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Search for Multipolar Instability in URu$_2$Si$_2$ Studied by Ultrasonic Measurements under Pulsed Magnetic Field

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The elastic properties of URu$_2$Si$_2$ in the high-magnetic field region above 40 T, over a wide temperature range from 1.5 to 120 K, were systematically investigated by means of high-frequency ultrasonic measurements. The investigation was performed at high magnetic fields to better investigate the innate bare 5$f$-electron properties, since the unidentified electronic thermodynamic phase of unknown origin, so called ‘hidden order’(HO) phase and associated hybridization of conduction and f-electron ($c$-$f$ hybridization) are suppressed at high magnetic fields. From the three different transverse modes we find contrasting results; both the $\Gamma_4(B_{2g})$ and $\Gamma_5(E_{x})$ symmetry modes $C_{66}$ and $C_{44}$ show elastic softening that is enhanced above 30 T, while the characteristic softening of the $\Gamma_3(B_{1g})$ symmetry mode $(C_{11} - C_{12})/2$ is suppressed in high magnetic fields. These results underscore the presence of a hybridization-driven $\Gamma_3(B_{1g})$ lattice instability in URu$_2$Si$_2$. However, the results from this work cannot be explained by using existing crystalline-electric field (CEF) schemes applied to the quadrupolar susceptibility in a local 5$f^2$ configuration. Instead, we present an analysis based on a band Jahn-Teller effect.

I. INTRODUCTION

The heavy-fermion unconventional superconductor URu$_2$Si$_2$ undergoes an enigmatic phase transition at $T_0=17.5$ K to the so called ‘hidden order (HO)’ phase$^{1-3}$, whose order parameter still remains unsolved$^4$. This compound has a body-centered-tetragonal (bct) ThCr$_2$Si$_2$-type crystal structure (space group No. 139, $I4/mmm; D_{4h}^{17}$). Recently, several experimental findings regarding a possible symmetry lowering of the electron and/or lattice system in the HO phase have been reported: including results of magnetic torque$^5$, synchrotron x-ray$^6$, Raman scattering$^7$, and elastoresistance measurements$^8$. However, the proposed broken symmetries conflict with each other. Many theories have been proposed to explain the HO phase; e.g., higher multipolar order from rank 3 to $5^9$-13, hystatic order$^{14}$, spin inter-orbital density wave$^{15}$, and dynamic antiferromagnetic moment fluctuations.$^{16}$ There is no comprehensive interpretation, which can explain all of the experimental observations.

With high magnetic fields applied along the [001] axis at low temperatures, URu$_2$Si$_2$ undergoes three metamagnetic transitions in the range between 35 and 39 T which are followed by a collapse of the HO phase.$^{17}$ In Fig. 1(b), we show a temperature-magnetic-field phase diagram of URu$_2$Si$_2$ for $H \parallel [001]$, which is constructed from the data of the present work and previous magnetization measurements.$^{18}$ First, the HO phase is suppressed at 35 T, followed by a cascade of transitions, where the spin-density wave with a propagation wave vector $\mathbf{k} = (0.6, 0, 0)$ is established in the intermediate phase.$^{19}$ Finally, the system enters the polarized paramagnetic (PPM) regime in the high-magnetic-field region above 40 T.$^{17}$ URu$_2$Si$_2$ also exhibits a strong hybridization between conduction and 5$f$ electrons ($c$-$f$ hybridization) below $T^* \sim 50$ K in low magnetic fields. This $c$-$f$ hybridization is also suppressed in association with the collapse of HO under high magnetic fields above 40 T for $H \parallel [001]^{18}$. Beyond 40 T, the electronic ground state of URu$_2$Si$_2$ changes from delocalized to a more localized 5$f$-electron regime.$^{18}$ Understanding the dual nature of the uranium 5$f$ electron that are neither fully localized nor itinerant will likely provide insight in the origin of the HO. No theory has been developed which fully describes both the hybridization effect and the localized electron degrees of freedom. There are two approaches to overcome these issues; either starting from the itinerant electron system (strong-coupling limit) or from the localized electron system (weak-coupling limit). A constraint is that the ‘symmetry’ of the order parameter itself must be the same, both in the itinerant and localized components of the 5$f$-electrons as they both play a role in developing the HO. Ultrasonic measurement is one of the sensitive probing techniques to investigate both itinerant band instabilities, such as the band-Jahn Teller effect, and the local anisotropic charge distribution, such as that found in multipolar ordering. Therefore the present work is aimed at obtaining better information on the dual nature of the 5$f$-electron states in URu$_2$Si$_2$. Our recent investigation of the elastic constant $(C_{11} - C_{12})/2$ of URu$_2$Si$_2$ under pulsed-magnetic fields strongly sug-
gests that the hybridized electronic state possesses an orthorhombic \((x^2 - y^2)\) lattice instability with \(\Gamma_3(B_{1g})\) symmetry. The origin of the lattice instability is considered to be either a potential deformation due to the Jahn-Teller effect of hybridized bands or a simple CEF effect of uranium’s 5f electrons; however, the origin of the \(\Gamma_3(B_{1g})\) lattice instability and its relation to the HO parameter are still open questions. In order to verify that the system does not exhibit a lattice instability for other symmetries, and to examine the theoretical predictions of CEF ground-state schemes for the high magnetic fields and related higher-multipolar order parameter scenarios for the HO phase as well, we study elastic responses of the other symmetry-breaking strains. In the present work, we report on the responses of \(C_{44}\) with \(\Gamma_5(E_g)\) symmetry and \(C_{66}\) with \(\Gamma_4(B_{2g})\) symmetry under high magnetic field, and compare these results with the previously reported \((C_{11} - C_{12})/2\) with \(\Gamma_3(B_{1g})\) symmetry.

**II. EXPERIMENTAL DETAILS**

We investigated two single crystals of \(\text{URu}_2\text{Si}_2\) grown using the Czochralski technique by a tetra-arc furnace at UC San Diego (sample \#1) and CEA Grenoble (sample \#2). For sample \#1, the dimensions are \(3.8 \times 1.8 \times 1.2\) mm\(^3\) with parallel [110] facets as grown. Residual resistivity ratio (RRR) \(\sim 10\) was used for \((C_{11} - C_{12})/2, C_{44},\) and \(C_{66}\) measurements. For sample \#2, \(3.38 \times 1.67 \times 1.5\) mm\(^3\) with parallel [100] facets, annealed in vacuum, RRR \(\sim 29\) is used for \(C_{11}, C_{44},\) and \(C_{66}\). Note, there is no obvious sample dependence in the magnetic field dependence of \(C_{44}\) for both samples, except for a difference in the signal-to-noise ratio. The sample surfaces were well polished and characterized by x-ray Laue diffraction to check the characteristic symmetries of the facets. Ultrasound was generated and detected by using LiNbO\(_3\) transducers with a thickness of 40-100 \(\mu\)m, which were fixed on the sample surfaces with RTV silicone or superglue. We used pulsed magnetic fields up to 68 T with pulse duration of about 150 ms at the Dresden High Magnetic Field Laboratory. The sound-velocity measurements were performed using a conventional phase comparative method using a digital storage oscilloscope. Ultrasound induces both linear strain and a rotation field (similar to Raman modes). A summary with \(D_{4h}\) point group are shown in Table I) in the solid, which behave as conjugate fields for the electric quadrupole or electric hexadecapole moments. These multipolar responses can be observed as a sound-velocity change and ultrasonic attenuation via electron-phonon interaction. The sound velocity \(v_\text{ij}\) is converted to the elastic constant \(C_{ij}\) by using the formula: \(C_{ij} = \rho v_{ij}^2\). Here, \(\rho = 10.01\) (g/cm\(^3\)) is the density of \(\text{URu}_2\text{Si}_2\).

**III. RESULTS**

In Fig. 1, we show the magnetic-field dependence of the following elastic constants \(C_{11}/10, (C_{11} - C_{12})/2, C_{33}, C_{44},\) and \(C_{66}\) at fixed temperatures of (a) 22-23 K and (c) 1.5 K for \(H \parallel [001]\) which are measured with ultrasonic frequencies of 75 MHz for \(C_{11}, 159.5\) MHz for \((C_{11} - C_{12})/2, 78.7\) MHz for \(C_{33}, 164\) MHz for \(C_{44},\) and 166 MHz for \(C_{66}.\) At 22-23 K, the elastic constants \(C_{33}, C_{44},\) and \(C_{66}\) decrease with increasing magnetic-field through the cross-over region of the \(c\)-\(f\) hybridization (below 30 T) and toward the polar-paramagnetic region (above 45 T), while \(C_{11}\) and \((C_{11} - C_{12})/2,\) both related to the \(\Gamma_3\)-symmetry response, increase above 35 T.

The magnetic field-temperature \((H-T)\) phase diagram is displayed in Fig. 1(b) for comparison, where the hor-
TABLE I. Symmetry, symmetrized strain and rotation, and multipole for different elastic constants.

| Symmetry (D4h group) | Strain and Rotation | Multipole | Elastic Constant |
|----------------------|---------------------|-----------|-----------------|
| Γ1(A1g)              | εxx, εyy            |           | C_{33} = -3C_{20} + 4C_u + 4C_{13} |
| Γ₁Γ₃(A₄g+B₁g)       | εxx = ϵ₁/3 - ϵ₁/2√3 |           | C₁₁ = 3C₁₈ - C_u + (C₁₁ - C₁₂)/2 - 2C₁₃ |
| Γ₃(B₁g)              | ϵ_ux = ϵ_x - ϵ_y   | O_x = √3(J_{2}^x - J_{2}^y)/2 | C_v = (C₁₁ - C₁₂)/2 |
| Γ₄(B₁g)              | ϵ_yx               | O_yx = √3(J_{2}^x - J_{2}^y)/2 | C_{66} |
| Γ₅(E₈)              | ϵ_yx               | O_yz = √3(J_{2}^z - J_{2}^y)/2 | C₄₄ |
| Γ₂(A₂g)              | ω_{yz}             |           | C_{66}, C_v |

One may consider the possibility of the magnetostriiction on the sound-velocity change, since the magnetic field change of the elastic constant looks very similar to the magnetization and magnetostriction change in pulsed-magnetic fields. However, by applying magnetic field along the [001] axis of URu₂Si₂, the c-axis length decreases only by ΔL/L_c ∼ 10^{-4} at 45 T and 1.5 K, and the a axis expands by the same order of magnitude due to the Poisson effect. In the present case, such an effect would mainly lead to enhance softening of the longitudinal C_{11} mode in the vicinity of the cascade transitions. C_{11} includes a contribution from the bulk modulus (volume strain). Based on the modified Ehrenfest equation, the estimated contribution of the magnetostriction to the sound-velocity change is Δv_{ij}/v_{ij} ∼ 10^{-4}, which is less than only 5% of the total velocity change ∼ 2 × 10^{-3} of the transverse ultrasonic modes C_{44}, C_{66}, and (C_{11} - C_{12})/2. The hardening of (C_{11} - C_{12})/2 at the collapse of the HO phase has a tendency opposite to the magnetostriction along [100], since it is equivalent to 1/√2 of the magnetostriction along [110]. Consequently, the elastic response originates from the drastic change of the transverse acoustic phonon dispersion due to strong coupling to the 5f-electrons.

In Figs. 2 (d) and (g), we show the isotherms of the modes C_{44} and C_{66} as a function of increasing and decreasing magnetic field applied along [001]. For comparison, our previous results21 for the (C_{11} - C_{12})/2 are also shown in Fig. 2 (a). From these data, we determined the elastic constants as a function of temperature in fixed magnetic field, shown in Figs. 2 (c), (f), and (i). The middle column combines 3-dimensional plots of the elastic constants vs. temperature and magnetic field H || c for the three different symmetries; (b) (C_{11} - C_{12})/2 for the Γ₃(B₁g), (c) C_{66} for the Γ₅(E₈), and (h) C_{11} for the Γ₄(B₁g) of the D₄h point group symmetry. The bottom of each cubic box shows the H - T phase diagram. The blue-white-red color gradation indicates the relative stiffness of each ultrasonic mode, stiffer in blue and softer in red. In the soft-mode regions, the system may indicate lattice instabilities of the corresponding symmetry. For example, for the (C_{11} - C_{12})/2 mode, the corresponding Γ₃(B₁g) lattice instability is enhanced in the low-temperature and low-magnetic-field region, where strong c-f hybridization occurs, and suppressed at high temperatures and high magnetic fields. The Γ₄(B₁g) and Γ₅(E₈) modes show the opposite tendency. Such a clear difference in the three transverse modes indicates the presence of the Γ₃(B₁g) lattice instability in the HO phase, and in the strong c-f hybridization region at low-magnetic fields in URu₂Si₂.

IV. DISCUSSION

A. Band Jahn-Teller Model: (Delocalized 5f-electron state)

In Figs. 3 (a), (b), and (c) the normalized elastic constants vs. temperature at various magnetic fields are shown for (a) Γ₃(B₁g): (C_{11} - C_{12})/2, (b) Γ₄(B₁g): C_{66}, and (c) Γ₅(E₈): C_{44}, with the phonon background subtracted. For simplicity, we made phenomenological fits to the elastic constants of ThRu₂Si₂ measured from 300 K to 1.5 K in zero magnetic field as the phonon background shown as the dotted lines in Figs. 2(c), (f), and (i). A similar subtraction was also performed in our previous work.24 First, we analyzed the softening of (C_{11} - C_{12})/2 by using the phenomenological theory of the band-Jahn-Teller (BJT) effect assuming a rigid degenerate two-band state.25 The solid lines in Fig. 3(a) were calculated from
the following equation:

\[
\frac{(C_{11} - C_{12})}{2} = C_{ph} - 2d^2N_0\left\{1 - e^{-(E_F - E_0)/k_B T}\right\}.
\]  

(1)

Here, \(C_{ph}\) is the phonon background [as shown in Fig. 2(c)], \(d\) is a deformation-potential coupling constant, \(N_0\) is the density of states at the Fermi energy \(E_F\), and \(E_0\) is the energy at the bottom of the conduction band. The term \(2d^2N_0\) is set to be temperature independent. Figure 4 shows the magnetic-field dependence of the fit parameters \((2d^2N_0)\) and \((E_F - E_0)\). We obtain \(E_F - E_0 = 43\) K at 0 T and \(E_F - E_0 = 28\) K at 35 T. The value of \(2d^2N_0 = 0.071 \times 10^{10}\) J m\(^{-3}\) at 0 T gradually decreases with increasing magnetic field, which is consistent with the reduction of \(c\)-\(f\) hybridization under magnetic field, where causes a weakening of the deformation-potential coupling. The parameters obtained below 30 T are comparable to the values reported for the typical band-Jahn-Teller system LaAg\(_{1-x}\)In\(_x\), where the compounds with \(x = 0\) and \(x = 0.11\) do not show structural transition but exhibit a softening in \((C_{11} - C_{12})/2\) due to \(\Gamma_3\) lattice instability. Here for URu\(_2\)Si\(_2\), the obtained deformation-potential coupling energy is less than 1/5 of the value of LaAg \((x = 0, 2d^2N_0 = 0.375 \times 10^{10}\) J m\(^{-3}\)), suggesting that the effect is too weak to induce a structural phase transition. Above 40 T, the gap and the fitting error bar drastically increase, which appears to be extrinsic and shows the limitations of this theory.
FIG. 3. Temperature dependence of the normalized elastic constants of (a) $\Gamma_3$: $(C_{11} - C_{12})/2$, (b) $\Gamma_4$: $C_{66}$, and (c) $\Gamma_5$: $C_{44}$ at various magnetic fields $H \parallel [001]$, where the phonon background is subtracted. Solid lines in (a) are calculated by using the band-Jahn-Teller model (see text), and the solid lines in (b) and (c) are visual aids. Calculated uniform quadrupolar susceptibilities of (d) $\Gamma_3$: $O_{2}^{2}$, (e) $\Gamma_4$: $O_{x\gamma}$, and (f) $\Gamma_5$: $O_{\gamma z}$ for different CEF schemes (see Table II) at 0 and 60 T.

FIG. 4. Magnetic field dependence of the BJF fit parameters for $(C_{11} - C_{12})/2$: The gap between the two levels $E_2 - E_0$ (red, left axis) and $2\delta^2 N_0$ (blue, right axis, see text for details). The dotted curves are visual aids.

B. Crystalline Electric Field Models: (Localized 5$f$-electron state)

We compare elastic responses obtained in the high-magnetic field region with uniform quadrupolar susceptibilities, which are calculated by using CEF schemes in the $5f^2$ configuration, proposed thus far. We have considered a variety of CEF level schemes, especially based on the U$^{4+}(5f^2)$ ionization and non-Kramers $3H_4$ ($J=4$). Hund’s rule ground-state multiplet; a non-Kramers configuration can easily reproduce the reported anisotropic magnetization along the $a$ and $c$ axis of this compound.$^{27}$ The details of the four CEF schemes considered are listed in Table II. It should be noted that the present CEF scheme 1 has two lowest-lying U-5$f$ singlets; $\Gamma_1^{(1)} = \alpha(|4\pm4\rangle) - \beta|0\rangle$ and $\Gamma_2 = i(|4\pm4\rangle)/\sqrt{2}$, which is identical to the level scheme in the theoretical models originally predicting the $A_{2g}$-type hexadecapolar order as the order parameter of the HO state, which have been proposed by Haule and Kotliar$^{10}$, or by Kusunose and Harima.$^{9}$

The present analysis allows us to qualitatively compare the measured normalized elastic constants [Figs. 3 (a), (b), and (c)] with the calculated quadrupolar susceptibilities as shown in Figs. 3 (d), (e), and (f) (Appendix A). At first glance, none of these CEF schemes successfully reproduces experimental observations. A detailed analysis follows below:

(i) $(C_{11} - C_{12})/2$, $\Gamma_3(B_{1g})$ symmetry: Only the Scheme 1 and Scheme 3 reproduce the temperature and magnetic field dependence of $(C_{11} - C_{12})/2$. Scheme 2 shows a steep softening below 20 K at $H = 0$ T and Scheme 4 shows a broad minimum at around 50 K at $H = 0$ and 60 T, inconsistent with the experimental data at low and high magnetic fields.

(ii) $C_{66}$, $\Gamma_4(B_{2g})$ symmetry:
Only Scheme 3 roughly reproduces the temperature dependence of $C_{66}$ at high magnetic field. However, the expected softening at 0 T in Scheme 3 is not seen in the experimental data. Scheme 2 again shows a steep softening at $H = 0$ below 20 K and Scheme 1 and Scheme 4 show local minima and upturns; inconsistent with the experiment.

(iii) $C_{44}$, $\Gamma_5(E_g)$ symmetry:

Only Scheme 4 reproduces the softening at 60 T, but its magnetic-field dependence shows an opposite tendency (no softening in magnetic field). All the other schemes (1-3) show neither low-temperature softening nor enhancement under magnetic fields.

Therefore, based on this logic, we conclude that the present experimental results cannot be fully explained by CEF schemes in the 5$f^2$ configuration. Note that other CEF schemes have been tested and also resulted in poor agreement with the experimental data. For example, $\Gamma_1(45 \text{ K})$-$\Gamma_2(51 \text{ K})$-$\Gamma_2(100 \text{ K})$ [32], which cannot be explained by tetragonal CEF since this theory is considering many-body effects, $\Gamma_1(42 \text{ K})$-$\Gamma_2(170 \text{ K})$ [27], and $\Gamma_4$-$\Gamma_5(44 \text{ K})$-$\Gamma_2(112)$ [30].

Here, we discuss conditions for the application of the CEF schemes to URu$_2$Si$_2$. As mentioned, the 5$f^2$ non-Kramers multiplet is the best assumption to reproduce the anisotropy in the magnetization. Because $J_z$ has diagonal matrix elements in doublet states and off-diagonal elements between singlet-singlet, doublet-doublet, while $J_x$ and $J_y$ only have off-diagonal elements between singlet-doublet. Thus, if the singlet and doublet states are separated in non-Kramers $J = 4$ CEF states (like Schemes 1 and 2), one can naturally get magnetic anisotropy. Indeed, CEF Schemes 3 and 4, where the singlet and doublet are relatively close ($\leq 300 \text{ K}$), cannot fully reproduce the anisotropic magnetization.

On the other hand, all CEF schemes above are inconsistent with the occurrence of the softening in $C_{44}$ mode, because the corresponding quadrupolar moments of $O_{yz}$ and $O_{zx}$, which has $\Delta J = \pm 1$ transition and always accompany with magnetic moment $J_z$. Thus, it is difficult to find a CEF scheme which satisfies the mutually exclusive features. Therefore, it is even more challenging to find a CEF scheme which balances the competing transitions of $O_{xy}$ with $\Delta J = \pm 2$, and $O_{yz}$ and $O_{zx}$ with $\Delta J = \pm 1$ and also reproduces all elastic constant softening at high magnetic fields, where the present system is not affected by both c-f hybridization and PPM states. Therefore, we need to find an appropriate CEF scheme and/or consider another origin or modulation to reproduce the experimental data.

One possibility is a rotation effect$^{33,34}$. A rotation invariant of the Hamiltonian describing quadrupole-strain interaction will produce a finite modulation of the transverse mode under magnetic field. In the present experiments, the geometry of the $C_{44}$ mode ($k \parallel [100], u \parallel H \parallel [001]$) is the case to consider this effect. This ultrasonic mode induces the strain field $\varepsilon_{xx}$ and also induces the rotation of $\omega_{zz}$, which will couple to the magnetic torque of the total angular momentum $J$. We tried to compute such an effect on CEF Scheme 3 which originally show no softening in $C_{44}$, but the rotation does not reproduce this. CEF Scheme 1, on the other hand, can generate the softening in $C_{44}$ when the rotation effect is considered (not shown). To verify whether this modulation exists or not, further measurements of $C_{44}$ with different geometries, for example ($k \parallel H \parallel [001], u \parallel [100]$) and ($k \parallel H \parallel [100], u \parallel [001]$), need to be performed.

### C. Consideration of Hexadecapolar Contribution

In contrast to $C_{44}$ and other modes, $C_{66}$ measured with ($k \parallel [100], u \parallel [010], \text{and } H \parallel [001]$), has no rotation-effect contribution. As mentioned, none of these CEF schemes could reproduce the low-temperature softening of $C_{66}$ in high magnetic field.

A possible explanation for this softening is a higher-rank multipolar contribution, such as an electric hexadecapole contribution to the elastic constant. As shown in Table I, the transverse ultrasonic mode $C_{66}$ and $(C_{11} - C_{12})/2$, which propagate in the $c$ plane ($k \perp \{001\}$) also induce the rotation $\omega_{xy}$, which couples to the electric hexadecapole $H_2^2 = \sqrt{35}(J_1^4 - J_1^2)/4i$, with $\Gamma_2$ (A$_{2g}$) symmetry (Appendix B). This is the theoretically predicted order parameter of Scheme 1 in Table II. It should also be noted that recent inelastic x-ray scattering measurements showed that the 5$f$ ground-state wave function is composed mainly of $\Gamma_1$ and/or $\Gamma_2$, which is consistent with CEF Scheme 1.$^{35}$

Additionally, from recent resonant x-ray scattering measurements, no superlattice reflections or azimuthal angle-dependences which evidence rank 2 and 3 multipolar order have been observed so far$^{36}$. Thus, the lower-rank electric quadrupole order and magnetic octupolar

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**Table II.** Labels, CEF level scheme, active multipoles, author and references

| Labels | Level Scheme (K) | Active Multipoles (Symmetry) | Authors Ref. |
|--------|------------------|------------------------------|--------------|
| Scheme 1 $\Gamma_1(1) - \Gamma_2(60) - \Gamma_3(178) - \Gamma_4(491)$ | $H_2^2(A_{2g})$ | Yanagisawa et al. [28] |
| Scheme 2 $\Gamma_1(1) - \Gamma_3(1) (404) - \Gamma_2(1076)$ | $O_2^2(B_{1g})$ | Ohkawa and Shimizu, Galatanu et al. [29] |
| Scheme 3 $\Gamma_3 - \Gamma_1(1) (44) - \Gamma_2(112) - \Gamma_5(485)$ | $O_2^2(B_{1g})$ or $T_{xy}(B_{1u})$ | Santini and Amoretti [30] |
| Scheme 4 $\Gamma_1(1) - \Gamma_5(2)(140) - \Gamma_2(300)$ | $T_{0}^4(E_u)$ | Hanzawa and Watanabe [31] |
order can be eliminated as candidates for the HO parameter. The remaining unsubscribed order is an electric hexadecapole order with $A_{2g}$ symmetry or a composite order corresponding to this symmetry such as the chiral density wave order with $A_{2g}$ parameter. The remaining unsubscribed order is an electric multipolar order with $A_{2g}$ symmetry as compared to this symmetry such as the chi-

Using a different approach, we also checked the hexadecapolar contribution on the elastic constant $C_{66}$ in a magnetic field applied perpendicular to c axis. Figure 6 shows the magnetic-field dependence of the elastic constant $C_{66}$ for $H \parallel [100]$ and $H \parallel [110]$ of URu$_2$Si$_2$ at 4.2 K and 20 K. There is no obvious difference of the data below and above $T_0$ and for both field orientations within the present measurement accuracy. The quadrupolar susceptibility was calculated using a mean-field approximation, which assumes the $H^2$-type antiferro-hexadecapolar interaction as the HO parameter, based on the theory of Kusunose et al., which predicts that a very tiny difference should appear between the [100] and [110] directions in the antiferro-hexadecapole (AFH) order state. The calculated uniform quadrupolar susceptibility using the mean-field theory with CEF Scheme 1 as described in the text.
D. Comments on the Low possibility of Rotational Symmetry Breaking in the HO

Finally, we comment on the recently proposed symmetry-breaking scenarios. Tonegawa et al. reported that the lattice symmetry is broken from tetragonal to orthorhombic only when using a sample with very high RRR as found in synchrotron x-ray measurements. Ultrasound is a highly powerful tool to detect symmetry-breaking lattice distortions even when the lattice distortions are staggered or small. For example, the tetragonal systems DyB$_2$C$_2$ and BaFe$_2$As$_2$ systems show a $\epsilon_{xy}$-type staggered/uniform lattice distortion due to antiferro/ferro-quadrupolar order. A clear softening towards the phase transitions was observed in the related symmetric ultrasonic modes. The absence of such softening in $C_{66}$ leaves a $\epsilon_{xy}$-type orthorhombic lattice distortion in the HO highly unlikely. Namely, there will be no tetragonal to orthorhombic (4- to 2-fold) symmetry breaking in the HO. Instead, the softening is enhanced above 37 T where the hidden order is suppressed. It should be noted that $C_{66}$ shows a relatively large jump at $T_0$ in the temperature dependence at 30 T for $H \parallel [001]$ [as indicated by the red arrowhead in Fig. 3(b)]. This fact may suggest the freezing of the related multipolar degrees of freedom $O_{xy}$ or $H^2_0$ at $T_0$. However, these features appear already above the region of the Fermi-surface reconstruction, which has been pointed out by Shishido et al. based on the Hall-effect measurement. Thus, it is not clear whether the enhancement of the elastic anomaly of $C_{66}$ at $T_0$ in magnetic field is related to the origin of pure HO parameter. To more precisely determine the response of $C_{66}$ in these magnetic field regions, further investigation, such as ultrasonic measurements under static magnetic field around 30 T, are needed.

V. SUMMARY

We performed ultrasonic measurements on URu$_2$Si$_2$ in pulsed magnetic fields to check the elastic responses of this compound and found that the $\Gamma_4(B_{xg})$-type lattice instability is dominant at low temperature and low magnetic fields. In contrast, we observed enhancements of the elastic softening of the $\Gamma_4(B_{xg})$ and $\Gamma_5(E_{g})$ symmetric modes towards low temperatures at magnetic fields above 40 T. We discussed the origin of these elastic responses based upon the D$_{4h}$ symmetry point group analysis, starting from a local multiplet state (crystalline electric field) assuming weak hybridization and used an itinerant scheme based on the deformation-potential coupling due to the band-Jahn-Teller effect of a strongly c-f hybridized band which becomes weaker as the field is increased. The present analysis revealed again that the itinerant-band Jahn-Teller model is more applicable and the c-f hybridization is important in HO. On the other hand, the results cannot be explained by the quadrupolar susceptibility based on the crystalline-electric-field schemes in the $5f^2$-configuration which have been proposed thus far. To conclude, this work revealed important information on the elastic response towards the crossover from the delocalized to the localized electric state of the present system. However, a comprehensive interpretation of these elastic responses is still pending, and further investigations will be required.

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Appendix A: Formulation of the Multipolar Susceptibility

We start from the CEF Hamiltonian with elastic-strain mediated perturbation,

$$\mathcal{H} = \mathcal{H}_{\text{CEF}} + \sum_{\Gamma} \frac{\partial \mathcal{H}_{\text{CEF}}}{\partial \epsilon_{\Gamma}} \epsilon_{\Gamma}. \quad (A1)$$

The tetragonal CEF Hamiltonian with Zeeman effect is written as

$$\mathcal{H}_{\text{CEF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_4^1 O_4^1 + B_6^0 O_6^0 + B_6^4 O_6^4 + g_J \mu_B \sum_{i=x,y,z} J_i H_i. \quad (A2)$$

Here, $B_n^m$ are the CEF parameters and $O_n^m$ are the Stevens operators. The numerical values of $B_n^m$, which were used in the present analysis, are listed in Table III.

The second term of Eq. (A1) is explained in terms of electric multipole-strain interaction. Especially for rank-2 multipoles (quadrupoles), this term is written as

$$\mathcal{H}_{\text{MS}}^{(2)} = -g_{12}^{(2)} O_2^{0} \epsilon_x - g_{14}^{(2)} O_{xy} \epsilon_{xy} - g_{15}^{(2)} \{ O_{yz} \epsilon_{yz} + O_{zz} \epsilon_{zz} \}. \quad (A3)$$

For rank-4 multipoles (hexadecapoles), we assume a bilinear coupling between hexadecapoles and rotations with the same $\Gamma_4(A_{2g})$ symmetry instead of using a symmetrized strain $\epsilon_{\Gamma}$ as a perturbation field,

$$\mathcal{H}_{\text{MS}}^{(4)} = -g_{12}^{(4)} H_z \omega_{xy}. \quad (A4)$$
Here, $g_T^{(2)}$ and $g_T^{(4)}$ are the coupling constants for the rank-2 and rank-4 multiplets, respectively. $O_T$ and $H^o$ are quadrupole and hexadecapole operators, respectively. Those are listed in Table I and the quadrupole operators are also defined in Appendix B. The free energy of the local 5f electronic states in the CEF can be written as

$$F = U = N k_b T \ln \sum_n \exp\{-E_n(\epsilon_T)/k_b T\}. \quad (A5)$$

Here, $N$ is the number of ions in a unit volume, $E_n(\epsilon_T)$ is a perturbed CEF level as a function of strain $\epsilon_T$. $n$ is the number index for $J$ multiplets and their degenerate states. $U$ gives the internal energy for the strained system, which is written in terms of the symmetry strains and elastic constants listed in Table I as,

$$U = \frac{1}{2} \left\{ C_B \epsilon_B^2 + C_{Bu} \epsilon_B \epsilon_u + C_{u} \epsilon_u^2 + C_{x} \epsilon_x^2 \right. \]
$$
$$+ \left. C_{44}(\epsilon_{yz}^2 + \epsilon_{zx}^2) + C_{66} \epsilon_{xy}^2 \right\}. \quad (A6)$$

Here, $C = -(C_{11} + C_{12} - C_{13} - C_{14})/\sqrt{3}$. In second perturbation, the temperature dependence of the elastic constant is given by

$$C_T(T, H) = C_T^0 - N(g_T^{(2)})^2 \chi_T(T, H). \quad (A7)$$

Here, $C_T^0$ is the background of the elastic constant. The single-ion multipolar susceptibility $\chi_T$ is defined as the second derivative of the free energy with respect to strain (in the $\epsilon_T \to 0$ limit),

$$-(g_T^{(2)})^2 \chi_T = -\left\langle \frac{\partial^2 E_n}{\partial \epsilon_T^2} \right\rangle \]
$$
$$+ \frac{1}{k_B T} \left\langle \left( \frac{\partial E_n}{\partial \epsilon_T} \right)^2 \right\rangle - \left\langle \left( \frac{\partial E_n}{\partial \epsilon_T} \right)^2 \right\rangle \quad (A8)$$

Here, the angle brackets mean the thermal average. Note that, when we use the rotation $\omega_{xy}$ as a conjugate field for the hexadecapole moment, we need to assume some mechanism of the anisotropic hexadecapolar interaction, e.g., a two electron state, as discussed in Ref. 38, because the rotation $\omega_{y}$ is a unitary transformation for the system, i.e., it does not change the single-ion Hamiltonian at zero magnetic field. If Eq. (A4) is valid, we can substitute $\omega_{xy}$ for $\epsilon_{xy}$ in the formulas above to determine the hexadecapolar susceptibility. Eq. (A6) can be rewritten in the form of a normalized elastic constant as shown in Fig. 3 (a), (b), and (c).

$$\Delta(C_T(T, H) - C_T^0) = \frac{C_T(T, H) - C_T^0(T)}{C_T^0(T=1.5K)} \]
$$
$$= \frac{N(g_T^{(2)})^2}{C_T^0(T=1.5K)} \chi_T(T, H). \quad (A9)$$

In the present analysis, we assume $C_T^0(T) = C_{ph}(T)$ as the phonon contribution, which is obtained from the elastic constant of ThRu$_2$Si$_2$ without 5f-electron contribution. We now have the tools to compare the temperature- and magnetic-field dependence of the normalized elastic constants with the quadrupole susceptibility by assuming $A = N(g_T^{(2)})^2/C_T^0(T=1.5K)$ being independent from $T$ and $H$.

### Appendix B: Definition of Multipolar Moments and Equivalent Operator Expression

The electric multipolar operators are defined by multipolar expansion of the electrostatic potential as,

$$Q_{lm} \equiv e \sum_{j=1}^{n_f} r_j^l Z_{lm}(r_j). \quad (B1)$$

Here, $e < 0$ is the electron charge, $n_f$ is the number of f electrons. $Z_{lm}(r_j)$ is written by using spherical harmonics $Y_{lm}(r_j)$ as,

$$Z_{lm}(r_j) \equiv \sqrt{4\pi/(2l+1)} Y_{lm}(r_j). \quad (B2)$$

Eq.(B1) can be rewritten by replacing $(x, y, z)$ in $Z_{lm}(r_j)$ with spherical tensor operators $J_{lm}$ with following transformations,

$$x^n y^n z^n \rightarrow \frac{1}{(n_x + n_y + n_z)!} \sum_{j \geq 0} \mathcal{P}(J_x^n J_y^n J_z^n). \quad (B3)$$

Here, $\mathcal{P}$ is a sum of all possible permutations. Operator $J_{lm}$ has the following commutation relation, with the ladder operator $J_\pm = J_x \pm i J_y$:

$$J_\pm = (-1)^l \sqrt{[(2l - 1)!!]/(2l)!} (J^0)^l, \quad (B4)$$
Following are quadrupolar and hexadecapolar operators, which are used in the present analysis:

\[ [J_-, J_{lm}] = \sqrt{(l + m)(l - m + 1)} J_{lm-1}. \]

\[ O_{2z} = \frac{i}{\sqrt{2}} [J_{22} + J_{2-2}] = \frac{\sqrt{2}}{2} (J_x^2 - J_y^2) \]

\[ O_{xy} = \frac{i}{\sqrt{2}} [-J_{22} + J_{2-2}] = \frac{\sqrt{2}}{2} (J_x J_y - J_y J_x) \]

\[ \Gamma_3 (B_{1g}) : \]
\[ \Gamma_4 (B_{2g}) : \]
\[ \Gamma_5 (E_g) : \]
\[ O_{yz} = \frac{1}{\sqrt{2}} [J_{21} + J_{2-1}] = \frac{\sqrt{3}}{2} (J_y J_z - J_z J_y) \]
\[ O_{xz} = \frac{1}{\sqrt{2}} [-J_{21} + J_{2-1}] = \frac{\sqrt{3}}{2} (J_x J_z - J_z J_x) \]

\[ \Gamma_2 (A_{2g}) : \]
\[ H_z^a = \frac{\sqrt{35}}{4i} [-J_{44} + J_{4-4}] \]
\[ = \frac{\sqrt{35}}{8} \left( J_x^4 J_y^4 + J_y^4 J_x^4 + J_x J_y J_x^2 J_y^2 + J_y J_x J_y^2 J_x^2 \right) \]
\[ - (J_x J_y J_x J_y + J_y J_x J_y J_x + J_x J_y J_x J_y + J_y J_x J_y J_x) \]
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