Observation of the Hybridization Gap and Fano Resonance in the Kondo Lattice URu_2Si_2

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The nature of the second-order phase transition that occurs in URu_2Si_2 at 17.5 K remains puzzling despite intensive research. A key question emerging in the field is whether a hybridization gap between the renormalized bands can be identified as the "hidden" order parameter. We report on the measurement of a hybridization gap in URu_2Si_2 employing a spectroscopic technique based on quasiparticle scattering. The differential conductance exhibits an asymmetric double-peak structure, a clear signature for a Fano resonance in a Kondo lattice. The hybridization gap opens well above 17.5 K, indicating that it is not the hidden order parameter. Our results put stringent constraints on the origin of the hidden order transition in URu_2Si_2 and demonstrate that quasiparticle scattering spectroscopy can probe the band renormalizations in a Kondo lattice via detection of a novel type of Fano resonance.

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The $5f$ orbital-based heavy electron system URu_2Si_2 has long puzzled researchers due to its enigmatic transition at $T_{HO} = 17.5$ K into the hidden order (HO) [1–4]. Despite gaplike behaviors [1–3,5], the exact order parameter remains unknown [4,6–9]. Static antiferromagnetism [1–3] is ruled out, because the magnetic moment is too small to account for the large entropy loss [10] and has been shown to be extrinsic [10]. Under pressure, the HO undergoes a first-order transition into an antiferromagnetic (AFM) state [10–12] and can be resurrected by a magnetic field [13]. Inelastic neutron scattering has established two magnetic excitations [14–17]: $\mathbf{Q}_0 = (1, 0, 0)$, $E_0 = 1.7$–2 meV and $\mathbf{Q}_1 = (1 \pm 0.4, 0, 0)$, $E_1 = 4$–5.7 meV. It has become evident that identifying the origin of the $\mathbf{Q}_0$ resonance, a unique feature of the HO, is critical [16]. Differentiating the consequences of the HO transition from its origin is also crucial, as demonstrated here.

Quasiparticle (QP) probes measuring tunneling and scattering conductance can provide direct electronic structure information. Recent investigations for a Kondo lattice, experimental [18–22] and theoretical [8,23–27], have brought new perspectives on the HO problem. A key question is whether a hybridization gap between the renormalized bands can be identified as the HO parameter [8]. In this Letter, we report spectroscopic measurements of a hybridization gap in URu_2Si_2 using quasiparticle scattering spectroscopy (QPS) or point-contact spectroscopy [19,28]. Our conductance spectra clearly exhibit characteristic features for a Fano resonance in a Kondo lattice. Analysis based on a recent theory [23] allows us to extract a hybridization gap: This gap opens well above T_{HO}, indicating that it is not the HO parameter.

Tunneling in a single Kondo adatom has been extensively investigated by using a scanning tunneling microscope (STM) [29,30] and well accounted for by the generic Fano resonance [31] formula $[dI/dV]_{KI} \propto (q_F + E')^2/(1 + E'^2)$, where $E' \equiv (eV - \varepsilon_0)/(W/2)$ with ε_0 and W being the resonance energy and full width at half maximum, respectively. As demonstrated in Fig. 1(a), the Fano factor $q_F \equiv A/B$ (A, tunneling probability into a localized orbital and B, into the conduction band) is a key parameter governing the conductance shape. According to the Kondo lattice model, the fate of localized moments is determined by the competition between the Kondo coupling and the Ruderman-Kittel-Kasuya-Yosida interaction [32]. Fermi surface (FS) topology plays important roles not only in itinerant magnetism such as a spin-density wave induced by FS nesting [33] but also in mediating the local magnetic interaction [34]. The periodic Anderson model, in a mean-field approximation considering on-site Coulomb interaction, gives two renormalized hybridized bands [35]: $E_{k\pm} = \frac{1}{2}\{\varepsilon_k + \lambda \pm \sqrt{(\varepsilon_k - \lambda)^2 + 4V^2}\}$. Here, λ is the renormalized f level and $V = z^{1/2}V_0$ is the renormalized hybridization matrix amplitude with $z = 1 - n_f$ (n_f, f-level occupancy). As shown in Figs. 1(b) and 1(c), a hybridization gap opens: a direct gap of $2V$ in $\boldsymbol{k}$ space and an indirect gap in the density of states (DOS) given as $\Delta_{hyb} = 2V^2/D$ ($2D$, conduction bandwidth). Based on this hybridization picture plus cotunneling, the differential tunneling conductance in a Kondo lattice was derived [23]:

$$[dI/dV]_{FR} \propto \mathrm{Im}\tilde{G}^{KL}_{\psi}(eV); \tilde{G}^{KL}_{\psi}(eV) = \left(1 + \frac{q_F W}{eV - \lambda}\right)^2 \ln\left[\frac{eV + D_1 - \frac{V^2}{eV-\lambda}}{eV - D_2 - \frac{V^2}{eV-\lambda}}\right] + \frac{2D/t_c^2}{eV - \lambda},$$

where $-D_1$ and D_2 are the lower and upper conduction band edges, respectively. $q_F = t_f V/t_c W$, where t_f and t_c are the tunneling matrix amplitudes for the f orbital

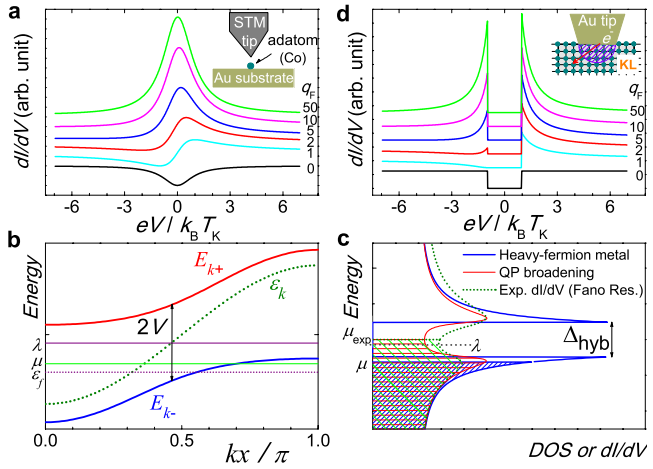

FIG. 1 (color online). (a) Single-impurity Fano resonance. T_K, Kondo temperature. (b) Hybridization between a conduction band (ε_k) and localized states (ε_f) (see the text). μ is the chemical potential. (c) DOS for the renormalized heavy bands (thick line), DOS broadened due to correlation effects (dotted line), and dI/dV (thin line) simulating our data at $T < T_{HO}$. (d) Fano resonance in a Kondo lattice. T_K, characteristic temperature for the Kondo lattice. Inset: Schematic for QPS.

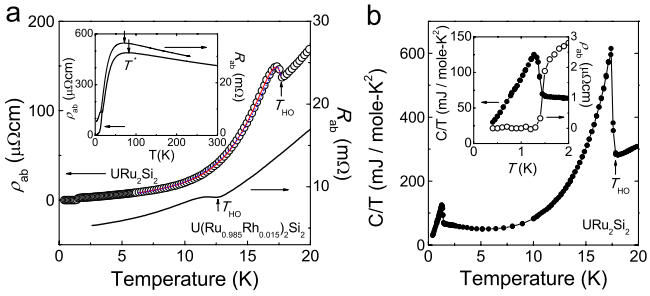

FIG. 2 (color online). (a) ab-plane resistivity (resistance) for URu_2Si_2 ($U(Ru_{0.985}Rh_{0.015})_2Si_2$). T^* for resistance maximum is 82 and 70 K, respectively. Note that the ratio $R_{300\,K}/R_{T\to 0\,K}$ is large: 248 for URu_2Si_2 and 5.8 for $U(Ru_{0.985}Rh_{0.015})_2Si_2$. T_{HO}, taken for minimum in dR/dT, is 17.56 and 12.8 K, respectively. The lines are best fits with gapped AFM excitations: ρ_{AFM1} (solid red line, $\Delta = 4.9$ meV) and ρ_{AFM2} (dotted blue line, $\Delta = 5.1$ meV) (see the text). (b) Specific heat divided by temperature for URu_2Si_2.

and the conduction band, respectively [23]. As shown in Fig. 1(d), for an intermediate q_F, an *asymmetric double-peak structure* is notable: the hallmark for a Kondo lattice, distinct from the single-impurity case.

Single crystalline URu_2Si_2 and $U(Ru_{0.985}Rh_{0.015})_2Si_2$ are grown by the Czochralski method and oriented by using a back-Laue CCD camera. The ab-plane resistivity and the specific heat of Fig. 2 show that our crystals exhibit distinct bulk HO and superconducting transitions. As-grown or cleaved crystals with mirrorlike surfaces normal to the c axis are used in QPS. Ballistic metallic junctions are formed at low temperature by using an electrochemically polished gold tip and differential micrometer [18,19]. They are formed on different spots *in situ* as resistance and pressure are controlled. Differential conductance is measured with a four-probe lock-in technique.

Figures 3(a) and 3(b) display a series of conductance curves for URu_2Si_2 and $U(Ru_{0.985}Rh_{0.015})_2Si_2$. A systematic evolution in the shape is clearly noticeable with a distinct double-peak structure appearing in curves 3–5, from which we conjecture on two parallel channels, one dominating the background and the other the asymmetric double-peak structure. We focus on the latter, leaving the background shape for future investigation. Andreev scattering [18,19] is ruled out since $T \gg T_c$. So is an AFM gap [36,37] (see also references in [38]) excluded [10]. To elucidate its origin, we further note that the positive-bias peak is always stronger and the conductance minimum occurs at a slightly negative bias at $T \ll T_{HO}$, supporting the Fano resonance origin [Fig. 1(d) [23] and Sec. S1 in [38]].

For a quantitative analysis, we start by considering strongly energy-dependent QP scattering into the renormalized heavy bands: The larger the DOS, the higher the transition rate. QPs passing through two channels, the heavy and the conduction band, interfere to produce a Fano resonance. In recent STM studies on Kondo adatoms [39], a single-impurity Fano resonance was observed in the metallic contact regime as well as in the tunneling regime, indicating that a similar quantum interference occurs in both regimes. Therefore, we conjecture that the aforedescribed Kondo lattice tunneling theory [23] can account for the characteristic features in our QPS data. The same Fano physics manifests in both QP tunneling and scattering with the conductance shape dictated by the universal parameter q_F. Thus, our model formula is $G(V) \equiv dI/dV = [dI/dV]_{FR} + \omega \cdot [dI/dV]_{bg}$, where the first term is Fano conductance and the second term accounts for the background shape with ω as a weighting factor. Figures 3(c)–3(f) show typical data for URu_2Si_2 and best fits obtained with a parabolic background and an energy-dependent QP broadening parameter [26]. Our model captures major conductance features accurately (Sec. S2 in [38]). The hybridization gap is extracted from the fitting parameters using the relation $\Delta_{hyb} = 2V^2/D$. It ranges from 11 to 14 meV with an average of 13 meV. The renormalized hybridization strength $V = 39$–45 meV and the Fano parameter $q_F = 9$–13. These values are reproducibly observed in many more conductance curves [38]. For $U(Ru_{0.985}Rh_{0.015})_2Si_2$ with $T_{HO} = 12.8$ K [Fig. 2(a)], $\Delta_{hyb} \approx 10$ meV, implying some correlation (proportionality) with T_{HO}.

The relation between the HO transition and the hybridization process [8] is addressed in Fig. 4(a) (Sec. S2 in [38]). The split peaks persist across T_{HO}, disappearing at a much higher temperature. As plotted in Fig. 4(b), the hybridization gap reproducibly opens at $T_{hyb} \sim 27$ K (Sec. S3 in [38]), well above T_{HO}, establishing that the gap opening well precedes the HO transition. Of the published QPS data, we note that the sharper the

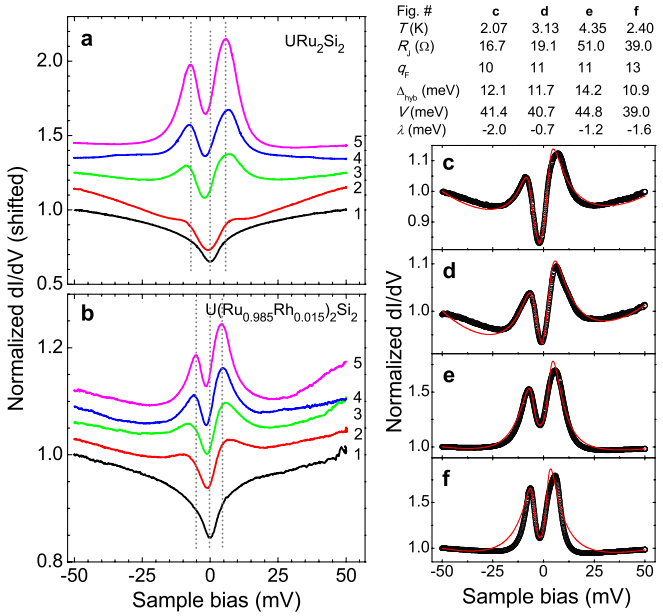

FIG. 3 (color online). (a),(b) Differential conductance (normalized by dI/dV at -50 mV) curves for junctions along the c axis of URu_2Si_2 and $U(Ru_{0.985}Rh_{0.015})_2Si_2$, respectively. Curves are shifted vertically. Dotted lines are a guide to the eye. (a) The measurement temperature (the differential junction resistance R_J at -50 mV) is 3.49 (12.3), 3.51 (18.7), 2.07 (16.7), 4.41 (55.6), and 4.35 K (51.0 Ω) for the curves from 1 to 5, respectively. (b) The measurement temperature is 4.34 K for all junctions, and R_J is 19.5, 25.0, 23.5, 20.4, and 19.7Ω for the curves from 1 to 5, respectively. (c)–(f) Typical conductance spectra for URu_2Si_2 and best fit curves with parameters shown.

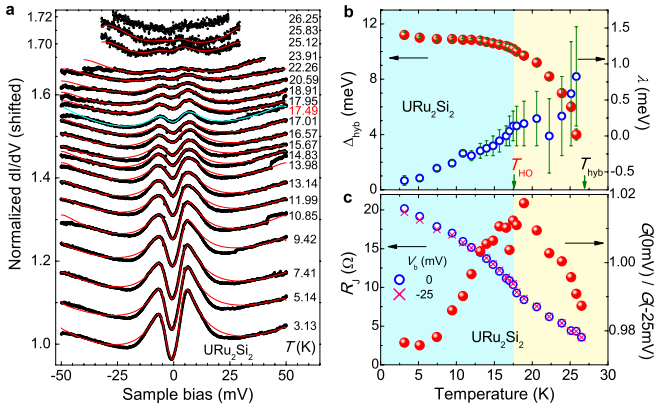

FIG. 4 (color online). (a) Temperature-dependent conductance (circles, normalized by dI/dV at -30 mV) and fit curves (lines). The R_J at the lowest temperature is 19.1Ω. The top three curves are plotted on an expanded vertical scale. (b) The hybridization gap (solid circles) Δ_{hyb}, opening at $T_{hyb} \sim 27$ K $\gg T_{HO}$ and the renormalized f level λ (right axis, open circles). (c) R_J at zero bias (open circles) and at -25 mV (crosses) and the NZBC (right axis, solid circles).

low-temperature gap structure is, the higher the gap opening temperature is observed (Sec. S3 in [38]). The renormalized f level λ appears to cross the chemical potential μ at $T \approx T_{HO}$ [Fig. 4(b)], reminiscent of a recent angle-resolved photoemission spectroscopy study [40]. If broadened, the hybridization-gap peaks can be merged into a single Kondo resonance peak [41]. By considering $\varepsilon_0 = W/2\tan[(1-n_f)\pi/2]$, the sign change in λ (ε_0) may signify an f-level occupancy change accompanying the HO transition. The normalized zero-bias conductance (NZBC) reveals a broad maximum around T_{HO} [37], as plotted in Fig. 4(c). A hallmark of a QPS junction being in the thermal (nonspectroscopic) regime is that $G(V)$ [also, ZBC(T)] strongly resembles the bulk conductivity [19,28,38]. That our data do not exhibit such a behavior indicates that the junctions are well within the spectroscopic limit [19,28,38]. To account for our NZBC, first note that it would be proportional to the DOS at μ for tunneling into the heavy band only and, thus, to the electronic specific heat coefficient (C_e/T) and effective mass. Indeed, C_e/T is found to show qualitatively similar temperature dependence [42]. A large contribution from the heavy band (large $q_F \sim 10$) as well as the ballistic nature enables us to observe such a behavior (Sec. S4 in [38]).

Our earlier QPS studies on $CeCoIn_5$ [18–20] have shown a single-impurity-like Fano line shape, contrary to URu_2Si_2. Considering $\Delta_{hyb} = 2V^2/D$ and $V = V_0(1-n_f)^{1/2}$, we conjecture that this discrepancy may arise from their different distances from the Kondo regime ($n_f \sim 1$) [43]: Δ_{hyb} would become larger away from it ($n_f < 1$), rendering the peaks less susceptible to merging. This agrees with $CeCoIn_5$ being considered closer to the Kondo limit than URu_2Si_2 [44]. The distinct double-peak structure seen in our data implies that the broadening effect, suggested to arise from intrinsic correlation [26], lattice disorder [23], and broken translation invariance [26], is not dominant in URu_2Si_2. In recent STM studies on URu_2Si_2 [21,22], a single-impurity Fano line shape is observed with $q_F < 2$, implying that the tunneling probability into the heavy band is much lower than in our QPS, which can account for the line shape (Sec. S4 in [38]). Other disparate STM observations are [21,22] (i) a gap opening at 16–17 K, (ii) a gap size of ~8 meV, and (iii) fine structures at low bias and temperature. To account for these discrepancies, one may consider surface effects, i.e., possible modifications in the hybridization [45] due to reduced near-neighbor coordination [30]. QPS [Fig. 1(d), inset] in the ballistic regime [19,28] probes scattering over the electronic mean free path, well beyond the surface. Thus, QPS is more likely to detect the bulk hybridized bands, as manifested by higher T_{hyb} and robust double-peak structure as predicted [23]. A recent optical spectroscopy, known as a bulk probe, has reported similar Δ_{hyb} and T_{hyb} values to ours [46].

We now address the widely varying gap values in other measurements [1–3,5] by focusing on resistivity (Sec. S5 in [38]). Despite no evidence for static magnetism, resistivity is frequently analyzed by considering scattering off gapped magnetic excitations (ρ_m): $\rho = \rho_0 + AT^2 + \rho_m$.

Furthermore, nearly all reports have adopted a formula for ferromagnetic (FM) excitations [47], $\rho_{\mathrm{FM}} = BT\Delta[1 + 2T/\Delta]e^{-\Delta/T}$, despite the close proximity to an AFM order, and ρ_{AFM} takes a quite different form due to linear, not quadratic, dispersion. Two known approximate formulas are $\rho_{\mathrm{AFM1}} = B\Delta^5[(T/\Delta)^5/5 + (T/\Delta)^4 + 5/3(T/\Delta)^3]e^{-\Delta/T}$ [48] and $\rho_{\mathrm{AFM2}} = B\Delta^2\sqrt{T/\Delta}[1 + 2/3(T/\Delta) + 2/15(T/\Delta)^2]e^{-\Delta/T}$ [49]. By adopting a generic T-dependent $\Delta(T) = \Delta_0 \tanh[\alpha\sqrt{T_{\mathrm{HO}}/T - 1}]$, the formulas based on FM excitations [47] and FS gapping [17] give diverging fits as $T \rightarrow T_{\mathrm{HO}}$, whereas the two AFM formulas produce reasonably good fits including the transition region, with $\Delta_0 \sim 5$ meV and $\alpha = 1.7$, as shown in Fig. 2(a) [50]. Our analysis extended to other published data [2,51,52] shows best fits with nearly the same $\Delta_0 \sim 4.7$ meV and $\alpha = 1.7$ for ρ_{ab} [2,52] (for ρ_c [2,51,52], $\Delta_0 \sim 3.3$ meV and $\alpha = 1.7$). Note that the inelastic neutron scattering resonance energy $E_1(T)$ [14] can be described well with these parameters, suggesting $\Delta_{ab} \sim E_1$. This association is likely, since a recent band calculation [7] identifies the $\mathbf{Q}_1$ resonance [15] as due to FS nesting and the gapped resistive behavior persists into the AFM phase [17]. Δ_{ab} decreases very little with increasing H when $H||I||ab$ [51], while both T_{HO} and Δ_c decrease but E_0 increases when $H||I||c$ [53,54] (cf. our Δ_{hyb} remains constant up to 4 T, Sec. S6 in [38]). This indicates that Δ_c (and Δ_{ab}) is not of the same origin as for the $\mathbf{Q}_0$ resonance associated with HO. Therefore, the resistive gaps are likely to be magnetic in nature and unlikely the HO gap (Sec. S5 in [38]). Our NZBC behavior in Fig. 4(c) may indicate the effect of magnetic excitations indirectly via interaction with charge carriers [55]. A second harmonic measurement with a current along the a or b axis (i.e., $||\mathbf{Q}_1$) at low T is planned.

The hybridization gap being distinct from the HO parameter is consistent with the general concept that the gradual hybridization process is unlikely to cause a phase transition. Following a generic argument, T_{hyb} may be the heavy-fermion coherence temperature, i.e., $T_{\mathrm{hyb}} = T_{\mathrm{coh}}$, distince from T^*, the resistivity peak temperature. For URu_2Si_2 [Fig. 2(a)], $T^* = 70$–80 K $\gg T_{\mathrm{hyb}}$. T^* may signify only a crossover in the dominant transport scattering channel, whereas T_{coh} is indicative of fully developed coherence among the renormalized Bloch states. Interestingly, our T_{hyb} is close to the temperature for the Fermi liquid behavior ($\propto T^2$) [56], supporting this speculation, but the nature of emergent heavy fermions in URu_2Si_2 is a topic of continued debate [52,56]. Additionally, the difference between T_{hyb} and T_{HO} is so large that the fluctuating HO scenario [57] may not account for our results.

We now discuss crucial elements to resolving the HO problem. Pressure and magnetic field play quite different roles in URu_2Si_2: Pressure induces AFM order, but magnetic field resurrects HO [13]. While both phases exhibit the $\mathbf{Q}_1$ resonance, the $\mathbf{Q}_0$ resonance is unique to the HO, albeit the AFM ordering occurs at the same wave vector [16]. Clearly, this points to the crucial roles played by the $\mathbf{Q}_0$ resonance. Our above analysis suggests that the $\mathbf{Q}_1$ resonance may cause the gapped resistive behavior but does not affect the conductance dramatically. Thus, we conjecture that the HO, which does not originate from itinerant bands, induces the FS nesting. However, no multipolar orders predicted to arise from localized f electrons have been detected. Even though crystal field effects are not established, they well deserve a revisit [6,9] to determine what crucial roles are played by the local degrees of freedom for (i) strong uniaxial magnetic anisotropy as observed by neutron scattering [58] and magnetic susceptibility [1] and (ii) the interplay of pressure and magnetic field in tuning the crystal field f levels and the intersite interaction [13].

In conclusion, our QPS on URu_2Si_2 unambiguously detects a novel Fano resonance as predicted for a Kondo lattice and probes the hybridization gap in the renormalized heavy bands. This gap opens at $T_{\mathrm{hyb}} \sim 27$ K $\gg T_{\mathrm{HO}}$, indicating that it is not the HO parameter. Our analysis of the gapped resistivity behavior suggests gapped magnetic excitations rather than a FS gapping as its origin, consistent with no dramatic change in QPS at T_{HO}. Further detailed studies as a function of magnetic field and pressure are planned, and expanding our investigation into another Kondo lattice system, UPd_2Al_3 [59], is of immediate interest. Also, other comparative studies will be fruitful, including intermediate valence vs the Kondo regime or Ce (one f electron) vs Yb (one f hole) compounds.

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