A Study of Elastic Properties of URu_2Si_2 in Comparison with the Non-5*f* Contribution of $ThRu_2Si_2$

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A Study of Elastic Properties of URu_2Si_2 in Comparison with the Non-5*f* Contribution of $ThRu_2Si_2$

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Abstract. Ultrasonic investigations of the elastic constants of non-*5f* compound $ThRu_2Si_2$ were performed in order to highlight possible quadrupolar response in the elastic constants of heavy fermion compound URu_2Si_2, by using the $ThRu_2Si_2$ for backgrounds to subtract the phonon contributions in the elastic constants of URu_2Si_2. We confirmed that the all-elastic constants of $ThRu_2Si_2$ show no elastic anomaly, *i.e.*, increasing monotonically and saturating toward low temperatures, which can be understood regular phonon contributions for the solid states. The background-subtracted $(C_{11}-C_{12})/2$ of URu_2Si_2 keeps decreasing with decreasing temperatures from 300 K, while other share modes C_{44} and C_{66} show little change. These contrasts suggest that the Γ_3-type quadrupole moment O_2^2 will be still dominant and active in URu_2Si_2 at low temperatures.

1. Introduction

Regarding a mysterious phase transition in URu_2Si_2 at T_O = 17.5 K, so-called 'Hidden Order (HO)', there are several experimental and theoretical approaches for the unknown order parameter [1, 2, 3]. In particular, non-magnetic multipolar orderings, including O_{xy}-type antiferro-quadrupole (AFQ) order [4], have recently been emerged as a candidate for the HO, where any intrinsic magnetic dipole moment and symmetry breaking lattice distortion should not be detected under ambient pressure in zero magnetic fields [5]. A direct verification experiment for the AFQ order has been performed by using resonant X-ray scattering (RXS), however, no significant scattering, which indicate quadrupole O_{xy} and O_2^2 order, have been detected thus far [6]. Since another theoretical predictions for the HO, such as antiferro-electric-hexadecapole order [7] or electronic-nematic phase [8, 9], have been proposed more recently, many experimental efforts such as RXS and neutron scattering under magnetic fields are ongoing in order to verify them microscopically.

On the other hand, ultrasonic measurement is one of the useful macroscopic methods to investigate the quadrupole response in the solid state. Generally in the localized 4*f*-electron system, a temperature dependence of the elastic constant, observed by ultrasound, can be understood as quadrupole susceptibility. When the system possesses quadrupole degree of freedom, a Curie-type decreasing (softening) appears on the elastic constant, corresponds to volume conservative and symmetry breaking ultrasonic modes [10]. Mostly in the case of U-based compound, such elastic softening in the shear mode is rarely found due to enhancement of its itinerant character of the *f*-electrons. However, if the hidden order parameter breaks the local symmetry of the crystal, some kind of elastic response via electron-phonon interaction would still be expected. In the present work, we have performed ultrasonic measurement both on URu_2Si_2 and on non-5*f* system $ThRu_2Si_2$ single crystals in order to search possible contributions of electric quadrupoles onto the elastic constants of URu_2Si_2.

Table 1. Absolute values of elastic constants at 80 K for URu_2Si_2 and $ThRu_2Si_2$ in the present study.

Elastic Constant	Quadrupole (Symmetry)	**URu_2Si_2** (10^{10} J m^{-3})	**$ThRu_2Si_2$** (10^{10} J m^{-3})
C_{11}	O_2^0 (Γ_1)	26.2	26.8
$(C_{11}-C_{12})/2$	O_2^2 (Γ_3)	5.7	6.2
C_{44}	O_{yz}, O_{zx} (Γ_5)	9.6	10.7
C_{66}	O_{xy} (Γ_4)	12.1	10.2

2. Experimental Details

Single crystalline URu_2Si_2 and $ThRu_2Si_2$ were prepared by Czochralski tri-arc technique and no annealing process was performed. A hexagonal-cylinder-shaped sample for URu_2Si_2 and two parallelepiped samples for $ThRu_2Si_2$ were cut from each bulk crystal by means of high-precision spark erosion. URu_2Si_2 sample has dimensions of 5.097 mm for [001], 4.161 mm for [110] and 3.864 mm for [100]. $ThRu_2Si_2$ samples have orientation of [001]-[010]-[001] and [110]-[1-10]-[001] with corresponding dimensions $1.970 \times 0.896 \times 0.799$ mm^3 and $2.229 \times 1.259 \times 0.697$ mm^3, respectively. A phase comparator using double balanced mixer can detect the relative change of sound velocity down to 4.2 K by using ^{4}He refrigerator. Piezoelectric $LiNbO_3$ wafers of 36°Y-cut with 100 μm thickness were used for generating and detecting longitudinal ultrasonic waves. The absolute value of sound velocity was estimated by monitoring ultrasonic echoes on a digital oscilloscope and calculated by sample length L and mass density ρ, which was calculated from the lattice constants.

3. Results and Discussions

The obtained absolute values of the elastic constants on URu_2Si_2 and $ThRu_2Si_2$ are summarized in Table 1 with corresponding quadrupole and symmetry, where measurement frequencies varying between 100-175 MHz were used. Comparing the elastic constants, $ThRu_2Si_2$ represents larger values in C_{11}, $(C_{11}-C_{12})/2$, C_{44} than that of URu_2Si_2. It is simply expected that additional 5*f*-electron contribution result the elastic constant in the U-based compound lower in general. Intriguingly, C_{66} mode, however, exhibits the opposite tendency of magnitude, which implies that the bonding force of the lattice has strong anisotropy due to some 5*f*-electron's contribution. Though it is a matter of speculation and open question.

Figure 1 shows a comparison of elastic constants C_{11}, $(C_{11}-C_{12})/2$, C_{44} and C_{66} of URu_2Si_2 and $ThRu_2Si_2$. The elastic constants C_{11} and $(C_{11}-C_{12})/2$ of URu_2Si_2 exhibit maximum at ~100 K while C_{44} and C_{66} do not show any anomaly in Para state above T_O, which are consistent with previous reports [11, 12]. On the other hand, all elastic constants of $ThRu_2Si_2$ increase monotonically with decreasing temperature and exhibit no anomaly. In the present study, the elastic constants of $ThRu_2Si_2$ were used for the backgrounds to subtract the phonon contributions in the elastic constants of URu_2Si_2.

Figure 2 represents the elastic constants of URu_2Si_2 with those of $ThRu_2Si_2$ subtracted, which are displayed by relative change as,

$$\frac{C_{ij(U-Th)}-C_{ij(U-Th@300K)}}{C_{ij(U-Th@300K)}} = \frac{\left\{C_{ij(URu_2Si_2)}(T)-C_{ij(ThRu_2Si_2)}(T)\right\}-\left\{C_{ij(URu_2Si_2)}(300K)-C_{ij(ThRu_2Si_2)}(300K)\right\}}{\left\{C_{ij(URu_2Si_2)}(300K)-C_{ij(ThRu_2Si_2)}(300K)\right\}}. \quad (1)$$

The softening appears in C_{11} down to 30 K implies that the system has the instabilities of Kondo volume collapse [13]. The background-subtracted $(C_{11}-C_{12})/2$ keeps decreasing almost linearly towards low temperature, while other share modes C_{44} and C_{66}, corresponding to Γ_5-type quadrupoles O_{yz} $(= J_yJ_z+J_zJ_y)$, O_{zx} $(= J_zJ_x+J_xJ_z)$ and Γ_4-type O_{xy} $(= J_xJ_y+J_yJ_x)$, respectively, show little change. These contrasts suggest that the Γ_3-type quadrupole moment will be active in URu_2Si_2, if one can assume that the softening of $(C_{11}-C_{12})/2$ is explained by quadrupole susceptibility.

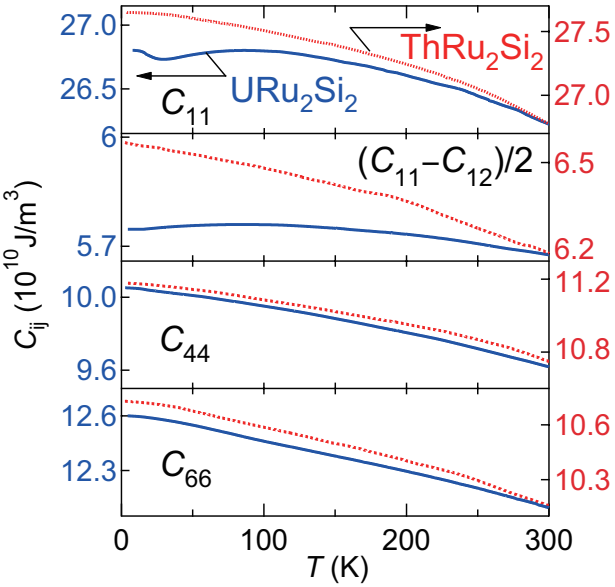

Figure 1. Elastic constants of URu_2Si_2 (Blue-line, left axis) and $ThRu_2Si_2$ (Red-dots, right axis).

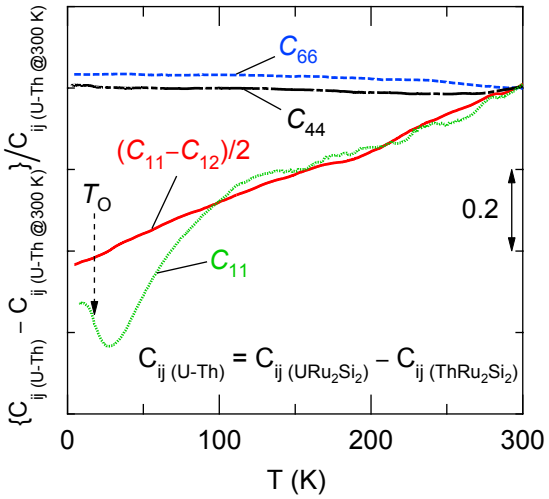

Figure 2. Relative change of the elastic constants as a function of temperature for URu_2Si_2 with the effect of $ThRu_2Si_2$ subtracted.

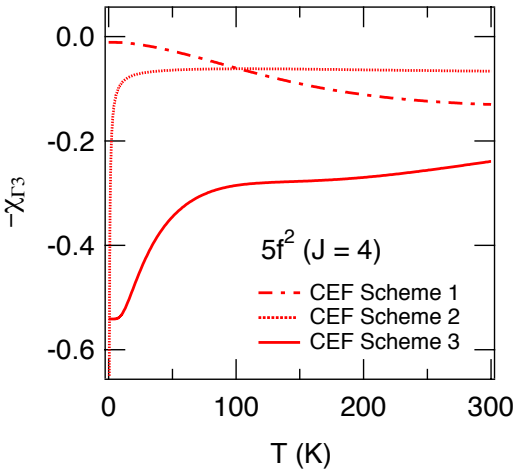

Figure 3. Calculated quadrupole susceptibility $-\chi_{\Gamma 3}$ vs. temperature for three proposed CEF level schemes of $5f^2$ ($J = 4$).

The CEF scheme-1 is $\Gamma_1^{(1)}$ (0 K) – Γ_2 (50 K) – $\Gamma_1^{(2)}$ (171 K) – $\Gamma_5^{(2)}$ (500 K) – Γ_4 (1254 K) – Γ_3 (1451 K) – $\Gamma_5^{(1)}$ (1727 K), proposed by Nieuwenhuys. [14] The CEF scheme-2 is $\Gamma_5^{(1)}$ (0 K) – $\Gamma_1^{(1)}$ (404 K) – Γ_2 (1076 K) – $\Gamma_1^{(2)}$ (1119 K) – $\Gamma_5^{(2)}$ (2017 K) – Γ_4 (3253 K) – Γ_3 (3613 K), proposed by Galatanu *et al*. [15] The CEF scheme-3 is Γ_4 (0 K) – $\Gamma_1^{(1)}$ (44 K) – Γ_2 (111 K) – $\Gamma_5^{(2)}$ (485 K) – Γ_3 (534 K) – $\Gamma_5^{(2)}$ (826 K) –$\Gamma_1^{(2)}$ (875 K), proposed by Santini *et al*. [16]

Temperature dependence of the elastic constant $(C_{11}-C_{12})/2$ can be described by CEF effect of localized *f*-electron's Γ_3 quadrupole susceptibility as,

$$\frac{(C_{11}-C_{12})}{2} = \frac{\left(C_{11}^0-C_{12}^0\right)}{2} - \frac{Ng_{\Gamma 3}^2\chi_{\Gamma 3}(T)}{1-g'_{\Gamma 3}\chi_{\Gamma 3}(T)} \qquad (2)$$

Here, $(C_{11}{}^0-C_{12}{}^0)/2$ is background, *i.e.*, other than the *f*-electron contributions such as phonon contributions on $ThRu_2Si_2$, N is number of the ions in a unit volume, $g_{\Gamma 3}$ and $g'_{\Gamma 3}$ is coupling constant of quadrupole-strain interaction and quadrupole-quadrupole intersite interactions for Γ_3-type quadrupole moment O_2^2 $(= J_x^2-J_y^2)$, respectively. Only the calculated quadrupole susceptibility by using the CEF scheme-3 shows decreasing for all temperature ranges, while the CEF scheme-2 gives broad local maximum at around 100 K and the CEF scheme-1 results increase in $-\chi_{\Gamma 3}$ from room temperature as shown in Fig. 3. However, the CEF scheme-3 still cannot be perfectly reproducing the temperature dependence of the background-subtracted $(C_{11}-C_{12})/2$ below 100 K. In the present stage, a strong *c-f* hybridization effect of URu_2Si_2 at low temperature region are not take into account. The possible Kondo effect had been evidenced by several physical properties such as log*T* increasing of resistivity around room temperature or strong suppression of resistivity and magnetic susceptibility below 60 K. Since the many-body effect might suppress not only the Γ_3-quadrupole susceptibility but also other quadrupole responses, it is rather difficult to understand the temperature dependence of the elastic constant of 5*f*-electron systems only from the localized picture. We could, however, conclude that at least the Γ_3-type quadrupole moment O_2^2 in URu_2Si_2 is still dominant in comparison to O_{yz}, O_{zx} and O_{xy} and/or this compound has some electric instability, which well couples to the strain wave with Γ_3 symmetry.

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