Element selectivity in second-harmonic generation by using a soft-X-ray Free Electron Laser

Significant developments of lasers have led us to discover novel optical responses of matter. A well-known example is the nonlinear effect that generates a signal from specific environments, such as inside a nonlinear crystal and a surface/interface system, that have hardly been probed in usual optical experiments. Nowadays, laser pulses of soft and hard X-rays can be used in experiments at X-ray free electron laser (XFEL) facilities such as the SPring-8 Angstrom Compact Free Electron Laser (SACLA). The energy of photons covers various absorption edges of elements and it can be tuned to satisfy the resonance condition in the interaction between light and matter. The resonance effect has been used to perform X-ray spectroscopy or X-ray scattering experiments with element selectivity. Using ultrahigh-intensity laser pulses of an XFEL, one can now examine the core-level resonance of the nonlinear optical effect (Fig.1).

In the present research, we investigated second-harmonic generation (SHG), one of the nonlinear optical frequency conversions, in the soft X-ray region [1]. The demonstration was performed at SACLA-SXFEL **BL1**. The FEL was operated at a nominal electron beam energy of 400 MeV in the experiment. The SXFEL beamline was chosen owing to the advantages of photon-energy tuning in the EUV-soft X-ray region and ultrahigh brilliance, enabling the examination of nonlinear optics at various photon energies. The sample was a non-centrosymmetric crystal of $GaFeO_3$. The resonance enhancement of SHG signals was expected when the photon energy with double the frequency 2ω was above the absorption edge of a sample. Thus, the SHG resonance condition ($2\omega = \omega_{ng}$) was satisfied by the energy difference between the Fe 3*p* level and the unoccupied band, as shown in Fig. 2.

Figure 3(a) shows an overview of the beamline and the measurement system. The measurement was made in the reflection geometry. The reflected light from the sample was incident on the grating and the diffracted intensity was measured using a microchannel plate (MCP) detector that could be moved along the energy-dispersion direction. The spectral intensities of the reflected light (I_ω and $I_{2\omega}$) were measured at various MCP positions, as shown in Fig.3.

Figure 3(b) shows the Fe 3*p* absorption spectrum of the $GaFeO_3$ crystal, showing the Fe 3*p* absorption edge above 54 eV. The photon energy of the SXFEL pulse was set to half of the Fe 3*p* absorption edge. Figure 3(c) shows the variation of the reflected intensity at a photon energy of $\hbar\omega = 27.5$ eV with respect to the incident intensity I_0. The dashed line represents the fitted curves obtained by using the power law $\propto I_0^\beta$. The exponent coefficient is $\beta = 1.0$, which verifies the linearity of the ω component. The intensity of the 2ω component ($2\hbar\omega = 55$ eV) clearly shows its nonlinear dependence on I_0, as shown in Fig. 3(d). Moreover, the 2ω signal intensity significantly decreases when the photon energy is tuned to $2\hbar\omega = 53$ eV ($\hbar\omega = 26.5$ eV), which is below the Fe 3*p* absorption edge (Fig. 3(e)). From the power-law fitting, the exponent coefficients are $\beta = 1.8$ ($2\hbar\omega = 55$ eV), $\beta = 2.0$ ($2\hbar\omega = 57$ eV), and $\beta = 2.0$ ($2\hbar\omega = 59$ eV). The experimental values consistently match the quadratic intensity dependence ($\beta = 2$) of the SHG light. These

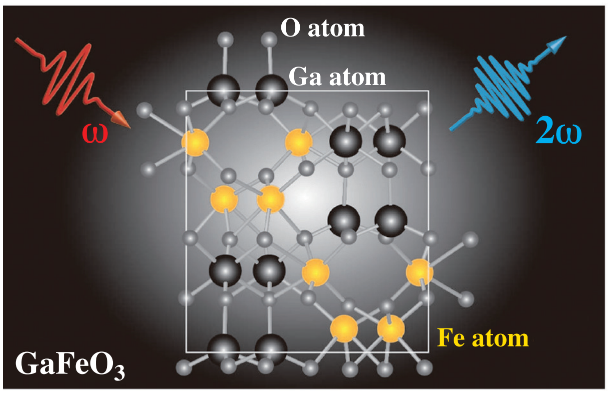

Fig. 1. Schematic drawing of second-harmonic generation in a $GaFeO_3$ crystal using Fe 3*p* resonance.

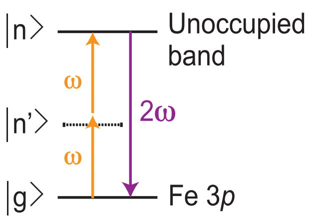

Fig. 2. Energy diagram of SHG in $GaFeO_3$. The notations g, n', and n represent the ground state, intermediate state, and excited state, respectively.

results unambiguously indicate the detection of SHG light from the $GaFeO_3$ crystal.

In summary, we demonstrated the possibility of element selectivity in SHG spectroscopy measurements in the soft X-ray range. We observed, for the first time, SHG in the reflected beam from a nonlinear crystal in the soft X-ray range. For a surface system, SHG in the soft X-ray region was observed at the XFEL facility of FERMI@ELETTRA in the transmission geometry [2]. The resonant SHG scheme is expected to promote new understanding of various samples, such as strongly correlated systems and the interfaces of heterojunctions in spintronics.

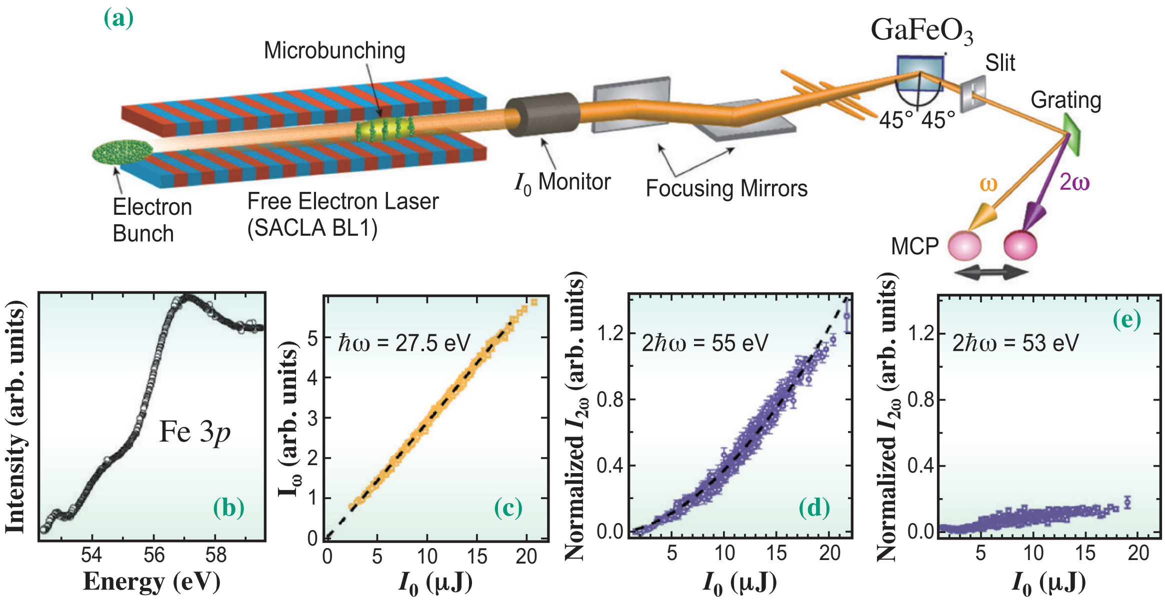

Fig. 3. **(a)** Schematic drawing of the SHG measurement at the SXFEL beamline in SACLA. **(b)** Fe 3*p* absorption spectrum of the $GaFeO_3$ crystal taken at BL-5B (UVSOR). **(c)** Intensity plot of the ω component, I_ω, of the reflected SXFEL pulses with respect to I_0 at the photon energy $\hbar\omega$ = 27.5 eV. **(d)** Plot of $I_{2\omega}$ at $2\hbar\omega$ = 55 eV. **(e)** Plot of $I_{2\omega}$ at $\hbar\omega$ = 53 eV. The dashed lines shown in (c) and (d) are the fitted curves.

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