Symmetry of the Excitations in the Hidden Order State of URu_2Si_2

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We perform polarized electronic Raman scattering on URu_2Si_2 single crystals at low temperature down to 8 K in the hidden-order state and under a 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 Γ 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_2Si_2.

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For almost three decades [1], the identity of the ordered phase found in URu_2Si_2 at a 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 order 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 to dotriacontapolar [12–16], local currents [17,18], unconventional density wave [19,20], modulated spin liquid [21,22], dynamical symmetry breaking [23], and hastatic order [24] are 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 [8,25,26] observe two magnetic excitations with a commensurate wave vector $Q_0 = (1,0,0) \equiv (0,0,1)$ and an incommensurate wave vector $Q_1 = (1.4, 0, 0)$ at 1.7 and 4.8 meV, respectively. The first one has been demonstrated to be a major signature of the HO state [27]. It is well accepted [6,28] that a partial Fermi-surface gapping with a strong reduction of the carrier number occurs at T_0 and, accordingly, the electronic structure abruptly reconstructs at T_0 [29–32]. It persists in the antiferromagnetic state under pressure [33] suggesting similar Brillouin zone folding in both states. Besides, a recent set of experiments has identified a fourfold symmetry breaking upon entering the HO state [34–36], steering some controversy [37,38]. Preliminary connections between the fingerprints of the HO transition have been made. The electronic structure of the HO state is consistent with a periodicity given by the commensurate wave vector Q_0 [39] and part of the gapping of the incommensurate spin fluctuations was related to the loss of entropy at the HO transition [26]. 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 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, which indicates a direct intimate relationship between them. They emerge from a broad A_{2g} quasielastic 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) [25] 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 transfers. This brings new and robust evidence for a Brillouin zone folding, which places the Z point on top of the Γ point as expected in a transition between a body-centered-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_2Si_2 were grown by the Czochralski method using a tetra-arc furnace [40]. Three samples from the same batch with a residual resistivity ratio of ~50 and freshly cleaved along the *ab* plane have been measured. Temperature and magnetic field dependencies have been performed in a closed-cycle ^{4}He cryostat with the sample in high vacuum and a ^{4}He pumped cryostat with the sample in exchange gas, respectively [41]. By combining different incident and scattered light polarizations and sample geometries, we have extracted the A_{1g}, B_{1g}, B_{2g}, and A_{2g} symmetries of the D_{4h} point group (space group n°139) [46].

Figure 1(a) shows the low-frequency Raman spectra of URu_2Si_2 in the pure A_{2g} symmetry [47]. Upon entering the HO state, a gap opens below $\Delta_G \sim 55$ cm^{-1} and a sharp excitation emerges at ~14 cm^{-1} deep inside the gap [48]. The subtraction of the Raman responses in the paramagnetic (PM) and HO state in various symmetries is reported in Fig. 1(b). Within our accuracy, no feature can be detected in the other probed symmetries [47,51]. The HO state is thus characterized by two Raman signatures of pure A_{2g} symmetry. The A_{2g} symmetry is intriguing. It is equivalent to the C_{4h} subgroup of the D_{4h} space group and transforms as $xy(x^2 - y^2)$ or R_z [52]. This symmetry is usually associated with time reversal and/or chiral symmetry breaking excitations [54–57]. Among the large families of compounds belonging to the D_{4h} space group, including the cuprates and the Fe-based superconductors, few measurements only have reported a sizeable A_{2g} Raman response [58].

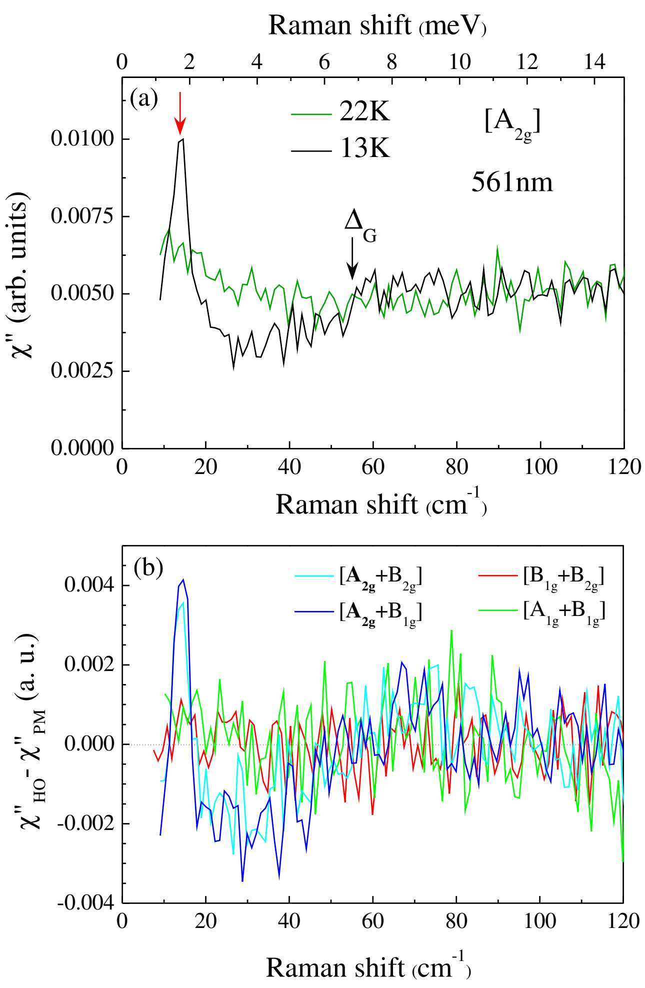

FIG. 1 (color online). (a) Raman spectra of URu_2Si_2 in the pure A_{2g} symmetry in the hidden-order (HO) phase (13 K) and in the paramagnetic (PM) phase (22 K). In the HO phase, a sharp peak at 14 cm^{-1}, indicated by the red arrow, is superimposed on a gap below $\Delta_G \sim 55$ cm^{-1}. (b) Subtracted Raman responses in the hidden-order phase and the paramagnetic phase for all probed symmetries. All spectra have been measured with the same laser power. Those containing the B_{1g} response have been normalized to the intensity of the B_{1g} phonon.

The energy of the gap is consistent with previous optical conductivity measurements [28,59–61]. In addition, the gaps Δ_G extracted from the resistivity (~56 cm^{-1}) [62] and from heat capacity measurements (~88 cm^{-1}) [63,64] are in the same energy range as our findings. Scanning tunneling microscopy experiments also report a gap of ~± 4 meV, i.e., $\Delta_G \simeq 65$ cm^{-1} [65,66].

We note that Raman scattering is also sensitive to double excitation processes, such as double phonon or two-spin excitations with $\pm\mathbf{Q}$ transferred wave vectors. A double excitation process involving the resonance at $\pm Q_1$ would be measured at ~75 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 ~2.5 meV at 20 K, i.e., 40 cm^{-1} for a double excitation [8,67] whereas the A_{2g} Raman gap depletion vanishes at T_0 while its energy remains roughly constant up to T_0 [68]. 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 excitation continuum as expected from a reconstruction of the Fermi surface inside the HO state [23,29–31]. We provide here new information on this electron-hole excitation continuum; i.e., it has the pure A_{2g} symmetry. From that, we surmise that the gap occurs in a continuum involving quasiparticles with strong spin-orbit character as described in Ref. [69].

Below the gap, the A_{2g} peak is sharp with a full width at half maximum (FWHM) of ~1 cm^{-1} at ~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 ~15 K. It abruptly drops to zero near T_0 with a temperature dependence stronger than expected for a mean field transition. It also 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 [25,27]. The A_{2g} Raman peak closely tracks the neutron resonance, which strongly suggests that the same excitation is coupled to both probes.

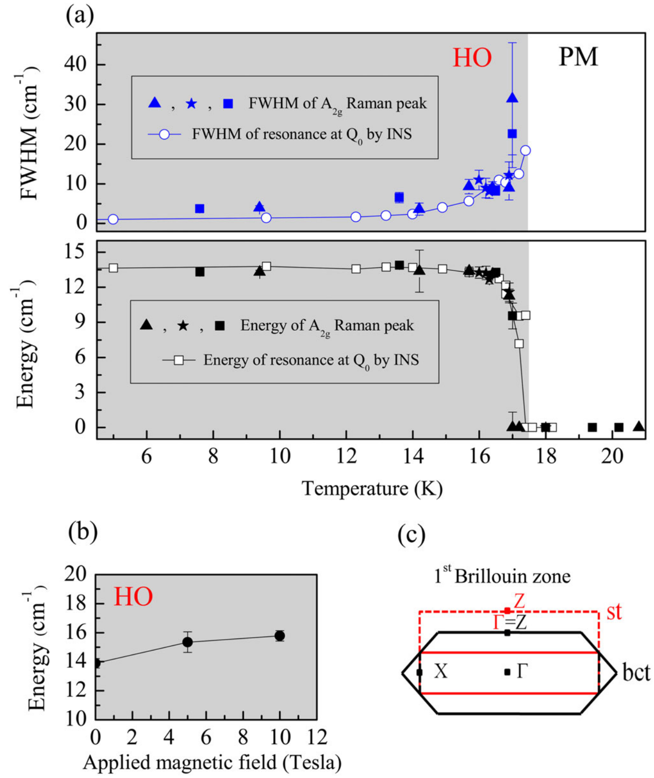

FIG. 2 (color online). (a) Temperature dependence of the energy and full width at half maximum (FWHM) [after deconvolution from the resolution of the spectrometer (2 cm^{-1})] of the sharp A_{2g} Raman peak (full symbols) and of the resonance at $Q_0 = (0, 0, 1)$ measured by inelastic neutron scattering (INS) (open symbols) [25]. The full symbols (stars, triangle, square) are extracted from various measurements on different samples from the same batch. The gray area corresponds to the temperature range of the HO phase. Both features are due to the same excitation probed by different techniques. The folding sketched in (c) would explain the observation of the same excitation at different **Q** wave vectors. (b) Magnetic field dependence of the energy of the sharp A_{2g} Raman peak. The magnetic field is applied at $30° \pm 5°$ of the c axis. (c) Sketch of the first Brillouin zone in the paramagnetic state [body-centered-tetragonal (bct) structure] and the folded one to the simple tetragonal (st) structure, which folds the Z point to the Γ point.

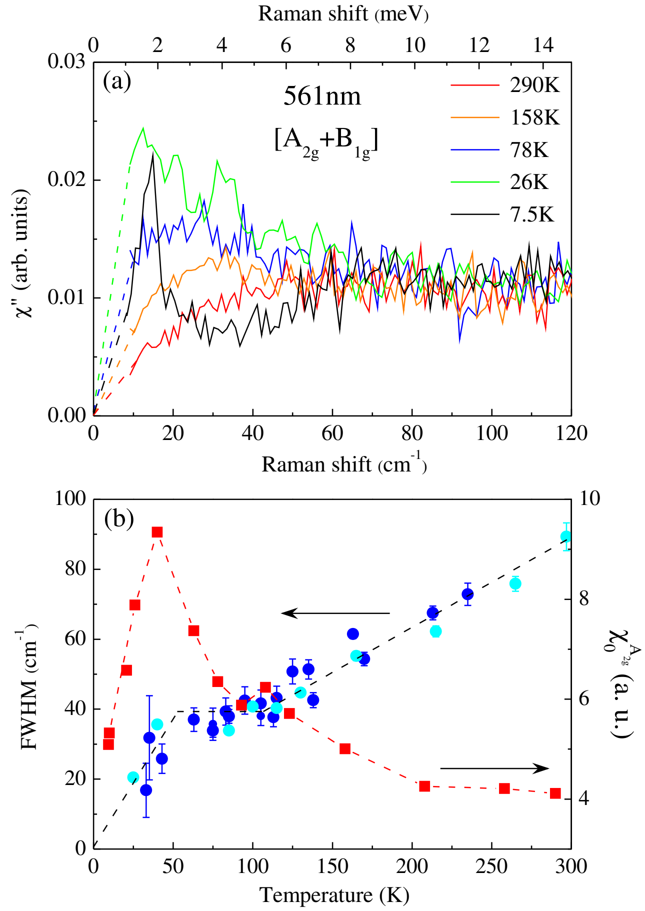

FIG. 3 (color online). (a) Raman susceptibility in the $A_{2g} + B_{1g}$ symmetry from 7.5 to 300 K. (b) Left axis: full width of the quasielastic response (cf. text) (blue filled circles) versus temperature. Right axis: Raman static susceptibility $\chi_0^{A_{2g}}$ (red filled squares) versus temperature (cf. text). Dashed lines are guides for the eyes.

In addition, as shown Fig. 2(b), the peak hardens slightly under a magnetic field up to 10 T [70], which is qualitatively consistent with the magnetic field dependence of the neutron resonance [72]. 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 $Q_0 = (0, 0, 1)$. 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. Fig. 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 [33] as well as from the comparison between Angle-resolved photoemission spectroscopy data at the Γ and the Z points [30–32,73,74]. 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 **Q** vectors.

Interestingly, the finite Raman response in the A_{2g} symmetry is not limited to the HO state. Indeed, as reported by Cooper *et al.* [75], already at 300 K, URu_2Si_2 exhibits a A_{2g} quasielastic peak (QEP) with an overdamped Lorentzian line shape. As shown Fig. 3(a), the quasielastic contribution sharpens with decreasing temperature before collapsing in the HO state. The FWHM, extracted from a simple relaxation model [75], is reported in Fig. 3(b). After a Korringa-like linear temperature dependence down to ~100 K, it exhibits a plateaulike behavior between ~100 and ~50 K before decreasing 3 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 [76], the A_{2g} static susceptibility $\chi_0^{A_{2g}}$ can be extracted as $\int \chi''_{A_{2g}}(\omega)/\omega d\omega$ with the integration spanning from 8 to 100 cm^{-1}, above which all spectra are on top of each other. As shown in Fig. 3(b), the A_{2g} static susceptibility exhibits a temperature dependence very reminiscent of the dc magnetic susceptibility along the c axis [77], 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 [78]. A similar Raman quasielastic response has already been discussed in the context of the $4f$ and $5f$ system, where it was attributed to either spin fluctuations or localized CEF excitations like in UBe_{13} [79]. In URu_2Si_2, 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 an A_{2g} symmetry, with different ground states and with an even or odd number of localized electrons [80].

Following this interpretation, a global and simple scenario, reminiscent of the results obtained across the metal-insulator transition of the skutterudite $PrRu_4P_{12}$ [82], can be given. The A_{2g} QEP as well as the sharp A_{2g} peak in the HO state are due to CEF excitations while the gap Δ_G is associated with the gapped itinerant electron-hole continuum. Because CEF excitations are coupled to itinerant carriers, the QEP is strongly damped and quasielastic above T_0. At the HO transition, A_{2g} decaying channels are quenched at low energy due to the opening of the A_{2g} gap in the electronic continuum and consequently, the CEF excitation becomes long lived. By the same process the CEF excitation becomes gapped (inelastic) because of the associated loss of hybridization with the delocalized quasiparticle continuum in the HO state. In this picture, the dual character of the phase transition in URu_2Si_2 appears naturally, with the gap Δ_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 with their local character. Within this scenario, a close relationship between the signatures of the itinerant and localized character of the HO state can be made, through the similarity of their A_{2g} symmetry.

Now, we discuss alternative explanations for each signature, the QEP and the sharp peak, independently. We inferred that the A_{2g} gap is a gapped electron-hole excitation continuum. As already noted above, the opening of this gap will help any excitation inside the gap to become long lived and sharp. In this context, the peculiar A_{2g} symmetry may emerge from local current loop excitations, which could give rise to the A_{2g} peak in the HO state. Indeed, similar anomalous orbital motion of charge carriers has been shown to have the A_{2g} symmetry [54] based on a Raman experiment in the insulating cuprates [55]. This hypothesis has also been recently brought up for URu_2Si_2 [3,69]. As for the QEP observed above T_0, in a pure itinerant picture perspective, a Drude-like Raman response of electron-hole excitations may explain it. We would need then to account for the strong spectral weight in the A_{2g} symmetry. This would certainly require us to go beyond the effective mass approximation [58] by taking into account the resonant contributions for the Raman vertex calculation [54,83,84].

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 ~55 cm^{-1}, as signatures of the hidden-order state in URu_2Si_2. Additionally, by determining accurate temperature and magnetic field dependencies of the sharp Raman peak, we have shown that the sharp peak matches the neutron resonance at Q_0. This brings new and robust evidence of the Brillouin zone folding along the c axis upon entering the HO state, consistent with a switch from a body-centered-tetragonal to a simple-tetragonal structure. Theoretical investigations, most probably accounting for both the local and the itinerant character of the quasiparticle in URu_2Si_2, are necessary to reach a global scenario of these Raman signatures of peculiar A_{2g} symmetry and to draw conclusions about the associated HO order parameter.

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[1] T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. **55**, 2727 (1985).
[2] J. A. Mydosh and P. M. Oppeneer, Rev. Mod. Phys. **83**, 1301 (2011).
[3] J. Mydosh and P. Oppeneer, Philos. Mag. **94**, 3642 (2014).
[4] W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B **62**, 171 (1986).
[5] K. Behnia, R. Bel, Y. Kasahara, Y. Nakajima, H. Jin, H. Aubin, K. Izawa, Y. Matsuda, J. Flouquet, Y. Haga, Y. Onuki, and P. Lejay, Phys. Rev. Lett. **94**, 156405 (2005).
[6] J. Schoenes, C. Schonenberger, J. J. M. Franse, and A. A. Menovsky, Phys. Rev. B **35**, 5375 (1987).
[7] A. de Visser, F. E. Kayzel, A. A. Menovsky, J. J. M. Franse, J. van den Berg, and G. J. Nieuwenhuys, Phys. Rev. B **34**, 8168 (1986).
[8] C. Broholm, H. Lin, P. T. Matthews, T. E. Mason, W. J. L. Buyers, M. F. Collins, A. A. Menovsky, J. A. Mydosh, and J. K. Kjems, Phys. Rev. B **43**, 12809 (1991).
[9] H. Amitsuka, K. Matsuda, I. Kawasaki, K. Tenya, M. Yokoyama, C. Sekine, N. Tateiwa, T. Kobayashi, S. Kawarazaki, and H. Yoshizawa, J. Magn. Magn. Mater. **310**, 214 (2007).
[10] N. P. Butch, J. R. Jeffries, S. Chi, J. B. Leão, J. W. Lynn, and M. B. Maple, Phys. Rev. B **82**, 060408 (2010).

[11] E. Hassinger, G. Knebel, K. Izawa, P. Lejay, B. Salce, and J. Flouquet, Phys. Rev. B **77**, 115117 (2008).
[12] H. Kusunose and H. Harima, J. Phys. Soc. Jpn. **80**, 084702 (2011).
[13] E. Ressouche, R. Ballou, F. Bourdarot, D. Aoki, V. Simonet, M. T. Fernandez-Diaz, A. Stunault, and J. Flouquet, Phys. Rev. Lett. **109**, 067202 (2012).
[14] K. Haule and G. Kotliar, Nat. Phys. **5**, 796 (2009).
[15] H. Ikeda, M.-T. Suzuki, R. Arita, T. Takimoto, T. Shibauchi, and Y. Matsuda, Nat. Phys. **8**, 528 (2012).
[16] J. G. Rau and H.-Y. Kee, Phys. Rev. B **85**, 245112 (2012).
[17] P. Chandra, P. Coleman, J. A. Mydosh, and V. Tripathi, Nature (London) **417**, 831 (2002).
[18] S. Fujimoto, Phys. Rev. Lett. **106**, 196407 (2011).
[19] P. S. Riseborough, B. Coqblin, and S. G. Magalhães, Phys. Rev. B **85**, 165116 (2012).
[20] T. Das, Phys. Rev. B **89**, 045135 (2014).
[21] C. Pépin, M. R. Norman, S. Burdin, and A. Ferraz, Phys. Rev. Lett. **106**, 106601 (2011).
[22] C. Thomas, S. Burdin, C. Pépin, and A. Ferraz, Phys. Rev. B **87**, 014422 (2013).
[23] S. Elgazzar, J. Rusz, M. Amft, P. M. Oppeneer, and J. A. Mydosh, Nat. Mater. **8**, 337 (2009).
[24] P. Chandra, P. Coleman, and R. Flint, Nature (London) **493**, 621 (2013).
[25] F. Bourdarot, E. Hassinger, S. Raymond, D. Aoki, V. Taufour, L.-P. Regnault, and J. Flouquet, J. Phys. Soc. Jpn. **79**, 064719 (2010).
[26] C. R. Wiebe, J. A. Janik, G. J. MacDougall, G. M. Luke, J. D. Garrett, H. D. Zhou, Y.-J. Jo, L. Balicas, Y. Qiu, J. R. D. Copley, Z. Yamani, and W. J. L. Buyers, Nat. Phys. **3**, 96 (2007).
[27] A. Villaume, F. Bourdarot, E. Hassinger, S. Raymond, V. Taufour, D. Aoki, and J. Flouquet, Phys. Rev. B **78**, 012504 (2008).
[28] D. A. Bonn, J. D. Garrett, and T. Timusk, Phys. Rev. Lett. **61**, 1305 (1988).
[29] A. F. Santander-Syro, M. Klein, F. L. Boariu, A. Nuber, P. Lejay, and F. Reinert, Nat. Phys. **5**, 637 (2009).
[30] R. Yoshida, Y. Nakamura, M. Fukui, Y. Haga, E. Yamamoto, Y. Onuki, M. Okawa, S. Shin, M. Hirai, Y. Muraoka, and T. Yokoya, Phys. Rev. B **82**, 205108 (2010).
[31] F. L. Boariu, C. Bareille, H. Schwab, A. Nuber, P. Lejay, T. Durakiewicz, F. Reinert, and A. F. Santander-Syro, Phys. Rev. Lett. **110**, 156404 (2013).
[32] J.-Q. Meng, P. M. Oppeneer, J. A. Mydosh, P. S. Riseborough, K. Gofryk, J. J. Joyce, E. D. Bauer, Y. Li, and T. Durakiewicz, Phys. Rev. Lett. **111**, 127002 (2013).
[33] E. Hassinger, G. Knebel, T. D. Matsuda, D. Aoki, V. Taufour, and J. Flouquet, Phys. Rev. Lett. **105**, 216409 (2010).
[34] R. Okazaki, T. Shibauchi, H. J. Shi, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, and Y. Matsuda, Science **331**, 439 (2011).
[35] S. Tonegawa, K. Hashimoto, K. Ikada, Y.-H. Lin, H. Shishido, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, Y. Matsuda, and T. Shibauchi, Phys. Rev. Lett. **109**, 036401 (2012).
[36] S. Tonegawa, S. Kasahara, T. Fukuda, K. Sugimoto, N. Yasuda, Y. Tsuruhara, D. Watanabe, Y. Mizukami, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, Y. Matsuda, and T. Shibauchi, Nat. Commun. **5**, 5188 (2014).
[37] S. Kambe, Y. Tokunaga, H. Sakai, T. D. Matsuda, Y. Haga, Z. Fisk, and R. E. Walstedt, Phys. Rev. Lett. **110**, 246406 (2013).
[38] H. Amitsuka, Workshop on Hidden Order, Superconductivity and Magnetism in URu_2Si_2, Leiden, 2013 (unpublished).
[39] P. M. Oppeneer, J. Rusz, S. Elgazzar, M.-T. Suzuki, T. Durakiewicz, and J. A. Mydosh, Phys. Rev. B **82**, 205103 (2010).
[40] D. Aoki, F. Bourdarot, E. Hassinger, G. Knebel, A. Miyake, S. Raymond, V. Taufour, and J. Flouquet, J. Phys. Condens. Matter **22**, 164205 (2010).
[41] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405, which includes Refs. [42–45], for details about the laser heating estimation.
[42] R. Hackl, R. Kaiser, and S. Schicktanz, J. Phys. C **16**, 1729 (1983).
[43] A. A. Maksimov, A. V. Puchkov, I. I. Tartakovskii, V. B. Timofeev, D. Reznik, and M. V. Klein, Solid State Commun. **81**, 407 (1992).
[44] A. Mialitsin, Ph.D. thesis, Rutgers University, New Brunswick, 2010.
[45] F. G. Aliev, V. Kovachik, V. V. Moshchalkov, V. V. Pryadun, N. E. Alekseevskii, A. V. Mitin, N. Agrait, S. Vieira, and R. Villar, J. Low Temp. Phys. **85**, 359 (1991).
[46] W. Hayes and R. Loudon, *Scattering of Light by Crystals* (Dover Publications, New York, 2004).
[47] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405 for details about the extraction of the pure A_{2g} symmetry.
[48] We are aware of a parallel Raman study by the group of G. Blumberg interpreted in the context of the theory developed by Haule and Kotliar [14]. Similar results were obtained in the A_{2g} symmetry with a small additional A_{1g} peak at 14 cm^{-1} [49,50].
[49] H.-H. Kung, R. E. Baumbach, E. D. Bauer, V. K. Thorsmølle, W.-L. Zhang, K. Haule, J. A. Mydosh, and G. Blumberg, arXiv:1410.6398.
[50] G. Blumberg, Polarized Electronic Raman Scattering Study of the Heavy Fermion Compound URu_2Si_2, Hidden Order, Superconductivity, and Magnetism in URu_2Si_2, Lorentz Center, Leiden, 2013 (unpublished).
[51] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405 for details about the measurements of the Raman response in other symmetries.
[52] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405, which includes Ref. [53], for details about the A_{2g} symmetry.
[53] H. Harima, K. Miyake, and J. Flouquet, J. Phys. Soc. Jpn. **79**, 033705 (2010).
[54] D. V. Khveshchenko and P. B. Wiegmann, Phys. Rev. Lett. **73**, 500 (1994).
[55] R. Liu, D. Salamon, M. V. Klein, S. L. Cooper, W. C. Lee, S.-W. Cheong, and D. M. Ginsberg, Phys. Rev. Lett. **71**, 3709 (1993).

[56] S. Yoon, M. Rübhausen, S. L. Cooper, K. H. Kim, and S. W. Cheong, Phys. Rev. Lett. **85**, 3297 (2000).
[57] P. E. Sulewski, P. A. Fleury, K. B. Lyons, and S.-W. Cheong, Phys. Rev. Lett. **67**, 3864 (1991).
[58] T. P. Devereaux and R. Hackl, Rev. Mod. Phys. **79**, 175 (2007).
[59] W. T. Guo, Z. G. Chen, T. J. Williams, J. D. Garrett, G. M. Luke, and N. L. Wang, Phys. Rev. B **85**, 195105 (2012).
[60] U. Nagel, T. Uleksin, T. Room, R. P. S. M. Lobo, P. Lejay, C. C. Homes, J. S. Hall, A. W. Kinross, S. K. Purdy, T. Munsie, T. J. Williams, G. M. Luke, and T. Timusk, Proc. Natl. Acad. Sci. U.S.A. **109**, 19161 (2012).
[61] J. S. Hall, U. Nagel, T. Uleksin, T. Room, T. Williams, G. Luke, and T. Timusk, Phys. Rev. B **86**, 035132 (2012).
[62] M. W. McElfresh, J. D. Thompson, J. O. Willis, M. B. Maple, T. Kohara, and M. S. Torikachvili, Phys. Rev. B **35**, 43 (1987).
[63] M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, M. S. Torikachvili, M. W. McElfresh, and J. D. Thompson, Phys. Rev. Lett. **56**, 185 (1986).
[64] R. A. Fisher, S. Kim, Y. Wu, N. E. Phillips, M. W. McElfresh, M. S. Torikachvili, and M. B. Maple, Physica (Amsterdam) **163B**, 419 (1990).
[65] P. Aynajian, E. H. da Silva Neto, C. V. Parker, Y. Huang, A. Pasupathy, J. Mydosh, and A. Yazdani, Proc. Natl. Acad. Sci. U.S.A. **107**, 10383 (2010).
[66] A. R. Schmidt, M. H. Hamidian, P. Wahl, F. Meier, A. V. Balatsky, J. D. Garrett, T. J. Williams, G. M. Luke, and J. C. Davis, Nature (London) **465**, 570 (2010).
[67] F. Bourdarot, S. Raymond, and L.-P. Regnault, Philos. Mag. **94**, 3702 (2014).
[68] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405 for details about the temperature dependence of the spectral weight of the A_{2g} features.
[69] P. M. Oppeneer, S. Elgazzar, J. Rusz, Q. Feng, T. Durakiewicz, and J. A. Mydosh, Phys. Rev. B **84**, 241102 (2011).
[70] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405, which includes Ref. [71], for details about the magnetic field dependence of the Raman peak.
[71] S. A. M. Mentink, T. E. Mason, S. Sullow, G. J. Nieuwenhuys, A. A. Menovsky, J. A. Mydosh, and J. A. A. J. Perenboom, Phys. Rev. B **53**, R6014 (1996).
[72] F. Bourdarot, B. Fak, K. Habicht, and K. Prokes, J. Magn. Magn. Mater. **272–276**, E31 (2004).
[73] R. Yoshida, K. Tsubota, T. Ishiga, M. Sunagawa, J. Sonoyama, D. Aoki, J. Flouquet, T. Wakita, Y. Muraoka, and T. Yokoya, Sci. Rep. **3**, 02750 (2013).
[74] C. Bareille, F. L. Boariu, H. Schwab, P. Lejay, F. Reinert, and A. F. Santander-Syro, Nat. Commun. **5**, 4326 (2014).
[75] S. L. Cooper, M. V. Klein, Z. Fisk, and J. L. Smith, Phys. Rev. B **34**, 6235 (1986).
[76] Y. Gallais, R. M. Fernandes, I. Paul, L. Chauvière, Y.-X. Yang, M.-A. Measson, M. Cazayous, A. Sacuto, D. Colson, and A. Forget, Phys. Rev. Lett. **111**, 267001 (2013).
[77] T. T. M. Palstra, A. A. Menovsky, G. J. Nieuwenhuys, and J. A. Mydosh, J. Magn. Magn. Mater. **54–57**, 435 (1986).
[78] P. Santini and G. Amoretti, Phys. Rev. Lett. **73**, 1027 (1994).
[79] S. L. Cooper, M. V. Klein, Z. Fisk, and J. L. Smith, Phys. Rev. B **37**, 2251 (1988).
[80] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.113.266405, which includes Ref. [81], for details about the CEF excitation selection rules.
[81] G. F. Koster, *Properties of the Thirty-Two Point Groups* (MIT Press, Cambridge, MA, 1963).
[82] K. Iwasa, L. Hao, K. Kuwahara, M. Kohgi, S. R. Saha, H. Sugawara, Y. Aoki, H. Sato, T. Tayama, and T. Sakakibara, Phys. Rev. B **72**, 024414 (2005).
[83] A. M. Shvaika, O. Vorobyov, J. K. Freericks, and T. P. Devereaux, Phys. Rev. B **71**, 045120 (2005).
[84] Indeed, in the effective mass approximation, the electron-hole excitation continuum is not A_{2g} active except by considering an unlikely electronic band structure on which $\partial^2 E/\partial k_x \partial k_y \neq \partial^2 E/\partial k_y \partial k_x$ in some k-space region of the Fermi surface.