Experimental report

30/04/2019

Proposal: **6-06-471** **Council:** 10/2016

Title: Local atomic and magnetic order inuranium oxide at low temperature

Research area: Physics

This proposal is a continuation of 6-06-453

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Samples: U_4O_9 (depleted uranium oxide)
UO_2 (depleted uranium oxide)

Instrument	Requested days	Allocated days	From	To
D4	6	5	28/05/2018	04/06/2018

Abstract:

Uranium oxide exhibits a complex phase diagram as a function of oxygen content and temperature, including magnetic phases at low temperature. In particular, the composition UO2 undergoes a first-order magnetic phase transition at T_N = 30.8 K that is accompanied by a structural distortion involving the oxygen atoms consistent with the Pa-3 space group, as opposed to the Fm-3m space group hitherto deduced for the high-temperature phase based on standard Rietveld refinement methods of diffraction data.
We have however evidence from PDF-analysis experiments on UO2 that the Pa-3 local structural symmetry should still exist, at least dynamically and perhaps statically, well above room temperature, and in very small domains that would be averaged-out out by Rietveld analysis. We propose a structural and magnetic PDF-analysis experiment on UO2 at low temperature in order to shed new light on the relation between local structural and magnetic order, including the phase transition at 30.8 K.

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Two samples were analysed during this experiment: a U_4O_9 sample and a UO_2 sample.

The U_4O_9 sample had already been analysed at ILL in a previous experiment. It consisted in a powder sealed in an airtight vanadium sheath, which we could not open after the previous experiment. Therefore, the powder sample and its vanadium sheath were taken altogether for this new experiment. We first performed measurements with an empty cryostat and with an empty vanadium sheath, at 10, 50, 100, 200 and 300K, that we have used for further background corrections. We then measurement the U_4O_9 sample in D4 cryostat at the following temperatures: 300, 70, 50, 35, 20, 10 K. A first analysis of the data showed that the experimental procedure was fine for data acquisition but that the U4O9 data were somehow complex to interpret. That is why the analysis of U_4O_9 data was postponed and the experiment then turned to UO_2 that was the focus of this experiment.

The UO_2 sample consisted in two ceramic polycrystalline pellets, previously sent from CEA to ILL, that were stacked into a vanadium sheath by the ILL SPR. The sample was set in D4 cryostat and the diffraction pattern was measured at the following temperatures: 300, 250, 200, 150, 100, 75, 50, 35, 20 and 10 K down, then 35K, and finally 25, 27.5, 30, 32.5, 40, 60, 175 up. A quick analysis of the results evidenced that we observed between 30 and 32.5K the magnetic transition reported in literature. Moreover, a remaining magnetic signal is still observed for T higher that the transition temperature (figure 1): this result is now analysed.

The average experimental time for each diffraction pattern varied between 3 and 6 hours.

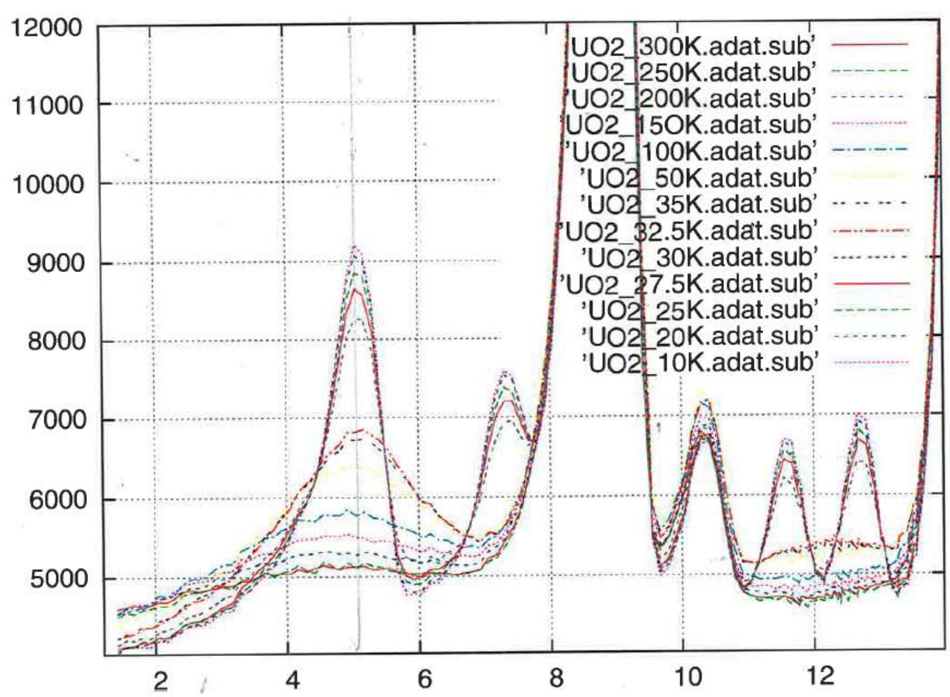

Figure 1: diffraction patterns of UO_2 at several temperatures between 10 and 300K. The peaks corresponding to the antiferro-magnetic phase are clearly visible at ≈ 5, 7.4, 11.5 and 12.6 °2th for temperature less than 30K. They become broader and less visible for temperature higher than 32.5K.