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Single-attosecond pulse generation using a seed harmonic pulse train

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We propose a method to generate a single-attosecond soft-x-ray or extreme ultraviolet pulse by use of two infrared (IR) laser pulses of different wavelengths, neither of which alone is sufficiently short for single-pulse generation. Our simulations based on the time-dependent Schrödinger equation show that, when a harmonic pulse train generated by one IR laser field is superposed with the other driving laser and applied to a target atom, only one pulse in the train significantly boosts harmonic generation and acts as an attosecond enhancement gate for isolated pulse generation.

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The recent progress in the high-order harmonic generation (HHG) technique [1] has enabled the production of ultrashort high-power coherent soft-x-ray (SX) and extreme ultraviolet (XUV) pulses [2–4] and raised significant interest in the generation of attosecond pulses. A single-attosecond pulse (SAP), in particular, is critical for the study of the electron dynamics inside atoms [5]. The general feature of HHG can be explained by a three-step semiclassical model [6]. According to this model, an electron is lifted to the continuum with no kinetic energy, the subsequent motion is governed classically by an oscillating electric field, and the electron recombines with its parent ion with a certain probability to generate an attosecond light burst. This process is repeated every half-cycle of the laser optical field and produces an attosecond pulse train.

If the driving laser pulse is sufficiently short that the effective HHG takes place only within one half-cycle, the cutoff region of the spectrum may become a continuum, corresponding to a single recollision. The first SAP's [7–9] were obtained on this basis. Kienberger *et al.* [9] generated single pulses (photon energy 93.5 eV) with a duration down to 250 as using a few-cycle laser pulse with a duration of 5 fs, a carrier wavelength of 750 nm, and a carrier-envelope phase (CEP) controlled in such a way that the pulse has a cosine wave form. Sekikawa *et al.* [10] obtained a single-subfemtosecond pulse as the ninth harmonic generated by a 8.3-fs blue laser pulse (photon energy 3.1 eV). Although the pulse is longer than in the previous case, they used a sufficiently high peak intensity of 5.5×10^{14} W/cm^2 to ionize nearly all atoms before the pulse peak. Thus, HHG was quickly shut off (*ionization shutter*), which enabled single-pulse generation.

Alternative schemes have also been proposed [11–15]. In particular, using a pulse with a time-dependent ellipticity constructed by two time-delayed countercircularly polarized pulses, we would expect that HHG takes place only at the center portion of the composed pulse, nearly linearly polarized (*polarization gate*), thus leading to SAP generation [11–13]. The generation of isolated 130-as pulses has been reported very recently [16,17]. A drawback is, however, that the center portion of the pulse is necessarily less intense than the preceding circularly polarized part, which may ionize a significant fraction of atoms before HHG occurs. SAP generation by adding a weak second-harmonic field [18] has recently been proposed, and generation of XUV continuum radiation driven by a sub-10-fs two-color field has recently been reported [19].

Since the methods used in Refs. [9,10,17] require a high-power sub-10-fs (typically 5-fs) laser pulse with CEP and/or polarization control, demonstrated SAP generation is still virtually limited to these authors.

In this Rapid Communication, we propose a scheme of SAP generation using multicycle laser pulses, based on enhanced HHG [20–22], by simultaneous irradiation of driving laser and seed harmonic pulses: namely, *attosecond enhancement gate for isolated pulse generation* (AEGIS). Let us first describe AEGIS qualitatively [Figs. 1(a) and 2(a)]. The theoretical study in Refs. [20,21] has revealed that the addition of a seed XUV or SX pulse of a photon energy smaller than the ionization threshold can enhance HHG efficiency by many orders of magnitude. Such a dramatic enhancement has recently been demonstrated experimentally [23–25]. The underlying mechanism [20,21] is that the seed pulse induces a transition to real (harmonic generation from a coherent superposition of states [26,27]) or virtual (two-color frequency mixing) excited levels, facilitating optical-field ionization (OFI), the first step of the three-step model. When the seed is composed of a train of attosecond pulses, this can be viewed as repeated attosecond enhancement gates. Let us consider that the fundamental pulse generating the seed harmonic pulse, referred to as the *seed fundamental* pulse hereafter, and the driving laser pulse which will be combined with the seed pulse have different wavelengths. For example, when the seed fundamental wavelength λ_{sf} and the driving wavelength λ_d are 800 nm and 2.1 μm, respectively, a seed harmonic pulse train even composed of several attosecond

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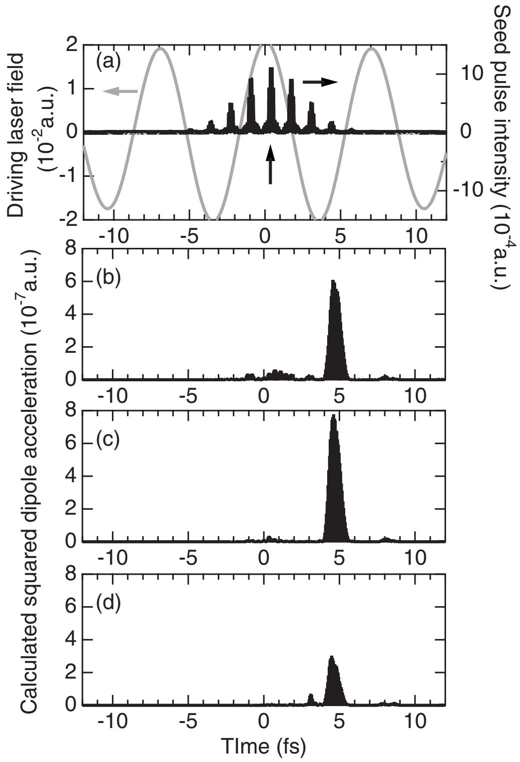

FIG. 1. Soft-x-ray pulse generation by the combination of $\lambda_d=2.1$ μm and $\lambda_{sf}=800$ nm. (a) Temporal profile of the seed harmonic pulse intensity (black, right axis) with a global pulse width of 5 fs and the driving laser field (gray, left axis) with a pulse duration of 30 fs. The vertical arrow indicates the pulse expected to act as a gate to enhance HHG. (b) Temporal profile of the calculated squared dipole acceleration (SDA), proportional to the generated pulse intensity, around 30 nm wavelength. (c) SDA when the seed pulse train has a global pulse width of 3 fs. (d) SDA when, in addition, the delay of the seed pulse train is a quarter cycle of the driving field.

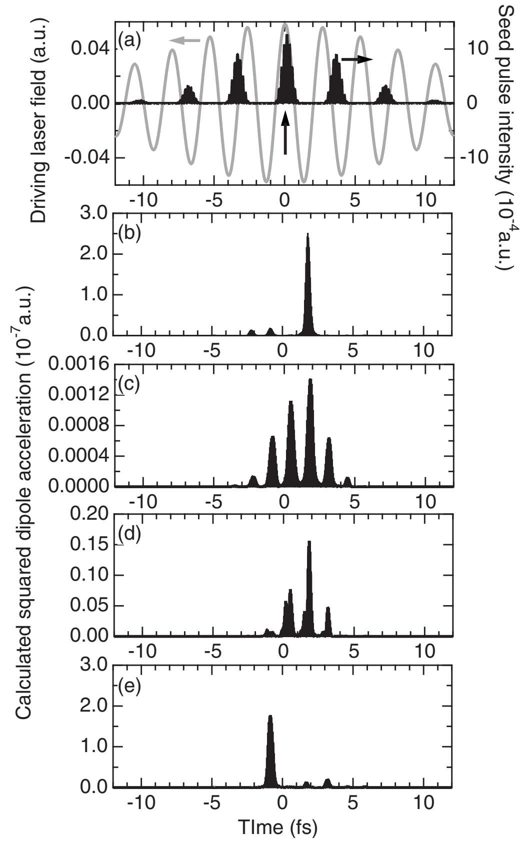

FIG. 2. Soft-x-ray pulse generation by the combination of $\lambda_d=800$ nm and $\lambda_{sf}=2.1$ μm. (a) Temporal profile of the seed harmonic pulse intensity (black, right axis) with a global pulse width of 10 fs and the driving laser field (gray, left axis) with a pulse duration of 15 fs. The vertical arrow indicates the pulse expected to act as a gate to enhance HHG. (b) Temporal profile of the calculated SDA, proportional to the generated pulse intensity, around 24 nm wavelength. (c) SDA in the absence of the seed pulse. (d) SDA when $\lambda_{sf}=800$ nm. (e) SDA when the delay of the seed pulse train ($\lambda_{sf}=2.1$ μm) is a quarter cycle of the driving field.

pulses repeated every 1.33 fs is confined within one cycle (7 fs) of the driving laser, as is schematically shown in Fig. 1(a). Hence, we would expect that the enhanced harmonic emission forms a SAP. When $\lambda_{sf}=2.1$ μm and $\lambda_d=800$ nm, conversely, seed harmonic pulses are separated by 3.5 fs, which is longer than the driving laser cycle (2.67 fs). If the seed harmonic pulse train and the driving pulse are superposed as shown in Fig. 2(a), only the central pulse would contribute to HHG in the cutoff region, resulting in SAP generation.

Furthermore, the kinetic energy of the recombining electron and, thus, the emitted photon energy in the three-step model depends on the electron's time of release, and that, in particular, an electron ionized by tunneling at $\omega t=\phi_c,\phi_c+180°,\ldots$ ($\phi_c\approx 17°$), around the pulse peak, contributes to cutoff emission [6]. Therefore, when only one pulse of the seed pulse train is adjusted to satisfy this relation, marked by vertical arrows in Figs. 1(a) and 2(a), only that pulse could contribute to harmonic emission in the cutoff region, which would further favor SAP generation, even though the driving pulse is relatively long and the seed contains multiple pulses. Although, independently of Refs. [20,21], Schafer *et al.* [28–30] have reported an increase and control of HHG through selection of a single quantum path using attosecond pulse trains, we would like to stress the important difference of the present idea in terms of SAP's that the driving field and the seed pulse train have different periodicity and that we mainly consider seed pulses of photon energy below the ionization threshold [20,21].

We now confirm this qualitative idea, using direct numerical solution of the time-dependent Schrödinger equation (TDSE) in the length gauge,

$$i\frac{\partial\psi(\mathbf{r},t)}{\partial t}=\left[-\frac{1}{2}\nabla^2+V(r)+z[E_d(t)+E_s(t)]\right]\psi(\mathbf{r},t), \quad (1)$$

for a model atom in the single-active-electron approximation, represented by an effective potential [31],

$$V(r)=-[1+Ae^{-r}+(Z-1-A)e^{-Br}]/r, \quad (2)$$

where Z denotes the atomic number. For Ne, we use a grid spacing of 0.25 a.u. and parameters $Z=10$, $A=4.341$, and B

=58.75, which faithfully reproduce the eigenenergies of the ground and first excited states. The harmonic spectrum is calculated by Fourier transforming the dipole acceleration, then filtering as is done in experiments with multilayer mirrors [32], and transforming back into the time domain to yield the temporal structure of the pulse radiated from the atom.

Let us first consider the case where $\lambda_{sf}=800$ nm and $\lambda_d=2.1$ μm. Figure 1(a) displays the seed harmonic pulse train used in the present simulation. The pulse train

$$E_s(t) = E_{s0}(t-\phi_c/\omega_d)\sum_{q(\mathrm{odd})=11}^{19} f_q \cos[q\omega_{sf}(t-\phi_c/\omega_d)] \quad (3)$$

is composed of the 11th–19th harmonics. We use experimentally observed values [3] for the harmonic mixing ratio $(f_{11}^2, f_{13}^2, f_{15}^2, f_{17}^2, f_{19}^2)=(0.50, 0.34, 0.07, 0.04, 0.05)$, and the common amplitude envelope $E_{s0}(t-\phi_c/\omega_d)$ is assumed to be of a Gaussian temporal profile centered at $t=\phi_c/\omega_d$ with a full width at half maximum (FWHM) of 5 fs, referred to as a global pulse width hereafter. The sum of the peak intensity of each component is 10^{13} W/cm^2. Such a train of ca. 7 pulses would typically be generated by applying a Ti:sapphire laser pulse of a duration $\approx$15 fs to a Xe gas, and even higher intensity has experimentally been realized using 20-mJ laser pulses [2–4]. The intensity of the resulting harmonic pulse is proportional to that of the seed harmonic pulse, but its relative temporal structure is not affected by the latter. The driving pulse $E_d(t)$, also shown in Fig. 1(a), is assumed to have a Gaussian temporal intensity profile centered at $t=0$ with a FWHM of 30 fs. Even shorter pulses (20 fs) with a stable CEP at 2.1 μm have recently been reported, based on optical parametric chirped-pulse amplification [32]. The peak intensity I_d is 1.5×10^{13} W/cm^2. Figure 1(b) presents the calculated squared dipole acceleration (SDA), which is proportional to the intensity of the pulse radiated from a Ne atom subject to the seed harmonic and the driving pulses simultaneously, as would be obtained after reflected by a multilayer x-ray mirror [33] whose reflectivity peaks in the cutoff region around 30 nm (H67–H75). As we have expected, we can see that only the central pulse in the seed [Fig. 1(a)] acts as a gate for dramatic enhancement of HHG in the cutoff region and that a practically single pulse with a FWHM of 800 as is obtained, even though small satellite pulses are present. If we use a seed pulse train with a 3-fs global pulse width, composed of ca. 5 pulses, we can suppress the satellite pulses, as is shown in Fig. 1(c). It should be noted that the driving pulse alone would generate virtually no harmonics for this driving intensity. This indicates that even if the driving laser is not sufficiently intense for HHG, the combination with a seed pulse can serve as efficient means to generate a harmonic single pulse. If we use a higher driving intensity, on the other hand, we obtain a single pulse of shorter wavelength and duration; for the case of $I_d=10^{14}$ W/cm^2; e.g., a 580-as single pulse would be obtained around 10 nm wavelength. Although the seed contains harmonic components (H15–H19) which may induce direct ionization, the obtained pulses are mainly due to H11 and H13 with a photon energy below the ionization threshold. We have confirmed this by simulations excluding H15–H19.

Let us next turn to the case where $\lambda_{sf}=2.1$ μm and $\lambda_d=800$ nm. Figure 2(a) displays the seed harmonic pulse train

$$E_s(t) = E_{s0}(t-\phi_c/\omega_d)\sum_{q(\mathrm{odd})=15}^{23} f_q \cos[q\omega_{sf}(t-\phi_c/\omega_d)], \quad (4)$$

containing ca. 5 pulses, composed of the 15th–23rd harmonics. The harmonic mixing ratio is $(f_{15}^2, f_{17}^2, f_{19}^2, f_{21}^2, f_{23}^2)=(0.0625, 0.25, 0.375, 0.25, 0.0625)$, and the common amplitude envelope with a FWHM of 10 fs peaks at $t=\phi_c/\omega_d$. The sum of the peak intensity of each component is 10^{13} W/cm^2. The driving pulse $E_d(t)$, also shown in Fig. 2(a), is assumed to have a FWHM of 15 fs and a peak intensity I_d of 1.2×10^{14} W/cm^2. Figure 2(b) presents the calculated SDA, as would be obtained after reflected by a multilayer x-ray mirror [33] whose reflectivity peaks in the cutoff region around 24 nm (H31–H35). The result, containing ca. 4 pulses, for the case of the driving pulse alone is shown in Fig. 2(c). From Fig. 2(b), we can see that only the middle pulse in the seed significantly boosts HHG in the cutoff region, thus leading to a single pulse with a duration of 350 as. This is shorter than in Fig. 1, probably because the phase range of the driving field relevant to the cutoff region translates to a shorter time interval due to a shorter driving wavelength. The small satellite pulses are further suppressed if the seed pulse is composed of a larger number of orders, for which each pulse in the train becomes shorter.

It is crucial for SAP generation that only one pulse in the seed pulse train contributes to the dramatic enhancement of HHG. This is enabled by the combination of the facts that the energy of a photon emitted in the HHG process depends on the phase of the driving field at which the electron is released and that λ_{sf} differs from λ_d. When they have the same value $\lambda_{sf}=\lambda_d$, each pulse in the seed pulse train, separated by a half laser cycle, contributes to the generation of harmonics of similar orders. This leads to multiple pulses instead of SAP, as can be seen from Fig. 2(d). The difference in intensity between Figs. 2(b) and 2(d) is mainly due to that in the number of photons contained in the seed pulse. Figures 1(d) and 2(e) display, on the other hand, the results for the case where $\lambda_d\neq\lambda_{sf}$ but the delay of the seed is a quarter cycle of the driving field. We can see that the SAP generation based on AEGIS is robust against the variation of the delay between the seed and the drive. In Fig. 2(e), especially, while the middle pulse in the seed cannot generate harmonics in the cutoff region, the left next one can now in its turn, which leads to a single pulse.

Although the seed photon energies considered in Fig. 2 [except for Fig. 2(d)] are below the first excitation energy of Ne, HHG is dramatically enhanced through OFI from virtual states [20,21]. With increasing seed harmonic orders with a fixed intensity, the obtained SX pulse intensity first gradually increases, since the photon energy approaches the first excitation energy. When they further increase into excited bound levels, however, the SX pulse intensity starts to decrease, and, more importantly, a pulse train, rather than a single pulse, is obtained, since electron population can remain in excited levels and be ionized later. When the seed photon energies are larger than the ionization threshold, the electron appears in the continuum, less localized, with a finite initial velocity. This alters the recombination timing and energy, again resulting in a pulse train with lower intensity. Thus, a seed pulse train of harmonic orders below the first excitation energy enhances HHG roughly as efficiently as that of higher orders and is even more convenient from the viewpoint of SAP generation. Moreover, by using harmonic photon energy below the ionization potential, one can avoid superfluous ionization by the pulses not involved in SAP generation.

In conclusion, we have theoretically established the possibility to generate a single-attosecond SX or XUV pulse, based on enhanced HHG by the combination of a seed harmonic pulse train and a relatively long driving IR pulse. Schemes with different seed fundamental and driving wavelengths are desirable, so that only one pulse in the seed train may contribute to enhanced HHG in the cutoff region. Since the origin of the HHG boost is the assist of ionization by the seed [20,21], the enhancement is especially dramatic when the driving pulse alone hardly ionizes the target atom and the emitted harmonic intensity is low. Although the harmonic intensity itself can be augmented by an increase of the driving intensity I_d, the cutoff energy is also necessarily increased. In the case of the AEGIS scheme proposed in the present study, on the other hand, I_d can be tuned so that the targeted wavelength may be in the cutoff region, convenient for SAP generation, and the intensity of the generated pulse can be enhanced by the addition of the seed harmonic pulse train. Thus, the AEGIS may become a flexible alternative to generate an ultrashort single pulse which is indispensable for emerging attosecond science.

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