A$_{2g}$ Signatures of the Hidden Order State of URu$_2$Si$_2$

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We have performed polarized electronic Raman scattering on URu$_2$Si$_2$ single crystals at low temperature down to 8 K in the hidden order state and under magnetic field up to 10 T. The hidden order state is characterized by a sharp excitation at 1.7 meV and a gap in the electronic continuum below 6.8 meV. Both Raman signatures are of pure A$_{2g}$ symmetry. By comparing the behavior of the Raman sharp excitation and the neutron resonance at $Q_0=(0,0,1)$, we provide new evidence, constrained by selection rules of the two probes, that the hidden order state breaks the translational symmetry along the c axis such that $\Gamma$ and $Z$ points fold on top of each other. The observation of these distinct Raman features with a peculiar A$_{2g}$ symmetry as a signature of the hidden order phase places strong constraints on current theories of the hidden order in URu$_2$Si$_2$.

For almost three decades [1], the identity of the ordered phase found in URu$_2$Si$_2$ at temperature below $T_0=17.5$ K has eluded researchers [2, 3] despite intensive experimental and theoretical investigations. The second order transition to this so-called hidden order (HO) appears clearly in the thermodynamic and transport quantities [1, 4–7]. This unique electronic state is not a simple long-range magnetic (dipolar) order since the associated tiny magnetic moment measured in the HO state cannot account for the large entropy release during the transition [8]. Nevertheless, the HO changes to a simple antiferromagnetic with a simple tetragonal structure under a small applied pressure of 0.5 GPa [9–11]. Many interesting theories have been proposed to explain the nature of HO, among which multipolar orders from quadrupolar [12–16], local currents [17, 18], unconventional density wave [19, 20] modulated spin liquid [21, 22], and hastatic order [23] for the most recent ones. Yet a complete understanding of the nature of the hidden order has not been reached.

A wide variety of experimental studies have succeeded in revealing salient features of the HO state. Inelastic neutron measurements [8] observed two magnetic excitations with a commensurate wave vector $Q_0=(1,0,0)=(0,0,1)$ and an incommensurate wave vector $Q_1=(1.4,0,0)$ at 1.7 meV and 4.8 meV, respectively. The first one has been demonstrated to be a major signature of the HO state [26]. It is well accepted [6, 24] that a partial Fermi-surface gapping with a strong reduction of the carriers number occurs at $T_0$ and accordingly, the electronic structure abruptly reconstruits at $T_0$ [25]. It persists in the antiferromagnetic state under pressure [31], suggesting similar Brillouin zone folding in both states. Besides, a recent set of experiments have identified a fourfold symmetry breaking upon entering the HO state [32–34], steering some controversy [35, 36]. Preliminary connections between the fingerprints of the HO transition have been made. The electronic structure of the HO is consistent with a periodicity given by the commensurate wave vector $Q_0$ [37] and part of the gapping of the incommensurate spin fluctuations was related to the loss of entropy at the HO transition [25]. However, these relationships remain indirect. Additionally, the question of the symmetry of the novel excitations emerging from the HO state has not been addressed experimentally.

In this letter using electronic Raman spectroscopy, we report a direct intimate relationship between the electronic gap and the commensurate magnetic excitation together with their explicit symmetry. We observe clear Raman signatures of the HO state, i.e. a gap below $\sim 55$ cm$^{-1}$ (6.8 meV) and a sharp excitation deep inside the gap at $14$ cm$^{-1}$ (1.7 meV). Both signatures are observed only in the A$_{2g}$ symmetry and emerge from a broad A$_{2g}$ quasi-elastic continuum which persists up to 300 K. Given the peculiarity of the A$_{2g}$ symmetry itself, our results give new and strong constraints on the nature of the HO state. We further demonstrate that the sharp Raman excitation tracks the resonance at $Q_0=(0,0,1)$ detected by inelastic neutron scattering (INS) [24] in the HO state as a function of temperature and magnetic field, indicating that both excitations have the same origin even if measured at different wave vector transfer. This brings a new and robust evidence for a Brillouin zone folding which places the $Z$ point on top of the $\Gamma$ point as expected in a transition between a body center tetragonal (bct) and a simple tetragonal (st) phase.

Polarized Raman experiments have been carried out using a solid state laser emitting at 561 nm and a Jobin Yvon T64000 triple subtractive grating spectrometer equipped with a nitrogen cooled CCD camera. Single crystals of URu$_2$Si$_2$ were grown by the Czochralski method using a tetra-arc furnace [38]. Three samples...
from the same batch with a residual resistivity ratio of \( \sim 50 \) have been freshly cleaved along the (ab) plane. Temperature and magnetic field dependencies have been performed in a closed-cycle \(^4\)He cryostat with sample in high vacuum and a \(^4\)He pumped cryostat with the sample in gas, respectively. By combining different incident and scattered light polarizations and sample geometry, we have extracted the \( A_{1g}, B_{1g}, B_{2g} \) and \( A_{2g} \) symmetries of the \( D_{4h} \) space group (n\(^o\)139) [49].

**Figure 1.** (Color online) (a) Raman spectra of URu\(_2\)Si\(_2\) in the pure \( A_{2g} \) symmetry in the hidden order (HO) phase (13 K) and in the paramagnetic phase (22 K). In the HO, a sharp peak at 14 cm\(^{-1}\) is superimposed on a gap below \( \sim 55 \) cm\(^{-1}\). In the PM, the gap is almost constant up to 100 cm\(^{-1}\). We also report a gap of \( \Delta \sim \pm 0.004 \) from the same batch with a residual resistivity ratio of \( \sim 50 \) have been freshly cleaved along the (ab) plane. The energy of the gap is consistent with previous optical conductivity measurements [27, 48–50]. In addition, the gaps \( \Delta_Q \) extracted from the resistivity (\( \sim 56 \) cm\(^{-1}\)) [51] and from heat capacity measurements (\( \sim 88 \) cm\(^{-1}\)) [52, 53] are in the same energy range than our findings. Scanning tunneling microscopy experiments also report a gap of \( \sim \pm 4 \) meV\( \sim 65 \) cm\(^{-1}\) [54, 55].

We note that Raman scattering is also sensitive to double excitations processes, such as double phonon or double magnetic excitations with \( \pm Q \) transferred wave-vectors. A double excitation process involving the resonance at \( \pm Q_1 \) would be measured from \( \sim 65 \) cm\(^{-1}\), in the energy range of the Raman gap. However, we can rule out this interpretation because the \( Q_1 \) resonance strongly shifts to lower energy with increasing temperature. It has been reported to be inelastic above \( T_0 \) reaching \( \sim 2.5 \) meV at 20 K, i.e. 40 cm\(^{-1}\) for a double excitation [8, 56] whereas the \( A_{2g} \) Raman gap depletion vanishes at \( T_0 \) while its energy remains roughly constant up to \( T_0 \) [57]. This comparison, as well as the observation of a similar gap by optical conductivity measurements, suggests that the depletion is not linked to the resonance at \( Q_1 \) but to a gapped electron-hole excitations continuum as expected from a reconstruction of the Fermi surface inside the HO state. The new significant information relies in its emergence only in the unique \( A_{2g} \) symmetry, implying that the gap occurs in a continuum involving composite quasi-particles with mixed Spin-Orbit character.

Below the gap, the low energy \( A_{2g} \) Raman peak is sharp with a full width at half maximum (FWHM) of \( \sim 1 \) cm\(^{-1}\) at \( \sim 10 \) K, showing it is a long lived excitation. Figure 2(a) presents the temperature dependence of its position and FWHM. Its energy and width are almost constant up to \( \sim 15 \) K. It drops drastically to zero and broadens when approaching \( T_0 \). We also report the energy \( E_0 \) and FWHM of the neutron resonance at \( Q_0=(0,0.1) \), the up-to-now major signature of the HO phase [24, 26]. The \( A_{2g} \) Raman excitation closely tracks the neutron resonance which strongly suggests that the same excitation is coupled to both probes. In addition, as
shown figure 2(b) the peak hardens slightly under magnetic field up to 10 T which is qualitatively consistent with the magnetic field dependence of the neutron resonance [69]. Raman spectroscopy probes the Γ point, i.e. the total transferred wave vector \( \mathbf{Q} = 0 \). The feature observed at 1.7 meV by neutron scattering is measured at the Z point, i.e. at \( \mathbf{Q}_0 = (0,0,1) \). Measuring the same excitation at the Γ and Z points can be explained by invoking a Brillouin zone folding along the c axis which occurs upon entering the HO state, as a bct to st transition would produce (Cf. Figure 2(c)). The same conclusion was previously made from the comparison between the Fermi surfaces at ambient pressure and under pressure in the antiferromagnetic state [31] as well as from the comparison between photoemission spectroscopy (ARPES) data at the Γ and the Z points [29] [30] [60]. The conclusion drawn here is robust as it results from measurements at zero magnetic field and zero applied pressure. Most of all it is based on the observation of a major signature of the HO state at different \( \mathbf{Q} \) vector.

Interestingly, the finite Raman response in the \( A_{2g} \) symmetry is not limited to the HO state. Indeed, as reported by Cooper et al. [61], already at 300 K, URu₂Si₂ exhibits a \( A_{2g} \) quasi-elastic peak (QEP) with a overdamped Lorentzian line shape. As shown Figure 3(a), the quasi elastic contribution sharpens with decreasing temperature before collapsing in the HO state. The FWHM, calculated with a simple relaxation model [61], is reported in Figure 3(b). After a Korringa-like linear temperature dependence down to \( \sim 100 \) K, it exhibits a plateau-like behavior between \( \sim 100 \) K and \( \sim 50 \) K before decreasing three times faster down to 20 K. The plateau is thus limited to the Kondo regime with an increase of the lifetime of the \( A_{2g} \) excitations most probably below the Kondo coherence temperature. Via the Kramers-Kronig relation [62], the \( A_{2g} \) static susceptibility \( \chi_{0}^{A_{2g}} \) can be extracted as \( \int \chi'^{A_{2g}}(\omega)/\omega d\omega \) with integration spanning from 8 to 100 \( \text{cm}^{-1} \), above which all spectra are on top of each other. As shown in Figure 3(b), the \( A_{2g} \) static susceptibility exhibits a temperature dependence very reminiscent to the dc magnetic susceptibility along the c axis [63], suggesting a link between the \( A_{2g} \) degree of freedom and the magnetic susceptibility. The temperature dependence of this last one has been tentatively explained considering various crystalline electric field (CEF) schemes [64]. A similar Raman quasi-elastic response has already been discussed in the context 4f and 5f systems, where it was attributed to either spin fluctuations or localized CEF excitations like in UBe₁₃ [65].

In URu₂Si₂, a tempting simple interpretation of the \( A_{2g} \) QEP would be to consider a CEF excitation between very broad (and partially delocalized) levels. Indeed, simple local CEF excitations on the U atoms can have a \( A_{2g} \) symmetry, with different ground state, such as \( \Gamma_1(A_{1g}) \), \( \Gamma_2(A_{2g}) \), \( \Gamma_3(B_{1g}) \), \( \Gamma_4(B_{2g}) \); or \( \Gamma_6 \) and \( \Gamma_7 \) with an even or odd number of localized electrons, respectively.

Following this interpretation, a global and simple scenario, reminiscent of the results obtained across the metal-insulator transition of the skutterudite PrRu₄P₁₂ [66], is that the high temperature \( A_{2g} \) QEP as well as the sharp \( A_{2g} \) peak in the HO state are due to CEF excitations. The first one is strongly damped and quasielastic. At the HO transition, \( A_{2g} \) decaying channels are quenched at low energy due to the opening \( A_{2g} \) gap and consequently, the CEF excitation becomes long-lived. By the same process it becomes gapped (inelastic) because of the associated loss of hybridization with delocalized quasiparticle continuum in the HO state. In this picture, the dual character of the phase transition in URu₂Si₂ ap-
pears naturally, with the gap $\Delta_G$ observed in the Raman continuum, directly linked to the itinerant nature of the 5f electrons and the CEF excitation observed as the Raman sharp peak, associated their local character. Here, for the first time, a close relationship between both signatures can be made, through the similarity of their $A_{2g}$ symmetry.

Regardless the exact origin of the sharp peak, we note that the opening of the gap will help any excitation inside the gap to become long-lived and sharp. In this context, the sharp peak in URu$_2$Si$_2$ might be attributed to a collective mode as it has already been suggested in the context of the theory developed by Haule and Kotliar [14] [6] . Alternatively, the peculiar $A_{2g}$ symmetry may be due to chiral local current loop excitations. Indeed similar anomalous orbital motion of charge carriers have been shown to have the $A_{2g}$ symmetry [13] based on a Raman experiment in the insulating cuprates [44]. This hypothesis has also been recently brought up for URu$_2$Si$_2$ [45].

If we take a pure itinerant picture perspective, a Drude-like Raman response of electron-hole excitations may express the $A_{2g}$ QEP at high temperature. In order to account for the strong spectral weight in the $A_{2g}$ symmetry, this would require an intriguing electronic band structure on which $\frac{\partial^2 E}{\partial k_x \partial k_y} \neq \frac{\partial^2 E}{\partial k_y \partial k_x}$ in some k-space region of the Fermi surface, leading to a non-zero $A_{2g}$ Raman vertex in the effective mass approximation [17]. We surmise that theoretical investigations accounting for both the local and the itinerant character of the quasiparticle in URu$_2$Si$_2$ are necessary to reach a global scenario of these Raman signatures of $A_{2g}$ symmetry.

In conclusion, we have reported two Raman features of pure $A_{2g}$ symmetry, a sharp peak at 14 cm$^{-1}$ and a low energy gap at $\sim$ 55 cm$^{-1}$, as signatures of the hidden order state in URu$_2$Si$_2$. Additionally, by performing accurate temperature and magnetic field dependencies of the sharp Raman peak, we have shown that the sharp peak matches the neutron resonance at $Q_g$. This brings new and robust evidence of the Brillouin zone folding along c axis upon entering the HO state, consistent with a switch from body center tetragonal to simple tetragonal. Theoretical investigations are needed to conclude about the order parameter from which such excitations with peculiar $A_{2g}$ symmetry can emerge.

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