Dynamic nanoimaging of extended objects via hard X-ray multiple-shot coherent diffraction with projection illumination optics

The functional properties of materials and devices often depend on their spatially hierarchical structures ranging in size from micrometer to nanometer scale. Thus, there is increasing importance in the *in situ/ operando* observation of their heterogeneous structures and structural changes during their operation. Coherent diffraction imaging (CDI) and its variants [1] in the hard X-ray regime are promising techniques for the nondestructive visualization of the internal structures of micrometer-thick objects at a nanometer-scale resolution owing to the short wavelength and high penetration depth. In CDI, a sample object is illuminated with spatially coherent X-rays, yielding Fraunhofer diffraction patterns, and then the spatial distribution of the complex refractive index of the object projected along the direction of the incident beam is reconstructed from the diffraction patterns via iterative phase retrieval (PR) calculation.

The measurement schemes of CDI are designed so that PR solution is efficiently constrained in real space. The local imaging of an extended object is achieved by a scanning variant of CDI, known as ptychography, where a set of diffraction patterns is collected such that illumination areas are sufficiently overlapped (Fig. 1(a)). In accordance with the constraints on the consistency of the object structures in the illumination-overlapped area (in real space) and the data fidelity to the Fourier magnitude of the exit wave (in reciprocal space), ptychographic PR reconstructs both the phase and absorption contrast image of the object together with the illumination wave field on the object. The "overlap constraint" yields the robust convergence of ptychographic PR, which greatly expands the application of ptychography [1]. However, the overlap constraint restricts the application to static objects. The overhead time included in the scanning measurement, which is typically 0.1–0.2 s per point, limits the temporal resolution of ptychography to several tens of seconds.

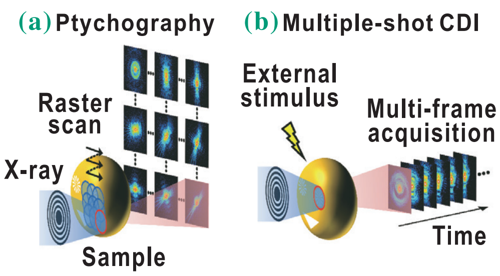

Fig. 1. Schematic illustration of ptychography (a) and multiple-shot CDI (b) experiments.

Therefore, nanometer-scale dynamics on the seconds to milliseconds time scale are still difficult to access in real space and are investigated by measurement in the reciprocal space such as by time-resolved small-angle X-ray scattering and X-ray photon correlation spectroscopy.

In this work, we develop multiple-shot CDI, an extension of CDI to dynamic nanoimaging, and demonstrate, with numerical simulations and a proof-of-concept experiment at SPring-8 **BL24XU**, that the proposed method is capable of achieving temporal resolutions of 10–100 ms, which is ~100 times faster than that of ptychography [2]. Figure 1(b) shows the experimental setup of multiple-shot CDI, where a movie of local structural changes in an object is reconstructed from a set of time-evolving diffraction patterns. The multiple-shot CDI system also allows the complementary use of ptychography, where the two methods can be switched by changing the measurement scheme (Fig. 1). The development includes a multiframe PR algorithm dedicated to a video reconstruction of a dynamic object

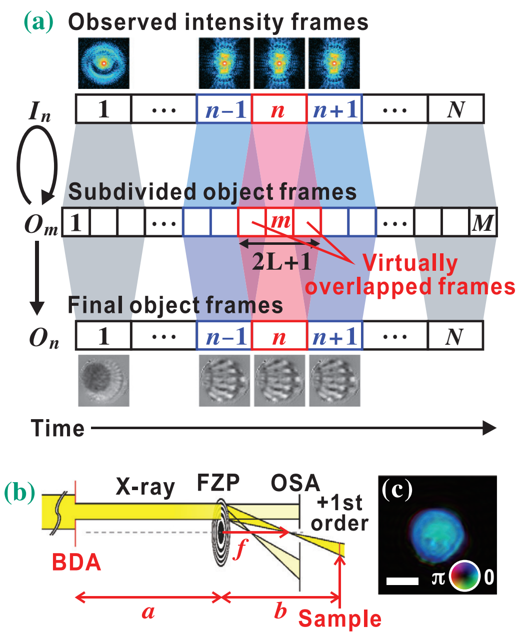

Fig. 2. Key developments in multiple-shot CDI. **(a)** Schematic illustration representing multiframe PR. Correspondences among the observed intensity frames I_n, numerically subdivided object frames O_m, and actual object frames O_n are shown. **(b)** Projection illumination optics. BDA beam-defining aperture, FZP Fresnel zone plate, OSA order-sorting aperture. BDA, FZP, and Sample are arranged in accordance with the thin lens formula $\frac{1}{a}+\frac{1}{b}=\frac{1}{f}$. **(c)** Illumination beam generated with projection illumination optics. Experimentally measured by ptychography. The scale bar indicates 1 μm.

and projection illumination optics for the generation of a top-hat-intensity beam to enhance the convergence of the proposed algorithm.

The key concepts of the proposed algorithm are illustrated in Fig. 2(a). (1) For a dynamic object, the diffraction pattern will change continuously even during a single-frame exposure, but previously, this was not taken into account. In the proposed algorithm, each frame of the object movie is divided into subframes, and each frame of measured diffraction patterns is represented as a sum of patterns calculated from the object subframes. (2) In real space, it is expected that the structural changes of the object will be smooth in both spatial and temporal dimensions. Thus, we introduced two types of spatiotemporal smoothness constraint. First, each object subframe between two adjacent measured frames is constrained by both frames to ensure temporal smoothness. Second, we applied the total variation regularization [3] along both the spatial and temporal directions. The total variation regularization restricts the spatiotemporal gradient of the object to be small, and thus, it facilitates the convergence of the object to the spatiotemporally smooth solution.

Illumination uniformity is also critical for multiframe PR. The main lobe of the conventional focused beam shows a Gaussian-like intensity profile, which causes the slow recovery of the object periphery because of the dim illumination and prevents the convergence to the optimum solution. We generated the top-hat-intensity beam by developing the projection illumination optics employing a zone plate as an imaging lens (Fig. 2(b)). Here, a beam-defining aperture is illuminated with a spatially coherent X-ray, and a real image of the aperture is formed on the sample with size reduction. Figure 2(c) shows an illumination beam generated by the proposed optics with 12.5-fold reduction, which was experimentally measured by ptychography. The generation of a nearly uniform illumination beam with a well-defined boundary is demonstrated.

We constructed a multiple-shot CDI system at BL24XU. First, the performance of multiple-shot CDI was investigated with numerical simulations based on the experiment at the photon energy of 8 keV. Brownian motions of colloidal gold particles were calculated with a time step of 1 ms and various diffusion coefficient values, yielding Brownian motions with root mean square displacements (RMSDs) ranging from 0.16 to 3.22 pixels/10 ms. The largest RMSD was ~30% of the particle size. Time-evolving diffraction patterns were simulated at a frame time of 10 ms, and real-space movies were reconstructed from the datasets. Compared with the conventional PR algorithm, the proposed algorithm markedly improves the reproducibility of the object. The fastest Brownian motion was also reconstructed with the twofold deterioration of spatial resolution, even though their diffraction patterns are blurred owing to accumulation over the frame time. This result indicates the robustness of the proposed method. Figure 3 shows the results of the proof-of-concept experiment, where a resolution test chart made of 500-nm-thick tantalum was continuously moved at a speed of 125 nm/s and imaged at a frame time of 100 ms via multiple-shot CDI. On average, ~79 nm lines and spaces are resolved in each frame. These results demonstrate the feasibility of visualizing dynamic phenomena, including those difficult to understand in reciprocal space, at temporal resolutions of 10–100 ms. It is also expected that the use of a partially coherent beam and/or an upcoming next-generation synchrotron radiation source will increase the spatiotemporal resolution. We expect that the proposed method will contribute to the understanding of the dynamical properties of heterogeneous systems in the materials and biological sciences and industry.

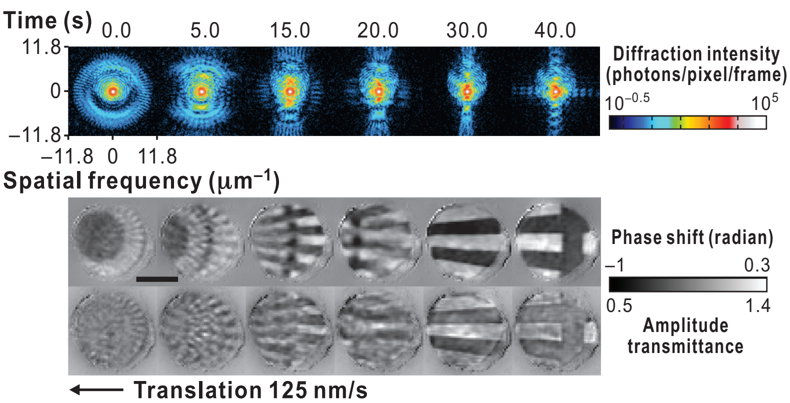

Fig. 3. Proof-of-concept experiment of multiple-shot CDI. The selected frames of the observed diffraction patterns (upper) and corresponding reconstructed frames of the phase (middle) and absorption (lower) images of the object are shown. . The scale bar indicates 1 μm.

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