

# FIRST-ORDER SUPERCONDUCTING TRANSITION AT THE UPPER CRITICAL FIELD IN $\text{CeCoIn}_5$ STUDIED BY DC MAGNETIZATION MEASUREMENT\*

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DC magnetization process of a high-quality single crystal of  $\text{CeCoIn}_5$  has been measured at low temperatures down to 50 mK. A sharp magnetization jump with a small hysteresis is observed at the upper critical field  $H_{c2}$  for both  $a$  and  $c$  directions, indicating the transition to the normal state to be of first-order. Although the results might suggest a strong Pauli paramagnetic effect, no Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) phase is observed.

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## 1. Introduction

The tetragonal heavy electron compound  $\text{CeCoIn}_5$  becomes superconducting at  $T_c = 2.3$  K [1]. Various experimental studies indicate that the electron pairing is spin singlet with  $k_x^2 - k_y^2$  gap symmetry [2, 3]. Due to its large Pauli paramagnetic susceptibility in the normal state,  $\text{CeCoIn}_5$  provides an interesting situation where the paramagnetic energy becomes a substantial  $f$  fraction of the superconducting condensation energy at high

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fields and may affect the transition at the upper critical field  $H_{c2}$ . Here we studied the magnetization process of a single crystal of  $\text{CeCoIn}_5$  at very low temperatures, with special interest in the  $H_{c2}$  transition behavior [4].

## 2. Experimental

The single crystal of  $\text{CeCoIn}_5$  has been grown by a flux method. Electron mean-free path  $l$  estimated by the de Haas–van Alphen effect was in excess of  $2000 \text{ \AA}$  [5], well in the clean limit  $l \gg \xi_{a,c} (< 100 \text{ \AA})$ . DC magnetization of the sample (6.9 mg weight) was measured by a capacitive Faraday magnetometer installed in a dilution refrigerator [6], at temperatures down to 50 mK in magnetic fields up to 125 kOe.

## 3. Results and discussion

Fig. 1 shows the magnetization curves obtained at the base temperature of 50 mK with slowly varying magnetic field applied parallel to the tetragonal  $a$  and  $c$  axes. The thick arrows indicate the direction of the field sweep. At low fields in the mixed state, the magnetization process is irreversible due to flux pinning. For  $H \parallel c$ , we observed an enhancement of the magnetization hysteresis at around  $2T$  (peak effect). The hysteresis becomes small as

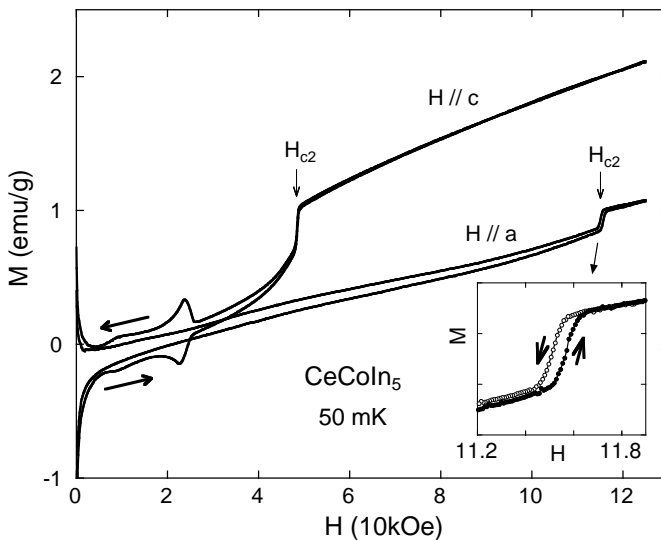


Fig. 1. Magnetization curves of  $\text{CeCoIn}_5$  measured at  $T = 50 \text{ mK}$ , with magnetic field applied parallel to the tetragonal  $a$  and  $c$  axes. The sharp jump of the magnetization implies that the  $H_{c2}$  transition is of first order. The inset shows the magnetization behavior near  $H_{c2}$  ( $H \parallel a$ ) in an expanded scale.

the upper critical field  $H_{c2}$  is approached. Very interestingly, magnetization exhibits a discontinuous jump at  $H_{c2}$  for both field directions. This behavior is in striking contrast to the case of ordinary type-II superconductors in which the magnetization continuously recovers the normal state value at  $H_{c2}$ . Our results indicate that the  $H_{c2}$  transition in CeCoIn<sub>5</sub> is of first-order at low temperatures. In fact, a small but distinct hysteresis is observed in the transition field (the inset of Fig. 1). Similar evidences for the first-order transition have been reported in the field dependence of the thermal conductivity [3] and the magnetic torque [7] measurements. When the temperature was raised, the magnetization jump at  $H_{c2}$  became smaller and eventually vanished at around 0.7 K for both field directions. Fig. 2 shows the resulting  $H_{c2}(T)$  phase diagrams of CeCoIn<sub>5</sub>. Open circles indicate the transition points which are considered to be of first order, whereas the solid squares denote the second order ones where the magnetization change is continuous. Thus, a critical point separating the first-order and the second-order transition lines seems to exist at  $T \sim 0.3T_c$  for both field directions.

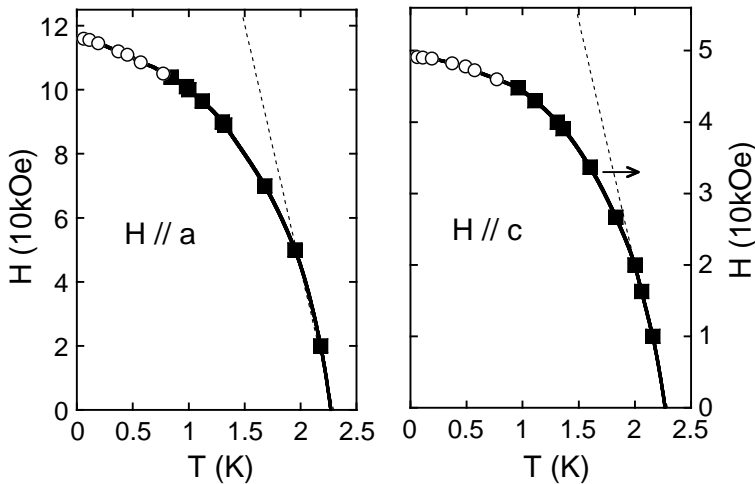


Fig. 2. Phase diagrams of CeCoIn<sub>5</sub> under magnetic field parallel to the *a* and *c* axes. Open circles are the first-order  $H_{c2}$  transition points where the magnetization shows a discontinuous jump, whereas the solid squares are the second order ones.

Although the origin of the first-order  $H_{c2}$  transition in CeCoIn<sub>5</sub> is not very clear at present, a possible mechanism would be the Pauli paramagnetic effect which comes from the difference in spin susceptibility between superconducting and normal states. The critical field to the normal state is suppressed at low temperature and the  $H_{c2}$  transition may become of first order, provided that the spin paramagnetism is strong enough [8–10]. In fact, the actual  $H_{c2}$  curves at low  $T$  appear to be suppressed well below the

orbital limit (the dashed lines in Fig. 2) which is estimated from the slope of  $H_{c2}(T)$  at  $T_c$ . CeCoIn<sub>5</sub> might therefore be the first system that exhibits a strong Pauli paramagnetic limiting of the upper critical field, though more careful study would be needed to confirm this point. Concerning this point, the Fulde–Ferrell–Larkin–Ovchinnikov phase, a partially spin-polarized superconducting state, has been predicted by several authors to show up near  $H_{c2}$  [11–13]. Looking carefully at our data, however, we could not find any new phase boundary branching from the  $H_{c2}(T)$  curves. Stability of the FFLO state might be very sensitive to impurities. By contrast, the first-order  $H_{c2}$  transition seems to be rather robust to doping. In our preliminary measurements on the mixed system CeCo<sub>1-x</sub>Rh<sub>x</sub>In<sub>5</sub>, we found that the magnetization jump clearly exists at least for  $x = 0.01$ . The detail of the results will be published elsewhere.

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