Recent progress in x-ray nonlinear optics and future perspectives

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X-ray nonlinear optics [1] has been considered as a frontier in both x-ray physics and nonlinear optics, and to be explored with x-ray free-electron lasers (XFEL). Recently, we succeeded in observing parametric down-conversion (PDC) of x-rays into extreme ultraviolet [2], vacuum ultraviolet [3], and soft x-rays [4] using an existing synchrotron-based x-ray source, SPring-8.

Our precision measurements reveal unique features of x-ray nonlinear optics [5]. X-ray PDC is observed indirectly through quantum mechanical interference known as the Fano effect [4]. The nonlinear susceptibility is sensitive to the resonance, and shows strong resonance enhancement. The second order nonlinear susceptibility has microscopic structure, which changes its shape as the energy of down-converted (idler) photon.

We review the physical background of nonlinear optical phenomena in the hard x-ray region, the recent experimental results, and discuss the following possibilities.

1. Pump-probe measurement

The second order nonlinear susceptibility is considered to reflect some microscopic property of the optical response at the idler energy, which cannot be measured with any linear optical process. X-ray PDC combined with the pump-probe technique can be a powerful tool to investigate dynamics of materials microscopically.

2. X →X+SX parametric conversion

Recently soft-x-ray resonant diffraction is recognized as a powerful tool to investigate materials [6]. However, the reachable resolution is in the nanometer range and stronger absorption limits the environment of sample. X-ray PDC into the soft-x-ray region could make it possible not only to obtain the same information as the soft-x-ray diffraction, but also to realize atomic resolution and to avoid the problem of absorption.

3. X →X+V parametric conversion and sum-frequency generation (SFG)

Observation of x-ray PDC into the visible region is still very challenging due to technical difficulty. The process is expected to give microscopic information about the optical property of solids [7]. If we find an efficient nonlinear material, it may be used for diagnosing x-ray free-electron lasers. A cross-correlator with fs-time resolution may be realized by SFG of XFEL and fs-Ti:sa laser to measure the pulse shape shot-by-shot.

4. Optical parametric amplifier (OPA)

XFEL oscillator (XFEL-O) is one of the next generation x-ray lasers, which provides fully coherent and very narrow-band x-rays by virtue of a cavity [8]. A drawback of the cavity is that it operates at a fixed energy. If we construct OPA with good nonlinear crystals, we may be able to produce x-ray laser with arbitrary photon energies. This could open up a spectroscopic application of XFEL-O.

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