

X-ray cross-correlation analysis and local symmetries of disordered systems

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The microscopic understanding of the structure of matter is of fundamental importance for materials design. Complexity in the structure is often associated with a sophisticated functionality of materials. Thus, study of the structural properties of complex matter is a challenging task for materials science on the way to development of high performance materials. Significant progress in theoretical studies of magnetic, electronic and other properties of the crystalline matter has been achieved due to a powerful and relatively simple description of a crystalline structure. Opposite to crystals, the concepts used for description of microscopic structural properties of disordered materials such as liquids and glasses did not go far beyond the pair distribution function [1-3]. This is an unfortunate gap considering that disordered matter may exhibit novel complex functional properties.

Availability of the state-of-the-art X-ray sources enable fascinating possibilities to access the microscopic structure of disordered matter. One of the potential approaches for the analysis of diffraction data from disordered materials is based on the application of the x-ray cross-correlation functions (CCFs) [4, 5], that in the simplest form can be defined as [4]

$$C_q(\Delta) = \frac{\langle I(q, \varphi)I(q, \varphi + \Delta) \rangle_\varphi - \langle I(q, \varphi) \rangle_\varphi^2}{\langle I(q, \varphi) \rangle_\varphi^2}. \quad (1)$$

Here $\langle \dots \rangle_\varphi$ is the angular average of the scattered intensity $I(q, \varphi)$ at different values of the momentum transfer $q = |\mathbf{q}_1| = |\mathbf{q}_2|$ [see Fig.1(a)]. It was demonstrated in the experimental study [4], that CCFs (1) can provide information on the local arrangements of polymethylmethacrylate (PMMA) spheres in a colloidal glass.

In our recent paper [5] we provide a theoretical basis for a more general class of CCFs defined at two different magnitudes of the momentum transfer vectors $q_1 \neq q_2$. We also propose to analyse the Fourier components C_q^n of the CCF

$$C_q(\Delta) = \sum_{n=-\infty}^{\infty} C_q^n \exp(in\Delta), \quad C_q^n = \frac{1}{2\pi} \int_0^{2\pi} C_q(\Delta) \exp(-in\Delta) d\Delta, \quad (2)$$

as a convenient way to determine the contribution to the CCFs defined by the local structure of a disordered system. According to our analysis, for a dilute disordered system only the Fourier components with specific n values, which are related to the local structure of the system, have nonzero contribution. As an illustration [6], in Fig. 2 the results of the cross-correlation analysis of a diffraction pattern modelled for a 2D disordered sample [see Fig. 1(b)], composed of 11 randomly positioned and oriented pentagonal PMMA clusters, is presented. Fourier spectra in Fig.2(b) indicate the dominating Fourier components C_q^n , with $n=10, 20$. These values of n can be defined as $n=5*(2*l)$, where l is an integer number, the coefficient 5 is defined by the 5-fold rotational symmetry of a pentagonal cluster, and factor 2 appears due to the fact that the diffraction pattern is centrosymmetric in this 2D case. Therefore, calculated CCFs provide an access to the local symmetry in the disordered system.

In our study [5], we also perform a detailed analysis of CCFs dependence on the number of particles (molecules, clusters) in a disordered system, density and the type of statistics of their orientational distribution.

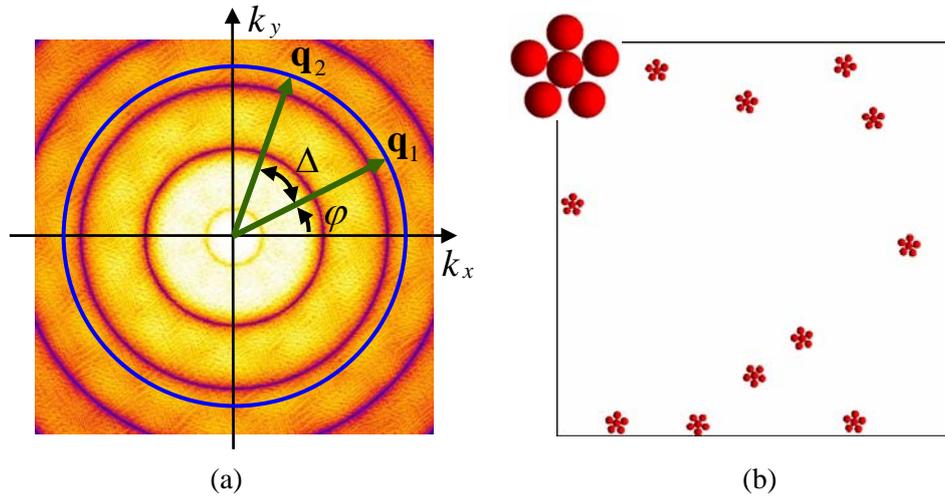


Figure 1: (a) The CCF $C_q(\Delta)$ defined on the intensity ring of a radius $q = |\mathbf{q}_1| = |\mathbf{q}_2|$. (b) 2D disordered sample composed of 11 pentagonal clusters in random positions and orientations.

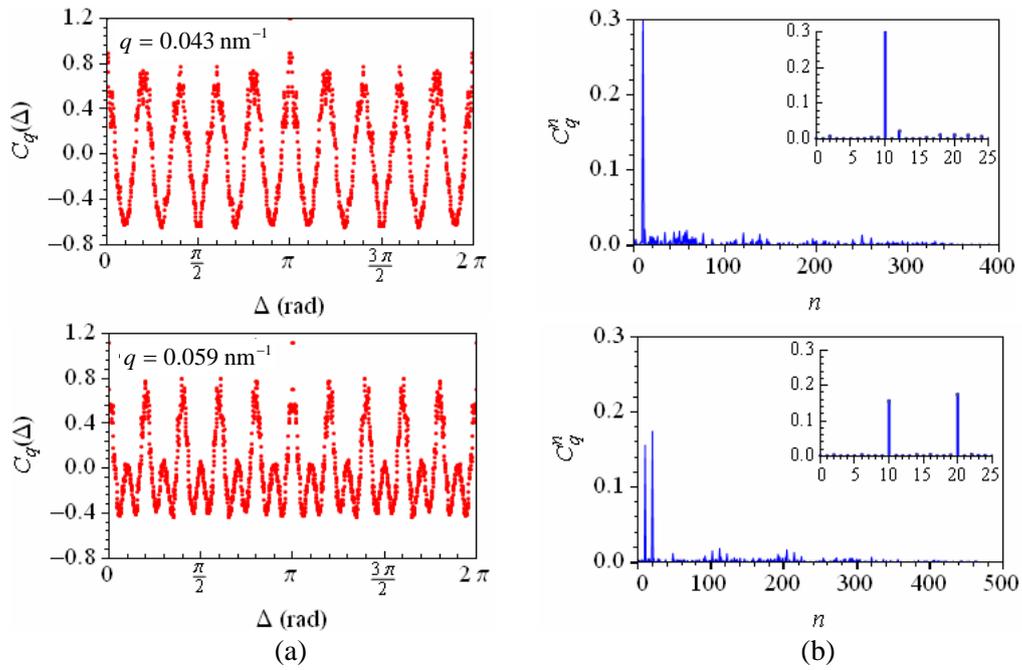


Figure 2: (a) CCFs (1) calculated at different values of the momentum transfer q , (b) The corresponding Fourier spectra (2).

References

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