Experimental Report

Proposal: **6-01-314** **Council:** 4/2010

Title: Dynamics across the liquid-liquid transition in supercooled tellurium

This proposal is continuation of: 6-01-307

Researh Area: Physics

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Samples: Tellurium, Te

Instrument	Req. Days	All. Days	From	To
IN11	12	7	27/11/2012	04/12/2012

Abstract:

Liquid-liquid transitions are found in many simple metals and semi-conductors, in the chalcogenides (S,Se,Te) and in network glasses and elemental liquids. How the thermodynamic signatures of these liquid-liquid transitions are related to the underlying structural and dynamical liquid properties is not well known. Tellurium, as several other chalcogenides, shows a clear thermodynamic transition with a peak in compressibility and heat capacity and a maximum in its density. This behaviour is related to a change in liquid structure and for tellurium the transition takes place in the supercooled regime at ~353 deg C. We want to know how the thermodynamic transition is manifested in the microscopic liquid dynamics. We thus propose to measure the dynamic structure factor across the liquid-liquid transition in Te using the neutron spin echo technique to reach the relevant time and length-scales. We have previously been granted beam time at IN11 for a test experiment (6-01-307) to prove the feasibility of using NSE at IN11 to study the dynamics in supercooled Te. We here propose the full experiment drawing on the experience of the test run.

Dynamics across the liquid-liquid transition in supercooled tellurium (6-01-314)

The aim of the proposed experiment was to investigate the dynamics near the liquid-liquid transition in tellurium. Tellurium, as several other chalcogenides, shows a clear thermodynamic transition in the liquid regime with a peak in its compressibility and heat capacity and a maximum in its density. This behaviour is related to a change in liquid structure and for tellurium the transition takes place in the supercooled regime at ~626 K. Using the IN11C Spin-Echo spectrometer the relevant energy and momentum transfer range can be detected. For this experiment we were granted seven days of beam-time.

In the experiment, the IN11C spectrometer was used with 3.8 Å incident neutron wave-length and the measurements were done at a wave-vector setting matching the first maximum of the structure factor at Q_p~2Å^{-1}. In an initial feasibility experiment (6-01-307) the sample was contained within a double-walled cylindrical quartz cell with a sample thickness of 6 mm and wall thickness of 2x4 mm (see the Experimental Report) and it was demonstrated that the structural relaxation can be observed in the liquids, but the data quality was poor due to the large elastic background contribution from the quartz sample cells. Thus, in these experiments, sample cells were designed with 5mm thick silicon windows fused onto a quartz body. A sample thickness of 6.5 mm was chosen as an optimum balance between signal strength and absorption. Based on the previous test experiments a shorter wavelength was chosen in order to reach the fast times needed, particularly at high temperatures.

Experiments were performed for temperatures in the range 649-744 K (the melting point of Te is T_m=723 K and the glass transition temperature T_g=303 K) in the non-magnetic high temperature oven. Two thermocouples were mounted on the sample to check for any inhomogeneities in the temperature profile. In order to be able to reach temperatures above the melting point of Te, the oven was modified and its temperature control fine tuned. With these modifications, we were able to both reach high enough temperatures and perform temperature quenches, reaching significantly into the supercooled range.

In the experiments, echoes could be observed and the signal was sufficient to make echo scans quickly enough. Typical data, $S(Q,t)/S(Q)$, are shown for 663 K liquid Te in Fig. 1a, here averaged over the full recorded wave-vector range. In contrast to the previously used quartz sample cells, the elastic background is very low, as seen in Fig. 1(a), and measurements with sufficient signal could be readily performed. For all data sets the analysis could be well described using a single exponential description as shown both in Fig. 1(a) for T=663 K and in a semi-log representation of all recorded data sets in Fig. 1 (b). It is here clear that as the temperature is reduced the time-scale slows down and we were able to obtain the structural relaxation time for tellurium at 8 temperatures in the range between 649 and 744 K.

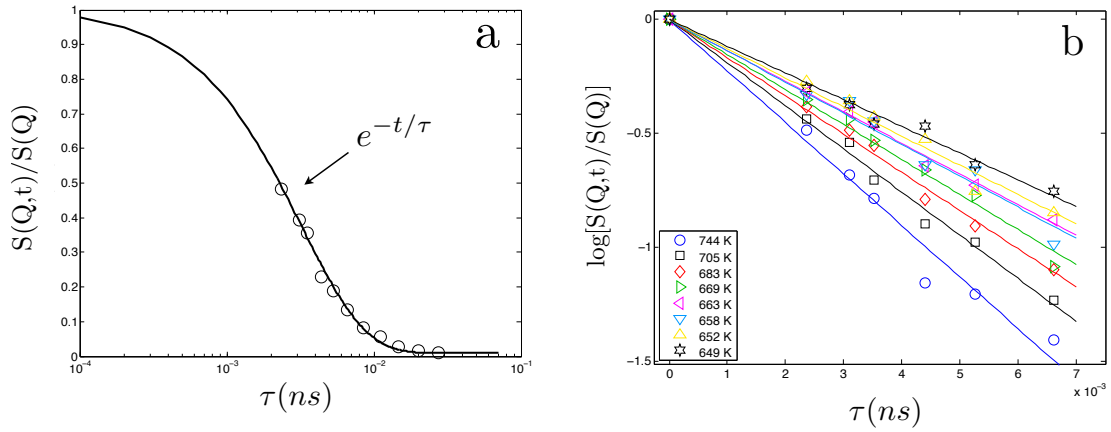

Figure 1 (a) Data for T=663 K together with a single exponential fit. (b) Data for the 8 different recorded temperatures represented in a semi-log plot to demonstrate that all data can be described using a single exponential description and the slowing down of the structural relaxation time as the temperature is reduced.

For measurements performed significantly below the melting point, particularly as the structural transition at 626 K was approached, the experiments were complicated by the strong tendency to crystallize. To address this, the sample was equilibrated above the melting point and then cooled quickly to the preferred temperature and data was subsequently collected until crystallization took place; crystallization could clearly be observed and monitored in the echo behaviour. We did manage to collect data for the 8 temperatures shown in Fig. 1(b), but on approaching lower temperatures during a cooling run the sample cell broke; the experiment was thus cancelled on Saturday 1 Dec. Due to the breaking of the sample cell, the oven was contaminated by the sample and had to be put into storage.