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Generation of harmonics via multiphoton resonant excitation of hydrogenlike ions in an x-ray free-electron-laser field

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The coherent radiation spectrum of highly charged hydrogenlike ions in an intense x-ray free-electron-laser field is considered. The spectrum corresponding to harmonic generation in the resonant multiphoton excitation regime is investigated both analytically and numerically, arising from the Dirac Hamiltonian. The obtained analytical results are based on the generalized rotating wave approximation and are in good agreement with performed numerical calculations. Estimations show that one can achieve efficient generation of coherent hard x-ray radiation using multiphoton resonant excitation by appropriate x-ray pulses.

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I. INTRODUCTION

The generation of coherent radiation of smaller wavelengths than optical ones on atomic transitions is an old goal of laser physics [1]. The recent remarkable progress in x-ray free-electron-laser (FEL) technology allows the production of supershort pulses exceeding the peak brilliance of conventional synchrotron sources by many orders of magnitude [2–4]. The interaction of such powerful x-ray radiation with matter has essentially multiphoton character and opens up a wide research field, where common nonlinear effects can be extended to high energy transitions [5–12]. The theory of the interaction of intense x-ray pulses with comparatively simple systems, such as atoms and small molecules, engages a special place in these studies. Its importance is specifically attributed to the recent experiments, as well as to the clarification of conceptual matters and the formation of a basis for studying interaction with the more complex systems such as biomolecules or solids [13–18].

The generation of harmonics of a strong laser radiation is one of the cornerstones of nonlinear light-matter interaction and has been studied extensively at infrared and optical driving frequencies [19]. Depending on the interaction parameters, harmonic generation may occur via bound-bound [20] and bound-free-bound transitions through the continuum spectrum [21]. For a light scattering process via bound-bound transitions, the resonant interaction is of interest. Apart from its pure theoretical interest as a simple model, the resonant interaction regime enables one to significantly increase the efficiency of frequency conversion [22,23]. For the high harmonics generation via bound-bound states one needs multiphoton resonant transition. The latter is effective when the atomic system has a mean dipole moment in the stationary states [24], or the energies of the two states of a three-level atomic system are close enough to each other and there is a nonvanishing transition dipole moment between these states [25,26]. A good example of such a configuration is the hydrogenlike atomic system where because of random degeneracy of an orbital moment the atom has a mean dipole moment in the excited stationary states. Furthermore, these systems have an advantage that allows one to generate coherent radiation with Rabi frequency [27] and moderately high harmonics by optical pulses [22]. The treatment in Ref. [22] was made in the scope of nonrelativistic theory for atoms or ions with a small nuclear charge and optical pumping. Note that a harmonic generation mechanism without ionization for these systems can be efficient for producing radiation up to VUV frequencies [22,23]. To reach the far x-ray region atoms or ions with the large nuclear charges are necessary. In this case the relativistic effects should be taken into account, specifically, the fine structure of the hydrogenlike atoms or ions. The quantum dynamics of highly charged hydrogenlike ions in a strong high-frequency laser field has been investigated in Ref. [28]. The obtained results have shown that despite complicated levels' linkages in the relativistic case one can effectively excite ions via multiphoton transitions. Thus, it is of interest to study the harmonic generation from hydrogenlike ions at multiphoton resonant excitation, when only a few resonant states are involved in the radiation generation processes. Here one can expect further up-conversion of existing x-ray FEL frequencies. The interest is also motivated by the success of x-ray free-electron lasers [2,3], where pump waves of sufficient intensities can be realized for multiphoton processes. In addition, ions may be produced with an arbitrary charge state via various effective methods [29].

In the present paper the coherent radiation spectrum of highly charged hydrogenlike ions in an intense x-ray free-electron-laser field is considered. The consideration is based on the Dirac Hamiltonian, which allows one to take into account the fine structure of the levels. The spectrum corresponding to harmonic generation in the resonant multiphoton excitation regime is investigated both analytically and numerically. The obtained analytical results are based on the generalized rotating wave approximation and are in good agreement with performed numerical calculations. Estimations show that one can achieve the efficient generation of hard coherent x-ray radiation using multiphoton resonant excitation by currently available soft x-ray pulses.

The paper is organized as follows: In Sec. II we describe our analytical model and derive the coherent x-ray scattering spectrum. Results are discussed in Sec. III, where we present some numerical calculations for the hydrogenlike ions and compare the obtained spectra with analytical results. Finally, conclusions are given in Sec. IV.

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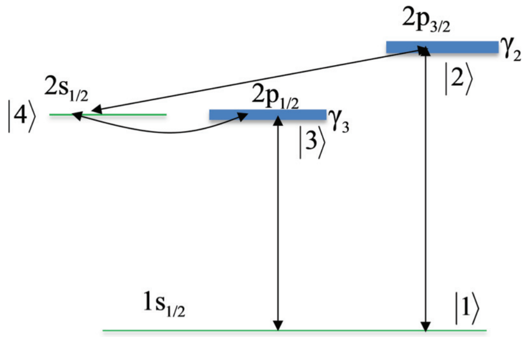

FIG. 1. (Color online) Atomic level structure with the dipole coupling transitions.

II. BASIC MODEL AND HARMONICS SPECTRUM

Let us consider resonant interaction of a hydrogenlike ion with a moderately strong x-ray coherent radiation field. In this case, if a radiation field is not so strong as to make dominant the ionization process, rather than to consider the whole atomic wave packet one can reduce the interaction dynamics to a few levels only. For a large charge number of the nucleus Z_a, the relativistic effects play an important role and, therefore, should be taken into account. For the considered x-ray frequencies and multiphoton resonances the dipole approximation is still applicable: $\lambda \gg a$, where a is the characteristic size of the atomic system and λ is the wavelength of the x-ray wave. Hence, we will take into account only electrical-dipole transitions $E1$ as the main coupling transitions between the states with the main quantum numbers $n_0 = 1,2$. It is supposed that the pump field is much smaller than characteristic atomic fields: $E_0 \ll E_{\mathrm{at}} \sim Z_a^3$ and ionization rates can be neglected. So, the Dirac equation in a linearly polarized x-ray radiation field with unit polarization vector $\widehat{\mathbf{z}}$, slowly varying amplitude E_0, and carrier frequency ω_X, reduces to two independent sets of four equations for each magnetic quantum number $M = \pm 1/2$, whole moment $j = 1/2, 3/2$, and the state parity $P = \pm 1$. The latter is defined via the orbital moment l . So, the resonant interaction of an x-ray coherent radiation field with a hydrogenlike ion can be described by the 4×4 effective Hamiltonian:

$$\widehat{H} = \begin{pmatrix} \varepsilon_1 & V_{12} & V_{13} & 0 \\ V_{12}^* & \varepsilon_2 & 0 & V_{24} \\ V_{13}^* & 0 & \varepsilon_3 & V_{34} \\ 0 & V_{24}^* & V_{34}^* & \varepsilon_3 \end{pmatrix}. \tag{1}$$

Here we have assumed the following basis $|\eta\rangle$, where $\eta = \{n_0, j, l, M\}$ indicates the set of quantum numbers as follows:

$$|1\rangle \equiv |1,1/2,0,-1/2\rangle\,, \quad |2\rangle \equiv |2,3/2,1,-1/2\rangle\,,$$
$$|3\rangle \equiv |2,1/2,1,-1/2\rangle\,, \quad |4\rangle \equiv |2,1/2,0,-1/2\rangle\,.$$

The atomic configuration with state couplings corresponding to Hamiltonian (1) is shown in Fig. 1. A similar Hamiltonian describes quantum dynamics for four other states with $M = 1/2$. In Eq. (1) ε_η is the energy of the stationary state $|\eta\rangle$ of the unperturbed hydrogenlike ion:

$$\varepsilon_1 = c^2\sqrt{1-(\alpha Z_a)^2}, \tag{2}$$

$$\varepsilon_2 = c^2\sqrt{1-(\alpha Z_a/2)^2}, \tag{3}$$

$$\varepsilon_3 = c^2\sqrt{(1+\varepsilon_1/c^2)/2}. \tag{4}$$

α is the fine structure constant and

$$V_{\eta\nu} = z_{\eta\nu} E_0 \cos \omega_X t \tag{5}$$

is the interaction part of the Hamiltonian with the electric-dipole moment $z_{\eta\nu}$ operator ($\eta,\nu = 1,2,3,4$) calculated by the known bispinor solutions of a stationary Dirac equation in the Coulomb field [28]. Here and below, unless stated otherwise, we employ atomic units ($\hbar = e = m_e = 1$).

As was shown in Ref. [28], this configuration is unitary equivalent to an atomic configuration with the mean dipole moments in the excited states, which are responsible for the multiphoton resonance due to the self-energy oscillating levels. Thus, in order to have a physically more transparent form of equations for multiphoton resonant transitions and harmonics radiation, we apply a unitary transformation [28]:

$$\hat{S} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1/\sqrt{3} & 1/\sqrt{6} & i/\sqrt{2} \\ 0 & -1/\sqrt{3} & 2/\sqrt{6} & 0 \\ 0 & 1/\sqrt{3} & 1/\sqrt{6} & -i/\sqrt{2} \end{pmatrix}, \tag{6}$$

which for the transformed Hamiltonian $\widehat{H}' = \hat{S}\widehat{H}\hat{S}^{+}$ gives

$$\widehat{H}' = \begin{pmatrix} \varepsilon_1 & id_{12}E & id_{13}E & id_{12}E \\ -id_{12}E & \varepsilon + d_{22}E & \varpi - d_{23}E & -\varpi \\ -id_{13}E & \varpi - d_{23}E & \varepsilon & \varpi + d_{23}E \\ -id_{12}E & -\varpi & \varpi + d_{23}E & \varepsilon + d_{44}E \end{pmatrix}. \tag{7}$$

Here the transition matrix elements are

$$\varpi = \frac{\varepsilon_3 - \varepsilon_2}{3}, \quad d_{12} = \frac{z_{13} + \sqrt{2}z_{12}}{i\sqrt{6}}, \quad d_{23} = \frac{z_{24} - \sqrt{2}z_{34}}{i\sqrt{6}},$$
$$d_{13} = \frac{\sqrt{2}z_{13} - z_{12}}{i\sqrt{3}}, \quad d_{22} = -d_{44} = \frac{z_{34} + \sqrt{2}z_{24}}{i\sqrt{3}}.$$

The matrix elements of the transitions d_{23}, d_{13}, and ϖ are proportional to fine structure splitting. The excited states have the same ε energy:

$$\varepsilon = \frac{\varepsilon_2 + 2\varepsilon_3}{3}, \tag{8}$$

with mean dipole moments in the excited states $|2'\rangle$ and $|4'\rangle$. Our method of solving the Dirac equation for Hamiltonian (1) has been described in detail in Ref. [28] and will not be repeated here. Hence, we will adopt the wave function obtained in the paper [28]. At the n-photon resonance and under the generalized rotating wave approximation, the time-dependent wave function can be expanded as

$$|\Psi(t)\rangle = e^{-i\varepsilon_1 t}\left\{[\overline{b}_1(t)+\beta_1(t)]|1'\rangle + [\overline{b}_2(t)+\beta_2(t)]\exp\left[-i\left(n\omega_X t - \int_0^t d_{22}Edt\right)\right]|2'\rangle + [\overline{b}_3(t)+\beta_3(t)]\exp[-in\omega_X t]|3'\rangle + [\overline{b}_4(t)+\beta_4(t)]\exp\left[-i\left(n\omega_X t - \int_0^t d_{44}Edt\right)\right]|4'\rangle\right\}, \tag{9}$$

where $\overline{b}_i(t)$ are the time-averaged probability amplitudes and $\beta_i(t)$ are rapidly oscillating functions on the scale of the pump wave period. The time-averaged amplitudes $\overline{b}_i(t)$ are

$$\overline{b}_i = \sum_{j=1}^{4} C_{ij}\exp(i\lambda_j t), \tag{10}$$

where C_{ij} are the constants of integration determined by the initial conditions, and the factors λ_j are the solutions of the fourth-order characteristic equation:

$$\begin{Vmatrix} 2\Delta+\lambda & L_{12}^{(n)} & 0 & L_{14}^{(n)} \\ L_{21}^{(n)} & \Delta-\delta+\lambda & 0 & \tilde{\Delta} \\ 0 & 0 & -\delta+\lambda & 0 \\ L_{41}^{(n)} & \tilde{\Delta} & 0 & \Delta-\delta+\lambda \end{Vmatrix} = 0, \tag{11}$$

with the terms

$$L_{12}^{(n)} = \left(L_{21}^{(n)}\right)^* = i(-1)^{n+1}\frac{d_{12}}{d_{22}}n\omega_X J_n(\zeta), \tag{12}$$

$$L_{14}^{(n)} = \left(L_{41}^{(n)}\right)^* = i\frac{d_{12}}{d_{22}}n\omega_X J_n(\zeta), \tag{13}$$

describing time-averaged probability amplitudes, and

$$\Delta = \omega_X\left(\frac{d_{12}}{d_{22}}\right)^2\sum_{k\neq n}\frac{k^2J_k^2(\zeta)}{k-n}, \tag{14}$$

$$\tilde{\Delta} = \omega_X\left(\frac{d_{12}}{d_{22}}\right)^2\sum_{k\neq n}\frac{(-1)^k k^2J_k^2(\zeta)}{k-n}, \tag{15}$$

dynamic Stark shifts. The argument of the ordinary Bessel function $J_n(\zeta)$ is the dipole interaction energy in the units of the pump wave photon energy: $\zeta = |d_{22}E_0/\omega_X|$. For the relatively small nuclear charges $(\alpha Z_a)^2 \ll 1$ one can neglect the terms $O((\alpha Z_a)^2)$ and obtain compact expressions in Eq. (11). In deriving these equations we have applied the well-known expansion of exponent through Bessel functions with real arguments [30]:

$$e^{i\zeta\,\sin\alpha} = \sum_{s=-\infty}^{\infty} J_s(\zeta)e^{\mathrm{i}s\alpha}, \tag{16}$$

and introduced the resonance detuning

$$\delta = \varepsilon_1 + n\omega - \varepsilon. \tag{17}$$

Assuming smooth turn-on of the pump wave, the relation between the rapidly and slowly oscillating parts of the probability amplitudes can be written as

$$\beta_1(t) = \overline{b}_2(t)\frac{d_{12}}{d_{22}}\sum_{k\neq n}\frac{(-1)^k kJ_k(\zeta)e^{i(k-n)\omega_X t}}{k-n} + \overline{b}_4(t)\frac{d_{14}}{d_{44}}\sum_{k\neq n}\frac{kJ_k(\zeta)e^{i(k-n)\omega_X t}}{k-n}, \tag{18}$$

$$\beta_2(t) = -\overline{b}_1(t)\frac{d_{12}^*}{d_{22}}\sum_{k\neq n}\frac{(-1)^k kJ_k(\zeta)e^{-i(k-n)\omega_X t}}{k-n}, \tag{19}$$

$$\beta_3(t) = O((\alpha Z_a)^2), \tag{20}$$

$$\beta_4(t) = -\overline{b}_1(t)\frac{d_{14}^*}{d_{44}}\sum_{k\neq n}\frac{kJ_k(\zeta)e^{-i(k-n)\omega_X t}}{k-n}. \tag{21}$$

The coherent part of the dipole spectrum in the Schrödinger picture has the form [31]

$$S_c = \left|\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\langle D(t)\rangle\right|^2, \tag{22}$$

where

$$\langle D(t)\rangle = \langle\Psi(t)|\,\hat{\mathbf{z}}\cdot\hat{\mathbf{d}}(\mathbf{0})\,|\Psi(t)\rangle \tag{23}$$

is the time-dependent expectation value of the dipole operator. With the help of expressions (18)–(21) one can analytically calculate (9) for arbitrary initial atomic state and, therefore, the expectation value of the dipole operator (23). Then the solution (10) for the system initially situated in the ground state, when the dynamic Stark shifts are compensated by appropriate detuning, is

$$\overline{b}_1(t) = e^{-i2\Delta t}\cos(\Omega_R t/2)$$

and

$$\overline{b}_3(t) = -\overline{b}_2(t) = \frac{e^{-i2\Delta t}}{\sqrt{2}}\sin(\Omega_R t/2). \tag{24}$$

Here

$$\Omega_R \equiv \left|2\sqrt{2}\frac{d_{12}}{d_{22}}n\omega_X J_n(\zeta)\right| \tag{25}$$

is the generalized Rabi frequency at n-photon resonance, which has a nonlinear dependence on the amplitudes of the wave fields through the Bessel functions.

Replacing the probability amplitudes in (9) by the corresponding expressions (24) and putting in (23) one can derive an analytical expression for $\langle D(t)\rangle$. Here, one can neglect second-order terms of the rapidly oscillating parts of the probability amplitudes, since $\beta_l^2(t) \sim (d_{12}/d_{22})^2 \ll 1$. This leads to the compact analytic formula:

$$\langle D(t)\rangle = \sum_k \{S_k \sin[(2k+1)\omega_X t] + C_k \cos[(2k+1)\omega_X t]\}, \tag{26}$$

where

$$S_k = \sqrt{2} d_{12} \frac{n J_{2k+1+n}(\zeta)}{2k+1} \sin(\Omega_R t), \tag{27}$$

$$C_k = \frac{d_{12}^2}{d_{22}} \sum_{s\neq n} \{(-1)^{n-s} - 1 - [3 + (-1)^{n-s}]\cos(\Omega_R t)\} \times \frac{s J_{2k+1+s}(\zeta) J_s(\zeta)}{s-n}. \tag{28}$$

The expression (26) for $\langle D(t)\rangle$ with Eqs. (27) and (28) show that intensities of the harmonics are mainly determined by the behavior of Bessel function $J_m(\zeta)$. Since the Bessel function exponentially decreases with the increasing of the index, one can conclude that for effective harmonic generation the dipole interaction energy $|d_{22}E_0|$ should be comparable to or larger than the pump wave photon energy ω_X. Then taking into account that $J_{-m}(\zeta) = (-1)^m J_m(\zeta)$, the cutoff harmonic s_c is determined from the condition $s_c - n \sim \zeta$, i.e., the cutoff position depends linearly on the laser field amplitude: $s_c \simeq n + |d_{22}E_0|/\omega_X$. From this estimation for a cut-off harmonic follows that the upper limit of the photon energy ω_c, which can be effectively generated by direct resonant excitation, is higher for the systems with a larger difference of energy in the stationary states ($\omega_c \simeq \varepsilon - \varepsilon_1 + |d_{22}E_0|$). The latter has a quadratic dependence on the nuclear charge Z_a.

III. NUMERICAL RESULTS AND DISCUSSION

In this section we apply our four-level model to study the numerically resonant interaction of hydrogenlike ions with a strong x-ray coherent radiation field of specific parameters. For the large nuclear charges the spontaneous decay of the excited states becomes significant since rates are $\sim Z_a^4$. Thus, in order to develop a microscopic theory of the multiphoton interaction of hydrogenlike ions with a strong radiation field we need to solve the master equation for the density matrix:

$$\frac{d\widehat{\rho}}{dt} = i(\widehat{\rho}\widehat{H} - \widehat{H}\widehat{\rho}) + \mathcal{L}\widehat{\rho}, \tag{29}$$

where

$$\widehat{\rho} \equiv \rho_{\mu\nu}; \quad \mu,\nu = 1,2,3,4 \tag{30}$$

is the density matrix. The decay processes with the rates

$$\gamma_i = \frac{4(\varepsilon_i - \varepsilon_1)^3}{3c^3}|z_{1i}|^2; \quad i = 2,3 \tag{31}$$

have been incorporated into evolution equation (29) by the damping term:

$$\mathcal{L}\widehat{\rho} = -\begin{pmatrix} -\gamma_2\rho_{22} - \gamma_3\rho_{33} & \frac{\gamma_2}{2}\rho_{12} & \frac{\gamma_3}{2}\rho_{13} & 0 \\ \frac{\gamma_2}{2}\rho_{21} & \gamma_2\rho_{22} & \frac{\gamma_2+\gamma_3}{2}\rho_{23} & \frac{\gamma_2}{2}\rho_{24} \\ \frac{\gamma_3}{2}\rho_{31} & \frac{\gamma_2+\gamma_3}{2}\rho_{32} & \gamma_3\rho_{33} & \frac{\gamma_3}{2}\rho_{34} \\ 0 & \frac{\gamma_2}{2}\rho_{42} & \frac{\gamma_3}{2}\rho_{43} & 0 \end{pmatrix}. \tag{32}$$

Here the operator $\mathcal{L}\widehat{\rho}$ represents the norm-conserving spontaneous decay of the population from the excited states $|2\rangle$ and $|3\rangle$ into the ground state $|1\rangle$. The decay process $|2\rangle \to |4\rangle$ has been neglected due to the smallness of its rate compared with $\gamma_{2,3}$. The time-dependent expectation value of the dipole operator now is determined by the density matrix $\widehat{\rho}$:

$$\langle \widehat{D}(t)\rangle = \mathrm{Tr}[\widehat{\rho}(t)(\widehat{\mathbf{z}}\cdot\hat{\mathbf{d}})] = \mathrm{Re}(\rho_{21}z_{12} + \rho_{31}z_{13} + \rho_{42}z_{24} + \rho_{43}z_{34}). \tag{33}$$

For comparison with obtained analytical results, semi-infinite pulses with smooth turn-on, in particular, with hyperbolic tangent $\tanh(t/\tau_r)$ envelope is considered. Here the characteristic rise time τ_r is chosen to be $\tau_r = 20T_X$, where $T_X = 2\pi/\omega_X$ is the x-ray wave period. For turn-on/off of the wave field the latter is described by the envelope function $E_0(t) = E_0 f(t)$:

$$f(t) = \begin{cases} \sin^2(\pi t/\tau); & 0 \le t \le \tau \\ 0; & t < 0, \quad t > \tau, \end{cases} \tag{34}$$

where τ characterizes the pulse duration. It should be noted that the current x-ray facilities, such as the Linac Coherent Light Source, operate in the self-amplified spontaneous emission regime [32] and produce pulses with partial temporal coherence and a spiky temporal profile. However, the rapid development of x-ray sources and self-seeding techniques makes the consideration of seeding pulses with high temporal coherence relevant.

For the numerical calculations we assume that a hydrogenlike atomic system is situated initially in the ground state [$\rho_{11}(0) = 1$]. The time evolution of system (29) is found with the help of the standard fourth-order Runge-Kutta algorithm [33] and for estimation of the power spectra the fast Fourier transform algorithm of expectation value of the dipole operator (33) is used.

Figures 2–4 show the coherent part of the spectrum for various Z_a and resonances calculated for semi-infinite pulses to compare with the spectrum calculated by the analytical expression (26). Here and below, to achieve almost complete population transfer the dynamic Stark shift is compensated by appropriate detuning. Figure 2 shows the coherent part of the spectrum as a function of harmonic order for five-photon ($n = 5$) and eight-photon ($n = 8$) resonant excitation of the hydrogenlike atomic system with nuclear charge $Z_a = 10$. The pump field strength and frequency are set to be $E_0 = 30$ a.u. and $\omega_X = 7.535$ a.u. for five-photon resonance, and $E_0 = 50$ a.u. and $\omega_X = 4.734$ a.u. for eight-photon resonance. The solid (red) line corresponds to numerical calculations, while the dashed (green) line corresponds to the analytical expression (26). For better visibility, the spectrum corresponding to analytical calculations has been slightly shifted to the left. As we can see from the Fig. 2, the analytical formula (26) is in good agreement with the numerical result. In Fig. 3 we plot the dipole spectrum as a function of harmonic order for five-photon ($n = 5$) and ten-photon ($n = 10$) resonant excitation of the hydrogenlike atomic system with nuclear charge $Z_a = 20$. The pump field strength and frequency are set to be $E_0 = 280$ a.u. and $\omega_X = 30.293$ a.u. for five-photon resonance, and $E_0 = 550$ a.u. and $\omega_X = 15.349$ a.u. for ten-photon resonance. In Fig. 4 we plot the dipole spectrum as a function of harmonic order for five-photon ($E_0 = 2400$ a.u.

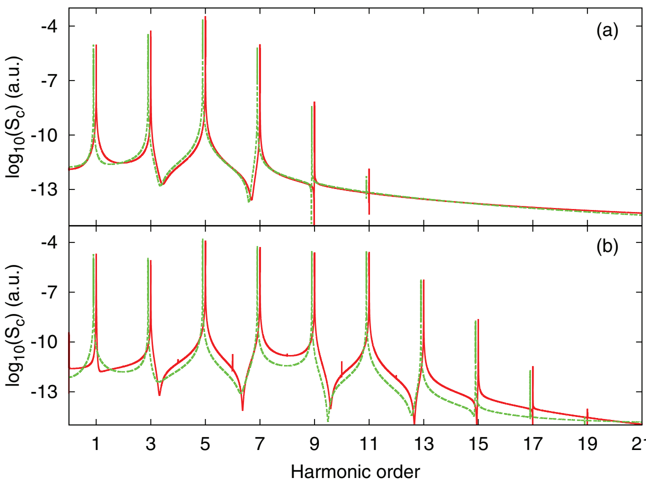

FIG. 2. (Color online) Coherent part of the harmonic emission as a function of the harmonic order at resonant excitation of the hydrogenlike atomic system with nuclear charge $Z_a = 10$. It is shown $\log_{10}(S_c)$ (a) for a five-photon resonance $E_0 = 30$ a.u., $\delta_n = 0.0165\omega_X$ and (b) for an eight-photon resonance $E_0 = 50$ a.u. and $\delta_n = 0.068\omega_X$. The solid red line corresponds to numerical calculations; the dashed green line corresponds to the approximate solution (for visual convenience the latter has been slightly shifted to the left).

and $\omega_X = 123.4$ a.u.) and eight-photon ($E_0 = 3800$ a.u. and $\omega_X = 77.49$ a.u.) resonant excitations of the hydrogenlike ion with nuclear charge $Z_a = 40$. As seen from the last two figures, with the increase of nuclear charge the spontaneous decay of the excited states becomes essential and numerical calculations with density matrix quantitatively differ from the analytical formula (26).

We have also performed calculations for the short x-ray pulse. Figure 5 shows the dipole spectrum as a function of harmonic order at the multiphoton resonant excitation of

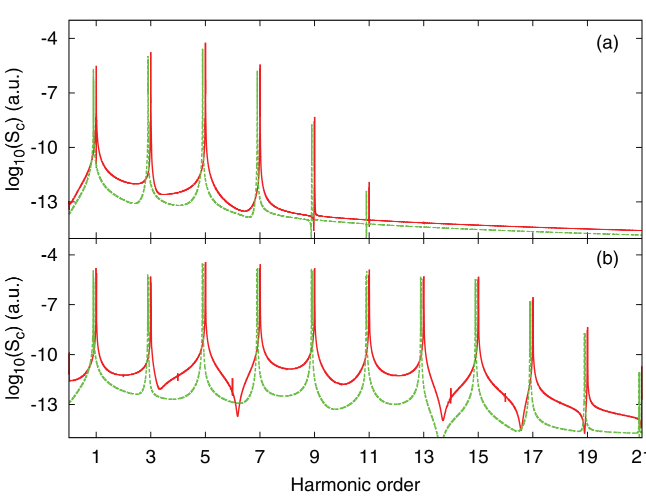

FIG. 3. (Color online) Coherent part of the harmonic emission as a function of the harmonic order at resonant excitation of the hydrogenlike atomic system with nuclear charge $Z_a = 20$: (a) for a five-photon resonance $E_0 = 280$ a.u., $\delta_n = 0.021\omega_X$ and (b) for a ten-photon resonance $E_0 = 550$ a.u. and $\delta_n = 0.173\omega_X$.

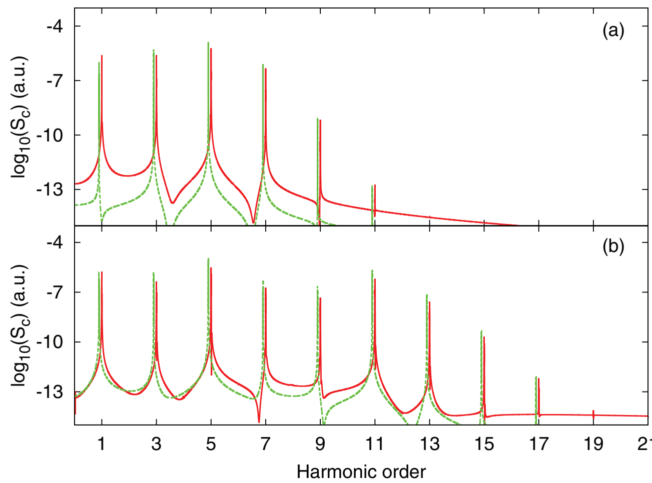

FIG. 4. (Color online) Coherent part of the harmonic emission as a function of the harmonic order at resonant excitation of the hydrogenlike atomic system with nuclear charge $Z_a = 40$: (a) for a five-photon resonance $E_0 = 2400$ a.u., $\delta_n = 0.026\omega_X$ and (b) for an eight-photon resonance $E_0 = 3800$ a.u. and $\delta_n = 0.08\omega_X$.

hydrogenlike atomic systems ($Z_a = 30$ and $Z_a = 40$) with an x-ray pulse of duration $\tau = 100T_X$. As seen from this figure, short laser pulses broaden the harmonics spectra.

Let us make some estimations for the total radiation power of the ensemble of hydrogenlike atoms. Thus, for considered x-ray pump fields the radiated wavelengths are much smaller than the transverse size of the interaction region. The latter is assumed to be limited due to the x-ray beam size with a waist of w_0, which is typically $w_0 \simeq 10^{-4}$ cm for currently available x-ray FELs. The longitudinal size of the interaction region is determined by Rayleigh length $L = \pi w_0^2/\lambda$, where λ is the pump x-ray wavelength. Thus, we have a cigar-shaped active medium ($L \gg w_0$) and the coherent radiation will occur

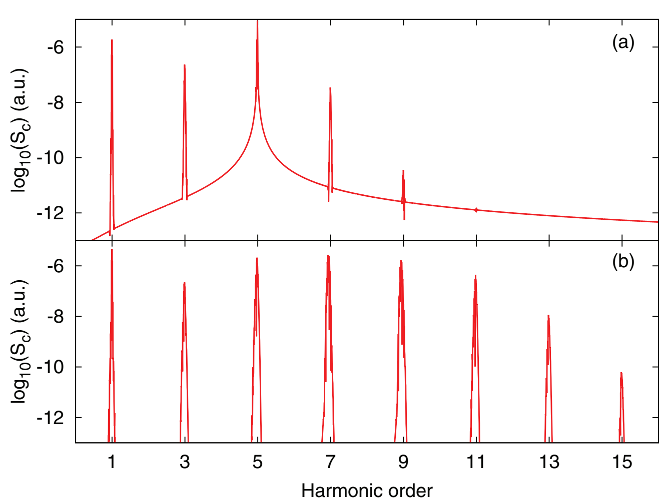

FIG. 5. (Color online) Harmonic emission rate as a function of the harmonic order for short x-ray pulses: (a) for the hydrogenlike atomic system with nuclear charge $Z_a = 30$ and at five-photon resonance ($E_0 = 1000$ a.u., $\omega_X = 68.68$ a.u.), and (b) $Z_a = 40$ and eight-photon resonance ($E_0 = 3800$ a.u. and $\omega_X = 77.49$ a.u.).

primarily along the propagation axis of the pump laser beam and will cover only a tiny solid angle $\sim\lambda_s^2/(\pi w_0^2)$ (λ_s is the sth-harmonic wavelength). Then, for the radiation power we have

$$P_s = P_s^{(1)} (\mathcal{V} N_0)^2 \mu_s,$$

where N_0 is the atomic density, and $P_s^{(1)} = 4s^4\omega_X^4 |d_s|^2 /3c^3$ is the single-atom total radiation power with the Fourier component of the dipole moment d_s. The interference factor μ_s is defined by the shape of an active medium. For the cylindrical system it can be estimated as $\mu_s = 3\lambda_s^2/(8\pi^2 w_0^2)$. In particular, for the setup of Fig. 5(a) $\lambda = 0.664$ nm and the interaction volume becomes $\mathcal{V} = \pi w_0^2 L \simeq 1.5 \times 10^{-8}$ cm^3. For the fifth harmonic $\mu_5 \simeq 6.7 \times 10^{-10}$ and $P_5^{(1)} \simeq 1.3 \times 10^{-2}$ W. Thus, for the atomic density $N_0 \simeq 10^{18}$ cm^{-3} the total power at the hard x-ray frequencies $\sim$10 keV is estimated to be $P_{10\ \mathrm{keV}} \simeq 2 \times 10^9$ W.

IV. CONCLUSION

We have presented a theoretical treatment of the coherent light scattering by hydrogenlike ions under the direct multiphoton resonant excitation. On the basis of the analytical solution of the Dirac equation for a four-level hydrogenlike ionic system driven by a strong x-ray field, we have obtained an analytical expression for the time-dependent expectation value of the dipole operator. Numerical investigation of the dipole spectrum of hydrogenlike systems with different nuclear charges has been performed. Numerical results are in good agreement with obtained analytical results. Our calculations suggest that by using ultrafast x-ray pulses with moderately strong intensities, when the rate of the concurrent spontaneous emission and ionization processes is relatively small, one can achieve efficient production of moderately high harmonics. The considered scheme may serve as a promising method for coherent hard x-ray radiation generation. While the intensities required for an efficient harmonic generation are of course very challenging, the recent experimental advances [4], where the 10^{20} W/cm^2 ($E_0 \simeq 50$ a.u.) intensity of the x-ray FEL is reached, promise clear prospects to reach the required fields.

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