Supplementary Information for:
Route to optimal generation of soft X-ray high harmonics with synthesized two-color laser pulses

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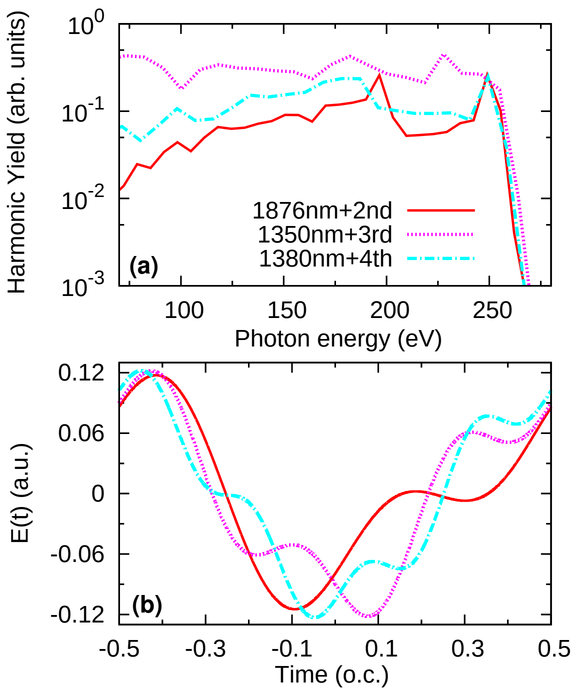

Supplementary Figure S1: Comparison of harmonics and waveforms optimally synthesized by using the fundamental and its 2nd, 3rd and 4th harmonics, respectively. (a) Single-atom HHG spectra of Ne by using three waveforms as shown in (b), showing that yields with 3rd harmonic as the supplementary is the highest. The cutoff energy was fixed at 250 eV. The spectra are smoothed by a Beizer curve [1]. Laser parameters are from Supplementary Table S1. (o.c. means optical cycle of the fundamental laser in the waveform.)

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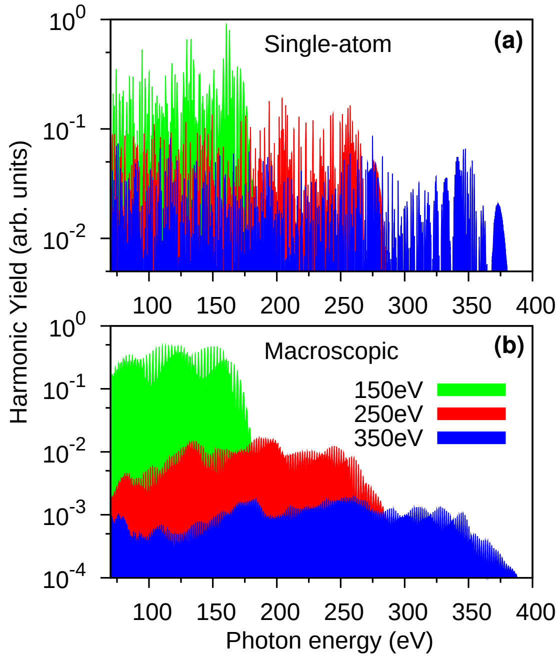
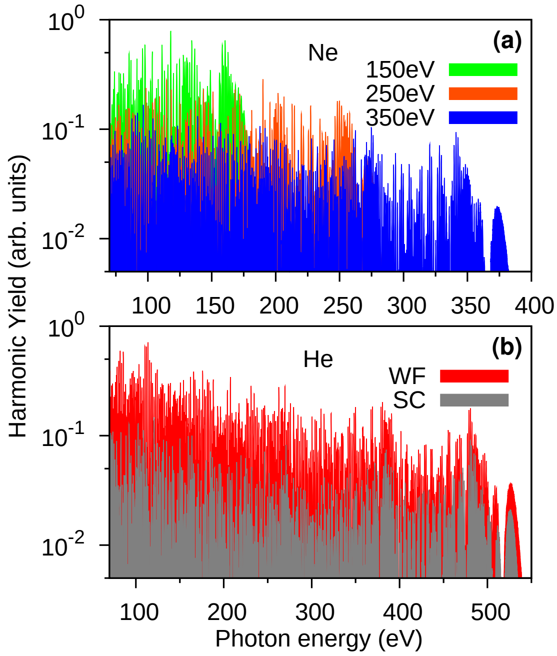
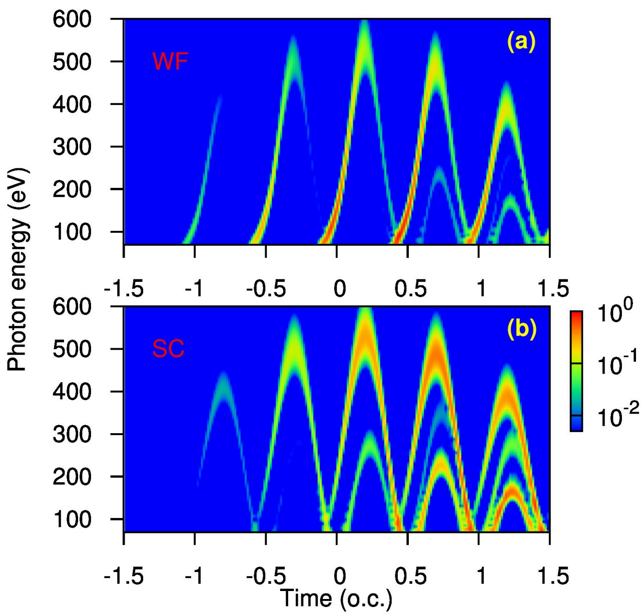


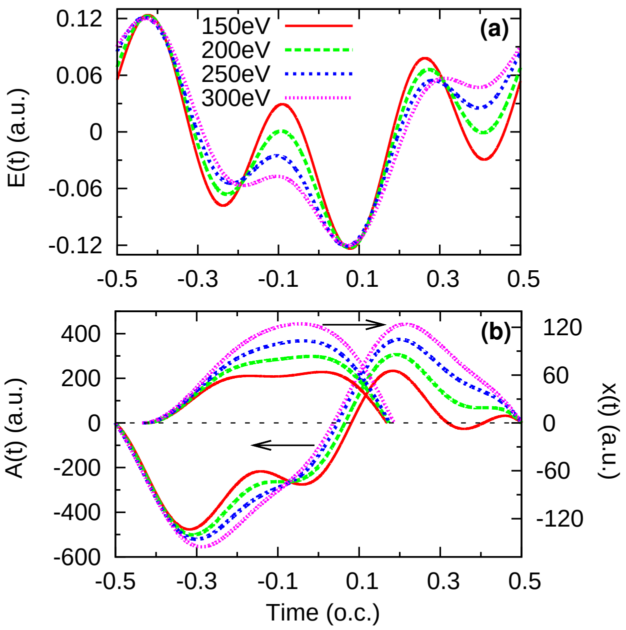
Supplementary Figure S2: Harmonic spectra of optimized single-color fields. (a) Single-atom and (b) macroscopic HHG spectra of Ne generated for three different cutoff energies. The corresponding maximum returning electron energies are 150, 250 and 350 eV, respectively. Laser parameters are from Table 2.



Supplementary Figure S3: Single-atom harmonic spectra of two-color waveforms. (a) Single-atom HHG spectra of Ne by using optimized waveforms to generate three different cutoff energies. The corresponding maximum returning electron energies are 150, 250 and 350 eV, respectively. Laser parameters are from Table 2. (b) Comparison of single-atom HHG spectra of He by using waveform (WF) and single-color (SC) wave. The maximum returning electron energy is 500 eV. Laser parameters are from Table 3.



Supplementary Figure S4: Time-frequency analysis of harmonics in Supplementary Fig. S3(b). (a) Waveform (WF) and (b) single-color (SC) wave. These figures show the relative strength of harmonic emission from "short"- and "long"-trajectory electrons. (o.c. stands for optical cycle of the fundamental laser, i.e., 4.74 fs for WF and 4.92 fs for SC.)



Supplementary Figure S5: Analysis of the the classical motion of an electron in two-color field that results in different cutoff energies. (a) The waveforms shown in Fig. 4(b) of the main text. Laser parameters are from Table 4. (b) The corresponding vector potential $A(t)$ and displacement $x(t)$ of the electron for electrons that return with the maximum energies. (o.c. stands for optical cycle of the 1600-nm laser.)

Supplementary Table S1: Optimized laser parameters for waveforms giving the cutoff energy of 250 eV. In the optimization, the second color is chosen as 2nd, 3rd or 4th harmonic, respectively, and $\phi_1 = 0$. Laser wavelength (λ_1) is given in nm, and peak intensities ($|E_1|^2$ and $|E_2|^2$) in 10^{14} W cm^{-2}. Target: Ne, $I_p = 21.56$ eV.

Combination	λ_1	$\lvert E_1\rvert^2$	$\lvert E_2\rvert^2$	ϕ_2
$\lambda_1 + \lambda_1/2$	1876	2.40	0.92	0.52π
$\lambda_1 + \lambda_1/3$	1350	3.50	0.46	1.41π
$\lambda_1 + \lambda_1/4$	1380	3.78	0.22	0.48π

Supplementary Note

S1. Wavelength scaling of fundamental laser assisted by its 3rd harmonic

Following Eq. (1) in the main text (MT), we search laser parameters of the optimized waveform using genetic algorithm (GA) by D. L. Carroll (FORTRAN genetic algorithm driver, version 1.7a, 2001, available at http://cuaerospace.com/carroll/ga.html). The ionization level of waveform in a cycle of the fundamental is set at 2%, and the 3rd harmonic is served as its complementary laser. Note that the 3rd harmonic is the best combination to efficiently produce high harmonics (shown in Sec. S2). For each cutoff energy and each target, we optimize the returning electron wave packet of the cutoff harmonic with constrains elaborated in the main text.

We showed laser parameters for the optimized waveforms extending harmonic cutoff energy from about 200 to 1000 eV using Ar, Ne and He as the targets, in Tables 1, 2 and 3, respectively.

Note that in this Supplementary Information (SI), a realistic Gaussian envelope with the full width at half maximum (FWHM) of 3 cycles of the fundamental laser is always applied for the single-color wave and the optimized waveform when we present either single-atom or macroscopic HHG spectrum. The single-atom induced dipole is calculated by quantitative rescattering (QRS) model [2–4].

S2. Waveforms synthesized by the fundamental laser with its 2nd and 4th harmonics

We have shown above the waveform synthesized using the fundamental laser and its 3rd harmonic. We can also synthesize the fundamental and its 2nd or 4th harmonic for this purpose, while the wavelength of the fundamental laser is optimized. Laser parameters for waveforms of the three combinations are listed in Supplementary Table S1. Among the three, combination of the fundamental and its 3rd harmonic has the smallest pulse energy and fundamental wavelength. The three waveforms are plotted in Supplementary Fig. S1(b). Except for peak structures around -0.2 o.c., they differ from each other in other parts. Note that the peak structure which is very common in our waveform can alter relative ionization rates of “short”- and “long”-trajectory electrons. We show single-atom HHG spectra of Ne by using three waveforms in Supplementary Fig. S1(a). The combination of a 1350-nm laser and its 3rd harmonic gives the strongest harmonic yields covering the photon energies from 70 to 270 eV. Our previous paper [5] has demonstrated that the combination of a 1600-nm laser and its 3rd harmonic could give the best harmonic yield in comparison with the combinations of the 1600-nm laser and any wavelength for the second laser. In a word, the most efficient combination for two-color waveform synthesis is the fundamental laser and its 3rd harmonic. This conclusion has been applied to obtain Fig. 1 of MT where the number of optimized parameters was reduced from 5 to 4.

S3. HHG spectra of Ne with optimized single-color waves

In Supplementary Fig. S2, we compare single-atom and macroscopic HHG spectra of Ne using optimized single-color (SC) waves. These spectra are generated by the SC waves with wavelengths (λ_1) of 1012, 1350 and 1611 nm (in order of increasing cutoff energy). For macroscopic calculations, laser beam waist w_0 is fixed as 50 μm, gas jet (1-mm long) is put at $z_R/2$ after the focus where $z_R = \pi w_0^2/\lambda_1$ is the Rayleigh range of the laser. Gas pressure with uniform distribution in the jet is 10 Torr. Gas jets are put at 3.9, 2.9 and 2.4 mm after the focus, respectively. This figure shows that with increasing cutoff energy the harmonic yield in the cutoff drops fast in the single-atom response, and it drops even much faster after macroscopic propagation. The drop of harmonic yield in the cutoff is very similar to the two-color optimized case, see Fig. 3(a) and Supplementary Fig. S3(a).

S4. Single-atom HHG with synthesized waveforms

In Supplementary Fig. S3(a), we compare single-atom HHG spectra of Ne for three different cutoff energies. These spectra are generated by the waveforms with fundamental wavelengths of 1075, 1367 and 1625 nm (in order of increasing cutoff energy). The corresponding macroscopic HHG spectra are shown in Fig. 3(a) of the MT. This figure indicates that longer wavelength is required to reach higher-energy photons. However, high harmonics after propagation scale even worse compared to single-atom response.

We compare single-atom HHG spectra of He using the waveform and single-color sinusoidal wave at the same ionization level in Supplementary Fig. S3(b). The fundamental wavelength in the waveform is 1422 nm and it is 1477 nm in the single-color wave. The corresponding macroscopic HHG spectra are shown in Fig. 3(b) of the MT. Supplementary Fig. S3(b) and Fig. 3(b) show that harmonic yield of the WF is slightly stronger than that of SC in single-atom response, it is much stronger than SC after macroscopic propagation. This is achieved by only adding a small amount of 3rd harmonic, whose intensity is about 5% of the fundamental laser.

Time-frequency pictures of single-atom harmonics in Supplementary Fig. S3(b) are shown in Supplementary Fig. S4. The "short"-trajectory electron emissions are much stronger than the "long"-trajectory ones in the two-color waveform while the opposite behaviors occur in the SC. This figure also indicates that ionizations of "short"- and "long"-trajectory electrons can be greatly altered by adding a small portion of another color. This is because ionization takes place in a very narrow time window.

S5. Waveforms synthesized by using 1600- and 533-nm lasers

The optimized waveforms used in Fig. 4 of MT are again plotted in Supplementary Fig. S5(a), and the corresponding vector potentials are plotted in Supplementary Fig. S5(b). (Vector potential is related to laser electric field by $E(t) = -(1/c)\partial A(t)/\partial t$, where c is the speed of light.) The classical trajectories for electrons with maximum returning energies are also shown in Supplementary Fig. S5(b). The peak in the waveform around -0.4 o.c. [see Supplementary Fig. S5(a)] is essentially the same for the four curves because the relative phase of the optimized wave is stable, thus ensuring that electrons corresponding to cutoff emissions are ionized at at the same time. Once into the laser field, they experience quite different electric fields from the different synthesized laser, see the waveforms near -0.1 o.c. in Supplementary Fig. S5(a). It is even more clearly seen from the vector potentials near -0.1 o.c. where high cutoff energy is related to high vector potential at the return time, see Supplementary Fig. S5(b).

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