EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

ESRF

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

Deadlines for submission of Experimental Reports

Experimental reports must be submitted within the period of 3 months after the end of the experiment.

Experiment Report supporting a new proposal ("relevant report")

If you are submitting a proposal for a new project, or to continue a project for which you have previously been allocated beam time, you must submit a report on each of your previous measurement(s):

- even on those carried out close to the proposal submission deadline (it can be a *"preliminary report"),*
- even for experiments whose scientific area is different form the scientific area of the new proposal,
- carried out on CRG beamlines.

You must then register the report(s) as "relevant report(s)" in the new application form for beam time.

Deadlines for submitting a report supporting a new proposal

- 1st March Proposal Round - **5th March**
- 10th September Proposal Round - **13th September**

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.

Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report in English.
- include the experiment number 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:** Electronic order and superconductivity in 1T-IrTe2 nano-flakes	**Experiment number**: HC-5276
Beamline: ID27	**Date of experiment**: from: 25.04.2023 to: 29.04.2023	**Date of report**: 5.9.2023
Shifts: 12	**Local contact(s)**: Gaston Garbarino, Bjorn Wehinger	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Experimental Report:

Summary:

The aim of this experiment was to investigate how the atomic structure and electronic order in exfoliated flakes of 1T-$IrTe_2$, a layered material with weak interlayer bonding, changes with different flake thicknesses. To this end we wanted to study the pressure-temperature-thickness phase diagram using single crystal x-ray diffraction (XRD). On the more general site, this experiment was also intended to demonstrate that the upgraded ESRF machine in combination with the new ID27 beamline is indeed capable to measure XRD data from nanoflakes with lateral dimensions in the range of 10µm and thicknesses well below 50nm making it a unique tool to explore how the complex electronic pressure-temperature phase diagrams in the wide class of layered vdW materials evolves when the thickness of these materials is reduced towards the 2D limit.
We were indeed able to not only measure the Bragg peaks in flakes with thicknesses below 20nm, but also the superlattice reflections stemming from the subtle lattice distortions due to the electronic order in these materials. To our knowledge this was the first time such a challenging experiment has been conducted, hence this result alone is a significant success, as it shows that such experiments have become possible with the ESRF upgrade. However, we were also facing a number of solvable technical problems (explained in more detail below) which, in the end prevented the intended quantitative mapping of the pressure-temperature-thickness phase diagram. Thus, the scientific case is yet to be resolved.

Details of the actual Experiment

To prepare the samples, we used the scotch tape method to peel off thin flakes from a high-quality bulk single crystal onto a silicon substrate. The thickness of a few flakes has been measured with an atomic fource microscope in order to correlate the optical contrast on the silicone substrate to the thickness of the flakes. Based on this optical appearance we than chose flakes with different thicknesses and used a dry transfer technique employing a Polycaprolactone (PCL) polymer stamp to pick up the flakes and position them on a diamond of a diamond anvil cell (DAC) to compare their structural evolution as a function of temperature and pressure. In addition to the proposed material 1T-$IrTe_2$ we also prepared flakes from the related material 1T-TaS_2.
In order to facilitate the alignment of the DAC we put a gold particle next to the flakes. A ruby which we wanted to use as a pressure gauge was also placed

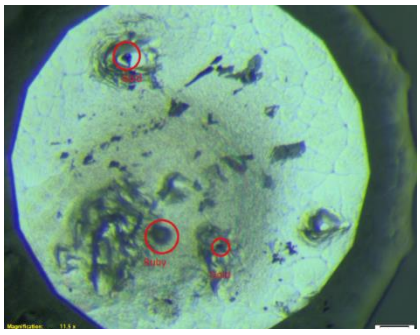

Figure 1: Microscope image of 1T-TaS2 flakes covered with a thin layer of PCL polymer.

on the diamond anvil. An image of the flakes covered with PCL with the ruby and gold particle next to it is shown in Fig. 1.
Initially we planned to measure the ambient-pressure data on flakes prepared on a TEM silicone nitride membrane (cf. proposal) to minimize the background signal as much as possible. However, this step would have required to use a different cryostat and since we only had four days of beamtime we decided to start with the DAC cryostat right away in order to avoid the time loss due to a change of setup.

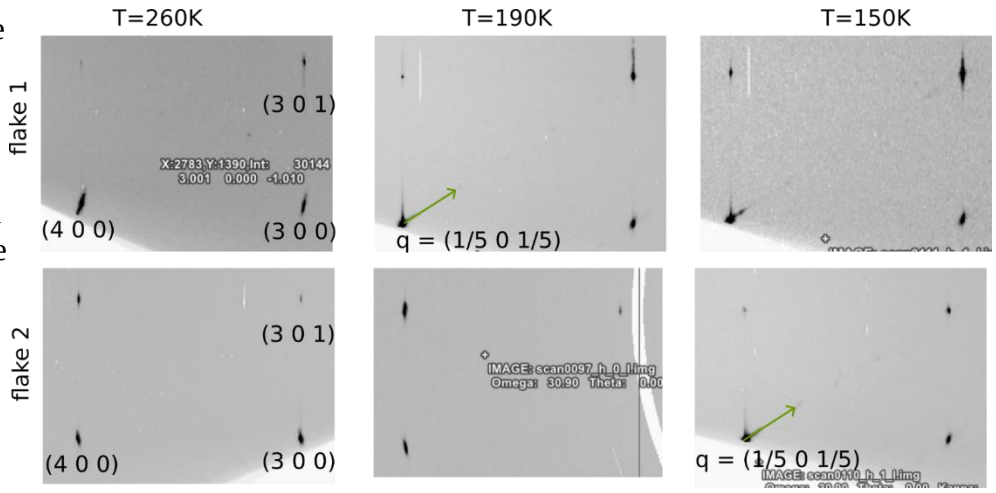

Figure 2: Reconstructed h1l-planes for two IrTe2 flakes with different thickness (flake 1: ~120nm and flake 2: ~40nm) as a function of temperature.

Thus, we started the experiment with 1T-TaS_2, as the flakes from this material have generally larger lateral dimensions. We used the KB2 optics at 30 keV photon energy and a beam focus of about 500nm diameter. Using the microscope camera of the ruby fluorescence spectrometer we were able to locate the flakes on the diamond anvil. We then aligned the gold particle to the center of rotation. Assuming that the surface of the diamond anvil is nearly perpendicular to the beam (at hdth=0) we then used the microscope camera of the ruby fluorescence spectrometer to bring the nano flakes into the beam. Using this procedure, we centered 18 different 1T-TaS2 flakes with varying thickness. For each of these flakes we collected one low- and one high flux single crystal datasets of 124 images taken from hdth=-31 deg to hdth=31 deg in steps of 0.5 deg. In these data it was possible to identify the superlattice reflections even for the thinnest flakes which had a thickness below 20nm. However, the background from the diamond and some residual PCL on top of the flakes makes a quantitative interpretation of this data very difficult and we are still working on the analysis. We then loaded the DAC with helium as pressure transmitting medium. Due to a technical problem during the loading process the starting pressure was already about 3GPa and it turned out that the residual PCL along with the nano flakes had detached from the diamond surface. This has prevented us from finding the samples again and we lost about 2 shifts.
We then decided to switch to the $IrTe_2$ samples. Since there was also some residual PCL on top of the flakes and because it would have taken about 12 hours to fully resolve the polymer in tetrahydofuran we decided to not close and load the DAC, but put it into the crysostat in order to measure the temperature dependency for different flake thicknesses at ambient pressure. For $IrTe_2$ the flakes have generally smaller lateral dimensions. However, using the same alignment procedure as for 1T-TaS_2 we were again able to measure XRD data for these samples. We then started a temperature dependence from RT down to 10K in steps of 40K. At 190K we could see the onset of electronic order in one flake and at 150K also in the other flake (see Fig. 2). The observed superstructure is qualitatively similar to the superstructure found in bulk samples but occurs at lower temperatures consistent with reported transport data. Unfortunately, at 110K and below a technical problem with the cryostat heater led to a drifting temperature of the shield and accordingly a misalignment of the sample position such that the data for these temperatures are not useable. We are also here facing the same challenges due to the large background from the diamond and the PCL and we are still working on the analysis.

Conclusion

In conclusion, the experiment surely demonstrated the feasibility to measure single crystal XRD on exfoliated nano-flakes in a DAC at ID27. Since this was the very first attempt, we think it is natural to face some technical issues. In particular the problem with the residual PCL can be easily solved with tetrahydofuran. In order to reduce the background, we can use cells with thinner diamonds which were developed for x-ray spectroscopy. For the vdW nano-flake materials the low-pressure region up to about 5GPa is most interesting which means that the reduced pressure range of these specialized cells does not cause a problem. The ultimate goal will be to improve the data quality up to a level which allows for full crystallographic refinements of the crystal structure. We want to emphasize that the development of this experimental method is very useful and important as it can not only be applied to the huge material class of layered vdW systems but also other thin film systems and even bulk systems with very light elements where the diamond background can be a problem, too.