ESRF	**Experiment title:** **Modulated Structure of $(TaSe_4)_2I$**	**Experiment number:** CH-480
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Report:

The aim of the proposed experiment was to determine the structure $(TaSe_4)_2I$ in its modulated low temperature phase by single crystal x-ray diffraction. The modulation of this compound is caused by the formation of a charge density wave at temperatures below 260K. In order to achieve this goal several thousand main Bragg reflections and satellites had to be measured at low temperatures around 120 K. For this experiment the 6 axis Kuma diffractometer was used on the Swiss-Norwegian Beamline (BM0l). Sample cooling was achieved by using a cryogenic gas flow cooling.

One of the major problems of the proposed experiment is the reported small value of the modulation wavevector $\mathbf{q} = (0.05,0.05,0.085)$ of $(TaSe_4)_2I$ which leads to very little separation of the weak satellites from the strong main Bragg reflections. This makes the experiment very demanding in angular spread of the incident beam as well as of angular resolution of the diffracted intensity. The angular spread of the incident beam was of the order of about 0.003" as typical for a third generation synchrotron source which was about a magnitude better than required for our experiment. A problem appeared to be the angular resolution of the detector. Because of the lack of a variable detector opening new apertures

had to be made to achieve the required angular resolution of around 0.2°. In addition to the high requirements on the experimental equipment extremely good sample quality in terms of a very small mosaic spread is needed. It turned out that our best samples had a mosic spread of around 0.07" at room temperature which was more than sufficient. Unfortunately some problems with diffractometer alignment in the beginning and continuous problems with the diffractometer control software made it difficult to determine accurate enough orientation matrices and modulation wavevectors in order to make best benefit of the achieved angular accuracy.

Nevertheless two datasets could be measured. One consists of the intensities of about 150 main reflections with eight surrounding satellites each corresponding to all possible modulation wavevectors. This data set will allow us to determine the symmetry of the modulated phase and will answer the question whether the modulated phase consists of a multi-domain phase with reduced symmetry. The second one consists of the intensities of around 800 main reflections with two surrounding satellites each corresponding to only one modulation wavevector. Due to the high number of measured reflections a complete determination of the modulation amplitudes of all atoms should be possible with this second data set. Both data collection have been performed on a half sphere in reciprocal space in order to get complete data even for triclinic symmetry of the modulated phase.

An unexpected result of the present experiment was the splitting of hk0 reflections below the phase transition temperature. The shape of 001 reflections appeared to be a lot less affected by cooling below the phase transition temperature. The observed splitting is reversible by warming the sample up to room temperature again. We attribute this splitting to twinning of the sample in its modulated phase supporting the assumed symmetry loss in this phase. The performed careful characterisation of the observed peak splitting will therefore support the search for the right symmetry of the modulated low temperature phase of $(TaSe_4)_2I$.

In conclusion the proposed experiment could be performed successfully despite the mentioned problems with the diffractometer control software. For future experiments we suggest further improvement of the software, in particular the handling of orientation matrix reflections including centering and the handling of reflections with broken indices which is essential for determining the structure of modulated compounds.