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Formation of the coherent heavy fermion liquid at the ‘hidden order’ transition in URu₂Si₂

Shouvik Chatterjee,¹ Jan Trinckauf,² Torben Hänke,² Daniel E. Shai,¹ John W. Harter,¹ Travis J. Williams,³ Graeme M. Luke,³ Kyle M. Shen,^{1,4} and Jochen Geck^{2,*}

¹Laboratory of Atomic and Solid State Physics, Department of Physics,
Cornell University, Ithaca, New York 14853, USA

²Leibniz Institute for Solid State and Materials Research IFW Dresden, Helmholtzstrasse 20, 01069 Dresden, Germany

³Dept. of Physics and Astronomy, McMaster University,
1280 Main St. West, Hamilton, Ontario, Canada L8S 4M

⁴Kavli Institute at Cornell for Nanoscale Science, Ithaca, New York 14853, USA

We present high-resolution angle-resolved photoemission spectra of the heavy-fermion superconductor URu₂Si₂. Detailed measurements as a function of both photon energy and temperature allow us to disentangle a variety of spectral features, revealing the evolution of the low-energy electronic structure across the ‘hidden order’ transition. Already above the transition our measurements directly reveal the existence of weakly dispersive states below the Fermi level that exhibit a large scattering rate, and which do not appear to shift from above to below the Fermi level, as previously reported. Upon entering the ‘hidden order’ phase these states rapidly hybridize with light conduction band states and transform into a coherent heavy fermion liquid, coincident with a dramatic drop in the scattering rate. This evolution is in stark contrast with the gradual crossover expected in Kondo lattice systems, which we attribute to the coupling of the heavy fermion states to the ‘hidden order’ parameter.

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The interactions between localized and delocalized electrons in the so-called heavy fermion materials result in fascinating and unexpected quantum phenomena that continue to challenge condensed matter researchers. One of the most prominent example is the enigmatic ‘hidden order’ (HO) state in URu₂Si₂, which is characterized by a large loss of entropy at T_{HO}=17.5 K [1, 2]. Although a multitude of theoretical scenarios have been proposed to explain the HO transition [3–9], our lack of knowledge of the complex and still debated electronic structure of URu₂Si₂ [10–16] remains the major obstacle to developing a definitive understanding of this phase. Here we disentangle the low-energy electronic structure of URu₂Si₂ by means of angle-resolved photoemission spectroscopy (ARPES) as a function of both excitation photon energy and temperature. Our findings not only provide new insights into the changes of the electronic structure at the ‘hidden order’ transition but also clarify the results of previous ARPES measurements. In particular, we demonstrate that the onset of hybridization and long range coherence of these heavy fermion states coincides precisely with T_{HO}, in contrast to a scenario where the occupation of the heavy band changes upon cooling through T_{HO}[13].

The present experiments were performed at the 1³-endstation on beamline UE112-PGM2 at the Berlin Synchrotron BESSY II, using a Gammadata R4000 analyzer with an overall energy resolution better than 7 meV and a base temperature lower than 2K. The Fermi energy was determined by measuring a polycrystalline gold film evaporated near the sample with a precision of better

than 1 meV. Single crystals of URu₂Si₂ were cleaved *in situ* at a base pressure of better than 4×10^{-11} torr, which yielded flat shiny surfaces parallel to the crystallographic *ab*-planes. None of the features reported in the following showed any dependence on samples or cleavage. The polarization of the incident photon beam was set to

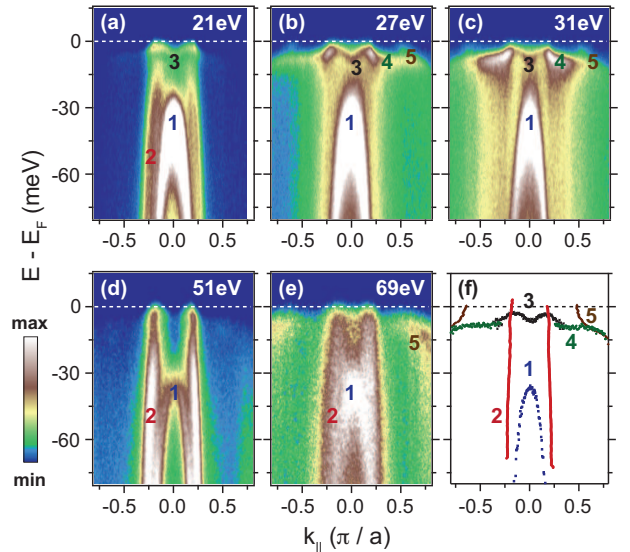


FIG. 1: (Color online) (a-e) ARPES spectra along the (0, 0) to (π , 0) direction for different excitation energies (noted in the top right of each image) measured at 2 K, deep inside the ‘hidden order’ (HO) phase. (f) Dispersions of all the different features obtained from fits to corresponding EDC/MDCs.

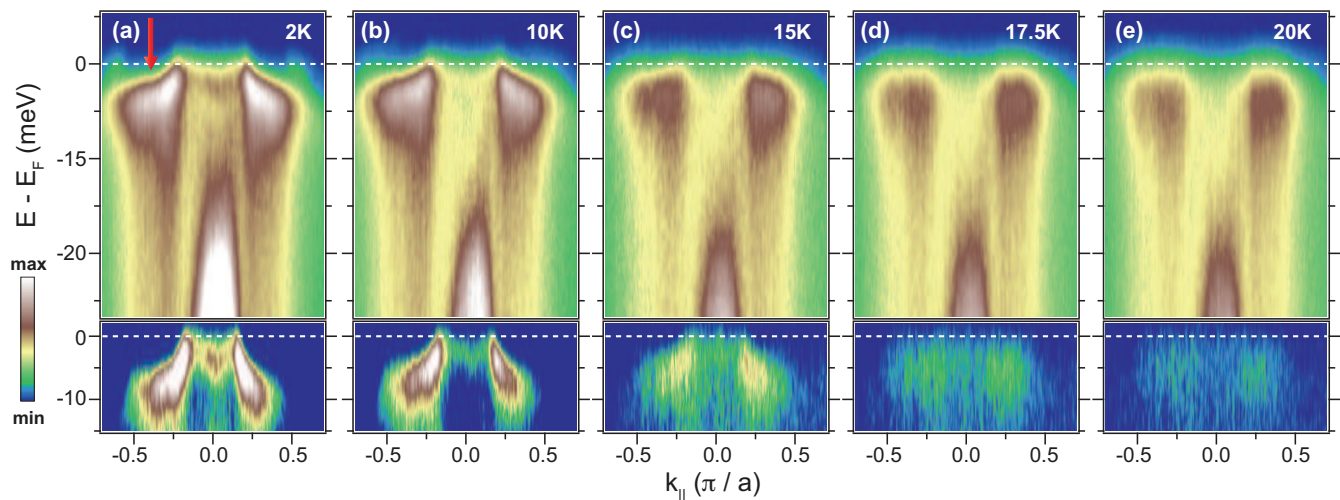


FIG. 2: (Color online) (a-e): Temperature evolution of the ARPES intensity plots of URu_2Si_2 measured along the $(0,0) - (\pi,0)$ direction at 31 eV photon energy over the temperature range 2-20 K. In the lower panel ARPES spectral maps obtained after subtracting the corresponding intensity map at 25 K is shown. Color scale has been adjusted to show only the positive part of the subtracted spectrum. Note that all the spectral maps in the lower panel are plotted keeping the range of the color scale fixed. Below T_{HO} a coherent heavy fermionic band rapidly emerges which simultaneously becomes sharper and more dispersive as the sample is cooled down. The red arrow in (a) indicates the momentum at which the EDCs shown in Fig. 3 are taken.

Linear Vertical (LV) unless mentioned otherwise.

In Fig. 1, we show ARPES spectra along the $(0,0) - (\pi,0)$ direction recorded deep within the HO phase at a variety of different photon energies. The spectra in Figs. 1 a-e, exhibit a dramatic dependence on the incident photon energy $h\nu$, revealing a multitude of electronic states near $(k_x = 0, k_y = 0)$, some of which have not been clearly delineated by previous photoemission studies. We emphasize that at no single photon energy are we able to clearly distinguish all five features, thus underscoring the importance of photon energy dependent measurements in revealing and disentangling the complete electronic structure of URu_2Si_2 .

A compilation of these different features is shown in Fig. 1 f. Feature 1 has been previously shown to be of surface origin and does not show significant changes across T_{HO} , while feature 2 corresponds to a light hole-like band which has been attributed to a bulk state [13, 14]. Feature 3 exhibits an ‘M’-shaped dispersion and was also reported for $h\nu = 7$ eV [15, 16]. It is connected, as is shown in our study, to a relatively flat band (feature 4) ostensibly of predominant $5f$ character. Finally, hole-like states (feature 5) that cross the Fermi level E_F at $k_x \approx 0.54 \pi/a$ form propeller-shaped Fermi surface (FS) sheets, also observed in quantum oscillation measurements [17, 18].

By changing the photon energy, we can probe different values of k_z along the (001) direction and can therefore determine the electronic dispersion perpendicular to the Ru_2Si_2 planes. We do not observe any appreciable dispersion along k_z for features 2, 3, and 4, while feature 1 has already been ascribed to a surface-derived origin [14]

and feature 5 is apparent at only very few photon energies. The main effect of varying photon energies observed here is to strongly modulate the photoelectron matrix elements of these different features, suggesting that these states have substantially different orbital character. This conclusion is further supported by a strong dependence on the photon polarization observed for the various features (not shown).

Here we will concentrate primarily on features 2,3, and 4, all of which undergo dramatic modifications across T_{HO} . Although we cannot distinguish whether these features correspond to quasi-two-dimensional bulk-derived states or surface-derived features based on their out-of-plane dispersion, the fact that all three features change dramatically precisely at the bulk T_{HO} clearly indicates that they are relevant to the physics of the ‘hidden order’ transition. Moreover, the absence of Feature 3 in Rh-doped samples, where the HO state is destroyed [15] further supports the notion that these states are directly tied to the HO phase and likely to be bulk-derived. In addition, we found that the features 2, 3, and 4 were significantly less sensitive to surface degradation than feature 1, the one established surface-derived feature near $(0,0)$. One of the FS sheets reported by Shubnikov-de Haas (SdH) oscillations [19] exhibits an extremal k_F similar to Feature 2, the light hole band, but these measurements also suggest that this FS sheet is closed along the (001) direction. At face value, this strong k_z dependence appears inconsistent with our data. However, this could be reconciled by the fact that our measurements are performed in the absence of a magnetic field, and that the

FS sheet observed in SdH only appears above a magnetic field of 21 T and could be a field-induced state. At present, the provenance of features 2, 3, and 4 is still not conclusive, but their strong changes upon crossing through T_{HO} clearly indicate that they are directly related to the ‘hidden order’ and the heavy-fermion physics of the bulk material.

Having identified the electronic states of interest, we now address their evolution across T_{HO} . In what follows we will refer to the states corresponding to feature 3 (‘M’ shaped band) and feature 4 (flat band) as heavy fermion states and to feature 2 as the conduction band. To investigate the temperature dependence of the heavy fermion states, we set $h\nu = 31\text{ eV}$, a photon energy at which these states can be easily tracked. As shown in Fig. 2, above T_{HO} only diffuse spectral weight is observed close to the Fermi level, indicating large scattering rates. The presence of such states is consistent with recent optical spectroscopy measurements [20], which again indicates that our ARPES results reflect bulk properties. As the temperature is lowered below T_{HO} , a well-defined heavy fermion band forms, which becomes progressively sharper

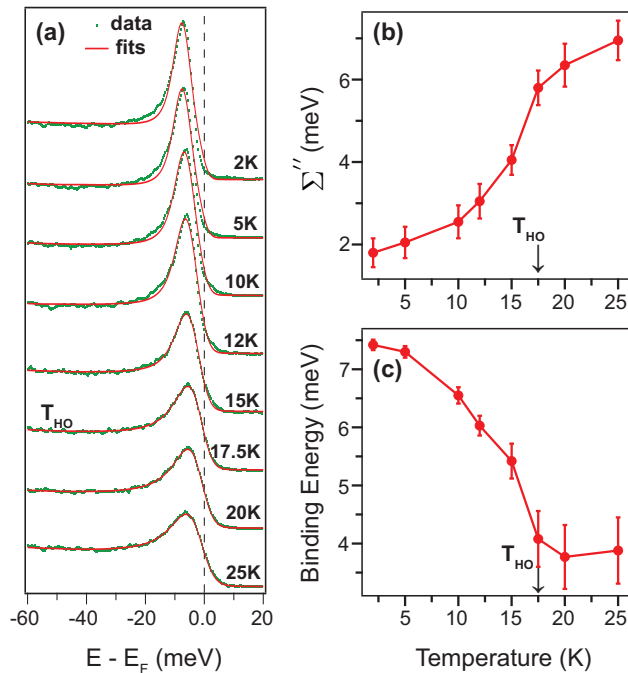


FIG. 3: (Color online) (a) Temperature dependence of the EDCs taken at the red arrow in Fig. 2 a, with corresponding fits (solid red lines). An instrumental resolution of 6 meV was used in the fits, as obtained from a gold reference. (b) Change in the imaginary part of the spectral function Σ'' . (c) Quasiparticle binding energy with temperature as extracted from the fits in a. A sharp drop in magnitude observed across T_{HO} shown in b, indicates a dramatic enhancement of the lifetime of the quasiparticles on entering the ‘hidden order’ phase.

and more dispersive upon cooling. This development is even more apparent in the lower panels of Fig. 2, where the corresponding spectrum taken at 25 K has been subtracted. In more conventional Kondo lattice systems, coherent heavy fermion bands develop only gradually below the Kondo temperature T_{K} , which is approximately 70 K for URu_2Si_2 . In contrast, we observe only incoherent states with no clear dispersion even below T_{K} , which suddenly gain coherence upon crossing T_{HO} .

To better quantify this temperature dependence, we have analyzed the energy distribution curves (EDCs) at the momentum indicated (red arrow) in Fig. 2 a. In Fig. 3 a, the data were fit to a lorentzian plus a temperature-independent Shirley background [21], multiplied by a Fermi-Dirac function and finally convolved with the instrumental resolution. As can be observed in Fig. 3 b, the scattering rate obtained from the width of the lorentzian exhibits a sharp drop precisely at T_{HO} . A similar temperature dependence has been observed in inelastic neutron scattering measurements, where the intensity of low energy spin excitations is greatly diminished upon entering the ‘hidden order’ phase [22]. Moreover, a decrease in the electronic relaxation rate upon entering the HO phase has also been reported in recent pump-probe experiments [23].

The development of the dispersion is reflected in the shift of the peak of the EDC by approximately 4 meV (Fig. 3 c), which is consistent with optical spectroscopy [24, 25], transport [26, 27] and tunneling measurements [28]. We note that this energy shift tracks the typical temperature dependence of an order parameter, supporting the notion that the observed changes in the electronic structure are directly related to the ‘hidden order’ parameter. Indeed, this suggests that the changes in the electronic density of states at the HO transition which are often referred to as the ‘hidden order’ gap are instead associated with the formation of a coherent and dispersing heavy fermion band and the onset of the hybridization with the conduction states.

We now turn to the temperature dependence of the conduction band states across T_{HO} . For this purpose we set $h\nu = 49\text{ eV}$, where the signal from the conduction band is strongly enhanced. In Figs. 4 a-b, we compare spectra measured at 2 K and 20 K, revealing very strong changes of the conduction band across T_{HO} due to the increasing hybridization with the heavy fermion states as they develop coherence. This is demonstrated clearly in Fig. 4 c, where the difference of the spectra measured at 2 K and 20 K is presented. The additional spectral weight below T_{HO} tracks exactly the dispersion of the ‘M’ band, showing that the formation of the coherent heavy fermion liquid goes hand in hand with the hybridization of the conduction band. This situation is summarized schematically in Fig. 4 e, showing how variations in the photoelectron matrix elements due to rapidly changing orbital characters can give rise to an apparent dispersion

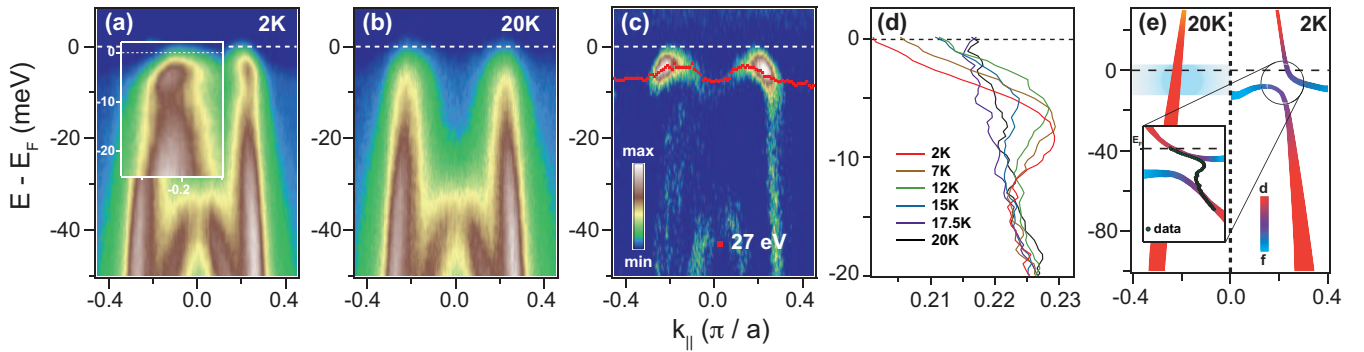


FIG. 4: (Color online) (a) Angle resolved spectra along the $(0, 0)$ to $(\pi, 0)$ direction for 49 eV photon energy below (a) and above (b) T_{HO} . Below T_{HO} , a break appears in the dispersion, shown more clearly in the inset of a. (c) Spectral map obtained after subtracting data in b from a clearly shows additional spectral weight appears below T_{HO} as the conduction band hybridizes with U 5f states. Dispersion of the ‘M’ shaped feature as observed with 27 eV excitation energy (cf. Fig. 1 b) is plotted on top of the subtracted spectrum showing the additional spectral weight follows exactly the dispersion of the ‘M’ shaped feature. (d) Temperature dependence of the MDC derived dispersion of the conduction band. The kink feature progressively gets stronger and shifts towards higher binding energy as temperature is lowered below T_{HO} . (e) A schematic illustrating the changes in the electronic structure taking place across T_{HO} . The inset shows how a kink-like feature appears in the dispersion as the bands develop mixed orbital character.

anomaly as the bands hybridize.

Although the dispersion anomaly in Fig. 4 a, resembles a kink feature, we believe it is *not* related to the coupling of the quasiparticles to a bosonic excitation. Apart from the arguments given above, there are a number of additional reasons why electron-boson coupling is unlikely to be responsible for the observed kink in the dispersion. First, the ‘kink’ energy is characteristic of the boson energy, but is shown here to be highly temperature dependent and to vanish above T_{HO} (Fig. 4 d). Second, the ratio of band velocity at higher binding energies to the velocity at E_F i.e. v_{HBE}/v_{EF} would be representative of the electron-boson coupling and mass renormalization, but the value of $v_{HBE}/v_{EF} \approx 4.0 \pm 0.2$ at 2 K would signify an unphysically large value of the coupling strength, particularly for such a soft mode.

The emergence of the ‘M’ feature observed here at $h\nu = 49, 27,$ and 21 eV agrees well with previous laser ARPES studies at $h\nu = 7$ eV [15, 16], where it was interpreted in terms of a symmetry reduction and the resulting zone folding in the HO phase. However, the spectral weight arising from zone folding is typically much weaker than the original bands, whereas we observe that at certain photon energies (e.g. 49 eV) the ‘M’ feature becomes as strong as the conduction band and the surface state. In addition, this feature coincides with the dispersion of the coherent heavy fermion band below T_{HO} . Our experiments therefore indicate that the emerging ‘M’ feature is due to the formation of the heavy fermion liquid and the onset of hybridization at T_{HO} , and not from zone folding. We note that neither in Refs. 15, 16 nor in the present study evidence was found for a band, which moves from above to below the Fermi level with cooling [13].

The onset of hybridization at T_{HO} was also observed

in recent scanning tunneling spectroscopy measurements [28, 29]. However, the ARPES data at hand yield essential new experimental insights, which were not known previously:

(i) Already above T_{HO} , “incoherent heavy fermion” states exist below the Fermi level. These states explain the observed drop in the resistivity at around 70 K and the enhanced effective mass deduced from specific heat measurements [1, 2], as those states contribute significantly to the electronic density of states close to the Fermi level. The new information provided here demonstrates that these states exhibit a large scattering rate, do not show a well-defined dispersion and do not hybridize significantly with the light conduction band above T_{HO} . Also, the consistency between ARPES and macroscopic measurements is an indication that the “incoherent heavy fermion” states are indeed a bulk feature.

(ii) Upon cooling through T_{HO} , the heavy fermion states hybridize with the conduction band and a coherent heavy fermion liquid rapidly forms. The formation of the latter therefore occurs in a thermodynamic phase transition which goes along with a dramatic reduction of the scattering rate. This evolution is in stark contrast with the gradual crossover expected for more conventional Kondo lattice systems suggesting that we are observing a new pathway to the formation of the heavy fermion state. Interestingly the onset of the HO is directly tied to the hybridization between the 5f states and the conduction band, an interaction which appears to be blocked above T_{HO} and only becomes active inside the HO phase.

The abrupt drop of the quasiparticle scattering rate at T_{HO} shows that the single particle electronic excitation spectrum is directly sensitive to fluctuations of the

‘hidden order’ parameter above T_{HO} . Our results therefore indicate an order-disorder transition, as opposed to a Stoner-type transition, where the degree of freedom that orders does not exist above T_{HO} . Hence, the fluctuations of the ‘hidden order’ parameter apparently suppress the coherence of the heavy fermion states and, at the same time, block the hybridization of the $5f$ states with the conduction band, a fingerprint that might help solve the ‘hidden order’ puzzle.

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* Author to whom correspondence should be addressed:
j.geck@ifw-dresden.de

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