Crossover from dilute-Kondo system to heavy-fermion system

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Ground-state properties of a Kondo lattice model with random configuration of f electrons are investigated with a variational Monte Carlo method. We show that the crossover from a dilute-Kondo system to a heavy-fermion system occurs when the density of c and f electrons (n_c, n_f) become comparable, $n_f \lesssim n_c$. In the heavy-fermion region, the correlation between f electrons is strong and the f electrons themselves greatly contribute to the screening of other f-electron moments. We propose that the character of Kondo screening changes from "individual" to "collective" across the crossover.

I. INTRODUCTION

Heavy-fermion materials show various interesting phenomena and have been extensively studied so far. As is well known, f electrons with a strong localized character play an important role in this system, especially for a large effective mass. Origins of this large effective mass are considered to be the following factors: hybridization between f electrons and other conduction (c) electrons, strong Coulomb interaction between f electrons, and a certain number of f electrons. In this Brief Report, we focus on the third factor, namely, relation between the large effective mass and the number of f electrons.

When some magnetic ions with f electrons are replaced by the nonmagnetic ions, e.g., Ce→La or Yb→Lu, the system gradually changes from the heavy-Fermi liquid (dense limit) to the local Fermi liquid (single-impurity limit).[1–3] However, the details of this crossover are quite different in each compound. For example, in $Ce_{1-x}La_xPb_3$, the resistivity shows local-Fermi-liquid behavior even at $x=0.2$.[2] It is interesting that the single-impurity picture is valid even in a rather "dense" region. On the other hand, the situation is rather complex in $Ce_{1-x}La_xCoIn_5$. Between the local Fermi liquid and heavy-Fermi liquid, there is an intermediate region where a temperature scale characterizing the Fermi liquid is greatly suppressed.[3] It is proposed that the specific heat, magnetic susceptibility, and resistivity can be described by a sum of single-impurity and heavy-fermion contributions.[4,5] This "two fluid description" indicates that the system is fluctuating between the local Fermi liquid and heavy-Fermi liquid in the intermediate region. However, the detailed properties of this region have not been clarified so far.

For this problem, several model Hamiltonians, such as a periodic Anderson model (PAM) (Refs. 6–9) and a Kondo lattice model (KLM),[10,11] have been investigated. A crossover from the dilute-Kondo system to the heavy-fermion system is well described by these studies. However, the character of Kondo screening and the correlation between f electrons have not been discussed in detail.

In this Brief Report, we study the ground-state properties of the KLM with random configurations of f electrons, particularly focusing on the Kondo screening and the correlation between f electrons. We show that the crossover from the dilute-Kondo system to the heavy-fermion system occurs when the density of c and f electrons (n_c, n_f) become comparable, $n_f \lesssim n_c$. In the dilute-Kondo region, the f electrons behave independently and the single-impurity picture is valid. In the heavy-fermion region, the correlation between f electrons becomes strong and the f electrons themselves greatly contribute to the screening of other f-electron moments. We show the change in the character of Kondo screening from "individual" to "collective" across the crossover. The variational Monte Carlo (VMC) method is used for calculation. This method makes it possible to treat the localized f electrons correctly and to take into account the effect of randomness beyond the averaged treatment of coherent-potential approximation.

II. MODEL AND METHOD

We consider the following KLM in a two-dimensional square lattice:

$$H = \sum_{k\sigma} \varepsilon_k c_{k\sigma}^\dagger c_{k\sigma} + J \sum_{i \in A} \boldsymbol{S}_i \cdot \boldsymbol{s}_i. \tag{1}$$

where ε_k is the energy dispersion of c electrons and $c_{k\sigma}^\dagger (c_{k\sigma})$ is a creation (annihilation) operator of c electrons with momentum $\boldsymbol{k}$ and spin σ. $\boldsymbol{S}_i$ and $\boldsymbol{s}_i$ represent the f- and c-electron spins, namely, $\boldsymbol{S}_i = \frac{1}{2}\sum_{\sigma\sigma'} f_{i\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} f_{i\sigma'}$ and $\boldsymbol{s}_i = \frac{1}{2}\sum_{\sigma\sigma'} c_{i\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{i\sigma'}$. $J(>0)$ denotes the antiferromagnetic exchange coupling between them. The f electrons are completely localized and treated as localized spins. We call the lattice sites with and without f electrons A and B sites, respectively. To calculate the physical quantities, we take the average over 10–40 random configurations of A and B sites.

First, we consider the strong-coupling limit ($J/t \gg 1$). We fix the density of c electrons per site (n_c) and change the density of f electrons n_f ($0 < n_f \leq 1$) on the lattice with N_s sites, as shown in Fig. 1. In the strong-coupling limit, c electrons at an A site forms an on-site singlet pair with the localized f-electron spin. When $n_f < n_c$, all localized spins form
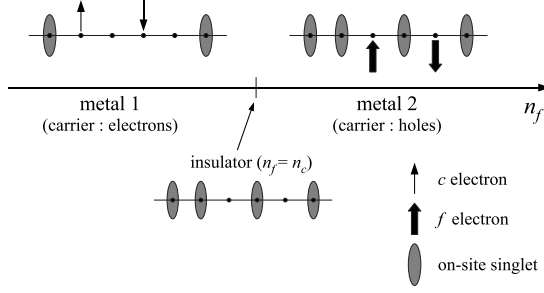
FIG. 1. Schematic phase diagram of the KLM in the strong-coupling limit as a function of n_f.

the on-site singlets and remaining $N_s(n_c-n_f)$ c electrons become mobile carriers (called as metal 1). On the other hand, when $n_f > n_c$, c electrons are "exhausted"[12] and $N_s(n_f-n_c)$ unscreened localized spins exist. In this case, when a c electron moves to a site with an unscreened localized spin (unscreened A site), the original on-site singlet disappears and another one is newly created. Thus the hopping of c electrons is described as a motion of on-site singlet among the A sites. It will cover all the A sites and will dynamically screen all the localized spins. We can say that this screening is "collective" in the sense that f electrons effectively participate the screening of other f-electron moments. The motion of on-site singlets can be regarded as a motion of mobile holes at the unscreened A sites and it is a different picture with metal 1 (metal 2). In the dense limit ($n_f = 1$), this system is identical to the $U = \infty$ Hubbard model with a reduced (half) band width.[13] Thus the metal 2 is considered to be a simple model for heavy-Fermi liquid. The metal 1 and metal 2 have different characters and separated by the special density of $n_f = n_c$ (see Fig. 1), where the system has a gap of order J/t and becomes insulator. For a finite value of J/t, this simple insulating state is not expected but it is possible that the character of metallic state changes around $n_f = n_c$.[10,11]

For finite J/t, we use the VMC method to calculate the physical quantities of the ground state. We prepare the Gutzwiller-type trial wave function $|\Psi\rangle$ for a trial state.[14] It consists of a projection operator $\hat{P}_f$ and a one-body part $|\Phi\rangle$ that is obtained from the solution of a certain one-body Hamiltonian. The specific form is as follows:

$$|\Psi\rangle = \hat{P}_f|\Phi\rangle, \quad \hat{P}_f = \prod_{i \in A} [\hat{n}_{i\uparrow}^f(1 - \hat{n}_{i\downarrow}^f) + \hat{n}_{i\downarrow}^f(1 - \hat{n}_{i\uparrow}^f)]. \quad (2)$$

$\hat{P}_f$ keeps the f-electron number of each A site exactly one. Since the f electrons must be treated as localized spins in the KLM, this constraint is necessary for the accurate calculation.[15] The one-body part $|\Phi\rangle$ is obtained by diagonalizing the $U = 0$ PAM in a real-space representation. The explicit form is as follows:

$$H' = \sum_{\langle i,j \rangle \sigma} \tilde{t}_{ij\sigma} c_{i\sigma}^\dagger c_{j\sigma} + \sum_{i,\sigma} E_{fi\sigma} f_{i\sigma}^\dagger f_{i\sigma} + \sum_{i,\sigma} \tilde{V}_{i\sigma}(f_{i\sigma}^\dagger c_{i\sigma} + \text{H.c.}). \quad (3)$$

Here, $\tilde{t}_{ij\sigma}$ are transfer integrals from site j to i with spin σ. $\tilde{V}_{i\sigma}$ and $\tilde{E}_{fi\sigma}$ are "effective" hybridization and f-electron en-
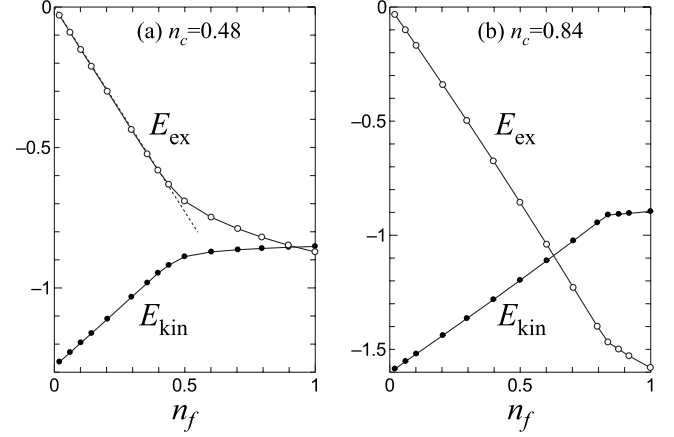

FIG. 2. n_f dependence of a kinetic energy E_{kin} and an exchange energy E_{ex} for $n_c = 0.48$ and $n_c = 0.84$ ($J/t = 3.0$).

ergy level for site i with spin σ. To avoid the occupancy of f electrons at B sites, we set $\tilde{E}_{fi\sigma} = \infty$ (or a sufficiently large value in the actual calculation) for $i \in$ B. $\tilde{t}_{ij\sigma}$, $\tilde{V}_{i\sigma}$, and $\tilde{E}_{fi\sigma}$ ($i \in$ A) are variational parameters and optimized so as to minimize the variational energy. Here, we set $\tilde{t}_{ij\sigma} = \tilde{t}$, $\tilde{V}_{i\sigma} = \tilde{V}$, and $\tilde{E}_{fi\sigma} = \tilde{E}_f$ for simplicity. Then we calculate physical quantities with the optimized wave function. To obtain the expectation values, we take 10–40 patterns of random configurations and average them. Although a large number of different configurations are necessary, this calculation procedure will take account of the effect of randomness more precisely than the coherent-potential approximation that is often used for disordered systems.

We perform a VMC calculation for a square lattice with $\varepsilon_{\mathbf{k}} = -2t(\cos k_x + \cos k_y)$. $t = 1$ is set to be a unit of energy in the following. The size dependence of results has been checked from 10×10 to 16×16. In the system with 10×10 sites, we have found some finite-size effects at $n_f \lesssim 0.16$,[16] but they disappear in the system with more than 14×14 sites. Since the 10×10 system is not sufficient for describing the dilute region, we show the results of 14×14 in the following.

III. NUMERICAL RESULTS

First, Fig. 2 shows the n_f dependence of kinetic energy E_{kin} and exchange energy E_{ex} that are expectation values of the first and second terms of right-hand side of Eq. (1). $n_f \to 0$ corresponds to the dilute limit and $n_f = 1$ corresponds to the usual KLM. For the case with $n_c = 0.48$ [Fig. 2(a)], E_{ex} shows almost linear behavior until $n_f \simeq 0.40$, namely, the exchange energy per f electron (E_{ex}/N_f) is almost unchanged even with increasing the number of f electrons. It means that the additional f electrons do not affect the screening of other f electrons so much. We can say that the f electrons in this region behave independently and the single-impurity picture is valid until $n_f \simeq 0.40$. Because of the finite value of J/t, this boundary is slightly different from that expected in the strong-coupling limit, 0.48. The effect of finite J/t is discussed later. For the case with $n_c = 0.84$ [Fig. 2(b)], as can be
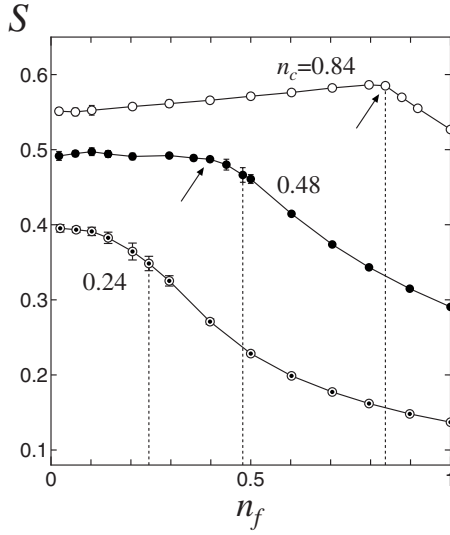
FIG. 3. n_f dependence of strength of on-site screening S for n_c=0.24, 0.48 and 0.84 (J/t=3.0). Dotted lines represent n_f=0.24, 0.48, and 0.84. Arrows indicate the crossover points.

seen by the abrupt changes in E_{kin} and E_{ex}, the single-impurity picture is valid until $n_f \simeq 0.84$. It is almost the same with the strong-coupling limit.

Next, we calculate the following quantity:

$$S = -\frac{E_{ex}}{JN_f} = -\frac{1}{N_f}\left\langle \sum_{i \in A} \mathbf{S}_i \cdot \mathbf{s}_i \right\rangle_{rnd}. \tag{4}$$

where $\langle \cdots \rangle_{rnd}$ denotes the random average. This quantity is always positive and corresponds to the strength of on-site screening per f electron. We fix the value of n_c as 0.24, 0.48, and 0.84 and show the n_f dependence of S for J/t=3.0 in Fig. 3. For n_c=0.84, as n_f increases, S gradually increases and rapidly decreases at $n_f \simeq n_c$=0.84. This crossover at $n_f \simeq n_c$ is consistent with the strong-coupling limit, and thus corresponds to the crossover from the dilute-Kondo system to the heavy-Fermion system. As shown later, the character of screening changes from individual to collective at this density. For n_c=0.48, on the other hand, the crossover occurs at $n_f \simeq 0.40$, slightly away from n_c. We consider that this is because the screening clouds have a finite spread for a finite value of J/t. In the strong-coupling limit, the screening clouds are completely localized at each site and have no overlap. In this case, the boundary between the dilute-Kondo system (individual screening) and heavy-fermion system (collective screening) is exactly at $n_f=n_c$. However, the screening can be collective if the neighboring screening clouds overlap and interact with each other, even though $n_f < n_c$. For n_c=0.48, the spread of screening cloud will exceed the lattice spacing and as a result, the region of heavy-fermion system extends to $n_f < n_c$. For n_c=0.24, the change in S becomes more gradual and we cannot determine the clear crossover point from these data. We conclude that the more n_c decreases, the more the screening cloud spreads and the crossover becomes ambiguous.
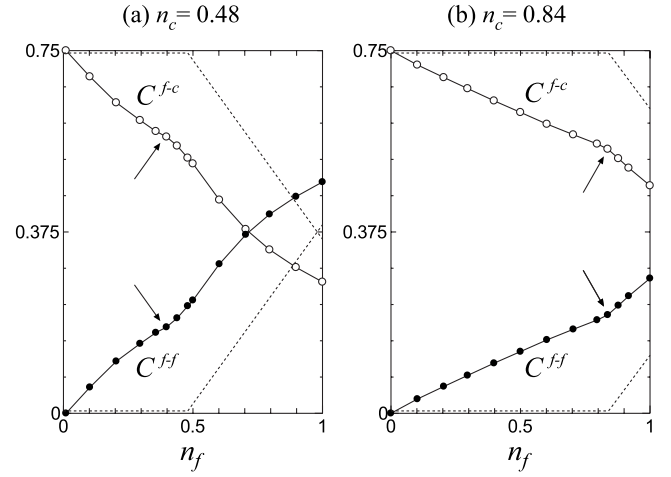

FIG. 4. n_f dependence of C^{f-c} and C^{f-f} for (a) n_c=0.48 and (b) n_c=0.84 (J/t=3.0). Dotted lines represent the strong-coupling limit. Arrows indicate the crossover points.

To observe the character of screening more clearly, we study the correlation between spins. For each f-electron spin $\mathbf{S}_i$, the following relation is satisfied:

$$C_i^{f-c} + C_i^{f-f} \equiv -\sum_j \mathbf{S}_i \cdot \mathbf{s}_j - \sum_{j \neq i} \mathbf{S}_i \cdot \mathbf{S}_j = +\frac{3}{4}, \tag{5}$$

because $\mathbf{S}_i$ forms singlet pairing with the rest of all spins in a paramagnetic state. We calculate $C^{f-c}=\langle C_i^{f-c} \rangle$ and $C^{f-f}=\langle C_i^{f-f} \rangle$, where $\langle \cdots \rangle$ denotes an average over both sites and random configurations. C^{f-c} (C^{f-f}) indicates how much the c (f) electrons are concerning with the screening. Figure 4 shows the n_f dependence of these quantities for n_c=0.48 and 0.84. In the single-impurity limit ($n_f \rightarrow 0$), screening is performed only by c electrons and $C^{f-c} \rightarrow 3/4$ (0.75) and $C^{f-f} \rightarrow 0$. As n_f increases, f electrons begin to participate the screening and C^{f-f} increases while C^{f-c} decreases. At a certain point around $n_f \simeq n_c$ (indicated by arrows in Fig. 4), an abrupt change is observed. It corresponds to the crossover point shown in Fig. 3 and represents the crossover from the dilute-Kondo system to the heavy-fermion system. In the heavy-fermion system, f electrons themselves are greatly participating the screening of other f-electron moments,[17,18] and therefore the correlation between f electrons is much stronger than in the dilute-Kondo system. We consider that this strong correlation between f electrons is an origin of large effective mass in the heavy-fermion system.

Note that we consider only the paramagnetic state because our aim is to describe the crossover within the paramagnetic state. However, some magnetic orders are expected in a certain condition. For example, the antiferromagnetic (AF) or spin-density-wave state is expected for small J where the Ruderman-Kittel-Kasuya-Yosida interaction overcomes the Kondo effect. Indeed, an AF transition at J/t=1.4–1.5 for $n_f=n_c$=1 is found in other theoretical studies.[19,20] To exclude the possibility of the AF state, we choose J/t=3.0 in the present study and discuss only the paramagnetic state. The ferromagnetic state is also expected for small n_c and large J at n_f=1 because of the equivalency between $J=\infty$ KLM and

$U=\infty$ Hubbard model. Here, we do not discuss the ferromagnetic state any more and leave it as an interesting future problem.

IV. DISCUSSION

Here, we compare our result with other theoretical works. Mean-field calculation by Kaul and Vojta for 20×20 square lattice[10] has shown the n_f dependence of $T^* = \langle b_r^2 \rangle / D$, where $2b_r = J \langle c_r^\dagger f_r \rangle$ denotes auxiliary fields and D denotes a half band width. T^* is almost the same quantity with S in our result and also shows the crossover around $n_f \simeq n_c$. Burdin and Fulde calculated T_0 / T_K using a coherent-potential approximation and a dynamical mean-field theory approach,[11] where T_0 denotes a coherence (Fermi liquid) temperature scale and T_K denotes a Kondo temperature. T_0 / T_K is an important ratio and related to the effective mass as $T_0 / T_K = m^*_{SIKM} / m^*_{KLM}$, where SIKM denotes single-impurity Kondo model ($n_f \rightarrow 0$ limit of KLM). Their result have shown the rapid decrease of T_0 / T_K, namely, the enhancement of effective mass around $n_f \simeq n_c$. It is consistent with our result showing that the crossover from the dilute-Kondo system to the heavy-fermion system occurs around $n_f \lesssim n_c$. The enhancement of effective mass for $n_f > n_c$ is also shown in the PAM by Pruschke *et al.*[21]

Finally, we give a brief comment on the relation between our result and the experiments. As mentioned in the introduction, an intermediate region with both the single-impurity and the heavy-fermion contributions are observed in $Ce_{1-x}La_xCoIn_5$.[4] This indicates that the crossover from the dilute-Kondo system to the heavy-fermion system is rather ambiguous and the system is fluctuating between them. In our result, the ambiguous crossover appears in the case of a small J/t and a small n_c because the screening cloud is fairly extended in these cases. We expect a smaller J/t and/or a smaller n_c for $Ce_{1-x}La_xCoIn_5$, compared to the materials with clear crossover, such as $Ce_{1-x}La_xPb_3$. In addition to the dilution effect, the substitution of atoms will cause various changes in the electronic state in real materials. We should consider these effects carefully for more precise comparison of theories and experiments.

V. SUMMARY

In this Brief Report, we have studied the ground-state properties of the KLM with random configuration of f electrons. The VMC method in real space is used for calculations. We have found that the crossover from the dilute-Kondo system to the heavy-fermion system occurs around $n_f \lesssim n_c$. In the dilute-Kondo region, the f electrons behave independently and the single-impurity picture is valid. In the heavy-fermion region, on the other hand, the correlation between f electrons is strong and the f electrons themselves participate the screening of other f-electron moments. It is considered to be an origin of large effective mass. We propose that the screening character changes from "individual" to "collective" across the crossover.

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