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Theory of Spin Resonance of Localized Moments in Normal Metals*

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The rf susceptibility of a normal metal containing one or a small concentration of magnetic impurities is calculated. We use Anderson's model, the Hartree-Fock approximation for the ground state, and the random-phase approximation for the susceptibility. The resulting g shift agrees with other semiphenomenological analyses.

I. INTRODUCTION

The g value measured in ESR in a dilute alloy containing paramagnetic impurities is derived from Anderson's model[1] of such impurities. Our result is independent of the parameters of the model when it is evaluated for the Cu-Mn system studied by Schultz *et al.*,[2] and agrees with the semiphenomenological analysis[2] based on the model of Hasegawa.[3]

The idea of our calculation is that the physical system, and Anderson's model, is invariant against rotations of the spin degrees of freedom in the absence of a magnetic field. (We neglect spin-orbit coupling, and therefore omit spin-lattice relaxation from the theory.) If the conduction-band g value g_s and the impurity state g value g_d were equal, the total magnetic moment would be proportional to the total spin, Larmor's theorem would hold exactly, and there would be no g shift. It is therefore incorrect to view the resonance as a transition between the quite broad virtual bound states associated with up and down spin whose energy separation is largely correlation energy. The correct view is that the whole structure that is the localized moment, with its associated self-consistent fields, precesses. If $g_s \neq g_d$, there is some dephasing, leading to an extra line width $T_2'^{-1}$ which has nothing to do with relaxation, and associated with this there is a g shift. It is plausible, and true, that both $T_2'^{-1}$ and the g shift are smooth functions of g_s/g_d.

We start from the Hartree-Fock approximation for the ground state. Then the desired precession of the self-consistent field and all of the good consequences of that precession are automatically incorporated in the random-phase approximation (RPA). In Sec. II, we consider the problem of a single impurity, first for a nondegenerate virtual bound state, then for the realistic degenerate case. For the mathematically simpler nondegenerate case, we perform the calculation of the rf susceptibility both by the summation of perturbation-theory diagrams, to pave the way for the discussion of a finite concentration of impurities in Sec. III, and by an elementary self-consistent-field method. When we discuss the degenerate case, we modify the Hamiltonian of Anderson[1] in a natural way to make it rotationally invariant; we show simply that the original version[1] is not rotationally invariant, and, as we have said, rotational invariance is crucial for a correct theory of spin resonance. In Sec. III, we discuss the problem of a finite concentration of impurities, and in Sec. IV we solve Eq. (3.24), which is the main result of Sec. III, for the g shift.

II. ONE-IMPURITY PROBLEM

A. Nondegenerate Case

As we have already mentioned, we choose to represent the transition impurity imbedded in the noble-metal matrix by means of the Anderson Hamiltonian,[1] which is written, in the case of a nondegenerate d level, and with the notation of Ref. 1:

$$\mathcal{H}=\sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}{}^{\dagger}c_{\mathbf{k}\sigma}+\sum_{\alpha} \epsilon_{d\sigma}c_{d\sigma}{}^{\dagger}c_{d\sigma}+\sum_{k\sigma}(V_{\mathbf{k}d}c_{\mathbf{k}\sigma}{}^{\dagger}c_{d\sigma}+\text{H.c.}) +Uc_{d\uparrow}{}^{\dagger}c_{d\uparrow}c_{d\downarrow}{}^{\dagger}c_{d\downarrow}. \tag{2.1}$$

Since a magnetic field $\mathbf{H}$ ($\|Oz$) is applied to the system,

$$\epsilon_{k\uparrow}=\epsilon_k-\mu_sH\,,\quad \epsilon_{d\uparrow}=\epsilon_d-\mu_dH\,,$$
$$\epsilon_{k\downarrow}=\epsilon_k+\mu_sH\,,\quad \epsilon_{d\downarrow}=\epsilon_d+\mu_dH\,, \tag{2.2}$$

where $\epsilon_k=k^2/2m-E_F$, ϵ_d is the energy of the free-atom d level measured with respect to the Fermi energy E_F, $\mu_s=\frac{1}{2}g_s\mu_B$ and $\mu_d=\frac{1}{2}g_d\mu_B$, g_s and g_d being the gyromagnetic ratios for an electron in the conduction band and the d level, respectively.

It is worth noticing that in actual situations, due, for instance, to the spin-orbit effects, μ_s and μ_d are not equal.

On the other hand, the transverse dynamic susceptibility is obtained by the usual linear-response argument.

The Hamiltonian which couples the system to the transverse field $h_+(\mathbf{r},t)$ is

$$\mathcal{H}_{\text{int}}=-\frac{1}{2}\int d^3r\,m_-(\mathbf{r},t)h_+(\mathbf{r},t)\,, \tag{2.3}$$

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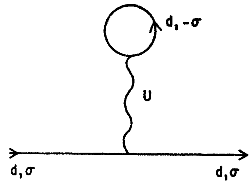

FIG. 1. Hartree-Fock self-energy for one nondegenerate impurity.

where m_- is the $(-)$ component of the magnetic-moment density operator.

This excitation induces a magnetic-moment density

$$\langle m_+(\mathbf{r},t)\rangle=\int d^3r'\int_{-\infty}^{\infty} dt'\chi(\mathbf{r},\mathbf{r}',t-t')h_+(\mathbf{r}',t'),\quad(2.4)$$

where

$$\chi(\mathbf{r},\mathbf{r}',t-t')=\tfrac{1}{2}i\theta(t-t')\langle[m_+(\mathbf{r},t),m_-(\mathbf{r}',t')]\rangle.\quad(2.5)$$

Here $\langle\;\rangle$ means the ensemble average on the equilibrium state of the system;

$$\begin{aligned}\theta(t-t')&=1\quad\text{for}\quad t>t'\\&=0\quad\text{for}\quad t<t'.\end{aligned}$$

The susceptibility (2.5) can then be calculated by means of the standard thermal-product technique.

Let us define the Fourier transform

$$\chi(\mathbf{q},\mathbf{q}',\omega)=\int d^3r\,d^3r'\int_{-\infty}^{\infty}dt\,\chi(\mathbf{r},\mathbf{r}',t)\times e^{-i(\mathbf{q}\cdot\mathbf{r}-\mathbf{q}'\cdot\mathbf{r}')+i\omega t},\quad(2.6)$$

and the thermal product

$$P(\mathbf{r},\mathbf{r}',\tau-\tau')=\tfrac{1}{2}\langle T_\tau[m_+(\mathbf{r},\tau)m_-(\mathbf{r}'\tau')]\rangle,\quad(2.7)$$

where τ is the usual imaginary time[4] and T_τ is the time-ordering operator. One calculates the Fourier transform $P(\mathbf{q},\mathbf{q}',\Omega_\nu)$ with $\Omega_\nu=2\nu\pi T$, where T is the temperature; then one takes its analytic continuation $P(\mathbf{q},\mathbf{q}',\Omega)$. The real frequency susceptibility is[4]

$$\chi(\mathbf{q},\mathbf{q}',\omega)=P(\mathbf{q},\mathbf{q}',-i\omega+\delta).\quad(2.8)$$

It is easy to check that

$$\begin{aligned}P(\mathbf{q},\mathbf{q}',\Omega_\nu)=-2[\mu_s^2&\sum_{\mathbf{k}\mathbf{k}'}S(\mathbf{k}\,\mathbf{k}-\mathbf{q}\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}'|\Omega_\nu)\\&+\mu_s\mu_dF(\mathbf{q})\sum_{\mathbf{k}'}S(dd\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}'|\Omega_\nu)\\&+\mu_s\mu_dF^*(\mathbf{q}')\sum_{k}S(\mathbf{k}\,\mathbf{k}-\mathbf{q}\,dd|\Omega_\nu)\\&+\mu_d^2F(\mathbf{q})F^*(\mathbf{q}')S(dd\,dd|\Omega_\nu)],\quad(2.9)\end{aligned}$$

$$F(\mathbf{q})=\int d^3r\,e^{-i\mathbf{q}\cdot\mathbf{r}}|\phi_d(\mathbf{r})|^2,$$

where $\phi_d(\mathbf{r})$ is the d-level wave function. $F(\mathbf{q})$ is the form factor of the d level.

$$S(1234|\Omega_\nu)=-\int_0^{1/T}d\tau\,e^{i\Omega_\nu\tau}\times\langle T_\tau[c_{1\uparrow}{}^\dagger(\tau)c_{2\downarrow}(\tau)c_{3\downarrow}{}^\dagger(0)c_{4\uparrow}(0)]\rangle.\quad(2.10)$$

We thus have to calculate four types of two-particle Green's functions which describe the propagation of electron-hole pairs with reversed spins; the electron and the hole can belong either to the conduction band or to the d level.

It is well known that, because of the presence of the Coulomb term on the impurity site, the one-particle Green's functions corresponding to the Hamiltonian (2.1) cannot be calculated exactly. Various approximate solutions[5] have been worked out; the simplest one consists in treating the Coulomb term in the Hartree-Fock approximation.

This corresponds to keeping in the self-energy only the term corresponding to the diagram of Fig. 1. We will confine ourselves here to this lowest-order approximation.

The one-particle thermal Green's function for the d electron is then given, from Anderson's original calculation, by

$$G_{d\sigma}(\omega_n)=1/(i\omega_n-E_\sigma+i\Gamma\,\mathrm{sgn}\,\omega_n).\quad(2.11)$$

Here $\omega_n=(2n+1)\pi T$, where n is any integer; $\Gamma=\pi N(0)\times|V|^2$, where $N(0)$ is the density of states in the conduction band at the Fermi energy for one spin direction; and following Ref. 1 we assume that $|V_{kd}|^2=|V|^2$ is a constant, for energies in the vicinity of E_F;

$$E_\sigma=\epsilon_{d\sigma}+U\langle n_{-\sigma}\rangle,\quad(2.12)$$

where $\langle n_{-\sigma}\rangle$, the number of electrons with spin $-\sigma$, is determined from the consistency equation (where we assume T to be very small compared to all the other energies in the problem, which are at least of the order of 1 eV)

$$n_{-\sigma}=\pi^{-1}\,\mathrm{arc\,cot}(E_\sigma/\Gamma).\quad(2.13)$$

Once an approximation has been chosen for calculating the one-particle Green's functions—or the self-energies—the way of evaluating the two-particle functions appearing in the Eq. (2.9) of the susceptibility is unambiguously defined. This follows from the fact that the conservation laws (i.e., the Ward identities) must be satisfied. In the present case the conserved quantity of interest is the total spin of the system. This in particular implies the well-known fact that, if

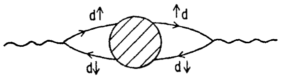

FIG. 2. Diagrammatic representation of $S(dddd|\Omega_\nu)$.

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[4] A. A. Abrikosov, L. P. Gorkov, and J. E. Dzailoshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, N. J., 1963).

[5] J. R. Schrieffer and D. C. Mattis, Phys. Rev. **140**, A1412 (1965); L. Dworin, Phys. Rev. Letters **16**, 1042 (1966).

$\mu_s = \mu_d = \mu$ and $\mathbf{q} = \mathbf{q}' = 0$ (uniform susceptibility), then the *s-d* interaction cannot induce any *g* shift (the electronic resonance must take place at $\omega = 2\mu H$). This condition can be used on the final expression of the susceptibility as a check of the consistency of the approximations.

In this section we will concentrate only on the calculation of the *d-d* two-particle function $S(dddd|\Omega_\nu)$. This is sufficient to formally define the approximations; furthermore, we are only interested in the finite concentration problem for comparison with the electronic resonance experiments.

It can be checked easily from the diagrammatic expansion—and it is well known—that if G has been calculated in the Hartree-Fock approximation, in order to satisfy the conservation laws one must calculate the two-particle Green's functions in the RPA scheme. In the present case, representing $S(dddd|\Omega_\nu)$ by the symbol of Fig. 2, this means that this quantity is the sum of the diagrams of Fig. 3.

In Fig. 3, each full line is the Hartree-Fock Green's function (2.11), ω_n, ω_n', $\cdots$ are internal frequencies on which sums have to be taken, so that the equation of Fig. 3 can be rewritten as

$$S(dddd|\Omega_\nu) = T \sum_{n=-\infty}^{\infty} G_{d\uparrow}(\omega_n) G_{d\downarrow}(\omega_n + \Omega_\nu) \times [1 - U\, S(dddd|\Omega_\nu)] \quad (2.14)$$

or

$$S(dddd|\Omega_\nu) = T \sum_n G_{d\uparrow}(\omega_n) G_{d\downarrow}(\omega_n + \Omega_\nu) / \times [1 + UT \sum_n G_{d\uparrow}(\omega_n) G_{d\downarrow}(\omega_n + \Omega_\nu)]. \quad (2.15)$$

Equation (2.14) can also be checked by making the RPA approximation directly on the equations of motion of the response functions. However, in order to make the physical content of our approximations clearer, it is of interest to rederive (2.15) in a slightly different way, which makes the treatment of the Hartree-Fock field appear more direct.

Making the Hartree-Fock approximation on the Hamiltonian (2.1) amounts to replacing the exact term $U n_{d\uparrow} n_{d\downarrow}$ by an effective one-particle Hamiltonian

$$\mathcal{H}_{\mathrm{HF}} = U(n_{d\uparrow}\langle n_{d\downarrow}\rangle + n_{d\downarrow}\langle n_{d\uparrow}\rangle - s_{d+}\langle s_{d-}\rangle - s_{d-}\langle s_{d+}\rangle), \quad (2.16)$$

with $s_{d+} = c_{d\uparrow}^\dagger c_{d\downarrow}$, $s_{d-} = c_{d\downarrow}^\dagger c_{d\uparrow}$.

This is the most general form of the effective one-particle term, which makes no assumption on the direction of the magnetic moment carried by the impurity. Let us now assume, as above, that at equilibrium this moment is along Oz; then $\langle s_{d+}\rangle = \langle s_{d-}\rangle = 0$, and the average fields $\langle n_{d\sigma}\rangle$ are defined by the self-consistency conditions (2.13).

Let us now apply a small transverse magnetic field $\delta h(r,t)$ to the system. If the Hartree-Fock Hamiltonian were the exact Hamiltonian, the induced magnetization would be given by the Kubo expression [Eqs. (2.4) and (2.5)], the correlation function $\langle[m_+, m_-]\rangle$ being calculated as an average on the Hartree-Fock states of the system.

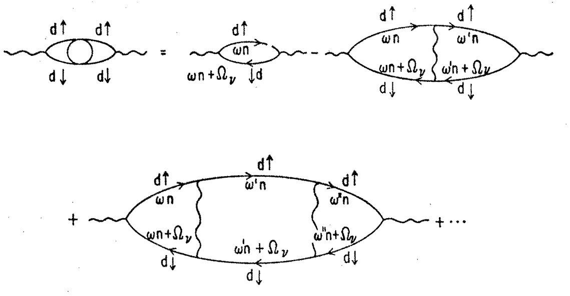

FIG. 3. Diagrammatic expansion of $S(dddd|\Omega_\nu)$ in RPA.

However, the presence of the excitation field δh_+ modifies the internal state of the system, including the values of the average fields $\langle n_{d\uparrow}\rangle \cdots \langle s_{d+}\rangle$; that is, δh_+ induces a motion of the impurity moment. The corresponding variation of $\mathcal{H}_{\mathrm{HF}}$ is

$$\delta\mathcal{H}_{\mathrm{HF}} = U(n_{d\uparrow}\delta\langle n_{d\downarrow}\rangle_t + n_{d\downarrow}\delta\langle n_{d\uparrow}\rangle_t - s_{d-}\delta\langle s_{d+}\rangle_t), \quad (2.17)$$

where $\delta\langle s_{d-}\rangle_t$ is obviously zero. Since we assume δh_+ to be very small compared to the static field H, it can tilt the impurity moment only by a small angle, so that we can assume $\delta\langle n_{d\uparrow}\rangle$ and $\delta\langle n_{d\downarrow}\rangle$ (i.e., $\delta\langle s_{dz}\rangle$) to be of order $(\delta h_+)^2$, while $\delta\langle s_{d+}\rangle$ is of order δh_+, and can be calculated as a linear response.[6]

Finally, the total coupling between δh_+ and the system is

$$\mathcal{H}_{\mathrm{int}} + \delta\mathcal{H}_{\mathrm{HF}} \cong \mathcal{H}_{\mathrm{int}} - U s_{d-}(t)\delta\langle s_{d+}\rangle_t. \quad (2.18)$$

$\delta\langle s_{d+}\rangle$ can itself be considered as the linear response of s_{d+} to the excitation (2.18), so that it is given by the Kubo formula

$$\delta\langle s_{d+}\rangle_\omega = \tfrac{1}{2}\langle[s_{d+}, m_-]\rangle_{\mathrm{HF},\omega}{}^R \delta h_{+\omega} + U\langle[s_{d+}, s_{d-}]\rangle_{\mathrm{HF},\omega}{}^R \delta\langle s_{d+}\rangle_\omega, \quad (2.19)$$

which plays the role of a self-consistency equation. Here $\langle[A,B]\rangle_{\mathrm{HF},\omega}{}^R$ is the Fourier transform of the retarded product $i\theta(t)\langle[A(\mathbf{r},t), B(\mathbf{r}',0)]\rangle_{\mathrm{HF}}$, where the ensemble average is taken on the Hartree-Fock equilibrium state of the system.

It is clear that for $\delta h_+ = 0$ the solution of Eq. (2.19) describes the precession of the impurity spin around the static field.

Finally, the total magnetization induced by the perturbation (2.18) is given, with the help of (2.19), by

$$\delta\langle m_+\rangle_\omega = \frac{1}{2}\left\{\langle[m_+, m_-]\rangle_{\mathrm{HF},\omega}{}^R + U\frac{\langle[m_+, s_{d-}]\rangle_{\mathrm{HF},\omega}{}^R \langle[s_{d+}, m_-]\rangle_{\mathrm{HF},\omega}{}^R}{1 - U\langle[s_{d+}, s_{d-}]\rangle_{\mathrm{HF},\omega}{}^R}\right\}\delta h_{+\omega}. \quad (2.20)$$

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[6] This would no longer be true in zero static field, where the moment would align itself along δh_+, and the present approximation would describe only the beginning of the motion.

The first term in (2.20) is the contribution to the susceptibility due to the noninteracting Hartree-Fock electron-hole excitations, while the second term is the contribution due to the possible flips of the impurity spin (in other words, to its precessional motion).

The retarded products $\langle[A,B]\rangle_{\mathrm{HF},\omega}{}^{R}$ can be calculated from the corresponding thermal products. Since the Hartree-Fock Hamiltonian is of a noninteracting-particles type, the two-particle Green's functions involved are simply products of one-particle Green's functions. It is then extremely easy to see that Eq. (2.20) is identical to the RPA approximation of (2.9).

For instance, we can extract from (2.20) the *d-d* contribution to the susceptibility, which is

$$P_{dd}(\mathbf{q},\mathbf{q}',\Omega_\nu) = -2\mu_d{}^2F(\mathbf{q})F^*(\mathbf{q}')\Big[T\sum_n G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu)\Big/ \times\Big[1+UT\sum_n G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu)\Big], \quad (2.21)$$

i.e., identical to the *d-d* susceptibility obtained from (2.15) by means of the diagram summation.

Let us finally explicitly calculate $\chi_{dd}(q,q',\omega)$, which value will be of use in the following. For the low temperatures of interest, we find

$$T\sum_n G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu) = \frac{\Delta n}{i\Omega_\nu+E_\uparrow-E_\downarrow} - \frac{N(0)|V|^2}{(i\Omega_\nu+E_\uparrow-E_\downarrow)(i\Omega_\nu+2i\Gamma+E_\uparrow-E_\downarrow)} \times\ln\left(\frac{(\Gamma-iE_\uparrow)(\Gamma+iE_\downarrow)}{(\Omega_\nu+\Gamma-iE_\uparrow)(\Omega_\nu+\Gamma+iE_\downarrow)}\right), \quad (2.22)$$

where

$$\Delta n=\langle n_\uparrow\rangle-\langle n_\downarrow\rangle,\quad E_\uparrow-E_\downarrow=-2\mu_dH-U\Delta n.$$

From (2.22), one immediately obtains $\chi_{dd}(\mathbf{q},\mathbf{q}',\omega)$. Since we will be dealing with frequencies $\omega\ll\Gamma$, we expand χ_{dd} to first order in ω/Γ. However, we do not expand expressions of the type $\omega+E_\uparrow-E_\downarrow$. (This is connected with the fact that the Hartree-Fock approximation does not give a reasonable value of the low-field static susceptibility, which will oblige us later to reinterpret $E_\uparrow-E_\downarrow$.) We then get

$$\chi_{dd}(\mathbf{q},\mathbf{q}',\omega) = -2\mu_d{}^2F(\mathbf{q})F^*(\mathbf{q}')\left(\Delta n-\frac{N(0)|V|^2\omega}{(E_\uparrow+i\Gamma)(E_\downarrow-i\Gamma)}\right)\Bigg/ \times\left(\omega-2\mu_dH-\frac{UN(0)|V|^2\omega}{(E_\uparrow+i\Gamma)(E_\downarrow-i\Gamma)}\right). \quad (2.23)$$

Let us note finally that, for $H=0$, $\chi_{dd}(\omega)$ goes to infinity when $\omega\to0$, as it should, since this simply means that in zero field the system is rotationally invariant.

B. Degenerate Case

The treatment of the single degenerate impurity case follows very simply from the one of the nondegenerate case. The only interesting question arises from the choice of the generalization of the Hamiltonian (2.1) suited to this situation.

The following Hamiltonian has been proposed by Anderson and used by several authors to describe the Coulomb and exchange terms on an impurity atom when one takes into account the orbital degeneracy of the *d*-wave functions[7]:

$$\mathcal{H}=\tfrac{1}{2}U\sum_{mm',\sigma} n_{m\sigma}n_{m',-\sigma}+\tfrac{1}{2}(U-J)\sum_{m\neq m',\sigma} n_{m\sigma}n_{m'\sigma}, \quad (2.24)$$

where *m* and *m'* are indices defining the *d* orbitals. $\mathcal{H}_A$ can be rewritten as

$$\mathcal{H}_A=\tfrac{1}{2}U\sum_{m\neq m'}(n_{m\uparrow}+n_{m\downarrow})(n_{m'\uparrow}+n_{m'\downarrow}) +\tfrac{1}{2}U\sum_m(n_{m\uparrow}+n_{m\downarrow})^2-(n_{m\uparrow}+n_{m\downarrow}) -\tfrac{1}{2}J\sum_{m\neq m'}\big[\tfrac{1}{2}(n_{m\uparrow}+n_{m\downarrow})(n_{m'\uparrow}+n_{m'\downarrow}) +2s_{mz}s_{m'z}\big]. \quad (2.25)$$

The Hamiltonian describing the impurity must obviously be rotationally invariant. $\mathcal{H}_A$ is invariant under a space rotation. However, it is clear from (2.25) that the term $-J\sum_{m\neq m'}s_{mz}s_{m'z}$ is not invariant under a rotation of the spin coordinates.[8]

$\mathcal{H}_A$ thus has to be replaced by an invariant Hamiltonian which we will take to be

$$\mathcal{H}'=\mathcal{H}_A-J\sum_{m\neq m'}(s_{mx}s_{m'x}+s_{my}s_{m'y}). \quad (2.26)$$

This keeps the meaning of *J* and *U* unchanged with respect to (2.24), and is simply the original Dirac formulation of Hund's rule. $\mathcal{H}'$ can be rewritten as

$$\mathcal{H}'=\tfrac{1}{2}U\sum_{mm'\sigma} n_{m\sigma}n_{m'-\sigma}+\tfrac{1}{2}(U-J)\sum_{m\neq m',\sigma} n_{m\sigma}n_{m'\sigma} -\tfrac{1}{2}J\sum_{m\neq m'} c_{m\sigma}{}^\dagger c_{m-\sigma}c_{m'-\sigma}{}^\dagger c_{m'\sigma}. \quad (2.27)$$

The one-particle Hartree-Fock Green's functions are still given by Anderson's expressions. For instance, when one neglects crystalline-field effects and orbital magnetism (which we will do in all that follows), the ones

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[7] Following Anderson, we have made the approximation that $U_{mm'}$ and $J_{mm'}$ are independent of *m* and *m'*. In fact the *U*'s and *J*'s are related to Slater's Integrals [see J. C. Slater, *Quantum Theory of Atomic Structure* (McGraw-Hill Book Co., New York, 1960).], and the above simplification violates the sum rules that $U_{mm'}$ and $J_{mm'}$ have to satisfy. In the present case where one is only interested in spin magnetism this approximation is not crucial. It has very important consequences in the case of orbital magnetism (see Ref. 11).

[8] This lack of invariance also appears in the fact that, if one uses $\mathcal{H}_A$ to perform an RPA calculation of the transverse susceptibility, one finds the unphysical result $\chi_{dd}{}^{-1}(H=\omega=0)\neq0$.

describing the d electrons are[9]

$$G_{mm'\sigma}(\omega_n)=\delta_{mm'}/(i\omega_n-E_\sigma+i\Gamma\,\mathrm{sgn}\omega_n)$$
$$\equiv\delta_{mm'}G_{d\sigma}'(\omega_n)\,,\qquad(2.28)$$

with

$$E_\sigma=\epsilon_{d\sigma}+5U\langle n_m\rangle-(U+4J)\langle n_{m\sigma}\rangle\,,\qquad(2.29)$$

and $\Gamma=\pi N(0)|V|^2$. [Precisely speaking, this choice of Γ corresponds to the expression for $V_{\mathbf{k}m}$

$$V_{\mathbf{k}m}=VY_{2m}(\hat{\Omega}_{\mathbf{k}})\,,$$

where the spherical harmonics $Y_{lm}(\hat{\Omega})$ are normalized, so that $\int(d\hat{\Omega}/4\pi)|Y_{lm}(\hat{\Omega})|^2=1.$] $\langle n_{m\sigma}\rangle$ is the Hartree-Fock occupation number of *one single orbital* with spin σ, and $\langle n_m\rangle=\langle n_{m\uparrow}\rangle+\langle n_{m\downarrow}\rangle$.

The transverse susceptibility is now [with the same notation as in (2.9)] the continuation of

$$\begin{aligned}P(\mathbf{q},\mathbf{q}',\Omega_\nu)=-2\Big(\mu_s^2\sum_{\mathbf{k}\mathbf{k}'}&S(\mathbf{k}\,\mathbf{k}-\mathbf{q}\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}'|\Omega_\nu)\\&+\mu_s\mu_d\sum_{m\mathbf{k}'}F_m(\mathbf{q})S(mm\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}'|\Omega_\nu)\\&+\mu_s\mu_d\sum_{\mathbf{k}m}F_m^*(\mathbf{q}')S(\mathbf{k}\,\mathbf{k}-\mathbf{q}\,mm|\Omega_\nu)\\&+\mu_d^2\sum_{mm'}F_m(\mathbf{q})F_{m'}^*(\mathbf{q}')S(mmm'm'|\Omega_\nu)\Big)\,.\end{aligned}\qquad(2.30)$$

The evaluation of the various S functions goes exactly along the same lines as in the nondegenerate case. One simply has to include the RPA diagrams corresponding to the J terms in $\mathcal{H}'$, so that $S(mmm'm'|\Omega_\nu)$ is now given by

$$\begin{aligned}S(mmm'm'|\Omega_\nu)=T\sum_n G_{d\uparrow}'(\omega_n)G_{d\downarrow}'(\omega_n+\Omega_\nu)&\\\times\Big(\delta_{mm'}-US(mmm'm'|\Omega_\nu)&\\-J\sum_{m''\neq m}S(m''m''m'm'|\Omega_\nu)\Big)\,,&\end{aligned}\qquad(2.31)$$

which gives

$$S(mmm'm'|\Omega_\nu)=\left(\delta_{mm'}-\frac{J\Lambda(\Omega_\nu)}{1+(U+4J)\Lambda(\Omega_\nu)}\right)\times\frac{\Lambda(\Omega_\nu)}{1+(U-J)\Lambda(\Omega_\nu)}\,,\qquad(2.32)$$

where

$$\Lambda(\Omega_\nu)=T\sum_n G_{d\uparrow}'(\omega_n)G_{d\downarrow}'(\omega_n+\Omega_\nu)\,.$$

We can then obtain, for instance, $\chi_{dd}(0,0,\omega)$ within the same approximation as in Sec. II A (low temperature, expansion in ω/Γ):

$$\chi_{dd}(0,0,\omega)=-10\mu_d^2\left(\Delta n_m-\frac{N(0)|V|^2\omega}{(E_\uparrow-i\Gamma)(E_\downarrow-i\Gamma)}\right)\Big/\left(\omega-2\mu_dH-\frac{(U+4J)|V|^2\omega}{(E_\uparrow+i\Gamma)(E_\downarrow-i\Gamma)}\right).\qquad(2.33)$$

Thus χ_{dd} has exactly the same structure as in the nondegenerate case; i.e., it is obtained from (2.23) in the case $\mathbf{q}=\mathbf{q}'=0$ by making the formal replacement $U\rightarrow U+4J$, and multiplying by an extra factor of $(2l+1)=5$, which comes from the orbital fivefold degeneracy.

We also note that $\chi_{dd}^{-1}(\mathbf{q},\mathbf{q}',H=\omega=0)-0$, which confirms that $\mathcal{H}'$ is invariant under spin rotations.

III. FINITE CONCENTRATION PROBLEM

We consider here the more realistic problem of a dilute alloy with a random distribution of impurities. As in Sec. II, and for reasons of simplicity, we will first study in detail the case of orbitally nondegenerate impurities, of which the degenerate case will be a very simple generalization.

A. Nondegenerate Case

The Hamiltonian (2.1) is now rewritten

$$\mathcal{H}=\sum_{k\sigma}\epsilon_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma}+\sum_{i\sigma}\epsilon_{d\sigma}c_{i\sigma}^\dagger c_{i\sigma}+\sum_{ki\sigma}(V_{\mathbf{k}i}c_{\mathbf{k}\sigma}^\dagger c_{i\sigma}+\text{H.c.})+U\sum_i c_{i\uparrow}^\dagger c_{i\uparrow}c_{i\downarrow}^\dagger c_{i\downarrow}\,.\qquad(3.1)$$

The index $i=1,\cdots,n$ labels the impurities, $V_{ki}=V_{\kappa d}\,e^{i\mathbf{k}\cdot\mathbf{R}_i}$, where $\mathbf{R}_i$ is the position of the ith impurity atom and V_{kd} is the same as in Sec. II. The interatomic interaction terms between different impurity atoms are neglected.

The multiple scattering problem corresponding to the Hamiltonian (3.1) has already been studied by Zuckermann[10] in the dilute limit. In order to clarify the calculation of the susceptibility, let us first recall very briefly how one obtains the average one-particle Green's functions

$$\bar{G}_{\mathbf{k}\sigma}=\langle\langle T_\tau[c_{\mathbf{k}\sigma}(\tau)c_{\mathbf{k}\sigma}^\dagger(0)]\rangle\rangle_{\text{imp}}\,,$$
$$\bar{G}_{ij\sigma}=\langle\langle T_\tau[c_{i\sigma}(\tau)c_{j\sigma}^\dagger(0)]\rangle\rangle_{\text{imp}}\,,$$

where the bar on $\bar{G}_{\mathbf{k}}$ and the $\langle\ \rangle_{\text{imp}}$ on the ensemble averages mean averaging over the positions of all impurities. In G_{ij} it means that the average is taken on the positions of all the impurities other than i and j. The calculation of the G's follows the same lines as in the well-known case of the one-band scattering.[4] The only difference is that, since V_{ki} cannot be treated here in the Born approximation, one has to retain diagrams

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[9] See, e.g., B. Caroli, thesis, University of Paris, 1966 (unpublished); and J. Phys. Chem. Solids 28, 1427 (1967).

[10] M. J. Zuckermann, Phys. Rev. 140, A889 (1965).

like the one of Fig. 4(a), including any number of scattering processes on the same impurity atom.

We also neglect diagrams of the type of Fig. 4(b) in which dashed lines corresponding to scattering on different impurities intersect; their contribution would as usual be of order $1/p_F l$ compared to diagrams without intesecting lines (p_F being the Fermi momentum and l the conduction-electron mean free path). Here, because of the fact that we are dealing with resonant scattering, l can be rather strongly energy-dependent, and $l(E_F)$ could be very small in the case of a very narrow d level centered very close to the Fermi energy. Thus one would have to be careful about the above approximation (especially when dealing with f instead of d impurity levels and not very dilute alloys), but this sort of problem does not arise for the very dilute d type of impurities that we consider here.

Finally, we will take

$$\bar{G}_{ij}=\bar{G}_d\delta_{ij}; \tag{3.2}$$

that is, we neglect the interactions between impurities (we assume T to be well above the ordering temperature of the impurity spins). With these approximations we find that $\bar{G}_{\mathbf{k}}$ and $\bar{G}_d$ obey the coupled system of equations

$$\begin{aligned}\bar{G}_{\mathbf{k}\sigma}(\omega_n)&=G_{\mathbf{k}\sigma}{}^0(\omega_n)+c|V|^2G_{\mathbf{k}\sigma}{}^0(\omega_n)\bar{G}_{d\sigma}(\omega_n)\bar{G}_{\mathbf{k}\sigma}(\omega_n)\,,\\ \bar{G}_{d\sigma}(\omega_n)&=G_{d\sigma}{}^0(\omega_n)+|V|^2G_{d\sigma}{}^0(\omega_n)\\ &\qquad\sum_k\bar{G}_{\mathbf{k}\sigma}(\omega_n)\bar{G}_{d\sigma}(\omega_n)\,,\end{aligned} \tag{3.3}$$

where c is the impurity concentration,

$$G_{k\sigma}{}^0(\omega_n)=(i\omega_n-\epsilon_{k\sigma})^{-1},$$

and

$$G_{d\sigma}{}^0=(i\omega_n-\epsilon_{d\sigma}-U\langle n_{-\sigma}\rangle)^{-1}.$$

As in Sec. II, the Coulomb term is treated in the Hartree-Fock approximation.

From (3.3), we obtain

$$\bar{G}_{d\sigma}=((G_{d\sigma}{}^0)^{-1}-|V|^2\sum_{\mathbf{k}}\bar{G}_{\mathbf{k}\sigma})^{-1}. \tag{3.4}$$

In Anderson's approximation (i.e., neglecting the energy shift due to the scattering), $\sum_{\mathbf{k}}\bar{G}_{k\sigma}=-i\pi N(0)\times\operatorname{sgn}\omega_n$, where $N(0)$ is the density of states in the conduction band of the alloy at the Fermi level. If we neglect the change in this density of states due to alloying, we get

$$\bar{G}_{d\sigma}(\omega_n)\equiv G_{d\sigma}(\omega_n)=(i\omega_n-E_\sigma+i\Gamma\operatorname{sgn}\omega_n)^{-1}, \tag{3.5}$$

(this is equivalent to neglecting the change in Γ, which is anyway a rather poorly known quantity[11]),

$$\bar{G}_{\mathbf{k}\sigma}=(i\omega_n-\epsilon_{\mathbf{k}\sigma}-c|V|^2G_{d\sigma})^{-1}. \tag{3.6}$$

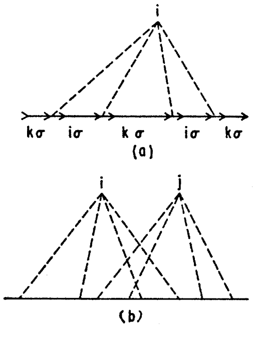

FIG. 4. Typical diagrams for the conduction-electron Green's function. The dashed line corresponds to a V_{ki} (or V_{ik}) scattering.

Let us now proceed to calculate the dynamic susceptibility. Because of the translational invariance of a disordered alloy, it is obviously of the form

$$\chi(\mathbf{q},\mathbf{q}',\omega)\equiv\chi(\mathbf{q},\omega)\delta_{\mathbf{qq}'}\,, \tag{3.7}$$

where $\chi(\mathbf{q},\omega)$ is the continuation of

$$\begin{aligned}P(\mathbf{q},\Omega_\nu)=-2[\mu_s{}^2&\sum_{\mathbf{kk}'}\bar{S}(\mathbf{kk}-\mathbf{qk}'\mathbf{k}'+\mathbf{q}|\Omega_\nu)\\ &+\mu_s\mu_dF(\mathbf{q})\sum_{i\mathbf{k}'}\langle S(ii\mathbf{k}'\mathbf{k}'+\mathbf{q}|\Omega_\nu)e^{-i\mathbf{q}\cdot\mathbf{R}_i}\rangle_{\text{imp}}\\ &+\mu_s\mu_dF^*(\mathbf{q})\sum_{\mathbf{k}i}\langle S(\mathbf{kk}-\mathbf{q}ii|\Omega_\nu)e^{-i\mathbf{q}\cdot\mathbf{R}_i}\rangle_{\text{imp}}\\ &+\mu_d{}^2F(\mathbf{q})|^2\sum_{ij}\langle S(iijj|\Omega_\nu)e^{i\mathbf{q}\cdot(\mathbf{R}_j-\mathbf{R}_i)}\rangle\rangle_{\text{imp}}].\end{aligned} \tag{3.8}$$

The S's are defined as in (2.9) and the bar and $\langle\ \rangle_{\text{imp}}$ again mean averaging over the positions of the impurities. As usual, even in the absence of interactions, the average of the two-particle propagators is not equal to the product of the averages of the corresponding one-particle propagators.

Finally, we will calculate only the uniform susceptibility $\chi(0,\omega)$ which is sufficient to interpret measurements of T_2 and of the g shift. (The generalization to finite $\mathbf{q}$ would be algebraically heavy but straightforward.) Let us define

$$\begin{aligned}\Gamma_{\mathbf{k}}&=\mu_s\sum_{\mathbf{k}'}\bar{S}(\mathbf{kkk}'\mathbf{k}'|\Omega_\nu)+\Omega_d\sum_i\bar{S}(\mathbf{kk}ii|\Omega_\nu)\,,\\ \Gamma_d&=\mu_s\sum_{\mathbf{k}'}\bar{S}(ii\mathbf{k}'\mathbf{k}'|\Omega_\nu)+\mu_d\sum_j\bar{S}(iijj|\Omega_\nu)\,.\end{aligned} \tag{3.9}$$

Then, keeping in the calculation of the Γ's only dia-

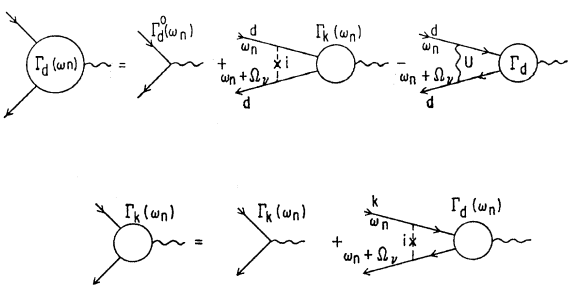

FIG. 5. Integral equations for the vertex functions. Solid lines represent renormalized average Green's functions.

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[11] See, e.g., B. Coqblin, thesis, University of Paris, 1967 (unpublished); and B. Coqblin and A. Blandin, Advan. Phys. 17, 281 (1968).

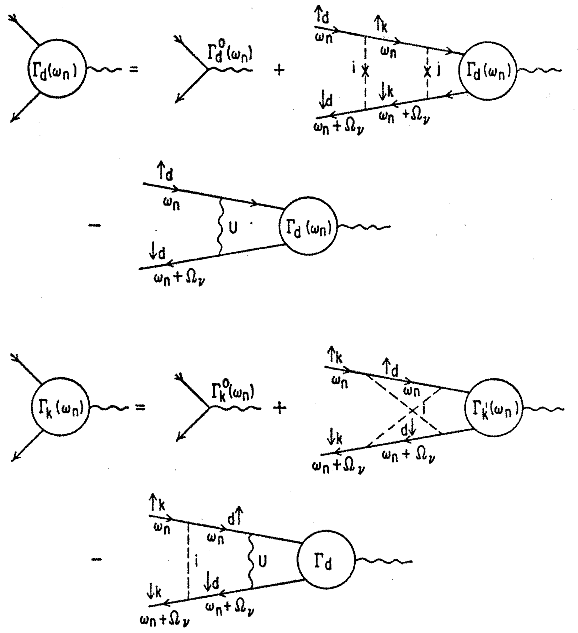

FIG. 6. Rearrangement of the equations for the vertex functions.

grams consistent with the approximations used in the evaluation of the G's, in particular treating again U in the RPA, we find that the Γ's satisfy the coupled system of equations of Fig. 5. Here $\Gamma_d{}^0(\omega_n)$ and $\Gamma_{\mathbf{k}}{}^0(\omega_n)$ are defined so that

$$\Gamma_d{}^0(\omega_n)=\mu_d G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu)+\mu_s|V|^2G_{d\uparrow}(\omega_n)$$
$$\times G_{d\downarrow}(\omega_n+\Omega_\nu)\sum_{\mathbf{k}}\bar{G}_{\mathbf{k}\uparrow}(\omega_n)\bar{G}_{\mathbf{k}\downarrow}(\omega_n+\Omega_\nu), \quad (3.10)$$

$$\Gamma_{\mathbf{k}}{}^0(\omega_n)=G_{\mathbf{k}\uparrow}(\omega_n)G_{\mathbf{k}\downarrow}(\omega_n+\Omega_\nu)[\mu_s+c\mu_d|V|^2$$
$$\times G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu)], \quad (3.11)$$

and $\Gamma_{\mathbf{k}}=T\sum_n\Gamma_{\mathbf{k}}(\omega_n)$, $\Gamma_d=T\sum_n\Gamma_d(\omega_n)$. The equations

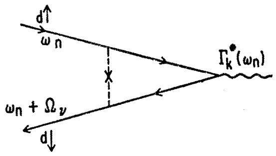

FIG. 7. In passing from Fig. 5 to Fig. 6, the double counting of this diagram must be avoided.

of Fig. 5 can be rewritten as shown in Fig. 6. Note that there is only one driving term in each equation [i.e., a diagram like the one of Fig. 7 must not appear in $\Gamma_d(\omega_n)$ because it is already included in $\Gamma_d{}^0(\omega_n)$].

We also neglect in $\Gamma_k(\omega_n)$ diagrams where dashed and U lines (referring to the same impurity) intersect. This approximation is good because the modification of the self-consistent field of the impurity (which is localized mainly in its atomic cell) due to the multiple scattering is negligible.

Using the notation

$$S_d(\omega_n)=G_{d\uparrow}(\omega_n)G_{d\downarrow}(\omega_n+\Omega_\nu),$$
$$S_{\mathbf{k}}(\omega_n)=\bar{G}_{\mathbf{k}\uparrow}(\omega_n)\bar{G}_{\mathbf{k}\downarrow}(\omega_n+\Omega_\nu),$$
$$\beta(\omega_n)=c|V|^4S_d(\omega_n)\sum_{\mathbf{k}}S_{\mathbf{k}}(\omega_n), \quad (3.12)$$
$$A(\omega_n)=\sum_{\mathbf{k}}S_{\mathbf{k}}(\omega_n)/[1-\beta(\omega_n)],$$

we get

$$\Gamma_d(\omega_n)=\Gamma_d{}^0(\omega_n)+c|V|^4S_d(\omega_n)$$
$$\times\Gamma_d(\omega_n)\sum_{\mathbf{k}}S_{\mathbf{k}}(\omega_n)-U\Gamma_dS_d(\omega_n),$$
$$\Gamma_{\mathbf{k}}(\omega_n)=\Gamma_{\mathbf{k}}{}^0(\omega_n)+c|V|^4S_{\mathbf{k}}(\omega_n)S_d(\omega_n)\sum_{\mathbf{k}'}\Gamma_{\mathbf{k}'}(\omega_n)$$
$$-c|V|^2US_{\mathbf{k}}(\omega_n)S_d(\omega_n)\Gamma_d. \quad (3.13)$$

After solving Eqs. (3.13), we obtain for the uniform susceptibility

$$P(0,\Omega_\nu)=-2(\mu_s\sum_{\mathbf{k}}\Gamma_{\mathbf{k}}+c\mu_d\Gamma_d),$$
$$=-2\mu_s{}^2\left\{T\sum_n A(\omega_n)-\left[c|V|^4\bigg/\left(1+UT\sum_n\frac{S_d(\omega_n)}{1-\beta(\omega_n)}\right)\right](T\sum_n A(\omega_n)S_d(\omega_n))^2\right\}$$
$$-2\mu_s\mu_d2c|V|^2T\sum_n A(\omega_n)S_d(\omega_n)\bigg/\left(1+UT\sum_n\frac{S_d(\omega_n)}{1-\beta(\omega_n)}\right)$$
$$-2c\mu_d{}^2T\sum_n\frac{S_d(\omega_n)}{1-\beta(\omega_n)}\bigg/\left(1+UT\sum_n\frac{S_d(\omega_n)}{1-\beta(\omega_n)}\right). \quad (3.14)$$

The explicit calculation of the various sums involved in Eq. (3.14) is completely straightforward in the low-temperature region [where $T\sum_n f(\omega_n)\to\int(d\omega/2\pi)\,f(\omega)$].[12]

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[12] As usual, one has to be careful in the calculation of $T\sum_nA(\omega_n)$ because of the slow convergence of the expression $T\sum_{n,k}G_{k\uparrow}(\omega_n)\times G_{k\downarrow}(\omega_n+\Omega_\nu)$. This is most easily dealt with by writing

$$T\sum_n A(\omega_n)=T\sum_n(A(\omega_n)-\sum_k G_{k\uparrow}{}^0(\omega_n)G_{k\downarrow}{}^0(\omega_n+\Omega_\nu))+T\sum_n G_{k\uparrow}{}^0(\omega_n)G_{k\downarrow}{}^0(\omega_n+\Omega_\nu).$$

The first term in the right-hand side is rapidly convergent; the exact calculation of the (slowly convergent) second term is trivial.

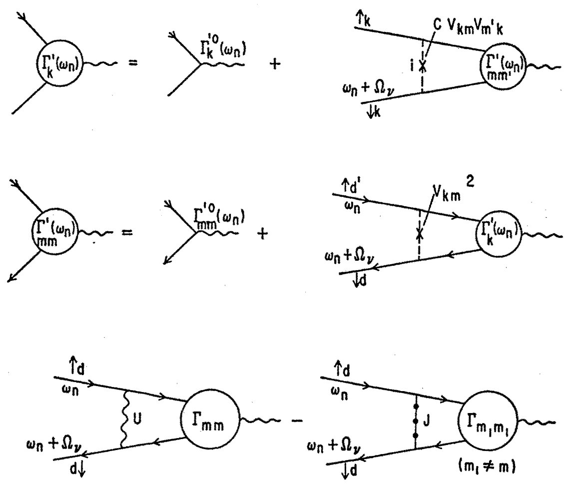

FIG. 8. Integral equations for the vertex functions in the degenerate case (see Fig. 5).

We finally obtain

$$\chi(0,\omega) =2\mu_s^2\left(\frac{N(0)\omega_s+c\Gamma I(\Delta E_d-\omega)}{\omega_s-\omega}+\frac{c\Gamma^2I^2U(\Delta E_d-\omega)}{\omega_d-\omega-U\Gamma I(\omega_s-\omega)}\right) -4c\mu_s\mu_d\frac{\Gamma I(\Delta E_d-\omega)}{\omega_d-\omega-U\Gamma I(\omega_s-\omega)} +2c\mu_d^2\frac{\Delta n+\Gamma I(\omega_s-\omega)}{\omega_d-\omega-U\Gamma I(\omega_s-\omega)},\quad(3.15)$$

where $\omega_{s,d}=2\mu_{s,d}H$, $\Delta E_d=E_\downarrow-E_\uparrow$,

$$I=\frac{1}{\pi(\omega_s-\omega)\sqrt{\mathfrak{D}}}\ln\left(\frac{E_\uparrow+E_\downarrow-\omega-\sqrt{\mathfrak{D}}}{E_\uparrow+E_\downarrow+\omega-\sqrt{\mathfrak{D}}}\times\frac{E_\uparrow+E_\downarrow+\omega+\sqrt{\mathfrak{D}}}{E_\uparrow+E_\downarrow-\omega+\sqrt{\mathfrak{D}}}\right),\quad(3.16)$$

$$\mathfrak{D}=(\Delta E_d-\omega-2i\Gamma)^2-\frac{4c|V|^2(\Delta E_d-\omega)}{\omega_s-\omega}.$$

Some checks can be made on Eq. (3.15):

(a) Let us assume first that $\mu_s=\mu_d=\mu$. We obtain after some manipulations

$$\chi(0,\omega)=2\mu^2\frac{2\mu HN(0)+c\Delta n}{2\mu H-\omega},$$

i.e., for $\mathbf{q}=0$ the electronic resonance shows no g shift and $T_2^{-1}=0$, which is in agreement with the condition of conservation of the total spin.

(b) $\chi^{-1}(\mathbf{q}=H=\omega=0)=0$, in agreement with the condition of rotational invariance.

(c) When $\mu_s\neq\mu_d$, one can check that $\omega=\omega_s$ is not a pole of the susceptibility. [Although it appears as a pole on expression (3.15), it is easy to show that it has a zero residue.]

Therefore, the resonance frequency is obtained from the condition

$$\mathrm{Re}[\omega_d-\omega-U\Gamma I(\omega_s-\omega)]=0.\quad(3.17)$$

B. Degenerate Case

It is now extremely easy to generalize the preceding results to the case of fully degenerate impurities without orbital magnetism. The exchange and Coulomb effects on each impurity site correspond to an interaction term $\mathcal{H}_i'$ [Eq. (2.27)]. Taking again $\bar{G}_{mm',ij}=\bar{G}_{mm'}\delta_{ij}$, and making the same assumptions as in Sec. III A, we find that $\bar{G}_{mm'}$ and $\bar{G}_k$ satisfy the system of equations

$$\bar{G}_{mm',\sigma}=G_{d\sigma}'^0\Big(\delta_{mm'}+\sum_{\mathbf{k}m_1}V_{m\mathbf{k}}\bar{G}_{\mathbf{k}\sigma}V_{\mathbf{k}m_1}\bar{G}_{m_1m'\sigma}\Big),$$

$$\bar{G}_{\mathbf{k}\sigma}=G_{\mathbf{k}\sigma}^0\Big(1+c\sum_{mm'}V_{\mathbf{k}m}\bar{G}_{mm'\sigma}V_{m'\mathbf{k}}\bar{G}_{\mathbf{k}\sigma}\Big),\quad(3.18)$$

with

$$(G_{d\sigma}'^0)^{-1}=i\omega_n-\epsilon_{d\sigma}-5U\langle n_m\rangle+(U+4J)\langle n_{m\sigma}\rangle=i\omega_n-E_\sigma.$$

Equations (3.18) are solved by

$$\bar{G}_{mm'\sigma}(\omega_n)=G_{d\sigma}'(\omega_n)\delta_{mm'} =\delta_{mm'}/(i\omega_n-E_\sigma+i\Gamma\,\mathrm{sgn}\omega_n),\quad(3.19)$$
$$\bar{G}_{\mathbf{k}\sigma}(\omega_n)=[i\omega_n-\epsilon_{k\sigma}-c(2l+1)|V|^2G_{d\sigma}'(\omega_n)]^{-1},$$

with $l=2$ (for a d level).

The uniform susceptibility is now obtained from

$$P(0,\Omega_\nu)=-2\mu_s\sum_{\mathbf{k}}\Gamma_{\mathbf{k}}'-2c\mu_d\sum_m\Gamma_{mm}',$$

where

$$\Gamma_{\mathbf{k}}'=\mu_s\sum_{\mathbf{k}'}\bar{S}(\mathbf{kkk'k'}|\Omega_\nu)+\mu_d\sum_{im}\bar{S}(\mathbf{kk}m_im_i|\Omega_\nu),\quad(3.20)$$

$$\Gamma_{mm}'=\mu_s\sum_{\mathbf{k}'}\bar{S}(m_im_i\mathbf{k'k'}|\Omega_\nu)+\mu_d\sum_{jm'}\bar{S}(m_im_im_j'm_j'|\Omega_\nu).$$

The index m_i refers to the mth orbital on the ith impurity atom. The equations satisfied by the Γ's in the RPA are represented graphically in Fig. 8.

From these equations we obtain (with notation analogous to that used in Sec. III A)

$$\begin{aligned}\Gamma_{mm}'(\omega_n)=\Gamma_{mm}'^0(\omega_n)+S_d'(\omega_n)\Big(c\sum_{\mathbf{k}m_1m_2}|V_{\mathbf{k}m}|^2S_{\mathbf{k}}'(\omega_n)\\ \times V_{\mathbf{k}m_1}V_{m_2\mathbf{k}}\Gamma'_{m_1m_2}(\omega_n)-J\sum_{m_1\neq m}\Gamma'_{m_1m_1}\\ -U\Gamma'_{mm}\Big),\\ \Gamma_{\mathbf{k}}'(\omega_n)=\Gamma_{\mathbf{k}}'^0(\omega_n)+cS_{\mathbf{k}}'(\omega_n)S_d'(\omega_n)\sum_{mm'}V_{\mathbf{k}m}V_{m'\mathbf{k}}\\ \times\Big(\sum_{\mathbf{k}'}V_{m\mathbf{k}'}V_{\mathbf{k}'m'}\Gamma_{\mathbf{k}'}'(\omega_n)\\ -U\Gamma_{mm'}'+J\delta_{mm'}\sum_{m_1\neq m}\Gamma_{m_1m_1}'\Big).\end{aligned}\tag{3.21}$$

Defining $\Gamma_d'(\omega_n)=\sum_m\Gamma_{mm}'(\omega_n)$, Eqs. (3.21) reduce to

$$\begin{aligned}\Gamma_d'(\omega_n)=\Gamma_d'^0(\omega_n)+S_d'(\omega_n)\Big(c(2l+1)|V|^4\\ \times\sum_{\mathbf{k}}S_{\mathbf{k}}'(\omega_n)\Gamma_d'(\omega_n)-(U+4J)\Gamma_d'\Big),\end{aligned}\tag{3.22}$$

$$\begin{aligned}\sum_{\mathbf{k}}\Gamma_{\mathbf{k}}'(\omega_n)=\sum_{\mathbf{k}}\Gamma_{\mathbf{k}}'^0(\omega_n)+cS_d'(\omega_n)\sum_{\mathbf{k}}S_{\mathbf{k}}'(\omega_n)\\ \times\Big((2l+1)|V|^4\sum_{\mathbf{k}'}\Gamma_{\mathbf{k}'}'(\omega_n)\\ -|V|^2(U+4J)\Gamma_d'\Big).\end{aligned}$$

The driving terms Γ'^0 are given by

$$\begin{aligned}\Gamma_d'^0(\omega_n)=(2l+1)\Big(\mu_dS_d'(\omega_n)\\ +\mu_s|V|^2S_d'(\omega_n)\sum_{\mathbf{k}}S_{\mathbf{k}}'(\omega_n)\Big),\end{aligned}\tag{3.23}$$

$$\sum_{\mathbf{k}}\Gamma_{\mathbf{k}}'^0(\omega_n)=[\mu_s+c\mu_d(2l+1)|V|^2S_d'(\omega_n)]\sum_{\mathbf{k}}S_{\mathbf{k}}'(\omega_n).$$

Finally, and after some very simple algebra, we find that $\chi(0,\omega)$ is given by an expression completely analogous to (3.15). That is, χ for the degenerate case is obtained from (3.15) by making everywhere the transformation

$$\begin{aligned}c\rightarrow c(2l+1),\\ U\rightarrow U+4J.\end{aligned}\tag{3.24}$$

Moreover, Δn is now to be understood as referring to *one single orbital*. It is then obvious that $\chi(0,\omega)$ has the same properties as in the nondegenerate case, namely, (a) For $\mu_s=\mu_d=\mu$, the uniform susceptibility has its only pole at $\omega=2\mu H$. (b) $\chi^{-1}(\mathbf{q}=\omega=H=0)=0$. (c) For $\mu_s\neq\mu_d$, $\omega=\omega_s$ is not a pole of χ and the resonance frequency is given by

$$\mathrm{Re}[\omega_d-\omega-(U+4J)\Gamma I'(\omega_s-\omega)]=0.\tag{3.25}$$

I' is obtained from (3.16) by the transformation (3.24).

IV. DISCUSSION

Equation (3.25) has many roots in general. We shall calculate the real, or almost real, root ω which we expect to be near ω_s and ω_d. The imaginary part of ω is $T_2'^{-1}$, the linewidth caused by the dephasing of the electron spin precession as it hops getween the conduction band, where it precesses with circular frequency ω_s, and the impurity levels, where it precesses with the slightly different circular frequency ω_d. This is expected to be a minute fraction of the total line width $T_2'^{-1}+T_1^{-1}$; note that spin-lattice relaxation T_1^{-1} has been omitted from our theory. Now the total line width is small: $\mathrm{Im}\omega/\mathrm{Re}\omega=\Delta\omega/\omega=\Delta H/H\approx0.01$. We are therefore justified in looking for a real root ω of (3.24).

The precise result for ω will depend only on rough orders of magnitude of the parameters of the model. We take $U+4J=4$ eV, $\Gamma=0.5$ eV, $E_\uparrow+E_\downarrow=-1.8$ eV, and all other parameters directly from experiment. We find that, for the desired root ω near ω_s and ω_d, the second term in $\mathfrak{D}$ is overwhelmingly larger than the first, and further that $(E_\uparrow+E_\downarrow\pm\omega)/\mathfrak{D}^{1/2}<1$ for temperatures under 100°K. Once these facts are accepted, we can forget the values of the parameters.

We have

$$\begin{aligned}I'=+\frac{4\omega}{\pi(\omega_s-\omega)\mathfrak{D}}\end{aligned}\tag{4.1}$$

$$=-\frac{\omega}{5c\pi|V|^2(\Delta E_d-\omega)}$$

$$=-\frac{N(0)\omega}{5c(U+4J)\Gamma\Delta n},$$

$$I'=-\frac{gg_d}{g_s^2}\frac{M_s}{(U+4J)\Gamma M_d},\tag{4.2}$$

$$I'\approx-\frac{M_s}{(U+4J)\Gamma M_d},\tag{4.3}$$

where M_s, M_d are the conduction-electron and impurity magnetizations per lattice site. We have put $gg_d/g_s^2\approx1$ in going from (4.2) to (4.3), which involves an error of order $|g_s-g_d|/g_s=1\%$ in I', and an error of order $|g_s-g_d|^2/g_s^2=10^{-4}$ in the g value. Finally,

$$\omega_d-\omega+(M_s/M_d)(\omega_s-\omega)=0$$

or

$$\omega=\frac{M_d\omega_d+M_s\omega_s}{M_d+M_s}.\tag{4.4}$$

This is exactly the result of the semiphenomenological theory of Schultz *et al.*,[2] based on the model of Hasegawa.[3]