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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:** Suppression of charge-density wave order in 2H-TaSe2 by pressure	**Experiment number**: HC-5083
Beamline: ID15B	**Date of experiment**: from: 05 Oct. 2022 to: 08 Oct. 2022	**Date of report**: 30 Jan. 2023
Shifts: 9	**Local contact(s)**: Gaston Garbarino (email: gaston.garbarino@esrf.fr)	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

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Report:

The aim of this experiment was to check the hypothesis that superconductivity (SC) and charge density wave (CDW) in 2*H*-TaSe2 transition metal dichalcogenide (TMD) are in strong exclusive relations and thus shed light on the mechanisms responsible for these phenomena.
The idea was to employ high-pressure x-ray diffraction (XRD) to determine the position of the quantum critical point (QCP), i.e. to find the critical pressure P_c at which the CDW transition temperature T_{CDW} suppressed to zero, and complete the P-T phase diagram of 2*H*-$TaSe_2$ in the pressure range passing through and beyond the QCP. Comparing the behavior of the CDW and earlier published SC transition temperature T_c [2] with pressure, we can confirm or disprove the assumption that the pressure at which T_c reaches its maximum corresponds to the P_c of the QCP.

This compound was previously intensively studied by means of XRD [1], susceptibility and resistance measurements [2, 3]. Under ambient pressure 2*H*-TaSe2 adopts the hexagonal ($P6_3/mmc$) structure at $T > T_{CDW} = 121$ K.
The published phase diagram, obtained from resistivity and susceptibility measurements [3], is shown in the inset to Fig. 2. The phase diagram illustrates CDW suppression with pressure along with a monotonic increase in the SC T_c. The SC dome reaches its maximum at $T_c = 8$ K at $P = 23$ GPa. Unfortunately, the signature of the CDW transition in the resistivity data vanishes at higher pressures and T_{CDW} (red triangles) could only be assessed up to 20 GPa. Thus, the position of a possible CDW QCP has not been determined prior to our experiment. A simple extrapolation of the data indicates that the P_c value for QCP is around 30 GPa or even higher, which is far from 23 GPa for the maximum SC T_c. In order to measure the position of the QCP with high accuracy and therefore test the hypothesis of a strong exclusive relationship between SC and CDW in this material, we performed high pressure XRD measurements. Our data show the behaviour of the CDW with pressure up to its complete suppression.

The crystals for this experiment were grown using the vapor growth technique in the group of K. Rossnagel (University of Kiel, Germany) and the high-quality and polytype purity of the 2*H* phase of our 2*H*-TaSe2 crystals were verified by the previous elastic and inelastic x-ray scattering experiments at APS. The experiment was performed at the ID15B beamline with 30 keV energy of the incoming beam. For the effective detection of the scattered photons the large area EIGER2 X 9M CdTe (340x370 mm) flat panel detector was used.
For high pressure XRD measurements, we prepared two small samples (in-plane < 50 µm, out-of-plane < 20 µm) suitable for a diamond anvil cell (DAC) measurements in the desired pressure range 10 – 35 GPa. Each sample was mounted in a diamond anvil cell (DAC), loaded with helium as a pressure transmitting medium. Pressure was varied *in situ* using a helium-pressurized membrane and monitored by the ruby fluorescence technique. The XRD measurements were done on a single crystals of 2*H*-$TaSe_2$ between room temperature and 2 K, and for pressures ranging from 0.19 to 30.27 GPa.

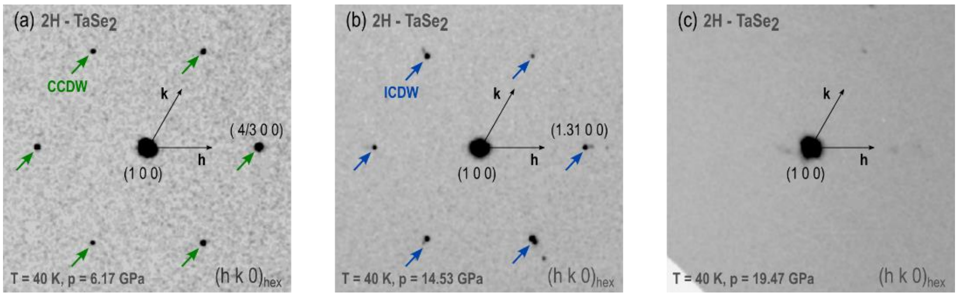

Fig. 1 Typical x-ray diffraction patterns of 2*H*-TaSe2 crystal at T = 40 K produced with CrysAlis Pro software. (a) CCDW state, P = 6.17 GPa, green arrows pointing to commensurate reflections $\vec{q}_{CCDW} = (0\ 1/3\ 0)_{\text{hex}}$, (b) ICDW state, P = 14.529 GPa, blue arrows pointing to incommensurate reflections $\vec{q}_{ICDW} \approx (0\ 0.31\ 0)_{\text{hex}}$, and (c) normal state (no CDW) at P = 19.47 GPa.

Typical diffraction patterns measured at T = 40 K and different pressures are shown in Fig. 1. The signature of a CDW in 2*H*-TaSe2 is a presence of order reflections at wave vectors $\vec{q}_{\text{CDW}} = \pm(1-\delta)\vec{a}^*/3$, where $\vec{a}^*$denotes the reciprocal lattice vector of the 2*H*-TaSe2 hexagonal lattice structure. Data shown in Fig. 1 (a) was measured at P = 6.17 GPa, here one can see six commensurate reflections $\vec{q}_{\text{CCDW}} = (1/3\ 0\ 0)_{\text{hex}}$ surrounding structural (1 0 0) Bragg peak related to the hexagonal symmetry of the high-temperature structure. Here, we find $\delta = 0$, i.e., a commensurate CDW order. The pattern shown in Fig. 1 (b) and measured at a higher pressure P = 14.529 GPa reveals an incommensurate CDW with $\vec{q}_{\text{ICDW}} \approx (0.31\ 0\ 0)_{\text{hex}}$ or $\delta \approx 0.066$. Data shown in Fig. (c) was taken at even higher pressure of 19.47 GPa where the CDW is suppressed at 40 K.
The high quality of the crystals enabled us to perform measurements up to pressures as high as 30 GPa, and to complete the phase diagram of 2*H*-TaSe2 in its most critical regime where the CDW is suppressed to zero temperature by hydrostatic pressure, confirming for the first time the existence of a quantum critical point (QCP) at $P_c \approx 24$ GPa. The resulting phase diagram, shown on Fig. 2., summarizes all the points measured during this experiment. At ambient pressure it consists of an incommensurate CDW (ICDW, red dots) below 122 K and a subsequent lock-in transition to a commensurate CDW (CCDW, blue triangles) below 90 K. With an increase in pressure at low temperatures, the commensurate phase undergoes a transition into an incommensurate one. Rising even higher in pressure, the system undergoes a lock-in transition and again enters a commensurate phase. From our measurements, we can make a preliminary conclusion that the maximum of the SC dome at T_c = 8 K strongly correlates with the QCP of the CDW which lies in the range 23 – 24.7 GPa.

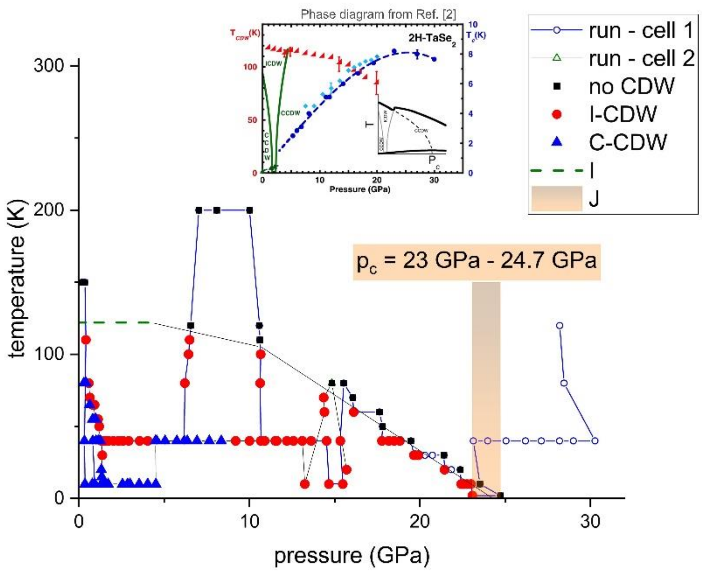

Fig. 2 2*H*-TaSe2 *T*-*P* phase diagram measured with high-pressure x-ray diffraction at ID15B. Inset: 2*H*-TaSe2 phase diagram taken from reference [3].

In summary, we performed high-resolution pressure-dependent x-ray diffraction measurements investigating the P-T phase diagram of CDW order in 2H-$TaSe_2$ TMD single crystals. At the end of the experiment, all the goals indicated in the proposal were achieved. The experiment revealed the details of a rich T-P phase diagram characterized by the interplay of commensurate and incommensurate CDW orders, and for the first time established the existence of a QCP at 24 GPa, where the CDW is completely suppressed by hydrostatic pressure. The obtained results indicate a strong exclusive relationship between the CDW and SC cooperative electron phenomena in 2H-$TaSe_2$.

1. Ph. Leininger et al., PRB **83**, 233101 (2011)
2. D. B. McWhan et al., PRL **45**, 269 (1980).
3. D. C. Freitas et al., PRB **93**, 184512 (2016).