High-fluence multilayer focusing optics attaining 2 nm spatial resolution in XFEL based coherent diffractive imaging

Femtosecond coherent diffractive imaging (CDI) is an analytical technique made possible by X-ray free electron lasers (XFELs). This is a microscopic technique in which an object image is reconstructed using an iterative algorithm from a coherent diffraction pattern obtained by illuminating a sample with a femtosecond XFEL pulse. Compared with CDI using synchrotron radiation, CDI using XFEL allows us to detect information under the native states of samples before radiation damage. On the basis of the theoretical calculations, femtosecond CDI is anticipated to take snapshots of biological molecules with a spatial resolution approaching an atomic resolution.

Various efforts have been made to achieve a high resolution; however, the spatial resolution of CDI in single-XFEL-pulse exposure has been limited to ~5 nm. A higher fluence to illuminate samples is required to obtain a higher spatial resolution in femtosecond CDI, because the signal-to-noise ratio obtained on a detector, i.e., a low amount of signal from a very small sample, is a critical problem. Our group has developed XFEL focusing mirrors to obtain a high fluence focus by solving technical problems in the fabrication of ultrahigh-precision mirrors with atomic-level precision. The developments have enabled us to realize an extremely high fluence focus at a beam size of 100 nm and to improve the spatial resolution of femtosecond CDI to an unprecedented 2 nm with single-XFEL-pulse exposure [1].

Our group designed a focusing optical system that matches CDI optics and includes X-ray energy. Our group employed multilayer focusing mirrors in the Kirkpatrick–Baez (K–B) geometry to focus X-rays efficiently with high demagnifications. A higher fluence at the focus can be produced by utilizing higher demagnification with smaller focal lengths, because multilayer mirrors can be designed to have larger glancing angles than general total reflection mirrors. However, multilayer focusing mirrors have steeply curved aspheric substrates, making them difficult to fabricate. The fabrication of the X-ray focusing mirrors requires extremely high precision technologies of a surface measurement system at the atomic level as well as an aspheric processing method.

Our group developed, in particular, a new high-precision surface measurement system of zero-method surface profilometry (ZSP) (see Supplementary Information in Ref. 1.) in addition to an ultraprecise processing technology used in the fabrication of X-ray mirrors at SPring-8 [2]. Using these fabrication technologies, substrates of the X-ray focusing mirrors with an elliptical-cylinder shape were finished with a precision of 0.2 nm in root mean square at the SPring-8 site. The mirror surfaces were then coated with $(Cr/C)_{30}$ multilayers in a lateral graded structure using a multilayer deposition system at SPring-8. The design of the focusing optics at an optimized X-ray energy of 4 keV is as follows. The focal length is 190 mm (110 mm) in the horizontal (vertical) direction, the two mirrors are 80 mm long, the two incident angles are 25 mrad, and the working distance is 60 mm. The mirrors have a large spatial aperture of 1850 μm square compared with the incident X-ray beam size of ~550 μm in full width at half maximum (FWHM).

Our group developed a femtosecond CDI instrument using the fabricated multilayer focusing mirrors at SACLA **BL2**. This system is called MAXIC-S. A photograph of the main vacuum chamber, which is equipped with X-ray focusing mirrors, an alignment mechanism, and sample scanning stages, is shown in Fig. 1. To avoid contaminants on the mirrors, air absorption, and unwanted scattering, all of the components, including the focusing mirrors, the sample, and the multiport charge-coupled device (MPCCD) detector to record diffraction patterns, are enclosed in a vacuum environment. The MPCCD detector can obtain diffraction signals at a full-period resolution of 2 nm at the edge.

Our group evaluated the focused beam produced by the multilayer mirrors using the knife-edge

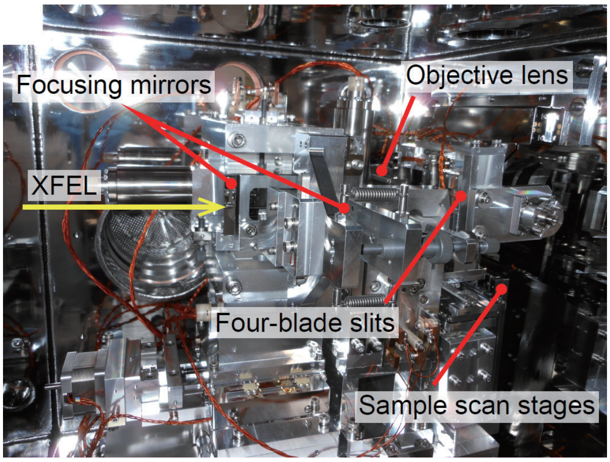

Fig. 1. Photograph of the CDI instrument at SACLA. [1]

scanning method at SACLA. Measured beam sizes were 110 nm and 60 nm (FWHM) in the horizontal and vertical directions, respectively, which are in agreement with the design. The reflectivity of the double reflection by the multilayer focusing mirrors was 52%, while the theoretical reflectivity is 62%. The optics achieved an extremely high fluence focus at 3×10^5 J/cm^2/pulse with the evaluated beam sizes and a pulse energy of 61 μJ. The photon density was 4×10^{12} photons/μm^2/pulse, and the intensity was 3×10^{19} W/cm^2 at a pulse duration of 10 fs. The photon density of XFEL was enhanced by a factor of 2×10^6 compared with the unfocused beam. This focus has a 50-times-higher photon density than the 1 μm focus [3] used for CDI at SACLA.

The CDI instrument utilizing the developed high-fluence beam was applied to observe Au nanoparticles (AuNPs) in water (Fig. 2). The sample AuNPs have a bipyramidal shape with a volume of ~20 nm cube, which includes electrons as small as the same order of magnitude as that of biological particles, such as a virus particle of ~70 nm diameter. As a result of the observation by single-XFEL-pulse exposure, the CDI system attained an unprecedented high spatial resolution of 2 nm (full-period resolution). Au particles become a highly functional material with unique physical and chemical properties when its size is reduced to the nanometer scale. In water, which is applicable to biological samples, the observation before changing with XFEL illumination was successfully demonstrated at this extremely high spatial resolution using ultrafast XFEL exposure. The observed result was a native structure of the material showing important information for elucidating the relationship between the structure and function of nanoparticles. In addition, the validity of the fluence at the focus evaluated by the knife-edge scanning method was confirmed by the agreement with the fluence estimated from the measured coherent diffraction pattern.

The CDI system developed in this study has been applied to observe biological samples and functional materials. The microscope is expected to be utilized for the ultrahigh-resolution and ultrahigh-speed observation of the native state of samples that has never been seen previously.

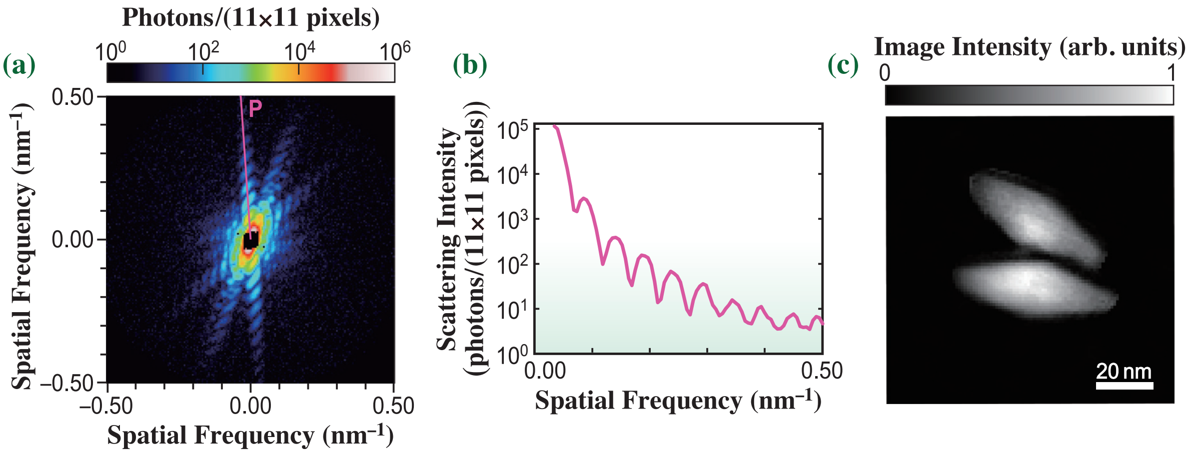

Fig. 2. Demonstration of femtosecond coherent diffractive imaging. **(a)** Measured coherent diffraction pattern. (**b**) Profile along line P in (a). (**c**) Reconstructed image. [1]

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