Anomalous two-photon Compton scattering

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\textbf{Abstract}

X-ray free-electron lasers can generate radiation pulses with extreme peak intensities at short wavelengths. This enables the investigation of laser–matter interactions in a regime of high fields, yet at a non-relativistic ponderomotive potential, where ordinary rules of light–matter interaction may no longer apply and nonlinear processes are starting to become observable. Despite small cross-sections, first nonlinear effects in the hard x-ray regime have recently been observed in solid targets, including x-ray-optical sum-frequency generation (XSFG), x-ray second harmonic generation (XSHG) and two-photon Compton scattering (2PCS). Nonlinear interactions of bound electrons in the x-ray range are fundamentally different from those dominating at optical frequencies. Whereas in the optical regime nonlinearities are predominantly caused by anharmonicities of the atomic potential in the chemical bonds, x-ray nonlinearities far above atomic resonances are expected to be due to nonlinear oscillations of quasi-free electrons, including inner-shell atomic electrons. While the quasi-free-electron model agrees reasonably well with the experimental data for XSFG and XSHG, 2PCS measurements have led to unexpected results: the energy of the nonlinearly scattered photons from non-relativistic electrons shows a substantial unexpected red shift in addition to the Compton shift that is well beyond that predicted by a nonlinear quantum electrodynamics model for free electrons.

A potential explanation for the spectral broadening is based on a previously unexplored scattering process that involves the whole atom rather than just quasi-free electrons. A first simulation that includes the atomic binding potential was successful in describing a broadening of the spectrum of the nonlinearly scattered photons to longer wavelengths for soft x-rays. However, the same model does not show any broadening at hard x-ray wavelengths, which is in agreement with other simulation approaches. To this point no calculation has been able to reproduce the experimentally observed broadening.

Here we present further experimental data of 2PCS for an extended parameter range using additional diagnostics. In particular, we present measurements of the electron momentum distribution during the interaction that strongly suggest that the spectral broadening is not caused by an increased plasma temperature. We extend our measurement of the magnitude of the red shift in beryllium to >1.9 keV in addition to the Compton shift expected for free electrons and expand the measurement of the angular distribution to include forward scattering angles. We also present first measurements of 2PCS from diamond.
1. Introduction

Linear x-ray scattering is typically used to investigate matter on the atomic scale. In particular, elastic scattering is the foundation of structural analysis with atomic spatial resolution and (inelastic) Compton scattering can be used to probe the momentum distribution of electrons. Compton scattering describes the inelastic scattering process of a photon from an electron, which results in an energy transfer to the recoiling electron. For an initially non-relativistic electron, this leads to a decrease in energy of the scattered outgoing photon [1]. When the energy transfer during the scattering process is large compared to the atomic binding energies, linear Compton scattering in a solid is well described by the impulse approximation (IA), which assumes that the atomic binding potential does not change during the interaction and the electrons can be treated as quasi free [2].

At the peak intensities that can be generated by x-ray free-electron lasers (XFELs), first nonlinear scattering effects in the hard x-ray regime have recently been observed, including x-ray-optical sum-frequency generation (XSFG) [3], x-ray second harmonic generation (XSHG) [4] and two-photon Compton scattering (2PCS) [5]. Pulses of the Linac Coherent Light Source (LCLS) XFEL that are focused to a 100 nm spot size can generate a peak intensity of up to \(4 \times 10^{15} \text{ W cm}^{-2}\) at hard x-ray wavelengths [6]. This enables the investigation of nonlinear light–matter interactions in a largely unexplored regime of high fields, yet small ponderomotive potential due to the high photon energy. Despite peak electric fields of \(E_0 \approx 5 \times 10^{15} \text{ V cm}^{-1}\), which is within nearly four orders of magnitude of the quantum-electrodynamical (QED) critical field, at a photon energy of \(\hbar \omega = 9 \text{ keV}\), the normalized Lorentz-invariant field strength parameter is only \(\eta = eE_0/(m_e\omega) \approx 2 \times 10^{-3}\) and the ponderomotive potential is only \(U_p = \eta^2 m_e c^2/4 \approx 0.5 \text{ eV}\), where \(m_e\) is the electronic rest mass and \(c\) the vacuum speed of light. At such high intensities, nonlinear Compton scattering is observed [5]. This process involves the simultaneous scattering of two (or more) photons from an electron to generate higher energy photons that are red-shifted from the second (or higher) harmonic of the incident photons. In the IA and for non-relativistic electrons, we expect the spectrum of the scattered photons as a function of observation angle \(\theta\) for \(n\) scattering photons each with energy \(\hbar \omega\) to be centered around the free-electron case [7] (up to Doppler broadening due to the momentum distribution of the initially bound electron)

\[
\hbar \omega_n(\omega, \theta) = \frac{n \hbar \omega}{1 + \left(\frac{n \hbar \omega}{\omega_n^{(1)}}\right)^2 (1 - \cos \theta)}.
\]

While the free-electron approximation works reasonably well to describe XSFG and XSHG, 2PCS yielded an unexpected result that is not compatible with this model. In particular, Fuchs et al [5] observed that the spectrum of the generated 2PCS photons from the simultaneously nonlinear scattering of two 9.75 keV x-ray photons in a beryllium target was measured to extend to significantly lower photon energies than expected: the observed spectrum is red-shifted from 19.5 keV by at least 800 eV in addition to the nonlinear Compton shift of 700 eV predicted by equation (1) at an observation angle of \(\theta = 90^\circ\). This also takes into account the ground-state momentum distribution of the bound electrons in Be, as evidenced from the linear Compton scattering of the XFEL second harmonic at low intensities. Thus, the observed redshift is not compatible with the IA of electrons interacting as quasi free in solids during nonlinear hard x-ray—matter interactions far from atomic resonances. The additional energy redshift also cannot be explained by ponderomotive effects, as \(\eta \ll 1\) such that the ponderomotive potential is much smaller than the incoming photon energy (see equation (1)). A hot electron momentum distribution as cause of the energy shift was estimated to be highly unlikely but could not completely be ruled out in reference [5]. Here we present further experimental evidence, namely electron temperature measurements during the interaction, that strongly suggests that the shift is not due to a hot plasma generated by x-ray absorption. At the same time we extend the previous measurement to include both smaller scattering angles and the measurement of larger red-shifts of the outgoing photons.

A potential explanation for the anomalous red-shift involves an extension of the scattering process beyond the free-electron case that includes the dynamical interaction with the atomic binding potential. In particular, processes with virtual intermediate excited atomic and electronic states have been proposed [5, 8, 9]. Specifically, Hopersky et al [8, 9] used a second order perturbation approach to theoretically demonstrate a broadening of the outgoing photon spectrum to longer wavelengths for soft x-ray photons scattering in helium and neon. Krebs et al [10] used a nonperturbative time-dependent Schrödinger equation approach to account for both the atomic system and the scattered photon, which is treated as a quantized mode. Surprisingly, in their time-dependent QED calculations the dominant contributions to the two-photon scattering process is associated with third-order \(\mathbf{p} \cdot \mathbf{A}\)-type Feynman diagrams, rather than predominantly from second-order \(\mathbf{A}^2, \mathbf{p} \cdot \mathbf{A}\) diagrams (although both contributions involve the same order...
in A). Here $\hat{p}$ is the canonical momentum operator and $\hat{A}$ the vector potential operator. While calculations for soft x-ray wavelengths (500 eV) in helium show an extension of the scattered photon energies that exhibit an anomalous redshift, such effects are absent for higher incoming photon energies such as used in Fuchs et al [5]. In particular, the energy of the nonlinearly scattered photon for 4 keV incoming photons in He and for 9.7 keV in Be are in good agreement with the expectation for nonlinear scattering in the IA and fail to reproduce the experimental observations of reference [5]. More recently Venkatesh and Robicheaux theoretically investigated 2PCS using an effective time-independent, local potential to study the effect of bound electrons on the 2PCS spectrum, including the effect of the binding energy, electron–electron correlations and photoionization [11]. Similar to Krebs et al, they were unable to reproduce the anomalous redshift seen in the experiment.

In order to further investigate this effect and rule out trivial causes for the anomalous redshift we have performed a more detailed experimental study of hard x-ray 2PCS. In particular, we have extended the measurement of the angular distribution and with higher energy photons. We find that the observed emission pattern of 2PCS has a double-peaked shape with an asymmetry peaked at higher (backward) scattering angles. The asymmetry is increasing with increasing FEL intensity. While a quadrupole-like pattern is expected for a free-electron interaction [12], the position of the peaks and its minimum cannot be explained by a free-electron model. We have also extended the measured lower bound of the anomalous redshift to $>1.9$ keV on top of the expected Compton shift for free electrons. Additionally, we measure the electron momentum distribution during the scattering process using linear Compton scattering. An additional redshift of the nonlinearly scattered photon of 800 eV would require an extremely hot plasma temperature with an electronic temperature of $kT > 320$ eV and $kT > 760$ eV for a 1.9 keV shift. The spectrum of the linear Compton scattering does not show a significant broadening, which indicates that the there is no appreciable heating of the electrons ($kT < 10$ eV) during the interaction. The unexpected small temperature can likely be explained by the x-ray interaction generating nearly 10 keV photoelectrons that propagate outside of the 100 nm focus before depositing most of their energy. The experiment also includes a first measurement of 2PCS from a dielectric, namely diamond. Although we observe a 2PCS signal in diamond, we cannot draw a quantitative comparison to Be due to limited statistics.

2. Experiment

The experiment was conducted at the CXI end-station of the LCLS XFEL [13]. The beam line provides hard x-ray pulses (5–11 keV) at 120 Hz, with a total energy of up to 2 mJ per pulse and a pulse duration of approximately 50 fs. The experimental setup is similar to that of reference [5] with an additional linear Compton scattering diagnostic, an extension of the observable scattering angles to include forward scattering and an extension to higher FEL photon energies (see figure 1). For this experiment the fundamental x-ray photon energy was varied from 8.8 keV to 10.05 keV. A grazing-incidence Kirkpatrick–Baez (KB) mirror system was used to focus the x-rays to a spot diameter of approximately 100 nm, providing an intensity of $I \approx 4 \times 10^{20}$ W cm$^{-2}$. The on-target intensity was varied by inserting Si attenuators into the unfocused beam. The FEL pulse energy is measured for every shot. The Si filters mainly attenuate the FEL fundamental, while the FEL second harmonic is largely transmitted. The FEL second harmonic is largely suppressed before reaching the target by multiple grazing incidence reflections on the hard x-ray offset mirror system and the KB focusing mirrors. We measure that the on-target FEL second harmonic is well below the nonlinear 2PCS signal.

Two targets were placed directly in the beam path of the x-ray pulses, one in the proximity of the focus and another 1 m further downstream in the expanded beam. Since the transmission through the first Be target is relatively high (over 90% at 8.8 keV), this geometry allows the simultaneous observation of nonlinear scattering from the high-intensity interaction and a measurement of the background using the low-intensity interaction. A 250 μm-thick beryllium sample or a 300 μm-thick CVD diamond sample was placed in the high-intensity interaction region, with the target normal 60° from the incoming x-ray beam. Assuming a Gaussian beam, the Rayleigh range of the focused x-ray beam at 9 keV is $z_R = 228 \mu$m, which means that the effective sample thickness is approximately equal to the confocal parameter and that the FEL peak intensity only varies by a factor of 2 over the interaction length. The high-intensity target was translated to a fresh spot before every shot due to damage. A second 250 μm-thick beryllium sample was kept in the low-intensity region for an on-shot low-intensity reference.

The photons scattered from the targets were measured as a function of angle and energy using an arc of four pixelated hard x-ray CSPAD-140k [14] detectors in single-photon counting mode. Depending on the run configuration these detectors observed back-scattering from 90°–135° and/or forward-scattering from 45°–90°. Note that the linear polarization of the x-ray beam is in the detector scatter plane. Each detector
Figure 1. Experiment setup: the x-ray beam (green) enters from the left and passes through (optional) Si attenuators for controlling the on-target x-ray pulse energy. Grazing incidence KB mirrors focus the beam onto a Be or diamond target (gray) to generate a high-intensity interaction region. The generated scattering signal (blue) is measured as a function of scattering angle and photon energy using 2D pixelated x-ray detectors. A second target is placed further downstream to simultaneously measure the background at low-intensity. The detectors are located in an arc around each interaction point primarily in the FEL polarization plane. The detectors can be located to measure backward and/or forward scattering. They are shielded by Zr foils to sufficiently attenuate the fundamental scattering signal and to ensure operation in single-photon counting mode. A HAPG-crystal-based spectrometer in 110 degree backscattering geometry (pink) is used to measure the linear scattering signal of the target in the high-intensity region to determine the plasma temperature during the interaction.

has 140 000 pixels in an active area of approximately $4 \times 4$ cm$^2$. The detectors are arranged in a horizontal arc at a distance of 20 cm around the interaction point. Each detector captures an angle of 12.2 degrees horizontally (in the FEL polarization plane) and $\pm 5.8$ degrees out of the polarization plane. The detector’s coarse intrinsic energy resolution of a few keV allows us to distinguish between a single (lower-energy) photon near the FEL fundamental and a (higher-energy) photon near the second FEL harmonic. A zirconium filter was used directly in front of the detectors to ensure single-photon counting and to sufficiently reduce photon pile-up (i.e. two fundamental photons hitting the same pixel during a single shot, mimicking the response of a single double-energy photon) well below the nonlinear scattering signal. Depending on the detector position, either 250 $\mu$m or 300 $\mu$m thick Zr filters were used, which reduces the scatter of fundamental photons by a factor of over $10^{-7}$. More importantly, the transmission around the K-edge of the zirconium filters (17.995 keV) decreases from 0.05 just below to $10^{-7}$ just above the K-edge. We use the Zr K-edge absorption in combination with varying the FEL fundamental photon energy as an integrating spectrometer for the nonlinear signal. In particular, we can investigate the red-shift of the two-photon Compton events by tuning the incoming photon energy so that the energy of the scattered photon lies above or below the K-edge. We used a highly annealed pyrolytic graphite (HAPG) crystal spectrometer [15] in 110 degrees backscattering geometry to measure the (linear) scattering of FEL fundamental photons from the plasma generated by photo-ionization in the high-intensity target. This allows us to estimate the plasma temperature during the interaction from the width of the inelastic scattering component.

For each CSPAD detector, the pixel response was normalized using a flat-field signal from fluorescence of a copper target. Masking algorithms were employed to remove pixels from the analysis that had an erratic signal response (more than one standard deviation from the mean) under a series of dark events (no x-rays). Furthermore, regions of relatively high count rates (for example from elastic Bragg scattering from the poly-crystalline sample) were removed from the analysis to reduce the possibility of photon pile-up. For every FEL shot, we record the x-ray camera image and generate a photon-energy histogram of the sparse signal. The coarse energy resolution of the camera allows us to isolate the FEL fundamental scattering signal. We integrate the histogram over energies well above the FEL fundamental to obtain the high-photon-energy scattering signal. We also ensure that the pile-up rate calculated through Poisson statistics is well below the level of the nonlinear signal.

3. Results

We have performed a systematic measurement of the double-differential 2PCS scattering rates as a function of scattering angle and scattered photon energy. We use the Zr absorption edge as a bandpass-pass filter for
the nonlinearly scattered signal around 18 keV and obtain integrated spectral information through varying the incoming photon energy. We have measured the signal as a function of intensity by (i) varying the incoming x-ray pulse energy and by (ii) translating the sample through the focus at full intensity (Z-scan) to measure the interaction under a similar number of incoming photons. We have measured the scattering signal for Be and diamond samples. For an overview of the run configuration, see table 1.

### 3.1. Nonlinearity of 2PCS signal

We verify the nonlinear dependence of the 2PCS signal on the incoming x-ray intensity by varying the x-ray pulse energy through inserting Si attenuators in the unfocused beam. We also translate the sample through the x-ray focus at the highest FEL intensity to measure the interaction for different intensities at constant incoming pulse energy.

#### 3.1.1. Be target

For the Be target, we observe a clear 2PCS scattering well above background. The signal has a quadratic dependence on the incoming x-ray intensity for all detector angles. As an example, the integrated detector histogram signal around 18 keV for an FEL photon energy of 9.300 keV at an observation angle of $\theta = 135^\circ$ as a function of x-ray intensity can be seen in figure 2. Similarly, we observe a quadratic intensity dependence for the other FEL fundamental photon energies used in the experiment. The measured background in the low-intensity interaction is mainly due to the linear Compton scattering of the FEL second harmonic. It shows a small linearly increasing signal which is due to the nearly constant transmission through the Si attenuators at the FEL second harmonic photon energy. The 2PCS signal of the ‘z’ scan of the target through the focus also shows a clear dependence on the FEL intensity, while the background signal is nearly constant.

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Table 1. Run configurations used in experiment. Comma-separated values indicate the different FEL photon energies. “z” scan type indicates a scan of the sample in the FEL propagation direction in the proximity to the focus, “T” indicates FEL transmission scans. The detector configuration and the range of their observation angle is given in the last column.

| Photon energies (keV) | Scan type | Target | Scatter angles |
|-----------------------|-----------|--------|----------------|
| 8.8, 9.3, 9.8         | z, T      | Be     | 90°–135°       |
| 10.05                 | T         | Be     | 90°–135°       |
| 9.8                   | z, T      | Be     | 45°–90°        |
| 9.3, 9.8              | z, T      | Be     | 45°–90° & 90°–135° |
| 10.05                 | T         | Be     | 45°–90° & 90°–135° |
| 9.8                   | T         | Diamond | 45°–90°       |
3.1.2. Diamond target
We also observe a 2PCS signal in poly-crystalline CVD diamond that indicates a quadratic intensity dependence (see figure 3). The observed signal is more than 20 times above the expected pile-up signal. The ratio of the signal from the high-intensity interaction and the FEL fundamental shows an increase with intensity, which indicates a super-linear intensity dependence of the 2PCS signal (see figure 3(b)). We have significantly less data than for Be due to a limited size of the diamond sample. For this reason we have only measured 2PCS for an FEL photon energy of 9800 eV and with the detectors in forward scatter geometry. Compared to Be, the maximum signal from diamond is about a factor of 5 lower both considering an observation angle of 60 degrees.

3.2. 2PCS spectrum
We measure the 2PCS signal as a function of the incoming FEL fundamental photon energy. We can get information about the integrated spectrum from this in combination with the absorption above the Zr K-edge at $E_{Zr} = 17.995$ keV of the filters in front of the detectors. From equation (1) we expect that for a given observation angle, the scattered photons are completely absorbed by the Zr filters when their energy $\hbar \omega'_n$ is above the Zr K-edge. For the low-intensity region, we observe a decrease in signal as a function of scattering angle and photon energy as expected from equation (1) (see figure 4(b)). Since the signal also does not significantly change for different Si attenuator settings (which are nearly transparent for the FEL second harmonic), this background is mainly due to linear Compton scattering of the residual second FEL harmonic.

Unlike the scattering from the low-intensity region, for the high-intensity region we do not see a significant decrease in signal for even up to highest photon energy 10.05 keV (see figure 4(a)). In particular, we still observe a 2PCS signal at 45 degrees forward scattering at a fundamental photon energy of $\hbar \omega = 10.05$ keV, for which from equation (1) we would expect a 2PCS ($n = 2$) photon energy of $\hbar \omega'_2 = 19.87$ keV. For the signal to be transmitted through the Zr filter requires a total redshift of at least 2.06 keV from $2\hbar \omega = 20.1$ keV, which corresponds to a redshift of $\Delta \hbar \omega = \hbar \omega'_2 - E_{Zr} = 1.87$ keV in addition to the Compton shift expected for free electrons.

While we do not have data for diamond at these highest FEL fundamental photon energies, we observe a nonlinear signal for a fundamental photon energy of 9.8 keV, which also indicates a significant broadening to lower photon energies.

3.3. Target heating
We infer the electron temperature during the interaction from linear inelastic scattering of the FEL fundamental. In particular, we spectrally analyze linear Compton scattering in backscattering geometry at a scattering angle of 110 degrees. In this geometry the width of the inelastic peak is mainly given by the Doppler shift due to the motion of the electrons, from which an electron velocity distribution can be inferred [15].

The relative width of our measured linear Compton peak at full intensity is comparable to that measured in previous synchrotron experiments performed at low x-ray intensity [16]. A fit of the Compton
profile using the free-electron approximation [17] for the Be\textsuperscript{2+} states suggests a plasma temperature of $kT < 5.5$ eV taking into account a 60 eV instrument function. A fit using the XRS code, which takes into account inter-particle coupling and a partially degenerate electron subsystem [18] leads to an electron temperature of $kT < 8$ eV. These measurements are well below the temperatures required to explain the redshift of the nonlinear scattering profile by an increased electron momentum of the target ($kT > 760$ eV). The measured linear Compton profiles do not show significant broadening for increasing x-ray intensities as would be expected for an increasing temperature of the supra-thermal electron distribution (see figure 5). Specifically, the data does not show any evidence of heating from either the bulk electrons or a supra-thermal population. A possible explanation could be that the high-energetic photo-electrons rapidly move outside the focal volume before equilibrating and depositing most of their energy. A simulation indicates that only 0.3% of the photo-electron energy is deposited within the 100 nm focal spot radius. Note that for photon energies around 9 keV over 90% of x-rays are transmitted through the Be sample and the leading interaction is due to photo-electric absorption. While in a non-equilibrated sample there likely are high-energetic photo-electrons in the focus volume, this is only a small fraction of the total electrons that interact with the FEL pulse and the probability of nonlinear scattering from these electrons (i.e. a single-photon ionization process followed by a non-sequential two-photon scattering event) is exceedingly small. We cannot rule out a non-ideal x-ray focus spot with a significant fraction of the pulse energy outside the main focus spot, in which case the Compton profile would be dominated by low-intensity scattering from the focus wings. However, this is unlikely since it would indicate that only a small fraction of the x-ray pulse energy contributes to the nonlinear scattering signal.
3.4. Angular 2PCS distribution

We measure the angular 2PCS distribution ranging from 45° (forward scattering) to 135° (backscattering). The 2PCS angular distribution has an asymmetric double-peaked shape with peaks around 65° and 125° that is stronger in the backscattering direction (see figure 6). The measured angular distribution does not agree with the angular emission pattern for a second-order \((n = 2)\) emission of a free electron driven by a linearly polarized electromagnetic field. The second-order free-electron pattern has an asymmetric quadrupole-like shape peaked near 130° and a forward scattering peak near 40° with a smaller amplitude (see dashed line in figure 6) [12]. Note that a calculation of the nonlinear x-ray scattering pattern for a bound electron to a \(Z = 4\) charged atom is in general agreement with that for free electrons [11]. The backward–forward asymmetry in cross section for the 2PCS signal increases with increasing FEL intensity. The asymmetry can be seen for all FEL fundamental photon energies (i.e. 9300 eV, 9800 eV and 10 050 eV) but is less pronounced for higher photon energies. However, this could be due to the intensity-dependence as the FEL was only able to generate pulses with lower pulse energy at higher photon energies.

4. Discussion and conclusion

The experimental results described here are an extension of the first measurement of 2PCS that has led to the observation of an unexpected anomalous red-shift in the energy of the outgoing photon [5]. 2PCS is a nonlinear light–matter interaction in a parameter regime of high electric fields yet low ponderomotive potential. We observe a clear signal well above background that depends quadratically on intensity. We have measured the dependence of the double-differential cross section on intensity and on incoming photon energy ranging from 8.8–10.05 keV. Our data extends the previous measurement of the anomalous red-shift in photon energy of the nonlinearly scattered photons from more than 800 eV to more than 1.9 keV in addition to the nonlinear Compton shift expected for free electrons and well beyond the bound-state Compton broadening measured for linear Compton scattering of the FEL second harmonic. We have also measured the electron temperature of the sample during the interaction. The measurement indicates that it is highly unlikely that the electron temperature in the sample is sufficiently high to explain the broadening. We have extended the measurement of the angular 2PCS distribution to also include forward scattering angles. The measured distribution has an asymmetric double-peaked shape, which is stronger in backscattering direction. The asymmetry increases with increasing intensity. The shape of the distribution and the location of the peaks do not agree with those of nonlinear scattering from a free electron [12], which is similar to that simulated for nonlinear scattering from bound electrons [11]. We have performed a first measurement of 2PCS in a dielectric, namely diamond. We also measure an anomalous red-shift in this case. However, due to limited data we cannot more quantitatively compare the nonlinear scattering in diamond to Be.

Although first attempts to explain the anomalous red-shift have been made, the detailed mechanism of this fundamental nonlinear processes is still not fully understood. Further investigations of the interaction are still required. This includes additional measurements at largely different wavelengths, in different materials and in different states of matter, such as liquids and gases. A complete measurement that has the ability to observe not only the nonlinearly scattered photons but also the momentum distribution of the scattered electrons and ions involved in the process will be able to give invaluable insights into the underlying physics.
A full understanding of this process will not only be beneficial for our fundamental understanding but also for potential future nonlinear x-ray applications of as those were so far largely prevented due to the small efficiencies of x-ray nonlinear processes.

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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