| ESRF | **Experiment title:**<br>Search for a Charge Density Wave in Cerium at High Pressure and Low Temperature | **Experiment number**:<br>HS-3900 |
|---|---|---|
| **Beamline**:<br>ID09a | **Date of experiment**:<br>from: 4 July 2009 to: 7 July 2009 | **Date of report**:<br>10 Sept. 2012 |
| **Shifts:** 9 | **Local contact(s)**: M. Hanfland | *Received at ESRF:* |

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## Report

At high pressure, in the range of 4–12 GPa, the element cerium adopts the same orthorhombic crystal structure as uranium at ambient pressure (α-U type, Pearson symbol *oC*4). α-U is well known to exhibit pronounced phonon anomalies and temperature-dependent mode softening, which leads to a series of charge-density wave (CDW) states at low temperatures below 43 K. Prior to the x-ray diffraction experiment HS-3900 on ID09a, we had investigated the lattice dynamics of the Ce-*oC*4 phase at high pressure and room temperature with inelastic x-ray scattering (IXS) on ID28 and observed pronounced phonon anomalies similar to those in α-U. This indicated the possibility that a CDW state may also exist in Ce at high pressure. Detailed structural studies at SRS, *performed at room temperature*, had not revealed any evidence of incommensurate satellite reflections. This had been not altogether surprising because the CDW in α-U is a low-temperature phenomenon. The principal aim of the experiment HS-3900 was therefore to extend the x-ray diffraction studies of cerium to low temperatures, by studying the high pressure *oC*4 phase of cerium down to 20 K at ~6 GPa, so as to look for the incommensurate satellite reflections that would arise from the appearance of a charge density wave.

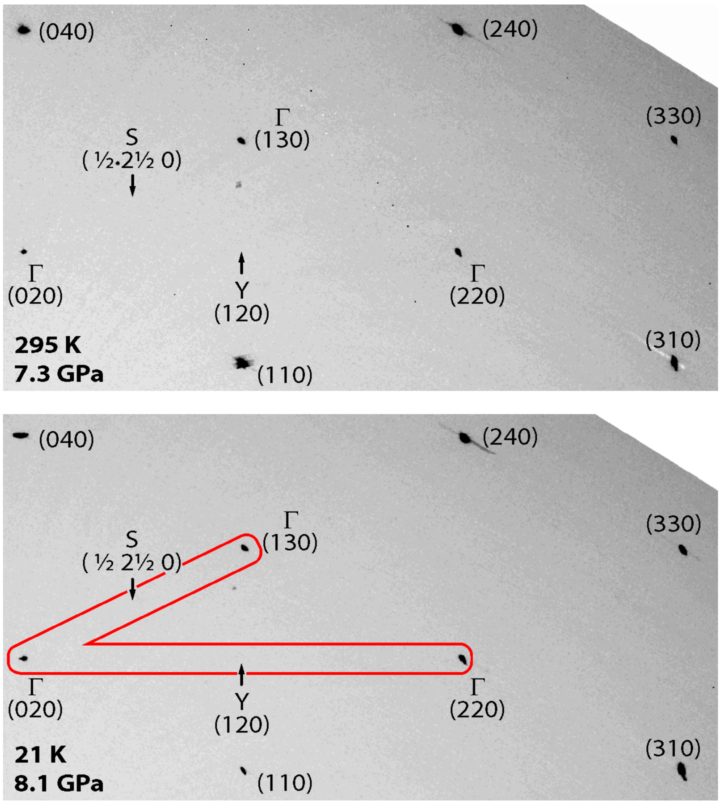


**Figure 1:** Reciprocal space mappings of the single crystal x-ray diffraction data of Ce-*oC*4 at high pressure and two temperatures, 295 K (top) and 21 K (bottom). The red outline marks the area of particular interest, around the S point of the Brillouin zone and along Γ–Y. There is no evidence for the appearance of satellite reflections after cooling.

A total of seven crystals of the high-pressure phase Ce-*oC*4 was produced for this experiment. The crystals were obtained by first cutting small chips from an ingot of distilled Ce that was immersed in mineral oil to minimize the formation of cerium oxide and hydroxide. Individual oil-covered chips were then loaded into Merrill-Bassett-type diamond anvil cells (DACs) with a 4:1 methanol/ethanol mixture as the pressure-transmitting medium. The pressure was then slowly increased at room temperature to above 6 GPa, where the chips of fcc Ce (α-phase) with a polycrystalline microstructure transform to single crystals of Ce-*oC*4.

The quality of the crystals was confirmed with single-crystal x-ray diffraction on ID09a. One of the cells with a Ce-*oC*4 crystal was then mounted in a He-flow cryostat and cooled to 50 K. Unfortunately, it turned out that the pressure in the DAC increased quite dramatically upon cooling — much more than expected based on previous experience — from 7 GPa at room temperature to 16 GPa at 50 K. Because of the usual pressure-induced phonon hardening we could not expect to observe a structural distortion or CDW at such high a pressure, and the run was aborted.

In three subsequent runs, using other DACs and new samples, we tried to limit the increase in pressure during cooling. The best result was obtained by using a gas membrane to adjust the pressure in the DAC, which allowed us to at least limit the pressure increase to 0.8 GPa. We were then able to collect several single-crystal diffraction data sets while the sample was cooled down to 21 K, and selected results are shown in Fig. 1. The reciprocal space mappings of the single-crystal data show the diffraction from the (*hk*0) layer of Ce-*oC*4 at room temperature ($P = 7.3$ GPa) and 21 K ($P = 8.1$ GPa). There is no sign of any additional reflections at low temperature.

After this experiment, we have performed additional electronic structure calculations [1], and they indicate that in Ce-*oC*4 phonon anomalies exist not only halfway along the Γ–Y line, like in α-U and as observed in our IXS experiments, but also near the S point of the Brillouin zone, **q** = (½ ½ 0). Here, one of the transverse acoustic branches shows soft-mode behaviour, and this is predicted to lead to a dynamical instability below 4.3 GPa [1]. These extended calculations predict also that the soft modes along the Γ–Y line lead to a dynamical instability only below 4 GPa. It is thus clear now that the pressure in our low-temperature diffraction experiment was still too high, and that better control over the sample pressure during cooling is needed to perform this experiment successfully.

Since we performed experiment HS-3900, significant progress has been made on the experimental side: On ID09a, mechanically-driven Stuttgart-type diamond anvil cells are now available for use in a cryostat. In future experiments, this will allow us to precisely control the pressure while the cell is cooled down and also to change the pressure on the sample at low temperature.

## Reference

[1] I. Loa, E. I. Isaev, M. I. McMahon, D.-Y. Kim, B. Johansson, A. Bosak, M. Krisch, Phys. Rev. Lett. **108**, 045502 (2012).