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GENERATION OF SHORT X-RAY PULSES

Influence of Detuning of the Seeding VUV Radiation from the Resonance on Formation of Subfemtosecond Pulses in the Active Medium of the Plasma-Based X-Ray Laser Dressed by an Intense IR Field

I. R. Khairulin[a, b, *], V. A. Antonov[a, c], M. Yu. Ryabikin[a, b, c], and O. A. Kocharovskaya[d]

[a] *Institute of Applied Physics, Russian Academy of Sciences, Nizhny Novgorod, 603950 Russia*
[b] *Nizhny Novgorod State University, Nizhny Novgorod, 603950 Russia*
[c] *Prokhorov General Physics Institute of the Russian Academy of Sciences, Moscow, 119991 Russia*
[d] *Department of Physics and Astronomy, Texas A&M Universityn, College Station, TX 77843-4242 USA*
**e-mail: khairulinir@ipfran.ru*

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Abstract—We have studied the influence of detuning of quasimonochromatic seeding vacuum ultraviolet (VUV)/X-ray radiation from the frequency of the inverted transition of the hydrogen-like active medium of the plasma-based X-ray laser dressed by an intense infrared (IR) laser field on the formation of a subfemtosecond pulse train. For the active medium (plasma) of Li^{2+} ions with the inverted transition wavelength of 13.5 nm, it is demonstrated that the use of the seeding radiation detuned from the resonance by double the frequency of the modulating field allows a fourfold increase in the peak intensity of the formed pulses relative to the case of the exact resonance, while the maximum contrast is preserved.

Keywords: subfemtosecond pulses, VUV radiation, plasma-based X-ray laser, strong optical field, Stark effect, generation of combination frequencies

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1. INTRODUCTION

To date great advances have been made in developing compact sources of vacuum ultraviolet (VUV) and soft X-ray radiation [1–7]. These developments are motivated by a wide range of possible applications of shortwave radiation in studies of atomic and molecular processes, in particular on the femto- and attosecond time scale with high spatial resolution, for diagnostics of dense plasma, studies of photochemical and biological processes, and various technological applications.

One of the promising laboratory sources of VUV/soft X-ray radiation is a plasma-based X-ray laser [4–7]. Its active medium is a plasma of multiply charged ions resulting from ionization of a material by an optical or IR laser field. Under certain conditions, during the plasma evolution the ions are predominantly at one of the excited energy levels. As a result, population inversion arises in the medium, and a VUV or soft X-ray pulse is formed depending on the type of ion. Note that plasma-based X-ray lasers are capable of generating shortwave radiation pulses with the energy of up to a few mJ, but the picosecond duration of the pulses limits their application to investigation and control of dynamics of ultrafast processes, which occur on a time scale comparable with or shorter than the optical field cycle.

Recently, we proposed a method of formation of a train of subfemtosecond pulses from the seeding quasimonochromatic resonant VUV/soft X-ray radiation in the hydrogen-like active medium of a plasma-based X-ray laser dressed by an intense optical or IR laser field [8–10]. As a result of the linear Stark effect, the energies of the excited states of the resonant ions oscillate in time and space following oscillations of the modulating optical/IR field. In addition, energy levels of the ions experience a constant shift due to the quadratic Stark effect. If the frequency of the seeding VUV/X-ray radiation coincides with the time-average frequency of the inverted transition of the medium in the laser field, combination spectral components separated from the incident resonant radiation frequency by even multiples of the modulating field frequency are generated during amplification of the seeding radiation due to modulation of the resonant transition frequency. Under certain conditions, the combination-frequency radiation is sufficiently intense and in phase with the amplified seeding radiation, which results in formation of subfemto-/attosecond pulse trains at the output of the active medium of the plasma-based X-ray laser. Note that all the above-mentioned works deal with the case where quasimonochromatic seeding radiation is resonant to the transition frequency of the active medium considering its changes caused by the quadratic Stark effect.

In this work we consider a more general formulation of the problem, when the incident quasimonochromatic radiation is detuned from the frequency of the inverted transition of the active medium (with allowance for its changes in the modulating field) by even multiples of the modulating field frequency, and analyze conditions of pulse formation in the hydrogen-like active medium of an X-ray laser based on resonant Li^{2+} ions with the inverted transition wavelength of 13.5 nm.

2. BASIC EQUATIONS

We will consider propagation of VUV radiation through the hydrogen-like active medium of a plasma-based X-ray laser, which has the form of a cylinder oriented along the x axis and is dressed by a linearly polarized laser field with the frequency Ω, much lower than frequencies of all transitions from resonant states of ions, and the amplitude $E_{\rm L}^{\rm (inc)}$

$$\mathbf{E}_{\rm L} = \mathbf{z}_0 E_{\rm L}^{\rm (inc)} \cos\left(\Omega t - \frac{\Omega n_{\rm pl}}{c} x\right). \qquad (1)$$

The laser radiation propagates along the x axis and is polarized along the z axis; c is the speed of light in vacuum, $n_{\rm pl} = \sqrt{1 - \omega_{\rm pl}^2/\Omega^2}$ is the refractive index of the plasma at the laser field frequency, $\omega_{\rm pl} = \sqrt{4\pi N_{\rm e} e^2/m_{\rm e}}$ is the (electron) plasma oscillation frequency, $N_{\rm e}$ is the concentration of free electrons in the plasma, and $m_{\rm e}$ and e are the electron mass and charge, respectively. It is worth noting that at the frequencies Ω under consideration the laser field (1) does not undergo considerable distortion and retains its characteristics while propagating in the medium.

Next, we assume that the VUV radiation at the entrance to the medium is a semi-infinite pulse with the amplitude $E_{\rm XUV}^{\rm (inc)}$ and the carrier frequency ω linearly polarized along the z axis, the front edge of which is incident on the medium at the time instant $t = 0$

$$\mathbf{E}_{\rm XUV}(x = 0, t) = \frac{1}{2}\mathbf{z}_0 E_{\rm XUV}^{\rm (inc)} \theta(t) e^{-i\omega t} + {\rm c.c.}, \qquad (2)$$

where $\theta(t)$ is the unit Heaviside step function. Note that we further assume that (i) the VUV radiation frequency is so high that the VUV radiation is not affected by plasma dispersion, and its phase velocity is equal to the speed of light in vacuum and (ii) the carrier frequency of the VUV radiation is close to the inverted transition frequency, which results in resonant interaction of the VUV radiation with the active medium.

The active medium of the plasma-based X-ray laser is taken to be plasma consisting of hydrogen-like Li^{2+} ions with the concentration $N_{\rm i} = 1.5 \times 10^{17}$ cm^{-3} and free electrons with the concentration $N_{\rm e} = 2N_{\rm i} = 3 \times 10^{17}$ cm^{-3}. We also assume that the characteristic temperature is 1 eV for ions and 2 eV for electrons. The active plasma medium with similar characteristics was experimentally obtained in [11]. In the medium under consideration, population inversion is achieved at the transition between the ions energy levels with $n = 2$ and $n = 1$, where n is the principal quantum number. For Li^{2+} ions the wavelength of the radiation resonant to the inverted transition is $\lambda_{\rm tr} \approx 13.5$ nm. At the considered ion and electron concentrations and temperatures, characteristic times of the collisional and emission relaxations at the inverted transition of ions are $\gamma_{\rm C}^{-1} \simeq 0.425$ ps [12] and $\Gamma_{\rm R}^{-1} \simeq 19.7$ ps, respectively.

Under the action of the laser field (1), the upper fourfold degenerate ion energy level is split into three sublevels due to the Stark effect. In the parabolic coordinate system with the main axis directed along the laser field polarization (z axis), two sublevels correspond to the states $\left(|2s\rangle + |2p, m = 0\rangle\right)/\sqrt{2} \equiv |2\rangle$ and $\left(|2s\rangle - |2p, m = 0\rangle\right)/\sqrt{2} \equiv |3\rangle$. Their energies follow the instantaneous value of the electric field (1) in space and time, i.e., they oscillate at the optical frequency due to the linear Stark effect and get a constant shift due to the quadratic Stark effect

$$\begin{aligned} \omega_2(t, x) &= \omega_2^{\rm (aver)} - \Delta\omega \cos\left(\Omega t - \frac{\Omega n_{\rm pl}}{c} x\right), \\ \omega_3(t, x) &= \omega_3^{\rm (aver)} + \Delta\omega \cos\left(\Omega t - \frac{\Omega n_{\rm pl}}{c} x\right), \end{aligned} \qquad (3)$$

where $\hbar\omega_2^{\rm (aver)} = \hbar\omega_3^{\rm (aver)} = -m_{\rm e} e^4 Z^2 \left(1 + 21F^2/4\right)/8\hbar^2$ is the energy of the states $|2\rangle$ and $|3\rangle$ averaged over the laser field cycle; $\hbar\Delta\omega = 3m_{\rm e} e^4 Z^2 F/8\hbar^2$ is the amplitude of the excited ion energy level splitting due to the linear Stark effect; $F = (2/Z)^3 E_{\rm L}^{\rm (inc)}/E_{\rm a}$ is the normalized laser field amplitude; Z is the nuclear charge number of the ion, which is 3 for Li^{2+}; $\hbar$ is the Planck constant; and $E_{\rm a} \simeq 5.14 \times 10^9$ V/cm is the atomic unit of the electric field. Note that higher-order corrections to the state energies at the laser field intensities under consideration can be neglected. Dipole moments of the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$, where $|1\rangle \equiv |1s\rangle$ is the ground state of the hydrogen-like ions with the energy $\hbar\omega_1 = -m_{\rm e} e^4 Z^2 \left(1 + 9F^2/256\right)/2\hbar^2$, are oriented along the polarization axis of the modulating field (1) and VUV radiation (2), that is, along the z axis. Thus, these transitions are efficiently excited by the radiation (2), which leads to resonant polarization of the medium

$$\mathbf{P}(x,t) = N_{\mathrm{i}}\left(\rho_{21}\mathbf{d}_{12} + \rho_{31}\mathbf{d}_{13} + \text{c.c.}\right), \tag{4}$$

where $\mathbf{d}_{12} = \mathbf{z}_0 d$, $\mathbf{d}_{13} = -\mathbf{z}_0 d$, $d = (2^7/3^5)ea_0/Z$, a_0 is the Bohr radius, and ρ_{21} and ρ_{31} are the nondiagonal elements of the medium density matrix.

The remaining sublevel of the excited ion energy level is twofold degenerate and corresponds to the states $|2p, m=1\rangle \equiv |4\rangle$ and $|2p, m=-1\rangle \equiv |5\rangle$. The energies of these states experience only a constant shift caused by the quadratic Stark effect: $\hbar\omega_4 = \hbar\omega_5 = -m_{\mathrm{e}}e^4Z^2\left(1+39F^2/8\right)/8\hbar^2$. Dipole moments of the transitions $|4\rangle \to |1\rangle$ and $|5\rangle \to |1\rangle$ are oriented along the y axis, and thus the incident VUV radiation does not interact with these transitions. However, since at the initial time there is positive population difference at these transitions, amplified spontaneously emitted y-polarized radiation is generated in the medium. At the same time, as shown in [9, 10, 13], if the intensity of the seeding VUV radiation (2) is high enough, generation of amplified spontaneous emission is suppressed, and its effect on generation of combination spectral components and on amplification of the z-polarized VUV radiation can be neglected. In what follows, we assume that this condition is satisfied and exclude the states $|4\rangle$ and $|5\rangle$ from the consideration.

Further, we can simplify the equations to obtain an analytical solution for the VUV radiation at the exit from the modulated active medium. We assume that the VUV radiation interacts with the medium linearly, so that the variation in the population difference at the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$ can be neglected. Then, in the approximation of slowly varying amplitudes for the resonant field $E_{\mathrm{XUV}}(x,\tau)$ and the medium polarization $P(x,\tau)$, where $\tau = t - x/c$ is the local time, and in the rotating wave approximation for the density matrix elements, the system of equations describing space–time dynamics of the VUV radiation (2) and the medium polarization (4) in the modulated active Li^{2+} plasma takes the form

$$\begin{cases} \dfrac{\partial \tilde{E}_{\mathrm{XUV}}}{\partial x} = i\dfrac{4\pi\omega N_{\mathrm{i}} d}{c}\left(\tilde{\rho}_{21} - \tilde{\rho}_{31}\right), \\ \dfrac{\partial \tilde{\rho}_{21}}{\partial \tau} = \left[-i\left(\omega_{\mathrm{tr}}^{(\mathrm{aver})} - \omega\right) - \gamma + i\Delta\omega\cos\left(\Omega\tau - \Delta K x\right)\right]\tilde{\rho}_{21} - i\dfrac{dn_0}{2\hbar}\tilde{E}_{\mathrm{XUV}}, \\ \dfrac{\partial \tilde{\rho}_{31}}{\partial \tau} = \left[-i\left(\omega_{\mathrm{tr}}^{(\mathrm{aver})} - \omega\right) - \gamma - i\Delta\omega\cos\left(\Omega\tau - \Delta K x\right)\right]\tilde{\rho}_{31} + i\dfrac{dn_0}{2\hbar}\tilde{E}_{\mathrm{XUV}}, \end{cases} \tag{5}$$

where $\tilde{E}_{\mathrm{XUV}}$ and $\tilde{\rho}_{21}$, $\tilde{\rho}_{31}$ are the slowly varying amplitudes of the VUV radiation and the nondiagonal elements of the medium density matrix, respectively, $\omega_{\mathrm{tr}}^{(\mathrm{aver})} = \omega_2^{(\mathrm{aver})} - \omega_1 = \omega_3^{(\mathrm{aver})} - \omega_1$ is the frequency of the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$ averaged over the laser field cycle, $\Delta K = \Omega(1 - n_{\mathrm{pl}})/c$ is the addition to the wave number of the modulating laser field caused by the plasma dispersion, $\gamma = \gamma_0 + W_{\mathrm{ion}}/2$ is the decay rate of coherences at the transitions $|2\rangle \to |1\rangle$ and $|3\rangle \to |1\rangle$ which defines the characteristic relaxation time of the medium polarization response, $\gamma_0 = \gamma_{\mathrm{C}} + \Gamma_{\mathrm{R}}/2$ is the decay rate of the coherences in the absence of the modulating field, W_{ion} is the rate of the tunnel ionization from the states $|2\rangle$ and $|3\rangle$ under the action of the modulating field [14], $W_{\mathrm{ion}} = \dfrac{m_{\mathrm{e}}e^4Z^2}{16\hbar^3}\sqrt{\dfrac{3F}{\pi}}\left[\dfrac{4}{F}e^3 + \left(\dfrac{4}{F}\right)^3 e^{-3}\right]e^{-2/(3F)}$, and n_0 is the initial population difference at the inverted transition, which is taken to be 0.25. In what follows, we assume that the laser field frequency is much higher than the medium polarization decay rate, $\Omega/\gamma \gg 1$.

Unlike the case in [8–10], where generation of combination spectral components and formation of a subfemtosecond pulse train were considered under the condition that the frequency of the incident quasi-monochromatic VUV radiation was equal to the time-average frequency of the resonant transition, $\omega = \omega_{\mathrm{tr}}^{(\mathrm{aver})}$, in this work we consider a more general case where the incident radiation is detuned from the resonance by an even multiple of the modulating field frequency, $\omega = \omega_{\mathrm{tr}}^{(\mathrm{aver})} + 2k_{\mathrm{inc}}\Omega$.

3. ANALYTICAL SOLUTION

Further, similar to [9, 10], we will seek the solution for the slowly varying amplitude of the electric field of the VUV radiation in the form

$$\tilde{E}_{\mathrm{XUV}}(x,\tau) = \sum_{k=-\infty}^{\infty} \hat{E}_{\mathrm{XUV}}^{(k)}(x,\tau)\, e^{-i2k\Omega\tau}, \tag{6}$$

where $\hat{E}_{\mathrm{XUV}}^{(k)}(x,\tau)$ is the amplitude of the spectral component with number k separated from the frequency of the incident field (2) by $2k\Omega$ and from the frequency of the inverted transition by $2(k_{\mathrm{inc}} + k)\Omega$.

Substituting (6) into (5) and assuming that $\left|\hat{E}^{(k)}_{\text{XUV}}(x,\tau)\right| \gg \tau\left|\partial\hat{E}^{(k)}_{\text{XUV}}(x,\tau)/\partial\tau\right|$ [9] and $\Omega/\gamma \gg 1$, after some transformations we get a system of equations describing variation in the amplitudes $\hat{E}^{(k)}_{\text{XUV}}(x,\tau)$ of the spectral components during propagation in the modulated active plasma medium

$$\frac{\partial\hat{E}^{(k)}_{\text{XUV}}}{\partial x} = g_{\max}\left(1-e^{-\gamma\tau}\right) \times \sum_{n=-\infty}^{\infty} J_{2(k_{\text{inc}}+n)}\left(P_\Omega\right) J_{2(k_{\text{inc}}+k)}\left(P_\Omega\right) e^{i2(n-k)\Delta K x}\hat{E}^{(n)}_{\text{XUV}}, \tag{7}$$

where $g_{\max} = 4\pi\omega n_0 d^2 N_i/(\hbar c\gamma)$ is the active medium gain coefficient in the absence of the linear Stark effect caused by the laser field, $P_\Omega = \Delta\omega/\Omega$ is the modulation index equal to the ratio between the amplitude of the linear Stark shift and the laser field frequency (and proportional to the ratio between the amplitude of the laser field and its frequency), and $J_n(x)$ is the Bessel function of the first kind of order n. Note that system of equations (7) describes variation in the amplitudes of the spectral components of the VUV radiation during its propagation in any hydrogen-like plasma medium. We will seek the solution of system (7) on the assumption that the amplitude of the VUV radiation at the frequency of the incident field is larger than the amplitudes of combination spectral components, i.e., $\left|\hat{E}^{(0)}_{\text{XUV}}(x,\tau)\right| > \left|\hat{E}^{(k)}_{\text{XUV}}(x,\tau)\right|$ for any k. This means that in the following we neglect scattering of combination spectral components into one another. Despite the fact that under certain conditions to be considered below, amplitudes of the combination spectral components are comparable with the field amplitude at the seeding radiation frequency, as shown in [9], the solution obtained within this approximation qualitatively correctly describes amplitudes and phases of the combination components. Thus, only one term corresponding to the index $n = 0$ should be left on the right-hand side of equations (7) and the solution can be written as

$$\hat{E}^{(0)}_{\text{XUV}}(x,\tau) = E^{(\text{inc})}_{\text{XUV}}\theta(\tau)\exp\left[g_{\max}J^2_{2k_{\text{inc}}}\left(P_\Omega\right)\left(1-e^{-\gamma\tau}\right)x\right], \tag{8a}$$

$$\hat{E}^{(k)}_{\text{XUV}}(x,\tau) = F_{\text{spectral}} \times F_{\text{spatial}} \times \hat{E}^{(0)}_{\text{XUV}}(x,\tau), \tag{8b}$$

where

$$F_{\text{spectral}} = \frac{g_{\max}}{2\Delta K}\left(1-e^{-\gamma\tau}\right)J_{2k_{\text{inc}}}\left(P_\Omega\right)J_{2(k_{\text{inc}}+k)}\left(P_\Omega\right), \tag{8c}$$

$$F_{\text{spatial}} = \frac{\exp(-i2k\Delta K x) - \exp\left[-g_{\max}J^2_{2k_{\text{inc}}}\left(P_\Omega\right)(1-e^{-\gamma\tau})x\right]}{\left(g_{\max}/2\Delta K\right)J^2_{2k_{\text{inc}}}(P_\Omega)(1-e^{-\gamma\tau}) - ik}. \tag{8d}$$

According to (8a), during the propagation through the medium the incident VUV radiation is amplified with the effective gain coefficient $g_{\max}J^2_{2k_{\text{inc}}}(P_\Omega)(1-e^{-\gamma\tau})$, which depends on (i) time due to finiteness of the medium polarization response time, (ii) the modulation index P_Ω, and (iii) the value of VUV radiation detuning from the frequency $\omega^{(\text{aver})}_{\text{tr}}$, which is characterized by the index k_{inc}. A decrease in the VUV radiation gain coefficient by a factor of $J^2_{2k_{\text{inc}}}(P_\Omega)$ relative to the active medium gain coefficient in the absence of the modulating field, $g_{\max}$, is due to the fact that under the action of the laser field the medium gain is distributed over the combination frequencies spaced at the double frequency of the laser field [13]. As a result, the effective gain coefficient for each of the combination frequencies with number k_{inc} turns out to be smaller than the gain coefficient of the unmodulated active medium.

In addition, according to (8b), combination spectral components of the VUV radiation are generated in the medium at frequencies $\omega_k = \omega^{(\text{aver})}_{\text{tr}} + 2(k_{\text{inc}} + k)\Omega$. As shown in [8–10], they result from coherent scattering of the seeding radiation by the modulation wave moving with the phase velocity of the optical/IR field. The amplitude of the kth combination spectral component of the field is defined by the amplitude of the central spectral component $\hat{E}^{(0)}_{\text{XUV}}(x,\tau)$ with allowance for its gain. The amplitudes of the combination components depend on two factors discussed below.

One of them, F_{spectral} (8c), has the meaning of the efficiency of the kth spectral component generation in the scattering of the seeding radiation by the modulation wave. As is seen in (8c), this efficiency is determined by (i) detuning of the incident field from the resonance (index k_{inc}), (ii) detuning of the kth spectral component from the incident field frequency (index k), (iii) the ratio between the unperturbed medium gain coefficient and the addition to wave number of the modulating field due to plasma dispersion, $g_{\max}/\Delta K$, and (iv) the modulation index, P_Ω. Also, according to (8c), the efficiency of generation of combination components increases with forming medium polarization response. If the concentration of free electrons in the plasma is high, and the inequality $g_{\max}/\Delta K \ll 1$ holds, the efficiency of generation of combination spectral components is close to zero; in other words, coherent scattering of the incident field by the modulation wave is suppressed. It also follows from (8c) that $F_{\text{spectral}}(-k_{\text{inc}},-k) = F_{\text{spectral}}(k_{\text{inc}},k)$. This means that the change of the seeding radiation frequency from $\omega^{(\text{aver})}_{\text{tr}} + 2k_{\text{inc}}\Omega$ to $\omega^{(\text{aver})}_{\text{tr}} - 2k_{\text{inc}}\Omega$, i.e., the change of the sign of the detuning from resonance with preservation of its value leads to mirror reflection of the scattering efficiency spectrum relative to the frequency of the incident VUV radiation.

Let us now analyze dependence of the spectral scattering efficiency F_{spectral} on the modulation index P_Ω at different values of detuning from the incident field frequency, i.e., different k_{inc}. The following remark should be made here. From the definition of the modulation index it follows that it can be increased in two ways, by increasing the intensity of the modulating field and/or by decreasing its frequency, i.e., by increasing its wavelength $\Lambda = 2\pi c/\Omega$. However, the following restrictions have to be applied to the characteristics of the modulating field. Its intensity should be below the ionization threshold from the excited states of the active medium. Otherwise, populations of excited states, and thus the medium gain coefficient $g_{\max}$, will decrease because of fast ionization, which, according to solution (8), will lead to a drop in efficiency of both the VUV radiation gain and the generation of the combination spectral components. In the case of the hydrogen-like Li^{2+} plasma, the maximum permissible intensity of the modulating field is $I_L = c(E_L^{(\text{inc})})^2/8\pi = 4\times10^{14}$ W/cm^2, while $W_{\text{ion}}^{-1} \simeq$ 3.3 ps. In the following we consider this laser field intensity since it allows obtaining the largest absolute width of the spectrum of combination components. In the considered plasma medium with the concentration of free electrons $N_e \simeq 3\times10^{17}$ cm^{-3} the plasma frequency will correspond to the wavelength $\lambda_{\text{pl}} = 2\pi c/\omega_{\text{pl}} \approx 61$ μm. Thus, for the optical field with the wavelength of about 1 μm, the refractive index of the plasma will be close to unity. In this case, for the optical modulating field we can take $\Delta K \simeq \omega_{\text{pl}}^2/(2c\Omega)$ and recast (8c) in the equivalent form

$$F_{\text{spectral}} = \alpha J_{2k_{\text{inc}}}(P_\Omega) J_{2(k_{\text{inc}}+k)}(P_\Omega)/P_\Omega, \qquad (9)$$

where $\alpha = cg_{\max}\Delta\omega(1-e^{-\gamma\tau})/\omega_{\text{pl}}^2$.

Figures 1a–1f show dependences of quantity (9) on the modulation index at different values of detuning of the seeding VUV radiation (2) from the resonant frequency $\omega_{\text{tr}}^{(\text{aver})}$. In the case of the exact resonance, where $k_{\text{inc}} = 0$ and $\omega = \omega_{\text{tr}}^{(\text{aver})}$, the efficiency of generation of combination spectral components, and thus their spectrum, turn out to be symmetric functions of the combination frequency number k (see Fig. 1c). If $k_{\text{inc}} \neq 0$, for the modulation index values $P_\Omega \leq 5$, at which the highest amplitudes of the combination spectral components are achieved, the generated spectrum turns out to be asymmetric with respect to the index k and, as in the case of $k_{\text{inc}} = 0$, localized around the resonance frequency $\omega_{\text{tr}}^{(\text{aver})}$. It is also seen in Fig. 1 that the maximum efficiency of generation of combination spectral components is achieved at $k_{\text{inc}} = 0$ and $k_{\text{inc}} = \pm1$. In these cases the maximum possible value of $|F_{\text{spectral}}|/\alpha$ is about 0.09, while at $k_{\text{inc}} = \pm2$ it is three times smaller and at $k_{\text{inc}} = \pm3$ it is six times smaller. Moreover, when $|k_{\text{inc}}| \geq 2$, large modulation indices have to be used for generation of combination frequencies (Figs. 1a, 1d), which leads to a smaller effective gain coefficient of the VUV radiation (Fig. 1f).

Thus, generation of combination spectral components is most efficient when the incident quasimonochromatic VUV radiation is in resonance with the inverted transition of the medium (with allowance for the change in its frequency in the modulating filed), $\omega = \omega_{\text{tr}}^{(\text{aver})}$, or is detuned from the transition frequency by the double frequency of the modulating field, $\omega = \omega_{\text{tr}}^{(\text{aver})} \pm 2\Omega$. The corresponding optimum values of the modulation index P_Ω fall within the interval $0 \leq P_\Omega \leq 5$ (see Figs. 1b, 1c, 1e). The spectrum of the formed combination components is localized in the vicinity of the frequency $\omega_{\text{tr}}^{(\text{aver})}$.

Further we will consider the second factor influencing the combination spectral components of the field, i.e., the spatial dependence of the kth spectral component amplitude. According to (8b) and (8d), this dependence is defined by the product of the exponential function characterizing the incident VUV radiation gain and the function F_{spatial}. The features of this spatial dependence are described in [9], and we will not reproduce them here. Note only the following. At the exit from the optically thick medium with the thickness x satisfying the condition $g_{\max}J_{2k_{\text{inc}}}^2(P_\Omega)\,x \gg 1$, the factor F_{spatial} leads to a change in the phase of the kth spectral component by $-2k\Delta Kx + \arctan\left\{2k\Delta K\Big/\left[g_{\max}J_{2k_{\text{inc}}}^2(P_\Omega)\left(1-e^{-\gamma\tau}\right)\right]\right\}$ and to a decrease in the amplitude of this spectral component by a factor of $\sqrt{\left[\frac{g_{\max}}{2\Delta K}J_{2k_{\text{inc}}}^2(P_\Omega)\left(1-e^{-\gamma\tau}\right)\right]^2 + k^2}$. It is worth noting that the phase of the kth spectral component also depends on the sign of the factor F_{spectral}. Thus, when the condition $g_{\max}J_{2k_{\text{inc}}}^2(P_\Omega)x \gg 1$ is fulfilled, the phase of the kth combination spectral component of the VUV radiation relative to the radiation at the seeding frequency ($k = 0$) has the form

$$\varphi_k = \pi\delta_{-1,S} + \arctan\frac{2k\Delta K}{g_{\max}J_{2k_{\text{inc}}}^2(P_\Omega)\left(1-e^{-\gamma\tau}\right)}, \qquad (10)$$

where $S = \operatorname{sgn}(F_{\text{spectral}})$, $\operatorname{sgn}(x)$ is the sign of the quantity x, and $\delta_{y,z}$ is the Kronecker symbol. Note that in (10) the addition to the phase $-2k\Delta Kx$ linear in frequency, i.e., in k, is omitted, because it only leads to a shift of the temporal dependence of the amplified VUV radiation and is equivalent to a change in the time origin.

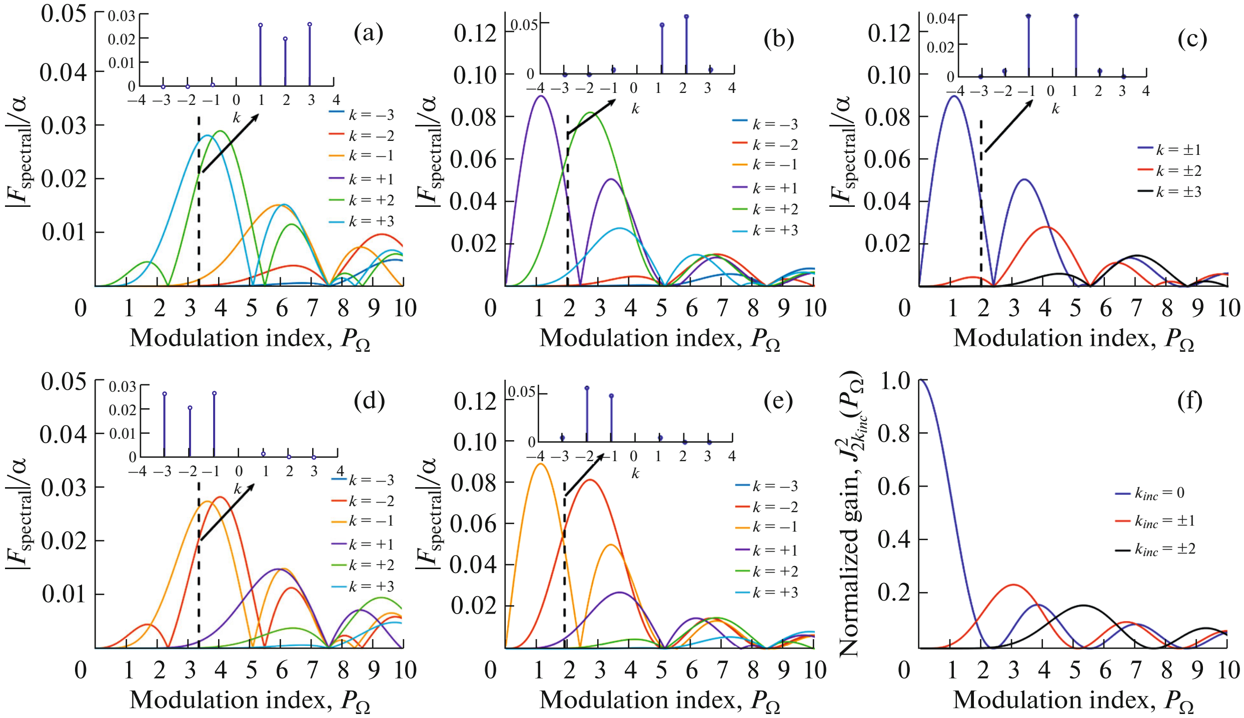

Fig. 1. (Color online) Modulus of the normalized efficiency of the combination frequency generation, $F_{spectral}/\alpha$, as a function of the modulation index, P_Ω, at the fixed laser field intensity $I_L = 4 \times 10^{14}$ W/cm^2 for different values of the seeding radiation detuning from the resonance: $k_{inc} = -2$ (a), $k_{inc} = -1$ (b), $k_{inc} = 0$ (c), $k_{inc} = 2$ (d), and $k_{inc} = 1$ (e). Insets: sections of the corresponding dependences at the fixed modulation index $P_\Omega = 3.4$ (a), $P_\Omega = 1.9$ (b), $P_\Omega = 2$ (c), $P_\Omega = 3.4$ (d), and $P_\Omega = 1.9$ (e). The normalization coefficient is $\alpha = cg_{max}\Delta\omega(1 - e^{-\gamma\tau})/\omega_{pl}^2$. Note that in Fig. 1a the lines corresponding to $k = 1$ and $k = 3$ coincide. The same is true for the $k = -1$ and $k = -3$ lines in Fig. 1d. (f) Dependence of the effective gain coefficient of the VUV radiation (8) on the modulation index for different k_{inc}.

4. ANALYSIS OF PULSE FORMATION CONDITIONS

Let us investigate optimum conditions for transformation of the quasimonochromatic seeding VUV radiation into a train of subfemtosecond pulses in the modulated plasma of excited Li^{2+} ions. Formation of pulses with high peak intensity and short duration requires maximization of amplitudes and matching of phases of spectral components of the field at the exit from the medium and generation of the maximum number of spectral components with a comparable amplitude.

As shown in the previous section, the highest generation efficiency (and maximum amplitudes) of the combination spectral components of the radiation are achieved at $k_{inc} = 0, \pm1$; the cases of $k_{inc} = 1$ and -1 only differ by the mirror reflection of the spectrum relative to the incident VUV carrier frequency ω and correspond to the same temporal dependence of the VUV radiation intensity. Consequently, we will consider only the cases of $k_{inc} = 0$ and $k_{inc} = 1$.

As a measure of phase ordering and of the number of radiation spectral components with the essentially nonzero amplitude, we use the contrast of the formed pulses equal to the ratio of the difference between the maximum and minimum intensities of the VUV radiation over the half-cycle of the modulating field to the average radiation intensity over the same interval of time. The thus defined contrast characterizes the pulse off-duty factor and shape: an increase in contrast corresponds to an increase in the off-duty factor and improvement of the pulse shape.

Figure 2a shows dependence of the VUV radiation contrast at the exit from the modulated active Li^{2+} plasma for $k_{inc} = 0$ on the modulation index P_Ω and the medium thickness x. The contrast is calculated on the basis of the analytical solution (6), (8) for the local times appreciably higher than the polarization response time, i.e., when $\gamma\tau \gg 1$. This dependence qualitatively agrees with the calculations [9] based on the numerical solution of the Maxwell—Bloch equations with allowance for the states $|4\rangle$ and $|5\rangle$ and the variation in the population difference at the inverted transitions. The analytical and numerical [9] solutions for the contrast are especially close to each other at medium thicknesses up to 2 mm in the region of appli-

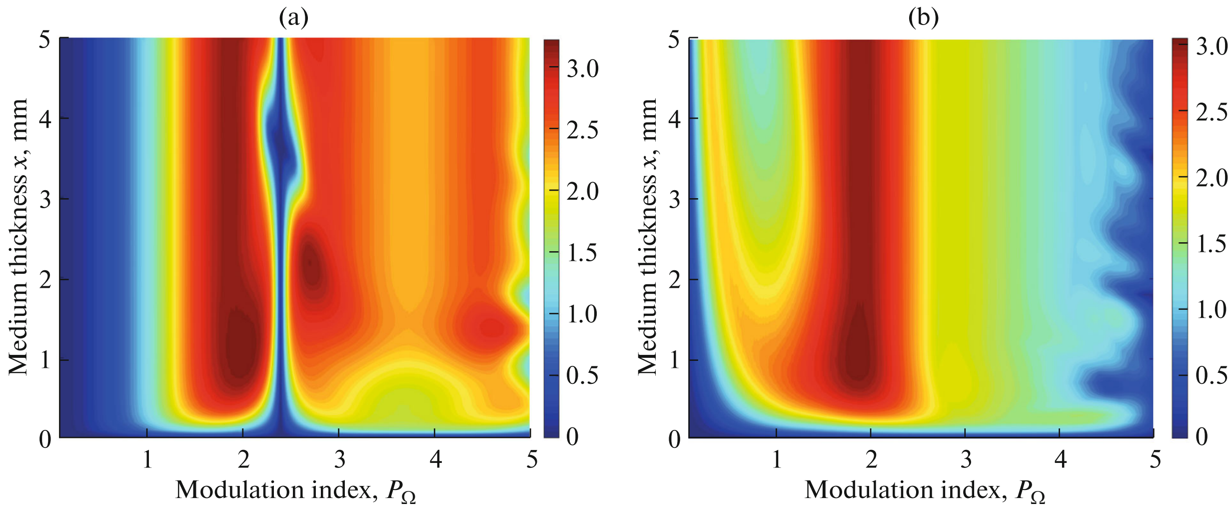

Fig. 2. (Color online) Dependence of the contrast of the VUV radiation (6), (8) at times $\tau \gg \gamma^{-1}$ on the medium thickness and the modulation index at $k_{\rm inc} = 0$ (a) and $k_{\rm inc} = 1$ (b); $g_{\max} \simeq 163.6$ cm^{-1}, $\alpha_{\gamma\tau\gg 1} \simeq 22.8$.

cability of the approximations used in this work. It is seen in Fig. 2a that the pulses with the highest contrast are formed when the parameters of the modulating field correspond to the modulation index $P_\Omega = 2$ (at the intensity $I_{\rm L} = 4\times 10^{14}$ W/cm^2 the required wavelength of the laser field is $\Lambda = 2\pi c/\Omega \simeq 850$ nm), and the medium thickness is $x = 1.1$ mm. In this case, the pulse contrast is about 3.3.

Figure 3a presents the temporal dependence of the output VUV radiation intensity for $k_{\rm inc} = 0$ and the optimum parameter values found above. As follows from the figure, the peak intensity of the formed pulses in the steady-state regime is about 47 times higher than the seeding VUV radiation intensity. The pulse FWHM is 410 as, and the pulse repetition period is 1.4 fs. The output radiation spectrum is shown in Fig. 3b. It contains three highest-intensity spectral components with a comparable amplitude at the frequencies $\omega_{\rm tr}^{(\rm aver)}$ ($k_{\rm inc} = 0$) and $\omega_{\rm tr}^{(\rm aver)} \pm 2\Omega$ ($k_{\rm inc} = \pm 1$). Since both the amplitude spectrum (see also Fig. 1c) and the phase spectrum (10) of the output VUV radiation are symmetric relative to the incident field frequency (which coincides with the resonant frequency in this case), the dominant components of the output radiation spectrum turn out to be phase-matched and form a train of bandwidth-limited pulses.

Now we consider the case of $k_{\rm inc} = 1$. Figure 2b presents the corresponding dependence of the contrast of the formed pulses on the modulation index and the medium thickness. While in the case of $k_{\rm inc} = 0$, apart from the above-considered global contrast maximum at $P_\Omega \simeq 2$, there are other regions of high contrast (in the vicinity of $P_\Omega \simeq 2.7$ and $P_\Omega \simeq 4.6$), in the case of $k_{\rm inc} = 1$ there is only one region of the highest contrast at $P_\Omega \simeq 2$, though at large modulation indices, e.g., $3 \le P_\Omega \le 4$, more combination spectral components with the essentially nonzero amplitude are generated (see Fig. 1e). This is because the detuning of the seeding radiation from the time-average transition frequency $\omega_{\rm tr}^{(\rm aver)}$ results in that the symmetry center of the phases of the VUV radiation spectral components, which is at the seeding field frequency, (10), does not coincide with the effective center of the amplitude spectrum of this radiation at the inverted transition frequency $\omega_{\rm tr}^{(\rm aver)}$ (Fig. 4b). As a result, phase matching of the spectral components deteriorates with increasing modulation index, and complicated time beats arise in the temporal dependence of the VUV radiation intensity at the exit from the medium. For large $k_{\rm inc}$, the phase mismatch of the spectral components increases. This is another reason why large values of the incident VUV radiation detuning, i.e., large values of $k_{\rm inc}$, turn out to be unsuitable for formation of short pulses.

It is seen in Fig. 2b that the maximum pulse contrast of about 3.1 is achieved at $P_\Omega = 1.9$ and $x = 1$ mm. It is slightly lower than the maximum contrast at $k_{\rm inc} = 0$. As in the case of resonant seeding radiation, $k_{\rm inc} = 0$, the frequencies $\omega_{\rm tr}^{(\rm aver)}$ ($k = -1$) and $\omega_{\rm tr}^{(\rm aver)} \pm 2\Omega$ ($k = 0, -2$) dominate in the output radiation spectrum (see Fig. 4b), while the spectrum center, $k = -1$, is shifted relative to the symmetry center of phase curve (10), $k = 0$. That is why the phase difference between the spectral components with numbers $k = -1$ and $k = -2$, $\Delta\varphi_{-1,-2} = \varphi_{-1} - \varphi_{-2} \approx 0.11\pi$, differs from the phase difference between the components with numbers $k = 0$ and $k = -1$, $\Delta\varphi_{0,-1} = \varphi_0 - \varphi_{-1} \approx 0.21\pi$. Together with the less homogeneous distribution of the amplitudes

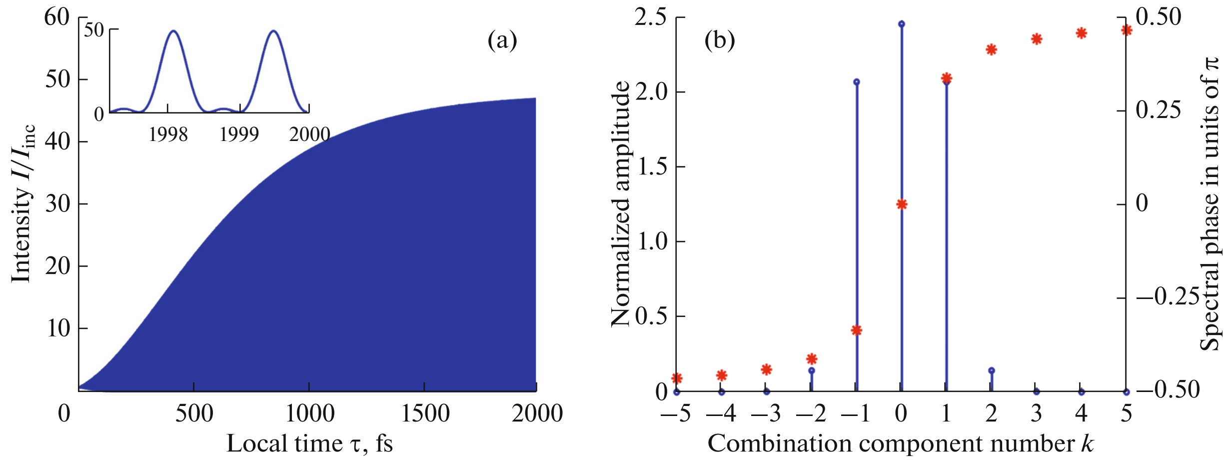

Fig. 3. (Color online) (a) Time dependence of the intensity of the VUV radiation (6), (8), $I = c|\tilde{E}_{XUV}|^2/8\pi$, at the exit from the active Li^{2+} plasma for $k_{inc} = 0$, $P_\Omega = 2$, and $x = 1.1$ mm. The concentration of ions is $N_i = 1.5 \times 10^{17}$ cm^{-3}, the concentration of free electrons is $N_e = 3 \times 10^{17}$ cm^{-3}, and the relaxation time of the medium polarization is $\gamma^{-1} \simeq 396$ fs. The seeding radiation intensity is $I_{inc} = c(E_{XUV}^{(inc)})^2/8\pi$. Inset: the pulse shape in the steady-state regime. (b) Dependence of the amplitude (8a) and (8b) normalized to $E_{XUV}^{(inc)}$ (blue lines) and the phase of the spectral components (red symbols) on the index k. The phases of the spectral components are calculated by formula (10). The other parameters are the same as in Fig. 3a.

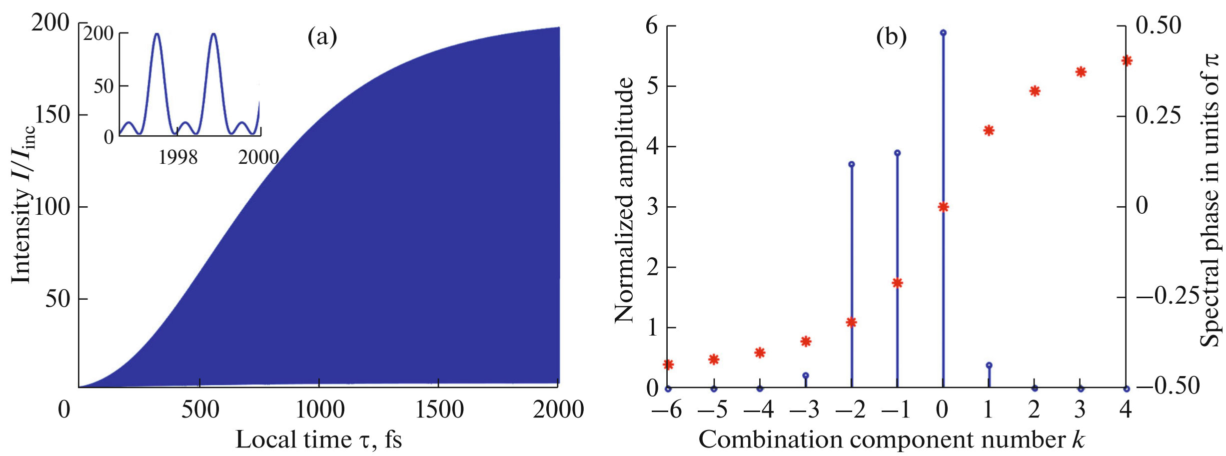

Fig. 4. (Color online) Same as in Fig. 3, but for $k_{inc} = 1$, $P_\Omega = 1.9$, and $x = 1$ mm; $k = 0$ corresponds to the seeding radiation frequency; $k = -1$, to the resonance frequency.

of the spectral components (cf. Figs. 3b and 4b), their phase difference leads to a decrease in the contrast of the formed pulses. At the same time, according to Fig. 4a, the peak intensity of the pulses at $k_{inc} = 1$ in the steady-state regime is about 200 times higher than the intensity of the seeding VUV radiation and 4.2 times higher than the peak intensity of the pulses at $k_{inc} = 0$. The increase in the pulse intensity is caused by enhancement of the interaction between the seeding radiation and the active medium at $k_{inc} = 1$ as a consequence of the fact that at the optimum modulation index value, $P_\Omega \approx 2$, which ensures generation of combination spectral components, $J_2(P_\Omega) > J_0(P_\Omega)$ (function $J_0(P_\Omega)$ achieves the maximum at $P_\Omega = 0$, i.e., in the absence of modulation). As a result, at the detuning by the double modulation frequency the effective gain coefficient of the VUV radiation in the modulated active medium increases (see Fig. 1f, in the vicinity of $P_\Omega \approx 2$), as does the efficiency of the generation of the combination spectral components (see Figs. 1c, 1e). It is also worth noting that since the required modulating field wavelength $\Lambda \simeq 805$ nm is smaller than at $k_{inc} = 0$, both the duration of a single pulse and the pulse repetition period turn out to be shorter, 370 as and 1.35 fs, respectively.

5. CONCLUSIONS

In this work we investigated a possibility of forming subfemtosecond pulses from the quasimonochromatic seeding VUV radiation in the hydrogen-like active medium of the plasma-based X-ray laser modulated by the intense optical or IR field. The influence of the detuning of the seeding radiation frequency from the inverted transition frequency averaged over the modulating field cycle on characteristics of the amplified VUV radiation was investigated. It is shown for the first time that subfemtosecond pulse trains can be formed from the seeding VUV radiation detuned from the resonance with the inverted transition of the active medium. In prospect, this opens up a possibility of using fixed-frequency narrow-bandwidth sources of the seeding VUV radiation (with the frequency detuned from the resonance of the active medium in the IR field by an even multiple of the IR field frequency). Explicit expressions are obtained for the amplitudes and phases of the combination spectral components generated during the propagation of VUV radiation through the modulated active medium. It is shown that formation of subfemtosecond pulses with the maximum contrast is achieved with the seeding VUV radiation resonant to the inverted transition of the active medium (with allowance for its change in the modulating field) or detuned from the resonance by the double frequency of the modulating field (irrespective of the detuning sign). Large detuning from the resonance impairs characteristics of the formed pulses because of decreasing efficiency of generation of combination spectral components and their phase mismatch caused by asymmetry of the generated spectrum relative to the seeding VUV radiation frequency. At the same time, it is shown that detuning of the seeding radiation frequency from the time-average frequency of the inverted transition by the double frequency of the modulating field allows the peak intensity of the formed subfemtosecond pulses to considerably increase relative to the case of the exact resonance with the retained pulse repetition period. It is demonstrated for the hydrogen-like active Li^{2+} plasma medium that it is possible to obtain a train of pulses with the duration of 370 as and the peak intensity 200 times higher than the intensity of the seeding VUV radiation at the central wavelength of 13.5 nm. Similar spectral combs and trains of subfemtosecond VUV radiation pulses can be used in spectroscopy, including measurements with the attosecond time resolution.

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