A_{\vec{q}}^{(2)} + (\gamma_l + \gamma_m)H]$, $\Delta B_{\vec{q}} = \frac{1}{2}(B_{\vec{q}}^{(1)} - B_{\vec{q}}^{(2)})$, (2.13)

$A_{\vec{q}}^{(3)} = \frac{1}{2}[A_{\vec{q}}^{(1)} + A_{\vec{q}}^{(2)} + (\gamma_l - \gamma_m)H]$, $B_{\vec{q}}^{(3)} = \frac{1}{2}(B_{\vec{q}}^{(1)} + B_{\vec{q}}^{(2)})$, (2.14)

and $A_{\vec{q}_1}^{(1)} = (2S-1)B[1 - (2/NS)\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle] - 2TJz + (2Jz/N)(T/S)^{1/2}\sum_{\vec{q}}\gamma_{\vec{q}} \langle a_{\vec{q}}^- b_{\vec{q}}^- \rangle + (2Jz/N)\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle$, (2.15a)

$A_{\vec{q}_1}^{(2)} = (2T-1)B[1 - (2/NT)\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle] - 2SJz + (2Jz/N)(S/T)^{1/2}\sum_{\vec{q}}\gamma_{\vec{q}} \langle a_{\vec{q}}^+ b_{\vec{q}}^+ \rangle + (2Jz/N)\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle$, (2.15b)

$B_{\vec{q}_1}^{(1)} = -2(TS)^{1/2} Jz\gamma_{\vec{q}_1} + (2Jz/N)(T/S)^{1/2}\gamma_{\vec{q}_1}\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle + (2Jz/N)\sum_{\vec{q}}\gamma_{\vec{q}+\vec{q}_1} \langle a_{\vec{q}}^+ b_{\vec{q}}^+ \rangle$, (2.16a)

$B_{\vec{q}_1}^{(2)} = -2(TS)^{1/2} Jz\gamma_{\vec{q}_1} + (2Jz/N)(S/T)^{1/2}\gamma_{\vec{q}_1}\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle + (2Jz/N)\sum_{\vec{q}} \gamma_{\vec{q}+\vec{q}_1} \langle a_{\vec{q}}^- b_{\vec{q}}^- \rangle$, (2.16b)

where $\gamma_{\vec{q}} = (1/z)\sum_j e^{i\vec{q}\cdot R_{ij}}$. (2.17)

The coupled equations (2.12) are diagonalized by the transformations

$$c_{\vec{q}}^+ = \cosh\theta_{\vec{q}} a_{\vec{q}}^+ + \sinh\theta_{\vec{q}} b_{\vec{q}}^- , \quad c_{\vec{q}}^- = (c_{\vec{q}}^+)^* , \quad (2.18a)$$

$$d_{\vec{q}} = \sinh\theta_{\vec{q}} a_{\vec{q}}^+ + \cosh\theta_{\vec{q}} b_{\vec{q}}^- , \quad d_{\vec{q}}^- = (d_{\vec{q}}^+)^* , \quad (2.18b)$$

$$\tanh 2\theta_{\vec{q}} = B_{\vec{q}}^{(3)}/A_{\vec{q}}^{(3)} , \quad (2.19)$$

and also $c_{\vec{q}}^+ c_{\vec{q}}^- = N_{\vec{q}}$, $d_{\vec{q}}^+ d_{\vec{q}}^- = M_{\vec{q}}$. (2.20)

The solutions to (2.12) are then (zero-point energies neglected)

$$E_{\vec{q}} = \pm \Delta A_{\vec{q}} + [(A_{\vec{q}}^{(3)})^2 - B_{\vec{q}}^{(1)} B_{\vec{q}}^{(2)}]^{1/2} . \quad (2.21)$$

For long-wavelength magnons, (2.21) reduces to the Nakamura-Bloch result[7] for ferrimagnets in the limit of vanishing applied field and anisotropy, and to Nagai's result[11] for an antiferromagnet in the limit of vanishing applied field.

The solution (2.21) will be examined for the special case of the uniform mode, and the results will be expressed in a form suitable for comparison with the macroscopic theories.

3. ANTIFERROMAGNETIC RESONANCE FREQUENCY

In this case

$$S = T, \quad \gamma_1 = \gamma_2 = \gamma . \quad (3.1)$$

The solutions for the excitation energies are given by (2.21), (2.15), and (2.16). Since $B_{\vec{q}}^{(3)} \gg \Delta B_{\vec{q}}$, a convenient expansion for the product $B_{\vec{q}}^{(1)} B_{\vec{q}}^{(2)}$ is

$$-B_{\vec{q}}^{(1)} B_{\vec{q}}^{(2)} \simeq -(B_{\vec{q}}^{(3)})^2 + (\Delta B_{\vec{q}})^2 , \quad (3.2)$$

whence (2.21) becomes, for the uniform modes,

$$E_{\vec{q}=0} = \pm(\Delta A_0) + [(A_0^{(3)} - B_0^{(3)})(A_0^{(3)} + B_0^{(3)}) + (\Delta B_0)^2]^{1/2} . \quad (3.3)$$

If $\theta = (2S-1)B/2S|J|Z \ll 1$, i.e., the anisotropy is small compared with exchange, we have, from (2.13) – (2.16) and (2.18) – (2.20),

$$\Delta A_0 = \Delta B_0 = -(2S|J|Z/2NS)\sum_{\vec{q}} \langle N_{\vec{q}} - M_{\vec{q}} \rangle , \quad (3.4)$$

$$A_0^{(3)} - B_0^{(3)} = 2S|J|Z\theta[1 - (1/NS)\cosh 2\theta_{\vec{q}} \langle N_{\vec{q}} + M_{\vec{q}} \rangle] , \quad (3.5)$$

$$A_0^{(3)} + B_0^{(3)} = 4S|J|Z[1 - (1/2NS) \times \sum_{\vec{q}} (\cosh 2\theta_{\vec{q}} - \gamma_{\vec{q}} \sinh 2\theta_{\vec{q}}) \langle N_{\vec{q}} + M_{\vec{q}} \rangle] . \quad (3.6)$$

It may be shown that, if zero-point energies are neglected, the temperature-dependent perpendicular susceptibility is given, in the spin-wave approximation, by[12]

$$\chi_\perp(T) = \frac{N\gamma^2}{2|J|Z}\left(1 - \frac{2|J|ZS}{2NS}\sum_{\vec{q}} \frac{\gamma_{\vec{q}}^2}{E_{\vec{q}}} \langle N_{\vec{q}} + M_{\vec{q}} \rangle\right) . \quad (3.7)$$

Using (2.18) and (2.19), (3.7) becomes

$$\frac{1}{\chi_\perp(T)} = \frac{2|J|Z}{N\gamma^2}\left(1 + \frac{1}{2NS}\sum_{\vec{q}} \gamma_{\vec{q}} \sinh 2\theta_{\vec{q}} \langle N_{\vec{q}} + M_{\vec{q}} \rangle\right) . \quad (3.8)$$

The anisotropy constant for an antiferromagnet is given by[5]

$$K(T) = NS(2S-1)B\left(1 - \frac{3}{2NS}\sum_{\vec{q}} \cosh 2\theta_{\vec{q}} \langle N_{\vec{q}} + M_{\vec{q}} \rangle\right) , \quad (3.9)$$

where the magnetization is

$$M(T)/M(0) = 1 - (1/2NS)\sum_{\vec{q}} \cosh 2\theta_{\vec{q}} \langle N_{\vec{q}} + M_{\vec{q}} \rangle . \quad (3.10)$$

The ratio of the parallel to perpendicular susceptibilities is given by[5]

$$\gamma H \frac{\chi_\parallel}{\chi_\perp} = \frac{2S|J|Z}{NS} \langle N_{\vec{q}} - M_{\vec{q}} \rangle = -2(\Delta A_0) = -2(\Delta B_0) . \quad (3.11)$$

From (3.5), (3.6), and (3.8), (3.9),

$$(A_0^{(3)} - B_0^{(3)})(A_0^{(3)} + B_0^{(3)}) = \gamma^2[2K(T)/\chi_\perp(T)] ; \quad (3.12)$$

therefore, from (3.3), (3.11), and (3.12),

$$E_{\vec{q}=0} = \pm\gamma H\left(1 - \frac{1}{2}\frac{\chi_{\parallel}}{\chi_{\perp}}\right) + \gamma\left[\frac{2K(T)}{\chi_{\perp}(T)} + \left(\tfrac{1}{2}H\frac{\chi_{\parallel}}{\chi_{\perp}}\right)^2\right]^{1/2} . \quad (3.13)$$

Equation (3.13) differs from the Keffer-Kittel result only in that $\chi_\perp(T)$ [Eq. (3.7)] is used rather than the molecular field result, and agrees with the Kanamori-Tachiki result in the limit of vanishing applied field. The present expression also agrees with the Oguchi-Honma result in the low- and high-temperature limits apart from the term in H^2. Oguchi and Honma claimed that it would be necessary to include terms up to the sixth order in spin operators to obtain this term, but the reason for its absence in their result is a consequence of their incomplete treatment of fourth-order terms.

4. FERROMAGNETIC RESONANCE FREQUENCY

In the ferrimagnetic case, useful evaluation of Eqs. (2.13)–(2.16) for the $\vec{q}=0$ modes may be made in low- and high-temperature limits.

A. High Temperatures

The spin-wave modes are uniformly populated, therefore[5]

$$\textstyle\sum_{\vec{q}} \gamma_{\vec{q}} \langle a_{\vec{q}}^- b_{\vec{q}}^- \rangle = \sum_{\vec{q}} \gamma_{\vec{q}} \langle a_{\vec{q}}^+ b_{\vec{q}}^+ \rangle \simeq 0 . \quad (4.1)$$

Then, from (2.15) and (2.16),

$$A_0^{(1)} = (2S-1)B[1 - (2/NS)\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle] + 2T|J|Z[1 - (1/NT)\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle] , \quad (4.2)$$

$$A_0^{(2)} = (2S-1)B[1 - (2/NT)\textstyle\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle] + 2S|J|Z[1 - (1/NS)\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle] , \quad (4.3)$$

$$B_0^{(1)} = 2(TS)^{1/2}|J|Z[1 - (1/NS)\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle] , \quad (4.4)$$

$$B_0^{(2)} = 2(TS)^{1/2}|J|Z[1 - (1/NT)\textstyle\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle] . \quad (4.5)$$

On the other hand, the sublattice magnetizations $M(T)$ and reduced magnetizations $m(T)$ are given by (the sublattices l, m are now denoted by 1, 2)

$$M_1(T) = \gamma_1 NS\left(1 - \frac{1}{NS}\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle\right), \quad m_1(T) = \frac{M_1(T)}{M_1(0)} , \quad (4.6a)$$

$$M_2(T) = \gamma_2 NT\left(1 - \frac{1}{NT}\textstyle\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle\right), \quad m_2(T) = \frac{M_2(T)}{M_2(0)} , \quad (4.6b)$$

and sublattice anisotropy constants and anisotropy fields may be defined as

$$k_1(T) = k_1(0)(m_1)^3, \quad k_1(0) = NS(S - \tfrac{1}{2})B ,$$
$$k_2(T) = k_2(0)(m_2)^3, \quad k_2(0) = NT(T - \tfrac{1}{2})B ; \quad (4.7)$$

$$\gamma_1 H_A^{(1)} = 2k_1(T)/(m_1) = 2k_1(0)(m_1)^2 ,$$
$$\gamma_2 H_A^{(2)} = 2k_2(0)(m_2)^2 . \quad (4.8)$$

The correspondence between the Weiss field and the Heisenberg Hamiltonian is given by

$$2S|J|Z \to M_1(0)\lambda\gamma_2 , \quad 2T|J|Z \to M_2(0)\lambda\gamma_1 . \quad (4.9)$$

Thus, from (4.2)–(4.9),

$$A_0^{(1)} - A_0^{(2)} = \gamma_1 H_A^{(1)} - \gamma_2 H_A^{(2)} + \lambda[\gamma_1 M_2(T) - \gamma_2 M_2(T)] , \quad (4.10a)$$

$$A_0^{(1)} + A_0^{(2)} = \gamma_1 H_A^{(1)} + \gamma_2 H_A^{(2)} + \lambda[\gamma_1 M_2(T) + \gamma_2 M_1(T)] , \quad (4.10b)$$

$$B_0^{(1)} B_0^{(2)} = M_1(T) M_2(T) \lambda^2 \gamma_1 \gamma_2 . \quad (4.10c)$$

Therefore, from (2.13), (2.14), (2.21), and (4.10) it may easily be shown that

$$2E_{\vec{q}=0} = \pm[\gamma_1(H_A^{(1)} + H) + \gamma_2(H - H_A^{(2)}) - \lambda(\gamma_2 M_1 - \gamma_1 M_2)] + \{\lambda^2(\gamma_2 M_1 - \gamma_1 M_2)^2 + 2\lambda(\gamma_2 M_1 + \gamma_1 M_2) \times [\gamma_1(H_A^{(1)} + H) + \gamma_2(H_A^{(2)} - H)] + [\gamma_1(H_A^{(1)} + H) + \gamma_2(H_A^{(2)} - H)]^2\}^{1/2} , \quad (4.11)$$

which is precisely Eq. (1.1), in agreement with the macroscopic theory.

B. Low Temperatures

Only long-wavelength spin waves are excited, hence[5]

$$\textstyle\sum_{\vec{q}} \gamma_{\vec{q}} \langle a_{\vec{q}}^{\pm} b_{\vec{q}}^{\pm} \rangle \to \sum_{\vec{q}} \langle a_{\vec{q}}^{\pm} b_{\vec{q}}^{\pm} \rangle . \quad (4.12)$$

Then, from (2.15), (2.16), (2.18), and (2.19),

$$A_0^{(1)} = H_A^{(1)}\gamma_1 + 2T|J|Z\{1 - (1/NT)\textstyle\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle + [1/2N(T+S)]\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- + b_{\vec{q}}^+ b_{\vec{q}}^- \rangle\} , \quad (4.13a)$$

$$A_0^{(2)} = H_A^{(2)}\gamma_2 + 2S|J|Z\{1 - (1/NS)\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle + [1/2N(T+S)]\sum \langle a_{\vec{q}}^+ a_{\vec{q}}^- + b_{\vec{q}}^+ b_{\vec{q}}^- \rangle\} , \quad (4.13b)$$

$$B_0^{(1)} = +2(TS)^{1/2}|J|Z\{1 - (1/NS)\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- \rangle + [1/2N(T+S)]\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- + b_{\vec{q}}^+ b_{\vec{q}}^- \rangle\} , \quad (4.13c)$$

$$B_0^{(2)} = 2(TS)^{1/2}|J|Z\{1 - (1/NT)\textstyle\sum_{\vec{q}} \langle b_{\vec{q}}^+ b_{\vec{q}}^- \rangle + [1/2N(T+S)]\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- + b_{\vec{q}}^+ b_{\vec{q}}^- \rangle\} . \quad (4.13d)$$

However, using (4.6)

$$1 + \frac{1}{N(S+T)}\textstyle\sum_{\vec{q}} \langle a_{\vec{q}}^+ a_{\vec{q}}^- + b_{\vec{q}}^+ b_{\vec{q}}^- \rangle = \frac{\gamma_2 M_1(0) + \gamma_1 M_2(0)}{\gamma_2 M_1(T) + \gamma_1 M_2(T)} . \quad (4.14)$$

Examination of (4.13) and (4.14) and comparison with (4.2)–(4.5) and (4.9) shows that at low temperatures the expression for the resonance frequency (4.11) may be used provided that $\lambda(T)$ is substituted for λ, where

$$\lambda(T)=\lambda\left[\frac{\gamma_2 M_1(0)+\gamma_1 M_2(0)}{\gamma_2 M_1(T)+\gamma_1 M_2(T)}\right]^{1/2} . \qquad (4.15)$$

5. CONCLUSION

The temperature dependence of the antiferromagnetic and ferrimagnetic resonance frequencies has been evaluated by spin-wave theory in a temperature-dependent Hartree-Fock approximation. Corrections to the macroscopic theory, due to spin correlations at low temperatures, are best represented for the antiferromagnet by a temperature-dependent perpendicular susceptibility, as is already well known. In ferrimagnets, when a static perpendicular susceptibility may not be defined in the absence of anisotropy, the same corrections are represented by an effective temperature dependence of the Weiss field parameter.

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NMR Study of Phase Transitions in Rochelle Salt*

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Nuclear-magnetic-resonance techniques have been used to measure the spontaneous polarization of ferroelectric Rochelle salt in the vicinity of the higher-temperature Curie point (T_C). The polarization was found to be proportional to $(T_C-T)^{1/2}$, but the technique did not allow measurements closer than 0.1 °C to T_C. In addition, the Na^{23} nuclear quadrupole coupling constant and field gradient asymmetry parameter in the low-temperature phase were found to be 1363 ± 3.4 kHz and 0.737 ± 0.004, respectively. These values are in accord with the model of Blinc, Petkovsek, Zupancic, and with the notion of an antiferroelectric low-temperature phase.

INTRODUCTION

The use of the Na^{23} nuclear magnetic resonance (NMR) to study ferroelectricity in Rochelle salt has been discussed in three earlier papers.[1–3] In the first of these,[1] the measurement of the Na^{23} electric quadrupole coupling constants in the ferroelectric phase and in both nonferroelectric phases of deuterated Rochelle salt is described. With the aid of a model proposed in the same paper, the experimental results are analyzed to determine the atomic displacements responsible for the ferroelectric behavior of the crystal. The second paper[2] makes use of the same model and a similar set of measurements on nondeuterated Rochelle salt to find values for the displacements causing ferroelectricity in that material. However, in the second case, the measurements were made only in the ferroelectric phase and the higher-temperature nonferroelectric phase. The third paper[3] describes methods of determining spontaneous polarization and domain characteristics from NMR measurements and the application of these methods to the study of radiation effects in Rochelle salt.

The present paper is concerned with further