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1993 J. Phys.: Condens. Matter 5 1947

(http://iopscience.iop.org/0953-8984/5/13/011)

J. Phys.: Condens. Matter 5 (1993) 1947–1956. Printed in the UK

On the thermodynamic properties of heavy-fermion systems in a magnetic field

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Received 3 June 1992, in final form 10 November 1992

Abstract. Using a two-conduction-band periodic Anderson model, the influence of a magnetic field on the thermodynamic properties of heavy-fermion systems is studied by means of the slave-boson mean-field theory. A depression of the heavy-fermion state by the magnetic field is found. A theoretical explanation for the anomalous volume magnetostriction of heavy-fermion systems is also given.

1. Introduction

The heavy-fermion systems (HFSs), which contain an entire class of rare-earth or actinide intermetallic compounds, are characterized by a very large density of states (DOS) at the Fermi level. Some of them show neither superconducting nor magnetic orders at extremely low temperatures, and most of the properties of these materials can be described in terms of a Fermi liquid with a very large effective mass [1, 2]. Recently, however, metamagnetic transitions have been observed in these normal-state compounds by alloying and applying a pressure [3–5], and the existence of weak magnetic correlation has been observed in neutron scattering experiments [6]. All these show a weak stability of the normal phase of HFSs, and the conventional Fermi liquid picture is facing a challenge. In order to understand more thoroughly the normal ground state of HFSs, much attention has been paid in the past few years to the effects of a magnetic field on the physical properties of HFSs, and especially of Ce-based compounds ($CeAl_3$, $CeCu_6$, $CeRu_2Si_2$ and $CeCu_2Si_2$). A large amount of experimental evidence reveals strong field dependences of the properties of these materials [7–14]. It has been found that a magnetic field suppresses the heavy-fermion state efficiently. The linear coefficient of the specific heat decreases at very low temperatures but increases above a certain temperature $T_{crossing}$ in a non-zero magnetic field, and a maximum emerges at a finite temperature when the magnetic field is sufficiently strong. $T_{crossing}$ and $T_{maximum}$ increase slowly with increasing magnetic field. Accompanying the suppression of heavy-fermion effects in the specific heat coefficient, the susceptibility at low temperatures is also reduced in a magnetic field. Another interesting experimental discovery is the volume magnetostriction of heavy-fermion compounds, which is found to be 10^2–10^3 times larger than that of a usual metal. Many anomalous magnetotransport properties have also been found in the coherent regime [15–17]. All these experimental results are significant in the study of the normal ground state of HFSs, especially when the applied field is strong enough to bring about evident modification of the quasi-particle band structure.

0953-8984/93/131947+10$07.50

Theoretical study of the anomalous field-dependent properties of HFSs seems to be an important and arduous task. A few studies using different approaches have been reported [14, 18]; nevertheless they are either phenomenological or only able to describe certain properties. Therefore a self-consistent microscopic theory which can clearly explain both thermodynamic and transport properties of heavy-fermion metals in a magnetic field in a unified picture is needed.

In the present paper, we report our theoretical result that most of the thermodynamic anomalies of HFSs in a magnetic field can be well described in a narrow-band picture. We have studied the influence of the magnetic field on thermodynamic properties of HFSs based on the widely accepted periodic Anderson model (PAM), and our results are in good agreement with experiments. Using this theory, we have also carried out research about the anomalous magnetotransport properties of HFSs [19]. The rest of this paper is organized as follows. In section 2, the two-conduction-band PAM is introduced, and the field dependence of the quasi-particle band structure is studied via the slave-boson mean-field approach. Results on the specific heat coefficient and magnetic susceptibility and the anomalous volume magnetostriction in a non-zero magnetic field of HFSs are presented in section 3. Finally, section 4 contains some conclusions and discussions.

2. Two-conduction-band PAM in a magnetic field

Generalized from the single-impurity Anderson [20] model, the $U = \infty$ periodic Anderson Hamiltonian is referred to as a suitable description for the normal state of heavy-fermion metals [21]. If the slave-boson technique introduced by Barnes [22] and Coleman [23] is utilized, the strong on-site correlation between f electrons is clearly described, and the Hamiltonian becomes easily solvable in the mean-field approximation which gives rise to exact results in a large-degeneracy limit, from which a Fermi liquid state with a large effective mass is obtained [24, 25]. As the system is metallic, we should consider two conduction bands at least. Therefore, as proposed by Ohkawa [26], we assume there are two conduction bands mixed with f electrons in the systems. The two-conduction-band slave-boson Hamiltonian can be written as [27]

$$\hat{H} = \sum_{i=1,2}\sum_{k\sigma} \epsilon_{ik} c_{ik\sigma}^{+} c_{ik\sigma} + \sum_{l\sigma} E_0 f_{l\sigma}^{+} f_{l\sigma} + V \sum_{i}\sum_{l\sigma} (c_{il\sigma}^{+} f_{l\sigma} b_l^{+} + f_{l\sigma}^{+} c_{il\sigma} b_l) \qquad (1)$$

with a constraint on each site, reflecting the strong on-site f–f correlation

$$\sum_{\sigma} f_{l\sigma}^{+} f_{l\sigma} + b_l^{+} b_l = 1 \qquad (2)$$

where $\sigma = \pm 1$ is the spin index and $c_{ik\sigma}$ ($f_{l\sigma}$) is the annihilation operator of the ith conduction band (localized) electron in Bloch (Wannier) representation. E_0 is the bare f-electron energy level, and V the conduction electron–f-electron mixing strength which is assumed to be site independent in our model. For simplicity, we introduce two conduction bands with the same dispersion $\epsilon_{k\sigma}$ and assume that the two conduction bands are expressed as

$$\epsilon_{1,2k\sigma} = \epsilon_{k\sigma} \pm \epsilon_0 \qquad (3)$$

with the constant DOS of unperturbed conduction-band electrons given by

$$\rho(\epsilon) = \frac{1}{N}\sum_k \delta(\epsilon - \epsilon_k) = \begin{cases} 1/2D & |\epsilon| \leqslant D \\ 0 & |\epsilon| > D \end{cases} \tag{4}$$

where N is the number of lattice sites. We suppose that the effect of the applied magnetic field is to add Zeeman energy in both conduction and f electrons without changing the mixing strength V:

$$\epsilon_{ik\sigma} = \epsilon_{ik} + \sigma g_c \mu_B B \tag{5}$$

$$E_{0\sigma} = E_0 + \sigma g_f \mu_B B. \tag{6}$$

For simplicity, we take $g_c = g_f = g$ and define a reduced magnetic field $H = g\mu_B B$. Then equations (5) and (6) can be rewritten as $\epsilon_{ik\sigma} = \epsilon_{ik} + \sigma H$ and $E_{0\sigma} = E_0 + \sigma H$.

In the mean-field approximation, both b and b^+ can be replaced by a c-number r. In order to include the constraint (2), we introduce a Lagrange multiplier λ, and the effective Hamiltonian in the slave-boson mean-field approximation is

$$\hat{H}' = \sum_{ik\sigma} \epsilon_{ik\sigma} c^+_{ik\sigma} c_{ik\sigma} + \sum_{l\sigma} E_{f\sigma} f^+_{l\sigma} f_{l\sigma} + rV \sum_{il\sigma} (c^+_{il\sigma} f_{l\sigma} + f^+_{l\sigma} c_{il\sigma}) + \sum_l \lambda(r^2 - 1) \tag{7}$$

with

$$E_{f\sigma} = E_{0\sigma} + \lambda \tag{8}$$

where the mean-field parameter λ and r are determined by the minimum condition of the free energy:

$$\sum_\sigma \langle f^+_{l\sigma} f_{l\sigma} \rangle + r^2 = 1 \tag{9}$$

$$2\lambda r + V \sum_i \sum_\sigma \langle f^+_{l\sigma} c_{il\sigma} + c^+_{il\sigma} f_{l\sigma} \rangle = 0. \tag{10}$$

The Green functions corresponding to the Hamiltonian (7) are

$$\begin{aligned} G^\sigma_{11}(\omega, k) &= [(\omega - E_{f\sigma})(\omega - \epsilon_{2k\sigma}) - r^2V^2]/A_\sigma \\ G^\sigma_{22}(\omega, k) &= [(\omega - E_{f\sigma})(\omega - \epsilon_{1k\sigma}) - r^2V^2]/A_\sigma \\ G^\sigma_{ff}(\omega, k) &= (\omega - \epsilon_{1k\sigma})(\omega - \epsilon_{2k\sigma})/A_\sigma \\ G^\sigma_{1f}(\omega, k) &= G^\sigma_{f1}(\omega, k) = rV(\omega - \epsilon_{2k\sigma})/A_\sigma \\ G^\sigma_{2f}(\omega, k) &= G^\sigma_{f2}(\omega, k) = rV(\omega - \epsilon_{1k\sigma})/A_\sigma \end{aligned} \tag{11}$$

where

$$A_\sigma = (\omega - E_{f\sigma})(\omega - \epsilon_{1k\sigma})(\omega - \epsilon_{2k\sigma}) - 2r^2V^2(\omega - \epsilon_{k\sigma}). \tag{12}$$

The quasi-particle spectrum is determined by

$$A_\sigma = 0. \tag{13}$$

It is easily seen from equations (12) and (13) that there are three quasi-particle bands $\omega_i(\epsilon_k)$ ($i = 1,2,3$) when $H = 0$. The reverse functions of $\omega_i(\epsilon_k)$ can be expressed analytically as

$$\begin{aligned}
\epsilon_{1k}(\omega) &= \omega - [1/(\omega - E_{\rm f})][r^2V^2 + \sqrt{r^4V^4 + \epsilon_0^2(\omega - E_{\rm f})^2}] \qquad \omega \in \omega_1 \\
\epsilon_{2k}(\omega) &= \omega - [1/(\omega - E_{\rm f})][r^2V^2 + \sqrt{r^4V^4 + \epsilon_0^2(\omega - E_{\rm f})^2}] \qquad \omega \in \omega_2 \\
\epsilon_{3k}(\omega) &= \omega - [1/(\omega - E_{\rm f})][r^2V^2 - \sqrt{r^4V^4 + \epsilon_0^2(\omega - E_{\rm f})^2}] \qquad \omega \in \omega_3 .
\end{aligned} \tag{14}$$

When a magnetic field is applied, each band is split by the Zeeman energy according to its spin orientation, and the DOS of electrons is

$$\rho_{\rm f(c)} = \sum_{i\sigma} \rho_{i{\rm f(c)}\sigma}(\omega) \tag{15}$$

where

$$\rho_{i{\rm f}\sigma}(\omega) = [r^2V^2/2D(\omega - E_{{\rm f}\sigma})^2][1 \pm r^2V^2/\sqrt{r^4V^4 + \epsilon_0^2(\omega - E_{{\rm f}\sigma})^2}] \qquad \omega \in \omega_{i\sigma} \tag{16}$$

and

$$\rho_{i{\rm c}\sigma} = 1/2D \qquad \omega \in \omega_{i\sigma}. \tag{17}$$

In equation (16), we use the + sign when $i = 1,2$ and the − sign when $i = 3$. In our model, we assume that there are one f electron and two conduction electrons on each site. Therefore we have another equation from which the chemical potential μ may be determined:

$$n_{\rm c} + n_{\rm f} = 3. \tag{18}$$

From self-consistent calculation of equations (9), (10) and (18), we can obtain the field-dependent parameters at a finite temperature. In the weak-field and low-temperature limit, we have

$$r^2(H,T) = r^2(0,T) - \{H^2/[1 + \rho_{\rm f}(\mu)]\}[\rho_{\rm f}'(\mu) + (\pi^2/6)\rho_{\rm f}'''(\mu)k_{\rm B}^2T^2] \tag{19}$$

and

$$\begin{aligned}
E_{\rm f}(H,T) - \mu(H,T) &= E_{\rm f}(0,T) - \mu(0,T) \\
&\quad + \{H^2/[1 + \rho_{\rm f}(\mu)]\}[\rho_{\rm f}'(\mu) + (\pi^2/6)\rho_{\rm f}'''(\mu)k_{\rm B}^2T^2]
\end{aligned} \tag{20}$$

where $\rho_{\rm f}'(\mu)$ and $\rho_{\rm f}'''(\mu)$ are the first and the third derivatives of the f DOS at the Fermi level.

In figure 1, we present the numerical results of the mean-field parameters r^2 and $E_{\rm F} - \mu$ in different magnetic fields at zero temperature. Here we take $\epsilon_0 = 0.9D$, $V^2 = 0.2D^2$, $E_0 = 2.2D$ and $D = 5000$ K. From equations (15) and (16), we find that in zero magnetic field the DOS of f electrons per spin is of a single-peak structure in the vicinity of the Fermi level. On one hand, when a magnetic field is applied, the band width for each spin is decreased owing to the reduction in r^2 with increasing H. On the other hand, because of the Zeeman splitting effect due to the magnetic field, the peak moves away from the Fermi surface, leading to a reduction in the electronic DOS at the Fermi level.

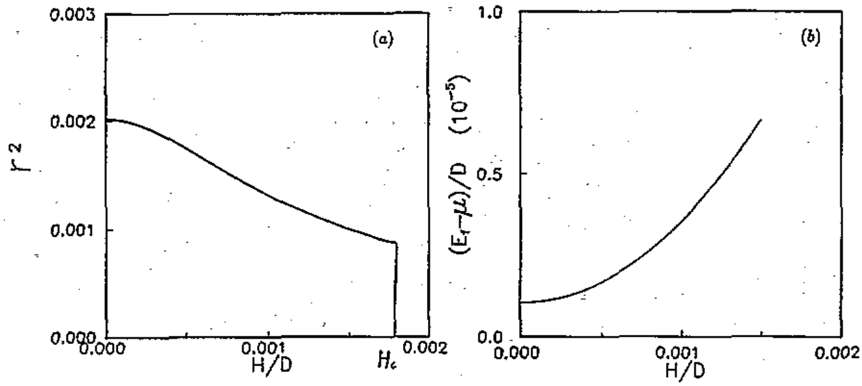

Figure 1. (*a*) Magnetic field dependence of the mean-field parameter $r^2(H)$ at zero temperature. (*b*) Magnetic field dependence of $E_f(H) - \mu(H)$ at zero temperature.

3. Thermodynamic properties at low temperatures in a non-zero magnetic field

3.1. The specific heat coefficient

A large specific heat is a common feature of the HFS, which shows the existence of a narrow band structure, and the study of specific heat is indispensable for probing the interaction between particles in the systems. In terms of the electronic DOS, the specific heat coefficient of the HFS in a magnetic field can be written as

$$\gamma(H,T) = \tfrac{1}{2}k_B^2\beta^3 \int_{-\infty}^{\infty} d\omega\, \omega^2[\rho_f(H,\omega) + \rho_c(H,\omega)]\, \mathrm{sech}^2\left(\frac{\beta(\omega-\mu)}{2}\right). \tag{21}$$

Since we have $\rho_f(\omega) \gg \rho_c(\omega)$ in the vicinity of the Fermi surface, the main contribution of $\gamma(H,T)$ comes from f electrons. By defining $\eta = \beta(\omega - \mu)$, where $\beta = 1/k_BT$, we obtain

$$\gamma(H,T) = \tfrac{1}{2}k_B^2 \int_{-\infty}^{\infty} d\eta\, \eta^2 \rho_f(H, \eta k_B T + \mu)\, \mathrm{sech}^2\left(\frac{\eta}{2}\right). \tag{22}$$

It is easy to understand that the relation between $\gamma(H,T)$ and T is totally determined by the shape of the DOS of f electrons near the Fermi surface. Because $\rho_f(H,\mu)$ reduces with increasing field, $\gamma(H, T = 0)$ reduces too, showing the suppression of heavy-fermion effects in the applied field. When the magnetic field is strong enough, two peaks of the f-electron DOS will emerge symmetrically on both sides of the chemical potential, resulting in a maximum $\gamma(H,T)$ at a finite temperature. The numerical results are presented in figure 2. In the experiment on $CeCu_6$ [17], it is found that the specific heat coefficient $\gamma_0(H = 0)$ in the absence of an external magnetic field is about 1.6 J mol^{-1} K^{-2} at $T = 0$ K. When a magnetic field of 5 T is applied, the zero-temperature specific heat coefficient $\gamma_0\,(H = 5\ \mathrm{T})$ is reduced to half the above zero-field value. If we take the effective magnetic moment $\mu_{\mathrm{eff}} = g\mu_B = \mu_B$ in the numerical calculation, it can be easily obtained that $H/D = 10^{-4}$ corresponds to a magnetic field of 0.75 T. Then our numerical results can be compared with the experimental measurements. Our calculation shows that the zero-temperature specific heat coefficient $\gamma_0(H = 0)$ in the absence of the magnetic field is about 2.8 J mol^{-1} K^{-2}, which is of the same order of magnitude as the experimental result in [17]. In the presence of the magnetic field, the zero-temperature specific heat coefficient γ_0 is reduced to its half-value at about 4.5 T. This is in quite good agreement with the experiment in [17]. It is also shown in figure 2 that for a definite external magnetic field a crossing between $\gamma(H, T)$ and $\gamma(H = 0, T)$ exists at T_{crossing}. When the temperature is below T_{crossing}, the specific heat is suppressed by the magnetic field, so that $\gamma(H, T) < \gamma(H = 0, T)$ for $T < T_{\text{crossing}}$. As the temperature increases to $T > T_{\text{crossing}}$, the non-zero-field specific heat is enhanced, i.e. $\gamma(H, T) > \gamma(H = 0, T)$ for $T > T_{\text{crossing}}$. As the magnetic field H increases, both T_{crossing} and T_{maximum} shift slowly towards higher temperatures. The above results are in qualitative coincidence with experiments [1, 7–10, 17].

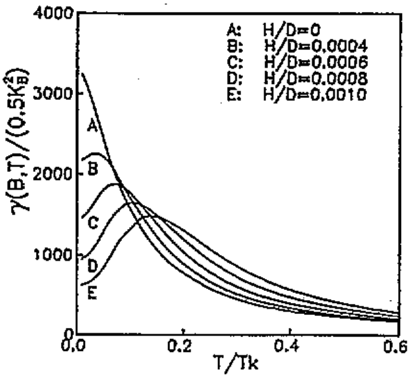

Figure 2. The specific heat coefficient $\gamma(H, T)$ in different magnetic fields.

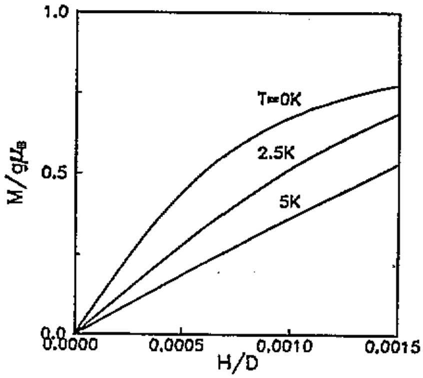

Figure 3. The variation in magnetization with magnetic field at various temperatures.

3.2. *Magnetization*

The magnetization of heavy-fermion metals in the presence of an external magnetic field is

$$M = g\mu_B(n_\downarrow - n_\uparrow) \tag{23}$$

where the direction of magnetization is the same as that of the applied magnetic field. As mentioned above, we consider only the contribution of f electrons, and equation (23) becomes

$$M = g\mu_B \int_{-\infty}^{\infty} d\omega\, f(\omega - \mu)[\rho_f^\downarrow(H, T, \omega) - \rho_f^\uparrow(H, T, \omega)] \tag{24}$$

where $f(\omega - \mu)$ is the Fermi function, and $\rho_f(\omega)$ may be determined from equations (15) and (16). In a non-zero magnetic field, the thermal fluctuation at finite temperatures will reduce the magnetization because the peaks of the f DOS shift away from the chemical potential with increasing H. Meanwhile, the narrowing of the band width at elevated temperatures will enhance the effect of the magnetic field. Since the magnitude of the Zeeman energy is only of the order of several kelvins, the former effect is more important, i.e. the magnetization will decrease at finite temperatures.

The numerical results obtained from equation (24) are sketched in figure 3, where we have calculated the variation in M with magnetic field at $T = 0$, 2.5 and 5 K. Obviously the susceptibility of heavy-fermion metals at low temperatures represented by the slope of the M–H curve is suppressed by the applied field, which is related to the previous result of the suppression of the specific heat coefficient in (1). These results can be compared with the experimental data in [9, 11, 12].

3.3. *Anomalous volume magnetostriction*

It is reported that the volume magnetostriction of these materials is anomalous at low temperatures (about 10^2–10^3 times larger than those of ordinary metals [13]). This phenomenon is difficult to explain in conventional theory, and thus a special mechanism should be used. Since $b^+(b)$ represents the production (annihilation) of a vacant site in the slave-boson theory, the quantity $\langle b^+b\rangle$ can be considered as the probability that a site is vacant. In the mean-field approximation, $\langle b^+b\rangle$ is replaced by r^2, and the probabilities of the f^0 and f^1 configurations are r^2 and $1-r^2$, respectively.

It is pointed out that the difference between the ionic radii of the rare-earth metal in various f configurations is much larger than those of transition or normal metals, and the anomalous volume change produced by an external field is mainly due to the change in the occupation probability of the different configurations [13, 14]. For Ce compounds, if we represent the cell volumes of f^0 and f^1 configurations as Ω_0 and Ω_1, respectively, the average volume per cell is $\Omega = r^2\Omega_0 + (1 - r^2)\Omega_1$ and the volume magnetostriction is

$$(\Delta\Omega/\Omega)_H = -[(\Omega_1 - \Omega_0)/\Omega](\Delta r^2)_H \simeq -[(\Omega_1 - \Omega_0)/\Omega_1](\Delta r^2)_H \tag{25}$$

where

$$(\Delta r^2)_H = r^2(H,T) - r^2(0,T). \tag{26}$$

Using equations (19) and (25), we find that at extremely low temperatures and in weak fields, the volume magnetostriction has the form

$$(\Delta\Omega/\Omega)_H = AH^2(1 - BT^2) \tag{27}$$

with

$$A = [(\Omega_1 - \Omega_0)/\Omega_0]\,\{\rho_f'(\mu)/[1 + \rho_f(\mu)]\} \tag{28}$$

$$B = -(\pi^2 k_B^2/6)[\rho_f'''(\mu)/\rho_f'(\mu)] \tag{29}$$

where A and B are both positive in our model. From equation (27) one can see that the volume magnetostriction is proportional to the square of the magnetic field as expected in a usual paramagnetic metal. However, owing to the large difference between the cell volumes of the two configurations, the volume magnetostriction is two to three orders of magnitude larger than that of a usual metal. In our calculation, we have taken $(\Omega_1 - \Omega_0)/\Omega_0 = 0.05$ as in [13]. It is found that, when the magnetic field $B = 2$ T and $T = 0$ K, the magnetostriction has the value 0.4×10^{-5}. This is in comparison with the experiment in [13], where the magnetostriction $\Delta V/V$ was found to be about 1×10^{-5} in $CeCu_6$ when $B = 2$ T and $T = 1.53$ K. Our results are in order-of-magnitude agreement. The factor $1 - BT^2$

in (27) indicates that the volume magnetostriction of the system has the feature of a Fermi liquid at very low temperatures. The numerical results are presented in figures 4 and 5. Figure 4 shows that the volume magnetostriction is a parabolic function of the magnetic field H at different temperatures. Figure 5 is the variation in volume magnetostriction with temperature in different fields, which shows that, in the temperature regime $0.05T_K < T < 0.4T_K$, the volume magneotostriction changes with temperature nearly in the form $1/(T+T_F)^2$. This result reproduces the scaling relation $\Delta\Omega/\Omega \propto [H/(T+T_F)]^2$ suggested by Zieglowski *et al* [13]. At extremely low temperatures ($T \ll T_K$), the volume magnetostriction is proportional to $1 - BT^2$ which can be considered as a feature of the Fermi liquid. Since the cell volume of the f^{14} ion is larger than that of the f^{13} ion (where f^{14} and f^{13} correspond to f^0 and f^1, respectively, owing to the electron–hole symmetry), the negative volume magnetostriction of Yb-based compounds can also be obtained in our theory.

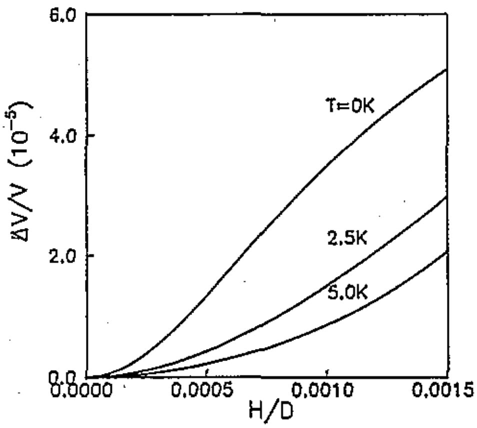

Figure 4. The magnetic field dependence of the volume magnetostriction at different temperatures.

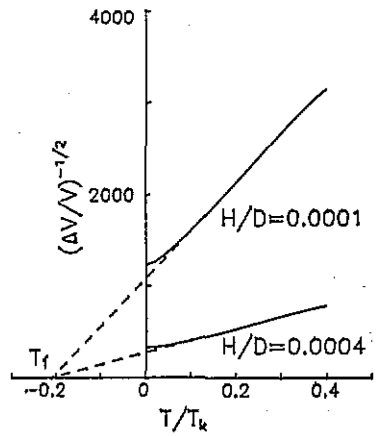

Figure 5. The temperature dependence of the volume magnetostriction in various magnetic fields.

4. Conclusions and discussion

From a two-conduction-band PAM, we have studied the thermodynamic properties of HFSs in a magnetic field. We find that the DOS at the Fermi surface is largely reduced in HFSs because of the Zeeman splitting of the quasi-particle bands. This is the reason for the suppression effect of the magnetic field on HFSs, which leads to reductions in the linear specific heat coefficient and the susceptibility at low temperatures in the presence of an external magnetic field. Since the occupation ratio of f configurations is changed by the applied field, and also the difference between the cell volumes of the two configurations (f^0 and f^1) is significant, a large volume magnetostriction can then be observed.

In the present paper, the influence of the magnetic field on the thermodynamic properties is studied in the scheme of the slave-boson mean-field theory. In the slave-boson theory of the Kondo lattice, the auxiliary boson field is introduced to describe the strong correlation between f electrons in the lattice Anderson Hamiltonian. The mean-field or the saddle-point approximation, which is exact in the large-degeneracy limit, involves some spurious results in the finite-degeneracy case. For example, an unphysical 'transition' appears at a temperature T_{MF}. Correspondingly, in the presence of the magnetic field, there is a sudden drop in the slave-boson mean-field parameter r^2 at a field $H = H_c$ as shown in figure 1(*a*). Therefore, our mean-field theory can be applied only to a relatively weak-field regime $H < H_c$. Nevertheless, our theory provides a good explanation of the thermodynamic properties in a relatively weak field.

Finally, the large DOS of quasi-particles of the HFS in the vicinity of the Fermi surface in the slave-boson mean-field theory is a result of the Kondo effect in the whole lattice, showing the many-body nature of the effective resonance between itinerant conduction electrons and localized f electrons. The quasi-particle DOS has a certain structure near the Fermi level and varies with the temperature and magnetic field. This description of the Fermi liquid with a large effective mass is valid for most of the thermodynamic properties in the HFS. However, to describe some other physical phenomena such as transport properties, we should go beyond the mean-field theory and take into account the many-body effects due to the fluctuations of the slave-boson field further.

Acknowledgment

This work was supported in part by the National Natural Science Foundation of China.

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