# Resonance near the Fermi energy in a Kondo lattice

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An anomalous increase (by a factor of 40–60) in the Hall coefficient $R_H$ has been observed in the $CeCu_2Si_2$ superconducting Kondo system as the temperature is lowered from 60 to 4 K. This effect is evidence of the formation of a narrow resonance near the Fermi level in $CeCu_2Si_2$.

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The transition from the magnetic regime ($T \gtrsim T_K$, where $T_K$ is the Kondo temperature) to the nonmagnetic singlet regime ($T \ll T_K$) has recently been attributed[1–3] in the theory of Kondo lattices to a radical restructuring of the energy spectrum near the Fermi energy $\epsilon_F$. Specifically, a narrow Kondo resonance appears near $\epsilon_F$, and its amplitude grows (while its width decreases) as the temperature is lowered from $T \gtrsim T_K$ to $T \ll T_K$. The appearance of a Kondo resonance may be accompanied, as a result of the coherence of the Kondo screening of the magnetic moments, by the formation of a correlation gap at the Fermi level for certain directions in the Brillouin zone.[1] To the best of our knowledge, however, these effects have not yet been observed.

In this letter we report an effort to experimentally test the theories of Refs. 1–3. We studied the temperature dependence of the Hall coefficient $R_H(T)$, that of the resistivity $\rho(T)$, and that of the magnetic susceptibility $\chi(T)$ for $CeCu_2Si_2$ single crystals and polycrystalline $Ce_XLa_{1-X}Cu_2Si_2$ samples ($0 < X \leqslant 1$) over broad ranges of the pressure ($p \leqslant 12$ kbar), the temperature ($0.05\ K \leqslant T \leqslant 100\ K$), and the magnetic field ($H \leqslant 40$ kOe).

The temperature dependence of the magnetic susceptibility of the $Ce_XLa_{1-X}Cu_2Si_2$ solid solutions, measured in a field $H \simeq 0.5$ Oe with a SQUID, has a local maximum (Fig. 1) which probably corresponds to a transition at $T = T_{SG}$ from a paramagnetic state to a spin glass. The temperature $T_{SG}$ (see the inset in Fig. 1) varies nonmonotonically with increasing concentration ($X$) of the magnetic component in

  

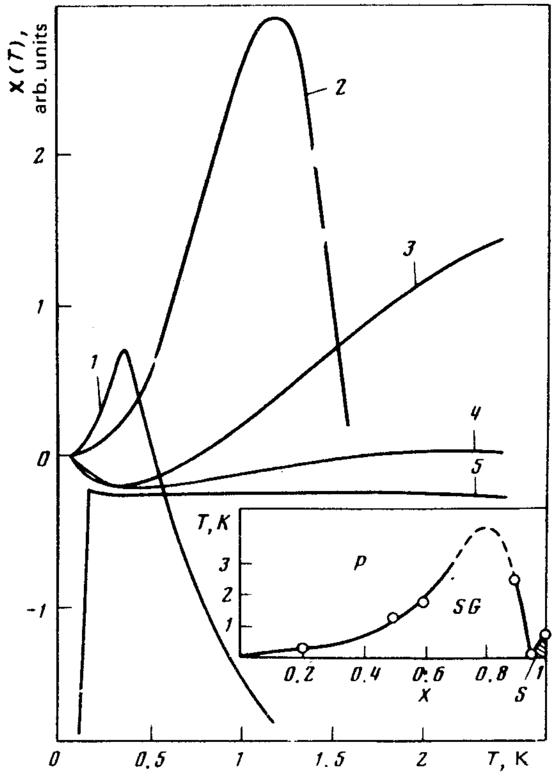


FIG. 1. Temperature dependence of the magnetic susceptibility, $\chi(T)$, of $Ce_XLa_{1-X}Cu_2Si_2$ at various concentrations $X$: 1—0.2; 2—0.5; 3—0.7; 4—0.9; 5—1.0. Inset: Phase diagram of the magnetic properties of $Ce_XLa_{1-X}Cu_2Si_2$. *P*—Paramagnetic state; SG—spin glass; *S*—superconductor.

$Ce_XLa_{1-X}Cu_2Si_2$. The amplitude of the local maximum at $X \geqslant 0.5$ falls off with increasing $X$, and the maximum becomes much broader. Near $X = 1$ the $Ce_XLa_{1-X}Cu_2Si_2$ polycrystalline samples become superconductors (curve 5 on Fig. 1).

As the temperature of the $CeCu_2Si_2$ single crystals ($X = 1$) is reduced from $\sim 60$ to $\sim 4$ K, we observe a dramatic increase (by a factor of 40–60) in the Hall coefficient $R_H$. This increase corresponds to a decrease in the free-carrier concentration from $n_H \simeq 1.5 \times 10^{22}$ cm$^{-3}$ at $T = 60$ K to $3 \times 10^{20}$ cm$^{-3}$ at $T = 1$ K (Fig. 2a). In a temperature interval below a certain characteristic $T_0$, which depends on the pressure $p$, the Hall coefficient remains essentially constant, while the resistivity $\rho$ decreases. The value of $T_0$ is approximately the same as the temperature ($T^{\rho}_{max}$) at which a maximum is observed on the $\rho(T)$ curve (Fig. 2b).

The anisotropy (at $\mathbf{H} \| \mathbf{J}$ and $\mathbf{H} \perp \mathbf{J}$) of the derivative of the upper critical field, $dH_{C2}/dT(T = T_C)$, in $CeCu_2Si_2$ decreases during hydrostatic compression (Fig. 3).

The observed increase in $R_H$ (Fig. 2) as the temperature is lowered from $T \gtrsim T_K$ to $T \ll T_K$ (in $CeCu_2Si_2$, $T_K \simeq 30$ K) can be attributed to the formation of a Kondo resonance corresponding to the appearance in the Kondo lattice of heavy fermions, with a concentration which is zero at $T \gg T_K$ and maximal at $T \ll T_K$ (see the inset in

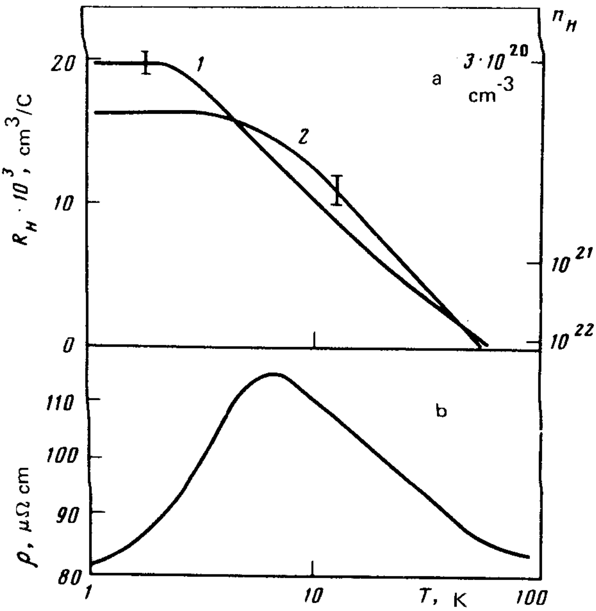


FIG. 2. Temperature dependence of the Hall coefficient $R_H$ (a) and that of the resistivity $\rho(b)$ for $CeCu_2Si_2$. The scale at the upper right shows the Hall concentration calculated from $R_H$. Curves 1 and 2 were measured at pressures of 3.8 and 6.0 kbar, respectively. The $\rho(T)$ curve was measured at 3.8 kbar.

Fig. 3). In the present study we determine $R_H$ from the field dependence of the Hall voltage $U(H)$ at $H \leqslant 40$ kOe. At such fields, the heavy fermions (with masses $m^* \sim 200m_0$; Ref. 4) are negligible. The measured Hall coefficient (Fig. 2a) thus does not give us the total concentration of electrons in the zone but only the concentration of free electrons—those which are not "stuck" at Kondo centers. If we assume that the decrease in the free-carrier concentration is caused exclusively by the redistribution of these carriers at the Kondo resonance, then the density of heavy fermions in $CeCu_2Si_2$ increases from $n \sim 10^{20}$ cm$^{-3}$ at $T \sim 60$ K at $n \sim 10^{22}$ cm$^{-3}$ at $T \leqslant 4$ K.

From this point of view, the data suggest that the giant electron specific heat $\gamma$ in Kondo lattices [$\gamma \simeq 1000$ mJ/(mole K$^2$) (Ref. 4) for $CeCu_2Si_2$ and $\gamma \simeq 1600$ mJ/(mole K$^2$) (Ref. 5) for $CeAl_3$; for normal metals, by way of comparison, the typical values are $\gamma \simeq 1$–$10$ mJ/(mole K$^2$)] results from the formation near $\epsilon_F$ of a narrow ($\sim kT_K$), large-amplitude Kondo resonance (see the inset in Fig. 3), rather than from the $f$ level, which lies 2 eV below the Fermi level in these systems.[6]

According to Ref. 1, the formation of a Kondo resonance and the appearance of a correlation gap at low temperatures can occur simultaneously. In this connection we do not rule out the possibility that the decrease in $\rho(T)$ is a consequence of a decrease in the effectiveness of the scattering of mobile carriers by the reduced magnetic moments of cerium as a result of the apperance of coherence in the Kondo scattering.

The anomalously high value of the derivative $dH_{C2}/dT(T = T_C)$ (Ref. 8; see Fig. 3 of the present paper) also implies a high state density $g(\epsilon)$ near $\epsilon_F$. The experimental value of $dH_{C2}/dT(T = T_C)$, we might note, is approximately the same as an estimate

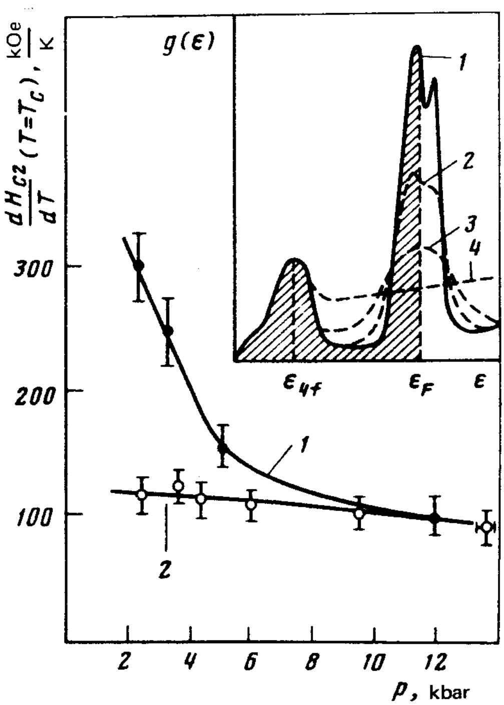


FIG. 3. Pressure dependence of the derivation of the upper critical field, $dH_{C2}/dT(T=T_C)$, for $CeCu_2Si_2$. 1—**H**‖**J**; 2—**H**⊥**J**. The measurement current $J$ is flowing in the direction perpendicular to the crystallographic $C$ axis in $CeCu_2Si_2$. Inset: Change in the structure of the energy spectrum of the Kondo lattice as the temperature is reduced from $T \gg T_K$ (4) through $T \sim T_K$ (3) and $T < T_K$ (2) to $T \ll T_K$ (1) ($T_K$ is the Kondo temperature).

found from the values of $\rho$ (Ref. 8) and $\gamma$ (at $T = 0.6$ K; Ref. 4); $dH_{C2}/dT(T=T_C) \simeq 140$ kOe/K.

The compound $CeCu_2Si_2$ differs from other known Kondo lattices involving cerium in that the valence of cerium is $\nu_{Ce} \simeq 3.08$ (Ref. 7; for an ideal Kondo lattice we would have $\nu_{Ce} = 3$), so that this compound is similar to intermediate-valence compounds. Hydrostatic compression triggers a transition from a Kondo lattice to a variable-valence compound by increasing the deviation of $\nu_{Ce}$ from an integer value. Consequently, the pressure-induced decrease in the concentration of heavy fermions can be attributed to a lowering and broadening of the Kondo resonance in the course of the transition. The decrease in the state density of heavy fermions is the reason for the pressure-induced decrease in the average value of $dH_{C2}/dt(T=T_C)$, which is proportional to $g(\epsilon_F)$.

The rapid growth of the Hall coefficient (Fig. 2a) is evidence that $CeCu_2Si_2$ has a Kondo lattice in which the relative concentration of free electrons (per magnetic center) is approximately one. This conclusion is supported by the nontrivial dependence of the magnetic properties of the solid solutions $Ce_XLa_{1-X}Cu_2Si_2$ (Fig. 1), in which there is a gradual transition from a Kondo impurity ($X \ll 1$) to a Kondo lattice ($X \sim 1$), which also erases the plateau on the $\rho(T)$ curves which is characteristic of an isolated Kondo impurity and gives rise to a maximum on the temperature dependence of the resistivity at $X \geqslant 0.5$ in the series $Ce_X La_{1-X} Cu_2 Si_2$ (Ref. 9).

As the concentration $X$ is raised above 0.5, the appearance of the Kondo resonance at $X \to 1$ causes a decrease in the relative concentration of free carriers (per magnetic center), which in turn weakens the RKKI interaction of the Kondo-reduced magnetic moment of cerium and reduces $T_{SG}$ (see the inset in Fig. 1). At the same time, the concentration of heavy fermions increases as $X \to 1$, giving rise to superconductivity in $CeCu_2Si_2$.

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