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Anisotropic Kondo pseudo-gap in URu₂Si₂

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A polarized electronic Raman scattering study reveals the emergence of symmetry dependence in the electronic Raman response of single crystalline URu₂Si₂ below the Kondo crossover scale $T_K \sim 100$ K. In particular, the development of a coherent Kondo pseudo-gap predominantly in the E_g channel highlights strong anisotropy in the Kondo physics in URu₂Si₂ that has previously been neglected in theoretical models of this system and more generally has been sparsely treated for Kondo systems. A calculation of the Raman vertices demonstrates that the strongest Raman vertex does indeed develop within the E_g channel for interband transitions and reaches a maximum along the diagonals of the Brillouin zone, implying a d -wave-like geometry for the Kondo pseudo-gap. Below the hidden order phase transition at $T_{HO} = 17.5$ K, the magnitude of the pseudo-gap is found to be enhanced.

The Kondo effect, in which an antiferromagnetic coupling between conduction electrons and local quantum impurities leads to the formation of a Kondo singlet, is well understood for single Kondo impurity systems and is characterized by the appearance of a Kondo resonance in the electronic density of states below a crossover temperature T_K [1]. By contrast, Kondo lattice systems containing rare-earth elements (for which the Kondo impurities are realized by a lattice of magnetic moments stemming from the localized f electron orbitals), appear much more complex [2] and exhibit surprising phenomena [3, 4]. Usually, the microscopic state of heavy-fermions is described with Anderson or Kondo lattice models [2] and most of the time the Kondo singlet state is considered to be fully isotropic (s -wave symmetry with zero angular momentum). In certain circumstances, however, a momentum-dependent Anderson hybridization, for which the f site hybridizes with its nearest-neighbor conduction sites [5], or a non-local Kondo coupling [6] can give rise to the formation of heavy quasiparticles with a strong momentum-space anisotropy - in such a case the singlet state will have a higher angular momentum rank (p , d , ...). As a consequence, the Kondo pseudo-gap (partial gapping of the Fermi surface) that develops in the coherent Kondo regime [7–10] may be strongly anisotropic, even exhibiting nodal and antinodal regions. Similarly to the widely discussed relationship between the pseudo-gap and the superconducting phase in the cuprates, an anisotropic (non s -wave) Kondo pseudo-gap may have significant consequences for the entire phase diagram of these Kondo systems. However, reports of

such momentum-dependent hybridization are scarce [11] and remain indirect.

A non-local Kondo hybridization may be particularly relevant for the complex Kondo system URu₂Si₂ where an enigmatic "hidden" order (HO) phase appears inside the Kondo coherent regime below $T_{HO} = 17.5$ K and an unconventional superconducting (SC) phase develops below $T_c = 1.5$ K [12, 13]. Many spectroscopic experiments have reported a Kondo pseudo-gap opening at a temperature scale $T_K = 50 - 100$ K, well above T_{HO} , suggesting that Kondo hybridization is not the cause of the HO transition [14–19]. On the other hand, scanning tunneling microscopy (STM) has revealed an asymmetric pseudo-gap opening at T_{HO} within the Kondo-Fano lattice structure that appears below T_K [20, 21]. The HO gap is found to be correlated on the atomic scale with the electronic signatures of the Kondo lattice state, suggesting that the two phenomena involve the same electronic states. Angle-resolved photoemission spectroscopy (ARPES), on the other hand, has shown that a Kondo pseudo-gap opens at the X point of the Brillouin zone, while the HO gap affects electronic states at the Γ and Z points [22]. Nevertheless, the electronic structure in the HO state at the Z point (M-shape dispersion) results from the hybridization of a light electron band with already hybridized bands characterizing the paramagnetic state. Moreover, high magnetic field measurements have demonstrated that the destabilization of the HO state (around 37 T) coincides with the "collapse" of the Kondo crossover ($T_{\rho,max}, H_{\rho,max} \rightarrow 0$) [23].

Significant theoretical works have been performed to

identify the correct order parameter for the HO state [13]. Some theories consider the Kondo hybridization as an important ingredient in the realization of the HO [24–28], with others even proposing the hybridization as the order parameter of the HO state itself [29, 30]. Moreover, under a pressure of ~ 0.5 GPa, the SC and HO states collapse together [12], promoting the HO as a precursor of SC pairing possibly of a chiral d -wave type [31–33]. The proper role played by the HO in the appearance of the SC, however, remains unclear.

In this Letter, we present Raman polarized spectroscopy measurements in both the Kondo phase and the HO phase of URu₂Si₂. We observe a Kondo pseudo-gap opening only in the E_g symmetry below T_K . These results, together with our accompanying Raman calculations, highlight a strong anisotropy in the Kondo physics in k -space and reveal a d -wave like geometry for the Kondo pseudo-gap that may play a key role in the realization of the HO phase.

Polarized Raman experiments have been carried out using a solid-state laser emitting at 532 nm and a K⁺ laser emitting at 647 nm. The scattered light has been analyzed by a Jobin Yvon T64000 simple grating spectrometer equipped with an ultra-steep long-pass edge filter to reject the Rayleigh scattering. Single crystals of URu₂Si₂ were grown by the Czochralski method using a tetra-arc furnace [34]. All the D_{4h} point group (space group $n^\circ 139$) symmetries [35] have been measured. To probe the E_g symmetry, we have used samples polished along the ac -plane. For completeness, A_{1g} , A_{2g} , B_{1g} and B_{2g} symmetries have been explored on samples freshly cleaved along the ab -plane. Temperature-dependent studies were carried out in a closed-cycle ⁴He cryostat with the sample in vacuum. All reported temperatures are corrected for laser heating following Refs. [36–39].

Figure 1 presents the Raman spectra of URu₂Si₂ in the E_g and $A_{1g}+B_{2g}$ symmetries. For the E_g symmetry (Fig. 1 a)), the sharp peaks at 210 cm⁻¹ and 390 cm⁻¹ are the two E_g phonon modes [40], while the small peak at 430 cm⁻¹ is a leakage of the A_{1g} phonon mode. The bump at ~ 800 cm⁻¹ has been associated with a crystal electric field excitation [41]. In the $[A_{1g}+B_{2g}]$ symmetry (Fig. 1 b)), the intense A_{1g} phonon mode is observed at ~ 430 cm⁻¹. Smaller features seen at 350, 760 and 832 cm⁻¹ are related to double phonon processes [41]. Finally, a tiny leakage of B_{1g} and E_g phonon modes are visible respectively at 165, 210 and 390 cm⁻¹.

Remarkable effects are observed in the electronic continuum below ~ 700 cm⁻¹ (~ 90 meV) in both the E_g and A_{1g} symmetries, i.e. a depletion in a large energy range upon cooling. As shown in Fig. 2 a) and b), such depletion is absent in the B_{1g} , B_{2g} or A_{2g} channels. In Figure 2 c), we illustrate this loss of spectral weight by integrating $\Delta\chi''(T)=\chi''(T)-\chi''(320K)$ from the lowest energy (100 cm⁻¹) to 1500 cm⁻¹ for all symmetries.

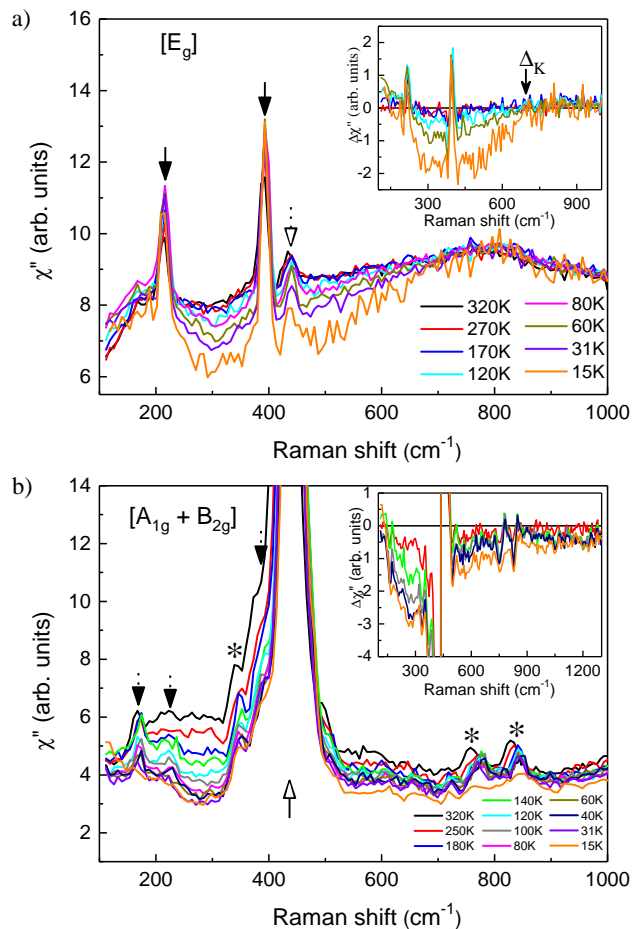


FIG. 1. (Color online) Raman spectra of URu₂Si₂ in the a) E_g and b) $A_{1g}+B_{2g}$ symmetries. The susceptibilities χ'' are normalized to the laser power. Full and open arrows indicate respectively the E_g and A_{1g} phonon modes. Dotted arrows indicate leakages of phonon modes. The symbol * denote double phonon processes [41]. Insets show the subtracted Raman responses $\Delta\chi'' = \chi''(T) - \chi''(T_{320K})$. The laser line wavelength used is 532 nm. Δ_K corresponds to the energy below which the Kondo pseudo-gap opens.

An example of the raw data is presented in the inset of Fig. 2 c). The large error bars in the $[B_{1g} + B_{2g}]$ channel prohibit us from drawing any definitive conclusions for the B_{2g} symmetry. Nevertheless, its spectral weight loss is found to be weaker than for the E_g response. This loss of spectral weight happens from 320 K in both E_g and A_{1g} symmetries. Below 100 K however, the depletion in the E_g symmetry begins to accelerate, while for A_{1g} , it decreases at a constant rate. This temperature scale of 100 K matches with T_K [20, 21, 42, 43]. Thus, the Raman response of URu₂Si₂ exhibits a clear signature of the Kondo crossover, but only in the E_g symmetry, i.e. a partial depletion in the electronic continuum below $\Delta_K \sim 700$ cm⁻¹ not associated with any symmetry breaking. This symmetry dependent pseudo-gap implies a strong anisotropy in the Kondo physics in

URu₂Si₂. Optical conductivity also shows anisotropic behavior between the *ab*-plane and *c*-axis responses [19, 44]. The *ab*-plane optical conductivity is strongly influenced by the formation of coherent Kondo singlets, reminiscent of what happens in the E_g Raman response, while the *c*-axis optical conductivity decreases continuously below 40 meV with no pseudo-gap signatures through the Kondo crossover, mimicking the Raman response seen in the A_{1g} channel.

Finally, it is worth noting that the total spectral weight loss in the E_g channel is most significant between 30 K and 15 K when entering the HO phase. Thus, the development of the HO state - for which the A_{2g} Raman signatures are fully developed below 16 K [45] - reinforces the Kondo pseudo-gap opening. This point will be discussed in more detail later on.

While a single energy scale $\Delta_K = 700 \text{ cm}^{-1}$ for the pseudo-gap is identified in both E_g and A_{1g} symmetries (see inset fig.2c), the temperature dependence of the Raman susceptibilities in these symmetries reveals two distinct temperature scales: one above 320 K (T_K^{high}) and a second at T_K. High-temperature Kondo ($T_K^{high} > T_K$) behavior has been observed previously in transport measurements at $\sim 370\text{K}$ [46]. T_K^{high} may result from crystal electric field effects tuning the degeneracy of the *f* orbitals. Indeed, it has been shown that T_K for a 4-fold degenerate Kondo model is significantly higher than T_K computed for the 2-fold degenerate equivalent model [2, 7]. Thus, a similar Kondo description could interpolate smoothly, for instance, from 4-fold degenerate (almost isotropic) impurities below T_K^{high} to 2-fold degenerate (anisotropic effective) impurities below T_K. Consequently, the Kondo physics may be observable in several Raman-active symmetries below T_K^{high} - here in both E_g and A_{1g} symmetries - and strengthen only in a specific symmetry - E_g in the present case - below T_K.

The energy scale for the spectral weight depletion in the E_g and A_{1g} symmetries is quite consistent with the previous observations of a Kondo pseudo-gap in URu₂Si₂ at $\sim 500 \text{ cm}^{-1}$ by optical conductivity measurements [14–17, 47] or at $\sim 250 \text{ cm}^{-1}$ by STM measurements [20, 21]. ARPES [22] and quasi-particle scattering spectroscopy [18] reported a lower hybridization pseudo-gap at 11 meV ($\sim 90 \text{ cm}^{-1}$). In Kondo models [7–10, 48], the electronic dispersion is characterized by a direct pseudo-gap (with zero transferred momentum) of order an effective hybridization Δ and an indirect pseudo-gap $\Delta^2/W \sim k_B T_K$ where *W* is the electronic bandwidth. Raman spectroscopy probes the direct Kondo pseudo-gap, while, for example, ARPES measures the indirect Kondo pseudo-gap. In the present case, $\Delta \sim \Delta_K = 700 \text{ cm}^{-1}$ and if we assume *W* = 1 eV, we extract an indirect pseudo-gap of 7.5 meV ($\sim 60 \text{ cm}^{-1}$) in agreement with the ARPES measurements. From these phenomenological considerations, the relevant tempera-

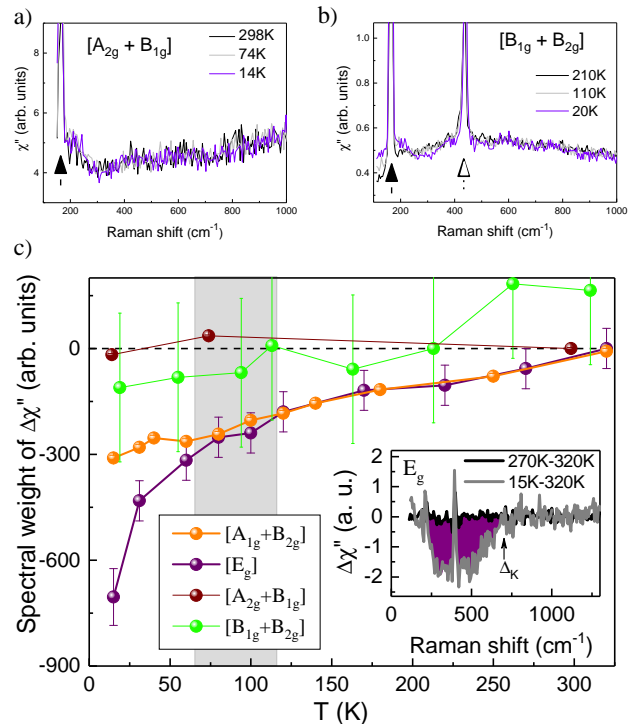


FIG. 2. (Color online) Raman spectra of URu₂Si₂ in the a) [A_{2g} + B_{1g}] and b) [B_{1g} + B_{2g}] symmetries. The laser line wavelengths are 647 nm and 532 nm, respectively. Open-dotted and full-dashed arrows indicate respectively the A_{1g} (leakage) and the B_{1g} phonon modes. c) Temperature dependence of normalized spectral weights of subtracted Raman responses $\Delta\chi'' = \chi''(T) - \chi''(T_{\text{ambiente}})$ in [A_{2g} + B_{1g}], [B_{1g} + B_{2g}], [A_{1g} + B_{2g}] and [E_g] symmetries. Insert shows an example of how the spectral weight was extracted in the E_g symmetry, in this case at *T* = 15 K. A Kondo pseudo-gap opens below ~ 100 K only in the E_g symmetry.

ture scale is found to be of order T_K. Interestingly, this description also accounts for the very large ratio between the Kondo direct pseudo-gap energy and the Kondo crossover temperature $\frac{\Delta_K}{k_B T_K}$. This ratio is of order 10 in URu₂Si₂, as found in Kondo insulators [49, 50].

In general and in contrast to optical conductivity measurements, electronic Raman scattering is not expected to follow spectral weight sum rules. However, some models show that interaction-dependent sum rules are possible [51]. For instance, in Kondo insulators such as FeSi or SmB₆, Raman electronic spectral weight that is suppressed at low frequencies by the charge gap is primarily recovered within an energy range equal to 6 Δ . This has been discussed theoretically in the framework of the Hubbard model [50] and extended to metallic systems for which a clear redistribution of spectral weight to a Fermi-liquid peak at low energy or in some cases to a high energy charge-transfer peak is expected [52]. However, according to our present and previous [45] results, URu₂Si₂ does not show any Raman spectral weight re-

distribution either above or below the depletion energy scale, which seems to indicate that the URu₂Si₂ Raman response does not follow any particular sum rule. Thus, our results call for further theoretical investigations in the context of Kondo metallic systems.

The E_g symmetry dependence of the Raman electronic continuum presented above implies k -space anisotropy in the quasi-particle excitations. Via the Raman vertex [53], polarized Raman scattering has the ability to provide information on the different regions of the Brillouin Zone probed for each selected symmetry. In order to elucidate the k -space dependence of the Kondo physics, we have therefore carried out Raman vertex calculations within the effective mass approximation [53, 54] [55].

The intraband and interband Raman vertices have been calculated using a tight-binding electronic band structure developed on the $5f^2$ uranium electron's configuration for URu₂Si₂ [56, 57]. This model is based on an effective one-particle picture with the $J=5/2$ states. The crystal electric field effect splits this sextet into three Kramers doublets $\Gamma_7^{(1)} \oplus \Gamma_7^{(2)} \oplus \Gamma_6$. For simplicity, the Γ_6 doublet is neglected. The main contribution to the Fermi surface comes from the two Γ_7 doublets that are described in the tight-binding approximation. Here, the Kondo lattice effect is considered as a mixing between the two Γ_7 doublets whose contribution to the Hamiltonian depends on a hopping term t_{12} between uranium sites at the zone center and at the corners [58]. The body-centered tetragonal (BCT) Brillouin zone of the paramagnetic state is shown in Figure 3 a). The Fermi surface obtained (Fig. 3 b)) from the electronic dispersion (Fig. 3 c)) is consistent with earlier *ab initio* calculations [59]. Only the small electrons pockets at Γ and X [60] are not reproduced by this simple model [57]. The electronic dispersion does not depend strongly on the hopping term t_{12} except around the N point where the energy between the two Γ_7 bands increases when t_{12} deviates from zero [61].

From our calculations, large values of the Raman vertices were found in the E_g and A_{1g} symmetries, while in the B_{1g} and B_{2g} symmetries, the vertices remain very weak [62]. The strongest value of the Raman vertex is in fact found in the E_g symmetry for interband transitions with an energy scale of ~ 40 meV (320 cm^{-1}), in reasonable agreement with our observations. The E_g vertex for intraband transitions, on the other hand, is weak. According to this result, the E_g Raman response mainly comes from vertical interband transitions along the diagonals of the Brillouin Zone. Indeed, as emphasized in Figure 3 b), the interband E_g Raman vertex is strong only at the X , Y and Y_1 points (as indicated by solid arrows in Fig 3 c)). Consequently, the Kondo pseudo-gap is linked to the electronic states at these k -points, reflecting the d -wave like geometry of the Kondo pseudo-gap.

The A_{1g} interband vertex is particularly strong at the N point [63] where the interband vertical electronic tran-

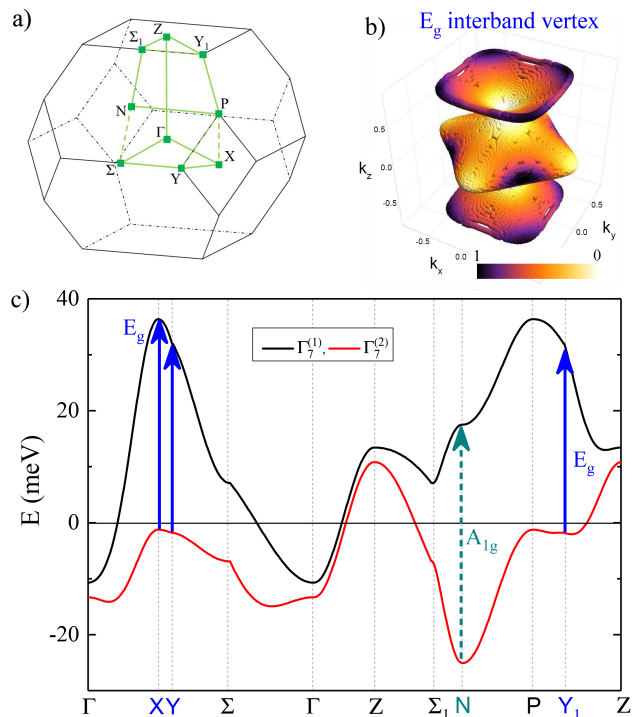


FIG. 3. (Color online) a) Body-centered tetragonal Brillouin zone of URu₂Si₂. b) Relative magnitude of the E_g Raman vertex at the Fermi surface for interband transitions calculated within the effective mass approximation. c) Calculated electronic dispersion of the $\Gamma_7^{(1)}$ and $\Gamma_7^{(2)}$ bands including hybridization. The solid (dashed) arrows at the X , Y and Y_1 (N) locations indicate the Brillouin Zone points where the E_g (A_{1g}) interband vertex reaches a maximum value (See text).

sition has typically the same energy scale. The Raman electronic depletion change is smooth at T_K in this symmetry thus confirming that the Kondo pseudo-gap does not affect the N point of the Brillouin zone; another indication of strong anisotropy in the Kondo physics in URu₂Si₂.

We now turn to discuss the relationship between the HO state and the Kondo physics in the light of our Raman results. The main observations are two-fold: the Kondo pseudo-gap in the E_g channel is reinforced below T_{HO} , confirming that the Kondo effect is not suppressed upon entering the HO state [26]. Moreover, our experimental results reveal no pseudo-gap opening in the A_{2g} symmetry despite the fact that this peculiar symmetry has been shown to exhibit all the Raman signatures of the HO state, including a sharp excitation (at 1.7 meV) and a gap (at 6.8 meV) [45, 64]. Raman experiments [45, 65] also showed that the width of the A_{2g} quasi-elastic continuum in the paramagnetic state is affected by the Kondo crossover, suggesting that the A_{2g} Raman channel is sensitive to the hybridization process. A definitive conclusion could be reached by going beyond the effective mass approximation so that the A_{2g} Raman

vertex is not expected to be zero. This would provide the k -dependent variation in the A_{2g} Raman response, notably the HO A_{2g} gap below 6.8 cm^{-1} , that may also have a d -wave geometry and by doing so, may be favorable to the chiral d -wave $k_z(k_x + ik_y)$ (E_g) pairing proposed for the SC state [31–33].

Interestingly, in a two-channel Kondo model such as the hastatic order model [30], the normal Kondo crossover at T_K may be converted into a phase transition at T_{HO} . Multiple components for the hybridization gap with different symmetries are expected: one that could open at T_K (as a crossover without symmetry breaking) and one that should manifest at T_{HO} as an order parameter for the HO [66]. A model inspired by a similar scenario might reconcile the E_g symmetry of the Kondo pseudo-gap with the A_{2g} symmetry of the HO gap.

In summary, we report the observation of an anisotropic Kondo pseudo-gap in a metallic Kondo lattice system by Raman spectroscopy. We observe a pseudo-gap opening within the E_g symmetry in URu_2Si_2 that appears to be linked to the Kondo crossover below 100 K. This Kondo pseudo-gap becomes even deeper upon entering the HO state pointing to a link between the Kondo physics and the HO state. From Raman vertex calculations, the interband Raman vertex in the E_g symmetry is strongest at the X , Y and Y_1 points of the Brillouin zone, suggesting that the Kondo pseudo-gap has a d -wave like symmetry. This anisotropy in the Kondo physics may play a significant role in the realization of the HO and henceforth should be considered as a key element in further theoretical developments.

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