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Report:

The experiment was performed at an incident photon energy of 17794 eV, using the silicon (999) setup, providing an overall energy resolution of 3 meV (FWHM). The surface-sensitive geometry was obtained by inserting a flat platinum coated mirror at 200 mm from the sample position, thus offering the possibility to vary the glancing angle of incidence on the sample, α , between 0 and 0.13°. By properly adjusting this deflecting mirror and the main focusing toroidal mirror, the standard spectrometer configuration could be preserved. For this first pioneering experiment, we used liquid indium as a sample. This choice was motivated by the low melting temperature T_m of 429.8 K, the low vapour pressure (<10⁻¹⁴ mbar at T_m), the favourable scattering signal, and the possibility to prepare very clean samples without any risk of surface oxidation [1]. The liquid indium samples were prepared *in situ* in a compact UHV chamber, especially designed for the IXS experiment. The whole set-up was mounted on an active anti-vibration table in order to ensure optimum stability during the course of the experiment. Most of the IXS spectra were recorded at $\alpha =$ $0.8\alpha_c$, corresponding to a glancing angle of incidence of 0.12°, and a scattering depth of 5 nm. The spanned momentum transfer range was 5-19 nm⁻¹.

Figure 1 shows a typical spectrum, together with the best fit result. This fit was obtained using a model, composed of a viscoelastic function (taking into account one structural relaxation process [2]) and a δ -function, convoluted with the experimentally determined resolution function. As a matter of fact, a viscoelastic model alone could not account for the observed intensity around zero-energy transfer. Alternatively, the data were fitted with a model function, consisting of two pairs of damped harmonic oscillators (DHO). Both model functions yielded comparable χ^2 -values and almost identical values for the energy position of the dispersive mode.



Fig. 1 (left): IXS spectrum of liquid indium in grazing angle geometry at Q=7.63 nm⁻¹. The experimental data points and their corresponding error bars are shown together with the best fit (see text for more details).

Fig. 2 (right): Results of the viscoelastic analysis. $\Omega_0(Q)$, $\Omega_L(Q)$, and $\Omega_{inf}(Q)$ are the energies (\hbar =1), corresponding to the adiabatic (c_0), apparent (c_{app}) and infinite sound dispersion (c_{inf}). The dotted line indicates the dispersion of the adiabatic sound velocity as derived from thermodynamic measurements. See text for more details.

Figure 2 reports the main results of the viscoelastic analysis, namely the Q-dependence of the Ω_0 , Ω_{inf} and Ω_{app} excitation energies, corresponding to the zero-, infinite frequency- and apparent sound velocity c_0 , c_{inf} and c_{app} , respectively. While the first two ones directly result from the fit, Ω_{app} was taken as the maximum of the longitudinal current spectrum $\omega^2 \cdot S(Q, \omega)$. With respect to previous IXS studies on bulk liquid metals [3], there is no evidence for an active structural relaxation process, which would be reflected by a positive dispersion of the excitation energies from the adiabatic to the infinite sound velocity value. We furthermore note that the thermodynamic sound dispersion below 12 nm⁻¹ closely follows c_{inf} , and that the extracted values for $\Omega_0(Q)$ lie systematically below it. Our results indicate that there are significant changes in the high-frequency dynamics of liquid surfaces, but our data set is not sufficient to draw definite conclusions. To this end further data have to be taken at even shallower angles of incidence ($\alpha = 0.6\alpha_c$, scattering depth 3nm) in order to enhance the effect of the surface, and at $\alpha = 1.5\alpha_c$ for which the bulk dynamics are probed.

References

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