Generation of harmonics of a strong field in a three-level atomic model

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We perform numerical calculations of the spectrum of one- and three-dimensional three-level atoms in a strong optical field. The position of the cutoff of the plateau in the harmonic spectrum of a three-level atom is shown to agree well with the exact numerical solution for a one-dimensional atom, as well as with experimental data available for a real xenon atom.

Experimental work of recent years has provided higher and higher optical harmonics under the action of strong optical field on atoms.[1–3] Although this effect is important from an optical standpoint, since it opens the possibility of producing coherent radiation sources in the ultraviolet and higher energy ranges, it is not clear yet what kinds of restrictions exist on limiting orders of the harmonics emitted. Most existing experimental work (in particular, Refs. 1–3) indicates the existence of a so-called plateau in the harmonic spectrum with a sharp intensity cutoff above the maximum harmonic order. However, the mechanism of harmonic cutoff is poorly understood at present. Existing theoretical work on the cutoff of emission in real atoms deals primarily with numerical calculation.[4–6] Although the results of that work agree well with experimental data, spectral analysis is strongly hampered in view of the complexity of the numerical models themselves. At the same time, it was shown in Refs. 7 and 8 that the plateau in the spectrum of harmonics appears even in model two-level systems. However, closer examination shows that the plateau width and the maximum harmonic order disagree in these models both with more accurate calculations and with experimental data. We show in this paper that the correct position of the plateau cutoff can be obtained in a comparatively simple model of a three-level atom in a strong field.

We consider a three-level system having a lower level with energy E_1 describing the atomic ground state and two levels with energies E_2 and E_3 located near the ionization limit, as shown in Fig. 1. Since in experimental work on the observation of the harmonic generation effect, the frequencies external photons were such that atomic ionization could occur only through absorption of a large number of photons (typically about ten), we also consider conditions under which the separation between the ground and the excited levels $\omega_{12}=E_2-E_1$ is greater that the frequency of the external radiation, and the condition $\omega_{21}^2 \gg \omega^2$ is met. The ground state will then experience a Stark shift, as shown in Ref. 9. Furthermore, it is important that the field strengths $\mathscr{E}$ actually attainable at present are considerably lower than atomic field strengths for the ground state, i.e., $\mathscr{E} < 0.1/n_1^4$, where n_1 is the principal quantum number of the ground state. Under these conditions, the Stark shift of the ground state will be small, and the state will be stable with respect to the ionization process.[10–12] As for the pair of upper states, the opposite conditions are usually fulfilled in experiments, with the separation between the levels $\omega_{32}=E_3-E_2$ being less than the external field frequency, $\omega_{32}^2 \ll \omega^2$, and the external field strength exceeding the atomic one, $\mathscr{E} \gg 0.1/n_2^4$. Then the upper states experience a Stark shift, as shown in Refs. 9 and 11. In this situation, we can expect that once the upper states are populated from the ground state under the action of an external field, harmonic radiation will appear due to spontaneous inverse transitions from the upper states to the ground state.

The population dynamics of three-level system states was determined in this work with the help of numerical calculations. The time-dependent wave function was chosen to be of the form

$$\Psi(\mathbf{r},t)=a_1(t)\psi_1(\mathbf{r})+a_2(t)\psi_2(\mathbf{r})+a_3(t)\psi_3(\mathbf{r}),$$

where $a_i(t)$ are the state population amplitudes, and $\psi_i(\mathbf{r})$ are the corresponding eigenfunctions. Obviously, for a one-dimensional atom, all the functions depend on the x-coordinate only, and the direction of the x-axis coincides with the external field direction. To simplify the statement of the problem, we assume that the lower and middle states have the same parity, and the upper state has the opposite parity. Dipole transitions between the lower and middle states are then forbidden by the selection rules.

Let us substitute the complete time-dependent wave function into the Shrödinger equation

$$i\partial\Psi/\partial t=(\hat{H}_0+\mathscr{E}xf(t))\Psi, \qquad (1)$$

where $\hat{H}_0$ is the Hamiltonian of the unperturbed atomic system, $x=r\cos\theta$ in the three-dimensional case, and the function $f(t)$ specifies the temporal dependence of the external field [the choice of the form of the function $f(t)$ will be discussed below]. Performing a standard change of variables $a_i(t)=b_i(t)\exp(-iE_it)$, we obtain a set of ordinary differential equations to determine the coefficients $b_i(t)$:

$$db_1/dt=-i\mathscr{E}x_{13}\exp(-i\omega_{31}t)b_3 f(t),$$

$$db_2/dt=-i\mathscr{E}x_{23}\exp(-i\omega_{32}t)b_3 f(t),$$

$$db_3/dt=-i\mathscr{E}x_{31}\exp(-i\omega_{13}t)b_1 f(t) -i\mathscr{E}x_{32}\exp(-i\omega_{23}t)b_2 f(t), \qquad (2)$$

where $\omega_{ji}=E_j-E_i$, $\omega_{ji}=-\omega_{ij}$, and the matrix elements are $x_{ij}=\langle i|x|j\rangle$ with $x_{ij}=x_{ji}^{+}$, $x_{ii}=x_{12}=0$.

We now discuss the form of the function $f(t)$, which determines the time dependence of the external field strength. If the field amplitude is small enough for this field to be considered weak, field turn-on and turn-off are poorly

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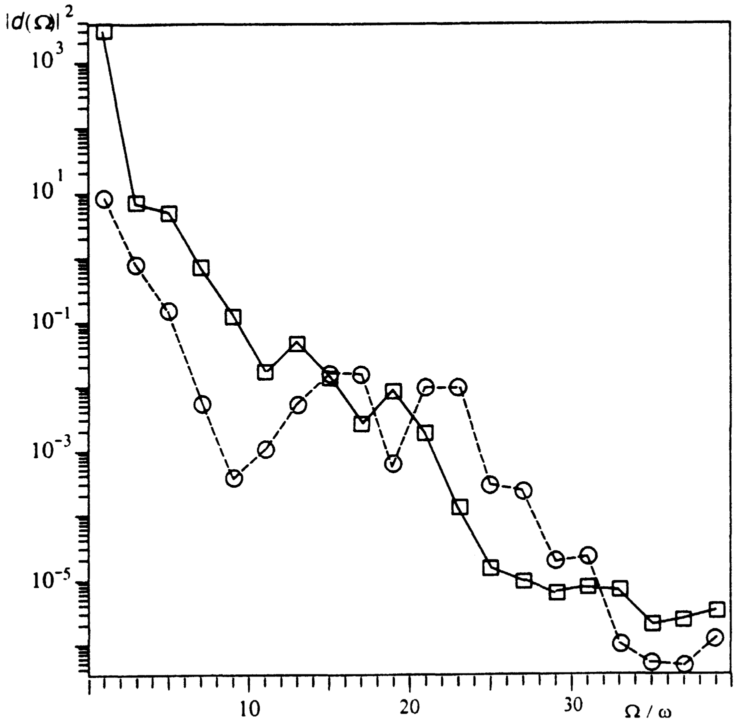

FIG. 1. Intensity $|d(\Omega)|^2$ of one-dimensional atomic harmonics as a function of harmonic number Ω/ω. The squares and the solid curve correspond to numerical calculations of the Schrödinger equation with a model Hamiltonian. The circles and the dashed curve correspond to calculations in the three-level model. The frequency of the external field is $\omega=0.0643$, and the field strength is $\mathscr{E}=0.05$.

defined, and we may set $f(t)=\sin\omega t$, as is usually assumed in calculations using perturbation theory. But when a strong field is acting on the atom, an atomic electron experiences a nonphysical impulse at the instant of field turn-on if the function $f(t)=\sin\omega t$ is used in theoretical calculations.[4,12] This phenomenon does not disappear even if the field is modulated by some envelope $\varphi(t)$ that vanishes at the instants of turn-on and turn-off. For example, we can employ a vector potential in the form $A(t)=-(\mathscr{E}c/\omega)\varphi(t)\sin\omega t$. Then, taking the relation $\mathscr{E}=-\dot{A}/c$ between the field strength and the vector potential into account, we find

$$f(t)=\varphi(t)\cos\omega t+\frac{\dot{\varphi}(t)}{\omega}\sin\omega t. \tag{3}$$

We can show that it is really possible to suppress electron impulses at field turn-on and turn-off if the field temporal dependence is chosen in the form (3). In our calculations, we used a trapezoidal envelope:

$$\varphi(t)=\begin{cases} t, & 0<t<(2\pi/\omega), \\ 1, & (2\pi/\omega)\leqslant t\leqslant 6(2\pi/\omega), \\ 7-(\omega/2\pi)t, & 6(2\pi/\omega)\leqslant t<7(2\pi/\omega), \end{cases} \tag{4}$$

i.e., the field grew linearly for one optical period, had a constant amplitude for five periods, and then decreased linearly for one period.

The differential equations (2) obtained with field temporal dependence in the form of (3)–(4) was solved numerically over a time interval $[0,T=7(2\pi/\omega)]$ with the constraint that only the ground state is initially populated, i.e., $b_1(0)=1$, $b_2(0)=b_3(0)=0$.

The dipole moment of an atomic electron was determined from

$$d(t)=\langle\Psi(\mathbf{r},t)|x|\Psi(\mathbf{r},t)\rangle. \tag{5}$$

It is easy to show that for a three-level system, Eq. (5) reduces to

$$d(t)=\sum_{ij}a_i^*(t)a_j(t)x_{ij}.$$

According to Refs. 4–6, the spectrum $|d(\Omega)|^2$ of radiated harmonics was determined from the Fourier components of the dipole moment as

$$|d(\Omega)|^2=\left|\int_0^T d(t)\exp(i\Omega t)dt\right|^2. \tag{6}$$

To test the appropriateness of the three-level model for analysis of the harmonic spectrum, it is convenient to compare with the results of more accurate calculations. With this in mind, we accurately integrated the Schrödinger equation for the one-dimensional problem with a frequently used[13] atomic Hamiltonian of the form

$$\hat{H}_0=-\frac{1}{2}\frac{\partial^2}{\partial x^2}-\frac{1}{\sqrt{x^2+1}}. \tag{7}$$

The Schrödinger equation (1) with Hamiltonian (7) was solved numerically using the combined spectral-shift method described in Ref. 14. Equations (5) and (6) were used to determine the spectrum of radiated harmonics.

In solving Eqs. (1) and (2), the ground state of Hamiltonian (7) with an energy $E_{1(+)}=-0.670295$ was chosen as the initial state of a one-dimensional atom. In solving Eqs. (2), the states $E_{2(+)}=-0.034676$ and $E_{3(-)}=-0.026942$ were chosen as the two upper states. The signs (+) and (−) denote the parities of the states. The transition matrix elements needed to solve system (2) were determined numerically: $x_{13}=-0.0584$, $x_{23}=17.71$. The amplitude of the external field was $\mathscr{E}=0.05$. The frequency corresponded to a 10-photon transition from the ground state to the upper state, $\omega=0.064335$.

The results of the calculations of the one-dimensional atomic harmonic spectrum are presented in Fig. 1. The harmonic intensity in the accurate solution of the Schrödinger equation is seen to decrease quite smoothly up to the 21st harmonic, after which a sharper drop occurs. In this case, it is conventional to say that the plateau cutoff occurs at the 21st harmonic. Completely analogous behavior is exhibited by the spectrum of a model three-level one-dimensional atom, only the plateau cutoff occurs at the 23rd harmonic. Moreover, it can be seen that the absolute values of the harmonic intensities from the 13th to 23rd harmonic agree well in order of magnitude for both calculations. Thus, in the one-dimensional case, the three-level model nicely describes the position of the plateau cutoff of harmonics as well as the spectral values near the cutoff. At low and medium harmonics, the three-level model underestimates the harmonic intensities, because there are no intermediate states between the ground state and the high excited levels in the three-level model. In the accurate model, these intermediate levels are also excited, and contribute significantly to spontaneous emission of harmonics at medium and low frequencies.

The calculations in the three-level model can also be compared with the existing experimental data. With this in mind, we oriented ourselves in this work to the experiments[5] in which harmonic generation was observed when Nd:YAG laser radiation with a wavelength $\lambda=1064$ nm ($\omega=0.043$) and intensity $I=4\cdot 10^{13}$ W/cm^2 ($\mathscr{E}=0.0336$) acted on xenon atoms. As the authors of the experiments[5] suggest, the laser radiation intensity exceeded the saturation intensity for their conditions of observing harmonics. This implied formation of a large number of singly ionized xenon ions. Therefore, we considered that the medium consists simply of atomic ions and the external field was described without a switching regime, i.e., $f(t)=\sin\omega t$. The ground state of a xenon ion has an energy $E_1(5p)=-0.774$. The states $E_2(9p)=-0.053$ and $E_3(10s)=-0.043$ were chosen as the two upper states. Thus, the transition from the ground state to the upper state comes about by virtue of the absorption of 17 photons, and absorption of the next photon results in the formation of a doubly-charged ion. The transition matrix elements for the indicated levels are $x_{12}=0$, $x_{13}=-0.2i$, $x_{23}=-4i$. The calculation time was $T=600$.

A Comparison of the numerical results with the experimental data[5] for the harmonic spectrum of a three-level xenon ion is presented in Fig. 2. It is seen that the three-level model provides a good description of the position of the harmonic plateau cutoff. A strong dip in the central part of the calculated spectrum is associated with two factors. First, our model does not account for the existence of intermediate discrete energy levels, which contribute to the final spectrum, as in the one-dimensional case. Second, both the xenon atom and ions produced radiation in the experiments, and this was not included in our model.

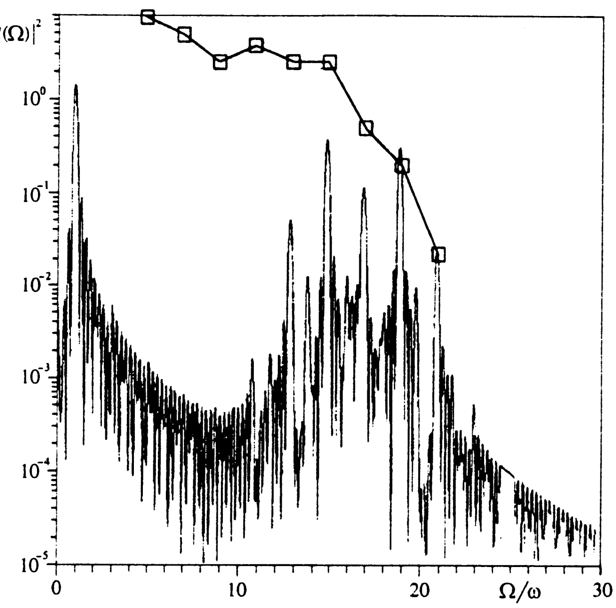

FIG. 2. Intensity $|d(\Omega)|^2$ of xenon ion harmonics as a function of their number Ω/ω. The solid oscillatory curve presents the calculated results in the three-level model. The squares denote the experimental results.[5] The frequency and strength of the external field are respectively $\omega=0.043$ and $\mathscr{E}=0.0336$. The numerical results are normalized to the experimental data at the frequency of 21st-harmonic radiation.

On the whole, the results presented here show that a three-level atomic model provides a good description of the position of the harmonic plateau cutoff for a one-dimensional atomic model as well as for actual experimental observations.

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