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Complex mixed state of the Pauli-limited superconductor CeCoIn₅

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Magnetization measurements were performed on CeCoIn₅ at temperatures down to 20 mK and magnetic fields up to 17 T applied along different crystallographic orientations. For field configurations nearly parallel to the *ab* plane ($\theta \lesssim 40^\circ$ and $T \leq 50$ mK), we have found an intriguing vortex dynamics regime revealed by a hysteretic and metastable anomalous peak effect (APE), which gives evidence of surface barrier effects enhanced by antiferromagnetic fluctuations in the mixed state of CeCoIn₅. Furthermore, we have observed crossover features in the torque and magnetization traces at fields below H_{c2} , which are consistent with vortices lattice phase transitions and with the anomalies speculated to be the Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) superconducting state in CeCoIn₅. All of the above features were found to be dramatically perturbed in Ce_{0.98}Gd_{0.02}CoIn₅.

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I. INTRODUCTION

The heavy-fermion superconductor (HFS) CeCoIn₅ is a clean unconventional superconductor ($T_c = 2.3$ K) that has been intensively investigated in last decade owing to several unusual properties of its superconducting (SC) state.^{1,2} For instance, it was established that the transition at the upper critical field H_{c2} changes from second to first order below a temperature T_0 ,³⁻⁵ which is considered strong evidence that CeCoIn₅ is a Pauli-limited superconductor.³⁻⁵ In addition, a number of studies controversially claimed that CeCoIn₅ exhibits the inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) SC state near H_{c2} ,⁴⁻¹⁰ which has been theoretically predicted^{11,12} but never unambiguously observed.

More recently, small-angle neutron scattering (SANS) experiments on CeCoIn₅ have revealed a complex phase diagram and an anomalous field dependence of the form factor, which is not consistent with the Abrikosov-Ginzburg-Landau scenario.¹³

These investigations motivate many theoretical models in an attempt to describe the complex low- T high- H mixed state of CeCoIn₅.¹⁴⁻¹⁷ Some of them take into consideration the presence of antiferromagnetic (AFM) fluctuations.¹⁷ In fact, high-field neutron diffraction and nuclear magnetic resonance results provided evidence of a magnetically ordered phase within the mixed state when the field is along the tetragonal basal plane.^{18,19}

The complexity of the mixed state of the HFS CeCoIn₅ is unprecedented for any type-II SC material, and its vortex dynamics have not been explored in detail by magnetization measurements. In fact, even concerning the occurrence of the FFLO state, results from magnetization studies are contradictory.^{3,4,20} Here we report magnetization studies on single crystals of CeCoIn₅ and Ce_{0.98}Gd_{0.02}CoIn₅ at temperatures down to 20 mK and fields up to 17 T applied along different crystallographic orientations.

II. EXPERIMENT

The single crystals used in this work were grown by In self-flux, and their phase purity and SC transition were checked, respectively, by x-ray diffraction and zero-field heat capacity experiments. The zero-field superconducting temperature $T_c = 2.3$ K and the heat capacity jump at T_c $\Delta C/\gamma T_c \approx 5$ for pure CeCoIn₅, and $T_c = 2.1$ K and the heat capacity jump at T_c $\Delta C/\gamma T_c \approx 4$ for Ce_{0.98}Gd_{0.02}CoIn₅ were in perfect agreement with the literature.^{1,5,21}

Our magnetization experiments were carried out in a diaphragm force magnetometer, and the sample magnetic response was detected by a capacitance technique. The measurement technique used in this work is different from that in Refs. 3 and 20, revealing new features of the magnetization of CeCoIn₅. Our experiments were carried out using a diaphragm force magnetometer^{22,23} inside a plastic diluted refrigerator operating in a 20-T SC magnet (see details in Refs. 22 and 23). The magnetic force on the sample was produced by a field gradient ($630 \text{ Oe/cm} \leq dH_z/dz \leq 1.8 \text{ kOe/cm}$) superimposed on the main magnetic field, and the sample magnetic response was detected by a capacitance technique. The contribution of the magnetic response caused by the torque was determined by repeating the measurement with no current in the gradient coil. From one set of runs (with and without field gradient), we were able to extract the component of the magnetization parallel to the magnetic field (M_z) by a simple subtraction of the contribution of the torque from the total response for $dH_z/dz \neq 0$. This is not clearly the case by cantilever measurements.²⁰ Our experimental method is similar to that in Ref. 3, but the force magnetometers are different. In the present work, the movable capacitor plate of the magnetometer is a diaphragm, which gives a stronger response to the torque than the apparatus used by Sakakibara *et al.*²⁴ The data were taken for increasing and decreasing magnetic field ($|dH/dt| \approx 35 \text{ Oe/s}$) after zero-field cooling the sample from well above T_c .

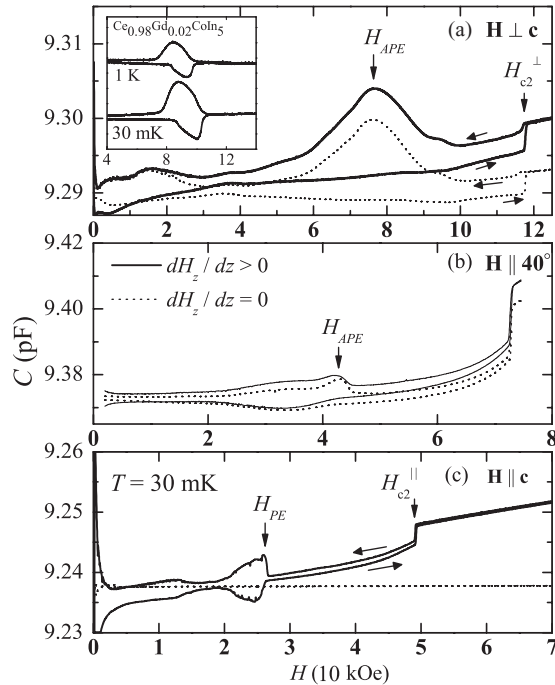


FIG. 1. Capacitance $C(H)$ loops for CeCoIn_5 at 30 mK taken with and without field gradient for (a) $\mathbf{H} \perp \mathbf{c}$, (b) $\mathbf{H} \parallel 40^\circ$, and (c) $\mathbf{H} \parallel \mathbf{c}$. The inset shows a similar set of data obtained at 30 mK and 1 K for $\text{Ce}_{0.98}\text{Gd}_{0.02}\text{CoIn}_5$ with $\mathbf{H} \perp \mathbf{c}$.

III. RESULTS AND DISCUSSION

Figure 1 shows the capacitance response of CeCoIn_5 for increasing and decreasing magnetic field at $T = 30$ mK and three orientations, measured with and without field gradient (dH_z/dz). For $\mathbf{H} \perp \mathbf{c}$ [Fig. 1(a)], \mathbf{M} is clearly not aligned to the field direction. The response with $dH_z/dz \neq 0$ shows two contributions: one caused by the torque (measured for $dH_z/dz = 0$) and the other due to M_z . For both up-sweep traces (with and without field gradient), the traces show a sharp jump at H_{c2}^\perp due to the first-order- superconducting normal-state transition (FOSNT). The data also show the existence of a broad peak [which we called the anomalous peak effect (APE)²⁵] at H_{APE} that shows a very intriguing behavior: it is observed *only* with decreasing field. The inset presents data for the $\text{Ce}_{0.98}\text{Gd}_{0.02}\text{CoIn}_5$ crystal, which we will discuss later.

Figure 1(b) shows data for H rotated 40° from the ab plane. The magnetic behavior is qualitatively the same as for $\mathbf{H} \perp \mathbf{c}$, but the APE becomes asymmetric and it is shifted to a lower field, while a small bump starts to develop on sweeping up. When $\mathbf{H} \parallel \mathbf{c}$ [Fig. 1(c)], the response for $dH_z/dz \neq 0$ is due only to the magnetization (\mathbf{M}) parallel to the field direction ($\mathbf{M} = M_z$). (The capacitance response is constant as a function of the field for $dH_z/dz = 0$.) The FOSNT manifests itself by a sharp jump at H_{c2}^\parallel . At lower fields we observed a hysteric peak effect (PE) near 25 kOe, which follows the same trend reported in Ref. 3.

The angular dependence of the magnetic behavior of CeCoIn_5 at $T = 30$ mK in the vicinity of H_{c2} can be seen in Fig. 2. The magnetization traces for $\mathbf{H} \parallel \mathbf{c}$ and $\mathbf{H} \perp \mathbf{c}$

are shown in Figs. 2(a) and 2(b), respectively. The sharp jump due to the FOSNT is clearly observed for both traces. From our data, we obtained $H_{c2}^\parallel = 49.2$ kOe and $H_{c2}^\perp = 117.7$ kOe with a hysteresis width of $\Delta H_{c2} \approx 0.4$ kOe, in very good agreement with the literature. In addition, for both directions, we have newly observed a change in the monotonic variation of the magnetization near H_{c2} . This crossover to a high-field phase (HFP) manifests itself in the derivative (dM_z/dH) trace by a step. The observed singularity takes place at a field H_{HFP} indicated by the vertical arrows in Figs. 2(c) and 2(d) for data obtained at different temperatures. For $\mathbf{H} \parallel \mathbf{c}$, H_{HFP} is found to be nearly independent on T . On the other hand, for $\mathbf{H} \perp \mathbf{c}$, H_{HFP} is clearly shifted to higher fields for increasing temperature. The angular evolution of the characteristic high- H anomalies as seen in the derivative traces are shown in Figs. 2(e) and 2(f).

To further explore the nature of the APE we have performed additional experiments. First, we repeated the measurements shown in Fig. 1 for H rotated 20° from the basal plane. Figure 3(a) shows the results for $dH_z/dz = 0$ up- and down-sweep traces at $T = 20$ and 50 mK. According to our data, the APE is rapidly suppressed by thermal effects and it is absent for $T > 50$ mK. Second, as the anomalous peak apparently starts to develop in the vicinity of the HFP crossover feature, we repeated the measurement at 20 mK by sweeping the field up to the value where the anomalous peak is centered, and then sweeping it down, that is, without passing through the HFP anomaly [dashed curve in Fig. 3(a)]. As can be seen, the up-sweep trace follows exactly the same behavior previously obtained, and, on reversing the sweeping direction, the anomalous peak is still observed but its size is smaller. This is strong evidence that the APE is not related to the high-field phase.

We also performed time relaxation measurements for selected H values in the region where the APE is observed ($0^\circ \leq \theta \leq 40^\circ$) and at the hysteretic peak found for $\mathbf{H} \parallel \mathbf{c}$. Before each measurement, the field was swept up to 140 kOe, subsequently swept down to the target value, and then the signal was measured as a function of time. The sweep rate was identical in all these measurements. The time-dependent $C(t) \propto \tau(t) \propto M(t)$ is depicted in Fig. 3(b) for distinct field orientations.

The results in Fig. 3 indicate that the APE in down-sweep traces for $0^\circ \leq \theta \leq 40^\circ$ is related to relaxation effects, however the APE relaxation time shows a nonmonotonic angle dependence. This result revealed another unusual aspect of the APE that indicates a very peculiar vortex dynamics regime in CeCoIn_5 in this T - H range. For $\mathbf{H} \parallel \mathbf{c}$, no relaxation effect for PE is observed in the same time window.

By mapping the temperature and angular dependencies of the distinct features identified in our $M(T, H)$ data, we constructed the phase diagrams depicted in Fig. 4. Figure 4(a) presents the high- H low- T phase diagram of CeCoIn_5 for $\mathbf{H} \perp \mathbf{c}$ and $\mathbf{H} \parallel \mathbf{c}$. Results from heat capacity⁵ and NMR^{8,26} experiments are also shown for completeness. The evolution of the H_{HFP} line determined from our data for both orientations is in good agreement with the line identified by distinct techniques and claimed by some authors to be the inhomogeneous FFLO SC state. However, from our data we cannot reach any conclusion about the microscopic nature of the HFP.

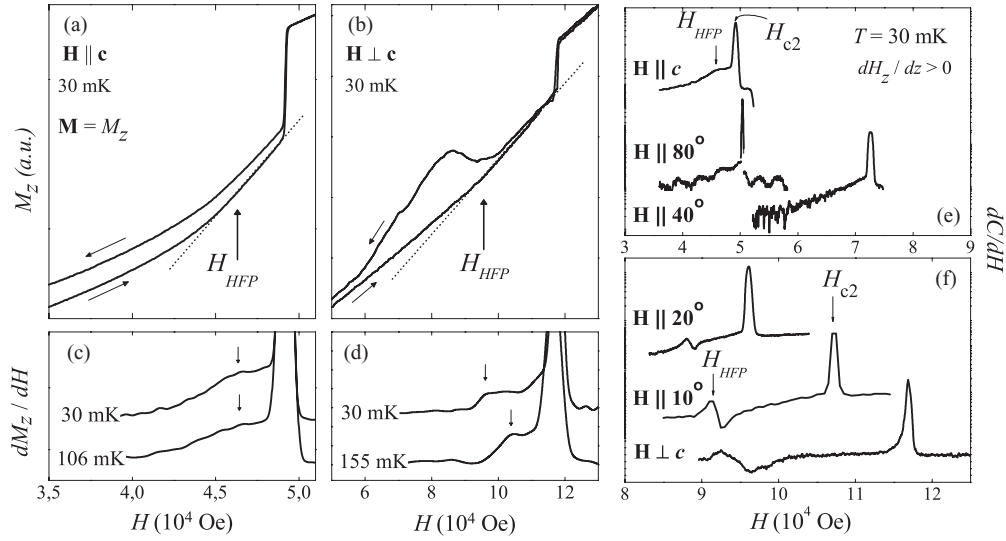


FIG. 2. Magnetic behavior of CeCoIn₅ near H_{c2} . $M_z(H)$ at $T = 30$ mK for (a) $\mathbf{H} \parallel \mathbf{c}$ and (b) $\mathbf{H} \perp \mathbf{c}$. The dotted lines emphasize the change in the monotonic variation of $M_z(H)$ near H_{c2} . For $\mathbf{H} \perp \mathbf{c}$, the APE is seen in the down-sweep trace. The respective dM_z/dH traces are shown in panels (c) and (d) for two temperatures. dC/dH versus H curves near H_{c2} show the angular evolution of the HFP anomaly for orientations (f) out and (e) nearly parallel to the ab plane. The vertical arrows indicate H_{HFP} while the other arrows indicate the direction of field sweep.

Concerning the low- H region of the diagram, when $\mathbf{H} \parallel \mathbf{c}$, the T dependence of the hysteretic peaks obtained from our measurements is consistent with the behavior reported in Ref. 3. As revealed by SANS experiments,¹³ this PE is related to transitions in the vortex lattice.

The θ dependence of the characteristic fields is displayed in Fig. 4(b). According to our data, the HFP anomaly persists only for fields slightly disorientated from the ab plane or from the c axis. In addition, the H - T area of the phase diagram corresponding to the HFP diminishes as the field is rotated away from the ab plane. On the other hand, the APE persists up to higher angles ($\sim 40^\circ$) and it is linearly shifted to lower

fields as the angle is increased, while the hysteretic PE takes place for field orientations almost parallel to the c axis.

Now we discuss the origin of the APE found below the HFP for H (nearly) parallel to the ab plane. In general, magnetic relaxation in type-II superconductors originates from the vortex motion driven by the gradient in the vortex density, ∇n_v , in the presence of vortex pinning (vortex creep regime). For our studies in field configurations (nearly) parallel to the ab plane, the torque is proportional to the c -axis component of the magnetization, $M_c(t)$. Then, in the vortex creep regime, the time-dependent signal $C(t) \propto \tau(t) \propto M_c(t)$. Because CeCoIn₅ is an anisotropic (layered) superconductor, the appearance of vortex kinks along the c axis and their interaction with vortex systems parallel to ab planes should also be taken into account;

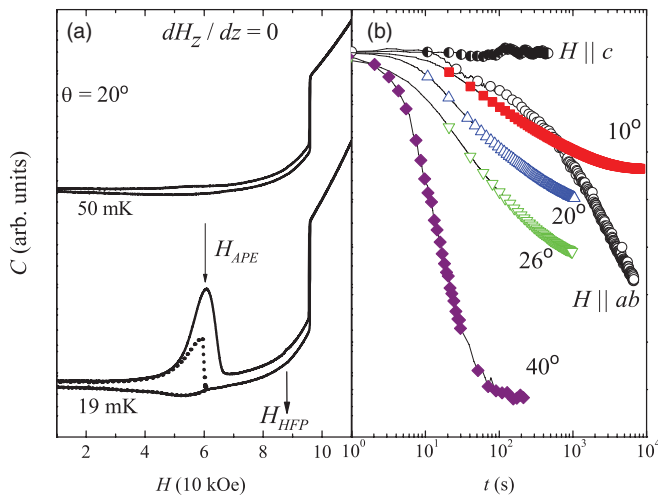


FIG. 3. (Color online) (a) $C(H)$ at two temperatures for $\mathbf{H} \parallel 20^\circ$. The dashed line denotes the trace obtained when reversing the sweeping direction at H_{APE} . (b) Results of $C(t)$ measured at H_{APE} for different orientations of the applied field.

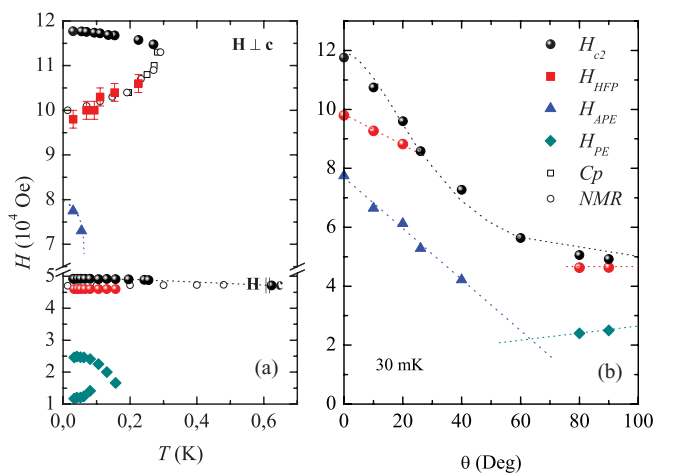


FIG. 4. (Color online) (a) H - T phase diagram of CeCoIn₅ for $\mathbf{H} \perp \mathbf{c}$ and $\mathbf{H} \parallel \mathbf{c}$. It includes previous $c_p(T)$ ⁵ and NMR^{8,26} data. (b) Angular dependence of the characteristic fields at $T = 30$ mK for CeCoIn₅. Dotted lines are guides to the eye.

this would provide information on the pinning of both in- and out-of-plane vortex systems.^{27–29} In principle, our observation of the time relaxation of the measured signal $C(t) \sim M(t)$ in the peak region for $\theta \leq 40^\circ$ (Fig. 3) suggests that the vortex pinning efficiency is enhanced in this field interval. However, in contrast to the more conventional PE behavior observed for $\mathbf{H} \parallel \mathbf{c}$, for which we have peak manifestation in both up and down-sweep traces [see Fig. 1(a) and Ref. 3], the APE measured for $\mathbf{H} \perp \mathbf{c}$ and \mathbf{H} near parallel [see Figs. 1(a) and 3], takes place only under decreasing field at very low T ($T \leq 50$ mK).

In attempts to shed light on the origin of the APE, we note that irreversibility under increasing and/or decreasing field state, characterized by a glassy-like time relaxation, has been also reported for other spin-paramagnetically limited superconductors, such as Al^{30,31} and Be³² films. Interestingly, the low- T high- H portion of the H - T plane (see Fig. 4) resembles very much the metastable H - T phases identified in Al and Be films, where SC and normal (N) states coexist.^{30–34}

Although the pinning mechanisms in Al or Be films certainly differ from that operating in highly pure CeCoIn₅, it is not impossible that the magnetization time relaxation occurring in CeCoIn₅ for \mathbf{H} nearly parallel to the ab planes may be governed by dynamics of coexisting SC and N domains, or possibly by BCS-like superconductivity [vortex state (VS) in our case] and the high-field phase identified as the FFLO phase by some authors. However, such an interpretation can be ruled out since the results presented in Fig. 3 for $\theta \leq 40^\circ$ indicate that the APE takes place well outside of the field hysteresis region associated with the FOSNT.

Irreversibility of the vortex dynamics has also been proposed in the context of a surface barrier.³⁵ In the model by Bean and Livingston,³⁶ two magnetic forces are considered to act on a vortex located near and parallel to the surface of a superconductor. The first one arises from the repulsion by the external magnetic field and tends to push the vortices into the interior of the sample. The second one tends to dominate very close to the surface. It originates from the image vortex on the outside of the surface and is directed in the opposite sense. Therefore, there is a distance from the surface at which the vortex energy is maximum, thus providing a barrier to vortex penetration. In the work by Walton *et al.*,³⁵ the authors discuss how vortex entry (or exit) is modified when the order parameter at the surface is strongly modulated, introducing the idea of *nascent vortices*. They assume that in thermal equilibrium in the mixed state, both the order parameter and the magnetic field at the surface are periodically modulated in the direction perpendicular to the magnetic field. The order parameter at the modulation minima is not necessarily equal to zero. Hence, the nascent vortices are lines of reduced surface order parameter and increased magnetic field penetration. The minima of this modulation act as nucleation and denucleation sites for vortices. Within this picture, irreversibility arises due to the fact that it is easier to get vortices into the sample than to get them

out when decreasing the magnetic field. It is also important to mention that the above scenario can be experimentally realized only in a very clean superconductor, since this effect cannot be detected if the mixed state dynamics is dominated by bulk pinning sites. Therefore highly clean CeCoIn₅ would be a perfect candidate to allow the formation of nascent vortices and reveal the irreversibility of the vortex dynamics dominated by surface barriers, as long as there is a modulated SC order parameter at the surface. Based on experimental evidence for a high-field-induced magnetic phase in CeCoIn₅ given by NMR¹⁹ and neutron diffraction experiments,¹⁸ a modulation of the order parameter at the surface may arise from surface nucleation of a field-induced magnetic phase at fields well below the HFP. Indeed, an x-ray magnetic diffraction study of antiferromagnetic GdIn₃³⁷ demonstrates that a magnetic phase can emerge in a submicrometric near-surface region with a Néel temperature higher than the bulk transition temperature. Therefore, our results allow us to claim that the APE found in this work provides an experimental realization of the nascent vortices and their related vortex dynamics regime. Furthermore, the SC order parameter modulation that keeps vortices parallel to the surface, enhanced by the field-induced AFM ordering nucleated at the surface, allows the observation of the nascent vortices at much higher angles than theoretically predicted.³⁵

Finally, if our interpretation is correct, the features observed in the magnetization curves in CeCoIn₅ should disappear as the sample is driven away from the clean limit. To verify this idea we have performed similar experiments for a Ce_{0.98}Gd_{0.02}CoIn₅ single crystal (inset of Fig. 1). According to our data, Gd doping suppresses the FOSNT at H_{c2}^\perp and the HFP anomalies. The PE becomes conventional due to the stronger pinning and remains to higher T , and magnetic relaxation does not take place for the same temperature and time window (not shown) as measured for pure CeCoIn₅.

IV. CONCLUSION

In conclusion, by torque and magnetization measurements in the mixed state of CeCoIn₅, we have found a peculiar vortex dynamics regime marked by an APE, possibly related to an enhancement of the surface pinning potential induced by AFM fluctuations initially nucleated at the surface. In addition, our data reveals crossover anomalies in the vicinity of H_{c2} that coincide with the $H_{\text{FFLO}}(T)$ line determined by others^{4–7} for $\mathbf{H} \perp \mathbf{c}$ and indicates that this HFP persists for out-of-plane field orientations.

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