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Kondo behavior of U in CaB_6

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Abstract

Replacing U for Ca in semiconducting CaB_6 at a few atomic percent level induces metallic behavior and Kondo-type phenomena at low temperatures, a rather unusual feature for U impurities in metallic hosts. For $Ca_{0.992}U_{0.008}B_6$, the resistance minimum occurs at $T \approx 17\,K$. The subsequent characteristic logarithmic increase of the resistivity with decreasing temperature merges into a T^2 dependence below 0.8 K. Correspondingly, the low-temperature specific heat indicates the formation of a ground state with a strongly enhanced electronic specific heat parameter.

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Keywords: Kondo effect; Actinide; Resistivity

A well-known dilute Kondo system is (La,-Ce)B_6, with a Ce-content on the few percent level, which exhibits all the traditional Kondo anomalies, as for example in the temperature dependence of the resistivity [1] or the specific heat [2]. By replacing La by U instead of Ce, no corresponding Kondo anomalies are observed [3]. It is most likely that this is caused by a broadening of the U 5f electron states via hybridization with the conduction band states, leading to a loss of their localization; distinctly different from the well-localized 4f electrons on the Ce ions. The situation seems to be different if U replaces cations in a low-carrier density matrix, such as CaB_6, with conduction electron densities of the order of 10^{-4} per unit cell [4].

Fig. 1 shows the temperature dependence of the resistivity $\rho(T)$ of $Ca_{0.992}U_{0.008}B_6$ in zero applied magnetic field and at temperatures between 0.4 and 300 K. The concentration of U-moments is established by measurements of the susceptibility at high temperatures and of the specific heat between 0.4 and 8 K (data not shown here). The decrease of ρ with decreasing T, reaches a minimum at $T = T_{min} \approx 17\,K$ followed by a logarithmic increase at even lower temperatures.

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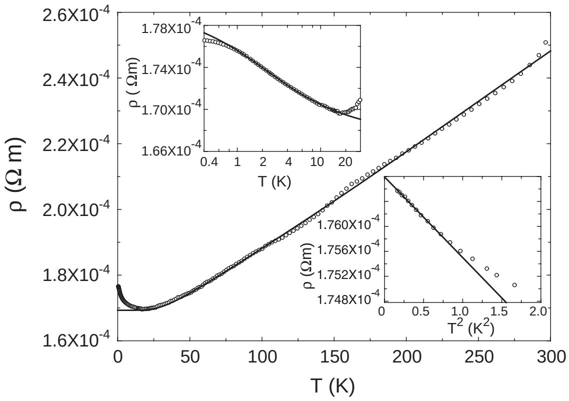

Fig. 1. Resistivity in zero magnetic field for $Ca_{0.992}U_{0.008}B_6$. The solid line in the main frame displays the calculation explained in the text. The left inset shows ρ for $T<30$ K and the fit according to Eq. (1). The right inset emphasizes the T^2 dependence of ρ at $T<0.8$ K.

At $T>30$ K, $\rho(T)$ is well described by a model where the conduction electrons are scattered by disordered magnetic moments, lattice vibrations and lattice defects [5]. The solid line in the main frame of Fig. 1 represents the corresponding fit assuming a constant spin-disorder and a defect resistivity of $\rho_d = 1.68 \times 10^{-4}\,\Omega$m.

Below $T \approx 17$ K, the resistivity increases again with decreasing T, exhibiting a nearly logarithmic dependence for $3\,\text{K} < T < 10\,\text{K}$. In non-magnetic host materials with magnetic impurities, such a T-dependence is attributed to virtual bound states of the conduction electrons at the magnetic site. According to Hamann [6], the magnetic impurity scattering leads to a resistivity

$$\rho_{\rm imp} = \frac{\rho_0}{2}\left\{1 - \frac{\ln(T/T_{\rm K})}{[(\ln(T/T_{\rm K}))^2 + \pi^2 S(S+1)]^{1/2}}\right\}, \tag{1}$$

where $T_{\rm K}$ is the Kondo temperature and S is the spin of the magnetic impurity. The upper inset in Fig. 1 shows $\rho(T)$ for $T<30$ K. The fit according to Eq. (1), displayed as the solid line, yields $T_{\rm K} = 2.0 \pm 0.1$ K and $S = \frac{1}{2}$, indicating a doublet ground state of the U 5f electron multiplet. Hence, U most likely adopts the $5f^3$ configuration and the ground state is the magnetic Γ_6-doublet [7].

It is expected that at $T \ll T_{\rm K}$, the Kondo system exhibits Fermi liquid behavior [8], thus implying that $\rho_{\rm imp}$ for the n-channel Kondo model with $n = 2S$ is [9]

$$\rho_{\rm imp}(T) = \rho_{\rm imp}(0)\left\{1 - \left(\frac{\pi^2}{12T_{\rm L}}\right)^2 (4n+5)T^2\right\}. \tag{2}$$

In our case, $n = 1$ and the corresponding fit results in $T_{\rm L} \sim T_{\rm K}\pi \approx 6.0$ K. This corresponds to $T_{\rm K} \approx 1.9$ K, in good agreement with the above-mentioned value. Unfortunately, $\rho_{\rm imp}$ depends strongly on the magnitude of the residual resistivity, and hence this analysis is less reliable.

Further support for the Kondo behavior of U in CaB_6 was obtained from measurements of the magnetic susceptibility, the magnetization as a function of external fields, and the specific heat at low temperatures and in external magnetic fields. At temperatures below 8 K, the measured specific heat $C_p(T)$ for $Ca_{0.992}U_{0.008}B_6$ is dominated by its magnetic contribution $C_m(T)$, which is obtained by subtracting the background lattice contribution from the measured $C_p(T)$ data. The maximum of the bell-shaped curve of $C_m(T)$ is, as expected, shifted in magnitude and temperature upon application of external magnetic fields. Our analysis of these data is based on the resonance level (RL) model of Schotte and Schotte [10], which assumes a Lorentzian shape of the electronic density of states, given by $D(\varepsilon) = \Delta/\pi(\varepsilon^2 + \Delta^2)$. We obtain consistent fits to the experimental curves $C_m(T,H)$ by postulating $S = \frac{1}{2}$, and setting $\Delta/k_{\rm B} = 1.6$ K and the g-factor $= 1.7$. This modification of the g-factor is equivalent to a reduced effective magnetic moment $\mu_{\rm eff}$ of the U ions, due to the magnetic screening by the conduction electrons [10,11]. Inserting the value for Δ, for $\varepsilon = 0$, yields the density of states $D(E_{\rm F}) = (\pi\Delta)^{-1}$ and consequently a specific heat parameter $\gamma = 92\,\text{mJ}/(\text{K}^2\,\text{mol U})$. For comparison, we calculated $\gamma_{\rm ord}$ for the case of an ordinary conduction band with a quadratic dispersion relation and populated by one electron per U-ion. The effective mass is taken as $m^* = 0.28m_0$, the same as for itinerant charge carriers in CaB_6 [12]. With these assumptions, we obtain $\gamma_{\rm ord} = 0.19\,\text{mJ}/(\text{K}^2\,\text{mol U})$, implying that the Kondo interaction leads to an enhancement of the electronic specific heat parameter by a factor of 480.

The model of Schotte and Schotte was also employed for the interpretation of the low-temperature data for $\chi(T)$ and the magnetization $M(H)$. In both cases, the parameters that were obtained from fitting the $C_m(T,H)$ data, gave equally good fits to these magnetic quantities.

Most earlier experimental attempts to demonstrate the onset of the Kondo effect induced by U impurities were either unsuccessful or did not provide conclusive results. Our data, however, indicate that the insertion of U at the low % level into a matrix with a low conduction electron concentration, such as CaB_6, favors the stability of the 5f local moment on the U site, leading to a well-developed Kondo effect at low temperatures. The complete experimental evidence for the Kondo behavior of this compound will be presented in detail elsewhere [3].

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