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A 250 Hz fast shutter with flexible sampling schemes for low-dose XPCS experiments at beamline P10 at PETRA III

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Abstract. Low-dose X-ray photon correlation spectroscopy experiments on biological samples can be used to investigate the structure formation and underlying dynamics during phase separation, denaturation, and gelation. However, the use of intensive X-ray beams as an investigative tool is limited due to the susceptibility of biological samples to radiation-induced changes. XPCS in particular requires recording a series of high-frame-rate images to capture fast dynamics. In such cases, a fast X-ray shutter system is required to minimize beam damage to the samples by allowing a flexible delay between two images. Here we show how a fast (250 Hz) X-ray shutter system, built by the University of Siegen in close collaboration with DESY, improves low-dose XPCS capabilities at the P10 beamline at PETRA III. We found that by employing this shutter system in a logarithmic sampling scheme, the absorbed X-ray dose on the samples could be significantly decreased.

1 Introduction

Radiation damage is a significant barrier in X-ray scattering investigations of dynamics in biological and soft matter systems via X-ray photon correlation spectroscopy (XPCS) [1–6]. Especially at synchrotron and X-ray free electron facilities providing highly intense X-ray radiation, sensitive protein samples can be irreversibly destroyed in a fraction of a second of X-ray exposure [2, 3]. Several methods such as cryocooling [7, 8], addition of cryoprotectants [2] or radical scavengers [9], and use of flowcells [10], are found to be effective to reduce the beam-induced damage in protein samples in macromolecular X-ray crystallography and small-angle X-ray scattering experiments. However, these methods can not be employed for all proteins or in all experimental configurations due to, (a) cooling to ultra-low temperatures can lead to changes in protein structures, (b) certain experiments require room temperature or high-temperature environments, (c) the radical scavengers or cryoprotectants can interact with proteins and change the dynamic properties of the system, etc. In particular, low-dose XPCS experiments require recording a series of high-frame-rate images to capture fast dynamics of biological samples at various physical conditions (like high temperature/pressure) with optimized signal-to-noise ratio for a given maximum tolerable dose [1, 11, 12]. This method has successfully been applied to probe structure formation and the underlying dynamics during phase separation, denaturation, and gelation [13–15]. However, to probe the fast dynamics in such systems at molecular length scales the samples need to be exposed to a relatively high photon flux to increase the scattering intensity (to increase the signal-to-noise ratio [16]) which would lead to radiation-induced effects.

For optimizing the signal-to-noise ratio (SNR) in low-dose XPCS experiments, a fast X-ray shutter system is needed which prevents beam exposure of the sample in between two images. Achieving an optimum in SNR requires the shutter to provide a highly flexible opening/sampling scheme in time as correlation functions usually decay on a logarithmic rather than a linear time scale. Here, we present the design, implementation, and testing of a fast (250 Hz) highly flexible in-vacuum X-ray shutter system which is now installed at the P10 beamline at PETRA III, DESY, Hamburg. This system consists of 20 piezo actuators (APA200M, CEDRAT TECHNOLOGIES) forming 10 'piezo-shutters', that can be individually controlled. Each piezo-shutter is capable of running with 10 Hz continuously and up to 25 Hz for limited periods allowing the system to reach frequencies of up to 250 Hz without any artefacts in the data collection. We tested the capabilities of the shutter by performing XPCS measurements on spherical silica nanoparticles where we observed a significant reduction in the absorbed X-ray dose by employing this shutter system in a logarithmic sampling scheme.

2 Shutter design

The main idea of the fast shutter concept is to control a series of piezo shutters so that each shutter module operates at a frequency of ≈ 10 Hz, but frequency multiplicity is achieved through the arrangement of the shutters in series. A schematic view of the fast shutter system is shown in Fig. 1. The system consists of 20 amplified piezo actuators (APA200M, CEDRAT TECHNOLOGIES) forming pairs of ten piezo-shutters, which are connected as depicted in Fig. 1a. The schematics of a single shutter perpendicular and parallel to the X-ray beam path are depicted in Fig. 1b and Fig. 1c, respectively. The open and closed position of the piezo actuators in a single shutter are depicted in Fig. 1d.

In addition, a 0.2 mm (v) x 0.2 mm (h) alignment hole allows translation of the shutter system easily out of the beam for beamline operation without using the fast shutter.

The shutter operation is controlled by an electronic control board designed and produced by the electrical workshop of the University of Siegen and housed in a vacuum vessel produced by the mechanical workshop also located at the University of Siegen. The shutter system is controlled by a TTL signal provided by the beamline control system. The TTL signal is passed to an SP75A, a bipolar (-20 / +150 V) switching power amplifier, and also serves as an input signal for the fast shutter controller. The high-voltage (HV) signal generated by the SP75A is also applied to the fast shutter controller and distributed to the outputs for controlling the 20 piezo actuators. Thus, the high voltage signal follows the TTL pulse (TTL signal down gives HV signal down; TTL signal up gives HV signal up). When the HV signal is applied, the piezo actuator retracts, and the piezo frame's height changes, causing the shutter to open.

3 Testing of the fast shutter operation

The fast shutter system is positioned in the X-ray beam path using a rotation and two translation stages (HUBER Diffraktionstechnik GmbH, Rimsting, Germany) as depicted in Fig. 2a. To test the fast shutter operation, we collected several time series of images with an EIGER X4M detector applying different opening (t_{exp}) and closing times (t_{sleep}) of the shutter. The increase (decrease) in transmitted intensity with t_{exp} (t_{sleep}) follows a linear trend, as expected (Fig. 2b). The deviation of data points at $t_{\mathrm{sleep}} < 0.004$ s from the linear behaviour indicates the maximum operation rate of 250 Hz of the shutter.

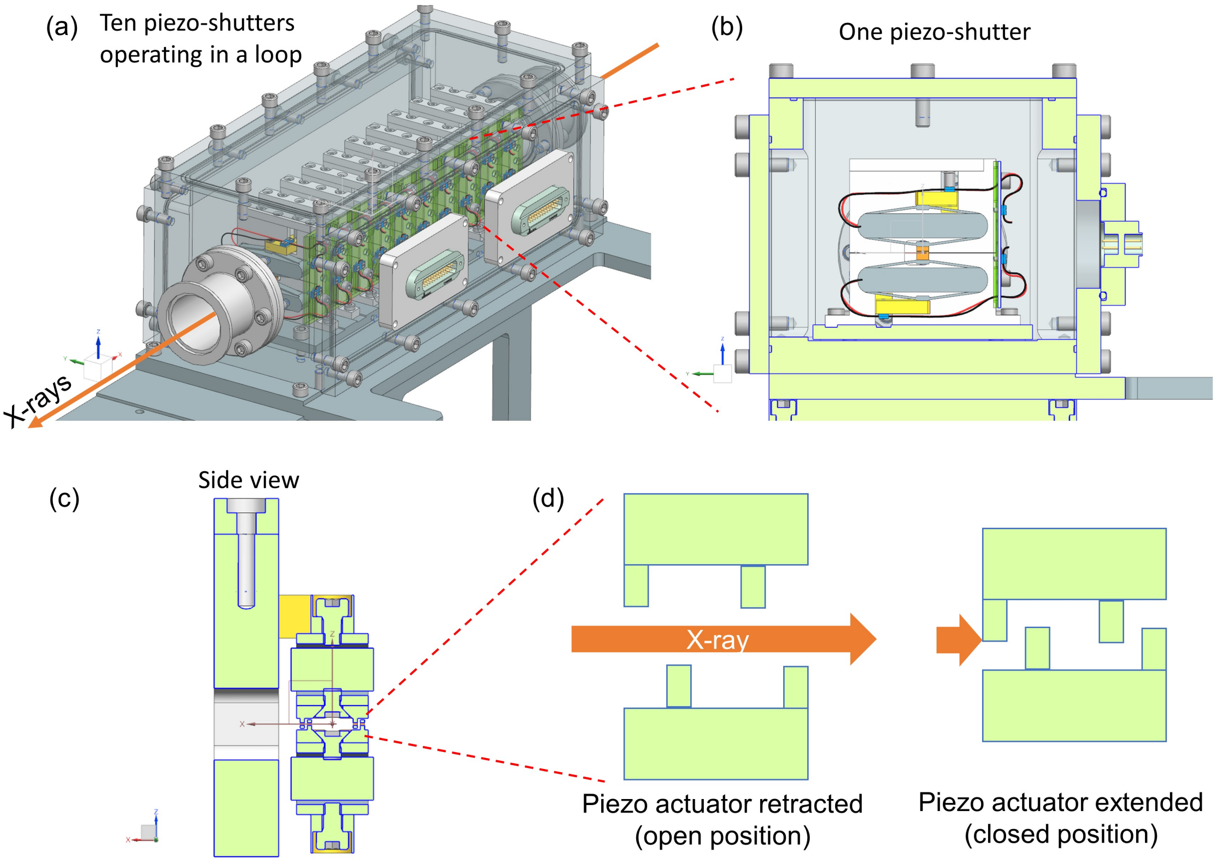

Figure 1: (a) computer model of the X-ray fast shutter system (the orange arrow indicates the direction of the X-ray beam). (b) Schematic of a single piezo shutter consisting of a pair of piezo actuators. (c) Cross-sectional side view (parallel to X-ray beam) of a single shutter. (d) Open and closed positions of the piezo actuators.

4 Testing of the fast shutter for XPCS experiments

We performed XPCS measurements of silica nanoparticles suspended in polypropylene glycol (PPG) with different shutter exposure times. t_{exp} and t_{sleep} are varied between 0.001–0.019 s, such that $t_{\text{exp}} + t_{\text{sleep}} = 0.02$ s. The intensity autocorrelation functions extracted from the XPCS data are given in Fig. 3a. The solid lines in Fig. 3a indicate the model fits using,

$$g_2(q,t) = 1 + \beta \mid \exp[-(t/\tau)^{\gamma}] \mid^2, \tag{1}$$

where τ, β, and γ are the relaxation time, speckle contrast, and Kohlrausch-Williams-Watts (KWW) exponent [17], respectively. The extracted relaxation times are shown in Fig. 3b. At all shutter speeds and exposure times, about the same relaxation time is observed, demonstrating that the fast shutter is operating as intended.

Next, we explored the capability of the shutter to sample correlation functions in a non-linear mode. For this, we applied a linear and a logarithmic data collection mode as depicted in Fig. 4. We obtain almost the same g_2 functions from these two data collection modes as depicted in Fig. 4. The dose, D, absorbed by the sample in both cases is estimated using the relation [1],

$$D = \frac{\Phi E A_{\text{cof}}}{d\, b^2\, \rho}, \tag{2}$$

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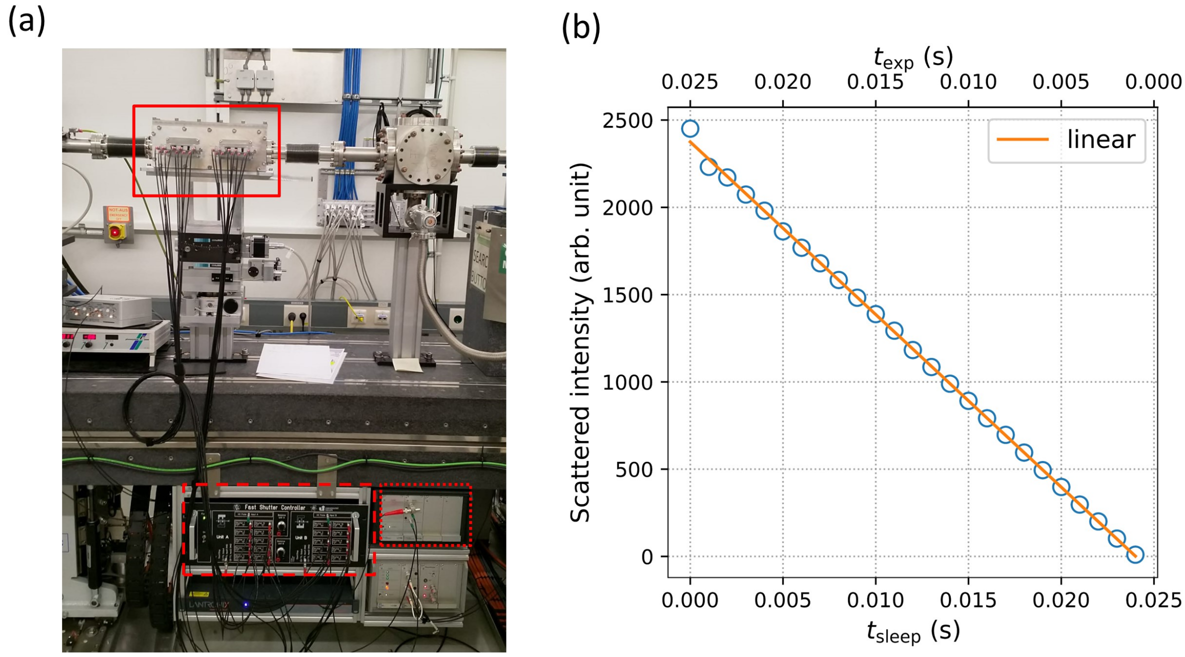

Figure 2: (a) Photo showing the fast shutter system positioned at the P10 beamline, PETRA III. Solid red rectangle: the vacuum vessel of the fast shutter, dashed red rectangle: the shutter control unit, and dotted red rectangle: the high voltage supply for the fast shutter. (b) The transmitted intensity collected on the detector at different opening (t_{exp}) or closure times (t_{sleep}). The orange curve indicates a linear fit to the intensity as a function of opening time.

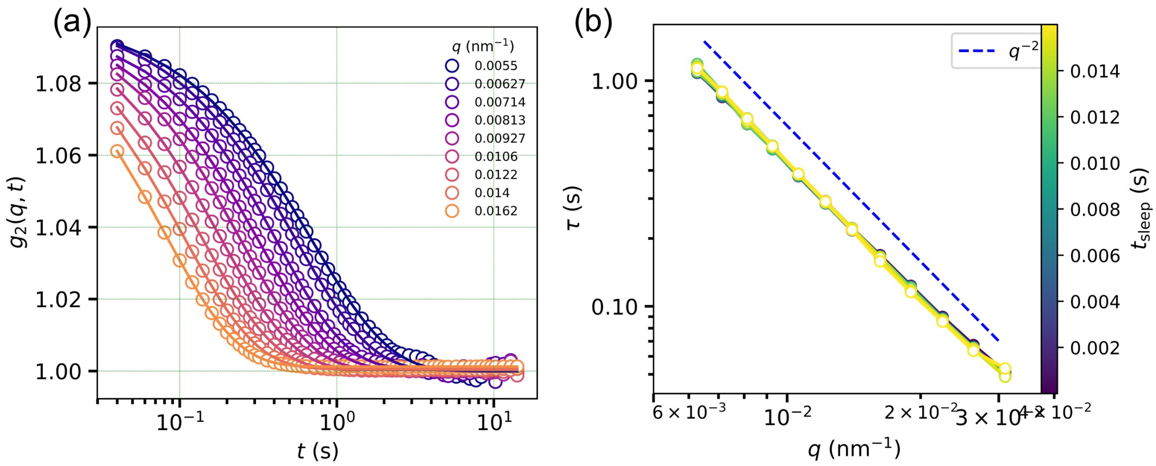

Figure 3: (a) Wave-vector dependent g_2 functions of silica nanoparticles in PPG collected when the fast shutter is used to add sleeping times between exposures. The $t_{exp} = 0.02$ s and $t_{sleep} = 0$ s, were used to collect g_2 functions given in (a). The solid lines indicate fits using Eq. 1, and the extracted τ at different t_{sleep} are shown in (b). The color bar indicates the t_{sleep}.

where Φ, E, A_{cof}, d, b, and ρ are the incident flux, the energy of the incident X-rays (8.54 keV), the X-ray attenuation coefficient of the sample (0.57 for propylene glycol), the thickness of the sample capillary (1.5 mm), the beam size (100 µm), and the density of silica (2.65 g/ml) respectively. The estimated absorbed dose for the linear and logarithmic cases are 5 kGy and 0.84 kGy, respectively. More systematic studies are needed to estimate the SNR [11, 12] of different sampling schemes to establish operational validity and broader applicability of the shutter. However, these first tests already demonstrate the capability of our shutter system to record correlation functions with a highly flexible sampling scheme.

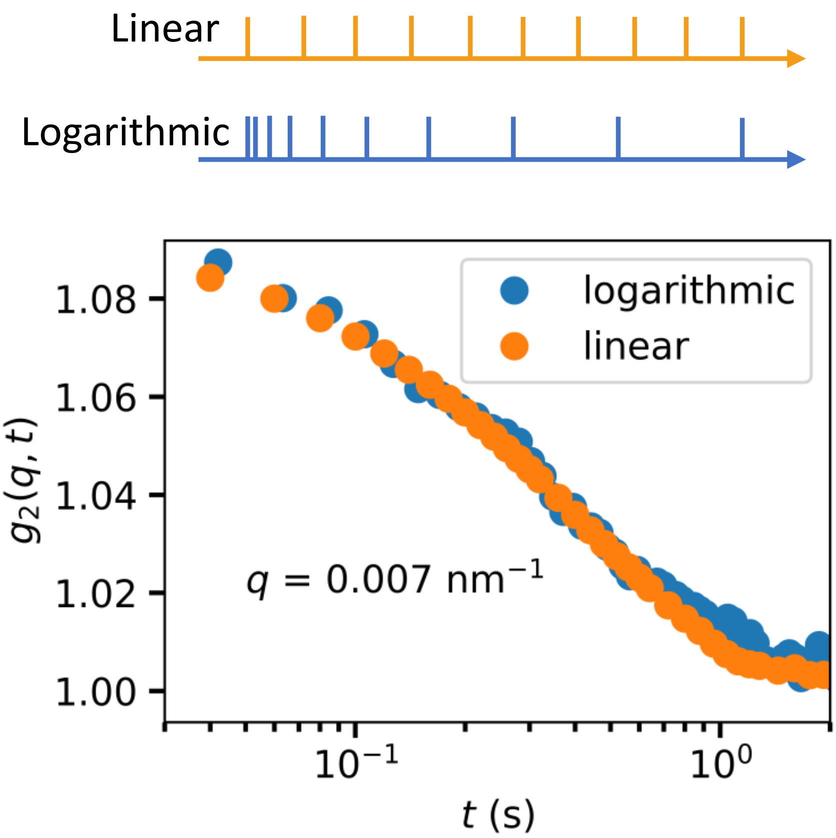

Figure 4: The top schematic illustrates the data collection in linear and logarithmic sampling modes. The vertical lines indicate the time point of a scattering image. Comparison of g_2 functions (bottom plot) of silica nanoparticles in PPG collected when the fast-shutter operated in a linear and logarithmic sampling scheme.

5 Summary

In summary, we report the design, implementation, and test of a 250 Hz fast shutter system installed at the P10 beamline, PETRA III, for enabling XPCS at low dose values. We established that the X-ray shutter is capable of sampling autocorrelation functions in linear and non-linear sampling schemes. This holds the potential to enhance the SNR in XPCS experiments while maintaining a tolerable dose level on the sample. However, future investigations are required to systematically estimate the SNR of different sampling schemes. Nonetheless, initial tests already showcase the ability of our shutter system to capture correlation functions with highly adaptable sampling schemes at low dose values.

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References

[1] Johannes Möller, Michael Sprung, Anders Madsen, and Christian Gutt. X-ray photon correlation spectroscopy of protein dynamics at nearly diffraction-limited storage rings. *International Union of Crystallography Journal*, 6(5):794–803, 2019.

[2] Shigeo Kuwamoto, Shuji Akiyama, and Tetsuro Fujisawa. Radiation damage to a protein solution, detected by synchrotron X-ray small-angle scattering: dose-related considerations and suppression by cryoprotectants. *Journal of Synchrotron Radiation*, 11(6):462–468, 2004.

[3] Mario Reiser, Anita Girelli, Anastasia Ragulskaya, Sudipta Das, Sharon Berkowicz, Maddalena Bin, Marjorie Ladd-Parada, Mariia Filianina, Hanna-Friederike Poggemann, Nafisa Begam, et al. Resolving molecular diffusion and aggregation of antibody proteins with megahertz X-ray free-electron laser pulses. *Nature Communications*, 13(1):5528, 2022.

[4] Giovanna Pintori, Giacomo Baldi, Beatrice Ruta, and Giulio Monaco. Relaxation dynamics induced in glasses by absorption of hard X-ray photons. *Physical Review B*, 99(22):224206, 2019.

[5] Yuriy Chushkin, Alessandro Gulotta, Felix Roosen-Runge, Antara Pal, Anna Stradner, and Peter Schurtenberger. Probing cage relaxation in concentrated protein solutions by X-ray photon correlation spectroscopy. *Physical Review Letters*, 129(23):238001, 2022.

[6] Sonja Timmermann, Nimmi Das Anthuparambil, Anita Girelli, Nafisa Begam, Marvin Kowalski, Sebastian Retzbach, Maximilian Darius Senft, Mohammad Sayed Akhundzadeh, Hanna-Friederike Poggemann, Marc Moron, et al. X-ray driven and intrinsic dynamics in protein gels. *Scientific Reports*, 13(1):11048, 2023.

[7] D. J. Haas. X-ray studies on lysozyme crystals at –50°C. *Acta Crystallographica Section B*, 24(4): 604, 1968.

[8] David J Haas. The early history of cryo-cooling for macromolecular crystallography. *IUCrJ*, 7(2): 148–157, 2020.

[9] Alexander Grishaev. Sample preparation, data collection, and preliminary data analysis in biomolecular solution X-ray scattering. *Current Protocols in Protein Science*, 70(1):17–14, 2012.

[10] Anne Martel, Ping Liu, Thomas M Weiss, Marc Niebuhr, and Hiro Tsuruta. An integrated high-throughput data acquisition system for biological solution X-ray scattering studies. *Journal of Synchrotron Radiation*, 19(3):431–434, 2012.

[11] Jan Verwohlt, Mario Reiser, Lisa Randolph, Aleksandar Matic, Luis Aguilera Medina, Anders Madsen, Michael Sprung, Alexey Zozulya, and Christian Gutt. Low dose X-ray speckle visibility spectroscopy reveals nanoscale dynamics in radiation sensitive ionic liquids. *Physical Review Letters*, 120 (16):168001, 2018.

[12] D Lumma, LB Lurio, SGJ Mochrie, and Mark Sutton. Area detector based photon correlation in the regime of short data batches: Data reduction for dynamic X-ray scattering. *Review of Scientific Instruments*, 71(9):3274–3289, 2000.

[13] Nimmi Das Anthuparambil, Anita Girelli, Sonja Timmermann, Marvin Kowalski, Mohammad Sayed Akhundzadeh, Sebastian Retzbach, Maximilian D Senft, Michelle Dargasz, Dennis Gutmüller, Anusha Hiremath, et al. Exploring non-equilibrium processes and spatio-temporal scaling laws in heated egg yolk using coherent X-rays. *Nature Communications*, 14(1):5580, 2023.

[14] Nafisa Begam, Anastasia Ragulskaya, Anita Girelli, Hendrik Rahmann, Sivasurender Chandran, Fabian Westermeier, Mario Reiser, Michael Sprung, Fajun Zhang, Christian Gutt, et al. Kinetics of network formation and heterogeneous dynamics of an egg white gel revealed by coherent X-ray scattering. *Physical Review Letters*, 126(9):098001, 2021.

[15] Anita Girelli, Hendrik Rahmann, Nafisa Begam, Anastasia Ragulskaya, Mario Reiser, Sivasurender Chandran, Fabian Westermeier, Michael Sprung, Fajun Zhang, Christian Gutt, et al. Microscopic dynamics of liquid-liquid phase separation and domain coarsening in a protein solution revealed by X-ray photon correlation spectroscopy. *Physical Review Letters*, 126(13):138004, 2021.

[16] Fivos Perakis and Christian Gutt. Towards molecular movies with X-ray photon correlation spectroscopy. *Physical Chemistry Chemical Physics*, 22(35):19443–19453, 2020.

[17] Graham Williams and David C Watts. Non-symmetrical dielectric relaxation behaviour arising from a simple empirical decay function. *Transactions of the Faraday Society*, 66:80–85, 1970.