Integral boundary conditions for the time-dependent Schrödinger equation: Superposition of the laser field and a long-range atomic potential

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We discuss long-range corrections for the integral boundary condition (IBC) introduced in A. M. Ermolaev, I. V. Puzynin, A. V. Selin, and S. I. Vinitsky, Phys. Rev. A **60**, 4831 (1999), in the case of the time-dependent Schrödinger equation with a long-range atomic potential. As in the work of Ermolaev *et al.* the laser-atom interaction is taken in the dipole approximation. The IBC techniques require the knowledge of the Green's function of the problem, beyond some surface σ remote from the atom. We consider the eikonal approximation (EA) for the Green's function in the asymptotic region and perform numerical tests on a one-dimensional problem with the soft Coulomb potential. We demonstrate that the account of long-range corrections, within the EA, allows us to reduce significantly the size of the space domain required for numerical integration and improves essentially on the accuracy of the computed spectral distribution for the ejected electrons.

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The numerical solution of the time-dependent Schrödinger equation (TDSE) for an atom interacting with an external time-dependent field is a powerful method particularly suitable for the case of short laser pulses. In our recent paper [1] we have suggested a general method for resolving the problems arising from the unphysical reflections (see, e.g., [2,3]) of the wave packet from the edge of a finite grid if the zero conditions are imposed there on the solution of the TDSE. The numerical example considered in [1] was a one-dimensional model atom with a short-range potential. It was shown there that the integral boundary condition (IBC) method allows for a substantial reduction of the grid size once an explicit account of the free Volkov propagator is made in the space outside the potential range.

The main purpose of the present paper is to discuss the implications for the formulated boundary conditions that may be caused by the presence of a long-range atomic potential. We note first that the integral condition used in [1] is a consequence of the Green's identity for the TDSE and in the three-dimensional case it is written in the form

$$\Psi(\mathbf{x}_\sigma,t)=2\int_{t_0}^{t}dt'\int_\sigma d\boldsymbol{\sigma}'\cdot\mathbf{j}'[\Psi(\mathbf{x}'_\sigma,t'),G_{as}(\mathbf{x}_\sigma,t;\mathbf{x}'_\sigma,t')]. \tag{1}$$

Here σ is a closed surface dividing the space $\mathbb{R}^3$ into an interior domain, with the atomic nucleus, and an exterior domain, $\mathbf{x}_\sigma\in\sigma$, and $G_{as}(\mathbf{x},t;\mathbf{x}',t')$ is a propagator that satisfies TDSE in the exterior domain. The vector $\mathbf{j}$ is a bilinear flux that for a pair of functions $\Psi(\mathbf{x},t)$ and $\Phi(\mathbf{x},t)$ has a form

$$\mathbf{j}[\Psi,\Phi]=\frac{i}{2}\{\Psi(\mathbf{x},t)\mathbf{D}^*_{\mathbf{x},t}\Phi(\mathbf{x},t)-\Phi(\mathbf{x},t)\mathbf{D}_{\mathbf{x},t}\Psi(\mathbf{x},t)\},$$

$$\mathbf{D}_{\mathbf{x},t}=\boldsymbol{\nabla}_{\mathbf{x}}-i\frac{e}{c}\mathbf{A}(\mathbf{x},t). \tag{2}$$

Here and below we use atomic units, $c\approx 137$ is light velocity, $e=-1$ is electron charge and $\mathbf{A}$ is the vector potential of the electric field. The prime at $\mathbf{j}$ in Eq. (1) means that the flux (2) is computed with Ψ and G_{as} taken as functions of the primed variables. It is assumed in Eq. (1) that the initial wave function $\Psi(\mathbf{x},t_0)$ is negligibly small beyond σ. Otherwise the term that takes into account the propagation by G_{as} of the tail of the initial wave function in the external region should be added to Eq. (1).

If the function Ψ is known on the boundary σ at times $t'<t$, then one can determine this function in the external region at time t with the help of G_{as} using the expression

$$\Psi(\mathbf{x},t)=\int_{t_0}^{t}dt'\int_\sigma d\boldsymbol{\sigma}'\cdot\mathbf{j}'[\Psi(\mathbf{x}'_\sigma,t'),G_{as}(\mathbf{x},t;\mathbf{x}'_\sigma,t')] \tag{3}$$

similar to one in Eq. (1). The term with $\mathbf{D}'G_{as}$ in Eqs. (1) and (3) represents the so-called parabolic potential of the double layer which is discontinuous on the surface σ [1]. This discontinuity accounts for an extra factor of two in Eq. (1).

The semiclassical approximation [4] will be used for G_{as} which is

$$G_{as}=(2\pi i)^{-n/2}\left|\det\left(\frac{\partial^2 S}{\partial x_i\partial x'_j}\right)\right|^{1/2}e^{iS(\mathbf{x},t;\mathbf{x}',t')}, \tag{4}$$

where n is the space dimension and S is the action evaluated along the classical path γ with the initial and final positions

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$\mathbf{x}'$ and $\mathbf{x}$, respectively. For such G_{as}, Eq. (1) assures the correct matching between the asymptotic semiclassical wave in the external domain and the solution in the interior region where it can be generated, e.g., by some numerical calculations.

For the case of a short-range potential considered in [1], the free Volkov propagator exactly satisfies the TDSE in the external region beyond the range of the potential and it can be used as G_{as}. The free Volkov propagator has a simple analytical representation convenient for implementing the boundary conditions (1) and has the form (4). The corresponding classical paths γ_0 for S are determined from the Hamilton equations for the canonically conjugate position $\mathbf{Q}$ and momentum $\mathbf{P}$,

$$\gamma_0: \ \dot{\mathbf{P}}(\tau)=0, \ \ \dot{\mathbf{Q}}(\tau)=\mathbf{P}-\frac{e}{c}\mathbf{A}(\tau). \tag{5}$$

In the case of a long-range potential $v_L(\mathbf{x})$ such as the Coulomb potential, the use of the free Volkov propagator in the integral (1) corresponds to cutting-off the potential tail beyond the surface σ and neglecting the phase distortion in the asymptotic region. As the exact form of the Coulomb-Volkov propagator is unknown we have to adopt an approximation. We shall use the fact that we need the semiclassical Green's function only in the asymptotic region. Hence it is natural to use the form (4) and to replace the exact classical paths γ for evaluating the action S, by the paths γ_0, Eq. (5). In this case, the action for the Hamiltonian

$$H(\mathbf{P},\mathbf{Q},\tau)=\frac{\mathbf{P}^2}{2}-\frac{e}{c}\mathbf{P}\mathbf{A}(\tau)+v_L(\mathbf{Q}) \tag{6}$$

along γ_0 differs from the action without $v_L(\mathbf{Q})$ by the long-range correction $Z[\gamma_0]$, that is

$$S=\frac{[\mathbf{x}-\boldsymbol{\xi}(t)-\mathbf{x}'+\boldsymbol{\xi}(t')]^2}{2(t-t')}-Z[\gamma_0], \tag{7}$$

where $\boldsymbol{\xi}(t)=-(e/c)\int^t\mathbf{A}(\tau)d\tau$, and

$$Z[\gamma_0]=\int_{t'}^{t}d\tau\, v_L(\mathbf{Q}(\tau)). \tag{8}$$

We note that the expression (7) is a solution of the Hamilton-Jacobi equation for S linearized in Z and is similar to the Glauber eikonal approximation [5]. The error of such an approximation is quadratic with respect to Z and is of order $(\nabla Z)^2$.

Setting the path density $|\det(\partial^2 S/\partial x_i\partial x_j')|\approx(t-t')^{-n}$, one can find that in the external region the eikonal approximation (EA) (4),(7) for G_{as} satisfies, at fixed time, the Schrödinger equation with the Hamiltonian (6) within an error of order $O((\nabla Z)^2+|\Delta Z|)$. Hence, for the Coulomb tail, the error is of order ϱ^{-4} in the three-dimensional case and ϱ^{-3} in the one-dimensional case, where ϱ is the closest distance between the path γ_0, connecting the points $\mathbf{x}$ and $\mathbf{x}'$, and the Coulomb center. This approximation for G_{as} loses accuracy for those γ_0 that lie close to the Coulomb center. However, the stationary phase analysis of action (7) shows that contributions to Eq. (1), which come from such γ_0, are small for sufficiently remote surfaces σ. Thus one can expect that the EA formulated above will be useful for the IBC provided that σ is not too close to the Coulomb center.

We now consider a one-dimensional atom where the long-range potential is modeled by the soft Coulomb potential $v_L(x)=-1/\sqrt{1+x^2}$ [6]. For the purpose of comparison with the advanced numerical method of Millack [3] where the wave-function-splitting technique and mask functions were employed (see also [2]) in order to reduce reflections from the edge of the grid, we shall solve the same TDSE as in [3] with the help of the IBC method.

Correspondingly, the laser pulse of duration T is chosen in the form

$$A(t)=\begin{cases} -\dfrac{c\mathcal{E}_0}{\omega}\sin^2\left(\dfrac{\pi t}{T}\right)\sin\omega t, & 0\leqslant t\leqslant T, \\ 0, \ \ t<0, \ \ t>T, \end{cases} \tag{9}$$

where $T=2\pi N/\omega$, N is the number of laser periods of angular frequency ω, $\mathcal{E}_0$ is the peak strength of the laser field, and $\xi_0=\mathcal{E}_0/\omega^2$ is the excursion amplitude of the electron.

We integrate numerically the TDSE with the parameters of the field $\mathcal{E}_0=0.075$ a.u., $\omega=0.152$ a.u., and $N=50$ periods taking the ground state as the initial state of the atom and applying a one-dimensional form of the integral boundary conditions (1).

For propagating the solution $\Psi(x,t)$ in the internal region, $-R\leqslant x\leqslant R$, we used the well-known Crank-Nicholson-Galerkin algorithm. The numerical procedures used in the present calculations as well as the finite-difference representation of the boundary condition (1a) were similar to those described in Ref. [1]. The present IBC calculations will be compared with the standard solution $\Psi_{\rm st}(x,t)$ of the same problem solved with the homogeneous Dirichlet boundary conditions, on a large integration grid $R_{\rm st}=3000$ a.u.

Let us turn now to the approximation for the Green's function G_{as}, Eq. (4), which uses the action (7). We note that for an arbitrary form of the function $\xi(t)$, the analytical evaluation of the integral in Eq. (8) is impossible and we need to develop an approximate method. The direct numerical calculation of the full distortion $Z[\gamma_0]$ for all t and t' required in Eq. (1) is impractical. A significant simplification can be achieved if one expands the functional $Z[\gamma_0]$ along a path γ_0 with the initial and final points to be taken as $x=x'=\pm R$ in the inverse powers of R. Thus

$$Z[\gamma_0]|_{x=x'=\pm R}=(t-t')\left\{-\frac{1}{R}\mp\frac{d(t,t')}{R^2}+\cdots\right\}, \tag{10}$$

where

$$d(t,t')=\frac{\xi(t)+\xi(t')}{2}-\frac{1}{t-t'}\int_{t'}^{t}d\tau\xi(\tau). \tag{11}$$

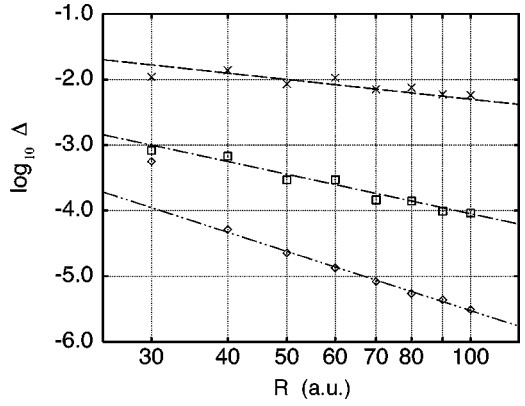

FIG. 1. The error Δ due to the approximations (10) made in the IBC, shown as a function of the grid size R. The error is defined as $\Delta=\max_t|\Psi_{\rm st}(\pm R,t)-\Psi(\pm R,t)|$, where $\Psi_{\rm st}$ is the solution of TDSE obtained on the large grid $R_{\rm st}=\pm 3000$ a.u. The crosses ($\times$) correspond to the short-range version of IBC (a); the boxes ($\Box$) correspond to version (b), and the diamonds ($\diamond$) correspond to version (c) of the IBC. The straight lines on the graph represent the overall uncertainty Δ which depends on R and on the approximation mode k, as R^{-k},$k=1,2,3$.

One can treat Eq. (10) as a multipole expansion in terms of a small parameter ξ_0/R.

Then we apply the EA for G_{as} at three different levels of accuracy. Namely, (a) with the distortion term $Z[\gamma_0]$ omitted from Eq. (7); (b) with only the first term in the expansion (10) retained in $Z[\gamma_0]$; and (c) with both expansion terms of (10) kept in $Z[\gamma_0]$. Case (a) is the crudest one because the potential tail is completely neglected beyond σ and the solution propagates in the potential-free space. In the cases (b) and (c), distortion in the external region is accounted for.

Figure 1 displays the maximum absolute error Δ in the IBC solution as obtained from the comparison with $\Psi_{\rm st}$ for three approximate forms of the boundary condition (1) [cases (a)–(c), see Eq. (10)]. It is clearly seen that the error depends on the grid size R as an inverse power of R, according to the approximation in question. In case (a), i.e., for the Coulomb tail neglected in the external region, $\Delta\sim R^{-1}$ at fixed time. As follows from the discussion above, for the Green function G_{as} in the approximation (4)–(7), $\Delta\sim|\partial^2 Z/\partial x^2|\sim R^{-3}$. Then it is clear that higher-order terms in the expansion (10) should be neglected. The numerical results of case (c) are in accord with this conclusion. For comparison, we note that in [3] the same level accuracy in the wave function requires a grid of $R=\pm 450$ a.u. which is larger by an order of magnitude than one sufficient in case (c) of the IBC method.

For obtaining the spectral distribution of atomic electrons ejected by the laser field, we need to know the wave function in a sufficiently large space domain. This is particularly true for the long-range atomic potentials of the present case. The method we are considering is completely self-consistent as the solution in the external region (extending into infinity) is constructed with the help of the solution in the internal region, by applying the extension relation (3).

The spectral distribution $w(E)$ of the electrons ejected with energy E is given as a sum, thus

$$w(E)=\frac{1}{2\pi}\sum_{s=1}^{2}|\langle\psi_{1,2E}^{(-)}|\Psi\rangle|^2, \tag{12}$$

over two directions of the final momenta, $k=\pm\sqrt{2E}$. The projection of the wave function Ψ is made onto eigenstates $\psi_{1,2E}^{(-)}$ of the continuum for the stationary problem, with the turned-off electric field.

The direct application of the relation (3) for extending the solution into the external region, as required in (12), will not be considered here [7]. Instead, we shall use the fact that at large distances the solution has a semianalytical form. Then we can apply the method of the stationary phase for evaluating the overlap integrals in Eq. (12), in order to obtain the leading part, in terms of the Planck's constant $\hbar$, of the expansion for the probability density $w(E)$.

The asymptotic behavior of $\psi_{1,2E}^{(-)}$ is given by a linear combination of the WKB solutions $|\partial^2 F/\partial x\partial k|^{1/2}e^{\pm iF(x,k)}$, where $F(x,k)=\int^x p(z)dz$ is the abbreviated action and the classical momentum $p(x)$ is given by $[k^2-2v_L(x)]^{1/2}$. An evaluation of the amplitudes $\langle\psi_{1,2E}^{(-)}|\Psi\rangle$ where Ψ is continued into the external region via relations (3) and (4) with the help of the stationary phase method, yields the following result:

$$\langle\psi_{1,2E}^{(-)}|\Psi\rangle\sim\int_{t_0}^{t}dt'\,j[\Psi(x',t'),\hat{G}_{as}^{\pm}(k,t;x',t')]_{x'=\pm R}, \tag{13}$$

where

$$\hat{G}_{as}^{\pm}(k,t;x',t')=\left|\frac{\partial^2 W^{\pm}}{\partial x'\partial k}\right|^{1/2}e^{iW^{\pm}(k,t;x',t')}, \tag{14}$$

The function $W(k,t;x',t')$ is defined as $S(x^0,t;x',t')\mp F(x^0,k)$ taken at the stationary point x^0, where S is assumed to be the exact form of the action. This function W can be obtained as a solution of the following Cauchy problem for the Hamilton-Jacobi equation [8]:

$$-\frac{\partial W^{\pm}}{\partial t'}+H\left(-\frac{\partial W^{\pm}}{\partial x'},x',t'\right)=0, \tag{15}$$

$$W^{\pm}(k,t;x',t'=t)=\mp F(x',k). \tag{16}$$

An approximate solution of the problem (15) accurate uniformly with respect to k within to $O(v_l'')$, can be written in the following form:

$$W^{\pm}(k,t;x',t')=-\frac{k^2}{2}(t-t')\mp F(x'-\xi(t')+\xi(t),k)+v_l'(x')\int_{t'}^{t}\{\xi(t)-\xi(\tau)\}d\tau+\cdots. \tag{17}$$

The IBC spectral distribution $w(E)$ [case (c)] evaluated via Eqs. (13)–(17) is shown in Figs. 2 and 3, together with the distribution $w_{\rm st}(E)$ directly obtained by projecting the

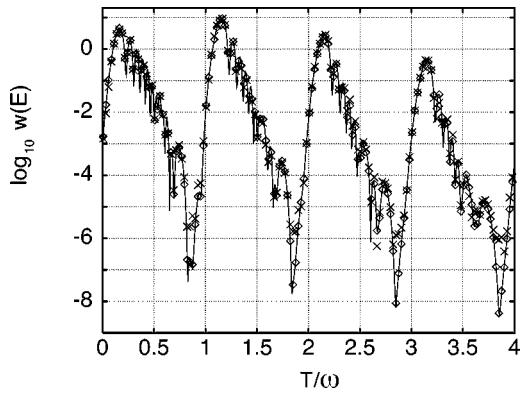

FIG. 2. The logarithm of the probability density $w(E)$ of ejected photoelectrons as a function of energy E (in units of ω). The solid curve $w_{st}(E)$ is obtained by projecting the soft Coulomb eigenstates of the solution Ψ_{st} evaluated on the large grid R_{st}. The crosses ($\times$) correspond to applying the short-range version (a) of IBC as in Ref. [1], and the diamonds ($\diamond$) correspond to version (c) of the IBC for $R=50$ a.u. with $w(E)$ evaluated by the stationary phase method via Eqs. (13)–(17), using the same grid size $R=50$ a.u.

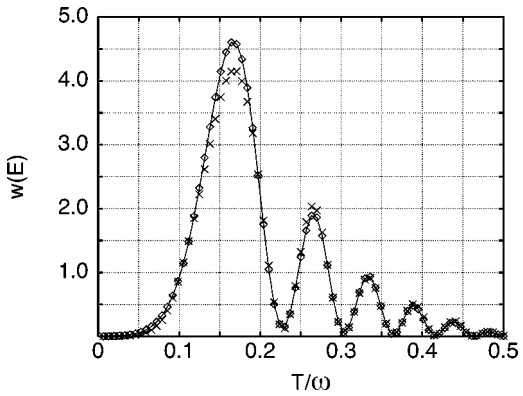

FIG. 3. The same as Fig. 2, but using a large scale to display the details of the first peak of the series. A ''rainbow'' structure that had been earlier discussed for a short-range potential in Ref. [9] and for a ''soft Coulomb'' potential in Ref. [10] is clearly seen in the declining slope of the peak.

standard solution Ψ_{st} onto the scattering eigenstates. Both agree with each other within 0.1% when the IBC is placed at $R=50$ a.u. At the same time, using the short-range version (a) of the IBC solution gives $w(E)$ accurate within some 10% only.

In conclusion, we have successfully aplied the IBC to treat numerically the TDSE with a long-range one-dimensional atomic potential and the atom-laser interaction. The main approximation introduced in the IBC was an asymptotic form of the G_{as} based on the EA. The numerical method (c) using the expansion (10) is shown to be very effective for relatively short-time integration intervals (50 laser periods in the present calculation). For much longer intervals, the method may need some modifications. These questions as well as the extension to the three-dimensional case will be considered separately.

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