Nuclear Instruments and Methods in Physics Research B xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Multiphoton ionization of many-electron atoms and highly-charged ions in intense laser fields: a relativistic time-dependent density functional theory approach

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ARTICLE INFO

Article history:
Received 15 November 2016
Received in revised form 25 February 2017
Accepted 4 April 2017
Available online xxxx

Keywords:
Highly charged ions
Intense laser fields
Ionization

ABSTRACT

We develop an efficient numerical implementation of the relativistic time-dependent density functional theory (RTDDFT) to study multielectron highly-charged ions subject to intense linearly-polarized laser fields. The interaction with the electromagnetic field is described within the electric dipole approximation. The resulting time-dependent relativistic Kohn–Sham (RKS) equations possess an axial symmetry and are solved accurately and efficiently with the help of the time-dependent generalized pseudospectral method. As a case study, we calculate multiphoton ionization probabilities of the neutral argon atom and argon-like xenon ion. Relativistic effects are assessed by comparison of our present results with existing non-relativistic data.

1. Introduction

Recent progress of laser technologies requires development of theoretical methods for study of the different processes in presence of a strong laser field. Heavy highly charged ions are ones of the most interesting objects, which can be studied in this case, since they have their own strong Coulomb field. The motivation of our research relates to the HILITE (High-Intensity Laser Ion-Trap Experiment) setup constructed at GSI (Darmstadt, Germany). The goal of these experiments is to study the interaction of atoms and ions confined in a Penning trap with highly intense laser field [1,2].

Since most of the experimental targets are many-electron systems, one has to take into account multielectron effects; for heavy atoms and highly-charged ions, relativistic description is also important. In this work, we make use of the relativistic time-dependent density functional theory (RTDDFT; for example, see [3] and references therein) to study relativistic dynamics of multielectron ions exposed to strong laser fields. In principle, time-dependent density functional theory provides an exact description of multielectron systems if an exact exchange–correlation functional is used. However, this functional is unknown, and only approximations are available for practical calculations. In our present work, we utilize adiabatic non-relativistic forms of the exchange–correlation potential. Adiabatic approximation means that the exchange–correlation potential at some specific time depends on the electron density at the same time only; this is well-justified in the case of low-frequency fields [4]. Non-relativistic form of the exchange–correlation potential means that the parameters of the potential have been adjusted in the non-relativistic density functional calculations. Nevertheless, such an approximation allows description of relativistic effects if relativistic electron density is used to evaluate the functional. Here we propose a numerical implementation of RTDDFT based on the time-dependent generalized pseudospectral (TDGPS) method in spherical coordinates (see [5] and references therein), which allows for accurate and efficient time propagation of the wave functions. The interaction with the external electromagnetic field is described within the electric dipole approximation, which is well justified for the laser field parameters used in the calculations. As a case study, we calculate multiphoton ionization probabilities of the argon atom and argon-like xenon ion exposed to strong laser fields. Atomic units ($\hbar = e = m = 1$) are used throughout the paper unless specified otherwise.

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http://dx.doi.org/10.1016/j.nimb.2017.04.018
0168-583X/

2. Theory

The basic equations of RTDDFT are the relativistic Kohn–Sham (RKS) equations for the one-electron orbitals $\Psi_k(\mathbf{r},t)$

$$i\frac{\partial}{\partial t}\Psi_k(\mathbf{r},t) = H(t)\Psi_k(\mathbf{r},t), \quad k = 1\ldots N, \tag{1}$$

where the relativistic Hamiltonian operator is defined as follows:

$$H(t) = c\boldsymbol{\alpha}\cdot\mathbf{p} + c^2(\beta - I) + V_{\text{eff}}(\mathbf{r},t) + V_{\text{ext}}(\mathbf{r},t). \tag{2}$$

Here $\boldsymbol{\alpha},\beta$ are the Dirac matrices and I denotes the unit 4×4 matrix, c is the speed of light, N is the number of electrons, $V_{\text{eff}}(\mathbf{r},t)$ is the effective single-particle potential of RTDDFT, and $V_{\text{ext}}(\mathbf{r},t)$ describes the interaction with the laser field. Using the electric dipole approximation and the length gauge, the potential $V_{\text{ext}}(\mathbf{r},t)$ can be expressed as

$$V_{\text{ext}}(\mathbf{r},t) = \mathbf{F}(t)\cdot\mathbf{r}. \tag{3}$$

Here $\mathbf{F}(t)$ is the electric field strength of the laser field. In the present study, we use linearly-polarized laser pulses with the sine-squared envelope:

$$\mathbf{F}(t) = \mathbf{F}_0 \sin^2\frac{\pi t}{T}\sin\omega_0 t, \tag{4}$$

where T and ω_0 denote the pulse duration and the carrier frequency, respectively; F_0 is the peak field strength.

The effective single-particle potential $V_{\text{eff}}(\mathbf{r},t)$ can be represented as a sum of three different contributions:

$$V_{\text{eff}}(\mathbf{r},t) = V_{\text{nucl}}(\mathbf{r}) + V_{\text{H}}(\mathbf{r},t) + V_{\text{xc}}(\mathbf{r},t). \tag{5}$$

Here $V_{\text{nucl}}(\mathbf{r})$ is the Coulomb interaction of the electron with the nucleus,

$$V_{\text{nucl}}(\mathbf{r}) = -\frac{Z(r)}{r}, \tag{6}$$

with $Z(r)$ being the effective nuclear charge (in this work we use Fermi distribution of the nuclear charge density [6] with parameters from [7]); $V_{\text{H}}(\mathbf{r},t)$ is the Hartree potential due to electron–electron repulsion,

$$V_{\text{H}}(\mathbf{r},t) = \int\frac{\rho(\mathbf{r}',t)d\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}, \tag{7}$$

where $\rho(\mathbf{r},t)$ is the total electron density:

$$\rho(\mathbf{r},t) = \sum_{k=1}^{N}\Psi_k^{\dagger}(\mathbf{r},t)\Psi_k(\mathbf{r},t). \tag{8}$$

The remaining term $V_{\text{xc}}(\mathbf{r},t)$ is the exchange–correlation potential. Its exact form is unknown but various good-quality approximations are available. In this work, we use an approximate exchange–correlation potential LB94 [8]. Originally, this potential was constructed for use with non-relativistic spin-polarized atomic systems. In our case (relativistic closed-shell atom or ion), it can be expressed as a functional of the total electron density:

$$V_{\text{xc}}(\mathbf{r},t) = V_{\text{x}}^{\text{LDA}}(\mathbf{r},t) + V_{\text{c}}^{\text{LDA}}(\mathbf{r},t) - \frac{bx^2(\mathbf{r},t)\left[\frac{1}{2}\rho(\mathbf{r},t)\right]^{1/3}}{1+3bx_\sigma(\mathbf{r},t)\ln\left\{x_\sigma(\mathbf{r},t)+\sqrt{1+x_\sigma^2(\mathbf{r},t)}\right\}}, \tag{9}$$

where $x_\sigma(\mathbf{r},t) = 2^{1/3}|\nabla\rho(\mathbf{r},t)|/\rho^{4/3}(\mathbf{r},t)$, $V_{\text{x}}^{\text{LDA}}(\mathbf{r},t)$ is the exchange potential in the local density approximation (LDA):

$$V_{\text{x}}^{\text{LDA}}(\mathbf{r},t) = -\left[\frac{3}{\pi}\rho(\mathbf{r},t)\right]^{1/3}, \tag{10}$$

and $V_{\text{c}}^{\text{LDA}}(\mathbf{r},t)$ is the correlation LDA potential. The latter potential has no simple analytic representation; we make use of numerical subroutines for the Perdew-Burke-Ernzerhof (PBE) functional [9] based on the representation of the correlation energy and potential by Perdew and Wang [10]. In Eq. (9), the parameter b has a value of 0.05; it was adjusted to fit the exact exchange–correlation potential for the beryllium atom [8]. As our results show, this potential is also quite accurate in calculations of the electronic structure of the argon atom.

3. Method

To solve the time-dependent RKS Eqs. (1), one has to supply the initial values for the orbitals $\Psi_k(\mathbf{r},t)$ and effective potential $V_{\text{eff}}(\mathbf{r},t)$. At $t=0$ (just before the laser pulse), these quantities can be obtained as solutions of the time-independent RKS equations

$$H_0\Phi_k(t=0) = E_k\Phi_k(t=0), \quad k = 1\ldots N, \tag{11}$$

where E_k are the orbital energies, and the Hamiltonian H_0 does not include the interaction with the laser field:

$$H_0 = c\boldsymbol{\alpha}\cdot\mathbf{p} + c^2(\beta - I) + V_{\text{eff}}(t=0). \tag{12}$$

The set of Eqs. (11) is solved self-consistently, starting from some appropriate approximation for $V_{\text{eff}}(t=0)$, until convergence is achieved. We should note that solving the eigenvalue problem for the Dirac Hamiltonian (such as Eqs. (11)) using finite basis set expansions results in generation of spurious non-physical eigenstates, if no additional constraint (dual kinetic balance (DKB) approach, see [11]) is imposed on the wave function. In the present work we do not follow the DKB approach: as our calculations show, presence of spurious states does not affect the final results since transitions to these states are negligibly small.

The time-dependent Hamiltonian $H(t)$ of Eq. (2) can be represented as a sum of time-independent and time-dependent contributions according to

$$H(t) = H_0 + V(t), \tag{13}$$

where

$$V(t) = V_{\text{ext}}(t) + V_{\text{eff}}(t) - V_{\text{eff}}(t=0). \tag{14}$$

To propagate the orbitals Ψ_k in time, we apply successively the following short-time split-operator propagator scheme:

$$\Psi_k(t+\delta t) = \exp\left(-i\frac{\delta t}{2}H_0\right)\exp\left[-i\delta t V\left(t+\frac{\delta t}{2}\right)\right]\exp\left(-i\frac{\delta t}{2}H_0\right)\Psi_k(t), \tag{15}$$

which is accurate up to the terms of the order of $(\delta t)^2$. To reduce the numerical effort, we extend the generalized pseudospectral (GPS) discretization of wavefunctions in spherical coordinates (see detailed description in [5]) to relativistic calculations. In this method, all multiplication operators are represented by diagonal matrices. In Eq. (15), diagonal is the matrix $\exp\left[-i\delta t V\left(t+\frac{\delta t}{2}\right)\right]$ in the coordinate representation. Although it is time-dependent and must be computed at each time step, this computation is not time-consuming. The matrix $\exp\left(-i\frac{\delta t}{2}H_0\right)$ is full but time-independent and computed only once before the start of the time propagation. We calculate it with the help of the spectral expansion:

$$\exp\left(-i\frac{\delta t}{2}H_0\right) = \sum_k \exp\left(-i\frac{\delta t}{2}E_k\right)|\Phi_k\rangle\Phi_k. \tag{16}$$

Using Eq. (16), we can also control the contributions of extremely high energy and negative continuum states thus improving the numerical stability of the calculations.

If the external laser field has a linear polarization, the projection of the total angular momentum onto the polarization axis is con-

Please cite this article in press as: D.A. Tumakov et al., Multiphoton ionization of many-electron atoms and highly-charged ions in intense laser fields: a relativistic time-dependent density functional theory approach, Nucl. Instr. Meth. B (2017), http://dx.doi.org/10.1016/j.nimb.2017.04.018