Dynamics of Localized Moments in Metals. I. Kubo Formalism and Relaxation

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The dynamic transverse susceptibility $\chi^-(\omega)$ is calculated microscopically for a random array of localized spins in a metal. Relaxation of both local and conduction-electron spins towards their respective instantaneous local fields is included using a new formalism for such instantaneous relaxation. The resulting equations for the relevant propagators are solved to first order in the exchange coupling constant J, and it is demonstrated that to this order, the results are identical to those obtained using a molecular-field model. Careful attention is paid to the correct analytic form of the spin propagators and to the sum rules which they must satisfy. It is shown that in this formalism an additional inhomogenous contribution to $\chi^-(\omega)$ arises from the requirement of relaxation to the local field. When this term is added to the usual Lorentzian form for the spin propagators, a form for $\chi^-(\omega)$ results which is distinctly non-Lorentzian, but which satisfies the physical requirement that the static limit ($\omega=0$) of the transverse susceptibility equal the longitudinal value.

1. INTRODUCTION

IN this paper we examine the dynamics of coupled spin systems, explicitly including in their equations of motion relaxation to an instantaneous self-consistent local field. Previous work on this problem, notably that of Peter *et al.*,[1] utilized a macroscopic molecular-field approach. Their results exhibited the physical requirement that in the static limit, the transverse and longitudinal spin susceptibilities were identical. They also discussed the significance of their result and its application to a wide variety of physical systems. We demonstrate here, by means of a microscopic calculation, the validity of their result to lowest order in the exchange coupling. In addition, we generate a framework which will enable methods of quantum field theory to be applied to the general problem of lattice relaxation in spin systems. In particular, we believe this method can be applied to extend the present calculation to higher order in the coupling strength. Other recent work, notably that of Giovannini *et al.*,[2] has also investigated the microscopic behavior of these systems, but they were forced to introduce an *ad hoc* modification of the usual Lorentzian spin Green function in order to obtain a correct[3] physical form for the transverse dynamic susceptibility, $\chi^-(\omega)$. We show in Sec. 4 that this modification, in fact, violates a fundamental sum rule which can always be constructed for any "one-particle" propagator. However, in Secs. 2 and 3 we demonstrate that the Lorentzian form of the Green's function need not be modified, but rather an inhomogeneous term must be added to the usual Kubo[4] linear response formula for the magnetization, which then yields the correct[3] form.

The starting point of this paper is the explicit introduction, into the quantum-mechanical operator equations of motion of the spin system, of a term corresponding to the relaxation of the system to the direction of the instantaneous local field. This technique somewhat artificially separates the relaxation term from the Hamiltonian which describes the dynamics of the spin system when no damping is present—the usual Heisenberg equation of motion. Thus, we regard the relaxation term as an "external" perturbation, the remainder of the dynamics we assume to be governed by the "internal" Hamiltonian. Although in principle such terms could be included in a complete "internal" Hamiltonian, eventually one must introduce contact with a heat bath in order to achieve thermodynamic equilibrium. We feel that it is easier, and more physical, to make the separation at this stage. These equations are an interpolation of the Heisenberg equations of motion for spin operators when no damping is present and the phenomenological Bloch[1,5] equations describing the dynamics of the average magnetizations driven by an effective magnetic field and undergoing "external" relaxation.

In Sec. 2, these equations are analyzed (for simplicity), for a single-spin system interacting with a coherent rf magnetic field in the presence of an external static magnetic field along the z axis. The results will be compared, at each stage, with corresponding expressions which only consider relaxation to the static magnetic field (z direction). Obviously the results for the dynamic transverse susceptibility must differ substantially between the two cases. As stated above, a fundamental physical requirement for the transverse susceptibility is that in the static limit it must reduce to the longitudinal susceptibility. This essentially follows from the

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[5] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford at the Clarendon Press, Oxford, England, 1961), especially p. 53.

long-time requirement (zero frequency) that the magnetization must follow vectorially the external field regardless of the relaxation rate. It is a simple matter[6] to show then that this results in an equality of static transverse and longitudinal susceptibilities.

Now, a Lorentzian result for the dynamic transverse susceptibility, which we show to obtain under the assumption of relaxation to only the external static magnetic-field direction, does not in fact reduce to the longitudinal susceptibility at zero frequency. In the case of a precessing free spin (after all transients have died away) this occurs because there is no preferred direction in the transverse plane. However, the requirement of relaxation to the instantaneous local field will be shown to result in a modified Lorentzian form for the transverse susceptibility which, in fact, reduces immediately to the longitudinal result in the static limit. Again, for the free-spin case, relaxation towards the instantaneous field defines (at any instant) a unique direction in the transverse plane and produces a net moment (complex) proportional to the perturbing field. This vanishes as the damping disappears but behaves like a static component as the damping becomes infinite. This contribution is really a coherence effect which extracts a net transverse component from the precessing motion of the magnetic moment around the static field. At the same time this additional transverse field is creating an extra transverse component by inducing spin flips of the actual z component of the moment, and for small fields this is given by the usual Kubo formalism. However, it is just this additional coherent contribution which modifies the basic Lorentzian Kubo result.

In Sec. 3, we treat the problem of a random collection of localized spins in a metal, interacting with the conduction-electron spins via a zero-range exchange coupling. We work to only first order in this coupling[7] and include the effects of "lattice" damping for both systems using the methods introduced in Sec. 2. We then solve the coupled dynamic susceptibilities and demonstrate their equality with the molecular-field results.

Section 4 contains a discussion of our results and of sum rules which must be satisfied by a Green's function. It is shown that part of the susceptibility which can be expressed as a retarded Green's function, as derived in Secs. 2 and 3, does satisfy the appropriate sum rule but the other part, originating from the assumption of relaxation to the instantaneous local field, does not belong in the propagator sum rule, and in fact, would destroy it if forcibly introduced into the propagator expressions.[2]

[6] C. P. Slichter, *Magnetic Resonance* (Harper and Row Publishers, Inc., New York, 1963), especially p. 39.

[7] Although results quoted in this paper include terms of order J^2, these arise from linear approximations in two coupled equations. Using the general methods derived in this paper, we have extended our treatment self-consistently to the next order in J; see following paper, Phys. Rev. **179**, 690 (1969).

2. RELAXATION EQUATIONS

In order to illustrate the ideas which are applied in Sec. 3 to the case of two coupled spin systems in a metal, we restrict ourselves in this section to the simple example of a single localized magnetic moment $\mathbf{M}$ or its quantum mechanical spin operator $\mathbf{S}$ ($\mathbf{M}=\gamma\mathbf{S}$) which is part of a system described by a Hamiltonian $\mathcal{H}$, and assumed to be affected by an external relaxation mechanism, characterized by a single inverse relaxation time Δ. In the absence of damping the dynamics of the moment is described by the Heisenberg equation of motion (with $\alpha=1,2,3$)

$$i(d/dt)S^\alpha(t)=[S^\alpha(t),\mathcal{H}], \tag{2.1}$$

where

$$[S^\alpha,S^\beta]=i\sum_\gamma \epsilon_{\alpha\beta\gamma}S^\gamma. \tag{2.2}$$

If an additional perturbation is applied adiabatically at $t=0$, the change in the averaged components of the spin are given by the usual Kubo linear response formula[4]

$$\langle\delta S^\alpha(t)\rangle_{\rm ext}=-i\int_0^t dt'\langle[S^\alpha(t),\mathcal{H}_{\rm ext}(t')]\rangle, \tag{2.3}$$

where $\langle S^\alpha\rangle_{\rm ext}$ is a canonical average with respect to the exact eigenstates of the total Hamiltonian, $\mathcal{H}+\mathcal{H}_{\rm ext}$, but weighted by the corresponding Boltzman factor containing only the energy eigenvalues of $\mathcal{H}$ (since the original population distributions are assumed not to have changed). Moreover, $\langle S^\alpha\rangle$ are canonical averages with respect to the original Hamiltonian $\mathcal{H}$. The change in the operator average is defined by

$$\langle\delta S^\alpha(t)\rangle_{\rm ext}=\langle S^\alpha(t)\rangle_{\rm ext}-\langle S^\alpha(t)\rangle. \tag{2.4}$$

We assume that $\mathcal{H}$ contains a large static magnetic field h_0, and that the z axis of spin quantization is taken parallel to the direction of h_0. We then impress on the system a small rf rotating transverse magnetic field resulting in a driving term

$$\mathcal{H}_{\rm ext}(t)=-\gamma\mathbf{S}\cdot\mathbf{h}_{\rm rf}(t). \tag{2.5}$$

Thus Eq. (2.3) takes the explicit form

$$\langle S^-(t)\rangle_{\rm ext}=\langle S^-(t)\rangle+\tfrac{1}{2}\gamma\int_{-\infty}^{\infty}dt'F_R(t-t')h_{\rm rf}^-(t'), \tag{2.6}$$

where the retarded spin-flip propagator $F_R(t-t')$ is defined by

$$F_R(t-t')=i\theta(t-t')\langle[S^-(t),S^+(t')]\rangle, \tag{2.7}$$

where $\theta(t)=1$ if $t>0$, and vanishes otherwise. Fourier transforming (2.6) according to the following convention, for any arbitrary function $A(t)$:

$$A(t)=\frac{1}{2\pi}\int_{-\infty}^{\infty}d\omega\, e^{-i\omega t}A(\omega); \tag{2.8}$$

we define a transverse susceptibility function $\chi^-(\omega)$ by the equation

$$\gamma^{-1}\chi^-(\omega)h_{\rm rf}^-(\omega)=\langle S^-(\omega)\rangle_{\rm ext} =\langle S^-(\omega)\rangle+\tfrac{1}{2}\gamma F_R(\omega)h_{\rm rf}^-(\omega). \quad (2.9)$$

It is now necessary to introduce the irreversible relaxation of the spin system to the lattice. We write, as a generalization of (2.1),

$$i(d/dt)S^\alpha(t)=[S^\alpha(t),\mathcal{H}] -i\Delta^\alpha\{S^\alpha(t)-\gamma^{-1}\chi_0 h^\alpha(t)\}, \quad (2.10)$$

where χ_0 is the usual static susceptibility, Δ^α is an inverse relaxation time for the α component of spin, and $h^\alpha(t)$ is the component of *total* external magnetic field (i.e., both static and rf) along that direction. In future we do not distinguish between the various Δ^α; in Bloch's notation this is equivalent to taking $T_1=T_2=1/\Delta$, which for the purposes of the following section, is certainly legitimate. Equation (2.10) forces the spin to relax to an equilibrium value (and direction) appropriate to the instantaneous local field.

In (2.10), $\mathcal{H}$ includes *all* the other interaction terms in the Hamiltonian [except, of course, the rf driving field whose lossless dynamic driving effect has already been included in (2.6) and (2.9)], which describes the lossless dynamics of the spin system. We believe this separation to be a physical one, and we find it difficult to imagine the spin system relaxing in a different manner to the lattice. Similar arguments, though in a quite different context, have been advanced by Wangsness.[3] The integration of (2.10) will then yield explicitly the "inhomogeneous" driving term $\langle S^\alpha(t)\rangle$ in the Kubo equations (2.6) and (2.9). Such terms are usually absent from these equations. In fact, when we consider what happens should we neglect the driving term in the relaxation part of (2.10), so that relaxation occurs to the direction of the static magnetic field, viz.,

$$i(d/dt)S^\alpha(t)=[S^\alpha(t),\mathcal{H}] -i\Delta\{S^\alpha(t)-\gamma^{-1}\chi_0 h_0\delta_{\alpha,3}\}, \quad (2.11)$$

we would find that indeed this "extra" term would vanish. This is demonstrated below.

We now assume that the dynamics of the non-interacting isolated moment are described by the Zeeman Hamiltonian $\mathcal{H}_0=\omega_0 S^z$, where $\omega_0=-\gamma h_0$. We first solve these equations for the transverse component of spin $S^-(t)$, $(S^\pm=S^x\pm iS^y)$, given by the driving Eq. (2.10). After canonically averaging this equation and taking the Fourier transform we find

$$\langle S^-(\omega')\rangle=-\frac{i\Delta\gamma^{-1}\chi_0 h_{\rm rf}^-(\omega')}{\omega_0-i\Delta-\omega'}. \quad (2.12)$$

We assume an adiabatic "switching on" of the driving field,

$$h_{\rm rf}^\pm(t)=\lim_{s\to 0^+}e^{-s|t|}h_1 e^{\pm i\omega t}, \quad (2.13)$$

which has the transform

$$h_{\rm rf}^\pm(\omega')=2\pi\delta(\omega'\pm\omega)h_1. \quad (2.14)$$

Using (2.14) in (2.12) and taking the inverse Fourier transform defined in (2.8) we obtain for the "local field relaxation" part of $\langle S^-(t)\rangle$:

$$\langle S^-(t)\rangle=-\frac{i\Delta\gamma^{-1}\chi_0 h_1 e^{-i\omega t}}{\omega_0-i\Delta-\omega}. \quad (2.15)$$

It is easily shown that, for an instantaneous applied perturbation, $h_{\rm rf}^\pm(t)=\theta(t)h_1 e^{\pm i\omega t}$, one obtains a result identical with (2.15) for times long compared to $1/\Delta$ (i.e., when transients have been allowed to die away).

Now, had we not insisted on relaxation to the local field, then we should have integrated (2.11) instead of (2.10). This equation has the averaged solution

$$\langle S^-(t)\rangle=\langle S^-(0)\rangle e^{-i\omega_0 t}e^{-\Delta t}. \quad (2.16)$$

Thus, even if for some reason $\langle S^-(0)\rangle$ were nonzero, after a time $t\gg 1/\Delta$, $\langle S^-(t)\rangle$ would vanish, and $\langle S^-(t)\rangle_{\rm ext}$ would be given by the usual result

$$\chi^-(\omega)=\tfrac{1}{2}\gamma^2 F_R(\omega). \quad (2.17)$$

Thus, we see that the requirement of relaxation to the instantaneous local field introduces into the usual Kubo expression for $\langle S^-(t)\rangle_{\rm ext}$ an additional "inhomogeneous" term. This contribution results from an extra transverse imaginary driving field made necessary by the equilibrium condition for lattice relaxation, i.e., the factor $i\Delta\gamma^{-1}\chi_0 h_{\rm rf}(t)$ in (2.10). It vanishes when the damping Δ vanishes, so that $\langle S^-(t)\rangle$ in (2.6) thereby vanishes and one is left with the usual Kubo formula. It is interesting to note that, as $\Delta\to\infty$ this extra term becomes real [see (2.15)] and one obtains a constant value for the length of the transverse magnetization, parallel to the driving field and always in phase with it. The second term in (2.6), as we show below, vanishes in this limit. This is what one would expect, of course, for the transverse magnetization in this limit, since the longitudinal magnetization has been "decoupled" from it by the assumption of instantaneous relaxation.

To complete this section, we calculate the usual Kubo term in (2.6). The equation of motion defined by (2.7) for $F_R(t)$ is simply

$$i(\partial/\partial t)F_R(t)=2\delta(t)\langle S^z\rangle +i\theta(t)\langle[i(\partial/\partial t)S^-(t),S^+]\rangle. \quad (2.18)$$

We are only considering the *linear* response of the spin system, so that we can use either (2.10) or (2.11) to compute $\dot{S}^-(t)$ in (2.18); that is, we can ignore that part of the relaxation term containing $h_{\rm rf}^-(t)$.

We have then,

$$(i(\partial/\partial t)-i\Delta)F_R(t)=2\delta(t)\langle S^z\rangle +i\theta(t)\langle[[S^-(t),\mathcal{H}_0],S^+]\rangle, \quad (2.19)$$

where again $\mathcal{H}_0=\omega_0 S^z$. The Fourier-transformed solu-

tion is simply

$$F_R(\omega) = -2\langle S^z\rangle(\omega_0 - i\Delta - \omega)^{-1}, \tag{2.20}$$

which is, as mentioned earlier, just the usual Lorentzian form for the propagator. Using (2.20), and (2.12) in (2.9) we obtain for the over-all spin susceptibility,

$$\chi^-(\omega) = -\frac{\gamma^2\langle S^z\rangle}{\omega_0 - i\Delta - \omega}\left(1 - i\frac{\Delta}{\omega_0}\right)$$
$$= \left(\frac{\omega_0 - i\Delta}{\omega_0 - i\Delta - \omega}\right)\chi_0. \tag{2.21}$$

In the (static) limit of zero driving frequency ω, this reduces immediately to χ_0, as stated in the Introduction. Had we not demanded relaxation to the instantaneous local field, i.e., had we used (2.16) so that the last term in the numerator of (2.21) would not have been present, we would have obtained the simple Lorentzian for $\chi^-(\omega)$, namely

$$\chi^-(\omega) = (\omega_0/\omega_0 - i\Delta - \omega)\chi_0. \tag{2.22}$$

It should be noted in comparing (2.21) and (2.22) that, in the limit of infinite damping, the Lorentzian form (2.22) vanishes, whereas the modified Lorentzian (2.21) reduces to the static limit, for all frequencies. Eq. (2.21) reflects the decoupling of the transverse spin component from the longitudinal component for very rapid relaxation to the instantaneous field. Moreover, in the zero-frequency ($\omega=0$) limit, $\chi^-(0)\neq\chi_0$ for the Lorentzian, whereas this is the correct limit for (2.21).

Thus, this section exhibits our central point. The *propagator* for the spin susceptibility in the presence of instantaneous local field damping does have the usual Lorentzian form, but to it must be added an inhomogeneous (complex) term arising from the rf driving field. The combination yields a distinctly non-Lorentzian form for the *susceptibility* itself. In the absence of the assumption of relaxation towards the instantaneous local field, such an additional term is not present, and the susceptibility is directly proportional to the propagator (Lorentzian form), a result we believe to be incorrect.

3. LOCALIZED CONDUCTION-ELECTRON EXCHANGE MODEL

We are now able, using the simple formalism developed in Sec. 2, to compute the self-consistent coupled local conduction-electron magnetic susceptibility in the presence of lattice damping of both systems. We formulate the solution in powers of J, the exchange coupling constant, and, in this paper, work to only first order in J.[7] We, thus, consider a system of conduction electrons in an N-point lattice interacting with a finite number n ($=cN$) of randomly situated localized magnetic impurities located at lattice sites R_j. Microscopically, the combined system is described by the Hamiltonian $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_1$, where

$$\mathcal{H}_0 = \sum_j \omega_s S_j^z + \tfrac{1}{2}\omega_e\sigma_0^z + \sum_{\mathbf{p},\lambda}(\epsilon_\mathbf{p} - \epsilon_F)a_{\mathbf{p},\lambda}^\dagger a_{\mathbf{p}\lambda} \tag{3.1}$$

and

$$\mathcal{H}_1 = -\frac{J}{N}\sum_\mathbf{q}\sum_j e^{-i\mathbf{q}\cdot\mathbf{R}_j}\mathbf{S}_j\cdot\boldsymbol{\sigma}_\mathbf{q}. \tag{3.2}$$

By construction, we are limiting ourselves to a δ-function range of the exchange coupling. Though this restrictive form is not necessary at this stage, it turns out later that, in order to effect the necessary decoupling of the Green's functions, an equivalent assumption would have to be made. In Eqs. (3.1) and (3.2) σ^α are the Pauli matrices and

$$\sigma_\mathbf{q}^\alpha = \sum_{\mathbf{p},\lambda\lambda'} a_{\mathbf{p}+\mathbf{q},\lambda}^\dagger \sigma_{\lambda\lambda'}^\alpha a_{\mathbf{p},\lambda'}. \tag{3.3}$$

It is straightforward to verify the commutation relations [compare with (2.2)]:

$$[\sigma_\mathbf{q}^\alpha, \sigma_{\mathbf{q}'}^\beta] = 2i\sum_\gamma \epsilon_{\alpha\beta\gamma}\sigma_{\mathbf{q}+\mathbf{q}'}^\gamma.$$

We define, for convenience, an average localized spin moment per atom $\mathbf{M}$ and an average conduction-electron moment $\mathbf{m}$ as, respectively,

$$M^\alpha = \frac{g_s}{N}\sum_j S_j^\alpha,$$
$$m^\alpha = (g_e/2N)\sigma_0^\alpha, \tag{3.4}$$

where for simplicity we have set $\mu_B=1$. For notational convenience we also define the canonical averages

$$R = \langle S_j^z\rangle,$$
$$\zeta = (2N)^{-1}\langle\sigma_0^z\rangle, \tag{3.5}$$

where

$$\omega_s = -g_s h_0, \quad \omega_e = -g_e h_0.$$

We must now extract the equivalent of (2.15) for the coupled case treated in this section. That is, we must calculate the "inhomogeneous" term introduced by the requirement of relaxation towards the instantaneous local field for the coupled spin systems. The relaxation equations analogous to (2.10) are

$$i(\partial/\partial t)M^\alpha = [M^\alpha, \mathcal{H}] - i\Delta_s\{M^\alpha - \chi_s(h^\alpha + \lambda m^\alpha)\}, \tag{3.6}$$

$$i(\partial/\partial t)m^\alpha = [m^\alpha, \mathcal{H}] - i\Delta_e\{m^\alpha - \chi_e(h^\alpha + \lambda M^\alpha)\}, \tag{3.7}$$

where Δ_s^{-1} and Δ_e^{-1} are, respectively, the impurity spin-lattice and electron spin-lattice relaxation times and χ_s and χ_e are the appropriate static susceptibilities. The coupling constant λ appearing in (3.6) is easily seen to be equal to

$$\lambda = 2J(g_e g_s)^{-1} \tag{3.8}$$

from the definition of the exchange coupling (3.2).

Had we included in the Hamiltonian $\mathcal{H}$ in (3.7) the rf driving term as well as converting $\mathcal{H}$ into an effective molecular field, the canonical averages of these equations would be formally identical to those of Peter *et al.*[1] It is our purpose here, however, to formulate the problem in such a way that it will be possible to go consistently beyond first order in J, and thus to extend the analysis beyond that of the molecular field.[7] In addition, by isolating the external driving term as we have done in Sec. 2, it is possible to derive an acceptable form for the propagators (in the sense of the sum rules of Sec. 4) and yet still obtain a correct physical result for the dynamic susceptibility.

It is necessary to first calculate the static z-component averages of the two-system magnetizations to first order in J (or λ). From (3.6)

$$[(\partial/\partial t)+\Delta_s]\langle M^z(t)\rangle=\Delta_s\chi_s\{h_0+\lambda\langle m^z(t)\rangle\}, \quad (3.9)$$

but to $O(J)$ [Eqs. (3.4), (3.5)], $\langle m^z(t)\rangle=g_e\zeta$; whence, in steady state ($t\gg\Delta^{-1}$),

$$g_s\langle M^z\rangle=-\chi_s(\omega_s-2J\zeta). \quad (3.10)$$

Similarly, from (3.7),

$$g_e\langle m^z\rangle=-\chi_e(\omega_e-2cJR). \quad (3.11)$$

These results will be used in our subsequent analysis. We now evaluate the commutator in (3.6) and canonically average. We find

$$[i(\partial/\partial t)-\omega_s+i\Delta_s]\langle M^-(t)\rangle=i\Delta_s\{h_{rf}^-(t)+\lambda\langle m^-(t)\rangle\}-\frac{g_sJ}{N^2}\sum_{\mathbf{q}j}e^{-i\mathbf{q}\cdot\mathbf{R}_j}\{\langle\sigma_{\mathbf{q}}^zS_j^-\rangle-\langle\sigma_{\mathbf{q}}^-S_j^z\rangle\}. \quad (3.12)$$

The molecular-field result is recovered by a simple decoupling, viz.,

$$\begin{aligned}\langle\sigma_{\mathbf{q}}^zS_j^-\rangle&\approx\langle\sigma_{\mathbf{q}}^z\rangle\langle S_j^-\rangle,\\ \langle\sigma_{\mathbf{q}}^-S_j^z\rangle&\approx\langle\sigma_{\mathbf{q}}^-\rangle\langle S_j^z\rangle.\end{aligned} \quad (3.13)$$

It should be pointed out that terms in (3.13) like $\langle S_j^-(t)\rangle$ are here nonzero because of the presence of the driving term in the relaxation part of the equations of motion. Because the impurities are distributed at random, the sum over j in (3.12) vanishes unless $\mathbf{q}=0$ (alternatively, since the driving field is assumed spatially uniform, $\langle\sigma_{\mathbf{q}}^{\alpha}\rangle$ vanishes unless $\mathbf{q}=0$). Hence, performing this separation, Fourier transforming (3.12) and inserting (3.10) we obtain

$$\begin{aligned}(\omega-\omega_s+2J\zeta+i\Delta_s)\langle M^-(\omega)\rangle&=i\Delta_s\chi_sh_{rf}^-(\omega)\\&+(g_s/g_e)2cJR[1-(i\Delta_s/(\omega_s-2J\zeta))]\langle m^-(\omega)\rangle. \quad (3.14)\end{aligned}$$

An entirely similar analysis for the conduction-electron magnetization yields

$$\begin{aligned}(\omega-\omega_e+2cJR+i\Delta_e)\langle m^-(\omega)\rangle&=i\Delta_e\chi_eh_{rf}^-(\omega)\\&+(g_e/g_s)2J\zeta[1-(i\Delta_e/(\omega_e-2cJR))]\langle M^-(\omega)\rangle. \quad (3.15)\end{aligned}$$

Note that these two coupled equations have imaginary inhomogeneous terms which are entirely equivalent to those discussed in Sec. 2. These equations have the solutions

$$\langle M^-(\omega)\rangle=-\frac{ih_{rf}^-(\omega)\{\Delta_s\chi_s(\omega_e-2cJR-i\Delta_e-\omega)-\Delta_e\chi_e(g_s/g_e)2cJ\tilde{R}\}}{\{(\omega_s-2J\zeta-i\Delta_s-\omega)(\omega_e-2cJR-i\Delta_e-\omega)-2J\tilde{\zeta}2cJ\tilde{R}\}} \quad (3.16)$$

and

$$\langle m^-(\omega)\rangle=-\frac{ih_{rf}^-(\omega)\{\Delta_e\chi_e(\omega_s-2J\zeta-i\Delta_s-\omega)-\Delta_s\chi_s(g_e/g_s)2J\tilde{\zeta}\}}{\{(\omega_e-2cJR-i\Delta_e-\omega)(\omega_s-2J\zeta-i\Delta_s-\omega)-2cJ\tilde{R}2J\tilde{\zeta}\}}, \quad (3.17)$$

and where

$$\tilde{R}=R\left[1-\frac{i\Delta_s}{\omega_s-2J\zeta}\right], \quad \tilde{\zeta}=\zeta\left[1-\frac{i\Delta_e}{\omega_e-2cJR}\right]. \quad (3.18)$$

Note that Eqs. (3.16) and (3.17) reduce immediately to (2.12) when $J=0$. Thus, we have now found the "inhomogeneous" terms in our calculation of the response of the coupled systems, introduced by the requirement of instantaneous local field relaxation. Next, it is necessary to treat the second (propagator) part of the Kubo equation, exactly as we did in Sec. 2. The analogous driving Hamiltonian to that of Sec. 2, Eq. (2.5) for the coupled system, is

$$\mathcal{H}_{ext}(t)=-\tfrac{1}{2}N\{h_{rf}^-(t)(M^++m^+)+\text{H.c.}\}. \quad (3.19)$$

This will result in four retarded propagators since there will now be two equations analogous to (2.5), one for $M^-(t)$ and one for $m^-(t)$. We define these propagators using a modified form of the Zubarev[8] notation. Thus for any two operators (in the Heisenberg picture) A,B we define

$$G_R^{AB}(t)=i\theta(t)\langle[A(t),B(0)]\rangle\equiv\langle\langle A(t);B\rangle\rangle. \quad (3.20)$$

Then the four propagators we need are

$$\begin{aligned}F_R(t)&=\sum_{ij}\langle\langle S_i^-(t);S_j^+\rangle\rangle, \quad K_R(t)=\tfrac{1}{4}\langle\langle\sigma_0^-(t);\sigma_0^+\rangle\rangle,\\ B_R(t)&=\tfrac{1}{2}\sum_i\langle\langle S_i^-(t);\sigma_0^+\rangle\rangle, \quad \tilde{B}_R(t)=\tfrac{1}{2}\sum_j\langle\langle\sigma_0^-(t);S_j^+\rangle\rangle.\end{aligned} \quad (3.21)$$

[8] D. N. Zubarev, Usp. Fiz. Nauk. **71**, 71 (1960) [English transl.: Soviet Phys.—Usp. **3**, 320 (1960)].

Using (3.19) and the fundamental relation (2.3) we can find expressions for the driven magnetizations in terms of the propagators (3.21). After Fourier transforming, we find

$$\langle M^-(\omega)\rangle_{\rm ext}=\langle M^-(\omega)\rangle + (2N)^{-1}\{g_s^2F_R(\omega)+g_sg_eB_R(\omega)\}h_{\rm rf}^-(\omega) \quad (3.22)$$

and

$$\langle m^-(\omega)\rangle_{\rm ext}=\langle m^-(\omega)\rangle + (2N)^{-1}\{g_e^2K_R(\omega)+g_eg_s\tilde{B}_R(\omega)\}h_{\rm rf}^-(\omega). \quad (3.23)$$

The equations of motion for these four propagators are easily derived in a manner identical to that which led to (2.18). Again, we omit the relaxation term proportional to $h_{\rm rf}^-(t)$ since we are constructing only a linear theory [see the discussion following (2.18)]. Thus, for the local moment propagator $F_R(t)$ we find

$$\left(i\frac{\partial}{\partial t}-\omega_s+i\Delta_s\right)F_R(t)=2nR\delta(t)+i\Delta_s\frac{g_e}{g_s}\lambda\chi_s\tilde{B}_R(t) -\frac{J}{N}\sum_{jl\mathbf{q}}e^{-i\mathbf{q}\cdot\mathbf{R}_l}\{\langle\langle\sigma_{\mathbf{q}}{}^zS_l^-(t);S_j^+\rangle\rangle -\langle\langle\sigma_{\mathbf{q}}{}^-S_l^z(t);S_j^+\rangle\rangle\}. \quad (3.24)$$

We see that this propagator equation couples $\tilde{B}(t)$ to $F(t)$ and introduces the two new Green's functions in the curly brackets. We treat these latter terms in this paper to lowest order (since they are multiplied by the coupling constant J) and decouple them as follows:

$$\langle\langle\sigma_{\mathbf{q}}{}^zS_l^-(t);S_j^+\rangle\rangle\approx\langle\sigma_{\mathbf{q}}{}^z\rangle\langle\langle S_l^-(t);S_j^+\rangle\rangle. \quad (3.25)$$

On Fourier transforming we have

$$(\omega-\omega_s+2J\zeta+i\Delta_s)F_R(\omega)=2nR+2cJ\tilde{R}\tilde{B}_R(\omega). \quad (3.26)$$

The exact equation of motion for the propagator $\tilde{B}(\omega)$ is

$$\left(i\frac{\partial}{\partial t}-\omega_e-i\Delta_e\right)\tilde{B}_R(t)=i\Delta_e\frac{g_s}{g_e}\lambda\chi_eF_R(t) -\frac{J}{N}\sum_{jl\mathbf{q}}e^{-i\mathbf{q}\cdot\mathbf{R}_l}\{\langle\langle S_l{}^z\sigma_{\mathbf{q}}{}^-(t);S_j^+\rangle\rangle -\langle\langle\sigma_{\mathbf{q}}{}^zS_l^-(t);S_j^+\rangle\rangle\}. \quad (3.27)$$

The same higher-order propagators appear in this expression as in that for $F(t)$. We decouple in the same manner to obtain, after Fourier transforming,

$$(\omega-\omega_e+2cJR+i\Delta_e)\tilde{B}_R(\omega)=2J\tilde{\zeta}F_R(\omega). \quad (3.28)$$

Thus, inserting (3.28) into (3.26), we obtain the localized spin propagator

$$F_R(\omega)=-\frac{2nR(\omega_e-2cJR-i\Delta_e-\omega)}{\{(\omega_s-2J\zeta-i\Delta_s-\omega)(\omega_e-2cJR-i\Delta_e-\omega)-2J\tilde{\zeta}2cJ\tilde{R}\}}. \quad (3.29)$$

A similar procedure completes the quartet; we find

$$(\omega-\omega_s+2J\zeta+i\Delta_s)B_R(\omega)=2cJ\tilde{R}K_R(\omega) \quad (3.30)$$

and

$$(\omega-\omega_e-2cJR-i\Delta_e)K_R(\omega)=2N\zeta+2J\tilde{\zeta}B_R(\omega); \quad (3.31)$$

so that, for example, the conduction-electron spin propagator $K(\omega)$ equals

$$K_R(\omega)=-\frac{2N\zeta(\omega_s-2J\zeta-i\Delta_s-\omega)}{\{(\omega_e-2cJR-i\Delta_e-\omega)(\omega_s-2J\zeta-i\Delta_s-\omega)-2J\tilde{\zeta}2cJ\tilde{R}\}}. \quad (3.32)$$

By definition $\langle M^-(\omega)\rangle_{\rm ext}=\chi_s^-(\omega)h_{\rm rf}^-(\omega)$, so that by use of (3.22), the relaxation driven term (3.16), and our expressions for $F(\omega)$ in Eq. (3.29), and $\tilde{B}(\omega)$ from Eq. (3.28), we can evaluate $\chi_s^-(\omega)$. We find

$$\chi_s^-(\omega)=\frac{-g_s{}^2c\tilde{R}(\omega_e-2cJR-i\Delta_e-\omega)+g_eg_s\tilde{\zeta}2cJ\tilde{R}}{\{(\omega_s-2J\zeta-i\Delta_s-\omega)(\omega_e-2cJR-i\Delta_e-\omega)-2J\tilde{\zeta}2cJ\tilde{R}\}}. \quad (3.33)$$

Similar arguments for $\langle m^-(\omega)\rangle_{\rm ext}$ yield

$$\chi_e^-(\omega)=\frac{-g_e{}^2\tilde{\zeta}(\omega_s-2J\zeta-i\Delta_s-\omega)+g_eg_s2J\tilde{\zeta}c\tilde{R}}{\{(\omega_s-2J\zeta-i\Delta_s-\omega)(\omega_e-2cJR-i\Delta_e-\omega)-2J\tilde{\zeta}2cJ\tilde{R}\}}. \quad (3.34)$$

Writing $\omega_e'=\omega_e-2cJR-i\Delta_e$, $\omega_s'=\omega_s-2J\zeta-i\Delta_s$ to conform to the notation of Peter *et al.*[1] we find

$$\chi_s^-(\omega)=\frac{\omega_s'(\omega_e'-\omega+\lambda\chi_e\omega_e')\chi_s}{\{(\omega_e'-\omega)(\omega_s'-\omega)-\lambda\chi_e\omega_e'\lambda\chi_s\omega_s'\}} \tag{3.35}$$

and

$$\chi_e^-(\omega)=\frac{\omega_e'(\omega_s'-\omega+\lambda\chi_s\omega_s')\chi_e}{\{(\omega_e'-\omega)(\omega_s'-\omega)-\lambda\chi_e\omega_e'\lambda\chi_s\omega_s'\}}. \tag{3.36}$$

These results are identical to those of Peter *et al.*[1] and demonstrate the connection between the modified Kubo approach [to $O(J)$] and the molecular-field result. The application of these equations to various physical problems was carried out extensively by Peter *et al.*[1] and we shall not go into such questions here. Before finishing this section, it should be noted, as was done at the end of Sec. 2, that the zero-frequency (static) limit of (3.35) and (3.36),

$$\begin{aligned}&\chi_s^-(0)=(1+\lambda\chi_e)\chi_s/(1-\lambda^2\chi_e\chi_s)\\ \text{and}\qquad&\\ &\chi_e^-(0)=(1+\lambda\chi_s)\chi_e/(1-\lambda^2\chi_e\chi_s)\end{aligned} \tag{3.37}$$

are identical to the expression for the longitudinal susceptibility easily derivable from (3.10) and (3.11). Hence, our solutions still obey the physical requirement of isotropy in the static limit.

4. DISCUSSION

We shall not consider in detail the results we have obtained here using the concept of instantaneous relaxation to the local field. We refer the reader to the work of Wangsness[3] who gave a detailed discussion of $\chi^-(\omega)$ for the case of isolated spins, comparing expressions equivalent to (2.21) and (2.22). The molecular-field result for the coupled system (3.37) has been extensively studied by Peter *et al.*[1] However, we reiterate that our results for the isolated spin and coupled spin systems both reduce to the longitudinal result in the static ($\omega=0$) limit, viz.,

$$\chi^-(0)=\chi^{\alpha\alpha}(0)=\chi^z. \tag{4.1}$$

The damped Lorentzian (2.22) fails to reduce to this limit and, in fact, vanishes for $\Delta\gg\omega_0$. In order to contrast these differences, we display the simple result for the transverse susceptibility function for an undamped freely precessing spin

$$\chi^-(\omega)=\lim_{s\to 0^+}\frac{\omega_0}{\omega_0-\omega-is}\chi_0. \tag{4.2}$$

Clearly, when the damping vanishes, the Lorentzian (2.22) and the modified Lorentzian (2.21) are indistinguishable from (4.2).

The final point of this section is concerned with the actual form of the spin propagators discussed in Secs. 2 and 3. Using a Lehman spectral representation for the Fourier transform of a general "one-particle" Green's function, as defined by (3.20), one can always prove the following sum rule, valid for $B=A^\dagger$:

$$\frac{1}{\pi}\int_{-\infty}^{\infty}d\omega\ \mathrm{Im}G_R{}^{AB}(\omega)=\langle[A,B]\rangle. \tag{4.3}$$

It turns out that all three forms for $\chi^-(\omega)$, namely, (4.2), (2.21), and (2.22) satisfy casuality, i.e., the Kramers-Kronig dispersion rule,

$$\mathrm{Re}G_R{}^{AB}(\omega)=\frac{\mathcal{P}}{\pi}\int_{-\infty}^{\infty}\frac{d\omega'}{\omega'-\omega}\mathrm{Im}G_R{}^{AB}(\omega'). \tag{4.4}$$

However, only (4.2) and (2.22) satisfy (4.3). Hence only the simple Lorentzian form for $\chi^-(\omega)$ satisfies the general requirement of (4.3). Thus, since we know that in the presence of damping only (2.2) gives the physically correct result for $\chi^-(\omega)$, then this cannot be represented by a simple propagator. We have seen in Secs. 2 and 3 that indeed the propagator part of $\chi^-(\omega)$ for relaxation to the instantaneous local field is of the form (2.22), so that the derived Green's function does, in fact, satisfy the sum rule (4.3). It is only when we add the necessary inhomogeneous term [e.g., (2.15) or (3.16)] to the propagator that the form for $\chi^-(\omega)$ is altered to (2.21). Hence, we see that it is not possible to represent relaxation to the instantaneous local field by simply altering the usual Lorentzian form of the propagator as was done by Giovannini *et al.*[2] Instead, the equations of motion dictate that, to the propagator contribution to $\chi^-(\omega)$, one must add an additional driving term which, in fact, cannot be put into correct propagator form if one describes the lattice damping phenomenologically. Thus we believe our present approach is essentially correct and is powerful enough to be useful in several complicated systems.

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