RESISTIVITY AND AC-CALORIMETRIC MEASUREMENTS OF THE SUPERCONDUCTING TRANSITION IN $CeCu_2Si_2$ UNDER VERY HIGH HYDROSTATIC PRESSURE IN A HELIUM-FILLED DIAMOND ANVIL CELL*

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In the ideal pressure conditions of a helium-filled diamond anvil cell, we have been able to probe the resistivity and, for the first time, the specific heat of the heavy fermion superconductor $CeCu_2Si_2$ at pressures over 6 GPa, down to temperatures below 100 mK, and in a magnetic field up to 8 T. We clearly observed the superconducting jump using the AC calorimetry technique, which provides a semi-quantitative measure of the sample specific heat. The evolution of the superconducting transition with pressure was observed quasi-simultaneously in a single sample in both the resistivity and heat capacity. The jump in C_p hints at changes in the coupling regime. When T_c is a rapidly varying function of pressure, the resistive transition is broadened and strongly dependent on the measuring current. When T_c has a maximum at 2 K, the residual resistivity shows a peak, and the resistivity is linear in temperature above T_c.

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1. Introduction

Pressure is a very useful technique for exploring the properties of strongly correlated systems such as $CeCu_2Si_2$. However, the hydrostaticity of the pressure medium can play an important role — hence the need for helium. $CeCu_2Si_2$ has commanded much interest since it was found to be a superconductor in 1979 [1]. Its phase diagram under pressure, $T_c(P)$, is one of

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$CeCu_2Si_2$'s least understood features. $T_c(P)$ has been measured in detail by susceptibility in a helium pressure transmitting medium [2], where it was found to vary in a highly non-monotonic way with pressure, with several sharp features. Resistivity measurements have been carried out in both a quasi-hydrostatic Bridgman anvil cell [3, 4], and in helium [6]. While in broad agreement, the resistivity results fail to conclusively reproduce T_c^{χ}; they show a marked sample dependence, with a lower T_c corresponding to a higher residual resistivity. Here we attempt to clear up some of this confusion by presenting resistivity and AC-calorimetric [5] measurements on $CeCu_2Si_2$ in a He-filled diamond anvil cell to over 6 GPa.

2. Experimental

Six 5 μm wires (four gold and two AuFe [0.07%]) were spot welded to a $230 \times 80 \times 20\,\mu m^3$ $CeCu_2Si_2$ sample. Two thermocouple junctions were formed, each from one AuFe and one Au wire. An alternating current was passed through one thermocouple, while the signal from the other was measured using a lock-in amplifier. The resulting temperature oscillations serve as a sensitive measure of the sample heat capacity. The signal also contains a contribution from the thermal coupling to the surroundings. At a high enough excitation frequency, the sample contribution dominates the signal. The cut-off frequency ω_c turns out to be very temperature dependent. Fortunately, while complicating the data analysis, the reduction in ω_c at the lowest temperatures allows the technique to be used down to $\sim 100\,\mathrm{mK}$.

3. Results and discussion

Fig. 1 shows the phase diagram determined by both resistivity and specific heat, both on increasing and decreasing the pressure. Previous resistivity measurements on $CeCu_2Si_2$ under pressure showed broad superconducting transitions whose widths depended strongly on pressure, an effect that can be partly blamed on pressure gradients. Unfortunately, while removing pressure gradients, helium uncovers further complications. Our measurements highlighted the presence of at least two qualitatively different types of behaviour in the same sample. The current dependence of the transition width implies that the sample contains a filamentary superconducting region with a higher T_c, while the majority of the sample is represented by the point at which $R = 0$. The bulk nature of the transition at $T_c^{R=0}$ was demonstrated by the calorimetry results, to be discussed below. If the filamentary superconductivity is defined by an onset criterion, $T_c^{\mathrm{onset}}(P)$ seems to show the sharp changes in slope found by susceptibility, while $T_c^{R=0}(P)$ is more in line with those resistivity measurements showing a lower T_c.

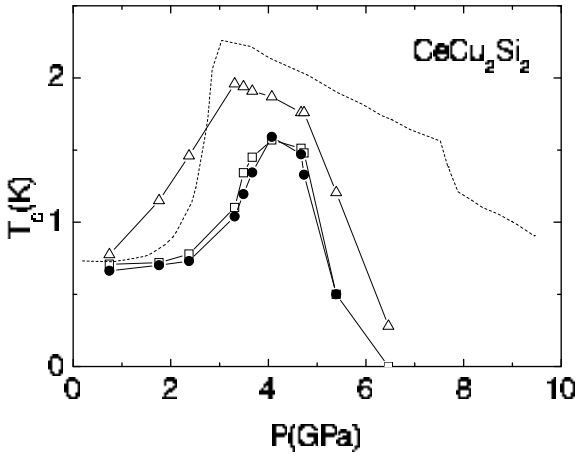

Fig. 1. $T_c(P)$ determined by resistivity (open symbols) and the C_p jump midpoint (filled circles). Triangles show $T_c^{\rm onset}$ and squares $T_c^{R=0}$. The dotted line shows the phase diagram determined by susceptibility [2]. Note the good agreement between the $R = 0$ and C_p onset criteria, and also the width of the resistive transition.

Fig. 2 shows the analysis of the specific heat data at 3.67 GPa. A simple one-dimensional model of the AC-calorimetry system predicts the amplitude and phase of the temperature oscillations ($T_{\rm AC}$) induced by AC heating. $T_{\rm AC} = P_0/(\kappa + i\omega C)$, where P_0 is the heating power, κ the thermal coupling to the bath, C the sample heat capacity, and ω the excitation frequency, assumed to be low enough that the thermometer can follow the temperature oscillations. At high enough frequencies, $T_{\rm AC}$ can be assumed to be inversely proportional to the sample heat capacity. However, $\omega \sim \omega_c = \kappa/C$, and ω_c in turn depends on the temperature. One can use the phase of the AC-calorimetry signal to correct for this. Alternatively, one can subtract a background signal taken at a different frequency. These two methods give a good agreement below ~ 2 K.

The shape and size of the specific heat jump should be treated with caution, but can provide useful information. If one compares the C_p jump at ~ 700 mK (2.4 GPa) and ~ 1.5 K (3.7 GPa), there appears to be an increase in $(C_s - C_n)/C_n$, suggesting a change in coupling regime. However, the measuring conditions are very different at the two temperatures. One can comment with fewer caveats on the shape of the peak: the specific heat jump remains sharp as the pressure is increased, even during the rapid rise in T_c, demonstrating the quality of the pressure medium. However, as the pressure is increased, the superconducting peak broadens and collapses, even before T_c starts to fall. At least above 2 GPa the physics is reversible, with the C_p peak regaining its shape and size on depressurisation. The electronic

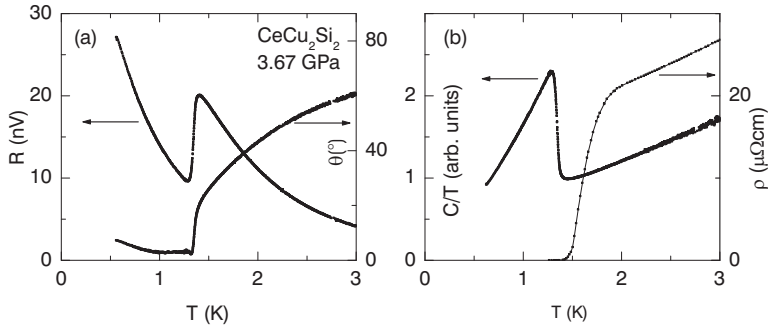

Fig. 2. (a) Modulus and phase of thermocouple signal at 3.67 GPa as a function of temperature. For $\omega C \gg \kappa$, $\theta \to 0°$; for $\omega C \ll \kappa$, $\theta \to 90°$. (b) Transition in C/T and ρ. $C = \cos\theta/(\omega T_{\mathrm{AC}})$, where T_{AC} is calculated from the thermocouple signal amplitude and the AuFe thermopower (*n.b.* a 90° phase offset is taken into account in the calculation). Note the onset of the specific heat transition when the resistive transition is complete.

specific heat γ can also be estimated from the normal-state calorimetry signal. It shows a peak close to the maximum T_{c}, superimposed on general decrease with pressure. The normal state resistivity confirms previous results, with a linear temperature dependence around the maximum T_{c}, along with a maximum in the residual resistivity.

4. Conclusions

We have extended the AC-calorimetry technique to higher pressures and lower temperatures in a helium medium. While this method is currently only semi-quantitative, our results should help further the understanding of superconductivity in $CeCu_2Si_2$. A more quantitative understanding of the AC-calorimetry results seems possible, by more accurate modelling, and/or by calibration with a well-characterised compound.

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