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Rapid Communications

Origin of gaplike behaviors in URu_2Si_2: Combined study via quasiparticle scattering spectroscopy and resistivity measurements

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We address two long-standing questions regarding the hidden order in URu_2Si_2: Is it associated with the hybridization process, and what are the distinct roles played by the localized and itinerant electrons? Our quasiparticle scattering spectroscopy reveals a hybridization gap ubiquitous in the entire phase space spanned by P and Fe substitutions in URu_2Si_2, including the no-order and antiferromagnetic regions, with minimal change upon crossing the phase boundary. This indicates its opening is not associated with the ordering, and thus localized electrons must be the major player. Towards a consistent understanding of all the other gaplike behaviors observed only below transition temperatures, we analyze the electrical resistivity using a model in which gapped bosonic excitations are the dominant scattering source. With their stiffness set to follow an unusual temperature dependence (decreasing with decreasing temperature), this model fits all of our resistivity data well including the jump at the transition. Remarkably, the extracted gap increases slowly with increasing Fe content, similarly to the gap detected by inelastic neutron scattering at $Q_1 = (1.4, 0, 0)$, suggesting a common origin. Such a model can also naturally explain the Hall effect temperature dependence without invoking Fermi surface gapping.

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Strongly correlated electron systems oftentimes exhibit seemingly similar phase diagrams. For their comprehensive understanding, it is not only necessary to identify the underlying interactions but also to elucidate the interplay among them. The f-orbital based heavy-fermion compounds are an archetypal correlated system, in which the hybridization between itinerant and localized electrons causes the emergence of heavy fermions [1,2]. What different roles are played by multiple f electrons is a key question in certain actinide compounds.

URu_2Si_2 is such a system, known for a phase transition at 17.5 K (T_{HO}) into the "hidden order" (HO) state [3]. Despite decades of intensive research [4], whether the HO is primarily associated with itinerant [5–7] or localized electrons [8–11] remains to be unambiguously determined. According to previous studies by some of us using quasiparticle scattering spectroscopy (QPS) [8,9], the hybridization gap opens well above T_{HO}, questioning the hybridization process being directly responsible for the HO [12–15]. This result also poses a challenge to the Fermi surface (FS) gapping picture, widely adopted to explain gaplike behaviors [5,7,16,17]. This is because the corresponding drastic change in the spectral density must be detected by QPS [18,19] as it exploits ballistic transport near the Fermi level [19,20], but no such signature was actually observed [8,9]. Another remaining issue is that the gap values extracted from different measurements are somewhat discrepant, e.g., in an analysis of electrical resistivity, heat capacity, and thermal expansion coefficient data [17]. The electrical resistivity has been frequently fit to expressions derived for the scattering off magnonlike excitations [17,21,22]. However, despite the likely existence of such collective modes, associating the extracted gap with the FS is questionable since it must be for the spin, rather than charge, sector. In addition, the resistivity jump at T_{HO}, taken widely as strong evidence for carrier depletion upon the FS gapping, needs to be explained quantitatively. After all, gaplike behaviors in URu_2Si_2 may reflect different aspects of the HO problem rather than sharing a single cause; thus, it is crucial to distinguish their origins.

Another approach is to investigate how the HO is related to other phases induced by tuning quantum critical control parameters. The effect of chemical substitution has been extensively studied including Rh [23,24] and Os [25]. In particular, P in $URu_2Si_{2-x}P_x$ nominally adds conduction electrons. However, unlike most other substituents, the phase diagram spans a no-order (NO, i.e., paramagnetic) region that separates the HO completely from an antiferromagnetic phase (AF-I) [26,27], as shown in Fig. 1(a). On the other hand, the isoelectronic substitution of Fe in $URu_{2-y}Fe_ySi_2$ causes a continuous transformation of the HO into another antiferromagnetic phase (AF-II) with a coexisting (CE) region in between [17], as shown in Fig. 1(b). This phase diagram closely resembles that of the parent compound under pressure [28], suggesting the Fe substitution effectively acts like applying hydrostatic pressure [29]. The pressure-induced large moment antiferromagnet (LMAF) was found to return to the HO under a strong magnetic field [30]. The smooth evolution between the HO and AF-II or LMAF suggests their underlying interactions may be rooted on the same ingredients, unlike

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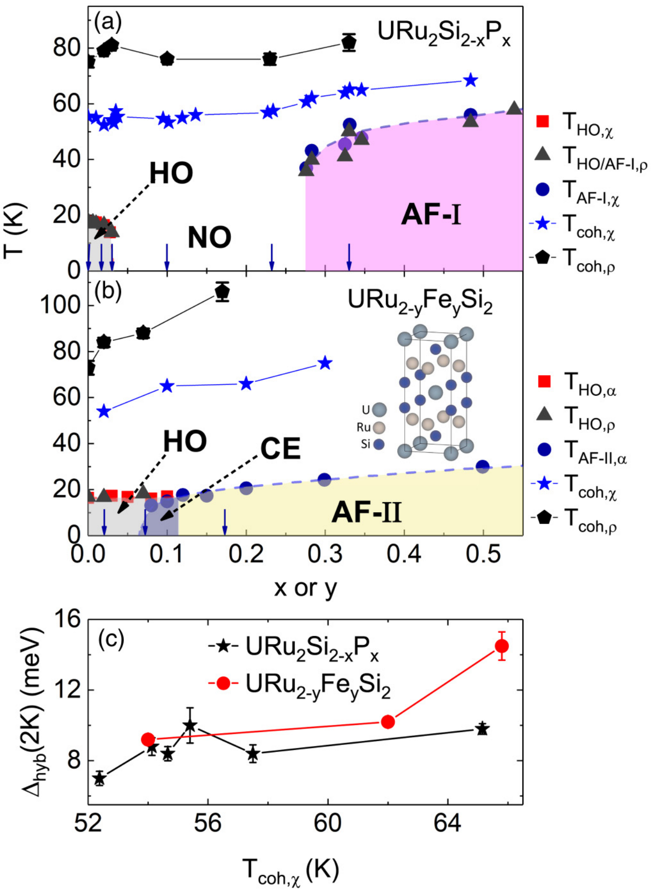

FIG. 1. (a) Temperature vs P-content (T-x) phase diagram of $URu_2Si_{2-x}P_x$, adapted from Ref. [27] and based on the measurements of magnetic susceptibility (χ) and electrical resistivity (ρ). AF-I stands for antiferromagnetic order and NO for no-order. T_{coh} denotes the coherence temperature. (b) T-y phase diagram of $URu_{2-y}Fe_ySi_2$ constructed based on thermal expansion coefficient (α) and resistivity [17], and magnetic susceptibility measurements [32]. AF-II stands for antiferromagnetic order and CE for coexisting orders. The inset depicts a unit cell of URu_2Si_2. In both panels, vertical arrows along the horizontal axis indicate substituent concentrations studied in this work. (c) Hybridization gap (Δ_{hyb}) at $T = 2$ K (from Figs. 2 and 3) vs coherence temperature ($T_{coh,\chi}$). The solid lines are a guide to the eyes.

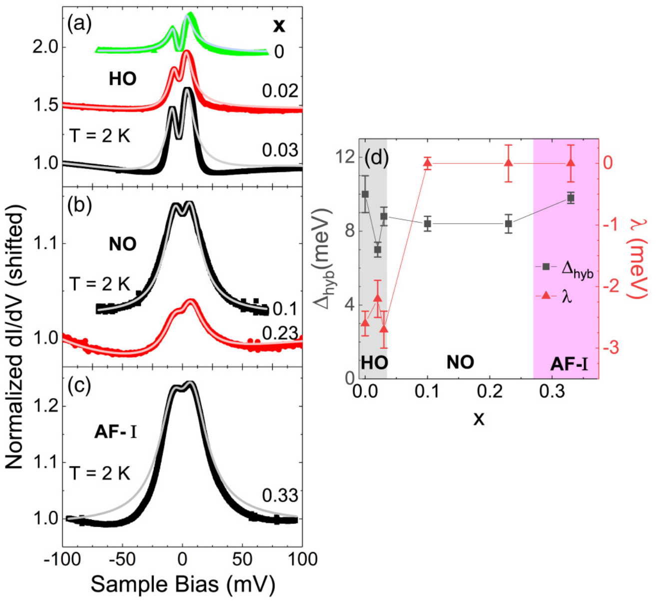

FIG. 2. (a)–(c) Normalized differential conductance taken from junctions on $URu_2Si_{2-x}P_x$ at $T = 2$ K (colored symbols) and best-fit curves (solid gray lines). In (a) and (b), data and fit curves are shifted vertically for clarity. (d) Hybridization gap (Δ_{hyb}) and renormalized f level (λ) extracted from an analysis using the MDC model. Labels denote different regions in the phase diagram.

for AF-I. Thus, comparative studies of all these phases should bring novel insights into the HO problem.

In this Rapid Communication, we report a combined study via QPS and resistivity measurements over the entire phase space for $URu_2Si_{2-x}P_x$ and $URu_{2-y}Fe_ySi_2$. The hybridization gap is observed in all phases including the NO region and evolves smoothly across phase boundaries, indicating the hybridization is a general process for heavy fermions rather than a driving force for phase transitions. For a consistent understanding of all gaplike behaviors, we advance a novel interpretation of the electrical resistivity by considering the scattering off gapped bosonic excitations in the ordered state [31]. By allowing an unusual temperature dependence of their *stiffness*, all of the characteristics including the jump can be nicely replicated. Our analysis also reveals the different nature of the AF-I from AF-II phases, for whose microscopic understanding we provide speculations on how differently P and Fe substitutions affect the underlying interactions.

$URu_2Si_{2-x}P_x$ and $URu_{2-y}Fe_ySi_2$ single crystals were grown by molten metal flux [26] and Czochralski methods [17], respectively. The (0 0 1) surface of the crystals was then manually polished to a few nanometers peak-to-dip roughness. Such a smooth surface is essential in making a spectroscopic junction (Sec. I in Ref. [33]) free from local heating effect that obscures intrinsic information (Sec. II in Ref. [33]). QPS junctions were formed using Au tips [34] in a custom-built rig [35] and differential conductance across the junction was measured using a standard four-probe lock-in technique. The conductance data were analyzed using the Maltseva-Dzero-Coleman (MDC) model ([36]; also see Sec. III in Ref. [33]), according to which the conductance curve can be asymmetric due to a Fano resonance [37] between the two *cotunneling* channels into a Kondo lattice. DC electrical resistance was measured with the four-probe method and analyzed using a model proposed by Jobiliong *et al.* [31].

The conductance spectra for $URu_2Si_{2-x}P_x$ with different P content (x) are displayed in Figs. 2(a)–2(c). They all exhibit an asymmetric double-peak structure resulting from the above-mentioned Fano resonance in a Kondo lattice [36]. It becomes smeared at large x as the electronic mean free path gets shortened due to increasing disorder, also reflected in the corresponding decrease of the residual resistivity ratio (RRR) (Sec. II in Ref. [33]). This structure has been established as signifying an indirect gap in the hybridized bands through recent theoretical [36,38–40] and experimental [8,9,20,41–43] studies. The solid lines are best-fit curves using the modified MDC model ([36,40]; also Sec. III in Ref. [33]). The extracted hybridization gap (Δ_{hyb}) and renormalized f

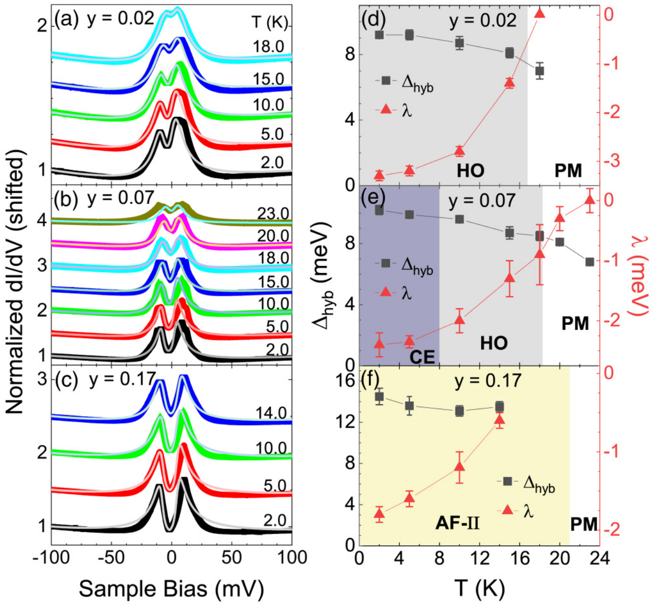

FIG. 3. (a)–(c) Temperature-dependent conductance spectra taken from junctions on $URu_{2-y}Fe_ySi_2$ (colored symbols) and best-fit curves (solid gray lines). Data and fit curves are shifted vertically for clarity. (d)–(f) Extracted hybridization gap (Δ_{hyb}) and renormalized f level (λ).

level (λ) are shown as a function of x in Fig. 2(d). The gap size for the parent compound is about 10 meV, similar to the values obtained from the previous QPS [8,9] and recent optical conductivity measurements [6,7]. Note Δ_{hyb} changes very little as a function of x, similarly to T_{coh} vs x shown in Fig. 1(a) and, thus, roughly conforming to the known correlation, $\Delta_{hyb} \propto T_{coh,\chi}$ [44], as plotted in Fig. 1(c). In particular, a hybridization gap is still observed in the NO region, similar in magnitude to those in the HO and AF-I regions, clearly indicating it is not associated with emergent ordering.

Temperature-dependent conductance spectra for $URu_{2-y}Fe_ySi_2$ are shown in Figs. 3(a)–3(c). Again, a hybridization gap is observed in all phases. In contrast to $URu_2Si_{2-x}P_x$, the conductance curve does not exhibit a noticeable change in the sharpness with increasing Fe content (y), in agreement with the RRR not changing much. In the HO and CE regions, the hybridization gap opens well above T_{HO}, in agreement with recent optical conductivity measurements of the parent compound [6,7]. In the AF-II region, the junction became unstable above 14 K but the sharpness of the double-peak structure implies that the hybridization gap may remain open well above $T_{AF\text{-}II}$. The temperature dependence of the hybridization gap and the renormalized f level are plotted in Figs. 3(d)–3(f). Like in $URu_2Si_{2-x}P_x$, the gap at 2 K roughly exhibits the correlation, $\Delta_{hyb} \propto T_{coh,\chi}$ [44], as shown in Fig. 1(c). With increasing temperature, in all three regions, Δ_{hyb} decreases and λ approaches zero (the Fermi level), similar to what occurs in the parent compound [8]. Furthermore, all properties including the conductance shape, λ, and Δ_{hyb} exhibit a smooth evolution without any anomaly upon crossing the phase transition temperature. While our result does not rule out a reconstruction of the FS at T_{HO}, it cannot be understood within the FS gapping picture [5,7]. Meanwhile, upon suppressing the HO in both $URu_2Si_{2-x}P_x$ and $URu_{2-y}Fe_ySi_2$ (by chemical substitution or temperature), λ goes from negative to zero, as shown in Fig. 2(d) and Figs. 3(d)–3(f). Inferring from the well-known single impurity Kondo resonance of width W, for which the resonance energy is expressed as $\varepsilon_0 = \frac{W}{2}\tan[(1-n_f)\frac{\pi}{2}]$ [1], the above-described behavior of λ may indicate an accompanying change in the f-level occupancy $\langle n_f \rangle$. This speculation is in line with a recent proposal invoking a possible valence change associated with the HO transition [45]. In addition, according to resonant x-ray emission and electron-energy loss spectroscopy measurements on URu_2Si_2 [46,47], the $5f$ electron count in the HO is quite far away from an integer, in agreement with our λ being finite in the HO region. It is also notable that observations similar to ours for λ and Δ_{hyb} in $URu_{2-y}Fe_ySi_2$ have been reported in a recent photoemission study on URu_2Si_2 [48]: a Π-shaped quasiparticle band at the Γ point shifts to the Fermi level from below with increasing temperature, whereas the hybridization gap at the X point does not change.

Previous QPS studies on URu_2Si_2 concluded that the hybridization gap is not the HO order parameter [8,9]. Furthermore, a hybridization gap is observed to open well above the Néel temperature in another related U-based compound, UPd_2Al_3, known to be a local-moment antiferromagnet [49]. In the present study, the opening of a hybridization gap even in the NO region corroborates that hybridization is a generic process underlying the formation of heavy-fermion bands and thus, is not correlated with the type of an emergent order. As speculated previously [8,9], our observation questions the plausibility of the FS gapping scenario. Related to this, it is important to note that while the FS topology undergoes no significant change in the HO-to-LMAF transition [50,51], the magnetic moment becomes finite abruptly upon the transition [52]. Within the itinerant picture, the LMAF must arise from FS nesting. In turn, the abrupt increase in magnetic moment should reflect a large change in the FS topology, in apparent contradiction with quantum oscillation results [50,51]. This suggests that both the HO and LMAF are more likely associated with localized electrons rather than itinerant electrons.

Within such a localized picture, some of us previously showed [8] that the gaplike behavior of the in-plane electrical resistivity in URu_2Si_2 is associated with the E_1 gap detected in inelastic neutron scattering (INS) by analyzing the resistivity with a model proposed by Jobiliong *et al.* [31]. This model explains the temperature dependence of resistivity in antiferromagnets in terms of scattering off gapped magnon excitations. In the previous analysis, similar gapped bosonic excitations were assumed to exist in the HO, and it was shown the entire resistivity curve, including the jump at T_{HO}, could be fit by this model. But the expression used in this fit is valid only in the low-temperature limit ($T \ll \Delta$). Thus, here (Sec. IV in Ref. [33]) we use a more general expression that is not subject to this constraint [31]:

$$\rho(T) = \rho_0 + AT^2 + \frac{B}{T}\int_0^\infty \frac{k^4\sqrt{\Delta_{ab}(T)^2 + D(T)k^2}}{\sinh^2[\sqrt{\Delta_{ab}(T)^2 + D(T)k^2}/2T]}dk, \quad (1)$$

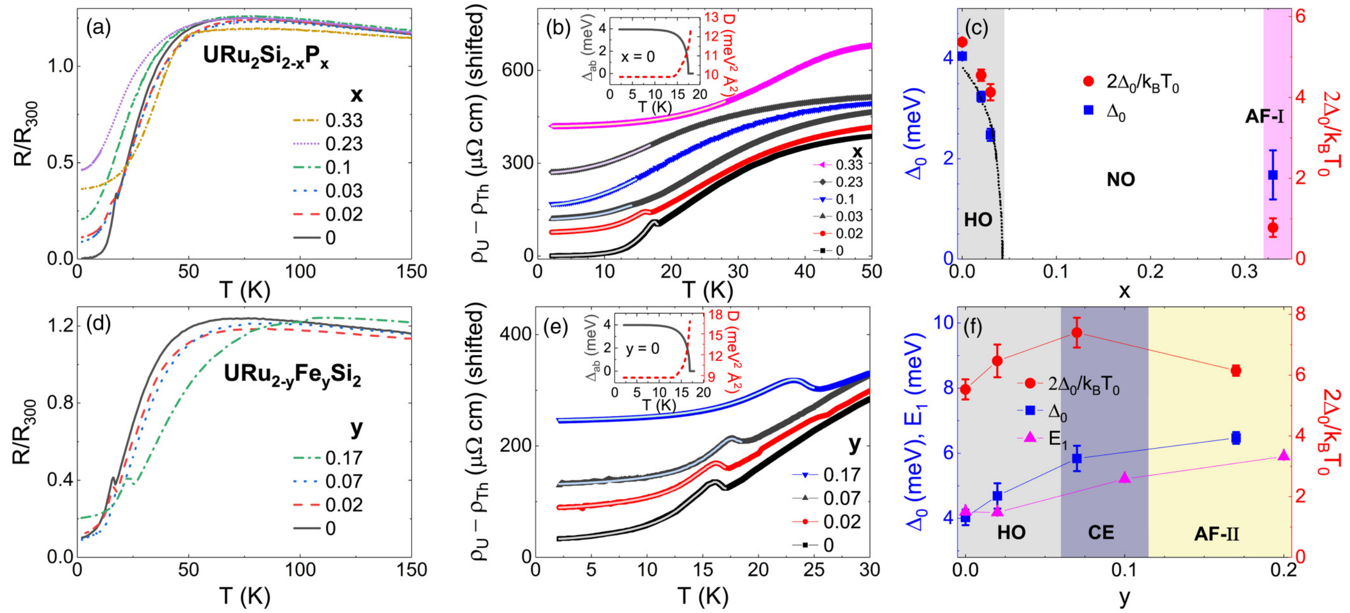

FIG. 4. (a) Normalized resistance vs temperature for $URu_2Si_{2-x}P_x$. (b) Resistivity in the low-temperature region with the phonon contribution subtracted out (colored symbols). ρ_U is the resistivity of $URu_2Si_{2-x}P_x$ and ρ_{Th} represents the resistivity of $ThRu_2Si_2$. Data and fit curves are shifted vertically for clarity. The inset shows the temperature-dependent bosonic excitation gap (left axis, solid line) and stiffness (right axis, dashed line) that are extracted for the parent compound. (c) Blue squares indicate the extracted gap value at zero temperature. The dashed line is a guide to the eye. Red circles represent the gap ratio, $2\Delta_0/k_BT_0$, where T_0 is the ordering temperature. (d)–(f) Same as (a)–(c) but for $URu_{2-y}Fe_ySi_2$. The E_1 gap for $URu_{2-y}Fe_ySi_2$ in (f) is from Ref. [54]. Solid lines are a guide to the eye.

where ρ_0 is the residual resistivity and the second term describes the Fermi-liquid behavior. Scattering off the bosonic excitations is accounted for by the third term, where k is the wave number, $D(T)$ is the stiffness, and the gap $\Delta_{ab}(T) = \Delta_0 \tanh(3.2\sqrt{T_0/T - 1})$. Δ_0 is the zero-temperature gap and T_0 is the ordering temperature. For more accurate estimation of the gap, the phonon contribution is eliminated by subtracting out the resistivity of $ThRu_2Si_2$, a compound isostructural to URu_2Si_2. By setting the stiffness as a free parameter, the entire resistivity curve including the jump at T_0 can be reproduced. Best-fit curves are shown in Figs. 4(b) and 4(e) with Δ_{ab} and D plotted in the insets for the parent compound for illustration (Sec. IV in Ref. [33]). While the data for AF-I with no jump at $T_{AF\text{-}I}$ were analyzed using the same approximate expression as in Ref. [8], the data for the HO, CE, and AF-II regions are all nicely fit by this general expression. Notably, the data for the NO region is fit well by an expression containing only the first two terms in Eq. (1) (Sec. IV in Ref. [33]), in agreement with the disappearance of bosonic excitations in this region.

The extracted zero-temperature gaps are plotted as a function of substituent concentration in Figs. 4(c) and 4(f). The gap ratio, $2\Delta_0/k_BT_0$, ranges from 4 to 7.5 except for AF-I, for which it is only ∼0.75, much smaller than 3.53 from the weak-coupling mean-field theory. This indicates that AF-I is of different nature from AF-II, as speculated earlier. A similarly small gap ratio (∼2) and the same kink-decay (instead of jump-decay) behavior are also observed in UPd_2Al_3 [43], suggesting that AF-I is more likely due to local moments, consistent with a recent nuclear magnetic resonance study [53]. The strongly contrasting properties between AF-I and AF-II might be due to the different roles played by different chemical substitutions, as mentioned earlier. As shown in the inset of Fig. 1(b), the Si sites are closer to the U sites than the Ru sites. Therefore, P substitution may affect the interaction that is responsible for the HO more drastically, transforming it into a rather conventional antiferromagnetic interaction. It is also notable that with increasing Fe content, the extracted gap closely follows the INS E_1 gap at $\mathbf{Q}_1 = (1.4, 0, 0)$ [54], as shown in Fig. 4(f), indicating that the INS E_1 gap may originate from the same gapped bosonic excitations as for the resistivity. Such association is also supported by the similar temperature dependence of the two gaps [10]. There is another gap detected by INS at $\mathbf{Q}_0 = (1, 0, 0)$. However, this gap is not only much smaller ($E_0 = 1.7$ meV for $y = 0$) but also detected only in the HO region [10], so it is unlikely to play a significant role in the resistivity jump-decay behavior. In contrast to the kink-decay behavior in the AF-I state, both HO and AF-II have the same jump-decay behavior and the bosonic excitation gap increases continuously when going from HO to AF-II, closely following T_0. These observations suggest that HO and AF-II may share a common order parameter. Such a model has been put forward recently to explain a resonance mode in Raman scattering observed in both the HO and AF-II phases [55]. Accordingly, one can imagine that similar bosonic excitations may exist in both phases, in line with our findings. Our analysis should also be applicable to gaplike behaviors in other experiments. For example, with decreasing temperature, the Hall coefficient in URu_2Si_2 abruptly increases at T_{HO}, then decays slowly at lower temperature, and this behavior was attributed to the depletion of charge carriers due to FS gapping [56]. Instead, the dominant scattering off gapped bosonic excitations in the HO or AF-II, in combination with a similarly anomalous temperature dependence of their stiffness, can qualitatively explain this behavior, similar to the case of $SrRuO_3$ where magnons are known to play a key role [57].

According to the working principle for QPS, such scattering off gapped bosonic excitations would show up as weak nonlinearity in the current-voltage characteristics at a bias voltage corresponding to the gap (~4 meV) [19]. Such a signature is not detected in our measurements presumably because it is buried in the conductance that varies rapidly due to the hybridization gap. It could be revealed in a second harmonic measurement, analogously to phonons in simple metals [19].

In conclusion, our QPS study on URu_2Si_2 containing P and Fe substituents reveals that a hybridization gap opens regardless of the emergent ordering including the NO without any anomaly upon crossing the phase boundary, indicating the hybridization is a general process instead of driving the phase transition. Because QPS detects quasiparticle scattering near the Fermi level, this result suggests the HO originates from localized (rather than itinerant) electrons. For a comprehensive understanding of all gaplike behaviors, we advance a new analysis of the electrical resistivity based on the scattering off gapped bosonic excitations, accounting for all of the characteristics, including the jump at the transition. The extracted gap is in agreement with the E_1 gap in INS. A similar approach can also provide a natural explanation for the Hall effect. Our results suggest the multitude of f electrons in URu_2Si_2 may play intriguing roles leading to intertwined orders (HO and AF), whose analogs can be found in other correlated systems [58–60].

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