# Generation of harmonic radiation by hydrogen atoms in intense laser fields

Marko Horbatsch and John K. Liakos
*Physics Department, York University, Toronto, Ontario, Canada M3J 1P3*
(Received 17 December 1990; revised manuscript received 10 May 1991)

A variational principle for the time-dependent Schrödinger equation with an anisotropic Gaussian wave packet as a trial wave function is used to calculate the nonperturbative response of a hydrogen atom to a strong linearly polarized laser field. This method leads to equations of motion for the position expectation value and allows one to study a classical ($\hbar=0$) limit as well as the first quantum corrections. These corrections are shown to be responsible for a strong enhancement in the generation of the third and fifth harmonic by the bound-state part of the electronic wave function for field intensities of the order of $10^{14}$ W/cm$^2$ and a laser frequency corresponding to three-photon ionization. For a low-frequency field a strong nonperturbative enhancement of harmonics of orders 9 to 15 is observed. It is concluded that the plateau at even higher harmonics observed recently in experiment and in numerical calculations is caused by the ionizing parts of the wave function that are not represented in the present calculation.

PACS number(s): 42.65.Ky, 32.80.Rm

## I. INTRODUCTION

Multiphoton ionization and the study of the generation of harmonic radiation in intense laser fields has become an active area of both experimental and theoretical research [1]. Many theoretical approaches are based on numerical solutions of the time-dependent Schrödinger equation (TDSE) in coordinate space, typically using a Crank-Nicholson scheme [2,3]. A complete treatment of the ionization problem including the projection of the time-evolved wave function onto discretized continuum states has been performed within a one-dimensional model [4]. It has been shown that the numerical approach is capable of describing the generation of high harmonics [5], above-threshold-ionization spectra [6,7], as well as ionization rates as a function of laser intensity.

The numerical solution of the TDSE requires large amounts of computer time. Many-electron targets, such as, e.g., Xe atoms, can be treated in an independent-particle model such as the TD Hartree-Fock approximation. Due to the large computational effort involved in solving the one-electron problem, however, calculations were restricted to the frozen-core approximation in which the inner orbitals are not responding to the external field [5].

We have performed nonperturbative calculations, which do not rely on a discretization of coordinate space. The method is based on the TD variational principle (VP), which has been used in the past to study atomic responses to strong TD external fields [8,9]. Using the TDVP with an isotropic Gaussian wave packet (GWP) as a trial function, we studied the ionization of hydrogen atoms in very intense laser fields [10]. This study was recently extended by allowing the GWP to become anisotropic during the interaction with the laser field. This additional freedom in the GWP is particularly important at intensities of $10^{14}$–$10^{16}$ W/cm$^2$. Ionization rates and electron emission spectra are presented elsewhere [11]. This paper focuses on the response of the GWP to the external field at intensities for which the GWP does not ionize, but for which the generation of harmonic radiation can be calculated from the oscillatory motion of the packet.

Two different ideas about the generation of harmonic radiation in a gas that is exposed to intense laser irradiation have been developed. We follow those studies, in which the response of an individual atom to the external field is investigated [12,13]. An alternative classical model has been proposed for the limit of high intensities and low frequencies of the laser field [14]. There it is shown that harmonic generation can be produced by free electrons released as bursts during tunneling at the time of the peak in the external field. This latter model ignores the bound-state contributions, which are the subject of this study. Common to both approaches is the source of the release of harmonic radiation: the strong oscillations in the position expectation value imply accelerations that lead to the emission of electromagnetic radiation.

The purpose of this paper is to clarify two issues. The calculation of the response of a purely bound wave function to the external dipole field demonstrates up to which order one can expect to generate radiation from this part of the wave function alone. This issue is obscured in the numerical approach to the TDSE. The second purpose is to study systematically the importance of quantum corrections to the $\hbar=0$ limit for this problem.

## II. THEORY

Starting with the TDSE for the hydrogen atom in a linearly polarized external field in dipole approximation, we have (atomic units $\hbar=m_e=e=1$ are used throughout)

$$i\frac{\partial}{\partial t}\psi(\mathbf{r},t)=\left[-\tfrac{1}{2}\nabla^2-\frac{Z}{r}-e_0(t)x\sin(\omega t)\right]\psi(\mathbf{r},t)\ . \quad (1)$$

 

In the TDVP we replace this equation by the equivalent action principle, which for a trial wave function normalized at all times reads

$$S=\int_{t_1}^{t_2}\left\langle\psi(\mathbf{r},t)\left|i\frac{\partial}{\partial t}-H(t)\right|\psi(\mathbf{r},t)\right\rangle dt \ . \tag{2}$$

The anisotropic GWP to be used in the TDVP (2) contains twelve real parameters:

$$\psi(\mathbf{r},t)=\frac{\exp\left[-\sum_{j=1}^{3}\left[A_j(r_j-q_j)^2+ip_jr_j\right]\right]}{[(2\pi)^{3/2}w_1w_2w_3]^{(1/2)}} \ , \tag{3}$$

where the parameters $A_j=(1-2iu_jw_j)/4w_j^2)$ are complex valued.

The real-valued $w_j$ are the width parameters associated with the three Cartesian coordinates ($r_1=x, r_2=y, r_3=z$) and the $u_j$ are the components of the corresponding expansion momentum. They measure the rate of delocation of the GWP along the directions of the $x$, $y$, and $z$ axes. The $q_j$ and $p_j$ are the components of the position and translational momentum vectors for the center of the packet, respectively.

The interaction between the electron and the laser field is given in dipole approximation for the linearly polarized case by

$$V_{\rm int}(t)=e_0(t)x\sin(\omega t) \ ,$$

$$e_0(t)=\begin{cases}E_0\sin^2(\pi t/2t_0) & \text{if } t<t_0\\ E_0 & \text{if } t\geq t_0 \ .\end{cases}$$

$e_0(t)$ contains the peak amplitude of the dipole field $E_0$ as well as a form for the envelope of the laser pulse. We use ten cycles to bring the laser field to full strength.

Using the trial wave function defined in Eq. (3) one can calculate the Lagrange function appearing in the action integral (2), perform the variations, and arrive at the following equations of motion for the variational parameters ($j=x$, $y$, or $z$) [11]:

$$\begin{aligned}\dot{w}_j&=u_j \ ,\\ \dot{u}_j&=\frac{\hbar^2}{4w_j^3}+\frac{\partial}{\partial w_j}Q(\mathbf{q},\mathbf{w}) \ ,\\ \dot{q}_j&=p_j \ ,\\ \dot{p}_j&=\frac{\partial}{\partial q_j}Q(\mathbf{q},\mathbf{w})-\frac{\partial V_{\rm int}}{\partial q_j} \ .\end{aligned} \tag{5}$$

Here the $\hbar$ dependence has been displayed explicitly in order to indicate how the classical ($\hbar=0$) limit is obtained in this calculation. The $Q$ function measures the potential energy of the trial GWP in the Coulomb field of the nucleus and is given by

$$Q(\mathbf{q},\mathbf{w})=\frac{Z}{\sqrt{2\pi}}\int_0^\infty ds\frac{\exp\left[-\frac{1}{2}\left[\frac{q_x^2}{w_x^2+s}+\frac{q_y^2}{w_y^2+s}+\frac{q_z^2}{w_z^2+s}\right]\right]}{[(w_x^2+s)(w_y^2+s)(w_z^2+s)]^{(1/2)}} \ , \tag{6}$$

where $Z$ is the atomic number. The partial derivatives of this integral can be expressed analytically [11] and were calculated at each time step by means of a Romberg integration [15]. The system of ordinary differential equations (5) was solved by either an extrapolation or the Runge-Kutta method with variable time step [15]. The initial conditions chosen at $t=0$ correspond to all momentum parameters set to zero, i.e., $p_j=u_j=0$, the center of the packet located at the nucleus ($q_j=0$) and the three width parameters are chosen such that the initial isotropic GWP minimizes the energy ($w_0=3\sqrt{2}\pi/8\approx 0.94$ a.u.). This yields a ground-state energy of $E_{1s}\approx -0.42$ a.u.

At $t=0$ the GWP is isotropic ($w_x=w_y=w_z=w$) and the effective interaction potential (6) can be expressed for this case in terms of the error function [10]

$$Q(q,w)=-\frac{Z}{q}\,\mathrm{erf}\left[\frac{q}{\sqrt{2}w}\right] \ . \tag{7}$$

For a linearly polarized dipole field oriented along the $x$ direction the two components $q_y$ and $q_z$ will remain zero at all times. Thus it is possible to compare the effective potential appearing in the equation of motion for $q_x$ with, e.g, the classical potential used in a one-dimensional model atom studied extensively in the numerical approach [4,6,7]:

$$V_{\rm at}(x)=-\frac{Z}{\sqrt{x^2+1}} \ . \tag{8}$$

Figure 1 provides such a comparison and also indicates how the effective Coulomb interaction is modified by a change in the (single) width parameter. Changes in the width parameters $w_x$ and $w_y=w_z$ will occur due to polarization of the atom in the laser field. It can be seen that an increase in the width parameter softens the interaction in the vicinity of the Coulomb center, but leaves the long-range behavior of the Coulomb interaction intact. It is also evident that the short-range behavior of the model potential (8) overestimates Coulomb binding, while our GWP approach slightly underestimates it, which results in a too strongly and too weakly bound ground state in each model, respectively.

One ought to be careful in comparing potentials (7) and (8). $Q(q,w)$ is an effective potential for the motion of the position expectation value. Therefore, it is not a classical potential, but contains contributions from averaging over

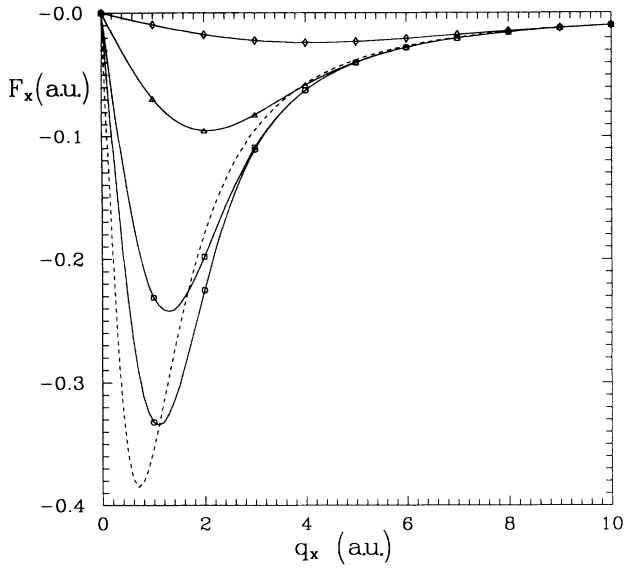


FIG. 1. $x$ component of the effective atomic binding force for the $q_x$ motion for fixed choices of the width parameter $w_0=w_x=w_y=w_z$ in a.u. ○, $w_0=0.8$; □, $w_0=0.94$; △, $w_0=1.5$, ◇, $w_0=3$. Dashed line, one-dimensional model of Javanainen *et al.* [4–7], Eq. (8).

the GWP.

The generation of harmonic radiation in nonperturbative situations can be calculated from the time evolution of the position expectation value, which is related to the dipole matrix element. A detailed discussion is given in Ref. [12]. The position expectation value of our trial wave function is simply given by $\langle \mathbf{r}(t) \rangle = \mathbf{q}(t)$. Time series of $q_x(t)$ were recorded and a harmonic analysis was performed using standard routines [15]. Overlapping and nonoverlapping analysis of the power spectrum agreed well in most instances and both are given in the graphs. A more accurate way to calculate the generation of harmonic radiation would be through the dipole correlation function $\langle \mathbf{r}(t)\mathbf{r}(t') \rangle$ (see [19]). It can be argued, however, that for the experimentally available results the factorization of the correlation function represents a good approximation.

## III. RESULTS

We concentrated in our calculations on cases similar to those treated by the numerical solutions of the TDSE. The intermediate laser frequency case of $w=0.2$ a.u. was chosen first, as we have obtained reasonable ionization rates at $3\times10^{14}$ W /cm$^2$ laser intensity while restricting the trial state to the isotropic GWP [10]. This calculation revealed that oscillations in the width parameter built up gradually. This delocalization of the GWP resulted in a softening of the effective potential for the $q_x$ motion and led to much larger amplitudes and eventual breakaway of the $q_x$ parameter from the origin.

A classical calculation results from setting $\hbar=0$ in the system of equations (5). This is equivalent to the restriction of the GWP to maintain a constant width. Such a calculation does not result in ionization for this intensity, but $q_x$ oscillates with fixed amplitude for large times. This is evident from the graph of the effective force acting on $q_x$ due to the Coulomb interaction for the $w_0=0.94$ a.u. case in Fig. 1 and the value of $E_0$ [$E_0=\sqrt{I_0/I_{\rm at}}$ (a.u.) where $I_{\rm at}=3.5\times10^{16}$ W/cm$^2$]. Note that this ($\hbar=0$) calculation is not to be confused with a classical trajectory calculation, in which one would propagate an ensemble of test-particle trajectories. Many such trajectories do ionize [16] due to sequential absorption of photons while a test particle is close to the nucleus. This type of calculation would be termed ($\hbar=0$) semiclassical as it involves the use of classical statistical mechanics in order to simulate quantum mechanics [17]. An analysis of ensembles of classical trajectories to compute the generation of harmonic radiation has been performed recently [20].

From our previous calculations [10,11] one can conclude that the GWP variational calculation with varied width parameter is able to describe ionization due to tunneling at intensities large enough that tunneling occurs in a coherent way [18] or where ionization by escape over the potential barrier is possible. At lower intensities more flexibility in the wave function is required in order to accommodate the possibility of small parts of the wave function tunneling through the barriers that appear alternatingly to the left and right of the Coulomb center. It is this range of intensities that we are interested in here, but we are only aiming for a description of the oscillations of the trapped part of the wave function and not of the ionization process which occurs at a slow rate.

Figure 2 shows results for the time evolution of some of the variational parameters in the anisotropic GWP calculation for $I_0=2\times10^{14}$ W/cm$^2$ and a turnon of the field over 10 cycles. The $q_x$ motion is dominated by oscillations with the external electric field, but a superimposed

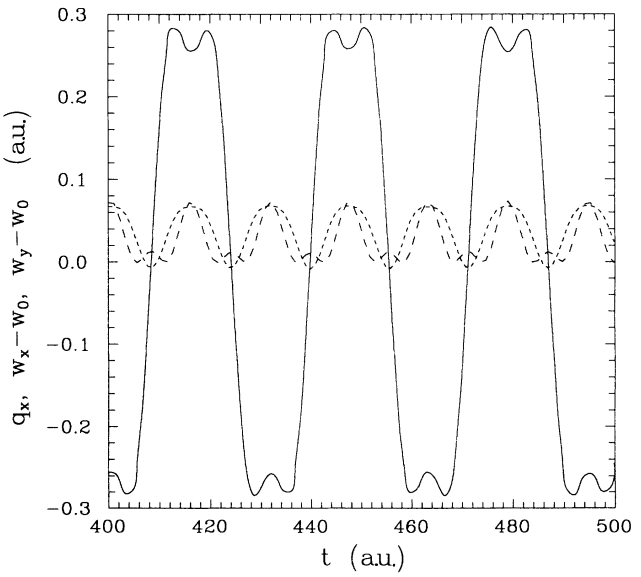


FIG. 2. Time evolution of the GWP parameters $q_x(t)$ (solid line), $w_x(t)-w_0$ (short dashes), and $w_y(t)-w_0=w_z(t)-w_0$ (long dashes) for the case of $\omega=0.2$ a.u. and $I_0=2\times10^{14}$ W/cm$^2$ after the laser field has reached full strength.

motion with the third harmonic is clearly visible. The oscillations of the two independent width parameters $w_x$ and $w_y = w_z$ shown as deviations from the initial value $w_0 \approx 0.94$ a.u. carry twice the laser frequency, such that the GWP is broadened at each peak of the external field. Note that the $w_y = w_z$ parameter does not decrease below $w_0$ while $q_x$ passes through zero, but the $w_x$ parameter does.

In Figs. 3(a) and 3(b) power spectra are shown for the generation of harmonic radiation in units of the external laser frequency. The power spectra were generated from recordings of the position expectation value of the GWP over 30 to 60 cycles of the laser field after it has reached full strength. The classical result (fixed width of the GWP) in Fig. 3(a) displays a strong first harmonic and a third harmonic suppressed by almost three orders of magnitude. The fifth harmonic is weaker than the third by four orders and, in fact, is barely visible in the noise. A classical calculation with the potential (8)—which in some sense can be compared to our effective potential with fixed width—shows that even the third harmonic is suppressed very strongly. This may explain the results found from a solution of the TDSE with this potential, which indicate a small presence of higher harmonics [5].

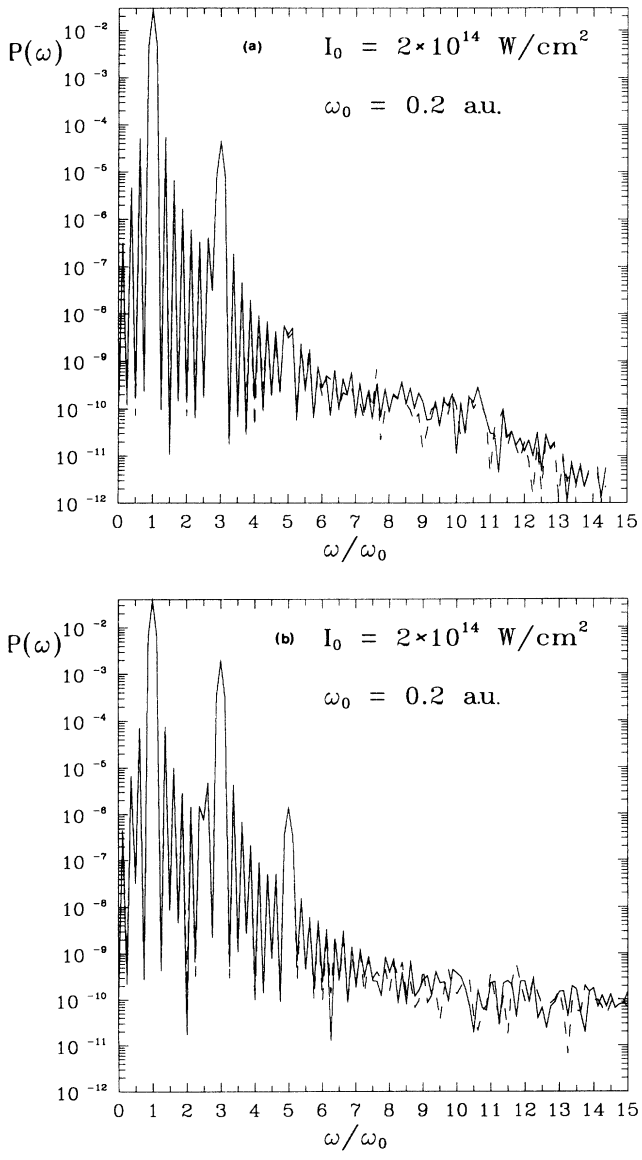


FIG. 3. Power spectrum for the case of $\omega = 0.2$ a.u. and $I_0 = 2 \times 10^{14}$ W/cm$^2$. Solid lines, nonoverlapping, dashed lines, overlapping analysis [15]. (a) Calculation with frozen width parameter ($u_j = 0$, $w_0 = 0.94$ a.u.); (b) anisotropic GWP calculation.

In Fig. 3(b) our results from the calculations with the anisotropic GWP are presented. The third harmonic is weaker than the first one only by slightly more than one order of magnitude. The fifth harmonic is three orders weaker than the third and is clearly visible. The seventh, however, is buried in noise. A mechanism for the strong enhancement of the third and fifth harmonic is visible from Fig. 2: the width parameters oscillate from their equilibrium value at twice the laser frequency. As a consequence the Coulombic restoring force is softened while the packet is away from the center position. Such a lowering of the potential barrier leads ultimately to the fact that ionization can be achieved at a weaker external field than in the classical calculation. At short distances, however, the $w_z$ width parameter falls slightly below the ground-state value giving rise to a stronger attraction. This modulation of the restoring force, which acts in addition to the external sinusoidal laser field, is responsible for the enhancement of the third and fifth harmonics.

These results can be compared to calculations where the TDSE was converted to a set of radial equations that were integrated numerically [13]. Harmonics up to order 23 were seen there, but the power spectrum is given only on a relative scale. The third harmonic in this calculation is suppressed by almost four orders of magnitude with respect to the first, but then the fifth comes in very closely. This change in the relative strength of the third harmonic could be related to the different turnon in the laser field. We do observe an enhancement of orders 7 to 13 in our calculation if only five cycles after turnon are used as opposed to the 40 cycles that were used for the analysis shown in Figs. 3(a) and 3(b).

In Figs. 4(a) and 4(b) we provide power spectra for $\omega = 0.05$ a.u. and two intensities $I_0 = 5 \times 10^{13}$ and $2 \times 10^{14}$ W/cm$^2$. Fast ionization of our model atom sets in at $I_0 = 4 \times 10^{14}$ W/cm$^2$ due to absorption of at least ten photons. Apart from an expected behavior of the strengths of the harmonics of order 1 to 5 we observe a strong appearance of harmonics of order 9 to 15 superimposed on a broad structure. This behavior is comparable to what is observed in calculations of harmonic generation in Xe atoms in similar fields [12]. In this case, however, such structures and higher harmonics as well are due to the response of a $5p_0$ orbital.

In Fig. 5 we show the behavior of the peak heights in the power spectrum as a function of laser intensity $I_0$. The first and third harmonics demonstrate the $I^n$ dependence as predicted by perturbation theory up to intensities of the order of $10^{14}$ W/cm$^2$, beyond which they rise more steeply. The fifth and seventh harmonics show such a dependence at intensities larger than $10^{14}$ W/cm$^2$. Very

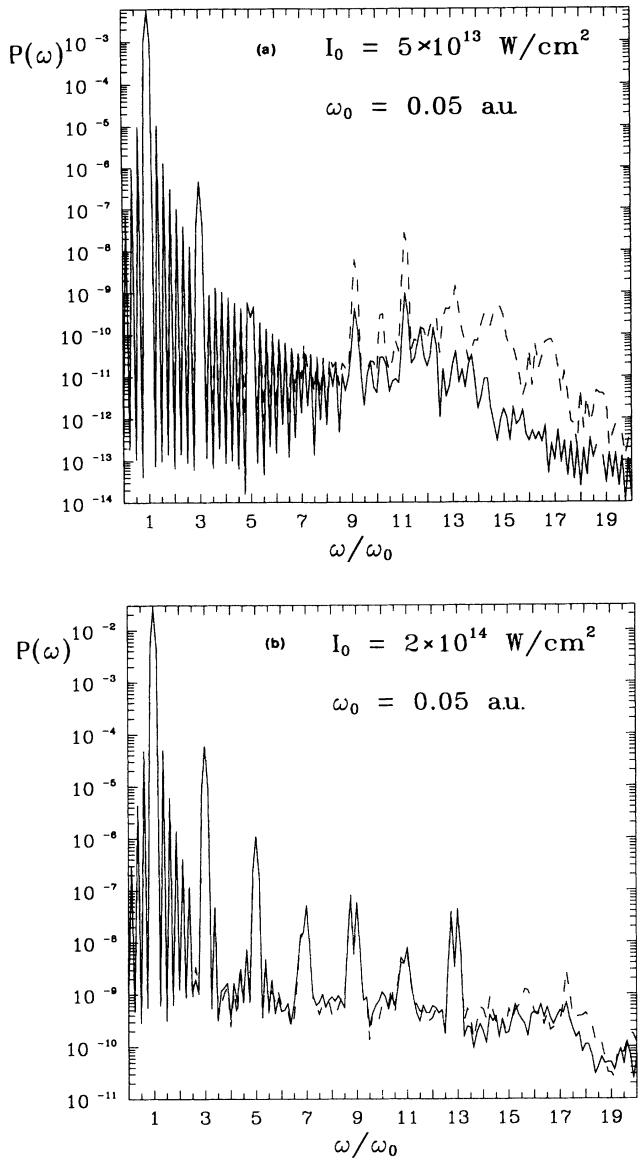


FIG. 4. Power spectra as in Fig. 3(b) for the cases of $\omega=0.05$ a.u. and $I_0=5\times10^{13}$ W/cm$^2$ (a) and $\omega=0.05$ a.u. and $I_0=2\times10^{14}$ W/cm$^2$ (b).

nonperturbative behavior is displayed by the higher harmonics in the range $10^{12}-10^{14}$ W/cm$^2$.

Another point of interest is the region close to $4\times10^{14}$ W/cm$^2$. For these intensities we notice that once the GWP ionizes quickly, only the first harmonic is clearly visible, i.e., the higher harmonics that rise steeply up to this point do not persist once the packet is set free rapidly. It will be interesting to study this regime in the numerical approach and to track the dependence of the strength of the harmonic peaks as a function of laser intensity.

## IV. CONCLUSIONS

We have studied in this work the generation of harmonic radiation due to the acceleration that a H(1$s$) orbital experiences in a laser field. Equations of motion for the position expectation value were derived from a TDVP. It has been shown that the freedom to vary the width parameters of the GWP results in a strong enhancement of harmonics of order 3 to 7 for fields where three photons can ionize the atom. We do not see a plateau in the power spectrum extending to very high harmonics. This suggests that the conjecture that the appearance of very high harmonics in the numerical TDSE calculations stems from the ionized parts of the wave function is true. The generation of harmonic radiation has been linked in the literature to the appearance of above-threshold ionization peaks [6]. Our bound-state wave function is not flexible enough to describe ionization at these intensities, yet it predicts the generation of harmonic radiation. The role of ionizing versus bound-state parts of the wave function contributing towards emitted radiation should be studied in the future by numerical calculations in which the position expectation values are calculated only over a finite range of space.

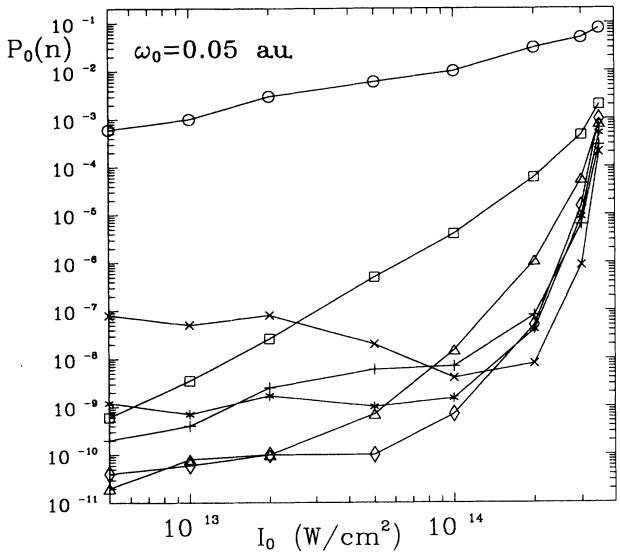


FIG. 5. Laser intensity dependence of the peaks for the harmonics of order $n=\omega/\omega_0$ in the power spectra for $\omega=0.05$ a.u. $\circ, n=1; \square, n=3; \triangle, n=5; \Diamond, n=7; +, n=9; \times, n=11; *, n=13$.

For low-frequency laser fields our calculation reveals a broad structure in the power spectrum and a strong nonperturbative enhancement of harmonics of order 7 to 15 at intensities between $10^{12}$ and $10^{14}$ W/cm$^2$. This is in accord with recent numerical calculations for Xe atoms [12].

The one-electron calculations described above were performed on a personal computer. Therefore, many-electron calculations in an independent-particle model without the frozen-core approximation are feasible and are planned to be the subject of future work.

## ACKNOWLEDGMENT

The financial support of the Natural Sciences and Engineering Research Council of Canada is gratefully acknowledged.

[1] An overview of the field is given in the topical issue *Theory of High-Order Processes in Atoms in Intense Laser Fields* [J. Opt Soc. Am. B **7**, 403-688 (1990)] and more recently by A. L'Huillier, K. J. Schafer, and K. C. Kulander, J. Phys. B **24**, 3314 (1991).
[2] A. Goldberg, H. M. Schey, and J. L. Schwartz, Am. J. Phys. **35**, 177 (1967).
[3] K. C. Kulander, Phys. Rev. A **35**, 445 (1987); **36**, 2726 (1987).
[4] J. Javanainen, J. H. Eberly, and Q. Su, Phys. Rev. A **38**, 3430 (1988).
[5] J. H. Eberly, Q. Su, J. Javanainen, K. C. Kulander, B. W. Shore, and L. Rosa-Franco, J. Mod. Opt. **36**, 829 (1989).
[6] Q. Su and J. H. Eberly, J. Opt. Soc. Am B **7**, 564 (1990).
[7] Q. Su, J. H. Eberly, and J. Javanainen, Phys. Rev. Lett. **64**, 862 (1990).
[8] M. Kleber and M. Unterseer, Z. Phys. A **292**, 311 (1979); Nucl. Instrum. Methods **192**, 35 (1982).
[9] M. Horbatsch, J. Chem. Phys. **79**, 4382 (1983); J. Phys. B **17**, 2591 (1984).
[10] J. K. Liakos and M. Horbatsch, J. Opt. Soc. Am. B **7**, 685 (1990).
[11] J. K. Liakos and M. Horbatsch, J. Phys. B **24**, 3387 (1991).
[12] B. W. Shore and K. C. Kulander, J. Mod. Opt. **36**, 859 (1989); J. Opt. Soc. Am. B **7**, 502 (1990).
[13] P. L. De Vries, J. Opt. Soc. Am. B **7**, 517 (1990).
[14] F. Brunel, J. Opt. Soc. Am. B **7**, 521 (1990).
[15] W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge University Press, New York, 1986).
[16] S. -I. Chu, K. Wang, and E. Layton, J. Opt. Soc. Am. B **7**, 425 (1990).
[17] M. Horbatsch, Z. Phys. D **1**, 337 (1986); Comput. Phys. Commun. **64**, 115 (1991).
[18] M. Razavy and A. Pimpale, Phys. Rep. **168**, 305 (1988).
[19] B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990).
[20] G. Bandarage, A. Maquet, and J. Cooper, Phys. Rev. A **41**, 1744 (1990).