Theory of spin resonance in dilute magnetic alloys. II

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The previous derivation by two of the authors of coupled Bloch equations appropriate to conduction-electron spin resonance is extended to the case where the conduction electrons and localized spins have different g values. As before, this derivation is carried out to second order in J, the exchange interaction between the two species. The derivation is also carried out in the presence of the hyperfine interaction between the nuclear spins and the electon spins of the localized impurities. Finally the form of the coupled equations when there exists "direct relaxation" is discussed and some rather unexpected features are found.

I. INTRODUCTION

This paper continues the discussion of the theory of local-moment resonance which two of us and Cowan[1] began a number of years ago and which was amplified by two of us more recently.[2] (This latter reference is henceforth referred to as I.) At the outset the results of I will be reviewed and contrasted with the work in this paper. In I we derived the Bloch equations for the conduction-electron magnetization density $\vec{M}_s$ and for a dilute concentration of localized magnetic impurities of magnetic-moment density $\vec{M}_d$. The spins of these two systems were characterized by the same gyromagnetic ratio γ and were coupled by an exchange interaction J. The resulting Hasegawa-like equations[3] were derived to second order in J in the limit $kT \gg \gamma H$.

A. Results of I

There were two essential features of the results of I. First, the disturbed magnetizations were shown to relax to the local instantaneous equilibrium magnetization. This led naturally to introduction of deviation magnetizations for the conduction electrons

$$\delta\vec{M}_s = \vec{M}_s - \chi_{s0}^0(\vec{H} + \alpha_1^0\vec{M}_d), \tag{1.1}$$

and the local spins

$$\delta\vec{M}_d = \vec{M}_d - \chi_{d0}^0[\vec{H} + (\alpha_1^0 + 2\alpha_2^0)\vec{M}_s]. \tag{1.2}$$

Here the susceptibilities are those for noninteracting species. Specifically the conduction-electron Pauli susceptibility is

$$\chi_{s0}^0 = \tfrac{1}{2}\rho(\hbar\gamma)^2, \tag{1.3}$$

where ρ is the density of states (for a single spin) at the Fermi surface, while the N local spins possess a Curie susceptibility

$$\chi_{d0}^0 = \frac{N\frac{1}{3}S(S+1)}{k_BT}(\hbar\gamma)^2. \tag{1.4}$$

The remaining quantities in (1.1) and (1.2) result from claculating the effective magnetization to second order in J, namely,

$$\alpha_1^0 = J/\gamma^2 n \tag{1.5}$$

and

$$\alpha_2^0 = (J/n)^2(\rho/\gamma^2)\ln(k_BT/D). \tag{1.6}$$

Here n is the density of electrons. A surprising result is the logarithmic dependence on the ratio of the thermal energy (k_BT) to the bandwidth (D) of the electrons. This manifestation of the Kondo effect[4] was one of the principal objectives of I. Another surprising feature was the asymmetric form of (1.1) and (1.2)—in particular, the absence of second-order, or indeed any high-order, term in (1.1). By setting $\delta\vec{M}_s$ and $\delta\vec{M}_d$ equal to zero, the correct susceptibility to second order in J does result. Our notation differs here from that of I in that we have used a *super*script zero to refer to the case of equal gyromagnetic ratios. Later on in the paper quantities not bearing that superscript can be assumed to have been calculated using the appropriate gyromagnetic ratio.

The second principal feature of I is that magnetizations are driven by different effective magnetic fields than appear as the coefficients of χ_{s0}^0 and χ_{d0}^0 in (1.1) and (1.2), respectively. This surprising effect only shows up in second order in J, and its occurrence has discouraged us from attempting higher-order calculations in J. The easiest way to illustrate this is simply to write down our equa-

tions [(1.24)–(1.26) in I]. We choose to do so in a way which will allow ready comparison with the results of this paper.

Our Hasegawa-like equations are

$$\frac{1}{\gamma}\frac{\partial M_s}{\partial t} - D\nabla^2(\delta\vec{M}_s/\gamma_s) + [\vec{H} + (\alpha_1^0 + \alpha_2^0)\vec{M}_d]\times\vec{M}_s = -\left(\frac{1}{T_{sl}} + \frac{1}{T_{sd}}\right)\frac{\delta\vec{M}_s}{\gamma} + \left(\frac{1}{T_{ds}} + \frac{\alpha_2^0\chi_{s0}^0}{T_{dl}}\frac{\delta\vec{M}_d}{\gamma}\right) \tag{1.7}$$

and

$$\frac{1}{\gamma}\frac{\partial\vec{M}_d}{\partial t} + [\vec{H} + (\alpha_1^0 + \alpha_2^0)\vec{M}_s]\times\vec{M}_d = -\left(\frac{1+\alpha_2^0\chi_{s0}^0}{T_{dl}} + \frac{1}{T_{ds}}\right)\frac{\delta\vec{M}_d}{\gamma} + \frac{1}{T_{sd}}\frac{\delta\vec{M}_s}{\gamma}\,. \tag{1.8}$$

The most important terms on the right-hand sides of (1.7) and (1.8) from the standpoint of the actual calculation in I are rates characterizing the loss of magnetization from the conduction electrons to the local spins

$$\frac{1}{T_{sd}} = \frac{2\pi}{\hbar}\left(\frac{J}{n}\right)^2 \rho N\tfrac{1}{3}S(S+1) \tag{1.9}$$

and the reverse rate, from local spins to the conduction electrons,

$$\frac{1}{T_{sd}} = \frac{\pi}{\hbar}\left(\frac{J}{n}\right)^2 \rho^2 k_B T, \tag{1.10}$$

where each rate has been claculated to second order in J. The other terms on the right-hand side involve the relaxation of the conduction-electron magnetization to the lattice via $1/T_{sl}$ and that of the local spins to lattice via $1/T_{dl}$. Our treatment of the latter process was heuristic in I, with the result that our Bloch equations weakly violated a symmetry relation. This flaw is discussed in detail at the outset of Sec. III, where the resolution of the difficulty also occurs. Turning now to the left-hand sides of (1.7) and (1.8), observe, first, the presence of a diffusion term in (1.7) associated with the itinerant character of the conduction electrons. More relevant to this paper are the effective driving fields causing precession of the magnetization.

B. Improvements in I

There are several aspects—frequently arising in physical systems—that have been left out of (1.7) and (1.8). The three taken up in this paper are as follows: (i) Differing gyromagnetic ratios γ_s and γ_d for the conduction and local spins, respectively, are included. To even the casual reader of I this might appear to involve a calculation of equal (and hence appalling) magnitude. By a trick and a little good luck we are able to use the voluminous appendixes of I to write the results down by inspection. Of course, the results to first order in J are already well known, the results to second order being not so intuitive. At the time of I the bulk of the experimental work[5,6] was on S-state ions for which γ_s is very close to γ_d. Since then a number[7–9] of non-S-state rare-earth ions have been studied in several host metals. For these $(\gamma_d - \gamma_s)H$ may exceed $1/T_{ds}$ and hence new phenomena not previously considered in I may be observed. Such a clearcut experimental situation does not motivate the remaining two aspects. (ii) We develop a more careful treatment of the manner in which a mechanism for directly relaxing the local spins to the lattice is included in the Bloch equations. Two classes of mechanisms are considered. (iii) The effect of the hyperfine interaction is included. A preliminary version of this work appeared in Ref. 1.

Let us first write down the derived Bloch equations in a general form for $\gamma_s \neq \gamma_d$ and then discuss several special cases. In this remember that (1.1) becomes

$$\delta\vec{M}_s = \vec{M}_s - \chi_{s0}(\vec{H} + \alpha_1\vec{M}_d), \tag{1.11}$$

where

$$\chi_{s0} = \tfrac{1}{2}\rho(\hbar\gamma_s)^2 \tag{1.12}$$

and

$$\alpha_1 = J/\gamma_s\gamma_d n. \tag{1.13}$$

On the other hand we will not specify $\delta\vec{M}_d$ since it depends so much on the case being considered. Further we note that in

$$\alpha_2 = (J/n)^2(\rho/\gamma_s^2)\ln(k_B T/D) \tag{1.14}$$

no factor of γ_d appears as it does in α_1. Finally we note that

$$\chi_{d0} = \frac{N\tfrac{1}{3}S(S+1)}{k_B T}(\hbar\gamma_d)^2. \tag{1.15}$$

The derivation to second order in J of the Bloch equations for $\gamma_s \neq \gamma_d$ yields

$$\frac{1}{\gamma_s}\frac{\partial\delta\vec{M}_s}{\partial t} - D_s\nabla^2(\delta\vec{M}_s/\gamma_s) + [\vec{H} + (\alpha_1 + \nu_{ds}\alpha_2)\vec{M}_d]\times\vec{M}_s + \alpha_2\left(\frac{\gamma_s}{\gamma_d}\nu_{ss} - \nu_{ds}\right)\vec{M}_d\times\delta\vec{M}_s = -\left(\frac{\mu_{ss}}{T_{sd}} + \frac{1}{T_{sl}}\right)\frac{\delta\vec{M}_s}{\gamma_s} + \frac{\mu_{ds}}{T_{ds}}\frac{\delta\vec{M}_d}{\gamma_d} \tag{1.16}$$

and

$$\frac{1}{\gamma_d}\frac{\partial \vec{M}_d}{\partial t}+[\vec{H}+(\alpha_1+\nu_{dd}\alpha_2)\vec{M}_s]\times\vec{M}_d+\alpha_2\left(\frac{\gamma_s}{\gamma_d}\nu_{sd}-\nu_{dd}\right)\delta\vec{M}_s\times\vec{M}_d=-\frac{\mu_{dd}}{T_{ds}}\frac{\delta\vec{M}_d}{\gamma_d}+\frac{\mu_{sd}}{T_{sd}}\frac{\delta\vec{M}_s}{\gamma_s}\ . \tag{1.17}$$

The reader is urged to compare these against (1.7) and (1.8) to see the many differences that occur. No alarm should be taken by the asymmetric occurrence of ν's as opposed to the μ's. This occurs because we have taken advantage of our knowledge of $\delta\vec{M}_s$ to organize the left-hand sides in a patently asymmetric fashion. Further T_{dl} does not explicitly appear but is hidden inside the ν's and μ's depending on the case, several of which we now consider.

1. No hyperfine and $1/T_{dl}=0$ (Sec. II)

In this case there is a considerable simplification:

$$\text{all } \nu\text{'s} = \text{all } \mu\text{'s} = (1-\alpha_2\chi_{s0})^{-1}, \tag{1.18}$$

whereas $\delta\vec{M}_s$ is given by (1.11) and $\delta\vec{M}_d$ by (2.40):

$$\vec{M}_d-\chi_{d0}(\vec{H}+\alpha_1\vec{M}_s+2\alpha_2(\gamma_s/\gamma_d)\{\vec{M}_s - \chi_{s0}[(\vec{H}+\alpha_1\vec{M}_s)-(\gamma_d/\gamma_s)(\vec{H}+\alpha_1\vec{M}_s)]\}) \ . \tag{1.19}$$

One can calculate the susceptibilities to linear order in α_1 or α_2 by setting $\delta\vec{M}_d=0$ and $\delta\vec{M}_s=0$:

$$\chi_d=\chi_{d0}[1+(\alpha_1+2\alpha_2)\chi_{s0}]$$

and

$$\chi_s=\chi_{s0}(1+\alpha_1\chi_{d0}),$$

which, allowing for changes in the definitions of χ_{d0}, χ_{s0}, α_1, and α_2, are the same as in I. While those results hold only for this case, the procedure works for every case.

2. No hyperfine; local spin relaxes by fluctuation mechanism (Sec. III)

In this case we imagine some fluctuation mechanism—such as spin orbit coupling to the phonons in the case of the conduction electrons—which relaxes the local spins to the lattice. Those objects off which the local spin flip (whatever they may be) are assumed to be in thermal equilibrium. While not defending this mechanism (since we do not know what the right one, if any, is), we believe that implications of this possibility should be worked out if only for the restraining influence they may have on the experimentalists.

In any case, we find that [see (3.13)]

$$\nu_{ss}=\mu_{ss},$$

$$\mu_{sd}=\nu_{sd}=\mu_{ds}=\nu_{ds},$$

and

$$\nu_{dd}=\mu_{dd}=1+T_{ds}/T_{dl}.$$

We have only exhibited the *dd* term since all the other terms' deviation from unity is reduced by an additional factor of $\alpha_2\chi_{s0}$. It is easy to show, for example by using the approach in Ref. 1, that ν_{dd} gives rise to an additional torque on the total magnetization so that even for $\gamma_s=\gamma_d$ there is a logarithmic g shift of order

$$\Delta\gamma/\gamma\sim\alpha_2\chi_{s0}T_{ds}/T_{dl}.$$

This rather surprising result would seem to be a way of uniquely identifying this mechanism for direct relaxation of the local spins. The ratio T_{ds}/T_{dl} also is seen in the formula for $\delta\vec{M}_d$ (3.15) which we do not reproduce here.

3. Hyperfine field (or frozen-in inhomogeneity) (Sec. IV)

The final case we consider is one where the local spin sees a hyperfine field, or alternately, a static inhomogeneous field, which can give rise to both a shift and width for the resonance. Here we find that

$$\text{all } \nu\text{'s} = \nu = (1-\alpha_2\chi_{s0})^{-1},$$

but all the μ's are different. The most significant μ, again, is

$$\mu_{dd}=\nu(1+T_{ds}/\tilde{T}_{dl}),$$

where now $\tilde{T}_{dl}$ is a complicated function of frequency and temperature.

There are two interesting extreme cases associated with complex structure of $\tilde{T}_{dl}$. On the one hand, if $\gamma_s\simeq\gamma_d$, we find both a nonlogarithmic g shift and an additional broadening. On the other hand, if the conduction electrons and local spin resonances are widely separated, then we find no additional g shift, but an additional broadening for the local spin which is proportional to T_{ds} (i.e., *not* $1/T_{ds}$!), so a quite anomalous temperature dependence might be observed under certain circumstances.

C. Connection to other work since I

The forms (1.1) and (1.2) for $\delta\vec{M}_s$ and $\delta\vec{M}_d$, respectively, were also arrived at about the same time as Ref. 1 by Cottet *et al.*[10] and have since been verified by other microscopic calculations[11,12] at least to first order in the exchange coupling J. However, there appeared a work by Sasada and Hasegawa,[13] based on the Anderson model[14] rather than on the s-d exchange model which we used, which came to the opposite conclusion, that is,

that the magnetization relaxed, respectively, to a full s or d susceptibility times the magnetic field H. This apparent contradiction was resolved in a note added in proof to I, and arose simply from a difference in the definition of the magnetization $\vec{M}_s$ and $\vec{M}_d$ in the two different models; indeed by applying the Schrieffer-Wolff transformation[15] we showed that the two apparently incompatible sets of equations could be transformed into each other. Schultz *et al.* and Fredkin[7,16] have extended our results to the case $\gamma_s \neq \gamma_d$; they find the same correspondence, although now the transformation introduces temperature dependence into the effective γ values. They also have applied symmetry relationships to find the most general form that such a set of coupled Bloch equation can take.

II. NONEQUAL G VALUES

A. Method of calculation

In this section we extend the derivation of I to nonequal g or γ values. As in I we prove that the exchange-coupled conduction electrons and localized spins are describable by a set of coupled Bloch equations in order $J^2 \ln T$, and we rigorously produce the coefficients to this order on either side of the equations. As a calculational device, instead of solving immediately the non-equal-γ problem, we first consider a fictitious problem in which the conduction electrons and localized spins have the same gyromagnetic ratio γ, but which interact with separately distinguishable magnetic fields $\vec{H}_s$ and $\vec{H}_d$, respectively. Then at the end of the calculation the solution to the physical problem is recovered by making the replacements:

$$\vec{M}_s \to (\gamma/\gamma_s)\vec{M}_s, \quad \vec{M}_d \to (\gamma/\gamma_d)\vec{M}_d \,, \tag{2.1a}$$

$$\vec{H}_s \to (\gamma_s/\gamma)\vec{H}, \quad \vec{H}_d \to (\gamma_d/\gamma)\vec{H}. \tag{2.1b}$$

We will eventually also redefine what we mean by the various symbols for the susceptibilities, but for clarity, the discussion of this is deferred until later. The replacement of the physical problem by the ficticious one has two advantages. The first is the vast majority of the term in the generalized Kadanoff-Baym equations[2,17] (GKB) remain unchanged from their values in I, and so a minimum of extra calculation is involved. The second is that one can automatically include the effects of the Hartree-like or mean-field self-energies [the diagrams of Figs. 1(a) and 1(c) in I] in the definitions of $\vec{H}_s$ and $\vec{H}_d$. Therefore we henceforth neglect these diagrams and at the end of the calculation, instead of making the replacement (2.1a) followed by (2.1b), we make the replacement (2.1a) followed by

$$\begin{aligned} &\vec{H}_s \to (\gamma_s/\gamma)\,[\vec{H} + (J/n\gamma_s\gamma_d)\vec{M}_d] \,, \\ &\vec{H}_d \to (\gamma_d/\gamma)\,[\vec{H} + (J/n\gamma_s\gamma_d)\vec{M}_s] \,. \end{aligned} \tag{2.1c}$$

The quantity $J/n\gamma_s\gamma_d$ is of course the non-equal-g version of what was called α_1 in I.

B. Expansion of the self-energies

To proceed further we note that the various self-energy functions can be expanded into pieces proportional to $\vec{M}_s$, $\vec{M}_d$, etc. For example, consider the imaginary part of the self-energy $\Gamma(p_0)$ for the local spins (as in I we use the upper case letters to denote local spin quantities while lower case letters refer to the conduction electrons—the one prominent exception being that to follow convention we denote the conduction-electron magnetizations by M_s; as before M_d refers to the local spin magnetization, whether it be d like, f like, or whatever). This function Γ may be written

$$\Gamma(p_0) = \Gamma_0(p_0) + \Gamma_{H_d}(p_0) + \Gamma_{M_s}(p_0) + \Gamma_{H_s\text{-}H_d}(p_0) \,, \tag{2.2}$$

where as in I, p_0 is the energy variable. The new term made possible by the distinctness of $\vec{H}_s$ and $\vec{H}_d$ is the final one, $\Gamma_{H_s\text{-}H_d}$, which is defined to be that piece of Γ proportional to $(\vec{H}_s - \vec{H}_d)$. All the other pieces must be the same functions as calculated in I, although Γ_{H_d} was called Γ_H there. It is shown in the Appendix that

$$\Gamma(p_0) = \Gamma_0(p_0) + \frac{\partial\Gamma(p_0)}{\partial p_0}\,\gamma\vec{S}\cdot\left(\vec{H}_d - \frac{\vec{M}_s - \chi^0_{s0}(\vec{H}_s - \vec{H}_d)}{S(S+1)\,\chi^0_{s0}}\right), \tag{2.3}$$

where $\Gamma_0(p_0)$ is the equilibrium zero field value [see Eq. (C4) of I]. The only new term is the $\chi^0_{s0}(\vec{H}_s - \vec{H}_d)$ term, which when multiplied out as indicated in (2.3) gives $\Gamma_{H_s\text{-}H_d}$. We note that the Γ_{M_s} implied above is the same as given in I [Eq. (C11)] although it was not written in this simple form. In writing (2.3) we have omitted the possibility of any direct relaxation of the local spins, and the discussion of this is deferred to Sec. III. The real and imaginary parts of the self-energy satisfy the usual dispersion relation

$$\Sigma(p_0) = \int \frac{dp_0'}{2\pi}\,\frac{\Gamma(p_0')}{p_0 - p_0'} \,, \tag{2.4}$$

where the Cauchy principal value is implied. This means, among other things, that $\Sigma(p_0)$ has the same functional relationship with $\Sigma_0(p_0)$ as implied by (2.3) for $\Gamma(p_0)$ and $\Gamma_0(p_0)$. Finally, one may also show that (see Appendix)

$$\Sigma^<(p_0) = \Sigma^<_0(p_0) + \Sigma^<_{M_d}(p_0)$$

$$+\frac{\partial\Sigma^<(p_0)}{\partial p_0}\,\gamma\vec{S}\cdot\left(\vec{H}_d - \frac{\vec{M}_s - \chi^0_{s0}(\vec{H}_s - \vec{H}_d)}{S(S+1)\,\chi^0_{s0}}\right). \tag{2.5}$$

Again, the only difference between (2.5) and the equivalent expression in I [$\Sigma^<_{M_d}$ is given by Eq. (C20) of I] is that $\vec{H}$ is replaced by $\vec{H}_d$ and then $\vec{M}_s$ is replaced by $\vec{M}_s - \chi^0_{s0}(\vec{H}_s - \vec{H}_d)$.

We treat the various self-energy functions for the conduction electrons in a similar manner. For example, defining (the proof that $\gamma_{M_s}=0$ given in I goes through unchanged)

$$\gamma(p_0)=\gamma_0(p_0)+\gamma_{H_s}(p_0)+\gamma_{M_d}(p_0)+\gamma_{H_d-H_s}(p_0)\ , \qquad (2.6)$$

one can show (see Appendix) that

$$\gamma_{H_d-H_s}(p_0)=\frac{\partial\gamma_0(p_0)}{\partial p_0}\gamma\vec{s}\cdot\frac{\vec{H}_d-\vec{H}_s}{s(s+1)}\ . \qquad (2.7)$$

Although this is identical in form to the analogous term for the local spins [Eq. (2.3)], in this case γ_{M_d} cannot be written in such a simple form as implied by (2.5) for Γ_{M_s} so that the whole γ function is no longer a simple function of γ_0. From the analog of the dispersion relation (2.5) for the conduction electrons, one can see that the functional relation (2.7) holds for the real part of the conduction self-energy σ as well. Finally, as shown in the Appendix, we can write

$$\sigma^<(p_0)=\sigma_0^<(p_0)+\sigma_{H_s}^<(p_0)+\sigma_{M_s}^<(p_0)+\sigma_{M_d}^<(p_0)+\sigma_{H_d-H_s}^<(p_0)\ , \qquad (2.8)$$

where

$$\sigma_{H_d-H_s}^<(p_0)=\frac{\partial\sigma^<(p_0)}{\partial p_0}\gamma\vec{s}\cdot\frac{\vec{H}_d-\vec{H}_s}{s(s+1)}\ . \qquad (2.9)$$

In Eqs. (2.6) and (2.8), the functions with subscripts other than H_d-H_s are the same as those in I, except that σ_{H_s}, γ_{H_s}, and $\sigma_{H_s}^<$ denote the functions called σ_H, γ_H, and $\sigma_H^<$ in I, with H replaced by H_s. The various self-energy relations given above neglect the direct relaxation of either the s or d spins; discussion of this is deferred to Sec. III.

C. Decompositions

The generalized Kadanoff-Baym equation (GKB) for the local spins is

$$-i[G_0^{-1}+\gamma\vec{S}\cdot\vec{H}_d-\Sigma),\ G^<]+i[\Sigma^<,G]=-\tfrac{1}{2}\{\Gamma,G^<\}+\tfrac{1}{2}\{\Sigma^<,A\}\ , \qquad (2.10)$$

where the square brackets represent commutation and the curly brackets anticommutation. The GKB for the conduction electrons is obtained by replacing capital letters by small letters as appropriate and $\vec{H}_d$ by $\vec{H}_s$. The symbols have the same meaning as in I. The general procedure is exactly as before: first we substitute the various self-energy functions given above into the GKB; and the GKB is Fourier transformed with respect to difference variables with the aid of the gradient expansion as described in Appendix A of I; finally to obtain the Bloch equation for the local spins, (2.10) is multiplied by $\gamma\vec{S}$ and summed over $\vec{p}$ and p_0 and traced over S; an analogous procedure is followed for the conduction electrons.

To simplify the process of carrying out this procedure we make use of decomposition of the functions $G^<$ and $g^<$. Just as in I, we write

$$G^<(p,p_0)=A(p,p_0)\,F(p_0)+\delta G^<(p,p_0)\ , \qquad (2.11a)$$

$$g^<(p,p_0)=a(p,p_0)f(p_0)+\delta g^<(p,p_0)\ , \qquad (2.11b)$$

where A and a are the nonequilibrium spectral weight functions for the local spins and conduction electrons, respectively, and F and f are appropriate Fermi functions. The decompositions (2.11) have the advantage that when $\delta G^<$ and $\delta g^<$ are neglected, the right-hand sides of the two GKB's vanish. As always

$$g^>+g^<=a=2\,\mathrm{Im}[p_0-\epsilon_p+\gamma\vec{s}\cdot\vec{H}_s-\sigma(p_0)-\tfrac{1}{2}i\gamma(p_0)]^{-1}\ , \qquad (2.12a)$$

$$G^>+G^<=A=2\,\mathrm{Im}[p_0+\gamma\vec{S}\cdot\vec{H}_d-\Sigma(p_0)-\tfrac{1}{2}i\Gamma(p_0)]^{-1}\ . \qquad (2.12b)$$

In deriving the Bloch equation one repeatedly has to evaluate

$$\mathrm{Tr}_S\,\gamma\vec{S}\sum_{\vec{p},p_0}\delta G^<(p,p_0)$$

and

$$\mathrm{Tr}_s\,\gamma\vec{s}\sum_{\vec{p},p_0}\delta g^<(p,p_0)\ ,$$

where $\delta G^<$ and $\delta g^<$ are the quantities defined by the decompositions (2.11). By referring to the analogous evaluations in I, the derivation can be accomplished with a minimum of effort. First, for the conduction electrons the result [Eq. (4.8) of I] still holds

$$\mathrm{Tr}_s\,\gamma\vec{s}\sum_{\vec{p},p_0}\delta g^<(\vec{p},p_0)=\vec{M}_s-\chi_{s0}^0\vec{H}_s \qquad (2.13)$$

in our model where the exchange coupling J and the density of states ρ are assumed constant; this is because the result (see Appendix D of I) depended only on the independence of σ and γ on the momentum and not their specific form, so that the sum in (2.13) is not changed by the presence of the extra piece (2.7) in the self-energy. On the other hand, for the local spins the functional form $\Gamma(p_0)$ [Eq. (2.2)] and hence A [Eq. (2.12b)] is the same as in I, except for the presence of the extra term $\Gamma_{H_s-H_d}$ whose effect may be incorporated, according to Eq. (2.3) by replacing M_s by $M_s-\chi_{s0}^0(H_s-H_d)$. Therefore, the appropriate sum, obtained by setting $\alpha_1=0$, $H\to H_d$, and then $M_s\to M_s-\chi_{s0}^0(H_s-H_d)$ in (4.15) of I is

$$\mathrm{Tr}_S\,\gamma\vec{S}\sum_{\vec{p},p_0}\delta G^<(\vec{p},p_0)=\vec{M}_d-\chi_{d0}^0\{\vec{H}_d+2\alpha_2^0[\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)]\}\ , \qquad (2.14)$$

where

$$\alpha_2^0=(J/n)^2(\rho/\gamma)^2\ln(k_BT/D) \qquad (2.15)$$

and

$$\chi_{s0}^0 = \tfrac{1}{2}\rho(\hbar\gamma)^2 , \tag{2.16a}$$

$$\chi_{d0}^0 = \tfrac{1}{3}S(S+1)N(\hbar\gamma)^2/k_B T . \tag{2.16b}$$

In equilibrium the sides of (2.14) and (2.15) vanish so that they may be solved to yield

$$M_s = \chi_s' H_s , \tag{2.17a}$$

$$M_d = \chi_d' H_d , \tag{2.17b}$$

where

$$\chi_s' = \chi_{s0}^0 , \tag{2.18a}$$

$$\chi_d' = \chi_{d0}^0(1 + 2\alpha_2^0 \chi_{s0}^0) . \tag{2.18b}$$

Note that χ_s' and χ_d' are not equal to the s and d susceptibilities χ_s and χ_d because $\vec{H}_s$ and $\vec{H}_d$ each contain the first-order exchange field from the other species. The actual susceptibilities are obtained as indicated following Eq. (1.19).

We now turn to the decompositions to be used in evaluating the left-hand sides of the GKB. As in I [Eqs. (3.1), (3.2), and (3.11)] we let

$$\begin{aligned} g^<(p,p_0) &= 2\pi\delta(p_0 - \epsilon_p + \gamma\vec{s}\cdot\vec{H}_s) f(p_0 + \gamma\vec{s}\cdot\vec{H}_s) + \Delta g^<(p,p_0), \\ g^>(p,p_0) &= 2\pi\delta(p_0 - \epsilon_p + \gamma\vec{s}\cdot\vec{H}_s)[1 - f(p_0 + \gamma\vec{s}\cdot\vec{H}_s)] - \Delta g^<(p,p_0) , \end{aligned} \tag{2.19}$$

and

$$G^<(p,p_0) = 2\pi\delta(p_0 - E_p + \gamma\vec{S}\cdot\vec{H}_d) F(p_0 + \gamma\vec{S}\cdot\vec{H}_d) + \Delta G^<(p,p_0) \tag{2.20}$$

and similarly for $G^>$. Note that the spectral weight function implied by (2.19) and (2.20) is a δ function which is correct to only zeroth order in J. It follows rather trivally from the definition of $G^<$ and $g^<$ that

$$\begin{aligned} &\mathrm{Tr}_S\, \gamma\vec{S}\sum_{\vec{p}p_0} \Delta G^<(p,p_0) = \vec{M}_d , \\ &\mathrm{Tr}_s \gamma\vec{s}\sum_{\vec{p}p_0} \Delta g^>(p,p_0) = \vec{M}_s . \end{aligned} \tag{2.21}$$

The advantage of the decomposition (2.19) is that if $\Delta G^<$ and $\Delta g^<$ are set equal to zero, a large class of terms on the left of the GKB identically vanish.

D. Derivation of the Bloch equations

We are now in a position to derive the Bloch equation with little further effort. Consider first the right-hand side of the conduction-electron equation. It is expanded using the decomposition (2.11) in terms $\delta G^<$ and $\delta g^<$, just as in Eq. (5.9) of I. The only difference is that when one multiplies by $\gamma\vec{s}$ and takes the trace, while summing over $\vec{p}$ and p_0, one must use (2.13) and (2.14) of this paper instead of (4.8) and (4.15) of I. The right-hand side of the conduction-electron Bloch equation is therefore

$$\begin{aligned} &-(1/T_{sd})(\vec{M}_s - \chi_{s0}^0 \vec{H}_s) \\ &+(1/T_{ds})(\vec{M}_d - \chi_{d0}^0\{\vec{H}_d + 2\alpha_2^0[\vec{M}_s - \chi_{s0}^0(\vec{H}_s - H_d)]\}) \end{aligned} \tag{2.22}$$

instead of the last two lines of (5.18) in I. The quantities T_{ds} and T_{sd} are the same as before:

$$1/T_{sd} = 2\pi(J/n)^2 \rho N \tfrac{1}{3}S(S+1) , \tag{2.23a}$$

$$1/T_{ds} = \pi(\gamma\rho J/n)^2 k_B T , \tag{2.23b}$$

and are in the ratio of the bare susceptibilities

$$\chi_{s0}^0/T_{sd} = \chi_{d0}^0/T_{d1} . \tag{2.24}$$

Similarly, the right-hand side of the local spin equation becomes

$$\begin{aligned} &-(1/T_{ds})(\vec{M}_d - \chi_{d0}^0\{\vec{H}_d + 2\alpha_2^0[\vec{M}_s - \chi_{s0}^0(\vec{H}_s - \vec{H}_d)]\}) \\ &+(1/T_{sd})(\vec{M}_s - \chi_{s0}^0 \vec{H}_s) \end{aligned} \tag{2.25}$$

instead of (5.25) of I.

The argument for the left-hand side of the conduction-electron equation is slightly more complicated, but equally straightforward. The only difference from I is (aside from setting $\alpha_1 = 0$ and replacing H by H_s) that now we have to evaluate the additional term [see Eqs. (2.6)–(2.9)]

$$i[\sigma_{H_d - H_s}, g^<]_{Ft} + i[\sigma^<_{H_d - H_s}, g]_{Ft} \tag{2.26}$$

and add them to the GKB of I [the reader should refer to Eqs. (6.3) and (6.10)–(6.13)]. It is shown in the Appendix however that the terms are small, and actually vanish within our approximation of a constant density of states and momentum-independent J. In other words there are *no* additional terms on the left-hand side of the conduction-electron Bloch equation. Therefore the left-hand side of the Bloch-like equation arising from the GKB for the conduction electrons is

$$\begin{aligned} &\frac{\partial\vec{M}_s}{\partial t} + \gamma(\vec{H}_s + \alpha_2^0\vec{M}_d)\times\vec{M}_s - D\nabla^2(\vec{M}_s - \chi_{s0}^0\vec{H}_s) \\ &\quad + \alpha_2^0\chi_{s0}^0\left(\frac{\partial\vec{M}_d}{\partial t} + \gamma\vec{H}_s\times\vec{M}_d\right) , \end{aligned} \tag{2.27}$$

which also equals

$$\frac{\partial\vec{M}_s}{\partial t} + \gamma\vec{H}_s\times\vec{M}_s + \gamma\alpha_2^0\vec{M}_d\times[(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \chi_{s0}^0\vec{H}_d]$$

$$+\alpha_2^0\chi_{s0}^0\left(\frac{\partial\vec{M}_d}{\partial t}+\gamma\vec{H}_d\times\vec{M}_d\right)-D\nabla^2(M_s-\chi_{s0}^0H_s)\ . \tag{2.28}$$

Equation (2.27) is taken directly from (7.10) of I, with $\alpha_1^0=0$ and $H=H_s$; Eq. (2.28) is the result of rearranging the terms for a purpose which will be obvious later.

The left-hand side of the Bloch equation for the local spins is trivial to write down, because according to (2.3)–(2.5) the only difference between the various self-energy functionals for the localized spins and those of I is the replacement of $\vec{M}_s$ by $\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)$. Therefore, since $\vec{M}_s$ appears in the localized spin equations only through the self-energy functionals, one can simply make the above replacement directly in the left-hand side of the Bloch equation for the local spins [i.e., (6.51) in I], obtaining

$$\left(\frac{\partial\vec{M}_d}{\partial t}+\gamma\vec{H}_d\times\vec{M}_d\right)(1-\alpha_2^0\chi_{s0}^0)$$
$$+\gamma\alpha_2^0(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\chi_{s0}^0\vec{H}_d]\times\vec{M}_d\ . \tag{2.29}$$

In fact the right-hand side of the local spin equation [see (2.25)] could also have been derived by replacing $\vec{M}_s$ by $\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)$. We now have two equations

(2.28) = (2.22)

and

(2.29) = (2.25)

which may be manipulated into the standard form for Bloch equations. This is done by using the second quality above to eliminate the last term in (2.28). We find that

$$\frac{\partial\vec{M}_s}{\partial t}+\gamma\vec{H}_s\times\vec{M}_s-D\nabla^2(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\gamma\alpha_2^0\nu_{ss}\vec{M}_d\times(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\gamma\alpha_2^0\nu_{ds}\vec{M}_d\times(\chi_{s0}^0\vec{H}_d)=$$
$$-\frac{\nu_{ss}}{T_{sd}}\delta\vec{M}_s+\frac{\nu_{ds}}{T_{ds}}\delta\vec{M}_d-\frac{1}{T_{sl}}\delta\vec{M}_s \tag{2.30}$$

and

$$\frac{\partial\vec{M}_d}{\partial t}+\gamma\vec{H}_d\times\vec{M}_d+\gamma\alpha_2^0\nu_{dd}(\chi_{s0}^0\vec{H}_d)\times\vec{M}_d+\gamma\alpha_2^0\nu_{sd}(\vec{M}_s-\chi_{s0}^0\vec{H}_s)\times\vec{M}_d=-\frac{\nu_{dd}}{T_{ds}}\delta\vec{M}_d+\frac{\nu_{sd}}{T_{sd}}\delta\vec{M}_s\ , \tag{2.31}$$

where

$$\delta\vec{M}_s=\vec{M}_s-\chi_{s0}^0\vec{H}_s\ , \tag{2.32}$$

$$\delta\vec{M}_d=\vec{M}_d-\chi_{d0}^0\{\vec{H}_d+2\alpha_2^0[\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)]\}\ , \tag{2.33}$$

and

$$\nu=\nu_{ss}=\nu_{sd}=\nu_{ds}=\nu_{dd}=(1-\alpha_2^0\chi_{s0}^0)^{-1}\ , \tag{2.34}$$

and $1/T_{sl}=0$. We have included the $1/T_{sl}$ term and the possibility of distinct ν's in anticipation of the requirements of Sec. III so that these long equations will not have to be reproduced again there.

We finally go from the ficticious problem of equal γ's and two different H's by making the replacements (2.1a) and (2.1c). These replacements would be sufficient to give the correct solution to the real problem if one remembered that χ_{s0}^0 and χ_{d0}^0 would still be given by (2.16) and thus would not be the bare susceptibilities for the real problem. However it will be eventually less confusing if we also redefine the susceptibilities,

$$\chi_{s0}=\tfrac{1}{2}\rho(\hbar\gamma_s)^2\ , \tag{2.35a}$$

$$\chi_{d0}=\tfrac{1}{3}S(S+1)N(\hbar\gamma_d)^2\ . \tag{2.35b}$$

Similarly we redefine α_2 so that the dimensionless parameter $\alpha_2^0\chi_{s0}^0$ (and hence the various ν's) are not affected:

$$\alpha_2=(J/n)^2(\rho/\gamma_s)^2\ln(k_BT/D) \tag{2.36a}$$

and we define the first-order exchange constant α_1 (cf. the definition in I) as

$$\alpha_1=J/n\gamma_s\gamma_d\ . \tag{2.36b}$$

The Bloch equations for the real problem are then

$$\frac{1}{\gamma_s}\frac{\partial\vec{M}_s}{\partial t}+\vec{H}\times\vec{M}_s-\frac{D}{\gamma_s}\nabla^2[\vec{M}_s-\chi_{s0}(\vec{H}+\alpha_1\vec{M}_d)]+\alpha_1\vec{M}_d\times\vec{M}_s+\nu_{ss}\frac{\gamma_s}{\gamma_d}\alpha_2\vec{M}_d\times[\vec{M}_s-\chi_{s0}(\vec{H}+\alpha_1\vec{M}_d)]+\nu_{ds}\alpha_2\vec{M}_d$$
$$\times\chi_{s0}(\vec{H}+\alpha_1M_s)=-\frac{\nu_{ss}}{T_{sd}}\frac{\delta\vec{M}_s}{\gamma_s}+\frac{\nu_{ds}}{T_{ds}}\frac{\delta\vec{M}_d}{\gamma_d}-\frac{1}{T_{sl}}\frac{\delta\vec{M}_s}{\gamma_s} \tag{2.37}$$

and

$$\frac{1}{\gamma_d}\frac{\partial\vec{M}_d}{\partial t}+\vec{H}\times\vec{M}_d+\nu_{dd}\alpha_2\chi_{s0}(\vec{H}+\alpha_1\vec{M}_s)\times\vec{M}_d+\alpha_1\vec{M}_s\times\vec{M}_d+\nu_{sd}\frac{\gamma_s}{\gamma_d}\alpha_2[\vec{M}_s-\chi_{s0}(\vec{H}+\alpha_1\vec{M}_d)]\times\vec{M}_d$$

$$= -\frac{\nu_{dd}}{T_{ds}}\frac{\delta\vec{M}_d}{\gamma_d} + \frac{\nu_{sd}}{T_{sd}}\frac{\delta\vec{M}_s}{\gamma_s}\ , \tag{2.38}$$

where

$$\delta\vec{M}_s = \vec{M}_s - \chi_{s0}(\vec{H} + \alpha_1\vec{M}_d)\ , \tag{2.39}$$

$$\delta\vec{M}_d = \vec{M}_d - \chi_{d0}\left\{\vec{H} + \alpha_1\vec{M}_s + 2\alpha_2\frac{\gamma_s}{\gamma_d}\left[\vec{M}_s - \chi_{s0}\left((\vec{H} + \alpha_1\vec{M}_s) - \frac{\gamma_d}{\gamma_s}(\vec{H} + \alpha_1\vec{M}_s)\right)\right]\right\}. \tag{2.40}$$

Note again that for the purposes of this paragraph (2.39), (2.40), (2.35), and (2.36) supercede (2.32), (2.33), (2.16) and (2.15), respectively. Equations (2.37) and (2.38) have been written in a form which is easily seen to be equivalent to that derived in I when $\gamma_s = \gamma_d$, the only differences being terms that are of order $\alpha_1\alpha_2$ or α_2^2 and hence negligible by assumption both here and in I. For practical use, however, it is usually more convenient to redefine the lifetimes and destination vectors so that the right-hand sides of (2.37) and (2.38) may be, respectively, written as

$$-\frac{\nu}{\gamma_s}\left(\frac{\gamma_s}{\gamma_d}\right)^2\frac{\chi_d'}{\chi_s}\frac{1}{T_{ds}}[\vec{M}_s - \chi_s'(\vec{H} + \alpha_1\vec{M}_d)]$$

$$+\frac{1}{\gamma_d}\frac{1}{T_{ds}}[\vec{M}_d - \chi_d'(\vec{H} + \alpha_1\vec{M}_s)] \tag{2.41}$$

and

$$-\frac{\nu}{\gamma_d}\frac{1}{T_{ds}}[\vec{M}_d - \chi_d'(\vec{H} + \alpha_1\vec{M}_s)]$$

$$+\frac{\nu}{\gamma_s}\left(\frac{\gamma_s}{\gamma_d}\right)^2\frac{\chi_d'}{\chi_s}\frac{1}{T_{ds}}[\vec{M}_s - \chi_s'(\vec{H} + \alpha_1\vec{M}_d)]. \tag{2.42}$$

This is the form espoused by Schultz *et al.*, whose definitions are such that for example our quantity $\chi_{d0}(1 + 2\alpha_2\chi_{s0})$ is called χ_{d0} by them, with other correspondences fairly obvious. If the left-hand sides of (2.37) and (2.38) are for the transverse components of $\vec{M}_s$ and $\vec{M}_d$ are linearized, then they can also be cast into the form of Ref. 7. We leave the demonstration of this as an exercise for any interested reader, along with the warning that the quantities which Schultz *et al.* denote by γ_s and γ_d are not the respective gyromagnetic ratios (*s* and *d* spins are unambiguously defined by the exchange Hamiltonian in I), but are rather the coefficients of the $\vec{H}\times\vec{M}$ terms in their form for writing the equations.

III. FORM OF EQUATIONS WITH DIRECT RELAXATION OF THE LOCAL SPINS

In real systems, there may exist mechanisms by which both the conduction and local spins relax directly and hence give up angular momentum to the solid as a whole, although their size is not well documented, not to speak of concentration or temperature dependence. This state of affairs led us in I to discuss the direct relaxation only generally with the aid of two assumptions: (a) that the direct relaxation was sufficiently weak that only the scattering functions of $\sigma^<, \gamma, \Sigma, \Gamma$ on the right-hand side of the GKB need to be modified, and that the spectral weight functions and the self-energy functions on the left-hand side of the GKB go through unchanged and (b) that the decompositions (2.11) with $\delta G^<$ and $\delta g^<$ equal to zero still cause the right-hand side of the GKB to vanish, implying that the only effect of the direct relaxation was to add terms $-\delta g^</T_{sl}$ and $-\delta G^</T_{dl}$ to the right-hand sides of the respective GKB. The second assumption was checked by reference to a specific model for $1/T_{sl}$. The only difference between equal and nonequal γ values with respect to these terms is that now we must use (2.13) and (2.14) (we adopt the ficticious model of equal γ's but nonequal H's for the two species as a calculational device in this section as well). Therefore, if the assumptions (a) and (b) are again adopted, we must add terms $-\delta M_s/T_{sl}$ and $-\delta M_d/T_{dl}$ to (2.22) and (2.25), respectively. Then if one combines (2.22) and (2.25) as before to yield Bloch equations, we find that the right-hand sides of (2.30) and (2.31) are to be replaced, respectively, by

$$-\left(\frac{\nu}{T_{sd}} + \frac{1}{T_{sl}}\right)\delta\vec{M}_s + \nu\left(\frac{1}{T_{ds}} + \frac{\alpha_2\chi_{s0}}{T_{dl}}\right)\delta\vec{M}_d \tag{3.1}$$

and

$$-\nu\left(\frac{1}{T_{ds}} + \frac{1}{T_{dl}}\right)\delta\vec{M}_d + \frac{\nu}{T_{sd}}\delta\vec{M}_s\ , \tag{3.2}$$

with $\delta\vec{M}_s$ and $\delta\vec{M}_d$ given by (2.32). To order J^2, (3.1) and (3.2) are identical to the right-hand sides of (7.16) and (7.17) of I, except for the redefinition of $\delta\vec{M}_s$ and $\delta\vec{M}_d$.

We are now in a somewhat embarrassing position, because according to the Onsager reciprocal relations the coefficient of $\vec{H}_d$ in Eq. (3.1) for $\partial\vec{M}_s/\partial t$ must be the same as the coefficient of $\vec{H}_s$ in Eq. (3.2) for $\partial\vec{M}_d/\partial t$. These coefficients are readily written down, noting that both $\delta\vec{M}_s$ and $\delta\vec{M}_d$ contain $\vec{H}_s$. The former coefficient is

$$-\nu\chi_{d0}\left(\frac{1}{T_{ds}}+\frac{\alpha_2\chi_{s0}}{T_{dl}}\right)(1+2\alpha\chi_{s0}) \ , \tag{3.3}$$

while the latter is

$$-\nu\chi_{d0}\frac{1}{T_{ds}}(1+2\alpha_2\chi_{s0})-\nu\chi_{d0}\frac{2\alpha_2\chi_{s0}}{T_{dl}} \ . \tag{3.4}$$

Even if terms of order α_2^2 are neglected, (3.3) and (3.4) differ[18] by the amount

$$-\nu\chi_{d0}\alpha_2\chi_{s0}/T_{dl} \ . \tag{3.5}$$

Thus we are faced with having to explain an obvious error.

On one level the explanation is simple. It is that (3.5) is very small and indeed is of the same order as the self-energy corrections which by assumption (a) were neglected. Therefore the term (3.5) is not significant in that there must be other terms of the same order which cancel it. As pointed out below Eq. (7.16) in I, a term this size is "hardly worth considering."

On the other hand one can ask what are the other terms not included. It would be comforting to produce them since one must be very suspicious of approximation schemes which fail to reproduce exact symmetries (in this case time-reversal invariance) exactly. In what follows we briefly consider the additional terms.

We would like to choose a model for $1/T_{dl}$ in which we have to do as little additional calculation as possible, since the real mechanism is not known, at least to us. To this end, we suppose that the local spins are coupled to a ficticious extra set of conduction electrons of spin $\tilde{s}$ with an exchange interaction $-(\tilde{J}/n)S\tilde{s}$, and assume that these are forced to be in thermal equilibrium of a temperature T. Since we assume a structureless $\tilde{J}$ and density of states, we can eliminate all extra parameters in terms of $1/T_{dl}$, so that the artificiality of this model is of no particular consequence, as it is typical of any spin flip coupling to any sort of density fluctuation excitation. It seems as if the mechanism recently considered by Yafet[19] would fit into this category, although there would be extra terms produced by the tranformation between the Anderson and the s-d exchange models. On the other hand, if $1/T_{dl}$ is due to some sort of static or frozen inhomogeneity, then it can be treated by the methods of Sec. IV.

We assign the ficticious coupling $\tilde{J}$, a bare susceptibility $\tilde{\chi}_{s0}$, and a second-order exchange coupling $\tilde{\alpha}_2$; we assume that $\tilde{\alpha}_1=0$ and take them to be acted on by a zero effective field ($\tilde{H}_{\tilde{s}}=0$) because we wish to introduce relaxation but not polarization effects.

First it is clear that the left-hand side of the conduction-electron equation is still given by (2.27) or (2.28) because there is no direct coupling between the conduction electrons and the fake conduction-electron field. (We defer discussion of $1/T_{sl}$ for the time being.) On the other hand, the left-hand side of the equation for the d spins is modified because these couple not only to the conduction electrons but to the extra field. Therefore (2.29) becomes

$$\begin{aligned}\left(\frac{\partial\vec{M}_d}{\partial t}+\gamma\vec{H}_d\times\vec{M}_d\right)&(1-\alpha_2^0\chi_{s0}^0-\tilde{\alpha}_2\tilde{\chi}_{\tilde{s}0})\\ &+\gamma\alpha_2^0[(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\chi_{s0}^0\vec{H}_d]\times\vec{M}_d\\ &+\gamma\tilde{\alpha}_2[(\vec{\tilde{M}}_{\tilde{s}}-\tilde{\chi}_{s0}\vec{\tilde{H}}_{\tilde{s}})+\tilde{\chi}_{\tilde{s}0}\vec{H}_d]\times\vec{M}_d \ .\end{aligned} \tag{3.6}$$

We note that in this model $1/T_{dl}\propto(\tilde{J}\tilde{\chi}_{\tilde{s}0})^2$, $1/T_{ds}\propto(J\chi_{s0}^0)^2$, $\tilde{\alpha}_2\tilde{\chi}_{\tilde{s}0}\propto(\tilde{J}\tilde{\chi}_{\tilde{s}0})^2$, and $\alpha_2^0\chi_{s0}^0\propto(J\chi_{s0}^0)^2$, so that

$$\tilde{\alpha}_2\tilde{\chi}_{\tilde{s}0}=\alpha_2^0\chi_{s0}^0(T_{ds}/T_{dl}) \ , \tag{3.7}$$

a relation which will be used repeatedly to eliminate the coupling parameters of the T_{dl} field wherever they occur. The expression (3.6) therefore becomes

$$\left(\frac{\partial\vec{M}_d}{\partial t}+\gamma\vec{H}_d\times\vec{M}_d\right)\left[1-\alpha_2^0\chi_{s0}^0\left(1+\frac{T_{ds}}{T_{dl}}\right)\right]+\gamma\alpha_2^0\left[(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\chi_{s0}^0\left(1+\frac{T_{ds}}{T_{dl}}\right)\vec{H}_d\right]\times\vec{M}_d \ , \tag{3.8}$$

where we have set $\tilde{M}_{\tilde{s}}=\tilde{H}_{\tilde{s}}=0$, as mentioned earlier. The right-hand side of the conduction electron (2.22) is modified only because the local spin spectral weight function, and hence the moment of $\delta G^<$, is changed by the extra field. Hence (2.22) becomes

$$\begin{aligned}-\frac{1}{T_{sd}}(\vec{M}_s-\chi_{s0}^0\vec{H}_s)+\frac{1}{T_{ds}}&(\vec{M}_d-\chi_{d0}^0\{\vec{H}_d+2\alpha_2^0[\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)]+2\tilde{\alpha}_2[\vec{\tilde{M}}_{\tilde{s}}-\tilde{\chi}_{\tilde{s}0}(\vec{\tilde{H}}_{\tilde{s}}-\vec{H}_d)]\})\\ &=-\frac{1}{T_{sd}}(\vec{M}_s-\tilde{\chi}_{s0}^0\vec{H}_s)+\frac{1}{T_{ds}}\left(\vec{M}_d-\chi_{d0}^0\left\{\vec{H}_d+2\alpha_2^0[\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)]+2\alpha_2^0\chi_{s0}^0\frac{T_{ds}}{T_{dl}}\vec{H}_d\right\}\right) .\end{aligned} \tag{3.9}$$

Finally, the right-hand side of the equation for the local spins differs from (2.25) both in the modification of the destination vector and in the additional rate:

$$-\left(\frac{1}{T_{ds}}+\frac{1}{T_{dl}}\right)\left(\vec{M}_d-\chi_{d0}^0\left\{\vec{H}_d+2\alpha_2^0[\vec{M}_s-\chi_{s0}^0(\vec{H}_s-\vec{H}_d)]+2\alpha_2^0\frac{T_{ds}}{T_{dl}}\chi_{s0}^0\vec{H}_d\right\}\right)+\frac{1}{T_{sd}}(\vec{M}_s-\chi_{s0}^0\vec{H}_s) \ . \tag{3.10}$$

For static fields the left-hand sides vanish; equating (3.9) and (3.10) to zero shows that (2.18b) must be replaced by

$$\chi_d' = \chi_{d0}^0[1 + 2\alpha_2^0\chi_{s0}^0(1 + T_{d1}/T_{d1})] , \qquad (3.11)$$

which expresses the fact that the direct relaxation mechanism affects the static susceptibility as well as the dynamic.

We thus have the required equations, (2.28) = (3.9) and (3.8) = (3.10). As before we combine these equations, using the second one to eliminate $\partial\vec{M}_d/\partial t$ in the left-hand side of the first. When this is done, we find with no further approximation, that (2.30) and (2.31) are still valid, except now (2.33) is replaced by

$$\delta\vec{M}_d = \vec{M}_d - H_{d0}^0\{\vec{H}_d + 2\alpha_2^0[\vec{M}_s - \chi_{s0}^0(\vec{H}_s - \vec{H}_d)] + 2\alpha_2^0\chi_{s0}^0(T_{ds}/T_{d1})\vec{H}_d\} \qquad (3.12)$$

and (2.34) is replaced by

$$\nu_{ss} = (1 - \alpha_2\chi_{s0}T_{ds}/T_{d1})/\Delta ,$$
$$\nu_{sd} = \nu_{ds} = 1/\Delta , \qquad (3.13)$$
$$\nu_{dd} = (1 + T_{ds}/T_{d1})/\Delta ,$$

with

$$\Delta = 1 - \alpha_2\chi_{s0}(1 + T_{ds}/T_{d1}) .$$

One notes that the largest effect of the direct relaxation, as expected, is the extra relaxation caused by the T_{ds}/T_{d1} term in ν_{dd} on the right-hand side of the local spin equation. Less obvious, but also important is the extra g shift arising from this same term in ν on the *left*-hand side of the local spin equation. This g shift is logarithmic, and does not go away when the s and d equations are added with $\gamma_s = \gamma_d$ as does the g shift predicted long ago by Spencer and Doniach.[20] The magnitude of the shift is approximately

$$\Delta\gamma/\gamma \simeq \alpha_2\chi_{s0}T_{ds}/T_{d1} , \qquad (3.14)$$

and since it depends on a second-order interaction (it is not a mean-field effect) it would seem to be a signature of a direct d-relaxation mechanism mediated by a coupling to density fluctuation interactions.

It is simple to show that the coefficient of H_d in the s equation is now *exactly* identical to the coefficient of H_s is the d equation, so that with due care, we note that our approximation scheme satisfies the reciprocal relation exactly. Note that this symmetry occurs as it must both in the cross-product terms on the left-hand side of the equation and in the relaxation terms on the right-hand sides of the equations.

We have not yet mentioned the relaxation of the s spins to the lattice. This is because at least in this the model presented in I, there are no particular difficulties or inconsistencies associated with it, and indeed with a wave-vector–independent interaction and constant density of states the self-energy effects neglected in I make no contribution. Therefore it is treated properly simply by adding a $1/T_{sl}$ just as we did in Eq. (2.30).

Finally, as before, there is no difficulty in going from our ficticious problem of different H's and identical γ's to the real problem of different γ's and identical H's. Equations (2.37) and (2.38) remain valid, except that now the γ's are given by (3.13) instead of (2.34) and $\delta\vec{M}_d$ is given by

$$\delta\vec{M}_d = \vec{M}_d - \chi_{d0}\left(\vec{H} + \alpha_1\vec{M}_s + 2\alpha_2\frac{\gamma_s}{\gamma_d}\left\{\vec{M}_s - \chi_{s0}\left[(\vec{H} + \alpha_1\vec{M}_d) - \frac{\gamma_d}{\gamma_s}\left(1 + \frac{T_{ds}}{T_{d1}}\right)(H + \alpha_1 M_s)\right]\right\}\right) \qquad (3.15)$$

and $\delta\vec{M}_s$ given by (2.39).

IV. HYPERFINE INTERACTION AND FROZEN-IN INHOMOGENIETIES

The Bloch equations which we presented in Ref. 1 a long while ago included the effect of the hyperfine interaction between the nuclear spin and the localized or d electron spins. Here we give the derivation of a slightly generalized form of these equations.

We assume, as an aid to starting the discussion, that the hyperfine interaction is of the form $A\vec{S}\cdot\vec{I}$, where $\vec{I}$ is the nuclear spin. We take the dc applied magnetic field H_0 to be in the z direction and write the hyperfine Hamiltonian above as $\mathcal{H}_{hf} = AS_zI_z + \delta\mathcal{H}_{hf}$. The essential approximation is to assume that $A/\gamma_d H_0 \ll 1$ in which case the spin-flip term $\delta\mathcal{H}_f$ can be neglected. The quantity A/γ_d is typically of order of 10's of gauss, while H_0 is of the order of 10 kG. Moreover, since the expectation value of $\delta\mathcal{H}_{hf}$ in any eigenstate of the Hamiltonian obtained by neglecting $\delta\mathcal{H}_{hf}$ is zero, we may expect any shift or broadening of the resonant frequency produced by it to be second order in this neglected quantity, that is, $\delta\omega_R/\omega_R \sim (A/\gamma H_0)^2$, and is thus quite small indeed. Therefore, the effects of the hyperfine interaction are to produce an effective field $H_n = -An\hat{z}$, where n is the azimuthal quantum number for the nuclear spin; a certain fraction f_n of the local spins feel the field H_n corresponding to each n; because of the large difference in nuclear and electronic gyromagnetic ratios it is a good approximation to take all the f_n's equal: $f_n = (2I+1)^{-1}$. Note that in general $\sum_n H_n = 0$; $\sum_n f_n = 1$.

It is evident now that as long as we are willing to approximate the hyperfine interaction by an effective field in the z direction, we are not forced to assume a hyperfine Hamiltonian $A\vec{S}\cdot\vec{I}$, and it can be generalized to include an anisotropic hyperfine interaction without affecting our scheme.

Before proceeding, we mention something that should become quite clear shortly, that the method we use can be applied not only to the hyperfine interaction, but also to problem of direct relaxation of the local spins (the $1/T_{d1}$ problem) provided that such relaxation arises from some sort of frozen-in or static inhomogeneities or impurities whose effect is to produce random effective fields which act on the local spins. In this case the subscript n above would probably be a continuous variable, and f_n would be the probability distribution function for the effective field H_n (or more precisely its projection in the direction of $\vec{H}_0$). The discussion which follows can therefore be regarded as a complement to that of Sec. III where the direct relaxation was assumed to arise from a dynamic spin flip mechanism.

Actually it requires no additional calculation to write down the appropriate Bloch-like equations. This is because we have already derived in Sec. II a set of coupled equations in which the local spins feel a different field from the conduction electrons, and we need only to generalize these to include a number of classes of local spins with magnetizations $\vec{M}_n$ with $\vec{M}_d = \sum_n \vec{M}_n$) each of which feel their own distinct hyperfine (or other) field H_n. This generalization is accomplished simply by inspection of Eqs. (2.22), (2.25), (2.27), and (2.29) as well as (2.32) and (2.33), and we find that

$$\frac{\partial}{\partial t}\vec{M}_s + \gamma\vec{H}_s\times\vec{M}_s - D\nabla^2(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \gamma\alpha_2^0\vec{M}_d\times[\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \chi_{s0}^0\vec{H}_d]$$
$$+\gamma\alpha^0 2\chi_{s0}^0\sum_n \vec{M}_n\times\vec{H}_n + \alpha_2^0\chi_{s0}^0\sum_n\left(\frac{\partial\vec{M}_n}{\partial t} + \gamma(\vec{H}_d+\vec{H}_n)\times\vec{M}_n\right) = \frac{-1}{T_{sd}}\delta\vec{M}_s + \frac{1}{T_{ds}}\delta\vec{M}_d \tag{4.1}$$

and

$$\left(\frac{\partial}{\partial t}\vec{M}_n + \gamma(\vec{H}_d+\vec{H}_n)\times\vec{M}_n\right)(1-\alpha_2^0\chi_{s0}^0) + \gamma\alpha_2^0[(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \chi_{s0}^0\vec{H}_d]\times\vec{M}_n + \gamma\alpha_2^0\chi_{s0}^0\vec{H}_n\times\vec{M}_n$$
$$= -(1/T_{ds})(\vec{M}_n - f_n\chi_{d0}^0\{\vec{H}_d + \vec{H}_n + 2\alpha_2^0[M_s - \chi_{s0}^0(\vec{H}_s - \vec{H}_d - \vec{H}_n)]\}) + (1/T_{sd})f_n\delta\vec{M}_s \ . \tag{4.2}$$

Note that (4.1) is identical to (2.7) as it must be because the conduction electrons do not directly feel the hyperfine field, the counter-term involving $\vec{M}_n\times\vec{H}_n$ has been added (and subtracted) in (4.1) in order to facilitate the manipulation below, which is the use of (4.2) to eliminate the term in large parentheses in (4.1). We find that (4.1) and (4.2) can be written exactly as

$$\frac{\partial}{\partial t}\vec{M}_s + \gamma\vec{H}_s\times\vec{M}_s - D\nabla^2\delta\vec{M}_s + \nu\gamma\alpha_2^0\vec{M}_d\times[(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \chi_{s0}^0\vec{H}_d] + \nu\gamma\alpha_2^0\chi_{s0}^0\sum_n\vec{M}_n\times\vec{H}_n = -\frac{\nu}{T_{sd}}\delta\vec{M}_s + \frac{\nu}{T_{ds}}\delta\vec{M}_d \tag{4.3}$$

and

$$\frac{\partial}{\partial t}\vec{M}_n + \gamma(\vec{H}_d+\vec{H}_n)\times\vec{M}_n + \nu\gamma\alpha_2^0[(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \chi_{s0}^0\vec{H}_d]\times\vec{M}_n + \nu\gamma\alpha_2^0\chi_{s0}^0\vec{H}_n\times\vec{M}_n$$
$$= -\nu/T_{ds}(\vec{M}_n - f_n\chi_{d0}^0\{\vec{H}_d + \vec{H}_n + 2\alpha_2^0[\vec{M}_s - \chi_{s0}^0(\vec{H}_s - \vec{H}_d - \vec{H}_n)]\}) + (\nu/T_{sd})f_n(\vec{M}_s - \chi_{s0}^0\vec{H}_s) \ , \tag{4.4}$$

where $\delta\vec{M}_s$, $\delta\vec{M}_d$, and ν are given by (2.32), (2.33), and (2.34) respectively.

The above equations, (4.3) and (4.4), are $n+1$ in number and represent the most general description of the effect of the hyperfine interaction that we shall present. In what follows we consider special cases. Before doing so we mention that for the case $I = \frac{1}{2}$ these equations can be solved by the methods of Ref. 1, and for a general spin by the methods developed and applied by Pifer and Longo[21] to a similar set of equations.

A reminder: to go to the physical problem one must make replacements and redefinitions analogous to (2.1) and (2.35) for each hyperfine component, that is, in addition to (2.1) and (2.35), we let

$$\vec{M}_n \to (\gamma/\gamma_d)\vec{M}_n \ , \tag{4.5a}$$

$$\vec{H}_n \to (\gamma_d/\gamma)\vec{H}_n \ . \tag{4.5b}$$

The reader should have no trouble writing down the resulting equations by inspection, because except for the slight added complication of separate hyperfine components, those equations are virtually identical with Eqs. (2.37)–(2.40).

For $\gamma_s = \gamma_d$ we will write the equations for comparison with Ref. 1:

$$\frac{\partial \vec{M}_s}{\partial t} + \gamma[\vec{H} + (\alpha_1+\alpha_2)\vec{M}_d]\times\vec{M}_s - D\nabla^2\delta\vec{M}_s + \gamma\alpha_2\chi_{s0}\sum_n \vec{M}_n\times\vec{H}_n = -\frac{1}{T_{sd}}\delta\vec{M}_s + \frac{1}{T_{ds}}\delta\vec{M}_d - \frac{1}{T_{sl}}\delta\vec{M}_s \quad , \tag{4.6}$$

where we have put the T_{sl} term back in, and

$$\frac{\partial\vec{M}_n}{\partial t} + \gamma[\vec{H}_d + (\alpha_1+\alpha_2)\vec{M}_s]\times\vec{M}_n + \gamma\alpha_2\chi_{s0}\vec{H}_n\times\vec{M}_n = -\frac{1}{T_{ds}}\{\vec{M}_n - f_n\chi_{d0}[\vec{H} + (\alpha_1+2\alpha_2)\vec{M}_s]\} + \frac{1}{T_{sd}}f_n\delta\vec{M}_s \quad . \tag{4.7}$$

In writing (4.6) and (4.7), which are valid for the transverse components of $\vec{M}_n$ and $\vec{M}_s$, we have set $\nu = 1$, because it only appears multiplying terms already of order J^2, and have noted that the field $\vec{H}_n$ is in the $\hat{z}$ direction. Equations (4.6) and (4.7) differ from (1a) and (1b) of Ref. 1 in three respects: (i) the equations in Ref. 1 were for simplicity written for the case $I = \frac{1}{2}$ and for the same reason the diffusion term was omitted there; (ii) the numerical coefficients of α_2 in the relaxation terms differ; this represents the same error in Ref. 1 which was pointed out in I; and (iii) the last term in the left-hand side of (4.6) and (4.7) involving $\vec{H}_n\times\vec{M}_n$ was not present in the equations of Ref. 1. These last terms represent the interference between the hyperfine interaction and the second-order exchange interaction, an effect not included in the derivation leading to the equations of Ref. 1. Since these terms are second order in both the hyperfine splitting and J, they are in some sense fourth order in small quantities, and should be negligible in most applications.

We now show how Eqs. (4.3) and (4.4) can be reduced to a pair of equations [rather than $(n+1)$ equations] when the hyperfine interaction is weak. In the process we derive the results for the g shift and broadening presented in Ref. 1.

For simplicity we consider the equations for one circular polarization of the transverse components, letting $\vec{M}_n \propto (M_{nx}\hat{x} + M_{ny}\hat{y})e^{-i\omega t}$ and similarly for $\vec{M}_s$, $\vec{H}_s$, and $\vec{H}_n$. Equation (4.2) then becomes

$$\begin{aligned} &-i\omega M_n + i(\omega_d+\omega_n)(M_n - f_n\chi_d' H_d) - \nu\alpha_2^0\chi_d' f_n i(\omega_d+\omega_n)(M_s - \chi_{s0}^0 H_s) + \nu\alpha_2\chi_{s0}^0 i(\omega_d+\omega_n)(M_n - f_n\chi_d' H_d) \\ &\quad = -(\nu/T_{ds})(M_n - f_n\chi_{d0}^0\{H_d + 2\alpha_2^0[M_s - \chi_{s0}^0(H_s - H_d)]\}) + (\nu/T_{sd})f_n\delta M_s \quad , \end{aligned} \tag{4.8}$$

where $\hbar\omega_d = \gamma H_0$ and $\hbar\omega_n = \gamma H_n$. The quantity χ_d' is still given by (2.18b) as the solution of (4.1) and (4.2) in the static limit would show, although (2.17b) must now be replaced by $M_n = f_n\chi_d'(H_d + H_n)$. When summed over n, this yields

$$\begin{aligned} &-i\omega M_d + i\omega_d(M_d - \chi_d' H_d) + i\nu\sum_n \omega_n M_n - i\nu\alpha_2^0\chi_d'\omega_d(M_s - \chi_{s0}^0 H_s) + i\nu\alpha_2^0\chi_{s0}^0\omega_d(M_d - \chi_d' H_d) \\ &\quad = -(\nu/T_{ds})\delta M_d + (\nu/T_{sd})\delta M_s \quad . \end{aligned} \tag{4.9}$$

This contains the unknown quantity $\sum_n \omega_n M_n$; to find it we multiply (4.8) by ω_n and then sum over n. The resulting equation for $\sum_n \omega_n M_n$ of course then contains the higher-order average $\sum_n \omega_n^2 M_n$; since $\sum_n \omega_n = 0$, there is only one nonvanishing contraction of this average. Accordingly, to order of the hyperfine interaction squared, we may write

$$\sum_n \omega_n^2 M_n \simeq \sum_n f_n\omega_n^2 M_d \quad . \tag{4.10}$$

With this approximation, then we may write

$$\begin{aligned} &-i(\omega - \nu\omega_d)\sum_n \omega_n M_n + i\nu\langle\omega_n^2\rangle(M_d - \chi_d' H_d) \\ &\quad - i\gamma\alpha_2^0\chi_d'\langle\omega_n^2\rangle(M_s - \chi_s' H_s) = -(\nu/T_{ds})\sum_n \omega_n M_n \quad , \end{aligned} \tag{4.11}$$

where $\langle\omega_n^2\rangle \equiv \sum_n f_n\omega_n^2$. If we now make the definition

$$\frac{1}{\tilde{T}_{dl}} = \frac{\langle\omega_n^2\rangle}{-i\omega(1 - \alpha_2^0\chi_{s0}^0) + i\omega_d + 1/T_{ds}} \quad ; \tag{4.12}$$

then solving (4.9) for $\sum_n \omega_n M_n$ gives

$$\sum_n \omega_n M_n = -\frac{1}{\tilde{T}_{dl}}(M_d - \chi_d' H_d) + \frac{i\alpha_2\chi_d'}{\tilde{T}_{dl}}(M_s - \chi_s' H_s) \, . \tag{4.13}$$

Note that $\tilde{T}_{dl}$ is not just a lifetime, but has real and imaginary parts and is frequency and field dependent. Now (4.11) is used to eliminate the unknown quantity $\sum_n \omega_n M_n$ which occurs both in (4.9) and also in (4.3) for the conduction electrons. When this is done our equations may be cast into the forms

$$\frac{\partial\vec{M}_s}{\partial t} + \gamma\vec{H}_s\times\vec{M}_s - D\nabla^2\delta\vec{M}_s + \nu\gamma\alpha_2^0\vec{M}_d\times(\vec{M}_s - \chi_{s0}^0\vec{H}_s) + \nu\gamma\alpha_2^0\chi_{s0}^0\vec{M}_d\times\vec{H}_d = -\delta\vec{M}_s\frac{\mu_{ss}}{T_{sd}} + \delta\vec{M}_d\frac{\mu_{ds}}{T_{ds}} \quad , \tag{4.14}$$

$$\frac{\partial \vec{M}_d}{\partial t} + \gamma \vec{H}_d \times \vec{M}_d + \nu\gamma\alpha_2^0 \chi_{s0}^0 \vec{H}_d \times \vec{M}_d + \nu\gamma\alpha_2^0 (\vec{M}_s - \chi_{s0}^0 \vec{H}_s) \times \vec{M}_d = -\delta M_d \frac{\mu_{dd}}{T_{ds}} + \delta M_s \frac{\mu_{sd}}{T_{sd}} \quad , \tag{4.15}$$

where δM_s, δM_d, and ν are given by (2.32)–(2.34) and where

$$\begin{aligned} \mu_{dd} &= \nu(1 + T_{ds}/\tilde{T}_{dl}) \quad , \\ \mu_{sd} &= \nu[1 - \alpha_2^0 \chi_{s0}^0 (1 - 2\alpha_2^0 \chi_{s0}^0) T_{ds}/\tilde{T}_{dl}] \quad , \\ \mu_{ss} &= \nu[1 - (\alpha_2^0 \chi_{s0})^2 (1 - 2\alpha_2^0 \chi_{s0}^0) T_{ds}/\tilde{T}_{dl}] \quad , \\ \mu_{ds} &= \nu[1 + \alpha_2^0 \chi_{s0}^0 (T_{ds}/\tilde{T}_{dl})] \quad . \end{aligned} \tag{4.16}$$

For familiarity, we have used a mixed notation in writing (4.14) and (4.15): that is to say the right-hand sides cannot simply be Fourier transformed back to the space-time vector notation we use on the left-hand side because $1/\tilde{T}_{dl}$ and hence the μ's have a frequency and field dependence which at least in many cases of interest is as important as the linear frequency and field dependence on the left-hand side of say (4.9). Said another way, Eqs. (4.14) and (4.15) are not a set of coupled Bloch equations, and if regarded as differential equations they would be of infinite order. In practice this means that their solution for the transverse susceptibility (which is as trivially effected as if they were Bloch equations) will have a different field and frequency dependence from the usual solution of a set of coupled Bloch or Hasegawa equations. This difference would show up when the actual linewidth of the local spin resonance is larger than or comparable to $1/T_{ds}$, as is normally the case when the g values for the two species are quite different.

Equations (4.14) and (4.15) have been written in a way which the variable replacements (2.1) may be made by inspection, on comparison of (2.30) and (2.31) with (2.37) and (2.38). We will not write the resulting equation, but merely say that their left-hand sides are identical with the left-hand sides of (2.37) and (2.38), [with all the ν's equal to $(1 - \alpha_2 \chi_{s0})^{-1}$], while their right-hand sides may be obtained from (2.37) and (2.38) by replacing ν_{dd}, ν_{sd}, ν_{ds}, and ν_{ss} by μ_{dd}, μ_{sd}, μ_{ds}, and μ_{ss}, respectively. No change in the definition of the μ's accompanies the variable replacements, except that ω_d in the definition of $1/\tilde{T}_{dl}$ now becomes $\gamma_d(1 + \alpha_1 \chi_{s0}) H_0$.

In writing (4.16) we have made no further approximation; using the values given plus the definitions of $\delta\vec{M}_s$ and $\delta\vec{M}_d$ [Eqs. (2.32) and (2.33)] it is a straightforward matter to show that the coefficient of $\vec{H}_d$ in (4.14) is exactly the same as the coefficient of $\vec{H}_s$ in (4.15); that is to say the reciprocal relations are satisfied exactly. After this it is certainly permissible to drop all terms of order α^2 or higher, because $\alpha_1 \propto J$ and $\alpha_2 \propto J^2$, so that such terms are beyond the limits of accuracy of our basic diagrammatic perturbation expansion.

By far the most important term in (4.16) is μ_{dd}, which produces the additional lifetime (and possibly a g shift) implied by $1/\tilde{T}_{dl}$, which after the variable replacement (2.1) becomes

$$\frac{\nu}{\tilde{T}_{dl}} = \frac{\nu^2 \langle \omega_n^2 \rangle}{-i\omega + i\omega_d(1 + \alpha\chi_{s0}) + \nu/T_{ds}} \quad , \tag{4.17}$$

where $\omega_0 = \gamma_d H$ and $\alpha = \alpha_1 + \alpha_2$; in writing the term in parentheses of (4.17) we have neglected terms of order α^2 or higher; except for the factors of ν, this is identical to the result quoted in Ref. 1; the difference between ν and unity in the denominator arises from the interference between the $\langle \omega_n^2 \rangle$ fluctuation and the second-order exchange term, and are included here for the first time, although as pointed out earlier, this is expected to be a small effect in most cases, and will for simplicity be neglected in the following discussion. For the case of hyperfine interactions $\hbar^2 \langle \omega_n^2 \rangle$ is given by

$$\hbar^2 \langle \omega_n^2 \rangle = \tfrac{1}{3} A^2 I(I+1) \quad , \tag{4.18}$$

and for the case of inhomogeneities it equals γ_d^2 times the mean-square fluctuation of the $\hat{z}$ component of the frozen-in "effective field."

In the case $\gamma_s = \gamma_d$ one generally has bottlenecked conditions, and the joint conduction-electron resonance occurs at $\omega \cong \omega_0$, in which case

$$1/\tilde{T}_{dl} = \langle \omega_n^2 \rangle / (i\omega_0 \alpha \chi_{s0} + 1/T_{ds}) \quad , \tag{4.19}$$

which predicts an additional line broadening as well as a g shift, in exact agreement with Eq. (10) in Ref. 1. [To find the actual contribution of (4.19) to the bottlenecked linewidth one must multiply this by a susceptibility ratio, as a complete solution to (4.14) and (4.15) would show.]

On the other hand, when γ_s and γ_d are widely different, then the local spin resonance occurs at $\omega \simeq \omega_0[1 + (\alpha_1 + \alpha_2)\chi_{s0}]$ and we have (when exactly on resonance)

$$1/\tilde{T}_{dl} = \langle \omega_n^2 \rangle T_{ds} \quad , \tag{4.20}$$

so that in this case there is no additional shift, but only additional broadening, which of course represents just an intrinsic width [$\sim (\langle \omega_n^2 \rangle)^{1/2}$] motionally narrowed by the s-d exchange interaction; as the temperature is lowered the s-d flips are inhibited by the Fermi statistics, and $1/T_{dl}$ becomes larger. At sufficiently low temperature the approximation (4.10) breaks down, and one must solve Eqs. (4.3) and (4.4) exactly. A word of warning however: even at temperatures high enough that (4.20) is

valid on resonance, the ω and ω_d dependence of (4.17) are important, because the actual linewidth of the local spin resonance is greater than $1/T_{ds}$; this point was discussed above.

In concluding, we emphasize that the two general types of relaxation methods for the d spins which we have considered in this section and the last give quite different predictions. For example the mechanism of Sec. III gives a logarithmic g shift, while the present one does not. On the other hand Eq. (4.20) predicts a $1/T_{dl}$ that decreases as the temperature is raised. Facts such as these should serve as a warning not to take too seriously numerical fits of the s-d exchange model to experimental data, at least until the actual T_{dl} mechanism is better understood.

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APPENDIX

Our first task is to prove Eq. (2.3). This amounts to proving that

$$\Gamma_{H_s-H_d}(p_0)=\frac{\partial\Gamma(p_0)}{\partial p_0}\,\gamma\vec{S}\cdot\frac{\vec{H}_s-\vec{H}_d}{S(S+1)} \tag{A1}$$

because the other pieces of (2.3) are unchanged from those given in I, although the particular form (which is true to order $J^2\ln T$)

$$\Gamma_{M_s}=\frac{-\partial\Gamma(p_0)}{\partial p_0}\,\gamma\vec{S}\cdot\frac{\vec{M}_s}{S(S+1)\chi_{s0}^0} \tag{A2}$$

was not written down there. Equation (A2) follows immediately upon making the variable change $q_0 \to -q_0$, $\epsilon_k \to -\epsilon_k$ in the first term in the curly brackets of Eq. (C11) in I and noting that $\vec{m}(q,q_0)\propto \partial f(q_0)/\partial q_0$ (an even function of q_0); then the two terms in the curly brackets are identical, and the proof of (A2) is completed by differentiating (C4) of I with respect to p_0 and then integrating by parts and comparing.

Equation (A1) is most easily proved by inspection of Eq. (C7) of I for the field-dependent part of Γ, which we rewrite as [using (C4) of I]

$$-[(2s+1)\tfrac{1}{3}s(s+1)S(S+1)]^{-1}\frac{\partial\Gamma_0(p_0)}{\partial p_0}$$

$$\times \mathrm{Tr}_s(\vec{s}\cdot\vec{S}\gamma\vec{S}\circ\vec{H}\vec{s}\circ\vec{S}+\vec{s}\cdot\vec{S}[\gamma\vec{s}\circ\vec{H},\,\vec{s}\circ\vec{S}])\,. \tag{A3}$$

What one must note is that the $\vec{H}$ in the first term of the spin trace arose from the expansion of a localized spin Green's function and hence becomes $\vec{H}_d$, while the $\vec{H}$ in the last commutator in the spin trace arose from an expansion of conduction-electron Green's functions and hence becomes $\vec{H}_s$. Therefore, upon performing the spin traces, the analog of (A3) for $\vec{H}_s\neq\vec{H}_d$ is equal to

$$\frac{\partial\Gamma(p_0)}{\partial p_0}\,\gamma\vec{S}\cdot\left(\vec{H}_d+\frac{(\vec{H}_s-\vec{H}_d)}{S(S+1)}\right)\,, \tag{A4}$$

which when combined with (A2) constitutes a proof of Eq. (2.3). The proof of Eq. (2.5) proceeds in exactly the same way.

For the conduction electrons, there is no analog of (A2), but we still must prove (2.7) and (2.9). This is done in a manner identical to that used above for the local spins, so we omit the details and simply refer the reader to Eq. (B9) of I and those equations immediately preceding it.

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