Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF.

Once completed, the report should be submitted electronically to the User Office via the User Portal:

https://wwws.esrf.fr/misapps/SMISWebClient/protected/welcome.do

Reports supporting requests for additional beam time

Reports can be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

Reports on experiments relating to long term projects

Proposers awarded beam time for a long term project are required to submit an interim report at the end of each year, irrespective of the number of shifts of beam time they have used.

Published papers

All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

ESRF

	Experiment title: Time resolved x-ray diffraction study of the newly discovered hidden quantum state in 1T-TaS2	**Experiment number**: HC2540
Beamline: ID09B	**Date of experiment**: from: 06/07/2016 to: 12/07/2016	**Date of report**: 23/08/2016
Shifts: 18	**Local contact(s)**: Norman Kretzschmar	*Received at ESRF:*

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Report:

The aim of this experiment was to study the microscopic structure of the newly discovered so-called hidden quantum state of 1T-TaS_2 by means of static and time-resolved elastic x-ray diffraction. Using resistivity and optical reflectivity measurements it was found that ultra-short laser pulses induce a new charge density wave (CDW) phase in 1T-TaS_2, which is persistent and does not exist under normal equilibrium conditions.

Up to now nothing is known about the microscopic structure of this new laser-induced CDW. Hence, our goal was to use x-ray diffraction (XRD) in order to determine the modulation wave vector (q-vector), the correlation length and amplitude of the hidden CDW state. Note that we had a preliminary attempt for this experiment in May 2015 (see experimental report HC1957). In the following paragraph we will explain the improvements and achievements that have been made with respect to this previous experiment.

Starting from high quality bulk single crystals we prepared thin films of 1T-TaS_2 by mechanical exfoliation. These samples were deposited on 200 nm thick $Si0_2$ membranes spanning a 1 x 1 mm^2 hole in a silicon waver. The thickness of the films was about 100 nm and typical lateral dimensions were 200 µm. The SiO_2 membranes provided much better support for the 1T-TaS_2 thin films then the previously used TEM grids and, hence, significantly improved the quality of the XRD data. In order to measure the electrical resistivity *in-situ* it was planned to fabricate contacts on the thin film samples using electron beam lithography. However, the fabrication of these contacts is very complex and demanding and needs to be developed iteratively. We managed to produce the first reliable contacts only several weeks after the beamtime, so that at the time of the beamtime the contacts were not available. Nonetheless, in order to get at least a qualitative macroscopic signature of the hidden state we contacted the samples using conductive paint which allowed for two point measurements (see figure 1 (a)).

The silicon waver was then clued on a sample holder which was mounted on the coldfinger of a continuous flow helium cryostat (figure 1 (b)). We passed a current of 10µA through the sample using a Keithley 6220 current source and measured the voltage with a Keithley 2000 multimeter. A simple one-circle goniometer was used to rotate the sample.

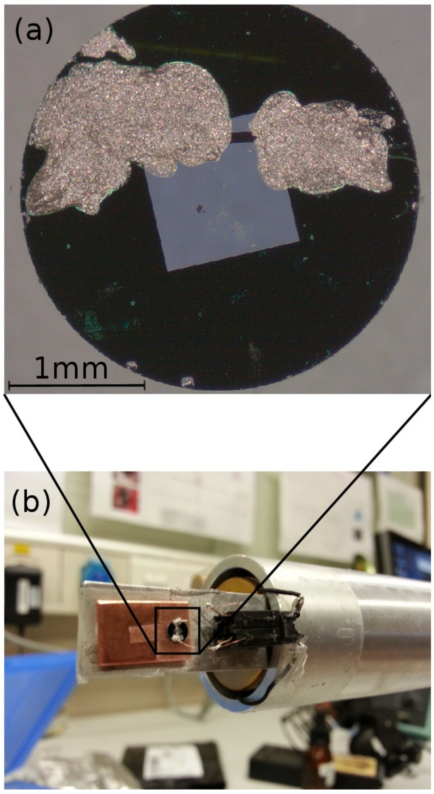

Figure 1: (a): 1T-TaS_2 sample on a SiO_2 membrane with conductive paint patches. (b): sample holder with contacted sample mounted on the cryostat cold finger.

The sample was exposed to a (60 x 100) µm^2 X-ray beam with a photon energy of 18keV. The XRD from the sample was collected with a rayonix MX170 detector. In order to optically pump the sample a Ti-sapphire laser with a wavelength of 800nm and pulse lengths of about 1ps was used. The laser beam had a size of about (1 x 1) mm^2. At first we checked the crystallographic quality of our samples at room temperature. Therefore, we collected single crystal datasets consisting of up to 160 images measured over a sample rotation of 40 degree. Typical reciprocal space maps deduced from these datasets are shown in the Figure 2 (a, b). Next we cooled the sample slowly down to 5K (1K/min) while measuring the resistivity. However, the conductive paint contacts were not stable enough and broke during cooling which prevented the resistivity measurement. We tried several cooling cycles with different samples but the contacts always failed. At 5K we again collected datasets over a sample rotation of 40 degree (see Figure 2 (c, d)). As in our earlier attempt, the data clearly confirm the high crystallographic quality of our thin film samples. In our previous beamtime (HC1957) we used a helium CryoJet to cool the sample which turned out to be inappropriate as it could not reach the needed temperature below 30K and lead to shacking of the sample which disturbed the XRD measurement. These problems have indeed been solved by using the cryostat which provided a very stable sample temperature without disturbing the diffraction experiment. With this improvement and the use of the SiO_2 membranes as sample substrates, the experiment yields the desired *q*-space resolution also for temperatures below 5K which is necessary in order to study the switching into the hidden state. We could observe the expected superlattice peaks corresponding to the nearly commensurate CDW (Figure 2 (a, b)) and the commensurate CDW (Figure 2 (c, d)) at room temperature and 5K, respectively. In particular the intensity distribution along the crystallographic *l*-direction, i.e. perpendicular to the TaS_2 -layers, is well resolved (see Figure 2 (b, d)). These information are indispensable in order to elucidate the CDW layer stacking order and are commonly difficult to obtain from electron diffraction.

However, technical difficulties with the beamline delayed the actual setup of this complex experiment by 5 shifts. We conducted several cooling cycles with small cooling rates in order to prevent breakage of the conductive paint contacts which also took a substantial amount of time. In addition, we had to ascertain that the cryostat mount was not stable enough so that rotating the sample led to unreproducible movements.

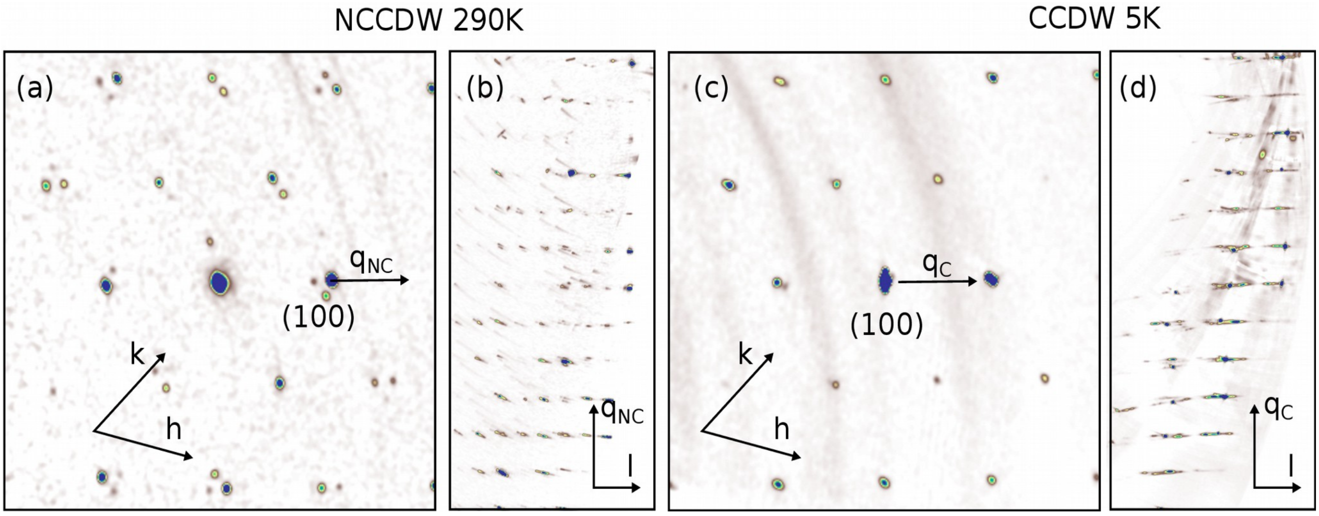

Figure 2: Reconstructed reciprocal space maps at room temperature ((a) and (b)) and 5 K ((c) and (d)).

Therefore, the actual measurements took much longer than expected because the sample position needed to be realigned every two degrees. With the little amount of time available to us it was not possible to reinforce the cryostat mount. In the end, we had only the last two shifts left to optically pump the sample. However, due to misalignment between the position of laser and x-ray beam we could not conduct the planned measurement . Nonetheless, this and our previous attempt clearly show that the proposed experiment is very well feasible. Many technical difficulties have been solved in this second attempt and we are in close contact with the beamline staff in order to address the remaining issues. It should be noted that the development of such a highly nonstandard and complex experiment is very challenging and certainly needs some iterations.