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Pseudospectral method with symplectic algorithm for the solution of time-dependent Schrödinger equations*

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A pseudospectral method with symplectic algorithm for the solution of time-dependent Schrödinger equations (TDSE) is introduced. The spatial part of the wavefunction is discretized into sparse grid by pseudospectral method and the time evolution is given in symplectic scheme. This method allows us to obtain a highly accurate and stable solution of TDSE. The effectiveness and efficiency of this method is demonstrated by the high-order harmonic spectra of one-dimensional atom in strong laser field as compared with previously published work. The influence of the additional static electric field is also investigated.

Keywords: pseudospectral method, symplectic algorithm, high-order harmonic generation
PACC: 0290, 3120A, 3280K

1. Introduction

In recent years, high-intense and ultra-short lasers have brought powerful sources of electromagnetic radiation into the laboratory. Laser intensities of $10^{16}\mathrm{W/cm^2}$ and more are routinely achieved in many laboratories all over the world. The length of the ultra-short laser pulse reaches femtosecond even attosecond order. So, many nonlinear novel phenomena in laser–atom interaction, such as high-order harmonic generation (HHG),[1,2] multiphoton ionization (MPI), above threshold ionization (ATI) and nonsequential ionization, are presented to us.[3] All these phenomena are not accessible through conventional perturbation theory where the external laser field is required to be small as compared with the atomic binding force acting on electron. Hence, a full *ab initio* solution of the time-dependent Schrödinger equation is desirable.[4] People have developed many ways, such as split-operator,[5] Crank–Nicholson,[6,7] Taylor series expansion method,[8] Runge–Kutta method, symplectic algorithm,[9] and so on, to deal with the time evolution of the TDSE. The split-operator and Crank–Nicholson are unconditional stable methods while the accuracy of them are $(\Delta t)^3$. The Taylor series expansion method, Runge–Kutta method and symplectic algorithm are flexible with accuracy from $(\Delta t)^3$ up to $(\Delta t)^5$ which is enough for usual needs. However, with the Taylor series expansion method and the classical Runge–Kutta method it is difficult to preserve the unitarity of the time evolution operator. The fundamental theorem of Hamiltonian mechanics says that the time-evolution of the Hamiltonian system is the evolution of symplectic transformation. In this sense, we say that the Hamiltonian system has a symplectic structure. Thus, Ruth and Feng presented the symplectic algorithm for solving the Hamiltonian system, and found a new method for solving the Hamiltonian system.[9−13] The high accuracy, stability and symplectic structure preserving property is the reason why we adopt it to deal with the time evolution.

To deal with the spatial part of the wavefunction, conventional methods mainly fall into two categories. The first is basis expansion, which may be selected from a complete orthogonal set or chosen for specific properties such as symmetry and spatial behaviour: for example, Sturmian basis, B-spline basis,[14,15] and so on. The second is the iterative method approximating Laplacian by second-order finite difference,[6] which is mostly much CPU and memory consuming because of dense and uniform grid. The Coulomb singularity problem is another trouble.

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Fortunately, the pseudospectral method reduces the problem to nonuniform grid points which are dense near the core while sparse far from the nucleus.[4,16,17] It has been successfully used not only in spherical system, but also in prolate spherical[18] and hyperspherical coordinates.[19] It greatly reduces the computation and avoids the Coulomb singularity problem, which is the reason why we alternatively adopt it to deal with the spatial part of the wavefunction.

People in the atomic and molecular field are still devoting effort to develop new methods to solve TDSE numerically with high precision.[20] It is our main aim to combine the pseudospectral method and symplectic algorithm to solve the TDSE. To our knowledge, no such method has been presented. In this paper, the method is introduced in Section 2; the HHG spectrum of a one-dimensional soft potential is shown in Section 3 in comparison with our previous work.[14] Conclusions are presented in Section 4.

2. The method

We consider a one-dimensional (1D) atom in a laser field. The TDSE with dipole approximation in length gauge is given as follows (atomic units are used throughout this paper):

$$\begin{aligned} \mathrm{i}\frac{\partial \Psi(x,t)}{\partial t} &= H(x,t)\,\Psi(x,t) \\ &= [H_0(x)+H_t(x)]\,\Psi(x,t), \end{aligned} \tag{1}$$

where

$$\begin{aligned} H_0(x) &= -\frac{1}{2}\frac{\mathrm{d}^2}{\mathrm{d}x^2}+V(x), \\ H_t(x) &= -xE(t). \end{aligned}$$

The central part of the pseudospectral method[4] is to approximate the exact function $f(x)$ defined on the interval $[-1,1]$ by Nth-order polynomial $f_N(x)$,

$$f(x) \cong f_N(x) = \sum_{j=0}^{N} f(x_j) g_j(x), \tag{2}$$

and requires the approximation to be exact at the collocation points x_j: $f_N(x_j) = f(x_j)$. In the case of Legendre pseudospectral method x_j are the roots of the first derivative of the Legendre polynomial with respect to x, namely, $P'_N(x_j) = 0$. The $g_j(x)$ in Eq.(2) is defined as

$$g_j(x) = -\frac{1}{N(N+1)P_N(x_j)}\frac{\left(1-x^2\right)P'_N(x)}{x-x_j},$$

which satisfy

$$g_j(x_{j'}) = \delta_{jj'}.$$

We use the nonlinear algebraic mapping in Ref.[4] to map $[-X_{\max}, X_{\max}]$ to $[-1,1]$,

$$r(x) = R_0\frac{x}{1+\dfrac{R_0}{X_{\max}}-x^2},$$

which makes the grid dense near the nuclear while sparse outward. It is reasonable to describe the physical problem and reduce CPU time consumption.

Introduce $\Psi(r(x),t) = \sqrt{r'(x)}f(x)$, and the Hamiltonian becomes a symmetric system:

$$\begin{aligned} \widehat{H}(x,t) = &-\frac{1}{2}\frac{1}{r'(x)}\frac{\mathrm{d}^2}{\mathrm{d}x^2}\frac{1}{r'(x)} + V(r(x)) \\ &+V_m(r(x)) + H_t(r(x)), \end{aligned} \tag{3}$$

where

$$V_{\mathrm{m}}(r(x)) = -\frac{1.5\left(1+\dfrac{R_0}{X_{\max}}\right)}{\left(1+\dfrac{R_0}{X_{\max}}+x^2\right)^2[r'(x)]^2}.$$

Then, Eq.(1) can be discretized into

$$\begin{aligned} \mathrm{i}\frac{\partial}{\partial t}A_{j'} = &\sum_{j=0}^{N} H_{j'j}A_j = \sum_{j=0}^{N}\Bigg[-\frac{1}{2}D^{(2)}_{j'j} + \delta_{j'j}V(r(x_j)) \\ &+\delta_{j'j}V_m(r(x_j)) + \delta_{j'j}H_t(r(x_j))\Bigg]A_j, \\ &(j' = 1,...,N-1) \end{aligned}$$

with

$$D^{(2)}_{j'j} = \begin{cases} [r(x_{j'})]^{-1}\left[-\dfrac{2}{(x_{j'}-x_j)^2}\right][r(x_j)]^{-1}, & j' \neq j, \\ [r(x_j)]^{-1}\left[-\dfrac{N(N+1)}{3(1-x_j^2)}\right][r(x_j)]^{-1}, & j' = j, \end{cases}$$

$$A_j = r'(x_j)f(x_j)[P_N(x_j)]^{-1} = [r'(x_j)]^{\frac{1}{2}}\,\Psi(r(x_j),t)[P_N(x_j)]^{-1}, \tag{4}$$

A_j is complex. We write it in real plus imaginary part in vector form:

$$A = (a + \mathrm{i}b),$$

with

$$A = [A_1, ..., A_{N-1}]^{\mathrm{T}},$$
$$a = [a_1, ..., a_{N-1}]^{\mathrm{T}},$$
$$b = [b_1, ..., b_{N-1}]^{\mathrm{T}}.$$

Here the superscript T represents the matrix transposition, then Eq.(1) can be written in the form of canonical equation as follows:

$$\frac{\mathrm{d}}{\mathrm{d}t}\begin{bmatrix} a \\ b \end{bmatrix} = B(t)\begin{bmatrix} a \\ b \end{bmatrix},$$

where

$$B(t) = \begin{bmatrix} 0 & S(t) \\ -S(t) & 0 \end{bmatrix},$$
$$S(t)^{\mathrm{T}} = S(t),$$
$$S_{ij}(t) = H_{ij}(t).$$

$B(t)$ is a symplectic matrix.

Defining

$$\begin{pmatrix} a(t) \\ b(t) \end{pmatrix} = U(H,t)\begin{pmatrix} a(0) \\ b(0) \end{pmatrix}, \tag{5}$$

we can prove that $U(H,t)$ is the symplectic matrix and the Hamiltonian system is a single variable symplectic group.[9,21] Since symplectic geometry is the basis of all conservative Hamiltonian system, symplectic methods play an essential role in both theoretical and computational Hamiltonian mechanics. On the contrary, some conventional numerical methods, such as the classical Runge–Kutta scheme, are dissipative and unstable for long time evolution.[22] Then, it is natural to calculate the time evolution in symplectic algorithm for stable and reliable solution.

The implicit 2nd-order Euler-centre method in symplectic scheme is presented in our previous work,[14] Now we choose the 2nd-order explicit scheme:

$$b^1 = b^k, \quad a^1 = a^k + \frac{\tau}{2}S_{t+\frac{\tau}{2}}b^1,$$

$$b^{k+1} = b^1 - \tau S_{t+\frac{\tau}{2}}a^1, \quad a^{k+1} = a^1 + \frac{\tau}{2}S_{t+\frac{\tau}{2}}b^{k+1},$$

where τ is the time step, and the superscripts k and $k+1$ represent the values at t_k and t_{k+1}, respectively. Higher order symplectic schemes are treated in Ref.[9].

Then using symplectic algorithm presented above we can obtain $A(t)$ at arbitrary time. $\Psi(r(x),t)$ can be obtained from Eq.(4), Then, we compute the induced dipole moment in the acceleration form[16]

$$d_A(t) = -\left\langle \Psi(x,t)|\frac{\partial V(x)}{\partial x}|\,\Psi(x,t)\right\rangle + E(t). \tag{6}$$

The corresponding power spectrum can be obtained by the Fourier transformation of the time-dependent dipole moments

$$P_A(\omega) = \left|\frac{1}{t_{\mathrm{f}}-t_{\mathrm{i}}}\frac{1}{\omega^2}\int_{t_{\mathrm{i}}}^{t_{\mathrm{f}}} d_A(t)\mathrm{e}^{-\mathrm{i}\omega t}\mathrm{d}t\right|^2. \tag{7}$$

The power spectra of the dipole moment in length and acceleration form should be the same if the wavefunction $\Psi(x,t)$ is fully converged. However, the usual time-dependent calculations showed that the length and acceleration power spectra for higher order harmonics can be quite different. The latter is found to be generally superior to the former because the dominant contributions to the HHG arise from the wavefunctions at short distances in the acceleration-form calculations,[16] the detailed discussion on this issue can be referred to Ref.[23].

3. Numerical results and discussion

To illustrate the present numerical method, we choose the 1D soft Coulomb potential. The parameter is the same as previous published works[4,14,24,25] for comparison.

$$V(x) = -\frac{1}{\sqrt{1+x^2}}.$$

The form of laser field is as follows:

$$E(t) = \begin{cases} E_0 \sin^2\left(\dfrac{\pi t}{6T}\right)\sin(\omega t), & 0 \le t \le 3T, \\ E_0 \sin(\omega t), & t \ge 3T. \end{cases}$$

The amplitude of the laser field E_0=0.1 a.u., laser frequency ω=0.148 a.u., period $T = \dfrac{2\pi}{\omega}$, in our calculation, the propagation time is $16T$. We adopt R_0=200, $X_{\max}$=300 a.u., time step τ=0.001 a.u., grid points N=600 and an absorber[4] to remove the influence of the boundary. The acceleration form HHG spectra of this work and our previous one[14] are shown in Fig.1. They are almost the same, so this method is reliable. The B-spline with symplectic method is a global way to deal with the spatial part. We use 600

B-splines and the time step 0.1 a.u. This work is a local method, the grid points is not uniform, which is the reason why we have to use very short time step to preserve the symplectic structure. This method is also fast because of avoiding the calculation of the matrix elements, which is a slightly long time consumption in our previous B-spline method.

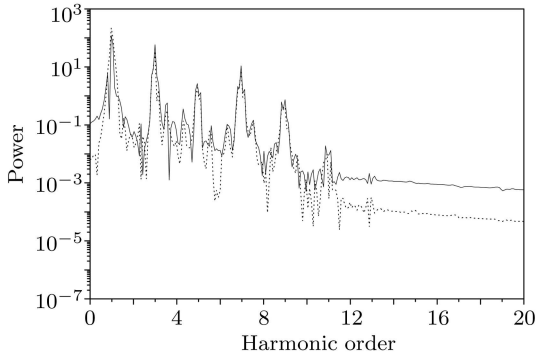


Fig.1. Harmonic generation spectra. The solid line is the result of pseudospectral with symplectic method; the dotted line is the result of B-splines with symplectic method.

Because of the inverse symmetry of the system, only the odd harmonics appear.[3] If we apply another static electric field to the system, the symmetry will be broken and the even harmonics will appear, even though the amplitude of the static field is several orders lower than that of the laser. We numerically solve the TDSE of the previous system by an addition of static electric field with strength 10^8V/m (2×10^{-4}a.u.), the result is shown in Fig.2, which is in good agreement with previously published results.[26,27]

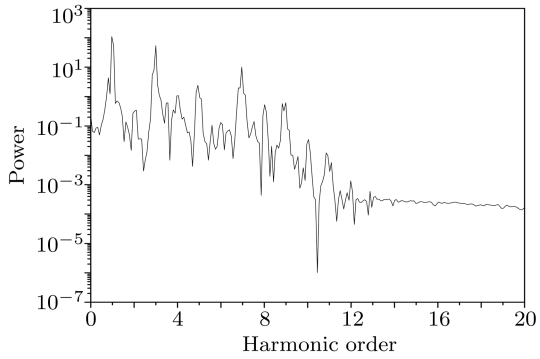


Fig.2. Harmonic generation spectra with an additional static electric field (10^8V/m).

4. Conclusions

While solving the TDSE, the pseudospectral is used to deal with the exact spatial part of the wavefunction by sparse grid, which reduces the memory consumption of computer; the symplectic method which preserves the symplectic structure of the Hamiltonian system in the long-time many-step calculations is stable and flexible to give high precision time evolution. The combination of them is a good way to solve the TDSE. The results show that it is reliable. Most of the calculation process is matrix and vector product, it is very easy to be realized by using BLAS library (basic linear algebra subroutines). The time consuming will be reduced greatly if parallel programming is used.

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