Attosecond Pulse Amplification in a Plasma-Based X-Ray Laser Dressed by an Infrared Laser Field

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High-harmonic generation (HHG) of laser radiation has led to attosecond pulse formation which offers unprecedented temporal resolution in observing and controlling electron and nuclear dynamics. But the energy of attosecond pulses remains quite small, especially for photon energies exceeding 100 eV, which limits their practical applications. We propose a method for amplification of attosecond pulses in the active medium of a plasma-based x-ray laser dressed by a replica of the laser field used for HHG. The experimental implementation is suggested in hydrogenlike C5+ x-ray laser at 3.4 nm wavelength in the "water window" range.

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Attosecond x-ray pulses produced by high-harmonic generation (HHG) of laser radiation offer a unique combination of unprecedented temporal and spatial resolution in observing and controlling electron and nuclear dynamics in atoms, molecules, and solids [1–4]. However, the energy of attosecond pulses remains quite modest, which limits their practical applications. In particular, it prevents carrying out the attosecond pump—attosecond probe experiments. This limitation is especially severe in the soft x-ray range above 200 eV corresponding to the fundamental absorption edges of matter. Attosecond pulses with down to tens of attosecond duration have been produced in this range recently via HHG of laser fields [5–15], but their energy does not exceed a few picojoules [12,13].

In this Letter, we suggest a technique for amplification of a train of attosecond pulses, produced by HHG, in the active medium of a plasma-based x-ray laser [16–24] in the presence of a strong copropagating infrared (IR) laser field. The amplification of a single high-order harmonic by the plasma-based x-ray laser has been widely studied before [18–20,22–24]. But narrow linewidths of x-ray lasers did not allow for joint amplification of several harmonics. In this Letter, we show that such amplification becomes possible if the active medium of an x-ray laser is simultaneously irradiated by a replica of a laser field used for the HHG.

As shown below, under the action of a strong IR field, the active medium amplifies x-ray radiation not only at the frequency of the resonance but also at its multiple sidebands, separated by twice the frequency of the modulating field. As a result, (i) if replicas of the same IR field are used for the HHG and for modulation of the active medium and (ii) the carrier frequency of the incident attosecond pulse train coincides with the time-averaged frequency of the modulated transition, then various spectral components of the incident x-ray field (harmonics of different orders) might be jointly amplified during their propagation through the medium. With the proper choice of parameters of the active medium (densities of the resonant ions and free electrons) and the modulating field (its amplitude and frequency), the spectral and temporal structure of the attosecond pulses can be preserved during the amplification process.

Let us consider amplification of a set of high-order harmonics in an active medium of a plasma-based x-ray laser with a population inversion at the transition between the ground and the first excited energy levels of hydrogenlike ions, $n = 1 \leftrightarrow n = 2$ (where n is the principal quantum number), simultaneously irradiated by the copropagating IR laser field of the fundamental frequency. If the harmonic of order $2k + 1$ is tuned in resonance with the inverted transition, that is $\omega_{2k+1} = \bar{\omega}_{\mathrm{tr}}$, then the frequency of any other harmonic of order $2(k + l) + 1$ can be represented as

$$\omega_{2(k+l)+1} = \bar{\omega}_{\mathrm{tr}} + 2l\Omega, \tag{1}$$

where Ω is the laser frequency, l is an integer number, $\bar{\omega}_{\mathrm{tr}} = \frac{3}{8}(m_e e^4/\hbar^3)Z^2[1 - (109/Z^6)(E_C^2/E_A^2)]$ is the frequency of the resonant transition accounting for the quadratic Stark effect [25], E_C is the laser field strength, $E_A = m_e^2 e^5/\hbar^4 \cong 5.14 \times 10^9$ V/cm is the atomic unit of electric field, e and m_e are the charge and mass of the electron, respectively, $\hbar$ is Planck's constant, and Z is the nucleus charge number of the ions. Based on Eq. (1), for

the sake of conciseness, we will call harmonic order $2(k+l)+1$ the "2lth" harmonic.

In order to gain insight into the process of high-harmonic amplification, we derive a simple analytical solution for the amplified x-ray field while assuming that the population difference at the resonant transition is constant. We also assume that the IR pulse used for both HHG and modulation of the active medium is sufficiently long so that it can be represented as a monochromatic wave, $\vec{E}_{\mathrm{IR}}(x,t) = \vec{z}_0 E_C \cos[\Omega(t - x n_{\mathrm{pl}}/c)]$, where the x axis is the propagation direction, $\vec{z}_0$ is a unit polarization vector along the z axis, c is the speed of light in vacuum, and n_{pl} is the plasma refractive index for the IR field. The incident x-ray field comprises the set of harmonics ranging from $2(k-l_{\min})+1$ to $2(k+l_{\max})+1$. For the sake of analytical study, it is assumed in the form

$$\vec{E}_{\text{x-ray}}(x \le 0, \tau) = \frac{1}{2}\vec{z}_0 \sum_{l=-l_{\min}}^{l_{\max}} E_{\mathrm{inc}}^{[2(k+l)+1]} \exp\{-i(\bar{\omega}_{\mathrm{tr}} + 2l\Omega)\tau\} + \text{c.c.}, \tag{2}$$

where $E_{\mathrm{inc}}^{[2(k+l)+1]}$ is a complex amplitude of the harmonic, $\tau = t - x/c$ is the local time, and c.c. stands for complex conjugation. Since the harmonics have the same z polarization as the modulating IR field, amplification of the harmonics can be described within the three-level approximation, taking into account the two excited states dressed by the IR field: $|2\rangle = (|2s\rangle + |2p, m=0\rangle)/\sqrt{2}$, and $|3\rangle = (|2s\rangle - |2p, m=0\rangle)/\sqrt{2}$, as well as the ground state $|1\rangle = |1s\rangle$ [26,27]. If the population differences between the states $|1\rangle$, $|2\rangle$, and $|3\rangle$ remain constant and a phase shift acquired by the IR field due to plasma dispersion is much larger than π (the IR and x-ray fields are not phase matched), then [as discussed in the Supplemental Material [28]; see Eqs. (S1)–(S12)] the scattering of the harmonics into each other is strongly suppressed so that each harmonic propagates through the medium independently from the others, and the output x-ray field acquires the form

$$\vec{E}_{\text{x-ray}}(x,\tau) = \frac{1}{2}\vec{z}_0 \sum_{l=-l_{\min}}^{l_{\max}} E_{\mathrm{inc}}^{[2(k+l)+1]} \exp\{g_{\mathrm{total}} J_{2l}^2(p_\omega) x\} \times \exp\{-i(\bar{\omega}_{\mathrm{tr}} + 2l\Omega)\tau\} + \text{c.c.}, \tag{3}$$

where $g_{\mathrm{total}} = (4\pi n_{\mathrm{tr}} N_{\mathrm{ion}} d_{\mathrm{tr}}^2 \bar{\omega}_{\mathrm{tr}}/\hbar\bar{\gamma}_{\mathrm{tr}} c)$ is the amplification coefficient for the resonant x-ray field in the absence of the linear Stark effect, $n_{\mathrm{tr}} = \rho_{22} - \rho_{11} = \rho_{33} - \rho_{11}$ is the population difference at the transitions $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |1\rangle$, d_{tr} and $\bar{\gamma}_{\mathrm{tr}}$ are the absolute value of the dipole moment and the decoherence rate at these transitions, N_{ion} is the density of the resonant ions, $J_m(\mathrm{x})$ is the Bessel function of the first kind of order m, $p_\omega = \Delta_\omega/\Omega$ is the modulation index, and $\Delta_\omega = 3(m_e e^4/\hbar^3 Z)(E_C/E_A)$ is an amplitude of the linear Stark shift induced by the modulating optical field.

As follows from Eq. (3), irradiation of the active medium of the x-ray laser by the modulating field results in the appearance of a gain for the x-ray field at the frequencies of the harmonics, $\bar{\omega}_{\mathrm{tr}} + 2l\Omega$, $l = \pm 1, \pm 2, \ldots$, at the cost of reduced gain at the resonance frequency $\bar{\omega}_{\mathrm{tr}}$; see Figs. 1(a) and 1(b). Since (i) each spectral component of the incident x-ray field is amplified independently from the others,

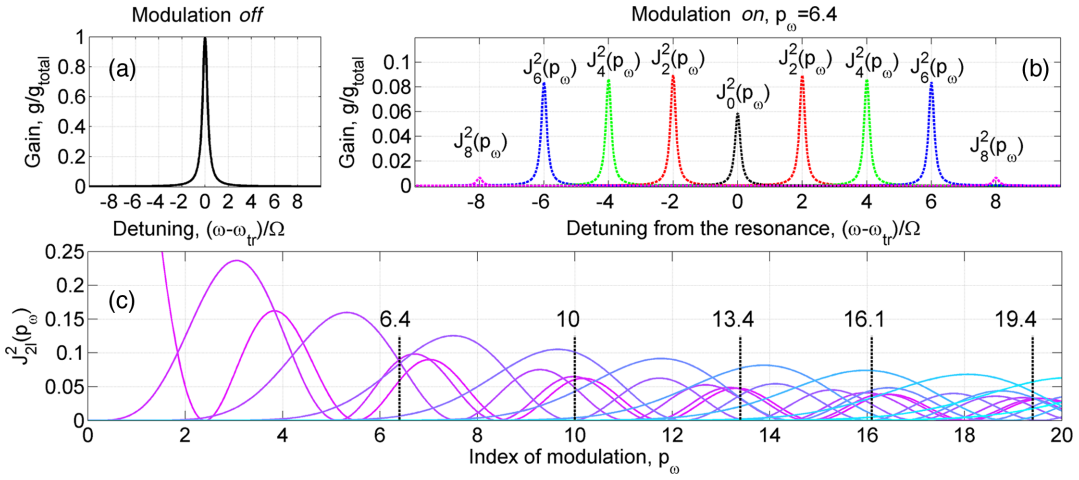

FIG. 1. Frequency dependence of the gain for the incident x-ray field (b) with and (a) without modulating field. In (b), the value of modulation index is $p_\omega = 6.4$; black, red, green, blue, and magenta curves correspond to the gain coefficients for 0th, ±2nd, ±4ht, ±6th, and ±8th sidebands of the inverted transition. (c) Dependence of the squared Bessel functions of even order (from 0th to 20th order) on the value of modulation index, p_ω. The color reflects the order of Bessel function and is gradually changed from magenta, corresponding to $J_0^2(p_\omega)$, to cyan, which corresponds to $J_{20}^2(p_\omega)$. Vertical dashed lines indicate the values of p_ω, at which the squares of Bessel functions of different orders are comparable to each other [compare with (b)].

(ii) the resonant interaction with the ions does not change the phases of the harmonics, and (iii) the plasma dispersion for the x-ray field is negligible; the relative phases of harmonics remain constant during propagation through the medium. Thus, if each harmonic experiences the same gain, then the incident field (2) will preserve its temporal shape during the amplification.

The gain for $2l$th harmonic is proportional to the squared Bessel function $J_{2l}^2(p_\omega)$ of order $2l$ of the modulation index p_ω. As follows from Fig. 1(c), the magnitudes of several Bessel functions of even orders are approximately equal at some particular values of the modulation index. For example, for $p_\omega = 6.4$, one has $J_0^2(p_\omega) \simeq 0.059$, $J_2^2(p_\omega) \simeq 0.090$, $J_4^2(p_\omega) \simeq 0.087$, and $J_6^2(p_\omega) \simeq 0.084$. Thus, the IR field providing such a value for the modulation index allows for nearly uniform amplification of "0th," "±2nd," "±4th," and "±6th" harmonics. The number of harmonics which can be amplified increases with increasing p_ω and is approximately equal to $p_\omega + 1$. Particularly, the values $p_\omega = 10$, $p_\omega = 13.4$, $p_\omega = 16.1$, and $p_\omega = 19.4$ are suitable for amplification of 11, 15, 17, and 21 high-order harmonics of the modulating field, respectively. However, with increasing value of the modulation index, the gain $g_{\text{total}} J_{2l}^2(p_\omega)$, averaged over the harmonic order $2l$, decreases as $1/p_\omega$, while the differences between the amplification coefficients of neighboring harmonics grow [see Fig. 1(c) and Figs. S1 and S6 in the Supplemental Material [28]].

The analytical theory of the linear amplification regime in the framework of the three-level model allows us to understand some general aspects of the high-harmonic amplification. However, in order to perform a quantitative analysis, one needs to take into account (i) the two additional degenerate upper states $|4\rangle = |2p, m = 1\rangle$ and $|5\rangle = |2p, m = -1\rangle$, leading to generation of the y-polarized amplified spontaneous emission (ASE) and (ii) variation of the population differences at all of the involved transitions. This is done on the basis of a complete set of density matrix equations for the five-level system described in the Supplemental Material [see Eqs. (S13)–(S16)] [28]. Here we present the results of calculations. Let us consider neutral plasma consisting of C^{5+} ions, electrons, and some other ions (for example, H^+, to maintain electric neutrality) with C^{5+} ion density $N_{\text{ion}} = 10^{19}$ cm^{-3} and electron density $N_{\text{el}} = 15 N_{\text{ion}}$. Lasing in inverted plasma with such parameters has being theoretically studied [21,29] and is under experimental investigation in the group of Professor Szymon Suckewer at Princeton University, New Jersey. As the modulating field and a source of the incident high-harmonic signal, let us consider 2.1 μm laser radiation, which is particularly suitable for HHG in the "water window" [14]. Let us study the case $p_\omega = 6.4$ and consider amplification of the harmonics of this laser field with orders ranging from 617 to 629, which are 0th, ±2nd, ±4th, and ±6th harmonics with respect to the resonant transition. The value $p_\omega = 6.4$ corresponds to the intensity of the modulating field $I_C = 2.7 \times 10^{15}$ W/cm^2. In order to tune the 0th harmonic (of order 623) in exact resonance with the transitions $|2\rangle, |3\rangle \leftrightarrow |1\rangle$, the laser wavelength should be $\lambda_C = 2102.9$ nm. For the numerical study, an incident x-ray field is assumed to have a Gaussian envelope centered at t_{peak} and the duration (the full width at half maximum of intensity) $t_{1/2}$:

$$E_z(t, x = 0) = \frac{1}{2}\vec{z}_0 E_{\text{hh}} \exp\{-2\ln 2 (t - t_{\text{peak}})^2 / t_{1/2}^2\} \times \sum_{l=-l_{\max}}^{l=l_{\max}} \exp\{-i(\bar{\omega}_{\text{tr}} + 2l\Omega)t\} + \text{c.c.}, \quad (4)$$

where $l_{\max} = 3$. Equation (4) implies that the incident harmonics are phase synchronized and have identical amplitudes. As initial conditions, we assume that at $\tau = 0$ all of the ions are excited to the states $|2\rangle$–$|5\rangle$ with equal probability by a pumping laser field propagating slightly ahead of the seeding x-ray and modulating IR fields, while spontaneous emission at the inverted transitions is taken into account via the initial values of quantum coherencies, which are randomly distributed along the medium [30] (see the Supplemental Material [28]).

The time dependencies of intensities of (i) the z-polarized amplified attosecond pulse train and (ii) the y-polarized ASE at the output from a plasma channel with length $L = 1$ mm and radius $R = 1$ μm are shown in Fig. 2. Figures 2(a)–2(c) correspond to different peak intensities, $I_0 = (c/8\pi)(2l_{\max} + 1)^2 E_{\text{hh}}^2$, of the incident field—namely, (a) $I_0 = 10^{13}$ W/cm^2, (b) $I_0 = 10^{12}$ W/cm^2, and (c) $I_0 = 10^{11}$ W/cm^2. For comparison, if the isolated attosecond pulse obtained in Ref. [13] is focused to $R = 1$ μm, the peak intensity in the water window range will be 5×10^{12} W/cm^2. Figure 2 is plotted for $t_{\text{peak}} = 10$ fs and $t_{1/2} = 35$ fs, which means that the incident attosecond pulse train (4) reaches the peak amplitude 10 fs after creation of the population inversion by a pumping laser pulse at $\tau = 0$ (the influence of t_{peak} on the amplification process is discussed in the Supplemental Material [28]; see Figs. S2–S4). The role of the ASE depends on the peak intensity of the incident x-ray field I_0. If the incident z-polarized field is strong enough, Fig. 2(a), then it is amplified and saturates the resonant transitions $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |1\rangle$ before the y-polarized ASE becomes substantial. As a result, population from the sates $|2\rangle$ and $|3\rangle$ drops down to the state $|1\rangle$, reducing population differences at the transitions $|4\rangle \leftrightarrow |1\rangle$ and $|5\rangle \leftrightarrow |1\rangle$ and decreasing amplification of the ASE. For this reason, in Fig. 2(a), the ASE is negligible. However, as intensity of the incident x-ray field decreases [see Figs. 2(b) and 2(c)], its amplification takes place in a linear regime (without saturation of the resonant transitions), resulting in (i) the larger ratio of the output intensity to I_0, and (ii) the

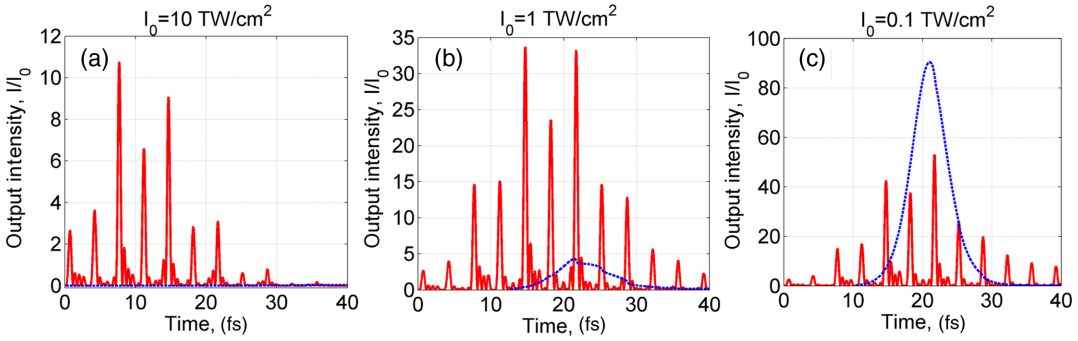

FIG. 2. Time dependence of intensity of an amplified attosecond pulse train of z polarization (red solid curve) and of an ASE of y polarization (blue dashed curve) at the output from an active medium of C^{5+} hydrogenlike x-ray laser. The length of active medium is $L = 1$ mm, the concentration of C^{5+} ions is $N_{\text{ion}} = 10^{19}$ $1/\text{cm}^3$. (a)–(c) correspond to peak intensities of the incident attosecond pulse train $I_0 = 10^{13}$ W/cm^2, $I_0 = 10^{12}$ W/cm^2, and $I_0 = 10^{11}$ W/cm^2, respectively.

stronger ASE, which in this case saturates the transitions $|4\rangle \leftrightarrow |1\rangle$ and $|5\rangle \leftrightarrow |1\rangle$ and thereby reduces population differences at the transitions $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |1\rangle$ (see Fig. S5 in the Supplemental Material [28]).

Amplification of shorter pulses is possible with stronger modulating fields. An increase in intensity of the modulating field to $I_C = 2.5 \times 10^{16}$ W/cm^2, which is still below the ionization threshold of the active medium from the upper lasing states, allows amplification of 140 as pulses, produced from 21 high-order harmonics of the laser field with 2.1 μm wavelength (in such a case, $p_\omega = 19.4$). But the gain for each harmonic decreases with increasing value of the modulation index. Thus, for the same parameters of the medium as in Fig. 2 and $I_0 = 10^{12}$ W/cm^2, the incident x-ray field (4) will be amplified merely 4.2 times (see Fig. S6 in the Supplemental Material [28]). However, if the laser frequency Ω is increased proportionally to the laser field strength E_C, then the modulation index is constant, and the amplification will be more efficient. This case is illustrated in Fig. 3, which corresponds to the amplification of seven harmonics (of orders 231–243) of a laser field with wavelength $\lambda_C = 801.53$ nm. The modulating intensity is $I_C = 1.9 \times 10^{16}$ W/cm^2, which corresponds to $p_\omega = 6.4$. The parameters of the incident x-ray field are the same as in Fig. 2, except for higher Ω, $I_0 = 10^{13}$ W/cm^2. If the length of the active medium would also be the same, the result of amplification would resemble that in Fig. 2. However, in Fig. 3, the medium length is increased to $L = 7$ mm. It results in a very large optical depth of the medium, $g_{\text{total}} J_0^2(p_\omega) L = 22.7$, and thus in the substantial shortening of the attosecond pulse train due to predominant amplification of its front edge, which extracts the major part of the energy, stored in the population inversion of the medium (see Figs. S7–S9 in the Supplemental Material [28]). For the considered parameters of the medium, the envelope of the amplified attosecond pulse train becomes shorter than the repetition period of the pulses in the train. As a result, the active medium isolates a single pulse with 130 as duration, which is amplified to $I_{\text{max}} = 261\, I_0$.

The lower limit for the duration of attosecond pulses which can be amplified is set by the upper limit of the intensity of the modulating field, which is determined by the threshold of ionization of the resonant ions from the upper lasing states. For the case of C^{5+} ions, the acceptable peak intensity of the modulating field is estimated as $I_C = 3.5 \times 10^{16}$ W/cm^2 and corresponds to the ionization time

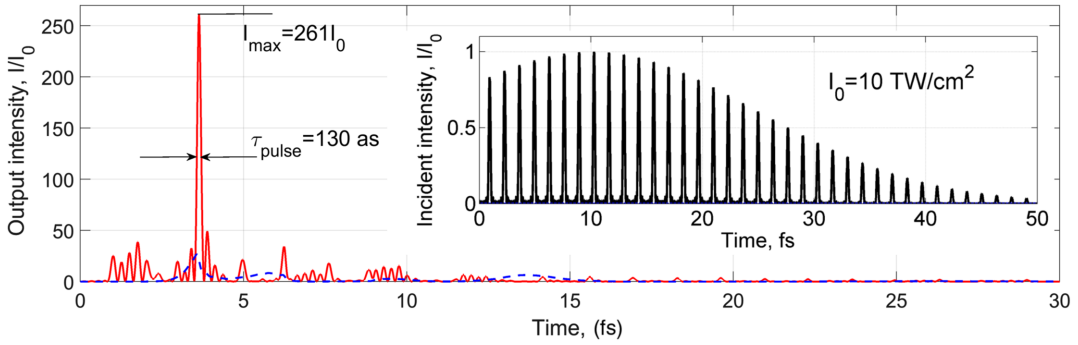

FIG. 3. A single attosecond pulse in the water window spectral range, which is produced as a result of amplification of the attosecond pulse train, shown in the inset, in an active medium of a C^{5+} hydrogenlike x-ray laser. Red solid curve is the amplified z-polarized x-ray signal; the blue dashed curve corresponds to the ASE of y polarization. The length of the active medium is $L = 7$ mm; the concentration of C^{5+} ions is $N_{\text{ion}} = 10^{19}$ $1/\text{cm}^3$.

from the states $|2\rangle$ and $|3\rangle$, $\tau_{\mathrm{ion}}^{(2),(3)} = 60$ fs. Scaling of the results, shown in Fig. 3, for this intensity of the modulating field and $\lambda_C = 590$ nm leads to duration of the amplified isolated attosecond pulse $\tau_{\mathrm{pulse}} \approx 100$ as.

In conclusion, we have shown the possibility of amplifying a set of high-order harmonics of an IR laser field in active medium of a hydrogenlike plasma-based x-ray laser, dressed by a replica of the same field as used for HHG. The amplification occurs due to periodic in time and space sub-optical-cycle Stark splitting of the upper lasing energy level by the IR field, which results in redistribution of the gain to the combination frequencies separated from the resonance by even multiples of the frequency of the IR field. For the specific intensities of the modulating field, nearly uniform gain may be provided for the whole set of harmonics. In a sufficiently dense plasma, the harmonics are amplified independently from each other so that their relative phases remain constant. Thus, if the incident x-ray field represents an attosecond pulse train, it will keep this form during the amplification. We have suggested an experimental implementation of this method in active medium of C^{5+} ions and have shown the possibility of amplifying by 2 orders of magnitude the attosecond pulses with duration down to 100 as at the carrier wavelength 3.4 nm in the "water window" range. In an optically deep medium, the duration of the amplified attosecond pulse train is reduced due to predominant amplification of its front edge. If the optical depth of the medium is high enough, the active medium selects a single attosecond pulse, which is amplified much more strongly than the other pulses from the train. The amplification of a set of harmonics does not rely on specific features of the hydrogenlike ions and potentially might be implemented in an arbitrary (not only hydrogenlike) active medium. The suggested approach for the attosecond pulse amplification is robust with respect to variation of major experimental parameters (such as population difference at the inverted transition and concentration of the resonant ions and free electrons, as well as intensities of the IR and x-ray fields), since (i) it does not rely on the phase matching of the IR and x-ray fields and (ii) preserves the relative phases of the amplified harmonics (see the Supplemental Material [28] for more details). To the best of our knowledge, this is the first proposal for using plasma-based x-ray lasers for the amplification of attosecond pulse trains.

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