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Search for the quadrupolar instability in URu_2Si_2

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Abstract

We have performed the thermal expansion $\alpha(T)$ and elastic constant $c(T)$ measurements of the heavy electron system URu_2Si_2, with special attention to the tetragonal symmetry of $\Gamma_3(x^2-y^2)$ and $\Gamma_4(xy)$. The experimental results of c_{66}, $\frac{1}{2}(c_{11}-c_{12})$ and $\alpha(T)$ along [±1 1 0] do not confirm a sizable uniform distortion for these two types of symmetry through a puzzling phase transition at $T_o = 17.5$ K. On the other hand, the $\frac{1}{2}(c_{11}-c_{12})$ data, which are missing in the previous detailed study, are found to show a weak but significant tendency of softening below about 70 K, suggesting an elastic response of the quadrupolar moment of $J_x^2 - J_y^2$.

Keywords: URu_2Si_2; Quadrupolar instability; Thermal expansion; Elastic constants

One of the most attractive issues regarding the heavy-electron (HE) superconductor URu_2Si_2 is to clarify the intrinsic order parameter (OP) of the phase transition at $T_o = 17.5$ K. Polarized neutron analyses have evidenced that 5f spin dipoles form an antiferromagnetic (AF) arrangement below T_o, excluding other prospective multipoles out of the primary OP [1]. Nevertheless, the extremely small amplitude of the observed staggered moments ($\mu_o \sim 0.04\,\mu_B$), which obviously contradicts large anomalies seen in the macroscopic property, still motivates us to search for a hidden mechanism of this phase transition [2].

Instead of the tiny dipolar ordering, a quadrupolar ordering [3] and an intermediate to $5f^2$ valence transition [4] have been argued as possible candidates for a hidden primary OP. The quadrupolar model gives a most convincing explanation to the macroscopic property of the system, although the tiny dipolar moments must be put aside as an unknown side effect. In the valence transition model, on the other hand, the tiny moments are ascribed to an admixture of a nonmagnetic state in $5f^2$ to excited spin states in $5f^1$ or $5f^3$. The order parameter is predicted to be a gap energy between two lower-lying crystalline electric field (CEF) singlets in the $5f^2$ configuration, which are merged with each other by valence fluctuations above T_o.

Interestingly, both models predict a lattice distortion in the tetragonal symmetry of $\Gamma_3(x^2-y^2)$ or $\Gamma_4(xy)$ to be involved in the phase transition, although no direct observation has been reported yet. The purpose of this paper is to investigate such possibility of lattice instability, supplementing the data of thermal expansion and elastic constants for the relevant symmetry.

A high-quality single-crystalline bar of URu_2Si_2 was prepared by the Czochralski tri-arc technique and no further heat treatment was performed. Two

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parallelepipeds were cut from the bulk crystal by means of high-precision spark erosion, in the orientation of [1 0 0]–[0 1 0]–[0 0 1] (corresponding dimensions $4.0 \times 2.0 \times 2.5\ mm^3$) and of [1 1 0]–[−1 1 0]–[0 0 1] ($3.0 \times 2.0 \times 2.8\ mm^3$). The coefficient of linear thermal expansion $\alpha = L^{-1}\,dL/dT$ was measured by a sensitive three-terminal capacitance technique in the temperature range 4.2–50 K. The ultrasonic velocity v was measured with a phase comparison method in the temperature range 4.2–150 K, using piezoelectric $LiNbO_3$ plates as ultrasonic transducers with resonance frequencies of 15 MHz (10 MHz) for longitudinal (transverse) waves. The absolute value of elastic constants $c = \rho v^2$ was calculated with the mass density at room temperature; $\rho \sim 10.01\ g/cm^3$.

First, we investigate the uniform lattice distortion in the Γ_4 symmetry by the thermal expansion along [1 1 0] directions. Previous results along a and b axes ($\alpha_{[1\,0\,0]}$) have evidenced that there is no lattice distortion uniform in the Γ_3 symmetry within the accuracy of $\Delta l/l < 10^{-7}$ [5]. This geometry of experiments however is insensitive to the Γ_4 distortion, since this distortion leads to an equivalent change in the capacitance gap for two [1 0 0] arrangements of the sample. If the Γ_4 distortion takes place, we would detect it most clearly by $\alpha_{[1\,1\,0]}$ as opposite variations between two orthogonal [±1 1 0] directions. However, the amplitude of such an anomaly estimated from the $\alpha_{[1\,0\,0]}$ data is equal to $\sim 10^{-5}$ in $\Delta l/l$. This may nearly be of the resolution limit in other techniques, and thus it is of interest to be checked by high-precision dilatometry. The results are shown in Fig. 1. The $\alpha_{[1\,1\,0]}$ data for two [±1 1 0] directions are superposed not only upon each other but also with the previous results of $\alpha_{[1\,0\,0]}$ [5]. We thus conclude that there is no sizable effect ($\Delta l/l < 10^{-7}$) in lowering the fourfold symmetry of URu_2Si_2 macroscopically.

In Fig. 2, we provide an overview of the temperature variations of elastic constant for three modes, c_{11}, c_{66} and $\frac{1}{2}(c_{11} - c_{12})$. Detailed studies have already been made for the former two modes [6], while the mode $\frac{1}{2}(c_{11} - c_{12})$ was measured for the first time. We found that this transverse mode decreases as the temperature drops, showing a maximum centered at $T_{max} \sim 70$ K. Upon further

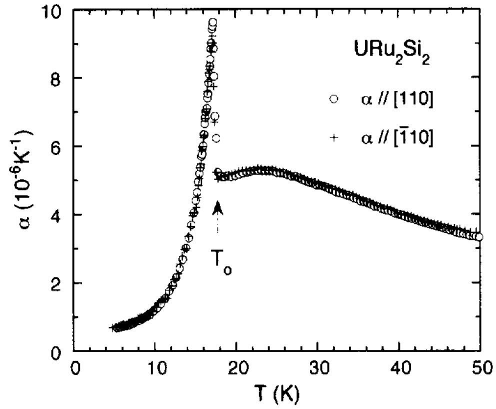

Fig. 1. The thermal-expansion coefficient along two orthogonal directions [± 1 1 0] of URu_2Si_2.

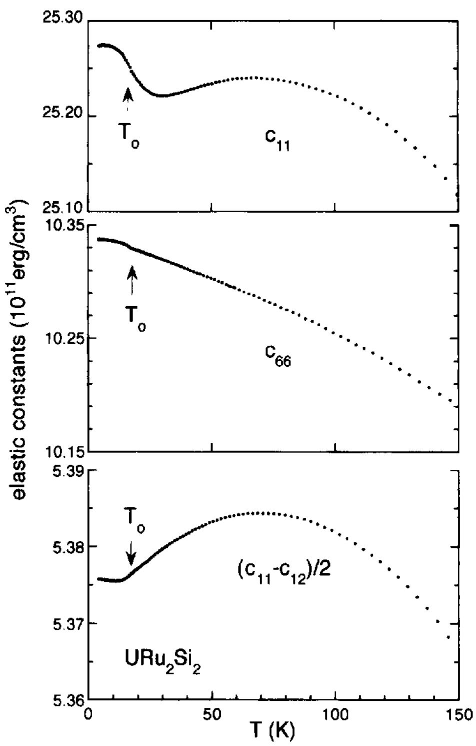

Fig. 2. Temperature variations of three elastic modes, c_{11}, c_{66}, and $\frac{1}{2}(c_{11} - c_{12})$ of URu_2Si_2.

cooling, it exhibits a small kink at T_o and turns up slowly again below ~ 12 K. The data for c_{11} and c_{66} modes, on the other hand, trace the previous results qualitatively: c_{66} monotonously increases as the temperature drops, and c_{11} exhibits a maximum at ~70 K, followed by a broad minimum at ~30 K.

The softening observed in $\frac{1}{2}(c_{11}-c_{12})$ below T_{max} ~ 70 K is weak (~ 0.2%) but could be remarkable in the sense that it occurs in the volume conserving transverse mode in the HE states. In general, the elastic property of HE materials can be characterized by a tendency of softening in the longitudinal modes in accordance with the volume effects due to c-f hybridization. In the previous reports [6], in fact, the anomalous behavior of the longitudinal c_{11} mode is treated in this line, since there is no anomaly visible in the transverse c_{44} and c_{66} modes. Our experiments, however, reveal a similar maximum appearing in the other transverse mode $\frac{1}{2}(c_{11}-c_{12})$ with nearly the same relative amplitude and T_{max} as those in c_{11}. Since c_{11} is the response to the combined strain $\varepsilon_{\Gamma_1}+\varepsilon_{\Gamma_3}$, we strongly suggest that the broad peak in c_{11} should be identical to that in $\frac{1}{2}(c_{11}-c_{12})$ belonging to the Γ_3 symmetry. The residual increase of c_{11} below ~30 K might be due to the behavior in Γ_1 symmetry, and/or due to the isothermal sound propagation as pointed out in Ref. [6]. We would like to stress that this transverse mode anomaly of URu_2Si_2 gives a particularly strong contrast to the isostructural HE material $CeRu_2Si_2$ [7] that exhibits a softening only in the longitudinal modes. This implies an essential difference in f states between these two compounds.

Assuming 5f electrons to be well localized, we could attribute the softening in $\frac{1}{2}(c_{11}-c_{12})$ to thermal fluctuations of quadrupolar moments $J_x^2-J_y^2$ in the CEF 5f states. In terms of the proposed CEF quadrupolar model [3], the $\frac{1}{2}(c_{11}-c_{12})$ data can be well described as a Van Vleck type of contribution to the strain susceptibility [8]. The absence of anomaly in the longitudinal c_{33} mode [6] is also consistent with the absence of coupling by J_z^2 between lower-lying CEF states. In this view, however, the possible quadrupolar ordering must form an AF type of structure because of the above discussion on $\alpha_{[1\,1\,0]}$, and because none of transverse modes tends to diverge at T_o. On the contrary, the CEF scheme for the induced dipolar ordering [9] seems unlikely to reproduce all modes consistently, because of the opposite selection rules assumed: a strong coupling for J_z^2 but none for $J_x^2-J_y^2$ between lower-lying levels.

The data for $\frac{1}{2}(c_{11}-c_{12})$ may also be consistent with the model of intermediate to $5f^2$ valence transition [4], in the qualitative sense that the system should have the structural instability in the symmetry of Γ_3 or Γ_4. Detailed calculations for the strain susceptibility in the paramagnetic intrmediate states would be desirable.

In conclusion, we have discussed the lattice instability for URu_2Si_2, focusing our attention on the symmetry of Γ_3 and Γ_4. Absence of uniform distortion for these two types of symmetry has been most accurately confirmed by means of dilatometry. The elastic response has been found to exhibit a tendency of softening in the symmetrized transverse mode $\frac{1}{2}(c_{11}-c_{12})$, suggesting the presence of lattice instability in the Γ_3 symmetry. Such anomaly in the volume conserving mode is quite rare in HE materials and thus expected to offer the key to an understanding of the 17.5 K phase transition. Within the existing theories, the observed elastic property is consistent with the quadrupolar model, and with the valence transition model, although further analyses need to be made. To further elucidate the relation between this elastic anomaly and the ground state, the systematic analyses for the doped materials, such as $U(Ru, Rh)_2Si_2$ and $(Th, U)Ru_2Si_2$, are planned.

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