

HIGH PRESSURE X-RAY DIFFRACTION STUDY
OF URu₂Si₂* **

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We have performed high pressure X-ray diffraction measurements on a powder sample of the tetragonal heavy-electron compound URu₂Si₂ at low temperatures and pressure up to 3 GPa, in order to investigate a pressure-induced phase transition at $P_c = \sim 1.5$ GPa, which was indicated in the neutron diffraction experiment under pressure. The pressure variations of the lattice parameters a and c at 15 K decrease monotonously with increasing pressure. No discontinuity of the lattice parameters of URu₂Si₂ around P_c is observed within experimental error.

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The heavy-electron compound URu₂Si₂ crystallizes in the body-centered tetragonal ThCr₂Si₂ type of structure, space group I4/mmm [1]. It undergoes two successive phase transitions; a puzzling phase transition at $T_0 = 17.5$ K and a superconducting transition at $T_c \sim 1$ K [2]. Recent NMR measurement under pressure ($P \leq 0.83$ GPa) revealed the coexistence of antiferromagnetic (AF) and paramagnetic (PM) regions below T_0 , where the two

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regions are spatially separated [3], although the intrinsic order parameter in the PM region below T_0 is still unknown. On the other hand, neutron diffraction (ND) experiment under pressure indicates that URu₂Si₂ undergoes a pressure-induced phase transition at $P_c = \sim 1.5$ GPa, from the puzzling phase at T_0 to AF phase with a large staggered moment ($\sim 0.4\mu_B/\text{U}$) [4]. Important finding in this ND experiment is a shrinkage of the lattice around P_c with a discontinuous change of the lattice parameter $-\Delta a/a \sim 0.2\%$, implying that this phase transition is of first order.

In previous high pressure X-ray diffraction measurements on a powder sample at room temperature, it is reported that the tetragonal structure of URu₂Si₂ is stable at very high pressure up to 50 GPa [5]. So far, however, no X-ray measurement around P_c under the combined conditions of high pressures and low temperatures has been done. Therefore, we have performed high pressure X-ray diffraction measurements on a powder sample of URu₂Si₂ at temperatures between 15 K and 100 K and pressure up to 3 GPa, focusing attention on the lattice anomaly.

A single crystal of URu₂Si₂ was grown in a tetra-arc furnace using the Czochralski method. The crystal was annealed at 1000°C for 10 days in a vacuum sealed quartz tube. In the preliminary X-ray diffraction experiment on fine powder of URu₂Si₂, we found that the powder process makes the shape of Bragg peaks very broad. Therefore, the single crystal was carefully crushed into rough powder for measurement of lattice parameters, although the effect of the preferred-orientation appears rather strongly. The powder sample was loaded into a He-gas driven diamond anvil cell (DAC) with stainless steel gasket with hole of 450 μm in diameter and thickness 200 μm . Small ruby chips were also placed with the sample for pressure determination with the ruby fluorescence method. A 4:1 methanol-ethanol solution was used as the pressure-transmitting medium. The DAC was mounted on a closed-cycle refrigerator. Synchrotron radiation X-ray diffraction experiments were performed at the BL-1B beamline of the Photon Factory [6]. The X-ray wavelength was 0.6884 Å calibrated with a CeO₂ powder diffraction pattern. The intensities of the Bragg reflections were measured in the range $2\theta \leq 38^\circ$. The obtained URu₂Si₂ powder diffraction patterns were analyzed by the Rietveld analysis program RIETAN-2000 [7]. The peak shape was modeled with a pseudo-Voigt function. The lattice, profile and preferred-orientation parameters were refined at each pressure.

X-ray powder diffraction pattern of URu₂Si₂ under highest pressure 2.93 GPa at 15 K is displayed in Fig. 1. The data can be fitted with ThCr₂Si₂ type of structure. The tetragonal lattice parameters at 2.93 GPa and 15 K are refined as $a = 4.1028(4)\text{\AA}$ and $c = 9.5065(16)\text{\AA}$. In the whole pressure range investigated, the Rietveld refinements indicate the absence of any low-ordering in the crystal symmetry.

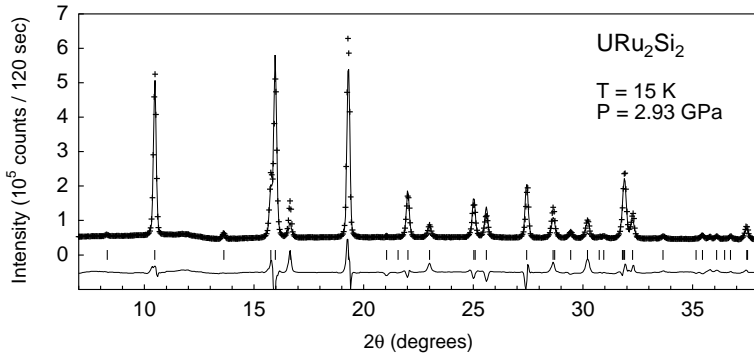


Fig. 1. X-ray powder diffraction pattern of URu_2Si_2 at 2.94 GPa and 15 K. The observed intensity data are shown by crosses. The solid line denotes the calculation results after Rietveld refinement. The lower solid line denoted the difference between the experimental and calculated intensities. The vertical markers denote positions of possible reflections for the body-centered tetragonal ThCr_2Si_2 type of structure.

Fig. 2 shows the pressure variations of the lattice parameters a and c , the volume V and the c/a ratio at 15 K ($\leq T_0$). Both lattice parameters decrease monotonically with increasing pressure. No discontinuous change of the lattice parameters around P_c is observed within experimental error. The lattice parameters at 100 K ($\geq T_0$) also decrease monotonically with increasing pressure without large discontinuity. From a linear fit of the relative volume $\Delta V/V$, we get an isothermal compressibility $\kappa_T = 5.2 \times 10^{-3} \text{ GPa}^{-1}$. The obtained κ_T value is smaller than the previous estimated value ($7.3 \times 10^{-3} \text{ GPa}^{-1}$) [8] and the value obtained from data of elastic constants at 4.2 K ($7.1 \times 10^{-3} \text{ GPa}^{-1}$) [9]. The obtained κ_T value is larger than the previous result at room temperature for the high pressure powder X-ray diffraction measurement ($4.0 \times 10^{-3} \text{ GPa}^{-1}$) [5]. On the other hand, the pressure variation of the c/a ratio is constant within experimental error. The compression is almost isotropic in the present pressure range.

The present result indicates that the pressure-induced phase transition at P_c is absent or is of second order in the powder sample, because the jump of lattice parameters at P_c is not seen. To clarify the discrepancy between the present result and the ND result, further microscopic measurements on URu_2Si_2 under pressure may be needed.

In conclusion, we have measured the pressure variations of the lattice parameters of URu_2Si_2 at low temperatures by using synchrotron X-ray diffraction technique, in order to investigate the pressure-induced phase transition at P_c , which was indicated in the ND experiment [4]. No discontinuity of the lattice parameters of URu_2Si_2 around P_c is, however, observed within experimental error.

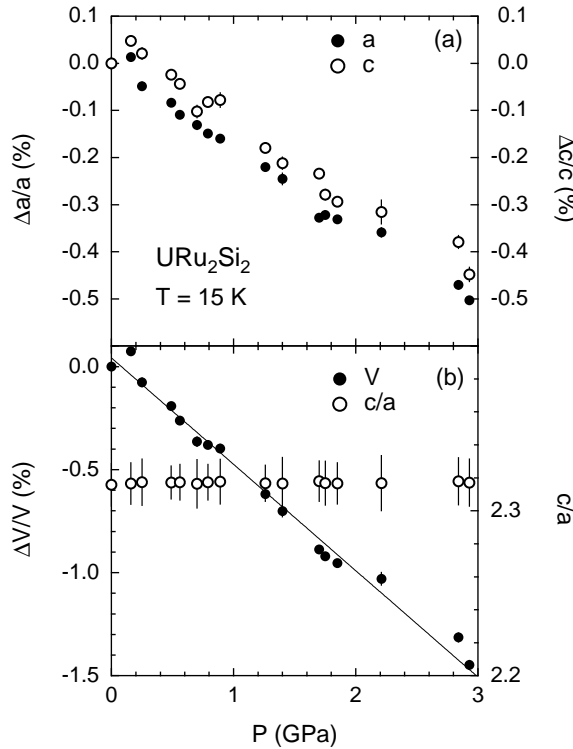


Fig. 2. Pressure variations of (a) the relative lattice parameters $\Delta a/a$ (●) and $\Delta c/c$ (○) and (b) the relative volume $\Delta V/V$ (●) and the c/a ratio (○) at 15 K. The solid line is a linear fit to the $\Delta V/V$ data.

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