Title

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HALL EFFECT, MAGNETORESISTANCE, AND CRITICAL FIELDS OF UBe$_{13}$ THIN FILMS

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We report the first Hall effect measurements on polycrystalline thin films of the heavy fermion superconductor UBe$_{13}$. Measurements have been extended to temperatures well below previous studies on single crystals. We find no sign change of the Hall coefficient when cooling through T_c in contrast to the earlier study. We also observe a "shorting" of the Hall voltage below T_c. As the field is swept up, a p-type Hall signal is observed above a threshold magnetic-field which we define as H_{c2}. The slope of the phase boundary defined by these values agrees well with that as defined by resistivity measurements. Below $T = 0.2$ K, the magnetoresistance and Hall signal have very little temperature or field dependence above H_{c2}. The resistivity as a function of field may be fit using a spin-1/2 Coqblin–Schreiffer model.

INTRODUCTION

UBe$_{13}$ is one of the most extensively researched systems in the family of heavy fermion materials. The present work extends Hall effect measurements on single crystal samples of UBe$_{13}$ to thin films. Investigators have concluded from ultrasonic [2], specific heat [3], and magnetization [4] studies that there is substantial evidence for unconventional superconductivity in these materials, including a non-trivial form for the order parameter [5]. Uranium based heavy fermion systems behave as dilute systems of localized magnetic moments at high temperatures. This is indicated by magnetic susceptibility measurements revealing a Curie–Weiss like temperature dependence [6]. Carriers scatter incoherently at these temperatures and transport properties are fit quite well by single impurity models [7]. Through a carrier mediated process, the lattice of "magnetic-impurities" behave collectively at temperatures below T_{coh} (2.4 K for UBe$_{13}$). A peak in the resistivity [8] and the Hall

coefficient [9], followed by a rapid decrease in both quantities, as well as a sharp increase in C/T [10] are signatures of the onset of the coherent region. This collective regime is physically inconsistent with the single ion picture. As a result, at temperatures below T_{coh}, the temperature dependence of the Hall coefficient is not very well understood [11, 12]. The temperature dependence of the magnetoresistance and magnetization yield inconsistent results in the coherent regime. While the magnetoresistance [13, 14] data is fit well with a Coqblin–Schreiffer $j = 1/2$ model [15], the magnetization is not [13]. The purpose of this work is to determine the transport properties of UBe$_{13}$ thin films as a function of temperature and magnetic-field.

Previous experimental work focussed in the coherent regime shows qualitatively similar results for several different heavy fermion materials. URu$_2$Si$_2$, which is superconducting and antiferromagnetic in its ground state, shows a temperature dependent Hall coefficient similar to those of UPt$_3$ [16] and UBe$_{13}$ [9], which have superconducting ground states only. Perhaps the most striking result is that heavy fermion materials which show no ordered ground state (to 20 mK) have similar $R_H(T)$. Although the qualitative

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features for the systems mentioned above are similar, they differ quantitatively. The temperatures at which the features occur as well as the signs and magnitudes of $R_H(t)$ vary. Each system shows an anomalously large p-type (two to three orders of magnitude larger than expected corresponding to 100–1000 carriers per unit cell) Hall coefficient at the onset of coherence and a sharp reduction below this temperature. However it is the region at temperatures below this sharp reduction where there exists a relatively temperature independent Hall coefficient and where a sign change sometimes occurs. For example, UAl$_2$ changes sign around 20 K and has a residual negative R_H, while CeAl$_3$ never changes sign [16].

Aliev et al. [1] have observed a sign change of R_H in UBe$_{13}$ below a temperature which corresponds to the superconducting critical temperature. They also note a "shorting" of the Hall voltage at small magnetic-fields. At larger magnetic-fields there exists a Hall signal, the threshold of which they define as H_{c2}. We extend these measurements to lower temperatures to investigate this definition of the upper critical field.

EXPERIMENTAL

In order to enhance the Hall signal by increasing the current density without complicating the measurement with contact heating problems, we have made measurements on 3000 Å thin films of the heavy fermion UBe$_{13}$. The films were fabricated using a d.c. sputtering technique with a bulk UBe$_{13}$ sample as a target. The films were sputtered on to single crystal sapphire substrates which were heated to 800°C during deposition. The deposition rate was 15–20 Å per minute. X-ray diffraction data indicates that the films are polycrystalline UBe$_{13}$ with only a uranium impurity phase. All physical properties of the films correspond to those of bulk samples with the one notable exception being the absence of a resistivity peak at $T = 2.4$ K [17]. A Hall probe configuration was scribed on the film using a carbide tool. Gold pads were evaporated on the films to insure low resistance contacts. Thin copper leads were then attached to the pads with silver epoxy. Typical contact resistances were less than one ohm. All measurements were made in a top-loading ^{3}He–^{4}He dilution refrigerator at the Francis Bitter National Magnet Laboratory. The dilution refrigerator was situated with either an 8 T superconducting magnet or a 20 T Bitter resistive high-field magnet. The samples were mounted on a sample holder that was placed directly in the mixing chamber. Temperature was monitored with a calibrated 220 Ω speer resistive thermometer which was also placed directly in the mixture. Considerable care

was taken when changing the temperature of the mixture to allow the entire contents of the mixing chamber to reach thermal equilibrium. Temperature sweeps were made recalibrating the thermometer to account for magnetoresistive effects. All measurements were taken using a four probe a.c. technique at a frequency of 40 Hz. Most measurements were made at a current density of 4.25 A cm^{-2}, while a check with a current density of 0.425 A cm^{-2} made no significant difference except for signal to noise ratio.

RESULTS

Figure 1 shows the low temperature resistivity of a film as a function of magnetic-field and temperature. The "foot" on the transition has a field dependence and is believed to be associated with weak Josephson coupling of the superconducting grains in the film. The magnetic-field dependence of the foot behaves roughly as a thermally activated process. Junctions, such as those occurring between superconducting grains, typically have critical currents which are orders of magnitude smaller than the intragrain regions. We have made I–V measurements on the films at several different temperatures and applied fields. These results, in addition to the activated behavior, support the above conjecture. Other possibilities for the occurrence of the "foot" include pressure effects due to substrate-film incompatibility or a small impurity phase leading to a two-step transition (a two-step transition is often observed when Josephson effects are present). Pressure has been shown to
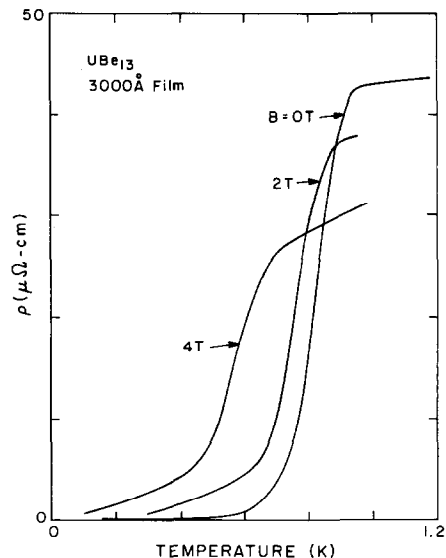

Fig. 1. Resistivity at fixed magnetic-field values as a function of temperature. Magnetic-field values are as indicated in the figure.
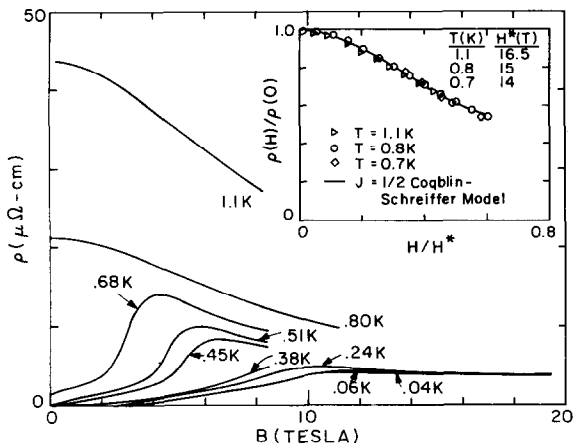
Fig. 2. Resistivity at fixed temperatures as a function of magnetic-field (Inset: fits to $j = 1/2$ model where only 5 percent of the data points used in the fit are shown for clarity).

Fig. 3. Hall voltages at fixed temperatures as a function of magnetic-field.

suppress the superconductivity in UBe$_{13}$ [18]. Disregarding the "foot", the interesting features of Fig. 1 are the rather large magnetoresistance in the normal state and the slightly broadened transition widths. Both of these effects have been observed in previous work [8, 13, 19].

In order to investigate the negative magnetoresistance more extensively, we have swept magnetic-field at fixed temperature. These results are shown in Fig. 2. Again we see the strong negative magnetoresistance in the normal state, which is consistent with previous work on polycrystalline materials [8] as well as single crystal samples [19]. This seems to be a characteristic of heavy fermion materials in general and for UBe$_{13}$ is fit well with the Coqblin–Schreiffer $j = 1/2$ model. Fitting our results to the same model, we derive a temperature dependent Kondo-field of 16.5 T, 15 T, and 14 T at temperature of 1.1 K, 0.8 K, and 0.7 K respectively, values larger than those of [13] but with a weaker temperature dependence. These fits are shown in the inset of Fig. 2. At large magnetic fields and low temperatures the resistivity is constant. This has been interpreted in previous work as a residual resistivity. In [14] they propose that a subtraction of the residual resistivity from the overall resistivity will yield a better fit to the spin-1/2 model and give a slightly different interpretation of the temperature dependent Kondo-field (reduces Kondo energy). We find that this subtraction degrades our fit. This is surprising since our residual resistivity of about 4 $\mu\Omega$ cm^{-1} is about a factor of three to four smaller than the values reported previously.

The Hall effect signal is shown in Fig. 3. Resistivity and Hall voltage have a strikingly similar magnetic-field dependence indicating a similar scattering
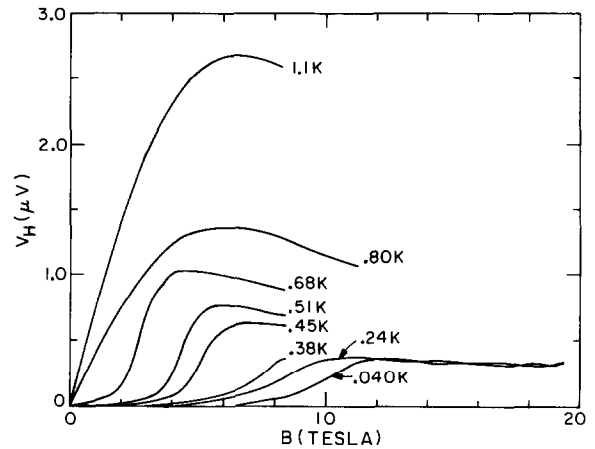
mechanism. At reduced temperatures, the Hall signal shows little temperature dependence when compared to the temperature region corresponding to the onset of coherence. In the Hall effect, this lack of temperature dependence corresponds to the constant residual background Hall coefficient referred to in the introduction. However, we detect no change of sign in the Hall coefficient, which is in sharp contrast to an earlier study by Aliev and coworkers [1].

The "switching on" of the Hall voltage has been observed before and the threshold field at which one sees a finite Hall voltage has been defined as H_{c2} [1]. If we construct a phase diagram in H–T space using this criteria, we develop a phase boundary with a slope consistent with published results [19]. This construction, a phase boundary generated from Fig. 2, and a phase boundary from single-crystal published work are shown in Fig. 4 for comparison. The threshold field lies roughly at the $R/R_N = 0.2$ point on the resistivity versus magnetic-field curves. There have been several attempts to explain the Hall effect in the mixed state of a type-II superconductor. Fukuyama et al. have calculated the signal due to fluctuation effects above T_c [20]. They expect a signal on the order of 10^{-8} V. This is two orders of magnitude smaller than our observed signal. Maki has calculated an expression for the Hall angle near H_{c2} in the mixed state [21]. Although we do not have all of the relevent parameters to check for quantitative agreement, this theory does not predict a sudden jump in the Hall angle. It predicts a maximum at H_{c2}, and a sharp drop at fields larger than this, while we observe the opposite. From these comparisons, it seems obvious that we can not attribute our Hall signal to flux motion or fluctuation effects.

Magnetic impurities (i.e., excess U atoms detected in X-ray spectra) which do not partake in the collec-
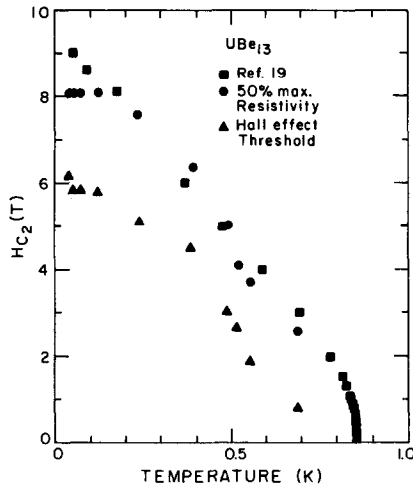
Fig. 4. H_{c2} vs T using the criteria indicated in the figure. Also shown is the phase boundary as defined in [19]. Symbols corresponding to each are defined in the figure.

tive behavior of the Kondo lattice could be responsible for the existance of the positive residual Hall effect as well as the absence of the peak in resistivity usually taken to be the onset of coherence. These considerations may indicate that Hall effect measurements on heavy fermion sytems yield very limited information regarding carrier concentration even at low temperatures due to the residual incoherent effects. The residual resistivity, which is considerably smaller than those values reported previously, is probably related to the absence of the peak. While it seems reasonable that the residual Hall effect may result from impurities, it is more likely the thin film geometry of our samples which obscures the resistivity peak. Many features in resistivity measurements are reduced in thin films due to structural defects influencing the transport mean free path.

CONCLUSIONS

The work described above indicates that the temperature dependent Hall effect observed in our studies is the result of temperature dependent skew and an enhanced residual incoherent scattering in the coherent regime. We believe the enhanced incoherent scattering results from an impurity phase of Uranium indicating that the residual Hall effect observed in UBe$_{13}$ and in other heavy fermion materials is sample dependent. However, the temperature dependence of the Hall coefficient is intrinsic to the system.

A comparison between the Hall and magneto-resistance signals indicates that the residual background terms of each are a result of the same phenomena, that is a saturation of the impurity mag-

netization. Thus, the residual resistivity is also sample dependent explaining our estimate of $4\,\mu\Omega\,cm^{-1}$ which is three to four times smaller than previous estimates.

From the longitudinal magnetoresistance data we see that the system can be described as $j = 1/2$ Coqblin–Schreiffer model after the applied magnetic-field has suppressed the superconductivity. We detect no positive magnetoresistance although it does approach zero as the temperature goes to zero.

Using the threshold field of the Hall effect as a definition for H_{c2} provides a criteria for the determination of the phase diagram from transport measurements. The threshold field corresponds to the onset of incoherent scattering and is not hampered by the ambiguities caused by Josephson effects or the large negative magnetoresistances present in these systems. We do not believe the measurements are complicated due to flux motion or superconducting fluctuations.

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