X → X+EUV Parametric Conversion Interfered with by Compton Scattering

The following year of the invention of laser in 1960, optical nonlinear response was demonstrated by generating the second harmonic of light. Now, nonlinear optics becomes an indispensable tool for science and engineering. On the other hand, there is little knowledge of nonlinear optics in the hard X-ray region to date. Scientists considered that nonlinearity in the X-ray region is too small to investigate and to find applications without X-ray lasers. Here, we show that some process in X-ray nonlinear optics is observable with sufficient accuracy for quantitative analysis by the currently available X-ray sources.

We investigated parametric down-conversion of X-rays into extreme ultraviolet (EUV), the lowest order nonlinear process. The X-ray pump photon decays spontaneously into the X-ray signal photon and the EUV idler photon, i.e., X→X+EUV, when both energy and momentum are conserved. The momentum conservation is usually referred to as phase-matching condition. The phase matching is realized by using a reciprocal lattice vector, **Q**, resulting in nonlinear diffraction (Fig. 1(a)). When the idler photon is in the EUV region, it was predicted that the efficiency of parametric down-conversion is determined by the **Q**-th Fourier component of the bond charge density [1,2], as is shown schematically in Fig. 1(b).

The experiment was performed at the 27-m in-vacuum undulator beamline **BL19LXU**. The pump energy was 11.0 keV, whereas the signal (idler) energy was changed from E_s = 10.96 keV (E_i = 40 eV) to 10.87 keV (130 eV) by a bent crystal analyzer (Fig. 2). Figure 3 shows the rocking curves of the 111 nonlinear diffraction of diamond. Rocking the crystal corresponded to scanning the phase-matching condition.

First we discuss the rocking curve at E_i = 100 eV [3]. On the exact phase-matching angle, there observed small enhancement of the signal intensity by ~10 counts/sec due to the X→X+EUV parametric conversion. We note that the incidence was eleven orders of magnitude higher. To our surprise, the measured rocking curve was asymmetric, having not only a peak but also a distinct dip. If there had been no interaction between the X→X+EUV parametric conversion and the Compton scattering, there should be a Lorentzian peak on the smooth Compton background. The high signal-to-noise ratio measurement made it clear for the first time that the Compton scattering interferes with the nonlinear process.

The rocking curve depended strongly on the signal and the idler energies [4]. The line shape, which was Lorentzian-like, became asymmetric, and finally characterized by a dip, as E_i (E_s) decreased (increased). The energy dependence is considered to relate to E_i, because there is no internal excitation in diamond around 11.0 keV, e.g., the nearest core excitation energy is 290 eV.

The physical picture of the interference is not clear at present. We pointed out similarity to the Fano effect, which is observed when a discrete state is buried in a continuum. The phase-matched parametric conversion and the Compton scattering may be regarded as the discrete transition and the

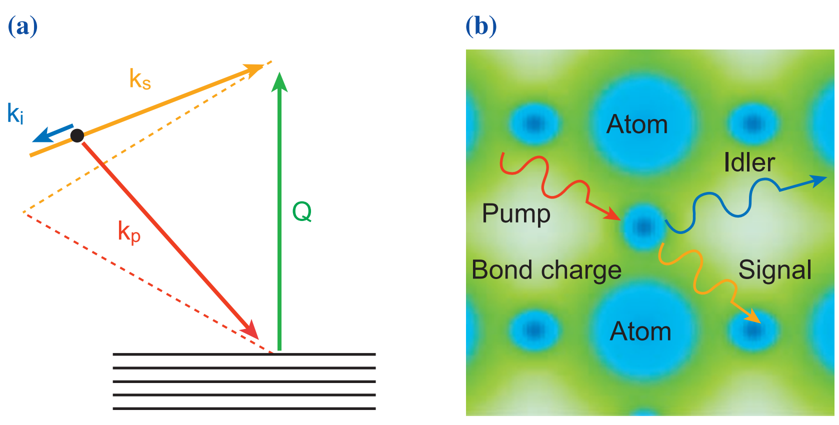

Fig. 1. Schematic view of **(a)** nonlinear (solid line) and linear (dashed line) diffraction, and **(b)** the X→X+EUV parametric conversion due to the bond charge.

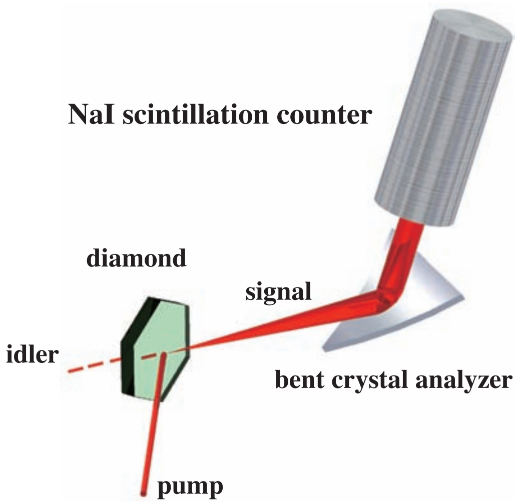

Fig. 2. Schematic view of the experimental setup. The signal photon was measured, whereas the idler photon was absorbed in the nonlinear crystal.

continuous transition, respectively. The difficulty of this picture is that quantum mechanical interference, such as the Fano effect, requires the identical final state for both the transitions. However, the final state of the parametric down-conversion is different from that of the Compton scattering, e.g., the number of photon is two in the former case, and one in the latter case. Further experimental and theoretical investigation is needed to understand the X→X+EUV parametric conversion and to apply it to structural analysis of the bond charge density.

The experimental findings shown here imply that the X-ray nonlinear optics has large frontiers. Also, the results show that the part of frontiers can be explored with the present X-ray sources.

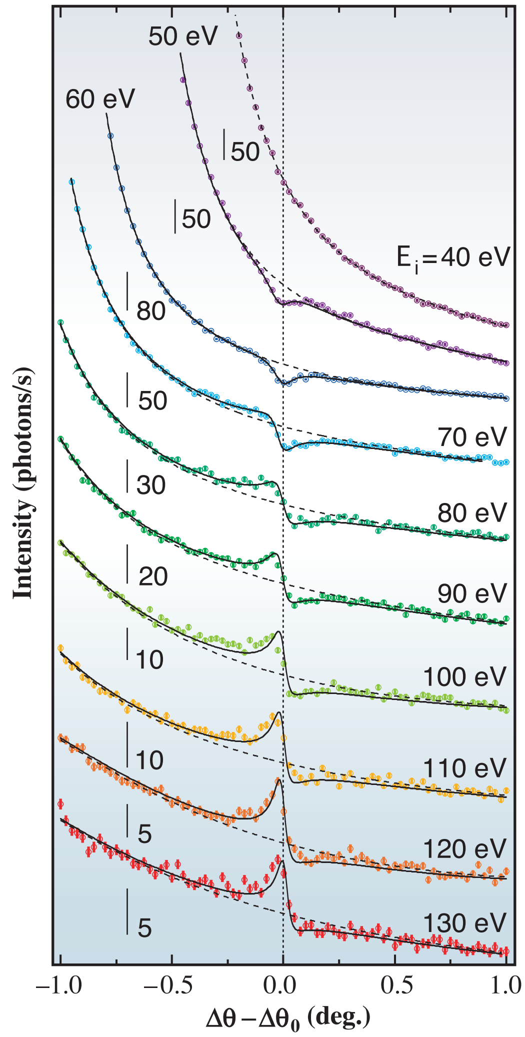

Fig. 3. Rocking curves of the 111 nonlinear diffraction of diamond for several combinations of E_i and E_s. The vertical dashed line indicates the exact phase-matching angle. The solid and the dashed line are the best fitting with the Fano formula and the estimated background, respectively.

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