Hard X-ray Fourier transform holography from an array of oriented referenced objects

The spatial resolution of optical imaging methods depends on the wavelength of the beam used. In the case of light microscopes, a spatial resolution obtained (~100 nm) is comparable to or even better than the wavelength. Magnified images are created by a set of lenses, which in mathematical terms, calculates the reverse Fourier transform of the light scattered from the object (the Fourier transform of the object). With our current technology we can fabricate lenses with precision good enough to reach the theoretical value of spatial resolution.

By using hard X-ray instead of visible light, one should in principle be able to create a microscope with an atomic resolution (~0.1 nm). However, the major difficulty is that we cannot fabricate lenses good enough to ensure atomic resolution. We therefore use computers to calculate the reverse Fourier transforms directly from the patterns of X-ray scattered from the objects. Here we encounter another obstacle, called the "phase problem". As an electromagnetic wave, an X-ray photon has its amplitude and phase, and both are needed to restore the original structure of the object. However, the phase information is lost when the scattering is recorded on a detector.

Then, what we get when we calculate a reverse Fourier transform without phase information? The Fourier transform calculated in this way is called a "Patterson function" (Fig. 1). A Patterson function is the autocorrelation function of the object, i.e., a collection of vectors representing all possible combinations of two points within the object. Although a Patterson function contains information about the structure of the object, it is usually not useful because it is nothing more than a heap of hopelessly overlapping vectors. However, a small trick can dramatically improve the situation. If one places a small dot near the object, and if the distance between the dot and the object is greater than the longest vector within the object, the vectors connecting the dot and the object (called cross-correlation terms) will never overlap with vectors within the object. In this way the structure of the object can be isolated from the rest of the Patterson function. This is a kind of holography, and because the structure is restored through the operation of Fourier transform, this method is called "Fourier transform holography".

The method of Fourier transform holography has been successfully implemented for soft X-ray, with wavelengths of >1 nm. The structures of microfabricated patterns or natural objects such as diatoms have been retrieved. The dots near the objects (reference dots) are also microfabricated, and the spatial resolution is determined by its size (~50 nm) rather than the wavelength.

If the technique of Fourier transform holography is to be extended to the realm of hard X-ray (wavelength ~0.1 nm) and the size of the reference dot is reduced to that of an atom (~0.1 nm), literally an atomic resolution should be obtained. Here we face a third obstacle: If the size of the reference dot is reduced, so is the amplitude of the cross-correlation terms, weakening the signal. To make the matters worse,

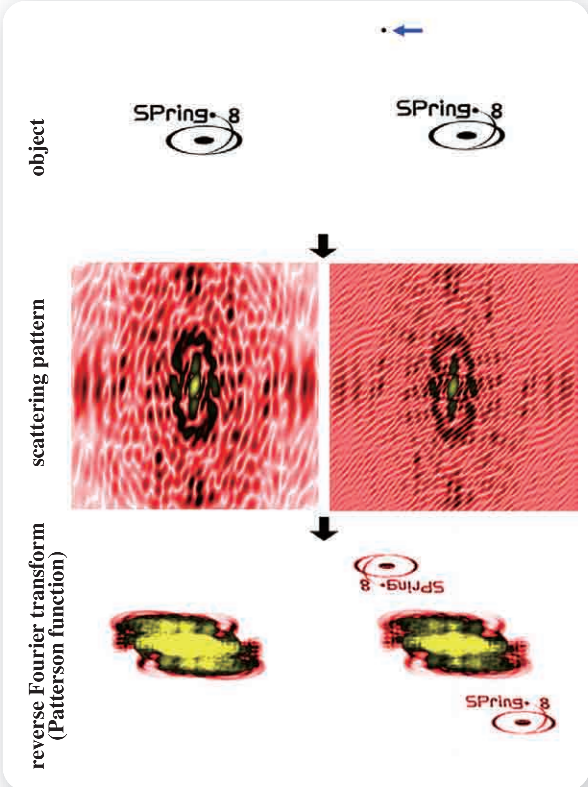

Fig. 1. Principle of Fourier transform holography. Top row, objects; middle row, scattering patterns recorded on the detector (Fourier transform of the objects); bottom row, reverse Fourier transforms of the scattering patterns calculated without phase information (Patterson function). Generally it is difficult to extract useful structural information from the Patterson function (left bottom), but if a reference dot is placed near the object (right top, blue arrow), the structure of the object is directly imaged in the Patterson function as a symmetric pair of cross-correlation terms (right bottom).

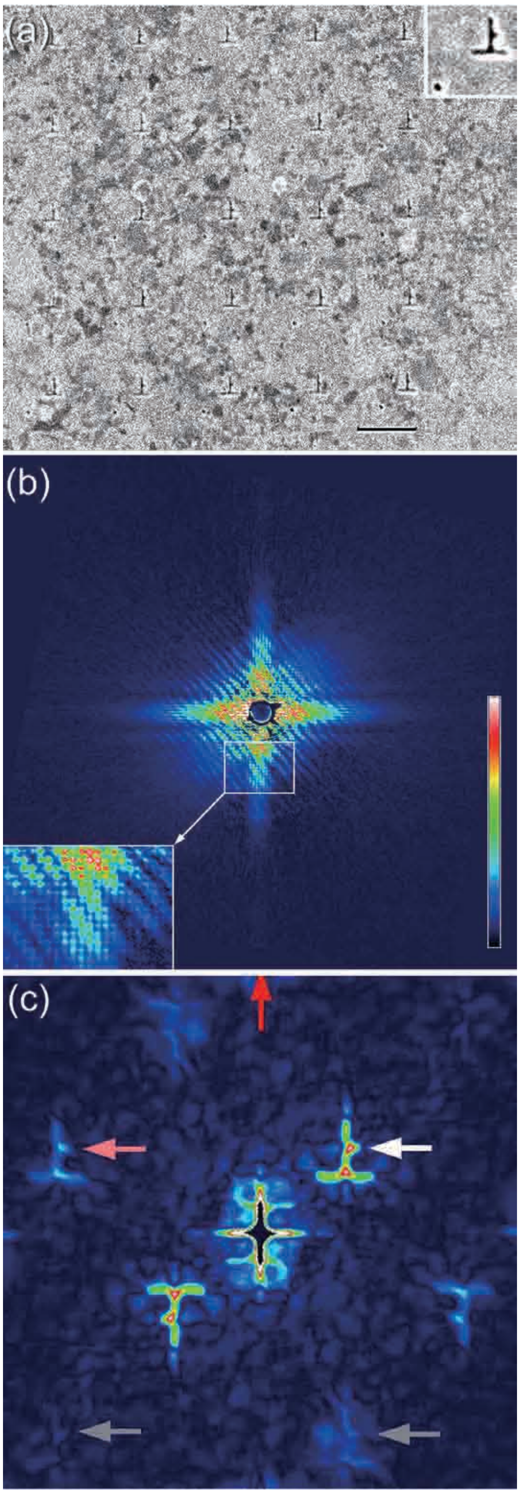

Fig. 2. Fourier transform holography from multiple pairs of reference and object. **(a)** A 5×5 matrix of pairs of reference and object (Chinese character meaning "up"), nanofabricated on gold substrate. **(b)** scattering pattern from the test pattern shown in **(a)**, recorded at BL40XU beamline by using hard X-ray (wavelength, 0.15 nm). **(c)** Patterson function calculated from the scattering pattern shown in **(b)**. Note that the Chinese characters are clearly retrieved. [1]

hard X-ray interacts with matters much more weakly than soft X-ray, further weakening the signal. As a result of these effects combined, practically no signal will be detected by any of the present-day detectors if, for example, a single atom is used as a reference and a single protein molecule is used as an object.

The paper by Iwamoto and Yagi [1] describes a method to overcome this obstacle. The idea is to prepare a large number of object-reference pairs oriented in the same direction, and illuminate them with hard X-ray simultaneously to obtain stronger signal. To test the feasibility of this method, they nanofabricated a test pattern, in which pairs of a reference dot and a Chinese character are arranged in a 5×5 matrix (Fig. 2(a)). Figure 2(b) shows the scattering pattern from it (hologram). In the scattering pattern, one can observe a series of coarse stripes coming from the interference between the reference dot and the Chinese character. The stripes further consist of fine dots arranged in a square lattice, representing the interference of reference-object pairs within the matrix. Figure 2(c) shows the Patterson function calculated from the scattering pattern (Fig. 2(b)). A pair of symmetrically arranged Chinese characters is clearly reproduced around the center, and weaker characters are also reproduced in the periphery. All experimental data were carried out at beamline **BL40XU**.

This study shows that the technique of Fourier transform holography using multiple oriented pairs of references and objects is as effective as in the single-object case, and can be used as an effective means to increase the signal level. This method will be especially useful for determining the atomic structure of weakly scattering biomolecules or their assemblies, which are not crystallizable but identical particles of which can be prepared in large numbers.

Hiroyuki Iwamoto

SPring-8/JASRI

E-mail: iwamoto@spring8.or.jp

References

[1] *H. Iwamoto and N. Yagi: J. Synchotron Rad.* ***18*** *(2011) 564.*