Experimental report

24/01/2018

Proposal: **DIR-143**

Council: 4/2015

Title: Neutron Laue Diffraction study with CYCLOPS of satellite reflections in Aplha-U at very low temperature

Research area: Physics

This proposal is a new proposal

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Samples: alpha-U depleted
concentration 235U: #0.4%

Instrument	Requested days	Allocated days	From	To
CYCLOPS	7	7	07/06/2016	14/06/2016

Abstract:

RAPPORT DIR-143.docx 24 January 2018
CYCLOPS

Neutron diffraction of α-U at very low temperature

The goal of the experiment was the search for the diffraction signature of the superconductivity transition at 480 mK and/or an a might be extra displacive phase instability.

The structural and lattice dynamical aspects of the incommensurate phase transition in α-U (T_c = 43 K) have already been studied by means of elastic [1] and inelastic neutron scattering [2]. The soft-mode transition and the phonon with the smallest energy were found to be located at $\mathbf{q}_{min} = \mathbf{q}_{cdw} = <q_x, q_y, q_z>$ incommensurate with the main lattice, with (: α_1), changing to (: α_2 with $<1/2, q_y, q_z>$) at 37 K and then to (: α_3 with $<1/2, 1/6, q_z$) at 22 K.

The goal here was to explore the 200 mK to 1.5 K domain to observe new satellites and shed some lights on the low temperature structure of α-U.

Sample
The sample was a cylindrical single crystal produced by grain coarsening (Fisher's sample n° 219-2, containing Si & Fe impurities, ILL storage number: 884) with dimensions Ø = 3.34(6) mm, h = 2.25(2) mm, m = 375(13) mg, v = 19.71(70) mm^3 and an activity of 9 kBq.

Experiment preparation

1- Uniaxial clamp pressure cell
In an effort to minimize the Kapitza resistance that could impair reaching the target temperature of 200 mK, a special sample holder able to accommodate an Orange cryostat was designed at the Institut Néel/MCBT/CNRS. It was calibrated to ensure a pressure well below the crystal deformation limit.
2- Cryostat
The RuO_2 thermometer of the Orange cryostat was calibrated by J. Bossy & X. Tenon (SANE) (10-13/05/2016) to account for the temperature shift introduced by the uniaxial clamp when attached to the dilution insert.
3- Chemical cleaning
The surface of the α-U sample is naturally covered with a 0.1 μm UO_2 layer, a thermal insulator that must be removed to reduce the Kapitza resistance.
With the help of Robert Dixon (SPR) and D. Heiss (ILL chemist), the end faces of the sample were chemically polished at the Alpha Lab using nitric acid at 30% concentration.
4- Mechanical polishing
A mechanical polisher was placed in the glove box of the Alpha Lab and used to shape the end faces of the sample. We used silicon carbide paper P600: 25 μm; P2400: 10 μm; P4000: 5 μm (supplied by J. Debray, CNRS/PLUM). The polished faces showed a microstructure (Fig. 1) but they seemed quite flat and parallel at a visual inspection.
5- Thermal contact
The crystal was immediately placed in the pressure clamp (Fig. 2) with the [010] direction roughly along the sample rod axis. The misalignment was of about ± 15° as indicated by prior OrientExpress tests.
6- Sample storage
The pressure clamp with the crystal inside were placed in a desiccator (24/05/2016) and stored in the Alpha Lab. until the experiment (06/06/2016):

The experiment n° DIR-143 on CYCLOPS (cycle n°178)
The room temperature diagrams immediately showed that the sharp Bragg peaks from the unperturbated sample (test experiment n° TEST-2536) were now much broader. The sample had developed in the clamp several grains (2 or 3) misaligned by about 1°.

Our analysis is that the amplitude of a "safe" uniaxial pressure was calculated for flat and parallel surfaces but a posteriori checks showed that one of the polished face was substantially curved. Thus the stress may have exceeded the plastic limit for some parts of the crystal. We clearly made the mistake of not having an optical standard for flateness control during the polishing phase!

The disappointment was huge but we thought that this would not totally hampered our chances to observe satellite reflections below T= 45 K.
Xavier Tonon and colleagues did a great job with the dilution insert. The target temperature was reached in no time and the temperature of the sample stabilised rapidly with an offset to the setpoint of only 20 mK (beam closed) and 55 mK (beam opened). In other words we can assume that the Kapitza resistance was reasonably small. During the experiment, the cryogenic setup proved to be very stable and reliable.
Unfortunately, the data processing showed that the satellites were much too blurred by the peak broadening to be observable without ambiguity. We must state here that during TEST-2536, the same sample, but unperturbated, showed many easy to observe satellites.

Conclusion

The experiment failed due to the damage to the sample but demonstrated the following:

- the Kapitza resistance was not a blocking point
- the cooling power of the insert is large enough to compensate for the neutron heating due to fissions in the uranium sample
- working at very low temperature on CYCLOPS is quite confortable.

The next step now is the study of a better sample holder that both respects the crystal integrity and offers a low enough Kapitza resistance.
In other words the experiment is worth a new try. We already have an excellent spare α-U sample was given back from Los Alamos. Since α-U single crystals are quite rare, we plan the annealing of the damaged sample under neutron control (CYCLOPS) to cure it.

References

[1] J. C. Marmeggi, G. H. Lander, S. van Smaalen, T. Brückel and C. M. E. Zeyen, Phys. Rev. B **42**, 9365 (1990).

[2] J. C. Marmeggi, G. H. Lander, A. Bouvet and R. Currat, J. of Physics: Conf. Series **92**, 012173 (2007); J. C. Marmeggi, P. Haen, A. Filhol, P. Bastie, J. of Physics: Conf. Series **592**, 012035 (2015).

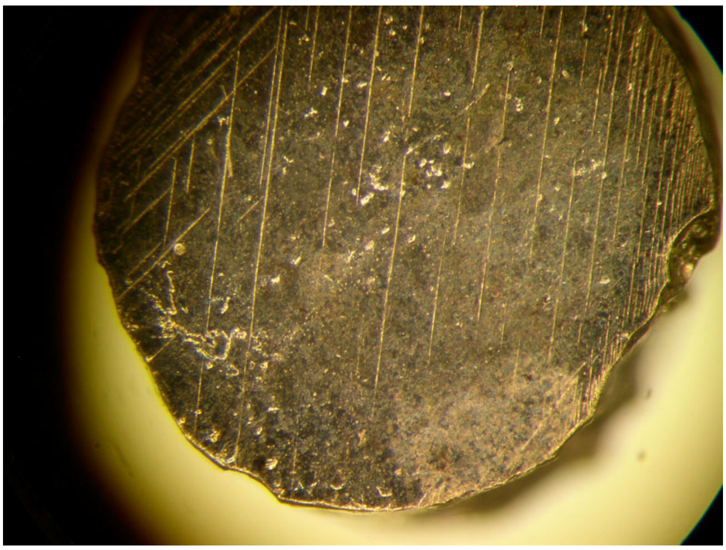

Fig. 1: Microstructure of one end of the sample; light lines are twin bands.

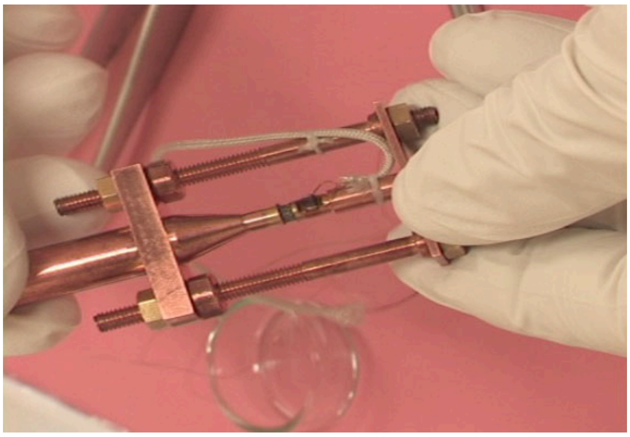

Fig. 2: The calibrated pressure clamp.
Pressure strength: F = 1.6 KN/turn; the applied strength was about 300 bar. On can see the RuO_2 thermometer sunk in epoxy resin (Stycast) located close to the sample.

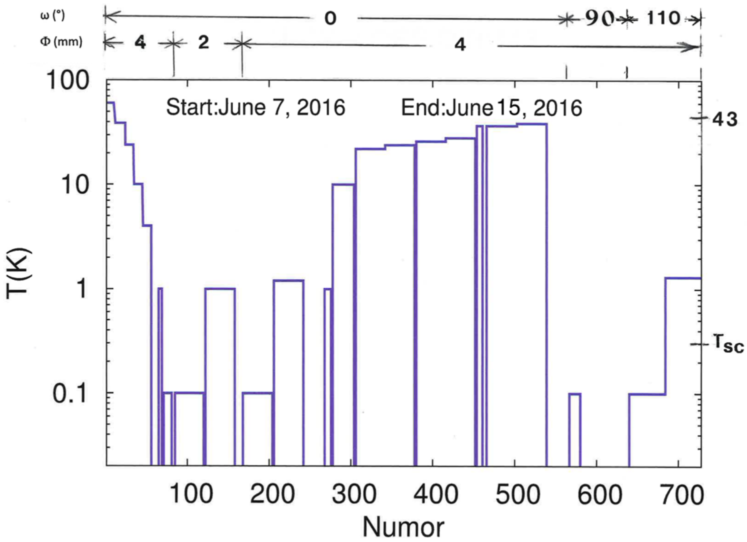

Fig. 3: Histogram plot of the low temperature experiment on CYCLOPS.
Each Numor corresponds to a data acquisition of 15'; the instrument parameters which were changed are: the diameter of the neutron beam Ø, the orientation of the crystal, ω. The transition temperatures are: 43 K, 37 K, 22 K and T_{SC} ~ 480 mK.