ESRF	**Experiment title:** Dynamic processes in glycerol near the glass transition temperature probed by XPCS	**Experiment number:** SC-813
Beamline: ID10A	**Date of experiment:** from: April 4, 2001 to: April 10, 2001	**Date of report:** September 6, 2001 *Received at ESRF:*
Shifts: 18	**Local contact(s):** Anders Madsen	

Names and affiliations of applicants (* indicates experimentalists):

Anders Madsen*, Gerhard Grübel (ESRF),

Tilo Seydel* (ILL),

Metin Tolan* , Christian Gutt* (Experimentelle Physik I, Otto-Hahn-Str.4, D-44227 Dortmund, Germany),

Werner Press (IEAP, Universität Kiel, D-24098 Kiel, Germany)

Report:

The experiment SC-813 was a continuation of the highly successful run SI-546 where surface dynamics of the prototype glass glycerol was probed with coherent x-rays [1, 2]. For the new run, it was our intention to test the limits of the new experimental technique by trying to go to very high lateral wave vector transfers and hence reaching molecular length scales. As expected, this was a difficult endeavour, and we probably have to conclude that the coherent flux available at the beamline is not yet sufficient for that purpose. Additionally, during the experiment we have encountered major problems due to beam damage which was probably at least partly related to a leak in the sample cell. Notwithstanding these difficulties, we can present new results from this beamtime, where we have used a CCD camera to record the full coherent scattering image. The rms roughness and long-range correlations of a liquid surface are essentially determined by thermally activated capillary waves [3, 4, 5]. Whether these capillary modes are propagating, overdamped or frozen-in as a function of temperature and wavevector cannot be distinguished in time averaged x-ray scattering. Coherent x-rays however may be used to determine this in an unambiguous way. The direct visualisation of the speckle pattern using a two-dimensional CCD camera is complementary to the XPCS experiment with a point detector [1] and gives additional information related to the spacial shape of the speckles and their q-dependence [6]. Furthermore, the CCD experiment provides a proof of a surface which is completely frozen-in on a time scale of at least several hours at a sufficiently low temperature. A time scale that large is not suitable for XPCS.

For our experiment a free surface of glycerol was prepared in a large trough to obtain a flat surface, unaffected by the meniscus at the borders. Glycerol is a prototypical glass former with a calorimetric glass transition temperature of $T_G \approx -87^oC$. The trough was kept in a hermetically sealed evacuated cell with appropriate temperature control [2]. A mirror was used to direct the monochromatic, partially coherent beam down towards the horizontal glycerol surface with an incidence angle $\theta_i = 0.08^o$ well below the critical angle for total external reflection. The envanescent wave has in this case a penetration depth of only $\sim 80\AA$. The radiation scattered from the surface was detected using a direct illumination CCD camera from Princeton Instruments. The CCD camera was placed to record the off-specular diffuse scattering image from the surface with the lateral component of the momentum transfer within the scattering plane being $q_{||} = 2\pi/\lambda(\cos\theta_f - \cos\theta_i)$, where θ_f is the take-off angle with respect to the surface. The slowing down of surface dynamics is directly visualized by the CCD images obtained at two different glycerol temperatures, $T = -34.3^oC$ and -80.0^oC in the figure below. Further analysis of the scattering profiles is in progress.

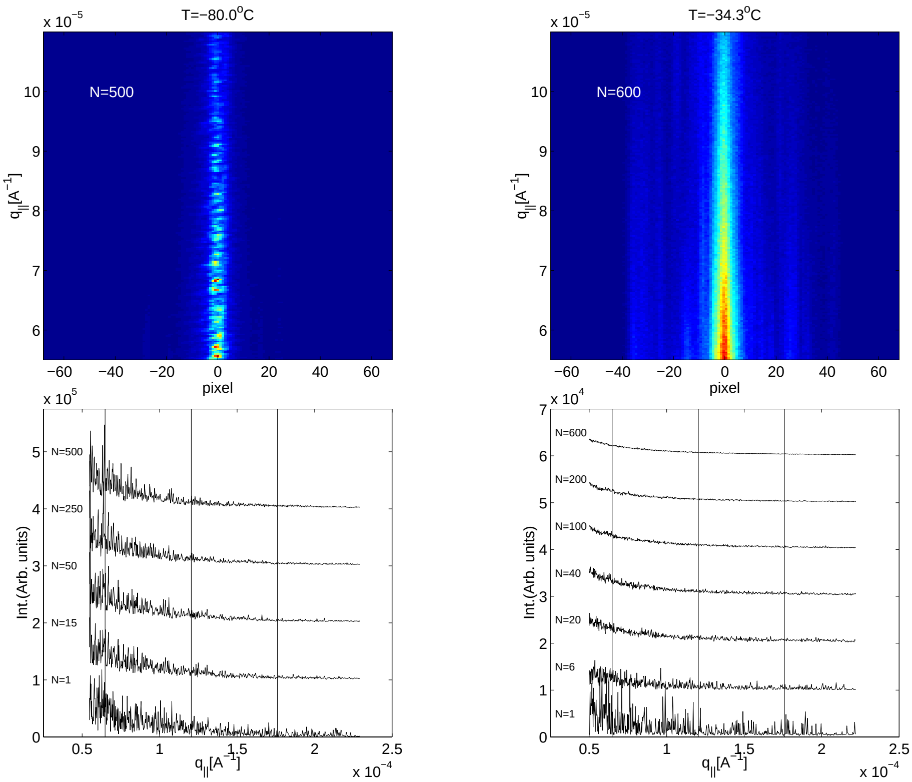

Figure 1. (Left): Image showing a speckle pattern obtained by summation of N=500 CCD exposures taken at $T = -80.0°$C(top). The evolution of the scattering profile as a function of N is also shown(bottom). (Right): Same as (left) but with $T = -34.3°$C. The difference between a dynamic and a static pattern(left) becomes evident.

References

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

[2] T. Seydel, PhD thesis, University of Kiel (2000) (http://www.ieap.uni-kiel.de/solid/ag-press/)

[3] J. Jäckle and K. Kawasaki, *J. Phys.: Condens. Matter* **7**, 4351 (1995)

[4] A. Braslau, P.S. Pershan, G. Swislow, B.M. Ocko, J. Als-Nielsen, *Phys. Rev. A* **38**, 2457 (1988)

[5] M.K. Sanyal, S.K. Sinha, K.G. Huang and B.M. Ocko, *Phys. Rev. Lett.* **66**, 628 (1991)

[6] A. Madsen, G. Grübel, T. Seydel and M. Tolan, submitted to *J. Sync. Radiation*