Spin-Spin and Spin-Phonon Interactions in the Heisenberg Antiferromagnet*

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The Dyson ideal spin-wave Hamiltonian for the two-sublattice Heisenberg antiferromagent is obtained using a Maleev transformation. A spin-phonon interaction Hamiltonian is derived by expanding the lattice coordinates in small displacements and retaining the linear terms. The static and dynamic properties of the system containing the spin field and the harmonic phonon field are studied using the double-time Green's-function method. This is done in a self-consistent manner, using symmetrized equations of motion, from which a Dyson equation is derived. It is found that if the zeroth-order Hamiltonian describing the Dyson equation contains contributions from the interacting ideal spin waves, the polarization operator is no longer simple. A zeroth-order approximation which contains all the static contributions arising from the interacting ideal spin waves is constructed. Contact is made with the work of previous authors and some aspects of the Callen decoupling procedure are clarified. Using an effective Hamiltonian and a canonical-transformation technique, expressions for the full polarization operator are developed. Finally, expressions for the frequency-dependent susceptibility are obtained and these are used to discuss the line shape for the absorption of energy from an oscillating field at frequencies near and far from resonance.

1. INTRODUCTION

OWING to the large number of studies on the Heisenberg ferromagnet,[1] it could at least be said that the behavior of this system far away from the critical region is well understood. Out of all the methods available for the study of this system, perhaps the most powerful and systematic approach is via the Green's-function technique. But most of the studies on the Heisenberg ferromagnet, and the parallel treatments of the antiferromagnet, have been devoted to a study of their static properties and how these could be more correctly approximated with the aid of improved techniques for decoupling the chain of Green's functions.

In a well-known study of the Heisenberg ferromagnet, Dyson[2] has investigated the deviations from simple spin-wave theory and shown that, at least at low temperatures, it is possible to use a non-Hermitian effective-boson Hamiltonian (e.b.H.), the so-called Dyson ideal spin-wave Hamiltonian, to discuss the properties of the system. This depended on his proof that the kinematical interactions are negligible in comparison to the dynamical interactions. The Dyson ideal spin-wave Hamiltonian can be directly obtained from the spin Hamiltonian using a transformation due to Maleev. This procedure was used by Tahir-Kheli and Ter Haar[3] in a study of the Heisenberg ferromagnet using the method of Green's functions. They succeeded in obtaining Dyson's results and also obtained expressions for the magnon lifetimes, but had to resort to a somewhat tortuous decoupling procedure. If a more clear-cut approach to the decoupling problem could be had, the use of the ideal spin-wave Hamiltonian affords a very convenient approach to the study of static and dynamical properties of the Heisenberg ferromagnet, at least in the low-temperature region.

In the present study a clearer approach to the decoupling problem will be presented using a "symmetrized" development of the equations of motion of the spin operators. The method is applied to the case of a spin-uncompensated two-sublattice antiferromagnet whose static and dynamical properties are studied. The main advantage of the mathematical formalism is brought out by showing how additional interacting fields, for example, the phonon field, could be incorporated in a general manner.

In Sec. 2 of this study the Hamiltonian of the system is discussed and, using a suitable Maleev transformation, the non-Hermitian e.b.H. is obtained. That part of the Hamiltonian which results from a simple Holstein-Primakoff approximation is used to derive a spin-phonon interaction Hamiltonian. For this purpose the phonon field is taken to be that of a harmonic crystal and, in developing the interaction term, only linear terms in the lattice displacements are retained.

In Sec. 3 a matrix form of the double-time Green's functions is defined and their equations of motion are derived with respect to both time arguments. This procedure[4] enables us to express the equations for the Green's functions in the form of a Dyson equation. In deriving the Dyson equation, unless the zeroth Green's functions are defined in terms of a noninteracting ideal spin-wave Hamiltonian, the polarization operator is found to have a complicated structure.

In Sec. 4 a "renormalized" zeroth-order approximation is derived. This approximation is adequate to discuss all the static effects arising from the scattering

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of spin waves as described by the e.b.H. In this section expressions are derived for the excitation energies, sublattice magnetizations, and the average energy of the system in this approximation. Contact is established with the results of Hewson and Ter Haar,[5] Anderson and Callen,[6] Lee and Liu,[7] and P'u Fu-Cho,[8] by taking suitable limits. Of the two possible ways of applying Callen's decoupling procedure (CD) to antiferromagnetism, Callen has shown that only one leads to satisfactory results. This choice is indeed found to be necessary in CD if the results are to agree with those obtained here, using the ideal spin-wave Hamiltonian. No attempt is made to discuss critical phenomena since the existing methods are, in our view, only of qualitative validity near the Néel temperature.

In Sec. 5 the method of evaluating the polarization operator is outlined. The details are presented in an appendix. Using a canonical transformation which diagonalizes the zeroth approximation, expressions are developed for the polarization operator in a self-consistent manner. In evaluating the contribution of the spin-phonon interaction to the polarization operator, it becomes necessary to decouple certain mixed Green's functions for which a clear-cut random-phase approximation becomes manifest. The expressions for the polarization operator are compared with those due to Kashcheev.[9]

In Sec. 6 the frequency-dependent susceptibility of the system is considered with and without the phonon field. Expressions for the line shape are obtained both in regions of frequency near and far from resonance. Finally, in the conclusion we have emphasized that the mathematical formalism has clear advantages, especially when a number of interacting fields have to be treated.

2. HAMILTONIAN OF SYSTEM

A. Spin-Spin Interaction

We assume that the crystal is composed of two equivalent cubic lattices interpenetrating each other, and that the nearest neighbors of the first lattice are on the second lattice. For simplicity, we consider only the exchange interaction J_{fg} between atoms f and g, which are on the two sublattices 1 and 2, respectively: Interactions of the type $J_{ff'}$ and $J_{gg'}$ will be disregarded for simplicity. The Hamiltonian of the spin subsystem is taken to be

$$H_s=\sum_{f,g} J_{fg}S_fS_g+A_{fg}S_f{}^zS_g{}^z-\mu B(\sum_f S_f{}^z+\sum_g S_g{}^z), \quad (1)$$

where the f and g summations are over the first and second sublattices, respectively. A_{fg} is an anisotropic constant along the z direction and adds an Ising-like contribution to the Hamiltonian. B is an external magnetic field along the z direction, and μ is a numerical factor containing the Landé factor associated with the spins S_f and S_g. J_{fg} and A_{fg} are positive and depend only on the distance between the lattice points f and g. We now introduce the following transformations.

Lattice 1:

$$\begin{aligned} S_f{}^+&=(2S_1)^{1/2}\left(\alpha_f-\frac{\alpha_f{}^\dagger\alpha_f\alpha_f}{2S_1}\right),\\ S_f{}^-&=(2S_1)^{1/2}\alpha_f{}^\dagger,\\ S_f{}^z&=S_1-\alpha_f{}^\dagger\alpha_f, \end{aligned} \quad (2a)$$

Lattice 2:

$$\begin{aligned} S_g{}^+&=(2S_2)^{1/2}\left(\beta_g{}^\dagger-\frac{\beta_g{}^\dagger\beta_g{}^\dagger\beta_g}{2S_2}\right),\\ S_g{}^-&=(2S_2)^{1/2}\beta_g,\\ S_g{}^z&=-S_2+\beta_g{}^\dagger\beta_g. \end{aligned} \quad (2b)$$

S_1 and S_2 are the intrinsic spins associated with the atoms of the lattices 1 and 2, respectively. Each lattice contains N atoms. In Eq. (2) we have assumed a system of units where $\hbar=1$. The operators α_f, $\alpha_f{}^\dagger$ (and β_g, $\beta_g{}^\dagger$) satisfy boson commutation rules, while all α_f operators commute with operators derived from β_g.

From (1) and (2) the Hamiltonian (1) can be written as

$$H_s=H_{pr}+H_{ma},$$

where

$$\begin{aligned} H_{pr}=&-\mu BN(S_1-S_2)-\sum_{f,g}(J_{fg}+A_{fg})S_1S_2\\ &+\sum_{f,g}(S_1S_2)^{1/2}(\alpha_f{}^\dagger\beta_g{}^\dagger+\alpha_f\beta_g)\\ &+\sum_f\{\mu B+\sum_g(J_{fg}+A_{fg})S_2\}\alpha_f{}^\dagger\alpha_f\\ &+\sum_g[-\mu B+\sum_f(J_{fg}+A_{fg})S_1]\beta_g{}^\dagger\beta_g\\ &-\sum_{f,g}(J_{fg}+A_{fg})\alpha_f{}^\dagger\alpha_f\beta_g{}^\dagger\beta_g, \end{aligned} \quad (3)$$

$$\begin{aligned} H_{ma}=-\tfrac{1}{2}\sum_{f,g}J_{fg}\Big[&\left(\frac{S_1}{S_2}\right)^{1/2}\alpha_f{}^\dagger\beta_g{}^\dagger\beta_g{}^\dagger\beta_g\\ &+\left(\frac{S_2}{S_1}\right)^{1/2}\alpha_f{}^\dagger\alpha_f\beta_g\Big]. \end{aligned} \quad (4)$$

H_{pr} corresponds to the Hamiltonian which would result from (1) if a linear Primakoff type of transformation had been carried out. H_{ma} gives the spin-spin interaction terms arising from the additional terms in the Maleev transformations. The Hamiltonian can be cast into the $\mathbf{k}$ representation as

$$H_s=\text{const}+H_0{}^0+H_{s\text{-}s},$$

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where

$$H_0^0=\sum_{\mathbf{k}}[B_1(0)\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}}+B_2(0)\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}}+A(\mathbf{k})\alpha_{\mathbf{k}}\beta_{-\mathbf{k}}+A(-\mathbf{k})\alpha_{-\mathbf{k}}^\dagger\beta_{\mathbf{k}}^\dagger], \quad (5)$$

$$H_{s-s}=\sum_{\mathbf{k}_1\mathbf{k}_2\mathbf{k}}[F(\mathbf{k})\alpha_{\mathbf{k}_1}^\dagger\alpha_{\mathbf{k}_1-\mathbf{k}}\beta_{\mathbf{k}_2-\mathbf{k}}^\dagger\beta_{\mathbf{k}_2}+F_1(\mathbf{k})\alpha_{\mathbf{k}}^\dagger\beta_{\mathbf{k}_1}^\dagger\beta_{\mathbf{k}_2}^\dagger\beta_{\mathbf{k}_1+\mathbf{k}_2+\mathbf{k}}+F_2(\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2)\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}_1}\alpha_{\mathbf{k}_2}\beta_{\mathbf{k}-\mathbf{k}_1-\mathbf{k}_2}], \quad (6)$$

and

$$\begin{aligned}B_1(0)&=\mu B+S_2(J_0+A_0),\\ B_2(0)&=-\mu B+S_1(J_0+A_0),\\ A_{\mathbf{k}}&=(S_1S_2)^{1/2}J_{\mathbf{k}},\\ F(\mathbf{k})&=-(J_{\mathbf{k}}+A_{\mathbf{k}})/N,\\ F_1(\mathbf{k})&=-(S_1/S_2)^{1/2}J_{\mathbf{k}}/2N,\\ F_2(\mathbf{k})&=-(S_2/S_1)^{1/2}J_{\mathbf{k}}/2N,\end{aligned} \quad (7)$$

with

$$\alpha_f=N^{-1/2}\sum_{\mathbf{k}}\alpha_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{r}_f},$$

$$J_{\mathbf{k}}=\sum_{f-g}J_{fg}e^{i\mathbf{k}\cdot(\mathbf{r}_f-\mathbf{r}_g)}, \quad \text{etc.} \quad (8)$$

We will define

$$\gamma_{\mathbf{k}}=\sum_{f-g}e^{i\mathbf{k}\cdot(\mathbf{r}f-\mathbf{r}_g)}$$

for subsequent use. In the above, sums over $\mathbf{k}$ are over the first Brillouin zone. This completes the description of the spin subsystem. Now we shall consider the phonon field and its interaction with the spin subsystem.

B. Spin-Phonon Interaction

We assume that the two sublattices, each of N atoms, can be regarded as a harmonic lattice of $2N$ atoms giving rise to a phonon subsystem,

$$H_{\rm ph}=\sum_q\omega_q(b_q^\dagger b_q+\tfrac{1}{2}), \quad (9a)$$

where $q=(\mathbf{q},\lambda)$, $\mathbf{q}$ and λ being the phonon wave vector and polarization index, respectively, ω_q is the frequency of acoustic vibrations of wave vector $\mathbf{q}$ and polarization λ; $b_q^\dagger$, b_q are the associated phonon creation and annihilation operators.

The interaction of the phonon subsystem with the spin subsystem is obtained in the usual way by expanding in power series the displacements of the atoms from their equilibrium positions and retaining only the linear terms in the expansion; the displacements $\delta\mathbf{r}_f$ (or $\delta\mathbf{r}_g$) are assumed to be small and are expressed in terms of $b_q^\dagger$ and b_q by

$$\begin{aligned}\delta\mathbf{r}&=N^{-1/2}\sum_q\mathbf{Y}(q)e^{i\mathbf{q}\cdot\mathbf{r}}A_q,\\ \mathbf{Y}(q)&=\mathbf{e}_q(4M\omega_q)^{-1/2},\\ A_q&=b_q+b_{-q}^\dagger,\end{aligned} \quad (9b)$$

M is the mass of an atom in the crystal, which contains $2N$ atoms in all. $\mathbf{e}_q$ is the unit vector defining the polarization of the mode q.

In deriving the spin-phonon interaction Hamiltonian, we shall retain the terms arising from the H_{pr} part, Eq. (3), of the total Hamiltonian and discard the contributions from the scattering terms in H_{ma}, Eq. (4).

The parameters of the displaced lattice are given by

$$\begin{aligned}J_{fg}'&=J_{fg}+\mathfrak{A}\cdot(\delta\mathbf{r}_f-\delta\mathbf{r}_g),\\ A_{fg}'&=A_{fg}+\mathfrak{A}''\cdot(\delta\mathbf{r}_f-\delta\mathbf{r}_g),\end{aligned} \quad (10)$$

with

$$\mathfrak{A}=\frac{\partial J_{fg}}{\partial\mathbf{r}_f}=-\frac{\partial J_{fg}}{\partial\mathbf{r}_g},$$

and similarly for $\mathfrak{A}''$.

For convenience we define $\mathfrak{A}'=\mathfrak{A}+\mathfrak{A}''$. In the above we have used the fact that J_{fg}, A_{fg} are translationally invariant.

On using the simplification

$$\begin{aligned}(S_1-\alpha_f^\dagger\alpha_f)\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}'}&\rightarrow\langle S_1^z\rangle\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}'},\\ -(S_1-\alpha_f^\dagger\alpha_f)(S_2-\beta_g^\dagger\beta_g)&\rightarrow\langle S_1^z\rangle\beta_g^\dagger\beta_g-\langle S_2^z\rangle\alpha_f^\dagger\alpha_f+\text{const},\end{aligned}$$

we finally obtain, for the spin-phonon interaction,

$$H_{s-p}=\sum_{\mathbf{k},q}[W_\alpha(q)\alpha_{\mathbf{k}+\mathbf{q}}^\dagger\alpha_{\mathbf{k}}A_q+W_\beta(-q)\beta_{-(\mathbf{k}+\mathbf{q})}^\dagger\beta_{-\mathbf{k}}A_{-q}+W_{\alpha\beta}(\mathbf{k},\mathbf{k}+\mathbf{q})\alpha_{\mathbf{k}+\mathbf{q}}^\dagger\beta_{-\mathbf{k}}^\dagger A_{-q}^\dagger+W_{\alpha\beta}^*(\mathbf{k},\mathbf{k}+\mathbf{q})\alpha_{\mathbf{k}+\mathbf{q}}\beta_{-\mathbf{k}}A_{-q}]. \quad (11)$$

The coupling terms occurring in (11) are given by

$$\begin{aligned}W_\alpha(q)&=\mathbf{Y}(q)\cdot[i\mathbf{q}z_1+m_2\mathfrak{A}'\gamma_{0q}]N^{-1/2},\\ W_\beta(q)&=\mathbf{Y}(q)\cdot[-i\mathbf{q}z_2+m_1\mathfrak{A}'\gamma_{0q}]N^{-1/2},\\ W_{\alpha\beta}(\mathbf{k},\mathbf{l})&=(S_1S_2)^{1/2}\mathbf{Y}(q)\cdot[i\mathbf{J}_{1\mathbf{k}}+\mathfrak{A}\gamma_{1\mathbf{k}}]N^{-1/2},\end{aligned} \quad (12)$$

where

$$\begin{aligned}z_1&=\mu B+m_2(J_0+A_0),\\ \mathbf{J}_{1\mathbf{k}}&=\mathbf{k}J_1-\mathbf{l}J_{\mathbf{k}},\quad \mathbf{l}=\mathbf{k}+\mathbf{q}\\ \gamma_{\mathbf{k}1}&=\gamma_{\mathbf{k}}-\gamma_1,\quad m_1=\langle S_1^z\rangle,\quad m_2=\langle S_2^z\rangle.\end{aligned} \quad (13)$$

The coupling constants $W_\alpha(q)$, $W_\beta(q)$, and $W_{\alpha\beta}(\mathbf{k},\mathbf{k}+\mathbf{q})$ go to zero as q tends to zero. The nondiagonal interaction $W_{\alpha\beta}$ is quite important as $\mathbf{k}\rightarrow 0$ in the sense that $|W_{\alpha\beta}|^2/|W_\alpha||W_\beta|$ tends to unity as $\mathbf{k}$ goes to zero.

Finally, we obtain for the total Hamiltonian of the system,

$$H=H_0^0+H_{\text{s-s}}+H_{\rm ph}+H_{\text{s-p}}, \quad (14)$$

where H_0^0 is given in Eq. (5), $H_{\text{s-s}}$ in Eq. (6), $H_{\rm ph}$ in Eq. (9), and $H_{\text{s-p}}$ in Eq. (12). Experimental data[10] on

[10] T. G. Phillips and H. M. Rosenberg, Rept. Progr. Phys. **29**, 285 (1966).

antiferromagnetic insulating crystals indicate that $H_{s\text{-}s}$ is usually more important than $H_{s\text{-}p}$.

3. DYSON'S EQUATION FOR GREEN'S FUNCTIONS

Most of the studies on spin-wave theory using Green's functions have been devoted to obtaining better decoupling approximations in order to obtain improvements on the random-phase approximation (RPA). In this study we follow a different approach in that we shall develop the expressions for the Green's functions in the form of a Dyson equation and then obtain an expression for the polarization operator of the system. This method enables us to study the dynamical effects in the system in a direct manner.

The Green's functions which we shall use are the double-time retarded and time-advanced Green's functions defined by Zubarev.

The Fourier components of the Green's functions are defined by

$$\langle\langle A(t),B(t')\rangle\rangle=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\langle\langle A,B\rangle\rangle_\omega e^{-i\omega(t-t')}d\omega,$$

and the subscript ω in $\langle\langle A,B\rangle\rangle_\omega$ will often be suppressed. We define the following symbols:

$$\begin{aligned}
&\boldsymbol{\alpha}_\mathbf{k}=\begin{pmatrix}\alpha_\mathbf{k}\\ \beta_{-\mathbf{k}}{}^\dagger\end{pmatrix},\quad \boldsymbol{\alpha}_\mathbf{k}{}^\dagger=(\alpha_\mathbf{k}{}^\dagger\ \beta_{-\mathbf{k}}{}^\dagger),\\
&\boldsymbol{\beta}_\mathbf{k}=\begin{pmatrix}\alpha_\mathbf{k}{}^\dagger\\ \beta_{-\mathbf{k}}\end{pmatrix},\quad \boldsymbol{\beta}_\mathbf{k}{}^\dagger=(\alpha_\mathbf{k}{}^\dagger\ \beta_{-\mathbf{k}}{}^\dagger),\\
&\bar{\alpha}_\mathbf{k}=\begin{pmatrix}\boldsymbol{\alpha}_\mathbf{k}\\ \boldsymbol{\beta}_{-\mathbf{k}}\end{pmatrix},\quad \bar{\alpha}_\mathbf{k}{}^\dagger=(\boldsymbol{\alpha}_\mathbf{k}{}^\dagger\ \boldsymbol{\beta}_\mathbf{k}{}^\dagger),
\end{aligned}\tag{15}$$

The operators are in the Heisenberg representation and their time arguments have been suppressed for convenience.

In the following we indicate vectors with four components and 4×4 matrices by a bar over the symbol. Vectors with two components and 2×2 matrices will be denoted in italic. This convention is indicated in Eqs. (15) and (16).

Further, we shall use the notation

$$\bar{X}=(\mathbf{A}:\mathbf{B})=\begin{pmatrix}\mathbf{A}&\mathbf{0}\\ \mathbf{0}&\mathbf{B}\end{pmatrix},$$

where $\mathbf{A}$ and $\mathbf{B}$ are 2×2 matrices and $\mathbf{0}$ is the null matrix.

The Fourier component of the Green's function $\bar{G}(\mathbf{k},\omega)\equiv\langle\langle\bar{\alpha}_\mathbf{k},\bar{\alpha}_\mathbf{k}{}^\dagger\rangle\rangle$ satisfies the equation

$$\omega\bar{G}(\mathbf{k},\omega)=\bar{I}+\langle\langle[\bar{\alpha}_\mathbf{k},H]_-,\bar{\alpha}_\mathbf{k}{}^\dagger\rangle\rangle.\tag{16}$$

By considering the equation of motion of $\bar{G}(\mathbf{k},t-t')$ with respect to t, for the Hamiltonian given in (14), it is easily established using Eq. (16) that the Fourier component $\bar{G}(\mathbf{k},\omega)$ is given by

$$[\bar{G}_0{}^0(\mathbf{k},\omega)]^{-1}\bar{G}(\mathbf{k},\omega)=\bar{I}+\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\bar{\alpha}_\mathbf{k}{}^\dagger\rangle\rangle,\tag{17a}$$

where $\bar{I}$ is the 4×4 unit matrix. Further,

$$\begin{aligned}
\bar{G}_0{}^0(\mathbf{k},\omega)&=[\mathbf{G}_0{}^0(\mathbf{k},\omega):\mathbf{G}_0{}^0(\mathbf{k},-\omega)],\\
\bar{\phi}(\mathbf{k})_q&=[\boldsymbol{\phi}(\mathbf{k})_q\quad\boldsymbol{\phi}'(\mathbf{k})_q]^T,
\end{aligned}\tag{17b}$$

where the superscript T indicates transposition to yield a column vector, with

$$(\mathbf{G}_0{}^0)^{-1}(\mathbf{k},\omega)=\begin{pmatrix}\omega-B_1(0)&-A(-\mathbf{k})\\ -A(\mathbf{k})&-\omega-B_2(0)\end{pmatrix},$$

$$\boldsymbol{\phi}(\mathbf{k})_q=\sum_q\mathbf{W}(\mathbf{k},\mathbf{k}-\mathbf{q})\boldsymbol{\alpha}_{\mathbf{k}-\mathbf{q}}A_q,$$

where

$$\mathbf{W}(\mathbf{k},\mathbf{k}-\mathbf{q})=\begin{pmatrix}W_\alpha(q)&W_{\alpha\beta}(\mathbf{k}-\mathbf{q},\mathbf{k})\\ W_{\alpha\beta}{}^*(\mathbf{k},\mathbf{k}-\mathbf{q})&W_\beta(q)\end{pmatrix}.$$

$\boldsymbol{\phi}'(\mathbf{k})_q$ is the transpose of $\boldsymbol{\phi}^\dagger(\mathbf{k})_q$, and is a column vector. The terms in $\bar{\phi}(\mathbf{k})_q$ arise from the spin-phonon interaction Hamiltonian.

The terms arising from the spin-wave scattering processes are given as follows.

$$\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=[\mathbf{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}\mathbf{F}'(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}]^T,\tag{17c}$$

with

$$\begin{aligned}
&\mathbf{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=\mathbf{F}_0(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\mathbf{F}_1(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\mathbf{F}_2(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\\
&\mathbf{F}'(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=\mathbf{F}_0{}^\dagger(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}{}^T+\mathbf{F}_3(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\\
&\mathbf{F}_0(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=\sum_{\mathbf{k}_1\mathbf{k}_2}F(\mathbf{k}_1-\mathbf{k}_2)\begin{pmatrix}\alpha_1\beta_{-\mathbf{k}_2}{}^\dagger\beta_{-\mathbf{k}_1}\\ \alpha_{\mathbf{k}_1}{}^\dagger\alpha_{\mathbf{k}_2}\beta_{-1}{}^\dagger\end{pmatrix},\\
&\mathbf{F}_1(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=\sum_{\mathbf{k}_1\mathbf{k}_2}\begin{pmatrix}F_1(\mathbf{k})\beta_{-1}{}^\dagger\beta_{-\mathbf{k}_2}{}^\dagger\beta_{-\mathbf{k}_1}\\ F_2(-\mathbf{k})\alpha_{\mathbf{k}_1}{}^\dagger\alpha_1\alpha_{\mathbf{k}_2}\end{pmatrix},\\
&\mathbf{F}_2(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=\sum_{\mathbf{k}_1\mathbf{k}_2}\begin{pmatrix}F_2(-\mathbf{k}_1)\alpha_1\alpha_{\mathbf{k}_2}\beta_{-\mathbf{k}_1}\\ F_1(\mathbf{k}_1)\alpha_{\mathbf{k}_1}{}^\dagger\beta_{-1}{}^\dagger\beta_{-\mathbf{k}_2}{}^\dagger\end{pmatrix},
\end{aligned}$$

where $\mathbf{l}=\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2$. Finally, we have

$$\mathbf{F}_3(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}=2\sum_{\mathbf{k}_1\mathbf{k}_2}\begin{pmatrix}F_2(-\mathbf{l})\beta_{-\mathbf{k}_2}\alpha_{\mathbf{k}_2}{}^\dagger\alpha_{\mathbf{k}_1}\\ F_1(\mathbf{l})\beta_{-\mathbf{k}_1}{}^\dagger\beta_{-\mathbf{k}_2}\alpha_1{}^\dagger\end{pmatrix}.\tag{17d}$$

The matrix Green's function $\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\bar{\alpha}_\mathbf{k}{}^\dagger\rangle\rangle$ occurs in Eq. (17) and is as yet unknown. Taking the equations of motion of the latter with respect to the time argument associated with $\bar{\alpha}_\mathbf{k}{}^\dagger$, we have the following result for the Fourier component:

$$\begin{aligned}
&\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\bar{\alpha}_\mathbf{k}{}^\dagger\rangle\rangle\bar{G}_0{}^{0-1}(\mathbf{k},\omega)\\
&\quad=\langle[\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\bar{\alpha}_\mathbf{k}{}^\dagger]_-\bar{X}\rangle_{t=t'}\\
&\qquad+\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\bar{F}'(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}{}^\dagger+\bar{\phi}(\mathbf{k})_{q'}\rangle\rangle,
\end{aligned}\tag{18a}$$

where

$$\bar{X}=(\mathbf{X}_-:\mathbf{X}^-)\tag{18b}$$

and $\mathbf{X}$ is the anti-unit matrix, viz.,

$$\mathbf{X}_{-}=\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix},\quad \mathbf{X}^{-}=\begin{pmatrix}-1 & 0\\ 0 & 1\end{pmatrix}. \tag{18c}$$

The commutator on the right-hand side of (18a) can be written as $\langle[\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\bar{\alpha}_{\mathbf{k}}^{\dagger}]_{-}\bar{X}\rangle$ as the phonon term gives no significant contribution except when phonon wave vector $\mathbf{q}$ becomes zero; but at this limit spin-phonon coupling terms occurring in $\bar{\phi}(\mathbf{k})_q$ vanish.

We let

$$\bar{K}(\mathbf{k})=\langle[\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\bar{\alpha}_{\mathbf{k}}^{\dagger}]_{-}\bar{X}\rangle. \tag{19}$$

Then it is easily proven that

$$\bar{K}(\mathbf{k})=(\mathbf{K}(\mathbf{k}):\mathbf{K}(\mathbf{k})),$$
$$\mathbf{K}(\mathbf{k})=\sum_{\mathbf{k}_1}\begin{pmatrix}F_0(0)n_{\mathbf{k}_1}{}^{\beta}+2F_2(-\mathbf{k}_1)n_{\mathbf{k}_1} & 2F_1(\mathbf{k})n_{\mathbf{k}_1}{}^{\beta}+F(\mathbf{k}-\mathbf{k}_1)n_{\mathbf{k}_1}\\ 2F_2(-\mathbf{k})n_{\mathbf{k}_1}{}^{\alpha}+F(\mathbf{k}-\mathbf{k}_1)n_{\mathbf{k}_1}{}^{\dagger} & F(0)n_{\mathbf{k}_1}{}^{\alpha}+2F_1(\mathbf{k}_1)n_{\mathbf{k}_1}{}^{\dagger}\end{pmatrix}, \tag{20}$$

where

$$n_{\mathbf{k}_1}{}^{\alpha}=\langle\alpha_{\mathbf{k}_1}{}^{\dagger}\alpha_{\mathbf{k}_1}\rangle,\quad n_{\mathbf{k}_1}{}^{\beta}=\langle\beta_{\mathbf{k}_1}{}^{\dagger}\beta_{\mathbf{k}_1}\rangle,\quad n_{\mathbf{k}_1}=\langle\alpha_{\mathbf{k}_1}\beta_{-\mathbf{k}_1}\rangle,\quad n_{\mathbf{k}_1}{}^{\dagger}=\langle\alpha_{\mathbf{k}_1}{}^{\dagger}\beta_{-\mathbf{k}_1}{}^{\dagger}\rangle.$$

Combining Eqs. (17) and (18) we obtain the equation for the Green's functions in the form of a Dyson equation by the following manipulation:

$$\bar{G}(\mathbf{k},\omega)\equiv\bar{G}_0{}^0(\mathbf{k},\omega)+\bar{G}_0{}^0(\mathbf{k},\omega)\bar{\pi}_0(\mathbf{k},\omega)\bar{G}(\mathbf{k},\omega) \tag{21a}$$

$$\bar{\pi}_0(\mathbf{k},\omega)=\bar{P}^0(\mathbf{k},\omega)/[1+\bar{G}_0{}^0(\mathbf{k},\omega)\bar{P}^0(\mathbf{k},\omega)], \tag{21b}$$

where

$$\bar{P}^0(\mathbf{k},\omega)=\bar{K}(\mathbf{k})+\bar{P}(\mathbf{k},\omega),$$
$$\bar{P}(\mathbf{k},\omega)=\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\ \bar{F}_0{}^{\dagger}(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}+\bar{F}_3(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}+\bar{\phi}^{\dagger}(\mathbf{k})_{q'}\rangle\rangle. \tag{21c}$$

The quantity given in Eq. (21b), $\bar{\pi}_0(\mathbf{k},\omega)$, is the polarization operator defined in terms of $\bar{G}_0{}^0(\mathbf{k},\omega)$. Now the problem is reduced to that of obtaining an expression for $\bar{\pi}_0(\mathbf{k},\omega)$ in an adequate approximation. The zeroth Green's function $\bar{G}_0{}^0(\mathbf{k},\omega)$ occurring in the polarization operator is the Green's function corresponding to the Hamiltonian $H_0{}^0$ which is a quadratic form in the boson operators $\alpha_{\mathbf{k}}$ and $\beta_{\mathbf{k}}$. The polarization operator can be developed in terms of an improved zero-order set of Green's functions $\bar{G}^0$ which correspond to the "equivalent Hamiltonian" to be discussed in the next section. However, if the equivalent Hamiltonian is used as the zeroth Hamiltonian, the polarization operator depends not only on $\bar{G}^0$ but on its inverse as well. This is considered in Appendix A.

4. RENORMALIZED ZEROTH-ORDER APPROXIMATION

Before proceeding to an evaluation of the polarization operator in detail, we will show how the zeroth-order Hamiltonian $H_0{}^0$, Eq. (5), can be replaced by an improved form, H^0, which takes into account all the static effects arising from the scattering processes described by the Maleev terms, given in Eq. (6). The new zeroth-order Green's functions, denoted by $\bar{G}^0(k,\omega)$, have temperature-dependent poles in contrast to those of $\bar{G}_0{}^0(k,\omega)$.

To obtain a first approximation to the polarization operator $\bar{\pi}_0(\mathbf{k},\omega)$ we note that $\bar{G}_0{}^0(\mathbf{k},\omega)\bar{P}^0(\mathbf{k},\omega)$ occurring in Eq. (21b) is small in comparison with unity; if the denominator of (21b) is nonzero, we let

$$\bar{\pi}_0(\mathbf{k},\omega)=\bar{P}^0(\mathbf{k},\omega)[1-\bar{G}_0{}^0(\mathbf{k},\omega)\bar{P}^0(\mathbf{k},\omega)+\cdots].$$

Thus we take the approximation

$$\bar{\pi}_0(\mathbf{k},\omega)=\bar{P}^0(\mathbf{k},\omega) \tag{22a}$$

when Eq. (21a) can be written as

$$[(\bar{G}_0{}^0)^{-1}(\mathbf{k},\omega)-\bar{K}(\mathbf{k})-\bar{P}(\mathbf{k},\omega)]\bar{G}(\mathbf{k},\omega)=\bar{I}. \tag{22b}$$

Thus we let

$$(\bar{G}^0)^{-1}(\mathbf{k},\omega)=(\bar{G}_0{}^0)^{-1}(\mathbf{k},\omega)-\bar{K}(\mathbf{k}). \tag{23}$$

Then it is easily established that

$$(\bar{G}^0)^{-1}(\mathbf{k},\omega)=[(\mathbf{G}_0{}^0)^{-1}(\mathbf{k},\omega):(\mathbf{G}_0{}^0)^{-1}(\mathbf{k},-\omega)] \tag{24a}$$

and

$$(\mathbf{G}^0)^{-1}(\mathbf{k},\omega)=\begin{pmatrix}\omega-E_{11}(0) & -E_{12}(\mathbf{k})\\ -E_{21}(\mathbf{k}) & -\omega-E_{22}(0)\end{pmatrix}, \tag{24b}$$

where

$$E_{11}(0)=\mu B-m_2J_0'-\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\left(\frac{S_2}{S_1}\right)^{1/2}n_{\mathbf{k}_1},$$
$$E_{12}(\mathbf{k})=m_2J_{\mathbf{k}}\left(\frac{S_1}{S_2}\right)^{1/2}-\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}'n_{\mathbf{k}_1},$$
$$E_{21}(\mathbf{k})=m_1J_{\mathbf{k}}\left(\frac{S_2}{S_1}\right)^{1/2}-\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}'n_{\mathbf{k}_1}{}^{\dagger}, \tag{25}$$
$$E_{22}(0)=-\mu B+m_1J_0'-\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\left(\frac{S_1}{S_2}\right)^{1/2}n_{\mathbf{k}_1}{}^{\dagger}.$$

Equation (24) corresponds to an equivalent Hamiltonian

$$H^0=\text{const}+\sum_{\mathbf{k}}[E_{11}(0)\alpha_{\mathbf{k}}{}^{\dagger}\alpha_{\mathbf{k}}+E_{12}(\mathbf{k})\alpha_{\mathbf{k}}{}^{\dagger}\beta_{-\mathbf{k}}{}^{\dagger}+E_{21}(\mathbf{k})\alpha_{\mathbf{k}}\beta_{-\mathbf{k}}+E_{22}(0)\beta_{\mathbf{k}}{}^{\dagger}\beta_{\mathbf{k}}], \tag{26}$$

and defines the renormalized zeroth-order approximation (RZOA).

We now discuss the RZOA in detail. The poles of the Green's function $\bar{G}^0(\mathbf{k},\omega)$ given in Eq. (24) are given by the solutions of

$$[\omega-E_1(\mathbf{k})][\omega+E_2(\mathbf{k})]\times[\omega-E_2(\mathbf{k})][\omega+E_1(\mathbf{k})]=0, \quad (27a)$$

where

$$E_1(\mathbf{k})=E_\mathbf{k}+Q_0, \quad E_2(\mathbf{k})=E_\mathbf{k}-Q_0, \quad (27b)$$

with

$$Q_0=\tfrac{1}{2}[E_{11}(0)-E_{22}(0)], \qquad E_\mathbf{k}^2=\tfrac{1}{4}[E_{11}(0)+E_{22}(0)]^2-E_{12}(\mathbf{k})E_{21}(\mathbf{k}). \quad (27c)$$

We give below the full expressions for Q_0 and $E_\mathbf{k}$:

$$Q_0=-\tfrac{1}{2}J_0'(m_1+m_2)+\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\left[\left(\frac{S_1}{S_2}\right)^{1/2}-\left(\frac{S_2}{S_1}\right)^{1/2}\right]n_{\mathbf{k}_1}+\mu B, \quad (28a)$$

$$(E_\mathbf{k})^2=\left\{\tfrac{1}{2}J_0'(m_1-m_2)-\frac{1}{2N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\left[\left(\frac{S_1}{S_2}\right)^{1/2}+\left(\frac{S_2}{S_1}\right)^{1/2}\right]n_{\mathbf{k}_1}\right\}^2+J_\mathbf{k}^2m_1m_2+\frac{J_\mathbf{k}}{N}\left[\left(\frac{S_2}{S_1}\right)^{1/2}m_1-\left(\frac{S_1}{S_2}\right)^{1/2}m_2\right]\times\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}n_{\mathbf{k}_1}-\left(\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}n_{\mathbf{k}_1}\right)^2, \quad (28b)$$

where we have used

$$J_\mathbf{k}'=J_\mathbf{k}+A_\mathbf{k}$$

and taken (cf. Eq. 20)

$$n_\mathbf{k}=n_\mathbf{k}^\dagger. \quad (28c)$$

From (28a) we note that Q_0 is smaller than $E_\mathbf{k}$ since, for an antiferromagnet, m_1 and m_2 have opposite signs. Thus the poles at $\omega=E_1(\mathbf{k})$, $\omega=E_2(\mathbf{k})$ gives the (positive) excitation energies of the system. The other two roots of (27b) are rejected.

It is interesting to compare these results with those of the simple RPA[5] and the CD scheme.[6]

In the case of RPA, we have

$$Q_0(\mathrm{RPA})=-\tfrac{1}{2}J_0'(m_1+m_2)+\mu B,$$
$$E_\mathbf{k}^2(\mathrm{RPA})=[\tfrac{1}{2}J_0'(m_1-m_2)]^2+J_\mathbf{k}^2m_1m_2,$$

and these are the leading terms of Eq. (28).

In treating antiferromagnetism by Callen's method, the decoupling to be used can be written as

$$\langle\langle S_f^zS_g^+,S_{f'}^-\rangle\rangle=\langle S_f^z\rangle\langle\langle S_g^+,S_{f'}^-\rangle\rangle-\alpha_1\langle S_f^-S_g^+\rangle\langle\langle S_f^+,S_{f'}^-\rangle\rangle,$$
$$\langle\langle S_g^zS_f^+,S_{f'}^-\rangle\rangle=\langle S_g^z\rangle\langle\langle S_f^+,S_{f'}^-\rangle\rangle-\alpha_2\langle S_f^-S_g^+\rangle\langle\langle S_g^+,S_{f'}^-\rangle\rangle,$$

where α_1 and α_2 are the CD parameters. In choosing α_1 (and α_2) according to the physical criteria given by Callen,[11] some ambiguity arises in that there are two possible choices for α_1 and similarly for α_2. Anderson and Callen[6] and Lee and Liu,[7] have treated the $S_1=S_2$ antiferromagnet and found that one of the possible choices leads to unsatisfactory results. For the more general case $S_1\neq S_2$ treated here, if the results of CD are to agree with Eq. (28), it is found necessary that α_1 and α_2 be chosen such that

$$\alpha_1=-\langle S_2^z\rangle/2S_1S_2, \quad \alpha_2=-\langle S_1^z\rangle/2S_1S_2.$$

When $S_1=S_2$, this is indeed the choice made by Anderson and Callen.[6] The results obtainable from Callen's theory for the present system are

$$Q_0(\mathrm{CD})=\tfrac{1}{2}J_0'(m_1+m_2)+\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\times\left[\left(\frac{S_1}{S_2}\right)^{1/2}\frac{m_1}{S_1}+\frac{m_2}{S_2}\left(\frac{S_2}{S_1}\right)^{1/2}\right]n_{\mathbf{k}_1}+\mu B,$$

$$E_\mathbf{k}^2(\mathrm{CD})=\left\{\tfrac{1}{2}J_0'(m_1-m_2)-\frac{1}{2N}\sum_{\mathbf{k}_1}J_{\mathbf{k}_1}\times\left[\left(\frac{S_1}{S_2}\right)^{1/2}\frac{m_1}{S_1}-\frac{m_2}{S_2}\left(\frac{S_2}{S_1}\right)^{1/2}\right]n_{\mathbf{k}_1}\right\}+J_\mathbf{k}^2m_1m_2-\frac{J_\mathbf{k}}{N}\left[\left(\frac{S_2}{S_1}\right)^{1/2}\frac{m_1m_2}{S_2}+\left(\frac{S_1}{S_2}\right)^{1/2}\frac{m_1m_2}{S_1}\right]\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}'n_{\mathbf{k}_1}+\frac{m_1m_2}{S_1S_2}\left(\frac{1}{N}\sum_{\mathbf{k}_1}J_{\mathbf{k}-\mathbf{k}_1}n_{\mathbf{k}_1}\right)^2. \quad (29)$$

At sufficiently low temperatures $m_1/S_1\approx1$, $m_2/S_2\approx-1$. Thus the expressions in (28) and (29) become equivalent at small spin deviations. This relationship between the results obtained from the ideal spin-wave Hamiltonian and the results of the Callen-decoupling approximation is easily established for the Heisenberg ferromagnet as well. We note in particular that the ground-state energy calculated with our RZOA will be identical with that from Callen's decoupling approximation.

The various expectation values, viz., m_1, m_2, and $n_\mathbf{k}$, which occur in (28), can be easily evaluated through the spectral representations of the Green's functions. Defining $\theta=k_BT$ to be the product of the Boltzmann constant and the absolute temperature, it is easily

[11] H. B. Callen, Phys. Rev. **130**, 890 (1963).

established that

$$n_{\mathbf{k}}{}^{\alpha}=\langle\alpha_{\mathbf{k}}{}^{\dagger}\alpha_{\mathbf{k}}\rangle=\frac{1}{4}\Big[\Big(\frac{R}{E_{\mathbf{k}}}+1\Big)\coth\Big(\frac{E_1(\mathbf{k})}{2\theta}\Big)+\Big(\frac{R}{E_{\mathbf{k}}}-1\Big)\coth\Big(\frac{E_2(\mathbf{k})}{2\theta}\Big)-2\Big],$$

$$n_{\mathbf{k}}{}^{\beta}=\langle\beta_{\mathbf{k}}{}^{\dagger}\beta_{\mathbf{k}}\rangle=\frac{1}{4}\Big[\Big(\frac{R}{E_{\mathbf{k}}}-1\Big)\coth\Big(\frac{E_1(\mathbf{k})}{2\theta}\Big)+\Big(\frac{R}{E_{\mathbf{k}}}+1\Big)\coth\Big(\frac{E_2(\mathbf{k})}{2\theta}\Big)-2\Big],\tag{30a}$$

$$n_{\mathbf{k}}=\langle\alpha_{\mathbf{k}}\beta_{-\mathbf{k}}\rangle=\frac{-E_{21}(\mathbf{k})}{4E_{\mathbf{k}}}\times\Big[\coth\Big(\frac{E_2(\mathbf{k})}{2\theta}\Big)+\coth\Big(\frac{E_1(\mathbf{k})}{2\theta}\Big)\Big],$$

$$n_{\mathbf{k}}{}^{\dagger}=\langle\alpha_{\mathbf{k}}{}^{\dagger}\beta_{-\mathbf{k}}\rangle=\frac{-E_{12}(\mathbf{k})}{4E_{\mathbf{k}}}\times\Big[\coth\Big(\frac{E_2(\mathbf{k})}{2\theta}\Big)+\coth\Big(\frac{E_1(\mathbf{k})}{2\theta}\Big)\Big],$$

where

$$R=\tfrac{1}{2}[E_{11}(0)+E_{22}(0)].$$

Finally,

$$m_1=S_1-\frac{1}{N}\sum_{\mathbf{k}}n_{\mathbf{k}}{}^{\alpha},\quad m_2=-S_2+\frac{1}{N}\sum_{\mathbf{k}}n_{\mathbf{k}}{}^{\beta}.\tag{30b}$$

A zeroth-order approximation to these quantities can be obtained by taking $E_{11}(0)=B_1(0)$, $E_{12}(\mathbf{k})=E_{21}(\mathbf{k})=A(\mathbf{k})$, and $E_{22}(\mathbf{k})=B_2(0)$, as in (18c), and then the matrix elements of the RZOA are obtained from (25). In using Maleev's transformation approach to problems where $S\neq\frac{1}{2}$, we note that, unlike in other methods, the very simple relations given in (30b) suffice to give the sublattice magnetizations.

5. EVALUATION OF POLARIZATION OPERATOR

In evaluating the polarization operator, we shall use the equivalent Hamiltonian H^0 of the RZOA to describe the spin subsystems. $H_{\rm ph}$ will describe the phonon subsystem. This zeroth-order evaluation of the polarization operator will be denoted by a superscript zero, viz., $\bar{P}^0(\mathbf{k},\omega)$. Thus for the total Hamiltonian of the system, we use the effective Hamiltonian

$$H_{\rm eff}=H^0+H_{\rm ph},\tag{31}$$

rather than (14).

Further, in evaluating $\bar{P}^0(\mathbf{k},\omega)$, we shall carry out the discussion entirely in terms of $\mathbf{P}^0(\mathbf{k},\omega)$, since the other component of $\bar{P}^0(\mathbf{k},\omega)$ is such that

$$\bar{P}^0(\mathbf{k},\omega)=[\mathbf{P}^0(\mathbf{k},\omega):\mathbf{P}^0(\mathbf{k},-\omega)].\tag{32}$$

It is convenient to cast H^0 into diagonal form using a Bogoliubov-Valantin transformation. Thus the operators $\alpha_{\mathbf{k}},\alpha_{\mathbf{k}}{}^{\dagger},\beta_{\mathbf{k}},\beta_{-\mathbf{k}}{}^{\dagger}$ are expressed in terms of the quasiparticle operators γ_1,γ_2 and their conjugates by

$$\boldsymbol{\alpha}\equiv\mathbf{C}(\mathbf{k})\boldsymbol{\gamma}(\mathbf{k})\equiv\begin{pmatrix}U_{\mathbf{k}} & V_{-\mathbf{k}}{}^{*}\\ V_{\mathbf{k}} & U_{-\mathbf{k}}{}^{*}\end{pmatrix}\begin{pmatrix}\gamma_1(\mathbf{k})\\ \gamma_2{}^{\dagger}(-\mathbf{k})\end{pmatrix},\tag{33}$$

where $\mathbf{C}_{\mathbf{k}}$ is the canonical transformation matrix. The elements of $\mathbf{C}_{\mathbf{k}}$ are easily determined to be given by

$$u_{\mathbf{k}}{}^2=(R+E_{\mathbf{k}})/2E_{\mathbf{k}},\quad v_{\mathbf{k}}{}^2=(R-E_{\mathbf{k}})/2E_{\mathbf{k}}.\tag{34}$$

The symbols R and $E_{\mathbf{k}}$ have already been defined in Eqs. (30) and (27).

$H_{\rm eff}$ is now given by

$$H_{\rm eff}=\sum_{\mathbf{k}}[E_1(\mathbf{k})\gamma_1{}^{\dagger}(\mathbf{k})\gamma_1(\mathbf{k})+E_2(\mathbf{k})\gamma_2{}^{\dagger}(\mathbf{k})\gamma_2(\mathbf{k})]+\sum_{q}\omega_q b_q{}^{\dagger}b_q.\tag{35}$$

An estimate of the importance of $v_{\mathbf{k}}$ in comparison to $u_{\mathbf{k}}$ is of interest. If the surd in the expression for $E_{\mathbf{k}}$ is expanded and the first term is retained, we have

$$v_{\mathbf{k}}{}^2/u_{\mathbf{k}}{}^2=x/(1-x),$$

with

$$x=\frac{E_{12}E_{21}}{(2R)^2}=\frac{S_1S_2}{(S_1+S_2)^2}\frac{\cos(\mathbf{k}\cdot\mathbf{b})}{(1+A/J)^2},$$

$$\mathbf{r}_f-\mathbf{r}_g=\mathbf{b},\quad J_{fg}=J,\quad A_{fg}=A,\quad A\ll J,$$

where only nearest-neighbor sums are taken. It is evident that when $S_1\approx S_2$, the two amplitudes $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ are of similar magnitude, especially at $\mathbf{k}$ close to zero, $v_{\mathbf{k}}{}^2/u_{\mathbf{k}}{}^2\approx\frac{1}{3}$. In Kashcheev's[9] treatment of the antiferromagnet, he seems to have ignored important nondiagonal terms involving $v_{\mathbf{k}}$ amplitudes.

Using the RZOA Green's function $\bar{G}^0(\mathbf{k},\omega)$, Eq. (23b) can be rewritten as

$$[\bar{G}^{0-1}(\mathbf{k},\omega)-\bar{P}^0(\mathbf{k},\omega)]\bar{G}(\mathbf{k},\omega)=\bar{I},\tag{36}$$

where we have already made the approximation given in Eq. (23a) for the polarization operator. From Eq. (22a) we have

$$\bar{P}^0(\mathbf{k},\omega)=\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2}+\bar{\phi}(\mathbf{k})_q,\ \bar{F}_0{}^{\dagger}(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}+\bar{F}_3(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}+\bar{\phi}^{\dagger}(\mathbf{k})_{q'}\rangle\rangle^0.$$

It is evident that $\bar{P}^0(\mathbf{k},\omega)$ could be written as

$$\bar{P}^0(\mathbf{k},\omega)=\bar{P}^0(\mathbf{k},\omega)_{\text{s-s}}+\bar{P}^0(\mathbf{k},\omega)_{\text{s-p}},$$

with

$$\bar{P}^0(\mathbf{k},\omega)_{\text{s-s}}=\langle\langle\bar{F}(\mathbf{k})_{\mathbf{k}_1\mathbf{k}_2},\bar{F}_0{}^{\dagger}(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}+\bar{F}_3(\mathbf{k})_{\mathbf{k}_1'\mathbf{k}_2'}\rangle\rangle^0,$$
$$\bar{P}^0(\mathbf{k},\omega)_{\text{s-p}}=\langle\langle\bar{\phi}(\mathbf{k})_q,\bar{\phi}^{\dagger}(\mathbf{k})_{q'}\rangle\rangle^0.\tag{37}$$

Thus the contribution to the polarization operator from the spin-spin scattering process can be evaluated separately from that due to the phonon field.

The detailed evaluation of the expressions for the polarization operator is given in Appendix B. In the following we shall outline the approach used therein.

Using the canonical transformation of Eq. (33), $\bar{P}^0(\mathbf{k},\omega)$ could be expressed in terms of the Green's functions defined below:

$$\begin{aligned}&\langle\langle\sigma_r(\mathbf{k}_i),\sigma_r^\dagger(\mathbf{k}_i')\rangle\rangle,\\&\langle\langle\sigma_r{}^{L\dagger}(\mathbf{k}_i),\sigma_r{}^L(\mathbf{k}_i')\rangle\rangle,\quad r=1,2,3\end{aligned}\tag{38}$$

and simpler forms like

$$\langle\langle\sigma_1(\mathbf{k}_i),\gamma_1^\dagger(\mathbf{k})\rangle\rangle,$$

where

$$\begin{aligned}\sigma_1&=\sigma_1(\mathbf{k}_i)=\sigma_1(\mathbf{k}_1,\mathbf{k}_2,\mathbf{l})=\gamma_1(\mathbf{l})\gamma_2^\dagger(-\mathbf{k}_2)\gamma_2(-\mathbf{k}_1),\\\sigma_2&=\sigma_2(\mathbf{k}_i)=\sigma_2(\mathbf{k}_1,\mathbf{k}_2,\mathbf{l})=\gamma_2^\dagger(-\mathbf{l})\gamma_2^\dagger(-\mathbf{k}_2)\gamma_2(-\mathbf{k}_1),\\\sigma_3&=\sigma_3(\mathbf{k}_i)=\sigma_3(\mathbf{k}_1,\mathbf{k}_2,\mathbf{l})=\gamma_1(\mathbf{l})\gamma_1(\mathbf{k}_2)\gamma_2(-\mathbf{k}_1),\end{aligned}\tag{39}$$

with $\mathbf{l}=\mathbf{k}+\mathbf{k}_1-\mathbf{k}_2$.

Given any expression, e.g., $\sigma(1,2,\mathbf{k}_i)$, which depends on the magnon branch numbers 1 and 2 and the wave vectors $\mathbf{k}_i$, the expression $\sigma(2,1,-\mathbf{k}_i)$ will be called a lattice dual and this is indicated by a superscript L, as in $\sigma^L(1,2,\mathbf{k}_i)$, for example.

The six Green's functions defined in Eq. (39) are easily evaluated using the effective Hamiltonian of Eq. (35). Expressions for these are given in Eq. (B4) of Appendix B. The Green's functions $\langle\langle\sigma_r,\sigma_r^\dagger\rangle\rangle$ and their lattice-dual forms describe three different physical processes and their conjugate processes. For example, the processes described by $\langle\langle\sigma_1,\sigma_1^\dagger\rangle\rangle$ involves the scattering of a magnon of a given type into two others belonging to the two spin-wave branches. The reverse process is the fusion of two magnons of types 1 and 2 into a single magnon. A crude examination of the probability amplitudes associated with the three processes indicates that for small wave vectors these are of comparable importance.

In the treatment by Kashcheev,[9] where a macroscopic Hamiltonian has been used, an expression for the spin-spin polarization operator has been given in his Eq. (A5). Unlike his energy denominators, the energy denominators appearing in Eqs. (B4) of this work are temperature-dependent. The factors involving the magnon and phonon occupation numbers obtained by us are the same as those of Kashcheev. An adequate discussion of the terms occurring in $\mathbf{P}(\mathbf{k},\omega)_{\text{s-s}}$ is possible only if suitable numerical computations are carried out. This will be the subject of a later publication.

The spin-phonon part of the polarization operator can be expressed in terms of the matrix Green's function

$$\boldsymbol{\Gamma}(\mathbf{k}-\mathbf{q},\mathbf{q},\omega)=\left\langle\!\!\left\langle\begin{pmatrix}\gamma_1(\mathbf{k}-\mathbf{q})A_q\\\gamma_2^\dagger(\mathbf{k}-\mathbf{q})A_{-q}{}^\dagger\end{pmatrix},\right.\right.$$
$$\left.\left.[A_q{}^\dagger\gamma_1^\dagger(\mathbf{k}-\mathbf{q})\;A_q\gamma_2(\mathbf{k}-\mathbf{q})]\right\rangle\!\!\right\rangle.\tag{40}$$

This is evaluated using the Hamiltonian

$$H^{(0)}=H_{\text{eff}}+H_{\text{ph}},$$

whereby both parts of the polarization operator, viz., $\mathbf{P}(\mathbf{k},\omega)_{\text{s-s}}$ and $\mathbf{P}(\mathbf{k},\omega)_{\text{s-p}}$ are evaluated self-consistently to the same order of accuracy. The Green's function (40), evaluated to this order of accuracy, is denoted by $\boldsymbol{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$ and is diagonal, viz.,

$$\begin{aligned}\boldsymbol{\Gamma}_{11}{}^0(\mathbf{k}-\mathbf{q},q,\omega)&=\frac{1}{2}\left(\frac{1+2n_{\mathbf{k}-\mathbf{q}}{}^{(1)}+N_q}{\omega-E(\mathbf{k}-\mathbf{q})-\omega_q}\right.\\&\qquad\left.+\frac{N_q-1-2n_{\mathbf{k}-\mathbf{q}}{}^{(1)}}{\omega-E(\mathbf{k}-\mathbf{q})+\omega_q}\right),\\\boldsymbol{\Gamma}_{12}{}^0(\mathbf{k}-\mathbf{q},q,\omega)&=0.\end{aligned}\tag{41}$$

The details of the derivation of the expressions for $\boldsymbol{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$, and also the higher-order result $\boldsymbol{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},\mathbf{q},\omega)$ are described in Appendix B. In (41), N_q denotes the c number $\langle A_q{}^\dagger A_q\rangle$.

The expressions given for the spin-phonon contribution to the polarization operator given by us may be compared with those of Kashcheev [Eq. (A6)] where our energy denominators are temperature-independent. Further, since the nondiagonal interactions have been consistently taken into account, the numerators in our expressions carry the magnon and phonon occupation numbers in a different manner. As in the case of the spin-spin interaction, a proper analysis of $\mathbf{P}(\mathbf{k},\omega)_{\text{s-p}}$ needs the support of numerical computation.

Equations (B4), (B5), and (B13) of Appendix B give the expressions for $\mathbf{P}(\mathbf{k},\omega)_{\text{s-s}}$ and $\mathbf{P}(\mathbf{k},\omega)_{\text{s-p}}$ to order $\mathbf{P}^0(\mathbf{k},\omega)$. Thus the first approximation to $\mathbf{G}(\mathbf{k},\omega)$, denoted by $\mathbf{G}^{(1)}(\mathbf{k},\omega)$, is given by

$$\begin{aligned}\mathbf{G}_{11}{}^{(1)}(\mathbf{k},\omega)&=\langle\langle\alpha_{\mathbf{k}},\alpha_{\mathbf{k}}{}^\dagger\rangle\rangle=[\omega+\mathbf{E}_{22}(\mathbf{k},\omega)]/D(\mathbf{k},\omega),\\\mathbf{G}_{12}{}^{(1)}(\mathbf{k},\omega)&=\langle\langle\alpha_{\mathbf{k}},\beta_{-\mathbf{k}}\rangle\rangle=-\mathbf{E}_{12}(\mathbf{k},\omega)/D(\mathbf{k},\omega),\\\mathbf{G}_{21}{}^{(1)}(\mathbf{k},\omega)&=\langle\langle\beta_{-\mathbf{k}}{}^\dagger,\alpha_{\mathbf{k}}{}^\dagger\rangle\rangle=-\mathbf{E}_{12}(\mathbf{k},\omega)/D(\mathbf{k},\omega),\\\mathbf{G}_{22}{}^{(1)}(\mathbf{k},\omega)&=\langle\langle\beta_{-\mathbf{k}}{}^\dagger,\beta_{-\mathbf{k}}\rangle\rangle=[-\omega+\mathbf{E}_{11}(\mathbf{k},\omega)]/D(\mathbf{k},\omega),\end{aligned}\tag{42}$$

where

$$\begin{aligned}\mathbf{E}_{ij}(\mathbf{k},\omega)&=E_{ij}(\mathbf{k})+\mathbf{P}_{ij}{}^0(\mathbf{k},\omega),\\D(\mathbf{k},\omega)&=[\omega-\mathbf{E}_{11}(\mathbf{k},\omega)][\omega+\mathbf{E}_{22}(\mathbf{k},\omega)]\\&\qquad+\mathbf{E}_{12}(\mathbf{k},\omega)\,\mathbf{E}_{21}(\mathbf{k},\omega).\end{aligned}$$

6. COMPLEX SUSCEPTIBILITY

In order to study the line shape associated with the absorption of energy from an alternating field, we shall derive an expression for the frequency-dependent susceptibility $\chi(\omega)$, of the system. We assume that a weak external magnetic field of frequency ω, linearly polarized and applied in the xy plane, is present in addition to the static field B, which, as before, defines the z direction. Then it can be shown that $\chi(\omega)$ is proportional to the Fourier component of a suitable

retarded Green's function, viz.,

$$\chi^{r,s}(\omega)=-2\pi\langle\langle M^r,M^s\rangle\rangle_\omega, \quad r,s=x,\ y,\ \text{or}\ z \quad (43a)$$

where M^r is the operator of the projection of the magnetic moment in the r direction. We thus have

$$M=\mu S^r=\mu(\sum_f S_f{}^r+\sum_g S_g{}^r). \quad (43b)$$

Expressing S^r in terms of the operators α_k, β_k, it is easily proven that

$$\langle\langle S^x,S^x\rangle\rangle_\omega{}^u=\langle\langle S^y,S^y\rangle\rangle_\omega{}^u$$
$$=\sum_{\mathbf{k}}[f^u(\mathbf{k},\omega)+f^u(-\mathbf{k},-\omega)]\gamma_{\mathbf{k}}, \quad (43c)$$

with

$$\gamma_{\mathbf{k}}=\gamma_{-\mathbf{k}}=\sum_{f-g} e^{i\mathbf{k}\cdot(\mathbf{r}_f-\mathbf{r}_g)},$$

as before. The superscript u indicates expressions for the undisplaced lattice, i.e., in the absence of the phonon field. The expression $f_{\mathbf{k}}{}^u(\omega)$ is given by

$$f^u(\mathbf{k},\omega)=m_1\left[\mathbf{G}_{11}{}^{(1)}(\mathbf{k},\omega)+\left(\frac{S_2}{S_1}\right)^{1/2}\mathbf{G}_{12}{}^{(1)}(\mathbf{k},\omega)\right]$$
$$-m_2\left[\mathbf{G}_{22}{}^{(1)}(\mathbf{k},\omega)+\left(\frac{S_1}{S_2}\right)^{1/2}\mathbf{G}_{21}{}^{(1)}(\mathbf{k},\omega)\right]. \quad (44)$$

In deriving (44) we have decoupled the Green's functions arising from the Maleev terms in the following manner. For example,

$$\langle\langle\alpha_{\mathbf{k}}+\alpha_{-\mathbf{k}}{}^\dagger,\alpha_{-\mathbf{k}_1}{}^\dagger\alpha_{\mathbf{k}_2}\alpha_{\mathbf{k}_1-\mathbf{k}_2-\mathbf{k}}\rangle\rangle\approx 2n_{\mathbf{k}_1}{}^\alpha\langle\langle\alpha_{\mathbf{k}}+\alpha_{-\mathbf{k}}{}^\dagger,\alpha_{-\mathbf{k}}\rangle\rangle,$$

where we have used the fact that $\langle\alpha_{\mathbf{k}}\alpha_{-\mathbf{k}}\rangle$ is zero. Also, we note that in (44) we have used the Green's functions $\mathbf{G}^{(1)}(\mathbf{k},\omega)$ of the first approximation.

Susceptibility in the presence of the phonon field. As the Green's functions have been evaluated with the effect of the phonon field taken into account, self-consistency demands that the susceptibility itself be calculated to the same accuracy. If (43b) is cast into the momentum representation and the linear terms in the displacements of the lattice vectors are retained, we obtain

$$\langle\langle S^x,S^x\rangle\rangle=\sum_{\mathbf{k}}[f(\mathbf{k},q,\omega)+f(-\mathbf{k},-q,-\omega)]\gamma_{\mathbf{k}}. \quad (45a)$$

If we define

$$\boldsymbol{\Omega}=\boldsymbol{\Omega}(\mathbf{k}-\mathbf{q},q,\omega)=\langle\langle\gamma_{\mathbf{k}},\gamma_{\mathbf{k}-\mathbf{q}}{}^\dagger A_q{}^\dagger\rangle\rangle-\langle\langle\gamma_{\mathbf{k}-\mathbf{q}}A_q,\gamma_{\mathbf{k}}{}^\dagger\rangle\rangle,$$

then $f(\mathbf{k},q,\omega)$ can be written as $f(\mathbf{k},q,\omega)=f(\mathbf{k},\omega)+:$

$$\sum_{q^i,j}\tfrac{1}{2}\mathbf{k}'N^{-1/2}\mathbf{Y}(q)[it_{ij}(\mathbf{k},\mathbf{k}')\boldsymbol{\Omega}_{ij}$$
$$+\mathbf{k}'\mathbf{Y}(q)N^{-1/2}t_{ij}(\mathbf{k},\mathbf{k}')\boldsymbol{\Gamma}_{ij}], \quad i=1,2, \quad j=1,2$$

where

$$\mathbf{k}'=\mathbf{k}-\mathbf{q},$$
$$t_{11}(\mathbf{k},\mathbf{l})=(S_1{}^{1/2}u_{\mathbf{k}}+S_2{}^{1/2}v_{\mathbf{l}})(S_1{}^{1/2}v_{\mathbf{k}}+S_1{}^{1/2}u_{\mathbf{l}}),$$
$$t_{12}(\mathbf{k},\mathbf{l})=(S_1{}^{1/2}u_{\mathbf{k}}+S_2{}^{1/2}v_{\mathbf{k}})(S_2{}^{1/2}u_{\mathbf{l}}+S_1{}^{1/2}v_{\mathbf{l}}). \quad (45b)$$

t_{21} is obtained from t_{12} by the interchange $u\leftrightarrow v$, and t_{22} from t_{11} by $S_1\leftrightarrow S_2$.

All the Green's functions occurring in (45) have been evaluated in the first approximation. It will be seen subsequently that a self-consistent treatment to order $1/N$ will require $\boldsymbol{\Omega}(\mathbf{k},\mathbf{k}-\mathbf{q})$ in the first approximation as in Eq. (B14), and $\boldsymbol{\Gamma}(\mathbf{k},\mathbf{k}-\mathbf{q})$ to zero order as in Eq. (B12).

A. Imaginary Part of $\chi(\omega)$ in RZOA

To begin with we shall examine the imaginary part of the susceptibility tensor in the RZOA, using Eqs. (43) and (44). In this approximation the imaginary part consists of a set of δ-function-like peaks, and when suitable limits are taken we easily recover the results of classical theory and those of, for example, P'u Fu-Cho.[8]

From (43), we have

$$\mathrm{Im}\chi^{xx}(\omega)=-2\pi\mu^2\,\mathrm{Im}\sum_{\mathbf{k}}\gamma_{\mathbf{k}}[f^u(\mathbf{k},\omega)+f^u(-\mathbf{k},-\omega)].$$

For simplicity, if we take the case where the two sublattices are equivalent, with $S_1=S_2$, we obtain, using the notation of Eq. (30),

$$(-i\pi/2E_{\mathbf{k}})\{[(m_1-m_2)R+(m_1+m_2)E_{\mathbf{k}}$$
$$+m_1E_{12}(\mathbf{k})-m_2E_{21}(\mathbf{k})]\delta(\omega-E_1)$$
$$+[(m_1-m_2)R-(m_1+m_2)E_{\mathbf{k}}$$
$$+m_1E_{12}(\mathbf{k})-m_2E_{21}(\mathbf{k})]\delta(\omega-E_2)\} \quad (46)$$

for the imaginary part of $[f^u(\mathbf{k},\omega)+f^u(-\mathbf{k},-\omega)]$.

Equation (46) may be compared with Eqs. (39) and (42) of P'u Fu-Cho and is seen to be equivalent to his results, except that our excitation energies and sublattice magnetizations include the correction terms which are not contained in P'u Fu-Cho's RPA result. The coefficients of the δ functions give the relative intensities of the two bands.

B. Line Shape

Since the RZOA does not contain any lifetime effects, the spectrum is a set of δ-function-like peaks. An expression for the line shape can be obtained in terms of the polarization operator previously derived; in the following we shall examine the broadening effects arising from spin-spin interactions in the absence of the phonon field, and then in the presence of the phonons as well.

Broadening due to spin-spin interactions. To study the line shape arising from spin-spin interactions, we use Eq. (44) containing the Green's functions in the first approximation and including only the spin-spin part of the polarization operator, $\mathbf{P}(\mathbf{k},\omega)_{\text{s-s}}$.

We use the following definitions:

$$\begin{aligned}
&R_{\mathbf{k}}(\omega)=\tfrac{1}{2}(E_{11}+E_{22})\,\mathrm{Re}[\mathbf{P}_{11}(\mathbf{k},\omega)+\mathbf{P}_{22}(\mathbf{k},\omega)],\\
&Q_{\mathbf{k}}(\omega)=(E_{11}-E_{22})+\mathrm{Re}[\mathbf{P}_{11}(\mathbf{k},\omega)-\mathbf{P}_{22}(\mathbf{k},\omega)],\\
&\hat{\gamma}(\mathbf{k},\omega)=\mathrm{Im}\{\omega[\mathbf{P}_{11}(\mathbf{k},\omega)-\mathbf{P}_{22}(\mathbf{k},\omega)]\\
&\qquad+E_{11}\mathbf{P}_{22}(\mathbf{k},\omega)+E_{22}\mathbf{P}_{11}(\mathbf{k},\omega)\\
&\qquad-E_{12}(\mathbf{k})\mathbf{P}_{21}(\mathbf{k},\omega)-E_{21}(\mathbf{k})\mathbf{P}_{12}(\mathbf{k},\omega)\},\\
&\phi(\mathbf{k},E_{ij})=m_1\left[E_{22}-\left(\frac{S_2}{S_1}\right)^{1/2}E_{12}(\mathbf{k})\right]\\
&\qquad-m_2\left[E_{11}-\left(\frac{S_1}{S_2}\right)^{1/2}E_{21}(\mathbf{k})\right],\\
&\hat{D}(\mathbf{k},\omega)=\{[\omega-Q_{\mathbf{k}}(\omega)]^2-[E_{\mathbf{k}}{}^2+R_{\mathbf{k}}(\omega)]\}^2\\
&\qquad+\hat{\gamma}^2(\mathbf{k},\omega).
\end{aligned}\tag{47}$$

The imaginary part of $\mathbf{P}_{ij}$ involves sums of δ functions while the real part involves principal-value integrals. Expressions for $\mathbf{P}_{ij}$ in the approximation $\mathbf{P}_{ij}{}^0$ appear in Appendix B. If terms involving products of principal-value integrals and similar small quantities be neglected, it is possible to express the imaginary part of $\chi(\omega)$ in the following manner:

$$\mathrm{Im}\chi^{x,x}(\omega)=-2\pi\mu^2\sum_{\mathbf{k}}\gamma_{\mathbf{k}}\{\mathrm{Im}[f^u(\mathbf{k},\omega)+f^u(-\mathbf{k},-\omega)]\}$$

and

$$\begin{aligned}
\mathrm{Im}f^u(\mathbf{k},\omega)=&[\phi(\mathbf{k},E_{ij})+(m_1+m_2)\omega]\hat{\gamma}(\mathbf{k},\omega)/\hat{D}(\mathbf{k},\omega)\\
&+[\omega-E_1(\mathbf{k})][\omega-E_2(\mathbf{k})]\\
&\times[\mathrm{Im}\phi(\mathbf{k},\mathbf{P}_{ij})]/\hat{D}(\mathbf{k},\omega).
\end{aligned}\tag{48}$$

The expression for $\hat{D}(\mathbf{k},\omega)$ given in (47) can be conveniently rewritten as

$$\hat{D}(\mathbf{k},\omega)=\{[\omega-\tilde{\omega}(\omega)][\omega+\tilde{\omega}_2(-\omega)]\}^2+\hat{\gamma}^2(\mathbf{k},\omega),\tag{49a}$$

where we let

$$\begin{aligned}
&\tilde{\omega}_1(\omega)=Q_{\mathbf{k}}(\omega)+E_{\mathbf{k}}(\omega),\\
&\tilde{\omega}_2(\omega)=-Q_{\mathbf{k}}(-\omega)+E_{\mathbf{k}}(-\omega),\\
&E_{\mathbf{k}}{}^2(\omega)=E_{\mathbf{k}}{}^2+R_{\mathbf{k}}(\omega).
\end{aligned}\tag{49b}$$

We note that $\hat{D}(\mathbf{k},\omega)^{-1}$ is strongly peaked around $\tilde{\omega}_1(\omega)$, while $\hat{D}(-\mathbf{k},-\omega)^{-1}$ is strongly peaked around $\tilde{\omega}_2(\omega)$. If those are considered to be slowly varying functions of ω, then $\tilde{\omega}_1$ and $\tilde{\omega}_2$ are the excitation energies inclusive of the shift due to the scattering processes. The expression for the shifts are immediately available from (49b) and (27c). Thus, for example,

$$\begin{aligned}
\tilde{\omega}_1(\omega)=&E(\mathbf{k})+\mathrm{Re}[\mathbf{P}_{11}(\mathbf{k},\omega)-\mathbf{P}_{21}(\mathbf{k},\omega)]\\
&-\left(\frac{E_{11}+E_{22}}{4E_{\mathbf{k}}}\right)\mathrm{Re}[\mathbf{P}_{11}(\mathbf{k},\omega)+\mathbf{P}_{22}(\mathbf{k},\omega)]+\cdots.
\end{aligned}$$

Equation (48) can be rewritten as

$$\mathrm{Im}f^u(\mathbf{k},\omega)=\mathfrak{L}(\mathbf{k},\omega)+\mathfrak{A}(\mathbf{k},\omega),$$

where $\mathfrak{L}(\mathbf{k},\omega)$ is the Lorentzian-like term,

$$\mathfrak{L}(\mathbf{k},\omega)=\phi(\mathbf{k},E_{ij})\gamma(\mathbf{k},\omega)/D(\mathbf{k},\omega)$$

and $\mathfrak{A}(\mathbf{k},\omega)$ is an asymmetric term, viz.,

$$\begin{aligned}
\mathfrak{A}(\mathbf{k},\omega)=&\{(m_1+m_2)\omega\hat{\gamma}(\mathbf{k},\omega)\\
&+[\omega-E_1(\mathbf{k})][\omega+E_2(\mathbf{k})]\,\mathrm{Im}\phi(\mathbf{k},\mathbf{P}_{ij})\}/\hat{D}(\mathbf{k},\omega).
\end{aligned}$$

Then

$$\begin{aligned}
\mathrm{Im}[\chi^{x,x}(\mathbf{k},\omega)]=&-2\pi\mu^2\sum_{\mathbf{k}}\gamma_{\mathbf{k}}\\
&\times[\mathfrak{L}(\mathbf{k},\omega)+\mathfrak{L}(\mathbf{k},-\omega)+\mathfrak{A}(\mathbf{k},\omega)+\mathfrak{A}(\mathbf{k},-\omega)].
\end{aligned}\tag{50}$$

Thus the spectrum may be thought of as consisting of two Lorentzian lines peaking at $\tilde{\omega}_1$ and $\tilde{\omega}_2$, onto which are superimposed two asymmetric lines, peaking at $\tilde{\omega}_1$ and $\tilde{\omega}_2$. In the region between the two peaks, viz., $\tilde{\omega}_1<\omega<\tilde{\omega}_2$, the term $[\omega-E_2(\mathbf{k})][\omega+E_2(\mathbf{k})]$ is negative, and hence the effect of the asymmetric term is to steepen the inside profiles of the two absorption lines. The function $\phi(\mathbf{k},E_{ij})$ is approximately $2|m|\times[E_{11}-E_{21}(\mathbf{k})]$, where m is the average sublattice magnetization; hence, if the offdiagonal contributions are large enough, the relative weight of the Lorentzian lines becomes correspondingly less. Thus it is possible that the negative contribution from the asymmetric terms may just overweigh those of $\mathfrak{L}(\omega)+\mathfrak{L}(-\omega)$ for some ω such that $\tilde{\omega}_1<\omega<\tilde{\omega}_2$, if the matrix elements E_{ij} and $\mathbf{P}_{ij}$ are favorable. Thus there could be an apparent "emissive" region between the two absorption peaks $\tilde{\omega}_1$ and $\tilde{\omega}_2$.

When the two sublattices become identical, with the external magnetic field tending to zero, the two peaks collapse into a single Lorentzian shape centered at the degenerate excitation energy.

Broadening in the presence of the phonon field. When the coupling with the phonons is of importance, we have to take into account all the terms which occur in (45) and also use the full polarization operator, inclusive of the contributions from the phonon field.

We write (45) as

$$f(\mathbf{k},q,\omega)=f(\mathbf{k},\omega)+f^{(1)}(\mathbf{k},\mathbf{l},\omega)+f^{(2)}(\mathbf{k},\mathbf{l},\omega),\tag{51}$$

where $f^{(1)}(\mathbf{k},\omega)$ is the term involving the Green's functions $\mathbf{\Omega}_{ij}(\mathbf{l},q,\omega)$, while $f^{(2)}(\mathbf{k},\omega)$ involves the Green's functions $\mathbf{\Gamma}(\mathbf{l},q,\omega)$, with $\mathbf{l}=\mathbf{k}+\mathbf{q}$.

Suppressing the arguments $\mathbf{l}$, q, and ω for brevity, it is easily established, from Eqs. (45) and (B9), that

$$\begin{aligned}
&\mathbf{\Omega}_{11}=\mathbf{\Gamma}_{11}{}^0(\chi_{11}\gamma_{11}+\chi_{12}\gamma_{12})iN^{-1/2}\mathbf{Y}(q),\\
&\mathbf{\Omega}_{12}=\mathbf{\Gamma}_{11}{}^0(\chi_{11}\gamma_{21}+\chi_{12}\gamma_{22})iN^{-1/2}\mathbf{Y}(q),\quad\text{etc.},
\end{aligned}\tag{52a}$$

where $\boldsymbol{\gamma}=\boldsymbol{\gamma}(\mathbf{k},\omega)=\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\boldsymbol{\gamma}^{\dagger}(\mathbf{k})\rangle\rangle$ and

$$\begin{aligned}\chi_{11}=\chi_{11}(\mathbf{k},\mathbf{k}-\mathbf{q})&=2[q(u_{\mathbf{k}}u_{\mathbf{l}}Z_1+v_{\mathbf{k}}v_{\mathbf{l}}Z_2)\\&\quad+(S_1/S_2)^{1/2}(v_{\mathbf{k}}u_{\mathbf{l}}+u_{\mathbf{k}}v_{\mathbf{l}})J_{1\mathbf{k}}],\\\chi_{12}=\chi_{12}(\mathbf{k},\mathbf{k}-\mathbf{q})&=2[q(v_{\mathbf{k}}u_{\mathbf{l}}Z_1+u_{\mathbf{k}}v_{\mathbf{l}}Z_2)\\&\quad+(S_1/S_2)^{1/2}(u_{\mathbf{k}}u_{\mathbf{l}}+v_{\mathbf{k}}v_{\mathbf{l}})J_{1\mathbf{k}}].\end{aligned}\tag{52b}$$

The expression for $\boldsymbol{\Omega}_{22}$ and $\boldsymbol{\Omega}_{21}$ are obtained from (52a) by interchanging suffix 1 with 2. The elements χ_{22} and χ_{21} are obtained from (52b) by interchanging u amplitudes with v amplitudes. Then, if we define the matrix

$$\mathbf{C}^u=\mathbf{C}^u(\mathbf{k},\mathbf{l})=\begin{bmatrix}t_{11}u_{\mathbf{k}}-t_{21}v_{\mathbf{k}} & 0\\ 0 & t_{12}u_{\mathbf{k}}-t_{22}v_{\mathbf{k}}\end{bmatrix}\begin{bmatrix}\chi_{11} & \chi_{12}\\ \chi_{21} & \chi_{22}\end{bmatrix}\begin{bmatrix}u_{\mathbf{k}} & -v_{\mathbf{k}}\\ v_{\mathbf{k}} & u_{\mathbf{k}}\end{bmatrix}$$

and the matrix $\mathbf{C}^v$ whose elements are obtained from $\mathbf{C}^u$ by interchanging u amplitudes with v amplitudes, it is easily shown that

$$f(\mathbf{k},q,\omega)=f(\mathbf{k},\omega)-\tfrac{1}{2}\sum_q\frac{(\mathbf{k}-\mathbf{q})V^2(q)}{N}(\boldsymbol{\Gamma}_{11}{}^0,\boldsymbol{\Gamma}_{22}{}^0)\times\left[\mathbf{C}^u\begin{pmatrix}\mathbf{G}_{11}\\ \mathbf{G}_{12}\end{pmatrix}+\mathbf{C}^v\begin{pmatrix}\mathbf{G}_{22}\\ \mathbf{G}_{21}\end{pmatrix}-(\mathbf{k}-\mathbf{q})\begin{pmatrix}t_{11}\\ t_{22}\end{pmatrix}\right].\tag{53}$$

This expression differs from that of the system without the phonon field not only in the presence of the term involving $\boldsymbol{\Gamma}^0(\mathbf{k},\mathbf{l},\omega)$, but also in that the full polarization operator inclusive of the phonon part has to be used. This does not change the qualitative form of the contribution to the line shape arising from $f(\mathbf{k},\omega)$, the first term on the right-hand side of (53). If products of principal-value integrals and δ-function-like terms be neglected, the contribution from the second and third terms on the right-hand side of (51) can be expressed as

$$\begin{aligned}\mathrm{Im}(f^{(1)}+f^{(2)})=-\tfrac{1}{2}\sum\frac{(\mathbf{k}-\mathbf{q})V^2(q)}{N}&\times\{[\theta(u,\omega)-(\mathbf{k}-\mathbf{q})t_{11}]\cdot\mathrm{Im}\boldsymbol{\Gamma}_{11}{}^0(\mathbf{l},q,\omega)\\&+[\theta(v,\omega)-(k-q)t_{22}]\cdot\mathrm{Im}\boldsymbol{\Gamma}_{22}{}^0(\mathbf{l},q,\omega)],\end{aligned}$$

where

$$\begin{aligned}\theta(u,\omega)=[&(\omega-E_1)(\omega+E_2)(\mathbf{C}_{11}{}^u-\mathbf{C}_{22}{}^u)\omega\\&+\mathbf{C}_{11}{}^uE_{22}+\mathbf{C}_{22}{}^uE_{11}-\mathbf{C}_{12}{}^uE_{21}-\mathbf{C}_{21}{}^uE_{12}]/\hat{D}(\omega).\end{aligned}$$

Also,

$$\begin{aligned}\mathrm{Im}\boldsymbol{\Gamma}_{11}{}^0(\mathbf{l},q,\omega)=\tfrac{1}{2}[&(1+2n_1{}^{(1)}+N_q)\delta(\omega-E_1(\mathbf{l})-\omega_q)\\&-(1+2n_1{}^{(1)}-N_q)\delta(\omega-E_1(\mathbf{l})+\omega_q)]\end{aligned}\tag{54a}$$

and

$$\begin{aligned}\mathrm{Im}\boldsymbol{\Gamma}_{22}{}^0(\mathbf{l},q,\omega)=\tfrac{1}{2}[&(1-2n_1{}^{(2)}-N_q)\delta(\omega+E_2(\mathbf{l})-\omega_q)\\&-(1+2n_1{}^{(2)}+N_q)\delta(\omega+E_2(\mathbf{l})+\omega_q)].\end{aligned}\tag{54b}$$

The full expression for the susceptibility will involve the complementary terms with ω replaced by $-\omega$, arising from $f(-\mathbf{k},-\mathbf{l},-\omega)$. However, it is evident from (54a) that the terms involving t_{11} and t_{22} will give rise to a series of strong δ-function-like peaks defined by (54b). The broadening in these very sharp lines will be manifest only in a higher-order theory. The terms involving the denominator $\hat{D}(\omega)$ will lead to an asymmetric contribution to the line shape, but, unlike $\mathfrak{A}(\mathbf{k},\omega)$ which occurs in (50), the contribution will be positive in the range $\tilde{\omega}_1<\omega<\tilde{\omega}_2$ and will tend to broaden out the line profiles which face each other, owing to the formation of the bands between the two main peaks. In the regions $\omega<\tilde{\omega}_1$, and $\omega>\tilde{\omega}_2$, the phonon side bands occur with much weaker intensity than those between the two peaks, as may be ascertained by a study of the sign of the contribution from the first term on the right-hand side of (54).

7. CONCLUSION

The development of the Green's function theory of the Heisenberg antiferromagnet via a Dyson equation has enabled us to achieve a systematic approach to the study of interacting magnon fields. In spite of the algebraic complexity of many of the results, useful qualitative conclusions regarding the line shape could be drawn and contact with established results could be easily made. A more detailed study of many of the results developed here needs the support of numerical computation and these will be presented in a subsequent study.

APPENDIX A

The non-Hermitian nature of the effective boson Hamiltonian of Dyson leads to a Dyson equation having a complicated structure if the zeroth-order Hamiltonian is taken to include other terms than those occurring in $H_0{}^{(0)}$.

For example, instead of Eq. (17), we could have

$$\begin{aligned}\mathbf{G}^0(\mathbf{k},\omega)^{-1}\mathbf{G}(\mathbf{k},\omega)&=\bar{I}+\sum_{\mathbf{k}_1,\mathbf{k}_2}{}'\begin{bmatrix}F(\mathbf{k}_1-\mathbf{k}_2) & 0\\ 0 & F(\mathbf{k}_1-\mathbf{k}_2)\end{bmatrix}\\&\quad\times\left\langle\left\langle\begin{bmatrix}\alpha_{\mathbf{l}}\beta_{-\mathbf{k}_2}{}^{\dagger}\beta_{-\mathbf{k}_1}\\ \beta_{-\mathbf{l}}{}^{\dagger}\alpha_{\mathbf{k}_1}\alpha_{\mathbf{k}_1}{}^{\dagger}\end{bmatrix},\boldsymbol{\alpha}^{\dagger}\right\rangle\right\rangle\\&\quad+\sum_{\mathbf{k}_1,\mathbf{k}_2}{}'\begin{bmatrix}0 & F_1(\mathbf{k})\\ F_1(-\mathbf{k}) & 0\end{bmatrix}\\&\quad\times\left\langle\left\langle\begin{bmatrix}\alpha_{\mathbf{k}_1}{}^{\dagger}\alpha_{\mathbf{k}_2}\alpha_{\mathbf{l}}\\ \beta_{-\mathbf{k}}{}^{\dagger}\beta_{-\mathbf{k}}{}^{\dagger}\beta_{-\mathbf{l}}{}^{\dagger}\end{bmatrix},\boldsymbol{\alpha}^{\dagger}\right\rangle\right\rangle\\&\quad+\sum_{\mathbf{k}_1,\mathbf{k}_2}{}'\begin{bmatrix}F_2(-\mathbf{k}_1) & 0\\ 0 & F_1(\mathbf{k}_1)\end{bmatrix}\\&\quad\times\left\langle\left\langle\begin{bmatrix}\alpha_{\mathbf{l}}\alpha_{\mathbf{k}_2}\beta_{-\mathbf{k}_1}\\ \beta_{-\mathbf{l}}{}^{\dagger}\beta_{-\mathbf{k}_2}{}^{\dagger}\alpha_{\mathbf{k}_1}{}^{\dagger}\end{bmatrix},\boldsymbol{\alpha}^{\dagger}\right\rangle\right\rangle,\end{aligned}\tag{A1}$$

where the prime on the summations indicates that $\mathbf{k}_1 \neq \mathbf{k}_2$ and $\mathbf{k}_2 \neq \mathbf{k}$, and $\mathbf{l} = \mathbf{k} + \mathbf{k}_1 - \mathbf{k}_2$. Further, we have

$$(\mathbf{G}^0)^{-1} = \begin{bmatrix} \omega - E_{11}(0) & -E_{12}(\mathbf{k}) \\ -E_{21}(\mathbf{k}) & \omega - E_{22}(0) \end{bmatrix}, \tag{A2}$$

which is the same as $\mathbf{G}^0$ of Eq. (24) and, unlike $(\mathbf{G}_0{}^0)^{-1}$ of Eq. (18b), unsymmetrical in that $E_{12} \neq E_{21}$ unless the two sublattices are equivalent and the external field is zero.

Differentiating (A2) with respect to the time argument of $\alpha^\dagger(t')$ occurring on the right, we have

$$(\mathbf{G}^0)^{-1}\mathbf{G} = \mathbf{I} + \mathbf{P}(\mathbf{G}^0)_T, \tag{A3}$$

where the subscript T denotes the transpose of $(\mathbf{G}^0)^{-1}$.

Thus, in casting (A3) into the form of a Dyson equation, we write

$$\begin{aligned} \mathbf{G} &= \mathbf{G}^0 + \mathbf{G}^0\mathbf{P}(\mathbf{G}^0)_T \\ &= \mathbf{G}^0 + \mathbf{G}^0\mathbf{\Pi}\mathbf{G}, \end{aligned}$$

when

$$\mathbf{\Pi} = \mathbf{P}(\mathbf{G}^0)_T\mathbf{G}^{0-1}/[\mathbf{I} + \mathbf{P}(\mathbf{G}^0)_T]. \tag{A4}$$

Thus the polarization operator will have the simple form $\mathbf{P}/(I + \mathbf{P}\mathbf{G}^0)$ only if $(\mathbf{G}^0)_T(\mathbf{G}^0)^{-1} = \mathbf{I}$. This is not the case, since (A2) is nonsymmetric except when $S_1 = S_2$ and the external field is zero.

APPENDIX B

The detailed derivation of the expressions for the polarization operator are presented in this appendix.

The spin-spin contribution to $\bar{P}^0(k,\omega)$. Applying the canonical transformation of Eq. (33) to $\bar{P}^0(\mathbf{k},\omega)_{s\text{-}s}$, we obtain

$$\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}s} = \langle\langle \chi(\mathbf{k}), \chi(\mathbf{k}) \rangle\rangle, \tag{B1a}$$

where

$$\chi(\mathbf{k}) = \sum_{\mathbf{k}_1\mathbf{k}_2} \begin{bmatrix} \sum_{r=1}^{3} (D_{ru}\sigma_r + D_{rv}\sigma_r{}^{L\dagger}) + d_1\gamma_1(\mathbf{k}) + d_2\gamma_2{}^\dagger(\mathbf{k}) \\ \sum_{r=1}^{3} (D_{ru}{}^L\sigma_r{}^{L\dagger} + D_{rv}{}^L\sigma_r) + d_2{}^L\gamma_1(\mathbf{k}) + d_1{}^L\gamma_2{}^\dagger(-\mathbf{k}) \end{bmatrix}, \tag{B1b}$$

$$\chi'(\mathbf{k}) = \sum_{\mathbf{k}_1'\mathbf{k}_2'} \begin{bmatrix} \sum_{r=1}^{3} (D_{ru}'\sigma_r{}^\dagger + D_{rv}'\sigma_r) + d_1'\gamma_1{}^\dagger(\mathbf{k}) + d_2'\gamma_2(-\mathbf{k}) \\ \sum_{r=1}^{3} (D_{ru}{}'^L\sigma_r{}^\dagger + D_{rv}{}'^L\sigma_r{}^\dagger) + d_2{}'^L\gamma_1{}^\dagger(\mathbf{k}) + d_1{}'^L\gamma_2(-\mathbf{k}) \end{bmatrix}, \tag{B1c}$$

where the notation of Eq. (39) has been used.

$\chi'(\mathbf{k})$ is a row vector of two components although it is written as a column vector for convenience. As before, the superscript L denotes the lattice-dual forms. D_{rv} is obtained from D_{ru} by interchanging all the u amplitudes with the corresponding v amplitudes. d_2 and d_2' are obtained from d_1 and d_1' by interchanging $u_\mathbf{k}$ with $v_\mathbf{k}$ for the $\mathbf{k}$ mode only. The expressions for all these coefficients will not be given here since they are easily derived. We display only the following examples:

$$\begin{aligned} D_{1u} = D_{1u}(\mathbf{k}_1,\mathbf{k}_2,\mathbf{l}) &= -(1/N)\{J_{\mathbf{l}-\mathbf{k}}'u_{\mathbf{l}}u_{\mathbf{k}_2} + v_{\mathbf{k}_2}v_{\mathbf{l}}J_{\mathbf{l}-\mathbf{k}_1}' + [(S_1/S_2)^{1/2}J_{\mathbf{k}}u_{\mathbf{k}_2}v_{\mathbf{l}} + v_{\mathbf{k}_2}u_{\mathbf{l}}J_{\mathbf{k}_1}(S_2/S_1)^{1/2}]\}u_{\mathbf{k}_1}, \\ d_1 = d_1(\mathbf{k},\mathbf{k}_1)\delta_{\mathbf{k}_1\mathbf{k}_2} &= -(1/N)\{J_0'u_{\mathbf{k}}v_{\mathbf{k}_1} + J_{\mathbf{k}-\mathbf{k}_1}'v_{\mathbf{k}}u_{\mathbf{k}_1}) + [(S_1/S_2)^{1/2}J_{\mathbf{k}}v_{\mathbf{k}}v_{\mathbf{k}_1} + u_{\mathbf{k}}u_{\mathbf{k}_1}J_{\mathbf{k}_1}(S_2/S_1)^{1/2}]\}v_{\mathbf{k}_1}, \end{aligned} \tag{B2}$$

where, as before, $J_\mathbf{k}' = J_\mathbf{k} + A_\mathbf{k}$.

In (B1a) we had

$$\bar{P}^0(\mathbf{k},\omega)_{s\text{-}s} = [\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}s} : \mathbf{P}^{0L}(\mathbf{k},\omega)_{s\text{-}s}]$$

and

$$\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}s} = \langle\langle \psi(\mathbf{k}), \psi'(\mathbf{k}) \rangle\rangle. \tag{B3}$$

From (B1b) and (B1c), it is evident that an evaluation of $\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}s}$ involves the evaluation of Green's functions like $\langle\langle \sigma_r, \sigma_r{}^\dagger \rangle\rangle^0$ $(r = 1,2,3)$ and the conjugates of their lattice-dual forms. All other Green's functions, e.g., $\langle\langle \sigma_r{}^{L\dagger}, \sigma_s{}^\dagger \rangle\rangle^0$, etc., which occur in (B3) are zero. We shall use $\mathbf{k}_i$ to denote $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{l}$. Then

$$\langle\langle \sigma_1(\mathbf{k}_i), \sigma_1{}^\dagger(\mathbf{k}') \rangle\rangle^0 = \delta_{\mathbf{k}_i\mathbf{k}_i'} \frac{(1 + n_1{}^{(1)} + n_{\mathbf{k}_1}{}^{(2)})n_{\mathbf{k}_2}{}^{(2)} - n_1{}^{(2)}n_{\mathbf{k}_1}{}^{(2)}}{\omega - E_1(\mathbf{l}) - E_2(-\mathbf{k}_1) + E_2(-\mathbf{k}_2)}, \tag{B4a}$$

$$\langle\langle \sigma_2(\mathbf{k}_i), \sigma_2{}^\dagger(\mathbf{k}_i') \rangle\rangle^0 = \delta_{\mathbf{k}_i\mathbf{k}_i'} \frac{2[(1 + n_1{}^{(2)} + n_{\mathbf{k}_2}{}^{(2)})n_{\mathbf{k}_1}{}^{(2)} - n_{\mathbf{k}_2}{}^{(2)}n_1{}^{(2)}]}{-\omega + E_2(-\mathbf{k}_1) - E_2(-\mathbf{l}) - E_2(-\mathbf{k}_2)}, \tag{B4b}$$

$$\langle\langle\sigma_3(\mathbf{k}_i),\sigma^\dagger(\mathbf{k}_i')\rangle\rangle^0=\delta_{\mathbf{k}_i\mathbf{k}_i'}\frac{2[(1+n_1^{(1)})(1+n_{\mathbf{k}_2}^{(1)}+n_{\mathbf{k}_1}^{(2)})+n_{\mathbf{k}_1}^{(2)}n_{\mathbf{k}_2}^{(1)}]}{\omega-E_2(-\mathbf{k}_1)-E_2(\mathbf{k}_2)-E_1(\mathbf{l})}, \tag{B4c}$$

where $n_{\mathbf{k}}^{(1)}=\langle\gamma_1^\dagger(\mathbf{k})\gamma_1(\mathbf{k})\rangle$ and similarly for $n_{\mathbf{k}}^{(2)}$.

The conjugate lattice-dual forms, viz., $\langle\langle\sigma_r^{L\dagger},\sigma_r^L\rangle\rangle$ ($r=1,2,3$) are obtained from the above by interchanging 1 with 2, $\mathbf{k}_i$ with $-\mathbf{k}_i$, ω with $-\omega$, and changing the sign of the whole expression. Thus, noting that $n_{\mathbf{k}}^{(1)}=n_{-\mathbf{k}}^{(1)}$, we have, by way of an illustration,

$$\langle\langle\sigma_3^{L\dagger}(\mathbf{k}_i),\sigma_3^L(\mathbf{k}')\rangle\rangle^0=-\delta_{\mathbf{k}_i\mathbf{k}_i'}\frac{2[(1+n_1^{(2)})(1+n_{\mathbf{k}_2}^{(2)}+n_{\mathbf{k}_1}^{(1)})+n_{\mathbf{k}_1}^{(1)}n_{\mathbf{k}_2}^{(2)}]}{-\omega-E_1(\mathbf{k}_1)-E_2(-\mathbf{k}_2)-E_1(-\mathbf{l})}.$$

Using these Green's functions, matrix elements of the polarization operator can be easily written down. Thus, for example,

$$\begin{aligned}\mathbf{P}_{11}^0(\mathbf{k},\omega)_{\text{s-s}}&=\sum D_{ru}D_{ru}'\langle\langle\sigma_r,\sigma_r^\dagger\rangle\rangle^0+D_{rr}D_{rr}'\langle\langle\sigma_r^{L\dagger},\sigma_r^L\rangle\rangle^0\\&\quad+f(\mathbf{k})/[\omega-E(\mathbf{k})]-f'(\mathbf{k})/[-\omega-E(-\mathbf{k})],\end{aligned} \tag{B5a}$$

with

$$\begin{aligned}f(\mathbf{k})=\sum_{\mathbf{k}_1\mathbf{k}_2}\sum_{\mathbf{k}_1'\mathbf{k}_2'}&(D_{1u}d_1'n_{\mathbf{k}_1}^{(2)}+n_{\mathbf{k}_1'}^{(2)}d_1D_{1u}'\\&+D_{2v}d_1'n_{\mathbf{k}_1}^{(1)}+n_{\mathbf{k}_1'}^{(1)}d_1D_{2v}'+d_1d_1')\delta_{\mathbf{k}_1\mathbf{k}_2}\delta_{\mathbf{k}_1'\mathbf{k}_2'}\\&+(D_{2v}d_1'n_{\mathbf{k}_1}^{(1)}+n_{\mathbf{k}'}^{(1)}d_1D_{2v}')\delta_{\mathbf{k}_1\mathbf{k}_2}\delta_{\mathbf{k}_1'1'}\end{aligned} \tag{B5b}$$

and with a similar expression for $f'(\mathbf{k})$, obtained by interchanging 1 with 2 and u with v. The $f(\mathbf{k})\{[\omega-E_1(\mathbf{k})]\}^{-1}$ and $f'(\mathbf{k})\{[-\omega-E_2(-\mathbf{k})]\}^{-1}$ terms are of the order N^{-2} and are in the nature of a first-order perturbation correction to the excitation energies given by the RZOA. The true dynamical part of the polarization operator is associated with the Green's functions $\langle\langle\sigma_r,\sigma_r^\dagger\rangle\rangle$ and $\langle\langle\sigma_r^{L\dagger},\sigma_r^L\rangle\rangle$. They describe three different physical processes and the corresponding conjugate processes. The various probability factors are largest in the neighborhood of small wave vectors and, on this basis, a very crude assessment of the magnitudes of the D coefficients can be given as

$$\begin{aligned}D_{1u}D_{1u}'\approx 4D_{2u}D_{2u}'\approx 4D_{3u}D_{3u}'\\\approx d_1\approx d_1'\approx[2N^{-1}(J_0+J_{\mathbf{k}})]^2,\end{aligned}$$

thus indicating that the three processes are of a similar order of importance.

The spin-phonon contribution to the polarization operator. In Eq. (27) we had

$$\mathbf{P}^0(\mathbf{k},\omega)_{\text{s-p}}=\langle\langle\boldsymbol{\phi}(\mathbf{k})_q,\boldsymbol{\phi}^\dagger(\mathbf{k})_{q'}\rangle\rangle.$$

On applying the canonical transformation to $\boldsymbol{\phi}(\mathbf{k})_q$, we obtain

$$\boldsymbol{\phi}(\mathbf{k})_q=\sum_q\mathbf{W}(\mathbf{k},\mathbf{k}-\mathbf{q})\mathbf{C}(\mathbf{k}-\mathbf{q})\boldsymbol{\gamma}_{\mathbf{k}-\mathbf{q}}A_q. \tag{B6}$$

We define the following operators:

$$B_q=b_q-b_q^\dagger,$$

$$\mathbf{A}(\mathbf{k}-\mathbf{q},q)=\boldsymbol{\gamma}(\mathbf{k}-\mathbf{q})A_q=\begin{bmatrix}\gamma_1(\mathbf{k}-\mathbf{q})A_q\\\gamma_2^\dagger(\mathbf{q}-\mathbf{k})A_{-q}^\dagger\end{bmatrix} \tag{B7a}$$

and

$$\mathbf{B}(\mathbf{k}-\mathbf{q},q)=\boldsymbol{\gamma}(\mathbf{k}-\mathbf{q})\,\mathbf{X}\mathbf{B}_q=\begin{bmatrix}\gamma_1(\mathbf{k}-\mathbf{q})B_q\\\gamma_2^\dagger(\mathbf{q}-\mathbf{k})B_{-q}^\dagger\end{bmatrix}, \tag{B7b}$$

where $\mathbf{X}$ is the anti-unit matrix defined in (18). Further, we let

$$\boldsymbol{\Gamma}(\mathbf{k}-\mathbf{q},q,\omega)=\langle\langle\mathbf{A}(\mathbf{k}-\mathbf{q},q),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle. \tag{B7c}$$

In order to evaluate $\mathbf{P}^0(\mathbf{k}-\mathbf{q},q)_{\text{s-p}}$, i.e., $\langle\langle\boldsymbol{\phi}(\mathbf{k})_q,\boldsymbol{\phi}^\dagger(\mathbf{k})_{q'}\rangle\rangle^0$, it is necessary to evaluate $\boldsymbol{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$ using the Hamiltonian H_{eff}. However, for use in a subsequent section, we directly evaluate $\boldsymbol{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)$, to achieve an accuracy equivalent to the evaluation of $\mathbf{G}(\mathbf{k},\omega)$, and recover $\boldsymbol{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$ *en passant*. Thus the Hamiltonian to be used is

$$H^{(1)}=H_{\text{eff}}+H_{\text{s-p}}. \tag{B8}$$

When the canonical transformation, Eq. (33), is applied to $H_{\text{s-p}}$, Eq. (11), it can be written in terms of γ operators as follows:

$$\begin{aligned}H_{\text{s-p}}=\sum_{\mathbf{k},q}&[\mathbf{W}_{11}(\mathbf{k},\mathbf{k}+\mathbf{q})\gamma_1^\dagger(\mathbf{k}+\mathbf{q})\gamma_1(\mathbf{k})A_q\\&+\mathbf{W}_{22}(\mathbf{k},\mathbf{k}+\mathbf{q})\gamma_2^\dagger(-\mathbf{k}-\mathbf{q})\gamma_2(-\mathbf{k})A_{-q}]\\&+\sum_{\mathbf{k},q}[\mathbf{W}_{12}(\mathbf{k},\mathbf{k}+\mathbf{q})\gamma_1^\dagger(\mathbf{k}+\mathbf{q})\gamma_2^\dagger(-\mathbf{k})A_q\\&+\mathbf{W}_{12}^*(\mathbf{k},\mathbf{k}+\mathbf{q})\gamma_1(\mathbf{k}+\mathbf{q})\gamma_2(-\mathbf{k})A_{-q}],\end{aligned} \tag{B9}$$

with $\mathbf{l}=\mathbf{k}+\mathbf{q}$. The coupling factors $\mathbf{W}_{ij}(\mathbf{k},\mathbf{k}+\mathbf{q})$, etc., now contain the various u,v amplitudes, W_α, W_β, $W_{\alpha\beta}$, and $W_{\alpha\beta}^*$.

Taking the equations of motion of $\boldsymbol{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,t-t')$, we have for the Fourier component $\boldsymbol{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)$ the following result:

$$\begin{aligned}\mathbf{G}_D^0(\mathbf{k}-\mathbf{q},\omega)^{-1}&\boldsymbol{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)\\&=N_q\mathbf{I}+\omega_q\langle\langle\mathbf{B}(\mathbf{k}-\mathbf{q},q),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)}\\&\quad+\sum_q\langle\langle\mathfrak{W}(\mathbf{k}-\mathbf{q}-\mathbf{q}',\mathbf{k}-\mathbf{q})\boldsymbol{\gamma}(\mathbf{k}-\mathbf{q}-\mathbf{q}')A_{q'}A_q,\\&\qquad\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)},\end{aligned} \tag{B10}$$

where

$$\mathbf{G}_D{}^0(\mathbf{k}-\mathbf{q},\omega)^{-1}=\begin{bmatrix}\omega-E_1(\mathbf{k}-\mathbf{q}) & 0\\ 0 & -\omega-E_2(\mathbf{k}-\mathbf{q})\end{bmatrix}$$

and

$$N_q=\langle A_qA_q{}^\dagger\rangle.$$

For the Green's function $\langle\langle\mathbf{B}(\mathbf{k}-\mathbf{q},q),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)}$ which occurs on the right-hand side of Eq. (B10), we have the equation

$$[\mathbf{G}_D{}^0(\mathbf{k}-\mathbf{q},\omega)]^{-1}\langle\langle\mathbf{B}(\mathbf{k}-\mathbf{q},q),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)}$$
$$=\begin{bmatrix}1+2n_{\mathbf{k}-\mathbf{q}}{}^{(1)} & 0\\ 0 & 1+2n_{\mathbf{k}-\mathbf{q}}{}^{(2)}\end{bmatrix}+\omega_q\mathbf{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)$$
$$+\sum_{q'}\langle\langle\mathbf{X}\mathbf{W}(\mathbf{k}-\mathbf{q}-\mathbf{q}',\mathbf{k}-\mathbf{q})\boldsymbol{\gamma}(\mathbf{k}-\mathbf{q}-\mathbf{q}')A_{q'}B_{q'}$$
$$+\mathbf{T}(\mathbf{k},\mathbf{k}-\mathbf{q})_{\mathbf{k}'},\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)},\quad\text{(B11)}$$

where $\mathbf{X}$ is the anti-unit matrix and $\mathbf{T}(\mathbf{k},\mathbf{k}-\mathbf{q})_{\mathbf{k}'}$ contains higher-order Green's functions like

$$\langle\langle\gamma_1(\mathbf{k}')\gamma_1(\mathbf{k}-\mathbf{q})\gamma_1{}^\dagger(\mathbf{k}'-\mathbf{q})\gamma(\mathbf{k}-\mathbf{q}-\mathbf{q}')A_{q'}B_{q'},$$
$$\gamma_1{}^\dagger(\mathbf{k}-\mathbf{q})A_q{}^\dagger\rangle\rangle.$$

These higher-order Green's functions can be decoupled in an obvious manner by using the averages

$$\langle A_qA_{q'}\rangle=N_q\delta_{q,-q'}\quad\text{in Eq. (B10)},$$
$$\langle A_{q'}B_q\rangle=\delta_{q,-q'}$$

and

$$\langle\gamma_1{}^\dagger(\mathbf{k}-\mathbf{q})\gamma_1(\mathbf{k}'-\mathbf{q})\rangle=n_{\mathbf{k}-\mathbf{q}}{}^{(1)}\delta_{\mathbf{k},\mathbf{k}'}\quad\text{in Eq. (B11)}.$$

Then, substituting Eq. (B11) in (B10), we obtain

$$\mathbf{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)=\mathbf{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)+\mathbf{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$$
$$\times\mathbf{W}(\mathbf{k},\mathbf{k}-\mathbf{q})\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle^{(1)},\quad\text{(B12a)}$$

where the zeroth result $\mathbf{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)$, Eq. (B12a), is a diagonal matrix, having the elements

$$\mathbf{\Gamma}_{11}{}^0(\mathbf{k}-\mathbf{q},q,\omega)=\frac{1}{2}\left[\frac{1+2n_{\mathbf{k}-\mathbf{q}}{}^{(1)}+N_q}{\omega-E(\mathbf{k}-\mathbf{q})-\omega_q}+\frac{N_q-1-2n_{\mathbf{k}-\mathbf{q}}{}^{(1)}}{\omega-E(\mathbf{k}-\mathbf{q})+\omega_q}\right],\quad\text{(B12b)}$$
$$\mathbf{\Gamma}_{12}{}^0(\mathbf{k}-\mathbf{q},q,\omega)=0.$$

The two elements $\mathbf{\Gamma}_{22}{}^0(\mathbf{k}-\mathbf{q},q,\omega)$ and $\mathbf{\Gamma}_{21}{}^0(\mathbf{k}-\mathbf{q},q,\omega)$ are obtained from the above by interchanging $n^{(1)}$ with $n^{(2)}$, ω with $-\omega$, and E_1 with E_2. Finally, using (B6), we have

$$\mathbf{P}_{11}{}^0(\mathbf{k},\omega)_{s\text{-}p}=\sum_q|\mathbf{W}_{11}(\mathbf{k}-\mathbf{q},q)u_{\mathbf{k}-\mathbf{q}}+v_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{12}(\mathbf{k}-\mathbf{q},q)|^2\times\mathbf{\Gamma}_{11}{}^0(\mathbf{k}-\mathbf{q},q,\omega)$$
$$+\sum_q|\mathbf{W}_{11}(\mathbf{k}-\mathbf{q},q)v_{\mathbf{k}-\mathbf{q}}+u_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{12}(\mathbf{k}-\mathbf{q},q)|^2\times\mathbf{\Gamma}_{22}{}^0(\mathbf{k}-\mathbf{q},q,\omega),\quad\text{(B13a)}$$

and, suppressing the $\mathbf{k}-\mathbf{q},q$ arguments in $\mathbf{W}(\mathbf{k}-\mathbf{q},q)$, we have

$$\mathbf{P}_{12}{}^0(\mathbf{k},\omega)_{s\text{-}p}=\sum_q|(\mathbf{W}_{11}u_{\mathbf{k}-\mathbf{q}}+v_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{12})(\mathbf{W}_{21}{}^*u_{\mathbf{k}-\mathbf{q}}+v_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{22}{}^*)|\times\mathbf{\Gamma}_{11}{}^0(\mathbf{k}-\mathbf{q},q,\omega)$$
$$+\sum_q|(\mathbf{W}_{11}v_{\mathbf{k}-\mathbf{q}}+u_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{12})(\mathbf{W}_{21}{}^*v_{\mathbf{k}-\mathbf{q}}+u_{\mathbf{k}-\mathbf{q}}\mathbf{W}_{22}{}^*)|\times\mathbf{\Gamma}_{22}{}^0(\mathbf{k}-\mathbf{q},q,\omega).\quad\text{(B13b)}$$

The other two elements are obtained by interchanging 1 with 2. These expressions may be compared with those obtained by Kashcheev [Eq. (A6)] where he has three energy denominators only. Our results contain four energy denominators (two from each $\mathbf{\Gamma}^0$ element) and, in addition, the numerators carry the magnon and phonon occupation numbers in a different manner, although they still occur linearly.

Equations (B4), (B5), and (B13), giving $\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}s}$ and $\mathbf{P}^0(\mathbf{k},\omega)_{s\text{-}p}$ complete the evaluation of the polarization operator $\mathbf{P}^0(\mathbf{k},\omega)$.

Spin-phonon Green's functions in the first approximation. In the remainder of this appendix we proceed to evaluate the Green's functions $\langle\langle\mathbf{A}(\mathbf{k}-\mathbf{q},q),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle$, $\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle$ in the first approximation.

In Eq. (B12a), the unknown Green's function $\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle$ is found to occur. Taking the equations of motion of the latter with respect to the right-hand time argument, we have, for its Fourier component,

$$\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\mathbf{A}^\dagger(\mathbf{k}-\mathbf{q},q)\rangle\rangle=\langle\langle\boldsymbol{\gamma}(\mathbf{k})\boldsymbol{\gamma}^\dagger(\mathbf{k})\rangle\rangle\mathbf{W}^\dagger(\mathbf{k},\mathbf{k}-\mathbf{q})\mathbf{\Gamma}^{(0)}(\mathbf{k}-\mathbf{q},q,\omega),\quad\text{(B14)}$$

where $\mathbf{W}^\dagger$ is the complex conjugate of the transpose of $\mathbf{W}$. Hence, from (B12a),

$$\mathbf{\Gamma}^{(1)}(\mathbf{k}-\mathbf{q},q,\omega)=\mathbf{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega)+\mathbf{\Gamma}(\mathbf{k}-\mathbf{q},q)\mathbf{W}(\mathbf{k},\mathbf{k}-\mathbf{q})\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\boldsymbol{\gamma}^\dagger(\mathbf{k})\rangle\rangle^{(1)}\times\mathbf{W}^\dagger(\mathbf{k},\mathbf{k}-\mathbf{q})\mathbf{\Gamma}^0(\mathbf{k}-\mathbf{q},q,\omega).\quad\text{(B15)}$$

The Green's function $\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\boldsymbol{\gamma}^\dagger(\mathbf{k})\rangle\rangle^{(1)}$ is already known in terms of $\mathbf{G}^{(1)}(\mathbf{k},\omega)$ which has already been determined. Thus we have

$$\langle\langle\boldsymbol{\gamma}(\mathbf{k}),\boldsymbol{\gamma}^\dagger(\mathbf{k})\rangle\rangle^{(1)}=\mathbf{C}^{-1}(\mathbf{k})\mathbf{G}^{(1)}(\mathbf{k},\omega)\mathbf{C}^{\dagger-1}(\mathbf{k},\omega),$$

with

$$\mathbf{C}^{-1}=\begin{bmatrix}u_{\mathbf{k}}, & -v_{\mathbf{k}}\\ -v_{\mathbf{k}} & u_{\mathbf{k}}\end{bmatrix}.$$