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Specific heat of heavy fermion systems in magnetic field—exchange field contribution

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Abstract

Magnetic field not only shifts and modifies the heavy fermion quasiparticle band, but also disturbs the distribution of residual exchange fields. An influence of the latter effect on the specific heat of non-magnetic heavy fermion systems is discussed. © 2002 Elsevier Science B.V. All rights reserved.

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Non-magnetic heavy fermion systems (HFS) (e.g. $CeCu_6$, $CeRu_2Si_2$) appear to be located close to magnetic instability. Slight deviations from stoichiometry or an application of pressure can induce cooperative magnetism [1]. In the early approaches a formation of a paramagnetic ground state was understood as a suppression of RKKY-type tendency to magnetic ordering by a single site Kondo screening effect [2]. In more recent discussions of Kondo lattice a paramagnetic ground state is considered as a collective state of the renormalized Fermi liquid. Nozières argues that a complete screening occurs for temperatures much lower than the single site Kondo temperature T_K, $T<T_c$ (T_c—coherence temperature) [3]. The singlet ground state results from the "isotropization" of local spin direction due to the dynamic exchange interaction with conduction electron which visits many sites. According to Nozières for $T_c<T<T_K$ a ground state is a dynamic mixture of a small number of inert singlets and a much larger amount of "bachelor" spins either up or down which can exchange with the singlets. A different picture which also

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stresses only partial screening of f-electron moments is presented in a series of papers by Kikoin et al. [4]. These authors propose the two-component Fermi liquid description with spin fluctuation responsible for thermodynamic properties and charge fluctuations accounting for transport phenomena. There is still a lack of a complete microscopic theory of magnetic fluctuations in HFS.

In the inelastic neutron scattering on $CeCu_6$ and $CeRu_2Si_2$, apart from a quasielastic line also a weak inelastic line which is q-dependent is observed [5]. The latter indicates the short-range magnetic correlations. These correlations can be destroyed by applying strong enough magnetic field (metamagnetic-like transition). The critical field value H_c correlates with the energy ($\hbar\omega_0$) of the inelastic line ($H_c \simeq \hbar\omega_0/2\mu$, μ is the magnetic moment value).

In the present paper, we discuss the contribution to the specific heat originating from the dynamically introduced disorder of exchange fields. We are not interested here in the region of quantum critical fluctuations where non-Fermi liquid behaviour is observed [6]. We skip in the following a discussion of Kondo contribution since this is widely discussed in the literature. Valence fluctuations prevent building of cooperative magnetism. Magnetic moment lifetime τ_K ($\tau_K \sim h/kT_K$) is much shorter than a typical time scale of long range magnetic correlations τ_M ($\tau_M < h/JS^2$, J is the n.n. exchange integral). In an oversimplified picture presented below we mimic the fluctuation of the exchange fields by introduction of fluctuating fields $\{c_i(t)\}$ taking values 1 or 0 with site average $\langle c_i(t)\rangle = 1 - v$, $v \ll 1$. Their average fluctuation lifetime τ

$$\tau = \int_0^\infty \langle c(0)\, c(t)\rangle \,\mathrm{d}t \tag{1}$$

is directly related to the half-width of the quasielastic peak and v measures the deviation from integer valence.

To illustrate the role of exchange field disorder we use Ising model with fluctuating fields

$$H = \sum_{ij} J_{ij}\, c_i(t)\, c_j(t)\, S_i S_j + h \sum_i c_i(t)\, S_i, \tag{2}$$

where $S_i = \pm\frac{1}{2}$, $h = h_0\mu|S|$ and h_0 is the external magnetic field. The Ising form of exchange is chosen, because the best-known non-magnetic HFS to which we address the present considerations—$CeCu_6$ and $CeRu_2Si_2$—have strong Ising-type anisotropy [7,8].

Let us first discuss the case where no magnetic correlations are included. The system behaves like a set of spins in the effective fields

$$h_i^{\mathrm{eff}} = H_i + h,$$
$$H_i = \sum_{j \neq i} J_{ij}\, c_i(t)\, S_j. \tag{3}$$

Hereafter, the field is measured in energy units. We approximate the effect of the fluctuating internal fields by the field distribution $P(H)$ which describes the probability that at any given time, the field acting on a spin S_i has a value H. We assume Gaussian form of distribution

$$P_h(H) = \frac{1}{\sqrt{2\pi}\,\Delta_h}(1-v)^z \exp\left[\frac{-(H-\bar{H})^2}{2\Delta_h^2}\right], \tag{4}$$

where z is the n.n. number. For $h=0$, $\Delta_0 = \sqrt{z}\,J|S|$ and $\bar{H}=0$ [9], $J>0$. For finite fields

$$\Delta_h = 2\sqrt{p\,(1-p)}\,\Delta_0,$$

$$\bar{H} = z\,(2p-1)\,J|S| \tag{5}$$

and the probability of parallel spin orientation p is related to the magnetization by

$$m = -(2p-1)\,\mu\,|S|. \tag{6}$$

An approximation of the distribution function by only one Gaussian is justified if a broadening caused by spin disorder on the n.n.n. shells is larger than a separation of the peaks corresponding to the different spin configurations of n.n. shell i.e. if $\sqrt{z_2}\,|J_2\,S| > 2\,|JS|$ (z_2 and J_2 denote coordination number and exchange integral of the n.n.n. shell). In the following, we will restrict to such a case. The specific heat which results from the distribution $P(H)$ reads

$$c = (1-v)\frac{1}{kT^2}\int_{-\infty}^{+\infty} P_h(H)\frac{[S(h+H)]^2\,\mathrm{d}H}{\cosh^2[(S(h+H)/kT)]} \tag{7}$$

and magnetization

$$m = -(1-v)\,\mu\int_{-\infty}^{+\infty} P_h(H)\tanh\left[\frac{|S|(h+H)}{kT}\right] \tag{8}$$

For small fields and low temperature

$$c \simeq \frac{\pi^2}{6}(1-v)\frac{P_h(-h)}{|S|}k^2T \equiv \gamma(h)T. \tag{9}$$

Taking the exchange energy parameter $JS^2 = h\omega_0 = 0.2\,\mathrm{meV}$ ($CeCu_6$) one gets the value of the linear temperature coefficient of the magnetic moment contribution to the specific heat $\gamma(h=0) = 0.65\,\mathrm{J\,mol^{-1}\,K^{-2}}$. The experimental value is $\gamma = 1.6\,\mathrm{J\,mol^{-1}K^{-2}}$ and we believe that the discrepancy is due to the neglect of Kondo effect in the present considerations. The square field dependence of c/T for small fields is a consequence of a shift of the field distribution. For higher fields magnetization changes more rapidly what results in the increase of the average internal field and the decrease of distribution width. Finally when magnetization saturates the distribution evolves into the function $\delta(H - z\,J\,|S|)$. A behaviour of the two-level system with the field dependent splitting is observed in this region. The results from Fig. 1 show that the disordered exchange fields picture can serve as a first approximation for $CeCu_6$. The weight of non–local magnetic scattering contribution for this system is of only 10% [5]. For $CeRu_2Si_2$ the inelastic peak contribution is 40% and the effects of correlations have to be included. A microscopic theory of a formation of quantum liquid where fluctuations allow for the existence of small magnetic clusters but prevent building of long-range order is

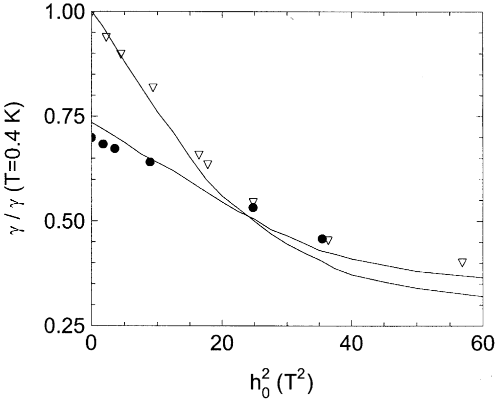

Fig. 1. The field dependence of $\gamma = c/T$ for the disordered exchange field system $CeCu_6$. The solid and broken lines are the calculated curves for $T = 0.4$ and 1 K, respectively, and triangles ($T = 0.4$ K) and dots ($T = 1$ K) are the corresponding experimental data [11].

still missing. This subtle effect can result from a minute disorder of any type, also the one induced by quantum fluctuations. An example of the latter are fluctuations of the values of magnetic moments caused by dynamic crystal field [10]. All these speculations however are outside the scope of the present paper.

Let us concentrate on the simple model, where only pairs with antiparallel ordered spins (AF) and "free spins" coexist. For clarity, we restrict to the case where for $h = 0$ the concentration of sites bounded in the AF pairs (x) is equal to the concentration of the free spin sites, i.e. $x = 1 - x = 0.5$. In the tetragonal $ThCr_2Si_2$ type structure ($CeRu_2Si_2$, $z = 4$) the free spin is surrounded on average by three spins from AF dimers and by one free spin. For sites from AF pairs the occupation of n.n. shell is opposite. Remembering our earlier assumption that the broadening of the peaks corresponding to the n.n. spin configurations resulting from the more distant exchange interactions is large enough to suppress the individual role of distinct peaks one can approximate the internal fields distribution by only one Gaussian. For the "free spin" sites it is taken in the form

$$P_s(H) = \frac{1}{\sqrt{2\pi}\,\Delta_s}(1-v)^z \exp\left(\frac{-(H-H_s)^2}{2\Delta_s^2}\right), \tag{10}$$

where

$$\Delta_s(h) = 2\sqrt{p(1-p)}\,\Delta_0,$$

$$H_s = z(2p-1)J|S|$$

and p is the average probability of parallel orientation of free spins (p_s) and spins bounded in AF pairs (p_p)

$$p = \frac{2(z-1)\,xp_p + (1+(z-1)(1-2x))p_s}{z}.$$

The distribution of exchange fields acting on the AF dimers is taken in the form

$$P_h(H_1, H_2) = AP_p(H_1)\,P_p(H_2)\Theta(J|S| - H_1 - h)\,\Theta(H_2 + h - J|S|), \tag{11}$$

where

$$P_p(H) = \frac{1}{\sqrt{2\pi}\,\Delta_p}(1-v)^{z^*}\exp\left(\frac{-(H-H_p)^2}{2\Delta_p^2}\right) \tag{12}$$

with

$$z^* = z - 1,$$

$$\Delta_p(h) = 2\sqrt{p_s(1-p_s)}\,\sqrt{z^*}J\,|S|,$$

$$H_p = z^*(2p_s - 1)J\,|S|.$$

The normalization constant

$$A = \left[\int P_{h=0}(H_1, H_2)\,\mathrm{d}H_1\,\mathrm{d}H_2\right]^{-1}.$$

The Θ functions introduced in Eq. (11) cut off the exchange fields which for $T = 0$ destroy the ground state with antiparallel spins. The specific heat of AF pairs under the influence of exchange fields reads

$$c = (1-v)^2\frac{1}{kT^2}\int P(H_1, H_2) \times \left[\frac{\sum_{\{\sigma\}}(E_\sigma^2 \mathrm{e}^{-\beta E_\sigma})(\sum_{\{\sigma\}}\mathrm{e}^{-\beta E_\sigma}) - (\sum_{\{\sigma\}}E_\sigma \mathrm{e}^{-\beta E_\sigma})^2}{(\sum_{\{\sigma\}}\mathrm{e}^{-\beta E_\sigma})^2}\right]\mathrm{d}H_1\,\mathrm{d}H_2, \tag{13}$$

where $\beta = kT$, $\{\sigma\} = \{(S_1, S_2)\}$ and $E_{\{\sigma\}} = E_{S_1S_2} = JS_1S_2 + H_1S_1 + H_2S_2 + h(S_1 + S_2)$.

The calculated specific heat coefficient γ for the system with magnetic correlations is presented in Fig. 2. Generally the dominant contribution to the low temperature specific heat give the spin subsystems with the first excited state lying close to the ground state. For the small external fields it corresponds in the case of free spins to the small exchange fields ($H \sim kT/|S|$, centre of the distribution) and for AF pairs ($H_2 + J|S| < kT/|S|$, $J|S| - H_1 < kT/|S|$, fields close to the edges of distribution). The field dependence of the free spin contribution to γ is weaker than for the disordered system. For low fields the magnetization of the AF pairs subsystem does not change rapidly. This quantity determines both the width of distribution (10) and the weight of the free spin contribution. The weak decrease of γ caused by a shift of distribution (10) with the increase of the field is partially compensated by the increase of the weight $(1-x)$. The value of the external field which destroys AF pair depends on the exchange field configuration. The stability region of the pair in the low temperature range is governed by $E_s(H_1, h) + E_s(H_2, h) - E_{\mathrm{pair}}(H_1, H_2, h) > 0$, where

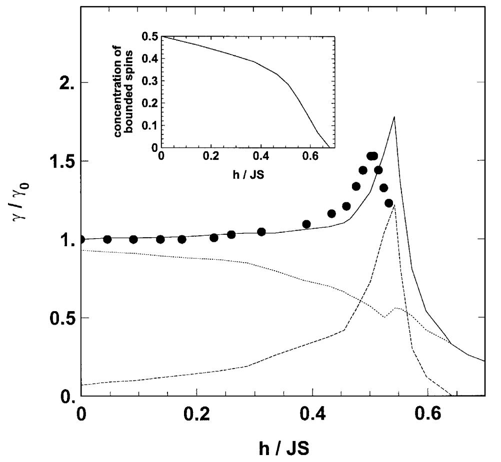

Fig. 2. The field dependence of γ for the non-magnetic HFS with short range magnetic correlations $CeRu_2Si_2$ ($T = 0.5\,K$). The dotted and broken lines are the "free spin" contribution and the contribution from the bounded pairs with antiparallel spins, respectively. The sum of the curves (solid line) is compared with the experimental data (dots) [13]. Inset shows the field dependence of the percentage of sites bounded in AF pairs.

E_s and E_{pair} are the free spin and pair ground states, respectively. For isolated dimer the destruction field takes a value $h_c \sim J|S|$. For partially polarized free spin subsystem ($h \neq 0$) h_c shifts towards smaller fields. One can roughly estimate the decreasing percentage of the sites bounded in the pairs by $x \sim \frac{1}{2} \int P_h(H_1, H_2)\, dH_1\, dH_2$ (Fig. 2). The pair concentration well correlates with the observed field dependence of the inelastic line intensity [5]. Increasing the external field one reaches the region of a rapid decrease of the number of AF pairs ($x \to 0$, metamagnetic-like transition). The peak of the field dependence of c/T reflects this transition. A very crude estimate of the average critical field H_c gives $H_c \sim J\,|S|(1 - z^*(2p_s(H_c) - 1))$. Although we compare the results with the data for $CeRu_2Si_2$ we realize that the above description is incomplete and only very crude. It accounts for only direct magnetic moment contribution. We do not discuss an influence of magnetic field on Kondo effect. The simple phenomenological analysis similar to the single-resonance-level model of Schotte and Schotte [12] is given in Ref. [13]. The main simplification of our approach is a separation of the proposed mechanism from the Kondo effect. In fact, magnetic fluctuations are essential for both of them. The discussed contribution to the specific heat with JS^2 taken as the energy of an inelastic line $h\omega_0 = 1.2\,meV$ gives $\gamma(0) = 85\,mJ\,mol^{-1}K^{-2}$. This value is about 10% underestimated due to the different from experimental assumed zero field value of the ratio of intra and inter site magnetic scattering contributions. The experimental value of γ for $CeRu_2Si_2$ is $\gamma = 350\,mJ\,mol^{-1}K^{-2}$ and thus the residual exchange field contribution to the linear coefficient of the specific heat for this system is of only about 30%.

In summary, we have tried, using an oversimplified Ising model with fluctuating fields, to expose the role of magnetic fluctuations in the low temperature specific heat of non-magnetic HFS above the coherence temperature. The proposed mechanism can be considered as complementary to the Kondo effect but can account for only one third of the observed enhancement of γ. Crucial for the presented discussion is an assumption of the incomplete screening of magnetic moments. This problem was recently widely discussed in literature [3,4], but a satisfactory theory is still missing. The conviction about the underscreening has its ground in the observation of magnetic correlations in non-magnetic HFS. It is believed that since inelastic neutron scattering mainly probes only the deep 4f component and not the weak 4f component at the Fermi level [5], the observed correlations are due to the residual moments and not due to the quasiparticle excitations.

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