

Numerical Simulation of Nonlinear Ionization in an Intense Laser Field

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Abstract—A code to integrate the three-dimensional time-dependent Schrödinger equation of an atomic system interacting nonlinearly with an intense laser pulse is developed based on the spectral method. The wave function is represented in terms of Coulomb-Sturmian basis functions. The simultaneous ordinary differential equations for the expansion coefficients are integrated by an implicit integration method with an adaptive time step to time propagate the atomic state. Numerical simulations of higher harmonic generation spectra and the so-called ionization stabilization for atomic hydrogen irradiated by a linearly polarized intense laser field is presented to verify the code.

1. Introduction

During past decade, the interaction of intense ultrashort laser pulses with atomic systems, which leads to highly nonlinear phenomena such as the so-called above-threshold ionization (ATI) and high harmonic generation (HHG), has been an experimental and theoretical subject of interest. In the ATI process, the energy spectrum of photoelectrons have peaks which are separated by the photon energy of the laser and correspond to the absorption of more photons than is required to overcome the binding potential. The HHG process is characterized by a spectrum of coherent radiation emitted at frequencies of odd multiples of the incident laser frequency. These phenomena have stimulated great interest in the possibilities of developing a new highly charged ion source, a new coherent source in the soft-x-ray region, or attosecond pulses from intense laser-atom interactions [1, 2].

However, these nonlinear interactions transcend the perturbative treatment. Since the interaction is so strong, *i.e.* the potential of the laser field is comparable to the binding energy of the atom, an ionizing atom does not immediately become a well-separated electron and ion. Therefore, the electron extracted from the atom returns and interacts in the vicinity of the ion for one or more laser periods. Floquet theory can be adopted in the analysis of atoms in periodic external fields with constant amplitude, however in reality intense fields are only produced in the form of short pulses. One nonperturbative approach has been formulated by Keldysh [3]. In this theory, the effect of the nuclear potential in the final state is assumed to be negligible after the interaction with the strong laser field, and a free electron

state in the time-dependent radiation field is taken as the final state. Keldysh's method has been refined by Faisal [4] and Reiss [5] by adopting the formal time-dependent scattering theory. On the other hand, Ammosov *et. al.* [6] proposed a closed form formula for the ionization probability as a function of the laser electric fields, based on the assumption that the wave function is approximated via the WKB method in the tunnel ionization regime. Although these methods have been extensively applied in the analysis of ATI and HHG process, there are still significant discrepancies in the estimated ionization probability among them [7].

Nowadays, it is widely recognized that simulations based on the direct integration of the time-dependent Schrödinger equation (TDSE) are extremely useful in elucidating these complex physical processes and predicting new effects. A large number of numerical methods have been developed, however most of them are one-dimensional models or use a modified nuclear potential to avoid numerical singularities. To the best of the authors' knowledge, computations even for real three-dimensional hydrogen have not been fully explored. We have developed a three-dimensional TDSE solver based on the spectrum method [8], and for the sake of illustration applied it to the ionization of atomic hydrogen by a strong ultrashort laser pulse.

2. Numerical Method

The Schrödinger equation which describes the time evolution of the wave function $\psi(\mathbf{x}, t)$ of an hydrogenic atomic system interacting with an electromagnetic field can be written as (we use atomic units $e = m = \hbar = 1$)

$$i \frac{\partial}{\partial t} \psi(\mathbf{x}, t) = (H_a + H_I) \psi(\mathbf{x}, t), \quad (1)$$

$$H_a = \frac{\mathbf{p}^2}{2} - \frac{Z}{r}, \quad (2)$$

where \mathbf{p} is the canonical momentum operator for the electron, Z is the nuclear charge, H_a is the unperturbed atomic Hamiltonian, and H_I is the interaction Hamiltonian. The interaction Hamiltonian using the dipole approximation can be written in the velocity gauge form as

$$H_I = \frac{1}{c} \mathbf{A}(t) \mathbf{p}, \quad (3)$$

where c is the speed of light, and $\mathbf{A}(t)$ is the (spatially independent) vector potential; we have removed the $\mathbf{A}(t)^2$ term by a simple gauge transformation. Here, we assume that the laser pulse propagates in the z -direction. Then the vector potential is expressed as

$$\begin{aligned}\mathbf{A}(t) &= f(t)(A_1 \cos(\omega t + \phi_1 t)\mathbf{e}_x + A_2 \sin(\omega t + \phi_2 t)\mathbf{e}_y) \\ &= A_x(t)\mathbf{e}_x + A_y(t)\mathbf{e}_y,\end{aligned}\quad (4)$$

where $f(t)$, ω , A_1 , A_2 , ϕ_1 , ϕ_2 are, respectively, the pulse envelope, the frequency, and the amplitude and the initial phase of the x and y component. Since the electric field $\mathbf{E}(t)$ is obtained from the relation $\mathbf{E}(t) = -(1/c)(d/dt)\mathbf{A}(t)$, linear, circular, or elliptical polarization of the laser field can be represented by properly setting the values A_1 , A_2 , ϕ_1 , ϕ_2 .

Since H_a is spherically symmetric, we expand the wave function with the basis consisting of products of Coulomb-Sturmian functions S_{nl}^κ and spherical harmonics Y_{lm} as [8]

$$\psi(\mathbf{x}, t) = \sum_{nlm} a_{nlm}(t) \frac{S_{nl}^\kappa(r)}{r} Y_{lm}(\mathbf{n}_x), \quad (5)$$

where \mathbf{n}_x is a unit vector which points along the direction \mathbf{x} . The Sturmian functions are eigenfunctions of the differential equation

$$\left(-\frac{1}{2} \frac{d^2}{dr^2} + \frac{l(l+1)}{2r^2} + \frac{\kappa^2}{2}\right) S_{nl}^\kappa(r) = \frac{\alpha}{r} S_{nl}^\kappa(r), \quad (6)$$

with the boundary condition

$$S_{nl}^\kappa(0) = S_{nl}^\kappa(\infty) = 0. \quad (7)$$

In this equation, κ is a constant real value to ensure the discreteness of the eigenvalue spectrum. Therefore, the eigenvalue is $\alpha = n\kappa$, where n is a positive integer larger than the angular momentum quantum number l , and the eigenfunctions are represented by using convenient hypergeometric functions as

$$S_{nl}^\kappa(r) = N_{nl}^\kappa r^{l+1} e^{-\kappa r} F_1(-n+l+1; 2l+2; 2\kappa r). \quad (8)$$

The normalization constant N_{nl}^κ is determined from the condition

$$\langle S_{nl}^\kappa | S_{nl}^\kappa \rangle \equiv \int_0^\infty S_{nl}^\kappa(r) S_{nl}^\kappa(r) dr = 1, \quad (9)$$

and the Sturmian functions satisfy the orthonormality condition

$$\langle S_{nl}^\kappa | \frac{1}{r} | S_{n'l}^\kappa \rangle = \frac{\kappa}{n} \delta_{nn'}. \quad (10)$$

With the above convention, the matrix elements of H_a , H_I may be expressed as follows:

$$\langle S_{nl}^\kappa Y_{lm} | H_a | S_{n'l}^\kappa Y_{l'm'} \rangle = \left((\kappa^2 - \frac{Z\kappa}{n} \delta_{nn'} - \frac{\kappa^2}{2} \langle S_{nl}^\kappa | S_{n'l}^\kappa \rangle \right) \delta_{l'l'} \delta_{mm'} \quad (11)$$

$$\begin{aligned}\langle S_{nl}^\kappa Y_{lm} | H_I | S_{n'l}^\kappa Y_{l'm'} \rangle &= -i \sqrt{\frac{2\pi}{3}} \kappa(n-n') \langle S_{nl}^\kappa | S_{n'l}^\kappa \rangle \\ &\times \left[\frac{A^-}{c} \langle lm | Y_{11} | l'm' \rangle - \frac{A^+}{c} \langle lm | Y_{1-1} | l'm' \rangle \right],\end{aligned}\quad (12)$$

where

$$\begin{aligned}A^+(t) &= A_x(t) + iA_y(t), \\ A^-(t) &= A_x(t) - iA_y(t),\end{aligned}\quad (13)$$

$\langle lm | Y_{11} | l'm' \rangle$ is the Clebsch-Gordan coefficient, and the non-zero matrix elements of $\langle S_{nl}^\kappa | S_{n'l}^\kappa \rangle$ are

$$\begin{aligned}\langle S_{nl}^\kappa | S_{nl}^\kappa \rangle &= 1, \\ \langle S_{nl}^\kappa | S_{n+1}^\kappa \rangle &= -\frac{1}{2} \sqrt{\frac{(n+l+1)(n-l)}{n(n+1)}}, \\ \langle S_{nl}^\kappa | S_{n-1}^\kappa \rangle &= -\frac{1}{2} \sqrt{\frac{(n+l)(n-l-1)}{n(n-1)}}, \\ \langle S_{nl}^\kappa | S_{n+l+1}^\kappa \rangle &= \frac{1}{2} \sqrt{\frac{(n+l+1)(n+l+2)}{n(n+1)}}, \\ \langle S_{nl}^\kappa | S_{n+l+1}^\kappa \rangle &= -\sqrt{\frac{(n+l+1)(n-l-1)}{n^2}}, \\ \langle S_{nl}^\kappa | S_{n-l+1}^\kappa \rangle &= \frac{1}{2} \sqrt{\frac{(n-l-1)(n-l-2)}{n(n-1)}}, \\ \langle S_{nl}^\kappa | S_{n+l-1}^\kappa \rangle &= \frac{1}{2} \sqrt{\frac{(n-l)(n-l+1)}{n(n+1)}}, \\ \langle S_{nl}^\kappa | S_{n-l-1}^\kappa \rangle &= -\sqrt{\frac{(n+l)(n-l)}{n^2}}, \\ \langle S_{nl}^\kappa | S_{n-l-1}^\kappa \rangle &= \frac{1}{2} \sqrt{\frac{(n+l)(n+l-1)}{n(n-1)}}.\end{aligned}\quad (14)$$

Therefore, the time-dependent Schrödinger equation may be written in the Sturmian basis as follows:

$$iS \frac{d}{dt} \mathbf{a}(t) = H(t) \mathbf{a}(t) \quad (15)$$

where H is the total Hamiltonian $H = H_a + H_I$, $\mathbf{a}(t)$ is a vector which contains coefficients $a_{nlm}(t)$, and S is the overlap matrix whose elements are

$$\langle S_{nl}^\kappa | S_{n'l}^\kappa \rangle = \delta_{l'l'} \delta_{nn'}. \quad (16)$$

Since the continuum states expanded by the discrete Coulomb-Sturmian basis require those with large eigenvalues, the above ordinary differential equation is stiff in many cases as seen from Fig. 1, *i.e.* the difference of the scale of eigenenergies amounts to 5. Therefore, we used the implicit solver (subroutine *dlsodis*) with general sparse Jacobian matrices to time propagate the solution developed by Hindmarsh and Balsaon [9], in which the step-size is adaptively controlled.

3. Numerical Results

Figure 2 shows the ionization probability of a hydrogen atom initially in its ground state as a function of the peak electric field (upper) and time (lower), where the incident laser is linearly polarized with a photon energy of 1.5 a.u. and the time profile of the

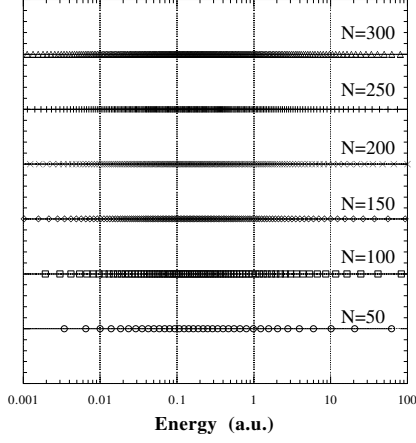


Figure 1:

Positive eigenenergies of hydrogen atom calculated with a Coulomb-Sturmian basis and $\kappa = 0.5$. N is the maximum of the principal quantum number n .

electric eld is a step function for 10 optical cycles. The ionization probability is calculated as

$$(1 - \sum_{nlm} |\langle \psi_{b,nlm} | \psi(t) \rangle|^2), \quad (17)$$

where $\psi_{b,nlm}$ is the eigenfunction of $H_a \psi = \lambda \psi$ with negative eigenvalues $\lambda < 0$. In this calculation, we set the parameters to be $\kappa = 0.5, n_{max} = 150$, and $l_{max} = 30$. The number of state is 3825. The ionization probability at the end of the irradiation increases linearly, when the electric eld is increased to 1 a.u. ($3.5 \times 10^{16} W/cm^2$). In this regime, the ionization probability also increases in time as seen in Fig. 2. However, we can observe that the ionization probability begins to decrease and wiggles when the electric eld is increased beyond 1 a.u., *i.e.* the so-called dynamic ionization stabilization.

In Fig. 3 we display the HHG power spectrum when the intensity of the laser is $1.4 \times 10^{14} W/cm^2$. Here, we obtained the HHG spectrum from the expectation value of the acceleration operator [10, 11] as

$$\begin{aligned} \langle \psi | \ddot{x} | \psi \rangle &= \langle \psi | p_x | \psi \rangle = \langle \psi | \nabla_x V(r) | \psi \rangle \\ &= \langle \psi(\mathbf{x}, t) | \frac{Z \cos \theta}{r^2} | \psi(\mathbf{x}, t) \rangle, \end{aligned} \quad (18)$$

where we assumed the electric eld is polarized in the x -direction, and $\cos \theta = \mathbf{e}_x \cdot \mathbf{n}_x$. The spectra of the HHG are proportional to the modulo square of the Fourier transform of the above expectation value. It is noted the acceleration operator is proportional to $\frac{1}{r^2}$. This means that the HHG spectrum is sensitive to the wave function in the vicinity of the nucleus. Therefore, a merit of this method is that the expectation value Eq.(18) is calculated using analytical matrix elements $\langle S_{nl}^k | \frac{1}{r^2} | S_{n'l'}^k \rangle$ avoiding the numerical singularity. This result displays the typical characteristics of the observed HHG spectrum including odd order radiation, a plateau, and cut-off.

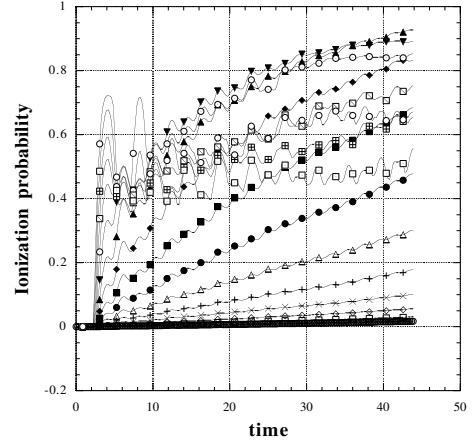
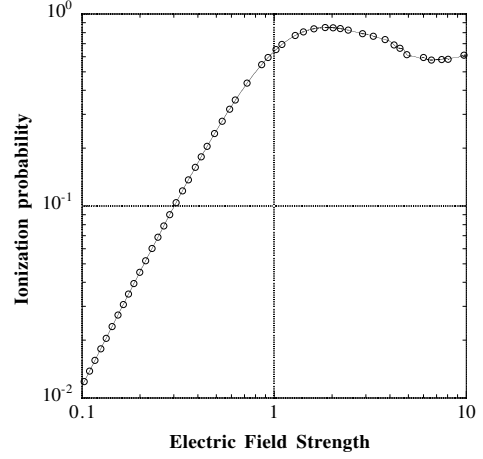


Figure 2:

Ionization probability as a function of the peak electric eld in a.u. (upper) and of time (lower). The linearly polarized laser pulse with a photon energy of 1.5 a.u. is irradiated for 10 optical cycles. In the calculation, we take $\kappa = 0.5, n_{max} = 150$, and $l_{max} = 30$.

4. Conclusion

We have developed a time-dependent Schrödinger equation solver, in which the wave function is represented in terms of Coulomb-Sturmian basis functions, and have demonstrated that nonlinear laser-hydrogenic atom interactions can be properly simulated.

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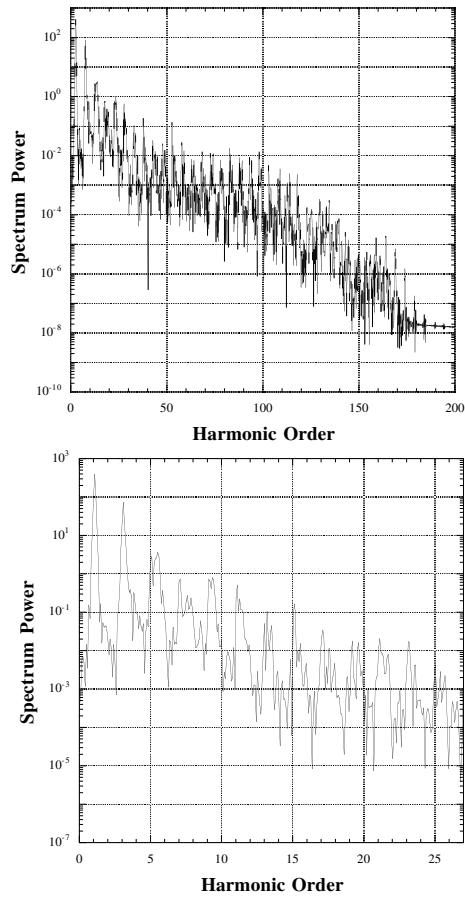


Figure 3:

High harmonic generation spectra of the hydrogen atom for the laser intensity $1.4 \times 10^{14} \text{ W/cm}^2$.

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