

X-ray photon correlation spectroscopy

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In recent years, X-ray photon correlation spectroscopy (XPCS) has emerged as one of the key probes of slow nanoscale fluctuations, applicable to a wide range of condensed matter and materials systems. This article briefly reviews the basic principles of XPCS as well as some of its recent applications, and discusses some novel approaches to XPCS analysis. It concludes with a discussion of the future impact of diffraction-limited storage rings on new types of XPCS experiments, pushing the temporal resolution to nanosecond and possibly even picosecond time scales.

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1. Introduction

X-ray photon correlation spectroscopy (XPCS) is a spatio-temporal coherent X-ray probe that measures nanoscale dynamics based on observations of intensity fluctuations within coherent X-ray speckle produced (typically detected in the far-field) due to interference from randomly distributed scatterers within the sample. A typical set-up for XPCS is shown in Fig. 1 (Cummins & Pike, 1974; Pecora, 1985; Sutton, 2008; Livet, 2007; Shpyrko *et al.*, 2007).

X-ray scattering intensity correlations in momentum space are directly related to fluctuation of the order parameter in real space. The advantage of XPCS is the ability to simultaneously characterize a volume of the sample containing a large number of scatterers, while at the same time retaining the spatial sensitivity defined by the dimensions of the speckle pattern, typically of the order of tens of nanometers. XPCS is therefore ideally suited for probing slow nanoscale dynamics in the bulk, not accessible with most scanning probes, over the range of length scales (wavevectors) not accessible by visible-light probes, and time scales (energy resolution) not accessible

by inelastic X-ray or neutron scattering techniques (see Fig. 2). Additional advantages of XPCS over optical probes is no complications due to multiple scattering, the ability to study opaque samples, surface sensitivity (in grazing-incidence geometry) as well as magnetic, orbital or chemical specificity due to resonant X-ray scattering.

The major current limitation of XPCS is the relatively small time window in which one can study slow fluctuations (typically ranging from time scales of the order of tens of milliseconds to thousands of seconds). This is primarily due to the relatively low coherent flux currently available even at state-of-the-art synchrotron facilities, and the fact that the XPCS requires measurement of correlations between pairs of photons rather than individual photons. The latter causes the

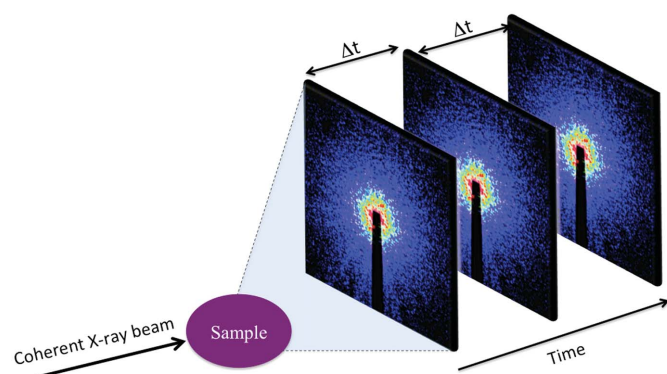


Figure 1
Schematic measurement procedure for XPCS measurements.

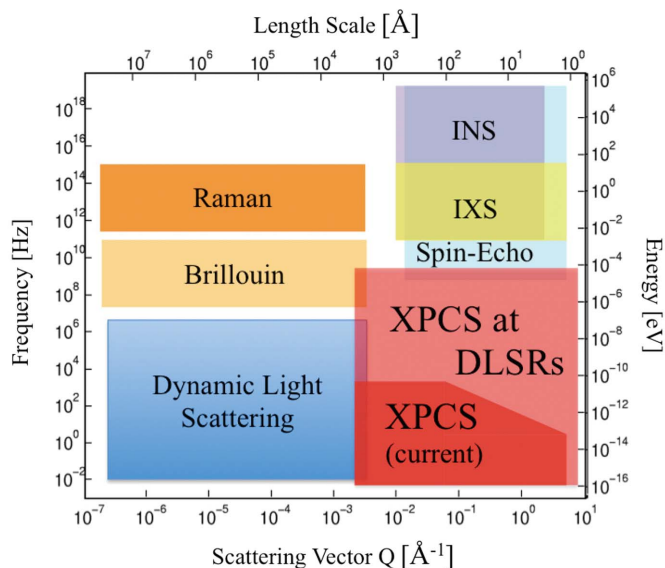


Figure 2
Phase map of spatiotemporal techniques accessing the dynamic structure factor $S(Q, \omega)$, showing the ranges of Q and ω typically accessible with XPCS at current sources and at DLSRs.

signal-to-noise ratio to scale with intensity instead of the square root of intensity, as discussed below. Low coherent flux can be partially mitigated by using a multi-speckle detection scheme, but then the fastest dynamics are generally limited by the readout rate of the X-ray area detector. X-ray damage is another important factor to consider, especially in soft matter and biological systems; this typically limits the longest time scales that can be probed (along with long-term beam/sample stability), but can be remedied at intermediate time scales by renewing the sample, *e.g.* by flowing it across the beam spot.

Correlations between series of two-dimensional speckle patterns is represented by the intensity autocorrelation function $g_2(\mathbf{Q}, t)$, calculated for each pixel of area detector and averaged over the pixels within the same range of wavevector transfers \mathbf{Q} ,

$$g_2(\mathbf{Q}, t) = 1 + A[S(\mathbf{Q}, t)/S(\mathbf{Q})]^2 = \frac{\langle I(\mathbf{Q}, t)I(\mathbf{Q}, t + \tau) \rangle_\tau}{\langle I(\mathbf{Q}, \tau) \rangle_\tau^2}, \quad (1)$$

where $I(\mathbf{Q}, t)$ and $I(\mathbf{Q}, t + \tau)$ are the intensities in a given pixel for frames taken at times t and $t + \tau$, respectively, and averaging is carried out over all τ while keeping the time lag t constant (Dierker *et al.*, 1995; Thurn-Albrecht *et al.*, 1996; Brauer *et al.*, 1995; Mochrie *et al.*, 1997). The resulting g_2 function, calculated for each individual detector pixel, is then averaged over a particular region of interest. It contains important information on spatial and temporal behavior of the system. The factor A in (1) is the so-called Siegert factor, equivalent to speckle visibility expected for a completely static sample, and directly related to the degree of X-ray beam coherence (Cummins & Pike, 1974; Pecora, 1985).

The intermediate scattering function $S(\mathbf{Q}, t)/S(\mathbf{Q})$, from now on assumed to be isotropic in reciprocal space, typically exhibits an exponential decay with a characteristic relaxation time constant τ (generally a function of Q): $S(\mathbf{Q}, t)/S(\mathbf{Q}) = \exp[-(t/\tau)^\beta]$, where β is the parameter that defines the shape of the exponential decay, either a stretched ($\beta < 1$) or a compressed ($\beta > 1$) component.

Since the coherent fraction of radiation from third-generation undulator sources is typically of the order of 0.1%, a spatial filtering of the full beam is usually needed in order to increase the Siegert factor, *i.e.* the speckle contrast, to values in the range 0.1–0.5. The contrast is reduced by relative sample-beam instabilities caused by beamline components such as optics or the sample environment. Therefore, a careful design is required to obtain a high degree of mechanical stability over long time scales (hours) (Sandy *et al.*, 2007).

XPCS has been applied to a number of condensed matter systems, including soft materials such as colloids (Dierker *et al.*, 1995; Narayanan *et al.*, 2007; Thurn-Albrecht *et al.*, 1996; Lu *et al.*, 2008; Mochrie *et al.*, 1997; Gao & Kilfoil, 2007; Duri *et al.*, 2009; Fluerau *et al.*, 2007; Lal *et al.*, 2001; Robert *et al.*, 2008; Spannuth *et al.*, 2011; Trappe *et al.*, 2007; Zhang *et al.*, 2011; Guo *et al.*, 2007; Chung *et al.*, 2006; Bandyopadhyay *et al.*, 2004), liquid crystals (Madsen *et al.*, 2003; Sikharulidze *et al.*, 2002, 2003; Relaix *et al.*, 2011), polymers (Falus *et al.*, 2005, 2006a,b; Guo *et al.*, 2009, 2012; Hernandez *et al.*, 2014; Hongyu

et al., 2012; Mochrie *et al.*, 1997; Narayanan *et al.*, 2007; Orsi *et al.*, 2011; Patel *et al.*, 2010), capillary fluctuations (Gutt *et al.*, 2003, 2007; Jiang *et al.*, 2007; Kim *et al.*, 2003; Madsen *et al.*, 2004; Alvine *et al.*, 2012; Frieberg *et al.*, 2013; Liu *et al.*, 2013) as well as metals and alloys (Brauer *et al.*, 1995; Brock & Sutton, 2008; Fluerau *et al.*, 2005; Livet & Sutton, 2012; Malik *et al.*, 1998; Sanborn *et al.*, 2011; Mueller *et al.*, 2011; Leitner *et al.*, 2009), metallic and molecular glasses (Leitner *et al.*, 2012; Ruta *et al.*, 2012, 2013; Sikorski *et al.*, 2010) and several electronic and magnetic systems (Holt *et al.*, 2007; Su *et al.*, 2012; Sutton *et al.*, 2002; Shpyrko *et al.*, 2007; Chen *et al.*, 2013; Konings *et al.*, 2011). While the vast majority of XPCS experiments have been conducted using hard X-rays (5–10 keV range), a few recent experiments extend XPCS to soft X-rays, mostly aiming to take advantage of resonant X-ray scattering at the magnetic resonant edges of transition metals and rare earths or coupling to helical magnetic structures (Konings *et al.*, 2011; Chen *et al.*, 2013). More detailed reviews of recent XPCS applications can be found elsewhere (Livet & Sutton, 2012; Sutton, 2006, 2008; Brock & Sutton, 2008; Livet *et al.*, 2006; Leheny, 2012; Grübel *et al.*, 2007, 2008; Ginzburg, 1958).

2. XPCS analysis beyond $g_2(\mathbf{Q}, t)$

2.1. Relaxation exponents: compressed versus stretched exponents

In addition to yielding the characteristic lifetime of the fluctuations, the line shapes of autocorrelation functions often have a deep physical meaning. For example, in XPCS studies of antiferromagnetic domain wall dynamics in chromium (Shpyrko *et al.*, 2007), the decay of $g_2(\mathbf{Q}, t)$ was found to follow a compressed exponential with the power law $\beta = 1.5$, similar to the compressed exponential found in a wide range of soft matter systems (gels, sponges, clays, emulsions) undergoing jamming transitions (Cipelletti *et al.*, 2000, 2003a; Bandyopadhyay *et al.*, 2004; Guo *et al.*, 2007; Falus *et al.*, 2006b). This is considered to be a universal signature of collective relaxation behavior (Cipelletti & Ramos, 2002, 2005; Cipelletti *et al.*, 2003a; Ruta *et al.*, 2013; Falus *et al.*, 2006a). Several experiments indicate that the nanoscale dynamics of many electronic and magnetic systems are intrinsically of a ‘glassy’ nature (Shpyrko *et al.*, 2007; Jaramillo *et al.*, 2007; Chen *et al.*, 2013; Su *et al.*, 2012; Kim *et al.*, 2013; Sutton *et al.*, 2002; Zhou *et al.*, 2012), and resembles the aging, memory, rejuvenation and other behavior that is traditionally associated with granular and soft matter systems as well as molecular or atomic glasses (Cipelletti *et al.*, 2000; Cipelletti & Ramos, 2002, 2005). In particular, relaxation dynamics observed in, for example, antiferromagnetic domains in chromium, dysprosium and charge density wave condensates exhibit compressed exponents ($\beta = 1.5$) that are commonly found in soft matter systems undergoing a jamming transition. More detailed understanding of the dynamics that give rise to stretched or compressed relaxation processes, including investigations of collective *versus* local dynamics and crossover from stretched

to compressed relaxations would require an expanded range of accessible structure factors $S(\mathbf{Q}, t)$, which will likely become available at diffraction-limited storage rings.

2.2. Two-time correlation function as a probe of non-equilibrium dynamics

The non-equilibrium ‘aging’ dynamics can also be probed directly by XPCS *via* the two-time correlation function $C(Q, t_1, t_2) = I(Q, t_1)I(Q, t_2)/\langle I(Q, t) \rangle^2$ (Fluerasu *et al.*, 2005; Sutton *et al.*, 2003; Malik *et al.*, 1998). For equilibrium fluctuations one can typically assume that the autocorrelation function depends only on the time lag $\tau = t_1 - t_2$ between the two states, but not on the actual time t_1 or t_2 ; hence the averaging performed over τ in equation (1). In non-equilibrium systems this assumption is no longer valid, and the relaxation time constants are generally a function of aging time, measured following a rapid quench or other thermodynamic parameter change. Additionally, intermittent dynamics (sudden spontaneous changes in local structural configurations) or temporally heterogeneous dynamics can also be revealed in analysis of two-time correlation functions, whereas this behavior generally remains ‘hidden’ in calculations that only involve $g_2(Q, t)$.

Summarizing, the standard analysis of XPCS data performed by calculating $g_2(t)$ is a special case providing information about time-average fluctuations. Analysis using two-time correlation functions can provide further details about the non-equilibrium nature, intermittency, temporal heterogeneities and time-resolved evolution of these fluctuations.

2.3. Higher-order correlation functions

In recent years much attention has been devoted to the issue of growing length scales accompanying the glass transition (Berthier *et al.*, 2005; Keys *et al.*, 2007) and growing dynamical heterogeneities (Trappe *et al.*, 2007; Duri & Cipelletti, 2006; Kawasaki *et al.*, 2007; Hartl, 2001), *i.e.* phenomena where the dynamics in nanoscale regions of cooperatively rearranging particles or molecules is orders of magnitude faster than in nearby regions of similar size. The heterogeneous dynamics have been observed in colloidal and granular systems undergoing a jamming transition (Weeks *et al.*, 2000, 2007; Berthier *et al.*, 2005; Mayer *et al.*, 2004; Keys *et al.*, 2007; Ballesta *et al.*, 2008; Dauchot *et al.*, 2005; Doliwa & Heuer, 1998; Gao & Kilfoil, 2007; Hartl, 2001) as well as in several molecular liquids undergoing a glass transition (Ediger, 2000; Glotzer, 2000). Recently it was demonstrated that in addition to the richness of the information obtained from XPCS and DLS measurements *via* the second-order autocorrelation function $g_2(Q, t)$, one could extract further highly valuable information from XPCS measurements. For example, intermittent dynamics (temporal heterogeneity) and spatial dynamical heterogeneity can be studied *via* the calculation of the dynamical susceptibility χ_4 , or fourth-order correlation function (Trappe *et al.*, 2007; Duri & Cipelletti, 2006; Duri *et al.*, 2005a,b; Cipelletti *et al.*, 2003b). XPCS can be

used to investigate whether the signatures of temporal heterogeneities and intermittent dynamics observed in soft glassy matter will also be found in electronic and magnetic systems; for example, recent microdiffraction measurements on spin- and charge-density wave domains in antiferromagnetic chromium show clear signatures of intermittent collective avalanche-like relaxation of the local distributions of the Q -vector values, pointing to metastability and non-equilibrium collective relaxation effects (Kim *et al.*, 2013).

The higher-order correlation function analysis of XPCS datasets could therefore produce important evidence of collective relaxation dynamics, which may provide additional information about the *dynamic* correlation length scales, similar to cooperatively rearranging regions in structural glasses. Higher-order correlation functions may reveal the relationship between dynamic correlations, *e.g.* the length and time scales over which systems may exhibit cooperative or collective dynamics, and the nature of the cross-over from individual local dynamics to collective fluctuations.

2.4. Driven domain dynamics and intermittency

XPCS also allows for detailed investigation of non-equilibrium driven dynamics, not only in soft matter systems but also in many hard condensed matter materials which are tunable with magnetic and electric fields, temperature and strain. For example, application of an electric field to a charge-density wave system (Monceau, 1985; Schlenker & Dumas, 1986; Thorne, 1996; Gruner, 1988) can often lead to ‘sliding’ of the charge-ordered condensate when the value of the applied field exceeds the threshold value defined by the pinning potential to the lattice defects (Fukuyama & Lee, 1978; Lee & Rice, 1979). The sliding dynamics are characterized by temporal coherence (Bhattacharya *et al.*, 1987, 1989), hysteresis and memory effects (Zettl & Gruner, 1982; Littlewood, 1986; Coppersmith & Littlewood, 1987), thermal and quantum creep regimes (McCarten *et al.*, 1991; Zaitsev-Zotov, 1993; Lemay *et al.*, 1999), long-range (micrometer-scale) periodic correlations (Bolloc’h *et al.*, 2008) and other intriguing effects. In strongly correlated oxides, *e.g.* in CMR manganites, the application of magnetic fields has been shown to produce intermittent avalanche-like dynamics (Hebert *et al.*, 2002; Mahendiran *et al.*, 2002; Woodward *et al.*, 2004; Voloshin *et al.*, 2007; Rana *et al.*, 2006; Rana & Malik, 2006; Alejandro *et al.*, 2006). In vanadites, in particular vanadium dioxide, the metal–insulator transition exhibits similar collective avalanche-like behavior, as a function of both the applied voltage and the temperature (Sharoni *et al.*, 2008). Similar avalanche-like intermittent dynamics have been observed in metals and alloys undergoing structural phase transitions (Mueller *et al.*, 2011; Sanborn *et al.*, 2011). The visibility of the X-ray speckle is directly related to the dynamical properties of the condensate or domain wall configuration; for example, rapidly moving phase defects in the sliding condensate would ‘smear out’ the speckle pattern and reduce the speckle visibility to zero (Sutton *et al.*, 2002). Therefore coherent X-ray scattering techniques based on speckle visibility could also be applied

to characterize the sliding of charge density waves recently observed in the manganite compound LCMO (Cox *et al.*, 2008), and in the near future to study the more elusive fluctuating order parameters, for example, charge stripes in high- T_c superconductors.

The speckle visibility approach is in fact a form of XPCS, since the observation of reduced speckle contrast due to a fluctuating order parameter is equivalent to a reduced correlation function in traditional sequential XPCS measurements. Even though in speckle visibility measurements one simply observes the reduction in speckle contrast as a function of time over which speckle is averaged, while calculations of autocorrelation functions in traditional XPCS requires recording of individual high-contrast speckle images and then calculating the decay of the correlations of intensities as a function of time delay between two such measurements, in systems with fluctuations that are governed by Gaussian statistics, the speckle visibility and autocorrelation function $g_2(Q, t)$ have a well defined mathematical relationship allowing equivalent information about dynamics in the sample to be obtained (Bandyopadhyay *et al.*, 2005). Even for non-Gaussian fluctuations, knowing the distribution, for example through higher-order moments of the autocorrelation function, there exists a well defined relationship between speckle visibility and $g_2(Q, t)$ (Bandyopadhyay *et al.*, 2005; Dixon & Durian, 2003).

The advantage of the speckle visibility approach is especially clear in situations where the dynamics are too fast to be able to capture individual speckle patterns, and instead the incoherent sum of several such speckle patterns is recorded, averaging speckle patterns (and therefore reducing speckle visibility) over time scales that are comparable with or even slower than the characteristic time scales on which the system evolves.

3. Outlook: XPCS at diffraction-limited storage rings

XPCS has so far been largely limited to relatively slow glassy relaxation processes, with characteristic time scales ranging from tens of milliseconds to minutes and hours. In part this is due to relatively low average coherent flux provided by the third-generation synchrotron sources, and in part due to limitations of area X-ray detector technology, which often results in relatively slow readout times, somewhat remedied by recent advances in detector technology and data compression schemes (Becker & Graafsma, 2012*a,b*; Hansen *et al.*, 2010; Hoshino *et al.*, 2012; Hromalik *et al.*, 2012; Johnson *et al.*, 2012; Lumma *et al.*, 2000; Madden *et al.*, 2010, 2011; Shinohara *et al.*, 2010; Westermeier *et al.*, 2009, 2013). The two-dimensional detector limitations can be circumvented by using a point (0D) detector. For strongly scattering systems, such as dynamics in liquid crystals and liquid surfaces, point detectors have yielded microsecond temporal resolution, a time scale that becomes comparable with the time structure of many synchrotron storage rings (typically 10–100 MHz repetition rate). This means that until now much of the interesting physics and materials science that occurs at sub-millisecond time scale

have remained out of reach of XPCS. This includes many of the fundamental questions in magnetism, with spin reorientation happening on the time scales of tens of picoseconds, or lattice dynamics, *i.e.* dynamics in ferroelectrics, structural phase transitions, *etc.*, with relevant time scales in the nanosecond or microsecond range. Much of the collective behavior, such as phase separations, coarsening, nucleation and growth, domain formation and domain fluctuations are expected to be slower than the relevant time scales measured over individual building blocks (spins, unit cells, *etc.*). Therefore, the crucial time scale range from 1 ps to 1 ms that is inaccessible by many scattering techniques (either time-domain probes, such as XPCS, or frequency-domain probes, such as inelastic X-ray scattering) represents much of the hidden physics. Revealing these nanoscale phenomena is likely to become possible at the next generation of synchrotron light sources.

Developments of diffraction-limited storage ring (DLSR) designs based primarily on the multi-bend achromat (MBA) lattice approach (Bei *et al.*, 2010; Borland, 2013; Cai *et al.*, 2012) have important implications for extending the temporal (and perhaps the wavevector) range that can be covered by XPCS. For example, preliminary upgrade plans at the Advanced Photon Source at Argonne National Laboratory in the USA and European Synchrotron Research Facility (ESRF) in Grenoble, France, the two synchrotron facilities that currently dominate as the originating location of most XPCS experiments, are both based on an MBA lattice and have projected a 100-fold increase in overall coherent X-ray flux over the current capabilities of these sources, depending on the X-ray energy and other characteristics of the experiments.

While many other X-ray techniques can benefit from such tremendous increase in coherent flux, the benefit is arguably among the strongest in the case of XPCS. The key figure of merit for XPCS, the shortest time scale of the fluctuations that can be accessed, scales with the square of the average coherent flux.

The signal-to-noise ratio R_{SN} for the autocorrelation function $g_2(Q, t)$ given in equation (1) in the low-count limit depends on the average coherent flux I_{COH} , the total accumulated time T , the shortest correlation time τ and the number of pixels $n_x \times n_z$ in the square detector array (Falus *et al.*, 2006*c*),

$$R_{SN} = I_{COH}(T\tau n_x n_z)^{1/2}. \quad (2)$$

As a result, an N -fold increase in average coherent flux I_{COH} enables a N^2 -fold decrease of τ for the same value of R_{SN} .

The origins of the scaling relationship expressed in equation (2) can be intuitively understood by considering that the probability of detecting a pair of photons in the same pixel within any given time window is the square of the probability of detecting a single photon in this pixel in the same time window, which scales with the coherent flux. In XPCS, the measurement relies on correlations between pairs of photons (separated in time but scattered into the same pixel), and the number of such pairs increases with the square of the coherent

flux and linearly with time, leading to the unusual scaling relationship for the signal-to-noise ratio shown in (2).

Therefore, a 100-fold increase in the average coherent flux at DLSRs will enable accessing time scales that are 10^4 times faster than currently possible, provided that future multipixel detector technology can accommodate these faster time scales. The exact time scales depend on the system and on experimental parameters such as X-ray energy, wavevector transfer Q , sample and scattering geometry. For a large subset of systems fluctuations can currently be measured at ~ 10 ms time scales. Access to 10^4 -fold shorter time scales then enables fluctuations to be measured in the ~ 1 μ s regime. These XPCS measurements would naturally require adequate detector technology with small pixels that can be read out at MHz rates. Additionally, since the correlation time scales approach the characteristic time structure of the synchrotron source (typically of the order of 10–100 MHz), one has to take special care to normalize for any differences in individual X-ray pulse intensities, since in this regime the source can no longer be considered quasi-continuous and uneven pulse intensities can produce varying intensity correlation functions that are X-ray source related and have nothing to do with the sample dynamics.

Considering that the current preliminary plans for DLSRs call for a repetition rate of approximately 10–100 MHz (corresponding to the temporal structure of the beam, defined by the duration between the bunches, of 10–100 ns), it is conceivable that for many experiments XPCS can achieve the temporal resolution defined by that time duration, corresponding to correlating the pairs of photons that come from two subsequent bunches. There are many examples of systems for which 1–10 ms dynamics can be currently measured routinely using XPCS at third-generation synchrotron sources, and for such systems DLSRs will present a unique opportunity of measuring fluctuations with time scales ranging from, say, 100 ns to seconds and hours, spanning ten orders of magnitude. For systems such as colloidal, molecular or metallic glasses, this wide time span would enable measuring both local dynamics (cage relaxations) that occur at fast time scales and glassy slow collective relaxations (so-called α and β relaxation modes) in a single measurement.

However, even faster temporal resolution is possible for XPCS, if one were to consider correlation between pairs of photons originating from the same pulse rather than from subsequent pulses. In the low-photon-counting limit, dominated by single-photon events, the autocorrelation function $g_2(t)$ is equivalent to the ratio of the total number of two-photon events in a specific pixel over a given time window and total average flux. By increasing the coherent flux, the total duration of the measurement decreases as the inverse square of the number of coherent photons per pulse, similar to the scaling described in equation (2). The most straightforward application of this approach, by statistical analysis of double-photon events from the same pulse, allows measurements of dynamics at a single time scale, defined by the duration of the pulse (currently of the order of 100 ps), which can then be combined with the sequential XPCS results at 10–300 ns time

scales and slower, obtained within the same measurement. If the duration of the X-ray pulse can be modified by either shortening or lengthening the electron bunch or the X-ray pulse itself, for example by using the split-and-delay line (Roseker *et al.*, 2009, 2011, 2012; Osaka *et al.*, 2013; Stetsko *et al.*, 2013), one could then expand the temporal window over which these correlations can be measured using such approach. Rather than obtaining a single $g_2(t)$ measurement for the time scale t defined by the duration of the pulse, varying that duration will enable measuring dynamics either at the sub-100 ps range (if the pulse duration can be shortened) or in the 100 ps to 10 ns window (using the split-and-delay approach). Dynamics in the 1 ps to 100 ps range could also be measured if a streak camera technology is developed that will allow multi-speckle (multi-pixel) detection of photons with readout approaching the repetition rate of the storage ring (Naylor *et al.*, 2001). The design of such a detector is a formidable task, which may be slightly simplified by the fact that only *pairs* of photons that arrive into the same pixel location from the same pulse need to be read out; most pulses will have zero such events for all pixels, and therefore will not require addressing or reading out any pixels. In case of the relatively low probability of obtaining a two-photon event in a single pixel (or row of pixels in the case of a streak camera), the readout rate of the detector is therefore relatively low and does not need to match the repetition rate of the source.

Note that for a fixed value of total coherent flux, or, equivalently, brightness (for a given X-ray energy), using the approach of correlating the pairs of photons arriving in the same pixel and originating from the same X-ray pulse will benefit from decreased repetition rate: (i) due to the increase in coherent photons per pulse, the probability of capturing a pair of photons within the same pixel and within the same pulse will scale as the square of the number of photons in the pulse, therefore resulting in a gain in total number of pairs of photons when averaged over many pulses, inversely proportional to the repetition rate; (ii) because of detector technology limitations, going towards lower repetition rates is likely to lead to a closer match between the pulse rate and the readout speed of the multi-pixel detector, *e.g.* using the streak camera. On the other hand, a decrease of the repetition rate will also increase the shortest time scale that can be measured by the traditional form of sequential XPCS since the shortest time is limited to correlation between pairs of photons arriving from two subsequent pulses.

4. Summary

XPCS has until now been largely limited to studies of slow glassy relaxation in condensed matter systems. However, the coherent flux available at DLSRs will result in a dramatic increase in the accessible range of time scales of XPCS, by at least four orders of magnitude. While there are several crucial obstacles to extending XPCS from the current fast limit of multi-pixel XPCS of a few milliseconds to tens of nanoseconds (sequential XPCS) and perhaps even sub-nanoseconds (correlations within the same pulse) at the DLSRs, it is

important to keep in mind that these obstacles are primarily technological rather than based on fundamental physics, and can probably be addressed over the next decade through concentrated research and development efforts in the area of ultrafast X-ray detectors, X-ray optics and accelerator physics. As can be seen from Fig. 2, achieving XPCS at, for example, the picosecond time scale means that the results can be directly compared with those obtained with inelastic neutron scattering, neutron spin-echo and soon even with inelastic X-ray scattering, which are techniques that operate in the frequency domain. The corresponding energy resolution in the meV range is achievable with state-of-the-art inelastic scattering spectrometers. Therefore, DLSRs offer the unique opportunity of bridging the experimental gap between temporal or energy resolution in $S(q, t)$ or $S(q, \omega)$ phase space maps. Filling this gap would enable future studies of the collective dynamics of spin, lattice and perhaps even charge degrees of freedom in many condensed matter systems over a hierarchy of temporal and spatial scales.

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