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Statistical-Mechanical Theory of Resonance Susceptibility in Heisenberg Antiferromagnetism*

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The method of double-time temperature-dependent Green functions is combined with results from equilibrium molecular-field theory in order to study magnetic resonance in a Heisenberg two-sublattice antiferromagnet with spin value S per site, having isotropic exchange interactions (both inter- and intrasublattice) of essentially arbitrary range, and having uniaxial crystal-field single-ion-type anisotropy. Expressions found for the transverse resonance susceptibility are examined at the absolute zero of temperature and, by series expansion, at temperatures closely surrounding the Néel transition temperature T_N. The rutile crystal-structure ferrous fluoride (FeF_2) is used as an example of application. Besides being in reasonable agreement with results found from previous spin-wave and macroscopic theories, the present results offer some supplementary features (e.g., thermal behaviors of absorption amplitudes) for resonance phenomena in Heisenberg antiferromagnetism.

INTRODUCTION

THE theory of antiferromagnetic resonance (AFMR) has been studied by various authors,[1-8] one of the more recent methods being that of double-time temperature-dependent Green functions[9] applied to the problem by P'u Fu-ch'o[10] and by Anderson and Callen.[11] The spin systems studied by P'u Fu-ch'o and by Anderson and Callen are two-sublattice Heisenberg models having isotropic antiferromagnetic exchange interactions between spins on different sublattices and having uniaxial anisotropy. The uniaxial anisotropy terms used by these authors differ in that Anderson and Callen use a crystal-field single-ion type, while P'u Fu-ch'o considers a form containing the squares of sublattice magnetization operators. Besides using different decoupling approximations for the equations of motion for the Green functions, the AFMR results of P'u Fu-ch'o are actually more appropriate for the case of spin $S=\frac{1}{2}$ (owing to the use of Pauli spin operators) while Anderson and Callen consider, in regard to AFMR, the temperature dependence of the external magnetic field required to bring the spin system $\mathbf{k}=\mathbf{0}$ antiferromagnetic spin-wave modes into resonance with a specified signal frequency ω incorporating into their study spin-wave renormalization effects.

In order to further investigate physical features of the magnetic resonance problem, the present paper combines the Green-function method with results from equilibrium molecular-field theory to treat spin systems having isotropic exchange interactions and crystal-field-type anisotropy. More specifically, the present Hamiltonian is taken to be that for a two-sublattice Heisenberg model with arbitrary spin value S per site, having *strong isotropic antiferromagnetic* exchange interactions between spins on *different* sublattices in addition to *weak isotropic* (either ferromagnetic or antiferromagnetic) exchange interactions between spins on the *same* sublattice, and where the system is taken to have uniaxial crystal-field single-ion-type anisotropy. The ranges for the exchange interactions are taken as arbitrary only to the extent that the ordered state should have a preferred antiferromagnetic spin configuration. In the absence of a uniform, static external magnetic field $\mathbf{H}$, the present method is used to find first-approximation AFMR results for circularly polarized radiation which are examined as functions of temperature and are numerically compared with FeF_2 experimental data at the absolute zero of temperature. In the presence of the external magnetic field $\mathbf{H}$ (applied along the axis of anisotropy), the resonance susceptibility results obtained are examined by series expansion for temperatures close to the Néel transition temperature T_N.

These results are in reasonable agreement with spin-wave theory[4-6,11] and offer some interpretation for results derived by macroscopic theories.[1-3] Because of the form of the Hamiltonian and the fact that earliest-stage "Tyablikov" decoupling approximations are entered into the hierarchy for the equations of motion for the Green functions along with molecular-field-type expressions, the results found are necessarily incomplete and only suggestive of a more correct treatment. For example, since relaxation and other irreversible effects have not been incorporated into the study because of the form of the Hamiltonian, the resulting

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expressions for the absorption-line profiles offer no linewidth (dissipation) information but rather show approximate thermal behaviors of the line locations and amplitudes. However, these results show some features (e.g., the latter amplitude information) not found in renormalized spin-wave[11] and macroscopic theories[1–3] of resonance susceptibility.

HEISENBERG SPIN-SYSTEM HAMILTONIAN

Consider a two-identical sublattice Heisenberg-model spin system with spin value S per site, having strong isotropic antiferromagnetic exchange interactions between spins situated on different sublattices in addition to weak isotropic (ferromagnetic or antiferromagnetic) exchange interactions between spins on the same sublattice, and having uniaxial crystal-field single-ion-type anisotropy. It is assumed, for the present,[12] that the exchange interactions are arbitrary in range only to the extent that the strong antiferromagnetic couplings offer a preferred spin configuration for the ordered state such that one sublattice may be termed the "up" sublattice and the other the "down" sublattice, where each is sublattice translationally invariant. Placing a uniform, static external magnetic field along the axis of anisotropy (taken as the z axis), the system Hamiltonian is given by

$$\begin{aligned}\mathcal{H}=-g\mu_\beta H\sum_f S_{fz}-\sum_{\alpha i,\beta j}J_{ij}\mathbf{S}_{\alpha_i}\cdot\mathbf{S}_{\beta_j}\\ -\sum_{\alpha i,\alpha j}J_{ij}'\mathbf{S}_{\alpha_i}\cdot\mathbf{S}_{\alpha_j}-\sum_{\beta i,\beta j}J_{ij}'\mathbf{S}_{\beta_i}\cdot\mathbf{S}_{\beta_j}\\ -D\Big(\sum_\alpha S_{\alpha z}^2+\sum_\beta S_{\beta z}^2\Big),\end{aligned}\tag{1}$$

where the **S** operators are localized spin angular-momentum operators, where α, β sites are sites on the up and down sublattices, respectively, where g, μ_B, and D are the splitting factor, Bohr magneton, and anisotropy constant, respectively, and where the following relations are assumed for the exchange constants:

$$J_{ij}=J_{ji}<0,\qquad J_{ij}'=J_{ji}',\qquad \Big|\sum_j J_{ij}\Big|\gg\Big|\sum_j J_{ij}'\Big|,\tag{2}$$

and

$$J_{ii}=J_{ii}'=0.$$

In (2), the first two expressions simply state, respectively, the existence of symmetric antiferromagnetic exchange coupling between spins situated on different sublattices and symmetric exchange coupling between spins on the same sublattice, while the last inequality states that the inter-sublattice antiferromagnetic coupling of any spin with the environment is much stronger than the intra-sublattice coupling of the same spin with the environment. The Hamiltonian (1) may be rewritten as

$$\begin{aligned}\mathcal{H}=-g\mu_B H\sum_f S_{fz}-\sum_{\alpha i,\beta j}J_{ij}\big[\tfrac{1}{2}(S_{\alpha_i}{}^+S_{\beta_j}{}^-+S_{\alpha_i}{}^-S_{\beta_j}{}^+)\\ +S_{\alpha_i z}S_{\beta_j z}\big]-\sum_{\alpha i,\alpha j}J_{ij}'(S_{\alpha_i}{}^-S_{\alpha_j}{}^++S_{\alpha_i z}S_{\alpha_j z})\\ -\sum_{\beta i,\beta j}J_{ij}'(S_{\beta_i}{}^-S_{\beta_j}{}^++S_{\beta_i z}S_{\beta_j z})\\ -D\Big(\sum_\alpha S_{\alpha z}^2+\sum_\beta S_{\beta z}^2\Big),\end{aligned}\tag{3}$$

where, as usual, $S^\pm\equiv S_x\pm iS_y$ and where use was made of the facts that $J_{ii}=J_{ii}'=0$ and spin operators associated with different lattice sites commute.

EQUATIONS OF MOTION FOR THE GREEN FUNCTIONS

The double-time temperature-dependent retarded Green function may be defined as[13]

$$\langle\langle A(t);B(t')\rangle\rangle_r\equiv-i\theta(t-t')\langle[A(t),B(t')]\rangle,\tag{4a}$$

where

$$\begin{aligned}A(t)&\equiv\exp(i\mathcal{H}t)A\exp(-i\mathcal{H}t),\\ B(t)&\equiv\exp(i\mathcal{H}t)B\exp(-i\mathcal{H}t)\end{aligned}\tag{4b}$$

are the time-shifted Heisenberg operators corresponding to the mechanical variables A and B, respectively, where

$$[A(t),B(t')]\equiv A(t)B(t')-B(t')A(t)\tag{4c}$$

is the commutator operator, where $\langle\cdots\rangle$ designates the canonical ensemble average

$$\langle\cdots\rangle\equiv Z^{-1}\,\mathrm{Tr}[\cdots\exp(-\mathcal{H}/kT)],\tag{4d}$$

Z being the canonical partition function, k the Boltzmann constant, T the absolute temperature, and where

$$\begin{aligned}\theta(t-t')&\equiv1,\qquad t>t'\\ &\equiv0,\qquad t<t'.\end{aligned}\tag{4e}$$

Since one may show, using (4) and the cyclic property of the trace, that the temporal behavior of the retarded Green function (4a) depends only on the time difference $(t-t')$, the temporal Fourier transform of the retarded Green function may be defined as

$$\langle\langle A;B\rangle\rangle_{r,E}=(2\pi)^{-1}\int_{-\infty}^{\infty}\exp(iE\tau)\langle\langle A(0);B(\tau)\rangle\rangle_r d\tau.\tag{5}$$

As shown in Ref. (9), the equation of motion for $\langle\langle A;B\rangle\rangle_{r,E}$ is given by

$$E\langle\langle A;B\rangle\rangle_{r,E}=(2\pi)^{-1}\langle[A,B]\rangle+\langle\langle[A,\mathcal{H}];B\rangle\rangle_{r,E}.\tag{6}$$

Since the right side of (6) contains higher-order Green functions, one obtains a hierarchy for the equations of

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[12] Later in the paper, the ranges of interaction appropriate for ferrous fluoride (FeF_2) will be entered as an example of application for the method.

[13] The double-time temperature-dependent advanced and causal Green functions can be defined in a similar manner as may be found in Ref. 9. The present paper, however, only considers the retarded Green function. Also, $\hbar=1$ will be assumed.

motion which, in practice, must be decoupled by introducing some approximations in order to close the system of equations. This paper considers only the most simple "Tyablikov" decoupling approximations, which will be discussed shortly.

For the resonance problem under investigation, it will be seen later that equilibrium time correlations are needed only between transverse spin components, which therefore implies the calculation of the following Green functions:

$$\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_{r,E}, \qquad \langle\langle S_{\alpha_1}{}^+; S_{\alpha_2}{}^-\rangle\rangle_{r,E},$$
$$\langle\langle S_{\alpha}{}^+; S_{\beta}{}^-\rangle\rangle_{r,E}, \qquad \langle\langle S_{\beta}{}^+; S_{\alpha}{}^-\rangle\rangle_{r,E}. \qquad (7)$$

For notational simplicity, the subscript r henceforth will be understood but omitted.

First, then, consider the equation of motion for $\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E$:

$$E\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E = (2\pi)^{-1}\langle[S_{\beta_1}{}^+, S_{\beta_2}{}^-]\rangle + \langle\langle[S_{\beta_1}{}^+, \mathcal{H}]; S_{\beta_2}{}^-\rangle\rangle_E. \qquad (8)$$

Using the spin commutation relations

$$[S_{\beta_i}{}^+, S_{\beta_j z}] = -S_{\beta_i}{}^+\delta_{\beta_i\beta_j}, \qquad [S_{\beta_i}{}^-, S_{\beta_j z}] = S_{\beta_i}{}^-\delta_{\beta_i\beta_j},$$
$$[S_{\beta_i}{}^+, S_{\beta_j}{}^-] = 2S_{\beta_i z}\delta_{\beta_i\beta_j}, \qquad (9)$$

where $\delta_{\beta_i\beta_j}$ is the Kronecker delta symbol, and the commutator property

$$[A, BC] = [\Psi, B]C + B[A, C], \qquad (10)$$

the equation of motion (8) becomes, upon substituting the Hamiltonian (3),

$$\begin{aligned} E\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E - \pi^{-1}\langle S_{\beta_1 z}\rangle\delta_{\beta_1\beta_2} &= g\mu_B H\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E \\ &- \sum_{\alpha i} J_{i1}\langle\langle S_{\alpha_i}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E \\ &+ \sum_{\alpha i} J_{i1}\langle\langle S_{\alpha_i z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E \\ &-2 \sum_{\beta i} J_{i1}'\langle\langle S_{\beta_i}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E \\ &+2 \sum_{\beta i} J_{i1}'\langle\langle S_{\beta_i z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E \\ &+D\langle\langle S_{\beta_1}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E + D\langle\langle S_{\beta_1 z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E. \end{aligned} \qquad (11)$$

In (11), the following Tyablikov decoupling approximations will be used:

$$\langle\langle S_{\alpha_i}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\beta z}\rangle\langle\langle S_{\alpha_i}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (12a)$$

$$\langle\langle S_{\alpha_i z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\alpha z}\rangle\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (12b)$$

$$\langle\langle S_{\beta_i}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\beta z}\rangle\langle\langle S_{\beta_i}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (12c)$$

$$\langle\langle S_{\beta_i z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\beta z}\rangle\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (12d)$$

$$\langle\langle S_{\beta_1}{}^+ S_{\beta_1 z}; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\beta z}\rangle\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (12e)$$

$$\langle\langle S_{\beta_1 z} S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E = \langle S_{\beta z}\rangle\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E. \qquad (12f)$$

The approximations (12a)–(12d) enter (11) within the exchange terms and may be interpreted (since $J_{ii} = J_{ii}' = 0$) as neglecting correlations between the longitudinal and transverse spin components of spins on *different* sites. However, the approximations (12e), (12f) enter (11) through the anisotropy terms and their physical interpretations are not as appealing in the sense that here one is neglecting correlations between the longitudinal and transverse components of the *same* spin. This "breaking" of spins in the anisotropy terms is known to give spurious anisotropy effects particularly noticeable for spin $S=\frac{1}{2}$, in which case the crystal-field terms appearing in the Hamiltonian (1) should actually offer no anisotropy since $S_z^2=\frac{1}{4}$. Discussion and derivations of more proper decouplings for the anisotropy terms are given by Lines[14] for the case having no external magnetic field **H** and small anisotropy. However, since the simple decoupling approximations (12) will be used in the present paper, one should interpret the results carefully, e.g., the results are taken as qualitatively correct only for large S spin values, as will be further discussed and compared with data for ferrous fluoride FeF_2($S=2$) later in the paper.

Substituting (12) into (11) gives

$$[E - g\mu_B H - J(0)\langle S_{\alpha z}\rangle - 2(J'(0)+D)\langle S_{\beta z}\rangle]\langle\langle S_{\beta_1}{}^+; S_{\beta_2}{}^-\rangle\rangle_E - \pi^{-1}\langle S_{\beta_1 z}\rangle\delta_{\beta_1\beta_2}$$
$$= -\langle S_{\beta z}\rangle \sum_{\alpha i} J_{i1}\langle\langle S_{\alpha_i}{}^+; S_{\beta_2}{}^-\rangle\rangle_E - 2\langle S_{\beta z}\rangle \sum_{\beta i} J_{i1}'\langle\langle S_{\beta_i}{}^+; S_{\beta_2}{}^-\rangle\rangle_E, \qquad (13a)$$

where

$$J(0) \equiv \sum_{\alpha i} J_{i1}, \qquad J'(0) \equiv \sum_{\beta i} J_{i1}'. \qquad (13b)$$

Since the two-identical sublattices were each assumed to be sublattice translationally invariant, one may introduce sublattice Fourier transforms and their corresponding inverses, respectively, as follows:

$$S_{\mathbf{k}}{}^{\pm} = (2/N)^{1/2} \sum_{\beta} \exp(\pm i\mathbf{k}\cdot\mathbf{r}_\beta) S_\beta{}^{\pm}, \qquad S_\beta{}^{\pm} = (2/N)^{1/2} \sum_{\mathbf{k}} \exp(\mp i\mathbf{k}\cdot\mathbf{r}_\beta) S_{\mathbf{k}}{}^{\pm}, \qquad (14)$$

where N is the total number of Heisenberg spins, $\mathbf{r}_\beta$ is the position vector of a site on the β sublattice, and **k** is any wave vector in the corresponding reciprocal sublattice first Brillouin zone. Similar transforms and corresponding inverse expressions are also understood for the α sublattice.

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[14] M. E. Lines, Phys. Rev. **156**, 534 (1967).

Using sublattice Fourier transformations of type (14) and similar, (13a) gives

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}=\frac{\pi^{-1}\langle S_{\beta z}\rangle-J(\mathbf{k})\langle S_{\beta z}\rangle\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}}{E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle}, \tag{15a}$$

where

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}\equiv\sum_{\beta_i}\langle\langle S_{\beta_i}{}^{+};S_{\beta_j}{}^{-}\rangle\rangle_{E}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\beta_i}-\mathbf{r}_{\beta_j})], \tag{15b}$$

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}\equiv\sum_{\alpha_i}\langle\langle S_{\alpha_i}{}^{+};S_{\beta_j}{}^{-}\rangle\rangle_{E}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\alpha_i}-\mathbf{r}_{\beta_j})], \tag{15c}$$

$$J(\mathbf{k})\equiv\sum_{\beta_j}J_{ij}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\beta_j}-\mathbf{r}_{\alpha_i})],^{15}\qquad J'(\mathbf{k})\equiv\sum_{\beta_j}J_{ij}{}'\exp[i\mathbf{k}\cdot(\mathbf{r}_{\beta_j}-\mathbf{r}_{\beta_i})].^{15} \tag{15d}$$

By similar calculations of introducing earliest stage Tyablikov approximations followed by sublattice Fourier transformations, the equation of motion (6) for $\langle\langle S_{\alpha}{}^{+};S_{\beta}{}^{-}\rangle\rangle$ gives

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}=-\frac{J(\mathbf{k})\langle S_{\alpha z}\rangle\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}}{E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle}, \tag{16}$$

where $\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}$, $\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}$, and $J(\mathbf{k})$ and $J'(\mathbf{k})$ are given again by (15b), (15c), and (15d), respectively.

From the symmetry of the Hamiltonian (3), similar results given by the equations of motion for $\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)}$ and $\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}$ may be found from (15) and (16), respectively, by simply interchanging α and β, namely,

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)}=\frac{\pi^{-1}\langle S_{\alpha z}\rangle-J(\mathbf{k})\langle S_{\alpha z}\rangle\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}}{E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle}, \tag{17a}$$

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}=-\frac{J(\mathbf{k})\langle S_{\beta}\rangle\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)}}{E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle}, \tag{17b}$$

where

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)}\equiv\sum_{\alpha_i}\langle\langle S_{\alpha_i}{}^{+};S_{\alpha_j}{}^{-}\rangle\rangle_{E}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\alpha_i}-\mathbf{r}_{\alpha_j})], \tag{17c}$$

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}\equiv\sum_{\beta_i}\langle\langle S_{\beta_i}{}^{+};S_{\alpha_j}{}^{-}\rangle\rangle_{E}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\beta_i}-\mathbf{r}_{\alpha_j})], \tag{17d}$$

$$J(\mathbf{k})=\sum_{\alpha_i}J_{ij}\exp[i\mathbf{k}\cdot(\mathbf{r}_{\alpha_i}-\mathbf{r}_{\beta_j})],\qquad J'(\mathbf{k})=\sum_{\alpha_i}J_{ij}{}'\exp[i\mathbf{k}\cdot(\mathbf{r}_{\alpha_i}-\mathbf{r}_{\alpha_j})]. \tag{17e}$$

Solving (15a) and (16) simultaneously gives

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}=\frac{\pi^{-1}\langle S_{\beta z}\rangle[E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle]}{\begin{gathered}\{[E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle]\\ \times[E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle]-[J(\mathbf{k})]^{2}\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle\}\end{gathered}} \tag{18a}$$

and

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}=-\frac{\pi^{-1}J(\mathbf{k})\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle}{\begin{gathered}\{[E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle]\\ \times[E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle]-[J(\mathbf{k})]^{2}\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle\}\end{gathered}}. \tag{18b}$$

Similarly, (17a) and (17b), solved simultaneously, give

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)}=\frac{\pi^{-1}\langle S_{\alpha z}\rangle[E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle]}{\begin{gathered}\{[E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle]\\ \times[E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle]-[J(\mathbf{k})]^{2}\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle\}\end{gathered}} \tag{19a}$$

and

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}=-\frac{\pi^{-1}J(\mathbf{k})\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle}{\begin{gathered}\{[E-g\mu_{B}H-J(0)\langle S_{\beta z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\alpha z}\rangle]\\ \times[E-g\mu_{B}H-J(0)\langle S_{\alpha z}\rangle+2(J'(\mathbf{k})-J'(0)-D)\langle S_{\beta z}\rangle]-[J(\mathbf{k})]^{2}\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle\}\end{gathered}}, \tag{19b}$$

where, as again expected from symmetry, (18) and (19) may be found from one another by interchanging α and β, i.e.,

$$\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta)}\underset{\alpha\leftrightarrow\beta}{\leftrightarrow}\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha)},\qquad\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\alpha\beta)}=\langle\langle S_{\mathrm{k}}{}^{+};S_{\mathrm{k}}{}^{-}\rangle\rangle_{E}{}^{(\beta\alpha)}. \tag{20}$$

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[15] One notices that the notations introduced in (15d) are consistent with those previously introduced in (13b).

RESONANCE-SUSCEPTIBILITY EXPRESSIONS

One now considers the case where the perturbing Hamiltonian $\mathcal{H}_1$ is due to a uniform rotating magnetic field $\mathbf{H}_1$ at right angles to the uniform steady magnetic field $\mathbf{H}$. Specifically, one chooses the static field $\mathbf{H}$ in the *negative* z direction and the circularly polarized field $\mathbf{H}_1$ to have components

$$H_{1x}=H_1\cos\omega t, \qquad H_{1y}=-H_1\sin\omega t, \qquad H_{1z}=0, \tag{21}$$

whereupon the perturbing Hamiltonian $\mathcal{H}_1$ is then given by

$$\mathcal{H}_1=-g\mu_BH_1\sum_f(S_{fx}\cos\omega t-S_{fy}\sin\omega t)$$
$$=Ve^{-i\omega t}+V^*e^{i\omega t}, \tag{22a}$$

where

$$V\equiv-\tfrac{1}{2}g\mu_BH_1\sum_f S_f^-, \quad V^*\equiv-\tfrac{1}{2}g\mu_BH_1\sum_f S_f^+. \tag{22b}$$

Letting $\delta\langle\cdots\rangle$ designate the deviation of an observable from its equilibrium value due to the action of a perturbation, the linear complex magnetic susceptibility per spin $\chi_\pm(\omega)$ is defined by

$$\delta\langle M_\pm\rangle\equiv N\chi_\pm(\omega)H_1e^{\pm i\omega t}, \tag{23a}$$

where

$$M_\pm\equiv M_x\pm iM_y=g\mu_B\sum_f S_f^\pm, \tag{23b}$$

$$\chi_\pm(\omega)\equiv\chi'(\omega)\pm i\chi''(\omega), \tag{23c}$$

$\chi'(\omega)$, $\chi''(\omega)$ being called the magnetic dispersion and absorption factor, respectively. The fact that (23a) agrees with other essentially equivalent expressions in the literature for $\chi_\pm(\omega)$ may be seen by verifying that W, the energy absorbed by the spin system from the circular field per unit time, is given by

$$W=N\omega H_1^2\chi''(\omega). \tag{24}$$

Considering the perturbation to be switched on adiabatically, (22a) may be written

$$\mathcal{H}_1=\sum_\omega e^{\epsilon t}e^{-i\omega t}V_\omega \qquad (\epsilon>0,\ \epsilon\to0). \tag{25}$$

Under the influence of such a perturbation [(25)], there exists a direct connection[9] between the linear steady-state response of a system observable, $\delta\langle A\rangle$, and the temporal Fourier transforms of appropriate retarded Green functions, namely,

$$\delta\langle A\rangle=2\pi\sum_\omega e^{\epsilon t}e^{-i\omega t}\langle\langle A;V_\omega\rangle\rangle_{E=\omega+i\epsilon} \qquad (\epsilon>0,\ \epsilon\to0). \tag{26}$$

For the case in hand, (26) may be written, using (22a), as

$$\delta\langle M_+\rangle=2\pi\sum_\omega e^{\epsilon t}e^{-i\omega t}\langle\langle M_+;V_\omega\rangle\rangle_{E=\omega+i\epsilon}$$
$$=2\pi e^{\epsilon t}(e^{-i\omega t}\langle\langle M_+;V\rangle\rangle_{E=\omega+i\epsilon}$$
$$+e^{i\omega t}\langle\langle M_+;V^*\rangle\rangle_{E=-\omega+i\epsilon}) \qquad (\epsilon>0,\ \epsilon\to0). \tag{27}$$

Comparing (27) and (23a) gives

$$NH_1\chi_+(\omega)=2\pi\langle\langle M_+;V\rangle\rangle_{E=\omega+i\epsilon} \qquad (\epsilon>0,\ \epsilon\to0), \tag{28}$$

where the factor $e^{\epsilon t}$ has been taken as unity. Substituting (22b) and (23b) into (28), one obtains

$$\chi_+(\omega)=-\pi(g\mu_B)^2\Big[\sum_{\alpha i}\langle\langle S_{\alpha_i}^+;S_{\alpha_j}^-\rangle\rangle_E+\sum_{\alpha i}\langle\langle S_{\alpha_i}^+;S_{\beta_j}^-\rangle\rangle_E+\sum_{\beta i}\langle\langle S_{\beta_i}^+;S_{\alpha_j}^-\rangle\rangle+\sum_{\beta i}\langle\langle S_{\beta_i}^+;S_{\beta_j}^-\rangle\rangle_E\Big]_{E=\omega+i\epsilon}$$
$$(\epsilon>0,\ \epsilon\to0). \tag{29}$$

Using (15b), (15c) and (17c), (17d), (29) may be written as

$$\chi_+(\omega)=-\pi(g\mu_B)^2\big[\langle\langle S_{\mathbf{k}}^+;S_{\mathbf{k}}^-\rangle\rangle_E^{(\alpha)}+\langle\langle S_{\mathbf{k}}^+;S_{\mathbf{k}}^-\rangle\rangle_E^{(\alpha\beta)}+\langle\langle S_{\mathbf{k}}^+;S_{\mathbf{k}}^-\rangle\rangle_E^{(\beta\alpha)}+\langle\langle S_{\mathbf{k}}^+;S_{\mathbf{k}}^-\rangle\rangle_E^{(\beta)}\big]_{\mathbf{k}=0,E=\omega+i\epsilon}$$
$$(\epsilon>0,\ \epsilon\to0). \tag{30}$$

Evaluating (18) and (19) at $\mathbf{k}=\mathbf{0}$ and substituting the resulting expressions into (30) gives

$$\chi_+(\omega)=-(g\mu_B)^2[N(E)/(E-\omega_1)(E-\omega_2)]\,|_{E=\omega+i\epsilon} \qquad (\epsilon>0,\ \epsilon\to0), \tag{31a}$$

where the numerator expression is given by

$$N(E)\equiv(E-g\mu_BH)(\langle S_{\alpha z}\rangle+\langle S_{\beta z}\rangle)-J(0)(\langle S_{\alpha z}\rangle^2+\langle S_{\beta z}\rangle^2)-2(J(0)+2D)\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle \tag{31b}$$

and where the roots of the denominator are calculated to be

$$\omega_{1,2}\equiv g\mu_BH+\tfrac{1}{2}(J(0)+2D)(\langle S_{\alpha z}\rangle+\langle S_{\beta z}\rangle)\pm\tfrac{1}{2}[(J(0)-2D)^2(\langle S_{\alpha z}\rangle+\langle S_{\beta z}\rangle)^2+16D(J(0)-D)\langle S_{\alpha z}\rangle\langle S_{\beta z}\rangle]^{1/2}. \tag{31c}$$

By the method of partial fractions, (31a) can be written

$$\chi_+(\omega)=-[(g\mu_B)^2/(\omega_1-\omega_2)]N(E)[(E-\omega_1)^{-1}-(E-\omega_2)^{-1}]\,|_{E=\omega+i\epsilon} \qquad (\epsilon>0,\ \epsilon\to0). \tag{32}$$

Using the symbolic identity

$$\lim_{\epsilon\to 0,(\epsilon>0)} (x\pm i\epsilon)^{-1}=P(1/x)\mp i\pi\delta(x), \tag{33}$$

where P denotes the principal part and $\delta(x)$ is the Dirac δ function, (32) gives, upon separating real and imaginary parts, the magnetic dispersion and absorption factor, respectively, as

$$\chi'(\omega)=-(g\mu_B)^2\frac{N(\omega)}{\omega_1-\omega_2}\left(\frac{1}{\omega-\omega_1}-\frac{1}{\omega-\omega_2}\right), \tag{34a}$$

$$\chi''(\omega)=(g\mu_B)^2\pi\left[\frac{N(\omega_1)}{\omega_1-\omega_2}\delta(\omega-\omega_1)-\frac{N(\omega_2)}{\omega_1-\omega_2}\delta(\omega-\omega_2)\right]. \tag{34b}$$

The thermal behaviors of the resonance-susceptibility expressions (34) will be examined and discussed shortly.

SUBLATTICE MAGNETIZATION EXPRESSIONS

If the *equilibrium* quantities $\langle S_{\alpha z}\rangle$, $\langle S_{\beta z}\rangle$ are now calculated and substituted into (34), the resonance susceptibility may be analyzed within this approximation for all values of temperature and magnetic field **H**. In the present paper, however, these equilibrium quantities will be calculated (by series expansion) only for certain ranges of temperature and magnetic field **H** using molecular-field theory.[16] In order to make the paper self-contained, the relevant parts of these molecular-field calculations will be outlined.

The molecular-field-approximation single-spin density operators for an α or β site are given, respectively, by

$$\rho^{(1)}(\alpha)=\frac{\exp\{[(g\mu_BH+J'(0)\langle S_{\alpha z}\rangle+J(0)\langle S_{\beta z}\rangle)S_{\alpha z}+DS_{\alpha z}{}^2]/kT\}}{\mathrm{Tr}\exp\{[(g\mu_BH+J(0)\langle S_{\alpha z}\rangle+J(0)\langle S_{\beta z}\rangle)S_{\alpha z}+DS_{\alpha z}{}^2]/kT\}}, \tag{35a}$$

$$\rho^{(1)}(\beta)=\frac{\exp\{[(g\mu_BH+J'(0)\langle S_{\beta z}\rangle+J(0)\langle S_{\alpha z}\rangle)S_{\beta z}+DS_{\beta z}{}^2]/kT\}}{\mathrm{Tr}\exp\{[(g\mu_BH+J'(0)\langle S_{\beta z}\rangle+J(0)\langle S_{\alpha z}\rangle)S_{\beta z}+DS_{\alpha z}{}^2]/kT\}}. \tag{35b}$$

Having chosen the α sublattice as the up sublattice, one now introduces the following *equilibrium assumption*:

$$\langle\mu_{\alpha z}\rangle=\langle\mu_{\alpha z}\rangle_0+\Delta\langle\mu_z\rangle, \tag{36a}$$

$$\langle\mu_{\beta z}\rangle=-\langle\mu_{\alpha z}\rangle_0+\Delta\langle\mu_z\rangle, \tag{36b}$$

where $\boldsymbol{\mu}$ is the magnetic-moment operator of a spin, $\langle\mu_{\alpha z}\rangle_0\geq 0$ is the α sublattice *spontaneous* magnetization per spin and, having previously chosen the applied field **H** in the negative z direction, $\Delta\langle\mu_z\rangle\leq 0$ is the *induced* magnetization per spin of either sublattice due to the field **H**. Since $\boldsymbol{\mu}$ will be considered as due to electron spin **S**,

$$\boldsymbol{\mu}=g\mu_B\mathbf{S},\qquad g\mu_B<0. \tag{37}$$

Using (37), (36) becomes

$$\langle S_{\alpha z}\rangle=\langle S_{\alpha z}\rangle_0+\Delta\langle S_z\rangle, \tag{38a}$$

$$\langle S_{\beta z}\rangle=-\langle S_{\alpha z}\rangle_0+\Delta\langle S_z\rangle, \tag{38b}$$

where

$$\langle S_{\alpha z}\rangle_0\leq 0,\qquad \Delta\langle S_z\rangle\geq 0. \tag{38c}$$

Substituting (38a) and (38b) into (35a), gives

$$\rho^{(1)}(\alpha)=\frac{\exp[\{[g\mu_BH+(J'(0)+J(0))\Delta\langle S_z\rangle+(J'(0)-J(0))\langle S_{\alpha z}\rangle_0]S_{\alpha z}+DS_{\alpha z}{}^2\}/kT]}{\mathrm{Tr}\exp[\{[g\mu_BH+(J'(0)+J(0))\Delta\langle S_z\rangle+(J'(0)-J(0))\langle S_{\alpha z}\rangle_0]S_{\alpha z}+DS_{\alpha z}{}^2\}/kT]}. \tag{39}$$

Considering small H and linear response, one may take

$$[g\mu_BH+(J'(0)+J(0))\Delta\langle S_z\rangle]/kT\ll 1 \tag{40}$$

and expand (39) to give

$$\rho^{(1)}(\alpha)=\rho_0{}^{(1)}(\alpha)\{1+(kT)^{-1}[g\mu_BH+(J'(0)+J(0))\Delta\langle S_z\rangle](S_{\alpha z}-\langle S_{\alpha z}\rangle_0)\}+o(H^2), \tag{41a}$$

where

$$\rho_0{}^{(1)}(\alpha)=\frac{\exp\{[(J'(0)-J(0))\langle S_{\alpha z}\rangle_0S_{\alpha z}+DS_{\alpha o}{}^2]/kT\}}{\mathrm{Tr}\exp\{[(J'(0)-J(0))\langle S_{\alpha z}\rangle_0S_{\alpha z}+DS_{\alpha z}{}^2]/kT\}} \tag{41b}$$

is the α-site molecular-field-approximation single-spin density operator in the absence of the external field **H**.

Using (41b), the spontaneous quantity $\langle S_{\alpha z}\rangle_0$ introduced in (38) may be found in lowest-order approximation from

$$\langle S_{\alpha z}\rangle_0=\mathrm{Tr}S_{\alpha z}\rho_0{}^{(1)}(\alpha). \tag{42}$$

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[16] Similar molecular-field-type calculations may be found in Ref. 8; also see A. Honma, J. Phys. Soc. Japan **15**, 456 (1960).

Also, using (41) and (42), one is able to calculate a lowest-order approximation expression for $\langle S_{\alpha z}\rangle$, since

$$\begin{aligned}\langle S_{\alpha z}\rangle &= \mathrm{Tr}\, S_{\alpha z}\rho^{(1)}(\alpha)\\ &= \langle S_{\alpha z}\rangle_0 + (kT)^{-1}[g\mu_B H + (J'(0)+J(0))\Delta\langle S_z\rangle][\mathrm{Tr}(S_{\alpha z}^2\rho_0^{(1)}(\alpha)) - \langle S_{\alpha z}\rangle_0^2]. \end{aligned}\tag{43}$$

Comparing (43) and (38a), one finds the expression for the induced quantity $\Delta\langle S_z\rangle$ to be

$$\Delta\langle S_z\rangle = g\mu_B \frac{\mathrm{Tr}(S_{\alpha z}^2\rho_0^{(1)}(\alpha)) - \langle S_{\alpha z}\rangle_0^2}{kT - (J'(0)+J(0))[\mathrm{Tr}(S_{\alpha z}^2\rho_0^{(1)}(\alpha)) - \langle S_{\alpha z}\rangle_0^2]} H. \tag{44}$$

The remainder of this section centers upon calculations for the equilibrium expressions (42) and (44). Although numerical calculations of (42) and (44) can be obtained for all temperatures using a computer, explicit calculations (using series expansion) will be made in this paper only for $|\langle S_{\alpha z}\rangle_0| \ll 1$, i.e., for temperatures above or slightly below the Néel transition temperature T_N defined as the temperature at and above which the sublattice spontaneous magnetization $\langle S_{\alpha z}\rangle_0$ disappears. Considering then $|\langle S_{\alpha z}\rangle_0| \ll 1$, (41b) may be expanded to give

$$\rho_0^{(1)}(\alpha) = \frac{(1+b\langle S_{\alpha z}\rangle_0 S_{\alpha z} + (1/2!)b^2\langle S_{\alpha z}\rangle_0^2 S_{\alpha z}^2 + (1/3!)b^3\langle S_{\alpha z}\rangle_0^3 S_{\alpha z}^3 + \cdots)\exp(aS_{\alpha z}^2)}{\mathrm{Tr}[(1+b\langle S_{\alpha z}\rangle_0 S_{\alpha z} + (1/2!)b^2\langle S_{\alpha z}\rangle_0^2 S_{\alpha z}^2 + (1/3!)b^3\langle S_{\alpha z}\rangle_0^3 S_{\alpha z}^3 + \cdots)\exp(aS_{\alpha z}^2)]}, \tag{45a}$$

where

$$a \equiv D/kT, \qquad b \equiv (J'(0) - J(0))/kT. \tag{45b}$$

Substituting (45a) into (42) gives

$$\begin{aligned}\langle S_{\alpha z}\rangle_0 &= \mathrm{Tr}\, S_{\alpha z}\rho_0^{(1)}(\alpha)\\ &= b\frac{m_2}{m_0}\langle S_{\alpha z}\rangle_0\left[1 + b^2\left(\frac{m_4}{3!m_2} - \frac{m_2}{2!m_0}\right)\langle S_{\alpha z}\rangle_0^2 + o(\langle S_{\alpha z}\rangle_0^4)\right],\end{aligned}\tag{46}$$

where

$$m_n \equiv \mathrm{Tr}[S_{\alpha z}^n \exp(aS_{\alpha z}^2)], \qquad n = 0, 1, 2, \cdots \tag{47a}$$

and where these m_n quantities may, when desired, be evaluated from

$$0 = m_1 = m_3 = m_5 = \cdots, \tag{47b}$$

$$m_0 = \mathrm{Tr}\exp(aS_{\alpha z}^2), \qquad m_2 = \partial m_0/\partial a, \qquad m_4 = \partial^2 m_0/\partial a^2, \cdots. \tag{47c}$$

Neglecting terms $o(\langle S_{\alpha z}\rangle_0^4)$, (46) gives

$$\langle S_{\alpha z}\rangle_0^2 = \frac{g(T)}{b^2(m_2/2!m_0 - m_4/3!m_2)}, \tag{48a}$$

where the numerator is defined as

$$g(T) \equiv 1 - b^{-1}(m_0/m_2). \tag{48b}$$

The Néel transition temperature T_N is found in this approximation by setting (48b) equal to zero, which gives

$$kT_N(m_0/m_2)_{T_N} = J'(0) - J(0), \tag{49}$$

where (45b) has been used. As is known in molecular-field-type theories, the solution of the transcendental equation (49) will locate T_N higher than actual experimental values.

For temperatures slightly below T_N, one may expand the right-hand side of (48a) with respect to T_N and conclude, upon using (49), that

$$\begin{aligned}\langle S_{\alpha z}\rangle_0 &= \left[\frac{\partial g/\partial T}{b^2(m_4/3!m_2 - m_2/2!m_0)}\right]_{T=T_N}(T_N - T)^{1/2}, &&\text{for } T \text{ slightly below } T_N\\ &= 0, &&\text{for } T \geq T_N\end{aligned}\tag{50}$$

where the critical index $\frac{1}{2}$ is the usual molecular-field value, again higher than actual experimental values.

In order to calculate $\Delta\langle S_z\rangle$ in this approximation, one substitutes (45) into (44) and finds, upon using (47) and (50), that, for $H \neq 0$, $\Delta\langle S_z\rangle$ has a ***maximum*** at $T = T_N$ and varies from this maximum value as $|T - T_N|$ for temperatures closely surrounding T_N, i.e.,

$$\begin{aligned}\Delta\langle S_z\rangle &= g\mu_B H\{[kT_N(m_0/m_2)_{T_N} - (J'(0)+J(0))]^{-1} + o(|T - T_N|)\}\\ &= [g\mu_B H/2|J(0)|][1 + o(|T - T_N|)], \qquad \text{for } T \text{ near } T_N\end{aligned}\tag{51}$$

where (49) was used to obtain the final form (51) which shows the maximum value to be independent of both the crystal-field anisotropy constant D and the intrasublattice exchange quantity $J'(0)$. The fact that $\langle S_{\alpha z}\rangle_0$ vanishes while $\Delta\langle S_z\rangle$ is nonvanishing at $T = T_N$ will be used to characterize a so-called transition region in the following section.

ANALYSES OF RESONANCE-SUSCEPTIBILITY EXPRESSIONS

One may now combine the results of the previous two sections to analyze the resonance susceptibility for certain ranges of temperature in both the absence and presence of the external magnetic field $\mathbf{H}$. Substituting (38) into (31b) and (31c) gives, respectively,

$$N(E) = 2(E - g\mu_B H)\Delta\langle S_z\rangle - 4(J(0)+D)(\Delta\langle S_z\rangle)^2 + 4D\langle S_{\alpha z}\rangle_0^2 \tag{52}$$

and

$$\omega_{1,2}=g\mu_BH+(J(0)+2D)\Delta\langle S_z\rangle$$
$$\pm[J(0)^2(\Delta\langle S_z\rangle)^2-4D(J(0)-D)\langle S_{\alpha z}\rangle_0{}^2]^{1/2}. \quad (53)$$

First, consider the situation where the external magnetic field $\mathbf{H}=\mathbf{0}$. Then (52) and (53) give, respectively,

$$N(E)=4D\langle S_{\alpha z}\rangle_0{}^2 \quad (54)$$

and

$$\omega_{1,2}=\pm2\,|\,D(J(0)-D)\,|^{1/2}\,|\,\langle S_{\alpha z}\rangle_0\,|. \quad (55)$$

Substituting (54) and (55) into (34) gives

$$\chi'(\omega)=-(g\mu_B)^2[D^{1/2}/|\,J(0)-D\,|^{1/2}]$$
$$\times|\,\langle S_{\alpha z}\rangle_0\,|\,P((\omega-\omega_1)^{-1}), \quad (56a)$$

$$\chi''(\omega)=(g\mu_B)^2\pi[D^{1/2}|\,J(0)-D\,|^{1/2}]\,|\,\langle S_{\alpha z}\rangle_0\,|\,\delta(\omega-\omega_1), \quad (56b)$$

where only the positive frequency ω_1 has been taken from (55). At this point, experimental data for FeF_2 will be entered as an example of application. These data are given by[8]

$$S=2, \qquad D=6.5\pm0.3\ \text{cm}^{-1},$$
$$J_{ij}\equiv J=-3.85\pm0.2\ \text{cm}^{-1},$$
$$J_{ln}'\equiv J'=(0.1\pm02.5)\tfrac{1}{2}J,$$
$$z\equiv8, \qquad z'=2, \quad (57)$$

where $(i,\,j)$ is a next-nearest-neighbor pair having antiferromagnetic exchange coupling, where $(l,\,n)$ is a nearest-neighbor pair having much weaker exchange coupling whose sign has not at present been clearly established, and because of the fact that FeF_2 has a body-centered tetragonal structure, it is found that there exist, for each spin, $z=8$ next-nearest-neighbor antiferromagnetically coupled spins and $z'=2$ nearest-neighbor coupled spins (the latter along the c axis of the rutile crystal structure). From (55), the value of the resonance frequency in the absence of the external field $\mathbf{H}$ and at $T=0$°K may be calculated, using (13b) and the FeF_2 data (57), to be

$$\tilde{\nu}\approx63\ \text{cm}^{-1}, \quad (58)$$

having taken $|\,\langle S_{\alpha z}\rangle_0\,|=2$ by neglecting any zero-point motion. The value given by (58) compares reasonably well with the experimental AFMR value[17] $52.7\pm0.2\ \text{cm}^{-1}$. The discrepancy can chiefly be traced in this case to the earlier Green-function decoupling used in the anisotropy terms, since (55) is the same expression as derived by Lines[8] using spin-wave theory, aside from a factor $(2S-1)/2S$ always multiplying D in his expression. One sees, therefore, that at low temperatures, the simple Tyablikov decoupling becomes better for large S, as shown by Lines[14] in the limit of small anisotropy. Also from (55), the resonance frequency ω_1 for $\mathbf{H}=\mathbf{0}$ is seen to have the same temperature dependence as the sublattice spontaneous magnetization and therefore, from (50), vanishes for $T\geq T_N$ and varies as $(T_N-T)^{1/2}$ for temperatures slightly below T_N. However, the power absorbed by the spin system at the circular frequency ω_1 is found, from (24) and (56b), to be

$$W=2\pi N(g\mu_B)^2H_1{}^2D\langle S_{\alpha z}\rangle_0{}^2\delta(\omega-\omega_1), \quad (59)$$

whose δ amplitude therefore vanishes, from (50), as (T_N-T) for temperatures slightly below T_N. In other words, the amplitude of power absorbed vanishes more rapidly than does the resonance frequency as functions of temperature slightly beneath T_N, which might suggest experimental AFMR detection difficulties for temperatures close to T_N.

Next, consider the situation where the external magnetic field $\mathbf{H}\neq\mathbf{0}$. Near T_N, (51) and (50) show, respectively, that $\Delta\langle S_z\rangle$ is constant to leading order, while $\langle S_{\alpha z}\rangle_0=0$ for $T\geq T_N$ and vanishes as $(T_N-T)^{1/2}$ for T slightly below T_N. As a result, a *transition region* (denoted by $T\sim T_N$) in the presence of the external field $\mathbf{H}$ will be defined as those temperatures near T_N for which

$$|\,\langle S_{\alpha z}\rangle_0/\Delta\langle S_z\rangle\,|\ll1 \quad (60)$$

and for which (50) and (51) both hold. These temperatures closely surrounding T_N which define the transition region $(T\sim T_N)$ correspond to a range of thermodynamic states in the *paramagnetic region* [since (38) and (60) taken together imply that the sublattice magnetizations point in the same direction], all of which are close to but not touching the phase-boundary curve separating the antiferromagnetic and paramagnetic regions for small H in an H-T phase diagram.[11]

Using (60), one may expand (53) to obtain expressions for the resonance frequencies (corresponding to $\mathbf{k}=\mathbf{0}$ paramagnetic modes) and their splitting at temperatures within and above the transition region as

$$\omega_1=g\mu_BH+2D\Delta\langle S_z\rangle$$
$$\times\left[1+\frac{J(0)-D}{J(0)}\left(\frac{\langle S_{\alpha z}\rangle_0}{\Delta\langle S_z\rangle}\right)^2+\cdots\right], \quad (61a)$$

$$\omega_2=g\mu_BH+2(J(0)+D)\Delta\langle S_z\rangle$$
$$\times\left[1-\frac{D(J(0)-D)}{J(0)\,(J(0)+D)}\left(\frac{\langle S_{\alpha z}\rangle_0}{\Delta\langle S_z\rangle}\right)^2+\cdots\right], \quad (61b)$$

$$\omega_1-\omega_2=2\,|\,J(0)\,|\,\Delta\langle S_z\rangle$$
$$\times\left[1-2\frac{D(J(0)-D)}{[J(0)]^2}\left(\frac{\langle S_{\alpha z}\rangle_0}{\Delta\langle S_z\rangle}\right)^2+\cdots\right]. \quad (61c)$$

Using (50) and (51), (61) gives, for temperatures within the transition region $(T\sim T_N)$, the results

$$\omega_1=g\mu_BH[1+D/|\,J(0)\,|+o(|\,T-T_N\,|)], \quad (62a)$$
$$\omega_2=g\mu_BH[D/|\,J(0)\,|+o(|\,T-T_N\,|)], \quad (62b)$$
$$\omega_1-\omega_2=g\mu_BH+o(|\,T-T_N\,|). \quad (62c)$$

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[17] R. C. Ohlmann and M. Tinkham, Phys. Rev. **123**, 425 (1961).

Substituting FeF_2 experimental data [(57)] into (62) as an example of application shows for $T \sim T_N$ that, to leading order, ω_1 is, in this case, about six times larger than ω_2 while their splitting is proportional to the external magnetic field H. In addition, (61) may be used to show for high temperatures in the paramagnetic region ($T \gg T_N$) that both frequencies tend to approach the same value $g\mu_B H$, since $\Delta\langle S_z\rangle$ can be expected to become a small and essentially temperature-independent residual quantity at these high temperatures.[8]

Using (34b), the expression (24) for the resonance power absorbed becomes

$$W = N(g\mu_B)^2 \pi H_1^2 \left[\frac{\omega_1 N(\omega_1)}{\omega_1 - \omega_2} \delta(\omega - \omega_1) - \frac{\omega_2 N(\omega_2)}{\omega_1 - \omega_2} \delta(\omega - \omega_2) \right]. \quad (63)$$

For temperatures within and above the transition region, one calculates, using (52), (60), and (61), that

$$\frac{N(\omega_1)}{\omega_1 - \omega_2} = 2\Delta\langle S_z\rangle \left[1 - \left(\frac{D}{J(0)}\right)^2 \left(\frac{\langle S_{\alpha z}\rangle_0}{\Delta\langle S_z\rangle}\right)^2 + \cdots \right], \quad (64a)$$

$$\frac{N(\omega_2)}{\omega_1 - \omega_2} = -2\left(\frac{D}{J(0)}\right)^2 \frac{\langle S_{\alpha z}\rangle_0^2}{\Delta\langle S_z\rangle} + \cdots. \quad (64b)$$

Substituting (64) and (62) into (63) and using (51), one can find the δ amplitude expressions for $T \sim T_N$, whereupon their ratio is then given by

$$-\frac{\omega_2 N(\omega_2)}{\omega_1 N(\omega_1)} = \frac{4D^3}{(g\mu_B H)^2 (D - J(0))} \langle S_{\alpha z}\rangle_0^2 \times [1 + o(|T - T_N|)] \quad \text{for} \quad T \sim T_N. \quad (65)$$

Using (50), the last result [(65)] enables one to conclude that the resonance power absorbed at the lower frequency ω_2 is much smaller than that absorbed at the higher frequency ω_1 and, in fact, vanishes for $T \geq T_N$. Also, recalling from (44) that $\Delta\langle S_z\rangle$ is proportional to the external field H, one finds, after substituting (61) and (64) into (63), that, for temperatures within and above the transition region, the resonance power δ amplitude at the higher frequency ω_1 varies as H^2 while the resonance power δ amplitude at the lower frequency ω_2 is independent of H. Finally, (64a) may be used to show that the resonance power absorption at the higher frequency ω_1 also tends to become small for high temperatures in the paramagnetic region ($T \gg T_N$) since, as mentioned previously, $\Delta\langle S_z\rangle$ can then be expected to become small and essentially temperature-independent. As before, these amplitude behaviors could suggest difficulties for experimentally detecting the lower frequency ω_2 at temperatures within the transition region using circularly polarized radiation. As mentioned in the Introduction, it is important to realize that any features of the resonance problem which are sensitive to relaxation or other irreversible processes have not been studied by the present treatment.

CONCLUSIONS

The method of double-time temperature-dependent Green functions was combined with results from equilibrium molecular-field theory in order to study resonance susceptibility in a two-sublattice antiferromagnetic Heisenberg model having isotropic exchange, uniaxial crystal-field single-ion-type anisotropy, and where the ranges of the exchange interactions and the spin value S per site are essentially arbitrary. Because of the form of the Hamiltonian and the nature of the approximations, no information is found for the absorption linewidths but rather for approximate thermal behaviors of the locations and amplitudes of the absorption profile.

In the *absence* of the uniform, static external magnetic field $\mathbf{H}$, the results show a single (positive) AFMR frequency having the *same* temperature dependence as the sublattice spontaneous magnetization $\langle S_{\alpha z}\rangle_0$ and where the absorbed-power δ amplitude varies with temperature as the *square* of the sublattice spontaneous magnetization. In a molecular-field approximation the latter results imply that, for temperatures slightly below T_N, the AFMR frequency varies as $(T_N - T)^{1/2}$ while the δ amplitude of power absorbed varies as $(T_N - T)$. Using FeF_2 experimental data, the calculated value of this AFMR frequency at $T = 0°K$ is in reasonably good agreement with experiment.

In the *presence* of the external field $\mathbf{H}$ and at temperatures within a so-called transition region ($T \sim T_N$), one finds a *doublet* of resonance frequencies (corresponding to $\mathbf{k} = \mathbf{0}$ paramagnetic modes) whose splitting is proportional to H and which are in ratio to one another by about a factor of 6 in the case of FeF_2. However, at these temperatures $T \sim T_N$, the resonance power absorbed at the lower frequency is much smaller than that absorbed at the higher frequency and actually vanishes for $T \geq T_N$ while, in regard to the field dependence at temperatures within and above the transition region, the higher frequency power absorbed varies as H^2 whereas that for the lower frequency is independent of H. Finally, for $T \gg T_N$, one may expect that the power absorbed at the higher frequency also tends to become small.

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