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Abstract: We present a new method to extract the intermediate scattering function from series of coherent diffraction patterns taken with 2D detectors. Our approach is based on analyzing speckle patterns in terms of photon statistics. We show that the information obtained is equivalent to the conventional technique of calculating the intensity autocorrelation function. Our approach represents a route for correlation spectroscopy on ultrafast timescales at X-ray free-electron laser sources.

OCIS codes: (320.7100)Ultrafast measurements; (300.6480) Spectroscopy, speckle; (140.2600) Free-electron lasers (FELs)

References and links

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1. Introduction

Complex dynamics on nanometer length scales is an omnipresent phenomenon which is investigated at the frontier of condensed matter research. Many systems display dynamic features on ultra-fast time scales and nanometer length scales. Systems of interest range from domain switching in correlated electron materials, crystalline phase transitions, magnetic dynamics, protein folding towards glassy non-equilibrium dynamics. As the time scales involved range from femtoseconds to seconds the method of choice to investigate the dynamic features is X-ray photon correlation spectroscopy (XPCS) using ultrafast X-ray sources.

XPCS utilizes a coherent X-ray beam to illuminate a disordered sample which results in a speckle pattern in direct relation to the sample's exact electron density distribution. Fluctuations within the sample cause this speckle pattern to fluctuate accordingly and the measured quantity

#101144 - $15.00 USD Received 5 Sep 2008; revised 14 Nov 2008; accepted 17 Nov 2008; published 22 Dec 2008

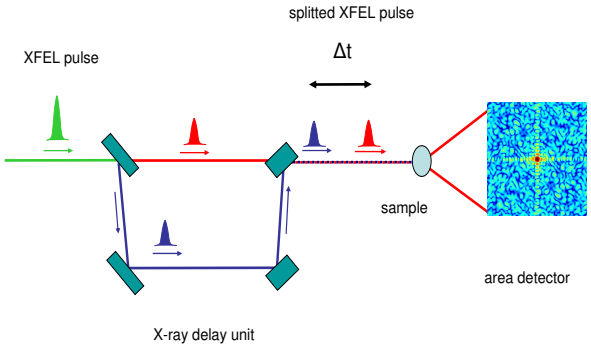

Fig. 1. Schematics of the split-pulse technique. A delay line unit consisting of mirrors and beam-splitters produces two equal intensity pulses travelling along the same path but delayed in time. Each pulse produces a speckle pattern and the sum is recorded on an area detector and analyzed in terms of speckle contrast.

in XPCS experiments is the intensity autocorrelation function $g_2(\tau) = < I(t)I(t+\tau) > / < I(t) >^2$. Assuming Gaussian fluctuations the intensity autocorrelation function can be rewritten with the help of the Siegert relation [1] as

$$g_2(\tau) = 1 + |f(\tau)|^2, \tag{1}$$

where $f(\tau)$ is the intermediate scattering function reflecting the dynamics of the sample.

XPCS has developed into a valuable tool for measuring slow dynamics (typically 10^{-2} to 10^6 Hz [2]) in the time domain as X-rays provide several advantages when compared to optical light such as, e.g., short wavelength, high penetration power, surface sensitivity and element specificity. However, the access to ultrafast dynamics on nanometer length scales is being hampered by the limited photon flux at 3rd generation synchrotron sources and the relatively long readout time of area CCD detectors.

In contrast to today's x-ray sources single-pass free-electron lasers based on self-amplified spontaneous emission (SASE) will provide uniquely intense, coherent, polarized, short-pulse radiation in the X-ray regime. At the European XFEL, for example, up to 3000 pulses will be delivered with a temporal spacing of 200 ns between each pulse and an overall repetition rate of 10 Hz [3]. This peculiar time structure of SASE-based sources excludes the classical sequential way of measuring intensity autocorrelation functions with arbitrary lag times τ. Therefore a different scheme based on a split-pulse technique has been proposed [4]. The concept of this technique is to split each x-ray pulse into two equal-intensity pulses separated in time, but propagating along the same path. The time separation is achieved via a delay path similar to delay line units used in optical laser technology (see Fig. 1). The scattering from the two pulses will then be collected during the same exposure on an area detector making ultrafast time resolution of area detectors unnecessary. Instead an analysis of the speckle contrast of the recorded image as a function of the delay time yields *direct* information about the intermediate scattering function of the sample.

Here we show with the help of coherent X-ray data taken at a 3rd generation synchrotron

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source that our proposed method is in fact working and the intermediate scattering function can be retrieved by analyzing the speckle contrast of summed CCD images. We thus prove that our approach allows to perform correlation spectroscopy experiments with femtosecond x-ray pulses.

2. Methodology

2.1. Sequential technique

The radiation scattered from a disordered sample produces a speckle pattern on an area detector with N pixels. Suppose that we take a series of K images with an exposure time t_e at an interval Δt which are read out and stored on a hard drive. The normalized intensity autocorrelation function sampled in the classical sequential way can then be calculated for each pixel n as

$$g_2(n,k) = \frac{1}{K-k}\sum_{i=1}^{K-k} I_n(t_i)I_n(t_i+k)/\bar{I}_n^2 \tag{2}$$

where k is the lag time, $I_n(t_i)$ the intensity of pixel n at time t_i and $\bar{I}$ the temporal averaged intensity of pixel n. Averaging over pixels leads to the intensity autocorrelation function $g_2(k) = 1/N\sum_{n=1}^{N} g_2(n,k)$. Effects of partial coherence are accounted for by introducing a contrast factor β^2 in the Siegert relation, yielding

$$g_2(k) = 1+\beta^2|f(k)|^2. \tag{3}$$

The fastest accessible time scale, i.e., the minimum lag time is given by $k_{min} = \Delta t + t_e$. Therefore the sequential scheme relies on area detectors with fast enough read out times and it is restricted to time scales on the order of 10^{-6} seconds [4].

2.2. Split-pulse technique

The split-pulse technique makes ultra-fast detection of intensities unnecessary and is therefore the ideal tool for measuring dynamics in the time domain faster than 10^{-6} s. In our split-pulse approach the area detector records the intensities of two equal intensity pulses delayed by a time k. The intensity at the detector is therefore $S(k) = I(t_i) + I(t_i+k)$, where $I(t_i)$ denotes the speckle pattern at time t_i. Neglecting shot noise the contrast C of a speckle pattern is given by the variance $\sigma^2(S)$ and the mean intensity of the speckle pattern via $C = \sqrt{\sigma^2(S)/S^2}$ [5]. For a fully coherent beam, C^2 lies between 1 for adding two identical speckle patterns and 0.5 for adding two completely uncorrelated patterns. In case the typical correlation time τ of the sample is much larger than the time delay k the two patterns $I(t_i)$ and $I(t_i+k)$ will be identical and the contrast of the summed image $S(k)$ is close to 1. In contrast, if the correlation time of the sample is much smaller than the delay time k then the two speckle patterns $I(t_i)$ and $I(t_i+k)$ will be very different and the contrast of the summed image is lower. By varying the delay time one can map out the dynamics of the sample.

More precisely, the measured quantity in such an experiment is the normalized variance of $S(k)$

$$c_2(k) = \frac{\langle S^2(k)\rangle - \langle S(k)\rangle^2}{\langle S(k)\rangle^2}. \tag{4}$$

$\langle S^2(k)\rangle$ can be expressed as

$$\langle S^2(k)\rangle = \langle [I(t)+I(t+k)][I(t)+I(t+k)]\rangle = 2\langle I^2\rangle + 2\langle I(t)I(t+k)\rangle, \tag{5}$$

and the squared mean intensity is

$$\langle S(k)\rangle^2 = \langle I(t)+I(t+k)\rangle^2 = 4\langle I\rangle^2. \tag{6}$$

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With this we obtain

$$c_2(k) = \frac{2\langle I^2\rangle + 2\langle I(t)I(t+k)\rangle - 4\langle I\rangle^2}{4\langle I\rangle^2}, \tag{7}$$

which can be rewritten as

$$c_2(k) = \frac{2\sigma^2(I) + 2\langle I(t)I(t+k)\rangle - 2\langle I\rangle^2}{4\langle I\rangle^2}. \tag{8}$$

As we are assuming a fully coherent beam the normalized variance of the single speckle pattern is one and the first term in Eq.(8) is

$$\frac{1}{2}\frac{\sigma^2(I)}{\langle I\rangle^2} = \frac{1}{2}. \tag{9}$$

Using the Siegert relation for Gaussian signals we write for the second term in Eq.(8)

$$\frac{\langle I(t)I(t+k)\rangle}{2\langle I\rangle^2} = \frac{1}{2}(1+|f(k)|^2), \tag{10}$$

where $f(k)$ is the intermediate scattering function of the system. So we find for the normalized speckle contrast

$$c_2(k) = \frac{1}{2}(1+|f(k)|^2). \tag{11}$$

Abberations due to extra optics will lead to a loss of coherence. Taking such effects of partial coherence (β) and shot noise (α) into account Eq. (11) yields

$$c_2(k) = \frac{\beta^2}{2}(1+|f(k)|^2) + \alpha. \tag{12}$$

This shows that the information obtained by the split-pulse technique is identical to the one obtained by the sequential technique.

In case the intensities of the two speckle pattern falling onto the detector are not equal we obtain the expression

$$c_2(k) = \beta^2\left(\frac{r^2+1+2r|f(k)|^2}{r^2+1+2r}\right), \tag{13}$$

with r denoting the ratio between the two intensity pattern. Maximum contrast can be achieved with a ratio $r = 1$.

2.3. Photon statistic of the split-pulse technique

Further insight can be gained by analyzing the summed speckle patterns S in terms of their counting statistics. The probability distribution function $P(i)$ for observing i counts in a speckle pattern is

$$P(i) = \frac{\Gamma(i+M)}{\Gamma(M)\Gamma(i+1)}\left(1+\frac{M}{\bar{i}}\right)^{-i}\left(1+\frac{\bar{i}}{M}\right)^{-M} \tag{14}$$

where $\bar{i}$ is the mean count rate and M is the number of modes [1]. The number of modes is a measure of the contrast of a speckle pattern and therefore reflects the underlying dynamics of the sample. That is for a delay time smaller than the sample correlation time the number of modes M contributing to the speckle pattern is smaller than for delay times larger than the sample correlation time. Thus the probability distribution function $P(i)$ will change as a function of delay time k.

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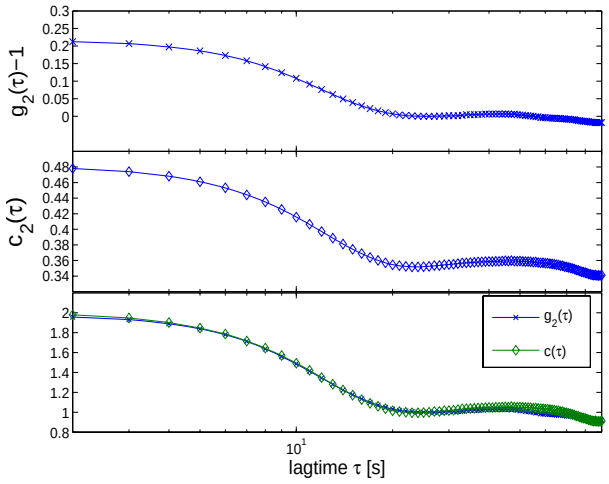

Fig. 2. Top: Normalized intensity autocorrelation function $g_2(\tau)-1$ deduced in the sequential mode. Middle: Speckle contrast correlation function $c_2(\tau)$ as calculated from the split-pulse technique. Bottom: Both correlation functions normalized to each other.

3. Experiment and analysis

In order to prove the concept we performed surface-sensitive XPCS experiments using the coherent X-ray beam of the ID10A beam line (ESRF) with a photon wavelength of 1.54 Å. The energy bandpass of the single bounce Si(111) monochromator leads to a longitudinal coherence length $\xi_l = \lambda^2/\Delta\lambda \approx 1\mu$m. A pinhole with a diameter of 20 μm is placed upstream of the sample in order to obtain a collimated and transversely coherent beam. The beam is tilted vertically by a mirror onto the sample surface resulting in an incident angle $\alpha_i = 0.12°$. The sample investigated was a colloidal suspension of silica particles immersed in a high viscosity polymeric liquid PPG-4000 [6]. The GISAXS signal from the colloidal particles at the surface is recorded by a Princeton direct illumination charge-coupled device (CCD) camera with 1242 $\times$ 1152 pixels and a pixel size of 22.5×22.5 μm^2. The exposure time was 1s and typically series of 500 - 1000 images were recorded and stored on hard disk. In addition a series of dark images with the same exposure time was recorded and its average subtracted from the speckle patterns. The sample was placed in a housing consisting of a two chamber design, where the outer cell was evacuated for thermal isolation and the inner one contained a stainless steel trough of 120 mm diameter and 0.2 mm height. The sample was filled into the trough ensuring a homogenous and flat surface. The air above the sample was replaced by helium gas. Sample cooling was achieved by evaporating liquid nitrogen in a heat exchanger underneath the sample chamber with a constant flow rate.

From the CCD images we selected a region of interest containing 2667 pixel. The mean count rate in the region of interest was 1.8 photons per pixel per second. The normalized intensity autocorrelation function recorded in the sequential way, Eq. (3), is shown in Fig. 2 (top). The correlation function displays a compressed exponential function representing the motion of the silica particles with a typical correlation time of 10 seconds. At larger lag times a second correlation is becoming visible due to a drifting of the overall intensity. The optical contrast of the correlation function is $\beta^2 = 0.21$. At large lag times the intensity correlation function lies below the unbiased value of 1.0 because Eq. (1) represents a *biased* estimator of the normalized intensity autocorrelation function [8].

Figure 2 (middle) shows the speckle contrast correlation function $c_2(k)$ obtained with the

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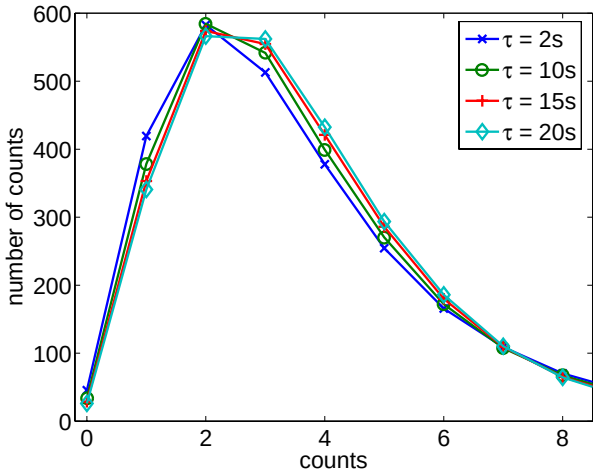

Fig. 3. Intensity distribution function P(i) for the sum of two speckle patterns with different delay times τ.

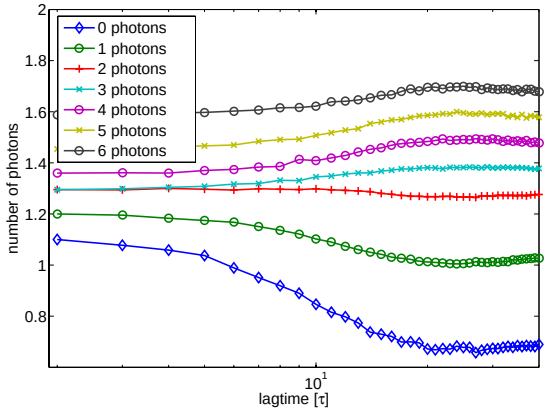

Fig. 4. Number of photons observed in the summed speckle patterns as a function of delay time. Curves are shifted and renormalized for reasons of clarity.

help of Eq. (4). This correlation function decays from a maximum value of 0.48 to a baseline of 0.35. As in the case of the sequential autocorrelation function a long term intensity drift is apparent and also values of c_2 below the apparent baseline at 0.35. The optical contrast of the correlation function $c_2(k)$ is $\beta^2 = 0.25$ which is in agreement with the contrast obtained from the sequential method. The bottom of Fig. 2 displays both correlation functions normalized to each other. The good agreement between both correlation functions for the short time scale dynamics is apparent. The long-term intensity fluctuations at lag times larger than 30 s are however slightly different in both correlation approaches which is attributed to the differences in the averaging procedure.

In connection with Eq. (14) it is also interesting to investigate the photon statistics of the summed speckle pattern. Figure 3 displays histograms of summed speckle patterns with lag times between 2 s and 20 s. Obviously, the probability distribution function $P(i)$ of the summed speckle patterns is shifting to the right as a function of lag time. This implies that the number

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of modes M of the summed speckle patterns is increasing with increasing lag time, i.e., the contrast is decreasing. Thus one may as well analyze the statistics of the photon distribution function to retrieve the lag-time dependent degree of coherence $M(k)^{-1}$. As an illustration we plot in Fig. 4 the number of photons contained in $S(t_i,k)$ as a function of delay time between the two summed intensities. An inspection of Eq. (14) reveals that $P(i)$ is not directly proportional to the number of modes M and therefore does not give direct access to the intermediate scattering function. However, the similarity between the curves is striking and it becomes clear that the basic feature of intensity correlation functions, namely the correlation time τ_0, is already accessible by counting the number of zero photons in a speckle pattern as a function of pulse delay time.

4. Conclusion

In conclusion, we have introduced a new method to analyze 2D speckle patterns in terms of speckle contrast. The speckle contrast correlation function allows to determine directly the intermediate scattering function and thus our approach represents the route for correlation spectroscopy experiments at future SASE-based ultrafast X-ray sources accessing picosecond timescales with nanometer spatial resolution.

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