Theory of the metamagnetic crossover in $CeRu_2Si_2$

Hiroyuki Satoh and Fusayoshi J. Ohkawa
Division of Physics, Graduate School of Science, Hokkaido University, Sapporo 060-0810, Japan
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Based on the periodic Anderson model, it is shown that the competition between the quenching of magnetic moments by local quantum spin fluctuations and a magnetic exchange interaction caused by the virtual exchange of pair excitations of quasiparticles in spin channels is responsible for the metamagnetic crossover in $CeRu_2Si_2$, cooperated with the electron-lattice interaction. The strength of the exchange interaction is proportional to the bandwidth of quasiparticles and its sign changes with increasing magnetizations; it is antiferromagnetic in the absence of magnetizations, whereas it is ferromagnetic in the metamagnetic crossover region. Experimental results of static quantities are well reproduced.

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I. INTRODUCTION

The metamagnetic crossover in $CeRu_2Si_2$ is an important issue. The compound has a large electronic specific-heat coefficient of $\gamma \simeq 360$ mJ/mol K^2,[1] and it shows a sharp increase of magnetization at the field of $H_M \simeq 7.7$ T.[2,3] Other physical properties such as magnetostriction[4,5] and specific heat[6] are also anomalous in this field region. One of the most crucial experimental results to be explained is the single-parameter scaling;[4,7] independent experimental quantities for different pressures are scaled with a single energy parameter $k_B T_K$, where T_K is called the Kondo temperature. The Kondo temperature is the energy scale of local quantum spin fluctuations and is approximately equal to the bandwidth of quasiparticles.

The present authors showed in a previous paper[8] that an exchange interaction caused by the virtual exchange of pair excitations of quasiparticles in spin channels plays a critical role in the metamagnetic crossover as well as the electron-lattice interaction called the Kondo volume-collapse effect.[9] This exchange interaction has the following two interesting properties. First, it changes from being antiferromagnetic at zero fields to being ferromagnetic in the metamagnetic crossover region because of a pseudogap structure in the density of quasiparticle states, which is characteristic of heavy-electron compounds. Second, the strength of the exchange interaction is proportional to the bandwidth of quasiparticles. Then, the single-parameter scaling can be easily explained.

After the previous paper was submitted, a detailed measurement of the field dependence of specific heat was reported by Aoki *et al.*[10] At sufficiently low temperatures, a single sharp peak was observed at H_M, while a double-peak structure was found at higher temperatures. They argued that such a result can be explained if a sharp peak exists in the density of states. A similar argument applies straightforwardly to our previous model; however, if the pseudogap is deep enough, the magnetization process shows a first-order transition within the theoretical framework of the previous paper. Recently, two theories were proposed.[11,12] They claimed that an anisotropic *c-f* mixing plays an important role in the metamagnetic behavior. In both theories, however, the peak structure in the density of states is too sharp because the **k** dependence of hybridization matrices is improperly treated; one-dimensional van Hove singularity is irrelevant. Such an extremely sharp peak is inconsistent with the experimental result of specific heat. Furthermore, it should give a discontinuous transition instead of the metamagnetic crossover if the exchange interaction mentioned above is properly taken into account. To reproduce the experimental results of magnetization and specific heat simultaneously has not been achieved so far.

A main purpose of this paper is to reformulate and improve the previous theory based on the periodic Anderson model so as to explain static properties of the system. We will also give a brief comment on intersite spin fluctuations around the zone center.

II. MODEL

The periodic Anderson model is written as

$$\mathcal{H}=\sum_{\lambda \mathbf{k}\sigma} E_\lambda(\mathbf{k}) a^\dagger_{\lambda \mathbf{k}\sigma} a_{\lambda \mathbf{k}\sigma} + \sum_{\mathbf{k}\sigma} (E_f - \sigma H^*) f^\dagger_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma} + \sum_{\lambda \mathbf{k}\sigma} [V_\lambda(\mathbf{k}) a^\dagger_{\lambda \mathbf{k}\sigma} f_{\mathbf{k}\sigma} + \text{H.c.}] + \frac{1}{2} U \sum_{i\sigma} n_{i\sigma} n_{i-\sigma}, \tag{2.1}$$

with λ the band index of conduction electrons, $n_{i\sigma} = f^\dagger_{i\sigma} f_{i\sigma}$, and $H^* = m_0 H$, where m_0 is the saturation magnetization per *f* electron. Other notations are standard. The kinetic energy of conduction electrons $E_\lambda(\mathbf{k})$ and the *f* electron level E_f are measured from the chemical potential, respectively.

When multiple conduction bands are assumed, a pseudogap structure is expected in the density of states because of the hybridization between the *f* band and conduction bands.[13] Since the property of the magnetic exchange interaction that plays a critical role in the magnetization process depends on the shape of the density of quasiparticle states as shown in subsequent sections, a phenomenological model for the density of quasiparticle states is employed instead of considering explicit forms of $E_\lambda(\mathbf{k})$ and $V_\lambda(\mathbf{k})$ for simplicity. Furthermore, in order to make our treatment easy, we assume

that the system is symmetrical. In other words, the Hamiltonian (2.1) is assumed to be invariant under the particle-hole transformation.

One of the most crucial issues in constructing a theory of strongly correlated electron systems is how properly local quantum spin fluctuations, which are responsible for the quenching of magnetic moments, are treated. They can be correctly taken into account in the single-site approximation (SSA) (Ref. 14) that is rigorous for paramagnetic states in infinite dimensions. Consider the zero-field case at first; $H=0$. Within the SSA, Green's functions for f electrons and conduction electrons are respectively given by

$$G_{ff\sigma}(i\varepsilon_n,\mathbf{k})=\frac{1}{i\varepsilon_n-E_f-\tilde{\Sigma}_\sigma(i\varepsilon_n)-\sum_\lambda \dfrac{|V_\lambda(\mathbf{k})|^2}{i\varepsilon_n-E_\lambda(\mathbf{k})}}, \tag{2.2}$$

with $\tilde{\Sigma}_\sigma$ the single-site self-energy function, and

$$G_{\lambda\lambda'\sigma}(i\varepsilon_n,\mathbf{k})=\delta_{\lambda\lambda'}g_\lambda(i\varepsilon_n,\mathbf{k})+g_\lambda(i\varepsilon_n,\mathbf{k})V_\lambda(\mathbf{k}) \times G_{ff\sigma}(i\varepsilon_n,\mathbf{k})V^*_{\lambda'}(\mathbf{k})g_{\lambda'}(i\varepsilon_n,\mathbf{k}), \tag{2.3}$$

with $g_\lambda(i\varepsilon_n,\mathbf{k})=[i\varepsilon_n-E_\lambda(\mathbf{k})]^{-1}$. Here, $i\varepsilon_n$ is an imaginary fermion energy with n an integer. To obtain the single-site self-energy function is reduced to solving a single-impurity Anderson model[14] that has the same localized electron level E_f and on-site Coulomb repulsion U as those in Eq. (2.1). Call this Anderson model a mapped Anderson model (MAM). Other parameters in the MAM are determined through the mapping condition

$$\tilde{G}_{ff\sigma}(i\varepsilon_n)=\frac{1}{N}\sum_{\mathbf{k}} G_{ff\sigma}(i\varepsilon_n,\mathbf{k}), \tag{2.4}$$

where N is the number of unit cells and $\tilde{G}_{ff\sigma}$ is the Green's function of the MAM written as

$$\tilde{G}_{ff\sigma}(i\varepsilon_n)=\frac{1}{i\varepsilon_n-E_f-\tilde{\Sigma}_\sigma(i\varepsilon_n)-L(i\varepsilon_n)}, \tag{2.5}$$

with $L(i\varepsilon_n)=(1/\pi)\int_{-\infty}^{\infty}d\varepsilon\,\Delta(\varepsilon)/(i\varepsilon_n-\varepsilon)$. Here, $\Delta(\varepsilon)$ is the hybridization energy of the MAM. Once a trial function for $\Delta(\varepsilon)$ is given, $\tilde{\Sigma}_\sigma(i\varepsilon_n)$ is obtained by solving the MAM numerically. Therefore, Eq. (2.4) is a self-consistency condition for $\Delta(\varepsilon)$. However, we do not perform this self-consistent calculation in this paper. Instead, several approximations will be taken in subsequent sections with the use of well-known results for the Kondo problem.

Consider the spin susceptibility of the MAM, $\tilde{\chi}_s(i\omega_l)$, where $i\omega_l$ is a boson energy with l an integer. The Kondo temperature for the periodic Anderson model, which is the energy scale of local quantum spin fluctuations, is defined by

$$\lim_{T\to 0}\tilde{\chi}_s(+i0)\equiv\frac{1}{k_BT_K}. \tag{2.6}$$

In the same way as the previous paper, the electron-lattice interaction is taken into account simply through the volume dependence of the Kondo temperature:

$$T_K(x)=T_K(0)e^{-x}, \tag{2.7}$$

with $x=\Gamma\,\Delta V/V_0$. Here, $\Gamma\sim 190$ is the Grüneisen constant of T_K. For the sake of simplicity, the argument x will be omitted unless particularly required hereafter.

III. FERMI-LIQUID DESCRIPTION

The self-energy function is expanded as

$$\tilde{\Sigma}_\sigma(i\varepsilon_n)=\tilde{\Sigma}_\sigma(+i0)+[1-\tilde{\phi}_m]i\varepsilon_n+\ldots, \tag{3.1}$$

for small $|\varepsilon_n|$, where $\tilde{\phi}_m$ is a mass enhancement factor in the SSA. Note that $\tilde{\Sigma}_\sigma(+i0)=-E_f$ in the symmetrical case. Then the coherent part of Eq. (2.2) is written as

$$G^{(c)}_{ff\sigma}(i\varepsilon_n,\mathbf{k})=\frac{1}{\tilde{\phi}_m i\varepsilon_n-\sum_\lambda \dfrac{|V_\lambda(\mathbf{k})|^2}{i\varepsilon_n-E_\lambda(\mathbf{k})}}, \tag{3.2}$$

and correspondingly, $G^{(c)}_{\lambda\lambda'\sigma}$ is given by Eq. (2.3) with replacing $G_{ff\sigma}$ by $G^{(c)}_{ff\sigma}$. Quasiparticles are defined as the poles of Eq. (3.2), namely, the dispersion relation of quasiparticles is obtained by solving the following equation

$$\tilde{\phi}_m z-\sum_\lambda \frac{|V_\lambda(\mathbf{k})|^2}{z-E_\lambda(\mathbf{k})}=0. \tag{3.3}$$

We write the solutions as $z=\xi_\nu(\mathbf{k})$ with ν representing the branch of quasiparticles. By dividing the right-hand side of Eq. (3.2) into partial fractions, it follows that

$$G^{(c)}_{ff\sigma}(i\varepsilon_n,\mathbf{k})=\sum_\nu \frac{1}{Z^f_\nu(\mathbf{k})}\cdot\frac{1}{i\varepsilon_n-\xi_\nu(\mathbf{k})}, \tag{3.4}$$

where the renormalization factor is given by

$$\frac{1}{Z^f_\nu(\mathbf{k})}=\frac{1}{\tilde{\phi}_m+\sum_\lambda \eta_{\lambda\nu}(\mathbf{k})}, \tag{3.5}$$

with $\eta_{\lambda\nu}(\mathbf{k})=|V_\lambda(\mathbf{k})|^2/[\xi_\nu(\mathbf{k})-E_\lambda(\mathbf{k})]^2$. Similarly, we have

$$G^{(c)}_{\lambda\lambda\sigma}(i\varepsilon_n,\mathbf{k})=\sum_\nu \frac{1}{Z^\lambda_\nu(\mathbf{k})}\cdot\frac{1}{i\varepsilon_n-\xi_\nu(\mathbf{k})}, \tag{3.6}$$

with

$$\frac{1}{Z^\lambda_\nu(\mathbf{k})}=\frac{\eta_{\lambda\nu}(\mathbf{k})}{\tilde{\phi}_m+\sum_\lambda \eta_{\lambda\nu}(\mathbf{k})}. \tag{3.7}$$

It immediately follows from Eqs. (3.5) and (3.7) that the renormalization factors satisfy the relation

$$\frac{\tilde{\phi}_m}{Z^f_\nu(\mathbf{k})}+\sum_\lambda \frac{1}{Z^\lambda_\nu(\mathbf{k})}=1. \tag{3.8}$$

With the use of Eq. (3.8), the density of quasiparticle states can be written in the form

$$\begin{aligned}\rho^*(\varepsilon)&\equiv\frac{1}{N}\sum_{\nu\mathbf{k}}\delta[\varepsilon-\xi_\nu(\mathbf{k})]\\&=-\frac{1}{\pi N}\sum_{\mathbf{k}}\mathrm{Im}\Big\{\tilde{\phi}_m G^{(c)}_{ff\sigma}(\varepsilon_+,\mathbf{k})\\&\quad+\sum_\lambda G^{(c)}_{\lambda\lambda\sigma}(\varepsilon_+,\mathbf{k})\Big\},\end{aligned} \tag{3.9}$$

with $\varepsilon_+=\varepsilon+i0$. The energy scale for quasiparticles, T_Q, is defined by

$$\rho^*(0)\equiv\frac{1}{4k_BT_Q}. \tag{3.10}$$

On the other hand, $Z^f_\nu(\mathbf{k})$ obeys the following condition:

$$\sum_\nu \frac{1}{Z^f_\nu(\mathbf{k})}=\frac{1}{\tilde{\phi}_m}, \tag{3.11}$$

which can be proved by comparing Eqs. (3.2) and (3.4) for the limiting case of $|\varepsilon_n|\to+\infty$. It should be noted that $Z^f_\nu(\mathbf{k})\simeq\tilde{\phi}_m$ for the quasiparticle band that is closest to the Fermi level and $Z^f_\nu(\mathbf{k})\gg\tilde{\phi}_m$ for other branches for a given $\mathbf{k}$, that is,

$$\frac{\tilde{\phi}_m}{Z^f_\nu(\mathbf{k})}\simeq\begin{cases}1 & \text{for} \quad |\xi_\nu(\mathbf{k})|\lesssim 2k_BT_Q\\ 0 & \text{for} \quad |\xi_\nu(\mathbf{k})|\gg 2k_BT_Q\end{cases}. \tag{3.12}$$

Consider a spectral function of renormalized f electrons, ρ^*_f, defined by

$$\frac{1}{N}\sum_{\mathbf{k}} G^{(c)}_{ff\sigma}(i\varepsilon_n,\mathbf{k})=\frac{1}{\tilde{\phi}_m}\int_{-\infty}^{\infty}d\varepsilon\frac{\rho^*_f(\varepsilon)}{i\varepsilon_n-\varepsilon}, \tag{3.13}$$

or equivalently

$$\rho^*_f(\varepsilon)=\frac{1}{N}\sum_{\nu\mathbf{k}}\frac{\tilde{\phi}_m}{Z^f_\nu(\mathbf{k})}\delta[\varepsilon-\xi_\nu(\mathbf{k})]. \tag{3.14}$$

Equation (3.12) tells that

$$\rho^*_f(\varepsilon)\simeq\begin{cases}\rho^*(\varepsilon) & \text{for} \quad |\varepsilon|\lesssim 2k_BT_Q\\ 0 & \text{for} \quad |\varepsilon|\gg 2k_BT_Q\end{cases}, \tag{3.15}$$

and it follows from Eq. (3.11) that ρ^*_f satisfies the normalization condition of $\int_{-\infty}^{\infty}d\varepsilon\,\rho^*_f(\varepsilon)=1$.

The Luttinger's argument[15] applies straightforwardly to the periodic Anderson model. Consider the number of electrons per unit cell:

$$n_\sigma=\frac{1}{N\beta}\sum_{n\mathbf{k}} e^{+i\varepsilon_n 0}\Big\{G_{ff\sigma}(i\varepsilon_n,\mathbf{k})+\sum_\lambda G_{\lambda\lambda\sigma}(i\varepsilon_n,\mathbf{k})\Big\}, \tag{3.16}$$

with $\beta=1/k_BT$. At $T=0$ K, Eq. (3.16) can be transformed to

$$n_\sigma=\int_{-\infty}^{0}d\varepsilon\,\rho^*(\varepsilon), \tag{3.17}$$

which is an exact relation and is identical to the Luttinger's theorem. Let us derive an expression for the specific heat. Recently an exact expression for the entropy of an interacting system was established.[16] For the periodic Anderson model, it is written as

$$\begin{aligned}S=-\frac{1}{\pi N}\sum_{\mathbf{k}\sigma}\int_{-\infty}^{\infty}d\varepsilon\frac{\partial f(\varepsilon)}{\partial T}\Big\{&\sum_\lambda \mathrm{Im}\ln[-g^{-1}_{\lambda\sigma}(\varepsilon_+,\mathbf{k})]\\&+\mathrm{Im}\ln[-G^{-1}_{ff\sigma}(\varepsilon_+,\mathbf{k})]\\&+\mathrm{Re}\ G_{ff\sigma}(\varepsilon_+,\mathbf{k})\cdot\mathrm{Im}\,\tilde{\Sigma}_\sigma(\varepsilon_+)\Big\},\end{aligned} \tag{3.18}$$

where $f(\varepsilon)=1/(e^{\beta\varepsilon}+1)$. In Eq. (3.18), $\mathrm{Im}\,\tilde{\Sigma}_\sigma(\varepsilon_+)$ can be ignored and $G_{ff\sigma}$ can be replaced by its coherent part because the derivative of $f(\varepsilon)$ is nonzero only for small $|\varepsilon|$ at low temperatures. Furthermore, the T dependence of $\tilde{\phi}_m$ can be neglected in the temperature region of interest because its T dependence should be given by $(T/T_K)^2$ at low temperatures as long as the system is in a normal Fermi-liquid state. Since the chemical potential does not change as a function of temperature in a symmetrical model, the specific heat at constant volume is given by

$$C_V=T\frac{\partial S}{\partial T}=\sum_\sigma\int_{-\infty}^{\infty}d\varepsilon\,\varepsilon\frac{\partial f(\varepsilon)}{\partial T}\rho^*(\varepsilon), \tag{3.19}$$

where the identity $T\partial^2 f/\partial T^2=-\partial/\partial\varepsilon(\varepsilon\partial f/\partial T)$ has been made use of.

The zero-temperature limit of Eq. (3.19) is given by $C_V=\gamma T$ with $\gamma=(2\pi^2k_B^2/3)\rho^*(0)$. On the other hand, a combination of Eqs. (2.4), (3.13), and (3.15) gives $\rho^*(0)\simeq\rho^*_f(0)=\tilde{\phi}_m/\pi\Delta(0)$. When $\Delta(\varepsilon)$ is constant, one can prove that[8]

$$\frac{\tilde{\phi}_m}{\pi\Delta}=\frac{1}{4k_BT_K}, \tag{3.20}$$

and then it follows $T_K\simeq T_Q$ in the SSA, where T_K and T_Q are defined by Eqs. (2.6) and (3.10), respectively. We take this approximation in order to estimate the Kondo temperature. From the experimental value of γ, we have $T_K(0)\simeq 38$ K.

Let us consider the finite-field case; $H\neq 0$. In the previous paper,[8] we constructed a perturbation method to derive a microscopic Landau free energy, one of whose independent variables is the magnetization $m=\Sigma_\sigma\sigma\langle f^\dagger_{i\sigma}f_{i\sigma}\rangle$. In the presence of magnetizations, physical quantities, such as self-energy and polarization functions, can be expressed as a function of m instead of the magnetic field H. The equation that defines quasiparticles becomes

$$\tilde{\phi}_m z - \delta\tilde{\Sigma}_\sigma(m) - \sum_\lambda \frac{|V_\lambda(\mathbf{k})|^2}{z - E_\lambda(\mathbf{k})} = 0, \tag{3.21}$$

with $\delta\tilde{\Sigma}_\sigma(m)$ a magnetic part of the self-energy. In Eq. (3.21), m dependence of the mass enhancement factor has been ignored.[17] The solutions for Eq. (3.21) are denoted by $\xi_{\nu\sigma}(\mathbf{k},m)$. All arguments for the finite-field case can be developed in parallel with those for the zero-field case with replacing $\xi_\nu(\mathbf{k})$ by $\xi_{\nu\sigma}(\mathbf{k},m)$. For example, $\tilde{\phi}_m / Z^f_{\nu\sigma}(\mathbf{k},m)$ satisfies a similar property as Eq. (3.12). Therefore it follows by comparing Eqs. (3.3) and (3.21) that

$$\xi_{\nu\sigma}(\mathbf{k},m) \simeq \xi_\nu(\mathbf{k}) - \sigma\Delta E(m), \tag{3.22}$$

where $\sigma\Delta E(m) \equiv -\delta\tilde{\Sigma}_\sigma(m)/\tilde{\phi}_m$. With the use of Eq. (3.22), the Luttinger's theorem gives

$$m = \sum_\sigma \sigma \int_{-\infty}^{0} d\varepsilon\, \rho_f^*[\varepsilon + \sigma\Delta E(m)], \tag{3.23}$$

where the polarization of conduction electrons has been ignored. In Eq. (3.23), we have replaced ρ^* with ρ_f^*; because the right-hand side of Eq. (3.23) is a difference between the contributions from up and down spin directions, only the low-energy part is relevant. From Eq. (3.23), the magnetic part of the self-energy, $\Delta E(m)$, can be determined as a function of m.

A similar treatment for the MAM is also possible. Consider the coherent part of Eq. (2.5), which is defined by $\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n) \equiv (1/N)\Sigma_{\mathbf{k}} G^{(c)}_{ff\sigma}(i\varepsilon_n,\mathbf{k})$ and written as $\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n) = [\tilde{\phi}_m i\varepsilon_n - L^{(c)}(i\varepsilon_n)]^{-1}$ at zero fields. In the presence of magnetizations, it becomes

$$\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n,m) = \frac{1}{\tilde{\phi}_m[i\varepsilon_n + \sigma\Delta E_A(m)] - L^{(c)}(i\varepsilon_n)}. \tag{3.24}$$

Note that $\Delta E_A(m)$ is different from $\Delta E(m)$ because the MAM is determined in the absence of magnetizations by Eq. (2.4) and then magnetic one-body potentials are added to both the periodic Anderson model and MAM separately.[8] In other words, $\Delta E_A(m)$ is a single-site term even with respect to m, whereas $\Delta E(m)$ includes multisite magnetizations. Because the magnetization in the MAM is also given by m to leading order in $k_B T_K/U$,[8] $\Delta E_A(m)$ can be approximately evaluated from

$$m = \sum_\sigma \sigma \int_{-\infty}^{0} d\varepsilon \left(-\frac{1}{\pi}\mathrm{Im} \right) \tilde{\phi}_m \tilde{G}^{(c)}_{ff\sigma}(\varepsilon_+,m). \tag{3.25}$$

In case of a constant hybridization energy, Eq. (3.25) is a rigorous relation and is nothing but the Friedel sum rule. It should be noted that the evaluation of ΔE and ΔE_A from Eqs. (3.23) and (3.25) is one of the most essential improvements. In the previous paper, we assumed that $\Delta E(m) = \Delta E_A(m) = (4k_B T_K/\pi)\tan(\pi m/2)$.

Before closing this section, it is helpful to mention the volume dependence of the physical quantities that have appeared above. First, it follows from Eqs. (2.7) and (3.20) that $\tilde{\phi}_m \propto e^x$. Taking notice of Eq. (3.12), we have $\rho_f^*(\varepsilon;x) = e^x \rho_f^*(\varepsilon e^x;0)$ and $L^{(c)}(i\varepsilon_n;x) = L^{(c)}(i\varepsilon_n e^x;0)$. Therefore Eqs. (3.23) and (3.25) give $\Delta E(m) \propto e^{-x}$ and $\Delta E_A(m) \propto e^{-x}$, respectively.

IV. MAGNETIC EXCHANGE INTERACTION

In this section, we study magnetic exchange interactions working between quasiparticles. Consider the magnetic susceptibility to this end. In the presence of magnetizations, spin and charge channels of the susceptibility are coupled with each other in general. In the symmetrical case, however, they are separated and the magnetic susceptibility can be expressed as

$$\chi_s(i\omega_l,\mathbf{q},m) = \frac{2\pi_s(i\omega_l,\mathbf{q},m)}{1 - U\pi_s(i\omega_l,\mathbf{q},m)}, \tag{4.1}$$

with $\pi_s = (1/2)\Sigma_{\sigma\sigma'}\sigma\sigma'\pi_{\sigma\sigma'}$, where $\pi_{\sigma\sigma'}$ is the irreducible polarization function. Similarly, the magnetic susceptibility of the MAM is given by

$$\tilde{\chi}_s(i\omega_l,m) = \frac{2\tilde{\pi}_s(i\omega_l,m)}{1 - U\tilde{\pi}_s(i\omega_l,m)}, \tag{4.2}$$

with $\tilde{\pi}_s$ the single-site polarization function for the spin channel. The static and zero-temperature limit of $\tilde{\chi}_s$ is approximately given by[8]

$$\lim_{T\to 0} \tilde{\chi}_s(+i0,m) = \frac{1}{k_B T_K}(1-m^2)^{3/2}. \tag{4.3}$$

Because χ_s is also of order $1/k_B T_K$, Eq. (4.1) can be rewritten in the form

$$\chi_s(i\omega_l,\mathbf{q},m) = \frac{\tilde{\chi}_s(i\omega_l,m)}{1 - \frac{1}{4} I_s(i\omega_l,\mathbf{q},m)\tilde{\chi}_s(i\omega_l,m)}, \tag{4.4}$$

where $I_s(i\omega_l,\mathbf{q},m)$ is the intersite exchange interaction, which is given by

$$I_s(i\omega_l,\mathbf{q},m) = 2U^2\{\pi_s(i\omega_l,\mathbf{q},m) - \tilde{\pi}_s(i\omega_l,m)\}, \tag{4.5}$$

to leading order in $k_B T_K/U$. Note that Eq. (4.4) is consistent with the physical picture of the Kondo lattice that local quantum spin fluctuations at each site are connected with one another by the intersite interaction. In order to calculate I_s, we consider a two-line diagram shown in Fig. 1 for the polarization part $\pi_{\sigma\sigma'}$, which remains finite even in infinite dimensions for specific $\mathbf{q}$'s. In other words, the two-line diagram is a leading term with respect to $1/d$ for specific $\mathbf{q}$'s, with d the spatial dimensionality. In Fig. 1, $\tilde{\lambda}^{(A)}_{\sigma\tau}$ is a three-point vertex function of the MAM and $\tilde{\lambda}_{\sigma\tau}$ denotes that of the periodic Anderson model of $d \to +\infty$ limit. Note that $\tilde{\lambda}^{(A)}_{\sigma\tau}$ is a single-site term, while $\tilde{\lambda}_{\sigma\tau}$ contains multisite terms with respect to both U and m. It follows from Fig. 1 that

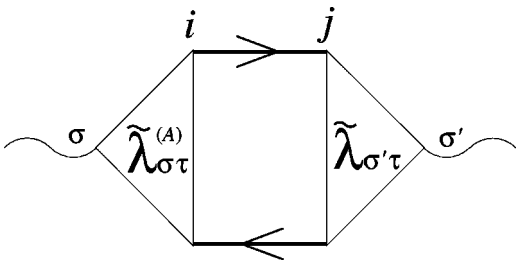

FIG. 1. Diagram for $\pi_{\sigma\sigma'}$ in the site representation. A thick line with an arrow stands for the Green's function. Overcounted contributions should be subtracted (see the text).

$$I_s(i\omega_l,\mathbf{q},m)=-\frac{U^2}{\beta}\sum_{n\sigma}\tilde{\lambda}_s^{(A)}(i\varepsilon_n,i\varepsilon_n+i\omega_l,m)\times\tilde{\lambda}_s(i\varepsilon_n,i\varepsilon_n+i\omega_l,\mathbf{q},m)\times\left\{\frac{1}{N}\sum_{\mathbf{k}}G_{ff\sigma}(i\varepsilon_n,\mathbf{k},m)G_{ff\sigma}(i\varepsilon_n+i\omega_l,\mathbf{k}+\mathbf{q},m)-\tilde{G}_{ff\sigma}(i\varepsilon_n,m)\tilde{G}_{ff\sigma}(i\varepsilon_n+i\omega_l,m)\right\},\tag{4.6}$$

where $\tilde{\lambda}_s^{(A)}\equiv\tilde{\lambda}_{\uparrow\uparrow}^{(A)}-\tilde{\lambda}_{\downarrow\uparrow}^{(A)}$ and $\tilde{\lambda}_s\equiv\tilde{\lambda}_{\uparrow\uparrow}-\tilde{\lambda}_{\downarrow\uparrow}$, respectively. In Eq. (4.6), the second term is necessary not only to subtract the single-site portion but also to avoid overcountings.

The main contribution to I_s is divided into two parts:[18]

$$I_s(i\omega_l,\mathbf{q},m)=J_s(\mathbf{q})+J_Q(i\omega_l,\mathbf{q},m).\tag{4.7}$$

The first term is an exchange interaction caused by the virtual exchange of high-energy spin excitations, which scarcely depends on m. It is composed of three terms; the conventional superexchange interaction, an extended superexchange interaction and an extended Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction.[18] Because the property of J_s depends on the whole band structure, we treat it as a phenomenological parameter. The second term, J_Q, is due to the virtual exchange of low-energy spin excitations within quasiparticle bands:

$$J_Q(i\omega_l,\mathbf{q},m)=-\frac{U^2}{\beta}\sum_{n\sigma}\tilde{\lambda}_s^{(A)}(+i0,+i0,m)\times\tilde{\lambda}_s(+i0,+i0,\mathbf{0},m)\times\left\{\frac{1}{N}\sum_{\mathbf{k}}G^{(c)}_{ff\sigma}(i\varepsilon_n,\mathbf{k},m)G^{(c)}_{ff\sigma}(i\varepsilon_n+i\omega_l,\mathbf{k}+\mathbf{q},m)-\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n,m)\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n+i\omega_l,m)\right\},\tag{4.8}$$

where energy dependence and $\mathbf{q}$ dependence of the vertex parts have been ignored. The vertex functions in Eq. (4.8) are related to the magnetic parts of the corresponding self-energy functions:

$$-\frac{U}{2}\tilde{\lambda}_s^{(A)}(+i0,+i0,m)=\tilde{\phi}_m\frac{\partial\Delta E_A(m)}{\partial m},\tag{4.9}$$

and

$$-\frac{U}{2}\tilde{\lambda}_s(+i0,+i0,\mathbf{0},m)=\tilde{\phi}_m\frac{\partial\Delta E(m)}{\partial m},\tag{4.10}$$

respectively. Therefore we have

$$J_Q(i\omega_l,\mathbf{q},m)=4\frac{\partial\Delta E_A}{\partial m}\cdot\frac{\partial\Delta E}{\partial m}\{P(i\omega_l,\mathbf{q},m)-\tilde{P}(i\omega_l,m)\},\tag{4.11}$$

with

$$P(i\omega_l,\mathbf{q},m)=\frac{1}{N}\sum_{\nu\nu'\mathbf{k}\sigma}\frac{f(\xi_{\nu'\sigma}(\mathbf{k}+\mathbf{q},m))-f(\xi_{\nu\sigma}(\mathbf{k},m))}{\xi_{\nu\sigma}(\mathbf{k},m)-\xi_{\nu'\sigma}(\mathbf{k}+\mathbf{q},m)+i\omega_l},\tag{4.12}$$

where Eq. (3.12) has been used and

$$\tilde{P}(i\omega_l,m)=-\frac{\tilde{\phi}_m^2}{\beta}\sum_{n\sigma}\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n,m)\tilde{G}^{(c)}_{ff\sigma}(i\varepsilon_n+i\omega_l,m).\tag{4.13}$$

In the following part of this section, we study the static and uniform component of Eq. (4.11), $J_Q(+i0,\mathbf{0},m)$. It is abbreviated to $J_Q(m,x)$ with the argument x explicitly written. Because excitations between different branches in Eq. (4.12) can be neglected for low-energy phenomena, we obtain

$$P(+i0,\mathbf{0},m)=\sum_{\sigma}\rho_f^*[\sigma\Delta E(m)].\tag{4.14}$$

On the other hand, Eq. (4.13) is expanded as

$$\tilde{P}(\omega_+,m)=\left[\frac{\partial\Delta E_A}{\partial m}\right]^{-1}+\frac{\pi}{2}i\omega\cdot\left[\frac{\partial\Delta E_A}{\partial m}\right]^{-2}+\ldots,\tag{4.15}$$

with the use of Eq. (3.25), where $\omega_+=\omega+i0$. It can be easily shown from Eqs. (4.11), (4.14), and (4.15) that the volume dependence of $J_Q(m,x)$ is the same as that of T_K, namely, $J_Q(m,x)=e^{-x}J_Q(m,0)$ and the magnitude of $J_Q(m,x)$ is of order k_BT_K. Thus $J_Q(m,x)$ is scaled with T_K, or the bandwidth of quasiparticles. This property is responsible for the single-parameter scaling.[4,8] Once an explicit form of $\rho_f^*(\varepsilon)$ is given, $J_Q(m,0)$ is straightforwardly calculated as a function of m. As a phenomenological model for $\rho_f^*(\varepsilon)$, we employ the following model in the same way as the previous paper:

$$\rho_f^*(\varepsilon)=\frac{1}{k_BT_K}\left\{\frac{c_1}{2}[D(\bar{\varepsilon};c_2,c_3)+D(\bar{\varepsilon};-c_2,c_3)]+(1-c_1)D(\bar{\varepsilon};0,c_4)\right\},\tag{4.16}$$

with $\pi D(y;a,b)=b/[(y-a)^2+b^2]$ and $\bar{\varepsilon}=\varepsilon/k_BT_K$. In this paper, $c_2=0.60$ and $c_3=0.15$ are assumed, which are different from previous ones. Because of the condition $\rho_f^*(0)\simeq 1/4k_BT_K$, only c_1 is variable. Figure 2 represents $\rho_f^*(\varepsilon)$

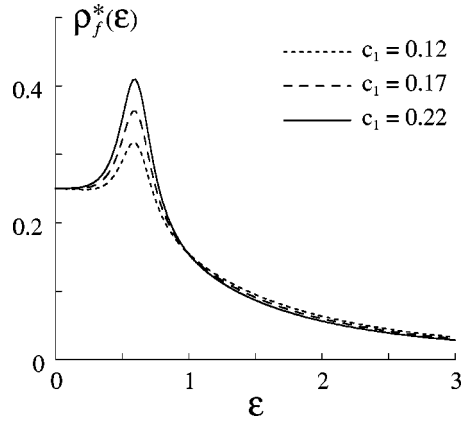

FIG. 2. Phenomenological model for $\rho_f^*(\varepsilon)$ for three values of c_1. Energies are in units of $k_B T_K$.

for three values of c_1, and the corresponding results for $J_Q(m,0)$ are shown in Fig. 3. The exchange interaction J_Q changes from being antiferromagnetic to being ferromagnetic with increasing m, which is consistent with the previous result. The maximum value of J_Q becomes larger when we raise the height of the peaks in ρ_f^*. Furthermore, it can be checked by changing c_2 with c_1 fixed that shifting of the location of the peaks to the band-edge side also enhances ferromagnetic instability.

V. THERMODYNAMIC QUANTITIES

The thermodynamic potential is expressed as

$$\Omega(m,x)=\Omega_{\rm para}(x)+\tilde{\Omega}(m,x)+\Delta\Omega(m,x). \tag{5.1}$$

Here, the first term is that for the paramagnetic state including a contribution from the lattice system. It is written as[8]

$$\Omega_{\rm para}(x)=\Omega_{\rm para}(0)+k_B T_K(0)\cdot\frac{x^2}{2\kappa}, \tag{5.2}$$

where κ is a dimensionless compressibility. The second term and the last term in Ω are the magnetic single-site and multisite terms; they are defined by $\partial^2\tilde{\Omega}/\partial m^2\equiv 1/\tilde{\chi}_s(+i0,m)$, and $\partial^2\Delta\Omega/\partial m^2\equiv -I_s(+i0,\mathbf{0},m)/4$, respectively. By integrating them, we obtain

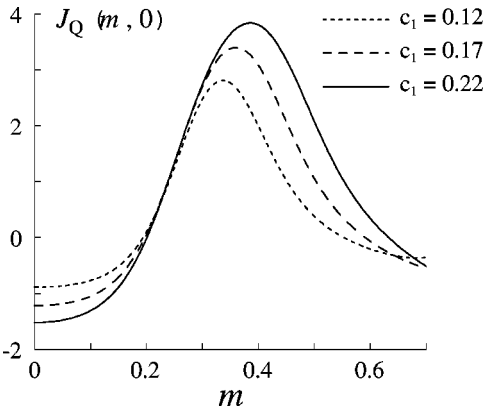

FIG. 3. Exchange interaction $J_Q(m,0)$ (in units of $k_B T_K$) as a function of m.

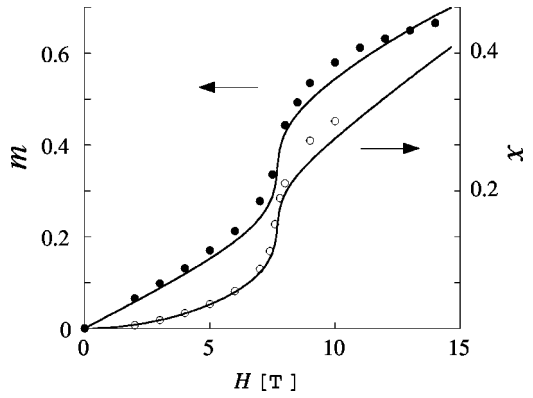

FIG. 4. Magnetization and magnetostriction. Experimental data are shown by dots and circles, respectively (Refs. 2 and 5).

$$\tilde{\Omega}(m,x)=k_B T_K(x)(1-\sqrt{1-m^2}), \tag{5.3}$$

and

$$\Delta\Omega(m,x)=-\frac{J_s(\mathbf{0})}{8}m^2-\int_0^m dm'\int_0^{m'}dm''\frac{J_Q(m'',x)}{4}. \tag{5.4}$$

In this section, we confine our study to the case of $c_1=0.17$. The equilibrium values of m and x are determined by solving the simultaneous equations, $\partial\Omega/\partial m=H^*$ and $\partial\Omega/\partial x=0$; they are shown in Fig. 4. Here, we have set the values of three parameters as $J_s(\mathbf{0})=0.3k_B T_K(0)$, $\kappa=2.4$, and $m_0=2.0\mu_B$ in order to have a better fit with experiments. The value of κ corresponds to 1.09 Mbar^{-1} in the ordinary unit. It should be mentioned that if the magnetization process is calculated with volume fixed constant against the magnetic field, the metamagnetic crossover is considerably suppressed.[8]

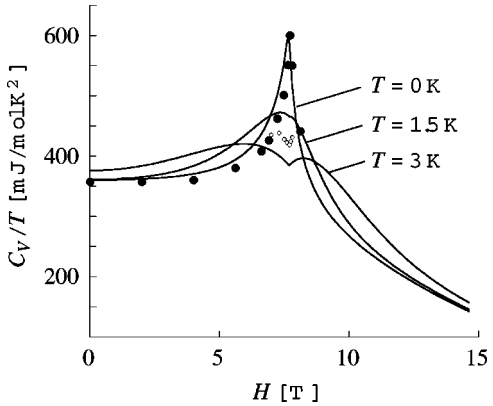

FIG. 5. C_V/T as a function of H. Experimental data at constant pressure for $T=0.25$ K are shown by dots (Ref. 10). Circles represent the values of C_V/T at $T=1.5$ K evaluated from experimental data (Ref. 19).

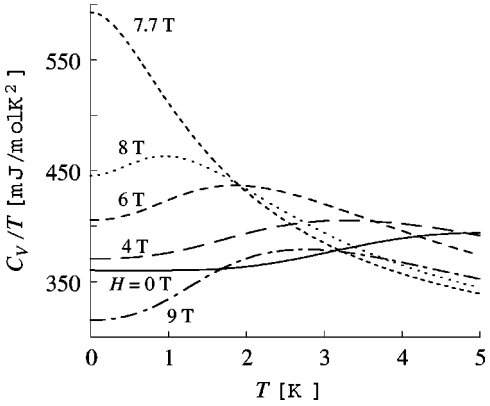

FIG. 6. C_V/T versus T for various values of the magnetic field.

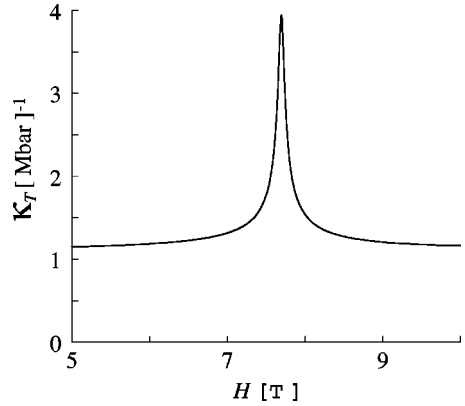

FIG. 7. Isothermal compressibility as a function of H.

Let us next study the specific heat. In the presence of magnetizations, it is given by Eq. (3.19) with $\rho^*(\varepsilon)$ replaced by $\rho_f^*[\varepsilon+\sigma\Delta E(m)]$. Figure 5 shows $C_V/T-H$ curves for three values of temperature; $T=0$, 1.5, and 3 K. The double-peak structure is qualitatively reproduced in the present model. Note that the experiment[10] was performed at constant pressure. From the experimental data, the specific heat at constant volume has been estimated;[19] it is also shown in Fig. 5 for $T=1.5$ K. In our calculation, no double peak appears at $T=1.5$ K, whereas it is observed above $T\simeq 1.0$ K in the experiment. A probable reason for this discrepancy is the easy estimation of the value of T_K based on Eq. (3.20). In addition to that, a contribution from ferromagnetic spin fluctuations or paramagnons, which is excluded in the SSA, may not be negligible. Figure 6 shows the temperature dependence of C_V/T for various values of the field, which is consistent with the experiment (Fig. 2 of Ref. 6).

Finally, the isothermal compressibility is given by

$$\frac{1}{\kappa_T(H)}=\frac{N\Gamma^2}{V_0}\left\{\frac{\partial^2\Omega}{\partial x^2}+\frac{\partial^2\Omega}{\partial x\partial m}\cdot\left(\frac{\partial m}{\partial x}\right)_H\right\}. \tag{5.5}$$

As shown in Fig. 7, it is strongly enhanced at H_M. Matsuhira *et al.*[19] have studied the field variation of the compressibility by using thermodynamic relations and assuming the single-parameter scaling. Our calculation shows a good agreement with their result.

VI. DISCUSSION

It is worthwhile to comment on spin fluctuations around the zone center, $\mathbf{q}\simeq 0$. The imaginary part of Eq. (4.12) is expanded as

$$\mathrm{Im}\,P(\omega_+,\mathbf{q},m)=c(m)\cdot\frac{\omega}{q}+\ldots, \tag{6.1}$$

for small $\mathbf{q}$ and small $\omega/|\mathbf{q}|$, where the coefficient $c(m)$ depends on the band structure. On the other hand, the inverse of the local susceptibility is expanded as[20]

$$\frac{1}{\tilde{\chi}_s(\omega_+,m)}=\frac{1}{\tilde{\chi}_s(+i0,m)}-\frac{\pi}{2}i\omega+\ldots. \tag{6.2}$$

Because the ω-linear terms in Eqs. (4.15) and (6.2) can be neglected compared with the ω/q term in Eq. (6.1) for small $\mathbf{q}$,[21] the total susceptibility is written in the form

$$\chi_s(\omega_+,\mathbf{q},m)\simeq\frac{1}{\mathrm{Re}[1/\chi_s(\omega_+,\mathbf{q},m)]-iC(m)\cdot\omega/q}, \tag{6.3}$$

with $C(m)=\partial\Delta E_A/\partial m\cdot\partial\Delta E/\partial m\cdot c(m)$. Since the system is close to ferromagnetic instability in the metamagnetic crossover region; $\mathrm{Re}[1/\chi_s(+i0,\mathbf{0},m)]\ll k_BT_K$, the imaginary part of Eq. (6.3) is expected to be enhanced around the metamagnetic point for small ω.

On the other hand, spin fluctuations in the Kondo lattice have been studied assuming the ω-independent RKKY interaction,

$$\chi_s(\omega_+,\mathbf{q},m)=\frac{\tilde{\chi}_s(\omega_+,m)}{1-I_{\mathrm{RKKY}}(\mathbf{q})\tilde{\chi}_s(\omega_+,m)}, \tag{6.4}$$

instead of Eq. (4.4) without any justification. In the low-energy region, however, Eq. (6.4) is expressed as

$$\chi_s(\omega_+,\mathbf{q},m)\simeq\frac{1}{\mathrm{Re}[1/\chi_s(\omega_+,\mathbf{q},m)]-\pi i\omega/2}, \tag{6.5}$$

and behaves quite differently from Eq. (6.3) around the zone center. Thus the argument based on Eq. (6.4) is apparently inadequate. Indeed, recent results of neutron-scattering measurements[22] cannot be understood from Eq. (6.4).

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