

Measuring Condensed Matter Dynamics Using Coherent X-Rays

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In the technique of x-ray photon correlation spectroscopy (XPCS) a coherent or partially coherent beam of x-rays illuminates a sample, and the resulting scattering pattern is modulated by a random speckle pattern. The speckles vary in time as the sample undergoes thermal fluctuations. Time autocorrelation of the speckle pattern yields information on the dynamics of a sample.

The first practical XPCS measurements were made in 1995 at the ESRF and NSLS^{1,2}. Since then the technique has been extensively developed both at the APS and ESRF and applied to a wide range of systems including, latex colloids^{3,4} ferro-fluids⁵, polymer blends⁶, surfaces^{7,8,9} spinodal decomposition of glasses¹⁰ and composition fluctuations in metal alloys¹. Measurements to date have studied either slow dynamics in glassy systems, or fast dynamics for very strongly scattering systems. New refinements in x-ray optics, and detector technology should permit future measurements on faster systems at shorter lengths, opening up the particularly exciting possibility of measuring the dynamics of biologically relevant material in aqueous solution.

Key to performing XPCS measurements is a partially coherent x-ray beam. The beam coherence has two components, transverse and longitudinal. For a source with an approximately Gaussian intensity distribution, the transverse coherence lengths depend on the source size, and source to sample distance: $\xi_{x,z} = \lambda R / 2\sigma_{x,z} \sqrt{\pi}$. Here, ξ is the coherence length at the sample in the x or z direction, λ the x-ray wavelength, R the sample to detector distance and σ is the Gaussian sigma of the source dimensions in the x or z direction. (assuming the y-axis lies along the beam). The longitudinal coherence length is given by $\Lambda \approx \lambda(E / \Delta E)$. Here, E is the energy of the x-ray beam and ΔE the band pass.

When the x-ray path length differences for all scattering events in the sample are smaller than the relevant coherence length, then the sample will give rise to a speckle pattern. This can typically be achieved by putting an aperture in front of the sample whose dimensions are comparable to the longitudinal coherence length (typically $\sim 20 \mu\text{m}$) and keeping the sample thickness small enough so that the scattering path length difference does not exceed the longitudinal coherence length. An example of a speckle pattern from a static scatterer (silica aerogel) is shown in figure 1.

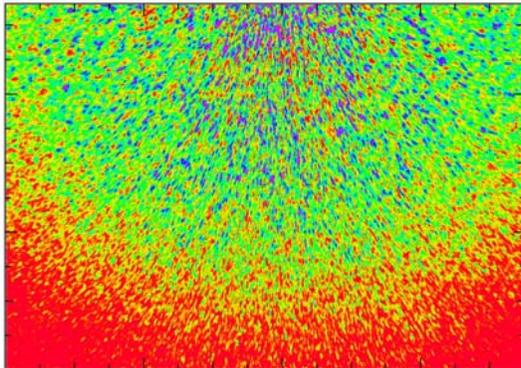


Figure 1. Coherent scattering pattern from a silica aerogel.

Dynamics in a sample can be probed by performing a time correlation of the intensity within a single speckle of the scattering pattern. In the case of a CCD detector, many correlation functions can be measured simultaneously and then averaged.¹¹ A typical fluctuation pattern for scattering from a dilute colloidal system, and its corresponding correlation function are displayed in figure 2. The intensity-intensity correlation function defined by $g_2(Q, \tau) = \langle I(Q, t)I(Q, t + \tau) \rangle / \langle I \rangle^2$, can be related to the dynamic structure factor of a sample via the relation $g_2(Q, \tau) = 1 + \beta [S(Q, \tau) / S(Q, 0)]^2$. The dynamic structure factor measures the fluctuations of the density within the sample and is defined via $S(\vec{Q}, \tau) = \langle \int e^{i\vec{Q}\cdot\vec{r}} \rho_e(0, 0) \rho_e(\vec{r}, \tau) d\vec{r} \rangle$, with ρ_e , the electron density.

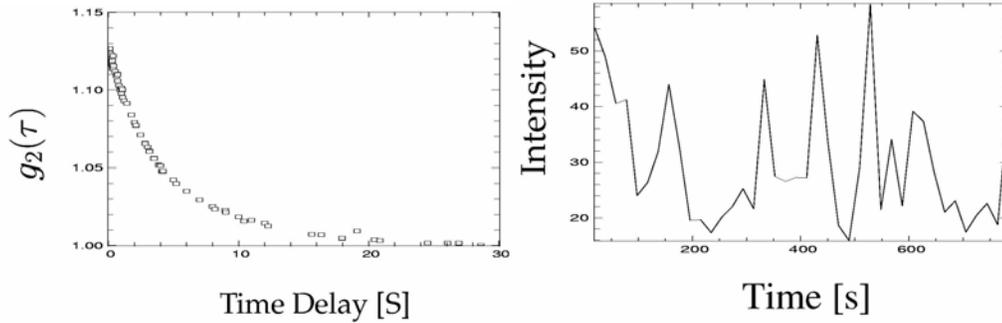


Figure 2. Intensity fluctuations from a speckle in the scattering pattern of a colloidal suspension, and its corresponding correlation function.

Consider the relatively simple example of the diffusion of a colloidal particle through a fluid (e.g. Brownian motion). In this case the normalized dynamics structure factor is an exponential decay; $S(Q, \tau) / S(Q, 0) \equiv f(Q, \tau) = e^{-\tau D Q^2}$. Here D is the particle diffusion coefficient. For dilute colloidal suspensions⁴ this form provides an excellent fit to the correlation functions, with the correct time constants. For more complicated systems the exponential form is often valid with a different Q dependence. Other behavior such as stretched exponentials, two exponentials decays or more complicated forms of the correlation function appear when the systems become densely packed and particle motions interfere.

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