X-Ray Photon Correlation Spectroscopy at a Third-Generation Synchrotron

The bright (high-flux) monochromatic and highly focused x-ray beams at third-generation synchrotron sources around the world are advancing the field of x-ray photon correlation spectroscopy (XPCS). In addition, newer CCD digital camera systems with high sensitivity and resolution have extended the lower-intensity range of x-ray detection in XPCS applications. This note describes several examples of XPCS in which high-performance CCD systems from Princeton Instruments can be used to capture microscopic, low x-ray-flux images.

XPCS at a Third-Generation Synchrotron

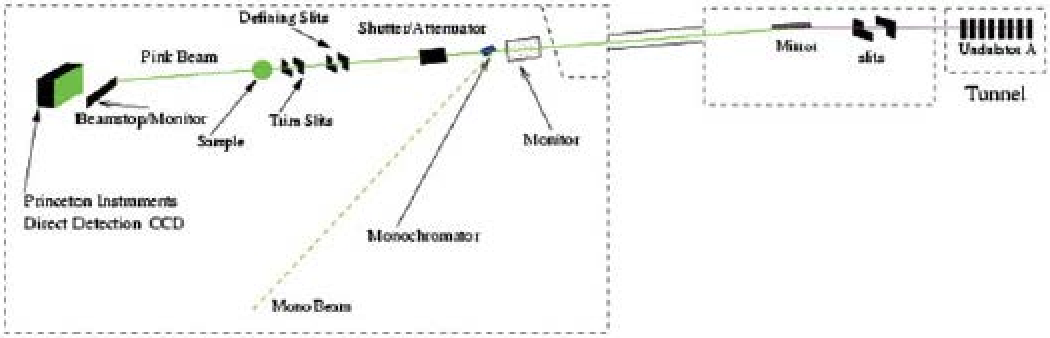
Recent experimentation has shown the potential for XPCS to become a powerful probe of sample dynamics in conditions of wave vector and frequency space that are inaccessible to various other light, neutron, or x-ray-scattering techniques. In particular, XPCS has several advantages over laser photon correlation spectroscopy. One of these is the ability to investigate smaller-scale phenomena using larger wave vectors. Another advantage is the smaller scattering cross section of x-rays compared to light, making measurements possible with almost no multiple scattering.

The Advanced Photon Source (APS) XPCS system at IMM-CAT (Sector 8) has a sufficiently coherent x-ray beam (yielding l_T^2 6μm in the horizontal and l_T^2 32μm in the vertical) that the time autocorrelation function of the resulting speckle pattern accurately yields the characteristic times of the sample. In addition, small-angle, coherent scattering experiments in the system are facilitated in two ways. First, approximately 10^5 more transversely coherent x-rays are available through a unit aperture than previously possible with second-generation sources. Second, the energy bandwidth of an undulator harmonic approximately matches the allowed energy bandwidth of the x-ray beam as derived from optical path-length difference considerations.

Figure 1 shows the APS XPCS setup. A Princeton Instruments PI·LCX camera (PI·LCX:1242) was used as the CCD detector. Additional methodological information is available at the web site http://www.imm.aps.anl.gov.

Figure 1.

APS small-angle, coherent x-ray-scattering setup.



Putting Coherent X-Rays to Use

Figure 2 shows the x-ray-induced speckle pattern from a 1.6-mm-thick, 95%-void SiO2 aerogel sample, obtained using a pink beam in the APS XPCS system. For this experiment, comparison of observed and expected (for the experimental configuration used) variation in speckle contrast and speckle widths with wave vector are shown graphically in Figure 3. The results indicated that there was fair agreement between the observed and expected values.

Figure 2.

Speckle pattern from SiO2 aerogel obtained using a pink beam.

Figure 3.

Top. Measured speckle contrast (circles) vs.wave vector compared to theory (line).

Bottom. Measured speckle widths (circles) vs. wave vector compared to theory (lines). Red circles are radial widths; violet circles are transverse widths.

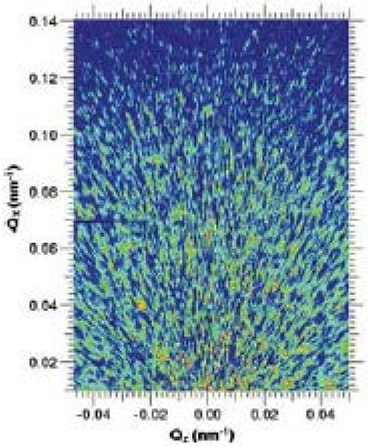


Figure 2

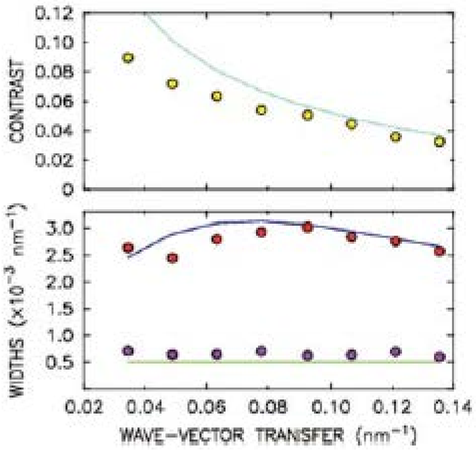


Figure 3

Colloidal suspensions are scientifically and technologically important from the perspective of paints, inks, cosmetics, and certain foods. In addition, a potential cutting-edge application is the possibility of creating self-assembling photonic bandgap materials made of colloidal crystals of semiconductor particles. A scattering pattern from a latex-in-glycerol colloid sample (particle radius R= 67 nm and volume fraction ø = 28%) is shown in Figure 4. Intensity autocorrelation functions for this experiment are depicted in Figure 5. It is apparent that the functions fit very well to single-exponential functions, as predicted by Brownian motion and Stokes-Einstein diffusion theories.

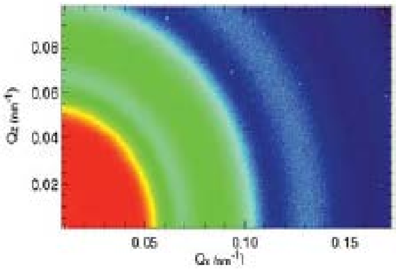


Figure 4

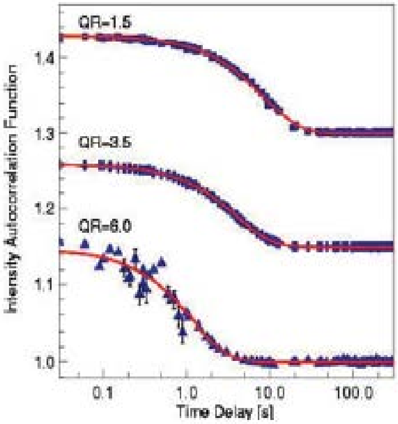


Figure 5

Figure 4.

Small-angle scattering pattern of latex in glycerol. The image represents the time average of scattering over 10 minutes at -5°C. Sample dynamics washed out the speckle contrast, making the image smoother than the one shown in Figure 2 above.

Figure 5.

Intensity autocorrelation functions for the experiment shown in Figure 4 at the indicated QR. Red lines are single- exponential fits.

Conventional incoherent x-ray scattering is insensitive to dynamics in systems where the average structure remains constant. By contrast, coherent x-ray beams allow measurements of microscopic dynamics in such cases. With the high brilliance available from third-generation synchrotrons (e.g., the APS), coherent scattering has become practical for a wide range of such applications. Figure 6 shows three sequential images of coherent x-ray-beam-induced speckle patterns from a borosilicate glass sample undergoing phase separation at high heat. When phase separation was initiated, a ring of scattering appeared at large wave numbers. As the domain coarsened, the ring moved to smaller wavelengths and became more intense with time.

For this type of system, previous work with incoherent x-ray beams showed that the average structure factors exhibited scaling behavior. In the dynamic measurements (such as those shown in Figure 6) carried out at the APS, it was observed that the correlation times of fluctuations about this average behavior also obeyed a scaling law. These results clearly indicate that correlation functions from matter illuminated by coherent x-rays are a useful quantitative probe of nonequilibrium system dynamics.

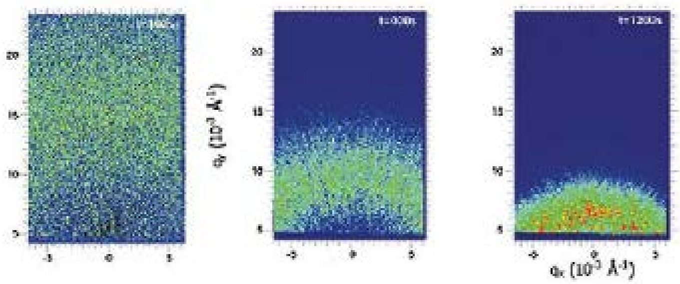


Figure 6.
X-ray-induced speckle patterns from a borosilicate glass sample undergoing phase separation at 963°K. Images are 1-sec exposures at the indicated time (t) after a quench from 1033°K. The horizontal and vertical components of q are indicated by qx and qy, respectively.

PI-LCX: 1300 Camera System

The Princeton Instruments PI-LCX: 1300 camera is a high-sensitivity, high resolution, deep depletion digital system designed for low x-ray flux imaging. With a 1340 x 1300 imaging array of 20.0 x 20.0 um pixels and 100% fill factor, this camera provides extremely high spatial resolution. A thin beryllium window in front vacuum seals the unit for deep cooling, protects the scientific-grade CCD, and reduces background by filtering low-energy x-rays. The thermoelectric cooling option provides dual speed control, whereas the liquid nitrogen cooling option offers single-speed control and very low dark current for long exposures. Other features and options include high frame-transfer capability with shutterless operation (optional), flexible binning and readout, 16-bit digitization, and PCI interface. The camera system runs under WinView, Princeton Instruments' versatile 32-bit Windows® software package designed specifically for high performance imaging.

A Selection of Cameras for X-ray Other Imaging

Princeton Instruments offers an array of high performance digital CCD systems for x-ray imaging applications. The Princeton Instruments Quad-RO, PIXIS-XO, PI-MTE and PIXIS-XF camera line offers X-ray detection for wide range of X-ray imaging and spectroscopy applications. They offer true 16 bit images at a readout rate of 2 MHz. These cameras also feature PI's world renowned low-noise electronics and cooling technology to allow detection of the faintest X-ray signals.

Princeton Instruments PIXIS CCD cameras were designed specifically for the most demanding, low-light imaging applications. They incorporate full-frame, front- or back-illuminated, scientific-grade CCDs, 16-bit digitization, a choice of thermoelectric or cryogenic cooling, and low noise electronics for maximum sensitivity and resolution. WinView software is included for easy operation and integration into application setups.

Custom fiber optic-coupling options for the CCDs are also available. High-performance CCD camera systems from Princeton Instruments offer the versatility to satisfy all sensitivity, dynamic-range, speed, and spatial resolution requirements for quantitative low-flux, x-ray imaging.

Appreciation is extended to the scientists at IMM-CAT, APS Sector 8, for providing information for this note. Some of the images in this note are reprinted from Synchrotron Radiation News (Copyright Overseas Publishers Association N.V.) with permission of Gordon and Breach Publishers.

References

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