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Possible Deduction of Ferromagnetic Intersite Correlations in the Heavy-Fermion Compounds

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Following our previous work on the effect of the anisotropy of the hybridization matrix element for the Periodic Anderson model, we find further consistency with the experiments. The linear specific heat coefficient exhibits a similar enhancement, and is at the same critical field, as the magnetic susceptibility. However, quantitative agreements require the assumption that there exist ***ferromagnetic*** correlations among the impurity electrons, as has been postulated by other authors in different theories. The strength of these correlations is the strongest at the critical field, in contrast to being at zero field for the observable antiferromagnetic correlations, and is quenched at large fields. Other mean-field results are also presented and discussed.

I. INTRODUCTION

Kondo effect describes the hybridization between the magnetic impurity-electrons, mainly from the f-orbits of the rare earth elements, and the conducting electrons of the metallic host. This gives rise to a hybridization gap at the mean-field level, on the sides of which a large density of states accumulates. There have been doubts on whether this gap is realistic: for instance, does the gap still exist after taking into account the higher order fluctuations which gradually fill the gap. Convincing evidence has now been found for some so-called Kondo semiconductors [1], which insulate due to the existence of such a gap for both the charge and spin excitations. However, for the metallic heavy-fermion compounds it is less conclusive besides the support from the optical reflectivity experiments [2].

Taking into account the large Coulomb repulsion at the impurity sites, it is easy to show that, via an intermediate double occupancy in the conduction bloch state, the spins of impurity- and conduction-electron can interchange with a coupling constant $J \approx V^2/|E_0|$ where V is the hybridization matrix element and E_0 is the bare impurity-electron one-body energy. This results in a singlet ground-state. Therefore it came as a surprise when magnetic states were found for the heavy-fermion compounds. These magnetic correlations

signal the insufficiency of the single-impurity model and its basic difference from the more appropriate lattice model. It is now known that, for the dense heavy-fermion compounds, magnetic states are more of a rule rather than an exception (even when they superconduct). This is commonly thought to derive from the interimpurity interactions made possible by the Ruderman-Kittel-Kasuya-Yosida (RKKY) effect via the conduction sea. Actually, the real physical mechanisms are more subtle, which is partly why they remains unsolved theoretically for many decades. For one thing, the RKKY interaction needs to be modified because the conduction sea is no longer fully polarizable due to the Kondo effect; secondly, the magnetic correlation may come from the impurity-electrons Kondo-screening each other, as has been demonstated in the underscreening case and the depletion theorem by Nozières [3].

By making the Schrieffer-Wolff transformation [4], the Periodic Anderson Hamiltonian (for the Anderson lattice) can be transformed into the Coqblin-Shrieffer type (for the Kondo lattice), i.e., the hybridization term between the impurity- and conduction-electrons is substituted by a coupling term between their spins. Although both hamiltonians have been widely studies, we choose to use the Anderson lattice for our derivations because, except in the Kondo limit, the effect of charge fluctuations is not confined solely in the spin-coupling term (for instance, it also changes the chemical potential [5] which is important since the density of states depends sensitively on energy at the Kondo resonance).

The technique we employ is the large-degeneracy expansion of the slave-boson method [6], and the mean field approximation is made. Although this approximation has been known to be insufficient for the prediction of a magnetic or/and superconducting ground-state, in view of its vast success in other properties, we expect to extract further information at this level (especially considering the benefit of its simplicity). This will be duely justified by our results and the discussion to follow.

II. POSSIBLE EXISTENCE OF FERROMAGNETIC CORRELATIONS

Many heavy-fermion compounds show metamagnetic-like behaviour [7], i.e., the magnetic susceptibility χ exhibits a peak at some critical field H_c. Since there is no critical temperature, this is not a real phase transition. This peak sharpens at lower temperature, while H_c remains roughly the same. We have shown [8] that a similar behaviour can be obtained at the mean-field level when the anisotropy of the hybridization matrix element V is considered. This is different from the view held by some people that the RKKY interactions between the impurity electrons, which do not appear until the second-order pertubation, might be responsible.

Consistent with the experiments [7], we find here, plotted in Fig. 1, a similar enhancement for the linear specific heat coefficient γ, with a peak at the same critical field

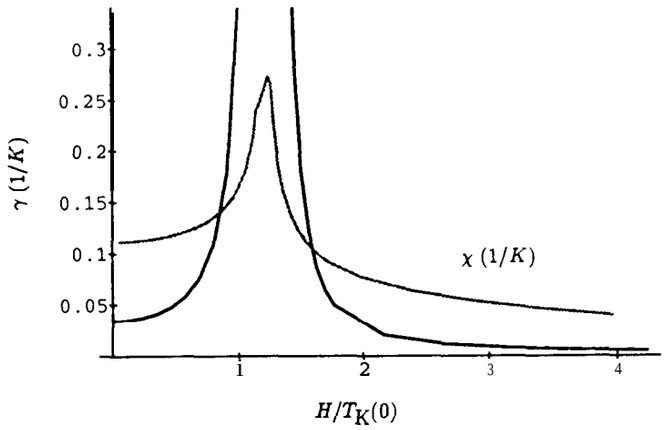

FIG. 1. Field dependence (normalized by $T_K(0)$) of the linear specific heat coefficient y (in black line) and the magnetic susceptibility χ (in grey line). Note that since we want to compare both quantities in the same plot, γ appears to diverge at the critical field, which value in fact is finite. The parameters chosen are: $V = 45000\ K$, bare conduction bandwidth $= 200000\ K$, bare one-body energy for the f-electron = -10000 K, and total number of electrons per unit cell = 1.7.

as for ***X,*** when ***V(6)*** obeys the anisotropic form [8]

$$V(\theta) = \sqrt{\frac{15}{8}} V(1 - \cos^2\theta)$$

appropriate for a doublet groundstate $m = \pm 5/2$ in the presence of crystal field splitting. Furthermore, the observed [7] asymmetry of the y(H) curve about the critical field and its saturation to a lower value at high fields than the zero-field value are reproduced. However, contrary to the experiments [9], the ratio of X and 7, or the Wilson ratio, does not exhibit a similar field dependence as of their individual curves. This can be understood from their definitions. Specific heat measures the state density D at the chemical potential, and it is easy to show that $7 = D_1 + D_2$ where the subscripts 1, 2 denote up and down spins. While

$$\chi(H) = \frac{M(H + \Delta H) - M(H)}{\mathrm{AH}} = 2Jg\frac{\Delta n_{f,\uparrow}}{\Delta H} \tag{1}$$

where $\Delta n_{f,\uparrow}$ denotes the increment of the up-spin electron occupation at the impurity site due to the small field increase AH, and J, g denotes the total angular momentum and

the Land& g factor of the impurity electrons (contribution from the conduction electron is neglected because of its small Jg value). For the second equality, we have neglected the change in the total n_f, valid in the heavy-fermion regime [10], and approximated $-\Delta n_{f,\uparrow} = \Delta n_{f,\downarrow}$. Since D_1 is normally bigger than D_2, the chemical potential μ has to shift by $\Delta\mu$ in order to satisfy the former condition:

$$\Delta n_{f,\uparrow} = D_1(Jg\Delta H + \Delta\mu)$$
$$-\text{An}_{f,\downarrow} = D_2(Jg\Delta H - \Delta\mu)$$

where the variation of $D_{1,2}$ has been neglected compared to the large $D_{1,2}$ characteristic of the heavy-fermion compounds. It is easy to solve for $\Delta\mu$ and rewrite Eq. (1) as

$$\chi(H) = \frac{4(Jg)^2 D_1 D_2}{D_1 + D_2} \tag{2}$$

Except at zero field, X(H) is normally smaller than y(H) in the mean-field approximation.

In order to further enhance X at the critical field, the intersite ferromagnetic correlations are needed. In spite of the experimental difficulty [11] in verifying their existence, the ferromagnetic correlations have been postulated by several authors [12] in different theories. Within the random phase approximation, the mean-field value χ_0 is enhanced by the ferromagnetic coupling, J_{ferro}, by a factor similar to the Stoner one:

$$\chi = \frac{\chi_0}{1 - \chi_0 J_{\text{ferro}}}. \tag{3}$$

From the experimental data [9] for the Wilson ratio in $CeRu_2Si_2$, we can deduce the field-dependence of J_{ferro}, which follows roughly the curve in Fig. 2. In contrast to the quenching of the antiferromagnetic correlations observed from the inelastic neutron scattering studies [13], these ferromagnetic correlations are strongest at the critical field. At large fields both correlations are quenched.

A physical explanation can be that the conduction sea, responsible for both the ferromagnetic and the antiferromagnetic RKKY couplings between the impurity-electrons, gradually loses its polarizability at high fields. This is equivalent to saying that the density of fully-polarizable conduction-electrons is gradually diminished. When this density is low, we know that the RKKY coupling is mainly ferromagnetic [14]. That probably gives rise to the peak in Fig. 2. At even higher fields, the conduction sea becomes totally rigid, and naturally we do not expect any of these two types of coupling. However, we would like to emphasize again that, since these ferromagnetic correlations have not been confirmed experimentally, these results should be treated with skepticism in spite of their plausibility from the physics point of view.

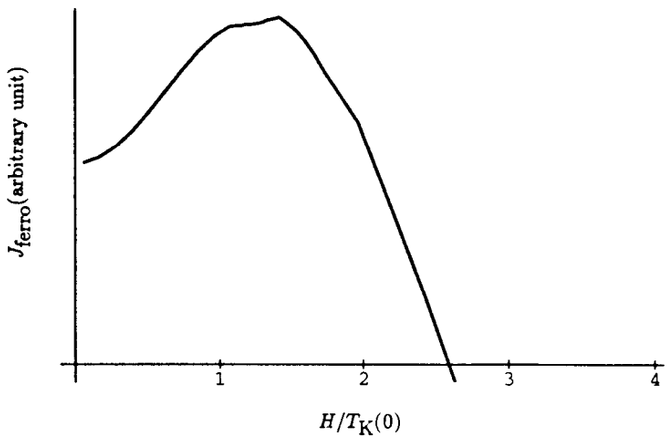

FIG. 2. Field dependence of the ferromagnetic coupling constant (in arbitrary unit) estimated from Eq. (3) and the experimental data [9] for the Wilson ratio in $CeRu_2Si_2$. Unlike the antiferromagnetic correlations [13] which decrease monotonically and are quenched strongest at the critical field, the ferromagnetic coupling is enhanced and reaches its maximum at the critical field before a similar quenching at large fields.

III. OTHER MEAN-FIELD RESULTS

If the hybridization matrix element is approximated by its root-mean-square average V *over* the Fermi surface, the mean-field equations become greatly simplified and analytic solutions are possible [8]. The latter can be achieved even at finite temperatures with the help of the recently proposed [15,16] "linear-temperature" approximation. The benefit of having an analytic solution can be examplified in the temperature dependence of the susceptibility χ. We find [16] that when χ exhibits a peak at some nonzero temperature T_m, as it always does for an isotropic hybridization [16], T_m obeys the following relation

$$T_m \exp\left(\frac{2.0843\,T_m}{\tau}\right) \approx 0.3733\,T_K(0) \exp\left(\frac{T_K(0)}{\tau}\right) \tag{4}$$

for $n = 2$ in a six-fold degenerate ground state, where $\tau\kappa V^4/W^3$, $2W$ is the conduction bandwidth, and T_K is the Kondo temperature [17]. When T_m is plotted against $T_K(0)$ in Fig. 3, the curve looks very linear as observed and concluded experimentally [18]. But Eq. (4) shows that this seemingly linear scaling, as opposed to the proven scaling for the linear specific heat coefficient and the Pauli susceptibility [19], is only approximate. Interestingly, T_m *always* corresponds [16] to when the tail of the linearized Fermi-Dirac distribution enters

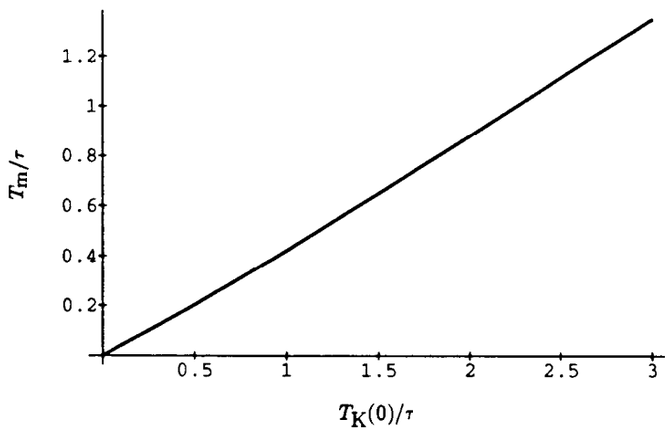

FIG. 3. T_m/τ is plotted as a function of $T_K(0)/\tau$ according to Eq. (4). T_m is the temperature when the susceptibility reaches its peak, $T_K(0)$ is the Kondo temperature at zero field, and $\tau \equiv V^4/W^3$ *as* defined in Eq. (4).

the hybridization gap. We also find X(T) $\approx$ X(0) + $O(T^2/T_K^3(0))$ at low T, and the Curie-Weiss behaviour at $T \gg T_K(0)$ with a negative $\theta \propto J$ signifying the antiferromagnetic nature of the Kondo effect. The scaling of the low-T variation of X with $T_K(0)$ is another support for the assertion that $T_K(0)$ is the only intrinsic energy scale at low T. However, it is important to point out that a second characteristic energy scale τ appears in most of the mean-field results [20,21], e.g. Eq. (4), and the validity of their scaling is based on the condition $T_K(0) << \tau$.

In addition to adjusting the energy parameters appearing in the Periodic Anderson Hamiltonian, it is important to consider the role of the total number of electrons n per unit cell that participate in the hybridization. Remembering that, due to the strong on-site Coulomb repulsion at the impurity sites for the heavy-fermion compounds, we have required $n_f \leq 1$. If we dope the compounds with p-type impurities, presumably n will increase. From the hybridization band picture, it is clear that n_f increases when μ rises with n. One interesting question to ask is: what happens to the compounds when n_f ultimately exceeds unity. One possible outcome will be for the crystal to undergo either a magnetic transition (because the mean-field theory breaks down) or a Jahn-Teller type of self-distortion in order to decrease the number of Fermi sheets crossing the impurity band.

We study the effect of n for a doublet ground state. The middle of the hybridization gap, approximately at $W(n-1-n_f)$, is normally not at zero energy, and therefore the number of accomodated impurity electrons is different in the two hybridization bands. For $n > 2$, a full lower band contains less than one impurity electron, which makes it possible for

μ to go into the upper band without violating the constraint $n_f \leq 1$. In this case, we find [16] that most of the thermodynamic properties remain unchanged except that T_m, at which $\chi(T)$ exhibits a peak, shifts to zero, providing the hybridization anisotropy is considered. In contrast, $T_m \neq 0$ for $n < 2$ with or without the anisotropy. To our knowledge, this is so far the only mechanism that can distinguish the equally frequent zero and nonzero T_m cases observed experimentally for the dense compounds. Note that our findings are different from the single-impurity problem where the size of the total angular momentum of the impurity-electron is known [22] to make the difference.

IV. DISCUSSION

The failure of the slave-boson mean-field approximation to predict a magnetic or/and superconducting ground state is well-known. It is not clear whether this is due to [23] the slave-boson technique which throws away the Coulomb repulsion term, or the mean-field approximation which loosens the local constraint into a global one. Although magnetism can be generated when second or higher order fluctuations are considered, its relevance to the real properties remains unconvincing [24]. The inelastic neutron scattering studies [13] show that, even when the heavy-fermion compounds do not order magnetically, there exists finite ranged magnetic correlations. So it comes as no surprise that we often have [16] difficulties obtaining a quantitative fit between the mean-field and the experimental results. In spite of these weaknesses, the mean-field approximation seems to give qualitatively correct descriptions for most other heavy-fermion features.

Even though the mechanism of the anisotropic $V(\theta)$ in causing a susceptibility peak [8] is unclear from the complicated angular averaging, from hindsight we know that the reason is probably that, when μ is driven toward smaller band gap by the external field (see Fig. 4 of Ref. 8), D_1 gets proportionally bigger [25] for those angles where μ is not in the gap. This overcomes the smaller weight from the angular averaging.

Although the exact functional form of y(H) close to the critical field H_c can not be determined from our numerical results, we find it misleading for γ to be empirically [26] fit to vary linearly with $\sqrt{H_c - H}$ when H is near, but not too close, to H_c. Because the T^2 coefficient of magnetization, proportional to $\partial\gamma/\partial H$ from the Maxwell relation, would then exhibit a positive curvature before the transition, contrary to the experimental results (see Fig. 3 in Ref. 26). Also we know that γ should be an even function of H from its invariance under the reversing of the field direction.

ACKNOWLEDGMENT

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