Visualizing heavy fermions emerging in a quantum critical Kondo lattice

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In solids containing elements with $f$ orbitals, the interaction between $f$–electron spins and those of itinerant electrons leads to the development of low–energy fermionic excitations with a heavy effective mass. These excitations are fundamental to the appearance of unconventional superconductivity and non–Fermi–liquid behaviour observed in actinide– and lanthanide–based compounds. Here we use spectroscopic mapping with the scanning tunnelling microscope to detect the emergence of heavy excitations with lowering of temperature in a prototypical family of cerium–based heavy–fermion compounds. We demonstrate the sensitivity of the tunnelling process to the composite nature of these heavy quasiparticles, which arises from quantum entanglement of itinerant conduction and $f$ electrons. Scattering and interference of the composite quasiparticles is used to resolve their energy–momentum structure and to extract their mass enhancement, which develops with decreasing temperature. The lifetime of the emergent heavy quasiparticles reveals signatures of enhanced scattering and their spectral lineshape shows evidence of energy–temperature scaling. These findings demonstrate that proximity to a quantum critical point results in critical damping of the emergent heavy excitation of our Kondo lattice system.

A local magnetic moment occurs when a strongly interacting quantum state, such as an atomic $d$ or $f$ orbital, cannot be doubly occupied owing to strong on–site Coulomb repulsions. In the presence of a dilute concentration of such magnetic moments in a metal, spin–flip scattering of conduction electrons from these local moments results in their collective magnetic screening below a characteristic temperature called the Kondo temperature, $T_K$. In materials where local moments are arranged in a dense periodic array, the so–called Kondo lattice, the deconfinement of localized orbitals through their hybridization with the conduction electrons results in composite low–energy excitations with a heavy effective mass (Fig. 1b). Tuning the hybridization between $f$ orbitals and itinerant electrons can destabilize the heavy Fermi–liquid state towards an antiferromagnetically ordered ground state at a quantum critical point (QCP). In proximity to such a quantum phase transition—between itinerancy and localization of $f$ electrons—many heavy–fermion systems exhibit unconventional superconductivity at low temperatures (Fig. 1c).

Thermodynamic and transport studies have long provided evidence for heavy quasiparticles, their unconventional superconductivity and non–Fermi–liquid behaviour in a variety of material systems. However, the emergence of a coherent band of heavy quasiparticles near the Fermi energy in a Kondo lattice system is still not well understood. Part of the challenge has been the inability of spectroscopic measurements to probe the development of heavy quasiparticles with lowering of temperature and to characterize their properties with high energy resolution. Such precise measurements of heavy–fermion formation are not only required for understanding the nature of these electronic excitations close to quantum phase transitions, but are also critical to identifying the source of unconventional superconductivity near such transitions.

**Composite heavy–fermion excitations**

The emergence of composite heavy fermions in a Kondo lattice can be considered as a result of the hybridization of two electronic bands: one

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dispersing band due to conduction electrons and one weakly dispersing band originating from localized $f$ electrons (dashed lines in Fig. 1d). This hybridization generates low-energy quasiparticles that are a mixture of conduction electrons and $f$ electrons with a modified band structure characterized by the so-called direct $(2\pi)$ and indirect $(\pi)$ hybridization gaps, as shown in Fig. 1d. Various theoretical approaches, including several numerical studies, reproduce the generic composite band structure shown in Fig. 1d. Recent theoretical modelling has also shown that tunnelling spectroscopy can be a powerful probe of this composite nature of heavy fermions. Depending on the relative tunnelling amplitudes to the light conduction ($t_c$) or to the heavy $f$-like ($t_f$) components of the composite quasiparticles, and due to their interference, tunnelling spectroscopy can be sensitive to different features of the hybridized band structure. Figure 1d–f shows examples of model calculations (see Supplementary Information section I) illustrating the sensitivity of the spectra to predominant tunnelling to the light (Fig. 1e) or heavy (Fig. 1f) electronic states.

Recent advances in the application of scanning tunnelling microscopy (STM) to heavy fermions are providing a new approach to examining the correlated electrons in these systems with high energy and spatial resolutions. STM and point-contact experiments on heavy-fermion compounds have shown evidence for hybridization of the conduction electrons with the $f$ orbitals and have been used to probe the so-called hidden order phase transition involving heavy $f$ electrons in URu$_2$Si$_2$ (refs 29–32). Sudden onset of the hidden order phase seems to give rise to strong modification of the band structure in URu$_2$Si$_2$ as detected by STM measurements. However, these changes are correlated with the phase transition into the hidden order at 17.5 K rather than being the generic physics of heavy Fermi liquids that should appear at higher temperatures and evolve smoothly with lowering of temperature. Direct experimental observation of the gradual formation of heavy quasiparticles with decreasing temperature and evidence of their composite nature, which is ubiquitous to all heavy fermions, as well as examination of their properties in proximity to QCPs, have remained out of the reach of STM and other spectroscopic measurements.

CeMIn$_5$ as a model heavy-fermion system

To provide a controlled study of the emergence of heavy-fermion excitations within a Kondo lattice system that can be tuned close to a QCP, we carried out studies on the CeMIn$_5$ (with $M = \text{Co, Rh}$) material system. These so-called 115 compounds (the chemical formula could be written Ce$_x$M$_1$In$_x$) offer the possibility of tuning the interaction between the $f$ orbitals of Ce and the itinerant $spd$ conduction electrons using isovalent substitutions at the transition metal site within the same tetragonal crystal structure. Consequently, the ground state of this system can be tuned (in stoichiometric compounds) between antiferromagnetism, as in CeRhIn$_5$ (Néel temperature $T_N = 3.5$ K), to superconductivity, as observed in CeCoIn$_5$ (superconducting transition temperature $T_c = 2.3$ K) and CeIrIn$_5$ ($T_c = 0.4$ K). Previous studies indicate that CeCoIn$_5$ is very close to a QCP, whereas CeIrIn$_5$ can be tuned close to this transition with application of pressure. These experiments confirm that superconductivity in the 115 system emerges at low temperatures close to a QCP from heavy low-energy excitations that developed at high temperature. More specifically, transport studies show a drop in the electrical resistivity of CeCoIn$_5$ around 50 K (which has been interpreted as evidence for the development of a coherent heavy quasiparticle band) followed by a $T$-linear resistivity at lower temperature (above $T_J$) – a behaviour that has been associated with the proximity to the QCP. Quantum oscillations and thermodynamic measurements find a heavy electron effective mass ($10$–$50 m_0$, where $m_0$ is the bare electron mass) for CeCoIn$_5$, whereas in the same temperature range the $f$ electrons in CeRhIn$_5$ are effectively decoupled from the conduction electrons.

Figure 2 shows STM images of a single crystal of CeCoIn$_5$ that has been cleaved in situ in our variable temperature ultrahigh-vacuum STM. In this family of compounds, the cleaving process results in the exposure of multiple surfaces terminated with different chemical compositions. The crystal symmetry necessarily requires multiple surfaces for cleaved samples, as no two equivalent consecutive layers occur within the unit cell. Therefore breaking of any single chemical bond will result in different layer terminations on the two sides of the cleaved sample. Experiments on many cleaved samples have revealed three different surfaces, two of which are atomically ordered (termed surface A and B in Fig. 2a, b) with a periodicity of $\sim 4.6$ Å corresponding to the lattice constant of the bulk crystal structure, whereas the third surface (termed surface C, Fig. 2b) is reconstructed. Comparison of the relative heights of the sub-unit-cell steps between the different layers (Fig. 2c, d) to the crystal structure determined from scattering experiments suggests that exposed surfaces A, B and C correspond to the Ce–In, Co and In$_2$ layers, respectively. Experiments on CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ and CeRhIn$_5$ reveal similar results, where cleaving exposes the corresponding multiple layers in those compounds (see Supplementary Information section II). Hg defects in CeCoIn$_5$ at this concentration have negligible influence on its thermodynamic and transport properties and are introduced for the scattering experiments described below.

Signatures of hybridization and composite excitations

Spectroscopic measurements of CeCoIn$_5$ show the sensitivity of the tunnelling process to the composite nature of the hybridized heavy-fermion states. Tunnelling spectra on surface A (identical to the Ce–In layer) of CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ show that on cooling the sample dramatic changes develop in the spectra in an asymmetric fashion about the Fermi energy (Fig. 3a). (The same behaviour is also observed in CeCoIn$_5$ see Supplementary Information section III.)
The redistribution of the spectra observed on this surface is consistent with a tunnelling process that is dominated by coupling to the light conduction electrons and displays signatures of the direct hybridization gap structure in CeCoIn$_5$ is also centred above the chemical potential (8 meV, see Fig. 3a), which makes access difficult for angle-resolved photoemission experiments$^{43-45}$—the typical technique used for probing electronic band structure in solids.

The composite nature of the heavy-fermion excitations manifests itself by displaying different spectroscopic characteristics for tunnelling into the different atomic layers. Figure 3b shows spectra measured on surface B (identified as Co) of CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ that look very different from those measured on surface A (Fig. 3a). In the temperature range where spectra on surface A (Fig. 3a) develop a depletion of spectral weight near the Fermi energy, surface B shows a sharp enhancement of spectral weight within the same energy window (Fig. 3b). With further lowering of temperature, the enhanced tunnelling on surface B evolves into a double-peak structure. As a control experiment, measurements on the corresponding surface in CeRhIn$_5$, once again, display no sharp features in the same temperature and energy windows (Fig. 3b, dashed line). The spectroscopic features of surface B of CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ display the characteristic signatures of dominant tunnelling to the $f$ component of the heavy quasiparticles, which reside near the Fermi energy and are expected to display the indirect hybridization gap ($A_f$; see Fig. 1d, f).

Modelling the tunnelling to composite heavy excitations can reproduce our spectroscopic measurements on the two different atomically ordered surfaces of CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$. Following recent theoretical efforts$^{26,27}$, we compute spectroscopic properties of a model band structure in which a single hole-like itinerant band of spd-like electrons hybridizes with a narrow band of $f$-like electrons (see Supplementary Information section I for details of the model).

The results of our calculations (Fig. 3c, d) are sensitive to the ratio of tunnelling ($t/f$) into the heavy $f$ states to tunnelling into the light conduction band—a behaviour that explains the differences between the tunnelling processes on the cleaved surfaces (Fig. 3a, b). Although naively one would expect that tunnelling to the heavy excitations would be more pronounced on the Ce–In layer, recent first principles calculations show that the amplitude of the hybridization of the $f$ states with the out-of-plane spd electrons can be remarkably larger than the amplitude of the hybridization with the in-plane spd electrons$^{21}$.

Visualizing quasiparticle mass enhancement

To directly probe the energy–momentum structure of heavy quasiparticles in the 115 material systems, we have carried out spectroscopic mapping with the STM that enables us to visualize the scattering and interference of these quasiparticle excitations from impurities or structural defects. Elastic scattering of quasiparticles from these imperfections gives rise to standing waves in the conductance maps at wavelengths corresponding to $2\pi/q$, where $q = k_f - k_i$ is the momentum transfer between initial ($k_i$) and final ($k_f$) states at the same energy. We expect that those $q$ with the strongest intensity connect regions of high density of states on the contours of constant energy, and hence provide energy–momentum information about the quasiparticle excitations. We characterize the scattering $q$ using discrete Fourier transforms (DFTs) of STM conductance maps measured at different energies. The presence of Hg substitutions in CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ provides a sufficient number of scattering centres to enhance signal to noise ratio for such quasiparticle interference (QPI) measurements.

Figure 4a shows examples of energy-resolved STM conductance maps on surface A of CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ measured at 20 K; the maps display signatures of scattering and interference of quasiparticles from defects and step edges. These conductance maps show clear changes of the wavelength of the modulations as a function of energy. Perhaps the most noticeable are the changes around each random defect (see Supplementary Information section II for the corresponding STM image showing the location of the Hg defects). Figure 4b shows DFTs of such maps; sharp non-dispersive Bragg peaks (at the corners, $(\pm 2\pi/a,0,0)$) corresponding to the atomic lattice are seen, as well as other features (concentric square-like shapes) that rapidly disperse with energy, collapse (Fig. 4b; 0 meV) and then disappear (Fig. 4b; 9 meV) near the Fermi energy. We have carried out such measurements both at low temperatures (20 K, Fig. 4b), where the spectrum shows signatures of hybridization between conduction electrons and $f$ orbitals, and at high temperatures (70 K, Fig. 4c), where such features are considerably weakened (for example, Fig. 4c; 2 meV, 10 meV). As a control experiment, we have also carried out the same measurements on the corresponding surface of CeRhIn$_5$ (Fig. 4d), for which signatures of heavy electron behaviour are absent (for example, Fig. 3a) in the same temperature window (20 K). Although understanding details of the QPI in Fig. 4 requires detailed modelling of the band structure of the 115 compounds, the square-like patterns observed in the data correspond to scattering wavevectors that can be calculated from the identified local-density approximation (LDA) band structure$^{26}$ (see Supplementary Information section V).

We find that analysing the features of the energy-resolved DFT maps provides direct evidence for mass enhancement of quasiparticles, in unison with related signatures in the tunnelling spectra. Figure 5a and b shows line sections of the DFT maps plotted along two high-symmetry directions (the thick white lines in Fig. 4b) as a function of energy for CeCo(In$_{0.9985}$Hg$_{0.0015}$)$_5$ at 20 K, and in Fig. 5c we show their
corresponding spatially averaged spectrum. The square-like regions of enhanced quasiparticle scattering in Fig. 4b appear in the line sections of Fig. 5a, b as energy-dependent bands of scattering, which become enhanced quasiparticle scattering in Fig. 4b appear in the line sections corresponding spatially averaged spectrum. The intensity is represented on a linear scale. PSD, power spectrum density.

Signatures of quantum criticality

The ability to tunnel through the f component of the heavy quasiparticles on surface B of CeCo(In_{0.9985}Hg_{0.0015})_5 provides an opportunity to probe the lifetime of the heavy quasiparticles as a function of temperature in a system that is close to a QCP. The narrow dispersion of the f band results in a direct connection between the experimentally measured width of the peak in the density of states near the Fermi energy (Fig. 3b) and the lifetime of the heavy quasiparticles. Analysis of this width measured at different temperatures is displayed in Fig. 6a (see Supplementary Information section VI), and shows a strong temperature dependence with a finite intercept (~3.5 meV) in the limit of zero temperature. The finite width at zero temperature can be understood as a consequence of a small but finite dispersion of the f bands as well as a finite probability of tunnelling into the spd electrons (see Supplementary Information section I). However, the large linear slope in Fig. 6a (that is, larger than 3/2 k_B T, where k_B is Boltzmann’s constant and T is temperature) indicates that the lifetime of the f electrons, as opposed to thermal broadening, is strongly influencing the spectra and its temperature dependence. Consistent with this observation, we also find that to capture the temperature evolution of the spectra in Fig. 3b, we have to use rather large values of scattering rate (or inverse lifetime) for the heavy quasiparticles, as opposed to thermal broadening, which is temperature independent.

A T-linear scattering rate (or inverse lifetime) for the heavy quasiparticles is consistent with the expectation that CeCoIn_5 is close to a QCP, because for systems tuned close to such transitions, temperature is the only relevant energy scale available to determine the quasiparticle lifetime, resulting in h/τ_f ~ k_B T (refs 18, 47). However, a more precise signature of a QCP would be the observation...
of energy–temperature scaling of experimental quantities near such transitions. In fact, recent theoretical work suggests that the instability of the Fermi surface near a QCP should result in scaling properties of the single-particle excitation that can be directly probed in measurements of the tunnelling density of states. To test this hypothesis, we examine the lineshape of the tunnelling spectra on surface B of CeCo(In0.9985Hg0.0015)5 near the chemical potential at different temperatures, and attempt to scale the data (Fig. 3b) by plotting \((d^2V/dE)^2(k_BT)^\alpha\) as a function of \((E/(k_BT))\beta\). (Here \((d^2V/dE)\) is the background-subtracted spectra of Fig. 3b (see Supplementary Information section VI) and \(E\) is the energy of the tunnelling quasiparticles.) We find that using the exponents \(\alpha = 0.53\) and \(\beta = 1\) results in a collapse of the data at different temperatures on a single curve in the low bias region (see Fig. 6b and Supplementary Information section VII). Although an understanding of the value of the exponent \(\alpha\) is currently lacking, the linear power \(\beta\) confirms our hypothesis of energy–temperature scaling associated with proximity to a QCP. These results indicate that the heavy quasiparticles in CeCoIn5 are damped because of critical fluctuations rather than the typical scattering that is expected in a Fermi liquid (\(T^2\) dependence). Similar energy–temperature scalings, with anomalous exponents \(\alpha\), have been previously observed in the dynamical spin susceptibility of other heavy-fermion systems near QCPs. However, here we show for the first time that the signatures of scaling and critical phenomena appear in the spectroscopic properties of the quasiparticle excitations.

**Conclusion and outlook**

The experimental results and the model calculations presented here provide a comprehensive picture of how heavy-fermion excitations in the 115 Ce-based Kondo lattice systems emerge with lowering of temperature or as a result of chemical tuning of the interaction between the \(f\) electrons and the conduction electrons. The changes in the scattering properties of the quasiparticles directly signal the flattening of their energy–momentum structure and the emergence of heavy quasiparticles near the Fermi energy. Such changes are also consistent with the predicted evolution from a small to a large Fermi surface as the localized \(f\) electrons hybridize with the conduction electrons. The sensitivity of the tunnelling to the surface termination and the successful modelling of these data provide direct spectroscopic evidence of the composite nature of heavy fermions and offer a unique method to disentangle their components.

Our experiments also demonstrate that the emergent heavy quasiparticles in our system are strongly scattered and show signatures of scaling associated with critical damping of excitations in proximity to a QCP. Like many other heavy-fermion systems, thermodynamic and transport studies of the 115 systems have shown evidence of quantum criticality, but such signatures have not been previously isolated in an electron spectroscopy measurement, as described here. Such spectroscopic signatures are direct evidence for the breakdown of coherent fermionic excitations approaching a QCP. Future extension of our measurements to lower temperatures could probe the interplay between quantum fluctuations and the appearance of superconductivity, an issue which continues to be one of the most debated in condensed matter physics.

**METHODS SUMMARY**

The single crystals of CeCoIn5, CeCo(In0.9985Hg0.0015)5 and CeRhIn5 used for this study were grown from excess indium at Los Alamos National Laboratory. Small, flat crystals were oriented along the crystallographic axes and cut into sizes suitable for STM measurements (~2 × 2 × 0.2 mm²). The samples were cleaved on a surface perpendicular to the \(c\) axis in ultrahigh vacuum (UHV) and transferred in situ to the microscope head. Differential conductance \((d^2V/dE)\) measurements were performed using standard lock-in techniques. Approximately 10 different samples of CeCoIn5, CeCo(In0.9985Hg0.0015)5 and CeRhIn5 were successfully cleaved and studied, and the spectroscopic data collected were reproducible on the corresponding identical exposed surfaces of the different samples. Spectra measured at different locations on each surface showed negligible variations. The spectra presented here (in the main paper) are averaged over approximately 200 individual spectra measured over an area of at least 100 Å × 100 Å. The spectroscopic lineshapes showed negligible variations as the tip height was varied (variation of the tunnelling current by two orders of magnitude).

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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