Dynamics of Localized Moments in Metals. II. Second-Order Exchange Effects

R. ORBACH* AND H. J. SPENCER†
Department of Physics, Imperial College, London, England
(Received 19 August 1968)

We have extended our previous microscopic treatment of the dynamic transverse susceptibility for a random array of localized spins in a metal to include terms of second order in the exchange coupling constant J. Lattice relaxation of the localized and conduction electrons is included, as before, in such a way as to ensure relaxation to the instantaneous local field. The results, in the limit of equal conduction-electron and localized-spin g values and no lattice damping, reduce to the correct ("bottlenecked") limit. The linewidth for frequencies close to the localized-spin resonance frequency agrees with previous calculations. Bottlenecking of both the longitudinal (frequency-modulation) (T_2') and transverse (spin-flip) (T_1') parts of the localized-spin resonance linewidth is demonstrated for equal g values and no lattice relaxation. Similarly, the linewidth for frequencies close to the conduction-electron resonance frequency exhibits both T_2'- and T_1'-type terms, and again bottleneck effects are present. The results are compared with previous macroscopic treatments. It is demonstrated that it is unnecessary to introduce detailed balance conditions *per se* in the microscopic theory. The relation between the conduction-electron-hole relaxation width and the one-electron width calculated by Overhauser is examined in an appendix.

I. INTRODUCTION

WE extend here the work of a previous paper[1] (to be referred to as I) to include the effects of localized-electron–conduction-electron exchange up to second order in the exchange coupling constant J. We include, as before, lattice relaxation of both spin species in such a way that relaxation occurs to the instantaneous value of the local field. The extension to $O(J^2)$ is important for, as Hasegawa[2] demonstrated, linewidths due to exchange effects enter only to this order. Hasegawa's treatment was, however, macroscopic and introduced a T_1-like broadening for both spin species (his T_{ds} and T_{sd}). We have demonstrated in an earlier letter[3] that in general the localized-spin linewidth was the sum of two terms, longitudinal (frequency modulation) (T_2') and transverse (spin-flip) (T_1') broadening, in the usual resonance sense. Further, the spin resonance linewidth $1/T_2=1/T_2'+1/T_1'$ equalled $1/T_1$ $(=1/T_{ds})$ only on the energy shell ($\omega=\omega_s$) and in the limit of isotropic g values and high temperatures ($kT\gg\hbar\omega_s$, the localized-spin resonance frequency). Because of the different character of T_2' and T_1', and because Hasegawa did not treat the lattice relaxation correctly (i.e., he did not require instantaneous local field relaxation) we have developed a microscopic theory which treats these effects properly. Also, very recently, Cottet *et al.*[4] have treated the macroscopic problem by forcing *both* the lattice *and* exchange relaxation terms to relax to the instantaneous local field. We believe the latter to be artificial, though to $O(J^2)$ this assumption has no effect on the transverse susceptibility, a limitation of our present treatment. Our treatment takes into account the dynamical effects of exchange in a microscopic manner, and as we shall show exhibits the correct limiting behavior without the necessity of explicit introduction of a detailed balance condition, a feature common to both the treatment of Hasegawa and Cottet *et al.*

It should be mentioned here that, in an interesting letter by Langreth *et al.*,[5] macroscopic equations identical to those of Cottet *et al.*[4] are derived using "the method of Kadanoff and Baym." Unfortunately, however, no details of the derivation are given in this letter. We are somewhat surprised by this result, because, as will be demonstrated in Sec. III, the quantities T_{ds} and T_{sd} are only approximate and imply energy shell-restricted self energies. We feel that our approach is worthwhile on a number of grounds. First, the dynamical effects of exchange are included in a straightforward way in our method, with no necessity for forcing the introduction of local field relaxation into these terms as proposed by Cottet *et al.* Second, the second-order exchange terms are *not* in general equivalent to T_1-like relaxation terms, as assumed by the above authors. Indeed, we demonstrate explicitly a separation into longitudinal (frequency modulation) and transverse (spin-flip) components and show that both (initially surprising for us) are bottlenecked to an equal degree under the appropriate circumstances. Thus, our approach displays the microscopic dynamical features of second-order exchange effects in a manner which we feel justifies the lengths to which we have gone in the present paper. In addition, quite apart from the specific results obtained here, some innovations in the decoupling method (e.g., symmetric decoupling, and com-

* N.S.F. Senior Postdoctoral Fellow, on leave of absence from the University of California, Los Angeles. Supported in part by the Office of Naval Research, Contract No. 233(88) and the National Science Foundation.
† Science Research Council (G.B.) Postdoctoral Fellow.
[1] H. J. Spencer and R. Orbach, preceding paper, Phys. Rev. **179**, 683 (1969).
[2] H. Hasegawa, Progr. Theoret. Phys. (Kyoto) **21**, 483 (1959).
[3] R. Orbach and H. J. Spencer, Phys. Letters **26A**, 457 (1968).
[4] H. Cottet, P. Donze, J. Dupraz, B. Giovannini, and M. Peter, Z. Angew. Phys. **24**, 249 (1968).
[5] D. C. Langreth, D. L. Cowan, and J. W. Wilkins, Solid State Commun. **6**, 131 (1968).

parison of the second-order equations for the higher-order propagators with the first-order equations) are presented, as well as a comparison between the one and two conduction-electron linewidths.

Our results demonstrate that, for equal g values and slow lattice relaxation, a "bottleneck" does in fact obtain. This bottleneck is interesting, for it appears to apply to *both* the T_2'- and T_1'-type terms with equal force. This result was unexpected, though the experiments of Gossard *et al.*[6] do demonstrate that in practice such must be the case. Thus, our results in a certain sense do demonstrate the essential validity of the Hasegawa[2] and Cottet *et al.*[4] approach, insofar as their assumptions regarding the bottlenecking of exchange relaxation are concerned. The similarities between the conclusions of their papers and of the present work will be described in detail in Sec. IV.

The basis of our approach is the double-time Green's function method of Zubarev,[7] as in I. In I only the lowest- (first-) order terms in J were retained. We were able to demonstrate that the effect of the requirement of instantaneous local field lattice relaxation was to introduce an imaginary inhomogeneous "driving" field term into the Kubo[8] response equations. In this paper, we continue this approach, but work to higher order in J before invoking the decoupling procedure. We have had to be very careful about the decoupling, for higher-order terms are known to be very dangerous. For example, we found it necessary not to include higher-order Knight shifts into the relaxation terms.[9] The inclusion of such terms destroys the first-order solution found in I. In general, we have checked at each stage of decoupling that our results reduce to the first-order results of I in the limit of small J. In addition, we have found it necessary to symmetrize our Green's functions in order to effect a decoupling which leads to what we believe are physically correct results. This procedure is justified in Appendix I and appears to be essential in problems of this type, and in fact whenever high-order Green's functions are important. In this way, we obtain linewidths for localized and conduction-electron spins which agree completely with the results of Spencer[10] and Ref. 3. The conduction-electron *spin* linewidth is tricky because previous calculations (e.g., Overhauser[11]) computed only *one*-electron lifetimes. However, because the transverse *magnetization* is important in a magnetic resonance experiment, the quantity which should be calculated is a *two*-electron (electron-hole) linewidth. We find, to order J^2, that this quantity is simply the sum of the particle-hole *one*-electron widths.[12] Because of this question we have also solved for the one-particle width using the Zubarev[7] method. Our results, appropriate to a random array of impurities in a metallic lattice, agree with those of Fullenbaum and Falk,[13] and Spencer[10] in the limit of $S=\frac{1}{2}$. The work is summarized in Appendix B, and the similarity to our result for the two-electron width is pointed out in Sec. II of this paper.

Section II of this paper begins with the equations from which the higher-order Green's functions are generated. The equation of motion for these new propagators are derived in this section in the absence of lattice damping, for simplicity, but, of course, using different localized and conduction-electron g values. A decoupling procedure is applied which we believe to be correct to the second order in J. It is demonstrated that the higher-order equations reduce to those of I in the limit of small J. Spatial averaging, appropriate to a random alloy, is then carried out, and the equations for the Green's functions of primary interest are derived and solved. The complex frequency-dependent transverse susceptibility is then found from these quantities for both the localized and conduction electrons. It is shown that the total susceptibility, in the limit of equal g values, reduces immediately to the uncoupled form. For differing g values, the second-order shifts and widths are found for both resonance roots.

In Sec. III, the dynamical equations are solved in the presence of instantaneous local field lattice damping and the full transverse susceptibility for both the localized and conduction-electron spin systems is constructed. The sum of the two susceptibilities is demonstrated to again reduce to the uncoupled form in the limit of equal g values and no lattice damping. Limits are taken which exhibit the character of the localized and conduction-electron line shift and width, respectively.

Section IV contains a comparison of our results with those of Hasegawa[2] and Cottet *et al.*[4]

II. COUPLED EQUATIONS IN ABSENCE OF LATTICE DAMPING

From I, the basic equations we must treat are

$$i(\partial/\partial t)M^\alpha=[M^\alpha,\mathcal{H}] \\ -i\Delta_s\{M^\alpha-\chi_s(h^\alpha+(2J/g_eg_s)m^\alpha)\} \quad (2.1)$$

and

$$i(\partial/\partial t)m^\alpha=[m^\alpha,\mathcal{H}] \\ -i\Delta_e\{m^\alpha-\chi_e(h^\alpha+(2J/g_eg_s)M^\alpha)\}. \quad (2.2)$$

[6] A. C. Gossard, A. J. Heeger, and J. H. Wernick, J. Appl. Phys. **38**, 1251 (1967).

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

[8] R. Kubo, J. Phys. Soc. Japan **12**, 570 (1957).

[9] Such terms have, for example, marred decoupling attempts in the theory of interacting spin waves. A discussion of this question is contained in a paper by H. J. Spencer, Phys. Rev. **167**, 434 (1968).

[10] H. J. Spencer, Phys. Rev. **171**, 515 (1968); H. J. Spencer and S. Doniach, Phys. Rev. Letters **18**, 994 (1967).

[11] A. Overhauser, Phys. Rev. **89**, 689 (1953).

[12] A similar result is also found for the localized spin linewidth in a very recent paper by M. B. Walker, Phys. Rev. **176**, 432 (1968), who used the Abrikosov technique (Ref. 16) to examine the damping of the single pseudofermions propagators in this model. Dr. C. B. Duke (private communication) informs us that, to the order we are working, we could have expected nothing else.

[13] M. S. Fullenbaum and D. S. Falk, Phys. Rev. **157**, 454 (1967).

Here M^α and m^α are the α components of the magnetizations of the localized spins and the conduction electrons, respectively, and, for simplicity, $\mu_B=1$. The $\sigma_0{}^\alpha$ and $S_j{}^\alpha$ are the usual second-quantized conduction-electron spin operator ($\sigma_{\mathbf{q}=0}{}^\alpha$) and the localized spin operator at the jth site, respectively, and h^α represents the total (static and rf) external magnetic fields acting on the spin systems. The Hamiltonian $\mathcal{H}=\mathcal{H}_0+\mathcal{H}_I$, where ($\hbar=1$),

$$\mathcal{H}_0=\sum_j \omega_s S_j{}^z+\tfrac{1}{2}\omega_e\sigma_0{}^z+\sum_{\mathbf{k},\lambda}(\epsilon_{\mathbf{k}}-\epsilon_F)a_{\mathbf{k},\lambda}{}^\dagger a_{\mathbf{k},\lambda} \quad (2.3)$$

and

$$\mathcal{H}_I=-\frac{J}{N}\sum_{\mathbf{q},j} e^{-i\mathbf{q}\cdot\mathbf{R}_j}\mathbf{S}_j\cdot\boldsymbol{\sigma}_{\mathbf{q}}. \quad (2.4)$$

The "bare" Zeeman energies ω_s and ω_e equal $-g_s h_0$ and $-g_e h_0$, respectively, where the static magnetic field h_0 is taken parallel to the z direction. In Eqs. (2.1) and (2.2), Δ_s and Δ_e are the lattice relaxation rates for the localized and conduction electrons, respectively. The quantities χ_s and χ_e are here assumed to be the localized and conduction-electron static longitudinal susceptibilities, respectively, which are equal to,

$$\begin{aligned}\chi_s&=-g_s{}^2cR/(\omega_s-2J\zeta),\\ \chi_e&=-g_e{}^2\zeta/(\omega_e-2cJR).\end{aligned} \quad (2.5)$$

We have used

$$M^\alpha=(g_s/N)\sum_j S_j{}^\alpha,\quad m^\alpha=(g_e/2N)\sigma_0{}^\alpha, \quad (2.6)$$

so that

$$R=\langle S_j{}^z\rangle,\quad \zeta=(1/2N)\langle\sigma_0{}^z\rangle.$$

In (2.5), c is the fractional concentration of the impurity spins.

For the remainder of this section, we shall omit for simplicity the lattice damping terms which occur on the right-hand sides of (2.1) and (2.2). These terms will be included in full, together with the inhomogeneous imaginary driving term [e.g., the rf component of h^α in Eq. (2.1)], in Sec. III. As in I, we define the various Green's functions using the notation,

$$\begin{aligned}G_R{}^{AB}(t)&=i\theta(t)\langle[A(t),B(0)]\rangle\\ &\equiv\langle\langle A(t);B\rangle\rangle.\end{aligned} \quad (2.7)$$

We shall require the four propagators

$$\begin{aligned}F_R(t)&=\sum_{ij}\langle\langle S_i{}^-(t);S_j{}^+\rangle\rangle,\\ B_R(t)&=\tfrac{1}{2}\sum_i\langle\langle S_i{}^-(t);\sigma_0{}^+\rangle\rangle,\\ \tilde{B}_R(t)&=\tfrac{1}{2}\sum_j\langle\langle\sigma_0{}^-(t);S_j{}^+\rangle\rangle,\end{aligned} \quad (2.8)$$

and

$$K_R(t)=\tfrac{1}{4}\langle\langle\sigma_0{}^-(t);\sigma_0{}^+\rangle\rangle.$$

Using (2.1) and (2.2) as the basic equations of motion, after Fourier transforming with respect to time we find the following (exact) equations of motion for these four propagators:

$$(\omega_s-\omega)F_R(\omega)+2J\{V_R(\omega)-W_R(\omega)\}=-2cNR, \quad (2.9a)$$

$$(\omega_e-\omega)\tilde{B}_R(\omega)-2J\{V_R(\omega)-W_R(\omega)\}=0, \quad (2.9b)$$

$$(\omega_e-\omega)K_R(\omega)+2J\{M_R(\omega)-N_R(\omega)\}=-2N\zeta, \quad (2.9c)$$

$$(\omega_s-\omega)B_R(\omega)-2J\{M_R(\omega)-N_R(\omega)\}=0, \quad (2.9d)$$

where, defining,

$$\mathcal{S}_{\mathbf{k}}{}^-(\mathbf{q})=a_{\mathbf{k}+\mathbf{q}\downarrow}{}^\dagger a_{\mathbf{k}\uparrow};\quad \mathcal{S}_{\mathbf{k},\lambda}{}^z(\mathbf{q})=a_{\mathbf{k}+\mathbf{q},\lambda}{}^\dagger a_{\mathbf{k},\lambda},$$

we have,

$$\begin{aligned}V_R(\omega)&=\frac{1}{N}\sum_{j,l,\mathbf{q},\mathbf{k}} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle\langle\mathcal{S}_{\mathbf{k}}{}^-(\mathbf{q})S_l{}^z;S_j{}^+\rangle\rangle_\omega,\\ W_R(\omega)&=\frac{1}{2N}\sum_{j,l,\mathbf{q},\mathbf{k},\lambda} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\lambda\langle\langle\mathcal{S}_{\mathbf{k},\lambda}{}^z(\mathbf{q})S_l{}^-;S_j{}^+\rangle\rangle_\omega,\\ N_R(\omega)&=\frac{1}{2N}\sum_{l,\mathbf{q},\mathbf{k}} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle\langle\mathcal{S}_{\mathbf{k}}{}^-(\mathbf{q})S_l{}^z;\sigma_0{}^+\rangle\rangle_\omega,\\ M_R(\omega)&=\frac{1}{4N}\sum_{l,\mathbf{q},\mathbf{k},\lambda} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\lambda\langle\langle\mathcal{S}_{\mathbf{k},\lambda}{}^z(\mathbf{q})S_l{}^-;\sigma_0{}^+\rangle\rangle_\omega.\end{aligned} \quad (2.10)$$

Note that $V_R(\omega)$ and $N_R(\omega)$ have the same left-hand structure, and so, apart from the averaged commutator term, will have identical equations of motion. A similar result obtains for $W_R(\omega)$ and $M_R(\omega)$. From (2.5), (2.9), and (2.10) we can obtain two exact relationships between the four propagators

$$(\omega_s-\omega)F_R(\omega)+(\omega_e-\omega)\tilde{B}_R(\omega)=-2cNR \quad (2.11)$$

and

$$(\omega_s-\omega)B_R(\omega)+(\omega_e-\omega)K_R(\omega)=-2N\zeta. \quad (2.12)$$

So far, all the equations presented have been exact. The first-order results (molecular field) obtained in I can be found in two ways, the second of which will be of prime importance in this paper.

The first method is to decouple the quantities appearing in (2.10) as in I. For example,

$$V_R(\omega)\cong\frac{1}{N}\sum_{j,l,\mathbf{q},\mathbf{k}} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle S_l{}^z\rangle\langle\langle\mathcal{S}_{\mathbf{k}}{}^-(\mathbf{q});S_j{}^+\rangle\rangle_\omega. \quad (2.13)$$

We now introduce spatial averaging, appropriate to a random array of impurity spins. This method of averaging to $O(J^2)$ distinguishes our results, for example, from those of Fullenbaum and Falk[13] (who consider only a single impurity spin) but, of course, our final results for physical quantities (e.g., electron lifetimes) will be the same as theirs for quantities involving scattering from essentially a single impurity (see Appendix B). We write

$$\sum_l e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle S_l{}^z\rangle=cRN\delta_{\mathbf{q},0}. \quad (2.14)$$

Using (2.14) in (2.13) we immediately obtain,

$$V_R(\omega)\cong cR\tilde{B}_R(\omega). \quad (2.15a)$$

Similar treatments for the remainder of the quantities in (2.10) yield

$$W_R(\omega)\cong\zeta F_R(\omega), \tag{2.15b}$$

$$M_R(\omega)\cong\zeta B_R(\omega), \tag{2.15c}$$

$$N_R(\omega)\cong cRK_R(\omega). \tag{2.15d}$$

It is a simple matter to insert (2.15) into (2.9) and obtain the first-order results presented in I. The approximations given in (2.15), however, are not very useful for the purposes of this paper because they do not display the dynamical properties of the higher-order Green's functions (2.10). It is possible to write down an equivalent first-order set of equations for these quantities which reduces to (2.15) but which also displays a form appropriate to an equation of motion result. These can then be used to check that our second-order forms (to be derived) for the quantities (2.10) reduce correctly to the first-order result in the limit of small J. Such checks are mandatory when utilizing an approximate decoupling procedure. We have

$$\frac{2}{N}\sum_{l,\mathbf{k},\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle\langle \mathcal{S}_{\mathbf{k}}^-(\mathbf{q})S_l^z;\alpha\rangle\rangle_\omega$$
$$\cong(\omega_e-\omega)^{-1}\left\{\frac{cR\gamma_e}{4}+2cJR \times[cR\langle\langle\sigma_0^-;\alpha\rangle\rangle_\omega-2\zeta\sum_l\langle\langle S_l^-;\alpha\rangle\rangle_\omega]\right\} \tag{2.16a}$$

and

$$\frac{1}{N}\sum_{l,\mathbf{k},\mathbf{q},\lambda} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\lambda\langle\langle \mathcal{S}_{\mathbf{k},\lambda}{}^z(\mathbf{q})S_i^-;\alpha\rangle\rangle_\omega$$
$$\cong(\omega_s-\omega)^{-1}\{2\zeta\gamma_s-2J\zeta \times[cR\langle\langle\sigma_0^-;\alpha\rangle\rangle_\omega-2\zeta\sum_l\langle\langle S_l^-;\alpha\rangle\rangle_\omega]\}, \tag{2.16b}$$

where

$$\gamma_e=0,\qquad \gamma_s=-2R \quad \text{for} \quad \alpha=S_j^+,$$
$$\text{and} \tag{2.16c}$$
$$\gamma_e=-2N\zeta,\quad \gamma_s=0 \quad \text{for} \quad \alpha=\sigma_0^+.$$

This result is easy to prove. From (2.9) and (2.15)

$$(\omega_e-\omega)\langle\langle\sigma_0^-;\alpha\rangle\rangle_\omega \cong\tfrac{1}{4}\gamma_e+2J\{cR\langle\langle\sigma_0^-;\alpha\rangle\rangle_\omega-2\zeta\sum_l\langle\langle S_l^-;\alpha\rangle\rangle_\omega\}. \tag{2.17}$$

Inserting (2.17) into (2.16a) immediately results in

$$\frac{2}{N}\sum_{l,\mathbf{k},\mathbf{q}} e^{-i\mathbf{q}\cdot\mathbf{R}_l}\langle\langle \mathcal{S}_{\mathbf{k}}^-(\mathbf{q})S_l^z;\alpha\rangle\rangle_\omega\cong cR\langle\langle\sigma_0^-;\alpha\rangle\rangle_\omega, \tag{2.18}$$

which are exactly the results (2.15a) and (2.15d). An identical argument for (2.16b) leads to (2.15b) and (2.15c). In summary, (2.16) leads to the following set of four first-order equations to which the higher-order equations of motion for the quantities (2.10) must reduce

$$(\omega_e-\omega)V_R(\omega)\cong 2cJR\{cR\tilde{B}_R(\omega)-\zeta F_R(\omega)\}, \tag{2.19a}$$

$$(\omega_s-\omega)W_R(\omega)\cong -2cNR\zeta -2J\zeta\{cR\tilde{B}_R(\omega)-\zeta F_R(\omega)\}, \tag{2.19b}$$

$$(\omega_s-\omega)M_R(\omega)\cong -2J\zeta\{cRK_R(\omega)-\zeta B_R(\omega)\}, \tag{2.19c}$$

$$(\omega_e-\omega)N_R(\omega)\cong -2cNR\zeta +2cJR\{cRK_R(\omega)-\zeta B_R(\omega)\}. \tag{2.19d}$$

In order to display the explicit decoupling procedure which we use in the equations of motion for the higher-order propagators (2.10), we examine the Green's function $V_R(\omega)$ in some detail. The equation of motion for this quantity leads to the following exact result:

$$(\epsilon_{\mathbf{k}\uparrow}-\epsilon_{\mathbf{k}+\mathbf{q}\downarrow}-\omega)\langle\langle \mathcal{S}_{\mathbf{k}}^-(\mathbf{q})S_i^z;S_j^+\rangle\rangle_\omega=\delta_{ij}\langle \mathcal{S}_{\mathbf{k}}^-(\mathbf{q})S_i^+\rangle$$
$$+\frac{J}{2N}\sum_{\mathbf{q}'} e^{-i\mathbf{q}'\cdot\mathbf{R}_i}\langle\langle \mathcal{S}_{\mathbf{k}}^-(\mathbf{q})(S_i^+\sigma_{\mathbf{q}'}{}^- - S_i^-\sigma_{\mathbf{q}'}{}^+);S_j^+\rangle\rangle_\omega$$
$$+\frac{J}{N}\sum_{l,\mathbf{q}'} e^{-i\mathbf{q}'\cdot\mathbf{R}_l}\langle\langle S_i^zS_l^z(a_{\mathbf{k}+\mathbf{q}+\mathbf{q}'\downarrow}{}^\dagger a_{\mathbf{k}\uparrow}+a_{\mathbf{k}+\mathbf{q}\downarrow}{}^\dagger a_{\mathbf{k}-\mathbf{q}'\uparrow});S_j^+\rangle\rangle_\omega$$
$$-\frac{J}{N}\sum_{l,\mathbf{q}'} e^{-i\mathbf{q}'\cdot\mathbf{R}_l}\langle\langle S_i^zS_l^-(a_{\mathbf{k}+\mathbf{q}+\mathbf{q}'\uparrow}{}^\dagger a_{\mathbf{k}\uparrow}-a_{\mathbf{k}+\mathbf{q}\downarrow}{}^\dagger a_{\mathbf{k}-\mathbf{q}'\downarrow});S_j^+\rangle\rangle_\omega. \tag{2.20}$$

We drop the inhomogeneous (first) term on the right since we are at present ignoring magnetic field independent Kondo terms as, for example, treated by Nagaoka.[14] These enter anyway only to $O(J^3)$, and our $O(J^2)$ perturbation procedure in the presence of a field is certainly valid at high enough temperatures ($T\gg T_K$). We do, of course, obtain "Kondo-like" terms to $O(J^2)$ in the g-shift expressions[10] [see Eq. (4.5)]. The first part of the second term on the right is also dropped for similar reasons. The remaining terms are approxi-

[14] Y. Nagaoka, Phys. Rev. 138, 1112A (1965); Progr. Theoret. Phys. (Kyoto) 37, 13 (1967).

mated in the following way:

$$\langle\langle S_i^z S_l^z a_{k+q+q',\downarrow}^\dagger a_{k\uparrow}; S_j^+\rangle\rangle_\omega$$
$$\cong\{(1-\delta_{il})\langle S_i^z\rangle\langle S_l^z\rangle+\delta_{il}\langle S_i^z S_l^z\rangle\}\times\langle\langle a_{k+q+q',\downarrow}^\dagger a_{k\uparrow}; S_j^+\rangle\rangle_\omega$$
$$=\{\langle S_i^z\rangle\langle S_l^z\rangle+\delta_{il}[\langle(S_i^z)^2\rangle-\langle S_i^z\rangle^2]\}\times\langle\langle a_{k+q+q',\downarrow}^\dagger a_{k\uparrow}; S_j^+\rangle\rangle_\omega, \quad (2.21a)$$

$$\langle\langle S_i^z S_l^-(a_{k+q+q',\uparrow}^\dagger a_{k\uparrow}-a_{k+q,\downarrow}^\dagger a_{k-q',\downarrow}); S_j^+\rangle\rangle_\omega$$
$$\cong\langle S_i^z\rangle\langle(a_{k+q+q',\uparrow}^\dagger a_{k\uparrow}-a_{k+q,\downarrow}^\dagger a_{k-q',\downarrow})\rangle\times\langle\langle S_l^-; S_j^+\rangle\rangle, \quad (2.21b)$$

$$\langle\langle \mathcal{S}_k^-(\mathbf{q})S_s^-\sigma_{q',}^+; S_j^+\rangle\rangle$$
$$\cong\langle\{\mathcal{S}_k^-(\mathbf{q}),\sigma_{q'}^+\}_s\rangle\langle\langle S_i^-; S_j^+\rangle\rangle_\omega. \quad (2.21c)$$

The first two decouplings are quite standard and need not be discussed further, but (2.21c) represents an interesting feature of the decoupling problem. The subscript s represents a symmetric product. Such a term would have arisen had we initially used a symmetric combination of operators on the left-hand side of our original Green's function $V_R(\omega)$. The other quantities appearing in (2.20) would not have been changed by such a symmetrization. It is demonstrated in Appendix A that we could have formulated the entire problem in this manner, and that it makes a difference only when the operators appearing in the yet higher-order Green's function do not commute. The expectation value in (2.21c) is easily seen to equal

$$\delta_{q,-q'}\{f_{k\uparrow}{}^+ f_{k+q\downarrow}{}^- + f_{k\uparrow}{}^- f_{k+q\downarrow}{}^+\},$$

where $f^-=1-f^+$ is the usual Fermi function. Inserting these results into (2.20), spatially averaging as in (2.14), dividing by the term in parenthesis on the left, multiplying by $\exp(-i\mathbf{q}\cdot\mathbf{R}_i)$ and summing over $i,j,\mathbf{k},\mathbf{q}$, we obtain

$$V_R(\omega)\cong(\omega_s-\omega)^{-1}2cJR\{cR\tilde{B}_R(\omega)-\zeta F_R(\omega)\}$$
$$-\tfrac{1}{2}J\Xi_T(\omega)F_R(\omega)+\tfrac{1}{2}cJ\langle(\delta S_i^z)^2\rangle\Lambda_L(\omega)\tilde{B}_R(\omega), \quad (2.22a)$$

where

$$\langle(\delta S_i^z)^2\rangle=\langle(S_i^z)^2\rangle-\langle S_i^z\rangle^2,$$

$$\Xi_T(\omega)=\frac{1}{N^2}\sum_{\mathbf{k},\mathbf{q}}\frac{\{f_{k\uparrow}{}^+ f_{k+q\downarrow}{}^- + f_{k\uparrow}{}^- f_{k+q\downarrow}{}^+\}}{\epsilon_{k\uparrow}-\epsilon_{k+q\downarrow}-\omega-i\eta}, \quad (2.23)$$

and

$$\Lambda_L(\omega)=\frac{1}{N}\sum_{\mathbf{q}}\left\{\frac{1}{\epsilon_{k\uparrow}-\epsilon_{k+q\downarrow}-\omega-i\eta}+\frac{1}{\epsilon_{k+q\uparrow}-\epsilon_{k\downarrow}-\omega-i\eta}\right\}, \quad (2.24)$$

where η is a positive infinitesimal. The quantities (2.23) and (2.24) will be evaluated later in this section, after their physical meaning has become clear.

Using very similar methods, one can also obtain expressions for the other higher-order propagators defined in (2.10). We find

$$W_R(\omega)$$
$$\cong-(\omega_s-\omega)^{-1}\{2cNR\zeta+2J\zeta[cR\tilde{B}_R(\omega)-\zeta F_R(\omega)]\}$$
$$+\tfrac{1}{2}J\Xi_L(\omega)F_R(\omega)-\tfrac{1}{2}cJ\{[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\times\Lambda_T{}^a(\omega)+\langle S_i^z\rangle\Lambda_T{}^b(\omega)\}\tilde{B}_R(\omega), \quad (2.22b)$$

where

$$\Xi_L(\omega)=\frac{1}{2N^2}\sum_{\mathbf{k},q,\lambda}\frac{\{f_{k,\lambda}{}^+ f_{k+q,\lambda}{}^- + f_{k,\lambda}{}^- f_{k+q,\lambda}{}^+\}}{\epsilon_{k,\lambda}-\epsilon_{k+q,\lambda}+\omega_s-\omega-i\eta} \quad (2.25)$$

and

$$\Lambda_T{}^a(\omega)=\frac{1}{N}\sum_{\mathbf{q}}\left\{\frac{1}{\epsilon_{k+q}-\epsilon_k+\omega_s-\omega-i\eta}+\frac{1}{\epsilon_k-\epsilon_{k+q}+\omega_s-\omega-i\eta}\right\} \quad (2.26a)$$

$$\Lambda_T{}^b(\omega)=\frac{1}{N}\sum_{\mathbf{q}}\left\{\frac{(f_{k+q\downarrow}{}^+-f_{k+q\downarrow}{}^-)}{\epsilon_{k+q}-\epsilon_k+\omega_s-\omega-i\eta}-\frac{(f_{k+q\uparrow}{}^+-f_{k+q\uparrow}{}^-)}{\epsilon_k-\epsilon_{k+q}+\omega_s-\omega-i\eta}\right\}. \quad (2.26b)$$

The expressions (2.22a) and (2.22b) are approximate, and can be generated in a satisfactory manner only when ϵ_k is near the Fermi energy. They arise from $\mathbf{k}$-dependent terms of the following form:

$$\sum_{\mathbf{k},j}\sum_{\mathbf{q}}\mathfrak{F}(\mathbf{k},\mathbf{q};\omega)\langle\langle a_{k\downarrow}^\dagger a_{k\uparrow}; S_j^+\rangle\rangle_\omega,$$

where $\mathfrak{F}(\mathbf{k},\mathbf{q};\omega)$ represents the curly bracketed terms appearing in (2.24) and (2.26). The sum over $\mathbf{q}$ of $\mathfrak{F}(\mathbf{k},\mathbf{q};\omega)$ is found to be dependent only on the magnitude of $\mathbf{k}$. The coupled resonance problem we are considering in this paper involves the Green's function $\tilde{B}_R(\omega)$, (2.8), which in turn depends only on those wave vectors whose length nearly equals the Fermi wave vector k_F. Thus we can separate the two terms to write

$$\sum_{\mathbf{q}}\mathfrak{F}(k_F,\mathbf{q};\omega)\sum_{\mathbf{k},j}\langle\langle a_{k\downarrow}^\dagger a_{k\uparrow}; S_j^+\rangle\rangle_\omega\equiv\sum_{\mathbf{q}}\mathfrak{F}(k_F,\mathbf{q};\omega)\tilde{B}_R(\omega).$$

Again, this approximation is only valid when $|\mathbf{k}|\approx k_F$, so that the separation leading to the quantities $\Lambda(\omega)$, (2.24), and (2.26), is only an approximate one. Otherwise, for ϵ_k far from ϵ_F, it is in fact impossible to effect such a separation of terms. As we shall see later in (2.39), this limits the validity of the concept of a conduction-electron exchange relaxation rate, $1/T_{sd}$.

In addition to (2.22a) and (2.22b), we also find

$$M_R(\omega)\cong-(\omega_s-\omega)^{-1}2J\zeta[cRK_R(\omega)-\zeta B_R(\omega)]$$
$$+\tfrac{1}{2}J\Xi_L(\omega)B_R(\omega)-\tfrac{1}{2}cJ\{[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\times\Lambda_T{}^a(\omega)-\langle S_i^z\rangle\Lambda_T{}^b(\omega)\}K_R(\omega) \quad (2.22c)$$

and

$$N_R(\omega)\cong-(\omega_e-\omega)^{-1}\times\{2cNR\zeta-2cJR[cRK_R(\omega)-\zeta B_R(\omega)]\}-\tfrac{1}{2}J\Xi_T(\omega)B_R(\omega)+\tfrac{1}{2}cJ\langle(\delta S_i^z)^2\rangle\Lambda_L(\omega)K_R(\omega)\,. \tag{2.22d}$$

Comparing (2.22) with the first-order results (2.19), we see that the effect of going to higher order in J has been to introduce the quantities $\Xi_{L,T}$ and $\Lambda_{L,T}$ into the equations for the fundamental propagators (2.9). Defining

$$\Xi(\omega)=\Xi_L(\omega)+\Xi_T(\omega)\,,$$
$$\tilde{\Lambda}(\omega)=\langle(\delta S_i^z)^2\rangle\Lambda_L(\omega)+[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\times\Lambda_T{}^a(\omega)+\langle S_i^z\rangle\Lambda_T{}^b(\omega)\,, \tag{2.27}$$

the Eqs. (2.9) for the propagators defined in (2.8) can be solved. Defining the quantities

$$a=2J\zeta;\quad b=2cJR; \tag{2.28}$$

we find

$$F_R(\omega)=-2cNR\{(\omega_e-\omega-b)\times[(\omega_s-\omega+a)(\omega_e-\omega+b)-ab]-(\omega_s-\omega)(\omega_e-\omega)cJ^2\tilde{\Lambda}(\omega)\}D^{-1}(\omega)\,, \tag{2.29a}$$

$$\tilde{B}_R(\omega)=2cNR\{a[(\omega_s-\omega+a)(\omega_e-\omega+b)-ab]+(\omega_s-\omega)(\omega_e-\omega)J^2\Xi(\omega)\}D^{-1}(\omega)\,, \tag{2.29b}$$

$$K_R(\omega)=-2N\zeta\{(\omega_s-\omega-a)\times[(\omega_s-\omega+a)(\omega_e-\omega+b)-ab]-J^2(\omega_s-\omega)(\omega_e-\omega)\Xi(\omega)\}D^{-1}(\omega)\,, \tag{2.29c}$$

$$B_R(\omega)=2N\zeta\{b[(\omega_s-\omega+a)(\omega_e-\omega+b)-ab]+(\omega_s-\omega)(\omega_e-\omega)cJ^2\tilde{\Lambda}(\omega)\}D^{-1}(\omega)\,, \tag{2.29d}$$

where

$$D(\omega)=\{(\omega_s-\omega-a)(\omega_e-\omega-b)-ab\}\times\{(\omega_s-\omega+a)(\omega_e-\omega+b)-ab\}-(\omega_s-\omega)(\omega_e-\omega)\times\{(\omega_e-\omega)J^2\Xi(\omega)+(\omega_s-\omega)c\tilde{\Lambda}(\omega)\}\,. \tag{2.30}$$

These rather formidable equations represent the dynamical propagators in the absence of lattice damping. The localized and conduction-electron transverse susceptibilities can be constructed from them, exactly as in I. In particular,

$$\chi_s^-(\omega)=(g_s{}^2/2N)\{F_R(\omega)+(g_e/g_s)B_R(\omega)\}\,, \tag{2.31a}$$

and

$$\chi_e^-(\omega)=(g_e{}^2/2N)\{K_R(\omega)+(g_s/g_e)\tilde{B}_R(\omega)\}\,. \tag{2.31b}$$

We do not write these quantities explicitly in this section but rather wait until Sec. III where lattice damping is introduced explicitly. It is interesting, however, to examine the form of the four propagators given by (2.29). As can be seen from (2.29) each propagator has the same denominator, so that both components of $\chi^-(\omega)$, given by (2.31), will also have the same denominator. If we follow Tahir-Kheli[15] and perform the following manipulation, the second-order linewidths and shifts caused by exchange will become evident. We note that $D(\omega)$, as given by (2.30), is accurate only to $O(J^2)$. Hence, to this order, we may change the last term in the expression for $D(\omega)$ to

$$-J^2[(\omega_s-\omega+a)(\omega_e-\omega+b)-ab]\times\{(\omega_e-\omega)\Xi(\omega)+(\omega_s-\omega)c\tilde{\Lambda}(\omega)\}\,. \tag{2.32}$$

Similarly, to the J^2 terms in the numerator, identical additions can be made. Then the square-bracketed term in (2.32) will exactly cancel against the numerator and the equations will greatly simplify. This trick has the effect of removing what we believe to be spurious roots in the denominator and is entirely consistent with our stated accuracy. The denominator $D(\omega)$ then becomes

$$D(\omega)=\{(\omega_s-\omega-a)(\omega_e-\omega-b)-ab\}-J^2\{(\omega_e-\omega)\Xi(\omega)+(\omega_s-\omega)c\tilde{\Lambda}(\omega)\}\,. \tag{2.33}$$

We now have the expected quadratic equation to solve in order to obtain the two resonant roots for $\chi^-(\omega)$. Again, to $O(J^2)$, we may rearrange (2.33) to read

$$D(\omega)=\{\omega_s-\omega-a-J^2\Xi(\omega)\}\times\{\omega_e-\omega-b-cJ^2\tilde{\Lambda}(\omega)\}-ab\,. \tag{2.34}$$

Finally, then, we see that the real parts of $J^2\Xi(\omega)$ and $cJ^2\tilde{\Lambda}(\omega)$ represent the shift of the localized and conduction-electron resonant frequencies, respectively, when ω_s and ω_e are well separated (i.e., when $|\omega_s-\omega_e|\gg a,b$). Likewise, the imaginary parts of $J^2\Xi(\omega)$ and $cJ^2\tilde{\Lambda}(\omega)$ represent the linewidths for each resonance under similar conditions. If, however, ω_s and ω_e are close to one another, then the full expression for $\chi^-(\omega)$, (2.31), must be examined, rather than just the denominator (2.34). In the limit of equal g values, the second-order widths and shifts present in the denominator cancel against the numerator. This cancellation, which must occur physically, also takes place for the original unsimplified expressions (2.29) when inserted into the sum of the expressions for the transverse susceptibilities (2.31), [i.e., for $\chi^-(\omega)=\chi_e^-(\omega)+\chi_s^-(\omega)$ and *not* for $\chi_e^-(\omega)$ or $\chi_s^-(\omega)$ separately].

It is straightforward to evaluate $\Xi(\omega)$ and $\tilde{\Lambda}(\omega)$. We find, for the former quantity, using the explicit forms (2.23) and (2.25): High-temperature limit ($kT\gg\omega,\omega_s$):

$$\Xi_T(\omega)=2\rho^2\left\{\omega\left[1+\ln\left(\frac{\gamma D}{\pi kT}\right)\right]+\omega_e\ln 2+i\pi kT\right\}\,, \tag{2.35a}$$

$$\Xi_L(\omega)=2\rho^2\left\{(\omega-\omega_s)\left[1+\ln\left(\frac{\gamma D}{\pi kT}\right)\right]+i\pi kT\right\}\,. \tag{2.35b}$$

Low-temperature limit (general result for imaginary

[15] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 88 (1962); **127**, 95 (1962); R. A. Tahir-Kheli, *ibid.* **159**, 439 (1967).

part; $kT \ll \omega, \omega_s$ required for real part):

$$\Xi_T(\omega) = 2\rho^2\{\omega[1+\ln|D/\omega|]+\omega_e \ln 2 + i\pi\tfrac{1}{2}\omega \coth(\tfrac{1}{2}\beta\omega)\}, \quad (2.35c)$$

$$\Xi_L(\omega) = 2\rho^2\{(\omega-\omega_s)[1+\ln|\tfrac{1}{2}D/(\omega-\omega_s)|] + i\pi\tfrac{1}{2}(\omega-\omega_s)\coth\tfrac{1}{2}\beta(\omega-\omega_s)\}. \quad (2.35d)$$

We have assumed a square-band model for the conduction electrons of width $2D$ centered at ϵ_F and constant (one-spin) density of states ρ. In (2.35), γ is Euler's constant $\cong 0.58$. This result is in agreement with Spencer's[10] second-order results, and displays the separability of Ξ into the $1/T_2' = \mathrm{Im}J^2\Xi_L(\omega_s)$ (frequency modulation) and $1/T_1' = \mathrm{Im}\Xi_T(\omega_s)$ (spin flip) parts found by Orbach and Spencer.[3] It is interesting that both contributions to the localized spin line width enter the equations of the propagators on equal terms. This implies that they are "bottlenecked" with equal efficiency, when ω_e is near ω_s.

The latter term $\tilde{\Lambda}(\omega)$ defined by (2.24), (2.26), and (2.27), gives the second-order linewidth and shift for the conduction electrons. Evaluating (2.24) and (2.26a) under the same assumptions which were used in (2.35), we find for all temperatures

$$\Lambda_L(\omega) = 2\rho\{\ln|(D+\omega_e-\omega)/(D-\omega_e+\omega)|+i\pi\}, \quad (2.36a)$$

$$\Lambda_T{}^a(\omega) = 2\rho\{\ln|(D+\omega_s-\omega)/(D-\omega_s+\omega)|+i\pi\}. \quad (2.36b)$$

For high temperatures, $kT \gg \omega, \omega_s, \omega_e$, (2.26b) becomes,

$$\Lambda_T{}^b(\omega) = 2\rho\left\{2\ln\left(\frac{\pi kT}{2\gamma D}\right)+\ln\left|\frac{D^2-(\omega_s-\omega+\frac{1}{2}\omega_e)^2}{D^2-(\omega_s-\omega)^2}\right| + i\pi \tanh\tfrac{1}{2}\beta(\omega_s-\omega+\tfrac{1}{2}\omega_e)\right\}. \quad (2.36c)$$

The imaginary part of (2.36c) is in fact valid for all temperatures, whereas at low temperatures $kT \ll \omega, \omega_e, \omega_s$ the first two terms (the real part) in the curly brackets are replaced by

$$-\ln\left|\frac{D^2-(\omega_s-\omega)^2}{(\omega_s-\omega+\frac{1}{2}\omega_e)^2}\right|. \quad (2.36d)$$

The physical regime in which we are interested involves frequencies $\omega, \omega_e, \omega_s \ll D$ so that the real part of (2.36a) and (2.36b) is negligible. However, a logarithmic dependence on temperature is present in the real part of (2.36c) which appears in the equation of motion multiplied by the localized electron magnetization $\langle S_i{}^z\rangle$ [see (2.27)]. Hence, we find a Kondo-like logarithmic g shift for the conduction electrons similar to that previously obtained for the localized spin, (2.35a).[10] The imaginary part of $\tilde{\Lambda}(\omega)$, using (2.36) in (2.27), becomes

$$\mathrm{Im}\tilde{\Lambda}(\omega) = 2\pi\rho\{S(S+1)-\langle S_i{}^z\rangle^2 + \langle S_i{}^z\rangle \tanh\tfrac{1}{2}\beta(\omega_s-\omega+\tfrac{1}{2}\omega_e)\}. \quad (2.37)$$

This is an interesting and quite new result. It implies at $T=0$ ($\langle S_i{}^z\rangle \to -S$ as $T \to 0$ in our notation, since we have taken $\omega_s = -g_s h^z$) that $\mathrm{Im}\tilde{\Lambda}(\omega) \to 0$. As we shall determine in Sec. III, $cJ^2\,\mathrm{Im}\tilde{\Lambda}(\omega)$ under certain circumstances can be identified with $1/T_{sd}$. Previous treatments[2,4] have used explicitly, or assumed implicitly, a form for $1/T_{sd}$ which is frequency and temperature-independent. Clearly, this is not the case.

In the vanishing magnetic field (or, equivalently, high-temperature) regime, (2.37) implies a conduction-electron spin-resonance linewidth equal to

$$\Delta\omega_e{}^{1/2} = cJ^2\,\mathrm{Im}\tilde{\Lambda}(\omega) = 2\pi S(S+1)c\rho J^2, \quad (2.38)$$

which, for a localized-electron spin $S=\frac{1}{2}$, equals,

$$\tfrac{3}{2}\pi c\rho J^2.$$

This is just twice the one-electron width found by Spencer[10] and Abrikosov[16] and derived by the present method in Appendix B. It is very interesting that, to order J^2, the two-electron propagator width appropriate to the conduction-electron-spin-resonance signal is just the sum of the individual electron-hole single electron widths identified with the two terms which comprise $\tilde{\Lambda}(\omega)$ in (2.24) and (2.26). Our result is not necessarily new for this type of problem, but it has not been recognized until now for this specific application. It is tempting to set

$$1/T_2' = cJ^2\langle(\delta S_i{}^z)^2\rangle\,\mathrm{Im}\Lambda_L(\omega_e),$$
$$1/T_1' = cJ^2\{[\langle(S_i{}^x)^2\rangle+\langle(S_i{}^y)^2\rangle]\,\mathrm{Im}\Lambda_T{}^a(\omega_e) + \langle S_i{}^z\rangle\,\mathrm{Im}\Lambda_T{}^b(\omega_e). \quad (2.39)$$

This implies, in zero magnetic field, that $1/T_1' = 2/T_2'$, a somewhat strange result. It is caused by the fact that a longitudinal fluctuation ($1/T_2'$) frequency shifts the two electrons in opposite senses. The associated linewidths are additive because they appear on the diagonal of the energy matrix. The spin-flip amplitude, however, for a given spin must be added in the off diagonal terms, before squaring, to the amplitude of its partner in the $\sigma_0{}^-(t)$ term in the propagator. This effectively doubles the efficacy of spin-flip broadening for the two-particle propagator as compared to frequency modulation in the same circumstances. The result for the conduction-electron resonance linewidth, (2.37), is magnetic field dependent because of the saturation of $\langle S_i{}^z\rangle$ when $\omega_s \gg kT$. It would be interesting to look for this effect experimentally.

Before our results can be compared in detail with previous work,[2,4] it is necessary to include the effects of lattice damping. This inclusion is also necessary when the g values of the localized and conduction electron are close to one another, as otherwise bottlenecking occurs and all of the above exchange terms will disappear in the combined expression for $\chi^-(\omega)$.

[16] A. A. Abrikosov, Physics 2, 5 (1965).

III. COUPLED EQUATIONS IN THE PRESENCE OF LATTICE DAMPING

We return to the fundamental equations (2.1) and (2.2) of this paper. If the equation of motion for the propagators (2.8) are now developed using (2.1) and (2.2), the exact results, analogous to (2.9) but including lattice damping, are

$$(\omega_s-\omega-i\Delta_s)F_R(\omega)+2J\{V_R(\omega)-W_R(\omega)\} = 2cNR-i\Delta_s g_s^{-2}\chi_s 2J\tilde{B}_R(\omega), \quad (3.1a)$$

$$(\omega_e-\omega-i\Delta_e)\tilde{B}_R(\omega)-2J\{V_R(\omega)-W_R(\omega)\} = -i\Delta_e g_e^{-2}\chi_e 2JF_R(\omega), \quad (3.1b)$$

$$(\omega_e-\omega-i\Delta_e)K_R(\omega)+2J\{M_R(\omega)-N_R(\omega)\} = -2N\zeta-i\Delta_e g_e^{-2}\chi_e 2JB_R(\omega), \quad (3.1c)$$

$$(\omega_s-\omega-i\Delta_s)B_R(\omega)-2J\{M_R(\omega)-N_R(\omega)\} = -i\Delta_s g_s^{-2}\chi_s 2JK_R(\omega). \quad (3.1d)$$

These equations reduce to Eqs. (2.9) in the case of zero damping ($\Delta_e=\Delta_s=0$).

In (3.1) all the symbols are defined as in Sec. II. Using (2.5), two exact equations relating the four propagators may be found;

$$(\omega_s-\omega-i\Delta_s)F_R(\omega)+(\omega_e-\omega-i\Delta_e)\tilde{B}_R(\omega) \equiv -2cNR+\frac{i\Delta_e a}{\omega_e-b}F_R(\omega)+\frac{i\Delta_s b}{\omega_s-a}\tilde{B}_R(\omega) \quad (3.2)$$

and

$$(\omega_s-\omega-i\Delta_s)B_R(\omega)+(\omega_e-\omega-i\Delta_e)K_R(\omega) \equiv -2N\zeta+\frac{i\Delta_e a}{\omega_e-b}B_R(\omega)+\frac{i\Delta_s b}{\omega_s-a}K_R(\omega). \quad (3.3)$$

Again, equivalent to (2.19) it can be shown that the second-order propagators must, to first order in J, satisfy the set of equations

$$(\omega_e-\omega-i\Delta_e)V_R(\omega)\cong 2cJR\{c\tilde{R}\tilde{B}_R(\omega)-\tilde{\zeta}F_R(\omega)\}, \quad (3.4a)$$

$$(\omega_s-\omega-i\Delta_s)W_R(\omega) \cong -2cNR\zeta-2J\zeta\{c\tilde{R}\tilde{B}_R(\omega)-\zeta F_R(\omega)\}, \quad (3.4b)$$

$$(\omega_s-\omega-i\Delta_s)M_R(\omega) \cong -2J\zeta\{c\tilde{R}K_R(\omega)-\zeta B_R(\omega)\}, \quad (3.4c)$$

$$(\omega_e-\omega-i\Delta_e)N_R(\omega) \cong -2cNR\zeta+2cJR\{cRK_R(\omega)-\tilde{\zeta}B_R(\omega)\}, \quad (3.4d)$$

where

$$\tilde{R}=R\{1-[i\Delta_s/(\omega_s-a)]\}; \qquad \tilde{\zeta}=\zeta\{1-[i\Delta_e/(\omega_e-b)]\}. \quad (3.5)$$

The utility of the first-order equations (3.4) can now immediately be seen. They demonstrate that it is essential to regard only the transverse component of the left-hand side of the second-order propagators (2.10) as being damped. Otherwise, there will enter, in their equations of motion, terms of the form $i(\Delta_s+\Delta_e)$ which will not reduce to the correct first-order result (3.4) in the limit of small J. This result is probably correct to order J^2 as we are, in our decoupling procedure, essentially ignoring the dynamics of longitudinal motion. Such an assumption is consistent with our use of the *static* longitudinal susceptibilities, χ_s and χ_e, in the fundamental equations of motion (2.1) and (2.2) in the lattice-relaxation terms.

The equation of motion in the presence of lattice damping for $V_R(\omega)$, analogous to (2.20), is then found to be

$$(\epsilon_{k\uparrow}-\epsilon_{k+q\downarrow}-\omega-i\Delta_e)\langle\langle \mathcal{S}_k^-(q)S_i^z;S_j^+\rangle\rangle_\omega = \delta_{ij}\langle \mathcal{S}_k^-(q)S_i^+\rangle-\langle\langle[\mathcal{S}_k^-(q)S_i^z,H_I];S_j^+\rangle\rangle_\omega + \frac{i\Delta_e a}{(\omega_e-b)}\frac{1}{N}\sum_l\langle\langle S_i^z S_l^-;S_j^+\rangle\rangle_\omega. \quad (3.6)$$

Using (2.4) and decoupling exactly as in Sec. II, [see (2.21)] we find, after a great deal of algebra,

$$V_R(\omega)\cong(\omega_e-\omega-i\Delta_e)^{-1}2cJR\{cR\tilde{B}_R(\omega)-\tilde{\zeta}F_R(\omega)\} -\tfrac{1}{2}J\Xi_T(\omega+i\Delta_e)F_R(\omega)+\tfrac{1}{2}cJ\langle(\delta S_i^z)^2\rangle \times\Lambda_L(\omega+i\Delta_e)\tilde{B}_R(\omega), \quad (3.7a)$$

$$W_R(\omega)\cong(\omega_s-\omega-i\Delta_s)^{-1} \times\{-2cNR\zeta-2J\zeta[c\tilde{R}\tilde{B}_R(\omega)-\zeta F_R(\omega)]\} +\tfrac{1}{2}J\Xi_L(\omega+i\Delta_s)F_R(\omega) -\tfrac{1}{2}cJ\{[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\Lambda_T{}^a(\omega+i\Delta_s) -\langle S_i^z\rangle\Lambda_T{}^b(\omega+i\Delta_s)\}\tilde{B}_R(\omega), \quad (3.7b)$$

$$M_R(\omega)\cong-(\omega_s-\omega-i\Delta_e)^{-1}2J\zeta[c\tilde{R}K_R(\omega)-\zeta B_R(\omega)] +\tfrac{1}{2}J\Xi_L(\omega+i\Delta_s)B_R(\omega) -\tfrac{1}{2}cJ\{[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\Lambda_T{}^a(\omega+i\Delta_s) -\langle S_i^z\rangle\Lambda_T{}^b(\omega+i\Delta_s)\}K_R(\omega), \quad (3.7c)$$

$$N_R(\omega)\cong(\omega_e-\omega-i\Delta_e)^{-1} \times\{-2cNR\zeta+2cJR[cRK_R(\omega)-\tilde{\zeta}B_R(\omega)]\} -\tfrac{1}{2}J\Xi_T(\omega+i\Delta_e)+\tfrac{1}{2}cJ\langle(\delta S_i^z)^2\rangle \times\Lambda_L(\omega+i\Delta_e)K_R(\omega). \quad (3.7d)$$

These expressions are formidable algebraically, but their essential similarity to the analogous set (2.22) in the absence of lattice damping is immediately seen. The second-order terms $J^2\Xi$ and $cJ^2\Lambda$ are now modified because of the presence of the complex argument which contains the lattice damping parameters. The evaluation of the quantities $\Lambda_L(\omega+i\Delta_e)$ and $\Lambda_T{}^a(\omega+i\Delta_s)$, defined by comparison with (2.24) and (2.26a), will not differ from (2.36a) and (2.36b) at high temperatures for damping coefficients which are small compared to any structure in the electronic density of states, as shown below. The expressions for $\Lambda_T{}^b(\omega+i\Delta_s)$ and $\Xi(\omega)$ will be affected, however, and will be discussed below.

Expressions for the propagators (2.8) derivable from (3.1) and (3.7) will not be written down explicitly be-

cause of their complexity and essential similarity to the results presented in Sec. II, Eq. (2.29). One complicating feature which arises now, but was absent in Sec. II, is the effect of the imaginary driving terms on the right-hand side of (2.1) and (2.2) which were extensively discussed in I. They arise from the coupled equations

$$\left(i\frac{\partial}{\partial t}-\omega_s+i\Delta_s\right)\langle M^-(t)\rangle = i\Delta_s\chi_s\left\{h_{\rm rf}^-(t)+\frac{2J}{g_e g_s}\langle m^-(t)\rangle\right\} + \frac{g_s J}{N^2}\sum_{\mathbf{q},i} e^{-i\mathbf{q}\cdot\mathbf{R}_i}\{\langle\sigma_{\mathbf{q}}^-(t)S_i^z(t)\rangle-\langle\sigma_{\mathbf{q}}^z(t)S_i^-(t)\rangle\} \tag{3.8a}$$

and

$$\left(i\frac{\partial}{\partial t}-\omega_e+i\Delta_e\right)\langle m^-(t)\rangle = i\Delta_e\chi_e\left\{h_{\rm rf}^-(t)+\frac{2J}{g_e g_s}\langle M^-(t)\rangle\right\} - \frac{g_e J}{N^2}\sum_{\mathbf{q},i} e^{-i\mathbf{q}\cdot\mathbf{R}_i}\{\langle\sigma_{\mathbf{q}}^-(t)S_i^z(t)\rangle-\langle\sigma_{\mathbf{q}}^z(t)S_i^-(t)\rangle\}. \tag{3.8b}$$

The first-order solutions of this set were discussed in I. The second order follow in the same manner as do the solutions for the propagators presented in this paper and we shall not go through the algebra here. Defining, for simplicity, (primed quantities include damping)

$$\omega_s'=\omega_s-\omega-i\Delta_s, \qquad \omega_e'=\omega_e-\omega-i\Delta_e;$$

$$\tilde{a}=a\left\{1-\frac{i\Delta_e}{\omega_e-b}\right\}, \qquad \tilde{b}=b\left\{1-\frac{i\Delta_s}{\omega_s-a}\right\};$$

$$\Xi'(\omega)=\Xi_T(\omega+i\Delta_e)+\Xi_L(\omega+i\Delta_s),$$

$$\tilde{\Lambda}'(\omega)=\langle(\delta S_i^z)^2\rangle\Lambda_L(\omega+i\Delta_e)+[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]\times\Lambda_T{}^a(\omega+i\Delta_s)+\langle S_i^z\rangle\Lambda_T{}^b(\omega+i\Delta_s); \tag{3.9}$$

our solutions for the transverse dynamic susceptibilities are as follows:

$$\chi_s^-(\omega)=-\{g_s^2c\tilde{R}\times[(\omega_e'-b)(\omega_s'\omega_e'+\omega_s'b+\omega_e'a)-J^2\omega_s'\omega_e'c\tilde{\Lambda}'] -g_e g_s\tilde{\zeta}[\tilde{b}(\omega_s'\omega_e'+\omega_s'b+\omega_e'a)+\omega_s'\omega_e'cJ^2\tilde{\Lambda}']\}D'^{-1}(\omega), \tag{3.10a}$$

and

$$\chi_e^-(\omega)=-\{g_e^2\tilde{\zeta}\times[(\omega_s'-a)(\omega_s'\omega_e'+\omega_s'b+\omega_e'a)-\omega_s'\omega_e'J^2\Xi'] -g_e g_s c\tilde{R}[\tilde{a}(\omega_s'\omega_e'+\omega_s'b+\omega_e'a)+\omega_s'\omega_e'J^2\Xi']\}D'^{-1}(\omega), \tag{3.10b}$$

where

$$D'(\omega)=[(\omega_s'-a)(\omega_e'-b)-\tilde{a}\tilde{b}][\omega_s'\omega_e'+\omega_s'b+\omega_e'a] -\omega_s'\omega_e'J^2\left\{\left[\omega_e'-\frac{i\Delta_s b}{\omega_s-a}\right]\Xi' +\left[\omega_s'-\frac{i\Delta_e a}{\omega_e-b}\right]c\tilde{\Lambda}'\right\}. \tag{3.10c}$$

These also are formidible equations, but they possess the entirety of the dynamics of the lattice damped coupled localized conduction-electron system to $O(J^2)$. It is easily demonstrated that, in the limit of equal g values ($\omega_e=\omega_s=\omega_0$) and no lattice damping, the sum of the susceptibilities (3.10a) and (3.10b) immediately reduces to the result

$$\chi^-(\omega)=\chi_s^-(\omega)+\chi_e^-(\omega)=-g^2(cR+\zeta)/(\omega_0-\omega), \tag{3.11}$$

where all second-order terms have exactly cancelled. The result (3.11) is exactly what one would obtain for a coupled system of two spins with identical g values and no external damping.

In order to examine this result in more detail we perform the same trick as in Sec. II which led to Eq. (2.33) for the resonance denominator. Namely, we change all products $\omega_s'\omega_e'$ which multiply J^2 terms to $(\omega_s'\omega_e'+\omega_s'b+\omega_e'a)$. This eliminates the extra double root which apparently is contained in $D'(\omega)$, (3.10c), and is again valid to $O(J^2)$. Then (3.10) reduces to

$$\chi_s^-(\omega)=-\{g_s^2c\tilde{R}[\omega_e'-b-cJ^2\tilde{\Lambda}'(\omega)] -g_e g_s\tilde{\zeta}[\tilde{b}+cJ^2\tilde{\Lambda}'(\omega)]\}\tilde{D}^{-1}(\omega), \tag{3.12a}$$

$$\chi_e^-(\omega)=-\{g_e^2\tilde{\zeta}[\omega_s'-a-J^2\Xi'(\omega)] -g_e g_s c\tilde{R}[\tilde{a}+J^2\Xi'(\omega)]\}\tilde{D}^{-1}(\omega), \tag{3.12b}$$

where

$$\tilde{D}(\omega)=[(\omega_s'-a)(\omega_e'-b)-\tilde{a}\tilde{b}] -J^2[\omega_e'\Xi'(\omega)+\omega_s'c\tilde{\Lambda}'(\omega)]. \tag{3.12c}$$

We have been forced to drop the imaginary terms $-i\Delta_s b/(\omega_s-a)$ and $-i\Delta_e a/(\omega_e-b)$ appearing as coefficients of Ξ' and $\tilde{\Lambda}'$ in (3.10c) in order to remain consistent to $O(J^2)$, since a and b (2.28) are proportional to J. To this order, it is easy to see that, for ω_s very different from ω_e (i.e., a large difference between conduction and localized electron g values) $J^2\Xi'$ and $cJ^2\tilde{\Lambda}'$ represent the frequency-dependent second-order widths and shifts for the localized and conduction electrons, respectively. To establish a connection with other authors, notably Hasegawa[2] and Cottet *et al.*,[4] respectively, we identify,

$$1/T_{ds}=\delta_{ie}=J^2\,\mathrm{Im}\Xi'(\omega), \qquad 1/T_{sd}=\delta_{ei}=cJ^2\,\mathrm{Im}\tilde{\Lambda}'(\omega). \tag{3.13}$$

These equalities are somewhat misleading for a number of reasons, some of which we have already set out

in this and a previous paper.[3] The quantities Ξ' and $\tilde{\Lambda}'$ contain contributions from both frequency modulation and spin-flip terms so the meanings of the symbols $1/T_{ds}$ and $1/T_{sd}$ (δ_{ie} and δ_{ei}) in this context seem to us to be obscure.

Some comments are also in order about the character of Ξ' and Λ'. As can be seen from (2.35) and (2.36), both quantities in the absence of damping are quite frequency-dependent. Thus, it is not correct to assume that T_{ds} and T_{sd} are frequency-independent terms proportional to, and independent of, $1/kT$, respectively. Only at high temperatures, zero field, and in the absence of lattice damping is $1/T_{ds}=\mathrm{Im}J^2\Xi'$ and $1/T_{sd}=\mathrm{Im}cJ^2\tilde{\Lambda}'$.

We examine first the situation for $\Xi'(\omega)$. From (3.9) it is seen that the relevant quantities are

$$\Xi'(\omega)=\Xi_T(\omega+i\Delta_e)+\Xi_L(\omega+i\Delta_s). \tag{3.14}$$

A glance at the explicit form of $\Xi_T(\omega)$ and $\Xi_L(\omega)$, (2.23) and (2.25), demonstrates that the presence of the damping terms Δ_e,Δ_s complicates their evaluation greatly. The imaginary part of Ξ' is essentially unaltered from (2.35) as long as $\Delta_e,\Delta_s\ll kT$. When the converse is true (an unlikely situation), Ξ' becomes temperature-independent and

$$\mathrm{Im}\Xi_T(\omega+i\Delta_e)=2\rho^2\left\{\Delta_e\ln\left[\frac{D}{2(\Delta_e^2+\omega^2)^{1/2}}\right]+\omega\tan^{-1}\left(\frac{\omega}{\Delta_e}\right)\right\}, \tag{3.15a}$$

$$\mathrm{Im}\Xi_L(\omega+i\Delta_s)=2\rho^2\left\{\Delta_s\ln\left[\frac{D}{2(\Delta_s^2+(\omega-\omega_s)^2)^{1/2}}\right]+(\omega-\omega_s)\tan^{-1}\left(\frac{\omega-\omega_s}{\Delta_s}\right)\right\}. \tag{3.15b}$$

We have not been able to evaluate the real parts of Ξ' in the presence of lattice damping.

We see, therefore, that one must be very careful using the idea of a T_{ds}. Even in the presence of lattice damping, the expression for Ξ' which can be related to an effective T_{ds} (or δ_{ie}), is strongly frequency-dependent. Thus, the absorption line shape will be altered from that derived by Hasegawa,[2] or Cottet *et al.*,[4] and it is necessary to evaluate $\Xi'(\omega)$ at the specific frequency (or field) in question in order to arrive at an appropriate magnitude for the absorption linewidth.

The situation for $\tilde{\Lambda}'(\omega)$ and hence the meaning of T_{sd} (or δ_{ei}) is quite similar. The imaginary parts of $\Lambda_L(\omega)$ and $\Lambda_T{}^a(\omega)$, as seen from (2.36a) and (2.36b) in the absence of damping, are frequency-independent. Thus, letting $\omega\rightarrow\omega+i\Delta_e$ and $\omega+i\Delta_s$, respectively, in (2.36) does not alter their value as long as any structure in the density of states at the Fermi surface does not take place within energy widths $\lesssim\Delta_e,\Delta_s$. Such variations are highly unlikely, and would represent an extremely pathological situation which we shall not consider further. The evaluation of $\mathrm{Im}\Lambda_T{}^b(\omega+i\Delta_s)$ is unaltered from (2.36c) as long as $\Delta_s\ll kT$. When the converse is true, an even more unlikely situation than for Ξ' (since Δ_s is usually $<\Delta_e$), $\mathrm{Im}\Lambda_T{}^b(\omega+i\Delta_s)$ becomes temperature-independent. We find, for $\Delta_s>kT$,

$$\mathrm{Im}\Lambda_T{}^b(\omega+i\Delta_s)=4\rho\tan^{-1}\left(\frac{\omega_s+(\omega_e/2)-\omega}{\Delta_s}\right). \tag{3.16}$$

The real part of $\tilde{\Lambda}'(\omega)$ (3.9) can be evaluated in a similar manner. The first two components, Λ_L and $\Lambda_T{}^a$, are always small in the absence of damping (2.36a) and (2.36b). In the presence of damping

$$\mathrm{Re}\Lambda_L(\omega+i\Delta_e)=\rho\ln\left\{\frac{\Delta_e^2+(D+\omega_e-\omega)^2}{\Delta_e^2+(D-\omega_e+\omega)^2}\right\}, \tag{3.17a}$$

$$\mathrm{Re}\Lambda_T{}^a(\omega+i\Delta_s)=\rho\ln\left\{\frac{\Delta_s^2+(D+\omega_s-\omega)^2}{\Delta_s^2+(D-\omega_s+\omega)^2}\right\}. \tag{3.17b}$$

Most assuredly $\Delta_e,\Delta_s\ll D$, so that these two terms remain small in the presence of damping. The $\mathrm{Re}\Lambda_L$ vanishes at $\omega=\omega_e$ while the $\mathrm{Re}\Lambda_T{}^a$ is at best of the order of $[(\omega_s-\omega)/D]^2$ or $[\Delta_s/D]^4$, and hence negligible. The last term, $\Lambda_T{}^b(\omega)$ in the expression for $\tilde{\Lambda}'(\omega)$, (3.9), is essentially unaltered for $\Delta_s\ll kT$. In the *opposite* limit,

$$\mathrm{Re}\Lambda_T{}^b(\omega+i\Delta_s)=2\rho\ln\left\{\frac{\Delta_s^2+(\omega_s+(\omega_e/2)-\omega)^2}{\Delta_s^2+(D-\omega_s+\omega)^2}\right\}. \tag{3.17c}$$

Comparing this result with (2.36c), we see that in the limit that $\Delta_s\gg kT$ the localized spin lattice damping has "smoothed out" the frequency dependence of $\mathrm{Re}\Lambda_T{}^b(\omega)$ and eliminated the logarithmic-temperature dependence, for all values of ω, ω_e, and ω_s.

IV. DISCUSSION

It is interesting to compare our solutions (3.12) for $\chi^-(\omega)$ with those of Hasegawa[2] and Cottet *et al.*[4] to see what differences exist between these approaches and our own. The most instructive method is to examine the resonance denominator $\tilde{D}(\omega)$ given by (3.12c) with the corresponding expressions of these two different (macroscopic) approaches. We shall, for our purposes, generalize Hasegawa's treatment to allow for $g_s\neq g_e$, and also admit a term representing lattice damping of the localized spin, $i\Delta_s$. Defining

$$\begin{aligned}\omega_s''&=\omega_s-a-i\Delta_s,\\ \omega_e''&=\omega_e-b-i\Delta_e,\end{aligned} \tag{4.1}$$

and setting $\tilde{D}(\omega)=0$, (3.12c) results in the following secular equation (with the coupling constant

$\lambda=2J/(g_e g_s)$):

$$\omega^2-\omega\{\omega_e''+\omega_s''-J^2\Xi'(\omega)-cJ^2\tilde{\Lambda}'(\omega)\}+\{\omega_e''\omega_s''-\lambda^2\omega_e''\omega_s''\chi_e\chi_s-J^2\Xi'(\omega)\omega_e''-\omega_s''cJ^2\tilde{\Lambda}'(\omega)\}=0. \quad (4.2)$$

The corresponding expression of Hasegawa [from his Eqs. (4.26) and (4.27)], where he allows only the z component of the localized electron magnetization to polarize the conduction electrons, re-expressed in our notation, and retaining terms only of $O(J^2)$, is

$$\omega^2-\omega\left\{\omega_e''+\omega_s''-\frac{i}{T_{sd}}-\frac{i}{T_{ds}}\right\}+\left\{\omega_e''\omega_s''-2J\zeta 2cJR-\frac{i}{T_{ds}}\omega_e''-\frac{i}{T_{sd}}\omega_s''\right\}=0. \quad (4.3)$$

We see that, if we make the identification (3.13) and absorb the real parts of Ξ' and $\tilde{\Lambda}'$ into ω_s'' and ω_e'', respectively, that (4.2) and (4.3) differ only in the second-order terms $\lambda^2\omega_e''\omega_s''\chi_e\chi_s$ and $2J\zeta 2cJR$, respectively. Rewriting the former, we have to $O(J^2)$,

$$\lambda^2\omega_e''\omega_s''\chi_e\chi_s=2J\zeta 2cJR\left(1-\frac{i\Delta_e}{\omega_e-b}\right)\left(1-\frac{i\Delta_s}{\omega_s-a}\right). \quad (4.4)$$

Hence, the resonance denominators differ to this degree, the importance of which will depend on the experimental situation. There is an important additional difference, of course, and that has to do with the numerators of our expressions for $\chi_s^-(\omega)$ and $\chi_e^-(\omega)$, (3.12a) and (3.12b). These will be quite different from Hasegawa's (unfortunately, he did not evaluate these quantities explicitly in his paper) because of his neglect of instantaneous local field relaxation. As shown in I, this neglect is a serious matter, causing grave difficulties in the limiting behavior of $\chi^-(\omega)$ at zero frequency.

The approach of Cottet *et al.*[4] does not suffer from this difficulty, in that relaxation to the instantaneous local field is put in everywhere. We are not happy about forcing the second-order exchange terms in this manner, but until Langreth *et al.*[5] have published the details of their proof of this point, we are not really in a position to comment on its use. The denominator for $\chi^-(\omega)$ given by Cottet *et al.*[4] in our notation, and including only terms of second order in J, is identical to (4.2) if we again make the identification (3.13) and absorb the real parts of Ξ' and $\tilde{\Lambda}'$ into ω_s'' and ω_e'', respectively. An intriguing point is that Hasegawa's case B [his Eq. (5.6), where he allows for instantaneous local field relaxation by setting $\mathbf{m}=\chi_s(h^z+\lambda\mathbf{M})$] does in fact lead to the same expression [to $O(J^2)$] for $\tilde{D}(\omega)$ as their result, and, of course, our (4.2). We believe that Hasegawa's result, insofar as the numerator of his expressions are concerned, is still wrong for the reasons outlined above, but it is interesting that the resoance roots in his second case are the same as those of Cottet *et al.*[4] and ourselves.

In summation, it would be very interesting to see if the points of difference between this paper and that of Cottet *et al.*[4] could be observed experimentally. In particular, it would be worthwhile to look for the exchange g shifts and linewidths near the Knight shifted localized and conduction-electron resonance frequencies as a function of field and temperature. Summarizing our results of Sec. II, as we believe that for all practical purposes $\Delta_e,\Delta_s\ll kT$, these are, respectively,

$$\Delta g_s(\omega)=\mathrm{Re}h_0^{-1}J^2\Xi'(\omega)=2(\rho J)^2\left\{\left(g_s+\frac{2\omega}{h_0}\right)\left[1-\ln\left(\frac{\pi kT}{\gamma D}\right)\right]-g_e\ln 2\right\}, \quad (4.5a)$$

$$\Delta\omega_s^{1/2}(\omega)=\mathrm{Im}J^2\Xi'(\omega)=\pi(\rho J)^2\left\{\omega\coth\left(\frac{\omega}{2kT}\right)+(\omega-\omega_s)\coth\left(\frac{\omega-\omega_s}{2kT}\right)\right\}, \quad (4.5b)$$

$$\Delta g_e(\omega)=\mathrm{Re}h_0^{-1}cJ^2\tilde{\Lambda}'(\omega)=2c\rho J^2h_0^{-1}\left\{\langle(\delta S_i^z)^2\rangle\ln\left|\frac{D+\omega_e-\omega}{D-\omega_e+\omega}\right|+\langle(S_i^x)^2+(S_i^y)^2\rangle\ln\left|\frac{D+\omega_s-\omega}{D-\omega_s+\omega}\right|+2\langle S_i^z\rangle\ln\left(\frac{\pi kT}{2\gamma D}\right)\right\}, \quad (4.5c)$$

$$\Delta\omega_e^{1/2}(\omega)=\mathrm{Im}cJ^2\tilde{\Lambda}(\omega)=2\pi c\rho J^2\left\{S(S+1)-\langle S_i^z\rangle^2+\langle S_i^z\rangle\tanh\left(\frac{\omega_s+(\omega_e/2)-\omega}{2kT}\right)\right\}. \quad (4.5d)$$

Our notation, again, sets $\omega_s=-g_sh_0$, $\omega_e=-g_eh_0$ so that, for example, for $S=\frac{1}{2}$, $\langle S_i^z\rangle=-\frac{1}{2}\tanh(\omega_s/2kT)$ and ω is negative at the resonances. Thus, in (4.5a), when $\omega=\omega_s$ the first term in parenthesis equals g_s. The g shifts in (4.5) are appropriate to $kT\gg\omega,\omega_e,\omega_s$ while the half-widths are correct for all T. The low-temperature forms, $kT\ll\omega,\omega_e,\omega_s$, for the g shifts can be found immediately by using (2.35c) and (2.36d), and (2.36a), (2.36b), and (2.36d) in (4.5a) and (4.5c), respectively, using the definitions of (3.9). We have gone to the trouble of exhibiting these results in order to emphasize the frequency and temperature dependences of the resonance shifts and widths. These are new results, and cannot be obtained from the usual macroscopic formulations utilizing T_{ds} and T_{sd}.

There is one other point which should be made concerning the underlying assumptions in this paper (and in I). As can be seen from (2.1) and (2.2), and also in (3.6), we have assumed a frequency- and wave-vector-independent lattice relaxation mechanism for the conduction electrons. We believe this to be the weakest part of our formulation. The algebraic complexity of treating this interaction explicitly by introducing yet another field, the phonons (and/or impurities), has prevented us thus far from pushing this part of the problem further. A very interesting recent paper by Fulde and Luther,[17] however, demonstrates that, in the long wavelength, low-frequency limit, it is possible to treat spin-orbit relaxation of the conduction-electron spin properly. Their result exhibits relaxation towards the instantaneous local field without the necessity of its explicit introduction, as we have been forced to do in (2.1) and (2.2). Their approach clearly leads to the next step in the analysis of this problem. However, it will be necessary to extend their method to arbitrary wave vector, as the quantities Ξ' and $\tilde{\Lambda}'$ involve sums over-all electron wave vectors, even at low frequencies. Again, our approach appears to be limited by the necessity of assuming an "external" relaxation width and thereby having to force instantaneous local field relaxation for such terms. We do not believe, however, that lifting this assumption will seriously modify the results of this paper.

APPENDIX A: SYMMETRIZED GREEN'S FUNCTIONS

In the equations of motion method for double-time Green's functions (GF) higher-order GFs are introduced with products of operators whose order depends on the original order of the operators in the simpler GF. In many cases the operators in the simpler GF commute so their order is unimportant. However, upon evaluating the commutator in the equation of motion (*q.v.*) it may occur that the new set of operators do not commute. This is especially important when subsequent decouplings are performed. The introduction of symmetrized Green's functions (SGF) ensures that no asymmetric decoupling is performed. Thus if two operators A and C commute we can define the (equal-time) symmetrized product by

$$\{A,C\}_s=\tfrac{1}{2}(AC+CA). \tag{A1}$$

If we use the same notation as in Eq. (2.7) for the retarded GF, then this GF satisfies

$$\langle\langle[A,H];B\rangle\rangle_\omega-\omega\langle\langle A;B\rangle\rangle_\omega=\langle[A,B]\rangle. \tag{A2}$$

It can be readily shown that the SGF also satisfies a similar equation, namely,

$$\langle\langle[\{A,C\}_s,H];B\rangle\rangle_\omega-\omega\langle\langle\{A,C\}_s;B\rangle\rangle_\omega=\langle[\{A,C\}_s,B]\rangle, \tag{A3}$$

where

$$[\{A,C\}_s,B]=\{[AC,B]\}_s, \quad \text{etc.} \tag{A4}$$

[17] Peter Fulde and Alan Luther, Phys. Rev. **175**, 337 (1968).

This procedure removes the degree of arbitrariness present in the standard (unsymmetrized) analyses of equations of motion. In the present paper, SGFs are important when we have more than one transverse operator of the same species appearing in the analysis. The resulting symmetric decoupling corresponds to the noncommutivity of the positive and negative time orderings in the self-energies of the GF's.

APPENDIX B: ONE-ELECTRON PROBLEM

In this Appendix, we examine the one-electron frequency shift and linewidth mentioned in the introduction for the coupled localized conduction-electron spin system. Using (2.4), the one-electron propagator satisfies the equation of motion, (A2)

$$(\epsilon_{\mathbf{k},\lambda}-\omega)\langle\langle a_{\mathbf{k},\lambda};a_{\mathbf{k}',\lambda}^\dagger\rangle\rangle_\omega=\delta_{\mathbf{k},\mathbf{k}'}+\frac{J}{N}\sum_{\mathbf{q},j}\exp\{-i\mathbf{q}\cdot\mathbf{R}_j\}\times\langle\langle\{a_{\mathbf{k}+\mathbf{q},-\lambda}S_j^{-\lambda}+\lambda a_{\mathbf{k}+\mathbf{q},\lambda}S_j^z\};a_{\mathbf{k}',\lambda}^\dagger\rangle\rangle. \tag{B1}$$

We thus define three Green's functions

$$G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)=\langle\langle a_{\mathbf{k},\lambda};a_{\mathbf{k}',\lambda}^\dagger\rangle\rangle_\omega; \tag{B2a}$$

$$\Psi(\omega)=\frac{1}{N}\sum_{\mathbf{q},j}\exp(-i\mathbf{q}\cdot\mathbf{R}_j)\times\langle\langle a_{\mathbf{k}+\mathbf{q},\lambda}S_j^z;a_{\mathbf{k}',\lambda}^\dagger\rangle\rangle_\omega; \tag{B2b}$$

$$\Phi(\omega)=\frac{1}{N}\sum_{\mathbf{q},j}\exp(-i\mathbf{q}\cdot\mathbf{R}_j)\times\langle\langle a_{\mathbf{k}+\mathbf{q},-\lambda}S_j^{-\lambda};a_{\mathbf{k}',\lambda}^\dagger\rangle\rangle_\omega. \tag{B2c}$$

The equations of motion for the higher-order Green's functions $\Psi(\omega)$ and $\Phi(\omega)$ are derived in exactly the same manner as in (2.20). After decoupling using the same technique as exhibited in (2.21), and then spatially averaging as in (2.14), we find

$$\Psi(\omega)\cong\frac{cR}{(\epsilon_{\mathbf{k},\lambda}-\omega)}\{\delta_{\mathbf{k},\mathbf{k}'}+\lambda cJRG_{\mathbf{k}\mathbf{k}',\lambda}(\omega)\}+\lambda cJ\{\langle(\delta S_i^z)^2\rangle\}R_\lambda(\omega)G_{\mathbf{k}\mathbf{k}',\lambda}(\omega), \tag{B3a}$$

$$\Phi(\omega)\cong cJ\{\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle\}R_{-\lambda}(\omega-\lambda\omega_s)G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)-\lambda cJR\mathfrak{F}_\lambda(\omega-\lambda\omega_s)G_{\mathbf{k}\mathbf{k}',\lambda}(\omega), \tag{B3b}$$

where

$$R_\lambda(\omega)=\frac{1}{N}\sum_{\mathbf{k}}(\epsilon_{\mathbf{k},\lambda}-\omega)^{-1},$$

$$\mathfrak{F}_\lambda(\omega)=\frac{1}{N}\sum_{\mathbf{k}}(f_{\mathbf{k},-\lambda^+}-f_{\mathbf{k},-\lambda^-})/(\epsilon_{\mathbf{k},-\lambda}-\omega). \tag{B4}$$

Being careful about the first-order solution, as discussed in Sec. II, we find for the second-order solution for the

Green's function $G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)$, using (B3) and (B4) in (B1),

$$(\epsilon_{\mathbf{k},\lambda}-\omega)^2 G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)\cong\delta_{\mathbf{k},\mathbf{k}'}(\epsilon_{\mathbf{k},\lambda}-\omega) +\lambda cJR\{\delta_{\mathbf{k},\mathbf{k}'}+\lambda cJRG_{\mathbf{k}\mathbf{k}',\lambda}(\omega)\} +cJ^2(\epsilon_{\mathbf{k},\lambda}-\omega)\{\langle(\delta S_i^z)^2\rangle R_\lambda(\omega) +[\langle(S_i^x)^2\rangle+\langle(S_i^y)^2\rangle]R_{-\lambda}(\omega-\lambda\omega_s) -\lambda cJR\mathfrak{F}_\lambda(\omega-\lambda\omega_s)\}G_{\mathbf{k}\mathbf{k}',\lambda}(\omega). \quad \text{(B5)}$$

The first two terms on the right-hand side of (B5) give the correct first-order limit, and the third the second-order correction. This can be verified by using the first-order decoupling procedure (2.15) immediately upon $\Psi(\omega)$ and $\Phi(\omega)$ as defined in (B2) and then inserting the resulting expressions in (B1).

Equation (B5) is the final result for $G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)$ to second order in J. To compare with previous work, and to Eq. (2.37) of this paper which gives the two-electron width, $cJ^2\,\mathrm{Im}\tilde{\Lambda}(\omega)$, we rewrite Eq. (B5) in the limit of equal g values [so that $R_{-\lambda}(\omega-\lambda\omega_s)=R_\lambda(\omega)$ which allows us to combine the appropriate two second-order terms in (B5)]

$$G_{\mathbf{k}\mathbf{k}',\lambda}(\omega)\cong\delta_{k,k'}\{\epsilon_{\mathbf{k},\lambda}-\omega+\lambda cJR\} \times\{[\epsilon_{\mathbf{k},\lambda}-\omega-\lambda cJR][\epsilon_{\mathbf{k},\lambda}-\omega+\lambda cJR] -(\epsilon_{\mathbf{k},\lambda}-\omega)cV_\lambda^{(2)}(\omega)\}^{-1}, \quad \text{(B6)}$$

where we have used Spencer's[10] notation. We have in our case

$$V_\lambda^{(2)}(\omega) =\frac{J^2}{N}\sum_{\mathbf{k}}\frac{\{S(S+1)-\langle S_i^z\rangle^2-\lambda\langle S_i^z\rangle(f_{\mathbf{k},-\lambda}{}^+-f_{\mathbf{k},-\lambda}{}^-)\}}{\epsilon_{\mathbf{k},\lambda}-\omega}, \quad \text{(B7)}$$

whereas Spencer finds, for $S=\frac{1}{2}$, the identical result with $S(S+1)$, of course, replaced by $\frac{3}{4}$. This result is also Abrikosov's[16] lowest-order result, for $\langle S_i^z\rangle=0$. Evaluation[10] of the imaginary part (B7) on the energy shell at the Fermi surface, $p\simeq p_F$ (note that here ω refers to the one-electron excitation energy, including the Zeeman energy $\frac{1}{2}\lambda\omega_e$, so that $\omega=\frac{1}{2}\lambda\omega_e$) leads to

$$\mathrm{Im}V_\lambda^{(2)}(\tfrac{1}{2}\lambda\omega_e)=\pi\rho J^2\{S(S+1)-\langle S_i^z\rangle^2 +\langle S_i^z\rangle\tanh[(\omega_s-\tfrac{1}{2}\omega_e)/2kT]\}, \quad \text{(B8a)}$$

and for zero magnetic field,

$$\mathrm{Im}V_\lambda^{(2)}(0)=\tfrac{3}{4}\pi\rho J^2. \quad \text{(B8b)}$$

Both results are just half the two-electron widths given by Eqs. (2.37) and (2.38) evaluated on the appropriate energy shells, namely, ω_e and zero, respectively. It is interesting that such equalities are also true for the real parts of $V_\lambda^{(2)}(\omega)$ and $\tilde{\Lambda}(\omega)$. In particular, from (B7) we find a logarithmic g shift near $\omega=0$,

$$\mathrm{Re}V_\lambda^{(2)}(0)=2\lambda\rho J^2\langle S_i^z\rangle\ln(\pi kT/2\gamma D), \quad \text{(B9)}$$

which for $S=\frac{1}{2}$ agrees with the results of Spencer.[10] This is to be compared with our results for the real part of $J^2\tilde{\Lambda}(\omega)$ given by Eqs. (2.27) and (2.36)

$$\mathrm{Re}J^2\tilde{\Lambda}(0)=4\rho J^2\langle S_i^z\rangle\ln(\pi kT/2\gamma D). \quad \text{(B10)}$$

In fact, this is a quite general result for the two-particle propagator if the frequencies in the corresponding self-energies are put onto their appropriate energy shells. Thus, the one-particle linewidths are *additive* while the *difference* in the g shifts are taken for the total resonance g shift; i.e.,

$$\mathrm{Im}\sum_\lambda V_\lambda(\tfrac{1}{2}\lambda\omega_e+i\eta)=\mathrm{Im}J^2\tilde{\Lambda}(\omega_e), \quad \text{(B11)}$$

and

$$\mathrm{Re}\sum_\lambda \lambda V_\lambda(\tfrac{1}{2}\lambda\omega_e)=\mathrm{Re}J^2\tilde{\Lambda}(\omega_e).$$

The real and imaginary parts of $\Xi(\omega)$ are in agreement with the results of Spencer.[10]

ACKNOWLEDGMENTS

The authors wish to especially thank Dr. B. Giovannini and Dr. L. Hirst for their very helpful suggestions. Dr. Hirst first pointed out to us the possibility that the imaginary part of $\Xi'(\omega)$ could saturate at low temperatures (i.e., could remain finite as $T\to 0$). We also wish to thank Dr. P. Fulde and Dr. A. Luther for a very helpful conversation on the question of the validity of introducing an external relaxation term in our equations of motion.