Search for Hidden Order Parameter of URu$_2$Si$_2$ by Neutron-Scattering Experiment under Uniaxial Stress

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Abstract. Elastic neutron-scattering experiment was performed on the heavy-fermion superconductor URu$_2$Si$_2$ under uniaxial stress in order to search a sign of a hidden order parameter. Contour maps of neutron-scattering at 1.5 K were achieved by mesh scanning. No additional magnetic signal exists in $(h0l)$ and $(hhl)$ planes except for a well-known weak antiferromagnetic signal with magnetic-propagation vector of $Q = (100)$ within the present experimental accuracy and scanned area.

1. Introduction

An elusive phase of URu$_2$Si$_2$ below $T_o = 17.5$ K is one of the most attractive interest in heavy-electron physics. URu$_2$Si$_2$ with a body-centered tetragonal ThCr$_2$Si$_2$-type structure was discovered as a heavy-fermion superconductor ($T_c \sim 1.4$ K) that coexists with antiferromagnetism with a simple type-I antiferromagnetic (AF) structure of $Q = (100)$ and an incredibly small staggered moment about 0.03 $\mu_B$/U below $T_o$ [1-4]. However, the tiny moment is incompatible with large bulk anomalies such as an entropy change at $T_o$ ($\Delta(C/T_o) \sim 300$ mJ/K$^2$mol). Recent progress for the paradoxical behavior was also obtained by microscopic experiments such as neutron-scattering (NS) [5, 6] and $^{29}$Si NMR [7] under hydrostatic pressure. These experiments suggest an existence of a first-order phase boundary around 0.5 GPa in a pressure-temperature ($p$-$T$) phase diagram of URu$_2$Si$_2$ between a nearly non-magnetic low-pressure phase and a high-pressure AF phase, which has a comparatively large-staggered moment of about 0.4 $\mu_B$/U and a modulation vector of $Q = (100)$. It is now understood that the weak-AF signal in low-pressure phase is extrinsic and originates in a little phase separation (mixing) of the high-pressure AF phase with large moment [6-8]. The order parameter in the elusive low-pressure phase has not been revealed yet.

In the present paper, we focus on a magnetic octupole model, which is based on the crystalline...
Table 1. Summary of experimental condition.

| Fig. # | Scattering plane | Scattering region, $q$ | Width of mesh, $\Delta_i$ | Area | $\sigma$ |
|--------|-----------------|------------------------|---------------------------|------|---------|
| (a)    | $(h0l)$         | (2.5→3.0, 0, 1.0→2.0)  | $\Delta_h=\Delta_l=0.025$ | 22.04| 0.2*, || [100] |
| (b)    |                 | (1.0→1.5, 0, 1.0→2.0)  | $\Delta_h=\Delta_l=0.0125$ |      |         |
| (c)    | $(hhl)$         | (0.5→1.0, 0.5→1.0, 1.0→2.0) | $\Delta_h=0.01, \Delta_l=0.025$ | 6.00 | 0.2*, || [110] |

* The magnitude of the stress were estimated from $\sigma = F/S$; $F$ is loading force, and $S$ is area of sample. A spread of the loading stress is about ±0.01 GPa in the experiment.

electric field and the group theory [9]. Although a direct observation of multipoles such as the octupole is generally difficult, experimental confirmations of a multipole order have gradually been increasing in recent years. Especially, the electric quadrupoles and the magnetic octupoles were observed directly or indirectly in (La,Ce)B$_6$ and NpO$_2$ etc. by means of resonant X-ray scattering and other microscopic techniques [10, 11, 12, 13].

The Kiss’s model suggests that the ‘hidden’ primary order parameter is $T^z$ or $T^{xyz}$-type octupole in URu$_2$Si$_2$. According to the model, an arbitrary magnetic dipole moment with the same $Q$ vector as a propagation vector of the octupole order is generated by applying uniaxial stress (but not hydrostatic pressure), which lowers the symmetry from tetragonal $D_{4h}$ to orthorhombic $D_{2h}$ in the ordered phase. However, quantitative predictions about the propagation vector $Q_{\text{oct}}$ and amplitude of the induced dipole moment were not described. To test this scenario, we searched the stress-induced dipole moment and its $Q_{\text{oct}}$ vector in some regions on reciprocal space by means of the NS experiment under uniaxial stress.

2. Experimental details

The elastic neutron-scattering experiments were performed on the triple-axis spectrometer ISSP-GPTAS (4G) located at the Japan research reactor JRR-3M of Japan Atomic Energy Agency in Tokai. Pyrolytic graphite PG(002) crystals were used for monochromating and analyzing the neutron beam with a momentum $k = 3.83$ Å$^{-1}$. We used a standard horizontal-collimation 40° - 80° - 40° - 80° and two pyrolytic graphite filters to reduce contamination from higher-order Bragg reflections.

A single-crystalline URu$_2$Si$_2$ was grown by the Czochralski-pulling method in a tetra-arc furnace. The grown single crystal was formed into two plates by means of spark erosion with parallel and flat surfaces with dimensions 22.04 mm$^2$ ($h0l$), and 6.00 mm$^2$ ($hhl$). The sample was installed in a $^4$He cryostat [14], which has pressure-tuning mechanism for loading uniaxial stress into the platy-formed sample by the stress method. Uniaxial stress was applied along [100] ($\perp (h0l)$), and [110] ($\perp (hhl)$) directions for the platy-formed ($h0l$) and ($hhl$) samples, respectively, and then scattering planes were taken in ($h0l$) and ($hhl$). We applied uniaxial stress $\sigma \approx 0.2$ GPa above $T_o$ in paramagnetic phase and cooled down to 1.5 K.

A stress-induced magnetic signal was searched by mesh scanning in some part of Brillouin zone of the scattering-planes ($h0l$) and ($hhl$). A width of the mesh, $\Delta_i$, was determined by measuring a full-width of half-maximum of nuclear Bragg peaks near the scan area, which is thought to be a resolution limit in the samples. Summary of the measurement condition is shown in Table 1.
Figure 1. Contour maps of the mesh scanning on the \((h0l)\) scattering plane for (a) and (b), and the \((hhl)\) for (c), respectively. Nuclear Bragg peaks exist at points of (301), (101) and (112). Clear peaks at (302), (102) and (111) are AF-Bragg peaks with \(Q = (100)\).

3. Results and Discussion
Results of the mesh scanning in some part of the Brillouin zone are plotted in (a), (b) and (c) of Figure 1. The vertical axis shows normalized counts of scattered neutrons, and the horizontal area corresponds to reciprocal-lattice space in \((h0l)\) or \((hhl)\) plane of \(\text{URu}_2\text{Si}_2\). Magnetic peaks were observed at (102) and (302) in the \((h0l)\) plane (in a and b), and at (111) in the \((hhl)\) plane (in c), respectively. These peaks exhibit the same \(Q\) vector as the well-known weak antiferromagnetism with \(Q = (100)\). In this experiment, estimated AF-scattering intensities that correspond to the volume-averaged staggered moment are \(\sim 0.07 \mu_B/U\) and \(\sim 0.12 \mu_B/U\) for \((h0l)\) and \((hhl)\) sample, respectively. These AF-scattering intensities at \(\sigma \sim 0.2\) GPa are roughly reproducing the results of previous NS experiments [8].

From the present results of the mesh scanning, we conclude that no additional magnetic signals are observed within the covered \(Q\)-range and the accuracy under uniaxial stress \(\sigma \sim 0.2\) GPa in the hidden ordered phase. Within the statistical analysis of a background and the weak-AF-peak intensities, a detection limit on present experiments are about \(1/2 - 1/10\) of the intensity of AF Bragg peak with \(Q = (100)\), namely an order of \(\sim 0.01 \mu_B/U\).

Finally, we comment a possibility that the octupole and the stress-induced dipole signal are obscured by the weak-AF signal, i.e. the hidden order parameter has the same propagation vector as the weak antiferromagnetism with \(Q = (100)\). In the previous NS experiments [8], the stress-induced AF-scattering intensities with \(Q = (100)\) varies in a linear fashion at least in a small stress region \(\sigma \leq 0.2\) GPa. This result might be able to take the induced-AF signal as the growth of the dipole moment that mixes with the spontaneous octupole moment as proposed by A. Kiss et al. [9]. However, we could not distinguish the information relating the growth of...
the dipole moment from the present data, which might be include development of the volume fraction of the large-moment AF phase. Therefore, to investigate the possibility of the octupole order with \( Q = (100) \), we should check the uniaxial-stress variation of the volume fraction of the AF signal, and of the growth of the dipole moment, separately by means of NMR and/or \( \mu \)SR measurements. It is now in progress.

4. Summary
In order to investigate the possibility of the octupole ordering in URu2Si2, we performed the NS experiment under uniaxial stress and searched the stress-induced dipole signal by mesh scanning in some part of Brillouin zone on the (h0l) and (hhl) planes. Under vertical-uniaxial stress \( \sigma \sim 0.2 \) GPa to the scattering planes, the contour maps of the neutron scattering indicate an absence of additional magnetic signal at 1.5 K except for the well-known weak-AF signal with \( Q = (100) \) within the present experimental condition and accuracy.

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