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Vortex liquid phase in the *p*-wave ferromagnetic superconductor UCoGe

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The upper critical field for the field along the **b** axis of the orthorhombic ferromagnetic superconductor UCoGe has a particular *S* shape, akin to the reentrant superconducting phase of URhGe. In order to explore the evolution of the superconducting phase under this transverse magnetic field, we report the thermal conductivity and resistivity measurements, revealing a possible field-induced vortex liquid phase, and supporting a field-induced change of the superconducting order parameter.

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I. INTRODUCTION

The homogeneous coexistence of ferromagnetism (FM) and superconductivity (SC) in the orthorhombic strongly correlated systems UGe_2, URhGe, and UCoGe [1–4] has been well established through NMR and muon spectroscopy [5,6]. Both orders emerge from the uranium $5f$ electrons, and the mere existence of a superconducting phase in the presence of the strong exchange field controlling the FM state, as well as the absence of Pauli depairing on their very large upper critical field [7–9], points to *p*-wave spin-triplet SC, with an "equal-spin-pairing" (ESP) state. URhGe and UCoGe share the same crystal structure with the space group $Pnma$ (No. 62, D_{2h}^{16}), and both show superconductivity at ambient pressure. They are weak ferromagnets with the spontaneous magnetization along the **c** axis.

A remarkable property of these ferromagnetic superconductors is the reentrant superconducting phase (RSC) observed in URhGe [10]. When the magnetic field is applied along (**H**//**b**), transverse to the easy magnetization axis, two SC phases are revealed: SC is first suppressed at around 2 T, and reappears at fields around 12 T, with an even higher transition temperature. The RSC in URhGe has a direct interplay with a field-induced FM instability at $H_R \sim 12$ T, for which the Curie temperature (T_{Curie}) decreases to zero (weak first-order transition) and the magnetic moments reorient completely along the applied field direction [10]. Despite the intense experimental and theoretical studies [11–16], it is not known whether the superconducting order parameter has the same symmetry in the low field and in the RSC phase. In the sister compound UCoGe, the situation is similar for the same field direction **H**//**b**: SC is reinforced and the upper critical field (H_{c2}) has an *S* shape [9]. Recent work [17] shows that in URhGe, under uniaxial stress larger than 0.2 GPa applied along the **b** axis, the low-field and RSC phases merge into a single phase, as in UCoGe. Despite the remarkable similarity between the two systems, no spin reorientation has been detected in UCoGe, and the mechanism for the *S*-shape H_{c2} also remains unknown, as well as the field-induced evolution of the SC order parameter. The question of field-induced phase transitions is recurrent for triplet superconductors: they have been observed in superfluid ^{3}He (A1 phase) [18] and UPt_3 [19], and in the well-known possible chiral *p*-wave superconductor Sr_2RuO_4 [20]. Here we show that for UCoGe, there is also strong experimental support for a deep change of the superconducting order parameter under transverse field.

II. EXPERIMENTAL

We report on thermal conductivity (κ) and resistivity (ρ) measurements in UCoGe under transverse magnetic field **H**//**b**, obtained on a first single crystal (sample 1), grown with the Czochralski pulling method in a tetra-arc furnace. It is bar-shaped with the heat current flowing along the **c** axis (same as used in Ref. [21], labeled S_{16}^c, and in Ref. [22]). It has a residual resistance ratio (RRR) of 16. κ has been measured down to 150 mK in a dilution refrigerator, and in magnetic fields **H**//**b** up to 15 T. We used the usual one-heater-two-thermometer method, and 15 μm diameter gold wires, spot-welded on the sample, to realize the thermal links. The temperature rise was limited to ∼1%. Four-wire ac-resistivity measurements were performed at the same time, through the same gold wires, allowing direct comparison of thermal and charge transport. A two-axis Attocube piezo alignment system (a goniometer $\sim \pm 3^\circ$, and a rotator $\sim 360^\circ$) has been used to orient *in situ* the sample **b** axis along the magnetic field (with a sensitivity better than 0.05°), by optimizing the T_{sc} of the resistive transition under field. Resistivity has also been measured on a second single crystal (sample 2, RRR 35, same growth technique and same geometry) for magnetic fields up to 16 T for **H**//**b**.

III. RESULTS

Figure 1 presents H_{c2} for **H**//**b** obtained from simultaneous thermal conductivity and resistivity measurements on sample 1. Raw data are presented in the Supplemental Material [23]. The rather low sample quality (RRR = 16) was chosen on purpose, to get a clear signature of the superconducting

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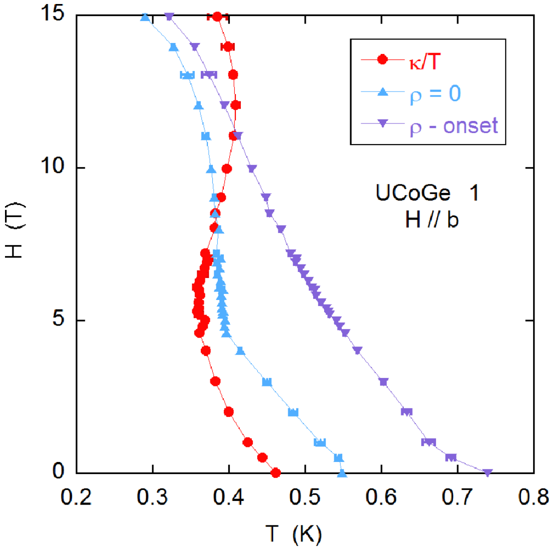

FIG. 1. H_{c2} for **H//b** of UCoGe, probed with thermal conductivity (red circles, shown already in Ref. [22]) and resistivity (blue triangles for $T_{\rho=0}$ and purple triangles for the onset), measured on sample 1.

transition in the thermal conductivity (suppression of the inelastic scattering [21]). For a precise determination of the transition temperature, κ/T data were first analyzed to extract the electronic quasiparticle from the phonon and magnon contributions, and then fitted to extract T_{sc} (same procedure as explained with more details in Ref. [22]). The resistive transitions were also fitted to extract systematically T_{sc} from an onset or $\rho = 0$ criterion. Figure 2 presents the SC transition on the thermal conductivity ($\kappa_{\text{elect}}/\kappa_n$), and on the resistivity (ρ/ρ_n) at 14 T. The same data at a few other selected fields are presented in the Supplemental Material [23]. Below ~8 T, the resistivity transition occurs at a temperature higher than the (bulk) superconducting transition on the thermal conductivity, for both the onset and the $\rho = 0$ criteria. This is usual, as any filamentary superconducting path in the sample can induce a resistivity drop, before the bulk superconductivity occurs. What is less usual, but well known for this system, is the fact that the resistive T_{sc} can be much larger than the bulk T_{sc} (almost a factor 2 for the onset criterion). A side effect of this very large resistive transition width is that the temperature dependence of H_{c2} deduced from the resistivity is strongly criterion-dependent, and is also known to be sample-dependent (notably the "S shape"). This was notably a major motivation for a bulk determination of H_{c2} [22]. Recently, the S-shape H_{c2} for **H//b** has also been confirmed by another bulk probe: thermal expansion [24].

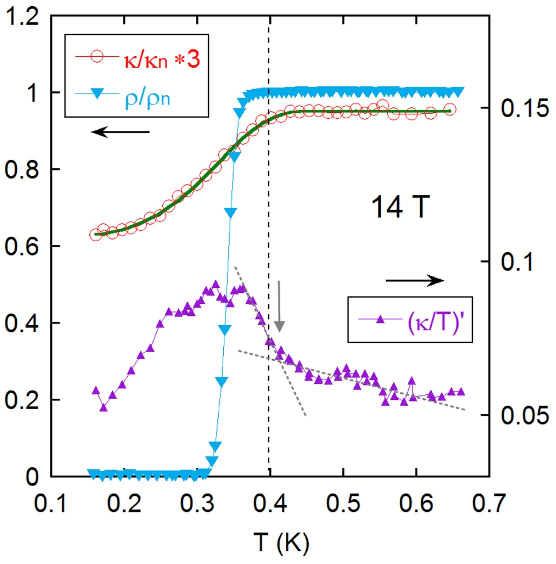

FIG. 2. Superconducting transition of sample 1, at 14 T for **H//b**. Red circles: Normalized electronic contribution κ/κ_n, with fit (green solid line). Blue triangles: Normalized resistivity (ρ/ρ_n), measured simultaneously on the same sample. Purple triangles: Derivative $\partial(\kappa/T)/\partial T$ in W K^{-3} m^{-1} (scale on the right). Vertical dashed line: T_{sc} from the (green) fit of κ/T; gray vertical arrows: T_{sc} from $\partial(\kappa/T)/\partial T$.

By contrast, a new phenomenon appears for fields above ~8 T: the resistive transition starts to overlap the bulk transition, and the $\rho = 0$ criterion leads to a transition temperature below the bulk transition. For fields above ~12 T, even the onset criterion leads to a resistive T_{sc} below the bulk determination: this is clearly seen in the raw data presented in Fig. 2. To show the robustness of this result, the derivative with respect to temperature $(\kappa/T)'$ of the raw data (without normalization) is equally presented in Fig. 2: $(\kappa/T)'$ is linear in temperature in the normal state in the presented temperature window, so the bulk SC transition would be marked by a change of slope of $(\kappa/T)'$ (gray arrows in Fig. 2). Although more arbitrary (notably for the noise averaging), this determination is in good agreement—yielding even a slightly higher T_{sc}—with that obtained from the fit on the normalized data (vertical dashed line). We also checked that the resistive transition does not shift back to larger T_{sc} when decreasing the measurement current: the crossing of the bulk and resistive transitions is not due to Joule heating.

When a type II superconductor shows an onset of the resistive transition at a temperature lower than the bulk T_{sc}, the prime suspect is current-driven vortex motion (flux creep or flux flow). This is most easily observed in 2D systems, such as organics or high-T_c cuprates, and in some other superconducting systems, where the resistive transition follows, instead of the real superconducting transition, a so-called irreversibility line. This line corresponds to the freezing of current-induced vortex motion [25–27]. In these systems, the resistive transition is significantly broadened with increasing magnetic field, and, in some cases, well below the bulk T_{sc}, it shows a sudden drop: this drop would arise from the "freezing transition," from a dissipative vortex-liquid state, to a vortex-lattice or glass-like state, where a much stronger pinning efficiency leads to zero resistance [28–34].

In UCoGe, the resistive transitions become much sharper in the high-field region for **H//b**. This is observed on all UCoGe samples, whatever the exact shape of H_{c2} for **H//b** (see for example data presented in Ref. [9], referred to as sample 3 in the following, and the raw data shown in the Supplemental Material for samples 1 and 2 of this study, together with H_{c2} for sample 2 [23]). The same phenomenon is completely absent for the two other field directions **H//a** and **H//c**: in these two cases, the resistive transition enlarges gradually with increasing field, and lies always above the bulk T_{sc} as

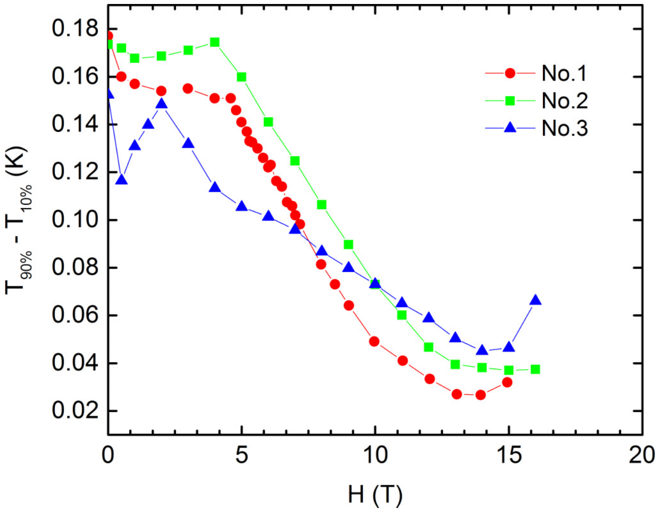

FIG. 3. Field dependence of the resistivity transition width ($T_{90\%} - T_{10\%}$) of UCoGe for **H//b**, on UCoGe samples 1, 2, and 3 reported in Ref. [9].

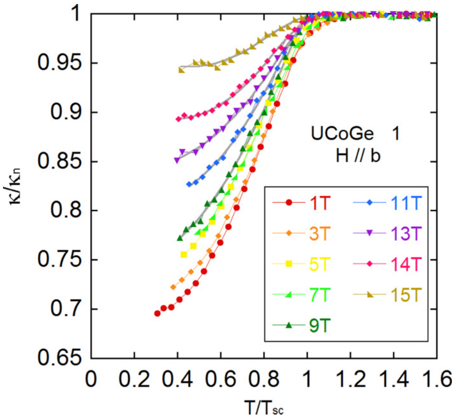

FIG. 4. Electronic contributions to the thermal conductivity normalized to its normal phase value κ/κ_n, as a function of temperature normalized to $T_{sc}(H)$, for fields along the **b** axis (sample 1). Solid lines are the fits used to determine H_{c2} (see Ref. [22]).

determined from thermal conductivity. Figure 3 shows the field dependence (**H//b**) of the resistive transition width of the three samples, defined as $T_{90\%} - T_{10\%}$. Here $T_{90\%}$ and $T_{10\%}$ are the transition temperatures defined by 90% and 10% resistivity compared to the normal state, respectively. We can observe a sudden drop of the transition width, starting at about 5 T in samples 1 and 2. In sample 3, the evolution of $T_{90\%} - T_{10\%}$ is more complicated, due to the several steps displayed in its resistive transition, but it is very similar to that of samples 1 and 2 above 5 T.

In that sense, UCoGe for **H//b** is similar to URu$_2$Si$_2$, where the authors of Ref. [32] have observed, with the same measurements, a resistive transition determining a melting transition T_m lower than the bulk T_{sc}. The peculiarities of URu$_2$Si$_2$ (low carrier density system with very heavy effective masses, 1D regime at high fields) put forward in Ref. [32] as favoring superconducting fluctuations (and so a vortex liquid phase) are certainly also present in UCoGe, which has a smaller Sommerfeld coefficient for even larger effective masses (according to the slope of H_{c2} at T_{sc} for **H//b** or **H//a**). Another sign of the importance of superconducting fluctuations in UCoGe could be, like for URu$_2$Si$_2$, the zero-field resistive transition width which is increasing with improvement of the RRR [35]. The most striking difference between the two systems lies in the fact that, while in the tetragonal system URu$_2$Si$_2$ the separation of T_m and T_c appears in the whole field range both for the in-plane and perpendicular field direction, in UCoGe such a behavior appears only in the high-field region for **H//b** (above around 8 T), as if the pinning mechanism was changing under field. The reinforcement of the pairing mechanism under field [22], leading to larger effective masses, larger T_{sc}, and smaller coherence length, goes in the right direction. But quantitatively, the effect on the critical fluctuation or the melting line as proposed in Ref. [32] is certainly not enough to explain the occurrence of the vortex liquid phase only at high fields. Another possibility could be a field-induced change in the vortex cores triggered by some change of the p-wave order parameter, which could lead to a reduction of the pinning force. Before discussing vortex structures in p-wave superconductors, let us first address the question of a field-induced transition in the superconducting state of UCoGe.

To study the evolution of the SC under magnetic field, we have further analyzed the κ/T data. Figure 4 presents the electronic contribution to the thermal conductivity normalized to the normal phase values (κ/κ_n), as a function of the normalized temperature (T/T_{sc}), at different magnetic fields along the **b** axis. For fields between 5 T and 9 T, κ/κ_n is almost field-independent. However, for fields above 9 T, κ/κ_n increases steadily with field and quickly approaches 1, even though 15 T seems still far from $H_{c2}(0)$. This can be quantified by reporting the extrapolated residual thermal conductivity $[\kappa/\kappa_n(T=0)]$ versus $H/H_{c2}(0)$. In UCoGe, because the pairing mechanism appears to be field-dependent, $H_{c2}(0)$ becomes also field-dependent (see [22]): Figure 5 displays κ/κ_n (with quadratic or linear extrapolation to $T = 0$) versus magnetic field (in the inset) or $H/H_{c2}(0)$ calculated according to the model in Ref. [22].

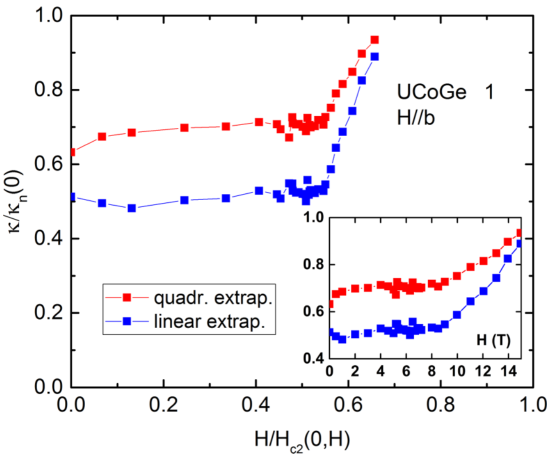

FIG. 5. $\kappa/\kappa_n(0)$ (sample 1) as a function of $H/H_{c2}(0,H)$, **H//b**, with $H_{c2}(0,H)$ obtained from the analysis in Ref. [22]. Red circles: From quadratic extrapolation to $T = 0$. Blue triangles: From linear extrapolation. Inset: Same data as a function of field.

IV. DISCUSSION

Figures 5 shows that, independently from the extrapolation procedure, there is an abrupt increase of the residual thermal conductivity above 9 T. Such an increase reflects a change of the electronic quasiparticle excitation spectrum in the superconducting state, leading to a strong enhancement of the residual density of states. This could arise from a field-induced change of the nodal structure of the superconducting order parameter, for example, due to a change from a line node in the (**a**, **b**) plane of the B state to a point node along the **c** axis of the A state, for an orthorhombic ferromagnetic superconductor [36], a situation similar to UPt$_3$ [19], or, alternatively, due to a rotation of the nodal structure with respect to the heat current direction (along the **c** axis in our case), as a result of a reorientation of the spin quantization axes. Note that a change of the vortex core structure alone is not likely to increase the residual thermal conductivity, owing to the geometry (field perpendicular to the heat current) and to the fact that UCoGe is in the clean limit (mean-free path much larger than the coherence length). Careful measurements of H_{c2} did not reveal (within experimental error) a kink of H_{c2}, which could have accompanied the transition between two different superconducting states.

There are many factors which could contribute to a field-induced transition inside the superconducting state. First of all, at zero field, UCoGe is generally considered to be in a spin-triplet-ESP state, with a quantization axis along the **c** axis imposed by the exchange field (H_{exch}). H_{exch} is in competition with the external field applied along the **b** axis, and for field values of order 10 T, the induced magnetization along **b** is of the same order as the spontaneous magnetization in zero field [37,38]. Meanwhile, due to the weakening FM order with increasing transverse field along the **b** axis, a reduction of H_{exch} is expected [39], resulting in a reduction of the energy gap between the spin-up and spin-down bands. For these reasons, even if no change of the order parameter symmetry occurs, a significant rotation of the d-vector is expected [16,39], as well as a recovery of a unitary state [39]. In URhGe, the possibility of a compensation of the exchange field (after the moment rotation along the **b** axis) by the external field (Jaccarino-Peter effect [40]) has been predicted to lead to non-ESP states [16].

Moreover, the nature of the pairing mechanism itself is also influenced by an external field along **b**. At low field, fluctuations are purely longitudinal, along the **c** axis [41]. But in the sister compound URhGe, NMR experiments have shown that ferromagnetic fluctuations also grow along the **b** axis on approaching H_R [11,12]. Still in URhGe, uniaxial pressure experiments suggest that fluctuations become more 2D in zero field under stress [17], and that a similar effect probably also arises for the RSC phase at H_R. A similar, and expected, field-induced evolution of the pairing mechanism in UCoGe could drive a change of the SC order parameter.

These effects suggest that, when the superconducting state is reinforced under field parallel to **b**, the spin direction of the Cooper pairs does not remain locked to the crystalline **c** axis as it certainly is in low field. This recovered spin degeneracy [16,39] is also favorable to the appearance of nonsingular vortices, i.e., vortices where the order parameter does not vanish completely in the core. Indeed, such vortices have been predicted to occur for superconductors with multicomponent order parameters, when the zeros of the two components of the superconducting order parameter are located at different points in space (see review [42]). The multicomponent character can arise from the orbital degrees of freedom [43] or from the spin degrees of freedom in a triplet state [44,45], and this field of research on exotic vortices notably in p-wave superconductors [46] or in Bose-Einstein condensates [47] is still very active. Most of these theoretical investigations concern isolated vortices (close to H_{c1}) [42], but the dissociation of singular vortices in pairs of half-quantum vortices [48] is also predicted to exist in high fields, in the regime of Abrikosov lattices, for multicomponent (orbital degrees of freedom) order parameters [48] or ESP triplet states [49]. In fact, any ESP p-wave state could support half-quantum vortices [50]. The appearance of such nonsingular vortices is an appealing hypothesis to explain the existence of a vortex liquid state through the weakening of the pinning force.

V. CONCLUSION

Both effects, the appearance of a "vortex liquid state" at high fields in UCoGe (pointed out by the crossing of bulk and resistive H_{c2} and by the narrowing of the resistive transition), as well as a possible change of the superconducting order parameter (detected by the abrupt increase of the residual thermal conductivity), whether they are related or not, show that the physics of p-wave ferromagnetic superconductors in transverse fields is extremely rich. They are worth further theoretical and experimental investigations of the superconducting phase diagram, which could reveal new field-induced phase transitions, and of the vortices, which could display nonsingular vortex cores, long predicted but only observed in superfluid ^{3}He [51,52] up to now.

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