Thermoelectricity of the ferromagnetic superconductor UCoGe

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UCoGe exhibits superconductivity in the presence of ferromagnetism. When a field is applied along the b axis (perpendicular to the easy axis), ferromagnetism is weakened, and superconductivity is enhanced. This enhancement has been attributed to an increase in coupling as observed in the enhanced effective mass produced by the critical fluctuations as the ferromagnetic transition is strongly suppressed. However, it is also important to know if and how the Fermi surface changes near the critical point. Here we report measurements of the thermoelectricity of UCoGe that reveal a low-carrier-density metal. Under magnetic field applied along the b axis, a sharp peak is observed in the thermopower of UCoGe at $H^* = 11.1$ T and low temperature that becomes broader at higher temperatures. At higher field, the thermopower changes sign, which suggests a modification of the Fermi surface. We analyze these results using a topological change in the Fermi surface and show that this can explain both the thermopower and the enhanced superconductivity.

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I. INTRODUCTION

The discovery of superconductivity coexisting with ferromagnetism in three uranium-based compounds, UGe_2,[1] URhGe,[2] and most recently UCoGe,[3] has attracted much interest due to the unusual properties of the superconducting state and the proximity to a critical pressure where the ground state of each compound switches from ferromagnetic (FM) to paramagnetic (PM). The enhanced ferromagnetic fluctuations in these Ising ferromagnets give rise to unconventional phenomena, such as reentrant superconductivity[45] and spin-triplet superconductivity.[6,7] In the cases of URhGe and UCoGe, a moderate field applied perpendicular to the easy axis can strongly suppress the Curie temperature to zero temperature.[4,8] For URhGe this reveals a reentrant superconducting phase and a reorientation of the magnetic moment[5] to be parallel to the applied field. For UCoGe, an enhancement of the superconducting temperature is observed.[4]

The phase diagram of a weak itinerant ferromagnet has been the subject of recent theoretical[9] and experimental work.[10,11] Measurements on UGe_2 under pressure P and field H (Ref. 10; applied along the easy axis) have demonstrated that the phase diagram has a tricritical point where the transition from the PM to the FM becomes first order and bifurcates in the H-P plane, resulting in quantum critical end points at high field and high pressure,[10] which is in rough agreement with the theoretical predictions.[9] However, the changes in the Fermi surface in UGe_2 (Ref. 12) are not accounted for in the theory, even though these changes can explain some of the features of UGe_2.[13] Also, it has been shown that topological transitions near quantum critical points can lead to unconventional critical behavior.[14]

In this paper, we focus on UCoGe. At ambient pressure, this compound is a weak ferromagnet with a Curie temperature $T_C \sim 2.8$ K associated with a spontaneous moment of $M_0 \sim 0.04\ \mu_B$ directed along the c axis at $T = 0$ K.[3] The superconducting transition temperature $T_{SC} \sim 0.6$ K in zero field. Applying pressure suppresses ferromagnetism and enhances superconductivity until $T_C \sim T_{SC}$ at around 1.25 GPa.[15] Above this pressure, the ferromagnetic transition is not observed. This pressure dependence could imply that UCoGe sits very close to the tricritical point. NMR measurements[16] have suggested that the PM-FM transition is first order in UCoGe. Applying a field along the b axis also suppresses ferromagnetism[17,18] and enhances superconductivity, resulting in an unconventional S-shaped upper critical field H_{c2} curve.[4]

Thermoelectric effects are very sensitive to changes in the Fermi surface topology and effective mass enhancements, as has been shown in several studies, for example, on $CeRu_2Si_2$ (Ref. 19) and $CeCoIn_5$.[20] Quantum oscillation experiments[21] on UCoGe have revealed an unusual field dependence of the cyclotron masses in fields greater than 20 T along the b axis. In this paper we report measurements of the thermopower of UCoGe as a function of field applied along the b axis to examine these effects in the region of reentrant superconductivity ($H_b \sim 10$ T). We suggest that the Fermi surface of UCoGe undergoes a topological change as the ground state switches from FM to PM under applied field and compare this with recent band-structure calculations. We also argue that this change in the Fermi surface can be used to explain the unusual H_{c2} curve.

II. EXPERIMENTAL METHOD

Single crystals of UCoGe were grown using the Czhochralski method in a tetra-arc furnace. The residual resistivity ratio (RRR) of the measured sample was around 30. The thermopower was measured using the two-thermometer, one-heater technique at temperatures down to 100 mK and in fields up to 18 T. All measurements were performed in both positive and negative fields and averaged to prevent contamination of the Nernst signal in the Seebeck. Thermometers were calibrated against a germanium thermometer in a field-compensated region up to 16 T and down to 100 mK. The temperature gradient was applied along the a axis of the crystal, and the field was applied along the b axis in all measurements. Based on the angular dependance of the upper critical field H_{c2} curves,[4] the crystals were aligned along the b axis to $<5^\circ$ with respect to the magnetic field H. The setup also allows *in situ* measurement of the resistivity; therefore all data presented below were taken on the same sample in the same conditions.

III. RESULTS

Figure 1 shows the temperature and field dependence of the thermopower divided by temperature S/T of UCoGe.

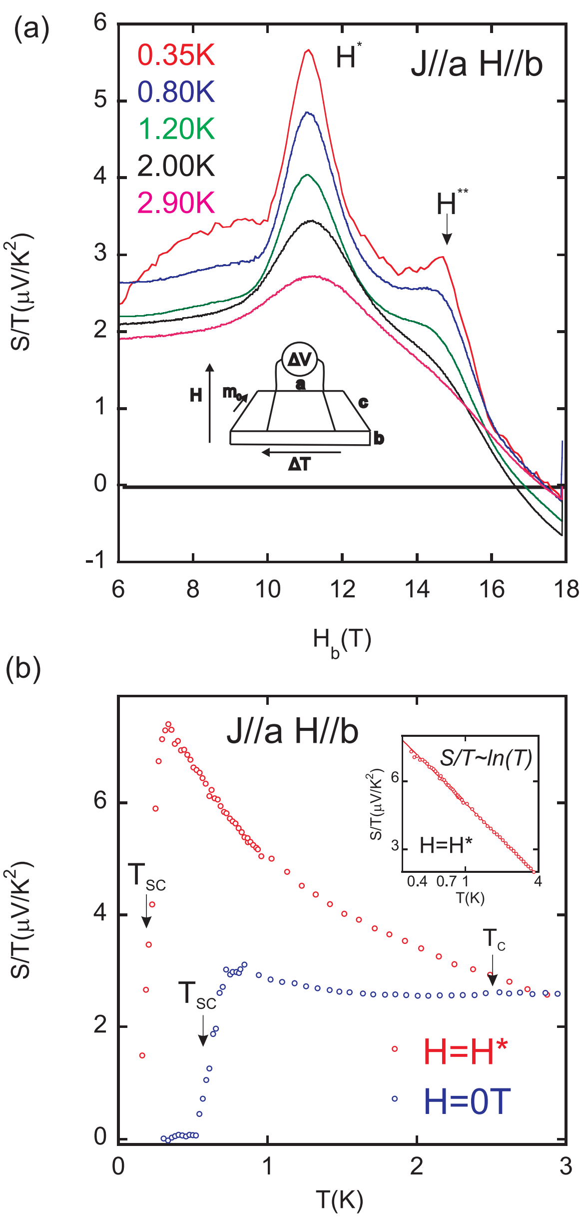

FIG. 1. (Color online) (a) Thermopower divided by temperature S/T as a function of field applied along the b axis H_b at several temperatures. A large peak is observed at $H^* = 11.1$ T at all temperatures, a smaller anomaly is observed at $H^{**} = 14.6$ T at low temperature, and a change of sign is observed above 16 T. The inset is a diagram of the setup showing the applied field H, the temperature gradient ΔT, the measured thermoelectric voltage ΔV, and the zero-field magnetization M_0 compared with the crystal axis. (b) Temperature dependence at two fields, 0 T and H^*. In 0 T both the Curie temperature T_c and the superconducting transition T_{SC} are observed. For $H = H^*$ a possible logarithmic divergence is observed above T_{SC}. The inset shows $H = H^*$ data on a semilog plot, illustrating the divergence above T_{SC}.

In 0 T, S/T shows a small anomaly at $T_C = 2.8$ K that is followed by a slight increase of S/T until the superconducting transition $T_{SC} = 0.6$ K. In a free electron gas, S/T is constant at low temperature and its amplitude is inversely proportional to the Fermi temperature, with a sign that reflects the type of charge carrier. For UCoGe, S/T is $\sim 3\ \mu\text{V/K}^2$ just above T_{SC}, indicating the dominant carriers are hole like. In a free electron gas, the ratio q $(= \frac{S}{T}\frac{N_a e}{\gamma}$, N_a is Avogadro's number) of the thermoelectric power to the Sommerfeld coefficient γ of the linear term in specific heat is a constant.[22] It has been observed in a large variety of strongly correlated systems that the low-temperature value of S/T scales with the Sommerfeld coefficient via ratio q, which is inversely proportional to the carrier number. Furthermore, despite the complexity of multiband systems, it has been shown[23] that even in compensated cases (such as URu_2Si_2) the thermoelectric response is dominated by one carrier type, and thus the scaling factor q holds. For UCoGe, $q = 5$ ($\gamma = 57$ mJ/mol K^2; Ref. 3), which indicates UCoGe has a low carrier density. This is similar to the case of URu_2Si_2, which also has $q \sim 5$ and is a low-carrier semimetal. Recent quantum oscillation measurements[15,24] and thermopower measurements[25] imply the Fermi surface of URu_2Si_2 is strongly modified in a moderate field (of the order of 20 T applied along the easy magnetization axis). This could be the same in UCoGe as the quasiparticle masses are heavy,[21,26] and this coupled with the low carrier density leads to a small bandwidth, which can be strongly modified by a moderate field.

As the field is increased, S/T remains roughly constant ($\sim 3\ \mu\text{V/K}^2$) up to ~ 10 T. At all measured temperatures (up to 3.5 K), a peak is observed centered on $H^* = 11.1$ T. This field is independent of temperature, but at higher temperatures the peak is broader and less pronounced. The peak is present even above the zero-field T_C. Above H^*, at low temperature a second anomaly is observed at $H^{**} = 14.6$ T that is smaller and indistinguishable from the background at temperatures above ~ 2 K. At higher field a change of sign of S/T is observed, indicating a change in the dominant carrier type and therefore a Fermi surface change. This is the field range where quantum oscillations have been observed.[21] There is a strong temperature dependence of S/T at H^*, with the magnitude of S/T increasing up to $\sim 7\ \mu\text{V/K}^2$ at the superconducting transition temperature. This could indicate one of two possibilities: H^* could be a critical point where one would expect an enhancement of the effective mass and hence thermopower[20,27] or H^* could be a topological change in the Fermi surface, which can give a large thermopower anomaly at low temperature as described by Lifshitz.[28] The fact that H^* is visible above the Curie temperature is interesting. If it is a Fermi-surface instability, then it implies that an element of the Fermi surface is constant in the ferromagnetic and paramagnetic state at low fields, which is contrary to recent band-structure calculations[29] but consistent with fact that the zero-field thermopower shows only a small anomaly at T_C. If it is a critical point, then the fluctuations are felt to high temperature, perhaps indicating a quantum critical point at H^*. It should be noted that the thermopower data imply there is a topological change between the zero-field PM state and the field-induced PM state as the change of sign occurs even at temperatures above T_C. This could be

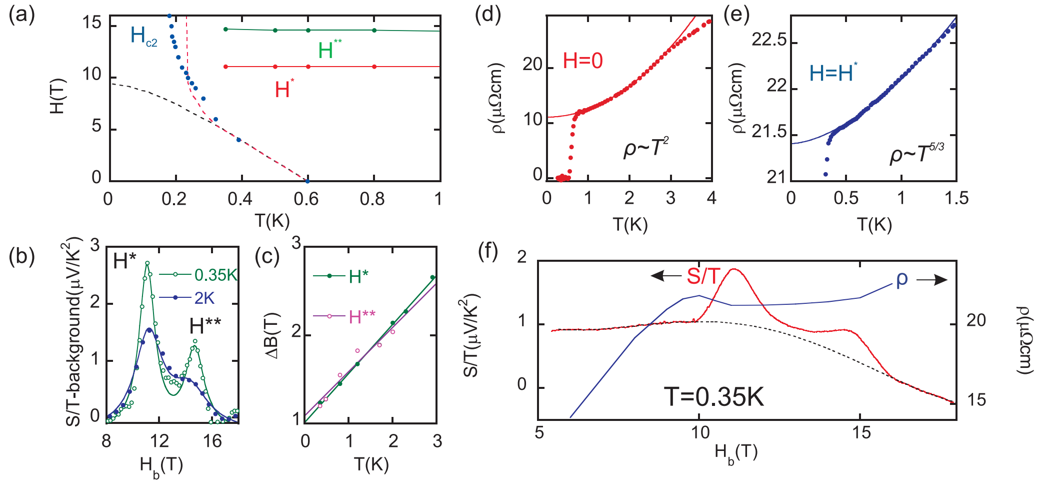

FIG. 2. (Color online) (a) Phase diagram of UCoGe in the T-H_b plane using thermopower data and resistivity data showing H_{c2}, H^*, and H^{**}. The dashed lines are a calculation for a polar p-wave state (black) and the same calculation assuming a variation of v_F with field (pink). (b) H^* and H^{**} at two different temperatures with background subtracted. Lines are a fit to two Lorentz distributions, one centered on H^{**} and one on H^* (some points not shown for clarity). (c) Width of H^* and H^{**} taken from the Lorentz distribution fit as a function of temperature. The lines are linear fits. (d) Resistivity data at 0 T as a function of temperature. The line is fit to $\rho = \rho_0 + AT^2$ below T_C. (e) Resistivity data at H^* as a function of temperature. The line is fit to $\rho = \rho_0 + aT^{\frac{5}{3}}$ below 1.5 K. (f) Field dependence of thermopower and resistivity taken at 0.35 K. The dashed line is the assumed background used for Lorentzian fits in (b).

evidence that the comparable scale of the Fermi energy and Zeeman energy is at least partly responsible for the modification of the Fermi surface such as is observed in URu_2Si_2 (Refs. 24 and 25). Finally, strong evidence for a Lifshitz transition is that the maximum thermoelectric response at H^* is enhanced compared with the response expected from the effective mass enhancements measured in a perfectly aligned crystal.[4]

Figure 2(a) shows the superconducting phase diagram of UCoGe derived from thermopower and resistivity data. The so-called S-shaped H_{c2} curve is slightly suppressed, probably due to the slight sample misalignment, and is visible as a sudden upturn in the H_{c2} curve. The calculated H_{c2} curve for a polar p-wave state is also shown calculated in Ref. 30 and following Ref. 6 based on the initial slope of H_{c2}. H^* seems to coincide with the near-vertical region of H_{c2}, suggesting that it drives the enhanced superconductivity phase. Figure 2(b) shows the thermopower in the critical region after a smooth background is subtracted. The two-peak structure can be reasonably well fitted with two Lorentzians centered on $H^*$and H^{**}. The fit is to the function $S = S^{**}(\frac{\Delta H^{**2}}{(H-H^{**})^2+\Delta H^{**2}}) + S^*(\frac{\Delta H^{*2}}{(H-H^*)^2+\Delta H^{*2}})$, where S^{**} (S^*) is the amplitude of the peak at H^{**} (H^*) and ΔH^{**} (ΔH^*) is the width of the peak at H^{**} (H^*). Both peaks are well fitted with a similar width but different amplitudes. Figure 2(c) shows ΔH as a function of temperature for both peaks and is linear in temperature with a finite intercept. This is consistent with a change in Fermi surface as temperature broadens the effect but the field required for the change is constant. As the temperature dependences of the widths of the H^* and H^{**} anomalies are very similar, it is reasonable to assume they have the same origin. One possible experimental origin is a small inclusion of misaligned crystal in the sample. Another possible origin is that the complexity of the Fermi surface leads to different characteristic fields, as recently observed in URu_2Si_2.[24]

Resistivity measurements taken on the same setup are also shown in Figure 2. In 0 T, the resistivity shows a T^2 dependence between T_C and T_{SC}, indicating a conventional Fermi liquid. As the field is increased, the temperature dependence of the resistivity becomes less like a Fermi liquid in the measured temperature range, which is expected as enhanced fluctuations drive the T^2 behavior to low temperature. At H^* the resistivity can be fitted with $\rho \sim T^{\frac{5}{3}}$ from 1.5 K to T_{SC}, in agreement with FM spin-fluctuation theory close to a FM critical point.[31] This coupled with $\frac{S}{T} \sim \ln(T)$ at H^*(see Fig. 1), which has been observed at an antiferromagnetic critical point[27] but can be applied to weakly ferromagnetic metals,[31] stresses that H^* is an FM critical point. Our experiment is only performed to 16 T, i.e., still close to H^*; in higher magnetic fields a $\rho \sim T^2$ law must be recovered, although this can be masked by quantum orbital effects when measuring magnetoresistivity in the transverse configuration. The enhanced FM correlations at this point would lead to an enhanced H_{c2} similar to the mechanism proposed[32] for reentrant superconductivity in URhGe. However, this scenario does not explain the presence of two anomalies in the thermopower. This plus the evidence of a Fermi surface modification in high magnetic field implies that the Fermi surface is strongly affected by the applied field. Enhancement of superconductivity through topological changes of the Fermi surface has already been considered for $CeCu_2Si_2$.[33] In the following we argue that the Fermi surface modification under field can lead to the enhanced T_{SC} in UCoGe.

In a metal the thermopower can be written as

$$\frac{S}{T} = -\frac{\pi^2 k_B^2}{3e}\left[\frac{1}{A_k}\frac{\delta A_k}{\delta E} + \frac{1}{l}\frac{\delta l}{\delta E}\right]_{E=E_f}, \tag{1}$$

where A_k is the area of the Fermi surface in reciprocal space, l is the mean free path, and E_f is the Fermi energy. For the following we assume l is energy independent, and therefore S/T is dominated by the first term in the equation. A large thermopower is observed when the area of the Fermi surface is strongly dependent on energy. This is the case near an extremum in the band where a small change in Fermi energy will change the Fermi surface area by a large amount. At finite temperature the observed increase in thermopower would be broadened by a factor kT, as observed in Fig. 2(c). If the Fermi energy is at an extremum in the band, then the Fermi velocity v_f is also very small ($v_f = |\frac{\delta E_k}{\delta k}|_{E_f}$). A decreased Fermi velocity will increase the orbital limit of superconductivity and therefore increase H_{c2} in the absence of the Pauli limiting (as is the case in UCoGe). In UCoGe, if the anomaly in S/T at H^* is the result of the band structure being modified by the field to a point with a reduced v_F, then the orbital limit of superconductivity will increase and H_{c2} will appear to increase. Figure 2(a) shows an H_{c2} curve calculated for a polar band state[30] with a v_F assumed to be related to S/T at low temperature ($S/T \sim 1/v_F$ normalized to the zero-field values) up to H^*. H^{**} would be a second band being modified, and then at higher fields the holelike Fermi surface has been suppressed, and an electron-like Fermi surface is recovered.

To date, there has been one band-structure calculation reported.[29] The calculation predicts there is a Fermi-surface change between the ferromagnetic and paramagnetic states. The paramagnetic state has three bands that cross the Fermi level, and the ferromagnetic state has four. Both states have a small Fermi surface, which is roughly consistent with the measured thermopower. In the ferromagnetic state, there are several extremum in bands close to the Fermi level particularly close to the Γ point of the Brillouin zone, which could result in an increased thermoelectric response. However, to confirm this band structure, a full quantum oscillation study needs to be performed. This is similar to UGe_2, where Fermi-surface reconstruction and critical spin fluctuations are associated, presumably due to the presence of flat bands near the Fermi level.[34] Recently, similar conclusions have also been reported for URhGe.[35]

IV. CONCLUSION

In conclusion, the zero-field thermopower of UCoGe implies a low-carrier metal in the ferromagnetic state. Applying a field perpendicular to the easy axis enhances the superconductivity, and the thermopower exhibits a sharp peak at low temperature in proximity to an enhanced T_{SC}. The temperature dependence of the thermopower is consistent with ferromagnetic fluctuations being driven to 0 K. At higher fields, a second anomaly is observed, and the thermopower changes sign, implying a Fermi surface reconstruction. Clearly, a Fermi surface change must also be included in theoretical descriptions. Our results stress the importance of taking into the account the Fermi surface in future theoretical studies of ferromagnetism around a critical point and emphasize the necessity to have a detailed understanding of the electronic band structure.

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