ESRF	**Experiment title:** Observation of a Charge Density Wave in Incommensurate Te-III	**Experiment number**: HS3200
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Names and affiliations of applicants (* indicates experimentalists):

M. I. McMahon, I. Loa*, S. R. Evans*, O. Degtyareva*, Lars F. Lundegaard

SUPA, School of Physics and Centre for Science at Extreme Conditions,

The University of Edinburgh, United Kingdom

Report:

Tellurium belongs to a group of elements that have recently been discovered to adopt an incommensurately modulated crystal structure at high pressure. In the pressure range of 8–29 GPa, Te adopts a body-centred monoclinic structure with an incommensurate transverse modulation, where the modulation vector is parallel to the crystal *b*-axis (Fig. 1). The same structure type was also observed in the chalcogens Se and S. Recent calculations have suggested that these incommensurate phases may arise through Fermi-surface nesting via a Kohn anomaly or charge density wave [1]. No experimental data has been published on the phonon dispersion relations in any of the incommensurate group-VI elements to test these calculations.

A single-crystal of the high-pressure phase Te-III was prepared by loading a small single-crystalline piece of Te in its ambient-pressure trigonal form into a diamond-anvil high-pressure cell and pressurising it slowly to 8.5 GPa. A 4:1 methanol/ethanol mixture was used as the pressure-transmitting medium. It is well known that single crystals tend to shatter into polycrystals when taken across a pressure-induced structural phase transition, and Te is no exception in this respect. A small Te-III crystal with lateral dimensions of 30 x 40 μm^2 and an estimated thickness of 10–20 µm was prepared previously in our group for diffraction experiments (Fig. 2). We attempted to obtain somewhat larger crystals of Te-III for the IXS experiment: Close to 100 crystals of Te were screened at ambient pressure and about 40 of them, which were high-quality

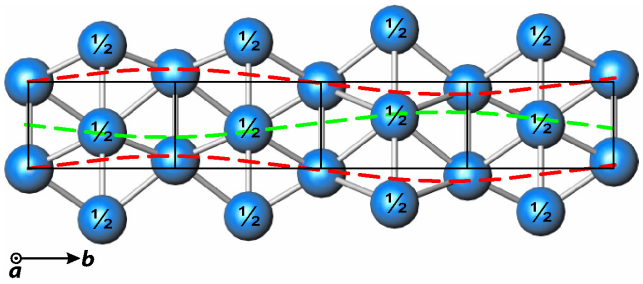

Figure 1: The incommensurately modulated crystal structure of Te-III at 8 GPa as viewed down the *a*-axis. The modulation wave (0*q*0) is shown by the dashed lines.

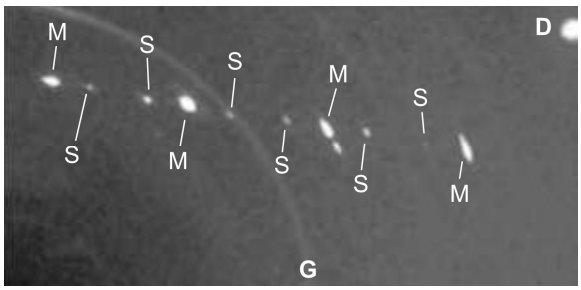

Figure 2: A diffraction image from a single crystal of Te-III at 7.4 GPa showing both the main (M) and incommensurate satellite (S) reflections. The features marked with 'G' and 'D' come from the gasket and one of the diamond anvils, respectively.

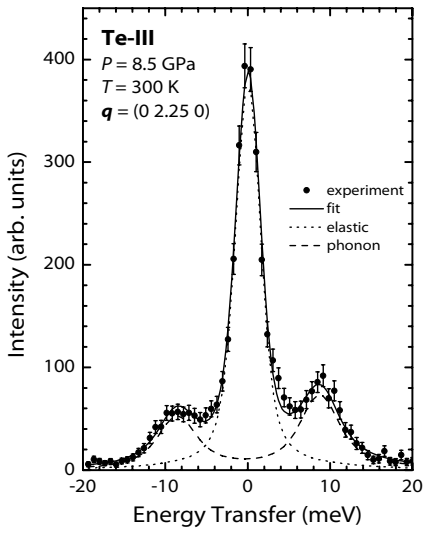

Figure 3: IXS spectrum of Te-III at 8.5 GPa. The measured intensities are shown by circles and the total fitted spectrum, the elastic contribution and the phonon contribution by solid, dotted and dashed lines, respectively.

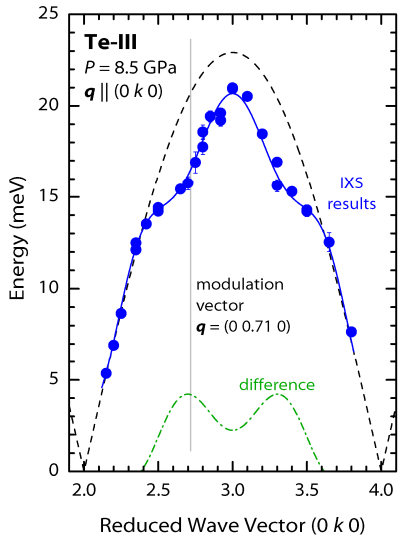

Figure 4: Dispersion relation of the longitudinal-acoustic phonon branch along (0*k*0) in Te-III, i.e., parallel to the incommensurate modulation vector.

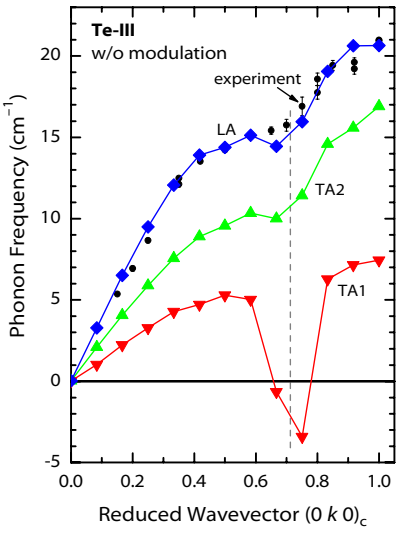

Figure 5: Comparison of the experimental LA phonon dispersion curve (small black symbols) with the first-principles theoretical results (coloured symbols and lines).

single crystals, pressurised to ~9 GPa. Unfortunately, the crystal quality degraded at the phase transitions to such an extent that we decided to use the initial Te-III crystal despite its small dimensions.

The inelastic x-ray scattering experiment was performed on beamline ID28 using the Si (999) configuration, i.e., with a photon energy of 18 keV and an energy resolution of 3 meV. A first set of spectra of longitudinal acoustic (LA) phonons was collected for momentum transfers along the (0 *k* 0) line in reciprocal space, i.e, parallel to the modulation vector. Figure 3 shows a selected spectrum together with the fitted model (the Fit28 software was used for the data analysis). Typical accumulation times were 90 s per data point. From these spectra, the LA phonon dispersion curve over a full Brillouin zone was constructed as shown in Fig. 4. An anomaly is observed near the modulation vector $\boldsymbol{q}$ = (0 0.71 0). In Fig. 4, the dashed line indicates a simple sine-type relation for comparison, and the dashed-dotted line shows the differences between the energies given by the dashed line and the observed phonon energies.

We have attempted to measure also the dispersion of the transverse acoustic (TA) branches. This was made difficult, however, by the fact that the Te-III crystal was twinned (which is hardly avoidable). Due to the particular nature of this twinning it does not affect measurements along (0 *k* 0), but it causes a superposition of two sets of dispersion branches for $\boldsymbol{q}$ = (0 *k* 1) and $\boldsymbol{q}$ = (0 *k* -2), where we tried to detect the TA branches. Further analysis of the data may allow us to extract at least parts of the TA branches.

We complemented the experimental study by first-principles phonon calculations based on a body-centred monoclinic unit cell *without* incommensurate modulation. Using linear-response theory as implemented in the *abinit* code [2], the three acoustic phonon branches were calculated along the (0 *k* 0) as shown in Fig. 5. The experimental and computational results for the LA are in excellent agreement. The theoretical results for the lower-energy TA branch evidence a lattice dynamical instability at the experimentally observed modulation vector. The anomaly observed in the experimental results of the LA branch can thus be considered a weaker image of this instability. *The combined experimental and theoretical results demonstrate for the first time that the incommensurately modulated structure in Te-III is the consequence of a lattice dynamical instability*. Preliminary calculations of the Fermi surface of Te-III suggest Fermi-surface nesting as a likely origin of the dynamical instability as suggested also in recent theoretical work [1]. These findings are also important to understand the behaviour of Se and S.

References

[1] Ackland and Fox, J Phys Cond. Matt. **17**, 1851 (2005).
[2] http://www.abinit.org