Resonant X-ray Scattering Study of Hidden Order in URu$_2$Si$_2$
using a Low-Stress Single Crystal

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Abstract. We have studied a puzzling phase below 17.5 K of URu$_2$Si$_2$ by means of resonant X-ray scattering (RXS) near the U $M_{IV}$ absorption edge ($E \sim 3.73$ keV), using a high-quality single-crystalline sample with low residual stress. We reconfirm that a commensurate (003) superlattice reflection comes purely from antiferromagnetic dipolar order with ordering wave vector $Q_{AF} = (1, 0, 0)$. No other traces of significant RXS have been detected in a major region of $(h0l)$ plane, indicating absence of quadrupolar order within a searched $Q$ range including a conceivable nesting vector $Q^* = (1.4, 0, 0)$.

1. Introduction

Hidden order (HO) phase below 17.5 K ($\equiv T_o$) in the heavy-electron superconductor URu$_2$Si$_2$ remains unidentified, despite substantial number of research efforts over the last two decades [1]. The transition is marked by abrupt changes in various bulk quantities such as specific heat, resistivity, thermal expansion, and hall coefficient [2-6]. However, microscopic studies have not yet succeeded in specifying not only the frozen 5f-electronic degree of freedom but also the ordering wave vector $Q$. Quadrupoles and octupoles are potential candidates of the hidden order parameter [7-11], firstly because almost no internal magnetic fields appear in HO [12-14], and secondly because emergences of longitudinal dipolar excitations below $T_o$ [15] and of antiferromagnetic (AF) order under hydrostatic pressure [16,17] are favorable for the ground state of $O_{x^2y^2}$, $O_{xy}$, $T_{2g}^0$, and $T_{3g}$, since those and $J_z$ all correspond to different irreducible representation of the $D_{4h}$ point symmetry.

In the present study, we investigate the possibility of quadrupolar order in URu$_2$Si$_2$ by means of resonant X-ray scattering (RXS) measurements. In previously published papers [18,19], authors have observed forbidden Bragg reflections at (003) and (005), and concluded from polarization and azimuthal analyses that the reflections correspond to a weak AF dipolar order with $Q_{AF} = (1, 0, 0)$ (the magnetic moments align along the tetragonal $c$ axis) [15]. This AF state had been believed to be a uniform order in early studies, but the accumulation of recent neutron-scattering (NS) data strongly suggests that the AF signal at ambient pressure is due to a phase coexistence of the high-pressure AF...
phase into HO with a volume fraction of ~ 0.1 – 1 % [20]. We recently found a crystal which shows a low AF NS intensity [20]. On the other hand, Nagao and Igarashi have theoretically predicted the detailed RXS spectra including azimuthal angular dependence for the expected quadrupolar ordered states [21]. Motivated by these advances, we have reinvestigated the RXS spectra using this particular sample under a condition of low AF contamination, and searched for \( \mathbf{Q} \) vector in an unexplored reciprocal-lattice region.

2. Experimental Procedure
A single crystal of URu2Si2 (ThCr2Si2-type bct structure, I4/mmm) with a cylindrical shape (\( \phi \) 2 mm x 25 mm) was prepared by the Czochralski pulling method in a tetra-arc furnace under a pure argon atmosphere. The cylindrical axis is parallel to the \( c \) axis. A part of this crystal (a \( c \)-plane-cleaved 6-mm bar, labeled #1) was used for our recent NS experiments performed under high pressure [20], where we found it showing the weakest AF scattering intensity ever reported for this system. A sample #2 used in the present study is a neighboring cleavage counterpart of the #1 piece, annealed in the same procedure (for 7 days at 1000 °C). The RXS measurements were performed at beamline BL22XU in SPring-8. The photon energy was the U Mn absorption edge 3.726 keV. The sample was mounted on the cold head of a commercial ⁴He closed cycle cryostat, which was installed on a standard four-circle diffractometer. The incoming beam was linearly polarized in the horizontal scattering plane (\( \pi \) polarization), and irradiated to the cleaved \( c \) surface. Orientation of the sample was checked using (002) and (103) reflections. The width of the (002) rocking curve \( \delta \omega_{(002)} \) was ~ 0.02°.

3. Results and Discussion
We first measured the energy spectrum of the (003) forbidden reflection intensity without polarization analysis (Fig. 1). The peak position and shape are similar to what have previously been reported for this material [18]. Tuning the photon energy to 3.726 keV, we then performed longitudinal scans through the (003) reflection for several azimuthal angles \( \Psi \). The scattered beam polarization was analyzed by using an Al (111) crystal. The measured RXS intensity was significant in the \( \pi-\sigma' \) scattering channel, while it fell to a background level in the \( \pi-\pi' \) channel (Fig. 2 (a)). Furthermore, no significant \( \Psi \) dependence was detected in both the channels. Although the measured \( \Psi \) window is limited, the observed behavior is apparently different from the theoretical predictions for the expected quadrupolar ordered states (Fig. 2 (b)) [21]. Thus it should be concluded, as in the previous reports [18,19], that the (003) RXS peak from the present sample also arises only from the AF dipolar (\( J_x \)) ordered state. We note that the observed (003) RXS intensity, if normalized to the (002) charge reflection, is roughly less than 1 % of our own previous data taken for a different sample with a larger crystal mosaic (\( \delta \omega_{(002)} \sim 0.2° \)). This supports the conjecture that the AF phase mixing is induced by the lattice distortion due to residual stress in the crystal [22], and the present results would confirm the absence of \( O_{xy} \) and \( O_{x'y'} \) quadrupolar order with \( \mathbf{Q}_{AF} = (1, 0, 0) \) in high precision.

Figure 1. The RXS spectrum of the (003) forbidden Bragg reflection vs. photon energy. The beam polarization is unanalyzed.
In order to explore the possibility of quadrupolar ordering with a different propagation vector, we further performed a two-dimensional mesh scan in the region of $0.25 \leq h \leq 0.42$ and $2.0 \leq l \leq 3.0$ in the principal $(h0l)$ plane that we could survey in the present experimental setup. The mesh ($\delta h \sim \delta l \sim 0.003$) was chosen to be about a half width at half maximum of the (003) reflection peak. As seen in Fig. 3, no significant peak was observed in the region within the statistical error of about one-tenth of the (003) peak intensity. The reciprocal-lattice point $(0.403)$ corresponds to the wave vector $Q^* = (1.4, 0, 0)$, where strong longitudinal spin fluctuations are known to appear in the inelastic NS spectra [15,23]. The phenomena reflect the collective excitations from the HO ground state, and their temperature variations compared with band calculations suggest a close relationship between HO and a Fermi-surface nesting instability with $Q^*$ [23,24]. Novel incommensurate quadrupolar order, which has actually been observed in PrPb$_3$[25], may be expected, but the present experimental results discard this possibility at least in the scanned $Q$ range as well as $Q^*$, within the experimental accuracy.
4. Summary

No indication for the quadrupolar $O_{2z}^2$ and $O_{xyz}$ order has been detected by the RXS measurements using a less-stress sample, in a surveyed region of $a^* - c^*$ plane including the prominent $Q$'s of (1, 0, 0) and (1.4, 0, 0). Nevertheless, we would reemphasize the fact that HO gives rise to almost no internal fields [14], and there remains a possibility of the quadrupolar order with a still unexplored $Q$ vector.

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