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Experimental discrimination of coherent and incoherent behavior in heavy-fermion materials

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The onset of heavy-fermion coherent-ground-state behavior is studied in $CeCu_6$ by Hall-effect and magnetoresistance measurements. $CeCu_6$ is an ideal system for this study since the fermions are extremely heavy and the system does not become either magnetic or superconducting. The strong temperature dependence of the Hall effect sets the scale for the high-temperature, single-Kondo-impurity, incoherent regime, a broad transition region, and the very-low-temperature coherent regime. In contrast to the resistivity, which shows a gradual transition to the coherent state, the Hall effect shows a rather sharp feature at the onset of coherence.

Some rare-earth and actinide intermetallic compounds show greatly enhanced electronic specific heats and Pauli susceptibilities at low temperatures.[1-4] They are called heavy-fermion systems because these enhanced properties would be characteristic of metals with Fermi-degeneracy temperatures as low as 10 K, and effective electron masses up to 1000 times the bare electron mass. At low temperature, heavy-fermion systems may be antiferromagnetic, superconducting, or normal-metal Pauli paramagnetic. An important unsolved theoretical problem is the nature of the normal coherent ground state. At high temperatures the resistivity of heavy-fermion systems is large. In many cases the resistivity increases approximately as log T, with decreasing temperature, as would a collection of independent Kondo impurities. However, as the temperature tends to zero, the resistivity of the normal heavy-fermion systems tends smoothly to a very small value. This behavior is in contrast to that of a system of independent Kondo impurities which would keep its maximum resistivity as the temperature went to zero. The vanishing of the resistivity at low temperature means that the scattering from the magnetic ions is coherent.

The purpose of this work is to study the development of the low-temperature coherent state and the transition from the high-temperature incoherent single-Kondo-impurity regime. The Hall effect is found to be extremely sensitive to this change. In $CeCu_6$ a large negative peak in the Hall constant, at several hundred millikelvin, sets the temperature scale for coherence. This peak distinguishes the low-temperature coherent regime from a transition region and from the high-temperature incoherent regime. $CeCu_6$ is an ideal choice for this study since it is a Pauli paramagnet and has extremely heavy fermions at low temperatures. Of the normal heavy-fermion systems, only $CeAl_3$ has as heavy fermions. In the magnetic and superconducting systems, the coherent state may be approached but the low-temperature limit may not be attained.

$CeCu_6$ is orthorhombic at high temperature and monoclinic below 165 K.[5] The orthorhombic c axis has the largest susceptibility at all temperatures below 300 K.[6] One of our samples was a single-crystalline bar in which current passed in the b direction, while the magnetic field was in the c direction. The other was a crystalline plate with a few small-angle grain boundaries. The c axis was nearly in the plane of the plate while the a and b axes were about 45° out of the plane. The magnetic field was applied perpendicular to the plate. Since the van der Pauw method was used, the resistivity for the plate is an average of the three directions. The plate is the same sample as that used, over a more limited temperature range, in our initial study.[7]

Hall and resistivity data are shown in Fig. 1. Both samples show the same qualitative behavior. On cooling from room temperature, the resistivity rises as would that for a collection of independent Kondo centers scattering incoherently. However, below about 15 K the resistivity decreases, indicating the breakdown of independent behavior. At still lower temperatures the resistivity smoothly approaches its small residual value. The Hall constant has a maximum near the resistivity maximum. It also has a relatively sharp negative peak at 200 mK for the plate and 350 mK for the bar. Since the Hall data are monotonic and smooth below the peak, a low-temperature region is defined. The coherent normal ground-state behavior is observed only below the negative Hall extremum. The extrema in the Hall data clearly divide the temperature scale into three regimes: coherent, transition, and incoherent.

Resistivity measurements on $CeCu_6$ at very low temperatures have shown a linear temperature dependence between 400 mK and 1 K and a T^2 dependence below about 100 mK.[8-10] This T^2 dependence was interpreted as due to electron-electron scattering, indicative of the limiting Fermi-liquid behavior of the coherent heavy-fermion ground state. The pronounced negative peak in the Hall constant correlates with the far more subtle change in power law of the resistivity and together mark the coherent-scattering regime. Hall measurements on the heavy-fermion compounds $CeRu_2Si_2$, UPt_3, and UAl_2,[11] do not show a negative extremum at low temperatures, but

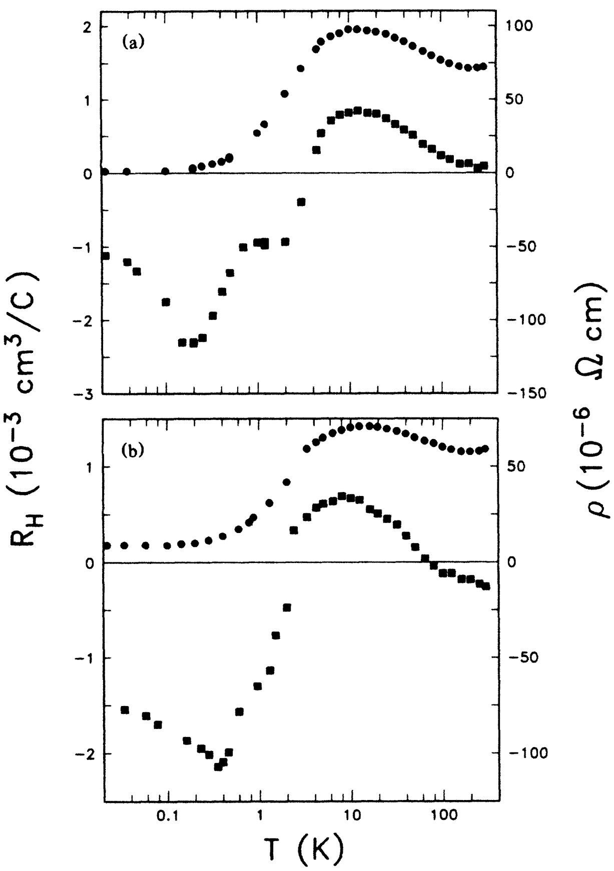


FIG. 1. Resistivity, dots, and Hall constant, squares, for (a) the plate and (b) the bar. The sharp negative extremum in the Hall effect marks the onset of the coherent state.

there is a change in behavior marking the coherence. The Hall constant has a strong temperature dependence at moderate temperatures but a weak temperature dependence at the lowest temperatures, where the dependence of the resistivity becomes T^2.

For $CeCu_6$ the temperature dependence of the Hall constant in the region of 100 mK and below should be characteristic of the coherent regime. In both our samples, with increasing temperature, the Hall constant increases negatively with approximately linear temperature dependence. The magnitude of the effect is very large, comparable to the size of the high-temperature peak. This behavior is not the same as that observed for $CeRu_2Si_2$, UPt_3, and UAl_2.[11] In these materials, at the lowest temperatures, the Hall constant has an extremely small temperature dependence, compared to the size of the high-temperature behavior. For $CeRu_2Si_2$ and UPt_3, which have positive Hall constants at all temperatures, the Hall constant increases weakly as $T^{1.4}$ and T^3, respectively. For UAl_2, which has a negative Hall constant at the lowest temperature, it increases weakly positively as T^3.

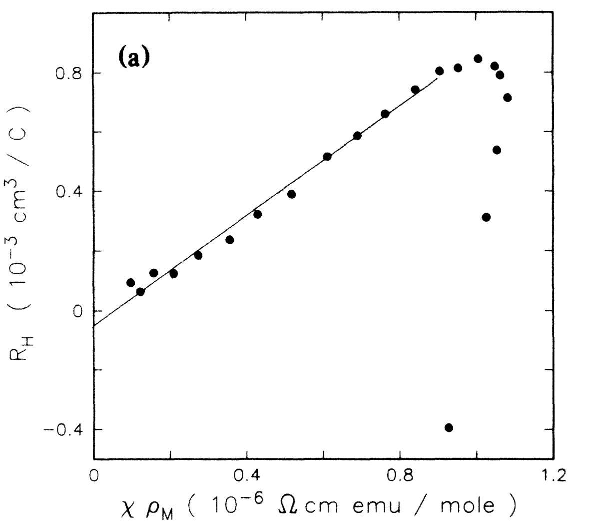
The limiting low-temperature value for the Hall effect may have contributions from the ordinary Lorentz-force term R_0 and from extraordinary scattering. Since the electronic structure renormalizes between the high- and low-temperature regimes, one would not expect the R_0 values determined at high temperature to be the correct values for the coherent regime. Apparently the existence of a coherent regime defined by the Hall effect is common to all of the systems studied, but the actual behavior is specific to the particular system. In the case of $CeCu_6$, the onset of the coherent state is found to be abrupt.

In the higher-temperature incoherent regime, the Hall effect has been measured in the heavy-fermion compounds, $CeCu_2Si_2$,[12,13] $CeAl_3$,[14] and UBe_{13}.[15] In all cases, the Hall constant was positive and increased with decreasing temperature. The intermediate-temperature transition region has been studied in $CeCu_6$ by us[7] and in UPt_3, UAl_2, $CeRu_2Si_2$, $CeAl_3$,[11] UPt_3,[16] U_2Zn_{17},[17] by others. In all cases, a positive peak was observed in the Hall constant. Since below this temperature, the resistivity dropped rapidly, the Hall peak and subsequent decrease were ascribed to the beginning of the transition to the coherent regime. This point of view is strongly supported by studies of $CePd_3$, a slightly-heavy-fermion compound.[13,18] In the pure material, there is a peak in the Hall constant and then at lower temperature a decrease and finally a change in sign from positive to negative. When either Y is substituted for Ce, or Ag for Pd, the resistivity remains high at low temperature and the decrease and sign change of the Hall constant are absent. This important result shows that, at least for this slightly-heavy-fermion system, the decrease in Hall effect is not a single-ion effect, but rather depends on the coherence of the system.

There is no theoretical model for the Hall effect in the coherent regime. There are theories for skew scattering from single magnetic impurities which may well be applicable in the high-temperature regime.[11,18-20] According to these theories the contribution to the Hall effect from single-ion skew scattering should be proportional to the product of the susceptibility χ and the magnetic contribution to the resistivity, ρ_M. Quite generally, for paramagnets with susceptibility $\chi \ll 1$ the Hall constant is $R_H = R_0 + 4\pi\chi R_S$, where R_0 is the ordinary Lorentz-force term and R_S is the extraordinary magnetic term. This expression can be rewritten as

$$R_H = R_0 + P\chi\rho_M$$

for $P = 4\pi R_S/\rho_M$. If skew scattering by a magnetic impurity is the origin of R_S, then since the number of scattering events is proportional to the magnetic resistivity and the average scattering angle is given by χH, the coefficient P will be temperature independent. If the electronic structure changes, P may change also. The magnetic resistivity is found by subtracting from the total resistivity the phonon contribution, which is taken as the resistivity of $LaCu_6$.[21] The magnetic resistivity, shown in Fig. 2(b), behaves approximately as ln T at high temperature, consistent with Kondo-impurity scattering. The Hall constant is plotted versus $\chi\rho_M$ in Fig. 2(a), for the $CeCu_6$ plate. The susceptibility was measured perpendicular to the plate in the same direction as in the Hall measurement. The linear dependence, shown by the solid line, shows the validity of the incoherent-skew-scattering picture in the high-temperature regime.[22] In Fig. 2(b), the fit determined in Fig. 2(a), extrapolated to lower temperature, is shown as a function of temperature and compared with the



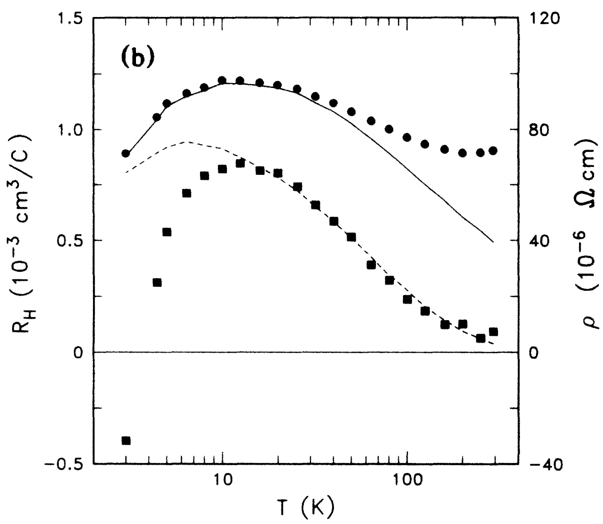


FIG. 2. (a) Hall constant vs the product of the susceptibility and the magnetic resistivity, $\chi\rho_M$ for the plate, (b) resistivity, dots; magnetic resistivity, upper line; Hall constant, squares; $\chi\rho_M$ fit, lower dashed line.

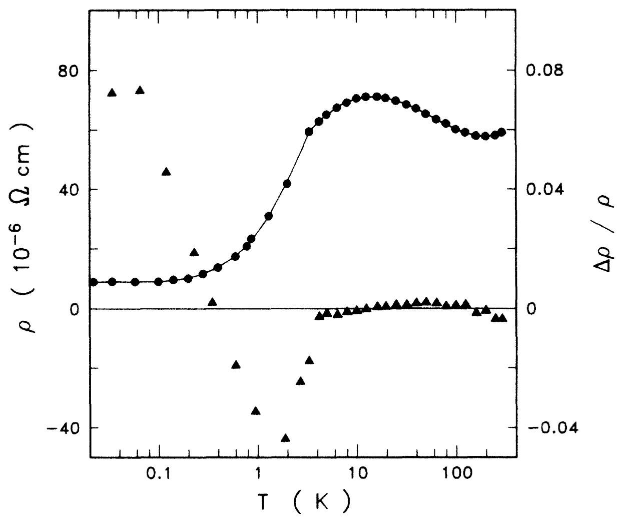


FIG. 3. Resistivity, dots, and magnetoresistance at 1 T, triangles, for the bar.

Hall constant. The fit fails in the neighborhood of the maximum and below. This deviation, found in both samples, is a good indicator of the breakdown of the single-ion, incoherent, high-temperature Kondo regime.

The transition region from incoherent to coherent scattering and from a strongly positive to a strongly negative Hall effect is complicated. It has been shown theoretically[10,20] that even for a single impurity model there can be a maximum and a sign change as the temperature falls below the Kondo temperature. However, the experimental evidence from $CePd_3$, if applicable to heavier-fermion compounds, supports the view that the sign change results from coherence. Furthermore, $CeCu_6$ samples with large residual resistivities (little coherence) due to impurities or partial substitution of La for Ce, did not show a maximum or sign change in the Hall effect.[5,23] These features have been observed in a sample with a small residual resistivity.[24] Specifically, it has been argued that in the intermediate-temperature regime, the Rudderman-Kittel-Kasuya-Yosida interaction between Kondo impurities can reduce scattering and cause a resistivity maximum.[25]

The magnetoresistance for the $CeCu_6$ bar, with a magnetic field of 1 T in the easy (c) direction, is shown in Fig. 3. These data, and field sweeps not shown, are in general agreement with previous studies of the magnetoresistance below 4 K.[26,27] The negative magnetoresistance in the neighborhood of 2 K has been shown to follow a single-ion Kondo model.[27,28] The lowest-temperature positive magnetoresistance is characteristic of the coherent state, without Kondo scattering. This positive magnetoresistance, expected generally for metals, occurs only below 350 mK, the temperature of the Hall extremum. On heating above 4 K, the magnetoresistance is relatively small.[29]

The extremely large electronic specific heats observed in heavy-fermion compounds at low temperature do not require coherence. In the case of $CePd_3$, substitution of 0.12 Ag for Pd increased the linear specific heat C/T from a modest 50 to a large 450 mJ/mole K^2, even though coherence was lost.[30] For $CeCu_6$, C/T is very large, about 1500 mJ/mole K^2, at low temperature both for a strongly coherent sample with very low residual resistivity and for a partially coherent sample with relatively large residual resistivity.[10,31] Coherence may cause structure in the electronic density of states, resulting in structure in the electronic specific heat but with little change in the magnitude.[32]

In conclusion, strong features in the Hall effect clearly set the temperature scale for the heavy-fermion system $CeCu_6$. The low-temperature coherent ground state is found only below a few hundred millikelvin. The onset of the coherent state is very sharp when viewed by the Hall effect. The transition between the pure coherent state and the single-ion high-temperature state is very broad, occurring over two decades in temperature. The high-temperature behavior is consistent with that of a collection of independent Kondo impurities.

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