Time-dependent multiphoton dynamics in intense short-pulse laser fields.
Interaction-representation approach

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We present an interaction-picture time propagation method for the numerical solution of the time-dependent Schrödinger equation in intense and superintense short-pulse laser fields. The method is tested on the problem of multiphoton and above-threshold ionization (MPI/ATI) of a model Cl⁻ ion. It is seen that the interaction-picture approach leads to localized strong-field wavepackets, allowing the elimination of the boundary reflection problem as well as efficient and accurate treatment of the MPI/ATI dynamics and spectra.

1. Introduction

Development of time-dependent methods for numerical solution of the time-dependent Schrödinger equations is a subject of much current interest in atomic, molecular, optical, and nuclear physics.¹ In this Letter, we address the problem of the nonlinear response of atoms and molecules to intense or superintense short-pulse laser fields.² In such strong fields, multiphoton and above-threshold ionization (MPI/ATI) of atoms or multiphoton and above-threshold dissociation (MPD/ATD) of molecules can take place. One known technical problem associated with the numerical solution of the Schrödinger equation in intense fields is that the wavepacket spreads and translates as it evolves. The spreading and translation of the wavepacket results in the use of an extensive grid in the dissociative coordinate and also makes it necessary the implementation of some ad hoc procedures (such as placing an absorber at the edge of the grid boundary) to avoid spurious reflection. Such procedures may not be ideal for MPI/ATI (or MPD/ATD) problems since the ionized (or dissociated) portion of the wavepackets (which move away from the interaction region rapidly and hit the grid boundary) are essential for analyzing the ATI(ATD) spectra. It is desirable to work in a framework which is free from these problems. In the following section, we discuss a new procedure based on the extension of the idea of the time propagation of the wavefunction in the interaction representation.

2. Interaction-picture time propagation method

The interaction picture has been recently found to be useful for the study of semiclassical wavepacket dynamics and quantal treatment of potential scattering. All these previous studies involve a time-independent Hamiltonian \( \hat{H} \). In this case, one first sets \( \hat{H} = \hat{H}_0 + V_m \), where the choice of \( \hat{H}_0 \) is usually (but not necessarily) the kinetic energy operator \( \hat{T} \). The interaction-picture wavefunction \( \psi_i \) is defined in terms of the Schrödinger wavefunction \( \psi_S \) by

\[
\psi_i(t) = \exp(iH_0 t) \exp(-iHt) \psi_S(t=0).
\]
The interaction wavefunction satisfies the following equation,

\[ \frac{\partial}{\partial t} \psi_i(t) = H_i(t) \psi_i(t), \tag{2} \]

where the interaction Hamiltonian is given by

\[ H_i(t) = \exp(iH_0t) \psi_{int} \exp(-iH_0t). \tag{3} \]

Eq. (1) shows that the interaction wavefunction can be viewed as a forward propagation in time under the full Hamiltonian \( H \), followed by a backward propagation in time under \( H_0 \). Thus the interaction wavefunction generally is more localized and compact, resulting in a more compact grid. We suggest below the development of a new time-dependent propagation technique in interaction representation for a time-dependent Hamiltonian \( H(t) \) suitable for the study of MPI/ATI processes in intense and su-

Fig. 1. Time-development of the wavefunction in the Schrödinger representation (\( \psi_S \)) for a model Cl\(^+\) ion subject to a laser field with intensity \( I = 7 \times 10^{14} \) W/cm\(^2\) and wavelength \( \lambda = 532 \) nm. The laser field is turned on with a 5-cycle smooth ramp. It is seen that wavepackets spread and translate, hit the grid boundary and reflect back.

Fig. 2. Time-development of the wavefunction in the interaction representation (\( \psi_I \)) for the same system and laser parameters considered in fig. 1. It is seen that the wavepackets become localized after about 20 optical cycles.
perintense short-pulse laser fields. The method can
be extended to MPD/ATD processes of molecules as
well with simple modification.

The Hamiltonian, in velocity gauge, can be written
as
\[
\hat{H}(t) = \left[ \hat{P} - eA(t) \right]^2 / 2\mu + V_c,
\]
where \( A(t) \) is the vector potential, and \( V_c \) the atomic
potential. Several possible choices of \( \hat{H}_0 \) can be made
depending upon physical conditions being consid-
ered. For very intense laser fields, an appropriate
choice is the time-dependent field-driven free-electron Hamiltonian,
\[
\hat{H}_0(t) = \left[ \hat{P} - eA(t) \right]^2 / 2\mu,
\]
whose eigenfunctions are the Volkov states, see, for example, ref. [6]. In this case, the interaction
Hamiltonian becomes
\[
\hat{H}_I = \exp[i\hat{Q}(t)] V_c(x) \exp[-i\hat{Q}(t)],
\]
where
\[
\hat{Q}(t) = \left( \hat{P}^2 / 2\mu \right) t - \hat{P} \cdot \alpha(t),
\]
and
\[
\alpha(t) = \frac{e}{\mu} \int_0^t A(t') dt'.
\]

The interaction wavefunction \( \psi_I \) is now related to the
Schrödinger wavefunction \( \psi_S \) by the equation
\[
\psi_S(t) = \exp[-i(\hat{P}^2 / 2\mu)t + i\alpha(t) \cdot \hat{P} - i\pi(t)] \psi_I(t),
\]
where
\[
\pi(t) = \frac{e^2}{2\mu} \int_0^t A^2(t') dt'.
\]

The time-dependent Schrödinger equation, eq. (2),
can now be solved in the interaction representation,
using eq. (6).

To test the usefulness of this idea, we have per-
formed a preliminary study of the MPI/ATI process
of a model negative ion described by the Gaussian
potential [7,8] \( \hat{V}_c = -V_0 \exp(-x/x_0)^2 \). We use
\( V_0 = 0.27035 \) au, and \( x_0 = 2.0 \) au to mimic the Cl-
ion [8]. Fig. 1 shows the time development of the
wavefunction, obtained directly in Schrödinger rep-

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ion [8]. Fig. 1 shows the time development of the
wavefunction, obtained directly in Schrödinger rep-
representation, for \( t = 0, 5, 20, \) and 30 optical cycles. The field parameters used are \( I = 7 \times 10^{14} \text{ W/cm}^2 \) and \( \lambda = 532 \text{ nm} \), and the laser field is turned on using 5-cycle smooth ramp and held constant in amplitude afterwards. It is seen that the ionized wavepackets move apart from the nucleus rapidly, hit the grid boundary and reflect back. The corresponding wavefunction in the interaction representation shown in fig. 2, on the other hand, shows some initial spatial development in the interaction region but then becomes completely localized after about 20 optical cycles. (In the interaction representation, it can be shown that the Volkov-state wavefunctions are completely localized.) An essential feature in this interaction-representation approach is that all the information about the electron dynamics is accurately treated and preserved in \( \psi_i \). For example, the ATI spectrum can be extracted directly from \( \psi_i \) as shown in fig. 3. Because of the spatial localization in the interaction-representation approach, a much smaller range of coordinate space needs to be considered, allowing orders of magnitude enhancement of the computational speed as well as the complete elimination of the boundary reflection problem.

The proposed procedure described above is expected to work best for superintense laser pulse fields, where the choice of \( \hat{H}_0 \) in eq. (5) is optimal and the laser-atom interaction time is brief. For less stronger fields, a different choice of \( \hat{H}_0 \) (such as the unperturbed atomic or molecular Hamiltonian) may be more appropriate. Extension of this work to 3D real systems is underway.

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References