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Pairing symmetry of $CeCoIn_5$ detected by in-plane torque measurements

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In-plane torque measurements were performed on heavy fermion $CeCoIn_5$ single crystals in the temperature T range $1.8\ \mathrm{K} \leq T \leq 10\ \mathrm{K}$ and applied magnetic field H up to 14 T. The normal-state torque is given by $\tau_n \propto H^4(1+T/T_K)^{-1} \sin 4\varphi$. The reversible part of the mixed-state torque, obtained after subtracting the corresponding normal-state torque, shows also a fourfold symmetry. In addition, sharp peaks are present in the irreversible torque at angles of $\pi/4$, $3\pi/4$, $5\pi/4$, $7\pi/4$, etc. Both the fourfold symmetry in the reversible torque and the sharp peaks in the irreversible torque of the mixed state imply d_{xy} symmetry of the superconducting order parameter. The field and temperature dependences of the reversible mixed-state torque provide further evidence for d_{xy} wave symmetry. The fourfold symmetry in the normal state has a different origin since it has different field and temperature dependences than the one in the mixed state. The possible reasons of the normal-state fourfold symmetry are discussed.

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I. INTRODUCTION

The superconductivity in the heavy fermion superconductor $CeCoIn_5$ is unconventional as exemplified by the non-Fermi-liquid behavior,[1–3] the giant Nernst effect present in the normal state,[4] its proximity to quantum critical points,[5,6] the Pauli limiting effect,[7–9] and the possible multiband picture in the superconducting state.[10–12] Unconventional superconductivity is always a subject of great interest. Knowing the pairing symmetry, which is related to the ground state and gap energy, is essential to the understanding of the pairing mechanism and the origin of superconductivity. For a conventional superconductor, which is described by the BCS theory, the pairing is phonon mediated and the pairing symmetry is s wave. For an unconventional superconductor, the quasiparticle gap vanishes at certain points on the Fermi surface. For example, most of the experimental evidence on high-temperature superconductors, such as $YBa_2Cu_3O_{7-\delta}$, indicates that the $d_{x^2-y^2}$ wave symmetry dominates in these materials.

It has been established that the superconducting order parameter of $CeCoIn_5$ displays d-wave symmetry. ^{115}In and ^{59}Co nuclear-magnetic-resonance measurements[13] and torque measurements[14] have revealed a suppressed spin susceptibility, which implies singlet spin pairing. A T^2 term is present in the low temperature T specific heat, consistent with the presence of nodes in the superconducting energy gap.[15] Nuclear quadrupole resonance and nuclear-magnetic-resonance measurements on $CeCoIn_5$ have revealed that the nuclear-spin-lattice relaxation rate $1/T_1$ has no Hebel-Slichter coherence peak just below the superconducting transition temperature T_c, and it has a T^3 dependence at very low temperatures, which indicates the existence of line nodes in the superconducting energy gap.[16]

Nevertheless, the direction of the gap nodes relative to the Brillouin-zone axes, which determines the type of d-wave state, namely $d_{x^2-y^2}$ or d_{xy}, is still an open question and an extremely controversial issue. For example, angular-dependent thermal-conductivity measurements in a magnetic field have revealed fourfold symmetry consistent with $d_{x^2-y^2}$ symmetry.[17] Neutron scattering experiments by Eskildsen *et al.* revealed a square lattice oriented along [110], also consistent with $d_{x^2-y^2}$ wave symmetry.[18] However, field-angle-dependent specific-heat measurements have found the symmetry of the superconducting gap to be d_{xy}.[19] More recent magnetoresistance[20] and neutron[21] data has shown $d_{x^2-y^2}$ symmetry. Furthermore, theoretical calculations by Ikeda *et al.* strongly suggest d_{xy} wave symmetry when taking into account the available T^* experimental data of the Fulde-Ferrell-Ovchinnikov states[22] [T^* is the temperature at which the upper critical field $H_{c2}(T)$ changes from second order to first order]. In contrast, recent calculations by Tanaka *et al.*[23] based on the Fermi-liquid theory and by Vorontsov *et al.*[24] based on a unified microscopic approach, support the $d_{x^2-y^2}$ gap symmetry.

This extremely controversial issue needs to be resolved through an experimental technique that allows the direct measurement of the nodal positions. The experiments that are phase sensitive usually include surface or boundary effects, while the experiments that detect bulk properties are not phase sensitive. In the study presented here, we use torque measurements to clarify the gap symmetry. Torque is a bulk measurement so it provides information on the order parameter of the bulk, not only the surface. (The order parameter of the surface might be different from that of the bulk). It also *directly* probes the nodal positions on the Fermi surface with high angular resolution since torque is the angular derivative of the free energy. Hence, such an experimental technique is ideal to determine the direction of the gap nodes relative to the Brillouin-zone axes and, in fact, it has already been successfully used to identify the nodal positions of untwined $YBa_2Cu_3O_{7-\delta}$ single crystals[25] and $Tl_2Ba_2CuO_{6+\delta}$ thin films.[26] In addition, recent theoretical calculations by Adachi have shown that low-field torque measurements can be used to detect the nodal positions of a *d*-wave superconductor with a small Fermi-surface anisotropy,[27] as is the case of CeCoIn_5.

In-plane torque measurements were performed on single crystals of CeCoIn_5 both in the normal state and in the mixed state. The reversible part of the angular-dependent mixed-state torque data, obtained after subtracting the corresponding normal-state torque, shows a fourfold symmetry with a positive coefficient. Sharp peaks in the irreversible torque data were observed at angles equal to $\pi/4$, $3\pi/4$, $5\pi/4$, and $7\pi/4$, etc. The symmetry of the free energy extracted from the fourfold symmetry of the reversible torque of the mixed state coupled with the position of the sharp peaks in the irreversible torque of the mixed-state point unambiguously toward d_{xy} wave symmetry in CeCoIn_5. Further support for the d_{xy} wave symmetry is provided by the H and T dependence of the mixed-state reversible torque. Normal-state torque shows a fourfold symmetry. Nevertheless, the T and H dependences of the coefficients of the fourfold torques in the normal and mixed states are different. Hence, these two fourfold symmetries have clearly different origin.

II. EXPERIMENTAL DETAILS

Single crystals of CeCoIn_5 were grown using the flux method. High quality crystals with regular shape and shiny surfaces were chosen to carry out the torque measurements. The single crystals were etched in concentrated HCl for several hours to remove the indium left on the surface during the growth process. The crystals were then rinsed thoroughly in ethanol.

A piezoresistive torque magnetometer was used to measure the angular dependence of the in-plane torque of CeCoIn_5 both in the normal state and mixed state. The torque was measured over a large temperature range ($1.8\ \mathrm{K} \leq T \leq 10\ \mathrm{K}$) and magnetic field H range ($1.5\ \mathrm{T} \leq H \leq 14\ \mathrm{T}$) by rotating the single crystal in fixed magnetic field. The angle φ for the in-plane rotation was defined as the angle made by the field with the *a* axis of the single crystal. The contributions of the gravity and puck to the total torque signal were measured and subtracted from it as discussed elsewhere.[12]

The experiments were carried out in a physical property measurement system (PPMS). In such a system with a one axis rotator, it is very difficult to ensure an in-plane alignment of better than about $\pm 3°$. If misalignment exists, i.e., the magnetic field is not completely within the *ab* plane of the single crystal, there should be a $\sin 2\varphi$ term [see Eq. (6) of Ref. 28]. Indeed, the angular-dependent in-plane torque signal has a $\sin 2\varphi$ term [α and φ of Eq. (6) are φ and $-23.7°$, respectively, in the present case], which we attribute to the misalignment of the single crystal, in addition to the $\sin 4\varphi$ term. In fact, the amplitude of the measured $\sin 2\varphi$ term gives a misalignment $\theta \approx 3.7°$. The torque data shown in this paper are after subtracting this $\sin 2\varphi$ term.

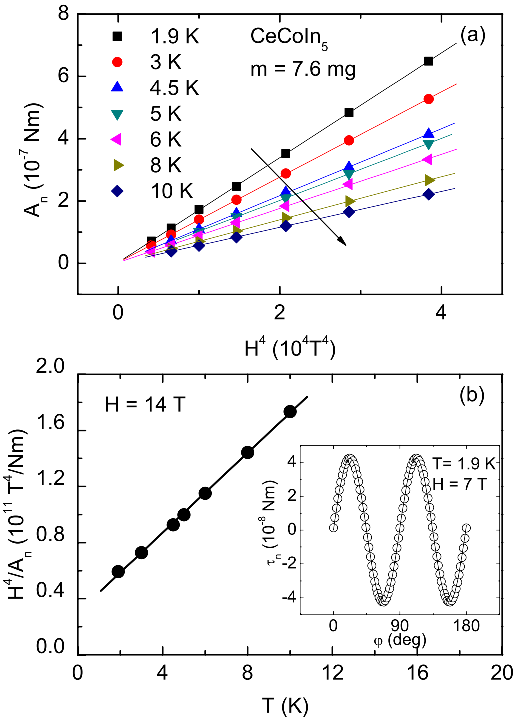

FIG. 1. (Color online) Field H and temperature T dependence of the amplitude A_n of the normal-state torque of CeCoIn_5 single crystals. (a) H dependence of A_n at measured temperatures of 1.9, 3, 4.5, 5, 6, 8, and 10 K. (b) T dependence of A_n at measured magnetic field of 14 T. Inset: Angular φ dependent torque τ_n measured in the normal state at 1.9 K and 7 T in CeCoIn_5 single crystals. The solid line is a fit of the data with $\tau_n = A_n \sin 4\varphi$.

III. RESULTS AND DISCUSSION

Previously, we have shown that the *b*-axis rotation torque signal measured in the mixed state has a paramagnetic component which is comparable with the diamagnetic component.[12] The former component is a result of the anisotropy of the susceptibilities along the *a* and *c* axes. Therefore, such a paramagnetic torque signal is absent in the present measurements in which the torque is measured while rotating the single crystal along the *c* axis, since $\chi_a \approx \chi_b$. Nevertheless, T and H dependent torque measurements in the normal state reveal that the normal-state torque signal is not negligible, is reversible, and it has a fourfold symmetry [see inset to Fig. 1(b)]. The solid line is a fit of the data with $\tau_n = A_n \sin 4\varphi$. The field and temperature dependence of the amplitude A_n gives the H and T dependence of the normal-state torque. The coefficient A_n has an H^4 dependence up to 14 T for all measured temperatures from 1.9 to 10 K [see Fig. 1(a)]. As the temperature increases, the slope of the plots in Fig. 1(a) decreases. This is consistent with the temperature dependence of H^4/A_n shown in Fig. 1(b); i.e., H^4/A_n increases, hence A_n decreases, with increasing T. A straight-line fit of the data with $H^4/A_n = c(1 + T/T_0)$ gives $T_0 = 2.2$ K, which has a value close to the single-ion Kondo temperature T_K [reported to be between 1 and 2 K, (Ref. 29)] and $c = 3 \times 10^{10}\ \mathrm{T}^4\ \mathrm{N}^{-1}\ \mathrm{m}^{-1}$. Hence, $A_n(H,T) \propto H^4(1 + T/T_K)^{-1}$. Therefore, in approaching the superconducting transition, A_n decreases with decreasing H and increases with decreasing T. We subtract the corresponding normal-state torque from the torque measured in the superconducting state.

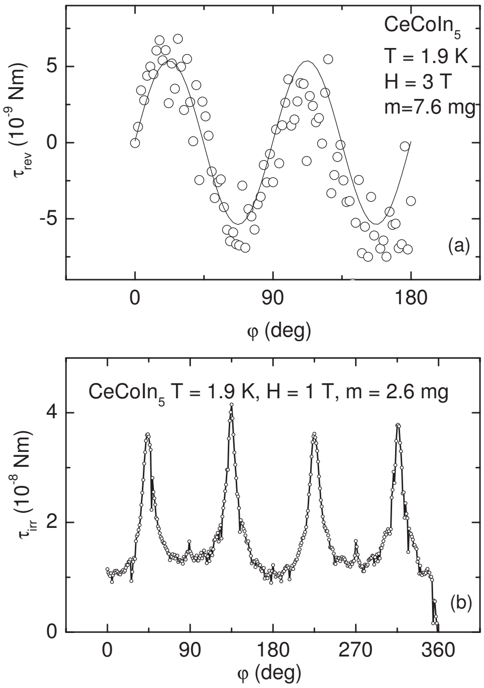

FIG. 2. (a) Angular φ dependence of the reversible torque τ_{rev} measured at 1.9 K and 3 T on $\mathrm{CeCoIn_5}$ single crystals. The solid line is a fit of the data with $\tau = A \sin 4\varphi$. (b) Angular dependence of the irreversible torque τ_{irr} measured at 1.9 K and 1 T on $\mathrm{CeCoIn_5}$ single crystals. Sharp peaks are present at $\pi/4$, $3\pi/4$, $5\pi/4$, and $7\pi/4$.

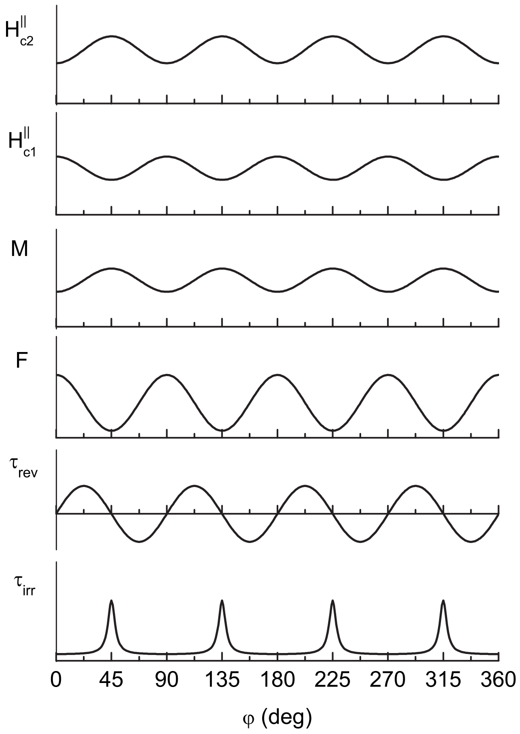

FIG. 3. Plot of the angular φ dependence of the upper critical field H_{c2} and lower critical field H_{c1} for magnetic field parallel to the *ab* plane, magnetization M, free energy F, reversible torque τ_{rev}, and irreversible torque τ_{irr} for d_{xy} wave symmetry.

The torque data measured in the mixed state of $\mathrm{CeCoIn_5}$, obtained by subtracting the corresponding normal-state torque from the measured torque, have both reversible and irreversible components. The reversible torque τ_{rev} is the average of the torque data measured in clockwise and anticlockwise directions, while the irreversible torque τ_{irr} is the average of the antisymmetric components of the torque data measured in clockwise and anticlockwise directions. Figure 2(a) shows the reversible part of the angular-dependent in-plane torque data measured in the mixed state at 1.9 K and in a magnetic field of 3 T. Clearly, there is a fourfold symmetry present in the torque data, although the data points are somewhat scattered. The solid line is a fit of these mixed-state data with $\tau_{\mathrm{rev}}(H,T,\varphi) = A_m(T,H)\sin 4\varphi$. The coefficient A_m is positive since the torque displays a maximum at $\pi/8$. Figure 2(b) shows the irreversible part of the mixed-state torque data $\tau_{\mathrm{irr}}(\varphi)$ measured at $T = 1.9$ K and $H = 1$ T for a single crystal with a mass of 2.6 mg. The $\tau_{\mathrm{irr}}(\varphi)$ data have sharp peaks at $\pi/4$, $3\pi/4$, $5\pi/4$, and $7\pi/4$, etc.

The nodal positions of $\mathrm{CeCoIn_5}$ can be obtained from the reversible and irreversible mixed-state torque data, as previously done in the study of $\mathrm{YBa_2Cu_3O_7}$.[25] Specifically, theoretical calculations predict that the in-plane upper critical field $H_{c2}^{\parallel}$ has a fourfold symmetry for a d-wave superconductor.[30] In the case of d_{xy} wave symmetry, the angular variation of the upper critical field $\Delta H_{c2}^{\parallel} \propto -\cos 4\varphi$; hence, it has maxima at $\pi/4$, $3\pi/4$, $5\pi/4$, $7\pi/4$, etc. Figure 3 shows the angular dependence of the reversible and irreversible torque obtained by starting from this angular dependence of H_{c2}, as follows. The lower critical field $H_{c1}^{\parallel}$ is out of phase with $H_{c2}^{\parallel}$ (see Fig. 3) since the thermodynamic critical field $H_c^2 = H_{c1}H_{c2}$ is independent of the magnetic-field orientation. Therefore, the magnetization M, given by $M \simeq -H_{c1}\ln(H_{c2}/H)/\ln\kappa$, has the same angular dependence as H_{c2} (see Fig. 3). The easy axis of magnetization (maximum magnetization) should correspond to free energy F minima. This implies that, for the d_{xy} symmetry, F has minima at $\pi/4$, $3\pi/4$, $5\pi/4$, $7\pi/4$, etc. (see Fig. 3). The torque is the angular derivative of the free energy F; i.e., $\tau = -\partial F/\partial\varphi$. Hence, the reversible torque data for a material with d_{xy} wave symmetry should display a fourfold symmetry with maxima at $\pi/8$, $5\pi/8$, $9\pi/8$, etc. (see Fig. 3). Also, the free-energy minima act as intrinsic pinning centers for vortices, so the irreversible torque data for a material with d_{xy} wave symmetry should display peaks at the same angles at which the free energy has minima; i.e., at $\pi/4$, $3\pi/4$, $5\pi/4$, $7\pi/4$, etc.

Notice that the φ dependence of reversible and irreversible torque data of $\mathrm{CeCoIn_5}$ shown in Figs. 2(a) and 2(b) are the same as the φ dependence of τ_{rev} and τ_{irr}, respectively, shown in Fig. 3, obtained from the theoretically predicted angular dependence of the upper critical fields for a material with d_{xy} symmetry. We hence conclude that the reversible along with the irreversible torque data in the mixed state unambiguously imply that the wave symmetry of $\mathrm{CeCoIn_5}$ is d_{xy}. The fact that the angular dependences of the reversible and irreversible torques, with the latter not being affected by the subtraction of the *reversible* normal-state torque in the mixed state, give the same angular dependence of the free energy indicates that there is no error in the subtraction of

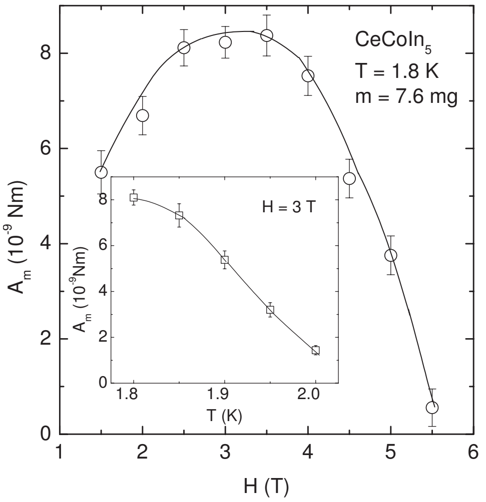

FIG. 4. Field H dependence of the amplitude A_m of the reversible torque in the mixed state of CeCoIn$_5$ single crystals measured at $T=1.8$ K. The solid line is a guide to the eye. Inset: Temperature T dependence of A_m measured at $H=3$ T. The solid line is a guide to the eye.

the normal-state contribution to obtain the reversible torque in the mixed state; i.e., the assumption we made that the normal-state contribution in the mixed state is just a simple extrapolation of its behavior above T_c is correct.

To further understand the fourfold symmetry displayed by the present torque measurements in the mixed state, we studied the field and temperature dependence of the amplitude A_m. Figure 4 is a plot of the H dependence of A_m, which gives the H dependence of the torque, obtained by fitting the angular-dependent torque data measured in different magnetic fields. Note that $A_m(H)$ increases with increasing H, reaches a maximum, and then decreases with further increasing H. Also note that the fourfold symmetry vanishes close to $H_{c2}^{\parallel}$ ($H_{c2}^{\parallel}=6$ T). This field dependence of the magnitude of A_m is the same as the field dependence of the basal-plane reversible torque in the mixed state of a layered $d_{x^2-y^2}$ wave superconductor [see Fig. 8(a) of Ref. 27] calculated by Adachi *et al.*[27] based on the quasiclassical version of the BCS-Gor'kov theory with a Fermi surface which is isotropic within the basal plane. The sign difference between the data of the present Fig. 4 and Fig. 8(a) of Ref. 27, which is for a $d_{x^2-y^2}$ wave symmetry, further indicates that the present data reflect d_{xy} symmetry since the torque data have opposite signs for the d_{xy} and $d_{x^2-y^2}$ wave symmetries.

The inset to Fig. 4 is a plot of the temperature dependence of the amplitude A_m. Note that A_m, hence the torque, decreases with increasing T and vanishes toward T_c. The fact that both the T and H dependences of the reversible mixed-state torque vanish at the superconducting-normal-state phase boundary further indicates that the observed fourfold symmetry is related with superconductivity; hence, it reflects the gap symmetry.

We note that the behaviors of $A_m(H,T)$ and $A_n(H,T)$ are totally different (compare Figs. 1 and 4). So the fourfold symmetries present in normal and mixed states have different origin. The origin of the fourfold symmetry in the normal state is not yet clear to us.

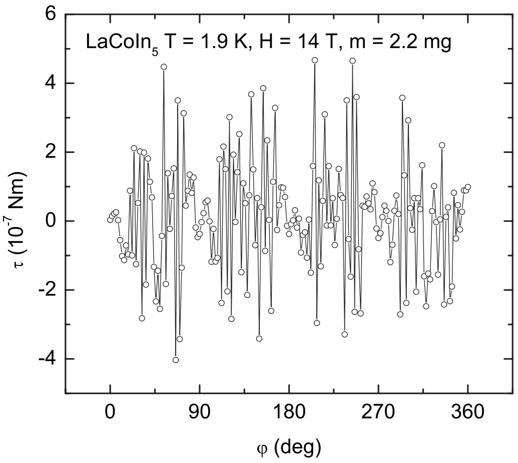

FIG. 5. Angular φ dependent torque τ measured at 1.9 K and 14 T on LaCoIn$_5$ single crystals.

Torque measurements on LaCoIn$_5$ single crystals, which also have a tetragonal structure but are not superconducting and the f electrons are absent, give some clues on the normal-state fourfold symmetry of CeCoIn$_5$. The angular-dependent torque data for LaCoIn$_5$ are shown in Fig. 5. Clearly, the sin 4φ symmetry observed in CeCoIn$_5$ is completely absent here. The difference in the normal-state torques between CeCoIn$_5$ and LaCoIn$_5$ could be due to the presence of heavy electrons in the former compound and their absence in the latter one. The crystalline electric field, which is important in heavy fermion systems, might be responsible for the fourfold symmetry in the normal-state torque. Also, the field induced order, possibly quadrupolar order, could be another reason for the fourfold symmetry in the normal-state torque of CeCoIn$_5$. No doubt, the origin of this normal-state fourfold symmetry present in the torque data requires further study. Nevertheless, this is beyond the scope of this paper.

Recently, Weickert *et al.* tried to directly determine the anisotropy of the upper critical field in the basal plane from measurements of resistance R as a function of the angle φ and magnetic field. Their results give a $d_{x^2-y^2}$ gap symmetry.[20] To further shed light on the discrepancy between these results and the torque results shown here, we also measured the resistance R_c of CeCoIn$_5$ in the c direction with the applied field in the ab plane at 1.8 K (same experimental condition as for the torque measurement reported here). Figure 6 is a plot of $R_c(H)$ measured at different φ values. Notice the $R_c(H)$ curves cross in the transition region such that the angular-dependent magnetoresistance $\Delta R \equiv R(\varphi)-R(\varphi=0)$ is positive at low dissipation and negative at high dissipation, all the way into the normal state of CeCoIn$_5$. The fact that the sign of the angular magnetoresistance changes in the superconducting transition region and since the thermodynamic upper critical field is ill defined show that one cannot determine $H_{c2}(\varphi)$ accurately from the $R(H,\varphi)$ curves. In fact, as shown in the inset of Fig. 6, the fourfold part of $R_c(\varphi)$ gives an angular dependence consistent with d_{xy} (black symbols) or $d_{x^2-y^2}$ (red symbols) symmetry when the upper critical field is chosen in the transition region close to the onset (high dissipation) or close to zero

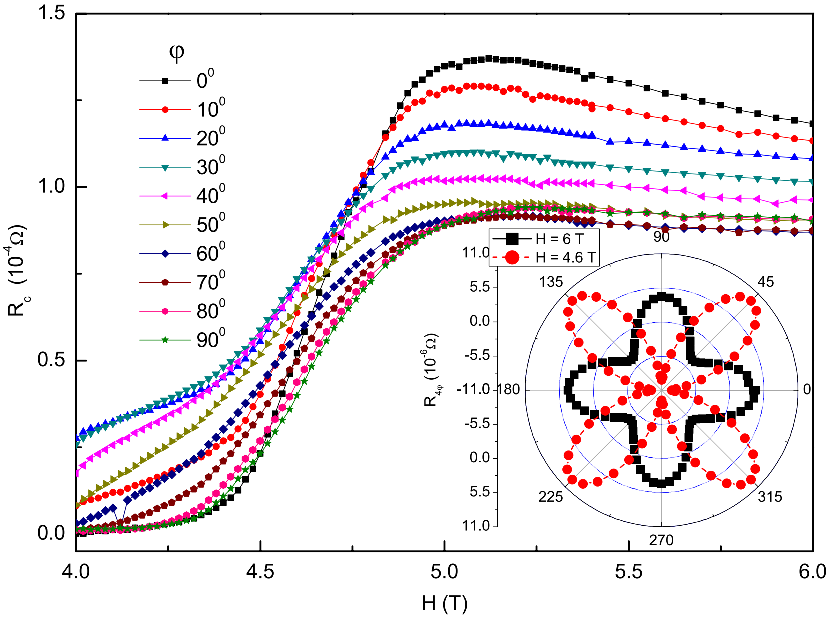

FIG. 6. (Color online) Plot of out-of-plane resistance R_c vs applied magnetic field H measured at a different in-plane angle φ between H and the a axis. Inset: Polar plot of the fourfold symmetric component $R_{4\varphi}$ of the out-of-plane resistance measured for two H values.

resistance (low dissipation, as Weickert *et al.* did), respectively. We note that if the in-plane angular-dependent resistance $R_c(\varphi)$ in the mixed state gives $H_{c2}(\varphi)$ of $\mathrm{CeCoIn_5}$, then the position of the maximum (minimum) resistance corresponds to the position of the minimum (maximum) H_{c2} for the following reason. T_c shifts to higher or lower temperatures as the angle of the applied field is varied in the basal plane. Therefore, by keeping the actual temperature $T=T_c$ fixed, a rotation of the magnetic field results in a variation of the measured resistance if H_{c2} changes with direction such that $R_c(\varphi)$ and $H_{c2}(\varphi)$ are out of phase.[31] We also note that, in addition to the fourfold symmetry in $R_c(\varphi)$, shown in the inset to Fig. 6, there is a twofold symmetry in $R_c(\varphi)$, which has previously been observed and associated with background.[20]

We want to emphasize that the measurement of resistance is not a thermodynamic measurement, hence the determination of the upper critical field and its angular dependence from such a measurement is not reliable. In fact it is well known that the extraction of H_{c2} from the resistive transition is inappropriate for unconventional superconductors such as both hole-doped and the lower T_c electron-doped cuprates. Actually, the magnetoresistance of a superconductor in the mixed state is due to vortex dissipation (as a result of vortex motion), hence it depends on the strength of the vortex pinning centers. Therefore, if one defines H_{c2} close to zero resistivity, as Weickert *et al.* did,[20] the such determined "$H_{c2}(\varphi)$" is in fact the irreversibility field $H_{\mathrm{irr}}(\varphi)$, hence it most likely reflects the pining anisotropy of vortices instead of the in-plane anisotropy of the thermodynamic upper critical field.

IV. SUMMARY

In-plane angular-dependent torque measurements were performed on $\mathrm{CeCoIn_5}$ single crystals both in the normal and mixed states. Normal-state torque measurements show a fourfold symmetry. The reversible torque in the mixed state, obtained after subtracting the corresponding normal-state contribution, also shows a fourfold symmetry with maxima at $\pi/8$, $5\pi/8$, $9\pi/8$, etc. Sharp peaks in the irreversible torque data were observed at $\pi/4$, $3\pi/4$, $5\pi/4$, etc. These latter peaks correspond to minima in the free energy of a d_{xy} wave symmetry. The mixed-state fourfold symmetry and the peak positions in the irreversible torque point unambiguously toward d_{xy} wave symmetry of the superconducting gap. The field and temperature dependences of the amplitude of the normal-state torque are different from that of the mixed-state torque, which indicates that the normal-state torque has a different origin.

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