Crossover from non-Fermi-liquid to Fermi-liquid behavior in the magnetoresistivity of $U_{0.9}Th_{0.1}Be_{13}$

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We have measured the temperature T and magnetic field H dependence of the electrical resistivity ρ of $U_{0.9}Th_{0.1}Be_{13}$ in the ranges $0.08\ \mathrm{K} \leqslant T \leqslant 10\ \mathrm{K}$ and $0\ \mathrm{kOe} \leqslant H \leqslant 80\ \mathrm{kOe}$. In zero applied magnetic field below 0.7 K, the resistivity displays a linear temperature dependence, characteristic of non-Fermi-liquid behavior, down to 0.23 K, where it then saturates as T^2, consistent with Fermi-liquid behavior, down to 0.08 K, the lowest temperature attained in the experiment. Magnetic fields up to 40 kOe have practically no effect on the electrical resistivity above 1 K, but dramatically influence the low temperature $\rho(T)$ behavior below 0.7 K. At temperatures below 0.7 K, an anomalously large positive magnetoresistivity is accompanied by a rapid expansion of the temperature region where Fermi-liquid behavior is observed. [S0163-1829(97)06542-9]

I. INTRODUCTION

A growing class of f-electron materials with anomalous physical properties at low temperatures has attracted considerable interest during the last several years. These materials are Ce and U intermetallics which, with a few exceptions, have been doped with a nonmagnetic element. Most prominent among these anomalous properties are a linearly saturating electrical resistivity, $\rho(T) \sim 1 - a(T/T_0)$, a logarithmically diverging electronic specific heat coefficient, $\gamma \equiv C(T)/T \sim (-1/T_0)\ln(T/bT_0)$, where $C(T)$ is the electronic specific heat, and a square-root divergence in the magnetic susceptibility, $\chi(T) \sim 1 - c(T/T_0)^{1/2}$, for temperatures $T \ll T_0$, where T_0 is a characteristic temperature that can often be identified as a Kondo temperature.[1,2]

These anomalous properties are commonly referred to as non-Fermi-liquid (NFL) properties since they do not conform to Fermi-liquid (FL) theory where $\rho(T) \sim 1 - a(T/T_F)^2$ and $C(T)/T \sim \chi(T) \sim$ const for $T \ll T_F$, where T_F is the effective Fermi temperature. The NFL properties observed are generally associated with a quantum critical point at $T = 0$ K, produced, for example, by an unconventional moment screening process, such as a multichannel Kondo effect,[3,4] or by fluctuations of an order parameter in the vicinity of a second-order phase transition.[5] While none of these models has been able to predict all of the physical properties observed, recent work has suggested that $U_{0.9}Th_{0.1}Be_{13}$, derived from the heavy fermion superconductor UBe_{13} by substituting uranium with nonmagnetic thorium, displays low temperature physical properties that are consistent with a two-channel, spin-1/2, electric quadrupolar Kondo effect (QKE).[6] In particular, the specific heat[7] and magnetic susceptibility[8] of $U_{0.9}Th_{0.1}Be_{13}$ for temperatures $0.8\ \mathrm{K} \leqslant T \leqslant 6$ K and $1.7\ \mathrm{K} \leqslant T \leqslant 10$ K, respectively, display temperature dependences which are consistent with the NFL power laws described above, and which are also predicted for the QKE.[9] Moreover, nonlinear susceptibility[10] and Hall effect measurements[11] also support the possibility of a quadrupolar Kondo ground state at least above 1.5 K in $U_{0.9}Th_{0.1}Be_{13}$. In the QKE model, the temperature dependence of the electrical resistivity is given[12] by $\rho(T) \sim 1 - a(T/T_0)^{1/2}$, a result that is at odds with the linear temperature dependence observed in virtually all of the f-electron systems found to exhibit NFL behavior. Recently, a $T^{1/2}$ dependence of $\rho(T)$ was observed[13] for $U_{0.9}Th_{0.1}Be_{13}$ in the temperature range $0.45\ \mathrm{K} \leqslant T \leqslant 6$ K. However, since the measurements did not extend below 0.45 K, it was not determined how low in temperature the $T^{1/2}$ dependence of $\rho(T)$ persists. If, in fact, the QKE is responsible for the NFL behavior of $U_{0.9}Th_{0.1}Be_{13}$, we might expect the resistivity to deviate from the $T^{1/2}$ dependence at low enough temperatures, since the QKE model suggests that at some finite temperature, the non-Fermi-liquid character of the QKE ground state may be suppressed by a cooperative Jahn-Teller (JT) transition.[4] In addition, the application of a magnetic field should result in a restoration of FL behavior below a characteristic temperature $T = T^{\star}$ with a concomitant change in the temperature dependence of the electrical resistivity from $T^{1/2}$ to T^2.[9,14] The applicability of the QKE as well as other models in describing the physical properties of $U_{0.9}Th_{0.1}Be_{13}$ can be explored further by studying the electrical resistivity at even lower temperatures.

In the present work, we report a detailed study of the electrical resistivity of $U_{0.9}Th_{0.1}Be_{13}$ down to millikelvin

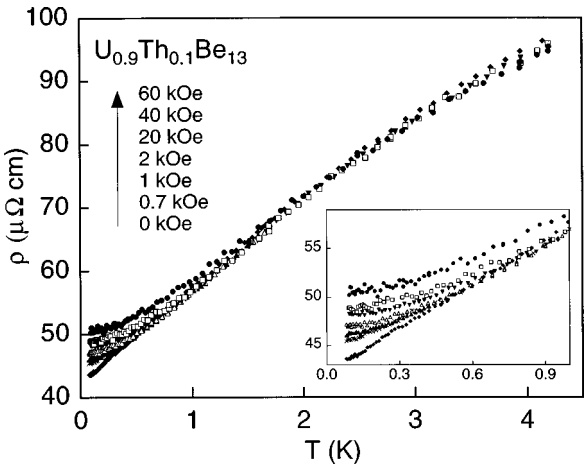

FIG. 1. Electrical resistivity ρ of $U_{0.9}Th_{0.1}Be_{13}$ vs temperature T below 4 K. The inset shows an enlarged view of the data for $T<1$ K.

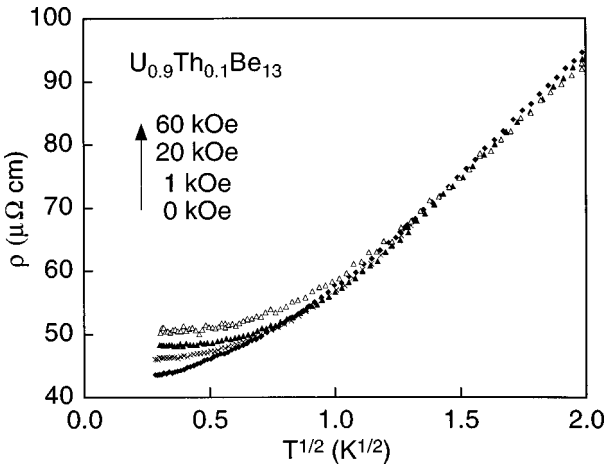

FIG. 2. Electrical resistivity ρ of $U_{0.9}Th_{0.1}Be_{13}$ vs square root of temperature $T^{1/2}$ between 0.08 K and 4 K in applied magnetic fields of 0, 1, 20, and 60 kOe from bottom to top.

temperatures and under applied magnetic fields up to 60 kOe. Our measurements show that non-Fermi-liquid behavior persists down to $T\sim 0.23$ K in zero field. An applied magnetic field dramatically affects the low temperature part of $\rho(T)$ by rapidly expanding the temperature interval where FL behavior occurs. The temperature $T^{\star}$ where the crossover from NFL to FL behavior takes place is field dependent, and for temperatures $T<T^{\star}$, a positive magnetoresistance is observed. The field dependence of the crossover could be used to distinguish between alternative theoretical models for non-Fermi-liquid behavior, but at present, none of the models predict the dependence seen in our measurements.

II. EXPERIMENTAL DETAILS

Details of the polycrystalline sample preparation and determination of the Th content were described elsewhere.[10] The electrical resistivity measurements were carried out in a transverse geometry using two cryostats: an SHE ^{3}He-^{4}He dilution refrigerator equipped with magnetic fields $H\leqslant 60$ kOe in the temperature interval 0.08 K$\leqslant T\leqslant 4$ K, and a Quantum Design PPMS, using the P400 resistivity option (a 7.5 Hz square wave) with magnetic fields $H\leqslant 80$ kOe for the interval 1.9 K $\leqslant T\leqslant 10$ K. A Linear Research LR 700 low-dissipation four-wire ac resistance bridge operating at a frequency of 16 Hz was used for the low temperature ($T<2$ K) measurements.

III. RESULTS

The electrical resistivity ρ of $U_{0.9}Th_{0.1}Be_{13}$ for 0.08 K $\leqslant T\leqslant 4$ K is plotted vs T in Fig. 1 and vs $\sqrt{T}$ in Fig. 2. Above approximately 0.7 K, $\rho(T)$ may be described by the following behavior: $\rho\approx\rho_0+B\sqrt{T}$ with $B\approx 38$ $\mu\Omega$ cm $K^{-1/2}$ and $\rho_0\approx 20$ $\mu\Omega$ cm. The values of both parameters, B and ρ_0, are in good agreement with those reported earlier.[13] Between ~ 0.23 K and ~ 0.7 K, the $\rho(T)$ data in zero field are consistent with a linear temperature dependence. The magnetoresistivity of $U_{0.9}Th_{0.1}Be_{13}$ was measured above 1.9 K and is in good agreement with previous isothermal dc measurements of the magnetoresistance for temperatures above 1.7 K.[13]

At low temperatures, $T<0.7$ K, a very different behavior of the magnetoresistivity of $U_{0.9}Th_{0.1}Be_{13}$ is found. This is illustrated in Fig. 3, in which the magnetoresistivity $\Delta\rho(H)/\rho(0)$, where $\Delta\rho(H)=\rho(H)-\rho(0)$, of $U_{0.9}Th_{0.1}Be_{13}$ is plotted vs T between 0.08 K and 2 K for selected applied fields between 1 kOe and 60 kOe. The magnetoresistivity, while small for $T>1$ K, shows anomalously large positive values as $T\to 0$ with $\Delta\rho(60\ \text{kOe})/\rho(0)\approx 0.16$ at $T=0.1$ K.

To examine the mechanism thought to be responsible for such a dramatic transformation of the magnetoresistivity of $U_{0.9}Th_{0.1}Be_{13}$, we plot the electrical resistivity as a function of temperature squared in Fig. 4. The zero field electrical resistivity is nonlinear in this coordinate down to approximately 0.23 K. Application of a magnetic field leads to the rapid expansion of the temperature interval where the resistivity is linear against T^2. For clarity, only data corresponding to two different applied fields are shown in Fig. 4. The

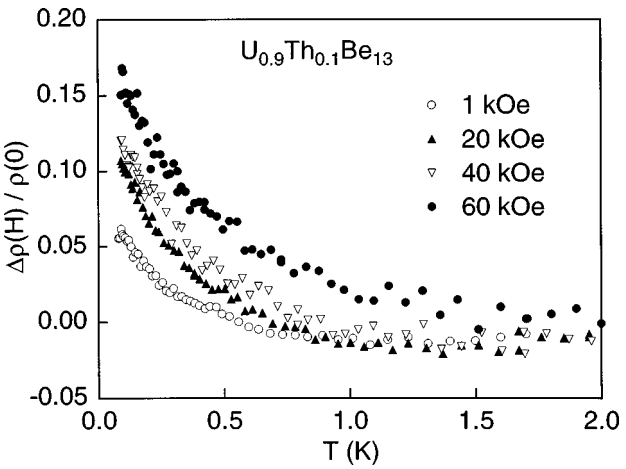

FIG. 3. Magnetoresistivity $\Delta\rho(H)/\rho(0)$ of $U_{0.9}Th_{0.1}Be_{13}$ vs temperature T down to 0.08 K in applied magnetic fields of 1, 20, 40, and 60 kOe.

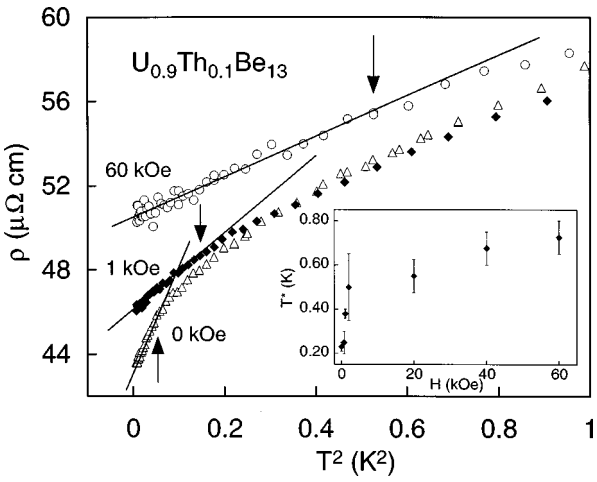

FIG. 4. Electrical resistivity ρ of $U_{0.9}Th_{0.1}Be_{13}$ vs temperature squared T^2 in applied magnetic fields of 0, 1, and 60 kOe below 1 K. The lines correspond to fits of the data, described in the text. The inset shows the field dependence of the characteristic Fermi-liquid temperature $T^\star$, indicated by arrows and explained in the text.

inset of Fig. 4 shows the magnetic field dependence of the upper limit of that range which may roughly be identified as a characteristic temperature $T^\star$ corresponding to an onset of the Fermi-liquid behavior. This characteristic temperature was determined from a linear least squares fit of the data. For each applied field, the fit was made starting with data from the very lowest temperatures and then extending the range of the fit to higher temperatures in the following manner. To test the goodness of the fit, the reduced chi square

$$\chi_\nu^2=\frac{1}{N-2}\sum_{i=1}^{N}\frac{[y_i-y(x_i)]^2}{\sigma_i^2} \tag{1}$$

was calculated.[15] In Eq. (1), N is the number of points being fit, y_i is the measured value of the resistance at the ith temperature, $y(x_i)$ is the value of the resistance predicted from the linear fit at the ith temperature, and σ_i is the standard deviation of the measured value of the resistance at the ith temperature. Next, an additional point was added to the fit and χ_ν^2 was recalculated. This procedure was continued until the fit included the entire data set for one field. Finally, $T^\star$ was determined as the temperature at which χ_ν^2 was a local minimum. The error bars associated with the inset to Fig. 4 reflect the uncertainty in choosing the location of the minimum.

IV. DISCUSSION

First, we note that in the region 0.7 K $\leqslant T \leqslant$ 6 K, where $\rho \sim T^{1/2}$, the resistivity is particularly insensitive to the application of a magnetic field. Both the $T^{1/2}$ temperature dependence and the extremely small magnetoresistance are consistent with the QKE model. In contrast, the results for $T<0.7$ K are not easily explained by a quadrupolar Kondo model. One important result of our measurements is the existence of a regime of linear temperature dependence in the resistivity at low temperatures. This is not entirely unexpected, since this type of low temperature behavior of $\rho(T)$ is displayed by a large number of other f-electron systems.[1,2] The main result of our work is the characterization of the crossover, indicated by $T^\star$, from non-Fermi-liquid to Fermi-liquid behavior at low temperatures. We find this crossover particularly interesting because the field dependence of $T^\star$ can be predicted theoretically, and thus the experimental results can be used to discriminate between competing explanations for the non-Fermi-liquid properties. Another informative result of our measurements is the large positive magnetoresistance seen at low temperatures. This large magnetoresistance is also potentially useful in distinguishing between applicable theories and, in this case, seems to indicate that the QKE alone is inadequate in describing the behavior of $U_{0.9}Th_{0.1}Be_{13}$ for $T<0.7$ K.

Similar behavior of the magnetoresistance has been observed in the $CeCu_{6-x}Au_x$ system. The resistivity of $CeCu_{5.9}Au_{0.1}$ varies linearly with T in zero field, but clearly displays T^2 behavior in applied magnetic fields.[16,17] For low fields, the crossover from NFL to FL behavior inferred from $\rho(T)$ measurements qualitatively agrees with a corresponding crossover determined from specific heat measurements in applied fields. An analysis of the specific heat measurements yielded a crossover temperature that varied roughly linearly with the applied field.[17] The magnetoresistance of $CeCu_{5.9}Au_{0.1}$ at low temperatures is positive in all applied fields, yet the magnitude of the magnetoresistance at 30 kOe is larger than the magnitude at 60 kOe, which is different than the monotonic increase of the magnetoresistivity with increasing field that we observe in the $U_{0.9}Th_{0.1}Be_{13}$ system. The non-Fermi-liquid behavior in $CeCu_{5.9}Au_{0.1}$ is attributed to the presence of antiferromagnetic ordering at $T=0$ K, as demonstrated by the detailed study of the thermal, magnetic, and transport properties of this system.[16] In fact, the QKE is ruled out as a description of the $CeCu_{6-x}Au_x$ system due to the incompatability of orthorhombic symmetry with the QKE model.[18] Thus, while the $\rho(T)$ behavior of $CeCu_{5.9}Au_{0.1}$ and $U_{0.9}Th_{0.1}Be_{13}$ are similar, different underlying mechanisms would appear to be responsible for the NFL behavior at low temperatures in these two systems.

We initiated this investigation to determine whether the low temperature magnetoresistivity of $U_{0.9}Th_{0.1}Be_{13}$ is consistent with the QKE in light of the evidence[6] for a QKE in this material. The QKE model makes specific predictions for the temperature dependence of the resistivity and the manner in which the system crosses over to FL behavior under the application of a magnetic field. In cubic symmetry, the QKE is expected to cross over from NFL to FL behavior in one of two ways. In the first scenario, an applied magnetic field acts as a ''channel field,'' which couples to the channel indices of the conduction electrons and lifts the degeneracy of the exchange coupling through splitting of the excited Γ_7 doublet.[9] According to Cox,[9] the exchange integrals for the different conduction channels should be split by an amount $\Delta J \propto H$, where H is the magnetic field strength. Assuming weak initial coupling and exact symmetry of the exchange integrals corresponding to each scattering channel, the crossover to FL behavior will then occur at a temperature $T_{ch} \approx (\Delta J)^2/T_K$. Hence, we expect the crossover temperature to have a quadratic field dependence.[3,9] The second manner in which FL behavior could manifest itself is by the application of a

uniaxial stress, which would be classified as a ''spin field,'' since it couples linearly to the impurity spin. The crossover temperature under the application of a spin field is also expected to follow a quadratic field dependence.[14] The experimentally obtained dependence of $T^{\star}$ on H, shown in the inset of Fig. 4, is quite different from the above quadratic form.

An additional complication in the QKE model arises when a strong enough applied magnetic field creates a uniaxial stress by magnetostriction. In this case, we expect a crossover between channel field physics at low fields, and spin field physics at high fields. An estimate of the channel field-spin field crossover point for the parent compound UBe_{13} was made by Cox,[14] who equated the exchange splitting of the channel states with the magnetostriction-induced splitting of the quadrupolar doublet to arrive at a crossover field of ≈ 40 kOe. It is possible that the rapid increase of $T^{\star}$ with H at low fields is due to a different mechanism than the moderate increase seen for 2 kOe $\leqslant H \leqslant 60$ kOe. However, if this is due to a channel field-spin field crossover, one would need to supply the reasoning why the crossover would occur at a field an order of magnitude smaller than the field postulated for the parent compound. In addition, considerably more data at different magnetic fields would be needed to determine if an H^2 dependence is reasonable, especially at high fields. If an H^2 dependence was not recovered at high fields, this could still be consistent with the view that the $U_{0.9}Th_{0.1}Be_{13}$ system is in the strongly coupled regime as evidenced by the positive slope of the $\rho(T)$ curves at low temperatures,[8] and hence the quadratic field dependences derived may not apply. Further theoretical work needs to be done to determine if the quadratic field dependences of the ''channel'' and ''spin'' field crossovers persist in the strong coupling limit.

Another possible explanation for the NFL behavior observed in f-electron materials is based on fluctuations of an order parameter above a $T=0$ K second-order phase transition. It is interesting to note that $U_{0.9}Th_{0.1}Be_{13}$ is near the Th composition where the unusual superconductivity observed in $U_{1-x}Th_xBe_{13}$ disappears.[19,20] Hence, critical fluctuations above a $T=0$ K superconducting phase transition could be responsible, at least in part, for the NFL behavior observed below 0.7 K. However, previous measurements of the specific heat[7] and magnetization[8] down to 0.3 K and 1.7 K, respectively, do not show any indication of the presence of superconducting fluctuations. Detailed studies of the thermodynamic properties at temperatures in the millikelvin range as well as studies of the ground-state properties of $U_{1-x}Th_xBe_{13}$ for $x<0.1$ need to be made in order to explore this possibility further.

In addition to the models mentioned above, several other explanations for NFL behavior in f-electron materials have been advanced, including, for example, disorder driven behavior[21–23] and an electronic polaron model.[24] The Kondo disorder model suggests that NFL behavior reflects a disordered material containing microscopic regions with different Kondo temperatures within which conventional (single channel local Fermi-liquid) Kondo physics takes place. The NFL properties arise from a superposition of the properties from the different regions with different Kondo temperatures. Since the behavior can be traced to conventional Kondo physics, we would expect that the magnetoresistance would be similar to that of a typical Kondo alloy; i.e., large and negative. Instead, we observe a large positive magnetoresistance at low temperatures. A detailed comparison of the Kondo disorder model and the properties of the $U_{0.9}Th_{0.1}Be_{13}$ system needs to be carried out to determine whether this model is viable. The electronic polaron model is an extension of the electronic polaron theory for heavy fermion systems which includes elastic scattering of the broad band electrons due to randomness inherent in the alloy. At this time, no comparison has been made between the properties of the electronic polaron model and our results for $U_{0.9}Th_{0.1}Be_{13}$.

V. CONCLUSION

In conclusion, we report the observation of a field-induced transformation of the resistivity from non-Fermi-liquid to Fermi-liquid behavior in $U_{0.9}Th_{0.1}Be_{13}$. In zero applied magnetic field, the electrical resistivity shows linear behavior between ~0.23 K and ~0.7 K. Application of a magnetic field has practically no effect on the electrical resistivity above 1 K, but dramatically affects $\rho(T)$ below 0.7 K, giving rise to a large, positive magnetoresistivity and a rapid expansion of the temperature interval where Fermi-liquid behavior is observed. This behavior cannot be described well by any of the theoretical models proposed for the non-Fermi-liquid f-electron materials at this time. The results described herein will be useful in the development of a consistent description of anomalous physical properties observed at low temperatures in this unique class of strongly correlated f-electron materials.

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