ESRF	**Experiment title:** Heterodyne mixing in surface XPCS experiments	**Experiment number:** SI942
Beamline: ID10A/C (TROÏKA)	**Date of experiment:** from: 2004-02-15 to: 2004-02-22	**Date of report:** 2004-02-23
Shifts: 18	**Local contact(s):** Anders Madsen	*Received at ESRF:*

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Report:

We report on a surface XPCS (x-ray photon correlation spectroscopy) experiment on liquid surfaces. The goal of the experiment was to explore the possibility of systematic heterodyne mixing. Heterodyning is a standard technique in visible light PCS, as it allows to measure correlation functions even at high $q_{||}$ where the scattered intensity is very low [1]. In this technique, part of the incident or of the specularly reflected beam is coherently "mixed" with the diffuse scattering, emphasizing the g_1 part of the measured correlation function. At the same time, the specular reflection is "modulated" by the diffuse scattering, drastically intensifying the signal.

It has been shown that surface-sensitive XPCS is indeed possible at third-generation synchroton sources, and the technique has already been successfully applied to e. g. the study of the capillary waves spectra on viscous liquids [4]. Recently, heterodyne mixing has been observed in the x-ray regime [2, 3]. The latter could be explained as an intrinsic interference effect at small in-plane wavevector transfers $q_{||}$. However, the goal of this experiment was to explore the possibility of heterodyne mixing in a systematic way.

The measurements were carried out on a liquid hexane surface in grazing incidence geometry with an incident angle of 0.1 degrees, thus below the critical angle of total external reflection. This setup provides for a typical penetration depth of about 10nm, making the measurements very surface sensitive. For heterodyne mixing, part of the primary or the specularly reflected beam has to be coherently superimposed on the diffuse scattering signal at a given $q_{||}$. To achieve this, two approaches were tried:

1. Deflecting the specularly reflected beam into the detector with a mirror. As mirror material Si single-crystals with and without Au coating have been used. Figure 1 shows the contrast of the correlation signal versus the amount of mixed specular signal. The more of the specular signal is superimposed, the more the contrast vanishes and no first order correlation function g_1 becomes visible in the signal. This shows that the signals were mixed incoherently. However, the results show that the specular beam can be successfully deflected into the region of diffuse scattering using a mirror.

2. Putting a diffraction grating inside the reflected beam. Gratings with lattice constants of 0.5μm and 1μm have been used. Figure 2 shows the diffraction pattern of the specular beam using a 1μm grating. In this setup, again no heterodyning could be observed. However, diffraction gratings could be verified as a a viable way of deflecting part of the specularly reflected beam.

It has therefore been shown that a part of the specularly reflected beam can successfully be deflected into the diffuse signal, using either gratings or mirrors. The reason why no heterodyne correlation functions at all were observed needs still to be investigated.
Usually, when measuring the time-time correlation function of the scattered signal near the specular reflexion you can expect intrinsic heterodyne mixing, yielding an almost purely heterodyne correlation function close to the specularly reflected beam [2]. However, in contrast to results of previous measurements, no intrinsic heterodyning could be observed during this experiment. The main difference to prior experiments was the beamline setup: the current measurements were done at ID10C, while previous experiments were done mainly at ID10A. Since the two setups use different monochromators and optics, there might have been a difference in the coherence properties of the x-rays. This has to be investigated further.

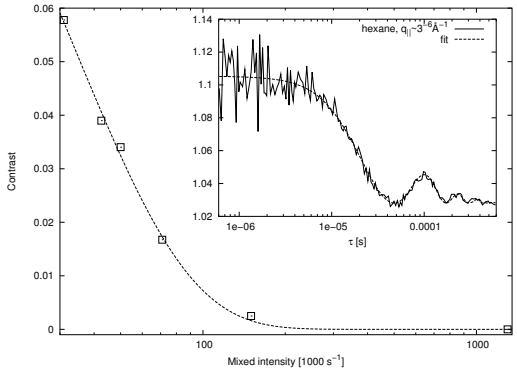

Figure 1: Correlation function contrast vs. intensity of specular signal superimposed by a Si mirror. Inset: XPCS signal from capillary waves on a liquid hexane surface.

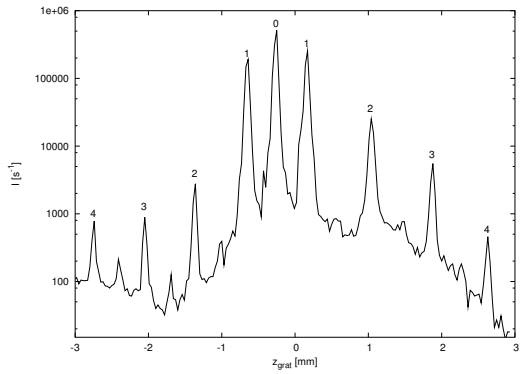

Figure 2: Diffraction pattern of a grating in the specularly reflected beam from the liquid hexane surface.

References

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