Mixing of states of conduction electrons and impurity *d*(*f*) ions

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A model of a metal with $d(f)$ ions as impurities is considered. The resonant scattering of the conduction electrons causes a restructuring of the electron spectrum to produce two quasiparticle bands separated by a gap. At a low impurity concentration, $c \ll 1$, the quasiparticles have an attenuation Γ, whereas the gap is proportional to Γc and hence becomes smeared out while the state density $g(E)$ has a peak, just as in Anderson's single-impurity model. In the other extreme case $1-c \ll 1$ the quasiparticle attenuation is proportional to Γc and the gap to Γ, so that $g(E)$ has a dip on whose slopes the state density is higher. The limiting transition to the regular case $c = 1$ is traced.

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1. INTRODUCTION

The interaction of the conduction electrons of a metal with magnetoactive $d(f)$ impurity ions has been repeatedly discussed in the literature in connection with the problem of the localized moment,[1] the Kondo effect, and others. In the description of this interaction account must be taken of the intra-atomic correlations of the electrons, which play the decisive role in the case of transition $d(f)$ metal impurities. Let, for example, ε_{n+1} and ε_n be respectively the energies of the atomic configurations d^{n+1} and d^n, where n is the number of electrons in the ground-state configuration of the ion. If the transition energy $\Omega = \varepsilon_{n+1} - \varepsilon_n$ turns out to be comparable with the energy of the conduction electron (c electron), i.e., $\Omega \sim \varepsilon_k$, competition sets in between two possible electron configurations in the metal, cd^n and d^{n+1}. The degeneracy connected with the mixing (hybridization) of the cd^n and d^{n+1} states is lifted by the interaction of the c electrons with the d ions, which can be regarded as a reaction channel via a "compound-ion" stage, $s+d^n \quad d^{n+1}$. This channel is due to the multiple absorption and emission of the conduction electron.

The single-impurity problem in the presence of a single-particle c–d mixing mechanism was solved by Anderson.[1] Using the Hartree–Fock approximation for the description of the d-electron states, he showed that the mixing leads to a smearing of the localized d level. The d-electron state density becomes equal to

$$g_d(E)=\frac{1}{\pi}\frac{\Gamma}{(E-\Omega)^2+\Gamma^2}, \quad \Gamma=\pi A^2 g_0, \tag{1}$$

where $\Omega = \varepsilon_d$ is the energy of the localized level, A is the c–d mixing interaction constant, and g_0 is the conduction-electron state density. We note that in Anderson's model the c-electron spectrum changes little. A similar broadening of the d level is obtained also when account is taken of the correlations.[2]

In the case of a transition or rare-earth metal, when the $d(f)$ ions form a crystal lattice and the atomic level (or the transition energy Ω) is inside the conduction band, the situation is entirely different. Because of the regular arrangement of the $d(f)$ ions, the mixing is coherent and leads to a substantial restructuring of the entire electron spectrum, wherein the localized level gives way to an energy gap that separates two new quasiparticle bands with dispersion laws[3,4]

$$E^{\pm}(k)={}^1/_2(\varepsilon_k+\Omega\pm\nu_k), \quad \nu_k^2=(\varepsilon_k-\Omega)^2+4A_{\rm eff}^2, \tag{2}$$

where $A_{\rm eff}$ is the effective mixing constant and $E_{\rm min} - E_{\rm max} \sim 2A^2\Omega$ is the energy gap. The state density is zero inside the gap and maximal near its edges—a picture quite the opposite of the one-impurity case.

It must be emphasized that in both cases the Hamiltonians of the system have identical operator structures. The difference between the results is the consequence of the difference in the dependence of the matrix element $A_f(k)$ of the mixing interaction on the wave vector k; this dependence takes the form $A_f(k) = A(k)e^{-ikf}$,[5] where f is the number of the site occupied by the d-ion. At low impurity concentration, when each impurity is independent, we can put $f = 0$, and then $A_f(k) = A(k)$ is a smooth function of k; in the regular case, however, the exponential factor is significant and plays the role of the coherence factor. Therefore the results obtained in the framework of the single-impurity model cannot be used to describe regular $d(f)$metals, as was done, e.g., in[6].

The purpose of the present paper is to consider the mixing of the electron states at a finite impurity concentration c. In two cases, $c \ll 1$ and $1 - c \ll 1$, we obtain the single-particle Green's functions and the densities of the electronic states. We trace the limiting transition both to the Anderson model and to the case of a regular crystal.

2. HAMILTONIAN AND GREEN'S FUNCTION

We consider a system of band and impurity atomic electrons described by the Hamiltonian

$$\begin{gathered}\mathscr{H}=\mathscr{H}_0+\mathscr{H}_Q+\mathscr{H}_{\rm Coul}+\mathscr{H}_{\rm exch},\\ \mathscr{H}_0=\sum_{k\sigma}\xi_k a_{k\sigma}{}^+a_{k\sigma}+\omega\sum_f p_f X_f{}^{22},\\ \mathscr{H}_Q=\sum_{fk\sigma}p_f\left[\frac{Q}{N^{1/2}}e^{-ifk}\eta(\sigma)a_{k\sigma}{}^+X_f{}^{-\sigma 2}+\frac{Q^*}{N^{1/2}}e^{ifk}\eta(\sigma)X_f{}^{2,-\sigma}a_{k\sigma}\right],\end{gathered} \tag{3}$$

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$$\mathscr{H}_{\text{Coul}}=\left(V-\frac{1}{2}J\right)\frac{1}{N}\sum_{fkk'\sigma}p_f e^{-if(k-k')}(1+X_f^{22})a_{k\sigma}^{+}a_{k'\sigma},$$

$$\mathscr{H}_{\text{exch}}=-\frac{J}{N}\sum_{\substack{fkk'\\ \sigma\sigma'}}p_f e^{-if(k-k')}2\hat{\tau}_{\sigma\sigma'}\hat{S}_f a_{k\sigma}^{+}a_{k'\sigma'}.$$

Here $a_{k\sigma}^{+}(a_{k\sigma})$ are the operators of creation (annihilation) of c electrons with quasimomentum k, spin projection σ, and energy ξ_k reckoned from the Fermi energy μ, and $X^{pq}\equiv|p\rangle\langle q|$ are Hubbard operators, where $|p\rangle$ and $|q\rangle$ are atomic states: $|2\rangle$—configuration d^{n+1} with spin $S=0$, $|\sigma\rangle$—configuration d^n with spin $\frac{1}{2}$ and its projection σ; $\eta(\sigma)=\pm 1$ at $\sigma=\pm\frac{1}{2}$; $p_f=1$ if f is a site with an impurity atom and $p_f=0$ in the opposite case. The constant Q of the "direct" mixing of the states is governed by the crystal field, so that in transition and rare-earth metals Q is much smaller than the constants V and J of the intra-atomic Coulomb and exchange c–d interactions.[7] The Hamiltonian $\mathscr{H}_0$ describes the system of non-interacting c and d electrons; $\omega=\Omega-\mu$

The equations for the Green's functions

$$G_{kk'}^{\sigma}(E)=\langle\langle a_{k\sigma}|a_{k'\sigma}^{+}\rangle\rangle_E,\quad F_{fk'}^{\sigma}(E)=\langle\langle\eta(\sigma)X_f^{-\sigma 2}|a_{k'\sigma}^{+}\rangle\rangle_E$$

are obtained in the generalized Hartree–Fock approximation[8] and are of the form

$$\begin{aligned}&(E-\xi_k)G_{kk'}^{\sigma}(E)-\sum_f A_{f\sigma}(k)p_f F_{fk'}^{\sigma}(E)=\delta_{kk'},\\ &(E-\omega)F_{fk'}^{\sigma}(E)-\sum_p n_f p_f A_{f\sigma}^{*}(p)G_{pk'}^{\sigma}(E)=0.\end{aligned}\tag{4a}$$

Here

$$A_{f\sigma}(p)=\frac{1}{N^{1/2}}e^{-ifp}A_{f\sigma},\quad A_{f\sigma}=\frac{Q-(V-J)\gamma_{f\sigma}}{n_{f\sigma}}-\frac{J\gamma_{f,-\sigma}}{n_{f,\sigma}},$$

where

$$\gamma_{f\sigma}=\frac{1}{N^{1/2}}\sum_p\langle X_f^{2,-\sigma}\eta(\sigma)a_{p\sigma}\rangle.$$

In the paramagnetic state, which will be considered from now on, we have

$$n_{f\sigma}\equiv 1-\langle X_f^{-\sigma,-\sigma}\rangle=n_{f,-\sigma}\equiv n_f,\quad A_{f\sigma}=A_{f,-\sigma}\equiv A_f.$$

The angle brackets denote Gibbs averaging with the Hamiltonian (3). Similar equations connect the functions

$$F_{kf'}^{+\sigma}(E)=\langle\langle a_{k\sigma}|\eta(\sigma)X_{f'}^{2,-\sigma}\rangle\rangle_E,\quad D_{ff'}^{\sigma}(E)=\langle\langle\eta(\sigma)X_f^{-\sigma 2}|\eta(\sigma)X_{f'}^{2,-\sigma}\rangle\rangle_E,$$

namely:

$$\begin{aligned}&(E-\omega)D_{ff'}^{\sigma}(E)-\sum_p n_f A_f^{*}(p)F_{pf'}^{+\sigma}(E)=n_f\delta_{ff'},\\ &(E-\xi_p)F_{pf'}^{+\sigma}(E)-\sum_m A_m(p)p_m D_{mf'}^{\sigma}(E)=0.\end{aligned}\tag{4b}$$

In the derivation of these equations we have neglected the correction that must be introduced in the transition energy Ω because of the direct mixing Q[9]:

$$Q^2 g_0\ln\frac{\Omega}{Q}<Q\ll\Omega\sim\mu,$$

and also the Hartree-Fock corrections to the electron energy ξ_k.

From the system (4) we readily obtain integral equations for the functions $G_{kk'}(E)$ and

$$D_{kk'}(E)=N^{-1}\sum_{ff'}p_f p_{f'}\exp(-ifk+if'k')D_{ff'}(E),$$

i.e.,

$$G_{kk'}(E)=G_k^0(E)\delta_{kk'}+G_k^0(E)D^0(E)\frac{1}{N}\sum_{fp}p_f n_f|A_f|^2 e^{-if(k-p)}G_{pk'}(E),\tag{5a}$$

$$\begin{aligned}D_{kk'}(E)=&D^0(E)\frac{1}{N}\sum_f p_f n_f e^{-if(k-k')}\\ &+D^0(E)\frac{1}{N}\sum_{pf}p_f n_f|A_f|^2 e^{-if(k-p)}G_p^0(E)D_{pk'}(E),\end{aligned}\tag{5b}$$

where

$$D^0(E)=(E-\omega)^{-1},\quad G_k^0(E)=(E-\xi_k)^{-1}.$$

Equations (5) are the basis for the study of two limiting cases: $c\ll 1$, $1-c\ll 1$, $c=(1/N)\sum_f p_f$, where c is the impurity concentration; we proceed now to investigate these cases.

3. LOW IMPURITY CONCENTRATION

To average the Green's functions over the positions of the impurities we use a diagram technique.[10] At $c\ll 1$ the average distance between the impurity atoms is much larger than the lattice period, so that the averaging can be carried out in volumes having dimensions much larger than a. If the impurity distribution is uniform, this averaging restores the translational invariance:

$$\overline{G_{kk'}(E)}=G_k(E)\delta_{kk'}.$$

Solving Eqs. (5a) and (5b) by iteration, we obtain G and D as sums of infinite series in which each term is diagrammed in accordance with the following rules[10]: the solid line is $G_k^0(E)$, the cross is the bare interaction

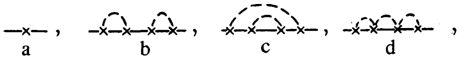
$$v_f(k-p,E)=|A_f|^2 n_f D^0(E)\exp[-if(k-p)]\equiv v_f^0(E)\exp[-if(k-p)],$$

the dashed line joins identical points, and the averaging rule is

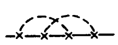
$$\overline{\frac{1}{N}\sum_f Z_f p_f e^{-if(k-k')}}=cZ\delta_{kk'},$$

where, in view of the identity of the impurity atoms, all the single-point characteristics Z_f are assumed to be independent of f.

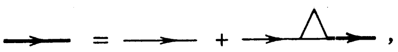
In the approximation linear in the impurity concentration it is necessary to retain the following diagrams:

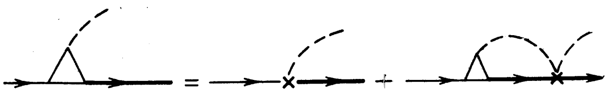


and to discard diagrams of the type



In contrast to the problem of potential scattering by impurities,[10] we retain diagrams of type a and d, inasmuch as the effective interaction $v_f^0(E)$ contains the resonant factor $D^0(E)$. We note also the summation of diagrams of type a only leads to the appearance of the spectrum of the type (2).[11] As a result the total Green's function $\overline{G_{kk'}}$ is obtained from the following system of equations:


(6)


(7)

Solution of Eq. (7) yields the effective interaction

$$v_f(k-p)=v_fe^{-if(k-p)},\quad v_f=|A_f|^2n_f/(E-\omega_f),\quad \omega_f=\omega+|A_f|^2n_f\langle G(E)\rangle. \tag{8}$$

Substituting (8) in (6) we obtain

$$G_k(E)=[E-\xi_k-v^0c-v^0cv\langle G(E)\rangle]^{-1}, \tag{9}$$

v^0 and v are the values of v_f^0 and v_f averaged over the impurity positions, and $\langle G(E)\rangle$ is obtained from the equation

$$\langle G(E)\rangle=\frac{1}{N}\sum_p G_p(E)=g_0\int_{-W}^{W}\frac{d\xi}{E-\xi-v^0c-v^0cv\langle G(E)\rangle}. \tag{10}$$

The integral in (10) can be calculated and a transcendental equation is obtained for $\langle G(E)\rangle$; a numerical solution of this equation shows that $\mathrm{Im}\langle G\rangle\approx-\pi g_0$ and $\mathrm{Re}\langle G\rangle/\mathrm{Im}\langle G\rangle\sim10^{-2}$, we therefore neglect $\mathrm{Re}\langle G\rangle$ (just as both in Anderson's solution[11]) and in the case of potential scattering by the impurity[10]), and then $\langle G(E)\rangle\approx-i\pi g_0$. Thus

$$G_k(E)=\frac{E-\omega+i\Gamma}{(E-\xi_k)(E-\omega+i\Gamma)-\Delta^2c}, \tag{11}$$
$$\Delta^2=|A|^2n,\quad \Gamma=\pi\Delta^2g_0.$$

From (5b) we obtain similarly

$$D_k(E)=\frac{c(E-\xi_k)n}{(E-\xi_k)(E-\omega+i\Gamma)-\Delta^2c}. \tag{12}$$

The Green's functions (11) and (12) differ from the regular case[8] in that the level acquires a width Γ, so that the quasiparticles (2) are attenuated and their spectrum is of the form

$$\tilde{E}^{\pm}(k)=E^{\pm}(k)\pm i\Gamma(E^{\mp}-\omega)/v_k,$$

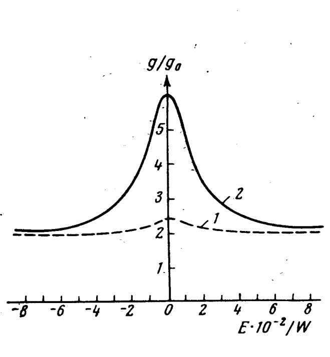


FIG. 1. Density of states at low impurity concentration; 1) $c=0.01$; 2) $c=0.1$ for $\Delta=0.1\ W$.

where $E^{\pm}(k)$ is defined in (2). On the edges of the gap between the subband E^+ and E^- the attenuation equals Γ and is independent of c, whereas the gap is $E_g\sim\Delta^2g_0c\sim\Gamma c\ll\Gamma$, so that attenuation leads to complete vanishing of the gap (see Fig. 1). For this reason, our previous analysis of this question,[11] where we neglected the attenuation of the quasiparticles, is incorrect.

Using expressions (11) and (12), we obtain the state density

$$g(E)=-\frac{1}{\pi}\mathrm{Sp}(\mathrm{Im}(G_k(E)+D_k(E)))=g_s(E)+g_d(E);$$

here

$$g_s(E)=\frac{2}{\pi}g_0\left\{\mathrm{arctg}\frac{(W-E)[(E-\omega)^2+\Gamma^2]+\Delta^2(E-\omega)}{\Delta^2\Gamma}\right.$$
$$\left.+\,\mathrm{arctg}\frac{(W+E)[(E-\omega)^2+\Gamma^2]-\Delta^2(E-\omega)}{\Delta^2\Gamma}\right\},$$
$$g_d(E)=\frac{c\Gamma}{\pi[(E-\omega)^2+\Gamma^2]}+$$
$$+\frac{c\Gamma}{\pi}g_0\left\{-\frac{\Delta^2(E-\omega)}{[(E-\omega)^2+\Gamma^2]^2}\ln\frac{[(W-E)(E-\omega)+\Delta^2]^2+\Gamma^2(W-E)^2}{[(W+E)(E-\omega)-\Delta^2]^2+\Gamma^2(W+E)^2}\right.$$
$$+\frac{\Delta^2}{\Gamma}\frac{[(E-\omega)^2-\Gamma^2]}{[(E-\omega)^2+\Gamma^2]^2}\left[\mathrm{arctg}\frac{(W-E)[(E-\omega)^2+\Gamma^2]+\Delta^2(E-\omega)}{\Delta^2\Gamma}\right.$$
$$\left.\left.+\,\mathrm{arctg}\frac{(W+E)[(E-\omega)^2+\Gamma^2]-\Delta^2(E-\omega)}{\Delta^2\Gamma}\right]\right\}.$$

A plot of $g(E)$ is shown in Fig. 1.

4. LOW HOLE DENSITY

In the case $1-c\ll1$, it is more convenient to change to the hole representation with the aid of the projection operator $h_f=1-p_f$, and then $c_h=1-c\ll1$ and we can average over the hole positions. In contrast to the preceding section, the null functions are here the Green's functions of the regular crystal with mixing. Indeed, Eqs. (5) can be rewritten in the form

$$G_{kk'}(E)=\delta_{kk'}\tilde{G}_k^0(E)-\tilde{G}_k^0(E)D^0(E)\frac{1}{N}\sum_{fp}|A_f|^2n_fh_fe^{-if(k-p)}G_{pk'}(E),$$
$$D_{kk'}(E)=\tilde{D}_k^0(E)\left[\delta_{kk'}-\frac{1}{N}\sum_f h_fe^{-if(k-k')}\right] \tag{13}$$
$$-\tilde{D}_k^0(E)\frac{1}{N}\sum_{fp}|A_f|^2n_fh_f\tilde{G}_p^0(E)e^{-if(k-p)}D_{pk'}(E),$$

where

$$\tilde{G}_k^0(E)=\frac{E-\omega}{(E-\xi_k)(E-\omega)-\Delta^2},\quad \tilde{D}_k^0(E)=\frac{(E-\xi_k)n}{(E-\xi_k)(E-\omega)-\Delta^2}$$

are the Green's functions of the regular problem.[8] These equations are averaged in analogy with Eqs. (5), and as a result we obtain

$$\overline{G_{kk'}(E)}=\frac{(E-\omega)\delta_{kk'}}{(E-\xi_k)(E-\omega)-\Delta^2(1-c_h)+i\Gamma\Delta^2c_h/(E-\omega-i\Gamma)}$$
$$\overline{D_{kk'}(E)}=\frac{(E-\xi_k)n\delta_{kk'}}{(E-\xi_k)(E-\omega)-\Delta^2(1-c_h)+i\Gamma\Delta^2c_h/(E-\omega-i\Gamma)} \tag{14}$$

In this case the gap in the spectrum is $E_g\sim\Delta^2g_0(1-c_h)\sim\Gamma\gg\Gamma c_h$, i.e., much larger than the quasiparticle attenuation.

From (14) we obtain the state density

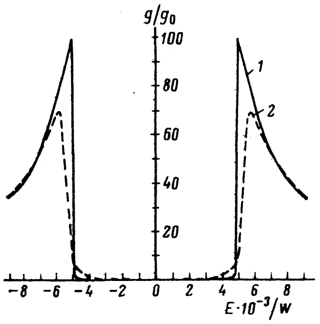


FIG. 2. State density at low hole concentration: 1) $c=0.001$; 2) $c=0.1$ for $\Delta=0.1\,W$.

$$g(E)=g_s(E)+g_d(E),$$

$$g_s(E)=\frac{2}{\pi}g_0\times$$

$$\times\left\{\operatorname{arctg}\frac{(W-E)[(E-\omega)^2+\Gamma^2]+\Delta^2[(E-\omega)(1-c_h)+\Gamma^2/(E-\omega)]}{\Gamma\Delta^2c_h}\right.$$

$$\left.+\operatorname{arctg}\frac{(W+E)[(E-\omega)^2+\Gamma^2]-\Delta^2[(E-\omega)(1-c_h)+\Gamma^2/(E-\omega)]}{\Gamma\Delta^2c_h}\right\},$$

$$g_d(E)=\frac{2}{\pi}g_0\frac{1}{(E-\omega)^2}$$

$$\times\left\{\frac{1}{2}b_1^2(E)\ln\frac{[(E-\omega)(E+W)-b^2(E)]^2+b_1^4(E)}{[(E-\omega)(E-W)-b^2(E)]^2+b_1^4(E)}\right.$$

$$\left.+b^2(E)\sum_{n=1}^{2}(-1)^n\operatorname{arctg}\frac{(E-\omega)[E+(-1)^nW]-b^2(E)}{b_1^2(E)}\right\},$$

where

$$b^2(E)=\Delta^2\left[1-c_h+\frac{\Gamma^2c_h}{(E-\omega)^2+\Gamma^2}\right]$$

$$b_1^2(E)=\frac{\Gamma c_h\Delta^2(E-\omega)}{(E-\omega)^2+\Gamma^2}.$$

A plot of $g(E)$ is shown in Fig. 2, from which it is seen that, in accordance with the general theorems on the spectrum of a disordered system,[12] the gap gives way to a pseudogap, and as the hole density increases the number of states inside the gap increases.

CONCLUSION

The difference between the state densities in the single-impurity ($c\ll 1$) and almost-regular ($1-c\ll 1$) states demonstrates once more the importance of the coherence factor in the investigation of impurity systems with c–d mixing

The single-particle Green's functions obtained in this paper, besides providing information on the state density, can be used to analyze the properties of spin glasses. Thus, for example, by the method of irreducible Green's functions[13] we can obtain from the correlation part of $\langle\mathscr{H}_{\text{exch}}\rangle$ also the effective spin Hamiltonian,[14] which in our case takes the form

$$\tilde{\mathscr{H}}=-\sum_{mf}I(m-f)p_m\mathbf{S}_mp_f\mathbf{S}_f,$$

where, owing to c–d mixing, $\mathbf{S}_m$ is the effective-spin Hamiltonian whose magnitude within the framework of the considered model is $S_{\text{eff}}=S(1-\langle X^{22}\rangle)$, and the exchange-interaction integral is

$$I(m-f)$$
$$=-\frac{J^2}{N^2}\sum_{kq}e^{-iq(m-f)}\int\frac{dE}{e^{E/T}+1}\left(-\frac{1}{\pi}\right)\operatorname{Im}[\bar{G}_{k+q}(E+iO)\bar{G}_k(E+iO)]. \tag{15}$$

Expression (15) is a generalization of a number of particular cases: in a regular metal, quasiparticles with spectrum (2) produce indirect exchange between the effective spins, and the result is magnetic ordering; in the case of "nonfluctuating" spin, when the atomic level $|2\rangle$ is absent or $\Omega\gg\mu$ (s–d model), (15) goes over into the known Ruderman–Kittel integral. A more detailed investigation of the magnetic properties of the considered model will be reported in another paper.

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