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Evidence of a hidden-order pseudogap state in URu_2Si_2 using ultrafast optical spectroscopy

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Ultrafast optical spectroscopy was utilized to investigate carrier dynamics in the heavy-fermion compound URu_2Si_2 from 5 to 300 K. The amplitude and decay time of the photoinduced reflectivity increase in the vicinity of the coherence temperature $T^* \sim 57$ K, consistent with the presence of a hybridization gap. At 25 K, a crossover regime manifests as a new feature in the carrier dynamics saturating below the hidden-order transition temperature of 17.5 K. This is indicative of a psuedogap region (17.5 K $< T <$ 25 K) separating the normal Kondo-lattice state from the hidden-order phase. Rothwarf-Taylor modeling of the data yields values of ∼10 meV (5 meV) for the hybridization gap (hidden-order gap).

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For more than two decades URu_2Si_2 has generated significant interest due to the puzzling nature of the "hidden-order" (HO) state observed above the superconducting transition temperature ($T_c = 1.5$ K) but below $T_H = 17.5$ K.[1] It is generally agreed that a hybridization gap accompanying the formation of a Kondo lattice explains the behavior of this material from the Kondo-lattice temperature ($T^* \sim 57$ K) down to T_H. However, to date, the nature of the hidden-order parameter below 17.5 K has not been determined: the exact origin and mechanism of symmetry breaking remain unknown. Theoretical studies have suggested different scenarios for the origin of the hidden order, including charge and spin density waves, dynamic symmetry breaking, lifting of the crystal-field degeneracy, and an incommensurate hybridization wave.[2–8]

The HO state in URu_2Si_2 has been extensively studied with multiple experimental techniques, including specific-heat measurements, scanning tunneling microscopy, optical conductivity, and (time-resolved) angle-resolved photoemission spectroscopy.[9–13] These experiments have verified the development of a ∼5-meV gap near the Fermi level E_F in the HO phase. Quite recently, a detailed theoretical analysis indicates that, prior to entering the HO state (starting at ∼25 K), strong fluctuations reminiscent of a pseudogap manifest in the electronic response.[14] This suggests that theoretical models must successfully account for HO fluctuations which in turn necessitates additional experiments to better characterize the precursor region between 17.5 and 25 K.

Ultrafast optical spectroscopy (UOS) has emerged as a powerful tool for probing the electronic structure in the vicinity of E_F for a broad range of correlated-electron materials. UOS enables temporal discrimination of various quasiparticle relaxation and coupling dynamics following gentle perturbation away from equilibrium by sub-100-fs optical pulses. UOS has been shown to be extremely sensitive to the opening of a small gap (from a few meV to tens of meV) in the electronic density of states as this impedes relaxation of the excited quasiparticles. Gap-influenced dynamics have been directly observed and quantified in heavy-fermion compounds, high-temperature superconductors, and charge and spin density wave materials.[15–23]

In this Rapid Communication, we present the results of high-sensitivity optical pump-probe spectroscopy (OPPS) to study the evolution of the low-energy electronic structure of single-crystal URu_2Si_2 as a function of temperature and excitation fluence in the vicinity of the HO phase transition. The amplitude and decay time of the photoinduced (PI) reflectivity $\Delta R/R$ increase in the vicinity of T^*, consistent with the presence of temperature-dependent hybridization gap as observed in other heavy-fermion compounds.[24,25] Most importantly, at 25 K, a crossover regime manifests as a new feature in the carrier dynamics and saturates below T_H. The observed dynamics in this crossover region (17.5 K $< T <$ 25 K) are indicative of a pseudogap state separating the normal Kondo-lattice state from the hidden-order phase. Rothwarf-Taylor modeling of the data yields values of ∼10 meV (5 meV) for the hybridization gap (hidden-order gap).

The URu_2Si_2 single crystals were grown using the Czochralski technique and were processed with electrorefinement. We utilized single crystals with residual resistivity ratios of ∼400 between 2 and 298 K.[26] The samples were cleaved in the ab plane and placed in a continuous-flow optical cryostat that allows for transient reflectivity measurements $\Delta R/R$. For comparison, we have also measured nonheavy-fermion $ThRu_2Si_2$ polycrystalline samples.

Our OPPS measurements employ 50-fs laser pulses generated by a Ti:sapphire oscillator operating at 80 MHz. The pump and probe pulses are centered at 820 nm. The pump beam is modulated at 300 kHz using an acousto-optic modulator. The pump-beam fluence is varied from 0.1 to ∼2 μJ/cm^2, while the probe-beam fluence is fixed at ∼0.02 μJ/cm^2. The experiments were performed over a broad temperature range (5–300 K) encompassing the normal paramagnetic metal → Kondo lattice → hidden-order states in URu_2Si_2.

Figure 1(a) displays the photoinduced reflectivity dynamics $\Delta R/R$ as a function of time as the temperature is decreased from 50 to 5 K. Three characteristic dynamics can be clearly distinguished in the optical response. The fast decay initially observed at high temperatures begins to slow down upon approaching the Kondo lattice transition at T^*. This is consistent with other ultrafast experiments on heavy-fermion

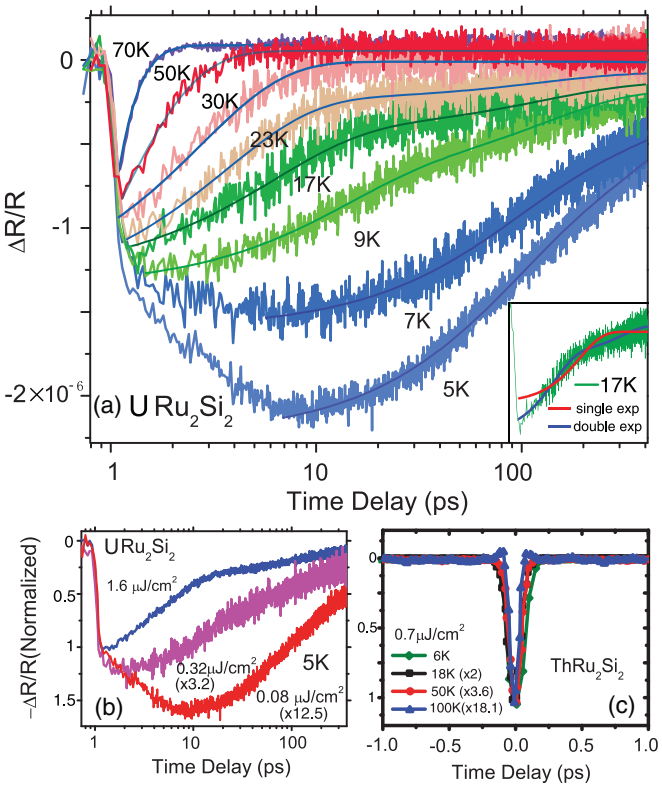

FIG. 1. (Color online) (a) Photoinduced reflectivity changes $\Delta R(t)/R$ of URu_2Si_2 as a function of time in the 5–50-K temperature range. The pump fluence for all temperatures is $\sim 0.1\ \mu J/cm^2$. The inset shows fits to the data at 17.5 K with single- and two-component exponential decay. (b) Intensity-dependent normalized reflectivity transients [$\Delta R(t)/R$ (normalized)] at 5 K. (c) Photoinduced reflectivity $\Delta R/R$ (normalized) of non-HF compound $ThRu_2Si_2$ at different temperatures. The decay time is less than 100 fs (limited by pulse width) at all temperatures; the pump fluence is $\sim 0.7\ \mu J/cm^2$.

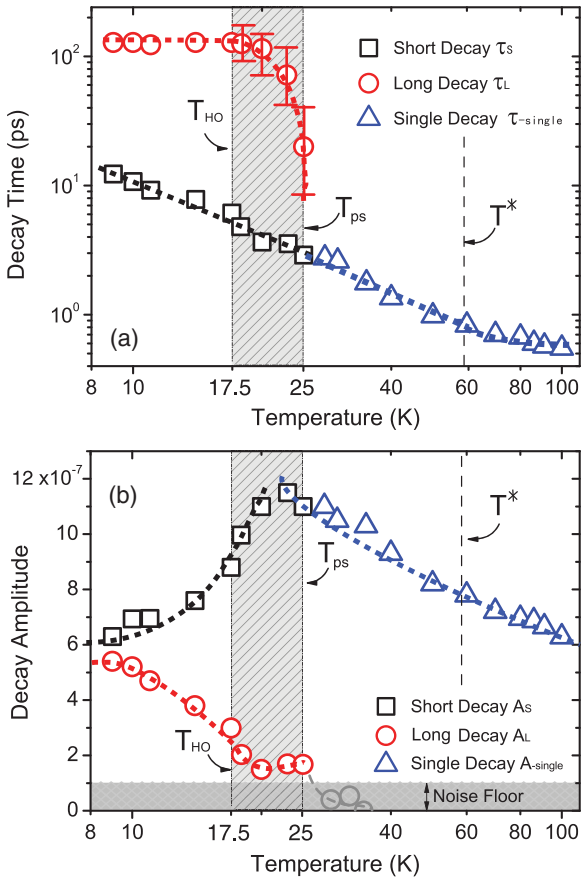

FIG. 2. (Color online) (a) Decay time τ as a function of temperature. τ_{single} is single-exponential decay time above 25 K, while τ_S and τ_L are the short and long decay times, respectively, which is evident below 25 K. (b) $\Delta R/R$ amplitude A as a function of temperature. A_{single} is single decay amplitude, while A_S and A_L are the short and long decay amplitudes, respectively. T_{HO} is the HO transition temperature, T_{PS} is the pseudogap temperature, and T^* is the Kondo temperature. The dashed lines are guides for the eyes.

(HF) compounds. A two-component decay emerges around 25 K, consisting of a fast decay and a slower decay. Finally, pronounced slow-rise-time dynamics appear in the transient reflectivity signal below ~9 K. The evolution of the decay at low temperatures is our primary concern since, as discussed below, it is this response that is indicative of a precursor or pseudogap regime upon approaching the HO state. The two-component-decay analysis is necessary and essential for the discussion of the pesudogap formation and the phase coexistence in low temperature below 25 K [see the inset in Fig. 1(a)].

As we show in Fig. 1(b), at 5 K, the dynamics of transient reflectivity of URu_2Si_2 are strongly dependent on the excitation fluence as it is varied from 0.08 to 1.8 $\mu J/cm^2$. Specific-heat analysis reveals that with 0.1-$\mu J/cm^2$ pump fluence, the crystal temperature is raised by ~1 K at low temperatures. Higher fluences will generate a phase transition (for example, from the HO state to the Kondo phase) via thermal heating. Thus at all temperatures, a pump fluence of $\sim 0.1\ \mu J/cm^2$ is used to minimize the heating effects. To further verify that the dynamics in Fig. 1(a) are related to Kondo and HO physics, we have measured $\Delta R/R$ dynamics on the the non-HF compound $ThRu_2Si_2$ at various temperatures, as shown in Fig. 1(c). There is no significant temperature or fluence dependence from room temperature down to 5 K. Therefore, the observed exotic dynamics of URu_2Si_2 result from the influence of the U $5f$ electrons on the electronic structure of this material.

Figure 2 shows the main results that derive from fitting the experimental data to multiexponential decay functions [solid lines in Fig. 1(a)]. In Fig. 2(a), we plot the temperature-dependent decay time from 100 down to 9 K. Above 25 K, the data are fitted to a single-exponential lifetime τ_{single}. At 100 K, τ_{single} is approximately 100 fs and begins to increase as the temperature approaches and then goes below T^*, reaching a value of ~3 ps at 25 K. Similar dynamics have been previously observed in other heavy-fermion metals and result from the opening of small hybridization gap in the electronic density of states (DOS) below the Kondo temperature.[27]

Upon approaching T_{HO} the relaxation dynamics no longer fit to a single-exponential decay and bifurcate to a dual exponential decay below ~25 K. This could be also inferred simply by the shape of the fits in Fig. 1(a). Specifically, the decay times τ_S and τ_L are determined by fitting the experimental curves in Fig. 1(a) with a double exponential decay:

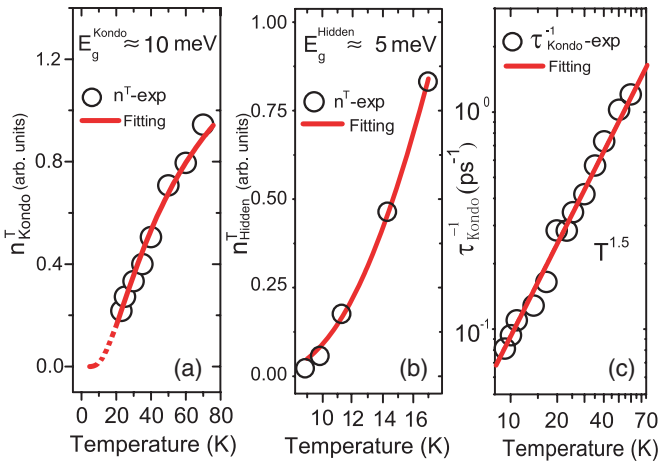

FIG. 3. (Color online) (a) RT model fitting to n^T_{Kondo} reveals a ∼10meV energy gap in the DOS of URu_2Si_2 above HO. (b) RT model fitting to n^T_{Hidden} indicates an opening of the energy gap of ∼5meV in the DOS. (c) τ^{-1}_{Kondo} versus temperature, where $\tau^{-1}_{\text{Kondo}} = 1/\tau_S$.

$\Delta R(t)/R = A_0 + A_S\exp(-t/\tau_S) + A_L\exp(-t/\tau_L)$, where τ_S and τ_L are the short and long decay times, respectively. A_S and A_L are the short and long decay amplitudes, respectively, and A_0 is a long time relaxation describing heat flow out of the excitation volume. As Fig. 2(a) shows, τ_S follows the trend of τ_{single}, while the longer decay time τ_L increases below 25 K and stabilizes at a constant value of approximately 100 ps below 17.5 K. As discussed below (Fig. 3), τ_L corresponds to the appearance of a second (smaller) gap close to E_F in the DOS of URu_2Si_2 at the HO transition temperature.

The decay amplitudes also show dramatic changes in the vicinity of T_{HO}. Figure 2(b) is a plot of the temperature-dependent decay amplitudes from 100 down to 9 K. The single decay amplitude A_{single} splits into two as the temperature approaches 25 K. A_S follows the trend of A_{single} in the range of 25 K down to 17.5 K but starts to decrease dramatically in the HO state. In contrast, A_L increases dramatically when the sample is cooled below the HO transition temperature of 17.5 K. In short, both the amplitude and lifetime dramatically evolve in the region from 17.5 K $< T <$ 25 K (shaded region in Fig. 2), which is consistent with the emergence of a HO gapped state. Above 25 K, A_L is too small to be addressed (below the noise level).

We use the Rothwarf-Taylor (RT) model to analyze the data in Figs. 1 and 2 based on its previous success and insight in describing ultrafast quasiparticle dynamics in other strongly correlated electron materials. The RT model describes how an energy gap ΔE in the quasiparticle DOS yields a bottleneck for the recombination of photoexcited quasiparticles.[28] Briefly, a quasiparticle must transfer excess energy to a high-frequency phonon (HFP) or other bosons with $h\nu > \Delta E$ in order to relax below the gap. However, the HFP can in turn excite another quasiparticle over the gap, making the overall quasiparticle relaxation dynamics controlled by the decay of HFP population. Therefore, the opening of a gap or temperature-dependent changes in an existing gap magnitude modify the lifetime and amplitude of $\Delta R/R$. In short, the RT model enables extraction of the magnitude of the Kondo-lattice hybridization and HO energy gaps.

For the RT model, the relaxation rate τ^{-1} is proportional to $n^S + n^T$, where n^S is the number density of photoexcited quasiparticles and n^T is the number density of thermally excited quasiparticles.[31] In addition, the amplitude of the transient reflectivity signal A is also related to n^S and n^T as $A \propto n^S - n^T$. Assuming the weak perturbation limit, n^T is proportional to $\mathcal{A}(T)^{-1} - 1$, where $\mathcal{A}(T) = A(T)/A(T \to 0)$.[31] We note that this analysis is applicable to both the hybridization gap (A_S, τ_S) and the HO gap (A_L, τ_L).

Following these arguments, we can attribute the increase of the single-exponential decay-time constant τ_S between 100 and 25 K to the opening of a hybridization gap as observed in other heavy-fermion materials[15,17,27]. As Fig. 3(a) shows, RT model fitting to $n^T_{\text{Kondo}} \propto \mathcal{A}_S(T)^{-1} - 1$ and $n^T_{\text{Kondo}} \propto T^{1/2}\exp(-E_g/2k_BT)$ yields a hybridization gap of approximately 10 meV. Notice that, using this analysis, the gap size is an averaged value over the three-dimensional (3D) Brillouin zone.

The slow-decay exponent [Fig. 2(a)] that emerges below $T \sim 25$ K results from the opening of another energy gap in the quasiparticle DOS. In particular, RT analysis of the decay dynamics below T_H indicates a HO gap of $\Delta E \sim 5$ meV. This can be clearly seen in Fig. 3(b), which shows a fit of n^T_{Hidden}. The results in Figs. 3(a) and 3(b) are in good agreement with other measurements.[1,10,12]

However, the behavior of A_L at temperatures between 17.5 and 25 K [Fig. 2(b)] shows that the number density of photoexcited quasiparticles (QPs) n^S_{Hidden} and thermally excited QPs n^T_{Hidden} across the HO gap has not been fully established until $T_H = 17.5$ K is reached. This indicates that the gap is not well stabilized in this narrow temperature range. This agrees with previous experimental and theoretical results.[4,8] In particular, Balatsky *et al.* recently offered a scenario where pre-transition ordering, best described as a HO pseudogap state (HOPG), would occur between 22 K (the pseudogap temperature T_{PS}) and 17.5 K.[14] In this picture, gap energy fluctuations must be taken into account. Clearly, in light of the analysis in Fig. 3, the experimental data in Fig. 2 are consistent with this interpretation.

Below T_H, the conventional Kondo-lattice effect (represented by A_S and τ_S) loses its dominance in favor of the smaller quasiparticle gap state. Indeed, our measurements demonstrate a continuous decrease of A_S and simultaneous rise of A_L starting at $\sim T_H$. This indicates the photoexcited quasiparticles n^S_{Hidden} relax toward the HO gap with decreasing temperature below 17.5 K. Also, it is interesting to see that the inverse decay time $1/\tau_S$ can be fitted by $1/\tau_S \sim T^{1.5}$ [Fig. 3(c)]. This suggests that the hybridization gap is anisotropic.[22] The preference of the HO gap at the lowest temperatures and the anisotropy of the Kondo gap merit additional investigation to better understand how the Kondo state evolves toward the HO phase.

Finally, we briefly address the onset of the slow-rise-time dynamics observed at temperatures below 9 K. It has previously been demonstrated in many heavy-fermion materials that this slow rise time is caused by an excessive HFP population at the initial stages of the relaxation process.[30,31] An initial excess of HFP in comparison to excited QPs results in continuous

excitation of QP over the energy gap. This leads to a slow rise time of $\Delta R/R$ at early delay times (typically several picoseconds). One plausible explanation to an HFP abundance was given by Kabanov *et al.*[31] In a material with gapped electronic structure in the vicinity of E_F, electrons from a photoexcited non-Fermi population preferentially transfer the excess energy to HFPs instead of other electrons. This is opposite to what normally happens in regular metals, where e-ph relaxation follows the e-e thermalization and can be described by a well-known two-temperature model (TTM).[32] Hence, immediately after excitation most of the electrons will relax toward the gap edge by emitting phonons yielding an excess of HFP. According to the RT model, these dynamics depend strongly on the excitation level at low temperatures. This behavior has also been observed in other gapped materials, including superconductors.[30] It is not surprising that at the lowest temperatures (<9 K) and low excitation levels (<0.2 $\mu J/cm^2$), the gap of a few meV strongly influences associated dynamics, which, as shown in Fig. 1(a), appear at the onset and subsequent relaxation of $\Delta R/R$.

In conclusion, we used ultrafast optical pump-probe spectroscopy to study the dynamics of quasiparticle relaxation in URu_2Si_2 over a broad temperature range. We found that (1) a well-defined 10-meV Kondo gap in the electronic DOS exists from above 60 K down to the pseudogap temperature $T_{PS} = 25$ K, (2) a pseudogap state emerges as a precursor to the HO state at 25 K and persists to 17.5 K, and (3) a 5-meV HO gap appears below 17.5 K. Our experimental data are consistent with the previous point contact and far-infrared optical conductivity measurement and are in accord with recent theoretical predictions.[14,29,33]

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