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Small angle neutron scattering at very high time resolution: Principle and simulations of 'TISANE'

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Abstract

The time resolution of SANS experiments is generally limited by frame overlap to some ms. We report on a new time-resolved stroboscopic SANS method, called TISANE, offering μs time resolution without a major sacrifice in intensity by making use of very large frame overlap. We may explore a new field in neutron scattering and complement the emerging field of time resolved small angle X-ray scattering. Here we discuss the principle of TISANE, its mathematical treatment and its limitations.

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1. Introduction

In this Letter we discuss a new technique, TISANE (Time dependent SANS Experiments), which allows time resolutions between ms and about 10 μs in small angle neutron scattering (SANS)—an improvement of two orders of magnitude. The technique avoids a dramatic drop in intensity; compared to static experiments only an order of magnitude is lost. The idea has been presented by the author and by co-workers since about 7 years at various conferences; however no attractive application had been in sight. The paper from Wiedenmann et al. [1] who, to our knowledge, applied the idea for the first time triggered this publication. TISANE should complement the emerging field of high time resolved small angle X-ray scattering (SAXS), which was opened by expanding the time domain far beyond the ms range. We cite some recent publications in this field:

- SAXS with time resolution in the 100 μs range allows tracing the dynamics of protein folding in rapid mixing devices [2,3].
- SAXS on shear flow observes transient flow phenomena [4], and the authors' statement '*the comparatively long exposure times necessary in neutron scattering limit its applicability to steady state flows*' should not hold anymore in case TISANE gets realized.
- Time resolved SAXS reveals micellar transformations within time constants of less than 10 ms [5].
- Precipitation processes in a liquid jet were observed in SAXS experiments at about 20 μs time resolution [6].
- Further examples were presented at a workshop on time resolved investigations with X-rays and neutrons [7].
- A comprehensive discussion on time resolved SAXS experiments with biomembranes and proteins is given in [8].

Due to the high intensity, SAXS experiments, in general, can directly follow transients in time. In contrary, TISANE is a Fourier technique achieving high time resolution even in the case of very low flux characteristic for neutron sources. How-

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ever, in this case, only periodic processes can be measured by the SANS-response of the sample as a function of the frequency and amplitude of applied fields or of modulated conditions (e.g. pressure or flow). This excludes experiments on singular events, e.g. on non-reversible fast reactions. TISANE, however, allows experiments with high contrast (H-D exchange) in realistic environments, often necessary to see subtle effects in biology. Applications for TISANE might certainly arise in the following fields:

- A proof of principle experiment with TISANE has recently been performed on magnetic nano-particles, showing different ordering processes for different frequency domains, extending into the kHz range [1].
- With polarized TISANE, the switching of magnetic domains in bulk can be visualized from the kHz close to the MHz range.
- The response of micelles or membrane proteins to external fields in various configurations and environments can be extended to the sub-ms range. Conformational changes of bacteriorhodopsin due to light absorption might become visible well below the ms limit [9].
- The study of dynamics of flux lines in superconductors can be far extended in time resolution [10].
- Combining TISANE with REFSANS (SANS + reflectometry), should allow experiments on the dynamics of magnetic domains near to 100 kHz over large correlation lengths on surfaces layers.

2. Principle of TISANE

In typical time dependent SANS experiments, the resolution Δt is limited by frame overlap. For a sample-detector distance, L, of typically 20 m and a neutron wavelength band of 10% at 10 Å, only experiments above $\Delta t \approx 5$ ms are usually feasible. Higher time resolution is possible with spin-echo based SANS techniques [11]; however its applications are very restricted (strong scatterers, no magnetic fields). In TISANE, the severe frame overlap is no longer a hindrance but actually enhances the intensity without sacrificing time resolution.

In the following, we outline the principle of TISANE, discuss a mathematical model, present computer simulations for a primitive model system and show the limitations of TISANE.

The TISANE principle can be deduced from the space–time diagram shown in Fig. 1, where we employ a cold neutron beam of mean wavelength λ, prepared by a velocity selector ($\Delta\lambda/\lambda \approx 10\%$). TISANE requires the addition of one fast chopper near the beginning of the collimator of the SANS instrument. In addition, TISANE requires a device, which modulates the scattering properties of the sample, and finally a detector with high time resolution. In Fig. 1, the lines represent the neutron trajectories of different wavelengths from successive chopper openings. By the time the neutrons reach the sample position, there is extreme frame overlap. In the ideal TISANE case, the intensity at the sample is constant and so the beam modulation due to the chopper is no longer visible at this position.

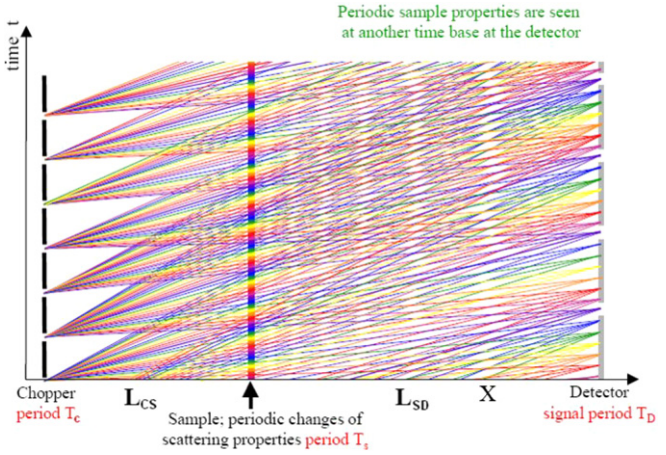

Fig. 1. Distance-time diagram for the TISANE setup at a SANS instrument. A broad wavelength band passes a chopper with repetition time T_C. The scattering properties of the sample are modulated with time period T_S, represented by the periodic change in colours. Neutron trajectories are coloured in the same way upon passing the sample. Shortly after the sample, the colours mix, but at a certain distance L_{SD}, where we place the detector, the colours re-order. In this way the SANS time response of the sample's scattering properties may be imaged at the detector. Note, we assume negligible inelastic scattering. For visualizing the principle of overlap, parameters pretty far off from reality had been used.[1] (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this Letter.)

The time modulation of the sample's scattering properties is illustrated by a sequence of rainbow colours (e.g. the distance from one 'blue' region of the sample to the consecutive 'blue' corresponds to the time period of the sample modulation) and the colours of the neutron trajectories are chosen to match the colour of the sample modulation at the instant of time when the neutron passes the sample. We can see that in a certain distance downstream of the sample all the colours reorder themselves, where we have naturally chosen to place our detector. In effect, each individual colour represents a physical observation of the sample in a specific configuration during one full time period. Recording the pattern at the detector as function of time (say 10–20 detector frames within one cycle) reveals the response function of the sample to the external field. Note that in general the frequency of the detector signal differs from the sample modulation frequency and the chopper frequency. The re-ordering at the detector is not affected by the angular spread of the neutron trajectories in the space–time diagram of Fig. 1, i.e. TISANE works for a very broad wavelength band.

The distance at which these colours reorder themselves can be easily deduced from geometry in Fig. 1:

$$\frac{T_S}{T_C} = \frac{L_{SD}}{L_{CD}}, \tag{1}$$

$$\frac{T_D}{T_S} = \frac{L_{CD}}{L_{CS}}, \tag{2}$$

[1] A typical time for neutron propagation from the chopper to the detector is about 100 times longer than the chopper repetition time and in that case appreciable overlap of trajectories occurs already at much narrower velocity bands than used in the drawing.