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INTERACTION OF ANDERSON IMPURITIES WITH HIGH ORBITAL ANGULAR MOMENTA : NON-RKKY BEHAVIOR AND INSTABILITY OF KONDO LATTICE

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Hybridization-induced interaction of Anderson impurities with orbital angular momentum l is revisited. At short distances $R < R_c \propto (l+1)/k_F$ the interaction has antiferromagnetic sign and decays as $(R_c/R)^{4l}$. At larger distances $R > R_c$ the RKKY-like oscillating interaction sets in. If l increases, then, sooner or later, the system occurs in the "short distance" domain, where the intersite magnetic interaction dominates over the screening processes. It means that, contrary to previous expectations, the nonmagnetic state of Anderson lattice is unstable at $l \to \infty$

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A permanent interest of theorists to the Anderson lattice model [1] is due to a non-trivial nonmagnetic ground state (Kondo lattice), which is expected to occur it this model under certain conditions (see reviews [2, 3]). Initially such a nonmagnetic state was viewed as a simple collection of basically independent Kondo ions, but it was quickly understood that the situation is not that simple. At any reasonable concentration of magnetic ions the Kondo clouds strongly overlap, so that the nonmagnetic state (if any) could only be a result of some sophisticated collective screening effect. A severe limitation to the non-magnetic scenario [4] is caused by the conduction-electrons-mediated magnetic interaction of Anderson ions [5, 6, 7, 8], tending to form a magnetically ordered state. The energy of magnetic interaction E_{mag} is proportional to the fourth order of the hybridization matrix element V, while the "Kondo energy" E_K, characteristic for screening processes, is exponentially small in $|V|^{-2}$. It means that a controlled theoretical analysis of the nonmagnetic state can only make sense if there is an additional parameter in the model, which can help to overcome the tendency to the magnetic order formation. It is widely believed now that the degree of "orbital" degeneracy N of Anderson ion may be such a parameter. It was first argued by Coleman [9] and Read, Newns, and Doniach [10] that the screening processes can dominate over the intersite interaction for the Anderson ions with high $N \gg 1$. The nonmagnetic state itself was extensively studied both for so called $SU(N)$ Anderson lattice model [11, 2] with unspecified external origin for the degeneracy, and for more realistic model with genuine orbital degeneracy, related to the orbital angular momentum l of a magnetic ion [12, 3]. For the former model the energy E_{mag} of the competing magnetic state is easy to find, and the criterion of the Kondo lattice stability can be easily checked: it is indeed fulfilled at large N. For the latter model only tentative estimates of E_{mag} were found (see [12, 3, 8]); they seemed to be also in favor of nonmagnetic state. In the papers [12, 8], however, only the angular dependence of matrix

elements was taken into account, while, as we will see in what follows, the dependence on $|\mathbf{k}|$ is of crucial importance for high angular momenta l.

The goal of this paper is (i) to study the dependence of effective magnetic interaction of Anderson ions on distance between ions, and (ii) to reconsider the question about stability of nonmagnetic state of Anderson lattice at high degeneracy N.

The mechanism for indirect interaction of magnetic atoms, originating from the hybridization of localized and delocalized electrons was proposed in the pioneering paper [5] by Coqblin and Schrieffer (see also [6]). This interaction is presumably essential (i.e. it can dominate over the conventional exchange-induced RKKY interaction [13]) in the resonant case, when the localized level ϵ_0 is situated only slightly below the Fermi level ϵ_F, which is supposedly the case for the metals with considerable valence fluctuations (e.g. Cerium). The original derivation [5, 6] of the interaction, based on the repeated application of the second order Schrieffer-Wolff transformation [14], was, however, incomplete. A straightforward fourth order Schrieffer-Wolff transformation (see [7, 8]), besides the RKKY-like (though highly anisotropic) Coqblin-Schrieffer interaction, gives rise to additional important term, reminiscent of usual superexchange [15]. This term alters the interaction quite dramatically, especially at short distances, where it dominates and changes the sign of the interaction to the antiferromagnetic one.

In this Letter we restrict our consideration to the case of one electron in the magnetic shell (like one f-electron for Cerium ions) and consider only the lower spin-orbital multiplet, characterized by the total angular momentum $J = l - 1/2$ or $l + 1/2$, the degeneracy of the localized state being $N = 2J + 1$. We describe a system of two magnetic ions (a and b) by the Anderson Hamiltonian $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{hyb}$, where

$$\mathcal{H}_0 = \sum_{\mathbf{k}\sigma} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \epsilon_0 \sum_{M,i=a,b} n_{iM} + \frac{U}{2} \sum_{i,M\neq M'} n_{iM} n_{iM'},$$

U is the energy of the Hubbard repulsion (we set $U = +\infty$ for simplicity); $n_{iM} = f^\dagger_{iM} f_{iM}$, $f^\dagger_{iM}$ creates an electron with $J_z = M$ in the unclosed shell of the ith magnetic ion; $c^\dagger_{\mathbf{k}\sigma}$ creates a conduction electron with momentum $\mathbf{k}$ and spin projection $\sigma = \pm 1/2$. The hybridization Hamiltonian is

$$\mathcal{H}_{hyb} = \sum_{i\mathbf{k}M\sigma} e^{i\mathbf{k}R_i} V_M(\mathbf{k}\sigma) c^\dagger_{\mathbf{k}\sigma} f_{iM} + \text{h.c.},$$

The initial Hamiltonian $\mathcal{H}$ can be reduced to an effective interaction $\hat{H}$ of magnetic moments, by means of a fourth order Schrieffer-Wolff transformation (see [8]). Consider degenerate groundstates of the unperturbed Hamiltonian $\mathcal{H}_0$, characterized by quantum numbers $\nu \equiv \{M_a, M_b\}$. Then, specifying all possible intermediate states $|i\rangle$, we obtain composite fourth order matrix elements between states $|\nu\rangle$ and $|\nu'\rangle$:

$$\hat{H} = \sum_{i_1 i_2 i_3 \neq \nu,\nu'} \frac{\langle\nu'|\mathcal{H}_{hyb}|i_3\rangle\langle i_3|\mathcal{H}_{hyb}|i_2\rangle\langle i_2|\mathcal{H}_{hyb}|i_1\rangle\langle i_1|\mathcal{H}_{hyb}|\nu\rangle}{(E_\nu - E_{i_3})(E_\nu - E_{i_2})(E_\nu - E_{i_1})} =$$

$$= -\sum_{\mathbf{k}\mathbf{k}'\sigma\sigma'} \hat{\Phi} \frac{\theta(\epsilon_{\mathbf{k}} - \epsilon_F)}{(\epsilon_{\mathbf{k}} - \epsilon_{\mathbf{k}'})(\epsilon_{\mathbf{k}} - \epsilon_0)^2}, \qquad (1)$$

$$\Phi^{M'_a M'_b}_{M_a M_b} \equiv \cos\{(\mathbf{k}-\mathbf{k}')\mathbf{R}\}\left\{V_{M'_a}(\mathbf{k}'\sigma')V^*_{M_a}(\mathbf{k}\sigma)V_{M'_b}(\mathbf{k}\sigma)V^*_{M_b}(\mathbf{k}'\sigma') + (a \leftrightarrow b)\right\}.$$

Note that expression (1) comprises both usual RKKY-like indirect exchange processes with electron-hole excitation in the intermediate state ($k > k_F, k' < k_F$), and the superexchange-like processes with two-electron excitations in the intermediate state ($k, k' > k_F$).

In order to proceed with calculation of matrix elements $\hat{H}$, described by general formula (1), we adopt the "free electron model" (see [3]), in which the conduction electrons are described by the plane waves and $\epsilon_{\mathbf{k}} = k^2/2m$. Having in mind an application to rare earths, we assume that the spatial size of the localized state r_0 is small (see [16]): much less than both R and k_F^{-1}, so that only the contributions of leading order in kr_0 should be kept. Then the matrix elements of the hybridization Hamiltonian are

$$V_M(\mathbf{k}\sigma) = C^{lJ}_{M\sigma}\int d^3\mathbf{r}\psi^*_l(r)Y^*_{lM-\sigma}(\Omega_{\mathbf{r}})V(r)e^{i\mathbf{k}r},$$

$$C^{lJ}_{M\sigma} = \frac{1}{\sqrt{2}}\left(1+\frac{4(J-l)\sigma M}{l+1/2}\right)^{1/2},$$

where $V(r)$ is the hybridization potential, which is spherically symmetric at relevant small distances $r \sim r_0$, $\psi_l(r)$ is the radial part of the localized wave function. Performing the angular integration, we get at $kr_0 \ll 1$

$$V_M(\mathbf{k},\sigma) = C^{lJ}_{M\sigma}(k/k_F)^l V_{k_F}\sqrt{4\pi}Y^*_{lM-\sigma}(\Omega_k), \tag{2}$$

where $V_{k_F} \propto (k_F r_0)^l$ is a constant. The factor $(kr_0)^l$, very important in the case of high l, arises due to tunneling under the centrifugal barrier. Choosing the quantization axis parallel to $\mathbf{R}$, we can now rewrite (1) in a form

$$\hat{H} = -\frac{2}{\pi}\hat{P}I_0\int_{k_F}^{\infty} g_{JM_a}(kR)\frac{(k/k_F)^{2l}k^2dk}{(k^2-2m\epsilon_0)^2}\times$$

$$\times\mathcal{P}\int_0^{\infty} g_{JM_b}(k'R)\frac{(k'/k_F)^{2l}k'^2dk'}{(k^2-k'^2)} + (a \leftrightarrow b). \tag{3}$$

where $I_0 = |V_{k_F}|^4(m/\pi)^3$, symbol $\mathcal{P}$ means the principal value of the integral, and $\hat{P} \equiv \delta_{M'_a M_b}\delta_{M'_b M_a}$ is the "exchange operator". The real function

$$g_{JM}(x) = \sum_{\sigma}\left(C^{lJ}_{M\sigma}\right)^2\int d\Omega|Y_{lM-\sigma}(\Omega)|^2\cos(x\cos\theta),$$

depends only on J and $|M|$, not on l, and not on the sign of M. It is convenient to represent $g_{JM}(x)$ as a real part of a complex function $\tilde{g}_{JM}(x)$ with appropriate analytical behavior

$$\tilde{g}_{JM}(x) = -\frac{(J-|M|)!}{(J+|M|)!}\int_1^{\infty}\left\{(J+|M|)^2|P^{|M|-\frac{1}{2}}_{J-\frac{1}{2}}(t)|^2 + |P^{|M|+\frac{1}{2}}_{J-\frac{1}{2}}(t)|^2\right\}dte^{it(x+i0)},$$

where P^m_n are associated Legendre polynomials. For the nondegenerate Anderson model ($J = 1/2$) one gets $\tilde{g}_{\frac{1}{2}\frac{1}{2}}(x) = e^{ix}/ix$. In general, $\tilde{g}_{JM}(x) = e^{ix}Q_{JM}(1/x)$, where Q_{JM}

are polynomials of power $2J$. Their explicit form for not very high J (e.g., for $J = 5/2$ in case of Cerium) can be easily found using a program of analytical calculations. The study of properties of $\tilde{g}$ for general J, M is quite involved mathematical exercise; here we give only few asymptotics without derivation:

$$\tilde{g}_{JM}(x) \approx \frac{B_{JM} e^{ix}}{(ix)^{|M|+1/2}}, \quad B_{JM} = \frac{(J+|M|)!}{2^{|M|-1/2}(|M|-1/2)!(J-|M|)!}, \quad \text{for } x \gg J, \tag{4}$$

$$\tilde{g}_{JM}(x) \approx J_0\left[x\sqrt{1-(M/J)^2}\right] - i\frac{A_{JM}}{x^{2J}} \exp\left(\frac{D_{JM}x^2}{8J}\right), \tag{5}$$

for $x \ll J$, where J_0 is a standard Bessel function, and

$$A_{JM} = \frac{(-1)^{J+M} M(2J)![(2J)!!]^2}{2J^2(J-M)!(J+M)!}, \; D_{JM} = \frac{J(J-2)+M^2}{(J-1)^2}.$$

Comparison of asymptotics (4) and (5) shows that they match at $x \sim J$ for all values of M and J.

Since $\tilde{g}_{JM}(x)$ is an analytic function in upper half-plane, one can perform the integration over k' in Eq(3) by residues; introducing dimensionless variable $z = kR$ we arrive at

$$\hat{H} = -\frac{\hat{P} I_0}{(k_F R)^{4l}} \mathrm{Im} \int_{k_F R}^{\infty} \frac{z^{4l+3} dz}{(z^2 - 2m\epsilon_0 R^2)^2} \tilde{g}_{JM_a}(z) \tilde{g}_{JM_b}(z). \tag{6}$$

We start a discussion from the case of nondegenerate Anderson model: $l = 0, J = 1/2$, where the interaction is isotropic: $\hat{H} = \left(\mathbf{J}^{(a)} \cdot \mathbf{J}^{(b)}\right) I(R)$. In the nonresonant case (when ϵ_0 is not especially close to ϵ_F) there is only one spatial scale $R_c \sim \pi/k_F$, and the "exchange constant" $I(R)$ has the following asymptotics: For $R \ll R_c$

$$I(R) = \frac{I_0}{k_F R}\left\{\frac{1}{\sqrt{\varepsilon}} \ln\frac{1+\sqrt{\varepsilon}}{1-\sqrt{\varepsilon}} + \frac{2}{1-\varepsilon}\right\} > 0,$$

where $\varepsilon = \epsilon_0/\epsilon_F$. This asymptotics is dominated by an antiferromagnetic contribution of the superexchange processes. For $R \gg R_c$ we get $I = I_0(\epsilon_F/\Delta)^2 (k_F R)^{-3} \cos 2k_F R$, which coincides with asymptotics of the conventional RKKY-interaction.

In the resonant case, when $\Delta \equiv \epsilon_F - \epsilon_0 \ll \epsilon_F$, the main contribution to the interaction comes from a narrow strip of width $< \Delta$ above the Fermi surface. As a consequence, a new spatial scale $R_{res} = \epsilon_F/k_F\Delta \gg R_c$, and a new intermediate asymptotics $I = I_0(\epsilon_F/\Delta)(k_F R)^{-2} \sin 2k_F R$, valid in the range $R_c \ll r \ll R_{res}$, arise. This asymptotics has a shifted phase of oscillations and slower decay of amplitude, compared to the RKKY one.

Let us now discuss the general case: $l > 0, J > 1/2$. It can be shown that, at "short" distances $k_F R \ll J$, the main contribution to (6) comes from the residues of the integrand, so that

$$\hat{H} = A_{JM_a} A_{JM_b} \frac{\pi I_0 \Lambda \hat{P}}{(k_F R)^{4l}} \exp\left\{\frac{m\epsilon_0 R^2}{4J}(D_{JM_a} + D_{JM_b})\right\}, \tag{7}$$

where $\Lambda = 2mR^2\epsilon_0$ for $J = l - 1/2$ and $\Lambda = (D_{JM_a} + D_{JM_b})/16J$ for $J = l + 1/2$. Note, that the interaction does not start to oscillate at $R \sim \pi/k_F$, as in conventional RKKY

interaction, but decreases monotonously, without changing its (antiferromagnetic) sign, up to $R \sim R_c \sim J/k_F$.

The physical interpretation of this result is as follows: The main contribution to the interaction comes from electrons which pass by the magnetic ions with impact parameters $\rho_a \sim \rho_b \sim R$. On the other hand, these electons should have angular momentum l, therefore their momenta $k \sim k^* = l/R$. The composite matrix element $\hat{H} \propto |V(k^*)|^4 \sim (l/k_F R)^{4l}$, which explains the principal features of (7). Note that for short distances, when $k^* \gg k_F$, the only processes, in which both electrons involved may have so high momenta, are the superexchange processes. Note also that the position of the Fermi level does not enter the expression (7). For large distances ($k_F R \gg J$), all factors in the integrand of (6), exept the oscillating ones, can be replaced by their value at $z = k_F R$. Then, using (4), we get

$$\hat{H} \approx -\hat{P}\left(\frac{\epsilon_F}{\Delta}\right)^2 \frac{I_0 B_{JM_a} B_{JM_b}}{2(k_F R)^{|M_a|+|M_b|+2}} \sin\left(2k_F R - \frac{\pi}{2}(|M_a| + |M_b|)\right). \tag{8}$$

For $|M_a| = |M_b| = 1/2$ it matches with the result of [6].

In the resonant situation we obtain, as in the nondegenerate case, an additional intermediate asymptotics: $\hat{H} \propto \mathrm{Im}\{\tilde{g}_{JM_a}(k_F R)\tilde{g}_{JM_b}(k_F R)\}$. In particular, at $R_c \ll R \ll R_{res}$ it leads to the phase shift $\pi/2$ and additional factor $(k_F R)\Delta/\epsilon_F$ in the amplitude of oscillations, exactly like in the nondegenerate case.

At moderate distances the spatial form of interaction differs considerably from the RKKY form even in the nonresonant case, though in the resonant case the difference is, of course, stronger. We have calculated the largest matrix element of interaction, $H^{1/2,1/2}_{1/2,1/2}(x = 2k_F R)$ numerically for two systems: the nondegenerate Anderson model, and f-levels (Cerium). The complete plots will be published elsewhere, here we only mention that the resulting dependences approach the standard one $F_{RKKY}(x) = x^{-3}\cos x - x^{-4}\sin x$ only for $x > 15$ (for $l = 0$) and for $x > 30$ (for $l = 3$). In rare earths, however, typically $2k_F R \approx 10$ for nearest neighbors, at these distances the spatial shape of interaction is quite far from RKKY even without resonance. Thus, neither long distance asymptotics (8), nor the simple RKKY function may be used for interpretation of experimental data for Cerium compounds; the k-dependence of matrix elements is essential for all practically important distances. Note, that this k-dependence arises here already in the leading order in $kr_0 \ll 1$, contrary to the case of the standard exchange-induced RKKY interaction, where the k-dependence of matrix elements occurs only due to corrections of higher order in kr_0 (see [13]).

There is an important message concerning the stability of the nonmagnetic Kondo-lattice state in the above results. Since $A_{JM} \sim (4l/e)^{4l}$ al large l (and small M), we can conclude from (7) that the energy (per site) in the magnetic state of a lattice of Anderson impurities is

$$E_{mag} \propto -|V_{k_F}|^4 (R_c/a_0)^{2N}, \quad R_c = 2N/ek_F, \tag{9}$$

provided the distance between nearest neighbors $a_0 \ll R_c$. The energy of the nonmagnetic (Kondo) state is $E_K \propto -\epsilon_F \exp(-\Delta/N\rho|V_{k_F}|^2)$, where ρ is the density of states at the Fermi surface [9]. Which energy is lower at high degeneracy $N \equiv 2l \to \infty$? To answer this question one rescales the parameters V_{k_F} and $k_F a_0$ so that E_K does not change with N, and then looks, whether E_{mag}/E_K goes to zero or to infinity at $N \to \infty$. It was argued [12, 3] that, to get a proper nonmagnetic state, the rescaling should be done in such

a way, that both effective coupling constant $N|V_{K_F}|^2/\Delta$ and the number of conduction electrons "per subband per site" $(k_F a_0)^3/N$ remain fixed. The latter means that R_c/a_0 scales as $N^{2/3}$ and, consequently, the condition $a_0 \ll R_c$ of the non-RKKY behavior is fulfilled at large N, so that one should indeed use Eq(9) for E_{mag}. Then one obtains $E_{mag}/E_K \propto N^{4N/3} \to \infty$, which means that the magnetic state, not the Kondo lattice, is preferable at large N, and the large-N Kondo-lattice scenario is inconsistent in the model, where the physical source for large N is the *orbital* degeneracy.

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