Lattice symmetry breaking at the hidden-order transition in URu_2Si_2

To elucidate the nature of a phase transition in materials, the most important step is to identify which symmetries are broken below the transition temperature. In 1985, a large anomaly in the specific heat was observed in the heavy-fermion metal, URu_2Si_2, at 17.5 K, indicating the presence of a phase transition at this temperature. Since then, tremendous efforts have been made to study the nature of this transition, but neither a symmetry change in the crystal structure nor a large magnetic moment that can account for the entropy change has been observed. This enigmatic order is thus called the "hidden order," and understanding this hidden-order transition is a long-standing issue in condensed matter physics [1].

Recently, magnetic torque measurements in small pure crystals under an in-plane magnetic field rotation have shown that the two-fold oscillation as a function of field angle in the *ab* plane starts to develop below the hidden-order transition temperature (T_{HO}). This two-fold oscillation indicates that the four-fold rotational symmetry associated with the body-centered tetragonal crystal structure (Fig. 1(a)) is broken in the hidden-order phase [2]. Similarly, cyclotron resonance [3] and nuclear magnetic resonance experiments [4] under an in-plane field rotation have provided evidence for four-fold rotational symmetry breaking below T_{HO}. However, these experiments were conducted under an in-plane magnetic field, which itself can break the rotational symmetry. Thus, direct observation of symmetry breaking in the absence of a magnetic field is required to identify the ground state of the hidden order.

To this end, we performed high-resolution synchrotron X-ray crystal structure analysis of URu_2Si_2 at beamline **BL02B1** [5]. We used ultraclean single crystals with very high residual resistivity ratios of ~670, which have become available recently [3]. First, out of more than ~30 crystals, we selected a high crystalline quality sample with the sharpest high-angle Bragg peak, which was measured using an imaging plate at room temperature. Then, we tuned the X-ray energy to 17.15 keV, which is just below the absorption edge of uranium so that the X-ray attenuation length is sufficient to obtain bulk information. To realize a high

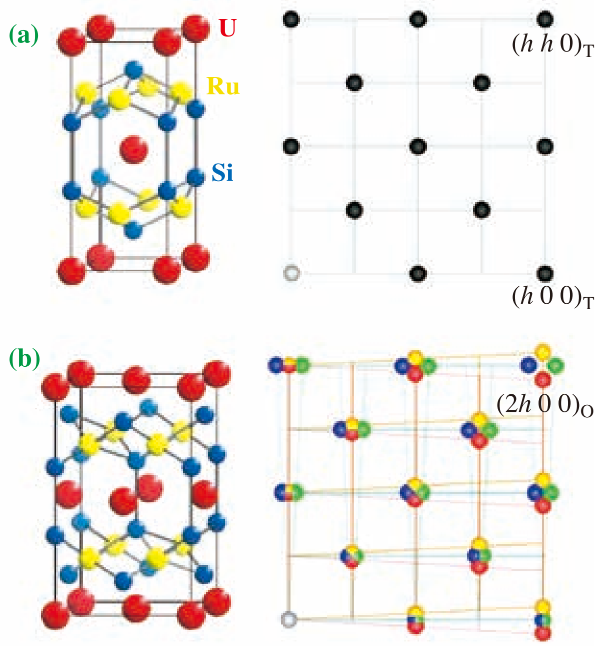

Fig. 1. **(a)** Crystal structure of URu_2Si_2 above the hidden-order transition temperature T_{HO}, which is *I*4/*mmm* body-centered tetragonal type. Schematic Bragg spots (black circles) in the ($h\,k\,0$) plane ($h, k \geq 0$) are also shown on the right. Left bottom is the origin. **(b)** Orthorhombic *Fmmm* structure in the ordered phase below T_{HO} identified in our study. Bragg spots split due to formation of the domains. Colored circles correspond to the four different domains.

resolution, we used a high-angle reflection set-up in which the four-circle diffractometer was equipped with a cryocooler. We focused on the high-angle $(880)_T$ Bragg peak at a reflection angle 2θ above 165 degrees, which corresponds to the experimental resolution of a lattice constant as good as 3×10^{-5}.

Figure 2(a) shows the temperature dependence of the $(880)_T$ Bragg peak measured by the 2θ/θ scattering mode. Below the hidden-order transition at T_{HO} = 17.5 K, clear peak splitting is observed, indicating that symmetry-breaking lattice distortion sets in just below the transition. We also performed two-dimensional scans in the (hk0) plane as shown in Figs. 2(b) and 2(c). The single peak at 19 K (above T_{HO}) clearly splits at 10 K (below T_{HO}). The integrated intensities of these data are identical within experimental error. Our analysis indicates that the profile at 10 K is consistent with the four-fold splitting of the single Bragg peak, as expected for the orthorhombic *Fmmm*-type crystal structure shown in Fig. 1(b) [5]. Hence, we conclude that the space symmetry of the hidden order belongs to this orthorhombic type, which breaks four-fold rotational symmetry. Our observation is fully consistent with previous high-field measurements [2-4], and indicates that rotational symmetry breaking is not field induced but is an intrinsic property of the hidden order.

To our knowledge, this is the first direct observation of a symmetry change in the hidden-order phase transition in URu_2Si_2 by scattering experiments. The clarified space symmetry places very strong constraints on the genuine hidden order parameter. Thus, the present results can be regarded as a big step toward the full resolution of this 30-year old mystery. In addition, we believe that understanding the origin of such spontaneous rotational symmetry breaking found in URu_2Si_2 may be important to uncover the nature of other unusual states of matter hidden in several strongly correlated electron systems.

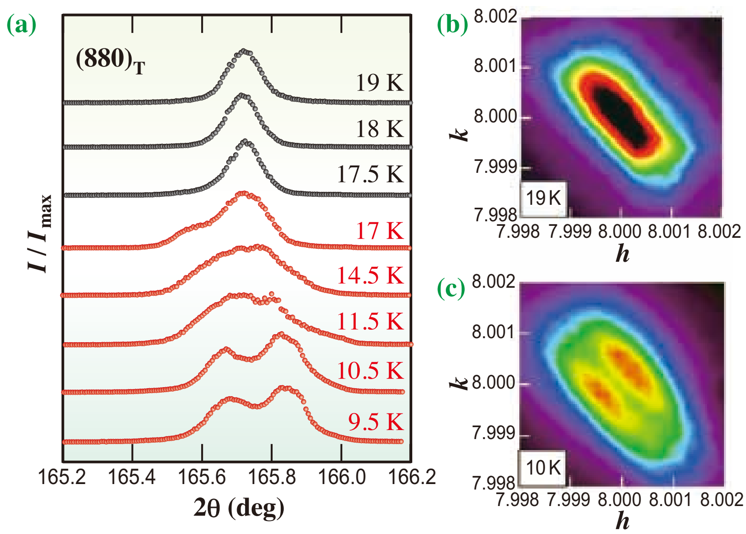

Fig. 2. **(a)** Temperature dependence of the Bragg peak $(880)_T$ in an ultraclean single crystal of URu_2Si_2. Each curve is shifted vertically for clarity. Below the hidden order transition at T_{HO} = 17.5 K, clear splitting of the Bragg peak is observed. **(b)** Two-dimensional intensity plot in the (hk0) plane near the $(880)_T$ Bragg peak at 19 K (above T_{HO}). **(c)** Similar data for 10 K (below T_{HO}).

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References

[1] See, for a review, J.A. Mydosh and P.M. Oppeneer: Rev. Mod. Phys. **83** (2011) 1301.
[2] R. Okazaki *et al.*: Science **331** (2011) 439.
[3] S. Tonegawa *et al.*: Phys. Rev. Lett. **109** (2012) 036401.
[4] S. Kambe *et al.*: Phys. Rev. Lett. **110** (2013) 246406.
[5] *S. Tonegawa, S. Kasahara, T. Fukuda, K. Sugimoto, N. Yasuda, Y. Tsuruhara, D. Watanabe, Y. Mizukami, Y. Haga, T.D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, Y. Matsuda and T. Shibauchi: Nat. Commun.* **5** *(2014) 4188.*