ESRF	**Experiment title:** Inelastic X-ray scattering of liquid As_2Te_3 at high pressures and temperatures.	**Experiment number**: HD-596
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Report:

The goal of this research was an Inelastic X-ray Scattering (IXS) study of As_2Te_3 liquid at high pressure – high temperature conditions. This liquid represents one of the chalcogenide melts displaying semiconductor – metal transition during temperature or (and) pressure increase. The structural modifications and changes of the electron transport properties were extensively studied for such kind of phase transitions. Recently the structural changes of As_2Te_3 liquid during heating at moderate pressures have also been studied in details[1].

At the same time there is no information about the modification of the dynamics of these liquids during semiconductor to metal transition. With the recent advances of the IXS technique it was found in last decades that the well-defined phonon-like collective excitations exists in most liquids. It is of great interest to understand how do the spectrum of these excitations and the dynamical viscosity change in region of semiconductor – metal transition in the melts.

We used a resistive heated DAC as high P-T apparatus. 99.999% purity As_2Te_3 glass has been used as a pristine material. The global DAC heating obtained by using an external band heater allowed the minimization of the temperature gradients and an accurate sample temperature measurement. In order to avoid chemical reactions with the metallic gasket material the sample was contained inside an NaCl ring, obtained by laser drilling a pre-pressed NaCl pellet inside the metallic gasket hole. The pressure has been measured by ruby and Sm-borate fluorescence technique as well as by measuring the XRD pattern of the NaCl. We have used a special vacuum chamber dedicated to experiments on liquids in the DAC at high pressure and temperature, developed in collaboration with the ID28 staff. This instrumentation has demonstrated to eliminate typical problems of measurements on samples in the DAC and allow quantitative measurements of the dynamical structure factor $S(Q,\omega)$ through IXS spectra[2].

The silicon (12 12 12) configuration provided enough energy resolution (1.3 meV) in order to perform a full viscoelastic analysis on the deconvoluted spectra. High enough statistics has been obtained with about

200 s of integration time per point even with a small sample thickness of about 80 μm. The scattering length of the sample is about 100 μm at 23.725 KeV.

We have used 18 shifts of beam-time (20-26 June 2012) at the ID28 beamline. We performed IXS measurements of liquid As_2Te_3 at high-temperature-high-pressure region in several P-T – points in the vicinity of the melting curve. The full set of good quality S(Q,ω) data for 0.5 GPa and 720K (in the metallization region) has been obtained and the dispersion curves were extracted from data analysis of the spectra (see Fig.1). Besides we have measured with less integration IXS spectra for lower pressures - P=0.25 GPa and T=690K (at the beginning of metallization, see dispersion curves in Fig.2), and at higher pressures - at 1 GPa and 800K (in metallic region). We have not managed to execute all the planned measurements of S(Q,ω) due to the experimental problems. The first problem was connected with the very low viscosity of melt and instability of sample inside the NaCl ring especially at very low pressure for the DAC, reason for which one sample was lost. The second problem was connected with the high temperatures (800K and higher) as the used heaters properly work in vacuum conditions only up to 600K. Nevertheless we have now acquired a certain skill to handle the sample at very high T conditions so that together with new heaters which can work in vacuum up to more than 1000 K (this project in collaboration with ID28 is in progress) we would now be able to complete the experiment in a potential Continuation project.

In this experiment we also observed an increase of the static structure factor S(Q) for small Q values (< 5 nm^{-1}) in liquid state at 0.5 GPa. This means a higher compressibility and larger fluctuations in the liquid state in the region of semiconductor-metal transition (with respect to the metallic state at higher pressures). Very recently a similar increase of S(Q) for small Q associated with static inhomogeneity and mesoscopic density fluctuations in the metallisation region has been observed for Se-Te liquid solution[3]. Strange "speckles" (intensity dips) in the static structure factor S(Q) in the vicinity of main peak (12-27 nm^{-1}) have also been observed, whose origin could be better investigated in the future.

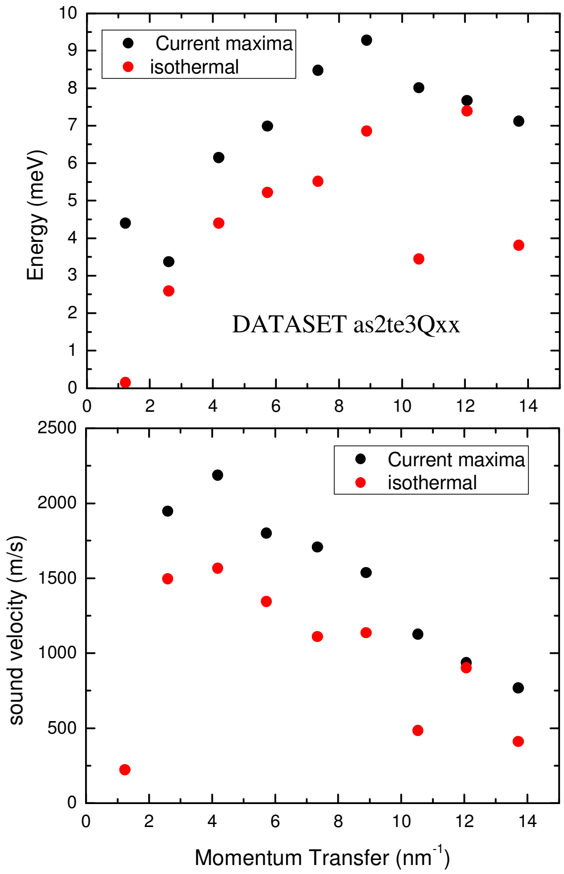

Figure 1: Dispersion curves at 0.5 GPa and 720 K

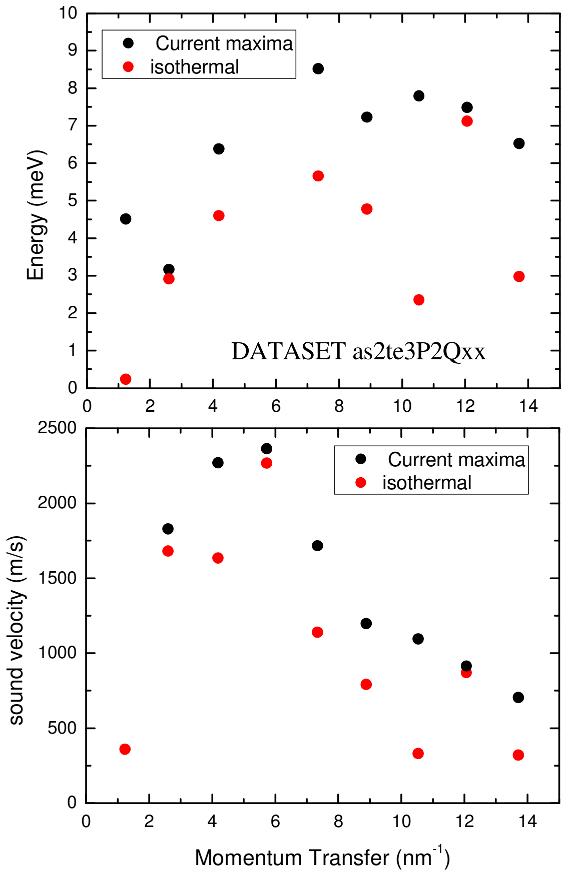

Figure 2: Dispersion curves at 0.25 GPa and 690 K

References:

1. C. Otjacques, et al., *Phys Rev. B* **82**, 054202 (2010).
2. F. Gorelli, et al. *Appl. Phys. Lett.* **94**, 074102 (2009).
3. Y. Kajihara et al., *EPJ Web of Conferences* **15**, 02002 (2011).