Emergence of higher order rotational symmetry in the hidden order phase of URu_2Si_2

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Electrical resistivity measurements were performed as functions of temperature, magnetic field, and angle θ between the magnetic field and the c-axis of a URu_2Si_2 single crystal. The resistivity exhibits a two-fold oscillation as a function of θ at high temperatures, which undergoes a 180°-phase shift (sign change) with decreasing temperature at around 35 K. The hidden order transition is manifested as a minimum in the magnetoresistance and amplitude of the two-fold oscillation. Interestingly, the resistivity also showed four-fold, six-fold, and eight-fold symmetries at the hidden order transition. These higher order symmetries were also detected at low temperatures, which could be a sign of the formation of another pseudogap phase above the superconducting transition, consistent with recent evidence for a pseudogap from point-contact spectroscopy measurements and NMR. Measurements of the magnetization of single crystalline URu_2Si_2 with the magnetic field applied parallel and perpendicular to the crystallographic c-axis revealed regions with linear temperature dependencies between the hidden order transition temperature and about 25 K. This T-linear behavior of the magnetization may be associated with the formation of a precursor phase or "pseudogap" in the density of states in the vicinity of 30-35 K.

Keywords: hidden order, rotational symmetry, heavy-fermion metals.

1. Introduction

Emergent phenomena are often manifested in systems with strong electronic correlations. The compound URu_2Si_2, a tetragonal heavy fermion f-electron system, exhibits two such phenomena, unconventional superconductivity below 1.5 K and an unidentified so-called "hidden order" (HO) phase below T_0 = 17.5 K that apparently coexist with one another [1–3] and occupy different parts of the Fermi surface [2]. In this mysterious HO phase, a small Ising-like antiferromagnetic moment of $0.03\mu_B$/U was detected by neutron diffraction [4, 5]; however, the moment is too small to account for the entropy of $0.2R\ln(2)$ derived from the striking mean-field like anomaly in specific heat $C(T)$ associated with the HO

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phase [2]. The HO phase transition is also manifested in the electrical resistivity $\rho(T)$ and magnetization $M(T)$ as an anomaly and a change in slope at T_0, respectively.

Almost three decades of research on URu_2Si_2 has demonstrated that the HO is related to a delicate interplay between magnetic and charge degrees of freedom, but has not succeeded in revealing the identity of its order parameter. Broadly, the puzzle of the hidden order starts with the dichotomy of order that involves itinerant vs. localized charge and spin degrees of freedom. By now, the preferred explanations seem to focus more on the itinerant degrees of freedom of carriers as a main channel where HO develops. As originally inferred from the specific heat [1, 2], and later confirmed by optical conductivity [6], a charge gap of $\Delta \approx 11$ meV opens over about 40% of the Fermi surface [2]. ^{29}Si nuclear magnetic resonance (NMR) experiments [7] show the presence of strong spin-fluctuations above the HO phase. These measurements suggest that the HO phase is preceded by the formation of a pseudogap phase that forms between $T_{\rm pg} = 30$ K and T_0, as previously proposed by Haraldsen *et al.* [8]. Features related to a pseudogap were also observed via optical [9] and point-contact spectroscopy measurements [10]. This putative pseudogap phase is interpreted as a precursor to a mean-field regime where a gap occurs without true long-range order, due to competition between the antiferromagnetic and HO phases [11].

In addition, inelastic neutron scattering experiments have revealed the existence of a spin gap (2 meV) at the commensurate wave vector $\boldsymbol{Q}_0 = (1, 0, 0)$, which is also related to the antiferromagnetic order that transforms to weak quasielastic spin fluctuations above T_0 [12]. More recent experiments [13] have shown that a second gap (4 meV) at the incommensurate wave vector $\boldsymbol{Q}_1 = (0.4, 0, 0)$ is due to itinerant-like spin excitations that are related to the heavy electronic quasiparticles that form below a coherence temperature $T^* \approx 70$ K. This also suggests that the hybridization of localized uranium $5f$ electrons with the itinerant conduction electrons is important for an understanding of the HO phase. This is underscored by angle-resolved photoemission spectroscopy (ARPES) [14], scanning-tunneling microscopy (STM) [15, 16], and point-contact spectroscopy (PCS) [17] studies, which reveal that the electronic structure of URu_2Si_2 is reorganized below T_0 where a heavy quasiparticle band shifts below the Fermi level, and the crossing with a light hole-like band at $Q^* = \pm 0.3\pi/a$ leads to the formation of a hybridization gap Δ_{Q^*} = 5 meV.

Several studies have shown that high magnetic fields can be used to tune the amount of hybridization in URu_2Si_2 [18–20], eventually resulting in the suppression of the HO phase and an accompanying reconstruction of the Fermi surface within the HO phase as revealed by Shubnikov-de Haas oscillation experiments [21, 22]. More recent angle-dependent Shubnikov-de Haas measurements [23] demonstrate that the Fermi surface branches are anisotropic, where the β branch is observed to split when the applied magnetic field is rotated from the crystallographic a- to the c-axis. This anisotropy is further reflected in the Zeeman energy of URu_2Si_2 [24, 25]. This anisotropic Ising-like g-factor has consequently been interpreted to point to an itinerant hidden order parameter involving quasiparticles whose spin degrees of freedom depart significantly from those of free electrons [24]. Correspondingly, the electrical resistivity and magnetization of URu_2Si_2 are both strongly anisotropic in which the c-axis is the easy axis [1, 26].

In this paper, we demonstrate that the electrical resistivity as a function of temperature T, magnetic field H, and angle θ between the magnetic field and the c-axis of URu_2Si_2 displays several new interesting features. In addition to the two-fold oscillations, even harmonics are also observed. Each $2n$-fold oscillation, where n is an integer, displays an anomaly at the HO transition temperature T_0. Finally, the magnetoresistivity changes sign at T = 35 K, in proximity to the pseudo-gap feature at $T_{\rm pg}$. Measurements of the magnetization with the magnetic field applied parallel and perpendicular to the c-axis reveal regions with linear temperature dependencies between T_0 and about 25 K. This T-linear behavior may be associated with the formation of a precursor phase or "pseudogap" in the density of states in the vicinity of 30-35 K.

2. Experimental Methods

The single crystal of URu_2Si_2 studied in these experiments was grown by the Czochralski method in a tetra-arc furnace and oriented by using a back-reflection Laue CCD camera. Electrical resistivity measurements were made for 1.8 K $\leq T \leq$ 200 K under magnetic fields H from 0 to 9 T with a standard 4-wire technique in a Quantum Design Physical Property Measurement System (PPMS) DynaCool with a rotator option. The current was applied perpendicular to the c-axis of the sample. The sample was rotated along the ac plane with θ ranging from 0° to 355°. Specific heat was measured using a thermal-relaxation technique in a PPMS DynaCool. The specific heat contribution from the Apiezon N-grease that was used to thermally and mechanically couple the single crystal to the calorimeter platform was measured in a separate experiment and subtracted. Magnetization was measured in a Quantum Design Magnetic Property Measurement System.

3. Results and Discussion

The electrical resistivity data exhibit a simple two-fold oscillation, $\cos(2\theta)$, at most temperatures. This is expected for the tetragonal crystal structure of URu_2Si_2. However, the data (not shown) cannot be described with a simple cosine function around the hidden order transition temperature T_0. By using Fourier analysis, the electrical resistivity data can be fitted with the expression,

$$\rho = \rho_0 + |A_{2\theta}|\cos(2\theta - \phi_{2\theta}) + |A_{4\theta}|\cos(4\theta - \phi_{4\theta}) + ..., \quad (1)$$

where ρ_0 is an average electrical resistivity, $|A_{n\theta}|$ is an amplitude for oscillations of the resistivity around ρ_0 with n-fold symmetry, and ϕ is a phase angle.

Figure 1(a) shows ρ_0 vs. T in various fields. The HO transition temperature T_0 is visible as an anomaly around 17 K, which decreases with increasing field as can be seen in Fig. 1(b). To further study the effect of magnetic field on ρ_0, the magnetoresistivity $\frac{\Delta\rho}{\rho_0} = \frac{\rho_0(H)-\rho_0(0)}{\rho_0(0)}$ was calculated. The HO transition in $\frac{\Delta\rho}{\rho_0}$ is manifested as a minimum (Figs. 1(c) and 1(d)), which reflects the shift of the feature associated with the HO, T_0, to lower temperatures with increasing magnetic field. Interestingly, $\frac{\Delta\rho}{\rho_0}$ changes sign at 35 K, which is very close to the temperature at which various physical properties exhibit behavior that has been suggested to be associated with a transition into a pseudogap phase [8]. The change of sign is associated with a 180°-phase shift of the phase angle ϕ, which is equal to either 0° or 180° for the whole temperature range (see Figs. 1(e) and 1(f)).

To compare the oscillation amplitude for different temperatures and magnetic fields, the amplitude of the oscillatory resistivity A, where $A_{n\theta} = |A_{n\theta}|\cos\phi$, over the average electrical resistivity ρ_0, $\frac{A_{n\theta}}{\rho_0}$, was calculated and plotted vs. temperature, as illustrated in Fig. 2. Here, negative values of $A_{n\theta}$ arise due to a phase angle of $\phi = \pi$. T_0 can be observed in $A_{2\theta}$ as a minimum (Fig. 2(a)). Moreover, four-fold, six-fold, and eight-fold oscillations were detected around T_0 as can be seen in Figs. 2(b), 2(c), and 2(d), respectively. $A_{4\theta}$, $A_{6\theta}$, and $A_{8\theta}$ are zero at most temperatures, except around T_0 and at $T \leq 4$ K, which could be an indication of another pseudogap phase related to the occurrence of superconductivity

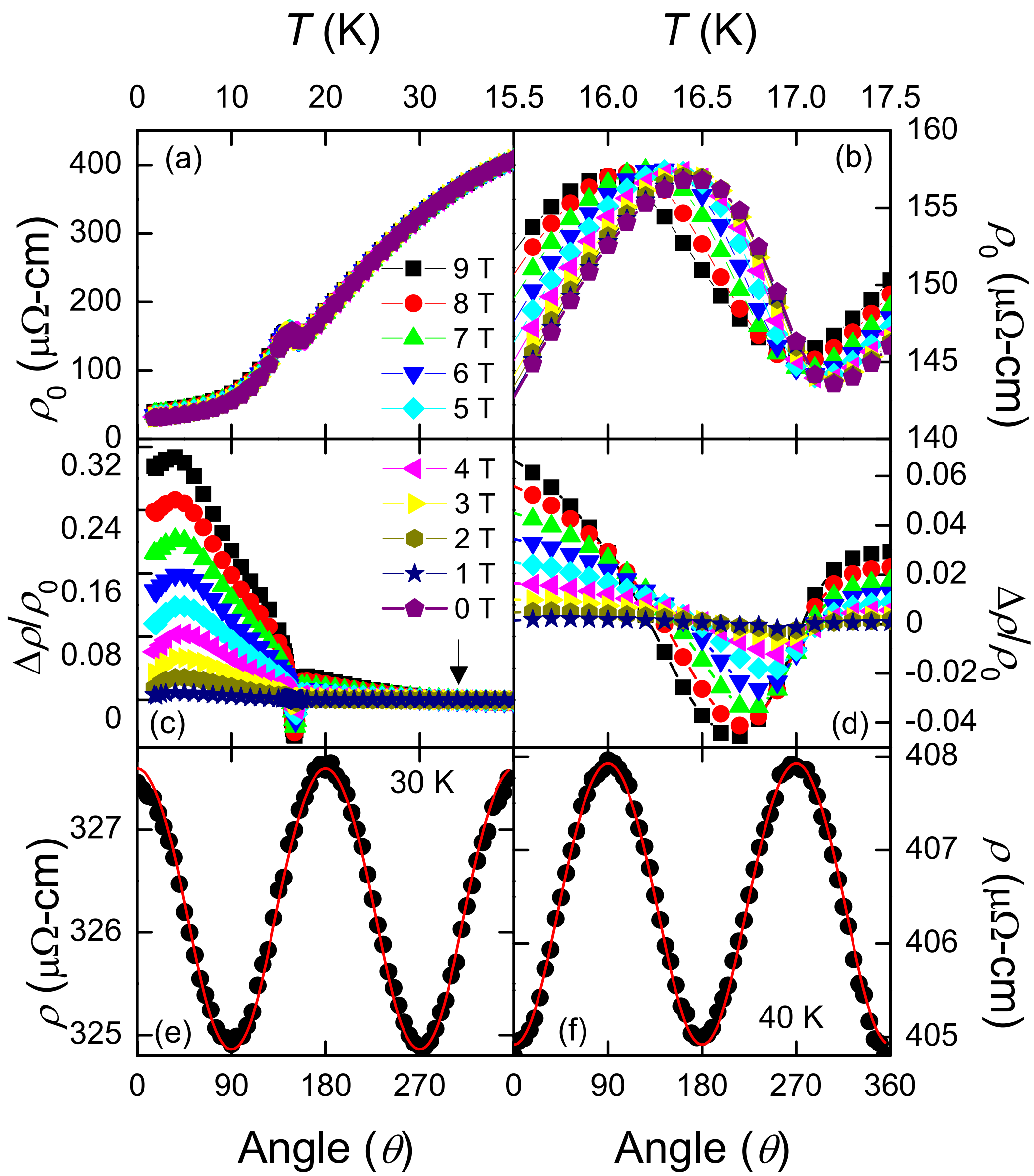

Figure 1. (Color online) (a) Average electrical resistivity ρ_0 vs. temperature T in various magnetic fields H. (b) Expanded view of the region around the hidden order transition temperature T_0 in ρ_0. (c) Magnetoresistance $\frac{\Delta\rho}{\rho_0}$ vs. T. The arrow indicates the temperature where $\frac{\Delta\rho}{\rho_0}$ changes sign (see text). (d) Expanded view of the region around T_0 in $\frac{\Delta\rho}{\rho_0}$. (e) Electrical resistivity ρ vs. angle θ at 30 K where the phase angle ϕ = 0°. (f) ρ vs. θ at 40 K where ϕ = 180°. Filled circles represent the data, and the red solid line represents the resulting fit using Eq. (1).

below 1.5 K [27]. We further note that the magnitude of the oscillation increases as a function of increasing magnetic field.

An expanded view of the angular and temperature dependence of the HO transition is shown in Fig. 3. T_0 can be identified with an anomaly in $\frac{A}{\rho_0}$ as can be seen in Figs. 3(a)-(d). In addition, the magnitude of the anomaly becomes smaller with increasing order of symmetry. Figures 3(e)-3(h) show the behavior of the derivative of A with respect to T. Interestingly, there is a striking similarity between the plots of $\frac{A_{2\theta}}{\rho_0}$ and $\frac{\Delta\rho}{\rho_0}$, $\frac{A_{4\theta}}{\rho_0}$ and $\frac{d(A_{2\theta}/\rho_0)}{dT}$, $\frac{A_{6\theta}}{\rho_0}$ and $\frac{d(A_{4\theta}/\rho_0)}{dT}$, as well as $\frac{A_{8\theta}}{\rho_0}$ and $\frac{d(A_{6\theta}/\rho_0)}{dT}$.

Figure 4 displays fits of Eq. (1) to the electrical resistivity data around T_0 at 9 T and the

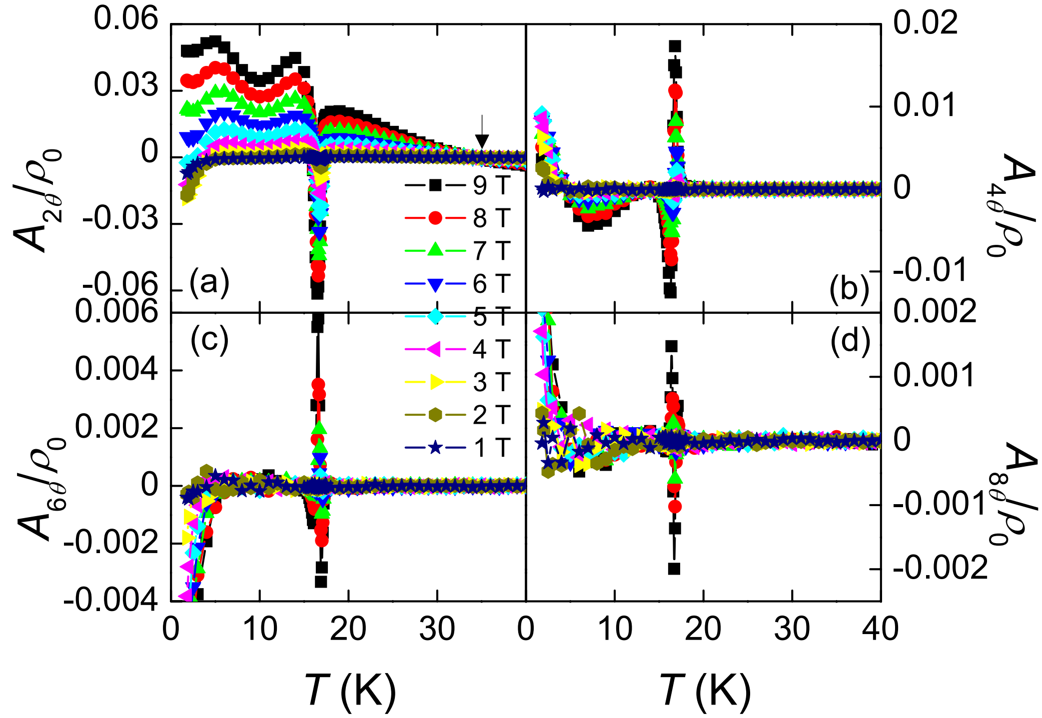

Figure 2. (Color online) Amplitude of the oscillatory resistivity A over the average resistivity ρ_0, $\frac{A}{\rho_0}$, vs. temperature T. (a) $\frac{A_{2\theta}}{\rho_0}$. The arrow indicates the temperature where $\frac{A_{2\theta}}{\rho_0}$ changes sign (see text). (b) $\frac{A_{4\theta}}{\rho_0}$. (c) $\frac{A_{6\theta}}{\rho_0}$. (d) $\frac{A_{8\theta}}{\rho_0}$.

value of $\frac{A}{\rho_0}$ extracted from the fits. As can be seen from Fig. 4(a), the magnitude of the anomaly decreases with increasing order of symmetry.

The origin of the $(n \neq 2)$ even-fold oscillations in the θ-dependence of the electrical resistivity remains to be determined. However, as the oscillations only occur for temperatures below the HO transition, it seems that they are not simply due to higher-order harmonics of the Ising-like anisotropy since that is already present in the high-temperature phase, but seem rather intimately related to the properties of the HO itself. One possibility is that these oscillations are due to the appearance of a high-order multipolar order parameter which opens up orientation-dependent scattering channels. Low-rank multipolar order parameters, such as quadrupolar and octupolar order parameters have been ruled out by resonant x-ray scattering experiments [28, 29] and the measured neutron scattering form factor [4]. Likewise, the upper bounds on the internal fields on the Si and Ru sites inferred from NMR measurements and a group theoretical analysis [30] of recent neutron diffraction data [31] make low odd-rank multipolar ordering seem improbable. However, higher-rank multipolar order parameters are difficult to discern since they would only show up in the high-momentum transfer tail of the neutron scattering form factors where the statistics are poor. Although there are theories that describe states with transverse magnetic anisotropy [32–35], there are no obvious mechanisms by which they could reproduce the oscillations reported here.

Another possibility is that the oscillations observed here are the result of gaps that open up in the electronic density of states due to a charge-density wave [36] or hybridization

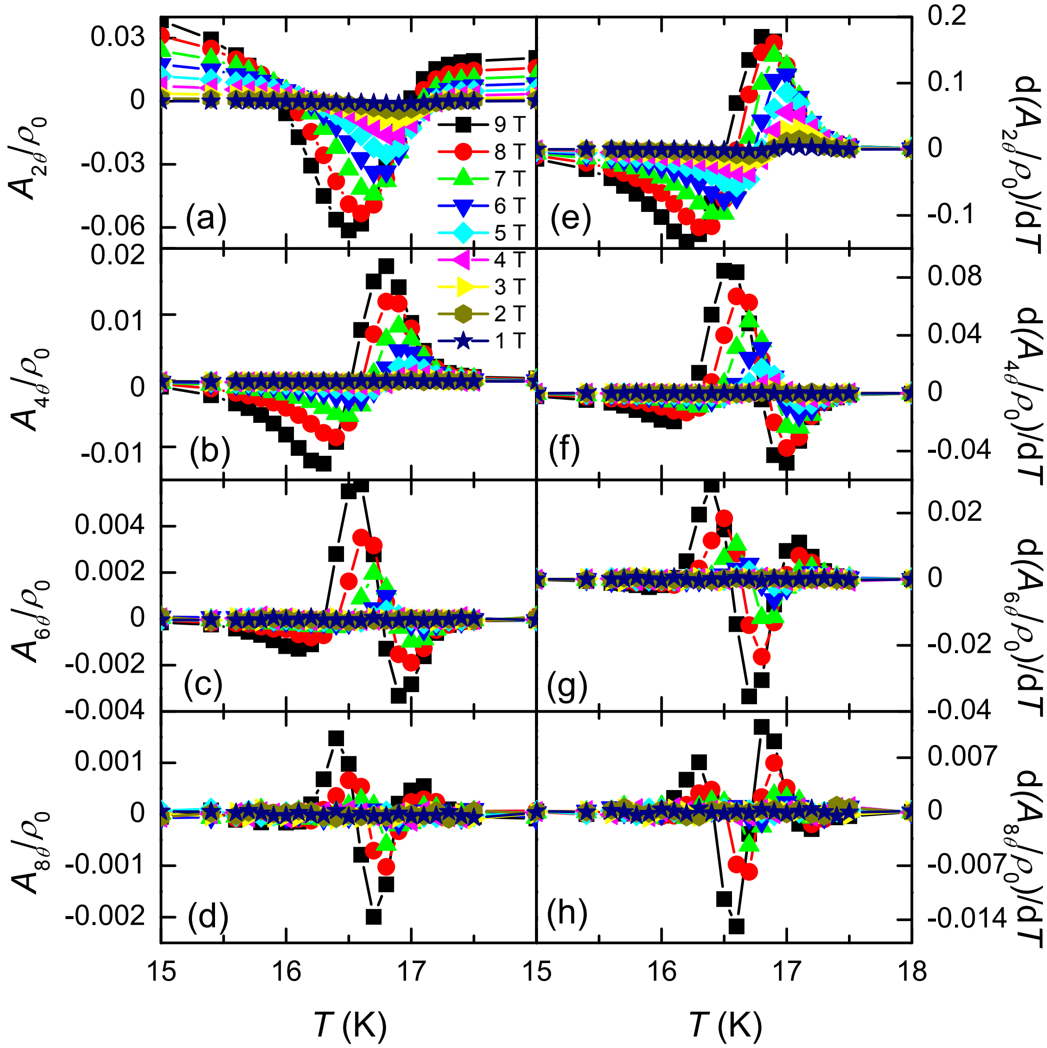

Figure 3. (Color online) (a)-(d) Amplitude of the oscillatory resistivity A over the average electrical resistivity ρ_0, $\frac{A}{\rho_0}$, vs. temperature T around the hidden order transition temperature T_0. (e)-(h) Derivative of $\frac{A}{\rho_0}$ with respect to T, $\frac{d(A/\rho_0)}{dT}$, vs. T.

wave [37]. Notably, an early analysis of the specific heat [2] has already demonstrated that only about 40% of the Fermi surface is gapped, and the symmetry of this gap may be at the origin of the observed higher-order harmonics. A density-wave order parameter could also encompass the hybridization of itinerant electronic degrees of freedom with an unconventional localized spin or multipolar degree of freedom as has been previously suggested based on the highly anisotropic g-factor of the heavy quasiparticles that drive the HO transition [24]. This latter scenario may also provide a natural explanation for the fact that the oscillations become stronger with increasing magnetic field; magnetic field is known to decrease the hybridization between localized and itinerant degrees of freedom, and could be imagined to increase the anisotropic scattering of the localized degrees of freedom. However, more measurements in higher magnetic fields that go beyond this study would be required to investigate this question.

In a recent paper [8], magnetic susceptibility measurements on a polycrystalline specimen of URu_2Si_2, previously reported by Maple *et al.* [2], were reanalyzed and found to contain a region in which the susceptibility was linear in temperature between T_0 and

$\sim$30 K. It was suggested that this T-linear region was associated with the occurrence of a precursor phase or "pseudogap" in the density of states in the vicinity of 30 K. Since the T-linear region of the magnetic susceptibility of the polycrystalline sample of URu_2Si_2 could be the result of a "polycrystalline average" over crystallites with different orientations and not an intrinsic property, we measured the magnetization $M(T)$ of a URu_2Si_2 single crystal for two orientations of the magnetic magnetic field, $H \parallel c$-axis and $H \perp c$-axis. The results of these two measurements are shown in Figs. 5(a) and (b), respectively. For both directions, there is a region between T_0 and $\sim$25 K where M has a linear T-dependence, suggesting that this is an intrinsic property. In an attempt to address this issue further, specific heat $C(T)$ measurements were performed on a URu_2Si_2 single crystal in zero field in the vicinity of T_0, the results of which are shown in Fig. 6. We have plotted the electronic contribution to the specific heat, C_e, extracted from the $C(T)$ data by subtracting the lattice contribution to specific heat, vs. T. Interestingly, $C_e(T)$ also contains a region between T_0 and $\sim$30 K that is linear in T, indicated by the straight line through the data in the shaded region in Fig. 6. However, this behavior is consistent with that expected for a Fermi liquid and therefore does not constitute clear evidence for the formation of a pseudogap in this region. Nonetheless, it seems possible that the T-linear behavior found in the magnetization M of single crystalline URu_2Si_2 for the two orientations of the magnetic magnetic field, $H \parallel c$-axis and $H \perp c$-axis, is associated with the formation of a precursor phase or "pseudogap" in the density of states in the vicinity of 30-35 K. As noted by Haraldsen $et\ al.$ [8], evidence for such a pseudogap has been found in a variety of measurements such as PCS, inelastic neutron scattering, NMR, and, as described in this paper, by the sign change in the magnetoresistance.

The challenge of determining the nature of the HO phase stems from what likely is its complicated character, where neither simple charge nor spin order would do. Hidden order is more likely of multiorbital or multipolar nature and exhibits neither simple d- nor f-electron character. Consequently, measurements that only probe single aspects of the complex dichotomy that is the HO, such as spin or charge scattering, are not suitable for identifying its order parameter (OP). Sophisticated types of characterization, including the angular dependency of the magneto-transport data presented here, seem to be more sensitive to the symmetry of the OP, and their understanding will be required to paint a full picture of the HO phase.

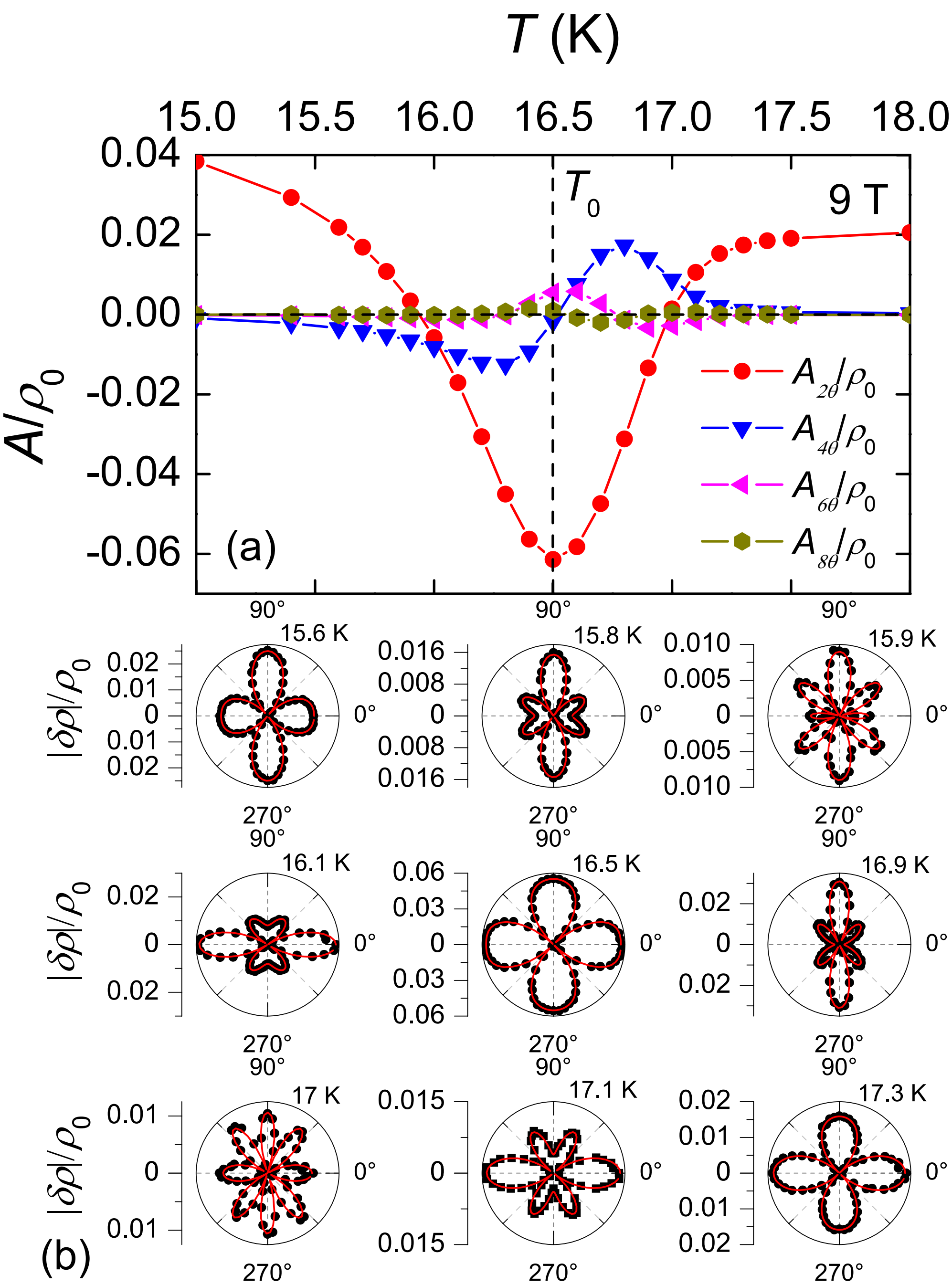

Figure 4. (Color online) (a) Amplitude of the oscillatory resistivity A over the average electrical resistivity ρ_0, $\frac{A}{\rho_0}$, vs. temperature T around the hidden order transition temperature T_0 (16.5 K at 9 T). The horizontal dashed line corresponds to $\frac{A}{\rho_0} = 0$, and the vertical dashed line is where T_0 is located. (b) Polar plots of the absolute value of the change in electrical resistivity divided by the average resistivity, $\frac{|\delta\rho|}{\rho_0}$, as a function of the angle between the applied magnetic field H of 9 T and the c-axis in the vicinity of the hidden order transition temperature. Filled circles represent the data, and the red solid line represents the resulting fit of Eq. (1) to the data.

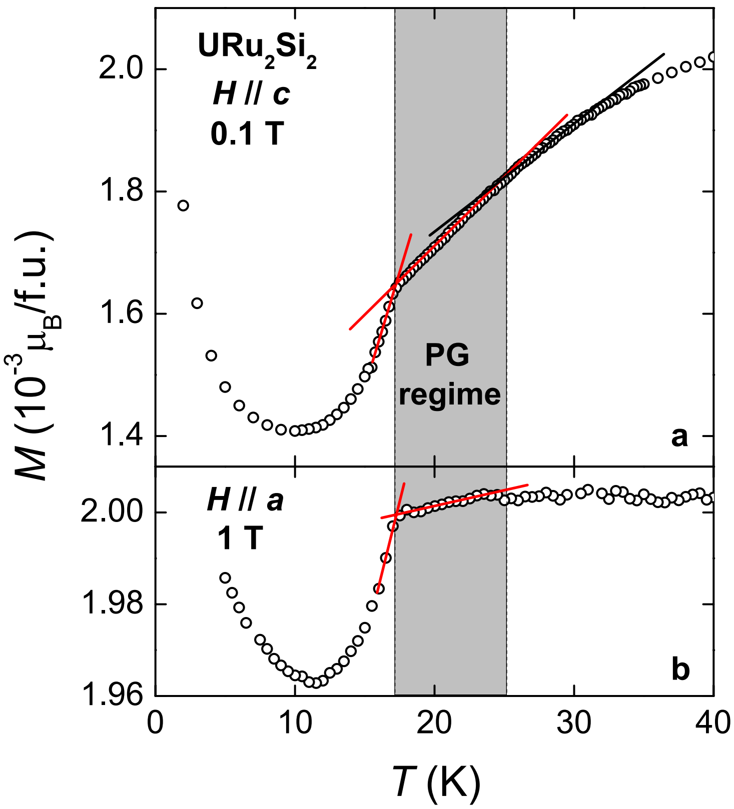

Figure 5. (Color online) Magnetization M as a function of temperature T with (a) magnetic field H = 0.1 T applied parallel to the crystallographic c axis, and (b) with H = 1 T applied parallel to the a axis. Lines are guides to the eye that demonstrate a temperature region just above T_0 where the $M(T)$ data are linear in temperature. The gray region delineates the boundaries of the linear temperature dependence of M, and is labeled pseudogap (PG) regime.

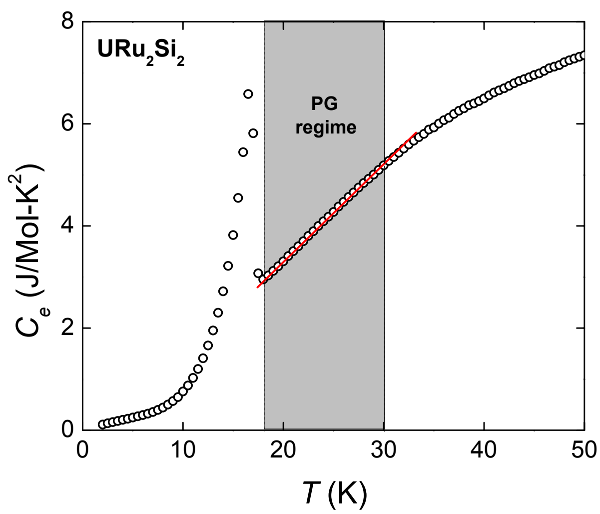

Figure 6. (Color online) Electronic contribution to specific heat, C_e, vs. temperature, T. The line through the data is a guide to the eye, emphasizing a linear temperature dependence of $C_e(T)$ at temperatures just above T_0. The gray region delineates the pseudogap (PG) regime within which there is evidence for a pseudogap, based on other measurements described in the text.

4. Concluding remarks

We have found that the electrical resistivity data for URu_2Si_2 exhibit two-fold rotational symmetry in the ac plane at most temperatures. However, higher order rotational symmetry emerges around the HO transition and at low temperatures. The magnetoresistivity changes sign at around 35 K, corresponding to where there is a $180°$-phase shift in the amplitude for two-fold symmetry, which may be related to the proposed phase transition to a pseudogap phase. Measurements of the magnetization of single crystalline URu_2Si_2 with the magnetic field applied parallel and perpendicular to the crystallographic c-axis reveal linear temperature dependencies between the hidden order transition temperature and about 25 K. Higher order rotational harmonics of electrical resistivity found in this study provide additional constraints for any proposed model of the hidden order. Further investigation will be required to unveil the origin of the higher harmonics in the oscillations of the electrical resistivity in the HO phase, which could be a key to solving the puzzle of the mysterious hidden order phase.

Acknowledgments

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