Efficient generation of moderately high harmonics by multiphoton resonant excitation of atoms

H. K. Avetissian,* B. R. Avchyan, and G. F. Mkrtchian
Department of Quantum Electronics, Yerevan State University, 1 A Manukian, Yerevan 375025, Armenia
(Received 20 November 2007; revised manuscript received 28 December 2007; published 11 February 2008)

The coherent scattering spectrum of a three-level atom driven by an intense laser field with multiphoton resonant excitation is considered. The spectrum corresponding to harmonic generation is investigated both analytically and numerically for different initial atomic states. The analytical calculations are based on the generalized rotating wave approximation, which allows one to explain obtained spectra containing in general both even and odd harmonics. The results applied to the hydrogen atom show that one can achieve efficient generation of moderately high harmonics using multiphoton resonant excitation by appropriate laser pulses.

DOI: 10.1103/PhysRevA.77.023409 PACS number(s): 42.50.Hz, 42.65.Ky, 32.50.+d, 82.53.Kp

I. INTRODUCTION

Harmonic generation and wave mixing are fundamental processes of traditional nonlinear optics which have been extensively studied both theoretically and experimentally with the appearance of lasers [1]. Since the first realization of second-order harmonic generation [2], continuous progress in this field with the advent of superintense laser sources has allowed generation of higher-order harmonics in the laser-atom interaction (see, e.g., [3] for a review, and references therein). The permanent interest in the processes of intense laser and atom or ion interaction can be largely attributed to the problem of high-order harmonic generation, and this is one of the most promising ways of producing coherent radiation in the uv and x-ray range.

Depending on the laser-atom interaction parameters, harmonic generation may occur via bound-bound and bound-free-bound transitions through the continuum spectrum. In bound-bound transitions, light resonant scattering when only a few resonant levels of an atom are involved in the interaction process is of interest. Apart from its pure theoretical interest as a simple model, the resonant interaction regime is important as it will provide high efficiency of harmonic generation.

The one-photon resonant excitation of atoms and associated light scattering have been comprehensively studied both theoretically and experimentally and described in numerous review articles and books (see, e.g., [4]). The spectrum of the light scattered by a two-level atom driven near resonance by a laser field displays a narrow elastic contribution at the laser frequency and an inelastic triplet structure, with the splitting proportional to the amplitude of the driving field [5]. A satisfactory explanation of the physics was given in the dressed-atom picture of the atom-field interaction [6,7]. In strong fields the spectrum is richer [8–15], and a large number of laser photons are converted into a single high-energy photon. However, in two-level atomic systems for efficient harmonic generation the laser field should be strong and the concurrent process of multiphoton ionization will dominate. The two-level model is not a good quantitative model of the interaction of atoms with intense laser fields when the resonance condition does not hold, or the Rabi frequency of the driving field is comparable to or larger than the atomic transition frequency. Therefore harmonic generation by a nonresonant laser field in the two-level model just remains a useful theoretical model for clarifying conceptual matters. Multiphoton resonant excitation of atoms for efficient harmonic generation by low-frequency laser fields also meets with difficulties in two-level atomic systems [16].

Nevertheless, in some atomic and molecular systems, one can avoid the difficulties with efficient multiphoton excitation based on the two-level model. As has been shown [17–20], the multiphoton resonant excitation of atoms subjected to a strong laser field is effective when the atom has a mean dipole moment in the excited states. Otherwise, the energies of the excited states of a three-level atom should be close to each other and the transition dipole moment between these states must be nonzero. As an example, we have considered the hydrogenlike atomic and ionic system where, because of random degeneracy of the orbital moment, the atom has a mean dipole moment in the excited stationary states. Other interesting applications of multiphoton resonant excitation concern dipolar molecules with a permanent dipole moment [21] and the method can also be applied to evenly charged homonuclear diatomic molecular ions [18,22].

Hence, haveing an efficient multiphoton resonant excitation scheme, it is of interest to consider light scattering by an atomic or molecular system under circumstances when only bound states are involved in the interaction process. In this connection, note that the harmonic radiation via bound-bound transitions in hydrogenlike atomic systems has been considered in Refs. [23,24]. In Ref. [23] the dynamics and radiation spectra of a classical hydrogen atom in an intense classical radiation field with a relatively low frequency as compared to the characteristic Kepler frequency of the atomic system have been investigated. In [24] the radiation of highly charged hydrogenlike ions in intense high-frequency laser pulses was investigated numerically, and efficient generation of moderately high harmonics was demonstrated.

In the present work, coherent light scattering by a three-level atom with multiphoton resonant excitation is investigated. Multiphoton resonance is important for effective excitation and to obtain effective harmonic generation in the considered scheme. For the collection of atoms, the resulting radiation may be incoherent or coherent depending on the

*avetissian@ysu.am

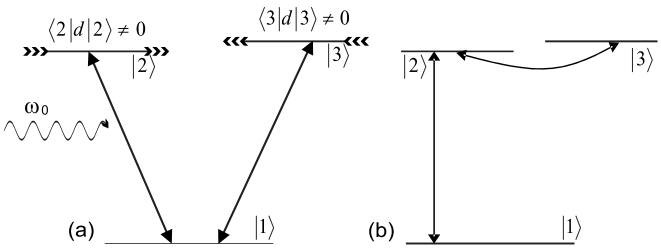

FIG. 1. Three-level atomic structures for (a) V-type configuration with mean dipole moments in the excited states and (b) atomic configuration with a coupling transition between the excited states.

emission direction and the phase matching among the atoms. In the present paper we investigate these issues with emphasis on the coherent part of the spectrum. Then the results obtained are applied to the hydrogen atom and numerical calculations are also made for various initial conditions. The nonperturbative resonant approach that we employ in calculations for the probability amplitudes has been described earlier in Ref. [17]. The spectra obtained by this approach coincide well with those obtained by numerical calculations. We compare the scattering spectrum of the emitted light by a three-level atom with the spectrum of the two-level atom and demonstrate quantitatively the advantages of the scheme considered.

The organization of the paper is as follows. In Sec. II we present the analytical model and derive the coherent contribution to the scattered light spectrum. In Sec. III we present some numerical calculations for the hydrogen atom and compare the obtained spectra with analytical results. Finally, conclusions are given in Sec. IV.

II. BASIC MODEL OF MULTIPHOTON RESONANT INTERACTION AND EMITTED LIGHT SPECTRUM

We consider a three-level atom driven by an intense laser field of frequency ω_0 as shown in Fig. 1(a). We assume the atom to be in a V configuration in which a pair of upper levels $|2\rangle$ and $|3\rangle$ with permanent dipole moments are coupled to a single lower level $|1\rangle$. Another possible three-level scheme is shown in Fig. 1(b). In this case the lower level $|1\rangle$ is coupled to an upper level $|2\rangle$ which has a strong dipole coupling to an adjacent level $|3\rangle$. If the difference between the energies of the excited states in this configuration is small compared with the laser-atom interaction energy then by a unitary transformation [17] the problem can be reduced to the V configuration [Fig. 1(a)]. The hydrogenlike atom is an illustrative example of this fact when one considers the problem in parabolic [25] and the usual spherical coordinates. In the first case, the atom has a mean dipole moment in the excited states, while in the second case the mean dipole moment is zero for the stationary states but because of the random degeneracy of the orbital moment there is a dipole coupling between degenerate states. Hence, we will consider the scheme of the V configuration.

The Hamiltonian describing the interaction of the laser field with the considered three-level atomic system in the dipole approximation is given by

$$\hat{H} = \epsilon_1|1\rangle\langle 1| + (\epsilon_2 + V_{22})|2\rangle\langle 2| + (\epsilon_3 + V_{33})|3\rangle\langle 3| + (V_{12}|1\rangle\langle 2| + V_{13}|1\rangle\langle 3| + \text{H.c.}), \quad (1)$$

where

$$V_{\eta\nu} = -\boldsymbol{\epsilon}\cdot\mathbf{d}_{\eta\nu}E\cos(\omega_0 t). \quad (2)$$

Here, $\mathbf{d}_{\eta\nu}$ is the matrix element of the electric dipole moment, E is the slowly varying amplitude of the linearly polarized pump wave, with a carrier frequency ω_0 and unit polarization vector $\boldsymbol{\epsilon}$. The terms V_{22} and V_{33} in Eq. (1) account for interaction due to the permanent dipole moments, and these terms are crucial for efficient multiphoton resonant excitation.

In general, for correct calculation of the power spectrum of the light scattered by an atom, one should start from the quantized radiation field concept, taking into account also the atom's radiation reaction effects, i.e., one should consider the self-consistent set of atom–quantized-field equations. But for our purpose a simpler ansatz is capable of describing the physics of the process considered. First, the pump field is assumed to be strong enough to be described with high accuracy as a classical Maxwell field in the given field approximation. Then the scattered radiation field and atom-pump field will be considered as dynamically isolated subsystems. This leads to the known scattering spectrum which has been obtained in general using the correspondence principle [10,11]. According to this ansatz the dipole spectrum can be calculated from the expectation value of the two-time dipole correlation function which in the Heisenberg picture reads

$$S(\omega) = \int_{-\infty}^{\infty} dt_2 \int_{-\infty}^{\infty} dt_1 e^{-i\omega(t_2-t_1)}\langle \boldsymbol{\epsilon}\cdot\hat{\mathbf{d}}(t_2)\boldsymbol{\epsilon}\cdot\hat{\mathbf{d}}(t_1)\rangle. \quad (3)$$

Since we are interested in obtaining the coherent part of the scattering spectrum in Eq. (3) we have taken the projection of the dipole operator $\boldsymbol{\epsilon}\hat{\mathbf{d}}(t)$. Note that the power spectrum relates to the dipole spectrum $S(\omega)$ (3) through a factor of ω^4.

The total spectrum can be separated into the coherent part $S_C(\omega)$, the spectrum of the average dipole moment, and the incoherent part $S_I(\omega)$, the spectrum of the quantum dipole fluctuations [10]. Thus, the spectrum (3) can be written as

$$S(\omega) = S_C(\omega) + S_I(\omega), \quad (4)$$

with

$$S_C(\omega) = \left| \int_{-\infty}^{\infty} dt\, e^{-i\omega t}\langle \Psi_i|\boldsymbol{\epsilon}\cdot\hat{\mathbf{d}}(t)|\Psi_i\rangle \right|^2 \quad (5)$$

and

$$S_I(\omega) = \sum_{k\neq i} \left| \int_{-\infty}^{\infty} dt\, e^{-i\omega t}\langle \Psi_i|\boldsymbol{\epsilon}\cdot\hat{\mathbf{d}}(t)|\Psi_k\rangle \right|^2. \quad (6)$$

Here $\{|\Psi_k\rangle\}$ is a complete and orthonormal set containing the initial state $|\Psi(0)\rangle = |\Psi_i\rangle$ of the atomic system. In the Schrödinger picture the coherent part of the spectrum can be written as

$$S_C(\omega) = \left| \int_{-\infty}^{\infty} dt\, e^{-i\omega t} \langle D(t) \rangle \right|^2, \tag{7}$$

where

$$\langle D(t) \rangle = \langle \Psi(t) | \boldsymbol{\epsilon} \cdot \mathbf{d} | \Psi(t) \rangle \tag{8}$$

is the time-dependent expectation value of the dipole operator and $|\Psi(t)\rangle$ is the time-developed initial state [we have abbreviated the initial-time dipole operator $\hat{\mathbf{d}}(0)$ as simply $\mathbf{d}$].

Thus, in order to calculate the coherent part of the spectrum given by Eq. (7) one needs the dynamic wave function of the atom in the pump laser field, i.e., the solution of the Schrödinger equation with Hamiltonian (1):

$$i\hbar \frac{\partial |\Psi(t)\rangle}{\partial t} = \hat{H} |\Psi(t)\rangle. \tag{9}$$

We assume the multiphoton resonant excitation regime, i.e., $|\delta_{2,3}| \ll \omega_0$, where the n-photon resonance detunings are given by

$$\delta_{2,3} = \frac{\epsilon_1 - \epsilon_{2,3}}{\hbar} + n\omega_0.$$

Our method of solving Eq. (9) has been described in detail in [17], and will not be repeated here. Hence, we will adopt the wave function obtained in the paper [17]. Under the generalized rotating wave approximation, the time-dependent wave function can be expanded as

$$\begin{aligned} |\Psi(t)\rangle = e^{-i/\hbar \epsilon_1 t} \Bigg\{ & [\bar{a}_1(t) + \alpha_1(t)] |1\rangle \\ & + [\bar{a}_2(t) + \alpha_2(t)] \exp\left[-i/\hbar \left(\hbar n\omega_0 t + \int_0^t V_{22} dt \right) \right] |2\rangle \\ & + [\bar{a}_3(t) + \alpha_3(t)] \exp\left[-i/\hbar \left(\hbar n\omega_0 t + \int_0^t V_{33} dt \right) \right] |3\rangle \Bigg\}, \end{aligned} \tag{10}$$

where $\bar{a}_j(t)$ are the time-averaged probability amplitudes and $\alpha_j(t)$ are rapidly oscillating functions on the scale of the pump wave period. With the help of wave function (10) the expectation value of the dipole operator (8) can be written as

$$\begin{aligned} \langle D(t) \rangle = & d_{22} |\bar{a}_2(t) + \alpha_2(t)|^2 + d_{33} |\bar{a}_3(t) + \alpha_3(t)|^2 \\ & + (d_{12} [\bar{a}_1^*(t) + \alpha_1^*(t)][\bar{a}_2(t) + \alpha_2(t)] \\ & \times \sum_{s=-\infty}^{\infty} J_s(Z_2) e^{i(s-n)\omega_0 t} + d_{13} [\bar{a}_1^*(t) + \alpha_1^*(t)] \\ & \times [\bar{a}_3(t) + \alpha_3(t)] \sum_{s=-\infty}^{\infty} J_s(Z_3) e^{i(s-n)\omega_0 t} + \text{c.c.}). \end{aligned} \tag{11}$$

In deriving Eq. (11) we have applied the following expansion of Bessel functions:

$$e^{iZ \sin \alpha} = \sum_{s=-\infty}^{\infty} J_s(Z) e^{is\alpha}.$$

In Eq. (11) $Z_l = d_{ll} E / (\hbar \omega_0)$ $(l = 2, 3)$ are the dipole interaction energies in units of the pump wave photon energy.

For arbitrary initial conditions the time-averaged amplitudes $\bar{a}_j(t)$ are

$$\bar{a}_j = \sum_{\nu=1}^{3} C_{j\nu} \exp(i\lambda_\nu t), \tag{12}$$

where $C_{\eta\nu}$ are the constants of integration determined by the initial conditions, and the factors λ_ν are the solutions of the third-order characteristic equation

$$\begin{Vmatrix} \Delta_2 + \Delta_3 - \lambda & \Omega_{12} & \Omega_{13} \\ \Omega_{12}^* & -\delta_2 - \Delta_2 - \lambda & \Delta_{23} \\ \Omega_{13}^* & \Delta_{23}^* & -\delta_3 - \Delta_3 - \lambda \end{Vmatrix} = 0, \tag{13}$$

with

$$\Omega_{1l} = -\frac{d_{1l}}{d_{ll}} \omega_0 n J_n(Z_l), \tag{14}$$

$$\Delta_l = \omega_0 \frac{|d_{1l}|^2}{d_{ll}^2} \sum_{s \neq n} \frac{s^2 J_s^2(Z_l)}{s - n}, \quad l = 2, 3, \tag{15}$$

$$\Delta_{23} = -\omega_0 \frac{d_{12}^* d_{13}}{d_{22} d_{33}} \sum_{s \neq n} \frac{s^2 J_s(Z_2) J_s(Z_3)}{s - n}. \tag{16}$$

Here the terms Δ_2, Δ_3, and Δ_{23} describe the dynamic Stark shifts.

Assuming adiabatic turn-on of the pump wave, the rapidly oscillating parts of the probability amplitudes can be written as

$$\begin{aligned} \alpha_1(t) = & \bar{a}_2(t) \frac{d_{12}}{d_{22}} \sum_{s \neq n} \frac{s J_s(Z_2) e^{i(s-n)\omega_0 t}}{s - n} \\ & + \bar{a}_3(t) \frac{d_{13}}{d_{33}} \sum_{s \neq n} \frac{s J_s(Z_3) e^{i(s-n)\omega_0 t}}{s - n}, \end{aligned} \tag{17}$$

$$\alpha_2(t) = -\bar{a}_1(t) \frac{d_{12}^*}{d_{22}} \sum_{s \neq n} \frac{s J_s(Z_2) e^{-i(s-n)\omega_0 t}}{s - n}, \tag{18}$$

$$\alpha_3(t) = -\bar{a}_1(t) \frac{d_{13}^*}{d_{33}} \sum_{s \neq n} \frac{s J_s(Z_3) e^{-i(s-n)\omega_0 t}}{s - n}. \tag{19}$$

Combining Eqs. (11), (12), and (17)–(19) one can calculate analytically the expectation value of the dipole operator for an arbitrary initial atomic state. The Fourier transform of $\langle D(t) \rangle$ gives the coherent part of the dipole spectrum. As is clear from Eq. (11) the spectrum contains in general both even and odd harmonics. However, depending on the symmetry of the system, in particular, for spherical symmetry when momentum conservation occurs, the terms containing even harmonics cancel each other and only the odd harmonics are generated.

From Eq. (11) it is also clear that for effective harmonic generation in certain resonance conditions one should provide at first a considerable population transfer between the atomic states $[\bar{a}_1(t) \sim \bar{a}_{2,3}(t)]$. Second, since the Bessel function exponentially decreases with increasing index, one can conclude that the dipole interaction energy $d_{\max}E$ (here $d_{\max}=\max\{|d_{22}|,|d_{33}|\}$) should be comparable to or larger than the pump wave photon energy $\hbar\omega_0$. Then the cutoff harmonic s_c is determined from the condition $s_c - n \sim Z_{\max}$, i.e., the cutoff position depends linearly on the laser field amplitude.

For the resonance conditions with large photon numbers $n \gtrsim 10$ the Bessel function $J_n(Z)$ reaches its maximal value when $Z \simeq n$, i.e., $d_{\max}E \simeq n\hbar\omega_0$. This means that for efficient multiphoton excitation of atoms at $n \gtrsim 10$ the laser field should be strong. On the other hand, in this case the dynamic Stark shift becomes of the order of the laser frequency, so the resonance condition is violated, and consequently Rabi oscillations vanish. The latter restricts harmonic generation by the scheme considered to harmonic numbers $s \sim 10$ and the optimal parameters of the pump field can be estimated from the condition $E \sim \hbar\omega_0/d_{\max}$. Then, for a given pump wave intensity one should take an appropriate detuning to compensate for the dynamic Stark shift [17,20].

As is seen from Eqs. (14)–(16), the Rabi frequency is proportional to the ratio d_{1l}/d_{ll}, while the dynamic Stark shifts are proportional to $|d_{1l}|^2/d_{ll}^2$. Therefore, for systems with $|d_{1l}|^2/d_{ll}^2 \ll 1$ the dynamic Stark shift plays a minor role. Hence, one can achieve efficient harmonic generation in systems with large mean dipole moments such as occur in hydrogenlike atoms. Note that polar molecules may be more favorable in this respect due to their larger permanent dipole moments.

For concreteness, let us consider a hydrogenlike atom. In this case, taking into account the problem's symmetry, it is convenient to use parabolic coordinates [17]. In these coordinates we have the V configuration illustrated in Fig. 1(a) with $\epsilon_2=\epsilon_3$. Without loss of generality one can take the polarization vector $\hat{\epsilon}$ aligned with the z axis of the parabolic coordinates. The excited states have opposite mean dipole moments $d_{33}=-d_{22}\equiv d=3$ a.u., and the transition matrix elements of the electric dipole moment are $d_{12}=-d_{13}\equiv d_{\mathrm{tr}}=2^7/3^5$ a.u. Then $\Delta_2=\Delta_3\equiv\Delta$, $\delta_2=\delta_3\equiv\delta$, $Z_3=-Z_2\equiv Z$, and

$$\Omega_{13}=(-1)^n\Omega_{12}\equiv\Omega=n\omega_0\frac{d_{\mathrm{tr}}}{d}J_n(Z). \tag{20}$$

For compensation of the dynamic Stark shift one should take an appropriate detuning. From Eq. (13) it follows that the desired detuning is

$$\delta=\delta_{\mathrm{St}}\equiv-3\Delta+(-1)^n\Delta_{23}. \tag{21}$$

In this case the solution (12) for the system situated initially in the ground state is

$$\bar{a}_1=e^{-i2\Delta t}\cos(\sqrt{2}\Omega t),$$

$$\bar{a}_3=(-1)^n\bar{a}_2=\frac{e^{-i2\Delta t}}{i\sqrt{2}}\sin(\sqrt{2}\Omega t). \tag{22}$$

Replacing the probability amplitudes in Eq. (11) by the corresponding expressions (22) one can derive the final analytical expression for $\langle D(t)\rangle$. Here, one can neglect second-order terms of the rapidly oscillating parts of the probability amplitudes, since $\alpha_l^2(t) \sim d_{\mathrm{tr}}^2/d^2 \ll 1$. This leads to a compact analytic formula, and we find

$$\langle D(t)\rangle=\sum_k\{S_k\sin[(2k+1)\omega_0 t]+C_k\cos[(2k+1)\omega_0 t]\}, \tag{23}$$

where

$$S_k=\sqrt{2}d_{\mathrm{tr}}\frac{nJ_{2k+1+n}(Z)}{2k+1}\sin\Omega_R t, \tag{24}$$

$$C_k=\frac{d_{\mathrm{tr}}^2}{d}\sum_{s\neq n}\{(-1)^{n-s}-1-[3+(-1)^{n-s}]\cos\Omega_R t\}\times\frac{sJ_{2k+1+s}(Z)J_s(Z)}{s-n}. \tag{25}$$

Here

$$\Omega_R\equiv 2\sqrt{2}n\omega_0\frac{d_{\mathrm{tr}}}{d}J_n(Z) \tag{26}$$

is the generalized Rabi frequency at n-photon resonance which has a nonlinear dependence on the amplitudes of the wave fields through Bessel functions. As is seen from Eq. (23), for these initial conditions ($|1S\rangle$ state with definite parity) only the odd harmonics appear in the spectrum. For the generation of even harmonics one should break inversion symmetry. The inversion symmetry of the atom can be broken by a weak static uniform electric field, or the initial stationary state should not have this symmetry allowing radiation of all even and odd harmonics. For the hydrogen atom both cases can be realized and will be considered in the next section.

III. NUMERICAL TREATMENT

In this section we present some numerical simulations for the hydrogen atom with specific parameters of the laser and initial conditions. The time evolution of the system was found with the Runge-Kutta method and the Fourier transformations were performed using the fast Fourier transform algorithm [26]. The calculations were made for a quasimonochromatic wave field providing only adiabatic turn on of the interaction. The latter is achieved by describing the envelope with the hyperbolic tangent function $\tanh(t/\tau)$, where τ characterizes the turn-on time. For all calculations we have taken $\omega_0\tau=20\pi$.

Figure 2 displays the harmonic emission rate via $\log_{10}[S_C(\omega)]$ (coherent part) as a function of the harmonic order (ω/ω_0) at four-photon resonance $(n=4)$ for the hydrogen atom situated initially in the ground state. The laser field

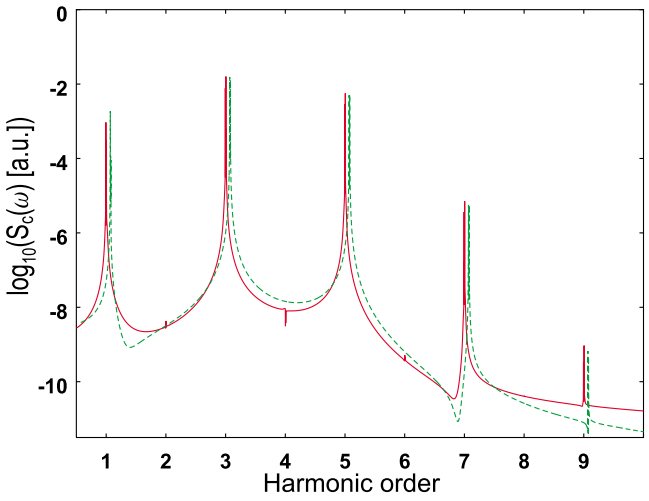

FIG. 2. (Color online) Harmonic emission rate via $\log_{10}[S_C(\omega)]$ (coherent part) as a function of the harmonic order (ω/ω_0) at four-photon resonance $(n=4)$ for a hydrogen atom situated initially in the ground state. The laser field strength is $E=0.032$ a.u., and frequency is $\omega=0.094$ a.u. 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 right).

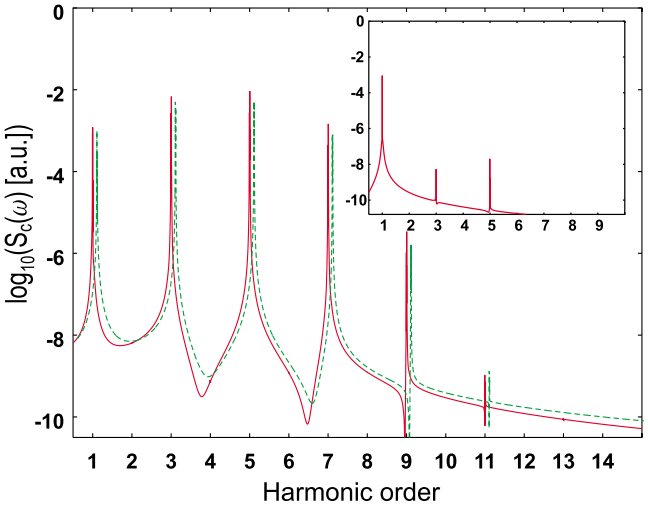

FIG. 3. (Color online) Harmonic emission rate via $\log_{10}[S_C(\omega)]$ as a function of the harmonic order (ω/ω_0) at five-photon resonance $(n=5)$ for a hydrogen atom situated initially in the ground state. The laser field strength is $E=0.04$ a.u., and frequency is $\omega=0.0754$ a.u. The solid (red) line corresponds to numerical calculations; the dashed (green) line corresponds to the approximate solution (slightly shifted). The inset represents the spectrum for a two-level atom with a zero dipole moment in a stationary state.

strength has been chosen to be $E=0.032$ a.u., and frequency is $\omega=0.094$ a.u. For these parameters the dynamic Stark shift is compensated. The latter provides almost complete population transfer. The solid (red) line corresponds to numerical calculations, while the dashed (green) line corresponds to the approximate expression (23). Note that the numerical and analytical calculations coincide with high accuracy, so for visual convenience (to distinguish these curves) the spectrum corresponding to analytical calculations Eq. (23) has been slightly shifted to the right. Hence, the obtained approximate solution is in good agreement with numerical results.

In Fig. 3 we plot the coherent part of the spectrum for five-photon resonance $(n=5)$ for the hydrogen atom situated initially in the ground state. The laser field strength is $E=0.04$ a.u., and frequency is $\omega=0.0754$ a.u. For the chosen parameters the dynamic Stark shift is compensated. The solid (red) line corresponds to numerical calculations; the dashed (green) line corresponds to our slightly shifted approximate solution. To show the advantage of the three-level system, we also present the coherent part of the spectrum for a two-level atom without a mean dipole moment in the excited state under the same conditions of excitation. The inset represents the spectrum for a two-level atom where the main contribution in the spectrum gives the first harmonic. The difference between these systems is significant. The main difference is the appearance of higher harmonics with peaks of much larger amplitudes at the same positions of the spectra. This is the result of multiphoton resonant interaction with the driving laser field, which is much more efficient in the three-level system than in the two-level one.

As has been mentioned in the previous section, if the inversion symmetry is broken, the atom may radiate all harmonics. Figure 4 displays the coherent part of the spectrum at four-photon resonance $(n=4)$ for the hydrogen atom situated initially in the excited state $|2\rangle$. The parameters are the same as in the case of Fig. 2. In contrast to Fig. 2, even harmonics appear in the spectrum. This is a consequence of the fact that in the state $|2\rangle$ (or state $|3\rangle$) the atom does not have definite parity since the state $|2\rangle$ is a superposition of $|2S\rangle$ and $|2P\rangle$ states: $|2\rangle=(|2S\rangle+|2P\rangle)/\sqrt{2}$.

The inversion symmetry may also be broken with an external perturbation, e.g., with an additional static uniform electric field. In the calculations we have applied a weak electric field of strength $E_0=5\times10^{-6}$ a.u. The linear Stark shift for the hydrogen atom excludes the degeneracy causing energy level splitting: $\epsilon_2-\epsilon_3=2dE_0$. As a consequence, the

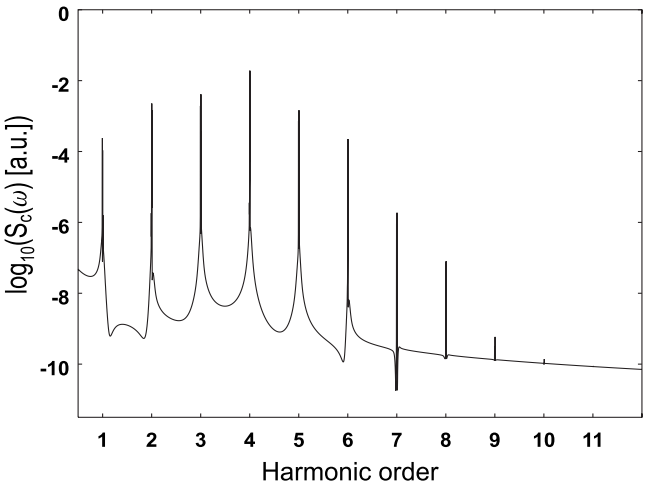

FIG. 4. Coherent part of the spectrum at four-photon resonance $(n=4)$ for a hydrogen atom situated initially in the excited Stark mixed superposition state. The laser field strength is $E=0.032$ a.u., and frequency is $\omega=0.094$ a.u.

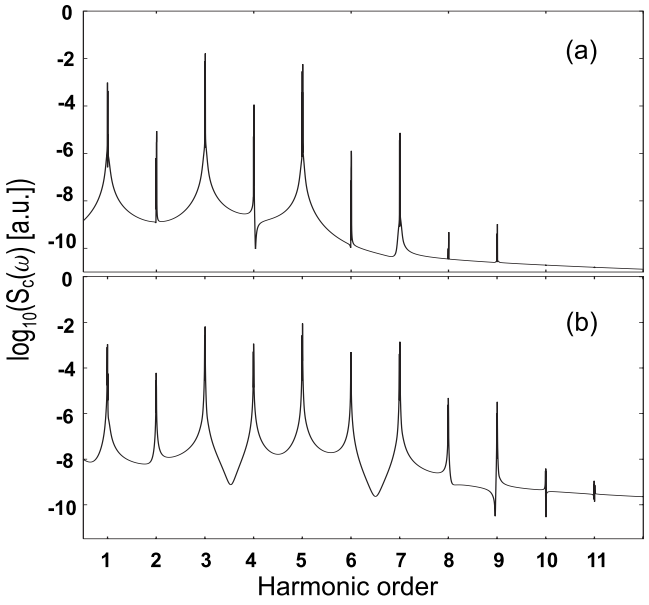

FIG. 5. Coherent part of the spectrum for a hydrogen atom situated initially in the ground state, with additional uniform electric field of strength $E_0=5\times10^{-6}$ a.u. Harmonic emission rate for (a) four-photon resonance ($E=0.032$ a.u, $\omega=0.094$ a.u.) and (b) five-photon resonance ($E=0.04$ a.u, $\omega=0.0754$ a.u.).

even harmonics appear in the scattering spectrum. In Fig. 5 we plot the coherent part of the spectrum for the hydrogen atom in a static electric field, situated initially in the ground state. Figures 5(a) and 5(b) display harmonic emission rate for four-photon ($E=0.032$ a.u., $\omega=0.094$ a.u.) and five-photon ($E=0.04$ a.u., $\omega=0.0754$ a.u.) resonances, respectively.

For comparison of the considered scheme with the conventional tunneling high-order harmonic generation (HHG), let us make some estimations for interaction parameters close to those of Refs. [27]. In these works tunneling HHG is considered, and it is shown that with Xe atoms and comparable laser intensities one can produce harmonics $s\sim10$ with a conversion efficiency of about 5×10^{-7}.

As is seen from Eqs. (23), the Fourier component of the dipole moment d_s varies much more slowly with the laser intensity than the perturbative power law E^s. Hence, one can make an estimation using known results for superradiation of a coherently excited multiatom ensemble [[4](a)].

Thus, if the observed wavelengths are much smaller than the transverse size of the interaction region, the coherent radiation from the atoms will occur primarily along the propagation axis of the pump laser beam and will cover only a tiny solid angle $\sim\lambda_s^2/S$, which will be defined by the transverse size of the interaction region S (λ_s is the sth-harmonic wavelength). The latter is assumed to be limited due to the focusing of the incident radiation. Then, for the radiation power we have

$$P_s \simeq P_{1s}N^2\mu, \tag{27}$$

where $P_{1s}=4s^4\omega_0^4|d_s|^2/(3c^3)$ is the single-atom radiation power with the Fourier component of the dipole moment d_s evaluated at the maximum intensity of the incident pulse. The interference factor μ is defined by the atomic form factor and depends on the interaction geometry [[4](a)]. For the cylindrical system it can be estimated as $\mu\simeq3\lambda_s^2/(8\pi S)$. In Eq. (27) N is the number of atoms in the interaction region $N=L_cSN_0$, where N_0 is the atomic density and L_c is the coherence length over which the sth harmonic field can be built up coherently. This coherence length L_c may be limited due to the dispersion in the medium, $L_c\sim1/\Delta k$, where Δk is the phase mismatch between the polarization and harmonic fields, due to the finite length of the atomic medium, or due to the focal size of the incident radiation, $L_c\sim L_R$ ($L_R=S/\lambda_s$ is the Rayleigh length). The last effect in general is dominant for moderate harmonics. Hence, for the radiation power we obtain

$$P_s \simeq (2\pi)^2 s\omega_0|d_s|^2N_0^2L_R^3. \tag{28}$$

The average number of photons emitted at each laser shot for the sth harmonic can be estimated as

$$N_s \simeq \frac{P_s\tau}{\hbar s\omega_0} = \frac{(2\pi)^2}{\hbar}|d_s|^2N_0^2L_R^3\tau, \tag{29}$$

where τ is the pulse duration.

The incident pulse duration is assumed to be $\tau\simeq40$ ps, the Rayleigh length is taken to be $L_R\simeq1$ mm, and for atomic density we assume $N_0\simeq5\times10^{17}$ cm^{-3}. For the setup of Fig. 3 with the chosen parameters, the average number of radiated photons per shot is $N_{3/7}\sim10^{13}$. This corresponds to a conversion efficiency of about 10^{-3}, which is three orders of magnitude larger than what one expects to achieve with tunneling HHG [27].

IV. CONCLUSION

We have presented a theoretical treatment of the coherent light scattering by a three-level atom under multiphoton resonant excitation. The coherent part of the dipole spectrum was investigated. With the help of an approximate analytical expression for the dynamic wave function of a three-level atom driven by an intense laser field, we obtained an analytical expression for the time-dependent expectation value of the dipole operator. Then the results obtained were applied to the hydrogen atom. We also made numerical calculations for the hydrogen atom, considering various initial conditions. In the case of broken inversion symmetry, the spectrum contains both even and odd harmonics. The spectrum shows a significant difference compared to the spectrum for a two-level atom without a mean dipole moment in the excited state under the same conditions of excitation. The main difference is the appearance of higher harmonics in the spectrum. These peaks have quite large amplitudes. The latter is the result of multiphoton resonant interaction of the atom with the driving laser radiation due to the mean dipole moment in the excited states. The cutoff position depends linearly on the laser field amplitude. Analytical calculations in

the generalized rotating wave approximation [17] allow an explanation of the obtained spectrum. The numerical simulations are in good agreement with the analytical results. The considered scheme may serve as a promising method for efficient production of moderately high harmonics. Note that for the coherent radiation of a multiatom ensemble one needs a more rigorous theory, which should account for collective effects in the medium arising from Maxwell's equations. This issue is currently under investigation and will be the subject of a future presentation.

ACKNOWLEDGMENTS

This work was supported by International Science and Technology Center (ISTC) Project No. A-1307. B.R.A. wishes to thank NFSAT for support.

[1] N. Bloembergen, *Nonlinear Optics* (Benjamin, New York, 1965); R. W. Boyd, *Nonlinear Optics* (Academic, San Diego, 1992).
[2] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Lett. **7**, 118 (1961).
[3] M. Protopapas, C. H. Keitel, and P. L. Knight, Rep. Prog. Phys. **60**, 389 (1997); P. Salières A. L'Huillier, P. Antoine, and M. Lewenstein, Adv. At., Mol., Opt. Phys. **41**, 83 (1999); T. Brabec and F. Krausz, Rev. Mod. Phys. **72**, 545 (2000); D. B. Milosevic and F. Ehlotzky, Adv. At., Mol., Opt. Phys. **49**, 377 (2003); Y. I. Salamin, K. Z. Hatsagortsyan, and C. H. Keitel, Phys. Rep. **427**, 41 (2006).
[4] (a) L. Allen and J. H. Eberly, *Optical Resonance and Two Level Atoms* (Wiley-Interscience, New York, 1975); (b) B. W. Shore, *Theory of Coherent Atomic Excitation* (Wiley-Interscience, New York, 1990); (c) M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, U.K., 1997).
[5] B. R. Mollow, Phys. Rev. **188**, 1969 (1969); Phys. Rev. A **12**, 1919 (1975); F. Y. Wu, R. E. Grove, and S. Ezekiel, Phys. Rev. Lett. **35**, 1426 (1975); R. E. Grove, F. Y. Wu, and S. Ezekiel, Phys. Rev. A **15**, 227 (1977).
[6] S. Haroche and F. Hartmann, Phys. Rev. A **6**, 1280 (1972); C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977).
[7] P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980).
[8] P. W. Milonni and J. H. Eberly, *Lasers* (Wiley, New York, 1988), Sec. 17.6.
[9] B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990).
[10] J. H. Eberly and M. V. Fedorov, Phys. Rev. A **45**, 4706 (1992).
[11] D. G. Lappas, M. V. Fedorov, and J. H. Eberly, Phys. Rev. A **47**, 1327 (1993).
[12] G. Compagno, K. Dietz, and F. Persico, J. Phys. B **27**, 4779 (1994).
[13] E. Conejero Jarque and L. Plaja, J. Phys. B **31**, 1687 (1998).
[14] F. I. Gauthey, C. H. Keitel, P. L. Knight, and A. Maquet, Phys. Rev. A **55**, 615 (1997).
[15] A. DiPiazza, E. Fiordilino, and M. H. Mittleman, Phys. Rev. A **64**, 013414 (2001).
[16] R. E. Duvall, E. J. Valeo, and C. R. Oberman, Phys. Rev. A **37**, 4685 (1988).
[17] H. K. Avetissian and G. F. Mkrtchian, Phys. Rev. A **66**, 033403 (2002).
[18] G. N. Gibson, Phys. Rev. Lett. **89**, 263001 (2002).
[19] H. K. Avetissian, G. F. Mkrtchian, and M. G. Poghosyan, Phys. Rev. A **73**, 063413 (2006).
[20] H. K. Avetissian, B. R. Avchyan, and G. F. Mkrtchian, Phys. Rev. A **74**, 063413 (2006).
[21] A. Brown, W. J. Meath, and P. Tran, Phys. Rev. A **63**, 013403 (2000).
[22] R. S. Mulliken, J. Chem. Phys. **7**, 20 (1939).
[23] G. Bandarage, A. Maquet, and J. Cooper, Phys. Rev. A **41**, 1744 (1990); G. Bandarage, A. Maquet, T. Menis, R. Taieb, V. Veniard, and J. Cooper, *ibid.* **46**, 380 (1992).
[24] M. Casu and C. H. Keitel, Europhys. Lett. **58**, 496 (2002).
[25] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics* (Butterworth-Heinemann, Oxford, 1981).
[26] W. H. Press, S. A. Teukolsky, W. T. Vetterling, and B. P. Flannery, *Numerical Recipes in C* (Cambridge University Press, Cambridge, U.K., 1992).
[27] A. L'Huillier, K. J. Schafer, and K. C. Kulander, J. Phys. B **24**, 3315 (1991); Ph. Balcou, C. Cornaggia, A. S. L. Gomes, L. A. Lompre, and A. L'Huillier, *ibid.* **25**, 4467 (1992).