Thermoelectric evidence for high-field anomalies in the hidden order phase of URu_2Si_2

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Measurements of the thermoelectric coefficients of URu_2Si_2 in a high magnetic field could imply topological Fermi surface changes even deep inside the hidden order state. A change is observed in the thermopower as a function of the field applied parallel to the easy axis of magnetization, which could signify a change in the Fermi surface characteristics. The maximum of the thermopower coincides with previously measured anomalies in resistivity at $H^* = 23$ T. We analyze our results in terms of a Lifshitz transition in URu_2Si_2 originating from the hidden order Pauli depairing on a given subband.

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I. INTRODUCTION

At $T_0 \sim 17$ K, URu_2Si_2 undergoes a second order phase transition into a hidden order (HO) state.[1] This transition demonstrates a large drop in entropy, but to date no order parameter has been conclusively observed. There are various theoretical suggestions[2–8] for the hidden order parameter. Transport and thermodynamic probes point to a significant Fermi surface reconstruction at T_0 (Refs. 9–11), which leads to a large drop in carrier density. This decrease in carrier density corresponds to a decreased Fermi temperature and furthermore this weakness is reinforced by the large effective mass of the quasiparticles. Thus a moderate magnetic field (H) field can have a large effect on the Fermi surface characteristics. In addition, a magnetic field, applied parallel to the **c** axis, suppresses the hidden order and induces a cascade of transitions above 35 T (Ref. 12) ending with a paramagnetic metal above 40 T. Recent resistivity and Hall measurements[13] on high-quality single crystals have revealed anomalies and additional quantum oscillation frequencies at $H^* \sim 23$ T implying additional structure in the HO phase or even a possible phase transition. Thermodynamic measurements[14,15] show no evidence of anomalies in this field range. This may point to a more subtle topological change in the Fermi surface or "Lifshitz transition." If this is the case then the interplay between the Fermi surface, the HO order parameter and magnetic field could lead to fascinating insights into this compound and a better understanding of Fermi surface changes in the presence of strong correlations.

Measurements of the thermoelectric coefficients at low temperature can provide valuable information of the Fermi surface characteristics. Measurements of thermoelectric power on compounds such as $CeColn_5$ (Ref. 16) and $YbRh_2Si_2$ (Ref. 17) have helped elucidate the nature of the electronic state. In the case of URu_2Si_2, previous measurements have concentrated on a large increase in the thermoelectric power and Nernst signal on entering the hidden order state,[10,18] which is a result of the Fermi surface reconstruction and on the non-Fermi liquid-like behavior observed for currents applied along the a axis.[19] However, no previous study has combined low temperature and high field to study H^*. Since Lifshitz[20] first proposed the "2.5" type transition, there have been several theoretical[21,22] and experimental studies[23] of thermopower around a topological transition which show the thermopower is very sensitive to Fermi surface changes. An early proof of this has been given in a study of $CeRu_2Si_2$ through its pseudometamagnetic transition.[24] Recent measurements of thermopower[25] on the high temperature cuprates are another example of this and have provided evidence for previously unexpected changes in the Fermi surface.

Figure 1 shows a schematic temperature-field phase diagram for URu_2Si_2 when the field is applied along the **c** axis.[15,26–28] The hidden order state can be suppressed with a field of 35 T. At this point several unknown phases (possibly due to the quantum critical point created when suppressing the HO state to $T = 0$) are observed and then at higher fields a paramagnetic metal is recovered. Recent resistivity and Hall measurements at low temperatures (down to 250 mK) show anomalies at $H^* \sim 23$ T (Ref. 13).

Several transport studies of the Fermi surface of URu_2Si_2 have been performed.[26,29–31] However, despite recent progress[31] they cannot account for the correlated Sommerfeld coefficient observed by heat capacity, implying there are still some unobserved Fermi surface branches. The angle dependence of the frequencies implies a roughly isotropic Fermi temperature. Angle-resolved photoemission experiments[32] report a significant Fermi surface change through T_0 due to the heavy electron bands crossing the Fermi surface, in good agreement with a symmetry change inside the tetragonal class.[2,3,8,31]

In this paper we report measurements of the thermopower of URu_2Si_2 in magnetic fields up to 28 T to observe H^*. A change in thermopower is observed approaching H^* when the field is applied along the c axis of the tetragonal crystal. These results could be explained by a field-induced Lifshitz transition at H^* originating from a specific sheet of the Fermi surface. Complimentary measurements with the field applied along the a axis show little change as a function of field. This raises

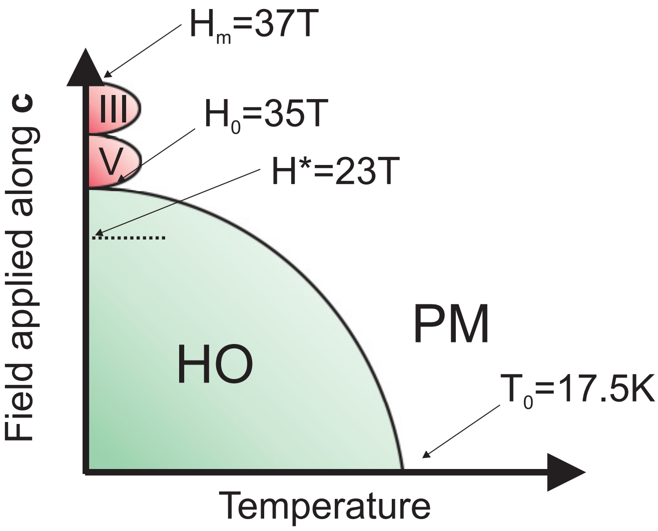

FIG. 1. (Color online) Schematic phase diagram of URu_2Si_2 with the field applied along the crystal **c** axis. The solid lines represent the well-established suppression of the hidden order phase (HO) into two unknown phases (V and III) and finally the paramagnetic phase (PM) measured by transport and thermodynamic probes (Refs. 26–28). The dashed line represents the resistivity anomalies observed by Shishido *et al.* (Ref. 13).

questions about the anisotropy of the HO phase and confirm a Fermi surface driven mechanism for the HO phase.

II. EXPERIMENTAL METHODS

High-quality single crystals were grown using the Czochralski pulling method in a tetra-arc furnace.[33] Sample 1 ($J \parallel a$, $H \parallel c$ configuration) had a residual resistance ratio (RRR) of 100, Sample 2 ($J \parallel H \parallel c$ configuration) and Sample 3 ($J \parallel H \parallel a$ configuration) had RRRs of 50. The thermopower was measured using the standard one heater, two thermometer setup, which also enabled measurements of the resistivity and Hall coefficients. Measurements up to 28 T and down to 1.3 K were performed in a resistive magnet at the Laboratoire National des Champs Magnétiques Intenses (LNCMI) Grenoble. These results were complemented with measurements at lower temperature and up to 16 T in a superconducting magnet. To eliminate any stray Nernst contributions, the magnetic field was applied in both positive and negative directions and the results averaged. Thermometer calibration was performed using a field compensated region up to 16 T and a capacitance thermometer at higher fields.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the thermoelectric power divided by temperature S/T as a function of magnetic field H up to 28 T. The field is applied along the c axis and the heat current J along the a axis. The thermopower is measured as the field is swept at different temperatures. The measured values of the thermopower below 12 T are in reasonable agreement with previous results.[10] S/T is negative and in the following discussion we refer to the magnitude of the thermopower $|S/T|$ (e.g., an increase/maxmimum in $|S/T|$ refers to an increase/maximum in the magnitude of S/T). At

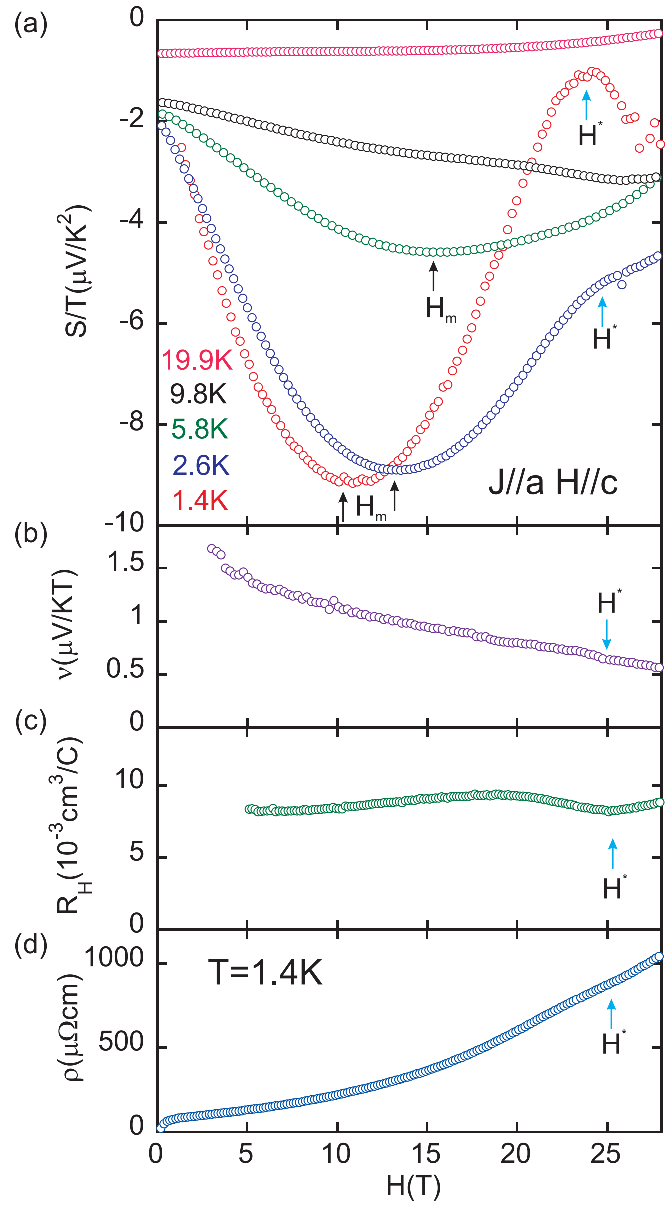

FIG. 2. (Color online) (a) Thermopower of URu_2Si_2 as a function of field for different temperatures. The black arrows correspond to H_m and the gray (blue) arrows to H^*. (b) The Nernst coefficient, (c) Hall coefficient, and (d) resistivity as a function of field showing more subtle anomalies at H^* all measured at 1.4 K.

the lowest temperature 1.4 K, $|S/T|$ shows an increase and a broad extremum at $H_m \simeq 11$ T followed by a decrease and minimum at $H^* \simeq 23$ T. The label H^* is taken from Ref. 13, where it corresponds to a extremum in the Hall resistance at ~23 T. As the temperature is increased both H^* and H_m are smeared out until at 10 K a monotonic increase is observed up to 28 T. Above T_0, in the paramagnetic state the thermopower is greatly reduced as previously reported.[10] Figure 2(b) shows the Nernst coefficient ν as a function of field, it monotonically decreases with increasing field as previously reported[10] and shows only a tiny change at H^*. Figures 2(c) and 2(d) show the Hall coefficient R_H and resistivity, respectively. There is a small kink in the resistivity and a modest change in the Hall coefficient at H^*, which is consistent with the previous

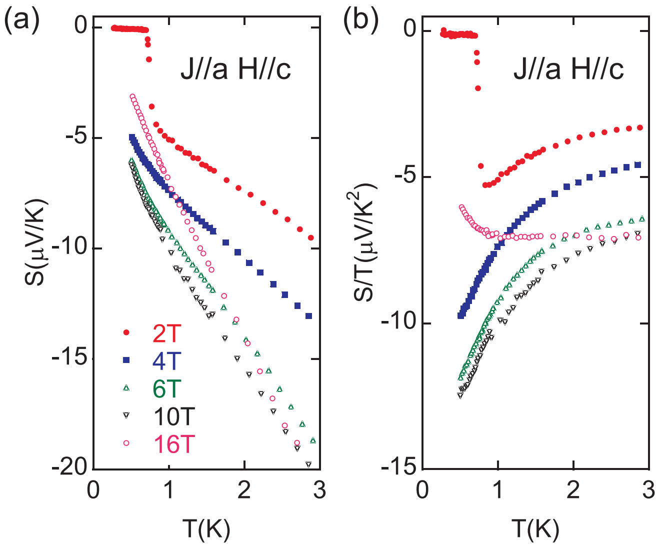

FIG. 3. (Color online) (a) Thermopower S of URu_2Si_2 measured with $J \parallel a$, $H \parallel c$ at several fields. (b) S/T vs temperature illustrating the change as a function of field of S/T.

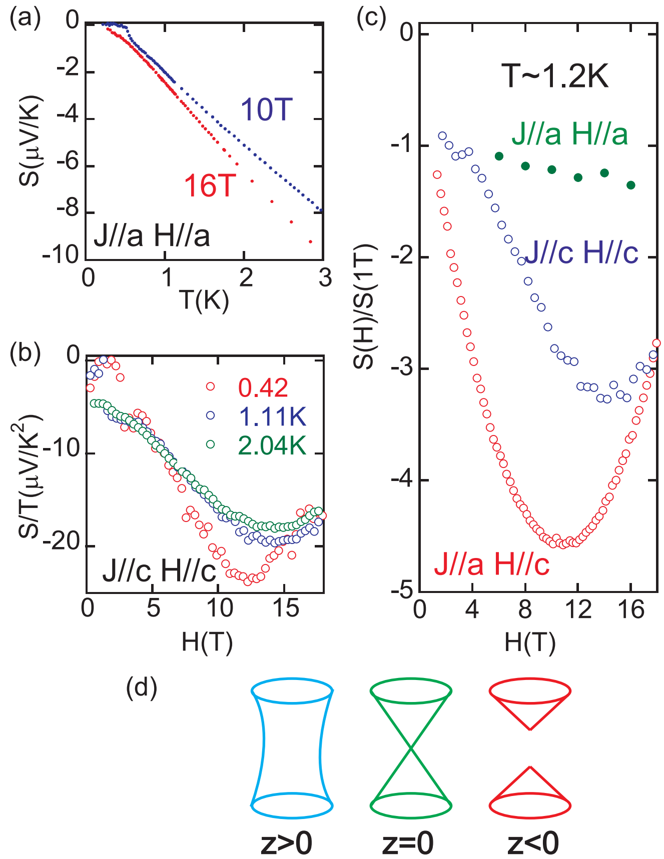

FIG. 4. (Color online) (a) Thermopower of URu_2Si_2 measured with $J \parallel H \parallel a$ in 10 and 16 T. There is little change in the amplitude between the two fields. (b) Thermopower of URu_2Si_2 measured with $J \parallel H \parallel c$ as a function of field at different temperatures. (c) A comparison of the different geometries measured showing the change in thermopower as a function of field at 1.2 K. The curves are normalized to the thermopower at 1 T. (d) Schematic of the topological change proposed by Lifshitz (Ref. 20), a Fermi surface neck is squeezed by tuning parameter z until at $z = 0$ it shuts completely.

measurements.[13] Resistivity measurements at 30 mK (Ref. 34) in the same geometry reveal a much sharper kink at H^*.

Figure 3 shows a more detailed temperature dependence of the thermopower in the same transverse configuration up to 16 T and down to lower temperature. There is a clear difference between the data in 10 T and 16 T. A maximum in $|S/T|$ is observed at $H_m \sim 10$ T followed by a decrease in magnitude up to 16 T. At 300 mK, the value of $|S/T|$ in 16 T is $-6\,\mu$ V/K^2 compared with $-12\,\mu$ V/K^2 in 10 T. The maximum of $|S/T|$ observed at 10 T and the minimum observed at 24 T could be the result of several contributions to the thermopower due to the multiband nature of URu_2Si_2. The possibility that different thermoelectric fields related to each specific Fermi surface of the different branches is an appealing one. This statement is supported by the most recent quantum oscillations study with high sensitivity[31] which observed a change in slope of resistivity above 8 T at very low temperature similar to that observed at 24 T. The cascade of associated phase transitions may occur at different temperatures. At the metamagnetic transition $H_0 = 35$ T, the full Fermi surface of the polarized paramagnetic phase of URu_2Si_2 will be restored.

To gain a better understanding of the observed thermopower, additional measurements were performed with $J \parallel H \parallel a$ and $J \parallel H \parallel c$ (Fig. 4). For $J \parallel H \parallel a$, only a small change is observed in $|S/T|$ as a function of field. There are no maximum or minimum up to 16 T in contrast to the measurements performed for $J \parallel a$, $H \parallel c$. However, for $J \parallel H \parallel c$ a large increase is observed with a maximum at $\simeq$13 T which is very similar to the change observed with $J \parallel c$, $H \parallel a$. Although the maximum is at a slightly higher field, this still shows that a change occurs in the Fermi surface. This implies that the change in thermopower is only present when the field is applied along the c axis. The HO phase will be destroyed at a much larger field along the a direction compared with the c direction. There is also a considerable anisotropy in the superconductivity, which is opposite compared with the conductivity to what is observed in more conventional systems.[19] Consequently, this topological change appears to be the result of the HO order parameter being strongly modified when a modest field is applied to the c axis, but less so when the field is applied to the a axis. A similar peak could be observed in the $J \parallel H \parallel a$ configuration at very high magnetic field, an estimate of the field based on the initial slopes of the two curves implies that it will occur in excess of 100 T. This strongly stresses that the polarization of the bands plays a major role. Finally, these measurements suggest that the scattering rate is not responsible for the change in thermopower as the longitudinal ($J \parallel H \parallel c$) measurements where $|S/T|$ is constant as a function of temperature[19] show a similar field dependence to the transverse ($J \parallel a, H \parallel c$) direction.

To consider the origin of these changes, the band structure must be examined. In URu_2Si_2, several bands have been observed by quantum oscillations[30,31] and there have been calculations of the band structure.[35] For $H \parallel c$, there are four observed quantum oscillation frequencies α, β, γ, and η with frequencies 1.05, 0.42, 0.19, and 0.09 kT, respectively. At high field (above H^*) an additional frequency ϵ is observed,[13] which is another indication of a Fermi surface change.

The cyclotron effective masses have been measured for all frequencies to be $m_\alpha^* = 13\,m_e$, $m_\beta^* = 25\,m_e$, $m_\gamma^* = 8.2\,m_e$, and $m_\eta^* = 20\,m_e$ with the field applied along the c axis. As expected from transport measurements, the bandwidth Δ_f of these bands are relatively low. The smallest bandwidths [$\Delta_f = \frac{\hbar eF}{m^* k_B}$, F is the relevant Shubnikov-de Haas (SdH) frequency] are the η band and β band (with values of 5.7 and 22 K, respectively). These bandwidths are low and comparable to the average gap involved in the Fermi surface reconstruction at T_0. Therefore a magnetic field of 20 T could create a Zeeman splitting comparable to the bandwidth and have a large effect on the HO gap structure. URu_2Si_2 is a compensated, multiband metal with different carriers (as evidenced by the different signs of the Hall and Seebeck coefficients). The large changes of $|S/T|$ (the increase to H_m and then decrease to H^*), which are observed with only modest changes in the Nernst and Hall coefficients, could be interpreted in a multiband model. The thermopower and Hall coefficient have two contributions (one hole and one electron) which have opposite signs. However, the Nernst coefficients of these two contributions have the same sign. As the HO is modified by the applied field on one subband then the carrier density increases and the thermoelectric coefficients for each band will decrease in magnitude. If the HO gap on one band is suppressed faster by the applied field then the relative contribution of each component to the thermopower will change (e.g., if the HO gap on a electron band is suppressed then the thermopower will become more positive). In this scenario, a small change would also be observed in the Hall coefficient as a change of the HO gap will change the carrier number. The Nernst coefficient would show a tiny change as both contributions are decreasing and have the same sign. If the HO gap is suppressed to zero then this could result in a change in band structure and a so-called Lifshitz transition. This also provides a natural explanation for the absence of an effect for a field applied along the a axis as no suppression can happen in the measured field range.

In proximity to a Lifshitz transition, one of the energy bands $\epsilon(k)$ is close to a singular point. Quantities, such as the thermopower, which are related to the derivative of the density of states, diverge at the singular point. The original Lifshitz paper[20] concentrated on a suppression of a neck in the Fermi surface [see Fig. 3(d)] and predicted thermoelectric quantities to diverge as $|z|^{-\frac{1}{2}}$ at $T = 0$ where z is the tuning parameter and $z = 0$ is the transition point. This transition only occurs at $T = 0$ and at finite temperature the divergences and anomalies are smeared out. It has been shown that impurities and temperature change the form of the divergence.[21] In addition, Lifshitz assumed a particular type of singular point in a simple band structure, but in a real metal the form of the divergence will depend on the details of the band structure around the singular point. It has also been shown[21] that the resistivity will show a kink-like feature close to a Lifshitz transition similar to the observed anomaly in Fig. 1(d).

In relation to the thermopower data of URu_2Si_2, the anomaly at H^* could be considered as a rounded divergence. In the HO state, the thermopower is large and negative,[10] however, at H^* it has become small at low temperature yet it is still in the hidden order state at 24 T. However, if there is an additional large, positive contribution from a band undergoing a Lifshitz transition then the total thermopower will be small. The fact that the anomaly at H^* is smeared out as the temperature is increased is consistent with a Lifshitz transition. It should be noted that the importance of Lifshitz transitions in heavy fermion compounds was first reported for $CeIn_3$ (Ref. 36) at the antiferromagnetic to paramagnetic boundary.

IV. CONCLUSION

In summary, we have measured the thermoelectric power of URu_2Si_2 in fields up to 28 T. Above 10 T, applied along the c axis, a dramatic in S/T is observed with a local maximum at 24 T. Small anomalies are also observed in resistivity and Hall effect measurements. These results suggest a Lifshitz transition in the HO state of URu_2Si_2 and explain the previously reported measurements of the resistivity and the Hall effect. Additional measurements with the field applied along the a axis show little change as a function of field. These results raise interesting questions of the interplay of the HO state with the Fermi surface and point to a Fermi surface driven mechanism for the HO order.

Recently, the confirmation of the feedback between spin polarization and Fermi surface evolution was given in a recent publication.[37]

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