

Modifications of High Harmonic Spectra by Ion Resonant Transitions

A. V. Birulin*, V. T. Platonenko*, G. Ferrante**, and E. Fiordilino***

*Faculty of Physics and International Laser Center, Moscow State University, Moscow, 119899 Russia

**Istituto Nazionale di Fisica della Materia and Dipartimento di Energetica ed Applicazioni di Fisica, Viale delle Scienze, Palermo, 90128 Italy

***Istituto Nazionale Fisica della Materia and Istituto di Fisica, via Archirafi 36, Palermo, 90123 Italy

e-mail: plat@femto.phys.msu.su

Received August 5, 1996

Abstract—High-order harmonic generation is considered in a system consisting of an ion with an internal degree of freedom plus an outer electron. The theoretical treatment is both quantum-mechanical and classical. The emphasis is on the core resonance effects, which can significantly modify the harmonic spectra, with appearance of anomalous peaks. Under some assumptions, the spectral amplitude of the resonant harmonic of the system dipole moment can be obtained by evaluation of such amplitude within a single-electron approximation and multiplication of the result by a correcting factor. The latter depends on the polarizability of the ion and of a free electron at the harmonic frequency.

1. INTRODUCTION

Generation of harmonics of high order, occurring in processes of interaction of short-pulse intense laser radiation with atoms and ions in gas jets, and with nondense plasmas, in recent years has become, and presently is, one of the most thoroughly investigated subjects in laser and laser-atomic physics. See [1–8] for useful summaries and discussions of the issues considered below.

Investigations are driven in an essential way by the perspective of creating coherent sources of electromagnetic radiation with increasingly shorter wavelength. Experiments are continuously reported in which harmonics of shorter and shorter wavelengths are observed. At the moment, the shortest wavelength seems to be about 6 nm [8, 9]. However, much effort needs still to be done to remove the practical limitations inherent in the goal of obtaining intense short-wavelength coherent light by the high-order harmonic generation. Among them, we quote, as an instance, the limit to the high-order harmonic conversion put by the ionization of a nonlinear medium exposed to a high-intensity laser field. This item poses the problem of optimizing the choice of the generating nonlinear medium. Experiments indicate that when neutral rare gases are used in nonlinear medium, species with higher ionization potentials should be preferred [10]. Alternatively, ionic species are becoming important, such as the alkali or other elements with a wide variety of atomic numbers and electron configurations [4, 5, 8]. Presently, most of the experimental investigations are concerned with the removal of this limitation.

Another important limitation is the very low conversion efficiency, to which, however, less attention has been devoted. This issue, in particular, was addressed

in [11, 12] where, optimizing the choice of experimental conditions, in a medium of Xenon rare gas the efficiency of conversion of a 1-ps 1053-nm Nd-glass laser radiation at 10^{15} W/cm² intensity into the 17th harmonic (20 eV) has been brought up to 10^{-6} , with an instantaneous power generated of about 30 kW. Theoretically, physical situations that are able, in principle, to yield more effective conversion efficiencies are considered in [13, 14]. In particular, in [13], using a simple phenomenological approach, the case when harmonic emission spectrum of an outer electron was significantly enhanced at selected frequencies, corresponding to resonant transitions of the core electrons, was addressed.

It is the aim of this paper to provide a quantitative treatment (mostly, quantum mechanical) of a physical process similar to that considered in [13], in which resonant transitions of core electrons alter the harmonic spectrum emitted by an outer electron. More precisely, we consider a model atom formed by a core with internal degrees of freedom plus an outer electron. In general, the atomic core states will undergo nonadiabatic changes if the position of the outer electron experiences fast changes. Accordingly, when high-frequency harmonics are generated, in the atomic core should take place effects bound to the nonadiabaticity of the process. In turn, they could significantly affect the harmonics spectrum. For instance, it is natural to expect that some exciting frequencies might provide resonant growth of harmonics with frequencies close to atomic core eigenfrequencies, or, the contrary, cause suppression. Such effects should manifest themselves in the energy distribution of the ionization products and in the spectra of the emitted radiation. To the best of our knowledge, in the context of high-order harmonic generation, such a mechanism and atomic models going

beyond the one-electron approximation have never been considered before, except for the phenomenological approach mentioned above [13]. We note also that some aspects of harmonic generation experiments implying the potential importance and role of ions have been discussed theoretically within one-electron treatments in various papers [15–17].

Below, a quantum-mechanical treatment of harmonic generation by a many-electron model atom experiencing resonant core electron transitions is worked out under the basic assumption that the geometrical dimensions of the atomic core are small as compared to the dimensions of the outer electron wave packet. Besides, we work out a classical model of high-order harmonic generation by a system of an “ion with internal degree of freedom plus an outer electron,” and discuss the results of numerical calculations based on this model. Combining the results of the classical model with those of the quantum-mechanical treatment helps to understand how resonant transitions in the atomic core may alter the harmonic spectrum.

2. QUANTUM-MECHANICAL TREATMENT

We treat the ion (the atomic core) and the outer electron as two subsystems with degrees of freedom \mathbf{x} and \mathbf{r} and Hamiltonians $H_{\mathbf{x}}$ and $H_{\mathbf{r}}$ interacting with each other through the potential $V(\mathbf{x}, \mathbf{r})$. The Hamiltonian of the whole system in the presence of the light wave field $\mathbf{E}_W(t)$ is then written as

$$H = H_{\mathbf{x}} + H_{\mathbf{r}} + V(\mathbf{x}, \mathbf{r}) + e\mathbf{r} \cdot \mathbf{E}_W - \mathbf{d} \cdot \mathbf{E}_W, \quad (1)$$

where \mathbf{d} is the dipole moment of the atomic core. Below, in working out the present treatment, the following assumptions are made:

(1) the probability of ionization of the ion is negligible [it puts a limitation on the field strength $\mathbf{E}_W(t)$]; (2) at the same time, the transition probability of the ion into one of the excited states of the discrete spectrum is finite; and (3) as anticipated, the geometrical dimensions of the ion in such states are small as compared to the dimensions of the outer electron wave packet.

Then, neglecting the particles identity, the state $\Psi(\mathbf{x}, \mathbf{r})$ of the whole system is written as

$$\Psi = \sum_j \exp\left(-i\frac{\epsilon_j}{\hbar}t\right) u_j(\mathbf{x}) \psi_j(\mathbf{r}, t), \quad (2)$$

where ϵ_j and u_j are the ion eigenenergies and eigenstates, i.e., $H_{\mathbf{x}}u_j = \epsilon_j u_j$. Below, we assume that in the absence of the external field, only the state $u_0\psi_0$ contributes to the sum in (2). According to (2), the dipole moment mean value of the whole system is written as the sum $-e\mathbf{r}_e + \mathbf{D}_c$, where

$$\mathbf{r}_e = \sum_j \langle \psi_j | \mathbf{r} | \psi_j \rangle, \quad (3)$$

$$\mathbf{D}_c = \sum_{jk} \exp(-i\omega_{jk}t) \mathbf{d}_{jk} \langle \psi_j | \psi_k \rangle, \quad (4)$$

$$\mathbf{d}_{jk} = \langle u_j | \mathbf{d} | u_k \rangle, \quad (5)$$

$$\omega_{jk} = \frac{\epsilon_j - \epsilon_k}{\hbar}, \quad (6)$$

and for the outer electron wavefunction ψ_j , the following equation is readily obtained:

$$i\hbar\dot{\psi}_j = [H(\mathbf{r}) + V_{jj}(\mathbf{r}) + e\mathbf{r} \cdot \mathbf{E}_W] \psi_j + \exp(i\omega_{jk}t) [V_{jk}(\mathbf{r}) - \mathbf{d}_{jk} \cdot \mathbf{E}_W] \psi_k, \quad (7)$$

where $V_{jk}(\mathbf{r}) = \langle u_j | V(\mathbf{x}, \mathbf{r}) | u_k \rangle$ and summation over the index k in the right-hand side is understood.

Let us assume that the ion transition frequencies ω_{jk} largely exceed the most significant frequencies of the harmonics present in (3). In such a case, any term on the right-hand side of (7) containing $\exp(i\omega_{jk}t)$ is rapidly oscillating and can be omitted as very small. Then (7) becomes a one-electron equation. If, instead, (3) contains significant harmonics with frequencies comparable to ω_{jk} , the function ψ_k has rapidly oscillating components, which, when multiplied by $\exp(i\omega_{jk}t)$, can give nonnegligible contributions to (7). In particular, it will cause the appearance of new not small terms in (4) for the ion dipole moment. Such terms may be estimated on the basis of the following considerations. First, exploiting the well-known expansion

$$\frac{1}{|\mathbf{r} - \mathbf{x}|} = \sum_{n=0}^{\infty} P_n[\cos(\widehat{\mathbf{r}, \mathbf{x}})] \min\left(\frac{x^n}{r^{n+1}}, \frac{r^n}{x^{n+1}}\right), \quad (8)$$

with P_n as the Legendre polynomials, we write

$$\begin{aligned} V(\mathbf{x}, \mathbf{r}) &= -\frac{Ze^2}{r} + \sum_{i=1}^{n_0} \frac{e^2}{|\mathbf{r} - \mathbf{x}_i|} \\ &= -\frac{z_c e^2}{r} - \frac{e\mathbf{d} \cdot \mathbf{r}}{r^3} + \sum_i \delta V_i, \end{aligned} \quad (9)$$

with Ze as the nuclear charge, $\mathbf{d} = -e\sum_i \mathbf{x}_i$ being the sum extended over all the core electrons, $z_c = Z - n_0$, and

$$\begin{aligned} \delta V_i &= e^2 \min\left(0, \frac{1}{x_i} - \frac{1}{r}\right) + e^2 (\mathbf{x}_i \cdot \mathbf{r}) \min\left(0, \frac{1}{x_i^3} - \frac{1}{r^3}\right) \\ &+ e^2 \sum_{n=2}^{\infty} P_n[\cos(\widehat{\mathbf{x}_i, \mathbf{r}})] \min\left(\frac{x_i^n}{r^{n+1}}, \frac{r^n}{x_i^{n+1}}\right). \end{aligned} \quad (10)$$

Exploiting now the assumption of small geometrical dimensions of the core ion as compared to the electron wave packet, δV_i is neglected and $V(\mathbf{x}, \mathbf{r})$ is written as

$$V(\mathbf{x}, \mathbf{r}) \approx V(r) - \mathbf{d} \cdot \mathbf{E}_e(\mathbf{r}), \quad (11)$$

where

$$V(r) = -\frac{z_c e^2}{r} \quad (12)$$

and

$$\mathbf{E}_e(\mathbf{r}) = \frac{e\mathbf{r}}{r^3} \quad (13)$$

is the field created by the outer electron at the center of the ion (center of coordinates).

From (11) it follows that the diagonal matrix element $V_{jj}(\mathbf{r})$ does not depend (within the employed approximation) on j , and that

$$V_{jk} = -\mathbf{d} \cdot \mathbf{E}_e(\mathbf{r}) \quad (14)$$

[expression (11), which is exact for a pointlike nucleus, may be viewed also as the result of the electron being on the average at a distance from the ion much larger than the ion dimensions]. We assume, next, that the ion remains in its ground state u_0 with a probability close to unity; accordingly, for $j \neq 0$, we have $\langle \psi_j | \psi_j \rangle \ll \langle \psi_0 | \psi_0 \rangle = 1$, and we can write

$$\psi_j = a_j(t)\psi_0 + \phi_j \quad (15)$$

with the term ϕ_j orthogonal to ψ_0 . Substitution of (15) into (3) and (4) causes the appearance of small terms proportional to $\langle \phi_j | \phi_k \rangle$ which in what follows will be neglected. As for a_j , from (1), (2), and (7)–(15) we have

$$i\hbar \dot{a}_j = -e^{(i\omega_{j0}t)} \mathbf{d}_{j0} \cdot \mathbf{E}, \quad (16)$$

with $\mathbf{E} = \mathbf{E}_W + \mathbf{E}_e(t)$ and

$$\mathbf{E}_e(t) = \langle \psi_0 | \mathbf{E}_e(\mathbf{r}) | \psi_0 \rangle, \quad (17)$$

and small terms containing $\langle \psi_0 | \mathbf{E}_e(\mathbf{r}) | \psi_k \rangle$ in the right-hand side of (16) neglected. Integrating (16) and using (15) and (4) for the harmonic amplitude, we obtain

$$\mathbf{D}_c(\Omega) = \alpha_{\text{ion}}(\Omega) \mathbf{E}_e(\Omega), \quad (18)$$

where

$$\alpha_{\text{ion}} = \sum_j d_{j0}^2 \frac{2\omega_{j0}}{(\omega_{j0}^2 - \Omega^2)} \quad (19)$$

and $\mathbf{E}_e(\Omega)$ is the spectral amplitude of the field $\mathbf{E}_e(t)$ (17). To be valid, expression (18) requires that the Stark shift of any level significantly contributing to it be small as compared to the value $\hbar|\omega_{k0} - \Omega|$:

$$\frac{1}{4} \alpha_k E(\omega)^2 < \hbar|\omega_{k0} - \Omega|. \quad (20)$$

In (20), $E(\omega) = E_e(\omega) + E_W(\omega)$ is the field amplitude at the fundamental frequency ω , and the polarizability α_k can be obtained from α_{ion} (19) substituting the index k for 0.

Somewhat more complicated is to obtain a relation between $\mathbf{E}_e(\Omega)$ and $\mathbf{d}_e(\Omega) = -e\mathbf{r}_e(\Omega)$. For the purpose, we use the Ehrenfest theorem for the average values of coordinates and moments. Using the Hamiltonian (1) and the assumptions formulated above, for the average value $\mathbf{r}_e = \langle \Psi | \mathbf{r} | \Psi \rangle$ the following equation is obtained:

$$m\dot{\mathbf{r}}_e = -e(\mathbf{E}_W + z_c \mathbf{E}_e + \mathbf{E}_d) \quad (21)$$

with

$$\mathbf{E}_d = \left\langle \psi_0 \left| \frac{3(\mathbf{r} \cdot \mathbf{d})\mathbf{r} - r^2 \mathbf{d}}{r^5} \right| \psi_0 \right\rangle. \quad (22)$$

From (21) one easily obtains

$$\mathbf{d}_e(\Omega) = z_c \alpha_e(\Omega) \mathbf{E}_e(\Omega) + \alpha_e(\Omega) \mathbf{E}_d(\Omega). \quad (23)$$

The first term in this expression can be treated as the moment evaluated in the single-electron approximation [as a matter of fact, under the condition $d_{jk} = 0$, (7) coincides with the equation of the single-electron theory]. We now write the whole moment of the system “ion plus electron” as the sum of the first term of (23) and of (18), it amounting to reproduce the dipole moment phenomenologically postulated in [13], namely

$$-e\mathbf{r}_e(\Omega) + \mathbf{D}_c(\Omega) = -e\mathbf{r}_e(\Omega) \left[1 + \frac{\alpha_{\text{ion}}(\Omega)}{z_c \alpha_e(\Omega)} \right]. \quad (24)$$

Thus, the present derivation constitutes a justification of (24), where now the basic assumptions and limitations are clearly and explicitly formulated. $\alpha_e = -e^2/(m\Omega^2)$ is the free electron polarizability.

In obtaining (24) we have, in practice, neglected the field \mathbf{E}_d . The role of this field is certainly decreasing with the increasing of the electron wave packet dimensions; but it is difficult to investigate this role quantitatively. Besides, the neglected term of (23) represents the influence of the ion dipole moment on the electron trajectory. Wishing to have an estimate defining the domain of validity of our approximation, the shortest way is to resort to a classical model (we note that we do not know ψ_0 defining \mathbf{E}_d). To build out a classical model, we need only to formulate some classical procedure for the calculation of the fields \mathbf{E}_e and \mathbf{E}_d instead of (17) and (22). It is done in the next section.

3. A CLASSICAL MODEL

The model is based on the following assumptions.

(1) The ion (the atomic core) is a fixed pointlike object placed in the center of coordinates and possessing constant charge Z and dipole moment \mathbf{D}_c determined by the equations

$$\mathbf{D}_c = \sum_j \mathbf{D}_j, \quad (25)$$

$$\ddot{\mathbf{D}}_j = -\omega_j^2 \mathbf{D}_j + \frac{e^2 f_j}{m} (\mathbf{E}_W + \mathbf{E}_e), \quad (26)$$

where ω_j and f_j are, respectively, a resonant frequency and the related oscillator strength; \mathbf{E}_e is the field created by the electron; and \mathbf{E}_W , as before, is the light wave field. The system of (25) and (26) yields the same dipole moment, (18), as the system of (4), (15), and (16). The condition $|a_j|^2 \ll 1$, implicit in (15), is equivalent to the condition

$$D_j^2 \ll \frac{\hbar e^2}{2m\omega_j} f_j, \quad (27)$$

which needs to be checked by calculations.

(2) The charge of the outer electron is distributed in space, and this distribution adiabatically changes with the motion of the electron center of mass. Accordingly, the field \mathbf{E}_e created by the electron in the center of coordinates is a function of the vector radius \mathbf{r}_e of the electron center of mass only. Under such assumptions, one can easily prove that the vector radius \mathbf{r}_e changes in accordance with (21), where now

$$\mathbf{E}_d = -\frac{1}{e}(\mathbf{d} \cdot \nabla)\mathbf{E}_e \quad (28)$$

and the dependence of \mathbf{E}_e on \mathbf{r}_e needs to be directly or indirectly postulated. It can be introduced as the mean value $\mathbf{E}_e(t)$ (17), assuming that $|\psi_0|^2 = \rho(\mathbf{r}_e, \mathbf{r} - \mathbf{r}_e)$ and that the density ρ is given in such a way to allow analytical integration in (17). It is done taking ρ as

$$\rho(\mathbf{r}_e, \mathbf{R}) = \frac{3r_0^2}{4\pi(r_0^2 + R^2)^{5/2}}, \quad (29)$$

where $\mathbf{R} = \mathbf{r} - \mathbf{r}_e$. Then

$$\mathbf{E}_e = \frac{e\mathbf{r}_e}{(r_0^2 + r_e^2)^{3/2}}. \quad (30)$$

We note that r_0 is the parameter determining in the present model the influence of the ion dipole moment on the electron trajectory. As it is an unknown parameter, in essence, we are still faced with the problem discussed at the end of the previous section. However, through (28) and (30) we have now a direct way of testing the influence of the dipole moment. The parameter r_0 has the meaning of a radius, and it may be considered representative of the electron wave packet spatial extent. Further, we set $r_0 = \text{const}$, the core charge $z_c = 1$, and the light linearly polarized, $\mathbf{E}_W = \mathbf{E}_a \sin \omega t$. This last choice makes equation (21) one-dimensional.

To avoid difficulties inevitably bound to widely varied parameters, we assume that the field was somehow switched on long ago, $\mathbf{E}_a = \text{const}$, and that the electron motion is established. To calculate the harmonics, we use the spatial representation

$$\mathbf{r}_e = \sum_k \mathbf{r}_e(k\omega) \sin k\omega t, \quad (31)$$

$$\mathbf{D}_c = \sum_k \mathbf{D}_c(k\omega) \sin k\omega t. \quad (32)$$

These equations, together with (21), (23), (28), and (30) and the equations

$$\mathbf{E}_W(k\omega) = \delta_{k1}\mathbf{E}_a, \quad (33)$$

$$\mathbf{E}_{e,d}(k, \omega) = \frac{1}{\pi} \int_0^{2\pi} \mathbf{E}_{e,d} \sin(k\omega t) d(\omega t), \quad (34)$$

form a closed system. Any solution of this system contains odd harmonics. Below only cases are considered

when the solution is made single-valued. As a matter of fact, parasitic effects may arise in the classical calculation. When the frequency ω is smaller than

$$\omega_{\text{cr}} = \left(\frac{e^2}{mr_0^3} \right)^{1/2},$$

the amplitude of the settled electron oscillations becomes a many-valued function of the amplitude \mathbf{E}_a and exhibits a kind of hysteresis loop between $\mathbf{E}_a = -\mathbf{E}_{\text{cr}}(\omega)$ and $\mathbf{E}_a = \mathbf{E}_{\text{cr}}(\omega)$, where \mathbf{E}_{cr} is some critical, frequency-dependent value of the field amplitude. If \mathbf{E}_a is slowly growing, when the critical value \mathbf{E}_{cr} is reached, the trajectory $\mathbf{r}_e(t)$ goes to infinity, the direction being determined by \mathbf{E}_a and changing by small variations of the latter. In the calculations reported below, the condition $\mathbf{E}_a > \mathbf{E}_{\text{cr}}$ is always fulfilled.

4. CALCULATIONS AND COMMENTS

Preliminarily, we have checked the importance of the parameter r_0 in the determination of the spectral amplitude of the dipole moment of the outer electron. In particular, we carried out some calculations assuming a single resonance, varying the radius r_0 and taking the oscillator strength $f = 0$ and 0.5. Besides, we varied the light amplitude and frequency and the ion resonant frequency $\omega_0 = k_0\omega + \delta$, where k_0 is some odd harmonic number and $-\omega < \delta < \omega$.

The results showed that for any reasonable set of the above parameters, the amplitude of the resonant harmonic $r_e(k_0\omega)$ remains almost unaffected by the ion dipole moment. It means that (24) holds for the resonant harmonic as well, with the amplitude $\mathbf{r}_e(k_0\omega)$ evaluated within the single-electron model.

Though this result is obtained by numerical calculations, it can be easily explained analytically. As a matter of fact, in the spectrum of $\mathbf{D}_c(k\omega)$, as a rule, only the resonant harmonic $\mathbf{D}_c(k_0\omega)$ is not small (i.e., only $\mathbf{D}_c(k_0\omega)$ can perturb the electron trajectory). In such a case, the amplitude $\mathbf{E}_d(k_0\omega)$ turns out to be inevitably small and does not perturb the amplitude $\mathbf{r}_e(k\omega)$. [We note that, if use is made of the quantum-mechanical equation (21), it is not easy to find out this effect, which, instead, is obvious from (26), keeping in mind that, in the spectrum of the field \mathbf{E}_e , the amplitude of the fundamental frequency strongly exceeds any other one].

As for the other amplitudes $\mathbf{r}_e(k\omega)$, the modifications are always small if $r_0 > 3r_B$ with r_B the Bohr radius. For $r_0 < (2-2.5)r_B$, the following features are observed:

(1) if $k \ll k_0$, the perturbations of the amplitudes $r_e(k\omega)$ are small (see the initial parts of the spectra reported in Fig. 1);

(2) if $\omega_0 < k_0\omega$ (the ion resonance is approached from above), the spectrum modifications are significant only when $k > k_0$, where, however, the amplitudes

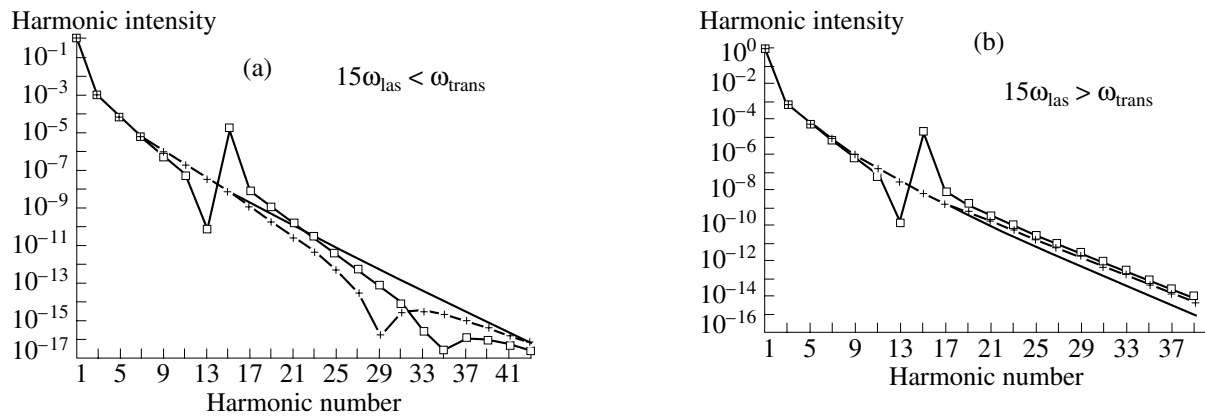


Fig. 1. Relative harmonic intensity vs. the harmonic number: (continuous curve) calculation of the dipole moment of the outer electron with oscillator strength $f_0 = 0$, (dashed curve with crosses) the same as the continuous one with $f_0 = 0.3$, (continuous curve with squares) calculation of the dipole moment of the whole system “ion plus outer electron” with $f_0 = 0.3$. Other parameters are $r_0 = 2r_B$ with r_B the Bohr radius, $\omega = 0.4\omega_e$ with $\omega_e = [e^2/(mr_0^3)]^{1/2}$, $E = 1.9er_0^2$, the resonant frequency is (a) $\omega_0 = 15.05\omega$ and (b) $\omega_0 = 14.95\omega$. The curves are meant to help visualization of the discrete points.

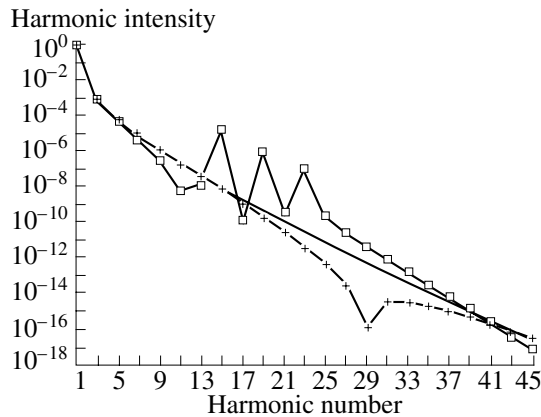


Fig. 2. Relative harmonic intensity vs. the harmonic number (continuous curve) calculation of the dipole moment of the outer electron with oscillator strengths $f_j = 0$ ($j = 0, 1, 2$); (dashed curve with crosses) the same as the continuous one with $f_0 = 0.3$, $f_1 = 0.5$, $f_2 = 0.5$; (continuous curve with squares) calculation of the dipole moment of the whole system “ion plus outer electron” with $f_0 = 0.3$, $f_1 = 0.5$, $f_2 = 0.5$. Other parameters are the same as in Fig. 1. Resonant frequencies are $\omega_0 = 15.05\omega$, $\omega_1 = 19.10\omega$, $\omega_2 = 23.10\omega$.

$r_e(k\omega)$ become significantly small (see the final part of the spectra of Fig. 1b);

(3) if $\omega_0 > k_0\omega$ (the resonance is approached from below), strong modifications turn out to be possible in the region of $k > k_0$ (Fig. 1a). The reason is that in such a case, the amplitudes $\mathbf{E}_e(k\omega)$ and $\mathbf{E}_d(k\omega)$ are close to each other as to the order of magnitude, but differ by sign, and may compensate for each other. Accordingly, small variations of some parameters can provide large changes in the spectrum $-\mathbf{r}_e(k\omega)$ and in the total spectrum where $k > k_0$ (final parts of the spectra of Fig. 1a). The number of harmonics $-\mathbf{r}_e(k\omega)$ strongly modified by

the resonance $\omega_0 \cong k_0\omega$ decreases with the increase of the number k_0 . Increasing k_0 in the region $k > k_0$ increases simultaneously the ratio

$$\frac{|D_c(k\omega)|}{|er_e(k\omega)|}. \quad (35)$$

Of course, the most striking changes in the total spectrum $-\mathbf{r}_e(k\omega) + \mathbf{D}_c(k\omega)$ occur at the resonance value $k \approx k_0$, whatever the sign of the difference $\omega_0 - k_0\omega$ (see Fig. 1). As a rule, at the frequency $(k_0 - 2)\omega$ one observes a sudden decrease of about four orders of magnitude with respect to the single outer electron spectrum. Here the amplitudes $-\mathbf{r}_e(k\omega)$ and $\mathbf{D}_c(k\omega)$ differ by sign and may compensate each other. Note also that, for a given value of r_0 , the variation of the field amplitude does not change the ratio (35), though it can cause dramatic changes in the total spectrum. In going from $k_0 - 2$ to k_0 , the spectrum undergoes a jump of about six or seven orders of magnitude, featuring an isolated peak.

Figure 2 reports the results of the generalization of the above calculations, when several resonant frequencies ω_j come simultaneously into play. The structure of the spectrum becomes rather complicated. Specific values of the resonant frequencies and of the oscillator strengths may yield a spectrum containing a plateau and cut-off. In the calculations of (Fig. 2), the three resonances are approached from below.

5. CONCLUDING REMARKS

We have worked out a theoretical model of high-order harmonic generation by the outer electron of an atomic system, taking into account the possibility that resonant transitions occurring in the atomic core modify the emitted spectrum. The reported calculations show that significant modifications may indeed take

place, the most peculiar signature being the appearance in the harmonic spectrum of anomalous peaks.

We note that, independently of their specific physical origin, similar anomalous peaks in spectra generated in plasma have been observed also experimentally [3, 4] and that inner-shell excitation mechanisms have also been invoked to account for some harmonic spectra [1].

(We would like to note that the frequency of the 7th harmonic of the KrF laser [4] is rather close to the frequency of the transition $3p-4s$ in K^{2+} and $2s-3p$ in C^{2+} [4].)

Two approximations, among others, made above are likely to restrict the domain of validity of our treatment. Mathematically, they are expressed by (11) and (20). The physical content of (11) is that the core must have very small spatial dimensions and/or the outer electron wave packet dimensions must be much larger than the core ones. Besides, the outer electron wave packet must spend most of its time far away from the core. We note that our model is expected to work well if the core is represented by a bare nucleus in which the internal degrees of freedom are accounted for. Naturally, as dipole moments of intranuclear transitions are small, they will hardly significantly affect high-order harmonic generation. Rather, in such a case the model can be used to estimate the probabilities of intranuclear transitions induced by fields under high-order harmonic generation or under multiphoton ionization. If, instead, the core contains electrons, then the applicability of our model depends essentially on the core dimensions, and on the smallness of such dimensions in comparison with other problem characteristic dimensions. The conditions of validity are improved if, during the interaction, the outer electron wave packet increases considerably. Such increase has been shown by means of a computer simulation to occur if the electron oscillates with a large amplitude [18].

We note, finally, that among the outcomes of our treatment there is the possibility of population inversion between some levels of the atomic core. The physical contents of (20) is that the Stark shift of any core level ϵ_k significantly contributing to the ion dipole moment must be small as compared to $\hbar|\omega k_0 - \Omega|$. Let us estimate the left-hand side of the inequality (20) for the level $((1s)^1(2p)^1)$ of the Li^+ ion assuming that $E_e(\omega) + E_W(\omega) \approx E_e(\omega)$, and that the field $E_e(t)$ coincides with (30) where $r_e \approx r_a \sin(\omega t)$. Taking $r_0 = 2.5r_B$ (approximately $e^2/(2I)$, with I the lithium atom ionization potential), $r_a = 2r_0$, and $\hbar\omega = 2.5-4.5$ eV, the value of the left-hand side of (20) is between 0.3 and 0.5 eV. It decreases increasing r_0 and r_a , and increases if the frequency ω approaches a strong resonance (up to 1.3-6 eV). Clearly, in the case of large atomic cores with several electrons one has many resonances and large oscillator strengths, so that the inequality (20) may well be violated. Altogether, our treatment is expected to work satisfactorily for relatively simple and compact atomic systems.

In conclusion we would like to point out the importance of addressing many-electron effects in the harmonic spectra, in particular electron core resonant transitions. Such effects are likely to be important and of growing interest for small and large atomic core as well.

ACKNOWLEDGMENTS

This research has been carried out in the framework of the General Agreement on Scientific Cooperation between Lomonosov Moscow State University (Russia) and the University of Palermo (Italy). Additional indirect support has been provided by the Russian Foundation for Basic Research, the University of Palermo Computation Center, The Italian Ministry of University and Scientific Researches, The National Group of Structure of Matter of the Italian National Research Council, and the Sicilian Regional Committee for Nuclear and Structure of the Matter Researches.

REFERENCES

1. McPherson, A., Gibson, G., Jara, H., *et al.*, 1987, *J. Opt. Soc. Am. B*, **4**, 595.
2. Ferray, M., L'Huillier, A., Li, X.F., *et al.*, 1988, *J. Phys. B*, **21**, L31.
3. Sarukura, N., Hata, K., Adachi, T., *et al.*, 1991, *Phys. Rev. A*, **43**, 1669.
4. Kubodera, S., Nagata, Y., Akiyama, Y., *et al.*, 1993, *Phys. Rev. A*, **48**, 4576.
5. Akiyama, Y., Midorikawa, K., Matsunawa, Y., *et al.*, 1992, *Phys. Rev. Lett.*, **69**, 2176.
6. Balcou, Ph., Cornaggia, C., Gomes, A.S.L., *et al.*, 1992, *J. Phys. B: Atom. Mol. Opt. Phys.*, **25**, 4467.
7. Lewenstein, M., Balcou, Ph., Ivanov, M.Yu., *et al.*, 1994, *Phys. Rev. A*, **49**, 2117.
8. Preston, S.G., Sampera, A., Zepf, M., *et al.*, 1996, *Phys. Rev. A*, **53**, R31.
9. Zhou, J., Peatross, J., Murnane, M.M., and Capteyn, H.C., 1996, *Phys. Rev. Lett.*, **76**, 752.
10. Lompré, L.A., L'Huillier, A., and Mainfray, G., 1991, *Proceedings on Short-Wavelength Coherent Radiation*, Bucksbaum, P.H. and Caglio, N.M., Eds. (Washington, DC: Opt. Soc. Am.), vol. 2, p. 2.
11. L'Huillier, A., Balcou, Ph., and Lompé, L.A., 1992, *Phys. Rev. Lett.*, **68**, 166.
12. L'Huillier, A. and Balcou, Ph., 1993, *Phys. Rev. Lett.*, **70**, 774.
13. Ferrante, G., Oleinikov, P.A., and Platonenko, V.T., 1994, *Pis'ma Zh. Eksp. Teor. Fiz.*, **60**, 235 [1994, *JETP Lett.*, **60**, 246].
14. Birulin, A.V., Platonenko, V.T., and Strelkov, V.V., 1996, *Zh. Eksp. Teor. Fiz.*, **110**, 63.
15. Perry, M.D., Szoke, A and Kulander, K.C., 1989, *Phys. Rev. Lett.*, **63**, 1058.
16. Xu, H., Tang, X., and Lambropoulos, P., 1992, *Phys. Rev. A*, **46**, R2225.
17. Krause, J.L., Schafer, K.J., and Kulander, K.C., 1992, *Phys. Rev. Lett.*, **68**, 3535.
18. De Vries, P.L., 1990, *J. Opt. Soc. Am. B*, **7**, 517.