Enhanced soft X-ray high-harmonic generation driven by two-color ($\omega+3\omega$) mid-IR laser pulses

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Abstract: We demonstrate the efficiency enhancement of soft X-ray high-harmonic generation from Ar in the range of 40-200 eV using two-color ($\omega+3\omega$) mid-infrared pulses with relative phase control, which is in qualitative agreement with 3D simulations.

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Mid-infrared (mid-IR) laser-driven high harmonic generation (HHG) has been proven to be a reliable way of producing coherent soft X-ray attosecond pulses in the water-window (280-540 eV) [1-3] and up to the keV regions [4] thanks to the cutoff energy scaling with λ^2 . The downside is the unfavorable wavelength scaling of single-atom HHG efficiency, approximately proportional to $\lambda^{-(5-7)}$. High gas pressure in the interaction region is needed to overcome the poor single-atom efficiency and obtain a reasonable phase matching, making differential pumping and minimal reabsorption in the HHG setup critical to the output soft X-ray flux and therefore demanding applications like spectroscopy and imaging. Two-color-driven HHG has shown a great potential for enhancing the efficiency in the EUV range (<100 eV) [5], but the experimental study on its efficiency has rarely been pursued in the soft X-ray range [6], where the efficiency greatly suffers. There have been recent investigations on the optimal waveform for HHG in terms of efficiency and isolated attosecond pulse generation [7-9]. Simulation studies have revealed that the two-color waveform made by mixing the fundamental (ω) and its third harmonic (3ω) is an optimal choice for dramatically enhancing the HHG efficiency due to the maximal return of short-trajectory electrons [9]. In this Contribution, we demonstrate the efficiency enhancement of soft X-ray HHG from Ar in the range of 40-200 eV using two-color ($\omega+3\omega$) mid-IR pulses with relative phase control and study the effect of gas pressure.

For the two-color experiment, the 1.7 mJ, 2.1 μm , ~30 fs output pulses from an optical parametric chirped pulse amplifier (OPCPA) [10] are frequency tripled via two-stage in-line frequency conversion based on second-harmonic generation (2ω) followed by type-II sum-frequency mixing of ω and 2ω in BBO crystals. The conversion efficiency to 3ω (700 nm) was as high as ~30%. To achieve the relative phase control, the two colors were separated and recombined using a dichroic mirror (HR@700 nm and HT@2.1 μm). In the 3ω arm of the interferometer, the residual ω beam was blocked by an ITO plate. In the ω arm, the delay was finely controlled using a piezo-stage with sub-cycle resolution. Due to the transmission and reflection loss, the available energies of the 3ω and ω pulses are ~65 μJ and ~700 μJ for HHG. The duration of the 3ω pulse is estimated as ~26 fs accounting for the dispersion of optics (~20 fs transform limit). Both beams were focused with ~65 μm waist diameter into a ~5 mm-long gas cell filled with Ar at 200 mbar and 400 mbar, respectively. The temporal and spatial overlap between ω and 3ω pulses were ensured by monitoring the 5ω beam at 420 nm via the four-wave mixing ($\omega+\omega+3\omega$) in the air plasmas. The relative delay, *i.e.*, relative phase, of the ω and 3ω pulses is scanned at around the zero delay position of the piezo stage. An XUV spectrometer and two photodiodes with Al and TiC_2 coating were used to detect the HHG spectra and photon flux, respectively. Either 0.5- μm -thick Al or Zr filter was used to block the optical beams.

We first optimized the 2.1 μm (ω) driven HHG signal by monitoring the photodiode signal and then added 3ω pulses to observe the enhancement of the HHG signal. The peak intensity of the ω and 3ω pulses were $\sim 2 \times 10^{14} \text{ W/cm}^2$ and $\sim 2 \times 10^{13} \text{ W/cm}^2$, respectively. Therefore, there is no HHG with the 3ω pulse alone. Fig. 1(a) shows the measured photo-current in the Al window (20-70 eV) with regard to the relative delay between ω and 3ω pulses at 400 mbar of Ar. The single-color HHG signal is ~5 pA as shown by dotted background line and the ~3x enhancement is observed clearly depending on the relative delay with the period of ~700 nm (3ω), indicating the sub-cycle effect of the driving

mid-IR waveform. Figure 1(b)-(d) show the single- and two-color HHG spectra in the different photon energy ranges covering the low-energy (b), plateau (c) and cutoff (d) regions. For the two-color cases, we compared the spectra having various relative phases. The $\sim 3\times$ enhancement in the low-energy (red curve in Fig. 1(b)) is consistent to Fig. 1(a), while the factor is reduced to ~ 2 and ~ 1.5 in the plateau and cutoff regions, as found in Figs. 1(c) and (d). With an optimal delay position we also observed a mild extension of the cutoff energy to ~ 210 eV, which is ultimately limited by the Zr filter transmission, as shown by the purple curve of the inset of Fig. 1(d) in log-scale.

Our 3D simulations in Figs. 1(e) and (f), plotted in log-scale, show a qualitative agreement with our experimental observations. The HHG enhancement with addition of the 3ω pulse is less dramatic at the plateau and cutoff regions than in the low-energy region, as indicated by Fig. 1(e). The pulse propagation effect in the relatively high pressure (400 mbar) degrades the two-color waveform such that the phase matching becomes more challenging for higher harmonics. Fig. 1(f) clearly shows that the relative phase control is critical to the efficiency enhancement of HHG.

Fig. 1: (a) Measured HHG photo current in 20-70 eV vs. relative delay between the ω and 3ω pulses. The ω pulse proceeds in a negative delay. (b)-(d) HHG spectra with low-energy, plateau, and cutoff regions. Black curves represent HHG spectrum with single color. The relative phase (converted from relative delay) is specified for each of two-color-driven HHG spectra. ϕ_1 and ϕ_2 are constant phase defined at a reference position of the piezo-stage. (e) Simulated HHG spectra with a single color and two colors. (f) Simulated HHG spectra with different relative phases.

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