Spectral fitting method for the solution of time-dependent Schrödinger equations: Applications to atoms in intense laser fields

Haoxue Qiao,[1] Qingyu Cai,[1] Jianguo Rao,[1] and Baiwen Li[1,2]
[1]*Laboratory of Magnetic Resonance and Atomic and Molecular Physics, Wuhan Institute of Physics and Mathematics, The Chinese Academy of Sciences, Wuhan, 430071, People's Republic of China*
[2]*Chinese Center of Advanced Science and Technology (World Laboratory), P.O. Box 8730, Beijing, People's Republic of China*
(Received 6 February 2002; published 5 June 2002)

A spectral fitting method for solving the time-dependent Schrödinger equation has been developed and applied to the atom in intense laser fields. This method allows us to obtain a highly accurate time-dependent wave function with a contribution from the high-order term of $\triangle t$. Moreover, the time-dependent wave function is determined on a small number of discrete mesh points, thus making calculations simple and accurate. This method is illustrated by computing wave functions and harmonic generation spectra of a model atom in laser fields.

DOI: 10.1103/PhysRevA.65.063403 PACS number(s): 32.80.Wr, 32.80.Rm, 32.80.Qk, 42.50.Hz

I. INTRODUCTION

The multiphoton phenomena of atoms in intense laser fields have been observed in experiments [1–3]. The perturbative quantum theoretical approaches are unable to explain all these phenomena. Among those quantum nonperturbative methods, the early analytic methods can obtain some analytic or semianalytic results by using a simple Coulomb potential model or rough approximations [4–12], of course, the results are not very accurate. The time-independent non-Hermitian Floquet Hamiltonian method [13–15] is an important one of accurate nonperturbative basis set expansion approaches. In this method, solving the time-dependent Schrödinger equation is reduced to the solution of a set of coupled linear equations. But this approach can be used only for atoms in periodic fields, and cannot be used to treat atoms in chirped laser fields. Another basis set expansion approach is the close-coupling method [16–19,21–24] in which the time-dependent wave function is expanded in some time-independent basis set and the problem is reduced to solving the time-dependent expansion coefficients by a set of coupled first-order differential equations in time. This approach is suitable for laser fields with any time and space profile. However, it does require the evaluation of a large number of matrix elements, and solving a set of coupled differential equations, which makes the calculations complex when large numbers of basis sets are used. The Crank-Nicholson method is one of the nonperturbative direct numerical grid approaches [25,26]. Evaluation of matrix elements can be avoided by this method, but the accurate representation of a rapidly oscillating wave function requires dense grid points, thus computer memory and time consuming is tremendous. The *R*-matrix-Floquet method is another commonly used approach [27–32]. It possesses good numerical accuracy, but needs much memory and CPU consuming. Recently, a linear-least-squares-fitting method has been proposed [33,34]. In this method, the time-dependent wave function is expanded in a basis set, but the expansion coefficients are determined by the linear-least-squares fitting of the wave function on discrete mesh points [34], thus avoiding complex calculations of many matrix elements. Another simple grid points method is split the operator one, in which the time-dependent wave function is propagated along time on discrete grid points [35–40,42], thus allowing the evaluation of matrix elements to be avoided too. By using the pseudospectral split-operator method [41,42], the time-dependent wave function can be more accurately determined on sparser grid points than on the traditional split operator. Both the linear-least-squares fitting and pseudospectral split-operator methods possess advantages of numerical accuracy and computation efficiency. But the orders of accuracy of the two methods are $(\triangle t)^2$ and $(\triangle t)^3$, respectively. It requires much more complex calculations to consider contribution from the high-order term of $\triangle t$.

In this paper we introduce a spectral fitting method. In this method, the time-dependent wave function is propagated on discrete and far between grid points, thus avoiding the calculations of many matrix elements. By using the Taylor series method [20], the high accuracy of wave function with a contribution from the high-order item of $\triangle t$, is obtained with simple calculations.

II. METHOD

We illustrate this method by a one-dimensional atom [43] in a laser field. In the dipole approximation, the time-dependent Schrödinger equation in atomic units is

$$i\frac{\partial\Psi(r,t)}{\partial t}=[\hat{H}_0(r)+\hat{H}_t(r)]\Psi(r,t), \tag{1}$$

where H_0 and H_t are

$$\hat{H}_0(r)=-\frac{1}{2}\frac{d^2}{dr^2}-\frac{1}{\sqrt{r^2+1}}, \tag{2}$$

$$\hat{H}_t(r)=-E(t)r\sin(\omega t). \tag{3}$$

Generally, the wave function is considered in the range of $[-R_{\max},R_{\max}]$. In order to map this large domain

$[-R_{\max},R_{\max}]$ to $[-1,1]$, a suitable algebraic mapping for atomic structure calculations is given by the following form:

$$r=r(x)=R_0\frac{x}{1+\frac{R_0}{R_{\max}}-x^2}, \tag{4}$$

where R_0 is the mapping parameter. This mapping allows the dense grid points near the nucleus and sparse ones far from the nucleus, thus leading to a more accurate wave function and the use of a considerably smaller number of grid points than those of the equal-spacing grid method. But it is necessary to choose a suitable value of R_0. For the cases where both localized and very extended wave functions are involved, it is suitable to increase the value of R_0. Then by defining

$$\psi(x,t)=\frac{\Psi[r(x),t]}{\sqrt{r'(x)}}, \tag{5}$$

the time-dependent Schrödinger equation can be transformed into the following form:

$$i\frac{\partial\psi(x,t)}{\partial t}=[\hat{H}_0(x)+\hat{H}_t(x)]\psi(x,t), \tag{6}$$

$$\hat{H}_0(x)=-\frac{1}{2}\frac{1}{[r'(x)]^2}\frac{d^2}{dx^2}-\frac{1}{\sqrt{[r(x)]^2+1}} -\frac{1.5\left(1+\frac{R_0}{R_{\max}}\right)}{\left(1+\frac{R_0}{R_{\max}}+x^2\right)^2[r'(x)]^2}, \tag{7}$$

$$\hat{H}_t(x)=-E(t)r(x)\sin(\omega t+\varphi), \tag{8}$$

where $r'(x)=dr(x)/dx$. The following absorber:

$$f[r(x)]=\begin{cases}\cos^{1/8}\left[\frac{r(x)+R_0}{2(R_{\max}-R_0)}\pi\right], & r(x)\leqslant -R_A\\ 1, & -R_A<r(x)<R_A\\ \cos^{1/8}\left[\frac{r(x)-R_0}{2(R_{\max}-R_0)}\pi\right], & r(x)\geqslant R_A,\end{cases} \tag{9}$$

is used to filter out the wave function reaching the outward boundary. So, the boundary condition of the wave function is $\psi(-1,t)=\psi(1,t)=0$. For the normal spectral method [44,45], the wave function can be approximately expanded with N Legendre polynomials,

$$\psi(x,t)=C_0(t)+\sum_{i=1}^{N-1}C_i(t)P_i(x)+C_N(t)P_N(x). \tag{10}$$

If N is odd, under the boundary condition, $C_0(t)$ and $C_N(t)$ are expressed as

$$C_0(t)=-\sum_{i=1}^{N-1}\frac{1+(-1)^i}{2}C_i(t), \tag{11}$$

and

$$C_N(t)=-\sum_{i=1}^{N-1}\frac{1-(-1)^i}{2}C_i(t). \tag{12}$$

The expansion coefficients $C_i(t)$ are obtained by Gauss-Legendre-Lobatto quadrature

$$C_i(t)=\int_{-1}^{1}\frac{2i+1}{2}\psi(x,t)P_i(x)dx =\sum_{j=1}^{N-1}\frac{2i+1}{2}W_j\psi(x_j)P(x_j), \tag{13}$$

where W_j and x_j are integral weights and collocation points, respectively. x_j are zeros of $P'_N(x)$, and W_j possesses the following form:

$$W_j=\frac{2}{N(N+1)}\frac{1}{[P_N(x_j)]^2}. \tag{14}$$

Equation (10) is substituted by Eqs. (11), (12), (13), and (14), and the wave functions are expanded on collocation points,

$$\psi(x,t)=\sum_{j=1}^{N-1}f_j(x)\psi(x_j,t), \tag{15}$$

where $f_j(x)$ is the cardinal function given by

$$f_j(x)=\frac{1}{N(N+1)P_N(x_j)}\frac{(x^2-1)P'_N(x)}{x-x_j}, \tag{16}$$

and possesses the unique property on grid points $f_j(x_i)=\delta_{ij}$. In the polynomial approximation, the time-dependent Schrödinger equation can be written on grid points $\{x_m,m=1,N-1\}$ as

$$i\frac{\partial\psi(x,t)}{\partial t}\bigg|_{x=x_m}=\sum_{n=1}^{N-1}[\hat{H}_0(x)+\hat{H}_t(x)]f_n(x)\psi(x_n,t)\bigg|_{x=x_m}. \tag{17}$$

For the matrix $H^t_{mn}=\hat{H}_t(x)f_n(x)|_{x=x_m}=-E(t)r(x_m)\sin(\omega t)\delta_{mn}$ is diagonal; it can be defined as a vector $H^t_m=-E(t)r(x_m)\sin(\omega t)$, and the values of the nth derivative of the wave function on grid points can be expressed as a vector $\psi^{(n)}=[\partial^n\psi(x_1,t)/\partial t^n,\partial^n\psi(x_2,t)/\partial t^n,\ldots,\partial^n\psi(x_{N-1},t)/\partial t^n]$. Defining an operation matrix $H^0_{mn}=\hat{H}_0(x)f_n(x)|_{x=x_m}$, the matrix elements can be given analytically as

$$H^0_{mn}=\begin{cases}\dfrac{N(N+1)}{6(1-x_n^2)}\dfrac{1}{[r'(x_n)]^2}-\dfrac{1}{\sqrt{r^2(x_n)+1}}-\dfrac{1.5\left(1+\dfrac{R_0}{R_{\max}}\right)}{\left(1+\dfrac{R_0}{R_{\max}}+x_n^2\right)^2[r'(x_n)]^2}, & m=n\\ (-1)^{n+m}\sqrt{\dfrac{W_n}{W_m}}\dfrac{1}{(x_m-x_n)^2}\dfrac{1}{[r'(x_m)]^2}, & m\neq n\end{cases}. \tag{18}$$

Imposing the time-dependent Schrödinger equation, the derivatives of the wave function can be obtained by simply multiplying matrix and vector, using the following recurrence formula:

$$i\psi^{(1)}(x_m,t)=\sum_{n=1}^{N-1}H^0_{mn}\psi(x_n,t)+H^t_m\psi(x_m,t), \tag{19}$$

$$i\psi^{(2)}(x_m,t)=\sum_{n=1}^{N-1}H^0_{mn}\psi^{(1)}(x_n,t)+H^t_m\psi^{(1)}(x_m,t)+H^{t(1)}_m\psi(x_m,t), \tag{20}$$

$$i\psi^{(K)}(x_m,t)=\sum_{n=1}^{N-1}H^0_{mn}\psi^{(K-1)}(x_n,t)+H^t_m\psi^{(K-1)}(x_m,t)+\sum_{j=1}^{K-1}(K-j)H^{t(j)}_m\psi^{(K-j-1)}(x_m,t), \tag{21}$$

where $H^{t(n)}_m=d^nH^t_m/dt^n$ is the nth derivative of H^t_m. Therefore, the wave function at $t+\triangle t$ can be obtained by the Taylor series method [20],

$$\psi(x_j,t+\triangle t)=\sum_{n=0}^{K}\frac{1}{n!}\psi^{(n)}(x_j,t)(\triangle t)^n+O[(\triangle t)^{K+1}]. \tag{22}$$

III. RESULTS AND DISCUSSION

To illustrate the present method, we use the laser field profile

$$E(t)=\begin{cases}E_0\sin^2\left(\dfrac{\pi t}{6T}\right)\sin\omega t, & 0\leqslant t\leqslant 3T\\ E_0\sin\omega t, & t>3T,\end{cases} \tag{23}$$

where E_0 is the amplitude of the laser field, and $T=2\pi/\omega$. At first, we consider these laser field parameters: $E_0=0.1$ a.u and $\omega=0.148$ a.u. At this frequency, it takes about five photons to ionize the model atom from the ground states [34]. In our calculations, we take mapping parameters $R_0=200$, $R_{\max}=300$ a.u, and propagate the time to $t=16T$. In Fig. 1, we show the harmonic generation spectra obtained by 600 grid points. The peaks occur at the odd harmonic orders only, and are quite visible up to the 13th harmonic. This agrees with other numerical calculations [34] and classic conclusions [46,47]. In Fig. 2, we show the convergence of the wave functions. The agreement between wave functions obtained by using 400 and 600 grid points is quite good. For other laser field parameters: $E_0=0.08$ a.u and $\omega=0.06$ a.u, we take the mapping parameters $R_0=200$, $R_{\max}=200$ a.u. The harmonic generation spectra obtained by using 600 grid points are presented in Fig. 3. The peaks of harmonic generation spectra are cut off quickly at the 35th harmonic order. This agrees with classic conclusions [46,47] too.

Now we analyze the errors of our method. The errors stem from two aspects. The first one is space operation, such as approximate expansion presented in Eq. (10), and multiplying matrix and vector presented in Eqs. (19), (20), and (21). The second one is from the time propagation of the wave function presented in Eq. (22). The errors from different sources are defined as space error and time error, respectively. In the Legendre-polynomials-expansion approximation presented in Eq. (10), Eq. (13) is exact according to the properties of Gauss quadrature. Imposing the time-dependent Schrödinger equation, Eqs. (18), (19)–(21) are also exact; therefore, the space-error stems only from Legendre-polynomials expansion, and has nothing to do with other calculation processes. It makes the time-propagation calculations highly numerically stable and highly accurate. On the other hand, the time propagation of the wave function from t

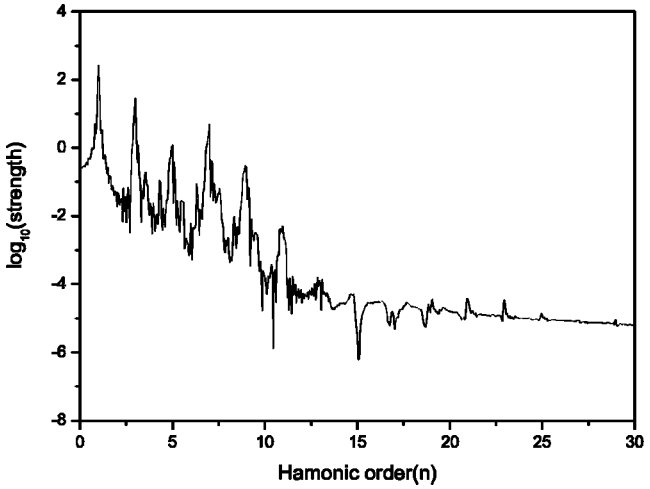

FIG. 1. Harmonic generation spectra obtained by using 600 grid points, with mapping parameters: $R_0=200$, $R_{\max}=300$ a.u., and laser field of strength $E_0=0.1$ a.u., $\omega=0.148$ a.u.; propagation time, $t=16T$.

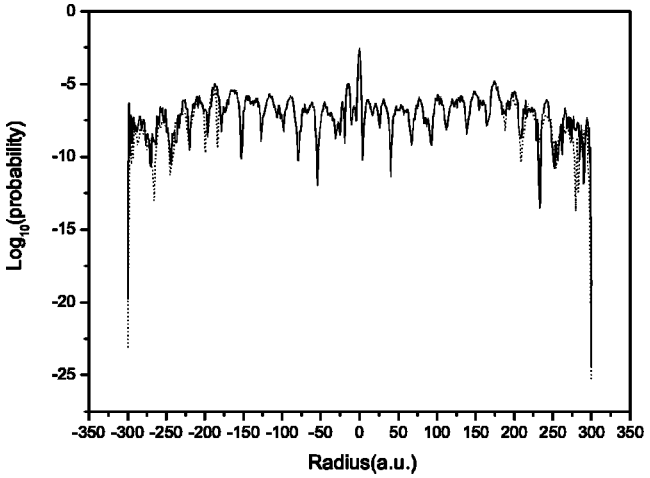

FIG. 2. Logarithm (to the base 10) probability for model atom in a laser field of strength $E_0=0.1$ a.u., $\omega=0.148$ a.u., and $t=16T$. The solid line presents the result obtained by using 600 grid points, and the dotted one presents the result obtained by using 400 grid points.

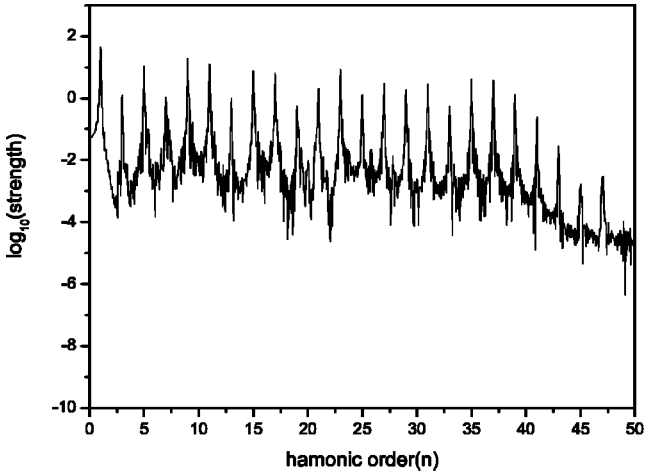

FIG. 3. Harmonic generation spectra obtained by using 600 grid points, with mapping parameters: $R_0=200$, $R_{\max}=200$ a.u., and laser field of strength $E_0=0.08$ a.u., $\omega=0.06$ a.u.; propagation time, $t=16T$.

to $t+\triangle t$ presented in Eq. (22), is an approximation of the Taylor series; therefore, the contribution from the high-order item of $\triangle t$ can be considered by using suitable K. This result in few time errors and a high accuracy of time-propagation calculations.

In summary, we have presented a spectral fitting method for solving the time-dependent Schrödinger equations. There are four advantages to our method: (a) the time-dependent wave function can be determined on sparse discrete grid points by using suitable mapping and Gauss quadrature collocation points, as is presented in Eqs. (5) and (13); (b) in the Legendre-polynomials-expansion approximation, the operation matrix can be exactly conducted by analytical methods, thus achieving numerical stability with little space error; (c) the contribution from the high-order term of $\triangle t$, which is difficult to obtain by other methods, can be easily obtained by using the Taylor series and a few multiplications of matrix and vectors, (d) our calculations are simple and less time consuming, because of the time propagation being reduced to a few matrix-vector products. We have compared the time consumption of the linear-least-squares-fitting method and our method. In our method, it takes about 15 min CPU to obtain the result presented in Fig. 1 by using 600 grid points and ''alpha500au'' workstation. But in linear least squares, it takes about 13 h CPU.

This method has been illustrated by calculations for a one-dimension problem, but it can be conveniently extended to a three-dimensional real atomic system. Detailed deductions and discussions are in progress.

[1] M. Gavrila, *Atoms in Intense Laser Fields*, edited by M. Gavrila (Academic, Boston, 1992), p. 435.
[2] K. Burnett, V.C. Reed, and P.L. Knight, J. Phys. B **26**, 561 (1993).
[3] M. Protopapas, C.H. Keitel, and P.L. Knight, Rep. Prog. Phys. **60**, 389 (1997).
[4] L.V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
[5] D.M. Volkov, Z. Phys. **94**, 250 (1934).
[6] F.H.M. Faisal, J. Phys. B **6**, L89 (1973).
[7] H.R. Reiss, Phys. Rev. A **22**, 1786 (1980).
[8] M.V. Ammosov, N.B. Delone, and Krainnov, Sov. Phys. JETP **64**, 1191 (1986).
[9] Chunlei Gou, Phys. Rev. Lett. **85**, 2276 (2000).
[10] M. Lewenstein, Ph. Balcou, M.Yu. Ivanov, Anne L. Huillier, and P.B. Corkum, Phys. Rev. A **49**, 2117 (1994).
[11] B. Wang, X. Li, and P. Fu, J. Phys. B **31**, 1961 (1998).
[12] M.W. Walser, C.H. Keitel, A. Scrinzi, and T. Brabec, Phys. Rev. Lett. **85**, 5082 (2000).
[13] R.M. Potvliege and R. Shakeshaft, Phys. Rev. A **38**, 4597 (1998).
[14] R.M. Potvliege and R. Shakeshaft, Phys. Rev. A **40**, 3061 (1989).
[15] D. Telnov, J. Wang, and S.-I. Chu, Phys. Rev. A **52**, 3988 (1995).
[16] W. Fritsh and C.D. Lin, Phys. Rep. **202**, 1 (1991).
[17] M. Kimura and N.F. Lane, Adv. At., Mol., Opt. Phys. **26**, 79 (1990).
[18] B. Sundaram and L. Armstrong, J. Opt. Soc. Am. B **7**, 414 (1990).
[19] S. Geltman, J. Phys. B **33**, 1967 (2000).
[20] Th. Mercouris, Y. Komninos, S. Dionissopoulou, and C.A. Nicolaides, Phys. Rev. A **50**, 4109 (1994).
[21] X. Tang, H. Rudolph, and P. Lambropoulos, Phys. Rev. Lett. **65**, 3269 (1990).
[22] S.M. Susskind and R.V. Jensen, Phys. Rev. A **38**, 711 (1988).
[23] Ph. Antoine, B. Piraux, and A. Maquet, Phys. Rev. A **51**, R1750 (1995).
[24] L.A. Collins and A.L. Merts, J. Opt. Soc. Am. B **7**, 647 (1997).
[25] K.C. Kulander, K.R. Sandhya, and S.E. Koonin, Phys. Rev. A **25**, 2968 (1982).

[26] W.H. Press, *Numerical Recipes in Fortran* (Cambridge University Press, Cambridge, England, 1992).
[27] E.P. Wigner, Phys. Rev. A **70**, 15 (1946); **70**, 606 (1946).
[28] M. Dorr, M. Terao-Dunseath, J. Purvis, C.J. Noble, P.G. Burke, and C.J. Joachain, J. Phys. B **25**, 2809 (1992).
[29] H.W. Van der Hart, J. Phys. B **29**, 2217 (1996).
[30] G. Gebarowski, P.G. Burke, K.T. Taylor, M. Dorr, M. Bensaid, and C. Joachain, J. Phys. B **30**, 1837 (1997).
[31] G. Gebarowski, K.T. Taylor, and P.G. Burke, J. Phys. B **30**, 2505 (1997).
[32] P.G. Burke, A. Hibbert, and W.D. Robb, J. Phys. B **4**, 153 (1971).
[33] E.Y. Sidky and C.D. Lin, J. Phys. B **31**, 2949 (1998).
[34] X. Zhou and C.D. Lin, Phys. Rev. A **61**, 053411 (2000).
[35] M.D. Feit, J.A. Fleck, and A. Steiger, J. Comput. Phys. **47**, 412 (1982).
[36] J.N. Bardsley, A. Szoke, and M.J. Comella, J. Phys. B **21**, 3899 (1988).
[37] T.F. Jiang and S.-I. Chu, Phys. Rev. A **46**, 7322 (1992).
[38] Q. Su, B.P. Irving, C.W. Johnson, and J.H. Eberly, J. Phys. B **29**, 5755 (1996).
[39] S.X. Hu and Z.Z. Xu, Phys. Rev. A **56**, 3916 (1997).
[40] L. Dimou and H.J. Kull, Phys. Rev. A **61**, 043404 (2000).
[41] X.M. Tong and S.-I. Chu, Chem. Phys. **217**, 119 (1996).
[42] X. Chu, S.-I. Chu, and C. Laughlin, Phys. Rev. A **64**, 013406 (2001).
[43] J. Javanainen, J.H. Eberly, and Q. Su, Phys. Rev. A **38**, 3430 (1988).
[44] J. Wang, S.-I. Chu, and C. Laughlin, Phys. Rev. A **50**, 3208 (1994).
[45] C. Canuto, M.Y. Hussaini, A. Quarteroni, and T.A. Zang, *Spectral Methods in Fluid Dynamics* (Springer, Berlin, 1988).
[46] P.B. Corkum, Phys. Rev. Lett. **71**, 1994 (1993).
[47] J.L. Krause, K.J. Schafer, and K.C. Kulander, Phys. Rev. Lett. **68**, 3535 (1992).