ESRF	**Experiment title:** **Surface Structure of glasses: Freezing of capillary waves? – An investigation with coherent x-rays.**	**Experiment number**: SI-546
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Report:

The experiment has resulted in a paper, submitted to PRL. In the following we include the abstract and selected parts of the text including some figures.

Capillary Waves in Slow Motion

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ABSTRACT

Capillary wave dynamics on glycerol surfaces has been investigated by means of x-ray photon correlation spectroscopy performed under grazing angles. The measurements show that thermally activated capillary wave motion is slowed down exponentially when the sample is cooled below 273K. This finding directly reflects the freezing of the surface waves. The wavenumber dependence of the measured time constants is in quantitative agreement with theoretical predictions for overdamped capillary waves.

The x-ray experiments were performed at the ID10A (Troika) beamline of the European Synchrotron Radiation Facility in Grenoble. The surfaces were illuminated with 8keV radiation (wavelength λ=1.548A) from a set of three undulators. The radiation was monochromatized by a Si(111) crystal with a wavelength bandpass of $\Delta\lambda/\lambda = 10^{-4}$ yielding a longitudinal coherence length of about 1μm. A pinhole with a diameter of 12 μm was mounted 278mm upstream from the sample in order to obtain a collimated and (partially) coherent beam. Typical transverse coherence lengths at the pinhole are 100 μm in the vertical and 10 μm in the horizontal direction. A guard slit in front of the sample was used to suppress the Fraunhofer fringes from the first pinhole. The data were collected with a scintillation counter at a distance 2368mm downstream from the sample. A collimating aperture with an adjustable size of 15 μm - 100 μm was chosen in front of the detector. The time-autocorrelation functions were recorded with a digital ALV5000/E correlator.

The glycerol (from Fluka inc., purity always better than 99.5%) was put into an evacuated inner sample cell. This cell was cooled by a constant flow of liquid nitrogen. A film of approximately 4.5mm thickness was filled into an aluminum trough of 140mm diameter. The size of the trough was chosen such that a large flat area of glycerol is obtained in the middle which is unaffected by the meniscus at the border. Furthermore the entire footprint of the impinging radiation should fit onto the sample surface in order to maximize the reflected intensity and to avoid parasitic scattering from the sample container. The inner cell was evacuated to about 10mbar and isolated against possible temperature variations introduced from the outside by a vacuum ($p < 10^{-5}$ mbar) that was maintained during the experiments. The temperature stability of this setup was better than 0.02K over several hours.

The glycerol surface was illuminated by x-rays under a grazing angle $\alpha_i=0.075^o$ and the scattered radiation was detected under different exit angles α_f within the scattering plane. Hence the lateral and perpendicular components of the wavevector transfer, q_x and q_z, are given by: $q_x=2\pi/\lambda(\cos\alpha_f - \cos\alpha_i)$ and $q_z=2\pi/\lambda(\sin\alpha_f + \sin\alpha_i)$. From the q_x-value the respective lateral length scale x_0 that is probed may be easily obtained by: $x_0=2\pi/q_x$. In our study length scales in the region 5 μm < x_0 < 150 μm were tested.

Figure 1

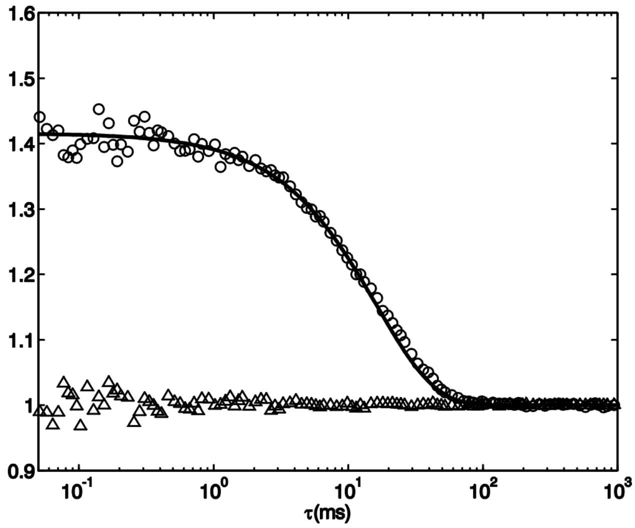
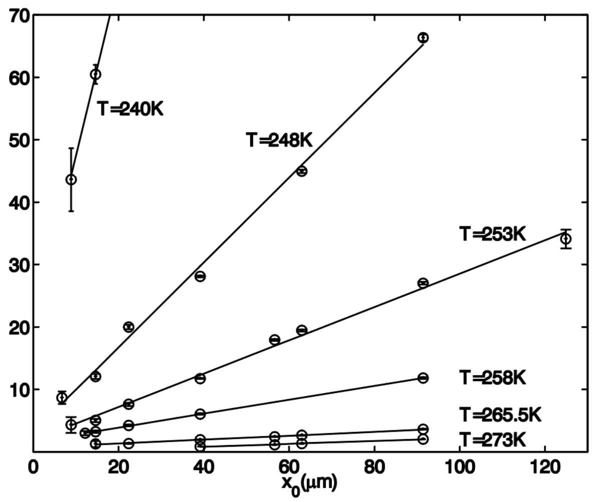


Figure 1 shows a correlation function $g_2(\tau)$ (circles) that has been obtained for a lateral length scale of x_0=56 μm and an opening of 30 x 30 μm of the exit aperture in front of the detector. The temperature of the glycerol surface was T=253K. For comparison the time correlation function of the monitor signal (triangles) is also plotted in Fig. 1. Oscillations at shorter times than those shown in Fig. 1 are due to the incident beam.

Between τ=0.1ms and τ=100ms a well-defined correlation function is visible. A fit of an exponential decrease to the data(solid line in Fig. 1) yields the time constant τ_0=17.9ms and a contrast of g_0=40.5%. The maximum value for the contrast in our measurements was g_0=69.5%.

Figure 2 shows the observed time constants τ_0 (in ms) as a function of the lateral length scale x_0 for different temperatures T. A linear relationship between τ_0 and x_0 is evident for all T. The slope of the curves depends almost exponentially on the temperature. This is shown in Figure 3 where the measured ratio τ_0/x_0 (in ms/µm) is plotted versus T on a log scale. Figure 3 directly reflects the slowing down of the motion on the glycerol surfaces with decreasing temperature.

Figure 2



Figures 2 and 3 can be understood quantitatively by the capillary waves theory of viscous liquids in the simple case of sinusoidal surface waves. This we will not elaborate further on here but rather summarize that for the first time we have demonstrated the possibility of measuring lateral surface motion using x-rays. We believe that this result is important for the further development of the XPCS field and experiments taking advantage of the new technique are already under preparation.

Figure 3