

Anomalously heavy fermions in YbPdSb

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A study of the specific heat of YbPdSb reveals that the electron component of the specific heat, C_e , divided by the temperature reaches values $C_e/T = 3.2 \text{ J}/(\text{mole} \cdot \text{K}^2)$ at $T = 0.5 \text{ K}$. Such values correspond to a record effective mass of delocalized quasiparticles for heavy-fermion systems. A study of the thermal expansion confirms that there is no magnetic transition down to 0.5 K . © 1994 American Institute of Physics.

Essentially all known heavy-fermion systems are based on compounds of Ce and U. An anomalously large contribution to the electron specific heat $\gamma = C_e/T$, on the order of $1 \text{ J}/(\text{mole} \cdot \text{K}^2)$, is reached not because of a magnetic transition but because of effective Kondo-scattering processes with a low Kondo temperature (T_K) which occur in a doublet spin-1/2 ground state.¹ One might also expect the realization of a “heavy-fermion” ground state for intermetallic compounds based on the Yb^{3+} ion ($4f^{13}$), which is a “hole” analog of $4f^1$ cerium. Until recently, the maximum value of γ has been less than $0.2 \text{ J}/(\text{mole} \cdot \text{K}^2)$ and has been observed in the compound YbCuAl (Ref. 1). Recently, Dhar *et al.*² reported seeing a heavy-fermion state in YbPdSb with γ exceeding $1 \text{ J}/(\text{mole} \cdot \text{K}^2)$, on the basis of measurements of the magnetic susceptibility and the specific heat at temperatures down to 2 K . This information, combined with the results of thermodynamic, transport, and thermoelectric studies,³ yielded an estimate of the Kondo temperature T_K : on the order of $5\text{--}7 \text{ K}$. With the help of the Kadowaki–Woods relation,⁴ a specific heat $\gamma \sim 1 \text{ J}/(\text{mole} \cdot \text{K}^2)$ was found from the magnitude of the Fermi-liquid coefficient in the electrical resistance for the interval $2 < T < 4 \text{ K}$. Further Mössbauer and thermodynamic studies carried out down to 0.1 K revealed a magnetic phase transition with $T_N = 1 \text{ K}$, with a spontaneous magnetic moment on the order of $1.3\mu_B$ per Yb^{3+} ion.⁵

In this letter we are reporting a study of the thermodynamic properties (specifically, the specific heat C and the thermal expansion coefficient α at $0.5 \text{ K} < T < 20 \text{ K}$) for a

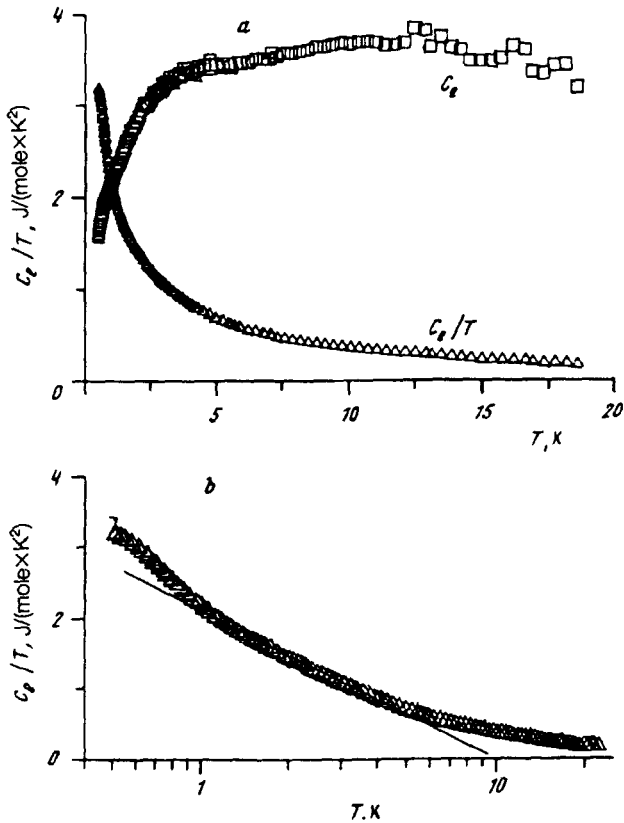


FIG. 1. Temperature dependence of the "electron" component of the specific heat, C_e , in the following coordinates: a— C_e versus T and C_e/T versus T ; b— C_e/T versus $\log T$.

new lot of YbPdSb samples. We have found that the electron specific heat γ reaches values of 3.2 J/(mole·K²) at $T=0.5$ K, without any indications of a magnetic transition. These values are evidence that we are seeing a record effective mass for delocalized quasiparticles for nonmagnetic heavy-fermion systems. A study of the thermal expansion confirms that there is no magnetic transition down to $T=0.5$ K.

The polycrystalline samples used in this study were prepared at the Physics Faculty of Tohoku University in Japan. The primary new and distinctive feature of the preparation was the arrangement of a preliminary Pd–Sb reaction in sealed-off quartz cells. Immediately thereafter, as in the earlier studies, we used a standard arc-melting furnace, followed by annealing at 1000° for a long time (48 h). An x-ray study confirmed the presence of a cubic crystal structure of the MgAgAs type. The measurement method⁶ and the method for preparing the samples⁷ are described in more detail elsewhere.

Figure 1 shows the temperature dependence of the "electron" component of the specific heat, $C_e = C(\text{YbPdSb}) - C(\text{LuPdSb})$, versus T (Fig. 1(a)) and also versus $\log T$ (Fig. 1(b)). The main distinctive features of these results are (a) the monotonic increase in the "heavy-fermion" contribution to the specific heat C_e/T without any hint of a magnetic phase transition down to $T=0.5$ K, and (b) the attainment, at $T=0.5$ K, of a record

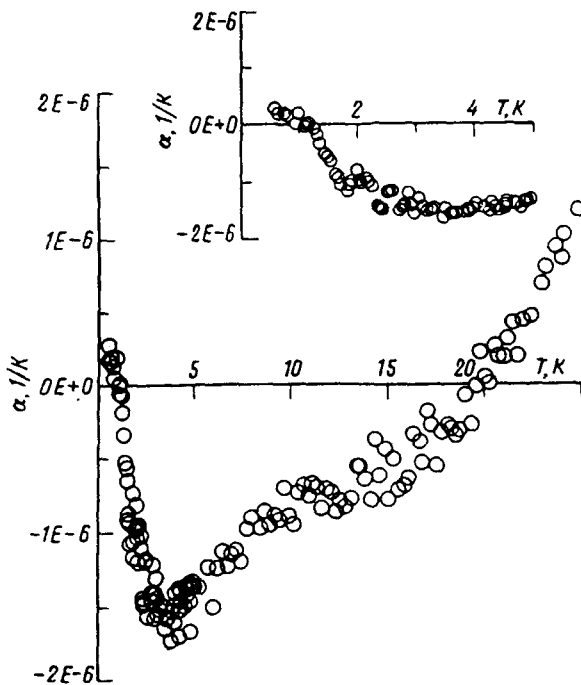


FIG. 2. Temperature dependence of the thermal expansion coefficient, $\alpha(T)$, of YbPdSb.

value of γ for heavy-fermion systems, more than $3 \text{ J}/(\text{mole} \cdot \text{K}^2)$. This value corresponds (in the model of free quasiparticles) to an effective mass on the order of 10^3 times the mass of a free electron. Curiously, a plot in a logarithmic temperature scale reveals a wide region ($1 < T < 5$ K) in which the electron contribution to the specific heat increases linearly (Fig. 1), followed by a deviation (of about 10%) from the straight line toward larger values of C_e/T at $T < 1$ K.

The temperature dependence of the thermal expansion coefficient, $\alpha(T)$, of YbPdSb (Fig. 2) shows a change in the sign of α , from positive to negative, at $T < 20$ K. There is a minimum near $T = 4$ K, and there is a quasilinear decrease in the absolute value of α at $T < 4$ K. At $T < 1.5$ K we find a stable tendency toward another change in the sign of α , to positive values, at $T < 1$ K. There are no indications of the magnetic transition which has been seen previously⁵ near $T = 1.3$ K.

The ground state of the free Yb^{3+} ion, with $J = 7/2$, is split by the cubic-symmetry field into two doublets (Γ_6 and Γ_7) and a quadruplet (Γ_8) (Ref. 8). A model has been proposed to explain the behavior of the specific heat at $T > 2$ K, where the results found for "magnetically ordered" YbPdSb were found to be approximately the same as those shown in Fig. 1 for "nonmagnetic" YbPdSb. In this model,⁵ a Kondo effect involving the Γ_8 quartet, with a Kondo temperature T_K on the order of 7 K, occurs at low temperatures, while the high-temperature maximum of C_e , at a temperature the order of 30 K, results from Kondo scattering by levels split by the crystal field. A study of inelastic neutron scattering has shown⁹ that the level nearest the ground state splits by about $\Delta_{\text{CF}} = 6-7$

meV \sim 70 K. The change in the sign of the thermal expansion coefficient at $T < 20$ K is apparently also due to a transition to the $T \ll \Delta_{CF}$ regime.

We now consider the reasons for the magnetic instability and anomalous large values of γ for YbPdSb. We start from the possibility of a partial interchange of the Yb and Sb atoms (as occurs in most intermetallic compounds with a crystal structure of the MgAgAs type¹⁰). The result of such a “perturbation” in the symmetry of the crystal lattice is then a lifting of the degeneracy from the quadruplet in the ground state. The characteristic splitting energy Δ must not exceed a few kelvins, so that the relation $\Delta < T_K$ can be satisfied and so that the specific heat for $T > \Delta$ in the triplet can correspondingly be modelled by a quadruplet. A study of the temperature dependence of the frequency of magnetic fluctuations by the method of μ_{SR} spectroscopy has shown that nonmagnetic YbPdSb has an activation law at $0.3 < T < 5$ K with a gap Δ on the order of 1–2 K (Ref. 11). In this situation, the quantity Δ , which depends on the degree of order in the sample, may determine the ground state in a critical way. For more disordered samples, with higher values of Δ (this category probably includes the samples which have been studied previously⁵), the time scale of the fluctuations of the magnetic moment near $T = T_N = 1$ K may be large enough that it does not prevent a transition to a magnetically ordered state. At the same time, the increased frequency of fluctuations of the magnetic moment near $T = T_N$ for the “more ordered” YbPdSb studied in the present experiments may suppress magnetism.

In summary, a nonmagnetic doublet ground state characterized by a low Kondo temperature $T_K^1 \sim 1$ K, gigantic values of γ , and correspondingly gigantic values of the effective mass of delocalized quasiparticles forms in the YbPdSb system at $T < 1$ K. The final formation of the heavy-quasiparticle band due to the onset of “coherence effects” should occur at T well below T_K^1 . We plan to carry out some further, detailed studies in this temperature range.

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