

Experimental report

09/08/2024

Proposal: 5-21-1184

Council: 4/2023

Title: Crystal Structure of Cerium Tantalum Nitride Perovskite

Research area: Materials

This proposal is a new proposal

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Samples: CeTaN₃ζx

| Instrument | Requested days | Allocated days | From | To |
|------------|----------------|----------------|------------|------------|
| D20 | 1 | 1 | 01/09/2023 | 02/09/2023 |

Abstract:

ABN₃ nitride perovskites are rarely reported. We propose to study the structure of bulk-phase nitride perovskite CeTaN₃ζx, which we prepared with a high-pressure reaction. The laboratory X-ray powder pattern can reasonably be fit with a cubic model (SG Pm3̄ζm, a = 4.009423 Å) but peak broadening on several reflections, e.g. <200>, <211>, suggest a structural distortion. We have measured the magnetic susceptibility of the as prepared CeTaN₃ζx, showing Curie-Weiss behaviour with an effective magnetic moment of 1.7 μB. The observed moment corresponds to a composition CeTaN₃ζx with 0.15 < x < 0.17. Powder neutron diffraction (PND) is required to investigate the octahedral tilt structure and to determine the nitrogen positions as well as occupancies in CeTaN₃ζx due to the high sensitivity of neutron for nitrogen. The D20 beamline of the ILL is optimal as the high neutron flux enables data collection from a small sample ~100 mg obtained from high-pressure experiments. Low temperature data will also help to decorrelate N-occupancies and thermal factors.

Experimental

Powder neutron diffraction was carried out on ~100 mg samples of CeTaN₃ on the D20 High-intensity two-axis diffractometer in order to obtain a nuclear structure model. Samples were loaded into small vanadium can sample holders. For the nuclear structure determination, temperature-dependent powder patterns were collected at 300K, 250, 200, 150, 50, and 10 K in high-resolution mode with a neutron wavelength of 1.54 Å. Temperatures were attained using a cryostat.

Results

High-quality powder patterns of CeTaN_{3-x} were obtained from diffraction experiments (Figure 1b). Small amounts of boron nitride were present from sample preparation, as well as contribution from the vanadium can was observed in the powder patterns. The structure refinement of CeTaN_{3-x} was tentatively tried in orthorhombic space group *Pnma* (no. 62) after refinement in the cubic aristotype structure was unsuccessful as several reflections were not explained by this model. The orthorhombic model allows an octahedral tilt structure shown in Figure 1a. However, the powder pattern could not fully be model with this structural model. This indicates a more intricate structure, which could not yet be solved from the neutron data alone. The presence of several unexplained reflections and the splitting of reflections may indicate a larger superstructure. For the determination of the structure metrics, synchrotron or single-crystal diffraction may be necessary.

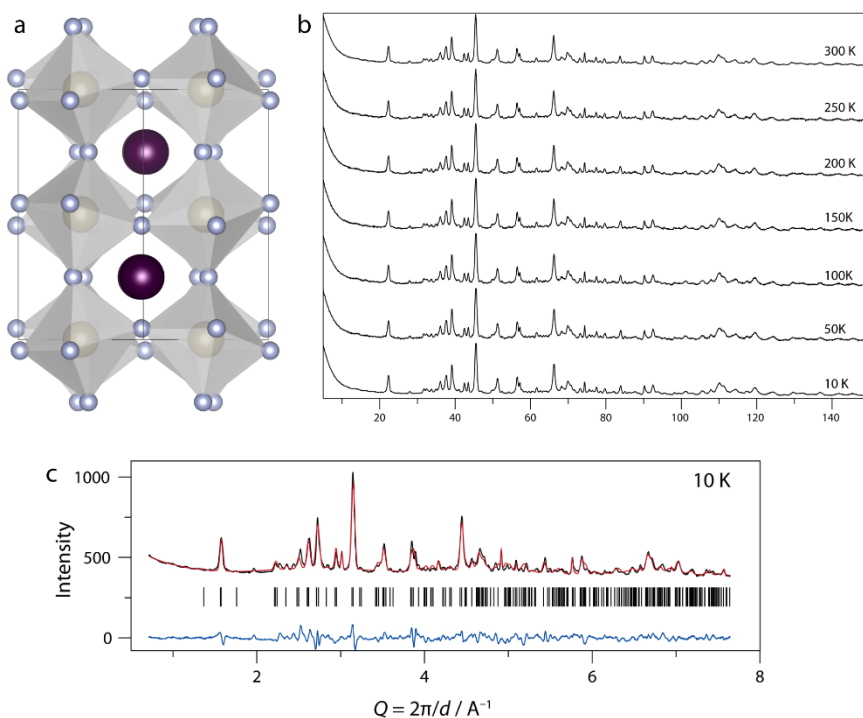


Figure 1: a) Tentative structural model of CeTaN_{3-x} obtained from powder neutron diffraction data at 10 K. b) Collected neutron diffraction data during the beam time. c) Rietveld refinement of the 10 K data, where the black line is the collected data, red is the refined data, blue is the difference data and vertical bars indicate theoretical Bragg positions.