sion that $|\Gamma|N(E_F) \simeq 1$ does not seem unreasonable. It follows from Knight-shift data for $CeAl_3$[4,5] that $\Gamma \simeq -3$ eV/at., whereas $N(E_F)$ can be close to 0.5 state/eV at., so that one finally arrives at the correct order of magnitude of the product $|\Gamma|N(E_F)$. Moreover, with the above values for Γ and $N(E_F)$ and using the conventional equation for spin-disorder resistivity, one arrives at a value of about 10 $\mu\Omega$cm for $\rho^{(1)}$ for temperatures relatively high compared to the total crystal field splitting. This is consistent with the above estimate of $\rho^{(1)}$ from experimental data. It follows that one "arbitrary unit," used in the theoretical first-order results (see Fig. 8), is equivalent to about 1 $\mu\Omega$cm. It also follows, then, that in the case of $Ce_{0.33}Th_{0.67}Al_3$ in the low-temperature region, the total variation of ρ in the experiment is at least one order of magnitude larger than expected from the first-order contribution in the situation (0, 2, 50) (compare the results of Figs. 3 and 8). In this case, too, the second-order contribution evidently remains dominant.

An alternative way to explain a decrease of ρ with increasing T seems possible, in principle, by means of Blatt's mechanism.[17] In that case, the $4f^1$ level has to be close to the Fermi surface, which is not unlikely. To obtain a maximum in the ρ-vs-T curve the Blatt mechanism has to be combined with effects of different origin (for instance, crystal field effects, if these could be treated simultaneously with the Blatt mechanism). It is seriously doubted, however, whether in such a model the occurrence of two maxima in the ρ-T curve could be explained, and whether the ρ data obtained for $CeAl_3$ upon increasing replacement of Ce by La, Y, or Th could consistently be accounted for.

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Accurate-Pairing Treatment of the Coulomb Interaction in the Anderson Model of a Localized Moment in a Metal*

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The treatment of the Anderson model of a localized moment in a metal has been extended beyond the effective-field approximation by a method recently developed for studying the Hubbard model. This leads to an accurate treatment of the Coulomb interaction associated with the localized center. The results are a simple generalization of those obtained by Anderson within the context of the effective-field approximation.

I. INTRODUCTION

In a previous paper, the author[1] studied the problem of an interacting system of localized moments in a dilute magnetic alloy. Each localized center containing a magnetic moment was represented by the model of Anderson.[2] The problem was treated by means of an equation-of-motion method, includ-

ing pairing.[3] In this approach, one pairs an electron in some single-particle orbital, spin up, with a hole in the same single-particle orbital, spin down. This amounts to leaving unspecified the *orientation* of the spin of the electron occupying the single-particle orbital, a procedure of great convenience in any magnetic problem.

An essential feature of the treatment of I was the use of a generalized effective field.[4] This led to an approximate treatment of the intracenter Coulomb interaction, associated with any center containing two opposite-spin electrons. In a recent paper devoted to a treatment of the Hubbard model,[5] the author[6] has developed a modification of the pairing equation-of-motion method which allows the Coulomb interaction to be treated accurately without making an effective-field approximation. In the present paper, we wish to apply this modified pairing method to the dilute magnetic alloy, thereby eliminating the major approximation both of I and of Anderson's original treatment.[2] In the interest of simplifying the matrix algebra, we restrict the present paper to a consideration of a *single* localized center in a metal.

In Sec. II we set up the modified form of the equations of motion, including pairing, to study the Anderson Hamiltonian. We obtain properties of the conduction-band quasiparticles in Sec. III and properties of the localized quasiparticles in Sec. IV. In particular, the energy spectrum and the magnetic moment associated with the localized center are simple functions of the two roots of a pair of coupled functional equations. Anderson's original treatment[2] can also be recast in terms of the roots of a pair of functional equations. The functional equations of the two theories are similar but not identical. The equations of both theories involve (in addition to temperature) the three parameters ϵ_0, U_0, and W; ϵ_0 being the bare one-electron energy of the center, U_0 being the Coulomb energy of the doubly occupied center, and W being the self-energy of the center due to coupling with the conduction band.

II. EQUATIONS OF MOTION

The Hamiltonian of our system is

$$H = H_0 + H_1 , \tag{2.1}$$

$$H_0 = \sum_{k,\sigma} \epsilon_k c_{k\sigma}^\dagger c_{k\sigma} + \epsilon_0 \sum_\sigma c_{i\sigma}^\dagger c_{i\sigma} + U_0 c_{i\uparrow}^\dagger c_{i\uparrow} c_{i\downarrow}^\dagger c_{i\downarrow} , \tag{2.2}$$

$$H_1 = \sum_{k,\sigma} (V_k c_{k\sigma}^\dagger c_{i\sigma} + V_{-k} c_{i\sigma}^\dagger c_{k\sigma}) . \tag{2.3}$$

The one-electron energies ϵ_k (for the conduction band) and ϵ_0 (for the localized center) are measured relative to the Fermi level. The total Hamiltonian represents a system of conduction-band electrons interacting with electrons in a localized center (the index i denoting the center). The positive Coulomb energy U_0 is associated with the center whenever it contains two opposite-spin localized electrons. (For simplicitly, we take the center to have a single localized s orbital.) We assume

$$V_k^* = V_{-k} , \tag{2.4}$$

so that H_1 is Hermitian.

The electron creation and destruction operators obey the usual anticommutation relations

$$\begin{aligned} &[c_{l\sigma}, c_{l'\sigma'}^\dagger]_+ = \delta_{ll'}\delta_{\sigma\sigma'} , \\ &[c_{l\sigma}, c_{l'\sigma'}]_+ = [c_{l\sigma}^\dagger, c_{l'\sigma'}^\dagger]_+ = 0 . \end{aligned} \tag{2.5}$$

Here we are using the notation

$$\{l\} = i + \{\vec{k}\} , \tag{2.6}$$

i.e., the set of indices l represents the sum of the index i plus the indices $\vec{k}$.

We wish to look for an operator $\mathcal{O}$ such that

$$[\mathcal{O}, H] = \hbar\omega\, \mathcal{O} . \tag{2.7}$$

If Eq. (2.7) is *exactly* satisfied, $\hbar\omega$ is necessarily *real*. If $\hbar\omega$ is *positive*, $\mathcal{O}$ is a quasiparticle *destruction* operator associated with an excited state of the system containing one quasiparticle of energy $\hbar\omega$. If $\hbar\omega$ is *negative*, $\mathcal{O}$ is a quasiparticle *creation* operator associated with an excited state of the system containing one quasiparticle of energy $|\hbar\omega|$. If Eq. (2.7) is only *approximately* satisfied, then $\hbar\omega$ may be *complex*. In this case the signature of the real part of $\hbar\omega$ determines whether $\mathcal{O}$ is a quasiparticle creation or destruction operator. In any case, $\hbar\omega$ must be in either the second or the fourth quadrant of the complex ω plane to ensure that the quasiparticle excitations are *causal* (i.e., decay with increasing time).

In order to determine what operators to use in constructing $\mathcal{O}$, we calculate the commutator of $c_{l\sigma}$ and H,

$$[c_{k\sigma}, H_0] = \epsilon_k c_{k\sigma} , \tag{2.8}$$

$$[c_{k\sigma}, H_1] = V_k c_{i\sigma} , \tag{2.9}$$

$$[c_{i\sigma}, H_0] = \epsilon_0 c_{i\sigma} + U_0 N_{i,-\sigma} c_{i\sigma} , \tag{2.10}$$

$$[c_{i\sigma}, H_1] = \textstyle\sum_k V_{-k} c_{k\sigma} . \tag{2.11}$$

In Eq. (2.10) we are using the notation

$$N_{i\sigma} \equiv c_{i\sigma}^\dagger c_{i\sigma} \tag{2.12}$$

for the particle-number operator, in terms of which we can write H_0 as

$$H_0 = \sum_{k,\sigma} \epsilon_k N_{k\sigma} + \epsilon_0 \sum_\sigma N_{i\sigma} + U_0 N_{i\uparrow} N_{i\downarrow} . \tag{2.13}$$

Note that $[c_{l\sigma}, H]$ is a linear combination of the various $c_{l'\sigma}$ plus $N_{i,-\sigma} c_{i\sigma}$. This suggests that we calculate the commutatior of $N_{i,-\sigma} c_{i\sigma}$ and H, getting

$$[N_{i,-\sigma}c_{i\sigma}, H_0] = (\epsilon_0 + U_0)N_{i,-\sigma}c_{i\sigma} , \tag{2.14}$$

$$[N_{i,-\sigma}c_{i\sigma}, H_1] = \sum_k V_{-k}(N_{i,-\sigma}c_{k\sigma} - c^\dagger_{i,-\sigma}c_{i\sigma}c_{k,-\sigma}) - \sum_k V_k c_{i,-\sigma}c_{i\sigma}c^\dagger_{k,-\sigma} . \tag{2.15}$$

We now introduce the basic approximation of this paper. We *linearize* the right-hand side of Eq. (2.15) with respect to the set of operators $c_{i\sigma}$ and $N_{i,-\sigma}c_{i\sigma}$ (all possible i and σ). In other words, in the right-hand side of (2.15) we replace all operator coefficients multiplying members of the set by the corresponding *thermal averages*. Furthermore, we assume that there are just two types of nonvanishing thermal averages, namely,

$$n_{i\sigma} \equiv \langle N_{i\sigma}\rangle , \tag{2.16}$$

$$b_i \equiv \langle c^\dagger_{i\downarrow}c_{i\uparrow}\rangle . \tag{2.17}$$

Thus Eq. (2.15) is replaced by

$$[N_{i,-\sigma}c_{i\sigma}, H_1] = \sum_k V_{-k}[n_{i,-\sigma}c_{k\sigma} - (b_i\delta_{\sigma\uparrow} + b_i^*\delta_{\sigma\downarrow})c_{k,-\sigma}] . \tag{2.18}$$

(Note that $c_{k\sigma}$ and $c_{k,-\sigma}$ are the only operators remaining after the linearization procedure.)

The actual calculation of $n_{i\sigma}$ and b_i in a self-consistent fashion will be discussed later. The assumption of finite b_i introduces the possibility of pairing. Here it is hole-electron pairing, as in the excitonic insulator,[7] rather than electron-electron pairing, as in the superconductor.[8] Unlike the excitonic insulator, however, here a finite b_i indicates only that an electron is occupying the orbital i with its spin pointing neither straight up nor straight down.

Let us, for the moment, consider some arbitrary set of operators $\psi_{p\sigma}$. We introduce the vector operators

$$\Psi_p \equiv \begin{pmatrix}\psi_{p\uparrow}\\ \psi_{p\downarrow}\end{pmatrix}, \qquad \Psi_p^\dagger \equiv (\psi^\dagger_{p\uparrow}, \psi^\dagger_{p\downarrow}) . \tag{2.19}$$

We define the 2×2 matrix

$$\langle[\Psi_p; \Psi_q^\dagger]_+\rangle \equiv \begin{pmatrix}\langle[\psi_{p\uparrow}, \psi^\dagger_{q\uparrow}]_+\rangle & \langle[\psi_{p\uparrow}, \psi^\dagger_{q\downarrow}]_+\rangle\\ \langle[\psi_{p\downarrow}, \psi^\dagger_{q\uparrow}]_+\rangle & \langle[\psi_{p\downarrow}, \psi^\dagger_{q\downarrow}]_+\rangle\end{pmatrix} . \tag{2.20}$$

We will have need of the general matrix notation

$$M \equiv \begin{pmatrix}m_{\uparrow\uparrow} & m_{\uparrow\downarrow}\\ m_{\downarrow\uparrow} & m_{\downarrow\downarrow}\end{pmatrix}, \qquad M^\dagger \equiv \begin{pmatrix}m^\dagger_{\uparrow\uparrow} & m^\dagger_{\downarrow\uparrow}\\ m^\dagger_{\uparrow\downarrow} & m^\dagger_{\downarrow\downarrow}\end{pmatrix} . \tag{2.21}$$

An arbitrary M can be expanded in terms of the four matrices

$$\tau_1 = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix} , \quad i\tau_2 = \begin{pmatrix}0 & 1\\ -1 & 0\end{pmatrix} , \tag{2.22}$$

$$\tau_3 = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix} , \quad \tau_4 = \begin{pmatrix}1 & 0\\ 0 & 1\end{pmatrix} .$$

Equations (2.20) and (2.21) imply that

$$\langle[\Psi_p; \Psi_q^\dagger]_+\rangle = \langle[\Psi_q; \Psi_p^\dagger]_+\rangle^\dagger . \tag{2.23}$$

We define the vector operators

$$\Psi_{1i} \equiv \begin{pmatrix}c_{i\uparrow}\\ c_{i\downarrow}\end{pmatrix} , \quad \Phi_i \equiv \begin{pmatrix}N_{i\downarrow}c_{i\uparrow}\\ N_{i\uparrow}c_{i\downarrow}\end{pmatrix} , \tag{2.24}$$

$$\Psi_{2i} \equiv \Phi_i - \mathfrak{U}_i\Psi_{1i} , \tag{2.25}$$

where

$$\mathfrak{U}_i \equiv \langle[\Phi_i; \Psi^\dagger_{1i}]_+\rangle . \tag{2.26}$$

We also define

$$\Psi_k \equiv \begin{pmatrix}c_{k\uparrow}\\ c_{k\downarrow}\end{pmatrix} . \tag{2.27}$$

The first of Eqs. (2.5) now implies

$$\langle[\Psi_k; \Psi^\dagger_{k'}]_+\rangle = \delta_{kk'}\tau_4 , \tag{2.28}$$

$$\langle[\Psi_{1i}; \Psi^\dagger_{1i}]_+\rangle = \tau_4 . \tag{2.29}$$

From the definition of Ψ_{2i}, we have

$$\langle[\Psi_{2i}; \Psi^\dagger_k]_+\rangle = \langle[\Psi_k; \Psi^\dagger_{2i}]_+\rangle = 0 , \tag{2.30}$$

$$\langle[\Psi_{2i}; \Psi^\dagger_{1i}]_+\rangle = \langle[\Psi_{1i}; \Psi^\dagger_{2i}]_+\rangle = 0 , \tag{2.31}$$

and

$$\langle[\Psi_{2i}; \Psi^\dagger_{2i}]_+\rangle = \langle[(\Phi_i - \mathfrak{U}_i\Psi_{1i}); (\Phi_i^\dagger - \Psi^\dagger_{1i}\mathfrak{U}_i^\dagger)]_+\rangle = \{\langle[\Phi_i; \Phi_i^\dagger]_+\rangle - \mathfrak{U}_i\mathfrak{U}_i^\dagger\} . \tag{2.32}$$

It follows from Eq. (2.5) that

$$[c_{i\sigma}, (N_{i,-\sigma'}c_{i\sigma'})^\dagger]_+ = [N_{i,-\sigma}c_{i\sigma}, c^\dagger_{i\sigma'}]_+ = [N_{i,-\sigma}c_{i\sigma}, (N_{i,-\sigma'}c_{i\sigma'})^\dagger]_+ = \delta_{\sigma\sigma'}N_{i,-\sigma} - \delta_{\sigma,-\sigma'}c^\dagger_{i,-\sigma}c_{i\sigma} . \tag{2.33}$$

Thus

$$\mathfrak{U}_i = \mathfrak{U}_i^\dagger = \langle[\Phi_i; \Phi_i^\dagger]_+\rangle = \begin{pmatrix}n_{i\downarrow} & -b_i\\ -b_i^* & n_{i\uparrow}\end{pmatrix} = \tfrac{1}{2}(n_{i\downarrow} + n_{i\uparrow})\tau_4 + \tfrac{1}{2}(n_{i\downarrow} - n_{i\uparrow})\tau_3 - \tfrac{1}{2}(b_i + b_i^*)\tau_1 - \tfrac{1}{2}(b_i - b_i^*)i\tau_2 . \tag{2.34}$$

Equation (2.32) now becomes

$$\langle[\Psi_{2i}; \Psi^\dagger_{2i}]_+\rangle = \mathfrak{U}_i(\tau_4 - \mathfrak{U}_i) . \tag{2.35}$$

Equations (2.8)–(2.11), (2.14), and (2.18) can be rewritten, respectively,

$$[\Psi_k, H_0] = \epsilon_k\Psi_k , \tag{2.36}$$

$$[\Psi_k, H_1] = V_k\Psi_{1i} , \tag{2.37}$$

$$[\Psi_{1i}, H_0] = \epsilon_0\Psi_{1i} + U_0\Phi_i , \tag{2.38}$$

$$[\Psi_{1i}, H_1] = \sum_k V_{-k}\Psi_k , \tag{2.39}$$

$$[\Phi_i, H_0] = (\epsilon_0 + U_0)\Phi_i , \tag{2.40}$$

$$[\Phi_i, H_1] = \sum_k V_{-k}\mathfrak{u}_i\Psi_k . \tag{2.41}$$

Making use of the fact that

$$\Phi_i = \mathfrak{u}_i\Psi_{1i} + \Psi_{2i} , \tag{2.42}$$

we can rewrite Eqs. (2.38)–(2.41) as

$$[\Psi_{1i}, H] = (\epsilon_0\tau_4 + U_0\mathfrak{u}_i)\Psi_{1i} + U_0\tau_4\Psi_{2i} + \sum_k V_{-k}\tau_4\Psi_k , \tag{2.43}$$

$$[\Psi_{2i}, H] = U_0\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i)\Psi_{1i} + [\epsilon_0\tau_4 + U_0(\tau_4 - \mathfrak{u}_i)]\Psi_{2i} . \tag{2.44}$$

Note that (2.44), unlike (2.41), does *not* couple an operator associated with the center i to an operator associated with the Bloch state $\vec{k}$. Equations (2.36) and (2.37) can be combined,

$$[\Psi_k, H] = \epsilon_k\Psi_k + V_k\Psi_{1i} . \tag{2.45}$$

We see that the commutators of the last three equations can all be written as linear combinations of Ψ_{1i}, Ψ_{2i}, and Ψ_k. Thus we assume that the operator $\mathcal{O}$ of Eq. (2.7) can be written in the form

$$\mathcal{O} = \sum_p A_p^\dagger\Psi_p , \tag{2.46}$$

where

$$A_p \equiv \begin{pmatrix} a_{p'} \\ a_{p'} \end{pmatrix} \tag{2.47}$$

is a vector coefficient. Here we are using the notation

$$\{p\} = 1i + 2i + \{\vec{k}\} . \tag{2.48}$$

If we now substitute Eq. (2.46) into (2.7), take the anticommutator with respect to $\Psi_{p'}^\dagger$, and thermally average, we get the set of equations

$$\sum_p A_p^\dagger[K(p,p') - \hbar\omega\delta_{pp'}C(p)] = 0 , \tag{2.49}$$

where we are defining the matrices

$$C(p) \equiv \langle[\Psi_p ; \Psi_p^\dagger]_+\rangle , \tag{2.50}$$

$$K(p,p') \equiv \langle[[\Psi_p, H]; \Psi_{p'}^\dagger]_+\rangle . \tag{2.51}$$

The previous results imply that

$$C(1i) = C(\vec{k}) = \tau_4 , \tag{2.52}$$

$$C(2i) = \mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) , \tag{2.53}$$

$$K(1i,1i) = \epsilon_0\tau_4 + U_0\mathfrak{u}_i , \tag{2.54}$$

$$K(1i,2i) = K(2i,1i) = U_0\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) , \tag{2.55}$$

$$K(2i,2i) = \mathfrak{u}_i(\tau_4 - \mathfrak{u}_i)[\epsilon_0\tau_4 + U_0(\tau_4 - \mathfrak{u}_i)] , \tag{2.56}$$

$$K(\vec{k},\vec{k}') = \delta_{kk'}\epsilon_k\tau_4 , \tag{2.57}$$

$$K(\vec{k},1i) = V_k\tau_4 , \tag{2.58}$$

$$K(1i,\vec{k}) = V_k^*\tau_4 , \tag{2.59}$$

$$K(\vec{k},2i) = K(2i,\vec{k}) = 0 . \tag{2.60}$$

In turn, setting p' equal to $1i$, $2i$, and $\vec{k}'$, we can write Eq. (2.49) as

$$A_{1i}^\dagger[K(1i,1i) - \hbar\omega C(1i)] + A_{2i}^\dagger K(2i,1i) + \sum_k A_k^\dagger K(\vec{k},1i) = 0 , \tag{2.61}$$

$$A_{1i}^\dagger K(1i,2i) + A_{2i}^\dagger[K(2i,2i) - \hbar\omega C(2i)] + \sum_k A_k^\dagger K(\vec{k},2i) = 0 , \tag{2.62}$$

$$A_{1i}^\dagger K(1i,\vec{k}') + A_{2i}^\dagger K(2i,\vec{k}') + \sum_k A_k^\dagger[K(\vec{k},\vec{k}') - \hbar\omega\delta_{kk'}C(\vec{k}')] = 0 . \tag{2.63}$$

Substituting in the explicit values of the K's and the C's, we get, respectively,

$$A_{1i}^\dagger[(\epsilon_0 - \hbar\omega)\tau_4 + U_0\mathfrak{u}_i] + A_{2i}^\dagger U_0\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) + \sum_k A_k^\dagger V_k\tau_4 = 0 , \tag{2.64}$$

$$A_{1i}^\dagger U_0\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) + A_{2i}^\dagger\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) \times [(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i] = 0 , \tag{2.65}$$

$$A_{1i}^\dagger V_{k'}^*\tau_4 + A_{k'}^\dagger(\epsilon_{k'} - \hbar\omega)\tau_4 = 0 . \tag{2.66}$$

Equation (2.65) gives immediately

$$A_{2i}^\dagger = -A_{1i}^\dagger U_0[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i]^{-1} . \tag{2.67}$$

Substituting this into (2.64), we get

$$A_{1i}^\dagger\{[(\epsilon_0 - \hbar\omega)\tau_4 + U_0\mathfrak{u}_i][(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i] - U_0^2\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i)\}[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i]^{-1} + \sum_k A_k^\dagger V_k\tau_4 = 0 . \tag{2.68}$$

This can be greatly simplified because of the identity

$$[(\epsilon_0 - \hbar\omega)\tau_4 + U_0\mathfrak{u}_i][(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i] - U_0^2\mathfrak{u}_i(\tau_4 - \mathfrak{u}_i) \equiv (\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)\tau_4 . \tag{2.69}$$

Thus

$$A_{1i}^\dagger(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i]^{-1} + \sum_k A_k^\dagger V_k\tau_4 = 0 . \tag{2.70}$$

Equations (2.66) and (2.70) represent a system of coupled equations for the unknown $A_{1i}^\dagger$ and $A_k^\dagger$'s. We can immediately eliminate the $A_{1i}^\dagger$ in terms of the $A_k^\dagger$'s, or vice versa.

III. CONDUCTION-BAND QUASIPARTICLES

From Eq. (2.70), we have

$$A_{1i}^\dagger = -(\epsilon_0 - \hbar\omega)^{-1}(\epsilon_0 + U_0 - \hbar\omega)^{-1} \times \sum_k A_k^\dagger V_k[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0\mathfrak{u}_i] . \tag{3.1}$$

Substituting this into Eq. (2.66), we get

$$A_k^\dagger[(\epsilon_k - \hbar\omega)(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)[(\epsilon_0 + U_0 - \hbar\omega)\tau_4$$

$$- U_0 \mathfrak{u}_i]^{-1} - |V_k|^2 \tau_4\} - \sum_{k' \neq k} A^\dagger_{k'} V^*_k V_{k'} = 0 \ . \quad (3.2)$$

In the limit of a macroscopic-sized crystal, the term $-|V_k|^2\tau_4$ becomes negligibly small, relative to the term to which it is added, so that (3.2) may be replaced by

$$A^\dagger_k(\epsilon_k - \hbar\omega)(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0 \mathfrak{u}_i]^{-1} - \sum_{k' \neq k} A^\dagger_{k'} V^*_k V_{k'} = 0 \ . \quad (3.3)$$

We look for a solution to Eq. (3.3) where one particular vector coefficient, say $A^\dagger_k$, is much larger than all the other $A^\dagger_{k'}$. Equation (3.3) can be supplemented by the equation for $A^\dagger_{k'}(\vec{k}' \neq \vec{k})$,

$$A^\dagger_{k'}(\epsilon_{k'} - \hbar\omega)(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0 \mathfrak{u}_i]^{-1} = \sum_{k'' \neq k'} A^\dagger_{k''} V^*_{k''} V_{k'} \cong A^\dagger_k V^*_{k'} V_k \ . \quad (3.4)$$

Here we are assuming that the localized center only weakly perturbs the conduction-band quasiparticles. Substituting (3.4) into (3.3), we get

$$A^\dagger_k\{(\epsilon_k - \hbar\omega)\tau_4 - (\epsilon_0 - \hbar\omega)^{-2}(\epsilon_0 + U_0 - \hbar\omega)^{-2}|V_k|^2 W \times [(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0 \mathfrak{u}_i]^2\} = 0 \ . \quad (3.5)$$

Here we have defined

$$W \equiv \sum_k |V_k|^2 (\epsilon_k - \hbar\omega)^{-1} \ . \quad (3.6)$$

The term involving $|V_k|^2$ in (3.5) is negligible for the same reason that the term $-|V_k|^2\tau_4$ in (3.2) was negligible. Thus we get

$$A^\dagger_k(\epsilon_k - \hbar\omega) = 0 \ , \quad (3.7)$$

so that

$$\hbar\omega = \epsilon_k \quad (3.8)$$

for the conduction-band quasiparticles.

The presence of the center causes a weak coupling between Bloch states $\vec{k}$ and $\vec{k}'$. If the center itself has no magnetic moment, then this coupling is between states of the same spin; but if the center has a net magnetic moment, then some mixing of opposite-spin states will occur. As a result, the conduction electrons in the vicinity of the center may be partially spin polarized. It is known that this spin coupling between the center and the conduction band is antiferromagnetic.[1,9] Thus any magnetic moment of the center will tend to be screened by the conduction band. We will not attempt to calculate such an effect in this paper.

IV. LOCALIZED QUASIPARTICLES

From Eq. (2.66), we have

$$A^\dagger_k = -(\epsilon_k - \hbar\omega)^{-1} V^*_k A^\dagger_{1i} \ . \quad (4.1)$$

Substituting this into Eq. (2.70), we get

$$A^\dagger_{1i}\{(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - W[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0 \mathfrak{u}_i]\} = 0 \ . \quad (4.2)$$

This represents a pair of linear equations in the unknown coefficients $a^*_{i\uparrow}$ and $a^*_{i\downarrow}$. The corresponding secular equation is

$$\det\{(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - W[(\epsilon_0 + U_0 - \hbar\omega)\tau_4 - U_0 \mathfrak{u}_i]\} = 0 \ . \quad (4.3)$$

Introducing the notation

$$p \equiv \epsilon_0 + U_0 - \tfrac{1}{2} U_0 (n_{i\downarrow} + n_{i\uparrow}) \ , \quad (4.4)$$

$$q \equiv \tfrac{1}{2} U_0 (n_{i\downarrow} - n_{i\uparrow}) \ , \quad (4.5)$$

$$\Delta \equiv U_0 b_i \ , \quad (4.6)$$

$$w \equiv (q^2 + |\Delta|^2)^{1/2} \ , \quad (4.7)$$

$$\mathfrak{W} \equiv W^{-1}(\epsilon_0 - \hbar\omega)(\epsilon_0 + U_0 - \hbar\omega) - (p - \hbar\omega) \ , \quad (4.8)$$

we can rewrite Eq. (4.3) as

$$\begin{vmatrix} (-\mathfrak{W} - q) & \Delta \\ \Delta^* & (-\mathfrak{W} + q) \end{vmatrix} = 0 \ . \quad (4.9)$$

Therefore, we have

$$\mathfrak{W} + \mu w = 0 \ , \quad (4.10)$$

where

$$\mu = \pm 1 \ . \quad (4.11)$$

Substituting (4.8) into (4.10), we get a quadratic equation for ω, the solution to which is

$$\hbar\omega = \hbar\omega_{\mu\nu} \equiv \epsilon_0 + \tfrac{1}{2}(U_0 - W) + \nu\{\tfrac{1}{4}(U_0 + W)^2 - W[\tfrac{1}{2} U_0 (n_{i\downarrow} + n_{i\uparrow}) + \mu w]\}^{1/2} \ , \quad (4.12)$$

where

$$\nu = \pm 1 \ , \quad (4.13)$$

it being understood that the choice of sign for ν is independent of that for μ.

Because of the two possibilities each for μ and ν, there are four roots $\hbar\omega_{\mu\nu}$ associated with localized quasiparticles. The reason for four roots can be understood in the following way: In the absence of coupling between the center and the conduction band (i.e., $W = 0$), there are two roots, namely, ϵ_0 and $\epsilon_0 + U_0$, these being the energies associated with putting an electron on an empty center, and on a center already occupied by an opposite-spin electron, respectively. However, each of these roots is doubly degenerate because of the two possible spin orientations of the electron being put on the center. Thus there are four roots. If we now reintroduce the coupling between the center and the

conduction band ($W \neq 0$), and if the center has a net average magnetic moment, then the spin degeneracy of the two pairs of roots will be lifted. For example, the energy required to put an electron on the empty site will depend on the spin of the electron, because of the coupling between the electron and the net spin polarization of the conduction band in the vicinity of the center. This spin polarization of the conduction band is, of course, in turn due to the average net moment on the center, as has already been discussed. Such a spin polarization is implied by Eq. (3.5), entering through the term $U_0 \mathfrak{u}_i$. Although the polarization of a single conduction electron is negligible, in the sense that Eq. (3.5) can be replaced by Eq. (3.7), the resultant polarization of *all* the conduction electrons is by no means negligible in its effect on the center. Precisely this effect is included implicitly in Eqs. (4.2) and (4.3).

In order to evaluate W we replace the k sum of Eq. (3.6) by the equivalent integration (assuming a crystal of unit volume)

$$W = (2\pi)^{-3} \int d^3k \, |V_k|^2 (\epsilon_k - \hbar\omega)^{-1} . \tag{4.14}$$

In this integral we make the usual replacement

$$\hbar\omega \to \hbar\omega + i\eta_\omega , \tag{4.15}$$

where η_ω is an infinitesimal having the same signature as the real part of ω. This implies that

$$\omega = \omega_1 - i\omega_2 \tag{4.16}$$

will lie in either the second or the fourth quadrant of the complex ω plane, as was discussed in Sec. II. Making use of the formal relation

$$\lim_{\eta \to 0+} (x \pm i\eta)^{-1} = \mathcal{P}(1/x) \mp i\pi\delta(x) , \tag{4.17}$$

where $\mathcal{P}$ denotes "principal part of," we can evaluate (4.14). Writing

$$W = W_1 - iW_2 , \tag{4.18}$$

we have

$$W_1 = (2\pi)^{-3} \mathcal{P} \int d^3k \, |V_k|^2 (\epsilon_k - \hbar\omega_1)^{-1} , \tag{4.19}$$

$$W_2 = -\tfrac{1}{2}(2\pi)^{-2} (\mathrm{sgn}\omega_1) \int d^3k \, |V_k|^2 \delta(\epsilon_k - \hbar\omega_1) . \tag{4.20}$$

It is often a good approximation to neglect the ω dependence of W by setting $\omega_1 = 0$ in the integrands of Eqs. (4.19) and (4.20). Note that W lies in either the first or the fourth quadrant of the complex plane. Leaving aside the factor of ν, the square root involving W in Eq. (4.12) should be taken in the same quadrant of the complex plane as that of W. All quantities other than W in the expression for $\hbar\omega_{\mu\nu}$ are real.

At this point we need to determine $n_{i\sigma}$ and b_i in a self-consistent fashion. Fortunately, this process can be carried out in a manner formally identical to that of the Appendix of II. First we need some definitions. The statistical factor

$$f_{\mu\nu} \equiv f(\omega_{\mu\nu}) \tag{4.21}$$

is given by

$$f(\omega) \equiv \pi^{-1} |\omega_2| \int_{-\infty}^{\infty} d\omega' [(\omega' - \omega_1)^2 + \omega_2^2]^{-1} [e^{\beta\hbar\omega'} + 1]^{-1} . \tag{4.22}$$

Next we define

$$b_{\mu\nu} \equiv [2(\epsilon_0 + \tfrac{1}{2}U_0 - \hbar\omega_{\mu\nu})(p - \hbar\omega_{\mu\nu} - \mu w) - (\epsilon_0 - \hbar\omega_{\mu\nu})(\epsilon_0 + U_0 - \hbar\omega_{\mu\nu})]^{-1/2} . \tag{4.23}$$

[Do not confuse $b_{\mu\nu}$ with b_i of Eq. (2.17).] Finally we define the *weighted* statistical factor

$$g_\mu \equiv (\textstyle\sum_\nu |b_{\mu\nu}|^{-2})^{-1} (\sum_\nu |b_{\mu\nu}|^{-2} f_{\mu\nu}) . \tag{4.24}$$

Note that both $f_{\mu\nu}$ and g_μ are real, and lie between zero and one. The results of the Appendix of II are that

$$(n_{i\downarrow} + n_{i\uparrow}) = (g_- + g_+) , \tag{4.25}$$

$$(n_{i\downarrow} - n_{i\uparrow}) = (q/w)(g_- - g_+) , \tag{4.26}$$

$$b_i = (\Delta/2w)(g_- - g_+) . \tag{4.27}$$

Substitution of these last three equations into Eqs. (4.4)–(4.6) gives, respectively,

$$p = \epsilon_0 + U_0 - \tfrac{1}{2}U_0(g_- + g_+) , \tag{4.28}$$

$$q = \tfrac{1}{2}U_0(q/w)(g_- - g_+) , \tag{4.29}$$

$$\Delta = \tfrac{1}{2}U_0(\Delta/w)(g_- - g_+) . \tag{4.30}$$

Note that Eqs. (4.29) and (4.30) are the same equation, namely,

$$w = \tfrac{1}{2}U_0(g_- - g_+) . \tag{4.31}$$

This means that q and Δ are not separately determinable. Subject only to Eq. (4.7) being satisfied, they are otherwise arbitrary. Of course q, by definition, is real, but Δ may be complex.

It is clear that

$$S_z \equiv -\tfrac{1}{2}\hbar(n_{i\downarrow} - n_{i\uparrow}) \tag{4.32}$$

is the z component of spin angular momentum on the localized center. From the defintion of b_i [Eq. (2.17)], it is similarly clear that

$$S_x - iS_y \equiv \hbar b_i \tag{4.33}$$

is the average value of the spin step-down operator Substituting (4.5) into (4.32), (4.6) into (4.33), we get

$$S_z = -(\hbar/U_0)q , \quad S_x - iS_y = (\hbar/U_0)\Delta . \tag{4.34}$$

Thus the *magnitude* of the spin on the center is

$$S = (\hbar/U_0)w = \tfrac{1}{2}\hbar(g_- - g_+) . \tag{4.35}$$

Note that this magnitude may be temperature dependent (through the β appearing in $f_{\mu\nu}$). The nonuniqueness of q and Δ indicates that the spin of the center can be *arbitrarily oriented*. There is a local moment when $g_+ < g_-$; there is no local moment when $g_+ = g_-$.

Using Eqs. (4.25) and (4.31), we can rewite Eq. (4.12) as

$$\hbar\omega_{\mu\nu} = \epsilon_0 + \tfrac{1}{2}(U_0 - W) + \nu[\tfrac{1}{4}(U_0 + W)^2 - U_0 W g_{-\mu}]^{1/2} . \tag{4.36}$$

With the aid of Eq. (4.20), and the fact that $0 \le g_\mu \le 1$, it is easy to check that all four roots $\hbar\omega_{\mu\nu}$ lie in either the second or fourth quadrant, as is required.

Equation (4.10) can be rewritten

$$(\epsilon_0 - \hbar\omega_{\mu\nu})(\epsilon_0 + U_0 - \hbar\omega_{\mu\nu}) = W(p - \hbar\omega_{\mu\nu} - \mu w) . \tag{4.37}$$

Substituting this into (4.23) gives

$$b_{\mu\nu} = \{2[\epsilon_0 + \tfrac{1}{2}(U_0 - W) - \hbar\omega_{\mu\nu}][p - \hbar\omega_{\mu\nu} - \mu w]\}^{-1/2} . \tag{4.38}$$

We note that

$$\epsilon_0 + \tfrac{1}{2}(U_0 - W) - \hbar\omega_{\mu\nu} = -\nu[\tfrac{1}{4}(U_0 + W)^2 - U_0 W g_{-\mu}]^{1/2}, \tag{4.39}$$

$$p - \mu w = \epsilon_0 + U_0(1 - g_{-\mu}) . \tag{4.40}$$

Thus

$$|b_{\mu\nu}|^{-2} = |(U_0 + W)^2 - 4U_0 W g_{-\mu}|^{1/2} \times |\epsilon_0 + U_0(1 - g_{-\mu}) - \hbar\omega_{\mu\nu}| . \tag{4.41}$$

Note that the square-root factor on the right-hand side of (4.41) is independent of ν. Thus this factor cancels out of the right-hand side of (4.24), and we get

$$g_\mu = \frac{\sum_\nu |\epsilon_0 + U_0(1 - g_{-\mu}) - \hbar\omega_{\mu\nu}| f_{\mu\nu}}{\sum_\nu |\epsilon_0 + U_0(1 - g_{-\mu}) - \hbar\omega_{\mu\nu}|} . \tag{4.42}$$

From Eq. (4.36) we see that $\hbar\omega_{\mu\nu}$, and thus $f_{\mu\nu}$ also, depends on $g_{-\mu}$, but not on g_μ. Combining this with Eq. (4.42) we see that g_μ is a function of $g_{-\mu}$, and vice versa. We define

$$G_\nu(x) \equiv \epsilon_0 + \tfrac{1}{2}(U_0 - W) + \nu[\tfrac{1}{4}(U_0 + W)^2 - U_0 W x]^{1/2} \tag{4.43}$$

and

$$F(x) \equiv \frac{\sum_\nu |\epsilon_0 + U_0(1 - x) - G_\nu(x)| f(G_\nu(x))}{\sum_\nu |\epsilon_0 + U_0(1 - x) - G_\nu(x)|} . \tag{4.44}$$

[The function f in (4.44) is that defined in Eq. (4.22).] Equation (4.42) can thus be written

$$g_\mu = F(g_{-\mu}) . \tag{4.45}$$

This is a pair of coupled functional equations, the solutions to which give g_+ and g_-.

From Eqs. (4.43) and (4.44) we note that

$$F(0) = f(\epsilon_0 - W) , \tag{4.46}$$

$$F(1) = f(\epsilon_0 + U_0 - W) , \tag{4.47}$$

so that

$$F(0) \ge F(1) . \tag{4.48}$$

This means that the curve $y = F(x)$ will always intersect the curve $y = x$ somewhere in the unit square $0 \le x \le 1$, $0 \le y \le 1$. Thus there is *always* a solution to Eq. (4.45) corresponding to $g_+ = g_-$ (no localized moment). In addition, there will be a solution $g_+ < g_-$ (finite localized moment) whenever

$$-\left.\frac{dF}{dx}\right|_{F(x)=x} > 1 . \tag{4.49}$$

The conditions under which (4.49) is satisfied must be investigated numerically. At the absolute zero of temperature ($T = 0$), for example, (4.49) implies certain conditions on the parameters ϵ_0, U_0, and W in order that there be a local moment. It is believed that whenever a local moment can exist it will have a lower free energy than the nonmagnetic solution. For given values of ϵ_0, U_0, and W, Eq. (4.49) also implies a maximum temperature above which no local moment exists. Note that such a temperature is not necessarily the Curie temperature associated with a finite density of localized moments in a crystal.

It is interesting to compare the results of this paper with the original Hartree-Fock treatment of the same problem by Anderson[2] (or with the results of I, equivalent to those of Anderson in the limit of vanishing density of centers). In this effective-field treatment there are just two (rather than four) localized quasiparticle energies, one for each direction of spin. These are

$$\hbar\omega_\mu = \epsilon_0 - W + U_0 f_{-\mu} , \tag{4.50}$$

where

$$f_\mu \equiv f(\hbar\omega_\mu) , \tag{4.51}$$

the symbols ϵ_0, W, U_0, μ, and f having the same meaning as before. Equation (4.50) can be recast in the following fashion. We replace Eq. (4.42) by

$$g_\mu \equiv f_\mu , \tag{4.52}$$

Eq. (4.43) by

$$G(x) \equiv \epsilon_0 - W + U_0 x , \tag{4.53}$$

and Eq. (4.44) by

$$F(x) \equiv f(G(x)) . \tag{4.54}$$

Equation (4.50) now becomes Eq. (4.45). Thus the difference between Anderson's treatment and that of the present paper lies in the difference in the two sets of definitions of $G(x)$ and $F(x)$. Note that Eqs. (4.46)–(4.49) still hold true in the Anderson treatment.

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Field Dependence of the Magnetization and Spin-Wave Correlations in Ferromagnetic $CrBr_3$ †

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Using the Cr^{53} NMR, the field dependence of the magnetization $M(T_0, H)$ at elevated temperatures of single-crystal ferromagnetic $CrBr_3$ has been determined. While an accurate fit to the $M(T_0, H)$-vs-H data can be obtained using a two-exchange-parameter first-order renormalized spin-wave theory, the parameters required differ appreciably from those required for an accurate fit to the previously obtained $M(T, 0)$-vs-T data with the same theory. A t-matrix two-parameter–model theory, correct to lowest order in the magnon density, but to all orders in the magnon-magnon interaction, was constructed. Although both the first-order and t-matrix corrections to the spin-wave energies are sizable, the full t-matrix results only change the large first-order corrections to $M(T, H)$ by 15%. However, even if one employs the full t-matrix renormalization there are no pairs of values of the two exchange parameters which simultaneously fit the $M(T, 0)$-vs-T and $M(T_0, H)$-vs-H data. We attribute the inability of the more sophisticated theory to provide agreement with measurements of more than one thermodynamic function to be an inadequacy of the two-parameter model rather than an intrinsic failure of the theoretical approach.

I. INTRODUCTION

$CrBr_3$ is one of the few ferromagnetic insulators.[1] The study of its magnetic properties has made possible detailed comparisons of experiment with the predictions of spin-wave theory for a Heisenberg ferromagnet.[2–4] Until recently, precise measurements of the temperature dependence of the magnetization using nuclear-magnetic-resonance (NMR) techniques have been confined to the case of no external magnetic field because of the difficulty in growing large single crystals.

A series of NMR studies have been made on a $CrBr_3$ single crystal which give information on the magnetization processes and the mechanisms involved in the enhancements of the NMR signals that