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Report:

We report on the first XPCS measurement of propagating capillary waves on a liquid water surface. The experiment has been carried out at a grazing angle of incidence below the critical angle of external reflection which results in an x-ray penetration depth of 10nm thus leading to a true surface sensitive experiment. Previous experiments [1] of liquid surface dynamics with XPCS concentrated on liquids with high viscosity exploring the region of overdamped capillary waves. The observation of propagating capillary waves on surfaces of liquids with low viscosity is considerably complicated by the presence of surface waves due to external vibrations, with a limited possibility of damping at a synchrotron source. As expected we found strong correlation functions of both the specular reflected beam and the signal at lateral wavevector transfer q_x we could extract the pure signal from the water surface. Figure 1 displays the measured correlation functions at $T=5^{\circ}C$ and different wavevector transfers q_x . The presence of propagating waves on the liquid water surface is obvious. From the correlation functions we obtained the dispersion relation (Fig 2) showing the $\omega \sim q^{3/2}$

dependence of capillary waves. It is interesting to note that the measured frequencies are somewhat lower than expected by simple elastic theory, while the damping coefficients are somewhat larger. Of special interest is the surface dynamics of the supercooled state. Fig 3 shows a correlation function measured at temperature T = -7° C and $q_x = 3.1 \cdot 10^{-6}$ Å-1. The observed correlation function is almost identical to the one measured at T=0.75 C (not shown). Thus on this length scales the capillary wave spectrum in the supercooled state is identical to the spectrum of the *normal* liquid water surface. There is no hint for an increase in damping coefficients or of wave frequencies in the supercooled state close to the phase transition.



Fig 1: Intensity auto correlation functions $(I(t)I(t+\tau))$ of the surface of liquid water at T=5C and different wave vector transfers q_x . As the incident angle of the beam is below the angle of total reflection the experiment is truly surface sensitve. The presence of slightly damped propagating capillary waves on the water surface is obvious.



Fig 2: Dispersion relation of capillary water waves at T=5° C and T=0.75° C obtained from the intensity auto correlation functions. The $\omega \sim q^{3/2}$ dependence is found as expected by elastic theory for capillary waves. However, it is also visible that the measured frequencies are somewhat smaller than predicted (solid line).

Fig 3. Intensity auto correlation function $(I(t)I(t+\tau))$ of the surface of supercooled liquid water at T=-7C and wavevector transfer $q_x = 3.1 \cdot 10^{-6} \text{ Å}^{-1}$.

[1] T. Seydel, A. Madsen, M. Tolan, G. Grübel and W. Press, Phys.Rev.B 63, 073409 (2001)