High-resolution three-dimensional crystalline microscopy

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Abstract—In this communication, we discuss how 3D information about the structure of a crystalline sample is encoded in Bragg 3DXCDI measurements. Our analysis brings to light the role of the experimental parameters in the quality of the final reconstruction. One of our salient conclusions is that these parameters can be set prior to the ptychographic 3DXCDI experiment and that the spatial resolution limit of the 3D reconstruction can be evaluated accordingly.

1 Introduction

Since its introduction in the early 2000s [12], three-dimensional X-ray Coherent Diffraction Imaging (3DXCDI) has widely demonstrated its ability to provide non-destructive three-dimensional (3D) images of complex nanostructures. Two key features of 3DXCDI are noteworthy: 1) 3DXCDI offers the possibility to measure data either in a Bragg [2] or in a forward geometry, the former case providing 3D images of strains in crystalline materials [13,4]; and 2) 3DXCDI can be executed with a ptychographic (spatial) scan, hence providing images of extended samples [5,6,2]. For these reasons, 3DXCDI opens a wide playground for x-ray microscopy. In this communication, we discuss how 3D information about the structure of a sample is encoded in Bragg 3DXCDI measurements. In particular, our analysis brings to light the role of the experimental parameters in the quality of the final reconstruction.

2 Spatial scan at a Bragg peak

Any 3DCDI approach is inherently a lens-less tomographic modality: it is lens-less because the dataset (a series of coherent intensity patterns) is numerically inverted by a phasing algorithm, and tomographic since each collected diffraction intensity is drawn from the sample via a tomographic measurement.

Some details about the Bragg ptychographical experiment are now provided. Let us introduce first the exit-field

$$\psi_m = p_m \times \rho$$

where $m \in \{0, \cdots M - 1\}$ is the position index during the spatial (ptychographical) scan. In the relation above, $\rho : \mathbb{R}^3 \rightarrow \mathbb{C}$ denotes the scattering density [11, Sec. 7.1.2] of the diffracting and $p_m : \mathbb{R}^3 \rightarrow \mathbb{C}$ is the $m$-th coherent probe illuminating the sample. When a Bragg condition is met, both quantities are conveniently expressed with coordinates $r := (r_\perp, r_z) \in \mathbb{R}^3$ within a 3D frame in (direct-)space matching the detection geometry, see Fig. 1. In addition, for the sake of simplicity, we consider that the probing field is generated from a probe function $p$ shifted along the scattering direction $e_z$ i.e., we have $p_m(r) := p(r - r_m)$ with $r_m := 0 \times e_\perp + 0 \times e_y + m \Delta e_z$ where $\Delta \in \mathbb{R}$ is the step size of the spatial scan. In Bragg ptychography, as in any coherent diffraction method, the measurement is the intensity of the scattered-field collected by an array detector. Under the first-order born approximation [2, Sec. 8.10.1], the scattered wave-field collected in the “far-field” reads at the detector plane

$$\Psi(q_\perp, r_z = m\Delta) = \tilde{\psi}_m(q_\perp, q_z = 0)$$

where $\tilde{\psi}_m$ is the 3D Fourier transform of $\psi_m$ and $q := (q_\perp, q_z)$ is the 3D frequency (or reciprocal-space) coordinates. Finally, the expected (i.e., noise-free) measurements at the detector plane are given by the intensity of the scattered field [2]

$$I(q_\perp, r_z = m\Delta) = |\Psi(q_\perp, r_z = m\Delta)|^2.$$

Although the relations (2) and (3) are useful in deriving actual Bragg ptychography reconstruction algorithms (see for instance [6,7], it does not tell anything about the spatial information extracted from the sample via the ptychographical measurements. A substantial leverage to address this question is provided by the following result, easily deduced from the Slice Projection Theorem [10, Sec 6.3.3]

$$F_\perp^{-1} \Psi = \rho \otimes \Psi.$$  

In the relation above, $F_\perp$ is the bi-dimensional (2D) Fourier transform with respect to $r_\perp$, and $\otimes$ is the one-dimensional convolution operator acting along the scattering direction $e_z$. In other words, the 3D quantity

$$g(r) := (\rho \otimes \Psi)(r)$$

is an approximation of the scattering-density $\rho$ built on a filtering by the probe profile along $e_z$. The scattering-density approximation $g$ given in [5] appears in a previous publication from the authors [12]. In this communication, this relation is a pivotal tool in deriving resolution limits and sampling conditions for Bragg ptychography experiments.

Let us assumed that the probe profile along $e_z$ is a band-limited function with its support strictly contained in the domain $\Omega_z := [-\bar{q}_z, \bar{q}_z]$ with $\bar{q}_z \geq 0$. We deduce from [2] and [5] that the scanning step-size $\Delta$ should be set at least such that the spatial information is preserved in $g(r_\perp, r_z = m\Delta), m \in \mathbb{N}$. The Shannon-Nyquist sampling rate is then driven by the maximal frequency $\bar{q}_z$ in the (assumed) band-limited probe profile. In addition, because the spatial sampling is performed over wave-field intensities [3], it is not difficult to show that

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1 According to the usual convention in coherent Bragg diffraction, the origin of reciprocal space ($q_x = 0, q_y = 0, q_z = 0$) corresponds to a reciprocal space Bragg peak denoted by the reciprocal space vector $\mathbf{G}_{HKL}$ for a given $(H, K, L)$ Bragg reflection, see for instance [8].

2 In general, the 3D shifting of the probe can be casted within a non-orthogonal frame ($e'_z, e'_y, e_x$) with $e'_z$ pointing along the direction of the incoming probe, see Fig. 1. In the x-ray regime, the probe $p(r)$ is invariant along $e'_z$ and any shift along this direction does not provide any spatial information about the sample.
lattice of the probed crystal, the whole 3D Bragg peak is probed with unusually small angular ranges. In addition, the sample rotation results in a cartesian resolution, rather than polar sampling of the Bragg peak, see Fig. 1-Lower. When Bragg 3DXCDI is performed with a scanning (focused) probe, the relation (5) clearly states that a 3D reconstruction can still be obtained without sample rotation. If the sample rotation is also performed, we can expect more spatial information to be extracted. This is the question we aim at addressing in this section.

In Bragg geometry, a small rotation of the sample, by an angle $\delta_\theta$, results in a frequency shift by $\mathbf{w}$ of the 3D Fourier transform of the scattering-density $\rho$, see Fig. 1-Lower. The scattered field at the camera plane reads then

$$\mathbf{w}(q_L; r_z = m\Delta) = \mathbf{\tilde{w}}_{m,w}(q_L, q_z = 0)$$  

(8)

where $\mathbf{\tilde{w}}_{m,w}(r) := \rho(r)e^{i2\pi\mathbf{w}\cdot r}$ is the modulated exit-field. The relation (4) is modified accordingly

$$F_{\perp}^{-1} \mathbf{w} = (\mathbf{w}(\cdot ; \mathbf{W}))\mathbf{r}.$$  

(9)

The relation above shows that the accessible frequency domain along $u_z$ is now $\Omega_z := \mathbf{\Omega}_z \oplus \mathbf{w}^* u_z$ (where $\oplus$ is the Minkowski sum). In practice, a series of $N$ tilts is usually performed, inducing an equivalent series of frequency shifts denoted $\mathbf{W} := \{\mathbf{w}_n\}_{n=1}^N$. The best approximation one can achieve is then of the form (5) and reads

$$g(r; \mathbf{W}) = [\rho \otimes w(l ; \mathbf{W})].$$  

(10)

where the equivalent probe $p(r ; \mathbf{W}) := \sum_n P(r) \times e^{i2\pi w_n^* r}$ defines the spatial information extracted from the joint spatial/angular scan. The set of frequency that are extracted by this equivalent probe are $\Omega_z(\mathbf{W}) := \Omega_z \oplus \sum_n w_n^* u_z$, and the resolution limit along $e_z$ is obviously better than (7) and reads

$$R_z = 1/|\bar{q}_z|.$$  

(11)

We underline that the sampling condition (6) is actually unchanged when angular diversity is considered. A remaining, potential issue is that $\Omega_z(\mathbf{W})$ may not be a compact set, hence creating un-probed “holes” in the frequency space of the approximation (10). The condition $|w_n| \cos \theta_B \leq \bar{q}_z, \forall n$, with $\theta_B$ the Bragg angle nevertheless ensures a continuous probing of the frequency domain.

This last section clearly connects Bragg ptychography to other super-resolved imaging techniques, e.g., structured illumination microscopy [13], synthetic aperture [14] strategies. We also stress that these resolution limits are reached only in the asymptotic limit, with noise-free intensity measurements. In practice, both the photon shot-noise and the physical extension of the camera will reduce the effective resolution in all three directions.

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3Bragg 3DXCDI was introduced as a natural extension of standard CDI techniques. In this context, the method aimed at imaging isolated nano-crystals, with restricted supports small enough so that the unfocused coherent beam illuminating the sample can be considered as a single plane wave. The method was then mostly understood as a 3D Fourier synthesis strategy [3][4].

4If the chosen (probed) Bragg peak is not the one that sits at the origin of 3D reciprocal lattice, the angular range required for a full 3D scan is $\sim 1^\circ$.
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