High-order harmonic generation in atomic hydrogen at 248 nm: Dipole-moment versus acceleration spectrum

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We present a study of the high-order harmonic-generation (HG) spectra of atomic hydrogen at 248 nm based on the Fourier transform of the expectation values of the induced dipole moment and acceleration. The calculations were performed by extending a fast-Fourier-transformation split-operator technique (in spherical coordinates) to the solution of the time-dependent wave functions in intense laser fields. It is seen that the acceleration form provides a more satisfactory and accurate framework for the study of the photoemission HG spectrum at very intense fields, particularly when ionization becomes appreciable.

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

Recently there is much interest, both theoretically and experimentally, in the study of high-order harmonic-generation (HG) phenomena of atoms in intense laser fields [1]. Almost all the theoretical calculations of the HG spectra so far were based on the Fourier transform of dipole moment [1]. It has been pointed out recently, however, that a more appropriate procedure for determining the HG spectra should be based on the Fourier transform of the acceleration vector [2,3], particularly in very intense laser fields. The one-dimensional model calculation by Burnett *et al.* [2] shows that the HG spectra based on the dipole moment can lead to spurious background appearing in the HG spectrum especially in the region where ionization is appreciable. There is as yet, however, no report on the acceleration power spectrum of any realistic systems.

The purposes of this communication are twofold: (i) We perform an *ab initio* study of the HG spectra of three-dimensional hydrogen atoms based on both dipole-moment and acceleration forms; and (ii) we examine the feasibility of extending the fast-Fourier-transformation split-operator spectral technique [4] in *spherical* coordinates (which has been previously used for dc field problems) to time-dependent intense-field problems.

The Hamiltonian for a hydrogen atom driven by linearly polarized fields can be written, in spherical coordinates, as ($\hbar = 1$)

$$\hat{H} = -(\tfrac{1}{2})\partial^2/\partial r^2 + \hat{L}^2/2r^2 - 1/r - Fzf(t)\sin\omega t \ , \qquad (1)$$

where F is the electric-field amplitude and $f(t)$ the pulse shape of the laser field. The two-dimensional split-operator technique in (r,θ) coordinates will be extended here for the time propagation of the wave function in the Schrödinger equation. Thus

$$\psi(t+\Delta t) = \exp[-i\Delta t\, \hat{p}_r^2/4]\exp[-i\Delta t(\hat{L}^2/4r^2 - 1/2r)]$$
$$\times \exp[-i\Delta t \cdot r\cos(\theta)E(t)]$$
$$\times \exp[-i\Delta t(\hat{L}^2/4r^2 - 1/2r)]$$
$$\times \exp[-i\Delta t\hat{p}_r^2/4]\Psi(t) + O(\Delta t^3) \ , \qquad (2)$$

where $\hat{p}_r = (\hbar/i)\partial/\partial r$ and $E(t) = Ff(t)\sin\omega t$. At each time step, we expand the wave function in Legendre polynomials,

$$\psi(r_i, x_j, t) = \sum_{l=0}^{L} g_l(r_i, t)P_l(x_j), \quad x_j = \cos\theta_j \ , \qquad (3)$$

where the P_l's are normalized Legendre polynomials. $g_l(r_i, t)$ can be determined accurately by the Gauss-Legendre quadrature,

$$g_l(r_i, t) = \sum_{k=1}^{L+1} w_k P_k(x_k)\psi(r_i, x_k, t) \ , \qquad (4)$$

where x_k's are the $L+1$ zeros of the Legendre polynomial $P_{L+1}(x)$ and w_k's are the corresponding quadrature weights. In performing the time propagation in Eq. (2), the evolution operator is factored into several propagators which are either functions of the coordinate r or momentum operators. Each propagator is evaluated in a representation for which it is diagonal and fast Fourier transformation is used to link efficiently back and forth between the coordinate and momentum space representation.

In our calculations, 512 evenly distributed space grid points in r, and up to 40 partial waves (and therefore up to 40th-order Gauss-Legendre quadrature points) are used. A filter function of the form $f(r) = \{1 + \exp[br - 90]\}^{-1}$ is used to filter out the wave packet reaching the outward boundary $r_{\max}$. The use of

$r_{max} = 100a_0$, $b = 1.25$ a.u and $\Delta t = 0.01$ a.u. is found to be satisfactory in the present calculation to yield converged HG results. The laser field is ramped on according to

$$E(t) = \begin{cases} F\sin\omega t\, \sin^2(\pi t/10T), & t \leq 5T \\ F\sin\omega t, & t \geq 5T \end{cases} \quad (5)$$

where $T = 2\pi/\omega$. The power spectrum in dipole-moment form is obtained from the Fourier transform of the expectation value of the time-dependent induced dipole moment $\langle z(t) \rangle = \langle \psi(\mathbf{r},t)|z|\psi(\mathbf{r},t)\rangle$. Similarly, the power spectrum in acceleration form is determined by Fourier transforming the expectation value of the acceleration $\langle \ddot{z}(t) \rangle$. Here the mean acceleration can be obtained directly by means of Ehrenfest's theorem,

$$\langle \ddot{z}(t) \rangle = \langle -\partial \hat{H}/\partial z \rangle = -\langle z/r^3 \rangle - E(t)\langle \psi(t)|\psi(t)\rangle \ . \quad (6)$$

The expectation values $\langle z \rangle$ and $\langle z/r^3 \rangle$ can be evaluated accurately by the Gauss-Legendre quadrature. Figure 1 shows the induced dipole moment $\langle z(t) \rangle$ (dotted line) and the acceleration $\langle \ddot{z}(t) \rangle$ (solid line) as a function of time t for the case of laser intensity $I = 4.375 \times 10^{13}$ W/cm^2 and $\lambda = 248$ nm. The hydrogen atom is initially ($t = 0$) prepared in the ground state. The corresponding power spectra are shown in Fig. 2: dipole spectrum (dotted line) and acceleration spectrum (solid line). At this field intensity, only three harmonic peaks are observed, and the two spectra are in good agreement, apart from a numerical factor (mainly due to the ω^4 factor [2]). The survival probability of the ground state (dashed line in Fig. 3) indicates that little ionization occurs before 20 optical cycles. This is also confirmed by the magnitude of the total multiphoton ionization width (rate) from H($1s$), $\Gamma/2 \simeq 1.294 \times 10^{-4}$ a.u., obtained from an independent calculation using the L^2 non-Hermitian Floquet matrix

FIG. 2. The dipole (dotted line) and acceleration (solid line) power spectra corresponding to the physical parameters of Fig. 1.

FIG. 3. The survival probability of H ($1s$) vs time. Dashed line, $I = 4.375 \times 10^{13}$ W/cm^2; solid line, $I = 7.0 \times 10^{14}$ W/cm^2.
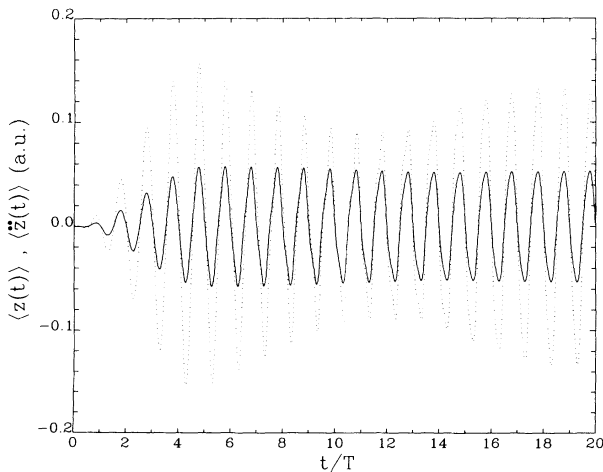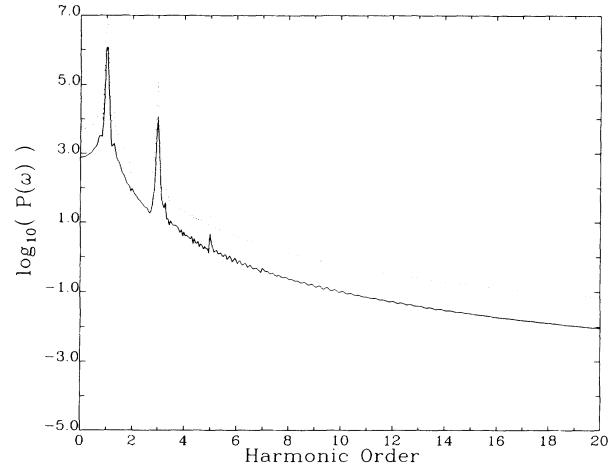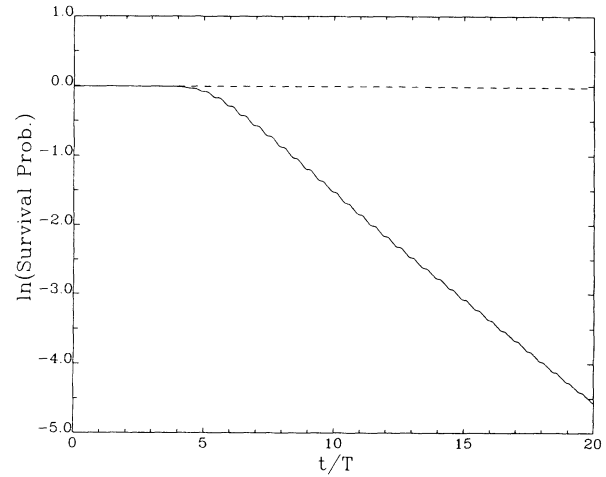

FIG. 1. Induced dipole moment $\langle z(t) \rangle$ (dotted line) and acceleration $\langle \ddot{z}(t) \rangle$ (solid line), in atomic units, as a function of time t. T is the period of field oscillation. The laser intensity is $I = 4.375 \times 10^{13}$ W/cm^2 and $\lambda = 248$ nm.
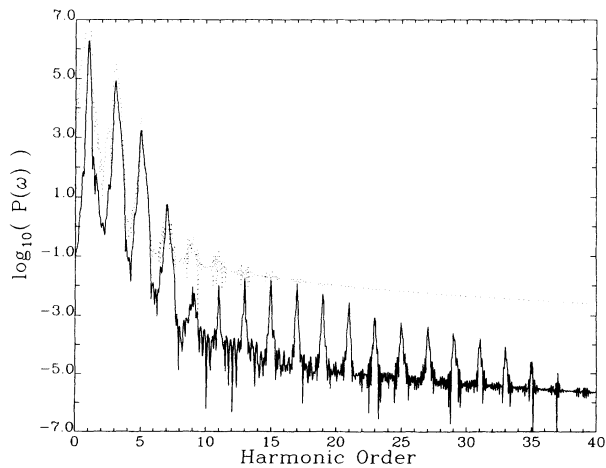

FIG. 4. The dipole (dotted line) and acceleration (solid line) power spectra at $I = 7.0 \times 10^{14}$ W/cm^2 and $\lambda = 248$ nm.
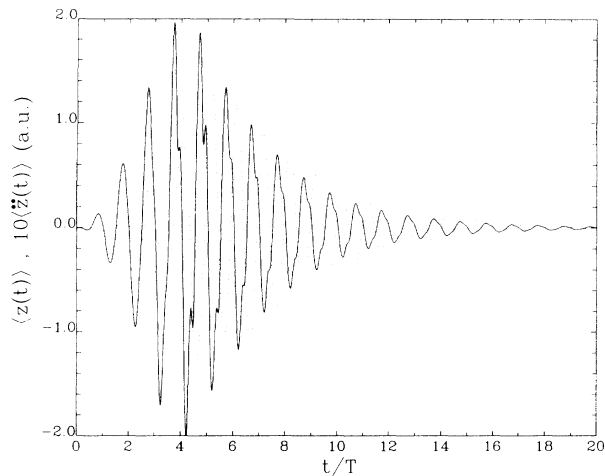
FIG. 5. $\langle z(t) \rangle$ (dotted line) and $\langle \ddot{z}(t) \rangle$ (solid line) correspond to the physical parameters of Fig. 4.

method [5]. This suggests that the dipole-moment and acceleration forms provide equivalent HG results at weaker fields when little ionization occurs.

Figure 4 shows the dipole-moment and acceleration power spectra for a stronger field $I = 7.0 \times 10^{14}$ W/cm^2 and at $\lambda = 248$ nm. The acceleration photoemission power spectrum (solid line) contains well-resolved high-order HG peaks up to 37th order, a feature similar to those observed in the rare-gas experiments [6]. However, the dipole power spectrum (dotted line) contains a spurious background and all higher-order HG peaks are completely masked. Floquet calculation at this intensity shows large ionization width ($\Gamma/2 \simeq 1.657 \times 10^{-2}$ a.u.).

This is also revealed from the survival probability plot (Fig. 3, solid line): large depletion of the ground-state population occurs before 20 optical cycles. As pointed out by Burnett et al. [2], the spurious background in the dipole spectrum can be attributed to the improper use of the dipole moment which is not vanishing at large times when ionization occurs. In our calculation, however, we have placed a filter function at r_{max} to filter out the ionized wave packets. Thus the induced dipole moment (dotted line) shown in Fig. 5 represents only the mean dipole moment obtained from the bounded portion of the wave packets whose magnitude decreases with time rapidly due to ionization. (There is no apparent "drift" in the mean dipole moments and acceleration shown in Figs. 1 and 5, since the drift motion is mainly contained in the ionized wave packets which have been filtered out. This is consistent with the behavior of the mean dipole-moment and acceleration results obtained from the L^2 non-Hermitian Floquet calculation [7].) Despite this, the dipole spectrum is not able to reveal the higher-order HG structure. Previous classical calculations [8] indicated that HG can occur only when the electron is close to the nucleus and any asymptotic value of the dipole moment is not relevant. The acceleration form Eq. (6) yields the correct result since it takes into account automatically and self-consistently the fact that the dominant contribution to HG arises from the portion of the wave function where the electron experiences the largest force.

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