Cheating the diffraction limit using X-ray nonlinear diffraction

Is there any ingenious method to see finer details than the diffraction limit? In 1878, E. Abbe first pointed out the diffraction limit that the spatial resolution of imaging system is determined ultimately by the half-wavelength, $\lambda/2$ [1]. Nevertheless, scientists have challenged to see smaller structure than $\lambda/2$. Recently a spatial resolution of around $\lambda/10$ was achieved by using the dispersion of Plasmon [2]. Further improvement along this scheme is, however, not feasible due to, for example, the collective nature of Plasmon, that requires a certain volume containing many electrons.

Now we propose a novel method to realize an atomic resolution in the optical region [3]. The unprecedented high resolution unveils the local optical response: how electrons in materials respond to the light. We think that the difficulty in improving the spatial resolution arises from the use of a single wavelength. Instead, we consider using two beams with different wavelengths and separating the spatial resolution from the probing wavelength of interest. However, even if we use two beams, they behave independently without giving any new information. The key piece is X-ray parametric down-conversion (PDC), which makes the two beams cooperate to image the local optical response. Our method does not beat the diffraction limit, but shifts it to X-rays, freeing the probing light from the Abbe's constraint.

X-ray PDC is one of the second order nonlinear optical processes, where an X-ray pump photon decays spontaneously into two photons (signal and idler). We investigate X-ray PDC for the idler photon in the optical region, and find that it is the optical response at the idler frequency that determines the efficiency of X-ray PDC. The underlying mechanism is considered to be the Doppler shift of the pump photon by oscillating electrons driven by the optical idler photon [3,4]. Thus, the X-ray second order nonlinear susceptibility, $\chi^{(2)}$, relates to the optical linear susceptibility, $\chi^{(1)}$, at the idler frequency. In addition, the spatial information obtained by X-ray PDC has the atomic resolution, because it is an X-ray coherent process. When we combine these two features, we expect that we can see the optical response at the idler frequency with the atomic resolution by X-ray PDC.

In fact, X-ray PDC is observed as a nonlinear

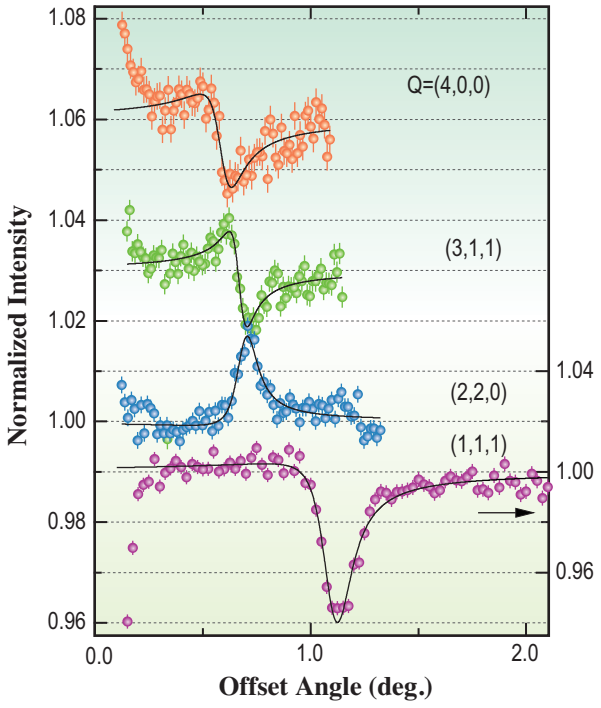

Fig. 1. Rocking curves of nonlinear diffraction measured for the signal wave of X-ray PDC with diamonds. The pump and the idler energies are 11.107 keV and 60 eV. The signal intensity is normalized to the background due to the Compton scattering.

diffraction. Figure 1 shows the rocking curve of the nonlinear diffraction, which is the glancing angle dependence of the intensity of the signal wave. The nonlinear crystal is synthetic type IIa diamonds. The idler photon energy is 60 eV in the extreme-ultraviolet (EUV) region. The asymmetric peaks are due to the Fano effect between X-ray PDC and the background Compton scattering [5]. From the Fano spectra, we estimate, $\chi^{(2)}_{\mathbf{Q}}$ the **Q**th Fourier coefficient of $\chi^{(2)}(\mathbf{r})$ [6]. Then, $\chi^{(2)}_{\mathbf{Q}}$ is converted to $\chi^{(1)}_{\mathbf{Q}}$, the **Q**th Fourier coefficient of $\chi^{(1)}(\mathbf{r})$ [3]. The reconstruction of $\chi^{(1)}(\mathbf{r})$ from $\chi^{(1)}_{\mathbf{Q}}$ follows the standard procedure of X-ray structural analysis.

Figure 2 shows the section of $\chi^{(1)}(\mathbf{r})$ on the (110) plane which contains both the atoms and the covalent bonds. The resolution of the reconstruction, 0.54 Å, is determined by the largest |**Q**| given by the 400 reflection. On the other hand, 203 Å (60 eV) is the idler wavelength at which $\chi^{(1)}(\mathbf{r})$ represents the optical response. As a result, the resolution in the wavelength unit reaches as fine as λ/380. This is the highest one ever achieved.

The reconstructed $\chi^{(1)}(\mathbf{r})$ reveals the local optical response of diamonds to the EUV light. The core electrons at the atomic site oscillate in phase to the EUV light, whereas the bonding electrons respond in the opposite phase. The opposite sign of oscillation between the atomic and the bonding electrons is due to the different resonance energies, 289 eV and 12 eV, respectively. The contribution from boding electrons is larger than that from the core electrons. So the bonding electrons determine the optical response at 60 eV. We note that these observations are found to be qualitatively consistent with the Lorentz model [3].

The present method opens a new window into the optical property of solids, and adds new structural information to the optical response. Such a microscopic structure cannot be obtained with the conventional spectroscopy, because of the longer wavelength of light than the size of the unit cell. In other words, the charge response at longer wavelengths can be investigated only in the vicinity of the origin in the momentum space. Now, X-ray PDC into optical region can access the local optical response and the charge response over the whole momentum space for better understanding of the optical property of solids.

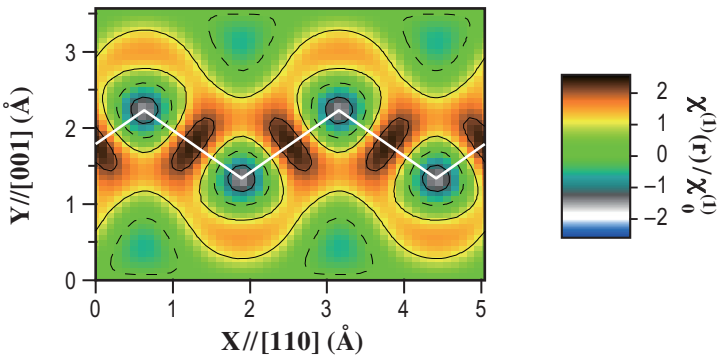

Fig. 2. Reconstructed $\chi^{(1)}(\mathbf{r})$ of diamonds at 203 Å (60 eV) on the (110) plane. The resolution is 0.54 Å, corresponding to λ/380. The white solid lines indicate the bonds between carbon atoms.

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References

[1] M. Born and E. Wolf: Principles of Optics (Cambridge University Press, 1999).
[2] S. Kawata *et al.*: Nature Photon. **3** (2009) 388.
[3] *K. Tamasaku, K. Sawada, E. Nishibori and T. Ishikawa: Nature Phys. 7 (2011) 705.*
[4] I. Freund and B.F. Levine: Phys. Rev. Lett. **25** (1970) 1241.
[5] K. Tamasaku and T. Ishikawa: Phys. Rev. Lett. **98** (2007) 244801.
[6] K. Tamasaku *et al.*: Phys. Rev. Lett. **103** (2009) 254801.