Second harmonic generation at hard X-ray wavelengths

Second harmonic generation (SHG) is a nonlinear effect in which an intense wave at frequency ω interacts with matter to generate another wave at frequency 2ω. It can be viewed as a nonlinear diffraction effect where energy and momentum are conserved.

Nonlinear optical phenomena have been vastly investigated in the last decades at wavelengths ranging from the infrared to the ultraviolet regimes. In this spectral range, concepts and techniques based on nonlinear optics are implemented in many diverse fields. For example, laser technology, optical communication, ultrafast science, quantum optics, and nonlinear spectroscopy techniques.

Due to the weakness of nonlinear interactions in the hard X-ray regime and insufficient peak brightness of all X-ray sources prior to the X-ray free-electron lasers (XFELs), the only nonlinear effect in this regime that has been observed until very recently is spontaneous parametric down conversion [1]. This effect relies on the interaction of the input beam with the large fluctuations of the vacuum field at X-ray wavelengths. The detection scheme, which utilizes fast coincidence measurements, enables the measurement of very low count rates. XFELs provide peak brightness, which is more than 10^{10} times of that of third generation synchrotrons, thus open new possibilities for observations of new nonlinear phenomena. Indeed, the number of publications related to nonlinear X-ray effects is increasing rapidly (for example [2-3]). The nature of X-ray nonlinearities is vey different from conventional nonlinearities in many aspects, thus the observations of those effects are interesting and form a basis for new spectroscopy and diffraction techniques.

Nonlinear interactions at photons energies, which are much larger than the electronic resonances, are dominated by free-electron nonlinearities. In the particular case of X-ray SHG, the strong electric field of the pump at frequency ω drives an electron velocity that oscillates at ω. This velocity mixes via the Lorentz force with the magnetic field of the same pump, which also oscillates at ω, to generate nonlinear current density that oscillates at frequency 2ω. The transverse component of the current density gives rise to the second harmonic radiation. The direction of the generated radiation is determined by momentum conservation (phase-matching). At hard X-ray wavelengths, phase matching is achieved by using the periodic structure of the generator crystal. The phase matching diagram is shown in Fig. 1. The wave vectors of the pump and the second harmonic are $\vec{k}_\omega$, $\vec{K}_{2\omega}$ respectively, and $\vec{G}$ is the reciprocal lattice vector. θ_ω and $\theta_{2\omega}$ denote the angles between the atomic planes and the pump and the second harmonic beams respectively. The phase matching condition for a SHG process at X-ray wavelengths is $2\vec{k}_\omega+\vec{G}=\vec{K}_{2\omega}$.

The experiment was performed at beamline **BL3** of SACLA. The incident beam was a 7.3 keV monochromatic beam and the nonlinear medium was a diamond crystal [4]. The average pulse-energy after a Si (111) monochromator was about 1 μJ. The beam was focused by using Kirkpatrick-Baez mirrors to the about 1.5 microns full width at half maximum (FWHM). The phase matching was achieved by slightly detuning the crystal from the orientation for the elastic Laue geometry diffraction from the (220) planes for the second harmonic. The scattered photons were detected by a YAP:Ce scintillation detector (30% energy resolution).

Due to the small-expected efficiency, the main challenge of the experiment was the exclusion of the residual second harmonic that is generated in the XFEL from the SHG measured signal. The most convincing test to distinguish between the contributions of the elastic scattering of the residual second harmonic and of the SHG signal is the dependence of the count rates of the measured signal on the pulse-energy of the input beam. The dependence of the SHG count rates is expected to be quadratic, while elastic scattered signals are linear with the pulse-energy of the input beam. We used two approaches: i) we insert different thicknesses (0.025 mm, 0.1 mm, and 0.2 mm, and no filter) Al foils before

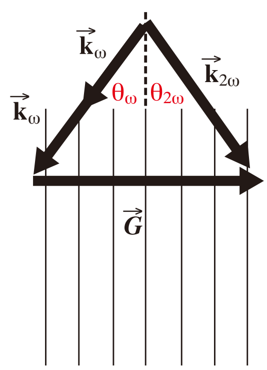

Fig 1. Phase matching diagram.

the diamond crystal and measured the count rates as a function of the average pulse-energy. The results are shown in Fig. 2(a). ii) We used the fluctuations of the XFEL pulse energy by building histograms of the number of pulses at a given pump pulse-energy. We scanned through the recorded data and counted the number of photons measured within each pump pulse-energy bin. We calculated the count rate by dividing the number of the second harmonic counts in each bin by the number of pulses with pulse-energy within the bin. The results are shown in Fig. 2(b). The inset shows the histogram of the pump-pulse energies. The results of the two approaches show clearly the quadratic dependence of the second harmonic signal on the pump, and agree within the experimental uncertainties.

Figure 3 shows the rocking curve of the SHG process, namely, the second harmonic count rate as a function of the angular deviation of the crystal from the phase matching angle. The Gaussian fit reveals that the FWHM of the rocking curve is about 180 μrad. The rocking curve confirms that the SHG is observed only in a narrow angular range near the phase matching angle.

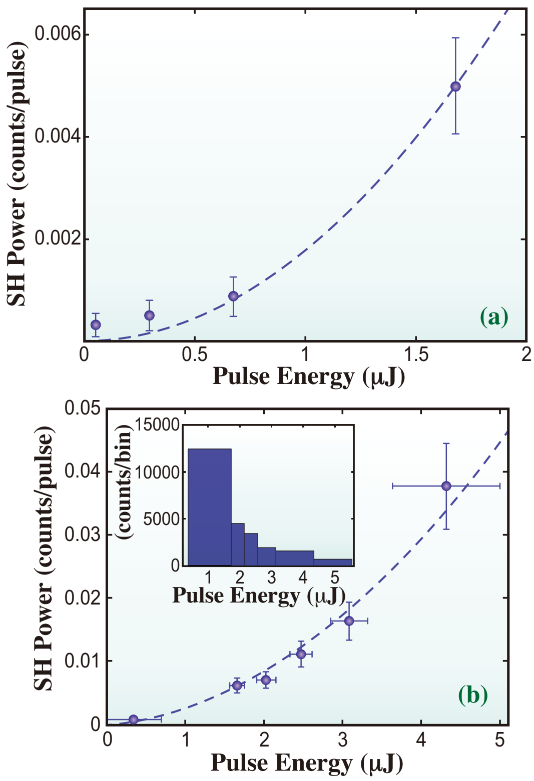

Fig. 2. Second harmonic count rate as a function of **(a)** Average pump pulse-energy where the pulse-energy is varied by inserting thin Al filters before the diamond crystal **(b)** The pulse-energy dependence is obtained from the pulse-energy fluctuations of the XFEL beam. The inset shows the energy fluctuations of the XFEL beam.

In conclusion, we have the observed the first SHG at hard X-ray wavelengths. As expected, the power of generated second harmonic scales quadratically with the pulse-energy of the pump, and the highest efficiency is obtained at the phase matching angle. The largest observed SHG efficiency was 5.8 ± 1.1 × 10^{-11} leading to a count rate that is more than ten times above the background. The observation of X-ray SHG opens possibilities for more general nonlinear studies at X-ray wavelen12 ptgths. For example, third harmonic generation, difference frequency generation, and the generation of quantum states of light via parametric down-conversion. SHG at X-ray wavelengths can be used in temporal correlation measurements for diagnostics of XFELs pulses on the femtosecond to sub-femtosecond scales and for nonlinear microscopy.

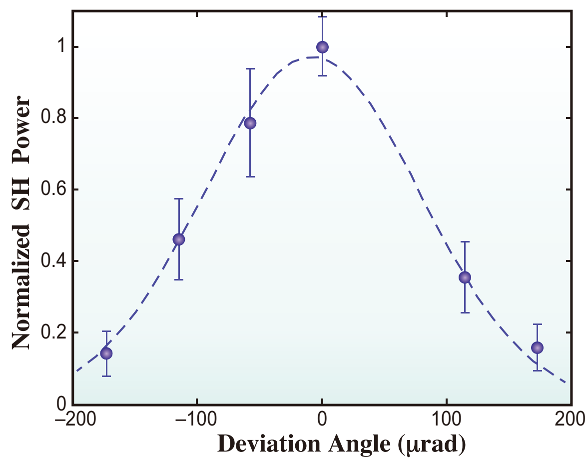

Fig. 3. Rocking curve of the second harmonic signal.

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References

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