Collapse of Ferromagnetism and Fermi Surface Instability near Reentrant Superconductivity of URhGe

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We present thermoelectric power and resistivity measurements in the ferromagnetic superconductor URhGe for a magnetic field applied along the hard magnetization b axis of the orthorhombic crystal. Reentrant superconductivity is observed near the spin reorientation transition at $H_R = 12.75$ T, where a first order transition from the ferromagnetic to the polarized paramagnetic state occurs. Special focus is given to the longitudinal configuration, where both the electric and heat current are parallel to the applied field. The validity of the Fermi-liquid T^2 dependence of the resistivity through H_R demonstrates clearly that no quantum critical point occurs at H_R. Thus, the ferromagnetic transition line at H_R becomes first order implying the existence of a tricritical point at finite temperature. The enhancement of magnetic fluctuations in the vicinity of the tricritical point stimulates the reentrance of superconductivity. The abrupt sign change observed in the thermoelectric power with the thermal gradient applied along the b axis together with the strong anomalies in the other directions is definitive macroscopic evidence that in addition a significant change of the Fermi surface appears through H_R.

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Quantum phase transitions (QPTs) are a central topic in contemporary condensed matter research. Their rich underlying physics plays an important role in explaining the exotic low-temperature properties of a variety of strongly correlated materials like high-T_C superconductors [1], quantum magnets [2], or heavy-fermion compounds [3,4]. Strictly speaking, a QPT is a zero-temperature instability, yet its manifestations can be observed at finite temperature within a rather wide temperature region as a function of a nonthermal control parameter. Recent theoretical [5–10] analyses of ferromagnetic (FM) QPTs have shown that generally the second order phase transition turns into a first order one at a tricritical point (TCP) in the proximity of a FM QPT when approaching absolute zero temperature in clean systems. Experiments in FM systems such as $ZrZn_2$ [11] or UGe_2 [12] confirm this trend. However, in other systems (such as $YbNi_4P_2$ [13]) a continuous second order QPT has been invoked. In principle, a control parameter can be tuned opportunely in order to move the TCP to zero temperature, generating a quantum critical end point. Some compounds are located close to a quantum critical end point at ambient conditions [12,14–16].

In the present Letter we study the magnetic phase diagram of the orthorhombic Ising-type ferromagnet URhGe and its interplay with superconductivity (SC) [17]. URhGe is one of the four uranium based compounds, besides UGe_2 [18], UCoGe [19], and UIr [20], where the microscopic coexistence of ferromagnetism and SC has been observed. In URhGe, the magnetic moments $M_0 \approx 0.4\ \mu_B$ are oriented along its easy c axis. A transverse magnetic field higher than the superconducting critical field H_{c2} applied along the hard magnetization b axis induces at low temperature a reorientation of the magnetic moments from the c to the b axis [21] at $H_R = 11.75$ T. A field reentrant superconducting phase (RSC) appears in a narrow field window around H_R below $T_{\mathrm{RSC}} = 410$ mK [22]. It has been suggested that the transverse magnetic field tunes the system in the vicinity of the TCP [23]. Thus, it is a key case to study a FM QPT. It allows us to investigate the interplay of magnetic fluctuations and possible Fermi surface (FS) changes with SC.

Thermoelectric power (TEP) is an excellent probe to detect electronic singularities and FS changes notably in strongly correlated electron systems as it is sensitive to the derivative of the density of states and electronic scattering with respect to the energy at the Fermi energy [24]. Pertinent examples are heavy-fermion compounds such as $CeRu_2Si_2$ [25,26], $CeRh_2Si_2$ [27], $YbRh_2Si_2$ [28,29], or URu_2Si_2 [30]. Here, we present systematic TEP and resistivity measurements on URhGe with different orientations of the thermal current J_Q and electric current J_e with respect to the magnetic field, which is always applied along the b axis. Experimental details are given in the Supplemental Material [31]. We will focus on the longitudinal response with the currents and field along the b axis.

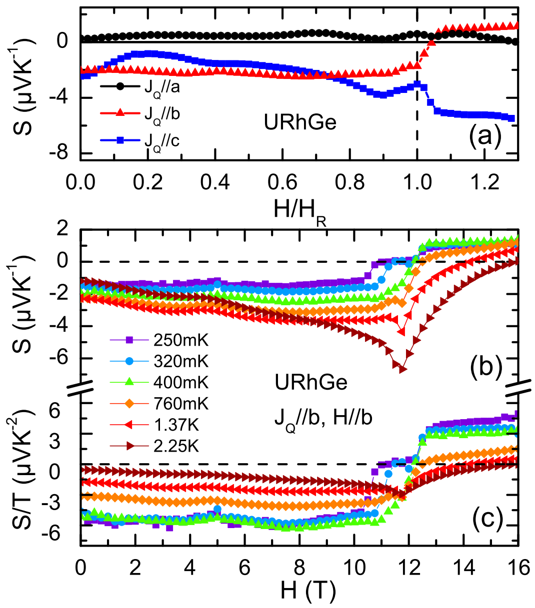

FIG. 1. (a) Thermoelectric power S as a function of magnetic field H along the b axis normalized by H_R at $T \approx 470$ mK for J_Q along the three crystallographic directions. (b) The TEP S and (c) S/T at different temperatures from 250 mK to 2.25 K for $J_Q \| b$ and $H \| b$.

Special attention is given on the temperature dependence of the resistivity at various magnetic fields. The validity of the Fermi-liquid T^2 dependence through H_R demonstrates clearly that no QCP occurs at H_R; thus, the FM transition line at H_R becomes first order implying the existence of a TCP at finite temperature. Evidence of a first order transition at H_R was reported by torque experiments [32] and Hall resistivity experiments [33], and has been recently confirmed by NMR experiments [34]. The abrupt variation in the TEP for the three directions of J_Q at H_R is a macroscopic signature of a drastic change of the FS. Previous signatures had been detected by quantum oscillation [35] and Hall effect [33] experiments.

Figure 1(a) shows the field dependence along the b axis of the TEP for J_Q along the three main crystallographic directions at $T = 470$ mK, just above the critical temperature of the RSC state ($T_{\mathrm{RSC}} = 410$ mK). The TEP is clearly anisotropic and shows very pronounced anomalies at H_R for the b and c direction. For $J \| a$, although the TEP is always positive and small, it shows small anomalies around H_R. In this direction the signature in the TEP of the scattering term is suspected to be small as J_Q stays perpendicular to the direction of the magnetic moments even above the reorientation at H_R. For the transverse configuration $J_Q \| c$, the TEP is always negative and decreases with increasing field. It shows a clear peak at H_R. In the longitudinal configuration $J_Q \| b$, the TEP is always negative in the FM state, has a steplike transition at H_R, and becomes positive in the polarized paramagnetic (PPM) state above H_R. As already reported in Hall resistivity experiments [33], small anomalies occur around 1.5 and 5 T suggesting minor changes in the FS. Without any orbital effect in the longitudinal configuration, the TEP change at H_R originates most likely from a FS reconstruction as suggested previously [33,35]. We will now focus on the results for $J_Q \| b$ with $H \| b$.

The magnetic field dependence of the TEP, $S(H)$, and the TEP normalized by temperature, $S/T(H)$, from 250 mK to 2.25 K for J_Q and $H \|$ b is represented in Figs. 1(b) and 1(c), respectively. S is negative below and positive above H_R. At 2.25 K, S shows a sharp negative peak at $H_R = 11.75$ T. With decreasing temperature the transition becomes sharper and finally steplike. At 250 mK S shows a two step transition with $S = 0$ from 10.5 to 12.5 T indicating the presence of the RSC in this system around H_R. In a simple two-band picture, the sign of the TEP is set by the product of the effective mass and the mean free path of the heat carriers [24]. Therefore, the observation of S/T with different signs below and above H_R [see Fig. 1(c)] implies that the nature of this pocket changes across the transition. While we cannot identify individually the pockets participating in this transition, the result finds a natural explanation if one assumes that the suppression of the FM state is accompanied by a substantial reconstruction of the FS without changing the compensated nature of the system. We can also notice [see Fig. 1(c)] that at H_R for $T > T_{\mathrm{RSC}}$, S/T is temperature independent with a value of $-2.8 \mu V K^{-2}$, indicating that the electronic singularity in the density of states occurs at a peculiar value of the entropy per carrier.

Figure 2 displays the temperature dependence of the TEP for $H = 0$ and 9 T. With decreasing temperature, a first minimum occurs around the Curie temperature $T_C \approx 9.5 K$. Inside the FM state, two other anomalies appear at $T^* \approx 4$ K

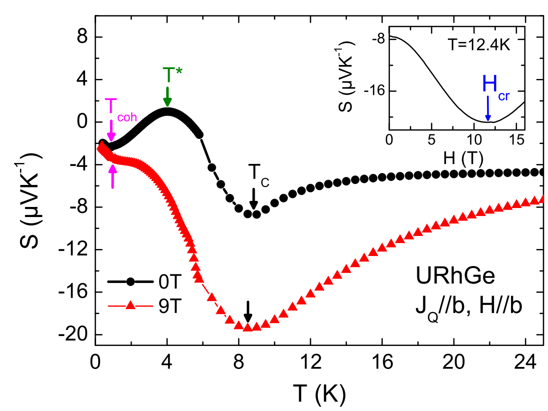

FIG. 2. Temperature dependence of the TEP between 1 and 25 K for $H = 0$ and 9 T. The black arrows mark T_C; the green and pink arrows indicate the position of the anomalies labeled T^* and T_{coh}, respectively. In the inset, the field dependence of S at 12.4 K (above T_C) shows a broad minimum indicated by the blue arrow at H_{cr}.

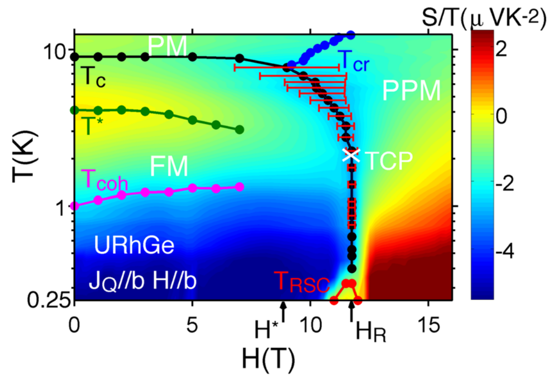

FIG. 3. Linear color map of S/T in the (T,H) plane. The Curie temperature T_C (black circles), the energy scales T^* (green circles) and $T_{\rm coh}$ (pink circles), the reentrant superconductivity $T_{\rm RSC}$ (red circles), and the crossover line $T_{\rm cr}$ between the PM and the PPM state (blue circles) are superimposed. The transition width observed in the TEP around H_R is also represented (red horizontal lines).

and $T_{\rm coh} \approx 1$ K. T^* may mark a characteristic energy of the interplay between the magnetic excitations and the establishment of the FM FS below T_C. $T_{\rm coh}$ indicates the entrance in the coherent low temperature Fermi-liquid regime in which the TEP is linear in T for $T \to 0$ K. In the inset, a typical field dependence of the TEP in the paramagnetic (PM) state at $T = 12.4\,{\rm K} > T_C$ is represented. The TEP still exhibits a broad minimum around $H_{\rm cr} \approx 12$ T defining a crossover $T_{\rm cr}(H)$ between the PM and PPM state. This crossover can still be observed at 36 K and 18 T.

Figure 3 presents S/T as a color plot in the (T,H) plane. We can clearly see that at low temperature S/T is strongly negative (dark blue) in the FM state (below H_R) and becomes positive (dark red) in the PPM state. The different anomalies obtained in the TEP measurements for J_Q, $H\|b$ are superimposed. The width of the FM transition (for details see Fig. S1 of the Supplemental Material [31]) observed in the H scans of the TEP around H_R is also represented (red horizontal lines). The sudden increase of the transition width with increasing temperature is a clear signature of crossing the TCP, which hence can be located precisely at $T_{\rm TCP} = 2$ K and $H_{\rm TCP} = 11.5$ T. Concomitantly, the low temperature energy scales T^* and $T_{\rm coh}$ seem to converge to the same point in the (T,H) plane, suggesting a link with the TCP. Magnetic torque measurements located a TCP at 11.45 T [32,36] for a perfect alignment along the b axis leading exactly to the same value of H_R. For $T < T_{\rm TCP}$, the FM transition becomes first order and is independent of the field.

The temperature dependence of the resistivity ρ is represented as a function of T^2 in Fig. 4(a). At very low temperatures $\rho(T)$ follows the Fermi-liquid theory with $\rho(T) = \rho_0 + AT^2$. With increasing temperature $\rho(T)$ deviates from the T^2 dependence with an exponent $n < 2$ for all fields except for $H = 0$. We fitted $\rho(T)$ such as

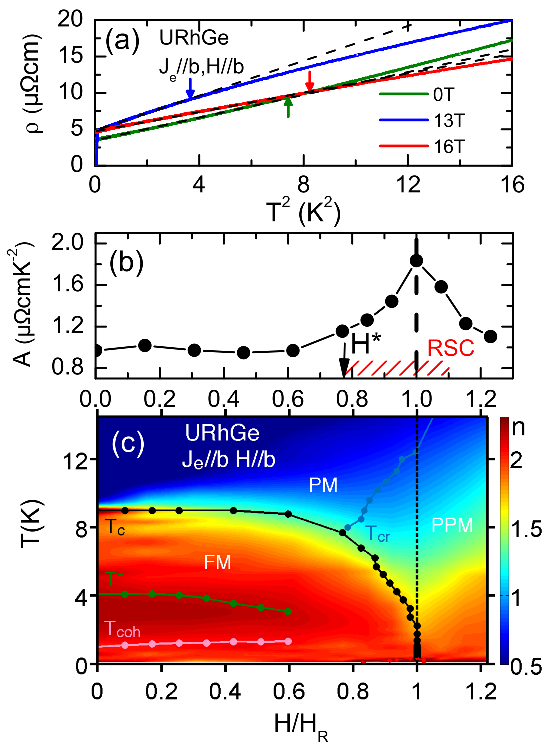

FIG. 4. (a) Resistivity as a function of T^2 for $J_e\|b$, $H\|b$ below 4 K for different magnetic fields. Linear fits at low temperature are represented by dashed lines. The vertical arrows indicate the deviation from T^2 dependence. (b) Field dependence of the A coefficient of the resistivity. (c) Linear color map of the exponent n of the resistivity $[\rho(T) = \rho_0 + AT^n]$ in the $(T,\ H/H_R)$ plane. The different anomalies observed in the TEP are superimposed on the phase diagram.

$\rho(T) = \rho_0 + AT^n$, on a sliding window of 400 mK below 14 K. ρ_0 is the residual resistivity and A is the coefficient characterizing the amplitude of the inelastic scattering. The field dependence of A determined at the lowest temperature is shown in Fig. 4(b). It exhibits a peak at H_R, indicating an increase of the effective mass associated with spin fluctuations. A similar behavior of $A(H)$ has been observed in the transverse configuration [37]. The enhancement in $A(H)$ starts roughly near the characteristic field where the crossover line $T_{\rm cr}(H)$ intercepts $T_C(H)$ at $H^* = 8.8$ T [black arrow in Fig. 4(b)] and where $T_C(H)$ starts to decrease. Astonishingly, the magnetization along the c axis, M_c, starts to decrease already at H^* [22], see Fig. S2 of the Supplemental Material [31]. The RSC in the TEP and in the magnetoresistance measurements is found at 270 mK between 10 and 12.5 T. The strong enhancement of A in the field range 8–15 T with a maximum at H_R is in excellent agreement with the observation of the enhancement of the nuclear relaxation rates $(1/T_1)$ and $(1/T_2)$ detected by NMR [34,38]. We notice that our $T_{\rm TCP}$ estimation is lower than that proposed in Ref. [34] where $T_{\rm TCP} \approx 4$ K.

A linear color plot of the exponent n of the resistivity in the $(T,H/H_R)$ plane is represented in Fig. 4(c) where the different anomalies observed in the TEP are superimposed. Remarkably, below 2 K $n \approx 2$ is found to be field independent and thus, no quantum critical behavior appears. This is in excellent agreement with the first order transition below the TCP close to H_R. We find $n \approx 2.3$ around 4 K where the anomaly T^* has been observed by TEP. This observation of $n > 2$ inside the FM state could be related to magnetic excitations.

The data reported in the different phase diagrams lead to an unambiguous determination of the position of the TCP of the FM to PPM transition, which is characterized by the c to b axis switch of the magnetization. Signatures of FS instabilities at the FM to PPM transition are clearly observed in the field variation of $S(H)$ through H_R. Furthermore, Hall effect [33] as well as angular resolved photoemission spectroscopy experiments [39] point out a FS change on crossing the PM-FM phase at T_C in a low field on cooling. Thus, three different FSs will correspond to the PM, FM, and PPM phases. The possibility of a Lifshitz transition at H_R in URhGe was proposed in Ref. [35] from Shubnikov–de Haas (SdH) measurements performed at an angle of 10° from the b axis to escape from the RSC [35]. For this angle, no first order transition and no RSC, but a crossover at H_R, is expected. The experimentally observed SdH oscillations below H_R, corresponding to a small orbit of only a few percent of the Brillouin zone, vanish on approaching H_R. A possible explanation is the collapse of the orbit. It is claimed that this Lifshitz-type transition, leading to the collapse of the Fermi velocity, is the driving force for the RSC. However, as shown in Fig. 5, the TEP in URhGe for $J_Q \| a$, $H \| b$ shows large quantum oscillations above 22 T, represented as a function of $1/H$ in the inset. The corresponding frequency, ≈500 T, is very similar to the frequency observed in the SdH measurements. Hence, a Lifshitz transition as the sole driving force for the RSC seems unlikely. In our study the misalignment is always less than 1° and the transition just above the RSC is clearly first order and thus it cannot be of sole Lifshitz nature. Instead, we give macroscopic evidence that the RSC is associated with both a FS instability and critical fluctuations when $T_C(H)$ vanishes. Surprisingly, neglecting the FS change, excellent agreement is found in the description of the RSC as a function of magnetic field and pressure in a crude phenomenological model where the enhancement of $A(H)$ reflects the enhancement of the effective mass and hence of the superconducting coupling constant [37,40]. An open question that remains is the field dependence of the FS inside the dome of the RSC and whether this dome can be described with a unique FS.

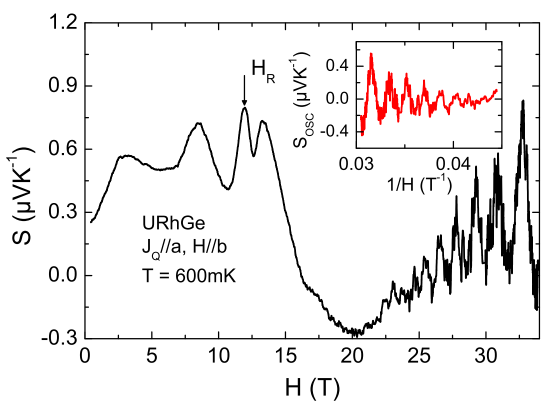

FIG. 5. Magnetic field dependence of the TEP in URhGe for $J_Q \| a$, $H \| b$ up to 34 T at 600 mK. S shows small anomalies around H_R and quantum oscillations above 22 T, represented in $1/H$ in the inset.

Recently, the RSC was described in a Landau approach taking into account a two-band approach due to the splitting of the bands in the FM domain [41] in the FM domain and stressing the importance of longitudinal fluctuations. In agreement with our experiments, it is shown that in a transverse field ($H \| b$) the PM-FM transition switches from second to first order at a TCP (H_{TCP}, T_{TCP}) close to H_R. The optimum of $T_{\mathrm{RSC}}(H)$ is predicted to be roughly half of T_{TCP}. In our experiment $T_{\mathrm{TCP}} \approx 2$ K and $T_{\mathrm{RSC}} \approx 0.4$ K; hence, $T_{\mathrm{RSC}} \approx T_{\mathrm{TCP}}/5$. Furthermore, as observed experimentally, $T_{\mathrm{RSC}}(H)$ is expected to fall down asymmetrically on both sides of H_R. The predicted decrease of $M_c \propto \sqrt{T_C(H)}$ cannot be properly tested due to the lack of accuracy of the existing magnetization data. The renormalized spin-fluctuation theory [42] predicts $M_c(H)$ varying as $T_C^{\frac{2}{3}}$ for the collapse of the FM state at a QCP. The vicinity of H_{TCP} from H_R ($H_{\mathrm{TCP}}/H_R \approx 0.97$) is close to what is observed in UGe_2 under pressure ($P_{\mathrm{TCP}}/P_C \approx 0.96$).

To summarize, we present clear evidence that on top of a large enhancement of the fluctuations detected here and very recently in NMR experiments [34,38], FS instabilities occur at H_R. These fluctuations associated with the energy scales converging to the TCP very close to H_R confirm the first order nature of the transition and the absence of a QCP. The role of longitudinal and transversal fluctuations observed close to H_R on the RSC is still under debate. Another interesting proposal is that soft magnons could possibly generate a new attractive pairing interaction for the RSC [43]. It is worthwhile to notice that the interplay of FS instabilities and SC is a quite challenging question as a similar problem remains unsolved for high-T_C materials as well as for the 115-Ce compounds [44]. The additional ingredient of FS instabilities in strongly correlated electronic systems, where the interaction of the quasiparticles themselves is responsible of the superconducting pairing, clearly deserves theoretical treatment. A further experimental challenge is to clarify possible differences in the superconducting phases on both sides of H_R.

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