tered at quite low temperatures if it were possible to apply a suitably oriented magnetic field of sufficient magnitude to give each Ni^{++} ion a substantial magnetic moment. The interesting possibility of observing long-range ordering of such induced moments depends critically on the structure of the salt which, unfortunately, is not yet known in detail.[15] We are unable, at present, to predict whether or not such effects should be observable. While A appears to be large in comparison with typically dipolar interaction energies, accurate estimates of the relative importance of exchange and dipolar coupling in $Ni(NO_3)_2 \cdot 6H_2O$ must also await the full determination of its structure. Further magnetic and thermal studies of single crystals of this substance at very low temperatures are being undertaken.

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[*] Work supported in part by the National Science Foundation and the Office of Naval Research.

[†] Present address: Department of Physics, Eindhoven University of Technology, Eindhoven, The Netherlands.

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Extra-Orbital Model of a Dilute Metallic Alloy*

R. H. Parmenter
Department of Physics, University of Arizona, Tucson, Arizona 85721

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Accurate values of both localized and conduction-band quasiparticle excitation energies are calculated for the extra-orbital model of a dilute metallic alloy. It is shown that this model can always be made self-consistent in the sense of satisfying the Friedel sum rule.

I. INTRODUCTION

In recent years, the most intensively studied model of a metallic alloy has been the so-called single-site approximation.[1,2] Here we wish to discuss a somewhat different model, one particularly appropriate where the minority constituents of the alloy are transition elements. We approximate the effect of each impurity atom by that of an *extra orbital*, coupled to the conduction band of the host metal. As Velický *et al.*[2] have pointed out, the single-site approximation is related to the many-electron Hubbard model[3] of a narrow-band solid. Similarly, the extra-orbital approximation is related to the many-electron Anderson model[4] of a localized magnetic center in a metal.

In this paper we restrict the discussion to a dilute alloy containing a single type of impurity element. We use the operator equation-of-motion method[5] to find accurate values for both localized and conduction-band quasiparticle excitation energies. Just as Stern[6] has done recently for the single-site model, we shall investigate the *self-consistency* of the extra-orbital model by means of the Friedel sum rule.[7] In contrast to Stern's conclusions, we shall find that the extra-orbital model can always be made self-consistent.

II. EQUATIONS OF MOTION

The Hamiltonian of our system is

$$H=\sum_k \epsilon_k c_k^\dagger c_k+\epsilon_0\sum_i c_i^\dagger c_i + \sum_{k,i}\left(V_{ki}c_k^\dagger c_i+V_{-ki}c_i^\dagger c_k\right) . \tag{2.1}$$

The k sum represents the Hamiltonian of the pure metal; the i sum that of the localized impurities; the k, i sum that of the coupling between conduction band and impurities. The one-electron energies ϵ_k (for the conduction band) and ϵ_0 (for the impurities) are measured relative to the Fermi level. For convenience, we restrict ourselves to the case where the conduction-band electrons have a positive effective mass. The total Hamiltonian represents a system of conduction-band electrons hopping in and out of localized s orbitals on the impurity centers. Since we have a one-electron problem, the spin indices have been suppressed. Since H is Hermitian,

$$V_{ki}^* = V_{-ki} . \tag{2.2}$$

We will later need the fact that

$$V_{ki}=V_k e^{-i\vec{k}\cdot\vec{R}_i} , \tag{2.3}$$

where $\vec{R}_i$ is the position of the ith center, and V_k is the matrix element for a center localized at the origin. The electron creation and destruction operators obey the usual anticommutation relations

$$\begin{aligned}&[c_q, c_{q'}^\dagger]_+ = \delta_{qq'} , \\ &[c_q, c_{q'}]_+ = [c_q^\dagger, c_{q'}^\dagger]_+ = 0 .\end{aligned} \tag{2.4}$$

The set of indices q represents the sum of the indices $\vec{k}$ plus the indices i.

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

$$[\mathcal{O}, H]=\hbar\omega\mathcal{O} . \tag{2.5}$$

If this equation 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. (2.5) 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). The lifetime of a quasiparticle is given by

$$\tau=(2|\omega_2|)^{-1} , \tag{2.6}$$

where $-i\omega_2$ is the imaginary part of ω.

Calculating the commutors of the various c_q with respect to H, we get

$$[c_k, H]=\epsilon_k c_k+\textstyle\sum_i V_{ki}c_i , \tag{2.7}$$

$$[c_i, H]=\epsilon_0 c_i+\textstyle\sum_k V_{-ki}c_k . \tag{2.8}$$

Since every $[c_q, H]$ is a linear combination of the various c's, we can write

$$\mathcal{O}=\textstyle\sum_q a_q c_q , \tag{2.9}$$

where the a_q are unknown coefficients. Substituting (2.9) into (2.5) and taking the anticommutator with respect to $c_{q'}^\dagger$, we get the set of equations

$$\textstyle\sum_q \{[[c_q, H], c_{q'}^\dagger]_+ - \hbar\omega\delta_{qq'}\}a_q=0 \tag{2.10}$$

or, specifically,

$$(\epsilon_k-\hbar\omega)a_k+\textstyle\sum_i V_{-ki}a_i=0 , \tag{2.11}$$

$$(\epsilon_0-\hbar\omega)a_i+\textstyle\sum_k V_{ki}a_k=0 . \tag{2.12}$$

Eliminating the a_k in favor of the a_i, we get

$$(\epsilon_0-W-\hbar\omega)a_i = \sum_k |V_k|^2(\epsilon_k-\hbar\omega)^{-1}\sum_{j\neq i} e^{i\vec{k}\cdot(\vec{R}_j-\vec{R}_i)}a_j , \tag{2.13}$$

where

$$W\equiv\textstyle\sum_k |V_k|^2(\epsilon_k-\hbar\omega)^{-1}=(2\pi)^{-3}\int d^3k\,|V_k|^2(\epsilon_k-\hbar\omega)^{-1} . \tag{2.14}$$

Alternatively, we can eliminate the a_i in favor of the a_k, obtaining

$$G_k a_k=\sum_{k'\neq k} V_k^* V_{k'} a_{k'}\sum_i e^{-i(\vec{k}'-\vec{k})\cdot\vec{R}_i} , \tag{2.15}$$

where

$$G_k\equiv(\epsilon_k-\hbar\omega)(\epsilon_0-\hbar\omega)-n_0|V_k|^2 . \tag{2.16}$$

Here n_0 is the number of impurities in the crystal. Since we are assuming a crystal of unit volume, n_0 is also the number density of centers. Since we are primarily interested in ω near the real axis, we make use of the formal relation

$$\lim_{\eta\to 0^+}(x\pm i\eta)=\mathcal{P}\left(\frac{1}{x}\right)\mp i\pi\delta(x) \tag{2.17}$$

in evaluating the self-energy W. Writing

$$\omega=\omega_1-i\omega_2 , \tag{2.18}$$

$$W=W_1-iW_2 , \tag{2.19}$$

we get

$$W_1=(2\pi)^{-3}\,\mathcal{P}\int d^3k\,|V_k|^2(\epsilon_k-\hbar\omega_1)^{-1} , \tag{2.20}$$

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

$$= -(2\pi)^{-1}(\operatorname{sgn}\omega_1)\left[k^2\left(\frac{d\epsilon_k}{dk}\right)^{-1} |V_k|^2\right._{\epsilon_k=\hbar\omega_1} . \tag{2.21}$$

III. LOCALIZED QUASIPARTICLES

We look for solutions to Eq. (2.13). In the limit of small n_0, we can always assume that one particular coefficient, say a_i, is much larger than all the other a_j. Thus for a_j ($j \neq i$), Eq. (2.13) becomes

$$(\epsilon_0 - W - \hbar\omega)\, a_j \cong \textstyle\sum_k |V_k|^2(\epsilon_k - \hbar\omega)^{-1} e^{i\vec{k}\cdot(\vec{R}_i-\vec{R}_j)} a_i . \tag{3.1}$$

Substituting this into (2.13), we get

$$(\epsilon_0 - W - \hbar\omega)^2 - I_i = 0 , \tag{3.2}$$

where we are defining

$$I_i \equiv (2\pi)^{-6}\int\int d^3k\, d^3k' |V_k|^2 |V_{k'}|^2 (\epsilon_k - \hbar\omega)^{-1} \times(\epsilon_{k'} - \hbar\omega)^{-1} \sum_{j\neq i} e^{i(\vec{k}-\vec{k}')\cdot(\vec{R}_j-\vec{R}_i)} . \tag{3.3}$$

When n_0 is sufficiently small, it is a good approximation to replace the sum over $j \neq i$ in (3.3) by the equivalent integral, whereupon $I_i = I$ becomes independent of the index i. It can be shown[8] that

$$I = \tfrac{1}{2} n_0 (2\pi)^{-6} \int d^3k\, |V_k|^4 (\hbar^2k^2/2m)^{-1}(\epsilon_k - \hbar\omega)^{-1} , \tag{3.4}$$

where m is the effective mass of the conduction electrons. Note that $(\hbar^2k^2/2m)$ causes no singularity in the integrand at $\vec{k} = 0$. The real and imaginary parts of I can be evaluated in a fashion equivalent to that used for W in Sec. II. Since we are assuming a positive effective mass, W and I lie in the same quadrant of the ω plane (the first or the fourth). We define $I^{1/2}$ to lie in the same quadrant as does I. Equation (3.2) now gives

$$\hbar\omega = \hbar\omega_i \equiv \epsilon_0 - W - I^{1/2} . \tag{3.5}$$

Here we have chosen that root of (3.2) which causes $I^{1/2}$ to contribute to the imaginary part of $\hbar\omega$ with the same signature as does that of W. Each impurity has its energy level lowered and broadened (as given by $-W$) by virtue of interaction with the conduction electrons. In addition, each impurity has its energy level further lowered and broadened (as given by $-I^{1/2}$) by virtue of its effective interaction with all the other impurity atoms (through the intermediary of the conduction band). Note that this latter lowering and broadening is proportional to $n_0^{1/2}$. This is a manifestation of the long-range nature of the effective interaction.

IV. CONDUCTION-BAND QUASIPARTICLES

We look for solutions to Eq. (2.15). We *tentatively* assume that one particular coefficient, say a_k, is much larger than all the other $a_{k'}$ ($\vec{k}' \neq \vec{k}$). Unlike the corresponding assumption in Sec. III, it is by no means obvious that this assumption is true here, even in the limit as n_0 vanishes. We are, in effect, invoking the Born approximation. For $a_{k'}$ ($\vec{k}' \neq \vec{k}$), Eq. (2.15) becomes

$$G_{k'}\, a_{k'} = V^*_{k'} V_k a_k \sum_j e^{-i(\vec{k}-\vec{k}')\cdot\vec{R}_j} + \sum_{k''\neq k,k'} V^*_{k'} V_{k''} a_{k''} \sum_j e^{-i(\vec{k}''-\vec{k}')\cdot\vec{R}_j} . \tag{4.1}$$

We assume

$$a_{k''} \cong G^{-1}_{k''} V^*_{k''} V_k a_k \textstyle\sum_p e^{-i(\vec{k}-\vec{k}'')\cdot\vec{R}_p} . \tag{4.2}$$

Substituting (4.2) into (4.1), we get

$$a_{k'} = G^{-1}_{k'} V^*_{k'} V_k a_k \left(\sum_j e^{-i(\vec{k}-\vec{k}')\vec{R}_j} + \sum_{k''\neq k,k'} |V_{k''}|^2 G^{-1}_{k''} \sum_{j,p} e^{i(\vec{k}'\cdot\vec{R}_j - \vec{k}\cdot\vec{R}_p)} \times e^{-i\vec{k}''\cdot(\vec{R}_j-\vec{R}_p)}\right) . \tag{4.3}$$

In turn, substituting (4.3) into (2.15), we get

$$G_k = |V_k|^2 \sum_{k'\neq k} |V_{k'}|^2 G^{-1}_{k'} \sum_{i,j} e^{-i(\vec{k}'-\vec{k})\cdot(\vec{R}_i-\vec{R}_j)} + |V_k|^2 \sum_{k'\neq k}\ \sum_{k''\neq k,k'} |V_{k'}|^2 |V_{k''}|^2 G^{-1}_{k'} G^{-1}_{k''} \times \sum_{i,j,p} e^{i\vec{k}\cdot(\vec{R}_i-\vec{R}_p)} e^{i\vec{k}'\cdot(\vec{R}_j\cdot\vec{R}_i)} e^{i\vec{k}''\cdot(\vec{R}_p-\vec{R}_j)} . \tag{4.4}$$

Equation (4.4) represents the *second* Born approximation. Because of destructive interference, the double sum over i and j reduces to a single sum ($i = j$); the triple sum over i, j, and p reduces to a double sum ($i = p$). We approximate the quantities $G^{-1}_{k'}$ and $G^{-1}_{k''}$ by dropping the terms proportional to n_0. Making use of the definitions of W and I, we thus get

$$G_k = n_0 |V_k|^2 (\epsilon_0 - \hbar\omega)^{-1} W + n_0 |V_k|^2 (\epsilon_0 - \hbar\omega)^{-2}(W^2 + I) . \tag{4.5}$$

Note that the terms involving W are *linear* in n_0, whereas that involving I is *quadratic* in n_0. Thus, in the dilute limit we drop the latter term, getting

$$G_k = n_0 |V_k|^2 (\epsilon_0 - \hbar\omega)^{-1} W [1 + (\epsilon_0 - \hbar\omega)^{-1} W] . \tag{4.6}$$

It can be seen that if we perform the iteration process for $a_{k'}$ an indefinite number of times (rather than twice as done above), then in the low-concentration limit, Eq. (4.6) is replaced by

$$G_k = n_0 |V_k|^2 \sum_{n=1}^{\infty} [(\epsilon_0 - \hbar\omega)^{-1} W]^n . \tag{4.7}$$

Keeping only the first n terms in this series represents the nth *Born approximation*. Fortunately, we can sum this Born series by inspection, getting

$$G_k = n_0 |V_k|^2 W(\epsilon_0 - W - \hbar\omega)^{-1} \tag{4.8}$$

or

$$\hbar\omega = \epsilon_k - n_0 |V_k|^2 (\epsilon_0 - W - \hbar\omega)^{-1} . \tag{4.9}$$

Solving this quadratic equation,

$$\begin{aligned}\hbar\omega = \hbar\omega_k \equiv {} & \tfrac{1}{2}(\epsilon_k + \epsilon_0 - W) \\ & + \tfrac{1}{2}\left[(\epsilon_k - \epsilon_0 + W)^2 + 4n_0 |V_k|^2\right]^{1/2} \\ = {} & \epsilon_k + n_0 |V_k|^2 \left(\frac{(\epsilon_k - \epsilon_0 + W_1) + iW_2}{(\epsilon_k - \epsilon_0 + W_1)^2 + W_2^2} \right) .\end{aligned} \tag{4.10}$$

In terms of the angle

$$\delta_k \equiv \arctan\left[W_2(\epsilon_k - \epsilon_0 + W_1)^{-1}\right] , \tag{4.11}$$

we have

$$\hbar\omega_k = \epsilon_k + n_0 |V_k|^2 \left[(\epsilon_k - \epsilon_0 + W_1)^2 + W_2^2\right]^{-1/2} e^{i\delta_k} . \tag{4.12}$$

The cross section for scattering of a quasiparticle of wave vector $\vec{k}$ by an impurity is

$$\sigma_k = (n_0 v_k \tau_k)^{-1} , \tag{4.13}$$

where τ_k is given by Eq. (2.6). We may approximate the quasiparticle velocity v_k by its value in the dilute limit, namely,

$$v_k = \hbar^{-1}\left(\frac{d\epsilon_k}{dk}\right) . \tag{4.14}$$

Combining Eqs. (2.6), (2.21), and (4.11)–(4.14), we get

$$\sigma_k = (4\pi/k^2)\sin^2\delta_k . \tag{4.15}$$

But, in general, we have

$$\sigma_k = \frac{4\pi}{k^2}\sum_{l=0}^{\infty}(2l+1)\sin^2\delta_{lk} , \tag{4.16}$$

where δ_{lk} is the phase shift of the lth partial wave associated with the quasiparticle of wave vector $\vec{k}$. Thus,

$$\sin^2\delta_k = \sum_{l=0}^{\infty}(2l+1)\sin^2\delta_{lk} . \tag{4.17}$$

It has been shown by Anderson and McMillan[9] that the Friedel sum rule[7]

$$Z = \frac{2}{\pi}\sum_{l=0}^{\infty}(2l+1)\,\delta_l \tag{4.18}$$

applies to the extra-orbital model of an impurity. Here δ_l is δ_{lk} evaluated on the Fermi surface, and Z is the excess number of conduction electrons introduced into the system by each impurity. If we make the approximation that only s-wave scattering is important (i.e., $\delta_{lk} = 0$ for $l > 0$), then Eq. (4.18) can be written

$$Z = (2/\pi)\arctan\left[W_{20}/(W_{10} - \epsilon_0)^{-1}\right] . \tag{4.19}$$

Here we are using the notation

$$W_0 = W_{10} - iW_{20} \tag{4.20}$$

for W evaluated at $\hbar\omega = 0$. Equation (4.19) expresses the condition for self-consistency in the choice of ϵ_0 and V_k in order that the excess charge Ze associated with an impurity be properly screened out by the surrounding conduction electrons. Equation (4.19) bears a close resemblance to the corresponding condition for the single-site model of an alloy.[10]

For Z even, Eq. (4.19) implies

$$W_{20} = |V_{k_F}| = 0 . \tag{4.21}$$

For Z odd, (4.19) implies

$$\epsilon_0 = W_{10} \equiv (2\pi)^{-3}\mathcal{P}\int d^3k\, |V_k|^2 \epsilon_k^{-1} , \tag{4.22}$$

which in turn implies that $\epsilon_0 > 0$ (since the conduction-band effective mass is positive). In contrast to the single-site model, there are nontrivial self-consistent choices of ϵ_0 and V_k for all values of Z.[11] The fact that Z and $Z+2$ have identical choices can be understood by the following argument.[12] In going from Z to $Z+2$, the s-wave phase shift δ_0 will increase by π. But whenever δ_0 crosses an integral multiple of π, a bound s level drops out of the bottom of the conduction band. This level is occupied by two opposite-spin electrons which serve to exactly screen out the increase in Z from the environment of the impurity.[13]

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[10] See Eq. (3) of Ref. 6.

[11] In fairness to the single-site model, it should be mentioned that the model's lack of a self-consistent choice of parameters for even Z appears to be an artifact of the assumption of only nearest-neighbor coupling between sites. If one assumes both nearest- and next-nearest-neighbor couplings, it appears that there should be self-consistent choices for even Z, although such choices might be difficult to determine.

[12] This argument is equally applicable to both the extra-

orbital and the single-site models of a dilute alloy.

[13]For a discussion of this point, see W. A. Harrison, *Solid State Theory* (McGraw-Hill, New York, 1970), p. 185.

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Spin-Wave Theory of Two-Magnon Raman Scattering in a Two-Dimensional Antiferromagnet*

S. R. Chinn, R. W. Davies, and H. J. Zeiger

Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173

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The graphical spin-wave approach to two-magnon Raman scattering which was developed in an earlier paper is applied to a study of the two-dimensional Heisenberg antiferromagnet K_2NiF_4. In the present treatment the one-magnon energies are taken to be renormalized in the Hartree-Fock approximation, with the two-magnon Green's function evaluated in the "ladder" approximation. It is found that the Hartree-Fock renormalization gives very good agreement with the experimental results for the temperature shift of the Raman peak up to the measured Néel temperature $T_N = 97.1\ ^\circ\mathrm{K}$, and yields satisfactory agreement above T_N up to the maximum temperatures for which Raman data are available. As in the three-dimensional case, a satisfactory explanation for the observed thermal broadening of the spectra remains to be given. Comparisons between the results for K_2NiF_4 and its three-dimensional analog $KNiF_3$ are made. In particular it is found that the renormalization of the zone-edge magnons for $T \simeq T_N$ is much less marked in the two-dimensional case. As has been discussed by other authors, this indicates a rather different temperature dependence of the "coherence length" for the two-dimensional system.

I. INTRODUCTION

In an earlier paper[1] (hereafter referred to as I), we presented a spin-wave approach to two-magnon Raman scattering and applied it to a study of simple three-dimensional antiferromagnetic systems (e.g., $KNiF_3$). The theory was based on the Dyson-Maleev boson representation of the spin operators, and proceeded through the application of the finite-temperature graphical perturbation theory. In this paper we present results of calculations based on the theory developed in I, but applied to the interesting case of a simple two-dimensional antiferromagnet. Probably the most widely studied example of such a system is the compound K_2NiF_4, which we shall consider in particular in this paper. At various points throughout the paper, we shall, for convenience, use the abbreviations [2] or [3] to stand for two- or three-dimensional systems.

Perhaps the most interesting theoretical question associated with the [2] systems involves the question of long-range ordering. Mermin and Wagner[2] have applied the Bogoliubov inequality to provide a rigorous proof that there can be no long-range order in a [2] system described by the Heisenberg exchange Hamiltonian for $T > 0$ in the absence of anisotropy. This instability is also suggested from simple spin-wave theory using a standard argument which we recall briefly here. Consider an isotropic [2] ferromagnet with a spin-wave branch having energy $\Omega_{\vec{k}} \propto k^2$ for $\vec{k} \to 0$. Calculation of the magnetization involves an integral of the form

$$\int k\,dk \frac{1}{e^{\Omega_{\vec{k}}/k_B T} - 1} ,$$

which diverges logarithmically in the neighborhood of $\vec{k} \to 0$. Next consider the case of an isotropic [2] antiferromagnet with a spin-wave branch having $\Omega_{\vec{k}} \propto k$ as $\vec{k} \to 0$. Here calculation of the sublattice magnetization involves an integral

$$\int \frac{k\,dk}{\Omega_k} \frac{1}{e^{\Omega_{\vec{k}}/k_B T} - 1} ,$$

where the extra factor of Ω_k in the denominator arises from the Bogoliubov transformation which diagonalizes the spin-wave Hamiltonian. Again one obtains a logarithmic divergence from the region $\vec{k} \to 0$. These considerations show that the question of long-range order in [2] depends sensitively on the behavior of the long-wavelength magnons. Also, since a small amount of anisotropy is sufficient to remove the singularity at $\vec{k} \to 0$, the ordering problem (at least in the simple spin-wave approximation[3]) involves a careful treatment of anisotropy effects.

In contrast to the discussion above, we now consider the problem of two-magnon Raman scattering in a [2] antiferromagnet. Here the cross section is determined almost exclusively by the behavior of the short-wavelength (zone-edge) magnons, which should be very insensitive to small effects due to anisotropy. Also one expects the zone-edge magnons to reflect the properties of short-range order in the system, and this can persist well above the