



	Experiment title: Diffusion of fluids in narrow pores	Experiment number: SC-4133
Beamline: ID10	Date of experiment: from: 17-July-2015 to: 20-July-2015	Date of report: 18-Feb-2016
Shifts: 12	Local contact(s): Federico Zontone & Yuriy Chushkin	<i>Received at ESRF:</i>
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Report:

How do dense fluids diffuse in narrow pores? This is a fundamental question of importance for many applications within the physical, chemical, materials, and life sciences, such as water transport in molecular sieves, hindered motion of colloidal systems in porous matrices, protein motion in cell membranes, transport of blood cells in narrow veins, and various micro- and nanofluidic applications. Spatial confinement induces microscopic ordering of the fluid constituents, which in turn alters the fluid's diffusive properties. However, confined fluids exhibit complex dynamical behavior, and a simple mechanistic description of diffusion in confined geometries is still missing.

We have recently developed a unique method to probe the microscopic structure of confined fluids at the fundamental level of pair densities, based on small-angle x-ray scattering (SAXS) from colloid-filled nanofluidic containers [1,2]. Here we extend the methodology to simultaneous determination of the microscopic static structure and collective diffusion, by carrying out high-energy SAXS and x-ray photon correlation spectroscopy (XPCS) experiments in parallel.

The data are exemplified in Fig. 1. In the inset we present the SAXS intensity obtained from a colloidal dispersion (182-nm-diameter silica particles dispersed in ethylene glycol) confined in narrow channels with a width of 2.8 particle diameters. The SAXS intensity is highly anisotropic, implying a strongly anisotropic structure factor similar as observed in Refs. [1,2]. Next we collected static and dynamic data simultaneously in three directions with respect to the confining walls, depicted by the white solid, dashed, and dashed-dotted lines. As an example, we present in the main figure intensity autocorrelation functions obtained from both the bulk and confined

fluid at a particular scattering vector magnitude of $q = 0.034 \text{ nm}^{-1}$. Clearly, spatial confinement leads to significantly slower dynamics compared to bulk.

Our main results can be summarized as follows. For bulk fluids the collective diffusion coefficient is known to scale as the inverse of the structure factor – a phenomenon known as de Gennes narrowing. For the confined fluid we observe the same effect. However, since the confined fluid’s structure factor is highly anisotropic [1,2], we observe anisotropic de Gennes narrowing. These experimental observations are essential for the development of a microscopic theoretical description of collective diffusion of dense confined fluids. We are in the process of publishing the results [3].

[1] K. Nygård et al., *Anisotropic pair correlations and structure factors of confined hard-sphere fluids: an experimental and theoretical study*, Phys. Rev. Lett. **108**, 037802 (2012).

[2] K. Nygård et al., *Density fluctuations of hard-sphere fluids in narrow confinement*, Phys. Rev. X **6**, 011014 (2016).

[3] K. Nygård et al., *Anisotropic de Gennes narrowing in confined fluids*, submitted to Phys. Rev. Lett. (2016).

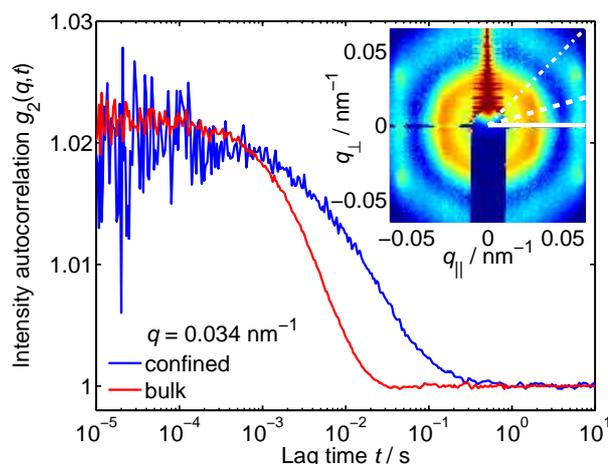


Fig. 1: Intensity autocorrelations collected from a dense colloidal dispersion (182-nm-diameter silica particles dispersed in ethylene glycol) at a scattering vector magnitude of $q = 0.034 \text{ nm}^{-1}$. Data are presented both in bulk (red) and narrow confinement (blue; slit width 2.8 particle diameters). Inset: Anisotropic SAXS intensity collected from the confined fluid, directly implying a highly anisotropic structure factor.