Formation and amplification of subfemtosecond x-ray pulses in a plasma medium of hydrogenlike ions with a modulated resonant transition

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Coherent intense attosecond x-ray pulses could lead to a fast dynamical imaging of biological macromolecules and other material nanostructures with a unique combination of a record high temporal and spatial resolution. Plasma-based x-ray laser sources are capable of producing high-energy x-ray pulses but with relatively long picosecond duration. The sources based on high-harmonic generation (HHG) of a laser field allow one to produce much shorter pulses but of lower energy. We suggest two different paths towards intense subfemtosecond x-ray sources: (i) via efficient transformation of the picosecond radiation of the x-ray plasma lasers into the trains of subfemtosecond pulses in a resonantly absorbing medium, and (ii) via amplification of HHG radiation in the active medium of the x-ray plasma lasers. We show that essentially the same technique can be used for realization of both paths. This technique is a modulation of the parameters of the resonant transition (accordingly in absorbing or amplifying medium) produced under the action of a sufficiently strong infrared or optical field. We propose experimental realization of the suggested technique in the passive and/or active media of (i) Li III ions modulated by the mid-IR laser field and (ii) C VI ions modulated by the optical laser radiation.

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I. INTRODUCTION

Coherent intense subfemtosecond (sub-fs) soft x-ray pulses would open extremely wide applications for dynamical, element-specific microscopy and diffraction imaging in chemistry, biology, medicine, nanoscience, and materials science, providing a unique combination of the unprecedented high spatial and temporal resolution, ultimately determined by the nanometer carrier wavelength and attosecond pulse duration, respectively (see reviews on x-ray lasers [1–10] and attosecond physics [11–19]). Production of the bright ultrafast coherent sources in a "water window" range (between the C and O K-shell absorption edges at 284–540 eV, i.e., 4.4–2.3 nm) is considered to be especially important for imaging of the protein dynamics in the living cells [20–26], including studies of protein-protein and protein-DNA interactions induced by intense ultrafast laser pulses [27,28].

Currently there are three main types of coherent sources in the soft x-ray wavelength range: free-electron lasers [29–36], plasma-based x-ray lasers [4–6,8,37,38], and high-harmonic generation (HHG) sources [25–28]. Free-electron lasers are located at large-scale state-of-the-art very expensive facilities and there are only few of them available in the world. They produce high-energy pulses, but the pulse duration is currently limited by femtoseconds and pulses are typically not transform limited due to the shot noise.

Table-top plasma-based soft x-ray lasers produce relatively high-energy pulses (up to several millijoules) but of rather long picosecond duration [37,38]. The HHG sources allow producing thousands of high harmonics, stretching into the x-ray range [23–26]. However, the individual harmonic energy in the soft x-ray range, in particular, in a water window range, does not exceed the nanojoule range [24–27,39] due to the low (less than 10^{-7}) conversion efficiency. The duration of a single harmonic is in a range of a few to hundreds of femtoseconds [27,40]. A set of harmonics might constitute the attosecond x-ray pulses (under the condition of an attochirp compensation), but the energy of such pulses is still quite limited [28,41].

This work provides a theoretical study and comparison of two possible paths to coherent, intense sub-fs x-ray sources: (i) via efficient conversion of picosecond pulses of plasma x-ray lasers into attosecond pulse trains without essential loss of energy, and (ii) via amplification of an individual high-order harmonic of laser radiation in an active medium of x-ray lasers accompanied by formation of attosecond pulses.

We show that both paths can be realized by using essentially the same technique recently theoretically developed by our group [42–46]. The basic idea is to use an interaction of the x-ray plasma laser or high-harmonic radiation with the plasma medium consisting of hydrogenlike ions (without or with a population inversion, respectively) at the resonant transition, the frequency of which is modulated by the infrared (IR) (for example, for low-Z ions, like Li III) or optical (for higher Z ions, like C VI) laser fields.

The outline of the paper is as follows. In Sec. II we formulate the basic set of the density matrix and wave equations, describing formation of sub-fs pulses in a plasma of hydrogenlike ions in the presence of a moderately strong IR or optical field. In Sec. III we study the possibility of an efficient transfer of the quasimonochromatic radiation of x-ray plasma lasers into the trains of sub-fs pulses in the passive (noninverted) plasma of hydrogenlike ions modulated by an IR or optical field. In Sec. IV we analyze the possibility of amplification of x-ray field, corresponding to an individual harmonic from a high-harmonic radiation source,

accompanied by the sub-fs pulse formation in an inverted plasma of hydrogenlike ions modulated by an IR or optical field. We also suggest an experimental realization of both techniques in Li III (Li^{2+} ions) and C VI (C^{5+} ions) plasmas in the vicinity of 13.5- and 3.4-nm wavelengths, respectively, corresponding to the resonant transitions from the first excited to the ground energy levels in these ions, modulated by the IR or optical laser fields correspondingly, and determine the ultimate duration and intensity of pulses which can be achieved by those techniques.

In the Conclusions section we briefly summarize the major results pointing out the advantages and disadvantages of both techniques, as well as the prospects for extension of these techniques to plasmas of non-hydrogen-like ions.

II. PROPAGATION OF X-RAY RADIATION THROUGH A MODULATED MEDIUM OF HYDROGENLIKE IONS

Let us consider propagation of x-ray radiation along the x axis through a medium of neutral plasma of hydrogenlike ions. At the entrance to the medium, $x = 0$, the radiation is quasimonochromatic and has the form

$$\vec{E}_{X\text{-ray, inc}}(t) = \tfrac{1}{2}\vec{z}_0\tilde{E}_0(t)\exp\{-i\,\omega_0 t\} + \text{c.c.}, \qquad (1)$$

where E_0 is the slowly varying envelope of incident radiation, ω_0 is its carrier frequencies, and c.c. stands for complex conjugation. The radiation (1) is chosen to be near resonant to a transition from the ground energy level corresponding to the state $|1\rangle$ to the first excited atomic energy level so that $\omega_0 \approx \omega_{21}^0$, where ω_{21}^0 is the unperturbed frequency of the resonant transition.

The medium is also irradiated by a moderately strong IR or optical laser field with the same polarization as x-ray radiation (1) and propagating in the same x direction with a phase velocity determined by the plasma refractive index:

$$\vec{E}_{\text{IR/opt}}(x,t) = \vec{z}_0\tilde{E}_{\text{IR/opt}}\cos\{-i\,\Omega(t - xn_{pl}/c)\}, \qquad (2)$$

where $\tilde{E}_{\text{IR/opt}}$ is the amplitude of the driving IR or optical field, Ω is its angular frequency, $n_{pl} = \sqrt{1-\frac{\omega_{pl}^2}{\Omega^2}}$ is the plasma index of refraction, $\omega_{pl}^2 = \frac{4\pi n_e e^2}{m_e}$, where n_e, m_e, and e are electron concentration, electron mass, and electron charge, and c is the speed of light in vacuum. The IR or optical field is far-detuned from all the relevant atomic resonances, and it traverses the medium without appreciable distortions.

Since x-ray radiation is z polarized and its frequency is close to resonance between the ground state and the first excited state of hydrogenlike ions, we may take into account only two excited states, $|2\rangle = (|2s\rangle + |2p,m=0\rangle)/\sqrt{2}$ and $|3\rangle = (|2s\rangle - |2p,m=0\rangle)/\sqrt{2}$ (the energies of which are harmonically modulated by z polarized by an IR or optical field due to the linear Stark effect, as may be seen from the density matrix equations below). We do not take into account the two other excited states $|4\rangle = |2p,m=1\rangle$, and $|5\rangle = |2p,m=-1\rangle$ (which are not modulated by an IR or optical field), corresponding to the first excited energy level of the ions, $n = 2$. In the case of a passive (noninverted) medium, studied in Sec. III, these excited states do not influence a propagation of a z-polarized field in any way because their dipole transition moments are oriented along the y axis. In the case of an active (inverted) medium y-polarized field may be generated starting from the spontaneous emission. Potentially, this field may influence an amplification of an incident z-polarized field via population of the ground state and hence reduction of the population inversion at the modulated $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |1\rangle$ transitions. Thus the possibility to use a three-level approximation can be easily justified only when transitions $|4\rangle \leftrightarrow |1\rangle$ and $|5\rangle \leftrightarrow |1\rangle$ remain unsaturated by a y-polarized field. (This is due to the fact that gain for z-polarized and y-polarized fields originates from the different populated excited states. Thus a y-polarized field does not influence gain for a z-polarized field until it appreciably changes a population of the ground state.) A possibility to use it for analysis of sufficiently short seeding pulse amplification in the case of high gain for the y-polarized field (even when the last one saturates $|4\rangle \leftrightarrow |1\rangle$ and $|5\rangle \leftrightarrow |1\rangle$ transitions) is proved in the Appendix. There, the equations for populations of those levels and coherencies at the corresponding transitions, as well as the wave equation for the y component of the quasiresonant x-ray field, are taken into account. As for the higher excited states, they may be safely neglected when both frequency and intensity of an IR or optical field are sufficiently small, as it was studied in detail in Refs. [46,47]. Within this approximation, the resonant polarization of the medium is defined by the density matrix elements ρ_{ij},

$$\vec{P}(\vec{r},t) = N(\vec{d}_{12}\rho_{21} + \vec{d}_{13}\rho_{31} + \text{c.c.}), \qquad (3)$$

where N is ion concentration and $\vec{d}_{ij}$ is the dipole moment of transition between states $|i\rangle$ and $|j\rangle$. Within the three-level model, all the nonzero dipole moments are given by

$$\begin{aligned}
\vec{d}_{12} &= \vec{d}_{1s\leftrightarrow 2p,m=0}/\sqrt{2} = \vec{z}_0 d_{\parallel},\\
\vec{d}_{13} &= -\vec{d}_{1s\leftrightarrow 2p,m=0}/\sqrt{2} = -\vec{z}_0 d_{\parallel},\\
\vec{d}_{22} &= \vec{d}_{2s\leftrightarrow 2p,m=0} = \vec{z}_0 d_{av},\\
\vec{d}_{33} &= -\vec{d}_{2s\leftrightarrow 2p,m=0} = -\vec{z}_0 d_{av}.
\end{aligned} \qquad (4)$$

In atomic units $d_{\parallel} = \frac{2^7}{3^5 Z}$ and $d_{av} = 3/Z$, where Z is an atomic number. Under the action of both x-ray and an IR or optical field, the evolution of the density matrix elements is given by

$$\begin{aligned}
\dot{\rho}_{11} &= +\gamma_{11}(\rho_{22}+\rho_{33}) - i[\mathrm{H},\rho]_{11}\\
\dot{\rho}_{ij} &= -\gamma_{ij}\rho_{ij} - i[\mathrm{H},\rho]_{ij}, ij \neq 11\\
\mathrm{H} &= \begin{pmatrix} \omega_1 & -E_z d_{\parallel} & E_z d_{\parallel}\\ -E_z d_{\parallel} & \omega_2 - \tilde{E}_{\text{IR/opt}} d_{av}\cos\{-i\,\Omega(t - xn_{pl}/c)\} & 0\\ E_z d_{\parallel} & 0 & \omega_3 + \tilde{E}_{\text{IR/opt}} d_{av}\cos\{-i\,\Omega(t - xn_{pl}/c)\}\end{pmatrix}.
\end{aligned} \qquad (5)$$

Here ω_i is the energy of atomic level $|i\rangle$. With the quadratic Stark effect taken into account, $\omega_1 = -\frac{Z^2}{2}(1+\frac{9}{256}F_c^2)$, $\omega_2 = \omega_3 = -\frac{Z^2}{8}(1+\frac{7}{4}F_c^2)$, where $F_c = (\frac{2}{Z})^3\tilde{E}_{\mathrm{IR/opt}}$ [43].

The decay rates γ_{ij} are defined as $\gamma_{12} = \gamma_{13} \approx \gamma_{\mathrm{coll}} + \Gamma_{\mathrm{ion}}/2 + \Gamma_{\mathrm{radiative}}/2$, $\gamma_{23} \approx \gamma_{\mathrm{coll}} + \Gamma_{\mathrm{ion}} + \Gamma_{\mathrm{radiative}}$, $\gamma_{22} = \gamma_{33} \approx \Gamma_{\mathrm{ion}} + \Gamma_{\mathrm{radiative}}$, $\gamma_{11} \approx \Gamma_{\mathrm{radiative}}$, where γ_{coll} and Γ_{ion} are collisional broadening and ionization decay rates, respectively. $\Gamma_{\mathrm{radiative}}$ are radiative decay rates, which could be found in Ref. [48]. Collisional broadening was estimated according to [49], $\Gamma_{\mathrm{ion}} \approx \frac{Z^2}{16}\sqrt{\frac{3F_c}{\pi}}[(\frac{4}{F_c})e^{+3} + (\frac{4}{F_c})^3 e^{-3}]\exp\{-\frac{2}{3F_c}\}$ [50]. We neglect Doppler broadening of transitions since we consider ion temperatures on the order of 1 eV, as is typical in recombination x-ray plasma lasers [35], resulting in Doppler broadening comparable to or less than collisional broadening. It is worth noting that for the IR or optical field parameters we consider in the paper, $\gamma_{ij} \ll \Omega$.

Let us seek a partial solution in the form

$$\begin{aligned}
\vec{E}(\vec{r},t) &= \tfrac{1}{2}\vec{z}_0\tilde{E}(\vec{r},t)e^{-i(\omega t-kx)} + \text{c.c.}\\
\rho_{12}(\vec{r},t) &= \tilde{\rho}_{12}(\vec{r},t)e^{i\omega t-ikx},\\
\rho_{13}(\vec{r},t) &= \tilde{\rho}_{13}(\vec{r},t)e^{i\omega t-ikx},\\
\rho_{ij}(\vec{r},t) &= \tilde{\rho}_{ij}(\vec{r},t), \quad ij = 11,22,33,23,
\end{aligned} \tag{6}$$

where $\tilde{E}(\vec{r},t)$ and $\tilde{\rho}_{ij}(\vec{r},t)$ are the slowly varying amplitudes of the field and decay matrix elements, respectively, that is, $|\frac{1}{\tilde{E}}\frac{\partial\tilde{E}}{\partial t}| \ll \omega$, $|\frac{1}{\tilde{E}}\nabla\tilde{E}| \ll k$, and $|\frac{1}{\tilde{\rho}_{ij}(\vec{r},t)}\frac{\partial\tilde{\rho}_{ij}(\vec{r},t)}{\partial t}| \ll \omega$, $|\frac{1}{\tilde{\rho}_{ij}(\vec{r},t)}\nabla\tilde{\rho}_{ij}(\vec{r},t)| \ll k$. In such a case, within the rotating wave approximation, we get for plane waves

$$\frac{\partial\tilde{E}}{\partial x} = i4\pi\frac{\omega N d_\parallel}{c\sqrt{\varepsilon}}(\rho_{21}-\rho_{31}), \tag{7}$$

where $\varepsilon \approx 1$ and

$$\begin{aligned}
\dot{\tilde{\rho}}_{11} &= +\gamma_{11}(\tilde{\rho}_{22}+\tilde{\rho}_{33}) + \frac{i\tilde{E}^* d_\parallel}{2}(\tilde{\rho}_{21}-\tilde{\rho}_{31}) + \frac{i\tilde{E}d_\parallel}{2}(\tilde{\rho}_{13}-\tilde{\rho}_{12}),\\
\dot{\tilde{\rho}}_{22} &= -\gamma_{22}\tilde{\rho}_{22} + \frac{i\tilde{E}d_\parallel}{2}\tilde{\rho}_{12} - \frac{i\tilde{E}^* d_\parallel}{2}\tilde{\rho}_{21}, \quad \dot{\tilde{\rho}}_{33} = -\gamma_{33}\tilde{\rho}_{33} + \frac{i\tilde{E}^* d_\parallel}{2}\rho_{31} - \frac{i\tilde{E}d_\parallel}{2}\rho_{13},\\
\dot{\tilde{\rho}}_{12} &= -\gamma_{12}\tilde{\rho}_{12} - i\tilde{\rho}_{12}(\omega_1-\omega_2+\omega+\tilde{E}_{\mathrm{IR/opt}}d_{av}\cos(\Omega[\tau+(1-n_{pl})x/c])) - \frac{i\tilde{E}^* d_\parallel}{2}(\rho_{32}-\rho_{22}+\rho_{11}),\\
\dot{\tilde{\rho}}_{13} &= -\gamma_{13}\tilde{\rho}_{13} - i\tilde{\rho}_{13}(\omega_1-\omega_3+\omega-\tilde{E}_{\mathrm{IR/opt}}d_{av}\cos(\Omega[\tau+(1-n_{pl})x/c])) - \frac{i\tilde{E}^* d_\parallel}{2}(-\rho_{23}+\rho_{33}-\rho_{11}),\\
\dot{\tilde{\rho}}_{23} &= -\gamma_{23}\tilde{\rho}_{23} - i\tilde{\rho}_{23}(\omega_2-\omega_3-2\tilde{E}_{\mathrm{IR/opt}}d_{av}\cos(\Omega[\tau+(1-n_{pl})x/c])) + \frac{i\tilde{E}^* d_\parallel}{2}\tilde{\rho}_{21} + \frac{i\tilde{E}d_\parallel}{2}\tilde{\rho}_{13},
\end{aligned} \tag{8}$$

where we have introduced local time $\tau = t - x/c$. Equations (7) and (8) along with initial conditions

$$\tilde{\rho}_{ij}(x,\tau=0) = \tilde{\rho}_{ij}^{(0)} \tag{9}$$

and boundary condition (1) describe propagation of a z-polarized x-ray field in x direction. In the case of a passive (noninverted) medium, which is discussed in the next section, initial conditions are as follows:

$$\tilde{\rho}_{11}(x,\tau=0) = 1, \quad \tilde{\rho}_{ij}(x,\tau=0) = 0, \quad ij \neq 11. \tag{10}$$

In order to model amplification of incident x-ray field (1) in an inverted medium of x-ray plasma lasers (considered in Sec. IV), we assume that at the moment $\tau = 0$ all the ions have equal probability to be in the excited states $|2\rangle,|3\rangle,|4\rangle,|5\rangle$ and there are no coherencies. Namely, the initial conditions become

$$\begin{aligned}
&\tilde{\rho}_{22}(x,\tau=0) = \tilde{\rho}_{33}(x,\tau=0) = 0.25,\\
&\tilde{\rho}_{ij}(x,\tau=0) = 0, \quad ij \neq 22,33.
\end{aligned} \tag{11}$$

The incident x-ray field (1) is considered to be a z-polarized quasimonochromatic pulse resonant to transitions $|1\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |3\rangle$.

As it can be seen from Eq. (8), the instantaneous frequencies of transitions are periodically modulated in space and time with a period equal to that of the IR or optical field. Physically it corresponds to the linear ac Stark effect, Fig. 1. Since transitions $|1\rangle \leftrightarrow |2\rangle, |1\rangle \leftrightarrow |3\rangle$ have nonzero components of dipole moment along z direction, the modulation of transition frequencies will result in a modulated polarization of the medium, leading to the appearance of the sidebands in the z-polarized field propagating in x direction. The neighboring sidebands are separated by the double IR or optical field frequency due to the atomic symmetry (Fig. 1). Both the phases and the amplitudes of the sidebands depend on the modulation amplitude and length of the medium. With a proper choice of these parameters it appears to be possible to have the sidebands in phase with each other as well as with the resonant component

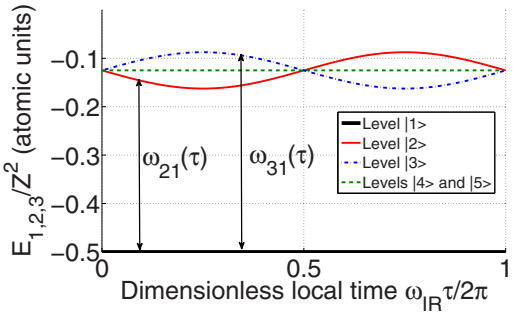

FIG. 1. Energy levels of the ground and first excited state of the hydrogen-like-ion dressed by an IR or optical field.

of a field at the exit of the medium. As a result, a train of the sub-fs pulses may be formed.

In the next sections we consider the formation of such sub-fs pulses in the passive (noninverted) and active (inverted) plasma of the hydrogenlike ions.

III. TRANSFORMATION OF X-RAY PLASMA LASER RADIATION INTO A TRAIN OF SUB-FS PULSES IN PLASMA OF NONINVERTED HYDROGENLIKE IONS MODULATED BY AN IR OR OPTICAL LASER FIELD

In the case of a passive (noninverted) optically thin resonant medium, when plasma dispersion can be neglected, a set of equations (1), (7), (8), and (10) has a simple analytical solution studied in Ref. [43]. The amplitudes and phases of the sidebands in this case are defined by the Bessel functions $J_{2n}(P_\omega)$ of the modulation index $P_\omega = \frac{d_{av}\tilde{E}_{\mathrm{IR/opt}}}{\Omega}$, $n = 0, \pm1, \pm2, \ldots$. In particular, for the modulation index $2.4 < P_\omega < 5.14$ all sidebands are in phase with respect to each other and with an incident field [43]. (The antiphased resonant scattered component is dominated by an incident resonant x-ray field.) As a result, a train of short pulses can be formed after attenuation of an incident field to the level of the sidebands. With an increase of the optical thickness of the medium, the amplitudes of the sidebands grow up while an amplitude of the incident field decreases, due both to the resonant absorption and transformation into the sidebands. The efficiency of transformation of the incident quasimonochromatic radiation into the train of the short pulses can be quite high. For example, the efficiency was shown to be as high as 76% for transformation of the quasimonochromatic VUV radiation with wavelength 122.1 nm into the train of 3.2-fs pulses in atomic hydrogen modulated with an IR field with a wavelength of 10.65 μm and intensity 1.4×10^{12} W/cm^2 ($P_\omega = 4.45$).

In this section we consider the possibility of application of this technique for transformation of quasimonochromatic x-ray radiation into trains of sub-fs pulses. The high frequency of radiation implies using a plasma of hydrogenlike ions instead of hydrogen atoms. Plasma dispersion results in an increase of the phase velocity of an IR or optical field with respect to the x-ray field. Thus the phase shifts between the neighboring sidebands determined by the phase difference between x-ray and IR or optical field change with the propagation distance. As a result, the sidebands formed at different points of the medium become out of phase with each other, which increases the pulse duration. Therefore the plasma dispersion limits the length of the medium from above. On the other hand, a sufficiently large interaction length is required for an efficient conversion of a quasimonochromatic radiation to the train of sub-fs pulses. Obviously, this leads to a tradeoff between pulse duration and efficiency of energy conversion.

Let us consider possible experimental implementation of this technique for transformation of quasimonochromatic x-ray radiation with a carrier frequency resonant to $|1\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |3\rangle$ ion transitions, near 13.5 nm in Li III and near 3.4 nm in C VI, into a train of sub-fs pulses and determine its ultimate capabilities in terms of the achievable pulse duration and transformation efficiency, taking into account the effect of the plasma dispersion.

In the case of Li III ions we consider the densities of ions and electrons to be equal to $N = 1.5 \times 10^{17}$ cm^{-3}, $n_e = 2N$ at ion temperature $\sim$1 eV and electron temperature $\sim$2 eV. Such electron density corresponds to a plasma frequency of $\sim$5 THz. These parameters are chosen to be the same as in the case of active plasma, considered in the next section, for convenient comparison of passive and active cases. In its turn, the parameters for an active plasma are chosen to be close with those used in experimental realization of a recombination laser at the $2p$-$1s$ transition in Li III [51]. The relaxation times (which are the inverse of the corresponding decay rates) under such plasma parameters are estimated as: $\tau_{\mathrm{collision}} \approx 0.425$ ps, $\tau_{\mathrm{radiative}} \approx 19.7$ ps. In the case of C VI (i.e., C^{5+}) ions we consider the densities of ions $N = 1.0 \times 10^{17}$ cm^{-3}, $n_e = 5N$ at ion temperature $\sim$3 eV and electron temperature $\sim$5 eV. The relaxation times are $\tau_{\mathrm{collision}} \approx 0.56$ ps, $\tau_{\mathrm{radiative}} \approx 1.23$ ps.

The plasma dispersion destroys the pulses after some critical length L_{crit}, at which the phase difference between neighboring sidebands acquires a π phase shift: $\frac{\Omega}{c}(1 - n_{pl})L_{\mathrm{crit}} \approx \pi$. Thus, in order to achieve the shortest possible duration of pulses, the condition $L \ll L_{\mathrm{crit}}$ needs to be satisfied. In the favorable case of sufficiently high frequency of the modulating field ($\Omega \gg \omega_{pl}$), the critical length L_{crit} is proportional to Ω. On the other hand, in order to form sidebands with amplitudes comparable with the amplitude of the incident field, the physical length of the medium L should be on the order of or higher than a characteristic absorption length $L_{ab} = \frac{\gamma_{12}\hbar c}{4\pi(\rho_{22}-\rho_{11})N\omega_{21}|d_{\mathrm{eff}}|^2}$, where $d_{\mathrm{eff}} = d_\parallel J_0(P_\omega)$ is an effective dipole moment of the resonant transition. An increase of the IR or optical field frequency at the fixed value of a modulation index implies an increase of intensity of this field. At sufficiently high intensity level an ionization rate from the resonant excited states (which grows exponentially with increase of the field strength) becomes dominant over the collisional decoherence rate. As a result, with increasing frequency (decreasing wavelength) of the IR or optical field the linewidths of the resonant transitions $\gamma_{12} = \gamma_{13}$ grow, leading to a fast increase of the absorption length L_{ab}. High efficiency of energy conversion into the pulses implies $L_{ab} \leqslant L_{\mathrm{crit}}$, which limits from below the wavelength of the modulating laser field. For the considered parameters of Li III and C VI ions, it implies a wavelength of the IR field larger than approximately 1400 nm for Li III and of the optical laser field larger than 400 nm for C VI. The minimum possible wavelength of the modulating laser, in its turn, for the fixed modulation index determines the minimum pulse duration to be achieved. The minimum achievable pulse duration can be estimated as half of the period of an IR or optical field divided by the number of sidebands of comparable amplitudes (which is approximately $2P_\omega + 1$). For the above wavelengths and $P_\omega = 4.45$, the estimate gives 330 as in Li III and 100 as in C VI. The intensities of the driving field required us to realize a modulation index $P_\omega = 4.45$ for those having wavelengths of $I_\Omega = 7.3 \times 10^{14}$ W/cm^2 and $I_\Omega = 3.6 \times 10^{16}$ W/cm^2 for Li III and C VI, respectively.

For the considered mechanism of pulse formation the ionization rate should satisfy the condition $\Gamma_{\mathrm{ion}} \ll \Omega$, so that the upper levels would not be depleted by tunneling ionization in the interval between the neighboring maxima of

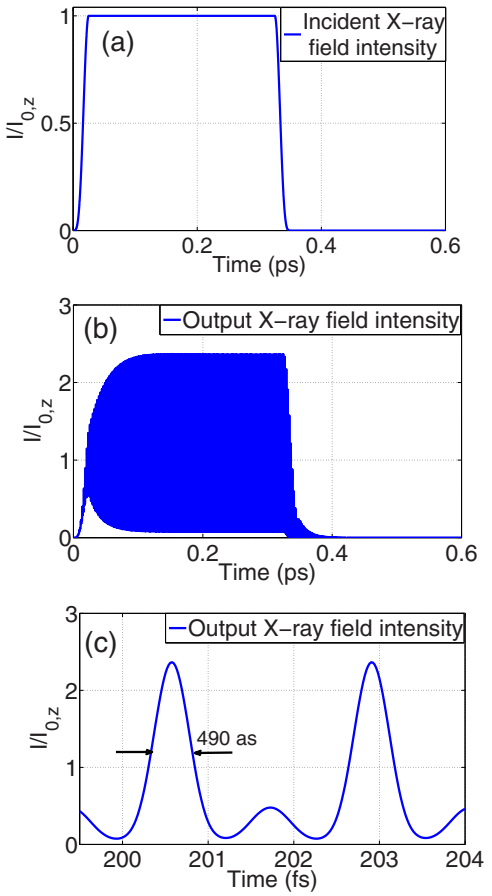

FIG. 2. Time dependence of x-ray radiation intensity: (a) incident x-ray field, (b) output field, and (c) the same as (b) but showing only a small part of the whole envelope. The parameters of the Li III plasma, an IR field, and an incident XUV field are provided in the text of the paper.

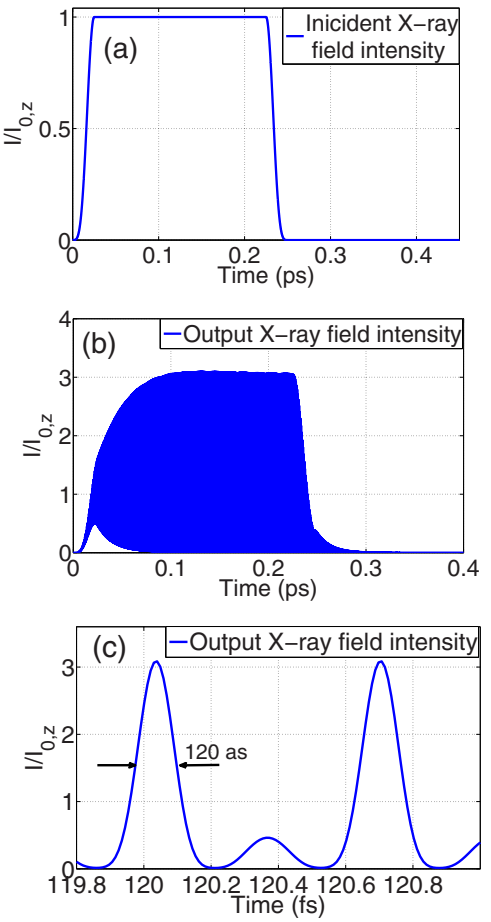

FIG. 3. Time dependence of x-ray radiation intensity: (a) incident x-ray field, (b) output field, and (c) the same as (b) but showing only a small part of the whole envelope. The parameters of the C VI plasma, as well as optical and incident XUV fields, are provided in the text of the paper.

a modulating field. For the chosen parameters of a modulating field, this condition is fulfilled.

A frequency of an incident x-ray field is chosen to be shifted from the frequency of the resonance, unperturbed by an IR or optical field, by a magnitude of the quadratic Stark shift. At the chosen field intensities of an IR or optical field this shift is equal to 0.16 eV in Li III and 0.56 eV in C VI. A peak intensity of an incident field is chosen to be small compared to the saturation intensity for the resonant transition, which was estimated from auxiliary calculations to be equal to 10^{10} W/cm^2 in Li III and 10^{11} W/cm^2 in C VI.

The incident x-ray field envelope is chosen in the form

$$\tilde{E}_z(x=0,t)=\tilde{E}_{z,0}f(t),$$
$$f(t)=\begin{cases}\sin^2\left(\frac{\pi t}{2t_{\rm ramp}}\right), & 0<t<t_{\rm ramp}\\ 1, & t_{\rm ramp}<t<T-t_{\rm ramp},\\ \sin^2\left(\frac{\pi(T-t)}{2t_{\rm ramp}}\right), & T-t_{\rm ramp}<t<T\end{cases} \quad (12)$$

where $t_{\rm ramp}=25$ fs, while $T=350$ fs for Li III and $T=250$ fs for C VI [see Figs. 2(a) and 3(a)]. The duration of the incident x-ray field is chosen to be on the same order of magnitude as a decay time. Such a field can be produced by an x-ray plasma laser seeded with a high-harmonic radiation [47,50] or by x-ray a free-electron laser.

The numerical solution of Eqs. (1), (7), (8), and (10) for the above-described set of parameters shows that trains of pulses with duration of 490 and 120 as could be formed in Li III and C VI plasma, respectively. An efficiency of transformation, defined as ratio of incident x-ray field energy to output field energy, for propagation lengths of 0.6 mm in Li III and 1.0 mm in C VI is about 60%.

IV. AMPLIFICATION OF AN INDIVIDUAL HIGH-HARMONIC RADIATION WITH ITS SIMULTANEOUS TRANSFORMATION INTO THE TRAIN OF ATTOSECOND PULSES IN AN ACTIVE MEDIUM OF HYDROGENLIKE RECOMBINATION X-RAY PLASMA LASER MODULATED BY AN IR OR OPTICAL FIELD

In this section we discuss the possibility of amplification of relatively weak quasimonochromatic incident x-ray radiation of a single high-harmonic in the inverted hydrogenlike medium accompanied by attosecond pulse formation due to modulation of the resonant transition by a moderately strong quasimonochromatic IR or optical field. Note that seeding of

x-ray plasma lasers with a single harmonic was widely studied for improvement of the spatial and temporal coherence of the produced pulses, reduction of the pulse duration up to a few hundred femtosecond duration, and polarization control [52–55]. However, seeding of x-ray lasers with a modulated active medium was not considered so far.

Similar to the case of a passive medium, let us consider the possibility of realization of the suggested technique in Li III and C VI with the same parameters of the plasma as in the passive medium but under the assumption that all ions are fully inverted (11). It is worth noting that lasing on the considered transition in Li III was theoretically studied [56] and experimentally demonstrated [51]. The above parameters closely correspond to those at which soft x-ray lasing was realized [51]. An inversion at the operating transition was achieved via a fast, three-body recombination process in the presence of intense laser pumping. Lasing at the considered transition in C VI was theoretically predicted by Princeton's group [37] and currently has been studied experimentally by the same group.

As it was discussed in the previous section, the pulse duration scales as $1/\Omega$ with IR or optical field frequency. Thus, increasing modulating field frequency might lead to formation of shorter pulses. However, there is a physical limit to it, namely, keeping the same modulation index $P_\omega = \frac{d_{av}\tilde{E}_{\mathrm{IR/opt}}}{\Omega}$ requires an increase in the IR or optical field strength with an increase of Ω, which leads to faster ionization of ions from the excited energy levels, and accordingly to vanishing population inversion and amplification of the x-ray field. The ionization rate from excited states Γ_{ion} grows exponentially with IR or optical field strength and, for $P_\omega = 4.45$, it reaches the magnitude of inverse incident soft x-ray pulse duration (which is chosen to be a few times smaller than the lifetime of population inversion in the absence of modulating field) for an IR wavelength of approximately 1660 nm in Li III and for an optical wavelength of approximately 440 nm in C VI. With further decrease of IR or optical field wavelength and, accordingly, an increase of its intensity, the large ionization rate leads to fast depletion of excited levels of the ions, making amplification impossible. This sets the lower limit for the duration of the pulses produced via this technique in an active medium.

We consider dressing of Li III ions by a quasimonochromatic 1900-nm IR field with intensity 4×10^{14} W/cm^2 and modulation of C VI ions by a quasimonochromatic 500-nm optical field with intensity 2.3×10^{16} W/cm^2. The chosen parameters both in Li III and C VI correspond to a value of the modulation index of $P_\omega = 4.45$.

Equations (1), (7), (8), and (11) were numerically solved for the chosen parameters. Contrary to the case of a passive medium, where for the value of the modulation index $P_\omega = 4.45$, all sidebands are produced in phase with each other and with the incident x-ray field. In an active medium for $P_\omega = 4.45$ there is a π phase shift between the incident (resonant) spectral component with respect to the sidebands. However, the plasma dispersion allows one to compensate for this phase shift at a specific length of the medium. Namely, there is a difference between phase velocities of the resonant x-ray field and the modulating IR or optical field due to the plasma dispersion. It leads to a phase shift between the spectral components of the field and the corresponding spectral components of the resonant polarization. This phase shift grows with the propagation distance and with the sideband number; the spectral components on different sides from the resonance acquire phase shifts of different signs, while the central spectral component has no shift. As it follows from the results of numerical simulations, the resonant (incident, central) spectral component of the x-ray field remains dominant during the whole process of pulse formation in the amplifying medium. Therefore, one may neglect rescattering of the generated sidebands and consider the resonant polarization of the medium to be determined solely by the incident spectral component. In this case, the phase shift between the sidebands of the x-ray field and the corresponding sidebands of the resonant polarization is equivalent to a phase shift between the sidebands of the field and its central spectral component. At a certain length of the medium, the sidebands become approximately phase-aligned with the central spectral component of the field. As a result, a train of well-shaped amplified attosecond pulses can be produced at the optimal length of the medium. In particular, a train of 960-as pulses with peak intensity about 20 times higher than a peak intensity of the incident pulse can be formed at length $L = 1.25$ mm in Li III (Fig. 4), while a train of 290-as pulses with peak intensity about 10 times higher than a peak intensity of an incident field can be formed at length $L = 2.7$ mm in C VI (Fig. 5). The estimated lengths of the active medium are experimentally achievable and are rather common for the x-ray lasers [1–6]. It is worth noting that a variation of the active medium length by about 10% does not appreciably influence the shape of the x-ray pulses. Further increase of the medium length leads to higher amplification, but the shape of the pulses becomes distorted.

A more general case when the incident field consists of both z- and y-polarized components and a saturation of the $|4\rangle \leftrightarrow |1\rangle$ and $|5\rangle \leftrightarrow |1\rangle$ transition by the y-polarized field takes place is studied in the Appendix. It is shown that the presence of a strongly amplified y-polarized component of an x-ray field does not produce any essential effect on amplification of a z-polarized pulse. It is due to the fact that this amplification originates from different populated excited states that the duration of an incident pulse is shorter than the gain duration, while the peak of a strongly amplified y-polarized component is delayed as compared to an incident pulse due to the superradiant character of amplification of the y-polarized field.

Note that an amplitude of the driving laser field $\tilde{E}_{\mathrm{IR/opt}}$ in formula (2) was considered as a constant. At the same time, rather high intensities of this field required for efficient modulation of the x-ray transition may be produced only in the pulsed regime. Therefore, our model implies that a pulse duration of the driving field largely exceeds the pulse duration of an incident x-ray field, and the pulses are synchronized in such a way that an x-ray pulse lags slightly behind the driving pulse. In its turn, an incident x-ray pulse duration is considered to be on the order of or larger than 100 fs (in order for its spectrum to fit the linewidth of the resonant transitions). The required intensities of the IR driving field 10^{14}–10^{16} W/cm^2 with a pulse duration in the range 300 fs to 10 ps are available both from commercial and homemade mode-locked lasers, such as Lu^{3+}-lasant (Nd^{3+},Yb^{3+},Er^{3+}) doped crystalline ceramics (yttrium aluminium garnet, yttrium lithium fluoride, yttrium orthovanadate, etc.), as well

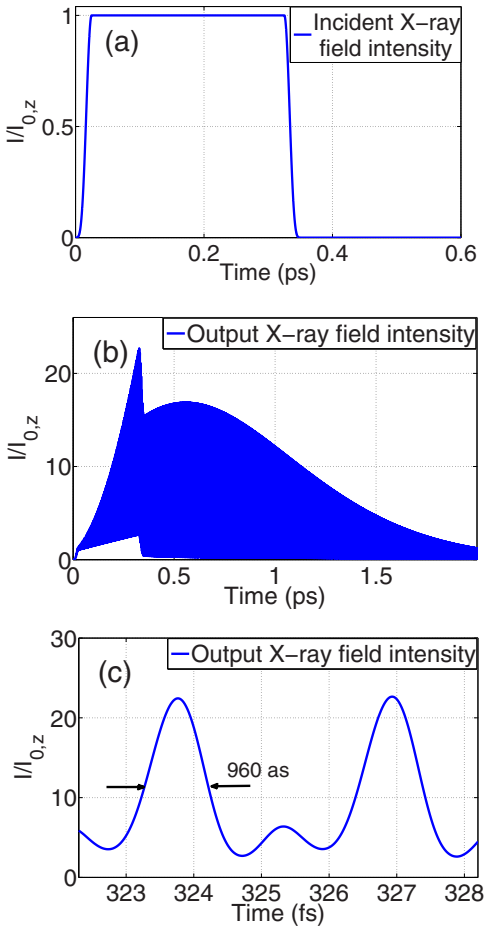

FIG. 4. Time dependence of an x-ray radiation intensity: (a) incident x-ray field, (b) output field, and (c) the same as (b) but showing only a small part of the whole envelope. The parameters of the inverted Li III plasma, an IR field, and an incident XUV field are provided in the text of the paper. An abrupt decrease in amplification at about 350 fs duration is due to the shortness of a seed pulse duration compared to the gain duration.

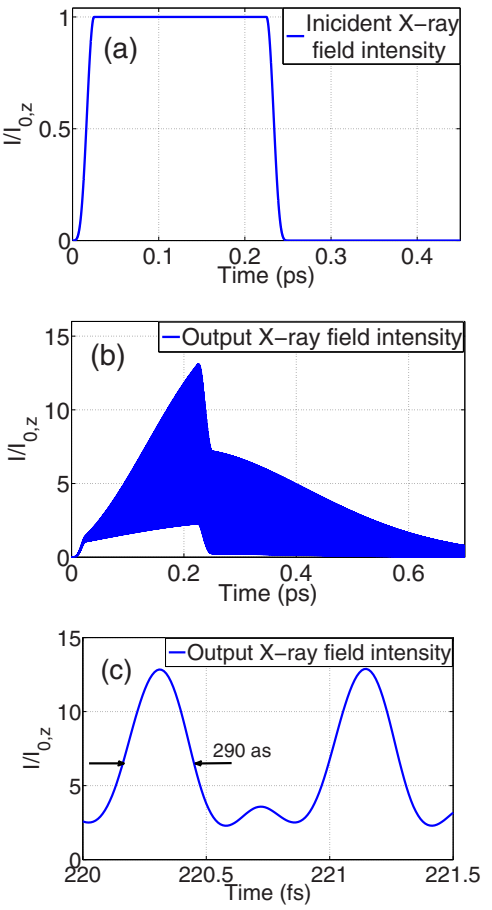

FIG. 5. Time dependence of an x-ray radiation intensity: (a) incident x-ray field, (b) output field, and (c) the same as (b) but showing only a small part of the whole envelope. The parameters of the inverted C VI plasma, optical field, and an incident XUV field are provided in the text of the paper. An abrupt decrease in amplification at about 250 fs duration is due to the shortness of a seed pulse duration compared to the gain duration.

as chirped pulse amplifiers (CPA) and optical parametric chirped pulse amplifiers (OPCPA) [57–61].

V. CONCLUSIONS

We studied two different paths on the way to producing intense coherent sub-fs x-ray radiation: (i) via efficient transformation of intense x-ray laser radiation into a train of sub-fs pulses, and (ii) via amplification of the single high-harmonic radiation accompanied by formation of sub-fs pulses. We have shown that both paths can be implemented by using the same technique, namely, via interaction of x-ray radiation with a plasma of the hydrogenlike ions with a modulated resonant transition. The first path implies using a noninverted medium while the second path requires using an inverted medium.

Since a limitation on the maximum intensity of the modulated field (and accordingly on the minimum modulation frequency for a given modulation index) is less stringent in the passive medium with respect to an active medium, shorter pulses can achieved via the first path.

It would be interesting to extend the obtained results to the wider range of ions. In the case of non-hydrogen-like ions, dressing by an IR or optical field is not reduced to a shift and modulation of the frequency of the resonant transition. Instead, it results in an effective modulation of its dipole moment [62], which also allows for very efficient transformation of a few hundred femtoseconds of x-ray laser radiation into the trains of sub-fs pulses in a passive medium of He or He-like ions [63]. One may expect that similar results can be obtained in the active medium.

Another interesting extension of this work would be seeding of an x-ray laser with a set of high harmonics rather than a single harmonic. Indeed, modulation of an active medium should allow for amplification of a set of harmonics within the bandwidth determined by the modulation depth of the parameters (frequency or dipole moment) of the resonant transition by the IR or optical field.

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APPENDIX: GENERAL CASE OF AN ARBITRARY POLARIZED INCIDENT FIELD

It is worth noting that the transitions $|1\rangle \leftrightarrow |4\rangle, |1\rangle \leftrightarrow |5\rangle$, which are not modulated by a z-polarized IR or optical field via the linear Stark effect, have nonzero dipole moment along y direction. They lead to amplification of a y-polarized component of an incident field, however, without formation of the sub-fs pulses, since the frequencies of these transitions are not affected by z-polarized IR or optical field. The modulation of $|1\rangle \leftrightarrow |2\rangle, |1\rangle \leftrightarrow |3\rangle$ transitions effectively reduces their dipole moments as compared to $|1\rangle \leftrightarrow |4\rangle, |1\rangle \leftrightarrow |5\rangle$ transitions by $J_0(P_\omega)$ times, resulting in smaller magnitude of the gain at those transitions. For the considered above value of modulation index $P_\omega = 4.45$ the effective dipole moment at the modulated transitions is approximately 3 times smaller compared to the unperturbed transitions. It results in approximately 9 times lower gain and in approximately 9 times higher saturation intensity for the modulated transition. Thus, even in the case of a high ratio between z-polarized and y-polarized components of the incident field, saturation of the unmodulated transitions occurs at the shorter distances than that of the modulated transitions. The saturation of the unmodulated transitions leads to an increase of the ground state population due to population depletion of levels $|4\rangle$ and $|5\rangle$, resulting in a decrease of population inversion at each of the two transitions $|1\rangle \leftrightarrow |2\rangle$ and $|1\rangle \leftrightarrow |3\rangle$ and, accordingly, to further reduced gain for a z-polarized component of an incident x-ray radiation.

Therefore, it is important to investigate an influence of an y-polarized component on amplification of a z-polarized component within a general 5-level model, taking all four excited degenerate levels into account.

Let us assume now that at the entrance to the medium, $x = 0$, the x-ray radiation has the form

$$\vec{E}_{X\text{-ray, inc}}(t) = \tfrac{1}{2}\vec{z}_0\tilde{E}_{z,0}(t)\exp\{-i\,\omega_{z,0}t\} + \tfrac{1}{2}\vec{y}_0\tilde{E}_{y,0}(t)\exp\{-i\,\omega_{y,0}t\} + \text{c.c.}, \quad \text{(A1)}$$

where $E_{z,0}$ and $E_{y,0}$ are the slowly varying amplitudes of z-polarized and y-polarized components of the incident radiation, $\omega_{z,0}$ and $\omega_{y,0}$ are their carrier frequencies, and c.c. stands for complex conjugation. The radiation (A1) is chosen to be near resonant to a transition from the ground state $|1\rangle$ to the first excited bound atomic energy level (which corresponds to four degenerate excited states, $|2\rangle$, $|3\rangle$, $|4\rangle$, $|5\rangle$, in the absence of the IR or optical field), $\omega_0 \approx \omega_{21}^0$ (where ω_{21}^0 is the unperturbed frequency of the resonant transition).

Propagation of x-ray radiation through the medium is described by the wave equation

$$\frac{\partial^2 \vec{E}_{X\text{-ray}}}{\partial x^2} - \frac{\varepsilon}{c^2}\frac{\partial^2 \vec{E}_{X\text{-ray}}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 \vec{P}}{\partial t^2}, \quad \text{(A2)}$$

where $\vec{E}_{X\text{-ray}} = \vec{z}_0 E_z + \vec{y}_0 E_y$ is the x-ray field strength and $\vec{P}$ is the resonant polarization.

We take into account now the following states of atomic hydrogen: the ground state $|1s\rangle$, denoted as $|1\rangle$, and four excited states: $|2\rangle = (|2s\rangle + |2p,m=0\rangle)/\sqrt{2}$, $|3\rangle = (|2s\rangle - |2p,m=0\rangle)/\sqrt{2}$, $|4\rangle = |2p,m=1\rangle$, and $|5\rangle = |2p,m=-1\rangle$, which are degenerate in the absence of the IR or optical field. Within this approximation, the resonant polarization is defined by density matrix elements ρ_{ij},

$$\vec{P}(\vec{r},t) = N(\vec{d}_{12}\rho_{21} + \vec{d}_{13}\rho_{31} + \vec{d}_{14}\rho_{41} + \vec{d}_{15}\rho_{51} + \text{c.c.}), \quad \text{(A3)}$$

where N is ion concentration and $\vec{d}_{ij}$ is dipole moment between states $|i\rangle$ and $|j\rangle$. Within the five-level model, all the nonzero dipole moments are given by

$$\begin{aligned}
\vec{d}_{12} &= \vec{d}_{1s\leftrightarrow 2p,m=0}/\sqrt{2} = \vec{z}_0 d_\parallel,\\
\vec{d}_{13} &= -\vec{d}_{1s\leftrightarrow 2p,m=0}/\sqrt{2} = -\vec{z}_0 d_\parallel,\\
\vec{d}_{22} &= \vec{d}_{2s\leftrightarrow 2p,m=0} = \vec{z}_0 d_{av},\\
\vec{d}_{33} &= -\vec{d}_{2s\leftrightarrow 2p,m=0} = -\vec{z}_0 d_{av},\\
\vec{d}_{14} &= \vec{d}_{1s\leftrightarrow 2p,m=1} = i\vec{y}_0 d_\perp,\\
\vec{d}_{15} &= \vec{d}_{1s\leftrightarrow 2p,m=-1} = i\vec{y}_0 d_\perp,\\
\vec{d}_{ij} &= \vec{d}_{ji}^{\,*}.
\end{aligned} \quad \text{(A4)}$$

In atomic units $d_\parallel = d_\perp = \frac{2^7}{3^5 Z}$ and $d_{av} = 3/Z$, where Z is the ion nucleus charge. Under the action of both x-ray and IR or optical fields, the evolution of density matrix elements is determined by the following equations:

$$\begin{aligned}
\dot{\rho}_{11} &= +\gamma_{11}(\rho_{22} + \rho_{33} + \rho_{44} + \rho_{55}) - i[\mathrm{H},\rho]_{11}\\
\dot{\rho}_{ij} &= -\gamma_{ij}\rho_{ij} - i[\mathrm{H},\rho]_{ij}, \quad ij \neq 11
\end{aligned}$$

$$\mathrm{H} = \begin{pmatrix}
\omega_1 & -E_z d_\parallel & E_z d_\parallel & i E_y d_\perp & i E_y d_\perp\\
-E_z d_\parallel & \omega_2 - \tilde{E}_{\mathrm{IR/opt}}\cos\{-i\,\Omega(t - x n_{pl}/c)\} & 0 & 0 & 0\\
E_z d_\parallel & 0 & \omega_3 + \tilde{E}_{\mathrm{IR/opt}}\cos\{-i\,\Omega(t - x n_{pl}/c)\} & 0 & 0\\
-i E_y d_\perp & 0 & 0 & \omega_4 & 0\\
-i E_y d_\perp & 0 & 0 & 0 & \omega_5
\end{pmatrix}. \quad \text{(A5)}$$

Here ω_i is the energy of atomic state $|i\rangle$. With the quadratic Stark effect taken into account, $\omega_1 = -\frac{Z^2}{2}(1+\frac{9}{256}F_c^2)$, $\omega_2 = \omega_3 = -\frac{Z^2}{8}(1+\frac{7}{4}F_c^2)$, $\omega_4 = \omega_5 = -\frac{Z^2}{8}(1+\frac{39}{8}F_C^2)$, where $F_c = (\frac{2}{Z})^3\tilde{E}_{\mathrm{IR/opt}}$ [43].

The decay rates γ_{ij} are defined as $\gamma_{12}=\gamma_{13}=\gamma_{\mathrm{coll}}+\Gamma_{\mathrm{ion}}/2+\Gamma_{\mathrm{radiative}}/2$, $\gamma_{14}=\gamma_{15}=\gamma_{\mathrm{coll}}+\Gamma_{\mathrm{ion},2}/2+\Gamma_{\mathrm{radiative}}/2$, $\gamma_{23}=\gamma_{\mathrm{coll}}+\Gamma_{\mathrm{ion}}+\Gamma_{\mathrm{radiative}}$, $\gamma_{24}=\gamma_{25}=\gamma_{34}=\gamma_{35}=\gamma_{\mathrm{coll}}+\Gamma_{\mathrm{ion}}/2+\Gamma_{\mathrm{ion},2}/2+\Gamma_{\mathrm{radiative}}$, $\gamma_{45}=\gamma_{\mathrm{coll}}+\Gamma_{\mathrm{ion},2}+\Gamma_{\mathrm{radiative}}$, $\gamma_{22}=\gamma_{33}=\Gamma_{\mathrm{ion}}+\Gamma_{\mathrm{radiative}}$, $\gamma_{44}=\gamma_{55}=\Gamma_{\mathrm{ion},2}+\Gamma_{\mathrm{radiative}}$, $\gamma_{11}=\Gamma_{\mathrm{radiative}}$, where γ_{coll} and Γ_{ion} are collisional broadening and ionization decay rates, respectively. $\Gamma_{\mathrm{radiative}}$ are radiative decay rates, which can be found in Ref. [48]. Collisional broadening was estimated according to [49,50]

$$\Gamma_{\mathrm{ion}} \approx \frac{Z^2}{16}\sqrt{\frac{3F_c}{\pi}}\left[\left(\frac{4}{F_c}\right)e^{+3}+\left(\frac{4}{F_c}\right)^3 e^{-3}\right]\exp\left\{-\frac{2}{3F_c}\right\},$$

and $\Gamma_{\mathrm{ion},2}$ can be found using Popov-Perelomov-Terentiev equations [64]. We neglect Doppler broadening of transitions since we consider ion temperatures on the order of 1 eV, as is typical for recombination x-ray plasma lasers [37], resulting in Doppler broadening comparable to or less than collisional broadening. It is worth noting that for the parameters we consider in the paper, $\gamma_{ij} \ll \Omega$.

Let us seek a partial solution in the form

$$\begin{aligned}
\vec{E}(x,t) &= \tfrac{1}{2}(\vec{y}_0\tilde{E}_y(x,t)+\vec{z}_0\tilde{E}_z(x,t))e^{-i(\omega t-kx)}+\mathrm{c.c.},\\
\rho_{12}(x,t) &= \tilde{\rho}_{12}(x,t)e^{i\omega t-ikx},\\
\rho_{13}(x,t) &= \tilde{\rho}_{13}(x,t)e^{i\omega t-ikx},\\
\rho_{14}(x,t) &= \tilde{\rho}_{14}(x,t)e^{i\omega t-ikx},\\
\rho_{15}(x,t) &= \tilde{\rho}_{15}(x,t)e^{i\omega t-ikx},\\
\rho_{ij}(x,t) &= \tilde{\rho}_{ij}(x,t), \quad ij \neq 12,13,14,15,
\end{aligned} \tag{A6}$$

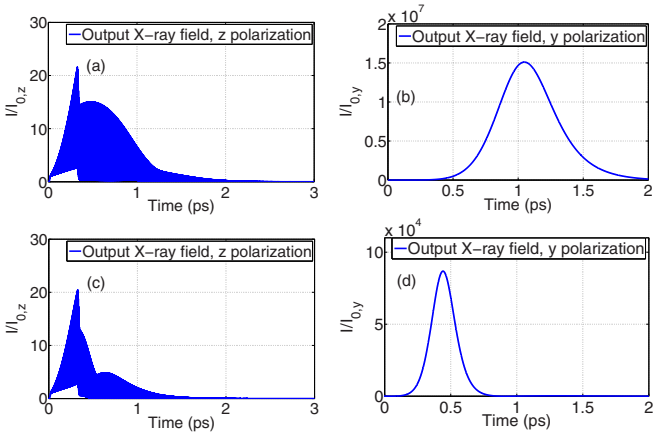

FIG. 6. Time dependence of intensities of z-polarized (a), (c) and y-polarized (b), (d) components of x-ray radiation after propagation through 1.25 mm of inverted Li III plasma. The parameters of the plasma, IR field, and envelope of the incident field are the same as in Fig. 4. (a) and (b) correspond to $I_y(x=0)=10^3$ W/cm^2, (c) and (d) correspond to $I_y(x=0)=10^6$ W/cm^2.

where $\tilde{E}_v(x,t)$, $v=\overline{y,z}$, and $\tilde{\rho}_{ij}(x,t)$ are the slowly varying amplitudes of the polarization components of the field and the density matrix elements, respectively, which means $|\frac{1}{\tilde{E}_v}\frac{\partial\tilde{E}_v}{\partial t}| \ll \omega$, $|\frac{1}{\tilde{E}_v}\nabla\tilde{E}_v| \ll k$, and $|\frac{1}{\tilde{\rho}_{ij}(\vec{r},t)}\frac{\partial\tilde{\rho}_{ij}(\vec{r},t)}{\partial t}| \ll \omega$, $|\frac{1}{\tilde{\rho}_{ij}(\vec{r},t)}\nabla\tilde{\rho}_{ij}(\vec{r},t)| \ll k$. In such a case, within the rotating wave approximation and approximation of plane waves, we get

$$\begin{aligned}
k &= \frac{\omega}{c},\\
\frac{\partial\tilde{E}_z}{\partial x} &= i4\pi\frac{\omega N d_\parallel}{c\sqrt{\varepsilon}}(\rho_{21}-\rho_{31}),\\
\frac{\partial\tilde{E}_y}{\partial x} &= -4\pi\frac{\omega N d_\perp}{c\sqrt{\varepsilon}}(\rho_{41}+\rho_{51}),
\end{aligned} \tag{A7}$$

and

$$\dot{\tilde{\rho}}_{11} = +\gamma_{11}(\tilde{\rho}_{22}+\tilde{\rho}_{33}+\tilde{\rho}_{44}+\tilde{\rho}_{55})+\frac{i\tilde{E}_z^* d_\parallel}{2}(\tilde{\rho}_{21}-\tilde{\rho}_{31})+\frac{i\tilde{E}_z d_\parallel}{2}(\tilde{\rho}_{13}-\tilde{\rho}_{12})-\frac{\tilde{E}_y d_\perp}{2}(\rho_{14}+\rho_{15})-\frac{\tilde{E}_y^* d_\perp}{2}(\rho_{41}+\rho_{51})$$

$$\dot{\tilde{\rho}}_{22} = -\gamma_{22}\tilde{\rho}_{22}+\frac{i\tilde{E}_z d_\parallel}{2}\tilde{\rho}_{12}-\frac{i\tilde{E}_z^* d_\parallel}{2}\tilde{\rho}_{21}$$

$$\dot{\tilde{\rho}}_{33} = -\gamma_{33}\tilde{\rho}_{33}+\frac{i\tilde{E}_z^* d_\parallel}{2}\rho_{31}-\frac{i\tilde{E}_z d_\parallel}{2}\rho_{13}$$

$$\dot{\tilde{\rho}}_{44} = -\gamma_{44}\tilde{\rho}_{44}-i\tilde{\rho}_{14}\frac{i\tilde{E}_y d_\perp}{2}-i\tilde{\rho}_{41}\frac{i\tilde{E}_y^* d_\perp}{2}$$

$$\dot{\tilde{\rho}}_{55} = -\gamma_{55}\tilde{\rho}_{55}-i\tilde{\rho}_{15}\frac{i\tilde{E}_y d_\perp}{2}-i\tilde{\rho}_{51}\frac{i\tilde{E}_y^* d_\perp}{2}$$

$$\dot{\tilde{\rho}}_{12} = -\gamma_{12}\tilde{\rho}_{12}-i\tilde{\rho}_{12}\{\omega_1-\omega_2+\omega+\tilde{E}_{\mathrm{IR/opt}}d_{av}\cos(\Omega[\tau+(1-n_{pl})x/c])\}-\frac{i\tilde{E}_z^* d_\parallel}{2}(\rho_{32}-\rho_{22}+\rho_{11})+\frac{i\tilde{E}_y^* d_\perp}{2}(i\rho_{52}+i\rho_{42})$$

$$\dot{\tilde{\rho}}_{13} = -\gamma_{13}\tilde{\rho}_{13}-i\tilde{\rho}_{13}\{\omega_1-\omega_3+\omega-\tilde{E}_{\mathrm{IR/opt}}d_{av}\cos(\Omega[\tau+(1-n_{pl})x/c])\}-\frac{i\tilde{E}_z^* d_\parallel}{2}(-\rho_{23}+\rho_{33}-\rho_{11})+\frac{i\tilde{E}_y^* d_\perp}{2}(i\rho_{53}+i\rho_{43})$$

$$\dot{\tilde{\rho}}_{14} = -\gamma_{14}\tilde{\rho}_{14} - i\tilde{\rho}_{14}(\omega_1 - \omega_4 + \omega) - \frac{i\tilde{E}_z^* d_\parallel}{2}(-\rho_{24} + \rho_{34}) + \frac{i\tilde{E}_y^* d_\perp}{2}(i\rho_{44} + i\rho_{54} - i\rho_{11})$$

$$\dot{\tilde{\rho}}_{15} = -\gamma_{15}\tilde{\rho}_{15} - i\tilde{\rho}_{15}(\omega_1 - \omega_5 + \omega) - \frac{i\tilde{E}_z^* d_\parallel}{2}(-\rho_{25} + \rho_{35}) + \frac{i\tilde{E}_y^* d_\perp}{2}(i\rho_{55} + i\rho_{45} - i\rho_{11})$$

$$\dot{\tilde{\rho}}_{23} = -\gamma_{23}\tilde{\rho}_{23} - i\tilde{\rho}_{23}\{\omega_2 - \omega_3 - 2\tilde{E}_{\mathrm{IR/opt}} d_{av}\cos(\Omega[\tau + (1 - n_{pl})x/c])\} + \frac{i\tilde{E}_z^* d_\parallel}{2}\tilde{\rho}_{21} + \frac{i\tilde{E}_z d_\parallel}{2}\tilde{\rho}_{13}$$

$$\dot{\tilde{\rho}}_{24} = -\gamma_{24}\tilde{\rho}_{24} - i\tilde{\rho}_{24}\{\omega_2 - \omega_4 - \tilde{E}_{\mathrm{IR/opt}} d_{av}\cos(\Omega[\tau + (1 - n_{pl})x/c])\} + \frac{i\tilde{E}_z d_\parallel\tilde{\rho}_{14}}{2} + \frac{\tilde{E}_y^* d_\perp\tilde{\rho}_{21}}{2}$$

$$\dot{\tilde{\rho}}_{25} = -\gamma_{25}\tilde{\rho}_{25} - i\tilde{\rho}_{25}\{\omega_2 - \omega_5 - \tilde{E}_{\mathrm{IR/opt}} d_{av}\cos(\Omega[\tau + (1 - n_{pl})x/c])\} + \frac{i\tilde{E}_z d_\parallel\rho_{15}}{2} + \frac{\tilde{E}_y^* d_\perp\rho_{21}}{2}$$

$$\dot{\tilde{\rho}}_{34} = -\gamma_{34}\tilde{\rho}_{34} - i\tilde{\rho}_{34}\{\omega_3 - \omega_4 + \tilde{E}_{\mathrm{IR/opt}} d_{av}\cos(\Omega[\tau + (1 - n_{pl})x/c])\} - \frac{i\tilde{E}_z d_\parallel\tilde{\rho}_{14}}{2} + \frac{\tilde{E}_y^* d_\perp\tilde{\rho}_{31}}{2}$$

$$\dot{\tilde{\rho}}_{35} = -\gamma_{35}\tilde{\rho}_{35} - i\tilde{\rho}_{35}\{\omega_3 - \omega_5 + \tilde{E}_{\mathrm{IR/opt}} d_{av}\cos(\Omega[\tau + (1 - n_{pl})x/c])\} - \frac{i\tilde{E}_z d_\parallel\tilde{\rho}_{15}}{2} + \frac{\tilde{E}_y^* d_\perp\tilde{\rho}_{31}}{2}$$

$$\dot{\tilde{\rho}}_{45} = -\gamma_{45}\tilde{\rho}_{45} - i\tilde{\rho}_{45}(\omega_4 - \omega_5) - i\tilde{\rho}_{15}\frac{i\tilde{E}_y d_\perp}{2} - i\tilde{\rho}_{41}\frac{i\tilde{E}_y^* d_\perp}{2}, \tag{A8}$$

where we have introduced the local time $\tau = t - x/c$. Equations (A7) and (A8) along with initial conditions

$$\tilde{\rho}_{ij}(x, \tau = 0) = \tilde{\rho}_{ij}^{(0)} \tag{A9}$$

and boundary conditions (A1) describe propagation of the x-ray field in the x direction.

In order to model amplification of incident x-ray field (A1) in an inverted active medium of x-ray plasma-based lasers, we assume that at the moment $\tau = 0$ all the ions have equal probability to be in the excited states $|2\rangle, |3\rangle, |4\rangle, |5\rangle$ and there are no coherencies, namely, initial conditions (A9) become

$$\begin{aligned}\tilde{\rho}_{22}(x, \tau = 0) = \tilde{\rho}_{33}(x, \tau = 0) = \tilde{\rho}_{44}(x, \tau = 0) \\ = \tilde{\rho}_{55}(x, \tau = 0) = 0.25, \\ \tilde{\rho}_{ij}(x, \tau = 0) = 0, \quad ij \neq 22, 33, 44, 55.\end{aligned} \tag{A10}$$

The results of the numerical solution of this set of equations in Li III plasma for the same parameters of the plasma as in Fig. 4 and for different values of intensity of an incident y-polarized component (with carrier frequency resonant to the perturbed $|1\rangle \leftrightarrow |4\rangle$ transition) are presented in Fig. 6. For all the figures, z-polarized component incident intensity is $I_z(x = 0) = 10^8$ W/cm^2.

Comparison of Figs. 4(a), 6(a), and 6(c) clearly shows that the presence of a highly amplified y-polarized component has little effect on amplification of the seeding z-polarized field, the duration of which is a few times shorter than a polarization decay time of the resonant transition. This is due to the fact that the peak of the y-polarized field is essentially delayed as compared to the peak of an amplified z-polarized seeding pulse. The reason for this delay is that a y-polarized emission occurs in a superradiant (SR) regime. Indeed, the cooperative frequency [65] for the chosen parameters of the medium $\Omega_c = \sqrt{\frac{3Nc\Gamma_{\mathrm{radiative}}\lambda_{41}^2}{16\pi}} \approx 5 \times 10^{12}$ Hz (where λ_{41} is the $|4\rangle \leftrightarrow |1\rangle$ transition wavelength) exceeds the line broadening $1/T_2 \approx 2.4 \times 10^{12}$ Hz. As a result of this delay, saturation of the unmodulated transitions and consequent reduction of gain at the modulated transition influences only an amplified stage of free induction decay of z-polarized radiation. It is worth noting that an increase of intensity of a y-polarized component of an incident field results in reduction of both delay time and SR pulse duration due to the induced nature of the superradiant process, enhancing the influence of an amplified y-polarized component on the shape of the amplified free induction decay in the vicinity of the peak of a y-polarized SR pulse [see the cusp in Fig. 6(c)] due to gain reduction, followed by a restoration of the gain at the back edge of the SR pulse. The investigation of the time dependence of the z-polarized component both at $I_y(x = 0) = 10^3$ W/cm^2 and at $I_y(x = 0) = 10^6$ W/cm^2 [Figs. 6(a) and 6(c)] with much higher resolution reveals the same structure of an ultrashort pulse train with a duration of about 900 as, as in the case of $I_y(x = 0) = 0$ [Fig. 4(a)]. The amplitude of the pulses in the train is defined by the total radiation pulse envelope, according to Figs. 6(a) and 6(c). The shape of the pulses starts to be destroyed only in the tail of the pulse envelope in the vicinity of 1-ps time.

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