# Generalization of an efficient procedure for calculating the evolution of the wave function of a system interacting with a short laser pulse

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The wave-function-splitting technique which has been developed for localized potentials is generalized to long-range potentials. This technique allows for a significant reduction of the grid size without losing information. The method is demonstrated for a standard one-dimensional model and some practical details are discussed.

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## I. INTRODUCTION

In recent years interest has grown in the direct numerical solution of the time-dependent Schrödinger equation for systems interacting with a short and strong laser pulse [1]. This is so because this approach provides the most flexible and straightforward solution of the dynamics of the problem. Many of these methods are based on the introduction of a box for the underlying space coordinates with a subsequent transformation of the differential equation into a matrix equation. The most serious drawback of confining the system in a finite volume is that the wave function spreads during the evolution of the system and one has to prevent the reflection of the wave function from the boundaries. There are essentially two ways to deal with this problem. The first is to choose the box so large that the wave function does not hit the boundary during the evolution. The problem here is that the calculation is often prohibitively large and long. The second is to use absorbing boundary conditions or the wave-function-splitting technique [2, 3]. The problem here is that it is no longer possible to calculate the photoelectron spectrum after the pulse if the external part of the wave function is thrown away. However, it was shown by Heather and Metiu [2] that the spectrum can be reconstructed if the interaction with the laser is localized. The latter condition has prevented a general use of this very efficient technique. It is the aim of this article to show how to adapt this method to the general case of long-range potentials. As a specific example a standard one-dimensional model with the potential $V(x) = -1/\sqrt{1+x^2}$ [4] will be studied, but it should become clear that the technique can be used for any one-dimensional or three-dimensional model.

## II. THEORY

The procedure of Heather and Metiu [2] was based on the fact that the outer parts of the spreading wave function are purely outgoing describing ionization and do no longer interact with the field if the atom-laser interaction is localized to a region around the nucleus. The wave function can therefore be split into two parts: the internal part which has to be propagated by the full numerical procedure chosen and the external part which can be propagated analytically by using its energy representation which is obtained by a Fourier transform. The splitting can be applied several times during the pulse and the different external parts can be added together. But considering the general form of the time-dependent Schrödinger equation within the dipole approximation (atomic units are used throughout)

$$i\frac{\partial}{\partial t}\Psi(\mathbf{x},t) = [-\tfrac{1}{2}\Delta + V(\mathbf{x}) + \mathbf{E}(t)\cdot\mathbf{x}]\Psi(\mathbf{x},t), \tag{1}$$

where $\mathbf{E}(t)$ describes the laser pulse and $V(\mathbf{x})$ the atomic potential, there are two problems preventing a direct application of the procedure described above. First the interaction with the laser is not localized so that it seems impossible to define an external part which is propagating freely. This, however, is an artificial problem as it is well known that the electron has to be near the nucleus in order to exchange energy with the laser. The second problem is the fact that if $V(\mathbf{x})$ is long ranged, e.g., has a Coulombic tail, the Fourier transformation does not give the proper energy representation. So the Fourier transformation has to be replaced by the projection onto the energy eigenstates of the unperturbed Hamiltonian.

In order to proceed one first has to transform (1) into a form which makes visible the localization of the interaction. This is done by a transformation into the Kramers-Henneberger frame [5–7] which leads to the form

$$i\frac{\partial}{\partial t}\Psi_{\mathrm{KH}}(\mathbf{x},t) = [-\tfrac{1}{2}\Delta + V(\mathbf{x}+\boldsymbol{\alpha}(t))]\Psi_{\mathrm{KH}}(\mathbf{x},t), \tag{2}$$

where

$$\boldsymbol{\alpha}(t) = -\int^t d\tau \int^\tau d\tau'\, \mathbf{E}(\tau'). \tag{3}$$

The point is that $\alpha(t)$ is bounded if $\mathbf{E}(t)$ is bounded [8]. Denoting the maximum amplitude of $\alpha(t)$ by $\alpha_0$ we have that those parts of the wave function which have their support at $|\mathbf{x}| >> \alpha_0$ are propagating in the unperturbed potential. So the effective scale for the interaction with the laser is given by $\alpha_0$. It might, however, not be the only relevant scale. The condition that the outer parts of

 

the wave function are purely outgoing sets another scale due to the size of the relevant bound states participating in the interaction. Denoting the maximal relevant scale by $R$ allows to implement the program of Heather and Metiu [2] by splitting the wave function into an internal and an external part

$$\begin{aligned}\Psi_{\mathrm{KH}}(\mathbf{x},t) &= \Psi_{\mathrm{in}}(\mathbf{x},t) + \Psi_{\mathrm{ex}}(\mathbf{x},t),\\ \Psi_{\mathrm{in}}(\mathbf{x},t) &= f(\mathbf{x})\Psi_{\mathrm{KH}}(\mathbf{x},t),\\ \Psi_{ex}(\mathbf{x},t) &= [1-f(\mathbf{x})]\Psi_{\mathrm{KH}}(\mathbf{x},t),\end{aligned} \tag{4}$$

where $f(\mathbf{x})$ is a splitting function chosen such that it is 1 for $|\mathbf{x}| < R$ and 0 for $|\mathbf{x}| >> R$. The wave function at some later time $t' > t$ can thus be calculated by

$$\Psi_{\mathrm{KH}}(\mathbf{x},t') = U(t',t)\Psi_{\mathrm{in}}(\mathbf{x},t) + \sum_{\nu} e^{-iE_\nu(t'-t)} < \phi_\nu|\Psi_{\mathrm{ex}}(t) > \phi_\nu(\mathbf{x}), \tag{5}$$

where $U(t',t)$ is the full evolution operator to (2) and $\phi_\nu$ are the eigenfunctions of $-\Delta/2 + V(\mathbf{x})$ with energy $E_\nu$. The propagation of the internal part has to be performed numerically, but the size of the corresponding box can be adapted according to the choice of $f(\mathbf{x})$. The propagation of the external part requires the calculation and storage of the overlap integrals once; its contribution to later times is then obtained simply by the multiplication of appropriate phase factors.

## III. APPLICATION TO A ONE-DIMENSIONAL MODEL

In order to demonstrate the usefulness of the method described in the preceding section I implemented the splitting technique in a standard one-dimensional model with the potential $V(x) = -1/\sqrt{1+x^2}$ [4]. The form of the laser pulse was $E(t) = E_0 \sin^2(\pi t/t_p)\sin(\omega t)$ with a maximal field strength of $E_0 = 0.075$ a.u., a frequency of $\omega = 0.152$ a.u., and a total pulse length of $t_p = 50$ cycles.

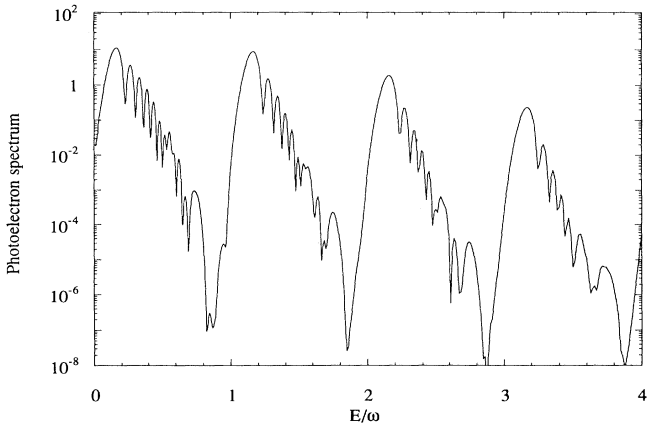


FIG. 1. The photoelectron spectrum for the one-dimensional $V(x) = -1/\sqrt{1+x^2}$ potential calculated with a box size of $\pm 3000$ a.u. The shape of the laser pulse was $E(t) = E_0 \sin^2(\pi t/t_p)\sin(\omega t)$ with $E_0 = 0.075$ a.u., $\omega = 0.152$ a.u., and $t_p = 50$ cycles. The initial state was the ground state.

These parameters led to a maximum amplitude of $\alpha(t)$ of $\alpha_0 = E_0/\omega^2 = 3.25$ a.u. Then Eq. (2) was solved in two different calculations both starting from the ground state and using a Crank-Nicholson algorithm. The first calculation, which was a standard calculation without the splitting technique, was done in a box with a maximum $x$ of 3000 a.u. requiring 40 000 grid points. This was large enough for the wave function not to hit the box boundaries during the 50-cycle evolution. At the end of the pulse the wave function was projected onto the unperturbed eigenstates to obtain the spectrum. In Fig. 1 the first four above-threshold ionization (ATI) peaks of this spectrum are shown. The dominant feature of it is the regular oscillations within each ATI peak. These are rainbow structures or Stueckelberg oscillations [9] caused by the interference of portions of the wavefunction ionized at different times in the pulse. They can be used as a nontrivial test of the splitting technique as the interference structures are very sensitive to a possible phase mismatch.

In the second calculation the splitting technique was used with a reduced grid size of 450 a.u. requiring 6000 points. So this calculation contained a saving in computer time of about a factor 7 compared to the previous calculation. The splitting function was chosen to be (for positive $x$)

$$f(x) = \begin{cases} 1 \text{ for } x < x_1 \\ \{1+\cos[\pi(x-x_1)/x_h]\}/2 \\ \text{for } x_1 \le x < x_1 + x_h \\ 0 \text{ for } x_1 + x_h \le x \le x_{\max} \end{cases} \tag{6}$$

and a similar function for negative $x$. A detailed discussion of how to choose the three parameters in (6) will be given in the next section. For the reconstruction of the first four ATI peaks of the spectrum 530 positive energy eigenstates were used and the time needed to calculate all overlap integrals was 2% of the total computer time. Figure 2 shows the reconstructed spectrum together with the contribution from the internal wave function only where

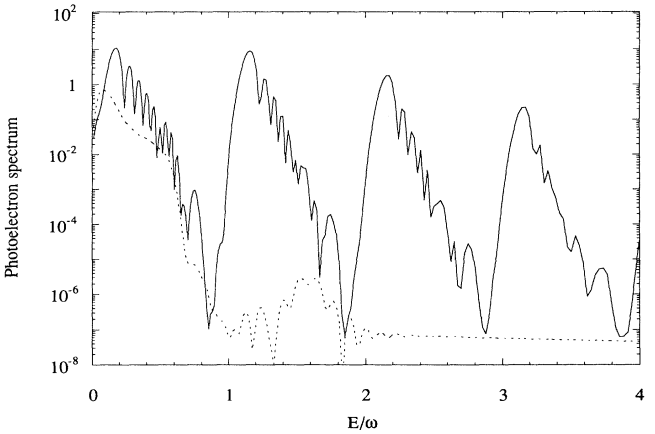


FIG. 2. The reconstructed photoelectron spectrum for the same system as in Fig. 1 calculated with a box size of $\pm 450$ a.u. and an internal region of the size $\pm 180$ a.u. (full line). The broken line indicates the contribution from the internal region only after the pulse.

the external parts are all thrown away. Comparing the full spectrum of Fig. 2 with Fig. 1 there is almost no difference between the two, especially since all Stueckelberg oscillations are reproduced correctly. The contribution from the internal wave function demonstrates that this spectrum is in fact dominated by the contributions from the external parts.

## IV. DISCUSSION

One of the biggest problems of the present implementation of the splitting technique is the need to calculate all overlaps of the external part of the wave function with the unperturbed eigenfunctions. This requires $n_E N$ multiplications per split where $n_E$ is the number of unperturbed eigenfunctions used and $N$ is the number of grid points needed. It is not only necessary to store this amount of data, but also the computer time needed to calculate this can easily exceed the time to propagate the wave function on a much larger grid, because most propagation algorithms (e.g., the Crank-Nicholson algorithm) are only proportional to the number of grid points per time step. The most efficient method to ensure that the overlap calculation does not dominate the computer time is to minimize the number of splits during the evolution. This can be achieved by introducing three zones as given in (6). In the third and external zone the internal part of the wave function is set to 0 after each split. In the subsequent evolution it spreads into this region, but it takes some time to reach the boundaries of the box again. The larger the external region, the longer it takes for the wave function to get to the boundaries and the smaller the number of splits required. So one has to compromize between the wish to reduce the grid size as much as possible and the need to minimize the number of splits. In the present calculation the size of the external zone was 160 a.u. which led to a total number of 27 splits. The time required to calculate the overlap integrals was therefore negligible compared to the time needed to calculate the 20 000 time steps of the total evolution.

The internal region which in (6) is given by $x_1$ has to be chosen according to two requirements. One is that it has to be much larger than $\alpha_0$. The other is due to the assumption that the external part of the wave function describes those parts of the system which ionize and which, therefore, do not return to the internal zone. So $x_1$ has to be large enough to accommodate all bound or resonance states which are important during the evolution of the system. In the present calculation the second requirement is stronger because $\alpha_0$ is relatively small. I varied $x_1$ between 180 and 300 a.u. without finding any differences in the results. The data shown in Fig. 2 were obtained with 180 a.u.

As discussed by Krause, Schafer, and Kulander [3] the form of the function applied in the intermediate zone [see (6)] should be as smooth as possible and the cutoff applied should not be too sudden. This is to prevent any reflections from that region. The value taken for the width of the intermediate zone was 100 a.u. and no attempt was made to systematically study the influence of this parameter on the result. But comparing this value to the value used by Heather and Metiu [2] it seems probable that it is possible to reduce it down to 10 or 20 a.u. if the requirements for computer time should be reduced further.

There is one final point which has to be discussed. In the calculation described in the preceding section 530 energy eigenstates were used to reconstruct the spectrum. But a grid of 6000 points only supports about 250 states in the same energy region. In order to increase the energy resolution the eigenstates had been created on a grid of 20 000 points with the same grid spacing. This is an equivalent procedure as the one used by Heather and Metiu [2] to increase the energy resolution. It might, however, lead to a problem. In using the Crank-Nicholson algorithm to propagate the wave function one implicitly diagonalizes the Hamiltonian at each timestep on the given grid. So the eigenfunctions used implicitly are different from the ones created on the larger grid, even in the external zone, especially since the eigenvalues are at a slightly different position. This can lead to a phase mismatch. There is no sign of this problem in the present calculation which might be due to the relatively small time periods over which the wave function is propagated. One should, however, check this point if the splitting technique is used for long propagation times.

## V. CONCLUSION

We have shown how to generalize the wave-function-splitting technique, originally developed by Heather and Metiu [2], for localized interactions to arbitrary potentials including the Coulomb potential. This technique allows for a significant reduction of the grid size without loss of the information necessary to calculate the final electron spectrum. The method was demonstrated in a one-dimensional model calculation and details for the implementation of this technique have been discussed.

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