

Angular-dependent torque measurements on CeCoIn₅ single crystals

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Angular-dependent torque measurements were performed on single crystals of CeCoIn₅, a heavy fermion superconductor ($T_{c0}=2.3$ K), in the temperature, T , range $1.9\text{ K} \leq T \leq 20\text{ K}$ and magnetic fields H up to 14 T. A large paramagnetic effect is found in the normal state. Torque measurements in the mixed state were also performed. The torque curves show sharp hysteresis peaks at $\theta=90^\circ$ (θ is the angle between H and the c -axis of the crystal), a result of intrinsic pinning of vortices. The anisotropy $\gamma \equiv \sqrt{m_c/m_a}$ in the mixed state was determined from the reversible part of the vortex contribution to the torque signal using Kogan's model [Phys. Rev. B **38**, 7049 (1988)]. The anisotropy γ decreases with increasing magnetic field and temperature. The fact that γ is not a constant points towards a multiband scenario in this heavy fermion material.

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I. INTRODUCTION

The heavy fermion compound CeCoIn₅ forms in the HoCoGa₅ tetragonal crystal structure with alternating layers of CeIn₃ and CoIn₂. It is superconducting at 2.3 K, the highest superconducting transition temperature T_{c0} yet reported for a heavy fermion superconductor.¹ Considerable progress has been made in determining the physical properties of this material. The superconductivity in this material is unconventional. The presence of a strong magnetic interaction between the $4f$ moments and itinerant electrons allows the possibility of nonphonon mediated coupling between quasiparticles.^{2,3} Angular-dependent thermal conductivity measurements show $d_{x^2-y^2}$ symmetry which implies that the anisotropic antiferromagnetic fluctuations play an important role in superconductivity.⁴ Bel *et al.* reported the giant Nernst effect in the normal state of CeCoIn₅, which is comparable to high T_c superconductors in the superconducting state.⁵ Non-Fermi liquid behavior was observed in many aspects.^{1,6,7} The unconventional superconductivity and the similarity to high T_c superconductors attract great interest to study this system.

Measurements of de Haas-van Alphen oscillations in both the normal and mixed states have revealed the quasi two-dimensional nature of the Fermi surface and the presence of a small number of electrons exhibiting 3D behavior.^{8,9} Through point-contact spectroscopy measurement, Rourke *et al.*¹⁰ found that in CeCoIn₅ there are two coexisting order parameter components with amplitudes $\Delta_1 = 0.95 \pm 0.15$ meV and $\Delta_2 = 2.4 \pm 0.3$ meV, which indicate a highly unconventional pairing mechanism, possibly involving multiple bands. This is very similar to the case of MgB₂, where two coexisting s -wave gaps were found by the same technique.¹¹ Thermal conductivity and specific heat measurements made by Tanatar *et al.* have revealed the presence of uncondensed electrons, which can be explained by an extreme multiband scenario, with a d -wave superconducting gap on the heavy-electron sheets of the Fermi surface and a negligible gap on the light, three-dimensional pockets.¹²

The presence of multibands (gaps) together with the reported field dependence of the cyclotron masses⁸ point towards a possible temperature and/or field dependent anisotropy in the superconducting state, which according to the standard anisotropic Ginzburg-Landau (GL) theory is given by $\gamma \equiv \sqrt{m_c/m_a} = H_{c2}^{\parallel c}/H_{c2}^{\parallel a} = \lambda_c/\lambda_a = \xi_a/\xi_c$ (c and a are crystallographic axes, and m, H_{c2}, λ , and ξ are the effective mass, upper critical field, penetration depth, and coherence length, respectively). Specifically, in the multiband scenario proposed by Rourke *et al.*¹⁰ and Tanatar *et al.*,¹² different gaps may behave differently in magnetic field, which may lead to a field dependent γ . Also an anisotropic gap may result in a temperature dependent γ . Reports up to date give values of the anisotropy of CeCoIn₅ in the range 1.5 to 2.47. For example, Petrovic *et al.* have reported an anisotropy of at least 2, as estimated from the ratio of the upper critical fields H_{c2} along the c and a directions.¹ Measurements of $H_{c2}(\theta)$ at 20 mK give a value for the anisotropy of about 2.47.¹³ Magnetization measurements of the lower critical field for temperatures between 1.5 and 2.1 K give the ratio of the out-of-plane and in-plane penetration depth $\lambda_c/\lambda_a \approx 2.3$ and of the in-plane and out-of-plane coherence length $\xi_a/\xi_c \approx 1.5$, which gives an anisotropy of 2.3 and 1.5, respectively.¹⁴

Magnetic torque is a sensitive tool for probing the anisotropy. It has been successfully applied to investigate the highly anisotropic high temperature superconductors and also the less anisotropic materials such as MgB₂.^{15,16} But all these previous torque measurements were made on materials which have negligible paramagnetism. On the other hand, CeCoIn₅ is a magnetic superconductor, so it may have large paramagnetism which cannot be ignored in the study of the mixed state. Here, we report torque measurements on single crystals of CeCoIn₅ both in the normal state and the superconducting state. Our results show large paramagnetism in this material in the normal state. Therefore, we assume that there are two contributions to the torque signal in the mixed state of CeCoIn₅ single crystals: one coming from paramagnetism and the other one coming from vortices. We determined γ from the reversible part of the vortex signal and

found that γ is not a constant, instead, it is field and temperature dependent. This provides evidence that the picture in this unconventional superconductor is not a simple single band scenario, supporting the conclusions of Rourke *et al.*¹⁰ and Tanatar *et al.*¹²

II. EXPERIMENTAL DETAILS

High quality single crystals of CeCoIn₅ were grown using a flux method. The superconducting transition temperature T_c , defined as the value of T at zero resistivity, is 2.3 K. The surfaces of the single crystals were etched in concentrated HCl for several hours and then thoroughly rinsed in ethanol in order to remove the indium present on the surface. The mass of the single crystal for which the data are shown is 0.75 mg. Angular dependent measurements of the magnetic torque experienced by the sample of magnetic moment M in an applied magnetic field H , were performed over a temperature range $1.9 \text{ K} \leq T \leq 20 \text{ K}$ and applied magnetic field range $0.1 \text{ T} \leq H \leq 14 \text{ T}$ using a piezoresistive torque magnetometer. In this technique, a piezoresistor measures the torsion, or twisting, of the torque lever about its symmetry axis as a result of the magnetic moment of the single crystal. The sample was rotated in the applied magnetic field between $H \parallel c$ -axis ($\theta=0^\circ$) and $H \parallel a$ -axis ($\theta=90^\circ$) and the torques τ_{inc} and τ_{dec} were measured as a function of increasing and decreasing angle, respectively, under various temperature-field conditions.

The contributions of the gravity and puck to the total torque signal were measured and subtracted from it. To measure the background torque due to gravity, we measured the torque signal at different temperatures in zero applied magnetic field with the sample mounted on the puck. The gravity torque is almost temperature independent and it is negligible at high applied magnetic fields. However, as the applied magnetic field decreases, the total torque signal becomes smaller and the effect of gravity becomes important, hence should be subtracted from the measured torque. To determine the contribution of the puck to the measured torque, we measured the torque without the single crystal on the puck at different magnetic fields and temperatures. The magnitude of the torque of the puck increases with increasing magnetic field. Also, the contribution of the puck to the measured torque is much larger than the contribution of the gravity. Therefore, the former contribution should always be subtracted from the total measured torque signal.

III. RESULTS AND DISCUSSION

Previous torque studies of Tl₂Ba₂CuO_{6+ δ} (Ref. 17) and MgB₂ (Refs. 15 and 16) systems have shown that the normal state torque coming from paramagnetism is small compared with the flux-vortex torque, therefore, one could neglect the former contribution to the total torque signal measured in the superconducting state. However, this is not the case for CeCoIn₅, for which the paramagnetic torque signal in the normal state is comparable with the total torque signal measured in the superconducting state, as shown below. Hence, one needs to subtract the former signal from the latter one

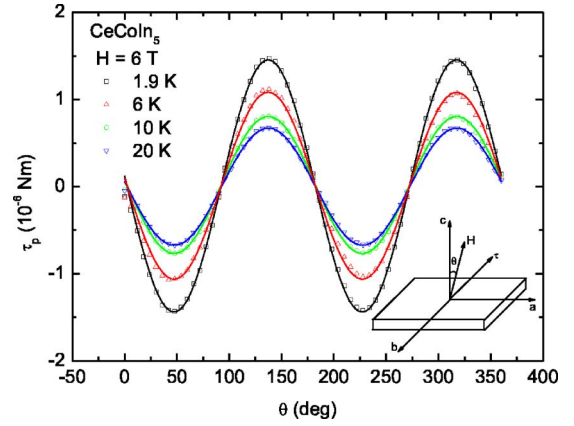


FIG. 1. (Color online) Angular θ dependence of the paramagnetic torque τ_p measured in the normal state of CeCoIn₅ at different temperatures T and applied magnetic field H values. The solid lines are fits of the data with Eq. (1). Inset, sketch of the single crystal with the orientation of the magnetic field H and torque τ with respect to the crystallographic axes.

in order to determine the torque due to vortices. This is similar to the case of the electron-doped high- T_c cuprate Nd_{1-x}Ce_xCuO₄, where a large paramagnetic contribution from Nd ions is discussed separately from a superconducting contribution.¹⁸ Therefore, we first discuss the field and temperature dependence of the magnetic torque in the normal state and show how we subtract this contribution from the measured torque in the mixed state, and then we return to the discussion of the torque signal in the mixed state and to the determination of the field and temperature dependence of the bulk anisotropy.

All the torque curves measured in the normal state, some of which are shown in Fig. 1, are perfectly sinusoidal and can be well fitted with

$$\tau_p(T, H, \theta) = A(T, H) \sin 2\theta, \quad (1)$$

where A is a temperature and field dependent fitting parameter. Indeed, note the excellent fit of the data of Fig. 1 with Eq. (1) (solid lines in the figure). The field dependence of A/H at 1.9, 6, 10, and 20 K is shown in Fig. 2. The solid lines are linear fits to the data, which show that A/H is linear in H with a negligible y-intercept and a slope which increases with decreasing T . So A is proportional to H^2 , i.e.,

$$A(T, H) = C(T)H^2, \quad (2)$$

with C a temperature dependent fitting parameter.

Next we show that the torque measured in the normal state and given by Eq. (1) is a result of the paramagnetism. Indeed, the torque of a sample of magnetic moment M placed in a magnetic field H is given by

$$\vec{\tau}_p(T, H) = \vec{M} \times \vec{H}. \quad (3)$$

The resultant magnetic moment \vec{M} can always be decomposed into a component parallel M_{\parallel} and one perpendicular M_{\perp} to the ab -plane of the single crystal. With the magnetic field H making an angle θ with the c -axis of the single crystal, Eq. (3) becomes

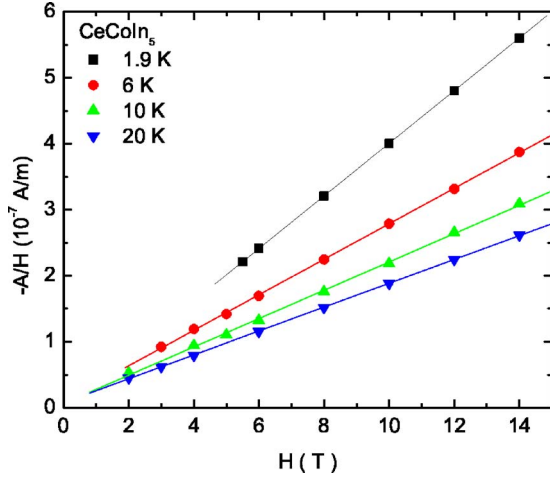


FIG. 2. (Color online) Field H dependence of A/H , where A is the fitting parameter in Eq. (1). The solid lines are linear fits of the data.

$$\vec{\tau}_p(T, H, \theta) = (M_{\parallel} H \cos \theta - M_{\perp} H \sin \theta) \hat{k}. \quad (4)$$

On the other hand, the experimental relationship of the torque, given by Eq. (1) with the fitting parameter A given by Eq. (2), becomes

$$\tau_p(T, H, \theta) = A(T, H) \sin 2\theta = 2C(T)H^2 \sin \theta \cos \theta. \quad (5)$$

Therefore, with $C \equiv (C_1 - C_2)/2$, Eqs. (4) and (5) give

$$M_{\parallel} = C_1 H \sin \theta \equiv \chi_a H_{\parallel},$$

where $C_1 \equiv \chi_a$, the a -axis susceptibility, and

$$M_{\perp} = C_2 H \cos \theta \equiv \chi_c H_{\perp}, \quad (6)$$

where $C_2 \equiv \chi_c$, the c -axis susceptibility. This shows that the torque measured experimentally is of the form

$$\tau_p(T, H, \theta) = \frac{\chi_a - \chi_c}{2} H^2 \sin 2\theta. \quad (7)$$

The fact that $A/H = (\chi_a - \chi_c)H/2 \equiv (M_{\parallel} - M_{\perp})/2$ [see Eqs. (5) and (7)] shows that A/H plotted in Fig. 2 reflects the anisotropy of the magnetic moments along the two crystallographic directions, a and c , while its linear field dependence shows that the magnetic moments are linear in H , hence the susceptibilities along these two directions are field independent. The temperature dependence of the magnetic moments is given by the temperature dependence of the parameter C . Therefore, Eq. (7) shows that the T , H , and θ dependences of the torque measured in the normal state of CeCoIn_5 reflect its paramagnetism and the anisotropy of its susceptibility $\Delta\chi \equiv \chi_a - \chi_c$ along the a and c directions.

To further check the consistency of the data and to precisely determine the paramagnetic value of the torque, we also measured the magnetic moment M of the same single crystal of CeCoIn_5 using a superconducting quantum interference device (SQUID) magnetometer. The magnetic moments measured at 4, 6, 10, 15, and 20 K are plotted in the main panel of Fig. 3 as a function of the applied magnetic field for both $H \parallel c$ -axis ($\theta = 0^\circ$) and $H \parallel a$ -axis ($\theta = 90^\circ$). The

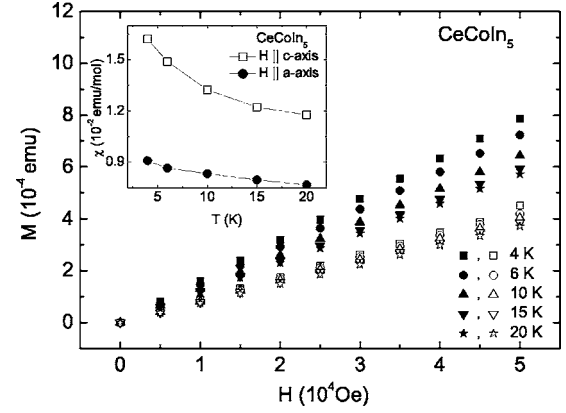


FIG. 3. Plot of the magnetic moment M vs applied magnetic field H , with $H \parallel c$ -axis (solid symbols) and $H \parallel a$ -axis (open symbols), measured at 4, 6, 10, 15, and 20 K. Inset, susceptibility χ vs temperature T , measured with $H \parallel c$ -axis and $H \parallel a$ -axis.

magnetic moments for both field orientations are linear in H with $M_{\parallel} < M_{\perp}$ for all temperatures measured, consistent with the torque data of Fig. 2 and with Eq. (6). Note that the units for the magnetic moments of Figs. 2 and 3 are different. We change the units and compare the results of the two types of measurements. For example, the torque measured at 6 K and 5 T gives $A/H = -1.42 \times 10^{-7} \text{ A m}^{-1}$. Since $A/H = (M_{\parallel} - M_{\perp})/2$, $\Delta M \equiv M_{\parallel} - M_{\perp} = -2.84 \times 10^{-7} \text{ A m}^{-1}$. The SQUID measurements give at the same temperature and applied magnetic field $M_{\perp} = 7.2 \times 10^{-4} \text{ emu} = 7.2 \times 10^{-7} \text{ A m}^{-1}$ and $M_{\parallel} = 4.2 \times 10^{-4} \text{ emu} = 4.2 \times 10^{-7} \text{ A m}^{-1}$; hence, an anisotropy $\Delta M = -3.03 \times 10^{-7} \text{ A m}^{-1}$. Therefore, the values of the magnetic moments obtained in the two types of measurements are within 5% of each other, an error well within our experimental error.

The inset to Fig. 3 is a plot of the susceptibilities along the two directions, calculated from the slopes of $M(H)$ of Fig. 3. Clearly $\chi(T)$ shows anisotropy with respect to the field orientation. Note that the susceptibilities for both directions increase with decreasing temperature. The continuous increase of $\chi(T)$ in the investigated temperature range may be related with the non-Fermi liquid behavior due to the proximity to the quantum critical field.¹⁹ These values of χ_c and χ_a are consistent with the ones reported by other groups.⁷

The above study of the magnetic torque in the normal state has shown that the contribution of paramagnetism to the torque signal is very large, it has a quadratic H dependence, and also a T dependence [see Eqs. (1) and (2)]. Therefore, to extract the vortex torque in the mixed state, one needs to account for this paramagnetic contribution and subtract the two background contributions from the measured torque. The gravity and puck contributions to the measured torque were determined and subtracted as explained in the Experimental details section. The resultant torque includes the paramagnetic τ_p and the vortex τ_v contributions and is plotted in the inset to Fig. 4. We need to mention here that the vortex and paramagnetic torque contributions have opposite signs since the magnetic moment representing the vortex torque is diamagnetic.

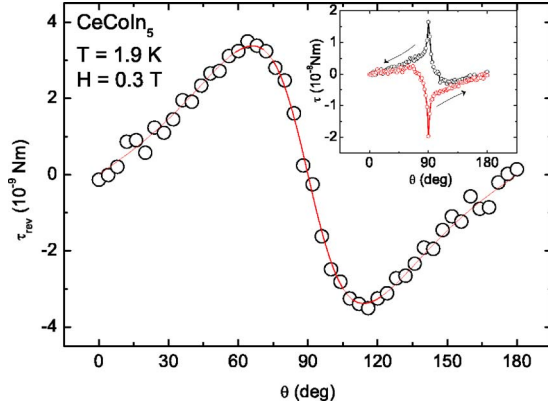


FIG. 4. (Color online) Angular θ dependence of the reversible torque τ_{rev} , measured in the mixed state of CeCoIn₅ at a temperature T of 1.9 K and an applied magnetic field H of 0.3 T. The solid line is a fit of the data with Eq. (9). Inset, θ dependence of the hysteretic torque τ , measured in increasing and decreasing angle at the same T and H .

The inset to Fig. 4 is the angular-dependent torque measured in the mixed state at 1.9 K and 0.3 T in increasing and decreasing angles. Again, this torque includes τ_p and τ_v . Note that $\tau(\theta)$ displays hysteresis. This hysteretic behavior is similar to the behavior in high T_c superconductors and is a result of intrinsic pinning.²⁰ The reversible component of the torque is determined as the average of the torques measured in increasing and decreasing angle; i.e.,

$$\tau_{\text{rev}} = (\tau_{\text{dec}} + \tau_{\text{inc}})/2. \quad (8)$$

A plot of $\tau_{\text{rev}}(\theta)$, obtained from $\tau_{\text{inc}}(\theta)$ and $\tau_{\text{dec}}(\theta)$ data is shown in the main panel of Fig. 4. The reversible component of the torque reflects equilibrium states, hence it allows the determination of thermodynamic parameters. In the three-dimensional anisotropic London model in the mixed state, the vortex torque τ_v is given by Kogan's model.²¹ We assume that the paramagnetic contribution in the mixed state is given by Eq. (1). Therefore,

$$\begin{aligned} \tau_{\text{rev}}(\theta) &= \tau_p + \tau_v \\ &= a \sin 2\theta + \frac{\phi_0 H V}{16\pi\mu_0\lambda_{ab}^2} \frac{\gamma^2 - 1}{\gamma} \frac{\sin 2\theta}{\epsilon(\theta)} \ln \left(\frac{\gamma\eta H_{c2}^{\parallel c}}{H\epsilon(\theta)} \right), \end{aligned} \quad (9)$$

where a is a fitting parameter, V is the volume of the sample, μ_0 is the vacuum permeability, λ_{ab} is the penetration depth in the ab -plane, $\gamma = \sqrt{m_c/m_a}$, $\epsilon(\theta) = (\sin^2\theta + \gamma^2 \cos^2\theta)^{1/2}$, η is a numerical parameter of the order of unity, and $H_{c2}^{\parallel c}$ is the upper critical field parallel to the c -axis [$H_{c2}^{\parallel c}(1.9 \text{ K}) = 2.35 \text{ T}$]. We define $\beta \equiv \phi_0 H V / (16\pi\mu_0\lambda_{ab}^2)$.

To obtain the field dependence of the anisotropy γ , we fit the torque data with Eq. (9), with a , β , and γ as three fitting parameters. The solid line in the main panel of Fig. 4 is the fitting result for $T=1.9 \text{ K}$ and $H=0.3 \text{ T}$. We need to mention here that the value of the fitting parameter a is 20% smaller than what we would expect from the extrapolation of the normal state paramagnetic torque data. Also, a has an H

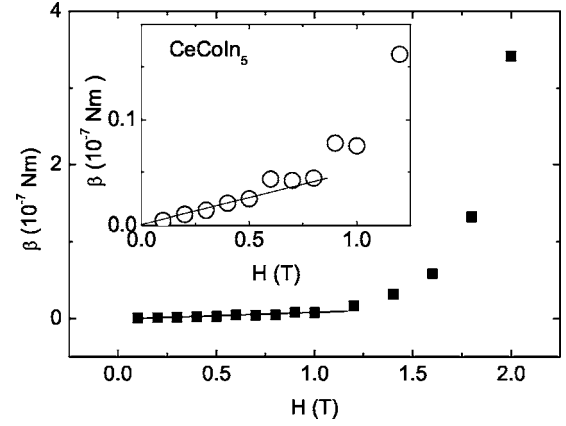


FIG. 5. Magnetic field H dependence of the fitting parameter β . The solid line is a guide to the eye. Inset, enlarged plot of the low field region of the data in the main panel.

dependence with an exponent of 2.3 instead of 2. So, either there is extra contribution from other physics which has a weak field dependence, in addition to the paramagnetism contribution, or maybe the paramagnetic contribution becomes smaller in the mixed state of CeCoIn₅. Further experiments are needed to clarify this issue.

Figure 5 is a plot of the field dependence of β . The Inset is an enlarged plot of the low field region. Note that β displays linear behavior up to a certain field with no y -intercept, then it deviates from linearity at $H \approx 0.5 \text{ T}$, and it increases fast in the high field region. Since, on the one hand λ should be field independent, and on the other hand, Eq. (9) is valid only for applied magnetic fields much smaller than the upper critical field, i.e., $H \ll H_{c2}(T)$ for a given temperature, we assume that 0.5 T, the field at which $\beta(H)$ deviates from linearity, is the cutoff field H_{cut} for the applicability of the above theory. The slope of $\beta(H)$ in the linear H regime gives $\lambda(T=1.9 \text{ K}) = 787 \text{ nm}$. This value is larger than previous reports, which give $\lambda_{ab} = 600 \text{ nm}$ from measurements using a tunnel diode oscillator²² and $\lambda_{ab} = 330 \text{ nm}$ from magnetization measurements.¹⁴

Next, we fix λ to the three values given above and fit the $\tau_{\text{rev}}(\theta)$ data with only two fitting parameters, a and γ . The resultant field dependence of γ is shown in Fig. 6 for the different λ values. The parameter γ is first decreasing with increasing H , reaches a minimum at $H=0.5 \text{ T}$, and then increases with further increasing field. As mentioned above, the cutoff field is 0.5 T. The data for $H > H_{\text{cut}}$ are not reliable due to the failure of Kogan's theory in this H region. So we conclude that the anisotropy γ decreases with increasing field. We note that this field dependence of γ in CeCoIn₅ is opposite with the one for MgB₂,¹⁶ in which γ increases with increasing field. We found that the value of γ is very sensitive to the value of λ , i.e., the larger the value of λ , the larger the value of γ , with no effect however on its H dependence.

To study the temperature dependence of γ , we performed torque measurements in the mixed state at 1.9 K, 1.95, 2.00 K in an applied magnetic field of 0.3 T, and determined γ from Eq. (9). Figure 7 is a composite plot of the temperature dependence of the anisotropy. The squares give $\gamma(T)$ determined from torque measurements as discussed above

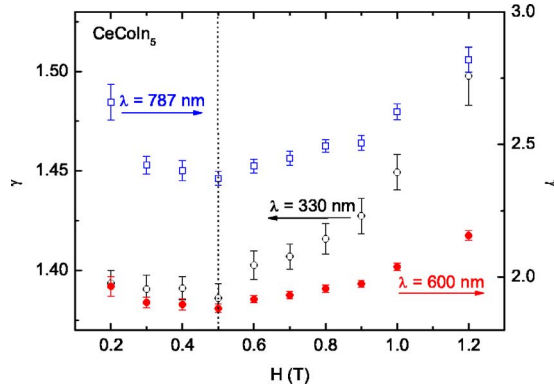


FIG. 6. (Color online) Field H dependence of the anisotropy γ measured at 1.9 K.

[with λ (1.9 K)=600 nm, λ (1.95 K)=670 nm, and λ (2 K)=740 nm taken from Ref. 25, the solid circles give $\gamma(T)$ calculated from the ratio of $H_{c2}^{\parallel c}/H_{c2}^{\parallel a}$ taken from previous reports,²³ while the triangles give $\gamma(T)$ determined from $\sqrt{\rho_c(T)/\rho_a(T)}$ measured in zero field [see $\rho_c(T)$ and $\rho_a(T)$ in inset to Fig. 7]. Note that the overall trend is a decrease of the anisotropy with increasing T , with a stronger dependence around $T_c(0.3 \text{ T})=2.23 \text{ K}$. The values of γ obtained from torque measurements are well within the range previously reported. The fact that the anisotropy depends both on temperature and magnetic field could explain its relatively wide range of values reported in the literature.

The field and temperature dependence of the anisotropy implies the breakdown of the standard anisotropic GL theory, which assumes a single band anisotropic system with a temperature and field independent effective-mass anisotropy. In fact, a temperature and field dependent anisotropy was previously observed in NbSe_2 ,²⁴ $\text{LuNi}_2\text{B}_2\text{C}$,²⁵ and MgB_2 ,^{16,26} the latter one having a value of γ similar with CeCoIn_5 . Very recently, Fletcher *et al.* reported measurements of the temperature-dependent anisotropies (γ_λ and γ_ξ) of both the London penetration depth λ and the upper critical field of MgB_2 .²⁷ Their main result is that the anisotropies of the penetration depth and H_{c2} in MgB_2 have opposite temperature dependences, but close to T_c they tend to a common value (see Fig. 4 of Ref. 27). This result confirms nicely the theoretical calculation of anisotropy based on a two-gap scenario.²⁸ So, the temperature dependence of γ in CeCoIn_5 and MgB_2 could have a similar origin. The field dependence of γ could also be related with a multiband structure. In this scenario, the response to an applied magnetic field may be different in the case of the d -wave superconducting gap on the heavy-electron sheets of the Fermi surface and the negligible gap on the light, three-dimensional pockets.

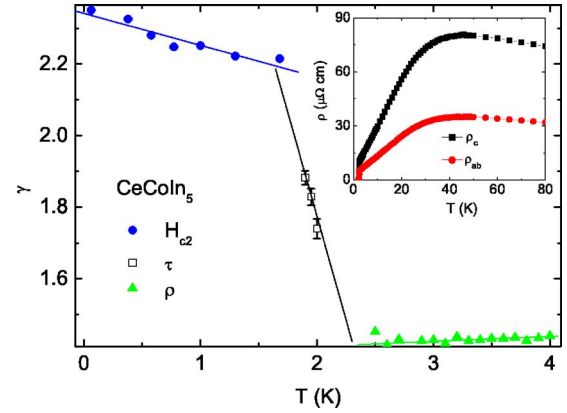


FIG. 7. (Color online) Composite plot of the temperature T dependence of the anisotropy γ . The circles show the results obtained from the upper critical field data (Ref. 26), the triangles are obtained from the resistivity data shown in the inset of this figure, and the squares are from the present torque data measured in an applied magnetic field of 0.3 T. Inset, T dependence of the in-plane ρ_a and out-of-plane ρ_c resistivities measured in zero field.

IV. SUMMARY

Torque measurements were performed on CeCoIn_5 single crystals in both the superconducting and normal states. Two contributions to the torque signal in the mixed state were identified: one from paramagnetism and the other one from the vortices. The torque curves show sharp hysteresis peaks at $\theta=90^\circ$ (θ is the angle between H and the c -axis of the crystal) when the measurements are done in clockwise and counterclockwise directions. This hysteresis is a result of the intrinsic pinning of vortices, a behavior very similar to high transition temperature cuprate superconductors. The temperature and magnetic field dependence of the anisotropy γ is obtained from the reversible part of the vortex torque. We find that γ decreases with increasing magnetic field and temperature. This result indicates the breakdown of the Ginzburg-Landau theory, which is based on a single band model and provides evidence for a multiband picture for CeCoIn_5 , which is highly possible due to its complex Fermi topology.^{8,9}

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