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# Restoring size of detonation nanodiamonds from small-angle x-ray scattering of polychromatic synchrotron radiation beam

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**Abstract.** Over the past two decades, LIH SB RAS and BINP SB RAS have jointly conducted experiments on time-resolved (TR) small-angle x-ray scattering (SAXS) with detonating high explosives. The purpose of these experiments is to restore the dynamics of carbon species condensation to diamond nanoparticles by analyzing series of SAXS patterns behind the detonation front measured in real time with fast detectors. This knowledge is crucial for the development of reliable detonation models. In this paper, we compare SAXS patterns of identical nanodiamond samples measured at the TR-SAXS extreme state of matter end-station (BINP SB RAS) in the static mode under realistic conditions simulating fast real-time measurements with polychromatic SR beam and traditional SAXS BioMUR beamline at the Kurchatov Synchrotron Radiation Source (NRC “Kurchatov Institute”) with monochromatic synchrotron radiation (SR) beam. These experiments confirm that the size of scattering inhomogeneities determined in dynamic experiments with single bunch exposure with polychromatic SR beam is correct.

## 1. Introduction

First experiments on the dynamic registration of small-angle x-ray scattering (SAXS) were made in the early 2000s. They showed a long signal intensity growth during the detonation of a 50/50 mixture of trinitrotoluene and hexogen [1].

A SAXS pattern can provide information about fluctuations of mean density inside a sample under investigation. In earlier time resolved small angle x-ray scattering (TR-SAXS) studies of explosion processes [1–3], it was shown that the integral SAXS signal intensity increases with increasing negative oxygen balance. Apparently, these conditions favor the formation of nanodiamonds since it is the carbon nanoparticles condensed from explosion gas products that play a role of density fluctuations in these experiments.

The ability to use short high-periodical synchrotron radiation (SR) pulses from a high-energy accelerator allowed us to reveal the SAXS signal time evolution during and immediately after the explosive detonation. This data can be converted into the average particle size of the condensed carbon as a function of time delay after the explosion initiation [4]. Presently, SAXS represents



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a very rare technique capable of direct real-time experimental measurement of the dynamics of the size of condensed carbon nanoparticles during detonation of high explosives (HEs) with required temporal resolution [5–7].

Similar SAXS experiments were carried out at Advanced Photon Source (Argonne National Laboratory, USA). In first studies they have shown that hexanitrostilbene detonation produces carbon particles with a radius of gyration of 2.7 nm, that was recorded in 400 ns and remains constant for several microseconds [7]. Further studies of larger charges (up to 2 g) [8–10] showed that the shape of SAXS patterns change and the carbon nanoparticles grows at least during the first 300 ns behind the detonation front. General description of their station and scheme of synchronization for studying time resolved SAXS at the APS is presented in [11], the size of particles was recovered in the Guinier approximation. For their experiments they used monochromatic SR beam from undulator (with energy about 23.6 keV) and scintillator with 4 charge-coupled device cameras for registration.

In this paper, we demonstrate the feasibility of determining the mean size of nanodiamonds using TR-SAXS measured for single-bunch duration with polychromatic radiation at the VEPP-4M storage ring (BINP SB RAS, Novosibirsk).

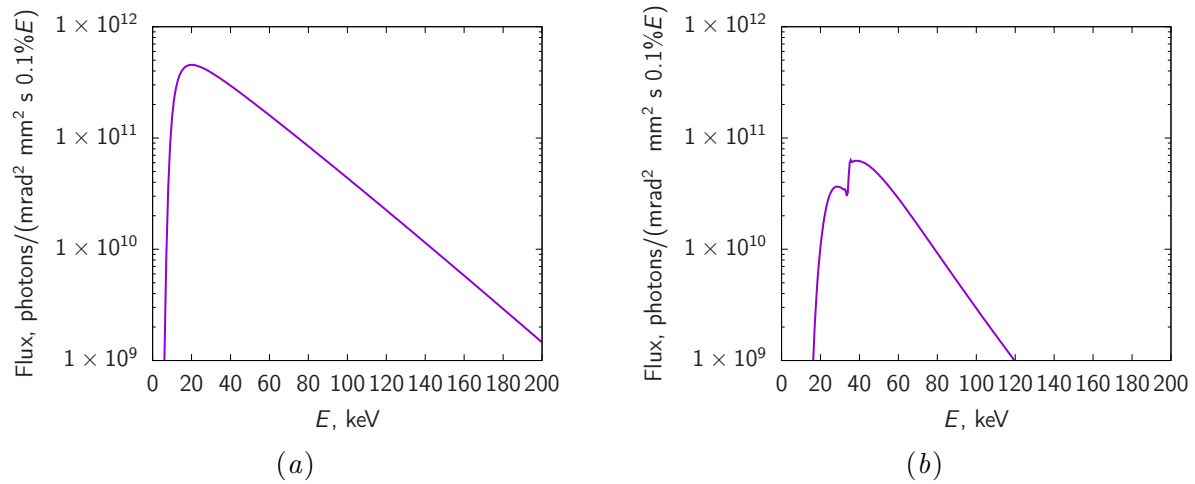
The role of carbon species condensation during the detonation of explosives under conditions of a negative oxygen balance and the possibility of condensation beyond the Chapman–Jouguet plane remain the central problems in the detonation transient kinetics. Tarver et al. showed [12] that an assumption that the carbon condensation goes on beyond the Chapman–Jouguet plane provides a more accurate description of the experimental data. Unfortunately, experimental verification of the assumption is extremely challenging since the required information can be gained only with TR-SAXS realized at a high-energy accelerator operating in a special storage mode filled with a train of bunches to generate a sequence of closely spaced intense pulses.

## 2. Extreme state of matter end-station

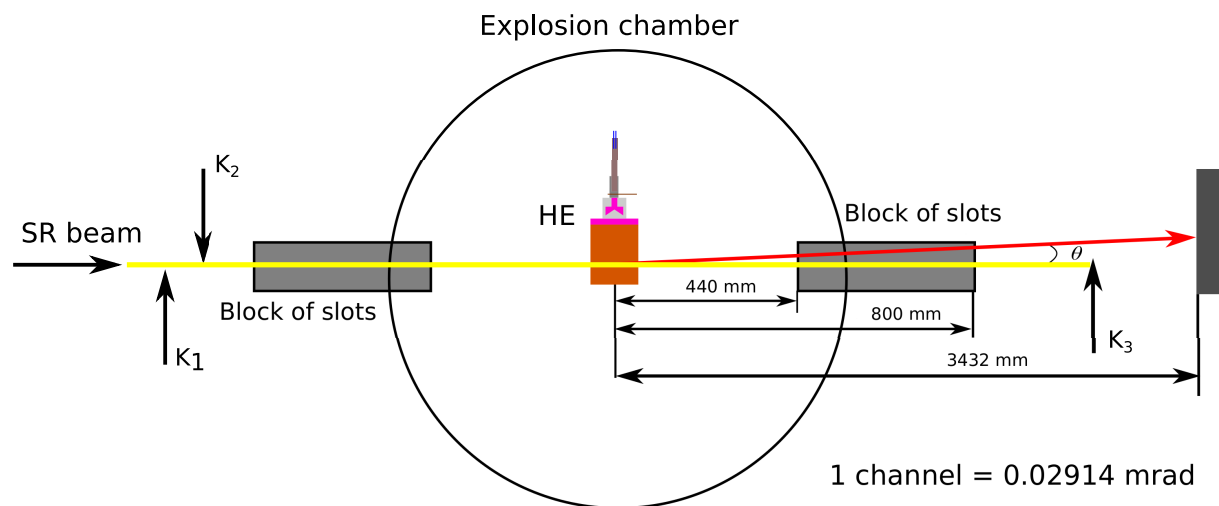
The end-station for the extreme state of matter (previously called as SYRAFEEMA end-station) at the shared-use center “Siberian Synchrotron and Terahertz Radiation Center (SSTRC)” based on VEPP-4M storage ring is a dedicated time-resolved SAXS and radiography instrument. To achieve a time resolution of 73 ps corresponding to a single bunch duration the instrument should use the real spectrum that is a convolution of three factors, viz, incident wiggler spectrum, sample transmission, and detector efficiency as a function of photon energy, rather than strictly monochromatic radiation as adopted by the majority of SAXS beamlines at SR sources worldwide. The apparent real spectrum was calculated according to the earlier described procedure [13, 14].

Typically, for the TR-SAXS measurements, the storage ring operated in a special two-bunch mode with a current of 10 mA and a period of 600 ns. The SR pulse duration was 73 ps [15]. The spectral characteristics of the radiation from the 9-pole 2 T wiggler are shown in figure 1(a). The explosion chamber had a beryllium entrance window 2 mm thick and a polycarbonate exit window 5 mm thick. The explosion chamber was not evacuated during the experiments, although that was feasible. The spectral characteristics of the x-ray beam at the extreme state of matter end-station are shown in figure 1(b).

The general scheme of the end-station is shown in figure 2. The SR beam is formed by two blades  $K_1$  and  $K_2$  (the so-called Kratky collimator) installed ahead of the explosion chamber. The direct beam was stopped by a knife  $K_3$  ahead of the detector. The scattered x-rays were detected by a detector DIMEX-3 [16] with an angular resolution of  $3 \times 10^{-5}$  rad and time resolution limited by the single bunch duration. The distance from the axis of the explosive charge to the detector was 3432 mm. The pitch between the strips of the detector DIMEX was 0.1 mm. Thus, a single channel of the detector DIMEX in these experiments on VEPP-4M corresponded to the following scattering angle  $2\theta$ : 1 detector channel = 0.029 14 mrad.



**Figure 1.** (a) Spectrum of the SR from the VEPP-4 wiggler. (b) Real radiation spectrum at the extreme state of matter end-station ( $E$  is the spectrum energy).



**Figure 2.** Scheme of experiments on SAXS measuring:  $K_1$  and  $K_2$ —blades forming  $5 \times 0.5 \text{ mm}^2$  SR beam;  $K_3$ —blade stopping direct SR beam.

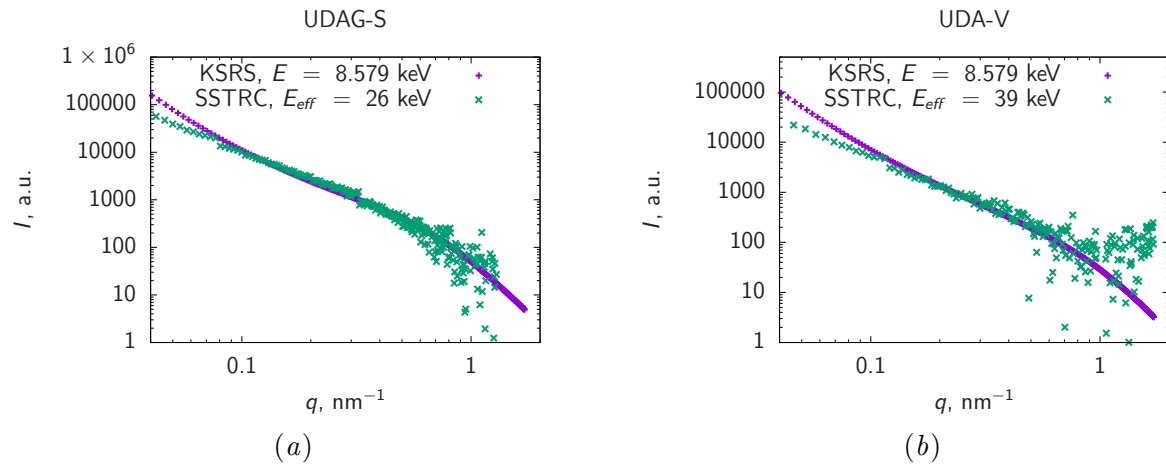
The extremum sizes of nanoparticles achievable at such a setup are defined by the following formulas:

$$D_{\min} = \pi/q_{\max} = \lambda/4 \sin(\theta_{\max}) \approx 2 \text{ nm},$$

$$D_{\max} = \pi/q_{\min} = \lambda/4 \sin(\theta_{\min}) \approx 100 \text{ nm}.$$

Here the scattering vector  $q = 4\pi \sin(\theta)/\lambda$ , where  $2\theta$  is the scattering angle.

In our experiments, the minimum registered angle corresponded to the 5-th channel:  $2\theta_{\min} = 0.15 \text{ mrad}$  (1 channel =  $0.02914 \text{ mrad}$ ); the maximum angle corresponded to the 150-th channel:  $2\theta_{\max} = 4.4 \text{ mrad}$ .



**Figure 3.** Measured SAXS patterns for (a) UDAG-S and (b) UDA-V samples:  $E$  is the energy (monochromatic) used in KRSR;  $E_{\text{eff}}$  is the effective energy calculated from the spectrum used in SSTRC, taking into account the absorption of the sample.

### 3. Experiments and their results

Although the feasibility of using polychromatic x-rays for SAXS measurements in the Guinier scheme and successful analysis to restore the size distribution of density inhomogeneities was considered theoretically [13, 14], we herewith verify this feasibility by a direct comparison of SAXS experimental patterns of identical samples measured using the TR-SAXS instrument described above and a traditional SAXS as implemented at the BioMUR beamline of the Kurchatov Synchrotron Radiation Source (KRSR) [17, 18].

Two samples with codes UDAG-S (raw explosion products containing both nanodiamonds and graphite) and UDA-V (aqueous suspension of purified detonation nanodiamond) were provided by the Federal Research and Production Center “Altai” (Biysk).

The samples for measurements at the BioMUR beamline were loaded into thin-walled glass capillaries. The evacuated sample-to-detector path was 2500 mm and the wavelength was  $\lambda = 1.445 \text{ \AA}$ , which made it possible to collect reliable SAXS data over the  $q$  range of  $0.04\text{--}2 \text{ nm}^{-1}$  for both samples (figure 4). The integration time was about 2 min.

The signal was recorded using a two-dimensional hybrid-pixel detector PILATUS 1M (Dectris, Switzerland). The 2D SAXS pattern was averaged over the azimuthal directions using the FIT2D program [19]. The PRIMUS program [20] was applied to subtract the background contribution of blank capillary. The particle size was estimated using the indirect Fourier transformation algorithm as implemented in the GNOM program [21].

For measurements at the extreme state of matter end-station, the samples were loaded into polypropylene vials with a diameter of 40 mm. The absorption of x-rays in the samples was taken into account in calculation of the “real” spectrum. Scattering angles were converted into scattering vectors by introducing an effective photon energy corresponding to the best approximation of monochromatic scattering, as described in detail elsewhere [13]. The SAXS patterns measured with two different aforementioned instruments are compared in figure 4. The measured curves are essentially identical.

### 4. Conclusions

Herewith, we compared SAXS patterns of two detonation nanodiamond samples measured using a dedicated TR-SAXS instrument with a polychromatic beam at the SSTRC and a traditional

one with the monochromatic beam at the KSRS and demonstrated their virtual equivalency. This justifies the applicability of the extreme state of matter end-station for real-time explosion dynamics studies. The mean size of scattering centers in the two samples was 15 nm (according to the Guinier method) for the SAXS patterns measured at the SSTRC and at the KSRS.

Thus, our experiments have demonstrated the possibility of determining the size of scattering inhomogeneities in dynamic experiments with single bunch exposure on polychromatic SR beam.

### Acknowledgments

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