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Anomalous femtosecond quasiparticle dynamics of hidden order state in URu_2Si_2

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At $T_0 = 17.5$ K an exotic phase emerges from a heavy fermion state in URu_2Si_2. The nature of this hidden order (HO) phase has so far evaded explanation. Formation of an unknown quasiparticle (QP) structure is believed to be responsible for the massive removal of entropy at the HO transition, however, experiments and *ab initio* calculations have been unable to reveal the essential character of the QP. Here we use femtosecond pump-probe time- and angle-resolved photoemission spectroscopy (tr-ARPES) to elucidate the ultrafast dynamics of the QP. We show how the Fermi surface is renormalized by shifting states away from the Fermi level at specific locations, characterized by vector $q_{\langle 110\rangle} = 0.56 \pm 0.08$ Å^{-1}. Measurements of the temperature-time response reveal that, upon entering the HO, the QP lifetime in those locations increases from 42 fs to few hundred fs. The formation of the long-lived QPs is identified here as a principal actor of the HO.

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Over the past 25 years, the nature of the hidden order (HO) transition in a heavy fermion system URu_2Si_2 has remained a mystery. The sharp second-order transition at $T_0 = 17.5$ K,[1–13] marked by a large jump in specific heat, corresponds to the removal of more than 10% of total entropy.[14] The phase transition might have been consistent with magnetism, but several years of intensive search showed the absence of magnetism in the HO phase. Antiferromagnetism is observed only under pressure.[15] Partial gapping of the Fermi surface (FS) was proposed in the context of itinerant models[8,9,11,12] and the HO gap was shown to behave similarly to a BCS order parameter.[16] Entropy removal at the HO transition also suggested that the Fermi-surface instability induces reconstruction of the density of states. It was proposed that a commensurate[6,9,10,17,18] or incommensurate[4,11,12,19] renormalization of the Fermi surface is the driver of the HO transition, with magnetic fluctuations[9] or a hybridization wave[11,12] as mechanisms for the gap formation. However, the underlying physics behind the key quasiparticles (QPs), the gap formation, symmetry, and momentum dependence remain elusive, despite several models of HO proposed over the years.[4,7–10,12,17,18]

The picture of the HO is also obscured by the formation of a hybridization gap, a typical feature of heavy fermion materials.[20] Such a gap formation is due to hybridization of the flat f band with a strongly dispersive d band, as evidenced by numerous experiments.[1–3,14,21,22] The hybridization gap onset is related to the coherence temperature T^*, usually from 60 to 100 K for uranium-based heavy fermion materials[23] and estimated at 70 K in URu_2Si_2. At T_0 the stage is already set by a well-established f-d hybridization gap structure evolving in a mean-field behavior.[24,25]

To reveal the nature of the HO transition within the hybridization gap, one needs to track the QPs responsible for the Fermi-surface renormalization. Several recent investigations using angle-resolved photoemission spectroscopy (ARPES), ion neutralization spectroscopy (INS), and scanning tunneling microscopy (STM) demonstrate that the HO transition is marked by modification of the density of states.[13,14,16,21] The two recent ARPES experiments[21,26] and this Rapid Communication can be reconciled with the three-dimensional nature of the electronic structure in URu_2Si_2.[27] These findings set the stage for resolving the critical unknowns: What is the nature of the QPs driving the HO transition? How does the HO gap structure evolve in energy-momentum space? Where does the missing entropy[14,17] go? And finally, is the hidden order gap related to the hybridization gap? To answer these questions, the temperature and time structure of the transition-driving QPs needs to be examined. Here we report a unique approach of time-resolved ARPES (tr-ARPES) measurements on the femtosecond scale, measuring the ultrafast QP dynamics in normal and HO states with 29.5-eV probe photons, at a position in momentum space of the body-centered-tetragonal (bct) Brillouin zone along k_z slightly below the Z point. Time-resolved ARPES allows us to populate the empty states with a pump pulse of 1.55 eV and measure the energy- and momentum-resolved electronic structure of the bands above the Fermi level. The populated electronic structure decays at a rate proportional to QP lifetime limited by electron-electron or electron-phonon scattering processes occurring in different time scales. The evolution of this structure is measured in small time intervals by a probe pulse to trace the decay. A temporal resolution of better than 50 fs is needed for analyzing the electron-electron processes, and a photon energy in the

range of cross sections favorable for 5*f* photoionization is crucial. The tr-ARPES instrument utilizing a high-harmonic generation (HHG) and time-compensating monochromator was constructed specifically for work with *f*-electron systems, and utilizes noble gas based HHG excited by a 30 fs, 1.55 eV Ti:sapphire laser with a 10-kHz repetition rate, coupled to the time-compensated double monochromator.[28] The differential pumping system maintains UHV conditions in the measurement chamber, where photoelectron spectra are acquired with a hemispherical electron energy analyzer. We use 1.55-eV, 10-kHz, 30-fs pump pulses with average power of 40 mW to avoid excessive heating of the electron gas, and 29.5-eV, 15-fs probe pulses to achieve a favorable cross section for *f*-orbital photoionization. The choice of temperature range, between 12 and 19K, was dictated by (a) the need to capture the transition at 17.5 K, and (b) the surface sensitivity of URu_2Si_2 requiring minimal sample exposure time and careful use of in-vacuum sample heaters. ARPES experiments were performed at the Synchrotron Radiation Center, utilizing the PGM beamline 71A and the SCES4000 hemispherical electron energy analyzer, with an energy resolution of 15 meV at 34-eV photons. Single crystals of URu_2Si_2 were grown via the Czochralski technique in electrical tri-arc and tetra-arc furnaces, followed by a 900 °C anneal in Ar for 1 week in the presence of a Zr getter.

Figure 1 shows the electronic structure of URu_2Si_2 in the ⟨110⟩ direction measured with tr-ARPES in the HO state at 12 K. Figure 1(a) shows the structure as seen with the probe pulse arriving 1 ps before the pump pulse. This is the semistatic structure, since the 10-kHz repetition rate allows all the states populated with the previous pump pulse to decay long before the probe pulse arrives. This structure is used as a baseline, and is subtracted from data taken at other delays, shown in all consecutive panels in Fig. 1. This subtraction allows us to see

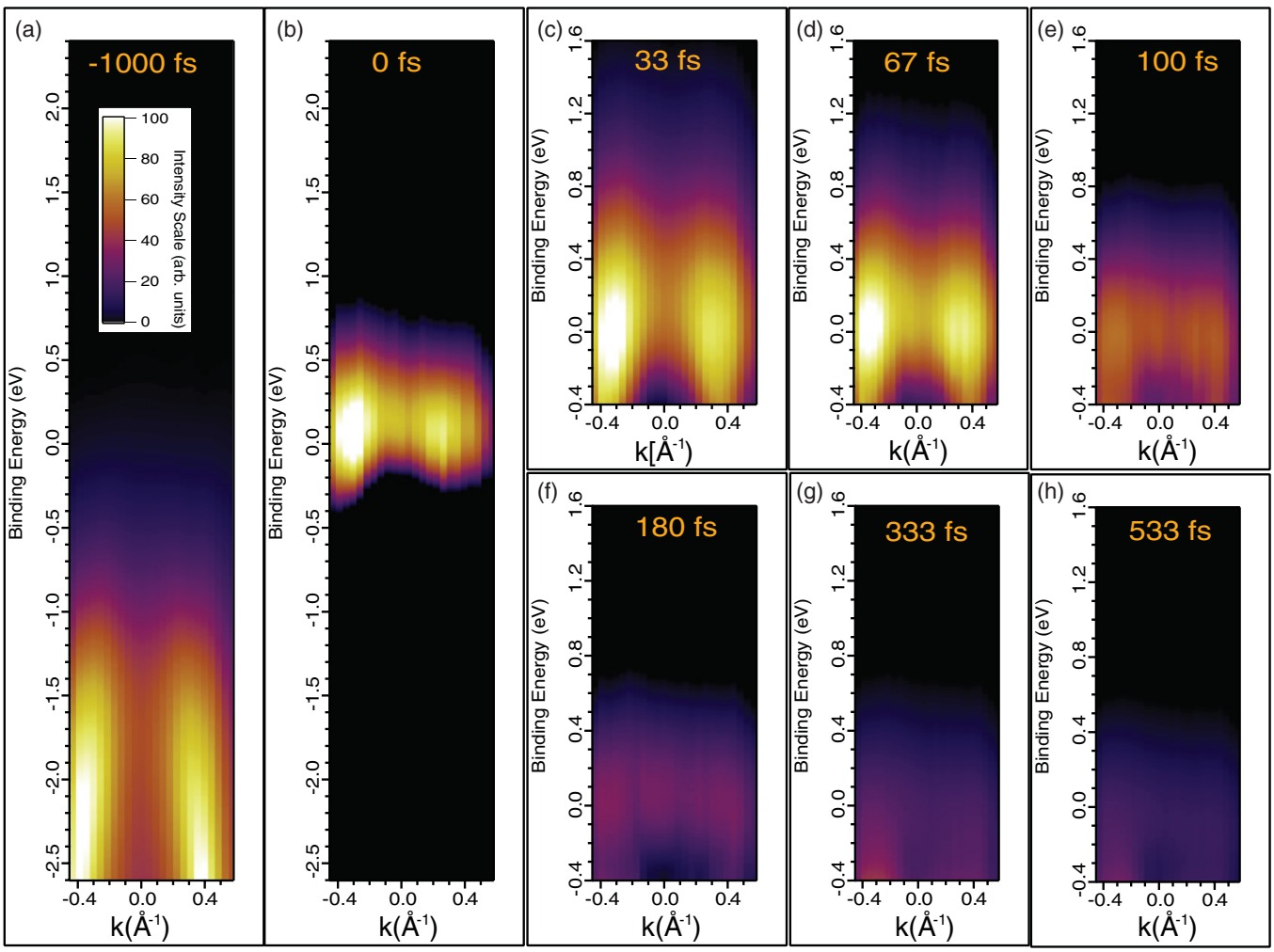

FIG. 1. (Color online) Time-resolved ARPES study of URu_2Si_2 in the hidden order state. (a) The case of a very long, "negative" 1-ps delay between probe and subsequent pump pulses. This spectrum is used to subtract a background from all other spectra collected for the probe pulse arriving after the pump pulse with delays between 0 and 533 fs, with (b) the "0-fs" case corresponding to the overlap of pump and probe pulses. The double structure seen above the Fermi level corresponds to the different, long-lived QPs, decaying in density between 0 and 533 fs, as shown in (b)–(h). The characteristic lifetime estimated from the decay in intensity as a function of delay is 213 ± 12 fs, corresponding to a QP peak width of 3 meV. Long-lived QPs are located right above the Fermi level and separated by the vector $q_{\langle 110 \rangle} = 0.56 \pm 0.08$ Å^{-1}. All pump-probed angle-resolved spectra shown here are taken below T_0 with a variable pump-probe delay.

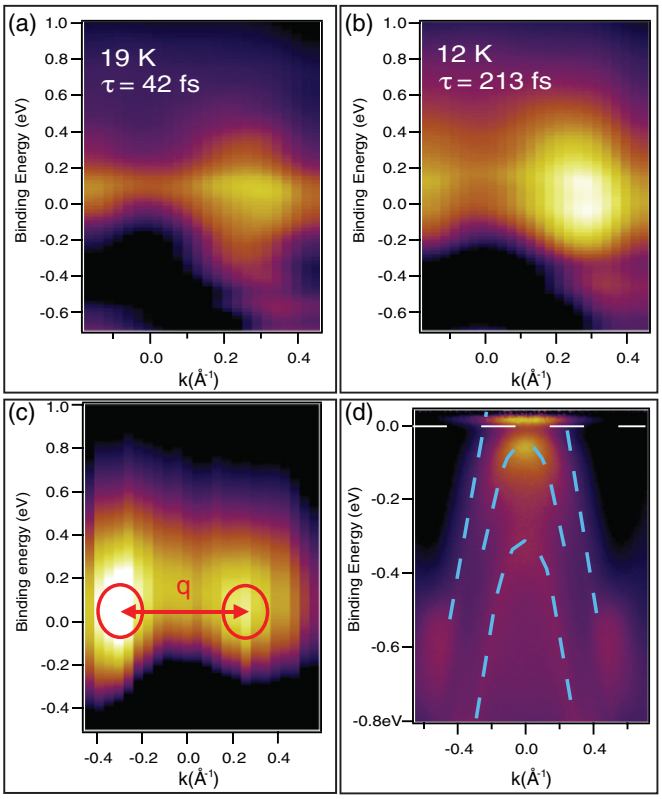

FIG. 2. (Color online) ARPES and tr-ARPES of URu_2Si_2. Time-resolved ARPES below and above T_0 is shown in (a) and (b), respectively, measured around zero delay between the pump and probe and normalized. We note the increase in QP density and lifetime upon HO transition. (c) The two QP spots as described in Fig. 1(b) are shown here, with the location of the vector $q_{\langle 110\rangle} = 0.56 \pm 0.08$ Å^{-1}. For comparison with static photoemission, the ARPES scan is shown in (d), with measured intensity divided by the Fermi function. This result provides evidence for the flat band above the Fermi level. ARPES data were collected with 34-eV photons at a position in momentum space of the bct Brillouin zone at the Z point with a sample temperature of 12 K. The dashed lines mark the location of dominant bands in the spectrum. The color intensity scale is the same as in Fig. 1.

only the part of the electronic structure being populated by the pump pulse. At delay zero [Fig. 1(b)], one can already see the very well-formed structure at the Fermi level, composed of two QP spots located at approximately $k_{HO} = \pm 0.28 \pm 0.04$ Å^{-1} and separated by a vector $q_{\langle 110\rangle} = 2k_{HO} = 0.56 \pm 0.08$ Å^{-1}. This value agrees with the STM estimate of $k_{HO} = 0.3$ Å^{-1} for the k_z-integrated case. In time, as seen in Figs. 1(c)–1(h), the QP density of states decreases, with a characteristic time constant of 213 ± 13 fs, extracted from the decay of intensity of the stronger of the two hotspots. These long-lived QPs are located around well-defined points at the upper edge of the HO gap, corresponding to the areas of highest susceptibility, where the formation of f-electron QPs drives the HO. The appearance of such localized, long-lived QPs is consistent with the partial gapping and removal of DOS from the Fermi level, and, consequently, entropy, at T_0. Observation of QPs localized in momentum space and with a dramatically increased lifetime is the central result of this Rapid Communication.

Measurements performed with tr-ARPES at 12 and at 19 K, below and above the HO transition, show a well-resolved QP structure, which exists only in the HO phase and a less pronounced QP structure existing above T_0 [see Figs. 2(a) and 2(b)]. The location of the $q_{\langle 110\rangle}$ vector is shown in Fig. 2(c). Here the density of states is only slightly elevated at 19 K, and significantly elevated at 12 K. ARPES was performed in order to establish the occupied side of the electronic band structure with high resolution [Fig. 2(d)] and indicate the existence of a narrow heavy band above the Fermi level. The lifetime measurement shown in Fig. 2 for the angle-resolved case and shown in Fig. 3 for the angle-integrated measurement on a different sample shows that the QP lifetime at 19 K is almost an order of magnitude shorter than at 12 K, indicating much broader peaks above T_0 than in the HO state. A lifetime of 213 fs is extracted only from the bright quasiparticle peak shown in Fig. 1, while a value of 301 fs in Fig. 3 is obtained from the decay of all the angle-integrated intensity. The possible enhancement of QP lifetime at T_0 was derived from transport measurements,[29] but with an unknown magnitude. We propose that the 19-K structure corresponds to the hybridization gap, which is different from HO gap. To summarize the differences, first, the hybridization gap

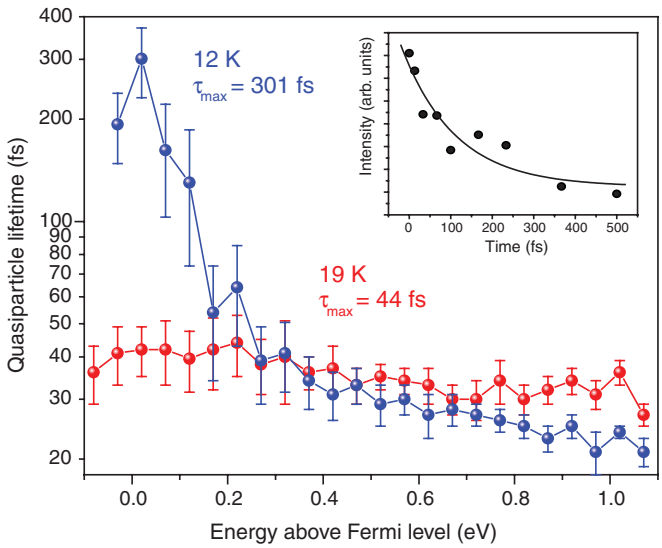

FIG. 3. (Color online) Quasiparticle lifetime. The maximum QP lifetime in and above the HO state is shown as a function of energy above the Fermi level. The maximum lifetime situated at the Fermi level corresponds to the formation of the long-lived QPs. These QPs are located at the HO "hotspots," as shown in Figs. 1 and 2, and can be identified as the principal driver and a fingerprint of the HO. We find 301 fs as the maximum QP lifetime at 12 K. This anomalous lifetime corresponds to a QP peak width of 2 meV and reflects the HO gap structure. The short-lived hybridization gap structure is seen above T_0, where the lifetime drops to a maximum value of 44 fs, with a corresponding width of 15 meV. The inset shows an example of the temporal evolution of integrated density used to create this plot; here we show the dynamics at 120 meV above the Fermi level in the HO state.

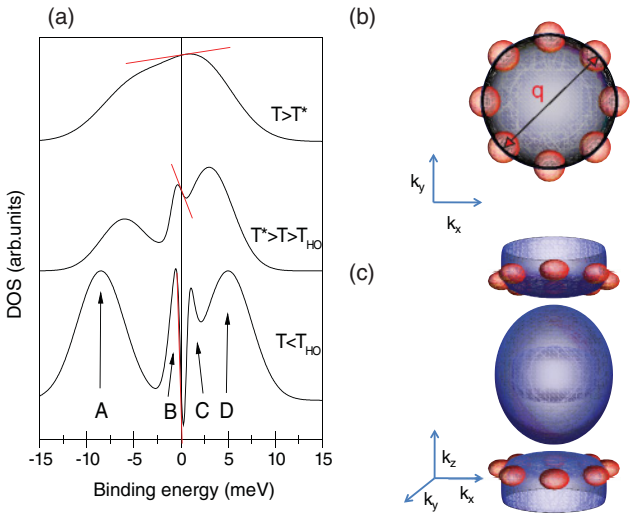

FIG. 4. (Color online) Model of the Fermi-surface renormalization in URu_2Si_2. (a) Evolution of the HO gap inside the hybridization gap. The red/gray line corresponds to the slope of the density of states extracted from thermoelectric power measurements (Ref. 30), a very sensitive probe of the integrated density of states (DOS) at the Fermi level. A Dip in DOS between B and C on the scale of a few meV is smaller than the resolution of our tr-ARPES instrument and hence we rely on transport measurements at these energies, and on time-resolved data for deduction of the gap structure. (b) In plane cut through the Fermi surface of URu_2Si_2 illustrating the location in k space of the two QP spots measured here, as indicated by vector q. Both these spots and the ones along the (100) direction are independently found by STM (Ref. 13). (c) The side view of the proposed Fermi surface in the HO state, modified after Ref. 27; the exact size of the pockets along the k_z direction remains unknown.

opens around the coherence temperature T^*, which is at least two times larger than T_0.[1–3] Next, the estimated size of the hybridization gap is roughly two times larger than the HO gap. Finally, the QPs contributing to the edges of the hybridization gap are short lived and hence much broader than the HO gap structure.

The model of Fermi-surface renormalization at T_0 based on various measurements can now be proposed as follows (Fig. 4). At high temperature $T > T^*$, the metallic system shows a full density of states around the Fermi level. Below T^* but above T_0, $T^* > T > T_0$, the hybridization gap opens in the density of states,[27] with a structure determined by two broad peaks, A and D, and some density of states at the Fermi level. The size of the hybridization gap between 10 and 20 meV is estimated from optical reflectivity and universal scaling.[20] At T_0 the HO gap opens, removing a large amount of the density of states from the Fermi level and significantly reducing entropy. The size of this gap determined by peaks B and C is between 5 and 10 meV.[13,14,16,21] Peaks C and D, essential in establishing the structure of the Fermi-surface renormalization, are evidenced by the QP lifetime change across the transition derived from the time-resolved and momentum-resolved data taken at 12 and 19 K, respectively. The momentum-resolved cut through the Fermi surface is shown in Fig. 4(b) and a full three-dimensional (3D) renormalization can be seen in Fig. 4(c).

We find that within the well-defined regions in momentum space the QP lifetime increases dramatically below T_0 in an anomalous manner not seen in other heavy fermion materials. Partial gapping of the QPs near the Fermi surface leads to suppressed recombination channels for the excited QPs. The structure of the QPs in momentum space strongly hints at momentum-dependent interactions being responsible for the HO gapping. A comparison between ARPES and magnetic torque measurements[31] in the context of rotational symmetry breaking might become possible in the future. In general, the momentum dependence observed here is not in disagreement with the recent proposal of a nematic phase in URu_2Si_2.[31] We conclude that the HO state is caused by a rapid renormalization of the FS through gapping at specific spots. Our unique tr-ARPES allows a direct, femtosecond-scale visualization of the formation of the long-lived QPs as the driver of the HO. The HO gap occurs as a momentum-dependent modification of a momentum-independent hybridization gap, and it constitutes the order parameter for the HO transition. These findings are not inconsistent with the itinerant character of the hidden order proposed by commensurate or incommensurate models,[4,9,11–17,27] and they underscore the need for alternate, advanced many-body modeling, taking into account the QP physics in the context of both T^* and T_0 energy scales.

We also sketch a model based on time-resolved ARPES data and transport measurements. Our model allows the reconciliation of apparently contradictory previous photoemission results, by acknowledging that the 21.2 eV,[21] 7 eV,[26] and our 34 eV photon energy datasets represent samplings of different parts of the 3D Fermi surface, and with very different $5f$ photoionization cross sections, ranging from 0 b at 7 eV to a few million barns at 34 eV.[32] Our observations provide an explanation for the entropy removal at T_0, and point toward the symmetry breaking through momentum-dependent scattering at $q_{\langle 110\rangle} = 0.56 \pm 0.08$ Å^{-1} as the source of second-order transition.

These results place further constraints on the possible theories of HO and will aid and stimulate the longstanding quest to uncover the exact nature of the enigmatic HO. The ability to visualize the evolution of the QP lifetime in energy-momentum space opens possibilities for heavy fermion and beyond research by investigating the complex time-dependent near-Fermi-level electronic structure via femtosecond dynamics.

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