STUDY OF LATTICE MODULATION IN CeP UNDER HIGH PRESSURE AND LOW TEMPERATURE

Ce-monopnictides with NaCl-type structure possess various physical properties such as Kondo effect and heavy-Fermion state, even though the carrier number is 10^{-2} to 10^{-3} times less than that in usual materials [1]. Among them, CeP exhibits complicated magnetic phase behaviour when subjected to magnetic fields and high pressure (Fig. 1). It has been shown by neutron diffraction that there exist several kinds of long-period magnetic structures due to the arrangement of two kinds of magnetic moments [2,3]. A current theory describing electronic states in Ce ions asserts that a magnetic polaron effect associated with the localization of low concentration holes, along with *p-f* mixing, takes place in CeP. Furthermore, the ground crystalline field level changes from a Γ_7-state to a Γ_8-like electronic state in some Ce ions. If Ce ions with different electronic states were to be spatially organized, modulation in a crystal lattice would be observed. Using X-ray diffraction, we observed for the first time the appearance of this superlattice structure, a result of the periodic lattice modulation accompanied by the magnetic ordering in CeP under high pressure and low temperature.

X-ray diffraction studies were performed at beamline **BL02B1**. The experiments were carried out twice, using a double Si 111 monochromator and a double mirror. X-ray wavelengths of 0.41364(2) Å for the first experiment and 0.41295(2) Å for the second experiment were used. Calibration was performed using CeO_2 powder diffraction. A single crystal of CeP was settled in a diamond anvil cell. The initial pressures were 2.0 GPa (first experiment), and 1.16 GPa and 0.98 GPa (second experiment) at room temperature. The applied pressures were measured by ruby fluorescence. The diamond anvil cell was placed in a cryostat equipped with a newly developed see-through vacuum shroud and was kept monitored by a micro-telescope while the crystal was cooled down (Fig. 2).

Figure 3 shows the reciprocal lattice scans along the [h 0 0] direction measured at 23.5 K and 34.0 K. Pressure was at 2.0 GPa, when it was measured at room temperature before cooling. New peaks were seen at the positions of (8.5, 0, 0) and (9.5, 0, 0) at 34.0 K. Reflections expressed by (n+1/2 0 0) (n: integer) were observed at several

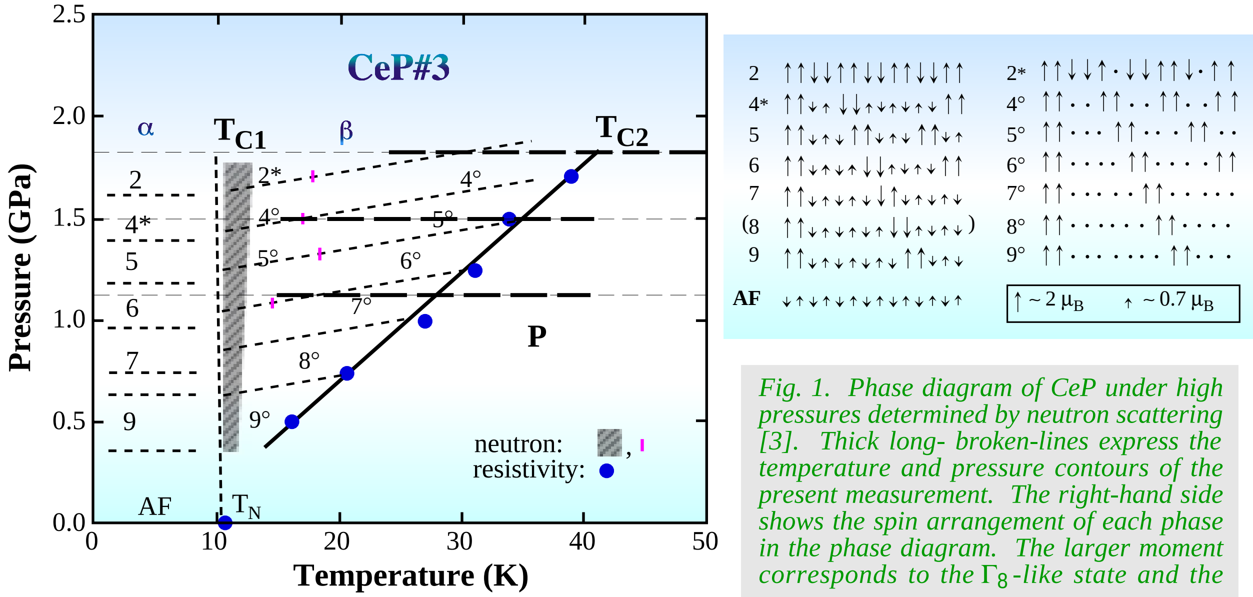

Fig. 1. Phase diagram of CeP under high pressures determined by neutron scattering [3]. Thick long- broken-lines express the temperature and pressure contours of the present measurement. The right-hand side shows the spin arrangement of each phase in the phase diagram. The larger moment corresponds to the Γ_8-like state and the smaller one corresponds to the Γ_7 state.

Fig. 2. A newly developed see-through vacuum shroud and radiation shields of a cryostat at BL02B1.

points in the reciprocal lattice. Since the symmetry of the lattice is an *fcc*, the reciprocal lattice point (9, 0, 0) is not the Γ-point but the zone boundary X-point in the Brillouin zone, so that the (8.5, 0, 0) point corresponds to q = 1/4 as the modulation wave in a primitive cell. The pattern of these superlattice reflections reveals that the Ce ion layer adopts a 4 period arrangement in the primitive cell. We have tried to measure a super-lattice reflection with 2q higher harmonics at (9 0 0) but we could not observe it probably because its intensity is too weak. Fig. 4(a) shows the temperature-induced change in the profile of the (8.5, 0, 0) superlattice reflection. The width of the superlattice reflection (8.5 0 0) is 0.005 in *h* of reciprocal unit, which is the same compared with that of the fundamental Bragg reflection (8 0 0). This demonstrates long-range ordering with the q=1/4 modulation. Figure 4(b) shows the temperature dependence of the integrated intensity of the (8.5 0 0) superlattice reflection. The superlattice reflection appears between T_L= 27.3 K and T_H= 41.7 K, where T_L and T_H indicate lower and higher phase transition temperatures, respectively. From Fig. 1, we can see that the 4 period phase of the magnetic order '4°' appears between 41.7 K and 33.0 K at 1.9 GPa. We estimated that the pressure in the low temperature region during this experiment is 1.9 GPa by comparing T_H in Fig. 4(b) with T_{C2} in Fig. 1. This novel reflection has a peak at 33 K, corresponding to a phase boundary between '2*' and '4°', and disappears upon cooling to 27.3 K. This result may indicate that a new phase, which has not yet been observed by neutron diffraction, appears in the temperature region from 27.3 K to 33.0 K. This point should be investigated more precisely in the future.

Similar results were obtained for pressures of 1.50 GPa and 1.15 GPa. The temperature dependence of the integrated intensity of (6+2/5 0 0) and (6+2/4 0 0) superlattice reflections at 1.50 GPa and that of (6+2/7 0 0) and (6+2/6 0 0) superlattice reflections at 1.15 GPa were measured. As shown in Fig. 1, the magnetic period changes from 5 to 4 times at about 16 K and 1.5 GPa At 1.15 GPa, the magnetic period similarly changes from 7 to 6 times at about 21 K. Both of these transitions are supported by the present experiment. Also, according to ref. [4], it is indicated that two phases coexist in the phase boundary, and it shows hysteresis. These are seen clearly in this measurement.

As a consequence, we confirmed the existence of the lattice modulation associated with the spin ordering in CeP at low temperature and high pressure. This modulation comes from the spatial ordering of different-sized Ce ions (Γ_7 and Γ_8 of 4*f* electrons) [5].

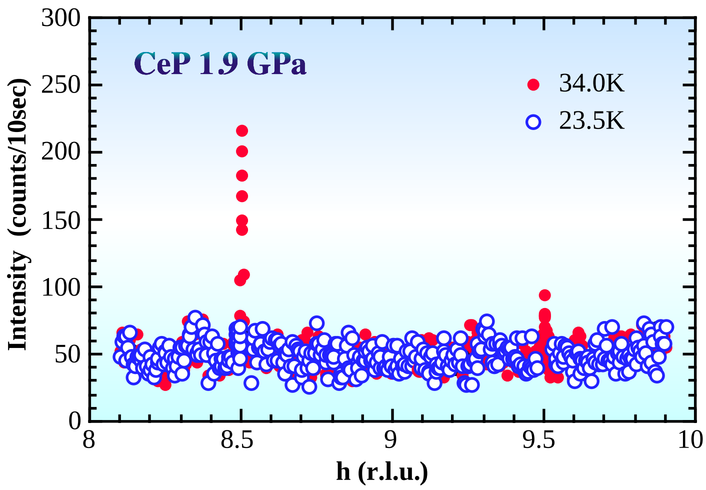

Fig.3. Scan along the [h 0 0] reciprocal lattice line at 1.9 GPa. Open circles show the scan at 23.6 K and the closed circles show the scan at 34.0 K, respectively. The superlattice reflections exist at (8.5, 0, 0) and (9.5, 0, 0) at 34.0 K.

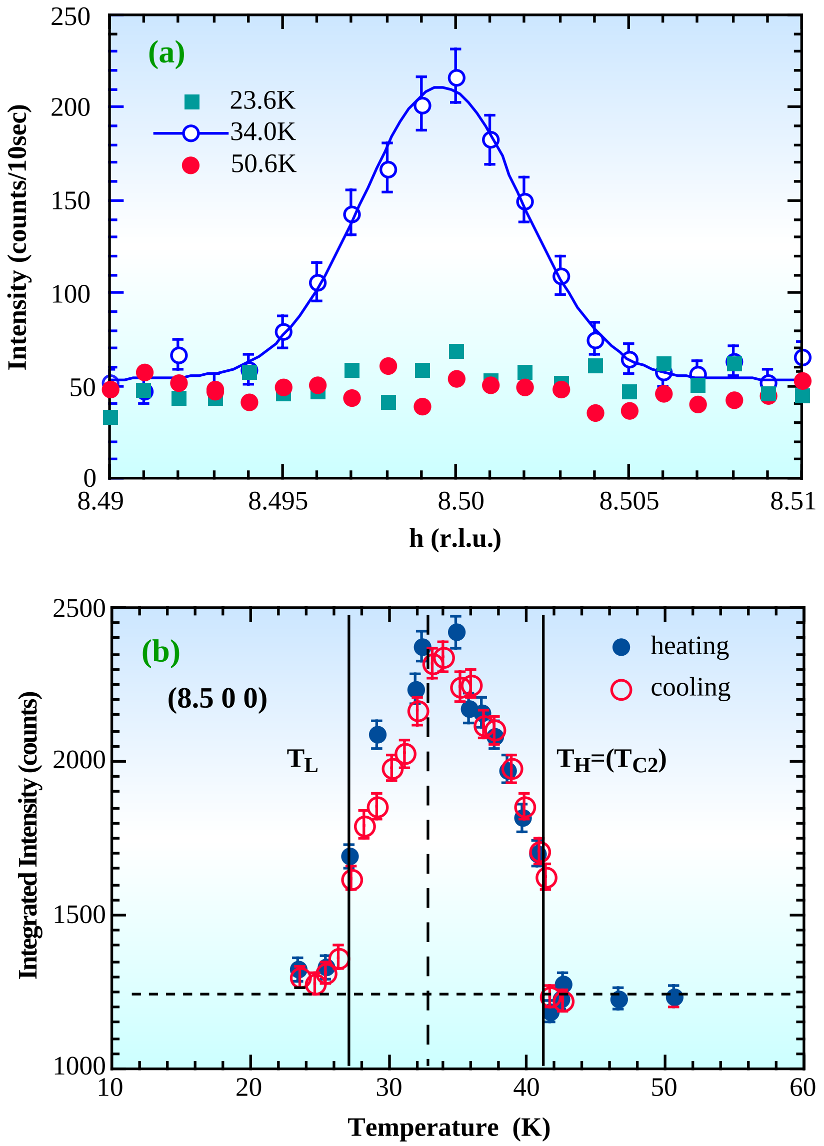

Fig. 4. ***(a)*** *Profiles of (8.5 0 0) reflection.* ***(b)*** *Temperature dependence of the integrated intensity of (8.5, 0, 0) reflection. Dashed line is the background.*

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