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Angle-Resolved X-Ray Second Harmonic Generation in Diamond

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Until recently, the investigation of X-ray light matter interaction was limited by available X-ray intensities. These days, X-ray Free Electron Lasers (XFELS) allow for observation and examination of many non-linear effects in the X-ray regime. One of the most fundamental non-linear effects is Second Harmonic Generation (SHG). While the effect is well understood in the optical regime, the fundamental physics behind X-ray second harmonic generation (XSHG), which was first demonstrated in 2013, need further investigation.

In this experiment, we investigated XSHG in Diamond using different geometries. Ultra-short, highly intense X-ray pulses with a photon energy of 9.831 keV were generated by the Linac Coherent Light Source (LCLS). The efficiency of the investigated SHG process is extremely low. Only for pulse energies around 170 μJ, more than 10 second harmonic photons per pulse were demonstrated. For the experiment, a 2D pixel array detector was used that allows for single photon counting, even when multiple photons per shot reach the detector. An advanced interpretation algorithm was developed in the context of this work. It allows for separation of the overlapping signal from multiple photons under certain circumstances.

Here, for the first time, XSHG was demonstrated for several phase-matched scenarios such that the efficiency of the process could be resolved angularly. The results for the angular dependence of the efficiency are in good agreement with the theory as shown by simulations. Further, the quadratic dependence of the number of generated second harmonic photons on the incident pulse energy was verified and the rocking curve widths of the process were investigated.

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