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Low-frequency modes in rare-earth magnets with degenerate crystal-field levels

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We develop a general theory of the low-frequency spin dynamics in rare-earth magnets with degenerate crystal-field levels. The analysis applies to the paramagnetic phase of those systems which have a well-defined separation between high- and low-frequency response. We calculate the low-frequency part of the zone-center dynamic susceptibility. Treating the static correlations in the molecular-field approximation we use the theory to interpret electron-paramagnetic-resonance studies of TbP. Critical anomalies in the electron paramagnetic resonance in singlet-ground-state ferromagnets are pointed out. The temperature dependence of the intensity and width of the central peak in Pr_3Tl is calculated assuming the broadening comes from the first-order interaction with the conduction electrons. Contact is made with other treatments of the low-frequency dynamics.

I. INTRODUCTION

The dynamical behavior of rare-earth magnets is a challenging problem. The existence of excited crystal-field levels at energies which can be comparable to the exchange interactions between the ions makes the problem much more complicated than the analysis of Heisenberg magnets such as $RbMnF_3$ where only the lowest crystal-field manifold is important. Because of the calculational difficulties associated with crystal-field effects it is important to identity features which can be analyzed in some detail without having to obtain a complete solution to the problem. In the characterization of dynamical effects in strongly interacting systems a separation into high- and low-frequency modes is sometimes possible. The latter are often associated with the long-wavelength fluctuations of conserved variables or critical variables if the system is near a second-order phase transition. If there is a separation into high- and low-frequency modes a "hydrodynamic" theory can be developed for the low-frequency response. In such a theory the characteristic frequencies are expressed in terms of the static correlation functions of the low-frequency variables while the linewidths are written as integrals of the corresponding dynamic correlation functions.[1]

In the case of rare-earth systems, a separation into high- and low-frequency modes is often possible if (i) the crystal-field symmetry is sufficiently high so that there are degenerate crystal-field levels or (ii) the ions in question have odd numbers of electrons and hence display Kramers degeneracy. The low-frequency modes are then associated with transitions between levels belonging to the same crystal-field manifold, while the high-frequency modes are intermanifold transitions. The separation into high- and low-frequency modes is precise only in the limit where the interaction between the ions is zero. As the interactions increase in strength both the low- and high-frequency modes broaden. For sufficiently strong interactions the two sets of resonances overlap and the distinction between high- and low-frequency response disappears. It should be noted that the extent of the broadening and overlap will in general depend on the wavelength of the modes in question. It can happen (e.g., TbSb) that the low-frequency modes at the center of the Brillouin zone are weakly damped while the corresponding modes at the zone boundary overlap the intermanifold excitations.

In this paper we develop a hydrodynamic theory for the low-frequency modes in rare-earth magnets with degenerate crystal-field levels. We calculate the frequency and linewidth of the modes and their contribution to the dynamic susceptibility. Since we do not deal with the intermanifold transitions we cannot put precise bounds on the range of applicability of the theory. These can only be defined operationally in the sense that the theory is appropriate whenever there is a well-defined separation in the dynamic susceptibility between the high- and low-frequency response. Our calculations are restricted to the paramagnetic phase, with the major focus on cubic systems having singlet ground states which undergo ferromagnetic or antiferromagnetic ordering. The detailed analysis is limited to the center of the zone ($\vec{q}=0$). However the theory can be applied to other points in the zone. We make contact with experiment in two types of studies: electron paramagnetic resonance (EPR) and inelastic neutron scattering. In the former case we interpret the $\Gamma_5^{(2)}$ EPR in the singlet ground-state antiferromagnet TbP.[2] We also calculate the linewidth and intensity of the central peak associated with the low-frequency

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modes in Pr_3Tl, which is observed in inelastic neutron scattering.[3] In addition to reproducing the qualitative, and to some extent, quantitative features of the experimental studies the theory also predicts anomalous behavior in the EPR in singlet ground-state systems undergoing ferromagnetic transitions.

Earlier studies of low-frequency modes in strongly coupled rare-earth systems have been carried out by Cheung,[4] by Lines,[5] and by Buyers.[6] We obtain Buyers's result for the relative weight of the low-frequency modes when we treat the static correlations in the mean-field approximation. Also, Cooper, Fedder, and Schumacher[7] have studied the influence of the exchange interaction on the effective g factor in rare-earth paramagnets. We recover their expressions in the weak coupling limit. Recently Becker, Fulde, and Keller[8] investigated the linewidths of crystal-field excitations in metallic rare-earth systems using an approach which has some points in common with our own. Their work will also be discussed.

The remainder of the paper is divided into four sections. In Sec. II we outline the general theory for the low-frequency modes using the formalism developed by Mori in Ref. 1. The theory presented in Sec. II is rigorous but involves many unknown functions. In order to bring the theory to the point where it can be utilized in the interpretation of experimental data it is necessary to introduce a number of approximations. These are discussed in Sec. III. In Sec. IV we analyze the EPR and neutron-scattering experiments. We summarize our work in Sec. V and comment on its relation to the theory developed in Ref. 8.

II. GENERAL THEORY

In developing the general theory for the low-frequency modes, we postulate a Hamiltonian of the form

$$\mathcal{H} = \sum_i V_i^c + \sum_{i>j} J_{ij} \vec{J}_i \cdot \vec{J}_j + g_J \mu_B H \sum_i J_i^z . \qquad (2.1)$$

Here V_i^c is the crystal-field Hamiltonian associated with the ith ion, J_{ij} is the exchange integral coupling ions i and j, $\vec{J}_i$ is the angular momentum of the ith ion, g_J is the Landé g factor, μ_B is the Bohr magneton, and H is the applied field. Our main interest is in non-Kramers systems which have cubic symmetry both with respect to the local crystalline field and the lattice of rare-earth ions. As a consequence V_i^c is interpreted as a cubic crystalline field operator with manifolds labeled according to the irreducible representations of the cubic group, i.e., $\Gamma_1, \ldots, \Gamma_5$. In addition, the exchange interaction is taken to be isotropic, a result also consistent with cubic symmetry. It is important to note that in most metallic rare-earth systems the exchange interaction is associated with the polarization of the conduction electrons due to $s-f$ coupling. We have not explicitly included conduction-electron terms in the Hamiltonian at this point. However, we will introduce them later in connection with the calculation of the damping.

We will be studying the behavior of the zone-center dynamic susceptibility $\chi_{J_T J_T}(\omega)$ associated with various components of the total angular momentum $\vec{J}_T = \sum_i \vec{J}_i$ whose imaginary part (in units of $g_J{}^2 \mu_B{}^2$) is given by an integral of the form[9]

$$\chi''_{J_T J_T}(\omega) = \omega \,\mathrm{Re} \int_0^\infty dt e^{-i\omega t} (J_T(t), J_T{}^\dagger) dt . \qquad (2.2)$$

Here Re means real part, the dagger denotes adjoint, and $(A(t), B)$ is a relaxation function defined by

$$(A(t), B) = \int_0^\beta d\lambda \langle e^{\lambda \mathcal{H}} e^{i\mathcal{H}t} A e^{-i\mathcal{H}t} e^{-\lambda \mathcal{H}} B \rangle - \beta \langle A \rangle \langle B \rangle , \qquad (2.3)$$

for operators A and B. In this expression, we have $\beta = 1/kT$, T being the temperature, while the angular brackets denote a thermal average.

The essential step in developing quasihydrodynamic theories such as the one outlined here is to identify the slowly varying variables. As noted in Sec. I the slowly varying variables in the rare-earth problem are associated with transitions between degenerate crystal-field levels. In order to make this more precise we utilize the standard-basis-operator formalism introduced by Haley and Erdos.[10] We introduce the standard-basis-operator $\hat{L}^i_{\alpha,\beta}$ associated with the α and β levels of V_i^c. These operators obey the commutation relations

$$(\hat{L}^i_{\alpha,\alpha'}, \hat{L}^j_{\beta,\beta'}) = \delta_{ij} (\delta_{\alpha'\beta} \hat{L}^i_{\alpha,\beta'} - \delta_{\alpha\beta'} \hat{L}^i_{\beta,\alpha'}) , \qquad (2.4)$$

with the consequence that $\hat{L}^i_{\alpha,\beta}$ can be interpreted as an operator taking the ith ion from level β to level α. Any operator can be written as a linear combination of the $\hat{L}^i_{\alpha,\beta}$ since they form a complete set. In particular the s component of the total angular momentum takes the form

$$J^s_T = \sum_i J^s_i = \sum_i \sum_{\alpha,\beta} \langle \alpha | J^s | \beta \rangle \hat{L}^i_{\alpha,\beta} , \qquad (2.5)$$

where $\langle \alpha | J^s | \beta \rangle$ denotes the matrix element of J^s_i between levels α and β of V^c_i. It is assumed to be the same for all sites.

As mentioned above the low-frequency modes in the dynamic susceptibility are associated with transitions between degenerate crystal-field levels. This suggests that an appropriate choice for the slowly varying variables will involve the $\hat{L}^i_{\alpha,\beta}$

with α and β referring to the same crystal-field manifold. The latter can be either Γ_4 or Γ_5 (Γ_3 does not appear since all matrix elements of $\vec{J}$ between states of this symmetry are zero). We designate the slowly varying variables associated with the μth manifold as Y^s_μ $(s=x,y,z)$. They are defined by

$$Y^s_\mu = \sum_i Y^s_\mu(i), \tag{2.6a}$$

$$Y^s_\mu(i) = \sum_{\gamma,\lambda} \langle \mu\delta | J^s | \lambda\mu\rangle \hat{L}^i_{\mu\delta,\mu\lambda}, \tag{2.6b}$$

where $\mu\delta$, $\mu\lambda$ refer to sublevels of the μth manifold.

Having identified the relevant variables for the susceptibility calculation we can begin to develop the Mori formalism. We introduce the projection operators for the low-frequency modes by means of the equation[1]

$$\mathcal{P}^s A = \sum_{\mu,\nu} (A, Y^{s\dagger}_\mu)(S^{-1})_{\mu\nu} Y^s_\nu, \tag{2.7}$$

where the sum on μ and ν is over the N_m Γ_4 and Γ_5 manifolds and $\underline{S}^{-1}$ denotes the inverse of the susceptibility matrix whose $\mu\nu$ element is given by

$$S_{\mu\nu} = \int_0^\beta d\lambda \langle e^{\lambda\mathcal{H}} Y^s_\mu e^{-\lambda\mathcal{H}} Y^{s\dagger}_\nu\rangle - \beta\langle Y^s_\mu\rangle\langle Y^{s\dagger}_\nu\rangle. \tag{2.8}$$

Note that we have dropped the explicit dependence on s in the definition of S since all equal time correlation functions are to be evaluated in the zero-field limit where the x, y, and z directions (which are assumed to coincide with the cubic axes) are equivalent. Our next step is to write J^s_T in the form

$$J^s_T = \mathcal{P}^s J^s_T + (1-\mathcal{P}^s) J^s_T. \tag{2.9}$$

The first term in (2.9) is the low-frequency projection of the total angular momentum; the remainder is associated with high-frequency transitions.

We are interested in both zero-field spin dynamics and EPR. The former is characterized by the susceptibility $\chi_{J_zJ_z}(\omega)$, the latter by $\chi_{J^+J^-}(\omega)$ ($J^\pm = J_x \pm iJ_y$). In both cases the susceptibility takes the form[4,11]

$$\chi''_{JJ}(\omega) = \omega\,\mathrm{Re}\int_0^\infty dt e^{-i\omega t}(\mathcal{P}J_T(t), \mathcal{P}J^\dagger_T) + f(\omega), \tag{2.10}$$

where the function $f(\omega)$, which we do not calculate, is assumed to have negligible spectral weight in the region where the first term on the right-hand side of (2.10) is significant. This assumption is extremely important. Were it not the case there would be no clear-cut separation into high- and low-frequency modes as hypothesized in the development of the theory. The first term in (2.10) comes from the low-frequency modes. It is seen to depend on the one-sided Fourier transform of relaxation functions of the type $(Y^z_\mu(t), Y^z_\nu)$ (for $\chi''_{J_zJ_z}$) or $(Y^+_\mu(t), Y^-_\nu)$, where $Y^\pm_\mu = Y^x_\mu \pm iY^y_\mu$, etc. (for $\chi''_{J^+J^-}$). We denote these transforms by

$$R_{\mu\nu}(\omega) = \int_0^\infty dt e^{-i\omega t}(Y_\mu(t), Y^\dagger_\nu), \tag{2.11}$$

temporarily suppressing the $z,+,-$ notation.

The $R_{\mu\nu}$ are solutions to the exact equation[1]

$$\sum_\nu [i\omega\delta_{\mu\nu} - i\Omega_{\mu\nu} + \Gamma_{\mu\nu}(\omega)]R_{\nu\lambda}(\omega) = \delta_{\mu\lambda}, \tag{2.12}$$

Here $\Omega_{\mu\nu}$ denotes the first moment frequency matrix

$$\Omega_{\mu\nu} = -i\sum_\lambda (\dot{Y}_\mu, Y^\dagger_\lambda)(S^{-1})_{\lambda\nu}, \tag{2.13}$$

with $\dot{A} = dA/dt$, etc., while $\Gamma_{\mu\nu}(\omega)$ is the damping matrix which is defined by

$$\Gamma_{\mu\nu}(\omega) = \sum_\lambda \int_0^\infty dt e^{-i\omega t} (\exp[it(1-\mathcal{P})\mathcal{L}](1-\mathcal{P})\dot{Y}_\mu, \times (1-\mathcal{P})\dot{Y}^\dagger_\lambda)(S^{-1})_{\lambda\nu}, \tag{2.14}$$

where $\mathcal{L}$ denotes the Liouville operator.

Equations (2.6)–(2.14) give the complete solution to the problem of characterizing the low-frequency part of $\chi''_{JJ}(\omega)$. There are a number of general statements that can be made about the solution. First, the first moment frequency matrix associated with the zz component of the susceptibility, $\Omega^{zz}_{\mu\nu}$, is identically zero. This can be seen by making use of the identity

$$(\dot{A}, B) = -i\langle [A,B]\rangle, \tag{2.15}$$

along with the commutation relations

$$[Y^z_\mu, Y^z_\lambda] = \delta_{\mu\lambda}[Y^z_\mu, Y^z_\mu] = 0. \tag{2.16}$$

In the case of the $+-$ component the first-moment matrix differs from zero only in a finite field. To see this we note that $\Omega^{+-}_{\mu\nu}$ involves commutators of the form

$$[Y^+_\mu, Y^-_\lambda] = \delta_{\mu\lambda}[Y^+_\mu, Y^-_\mu]. \tag{2.17}$$

From their definition (2.6), it can be established that variables belonging to either the Γ_4 or Γ_5 manifolds obey angular-momentumlike commutation relations, i.e.,

$$[Y^+_\mu, Y^-_\mu] = 2C_\mu Y^z_\mu, \tag{2.18}$$

where the constant C_μ is connected with the crystal-field-only g factor for the μth manifold, g^0_μ, through the equation

$$g^0_\mu = g_J C_\mu. \tag{2.19}$$

As a result we have

$$\Omega^{+-}_{\mu\nu} = -(g^0_\mu/g_J)\langle Y^z_\mu\rangle(S^{-1})_{\mu\nu}\,, \tag{2.20}$$

where $(S^{-1})_{\mu\nu}$ is given by (2.8) with $s=z$. Since Y^z_μ transforms like an axial vector $\langle Y^z_\mu\rangle$ will be zero when $H=0$. For small fields, which is the only case being considered, we can use thermodynamic perturbation theory to obtain

$$\langle Y^z_\mu\rangle = -g_J\mu_B H(Y^z_\mu, J^z_T)\,, \tag{2.21}$$

where it is understood that the correlation function (Y^z_μ, J^z_T) is evaluated in zero field. Using (2.21) we can rewrite (2.20) in the form

$$\Omega^{+-}_{\mu\nu} = g^0_\mu\mu_B H(Y^z_\mu, J^z_T)(S^{-1})_{\mu\nu}\,. \tag{2.22}$$

We will see later that the $\Omega^{+-}_{\mu\nu}$ are connected with the EPR frequencies.

A second point concerns the relative weight of the low-frequency modes, designated by $\mathcal{R}$, which we define as the integral over all frequencies of the low-frequency part of $\chi''_{JJ}(0)/\pi\omega\chi_{JJ}(0)$:

$$\mathcal{R} = \frac{1}{\pi\chi_{JJ}(0)}\int_{-\infty}^{\infty} d\omega\,[\mathrm{Re}\int_0^\infty dt e^{-i\omega t}(\mathcal{P}J^z_T(t), \mathcal{P}J^z_T)], \tag{2.23}$$

where $\chi_{JJ}(0)$, equal to (J^z_T, J^z_T), is the static susceptibility (in units of $g_J^2\mu_B^2$). It is related to $\chi''_{JJ}(\omega)$ through the Kramers-Kronig relation

$$\chi_{JJ}(0) = \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega\ \omega^{-1}\chi''_{JJ}(\omega)\,. \tag{2.24}$$

If the high-frequency term in (2.10) were absent, we would have $\mathcal{R}=1$, i.e., all the spectral weight would be in the low-frequency modes. However, in general this is not the case and we find

$$\mathcal{R} = \chi_{JJ}(0)^{-1}(\mathcal{P}J^z_T, \mathcal{P}J^z_T)\,, \tag{2.25}$$

which reduces to

$$\mathcal{R} = \chi_{JJ}(0)^{-1}\sum_{\mu,\nu}(J^z_T, Y^z_\mu)(S^{-1})_{\mu\nu}(Y^z_\nu, J^z_T)\,, \tag{2.26}$$

in light of Eq. (2.7) and the fact that Y^z_μ and J^z_T are Hermitian operators.

Although providing the general solution to the problem of the low-frequency modes, the equations developed in this section are largely of formal value. In order to make contact with experiment it is necessary to introduce a number of approximations. These are discussed in Sec. III.

III. APPROXIMATIONS

When applying the theory developed in Sec. II we are forced to make a number of approximations. These fall into two categories depending on whether they pertain to the static or the dynamic correlation functions. We consider the static problem first. As noted previously, we calculate all static correlation functions in the zero-field limit where we have

$$(J^z_T, J^z_T) = \tfrac{1}{2}(J^+_T, J^-_T) \equiv (J_T, J_T)\,, \tag{3.1}$$

$$(Y^z_\mu, J^z_T) = \tfrac{1}{2}(Y^+_\mu, J^-_T) \equiv (Y_\mu, J_T)\,, \tag{3.2}$$

$$(Y^z_\mu, Y^z_\nu) = \tfrac{1}{2}(Y^+_\mu, Y^-_\nu) \equiv (Y_\mu, Y_\nu)\,. \tag{3.3}$$

We use the mean-field approximation (MFA) for the correlation functions (3.11)–(3.3). In the MFA, we have

$$\chi_{JJ}(0)_{\mathrm{MFA}} = (J_T, J_T)_{\mathrm{MFA}} = N\chi^0_{JJ}/(1-\mathcal{J}_0\chi^0_{JJ})\,, \tag{3.4}$$

$$(Y_\mu, J_T)_{\mathrm{MFA}} = N\chi^0_{\mu J}/(1-\mathcal{J}_0\chi^0_{JJ}) = N\chi^0_{\mu\mu}/(1-\mathcal{J}_0\chi^0_{JJ})\,, \tag{3.5}$$

$$S^{\mathrm{MFA}}_{\mu\mu} = (Y_\mu, Y_\mu)_{\mathrm{MFA}} = N\left(\chi^0_{\mu\mu} + \frac{\mathcal{J}_0\chi^0_{\mu J}\chi^0_{\mu J}}{1-\mathcal{J}_0\chi^0_{JJ}}\right) = \frac{N\chi^0_{\mu\mu}}{1-\mathcal{J}_0\chi^0_{JJ}}\,[1-\mathcal{J}_0(\chi^0_{JJ}-\chi^0_{\mu\mu})]\,, \tag{3.6}$$

and for $\mu\neq\nu$

$$S^{\mathrm{MFA}}_{\mu\nu} = (Y_\mu, Y_\nu)_{\mathrm{MFA}} = N\mathcal{J}_0\chi^0_{\mu J}\chi^0_{\nu J}/(1-\mathcal{J}_0\chi^0_{JJ}) = N\mathcal{J}_0\chi^0_{\mu\mu}\chi^0_{\nu\nu}/(1-\mathcal{J}_0\chi^0_{JJ})\,. \tag{3.7}$$

Here $\mathcal{J}_0$ denotes the total exchange,

$$\mathcal{J}_0 = \sum_j J_{ij}\,, \tag{3.8}$$

and N is the number of rare-earth ions. The symbols χ^0_{JJ}, $\chi^0_{\mu J}$, and $\chi^0_{\mu\mu}$ denote single-ion susceptibilities calculated in the absence of exchange interactions, i.e.,

$$\chi^0_{JJ} = Z^{-1}\sum_{\alpha,\beta} e^{-\beta E_\alpha}|\langle\alpha|J^z|\beta\rangle|^2\frac{e^{\beta(E_\alpha-E_\beta)}-1}{E_\alpha-E_\beta}\,, \tag{3.9}$$

where $Z=\sum_\alpha\exp(-\beta E_\alpha)$ and α and β refer to eigenstates of V^c. In the case of the other correlation functions we have

$$\chi^0_{\mu J} = Z^{-1}\sum_{\alpha,\beta} e^{-\beta E_\alpha}\langle\alpha|\sum_\delta\langle\mu\delta|J^z|\delta\mu\rangle\hat{L}_{\mu\delta,\mu\delta}|\beta\rangle \times\langle\beta|J^z|\alpha\rangle\frac{e^{\beta(E_\alpha-E_\beta)}-1}{E_\alpha-E_\beta}\,. \tag{3.10}$$

Since the standard basis operators have the property $\hat{L}_{\alpha\beta}|\gamma\rangle = \delta_{\beta\gamma}|\alpha\rangle$ the sum in (3.10) collapses to a sum over the levels of the μth manifold, and we have the result

$$\chi^0_{\mu J} = (kTZ)^{-1}e^{-\beta E_\mu}\sum_\delta|\langle\mu\delta|J^z|\delta\mu\rangle|^2 = \chi^0_{\mu\mu}\,, \tag{3.11}$$

which has been anticipated in obtaining the last lines of Eqs. (3.5)–(3.7).

It is difficult to establish the extent of the errors

introduced in making the mean-field approximation. It appears to work quite well for many rare-earth systems presumably because of the long-range interactions. However, the MFA does breakdown near the ordering temperature. Even here in the case of antiferromagnets the approximation may not be too bad for our purposes since we are carrying out calculations at the center of the Brillouin zone, where the susceptibility remains finite, as opposed to the antiferromagnetic superlattice point where the susceptibility diverges.

Using the expressions given in Eqs. (3.4)–(3.7) we can obtain the MFA approximation for the relative weight of the low-frequency modes [Eq. (2.26)]. As shown in the Appendix we have

$$\mathcal{R}=\frac{\sum_{\mu}\chi^{0}_{\mu\mu}}{\chi^{0}_{JJ}[1-\mathcal{J}_{0}(\chi^{0}_{JJ}-\sum_{\mu}\chi^{0}_{\mu\mu})]}. \tag{3.12}$$

Equation (3.12) is identical to an expression derived by Buyers[6] in a hybrid random-phase mean-field calculation. It has the property that $\mathcal{R}=1$ whenever $\mathcal{J}_{0}\chi^{0}_{JJ}=1$. Since the latter equation determines the transition temperature in the MFA treatment of a ferromagnet we conclude that there is a large increase in the intensity of the low-frequency modes near the Curie point.

The paramagnetic resonance is described by the transverse susceptibility $\chi_{J^{+}J^{-}}(\omega)$. Whenever the linewidth is small in comparison with the resonance frequency we can use (2.12) with $\Gamma_{\mu\nu}$ set equal to zero to calculate the resonance spectrum. The frequencies are the eigenvalues of the first-moment frequency matrix, while the intensities of the resonances are determined by the corresponding eigenvectors. The general solution involves standard numerical and algebraic techniques. However, there are two limiting cases which have simple solutions. The first corresponds to a situation where $|\mathcal{J}_{0}|\chi^{0}_{JJ}\ll 1$. In this limit the manifolds become decoupled from one another. The effective g factors are then given by

$$g^{\mathrm{eff}}_{\mu}\approx g^{0}_{\mu}[1+\mathcal{J}_{0}(\chi^{0}_{JJ}-\chi^{0}_{\mu\mu})]. \tag{3.13}$$

and the relative weights by $\chi^{0}_{\mu\mu}/\chi^{0}_{JJ}$. Equation (3.13) is the same as a result obtained earlier by Cooper *et al.* in a first-order calculation.[7] The other case corresponds to having only one populated degenerate level. In this situation, where the system can be described by a singlet-triplet model, we have[12]

$$g^{\mathrm{eff}}_{1}=g^{0}_{1}[1-\mathcal{J}_{0}(\chi^{0}_{JJ}-\chi^{0}_{11})]^{-1}, \tag{3.14}$$

and

$$\mathcal{R}=\chi^{0}_{11}/\chi^{0}_{JJ}[1-\mathcal{J}_{0}(\chi^{0}_{JJ}-\chi^{0}_{11})]. \tag{3.15}$$

The damping of the low-frequency modes is a complicated problem. In the analysis of relaxation effects it is important to distinguish between intrinsic and extrinsic damping. Intrinsic damping, which is associated with Hamiltonians of the type given by (2.1), arises from interactions among the spin fluctuations. Extrinsic damping comes from direct interactions between the spin fluctuations and the conduction electrons and phonons. In systems with direct or superexchange intrinsic relaxation usually dominates if the exchange interaction is strong enough to induce a phase transition. However, in systems with indirect exchange, extrinsic damping coming from first-order interactions with the conduction electrons is frequently the most important relaxation mechanism.[13] Since we are primarily interested in metallic rare-earth systems with indirect exchange interactions we will mainly consider extrinsic damping.

We introduce extrinsic damping using an approximation which is similar in spirit to the MFA. The integral in the equation for $\Gamma_{\mu\nu}$ (2.14) is evaluated treating the ions as independent, while the matrix $\underline{S}$ is calculated in the MFA. We obtain the result

$$\Gamma_{\mu\nu}(\omega)=N\gamma^{\mathrm{SI}}_{\mu\mu}\chi^{0}_{\mu\mu}(S^{-1})^{\mathrm{MFA}}_{\mu\nu}, \tag{3.16}$$

where $\chi^{0}_{\mu\mu}$ is given by (3.11) and $(S^{-1})^{\mathrm{MFA}}_{\mu\nu}$ by Eq. (A9) in the Appendix. Here the symbol $\gamma^{\mathrm{SI}}_{\mu\mu}$ denotes the low-frequency limit of the single-ion linewidth associated with the μth manifold. (Note that as $\mathcal{J}_{0}\to 0$, $S_{\mu\nu}\to N\delta_{\mu\nu}\chi^{0}_{\mu\mu}$ so that $\Gamma_{\mu\nu}\to\delta_{\mu\nu}\gamma^{\mathrm{SI}}_{\mu\mu}$.) Since a general formula for $\gamma^{\mathrm{SI}}_{\mu\mu}$ has been given earlier[14] we will not discuss it in any detail.

The appearance of the single-ion linewidth in (3.16) is indicative of the fact that we have tacitly assumed that the fluctuations of the conduction-electron polarization at different sites are uncorrelated so that each ion contributes independently to the integral in (2.14). Moreover, we have also assumed that the fluctuations in the polarization have a decay rate which is large in comparison with the relaxation rates of the low-frequency modes. We have also omitted off-diagonal single-ion damping associated with time-dependent correlations involving $\dot{Y}_{\mu}$ and $\dot{Y}_{\nu}$, an approximation which is consistent with the vanishing of the static correlation function (Y_{μ}, Y_{ν}) $(\mu\neq\nu)$ when $\mathcal{J}_{0}=0$.

The calculation of the low-frequency part of $\chi''_{JJ}(\omega)$ in zero-field involves the solution of (2.12) with $\Omega_{\mu\nu}=0$ and $\Gamma_{\mu\nu}$ approximated as shown in (3.16). As in the resonance problem the modes become decoupled when $|\mathcal{J}_{0}|\chi^{0}_{JJ}\ll 1$. The low-frequency part becomes a sum of Lorentzians with widths $\gamma^{\mathrm{SI}}_{\mu\mu}$ and relative weights $\chi^{0}_{\mu\mu}/\chi^{0}_{JJ}$. The other simple case corresponds to the singlet-triplet model. Here the low-frequency term consists of a single Lorentzian with a width given by

$$\Gamma_{11}=\frac{\gamma_{11}^{SI}(1-\mathcal{J}_0\chi_{JJ}^0)}{1-\mathcal{J}_0(\chi_{JJ}^0-\chi_{11}^0)}, \tag{3.17}$$

and relative weight by (3.15).

It is beyond the scope of this paper to discuss intrinsic damping in any detail. However, there is one aspect of the problem that deserves comment. The damping matrix involves correlations of $\dot{Y}_\mu^s$ which is identified with $i(Y_\mu^s,\mathcal{H})$ with $\mathcal{H}$ given by (2.1). If the exchange interaction is written in terms of standard basis operators two types of terms can be recognized as possible contributors to $\dot{Y}_\mu^s$, those involving operators $\hat{L}_{\mu\delta,\mu\lambda}$ which are associated with transitions within the μth manifold and terms involving $\hat{L}_{\mu\delta,\beta}$, where β is any level outside the μth manifold. It can be shown that the terms with $\hat{L}_{\mu\delta,\mu\lambda}$ do not contribute to $\dot{Y}_\mu^s$ when the exchange interaction is isotropic. The reason is that the Y_μ^s act like (total) pseudospin operators with respect to the μth manifold [Eq. (2.18)]. If the exchange interaction is isotropic that part of the Hamiltonian which refers entirely to the μth manifold can be written as a combination of scalar products of pseudospin operators and hence commutes with Y_μ^s. Since only the intermanifold terms contribute to $\dot{Y}_\mu^s$ the intrinsic damping of the low-frequency modes at the center of the zone will be very small when the crystal-field splittings are large in comparison with the exchange splitting. This of course is the situation in many $3d$ insulators. Here the intermanifold contribution to the damping is negligible so that the EPR linewidth comes from the anisotropic terms in the Hamiltonian. It should be noted that because of dispersion or "soft-mode" effects, in some systems the energy of the crystal-field excitons can be much less than the corresponding single-ion crystal-field splittings. Such a situation is likely to lead to an enhancement of the intermanifold damping over what one would calculate using single-ion levels.

Having developed the general theory and appropriate first approximations, we are in a position to carry out detailed calculations for specific systems. The results of these calculations are discussed in Sec. IV.

IV. APPLICATIONS

In this section we discuss several applications of the theory. We begin with the EPR in TbP. Terbium phosphide is a singlet ground-state antiferromagnet which orders at 8 K. Despite the fact that the magnetic excitations near the superlattice point are probably overdampled as is the case for TbSb,[15] where EPR is also observed,[2] the EPR of the $\Gamma_5^{(2)}$ triplet in TbP is detectable down to temperatures on the order of 10 K indicating that there is relatively weak damping of the low-frequency modes at the center of the zone. Huang *et al.*[2] have reported measurements of the intensity, linewidth, and g factor of the $\Gamma_5^{(2)}$ resonance in the range 10–50 K. They find that the intensity is quite well described by the single-ion expression $e^{-35/T}/TZ$ corresponding to an energy gap of 35 K between the $\Gamma_5^{(2)}$ manifold and the ground state. The linewidth is observed to increase with temperature from 200 G(10 K) to 800 G(50 K). They also report an anomalous increase in the g factor near the ordering temperature. However, the anomaly is now believed to be an artifact of their criterion for determining the resonance field.[16]

Perhaps the most unusual feature of the data for TbP is the applicability of the single-ion expression for the intensity in a system where the ions interact strongly enough to bring about a phase transition. The explanation lies in the small value of the paramagnetic Curie temperature, 1 K, reported by Busch *et al.*[17] From the paramagnetic Curie temperature, T_p, we infer a value of $\mathcal{J}_0$ $[=3kT_p/J(J+1)]$ equal to 0.2 K. As is evident in Sec. III the relative importance of the effects of the exchange interaction on the dynamics of low-frequency modes at the center of the zone is determined by the value of $|\mathcal{J}_0|\chi_{JJ}^0$. In the case of TbP, we have $|\mathcal{J}_0|\chi_{JJ}^0 \leq 0.1$ throughout the entire paramagnetic region. As a consequence the exchange interaction has only a weak effect on the $\Gamma_5^{(2)}$ resonance so that the g factor is adequately approximated by (3.13) and the intensity by $\chi_{\Gamma_5\Gamma_5}^0$.[12]

The behavior seen in TbP contrasts strongly with what we predict for the EPR in singlet ground-state ferromagnets. Near the Curie point the exchange interaction has a strong effect on the resonance. This is evident from the fact that $\mathcal{J}_0\chi_{JJ}^0$ reaches a maximum value of unity at the mean-field transition temperature. We have calculated the g factors and spectral weights [in $\chi_{J^+J^-}''(\omega)/\omega$] of the EPR modes in the singlet ground-state ferromagnet Pr_3Tl. A fourth-order crystal field was assumed with $E_{\Gamma_4}-E_{\Gamma_1}=77$ K. We also took $\mathcal{J}_0$ $=5.8$ K corresponding to a mean-field transition temperature of 16 K.[18] Omitting damping, we obtain results for the g factors and weights which are shown in Figs. 1 and 2, respectively. For $T>100$ K the two g factors approach the crystal-field-only values of 2.0 and 0.4. Below 40 K both $g_1^{\text{eff}}(T)$ and $g_2^{\text{eff}}(T)$ increase rapidly. Although $g_1^{\text{eff}}>g_2^{\text{eff}}$ for all $T>T_C$ the spectral weight of resonance (2) equals that of (1) at 35 K; below this temperature the spectrum is increasingly dominated by the (2) resonance whose g factor approaches a limiting value ≈ 300 at the (MFA) T_C.

While it is clear that not too much quantitative significance can be attached to predictions of crit-

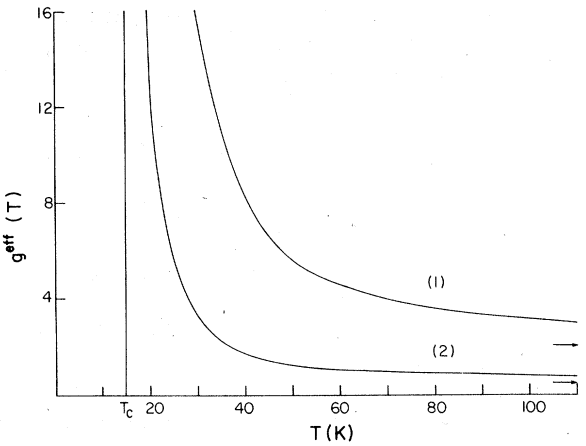

FIG. 1. Effective g factors in Pr_3Tl vs temperature. The arrows indicate the values in the absence of the exchange interaction, where (1) and (2) are identified with the Γ_5 and Γ_4 crystal-field multiplets, respectively. T_C denotes the mean-field Curie temperature.

ical anomalies in ferromagnets which are based on mean-field approximations we expect the qualitative features of Figs. (1) and (2) to be preserved in more realistic calculations. We are also led to predict critical anomalies in the g factors of other singlet ground-state ferromagnets provided the resonances are not overdamped (as may actually be the case[16] in Pr_3Tl).

Although not obvious in Figs. 1 and 2, there is another feature of the EPR which deserves mention. The exchange interaction induces a coupling between the crystal-field resonances so that the EPR modes are no longer purely Γ_4 or Γ_5. This is evidence in the limiting expression for the g factor of the (2) resonance at the mean-field ordering temperature. We find

$$g_2^{\mathrm{eff}}(T_c)=\frac{g^0_{\Gamma_4}g^0_{\Gamma_5}}{g^0_{\Gamma_4}\mathcal{J}_0\chi^0_{\Gamma_5\Gamma_5}(T_C)+g^0_{\Gamma_5}\mathcal{J}_0\chi^0_{\Gamma_4\Gamma_4}(T_C)}. \quad (4.1)$$

Equation (4.1) formally treats the Γ_5 and Γ_4 levels on an equal footing, although in Pr_3Tl we have $\chi^0_{\Gamma_4\Gamma_4}(T_C)\gg\chi^0_{\Gamma_5\Gamma_5}(T_C)$ so that $g_2^{\mathrm{eff}}(T_C)\approx g^0_{\Gamma_4}/(\mathcal{J}_0\chi^0_{\Gamma_4\Gamma_4}(T_C)$. It should be pointed out that the (1) resonance has a g factor which is infinite at T_C. However, its intensity is finite at the critical point whereas the intensity of resonance (2) diverges.

We have also studied the low-frequency modes in Pr_3Tl in zero field assuming extrinsic damping due to interactions with conduction electrons. The damping matrix was calculated using Eq. (3.16) together with the results for the single-ion damping rate given in Ref. 14. Since the s-f exchange integral is not known for this material, our widths are calculated only to within an overall multiplicative factor. In Fig. 3(a) we show the temperature dependence of the half-width at half-height of the central peak in the dynamic structure factor of Pr_3Tl, while in 3(b) we display our results for the relative intensity of the central peak as given by Eq. (3.12). For $T>40$ K the half-width varies approximately as $a+bT$ as might be expected for a single ion. Below 20 K critical effects set in. The linewidth drops to zero at T_C while the relative intensity rapidly increases to unity.

Just as in the case of the EPR modes the relaxational modes characterizing the dynamics in zero field are a combination of modes associated with the Γ_4 and Γ_5 manifolds. At high temperatures, where $\mathcal{J}_0\chi^0_{JJ}\ll 1$, the two manifolds contribute

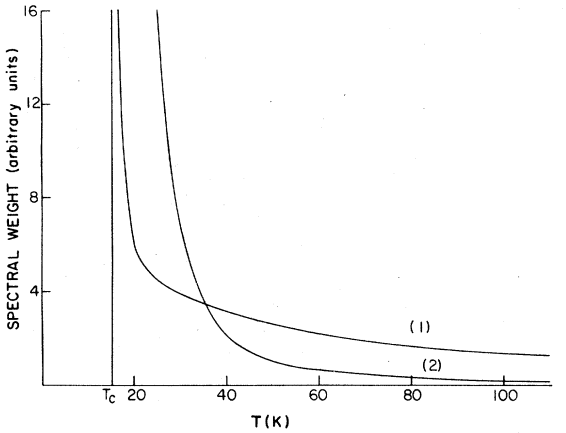

FIG. 2. Spectral weight [in $\chi''_{J^+J^-}(\omega)/\omega$] of the EPR modes in Pr_3Tl vs temperature. (1) and (2) are associated with the corresponding g factors in Fig. 1.

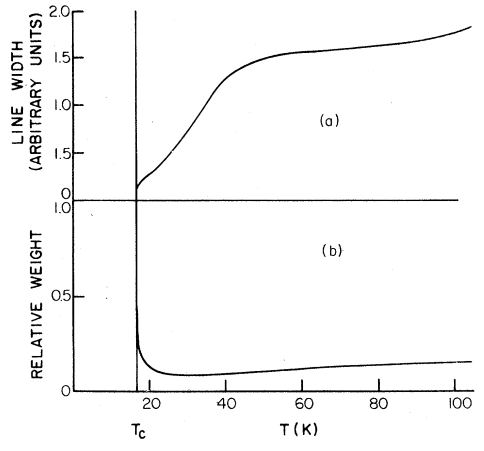

FIG. 3. (a) Temperature dependence of the half-width at half-height of the central peak in the dynamic structure factor for Pr_3Tl. (b) Relative weight of the central peak [Eq. (3.12)] in Pr_3Tl. T_C denotes the mean-field Curie temperature.

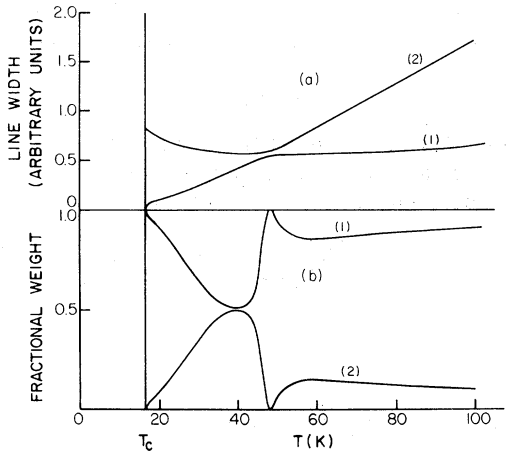

FIG. 4. (a) Temperature dependence of the half-widths of the two low-frequency modes in Pr_3Tl. (b) Fractional weights of the two low-frequency modes in Pr_3Tl. In the high-temperature limit, $\mathcal{J}_0\chi^0_{JJ} \to 0$, (1) and (2) are identified with the Γ_5 and Γ_4 multiplets, respectively. Note that the sum of the two fractional weights is equal to unity.

independently to the response. However, as the temperature decreases the coupling between the manifolds becomes stronger. In the limit $T \to T_C$ the response is dominated by the mode with the smallest relaxation rate, $\Gamma_{\min}$, which is found to vary as

$$\Gamma_{\min} = \frac{\gamma^{SI}_{\Gamma_4\Gamma_4}\gamma^{SI}_{\Gamma_5\Gamma_5}(1-\mathcal{J}_0\chi^0_{JJ})}{\gamma^{SI}_{\Gamma_4\Gamma_4}\mathcal{J}_0\chi^0_{\Gamma_5\Gamma_5}+\gamma^{SI}_{\Gamma_5\Gamma_5}\mathcal{J}_0\chi^0_{\Gamma_4\Gamma_4}}, \tag{4.2}$$

for $T \approx T_C$. Note that as T approaches the (mean-field) transition temperature the linewidth vanishes as $(1-\mathcal{J}_0\chi^0_{JJ})$, i.e., as the inverse of the static susceptibility. The latter behavior is characteristic of "conventional critical slowing down" and is likely to be modified by nonlinear interactions among the Y_μ.[19] In contrast, the linewidth of the fast relaxing mode remains finite at T_C. However, its relative intensity goes to zero so it makes no contribution to the response. This behavior is illustrated in Fig. 4 where we show the linewidths and fractional intensities of the two modes contributing to the central peak whose overall half-width and relative intensity are displayed in Fig. 3. In addition to the critical anomalies we note the interference effects associated with the anticrossing at 48 K.

Recently, inelastic neutron scattering studies of the central peak in Pr_3Tl have been reported by Als-Nielsen *et al.*[3] Their experiments show an increase in the intensity near T_C, as predicted by (3.12). Unfortunately there is insufficient data in the paramagnetic phase to make a comparison with the linewidth curve in Fig. 3.

V. DISCUSSION

There are a number of additional points to be made in connection with the theory outlined in Sec. II. First, although the analysis we have given is for $\vec{J}_T$ it could equally well have been given for the Fourier transform $\vec{J}(\vec{q}) = \sum_n \exp(i\vec{q}\cdot\vec{r}_n)\vec{J}_n$ in which case the operators Y^s_μ would become $Y^s_{\mu\vec{q}} = \sum_n \times \exp(i\vec{q}\cdot\vec{r}_n)Y^s_\mu(n)$ with corresponding changes in the definitions of the susceptibilities, relaxation functions, etc. Second, despite being developed for non-Kramers ions the formalism carries over almost intact to Kramers ions. The only complication that arises is in connection with the behavior of the Γ_8 manifold in an external field. In some cases, e.g., $J = \frac{5}{2}$ and $\frac{7}{2}$, the Y^s_μ associated with this manifold do not obey angular momentumlike commutation relations [Eq. (2.17)]. When this situation occurs the Γ_8 manifold has multiple resonance frequencies rather than a single resonance frequency as is always the case for the Γ_6 and Γ_7 manifolds. The multiplicity of resonances leads to an increase in the dimensionality of Eq. (2.12) since each of the modes has to be treated separately.

One of the most important questions to be addressed concerns the applicability of the theory to metallic systems where the rare-earth ions interact indirectly through the conduction electrons. Becker *et al.*[8] have argued that the response of the conduction electrons cannot be neglected in calculating the rare-earth dynamic susceptibility. In the case of the singlet-triplet system they obtain an expression for the dynamic susceptibility which has two low-frequency poles, one of which is associated with the crystal field excitations and corresponds to what we would obtain using the formalism of Secs. II and III. The other pole is associated with the conduction electrons and describes paramagnon excitations. It is important to note that their results were obtained at finite wave vector using an expression for the dynamic susceptibility of the conduction electrons which is valid only when $ql \gg 1$, where l is the electron mean free path. As pointed out by Fulde and Peschel[20] the character of the susceptibility changes in the small momentum range, $ql \ll 1$. In this regime in zero-field we have

$$\chi_e(q,\omega) = \chi^0_e \frac{1/\tau + Dq^2}{1/\tau + Dq^2 + i\omega}, \tag{5.1}$$

where χ^0_e is the static susceptibility, D is the diffusion constant, and $3/4\tau$ is the rate for the spin-flip scattering of the conduction electrons by the rare-earth ions. The essential point is the relative magnitude of $1/\tau$ in comparison with the relaxation rates of the low-frequency rare-earth modes.

We can obtain a rough estimate of this ratio from the detailed balance relation for the rare-earth–conduction-electron system[21]:

$$\gamma_{\mathrm{RE}}\tau = 3kT\chi_e^0/NJ(J+1), \tag{5.2}$$

where γ_{RE} is the rare-earth relaxation rate due to conduction electron scattering and χ_e^0 is in units of $g_e^2\mu_B^2$, g_e being the electronic g factor. Equation (5.2) is obtained from a theory which omits crystal-field effects. However, the single-ion relaxation rates in the presence of the cubic crystalline field are usually of the order of magnitude $(g_\mu^0/g_J)^2\gamma_{\mathrm{RE}}$. Since χ_e^0 is approximately $3N_e/2E_F$, where N_e is number of conduction electrons and E_F is the Fermi energy we have

$$\gamma_{\mathrm{RE}} \approx \left(\frac{9kTN_e}{2NJ(J+1)E_F}\right)\frac{1}{\tau}. \tag{5.3}$$

In the case of concentrated systems $N_e \approx N$ so that $\gamma_{\mathrm{RE}} \sim (kT/E_F)1/\tau \ll 1/\tau$.

Since the relaxation rates for the low-frequency modes are likely to be much less than $1/\tau$ we believe we are justified in approximating $\chi_e(q,\omega)$ by χ_e^0 in calculating the effective interaction between the rare-earth ions in the low-frequency–long-wavelength regime. When this is done we are left with an expression for the low-frequency part of $\chi_{JJ}(\omega)$ which has only crystal-field poles. On the basis of this argument we conclude that as long as $ql \ll 1$ our theory which is based on the Hamiltonian (2.1) with extrinsic damping introduced through Eq. (3.16), is a valid approach for characterizing the low-frequency relaxation modes in metallic rare-earth systems.

In summary, we have developed a general theory for the low-frequency modes in rare-earth systems with degenerate crystal-field levels. From a comparison with experimental data we conclude that the theory is successful in explaining the qualitative features of the results. However additional work is needed. It is important to go beyond mean-field theory in obtaining approximations for the static correlation functions. Further studies of the linewidth mechanisms are also called for. In particular, one would like to know the limitations of the approximate treatment of extrinsic damping. On the experimental side, additional measurements of the linewidths and g factors in well-characterized systems are needed. Paramagnetic resonance studies in singlet ground-state ferromagnets could be an important test of the validity of the coupled-mode picture.

ACKNOWLEDGMENTS

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APPENDIX

In this Appendix, we outline the proof of Eq. (3.12). In the MFA, the general expression for the relative intensity, (2.26), can be written

$$\mathcal{R} = \frac{1}{\chi_{JJ}(0)}\sum_{\mu,\nu}\frac{NY^0_{\mu\mu}}{1-\mathcal{J}_0\chi^0_{JJ}}(S^{-1})^{\mathrm{MFA}}_{\mu\nu}\frac{NY^0_{\nu\nu}}{1-\mathcal{J}_0\chi^0_{JJ}},$$

$$= \frac{1}{\chi^0_{JJ}}\sum_{\mu,\nu}(Z^{-1})_{\mu\nu}, \tag{A1}$$

where the matrix $\underline{Z}$ has elements

$$Z_{\mu\nu} = A_\mu\delta_{\mu\nu} + J_0, \tag{A2}$$

with

$$A_\mu = \chi^{0-1}_{\mu\mu}(1-\mathcal{J}_0\chi^0_{JJ}). \tag{A3}$$

We write $\underline{Z}$ as the sum of two matrices:

$$\underline{Z} = \underline{A} + \mathcal{J}_0\underline{B}, \tag{A4}$$

where $\underline{A}$ is a diagonal matrix with elements A_μ while $\underline{B}$ has all elements equal to unity. The inverse of $\underline{Z}$ is then given by the equation

$$\underline{Z}^{-1} = \underline{A}^{-1} - \mathcal{J}_0\underline{A}^{-1}\underline{B}\underline{Z}^{-1},$$

$$= \underline{A}^{-1} - \mathcal{J}_0\underline{A}^{-1}\underline{B}\underline{A}^{-1} + \mathcal{J}_0^2\underline{A}^{-1}\underline{B}\underline{A}^{-1}\underline{B}\underline{A}^{-1} - \cdots, \tag{A5}$$

upon iteration. As a direct consequence of the properties of $\underline{A}$ and $\underline{B}$ we obtain the result

$$\sum_{\mu,\nu}(Z^{-1})_{\mu\nu} = \sum_\mu A_\mu^{-1} - \mathcal{J}_0\sum_{\mu,\nu}A_\mu^{-1}B_{\mu\nu}A_\nu^{-1}$$

$$+\mathcal{J}_0^2\sum_{\mu,\nu,\lambda}A_\mu^{-1}B_{\mu\lambda}A_\lambda^{-1}B_{\lambda\nu}A_\nu^{-1} - \cdots,$$

$$+\mathrm{Tr}\underline{A}^{-1} - \mathcal{J}_0(\mathrm{Tr}\underline{A}^{-1})^2 + \mathcal{J}_0^2(\mathrm{Tr}\underline{A}^{-1})^3 - \cdots,$$

$$= \frac{\mathrm{Tr}\underline{A}^{-1}}{1+\mathcal{J}_0\mathrm{Tr}\underline{A}^{-1}}, \tag{A6}$$

where Tr denotes the trace operation

$$\mathrm{Tr}\underline{A}^{-1} = \sum_\mu A_\mu^{-1} = (1-\mathcal{J}_0\chi^0_{JJ})^{-1}\sum_\mu\chi^0_{\mu\mu}. \tag{A7}$$

Using (A7), we can rewrite (A6)

$$\sum_{\mu,\nu}(Z^{-1})_{\mu\nu} = \frac{\sum_\mu\chi^0_{\mu\mu}}{1-\mathcal{J}_0(\chi^0_{JJ}-\sum_\mu\chi^0_{\mu\mu})}, \tag{A8}$$

which is equivalent to (3.12).

Essentially the same arguments can be used to evaluate S^{-1} in the MFA. We find

$$N(S^{-1})^{\mathrm{MFA}}_{\mu\nu} = \chi^{0\ -1}_{\mu\mu}\delta_{\mu\nu} - \frac{\mathcal{J}_0}{1-\mathcal{J}_0(\chi^0_{JJ}-\sum_\lambda\chi^0_{\lambda\lambda})}. \tag{A9}$$

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