EUROPEAN SYNCHROTRON RADIATION FACILITY

INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

	Experiment title: Pressure dependence of the lattice anharmonicity in 3D CDW VSe2	**Experiment number**: HC-4513
Beamline:	**Date of experiment**: from: 01/06/2021-07/06/2021to:	**Date of report**: 12/08/2021
Shifts: 18	**Local contact(s)**: Alexei Bosak	*Received at ESRF:*

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Report:

In the HC-4513 project, we have proposed to study the suppression of the CDW under hydrostatic pressure and its effect on the lattice anharmonicity in the canonical 3-dimensional (3D) CDW transition metal dichalcogenide, VSe_2. We had previously observed that van der Waals interactions account for the prevention of the CDW formation at high temperature and its temperature dependence is described by anharmonic effects [1]. In the follow-up experiment, we intended to track the pressure and temperature dependence of the renormalized soft phonon mode (energy and linewidth) in VSe_2 and model the anharmonicity dominating the low-energy phonon spectrum by means of state-of-the-art lattice dynamic calculations.

Due to the present pandemic situation, the beamtime was carried out remotely. We have prepared high quality VSe_2 single crystals ("*flakes*") using chemical vapour deposition (CVD) with T_{CDW}=110 K and shipped them to ESRF. The layered nature of the material allowed an easy preparation of the sample for IXS measurements. ID28 was configured in the Si (9 9 9) 17.794 keV reflection setup which gave 3 meV energy resolution. The sample was loaded-in using a gasketed diamond anvil cell (DAC) positioned in a custom-designed continuous flow helium cryostat. The pressure was varied in situ using a helium-pressurized membrane.

Once we have loaded the sample in the pressure cell, we have taken IXS scans at room temperature and 0.5 GPa. A representative set of results are plotted in the figure 1(a), where a low energy optical phonon is visible at 6 meV energy loss. Despite the lower resolution of the ID28 spectrometer, we still see the broadening of the low energy soft mode at h=2.25 rlu., and the spectra shows high statistics, demonstrating the feasibility of the experiment.

Next, we have cooled the pressure cell down to 150 K. As we can see in figure 1(b), the soft phonon mode around the $\boldsymbol{Q}_{CDW}$ is visible in VSe_2 crystals loaded in the pressure cell. Unexpectedly, the elastic line increases (see figure 1(c)), demonstrating a pressure enhancement of the CDW correlations, unlike the theoretical calculations we have attached to the HC-4513 proposal and the experimental reports on similar 2 dimensional transition metal dichalcogenides [3].

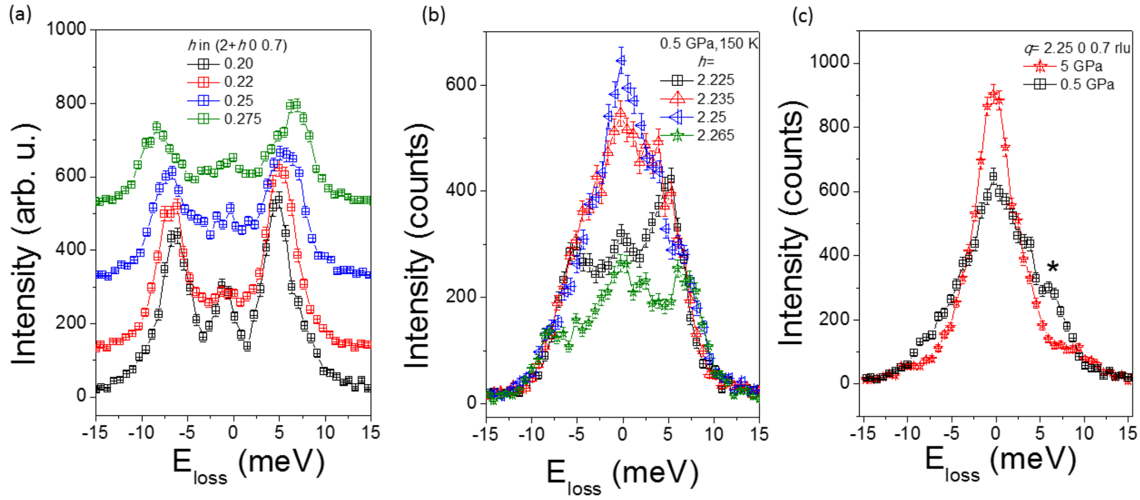

Figure 1. Representative scans of VSe_2 taken at different temepratures and pressure. (a) Room temperature and room pressure IXS spectra in VSe_2. (b) Momentum dependence of the soft phonon mode at 150 K and 0.5 GPa. (c) IXS spectra at $\boldsymbol{Q}_{CDW}$ and 5 GPa. The data shows a clear enhancement of the elastic line, indicating that the CDW increases with pressure, unlike similar 2D TMDs and our own DFT calculations.

Recently, several works report the enhancement of the CDW transition up to room temperature based on high pressure resistivity measurements [2]. Our data taken at ID28 seems to corroborate those arguments and also give an explanation for the collapse of the VSe_2 lattice at room temperature we have seen in powder samples at 12 GPa. Indeed, to corroborate this point of view, we have increased the pressure up to 12 GPa and observed neither the elastic line nor low energy phonons, suggesting that VSe_2 has entered in a new crystallographic phase.

Our preliminary experimental data certainly indicates that the CDW increases with pressure. This is, at first, at odds with the $NbSe_2$ crystals, where pressure was reported to drop the CDW down to zero temperature, highlighting the critical role of the lattice anharmonicity. Nevertheless, the 3D nature of VSe_2 and the importance of the van der Waals interactions to describe the temperature dependence of the soft mode seem to be at the root of the VSe_2 under pressure. VSe_2 seems to be an unique case among all the TMDs (and perhaps an unique case among all CDW systems. Note that in cuprates [4], the CDW dramatically drops under very moderate pressures), where van der Waals interactions and lattice anharmonicity are competing forces in the ground state of VSe_2. In this sense, we have found that the uniaxial out-plane-pressure is a mean to tune the balance between the two energies and, therefore, a way to delve into the physics of TMDs. We believe that, once pressure is increased, van der Waals forces start to become less effective and the 3D CDW increases its onset temperature, as we report in our Nat. Comm. paper. Once the lattice anharmonicity becomes energetically comparable to the van der Waals forces at room temperature, the lattice suddenly collapses. Indeed, until the drafting of this report, the group of Ion Errea and his collaborators (Mateo Calandra, Francesco Mauri,…) have been struggling to theoretical model the pressure dependence of the soft mode under pressure assuming the van der Waals-anhamonicity competition scenario. This is still fruitless and unsuccessful, since we lack the knowledge of the effect of the van der Waals forces under pressure. A fluid and continuous feedback from theoreticians says that experimental input from IXS is crucial to understand the physics of VSe_2. Therefore, we will continue with the project and submit the proposal again to measure the pressure dependence of the soft mode under pressure and room temperature.

References

[1] J. Diego, I. Errea, S. Blanco-Canosa et al Nature Communications 12, 598 (2021).
[2] S. Sahoo, et al Phys. Rev. B 101, 014514 (2020).
[3] M. Leroux et al., Phys. Rev. B 92, 140303(R) (2015).
[4] M. S. Souliou et al. Phys. Rev. B 97, 020503(R) (2018).