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Letter

Prospects of odd and even harmonic generation by an atom in a high-intensity laser field

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Abstract

A new approach for studying the spontaneous emission of an atomic system in the presence of a high-intensity laser field is used to study the process of harmonic generation. The analysis is based on consideration of quantum system interaction, with the quantized field modes being in the vacuum state, while the intense laser field is considered to be classically beyond perturbation theory. The numerical analysis of the emission from the single one-electron 1D atom irradiated by the femtosecond laser pulse of a Ti:Sa laser is discussed. It is demonstrated that not only odd, but also even harmonics can be emitted if the laser field is strong enough. The origin of the appearance of even harmonics is studied. The obtained results are compared with those found in the framework of the semiclassical approach that is widely used to study harmonic generation. It is found that the semiclassical approach is inapplicable in the strong-field limit.

Keywords: strong-field atomic dynamics, numerical modeling, harmonic generation, nonlinear optics

(Some figures may appear in colour only in the online journal)

1. Introduction

Strongly nonequilibrium plasma produced in gases by high-intensity femtosecond laser pulses has attracted interest because of a number of specific electrodynamic properties that can be used for the generation and amplification from x-ray and EUV radiation to the THz frequency band [1–13]. In particular, the filamentation phenomenon can be very important in the physics of the generation and (or) amplification of radiation [14].

Typically, spontaneous emission from atoms and ions is not taken into account when the strong-field dynamics of an atomic system is studied. In reality, the value of the vacuum modes of the electric field strength is negligible in comparison with those in high-intensity femtosecond laser pulses. The typical time allowed in dipole approximation transitions for visible or UV radiation is in the range 10^{-6}–10^{-8} s, which is shorter than the duration of a femtosecond pulse by many orders of magnitude. Hence, it seems that there is no necessity to take such transitions into account. On the other hand, any nonlinear process in plasma starts from spontaneous emission and this emission should at least be taken into account in the early stages of generation. To overcome this difficulty, the semiclassical approach [15] is now widely used. This approach is based on the calculation of the response of the quantum system driven by the laser field using the average over the atomic wave function $\psi(\vec{r}, t)$ of the dipole moment

$$\vec{d}(t) = -e\int|\psi(\vec{r},t)|^2\,\vec{r}\,\mathrm{d}^3r, \quad (1)$$

where $\psi(\vec{r},t)$ describes the evolution of an atomic system in a laser field. If a laser field is weak enough, i.e. its strength is much smaller than that of the atomic field, perturbation theory can be applied and one obtains the decomposition of the response over the field strength powers [16]. If the field is strong and perturbation theory is not applicable to study the quantum dynamics, the numerical solution of the non-stationary Schrödinger equation for the quantum system in a laser field can be used. Such an approach for determining the response and studying the contribution of different processes to filamentation was used in [17, 18].

A similar approach was applied to study the atomic high-frequency response and high-order harmonic generation (HHG) in the last two decades [19–23]. In this case the intensity of emission was calculated as

$$I_\omega \sim \omega^4\left|\vec{d}_\omega\right|^2, \quad (2)$$

where d_ω is the Fourier transform of the dipole moment (1):

$$\vec{d}_\omega = \frac{1}{\sqrt{2\pi}}\int\vec{d}(t)\exp(-\mathrm{i}\omega t)\mathrm{d}t. \quad (3)$$

Such an approach was found to be extremely fruitful—see, for example, a recent review [5] and references therein.

Nevertheless, the possibility of applying the semiclassical approach to study harmonic generation and the polarization response in the case of a high-intensity laser field when the population of the initial (ground) state is depleted during the laser pulse action was questioned recently in [24]. It was demonstrated that the application of the semiclassical approach to study the emission of a quantum system driven by a high intensity laser field is generally in contradiction with quantum electrodynamical calculations. In [25], a new approach for studying the spontaneous emission of an atomic system in the presence of a high-intensity laser field was developed. This approach is based on first order perturbation theory applied to the interaction of an atomic system dressed by an external laser field, with many quantized field modes under the assumption that initially all the modes are in a vacuum state.

Here we reformulate this approach for the velocity gauge and apply it to study the emission of a model single-electron atom driven by a femtosecond pulse from a Ti:Sa laser ($\hbar\omega = 1.55$ eV). We find that odd harmonics of fundamental frequency are only emitted by the atom in rather weak external laser fields when the strong-field atomic dynamics can be studied in the framework of quantum-mechanical perturbation theory. Beyond the applicability of perturbation theory in the regime of effective atomic ionization, both odd and even harmonics of fundamental frequency are found to exist as a result of the electron bremsstrahlung in a strong laser field.

Comparative analysis of the obtained data with those derived in the semiclassical approximation is performed. It is found that the semiclassical approach that is widely used to study high-order harmonic generation fails in the strong field limit when the population of the ground state is depleted and essential ionization takes place.

2. Theoretical model of the interaction of an atomic system driven by a classical external field with quantized field modes

We start the analysis of the spontaneous emission of an atomic system using the Hamiltonian

$$H = H_0(\vec{r},t) + H_f(\{a\}) + V(\vec{r},\{a\}), \quad (4)$$

where $H_0 = H_{\mathrm{at}}(\vec{r}) + W(\vec{r},t)$; $H_{\mathrm{at}}(\vec{r})$ is the atomic Hamiltonian, and

$$W = -\frac{e}{mc}\vec{A}(t)\vec{p} + \frac{e^2A^2(t)}{2mc^2} \quad (5)$$

is the interaction of an atom with a classical laser field in the velocity gauge in the dipole approximation, $\vec{A}(t)$ is the vector potential of the classical field, $\vec{p} = -\mathrm{i}\hbar\nabla$ is the momentum operator, $H_f(\{a\})$ is the Hamiltonian of the set of field modes excluding the laser field mode, $V(\vec{r},\{a\})$ stands for the interaction of an atomic electron with the quantized electromagnetic field, $\vec{r}$ is the electron radius vector and $\{a\}$ is the set of quantized field mode coordinates.

Let us assume that we know the solution of the nonstationary problem for the atomic dynamics in the classical field

$$\mathrm{i}\hbar\frac{\partial\psi(\vec{r},t)}{\partial t} = H_0(t)\psi(\vec{r},t), \quad (6)$$

with the initial condition $\psi(\vec{r},t=0) = \varphi(\vec{r})$, where $\varphi(\vec{r})$ is a given stationary or unstationary state of the atomic discrete spectrum or continuum.

We will also suppose that at the initial instant of time all the field modes are in the vacuum state $|\{0\}\rangle$. Then the solution of the general equation with the Hamiltonian (4)

$$\mathrm{i}\hbar\frac{\partial\Psi(\vec{r},\{a\},t)}{\partial t} = (H_0(t) + H_f + V)\Psi(\vec{r},\{a\},t) \quad (7)$$

and initial condition $\Psi(\vec{r},\{a\},t=0) = \varphi(\vec{r})\times|\{0\}\rangle$ can be found by means of perturbation theory.

The wave function of the zero-order approximation, excluding interaction with the quantum field modes, reads as

$$\Psi^{(0)}(\vec{r},\{a\},t) = \psi(\vec{r},t)\times|\{0\}\rangle. \quad (8)$$

We are going to find the solution of (7) in the form

$$\Psi(\vec{r},\{a\},t) = \Psi^{(0)}(\vec{r},\{a\},t) + \delta\Psi(\vec{r},\{a\},t), \quad (9)$$

with $\delta\Psi \ll \Psi^{(0)}$.

For further analysis let us recall that initially we have vacuum in all of the field modes. Therefore, in the first order of perturbation theory, $\delta\Psi$ contains only one-photon excitations in a field mode:

$$\delta\Psi(\vec{r},\{a\},t) = \sum_{k,\lambda}\delta\psi_{k\lambda}(\vec{r},t)\times\{0,0,....1_{k\lambda},0,...0,0\}. \quad (10)$$

Here $\delta\psi_{k\lambda}(\vec{r},t)$ is the electron wave function, provided that one photon with wave vector $\vec{k}$ and polarization λ has appeared.

As the interaction of the atom with quantized field can be written in a form

Table 1. Xenon energy levels obtained in the numerical simulations for potential (18).

Principal quantum number, n	Energy (eV)
1	−12.134
2	−5.910
3	−3.457
4	−2.220
5	−1.550
6	−1.135
7	−0.870
8	−0.685
9	−0.555
10	−0.457
11	−0.385

$$V(\vec{r},\{a\}) = \sum_{k,\lambda} V_{k\lambda} = -\frac{e}{mc}\sum_{k\lambda}(\vec{e}_{k\lambda}\vec{p})a_{k\lambda}, \tag{11}$$

where $a_{k\lambda}$ is the vector potential operator of the mode $\{k,\lambda\}$ and $\vec{e}_{k\lambda}$ is the polarization vector, for a given mode with one-photon excitation one has the problem

$$\mathrm{i}\hbar\frac{\partial\delta\psi_{k\lambda}(\vec{r},t)}{\partial t} = H_0(t)\delta\psi_{k\lambda}(\vec{r},t) - \frac{e(\vec{e}_{k\lambda}\vec{p})}{mc} \times \frac{a_{\mathrm{norm}}}{\sqrt{2}} \times \psi(\vec{r},t)\times\exp(\mathrm{i}\omega_{k\lambda}t), \tag{12}$$

where $a_{\mathrm{norm}} = \sqrt{4\pi\hbar c^2/(\omega_{k\lambda}L^3)}$, L^3 is the normalization volume, and the initial condition is $\delta\psi_{k\lambda}(\vec{r},t=0)=0$.

Thus, we have the set of equations for atomic system evolution, provided that one photon in the mode $\{\vec{k},\lambda\}$ has appeared. It is obvious that the expression

$$W_{k\lambda}(t) = \int|\delta\psi_k(r,t)|^2\mathrm{d}^3r \tag{13}$$

represents the probability of finding a photon in the mode $\{\vec{k},\lambda\}$ as a function of time. Then the total probability of emitting a photon of any frequency and polarization during the transition $f\rightarrow i$ is

$$W_{fi}(t) = \sum_{k,\lambda} W_{k\lambda}(t). \tag{14}$$

As the spectrum of field modes is dense, we can replace the sum in (14) by the integral over field modes. After integration over the angular distribution of the photons and summation over the possible polarizations, the probability of spontaneous decay in the spectral interval $(\omega,\omega+\mathrm{d}\omega)$ can be expressed in the form

$$W_\omega\mathrm{d}\omega = \frac{L^3}{3\pi^2c^3}\omega^2\mathrm{d}\omega\times W_{k=\omega/c,\lambda}, \tag{15}$$

where $W_{k,\lambda}$ is given by (13). One should note that expression (15) does not depend on the normalization volume, as $W_{k,\lambda}\sim 1/L^3$.

To provide more insight into the physics of spontaneous emission in the presence of a strong laser field, the wave functions $\delta\psi_{k\lambda}(\vec{r},t)$ should be represented as a superposition of the stationary states of the atomic Hamiltonian:

$$\delta\psi_{k\lambda}(\vec{r},t) = \sum_n C_n^{(k\lambda)}(t)\phi_n(\vec{r})\exp\left(-\frac{\mathrm{i}}{\hbar}E_nt\right). \tag{16}$$

The squared coefficients of the decomposition (16) $\left|C_n^{(k\lambda)}\right|^2$ find the atom in the states $|n\rangle$ under the assumption that the emitted photon is in the definite mode $\{k,\lambda\}$. We will further use these values to interpret the results of the numerical simulation.

We would like to note that the discussed problem can also be formulated in the length gauge [25]. In this case

$$W = -\vec{d}\vec{\varepsilon}(t), \tag{17}$$

where $\vec{d}$ is the dipole operator and $\vec{\varepsilon}(t) = -\frac{1}{c}\frac{\mathrm{d}\vec{A}}{\mathrm{d}t}$ is the electric field strength. However, it is known [15] that if the dipole approximation is valid, both gauges are equivalent to each other.

3. Numerical model

In this section we briefly describe the numerical model that was used to study the spontaneous emission of a quantum system driven by a high-intensity laser field. We studied a 1D single-electron atomic system with a Coulomb-screened potential [26]

$$V(x) = -\frac{e^2}{\sqrt{\alpha^2+x^2}}, \tag{18}$$

with a screening parameter $\alpha = 1.6165a_0$, where a_0 is the Bohr radius. For such a value of α the ionization potential is 12.13 eV, which corresponds to the ionization potential of a xenon atom. The set of energy levels in such a xenon-like atom can be found in table 1.

We will also discuss the set of calculations for an atom initially prepared in the ground state $n=1$ and exposed to the radiation of the Ti:Sa laser ($\hbar\omega = 1.55$ eV) with a trapezoidal sine-squared pulse with a plateau of duration t_{p} and smoothed sine-squared fronts of duration t_{f}, so that the total pulse duration was $\tau_{\mathrm{p}} = t_{\mathrm{p}} + 2t_{\mathrm{f}}$. The parameters t_{p} and t_{f} were chosen to be equal to two and ten optical cycles (OC).

According to the above-mentioned model, we solved equation (6) for the evolution of the atomic wave function self-consistently with the set of equations (12) for one-photon excitations in different quantized field modes. The procedure is described briefly in [25]. The frequency interval $\Delta\omega$ between the quantized field modes was typically 0.02 of the fundamental frequency, while the number of modes was 400–1200, depending on the chosen laser intensity value. The modeling was performed for a time interval that exceeded 2× the duration of pulse $\tau = 2\tau_{\mathrm{p}}$. This allowed one to distinguish the spontaneous transitions that are possible without laser field from the stimulated transitions, such as the Raman and Rayleigh types, or the stimulated bremsstrahlung, when spontaneous photons only appear during the laser pulse action.

To compare the obtained spectra with those obtained in the semiclassical model, the semiclassical probability of emission in the spectral interval $(\omega,\omega+\mathrm{d}\omega)$ was also calculated

Table 2. Ionization yield and probability of staying in the ground state ($\hbar\omega = 1.55$ eV).

Intensity (W cm^{-2})	Ionization yield	Ground state population
1×10^{13}	4.37274×10^{-4}	0.99927
2×10^{13}	0.00836	0.99086
3×10^{13}	0.08737	0.84413
4×10^{13}	0.28431	0.70324
5×10^{13}	0.52553	0.45724
6×10^{13}	0.45933	0.47493
8×10^{13}	0.58327	0.16376
1×10^{14}	0.94116	0.01716

$$W_{\omega}^{(\mathrm{semi})} = \frac{4\omega^3}{3\hbar c^3}|d_{\omega}|^2, \qquad (19)$$

where d_{ω} was taken from (1) and (3).

4. Discussion

We will start the discussion of atomic dynamics in a strong laser field and harmonic generation with the data for the ionization yield and the probability of staying in the ground state in dependence on laser intensity (see table 2).

First, we note that up to the intensity values $\leqslant 1-2\times10^{13}$ W cm^{-2} the atom predominantly stays in its ground state. According to [24], that is the regime where the semiclassical approach can really be used to study spontaneous emission. Our simulations partially confirm this statement. The semiclassical calculations for atomic emission are presented in figure 1(a). Due to the definite parity of the atomic stationary states, the peaks in the spectra only correspond to the odd harmonics. Three additional peaks are observed at energies ~6.22, 9.92 and 11.01 eV. These lines mainly appear in the after-pulse regime due to the population of states $|n=2\rangle$, $|n=4\rangle$ and $|n=6\rangle$ and hence represent the oscillation of the dipole moment at consequent frequencies. These lines can be interpreted as spontaneous transitions to the ground state.

The results of the quantum-mechanical simulations of the atomic spontaneous emission for the same laser intensity are presented at figure 1(b). One can see that there are two groups of lines in the spectra: one group corresponds to the lines appearing during the laser pulse, while the other includes the lines emitted predominantly in the after-pulse regime. The first group of lines mainly contains the emission at the fundamental frequency and its third and fifth harmonics. We can state that these harmonics are the result of Rayleigh $|n=1\rangle+\hbar\omega\rightarrow|n=1\rangle+\hbar\omega_{k\lambda}$ ($\omega=\omega_{k\lambda}$) and hyper-Rayleigh scattering when three of the five laser photons are absorbed and one phonon is emitted. These lines are in rather good agreement with the semiclassical calculations.

In addition to the lines corresponding to the generation of odd harmonics, several lines are found to emerge in the after-pulse regime that in part do not appear in the semiclassical calculations. Lines $\hbar\omega_{k\lambda}=6.22$, 9.92, 11.01 eV correspond to the $|n=2,4,6\rangle\rightarrow|1\rangle$ series of the spontaneous emission from levels excited during the laser pulse action. These lines are also observed in the semiclassical model. Nevertheless, one can distinguish a number of additional lines that do not exist in the semiclassical model. Among these are the lines with $\hbar\omega_{k\lambda}=2.45$, 4.31, 5.02, 5.36, 5.58 eV that form the series $|n=3,5,7,9,11\rangle\rightarrow|2\rangle$. The lines with $\hbar\omega_{k\lambda}=1.23$ eV can be associated with the $|n=4\rangle\rightarrow|3\rangle$ transition. In both cases, all of these lines correspond to the transitions to unpopulated excited states and hence cannot be observed in the semiclassical model [25].

The next point in our discussion concerns spontaneous emission in the strong field limit when an atom is predominantly ionized during the laser pulse. The semiclassical and quantum-mechanical spectra for the intensity value 10^{14} W cm^{-2} are presented in figure 2. For this intensity value the probability of ionization is ≈ 0.94 while the ground state is depleted up to ≈ 0.017. In both cases one observes harmonics of the fundamental frequency with a plateau-like structure up to energies $\approx 3.17U_p + I_i$ (U_p is the ponderomotive potential and I_i is the ionization potential of the atom). We would also like to mention the existence of spontaneous emission lines that correspond to the transitions $|4\rangle\rightarrow|1\rangle$ and $|2\rangle\rightarrow|1\rangle$. Nevertheless, one can see that the results of the quantum-mechanical calculations differ dramatically from those of the semiclassical analysis. First, not only odd, but also even harmonics are observed in the emission spectra in a relatively low-energy part of the spectra $\hbar\omega_{k\lambda}\leqslant 10$ eV. In the high-energy part of the spectra, odd-order harmonics still dominate.

To provide an insight into the physical processes of harmonic emission in the regime of strong field ionization, hereafter we analyze the distribution of the population of the ground state in dependence on harmonic frequency.

First, for a strong laser field when the probability of ionization is high, the peak with $\omega_{k\lambda}=\omega$ can have a different physical nature. In addition to the process of Rayleigh scattering, it can arise from the near-free electron oscillations at the radiation frequency in the area of the atomic potential. To distinguish these mechanisms, the distribution of probabilities $\left|C_n^{(k\lambda)}\right|^2$ under the assumption that the photon is in the mode $\{k,\lambda\}$ with $\hbar\omega=1.55$ eV was calculated. It was found that for $\hbar\omega_{k\lambda}=1.55$ eV the probability of finding the atom in the ground state is ≈ 0.003, and hence the bremsstrahlung dominates. A similar situation is also realized for a number of higher harmonics with $\omega_{k\lambda}=m\omega$, $(m=2-5)$. All of them come from the bremsstrahlung. To the extent that the continuum states are degenerated and have different spatial parity for the same energy, both odd and even harmonics can be emitted.

From our point of view, the oscillating and spreading of the ionized wave packet near the parent center is similar to the potential scattering in the presence of a strong external laser field when the quiver velocity is greater than the translational one. The spontaneous emission for such a bremsstrahlung regime was analyzed in [27]. The typical spectrum for spontaneous emission that was obtained in [27] is presented in figure 3 and consists of a number

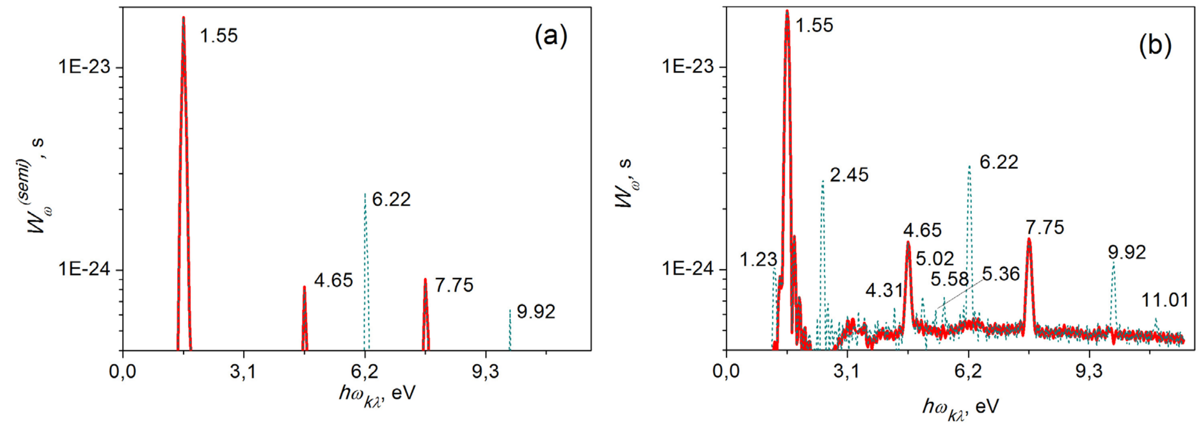

Figure 1. Semiclassical (a) and quantum-mechanical (b) spectra of spontaneous atomic emission for a laser intensity of 10^{13} W cm^{-2}. The solid curve is the emission spectrum by the end of the laser pulse and the dashed curve corresponds to the instant of time equal to the two-pulse duration. The values near the peaks indicate their position in eV.

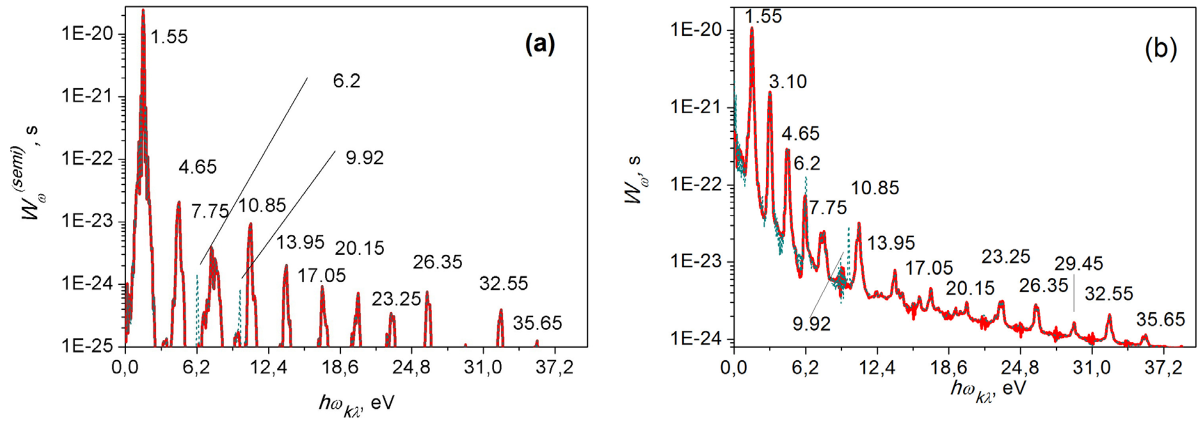

Figure 2. The same as figure 1, but for a laser intensity of 10^{14} W cm^{-2}.

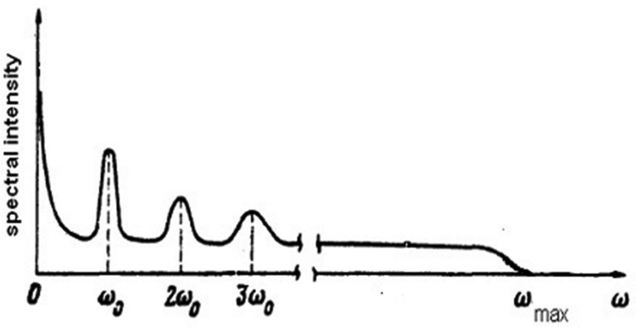

Figure 3. Spectral intensity of spontaneous bremsstrahlung in the field of an intense external wave.

of both odd and even harmonics. It is in a good qualitative agreement with our data for low-order harmonic emission.

For high-order harmonics in the plateau regime, odd harmonics were still predominantly emitted (see figure 2(b)). This part of the harmonic spectra was in qualitative agreement with the semiclassical model. To explain this peculiarity the decomposition (16) of the wave function $\delta\psi_{k\lambda}(\bar{r}, t)$ was analyzed for field modes corresponding to $\omega_{k\lambda} = m\omega$, $(m = 6 \div 24)$ in the intensity range $4 \times 10^{13} \div 10^{14}$ W cm^{-2} when the plateau-like structure of harmonics is formed. It was found that both photorecombination to the ground state and bremsstrahlung contribute to the odd harmonics, although photorecombination provides the greater contribution to the plateau of odd harmonics. According to [27] we can conclude that the bremsstrahlung acts predominantly in the part of the spectra that is located before the plateau (see figure 2(b)) and as a result even harmonics are produced there.

It is also necessary to mention that the existence of the bremsstruhlung continuum was not observed in the semiclassical calculations. It was found that the quantum-mechanical calculation provided the intensity of the harmonics and lines corresponding to transitions $|4\rangle \to |1\rangle$ and $|2\rangle \to |1\rangle$ up to the order of the values of—and above—the semiclassical ones. Both of these peculiarities result from the essential depletion of the ground state during the ionization process, followed by the suppression of the transition to the ground state, studied in the framework of the semiclassical approach. In the semiclassical approach the probability of photorecombination is proportional to the population of the final state [25], and hence it underestimates the efficiency of high-order harmonic generation in the strong-field limit.

It should be also noted that the emission of a single atom was analyzed in this study. To produce the effective process for HHG one needs to analyze the emission from the atomic ensemble and the phase-matching effect. These problems are outside the scope of this paper. Nevertheless, we can suppose that phase matching would suppress the emission arising from transitions between discrete atomic levels and to a lesser degree those coming from bremsstrahlung and photorecombination.

5. Conclusions

We analyzed the spontaneous emission of an atom in the presence of a high-intensity laser field. Our analysis is based on consideration of quantum system interaction, with the quantized field modes being in the vacuum state, while the intense laser field is considered to be classically beyond perturbation theory. It was demonstrated that not only odd, but also even harmonics, as well as lines associated with the transitions between different discrete levels, can be emitted if the laser field is strong enough. The origin of even harmonics was studied. It was determined that they result from the bremsstrahlung, which becomes efficient in the regime of strong ionization. The obtained results were compared with those found in the framework of the semiclassical approach. One can conclude that the semiclassical approach is inapplicable in the strong-field limit.

Acknowledgments

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