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Fast two-dimensional detection for X-ray photon correlation spectroscopy using the PILATUS detector

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The first X-ray photon correlation spectroscopy experiments using the fast single-photon-counting detector PILATUS (Paul Scherrer Institut, Switzerland) have been performed. The short readout time of this detector permits access to intensity autocorrelation functions describing dynamics in the millisecond range that are difficult to access with charge-coupled device detectors with typical readout times of several seconds. Showing no readout noise the PILATUS detector enables measurements of samples that either display fast dynamics or possess only low scattering power with an unprecedented signal-to-noise ratio.

Keywords: XPCS; X-ray instrumentation; two-dimensional pixel detector; colloids; diffusion in liquids.

1. Introduction

Using partially coherent X-rays, X-ray photon correlation spectroscopy (XPCS) (Grübel & Zontone, 2004) measures the temporal intensity fluctuations of a speckle pattern caused by changes in the spatial arrangement of the scatterers in the sample. However, the need to select the coherent part of the incident X-ray beam reduces the scattered intensity. It is therefore highly desirable to maximize the signal-to-noise ratio (SNR) and to possibly ensure the detection of every scattered photon in an XPCS experiment, which can be achieved by the use of two-dimensional area detectors.

As the detection of scattered photons has to be as fast as the dynamics investigated, the time resolution of area detectors is a limiting factor. While for point detectors a time resolution down to 50 ns has been explored by Sikharulidze *et al.* (2002), for two-dimensional detectors the investigated time scales are of the order of seconds down to hundreds of milliseconds, and in the fastest case down to 2 ms (Falus *et al.*, 2004).

In recent years considerable effort has been put into the development of area detectors (Falus *et al.*, 2004; Johnson *et al.*, 2009) such as the Medipix2 detector (Llopart *et al.*, 2002) and area-detector data acquisition schemes (Lumma *et al.*, 2000) suited to XPCS experiments. At the Paul Scherrer Institute (PSI), Switzerland, a novel type of area detector has been developed: the PILATUS detector is a two-dimensional hybrid pixel array detector (Schlepütz *et al.*, 2005; Broennimann *et al.*, 2006) which can be operated with a frame rate of up to 300 Hz. As the PILATUS detector is a single-photon-counting detector, it offers unprecedented noise characteristics owing to the absence of dark current and readout noise, which is extremely important in an XPCS experiment, where the average number of photons per pixel per accumulation time is often smaller than unity.

2. Experimental details and results

As a model system for XPCS we used a hard-sphere silica system (volume fraction $\varphi \simeq 0.24$) suspended in polypropylene glycol with an average molecular weight of 4000 g l^{-1} (PPG 4000; Aldrich). The experiment was carried out at the coherent small-angle X-ray scattering beamline, cSAXS, at the Swiss Light Source, Switzerland, which is equipped with a PILATUS 2M detector consisting of 3×8 single modules. While the whole detector can be read out with a frame rate of 10 Hz, an increase of the frame rate can be achieved by reading out the two central modules of the detector, resulting in a maximum frame rate of 100 Hz. During the experiment we took 5000 pictures using an exposure time of 10 ms, followed by a readout time of 10 ms, thus resulting in a frame rate of 50 Hz. X-rays were monochromated to a photon energy of 8 keV, and the sample was placed 90 mm upstream of a circular pinhole aperture of diameter 12 µm. The PILATUS 2M detector was placed 7050 mm downstream of the sample.

The speckle size is given by $\sigma = (\lambda d)/a$, where λ is the wavelength, d is the distance between sample and detector, and a is the width of the illuminated area. For this set-up the speckle size was 91 µm both in the horizontal and the vertical. The speckle contrast β can be approximated by $\beta = 1/[1 + (P/\sigma^2)^2]$ where P is the pixel area and σ^2 is the speckle area. As the PILATUS detector has a pixel size of 172 µm $\times$ 172 µm, the calculated contrast of this set-up is 0.07.

By summing up 5000 images (Fig. 1*a*) and azimuthally integrating, the Q-dependent static scattering intensity is obtained. Fitting a sphere form factor to the high Q-values of the data gives a mean radius of R = 270 Å and a polydispersity of $\Delta R/R$ = 0.13. The static intensity distribution $I(Q)$ of a concentrated sample is given by $I(Q) = S(Q)\,P(Q)$. Here, $S(Q)$ is the static structure factor and $P(Q)$ is the

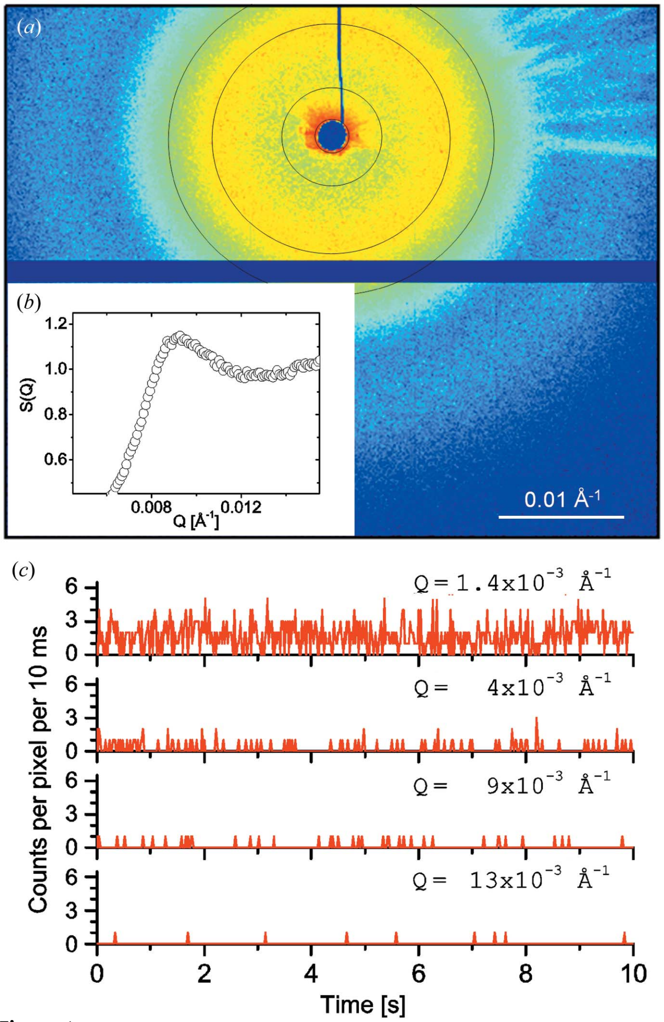

Figure 1
(*a*) Sum of 5000 images. The four black circles mark the Q-values of the pixels shown in (*c*). (*b*) Static structure factor of the sample with $S(Q)_{\rm max}$ at Q = 0.009 Å^{-1}. (*c*) Counts per pixel per frame (10 ms of data acquisition time) shown for four arbitrarily chosen pixels at Q-values of 0.0014, 0.004, 0.009 and 0.013 Å^{-1} during the first 10 s of the experiment.

particle form factor. By dividing the measured intensity values by the intensity of a dilute sample (volume fraction $\varphi \simeq 0.01$) the static structure factor is obtained as shown in Fig. 1(*b*). The maximum of the structure factor $S(Q)_{\rm max}$ is found at Q = 0.009 Å^{-1} corresponding to a mean particle distance of ~700 Å.

For the dynamic analysis of the images the pixels between $Q_{\rm min}$ = 0.0022 Å^{-1} and $Q_{\rm max}$ = 0.012 Å^{-1} have been analyzed. In Fig. 1(*c*) the counts per pixel per frame for four different pixel positions on the detector are shown. Even at Q = 0.0014 Å^{-1}, a position close to the direct beam, the counts are fluctuating between 0 and 5. At larger momentum transfers the count rate decreases and the most likely value is 0.

Noise-free photon detection leads to a superior SNR. The SNR in an XPCS experiment is given by an expression derived by Jakeman (1973) and adapted for area detectors by Falus *et al.* (2004): SNR = $\beta I(T\tau_{\rm a})^{1/2}\eta(\tau_{\rm a}F)^{1/2}(n_{\rm pix})^{1/2}$. Here η is the quantum efficiency, $n_{\rm pix}$ is the number of pixels per frame, β is the speckle contrast, I is the intensity per pixel, T is the total measurement time, and $\tau_{\rm a}$ is the accumulation time per frame. F, the frame rate, is related to the delay time between consecutive pictures *via* $F = 1/(\tau_{\rm a} + \tau_{\rm t})$ where $\tau_{\rm t}$ is the readout time of the detector. Calculating the SNR in the region used for the dynamic analysis yields a SNR of 3872. In the case of an ideally efficient ($\eta = 1$) point detector ($n_{\rm pix} = 1$) showing no readout time ($\tau_{\rm t} = 0$) the formula reduces to SNR = $\beta I(T/F)^{1/2}$, yielding a SNR of 19 for the same experimental parameters. The comparison between the SNR of the PILATUS detector and an ideal point detector yields a factor of ~200 in favour of the PILATUS detector.

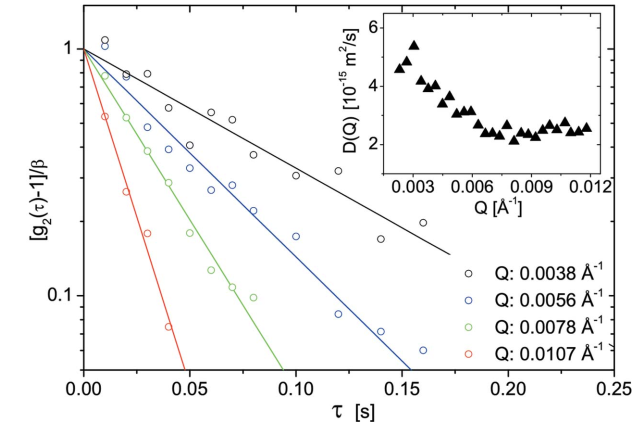

Figure 2
Normalized intensity autocorrelation functions $g_2(\tau) - 1$ and corresponding fits at Q-values of 0.0038, 0.0056, 0.0078 and 0.0107 Å^{-1}. The inset shows the wavevector-dependent diffusion coefficient $D(Q)$.

For the dynamics analysis the region of interest was divided into 27 equidistant constant-Q rings. All pixels inside a ring were binned to a mean Q-value. The intensity autocorrelation function was calculated for each pixel according to (Lumma *et al.*, 2000)

$$g_2(k) = \frac{(1/N)\sum_{k=0}^{N-1} I(t)\,I(t+k)}{\left\langle \left[(1/N)\sum_{k=0}^{N-1} I(t+k) \right]^2 \right\rangle}, \qquad (1)$$

where N is the total number of frames and k is the frame number. $\langle \ldots \rangle$ denotes the azimuthal average of all pixels binned to a mean Q-value with a width five times narrower than that used for the dynamics analysis. The intensity autocorrelation functions can be fitted by $g_2(\tau) = 1 + \beta\exp[-2\Gamma(Q)\tau]$ where β denotes the speckle contrast and $\Gamma(Q)$ is the relaxation rate. The experimentally measured speckle contrast was about $\beta = 0.07$, in good agreement with the theoretical predicted value.

The dynamics becomes faster with increasing momentum transfers, as shown by the normalized intensity autocorrelation functions in Fig. 2. In the absence of interparticle interactions, the particles migrate freely with the Stokes–Einstein diffusion coefficient D_0. At higher colloid concentrations, interparticle interactions as well as indirect hydrodynamic interactions mediated by the solvent come into play, which can be described by the wavevector-dependent diffusion coefficient $D(Q) = \Gamma(Q)/Q^2$ shown in the inset of Fig. 2.

3. Conclusions

The PILATUS detector opens the possibility of investigating samples showing dynamics in the millisecond range and thus allows new types of XPCS experiments to be conducted using area detectors. Two developments can account for this achievement: first, the PILATUS detector is able to record images with a frame rate of up to 300 Hz; second, the PILATUS as a noise-free single-photon-detecting hybrid pixel array detector allows accurate measurements of images showing a relatively low scattering signal. This development is crucial for the fast recording of two-dimensional images as the intensity per image is simultaneously decreasing as the data acquisition time becomes shorter. In particular, the extremely high SNR achieved in this experiment yields intensity autocorrelation functions of high precision. Further developments of the detector are planned: a novel PILATUS detector will make it possible to record images with a frequency of up to 10 kHz. This progress in detector technology will make it possible to explore systems showing dynamics over the whole millisecond range.

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