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N-electron dynamics of a heavy atom in strong fields

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Abstract

A semi-classical model is built to study the dynamical behaviour of many-electron wavefunctions interacting with strong fields for applications such as the interaction of a laser beam with a high-Z gas at high intensity or the collision between two heavy ions in the intermediate velocity regime. Within the semi-classical model, quantum dynamics of the electrons is described from generalized coordinates, which, as a result of a variational principle, obey classical Lagrange equations. The formalism is first applied to hydrogen ions and then extended to many-electron systems.

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1. Introduction

For various applications there is a need to accurately describe the dynamical behaviour of many-electron atomic systems interacting with strong fields. When the field strength is comparable with the nuclear one during a time larger than the inverse of the electron binding energy, expressed in atomic units, perturbation theories can no longer be applied. In this case, it is therefore necessary to numerically solve time-dependent self-consistent equations. A high-Z ion is obviously a strongly coupled Coulomb system (SCCS). Therefore, when considering the dynamics of heavy-ion electrons in a strong external field it is often required to go beyond the independent electron approximation. It concerns, for example, heavy-atom collisions in the intermediate velocity regime. Using Monte Carlo classical trajectories, it was shown in [1] that multi-ionization processes could significantly affect heavy-ion beam dynamics. Another important application is the interaction of heavy atoms with high-intensity laser beams. For XUV lasers created by optical field ionization (OFI) of a high-Z gas [2], it is necessary to determine precisely the energy distribution of the hot pumping electrons created by OFI, currently calculated within the one active electron model of [3]. One powerful model to treat the dynamics of a SCCS is the time-dependent density functional theory (TD-DFT)

that has been applied both in atomic systems and in plasmas. The main problem with the TD-DFT is that it can describe the evolution of only an average configuration and also that the calculation is performed on a fixed grid, with which it is difficult to consider large variations of the electronic properties such as those induced by a strong external field. Recently, a new approach was applied in a strongly coupled plasma based on the Gaussian wave packet molecular dynamic (GWPMD) [4]. The main advantage of the GWPMD is that the electron wavefunction in phase space is centred at the average values both for the position and for the velocity spaces. As a first approach of GWPMD for atomic systems, we restrict ourselves here to the same model as in [4] of one Gaussian wave packet per electron. As shown below this model already yields interesting results for treating an atomic system interacting with a strong field. (Atomic units will be used except when specified.)

2. Time-dependent variational principle (TDVP)

We consider a system characterized by a Hamiltonian $\hat{H}$ and a wavefunction φ, which depends on several parameters q_j. The evolution of the wavefunction is given by the TDVP [5] using the following Lagrangian,

$$\mathcal{L}(q_j, \dot{q}_j) = \sum_j p_j(q_j)\dot{q}_j - \mathcal{H}(q_j), \tag{1}$$

where $p_j = \langle\varphi|\mathrm{i}\frac{\partial}{\partial q_j}|\varphi\rangle$ is the conjugated momentum of q_j, and $\mathcal{H} = \langle\varphi|\hat{H}|\varphi\rangle$ is the semi-classical Hamiltonian. This variational principle allows the wavefunction evolution to be the closest to that of the exact solution.

Equation (1) yields equations similar to the Hamilton ones with the help of the so-called *norm-matrix* $\mathcal{N}$ defined by $\mathcal{N}_{j,k} = \frac{\partial p_j}{\partial q_k} - \frac{\partial p_k}{\partial q_j}$:

$$\sum_j \mathcal{N}_{j,k}\dot{q}_j = \frac{\partial \mathcal{H}}{\partial q_k} \Rightarrow \dot{q}_j = \sum_k \mathcal{N}^{-1}_{k,j}\frac{\partial \mathcal{H}}{\partial q_k}. \tag{2}$$

This set of coupled differential equations is solved by an adaptive step size RK4 method [6]. The accuracy of the TDVP method depends heavily on the form of the wavefunction and on the set of parameters used.

In [4], it was shown that the TDVP based on Gaussian wavefunctions is an efficient way to describe a SCCS, such as dense hydrogen plasmas. In this paper, the N-electron wavefunction is described by a Slater sum of Gaussian wave packets φ_{G} (GWP):

$$\varphi_G(\vec{q};\vec{x}) = \left(\frac{\omega}{\pi}\right)^{\frac{3}{4}} \mathrm{e}^{-(\frac{\omega}{2}+\mathrm{i}\gamma)(\vec{x}-\vec{r})^2}\,\mathrm{e}^{\mathrm{i}\vec{p}\cdot(\vec{x}-\vec{r})} \qquad \text{where} \quad \vec{q} = \{\omega, \gamma, \vec{r}, \vec{p}\}. \tag{3}$$

In this equation $\vec{r} = \langle\vec{x}\rangle$ and $\vec{p} = \left\langle -\mathrm{i}\frac{\partial}{\partial\vec{x}}\right\rangle$; these parameters are analogous to classical position and momentum. The parameters $\{\omega, \gamma\}$ allow for quantum behaviour of the electron such as charge delocalization.

3. Results

3.1. Hydrogen in a laser field

The Hamiltonian of the system proton+electron+laser field is $H = -\frac{1}{2}\frac{\partial^2}{\partial\vec{x}^2} - \frac{1}{|\vec{x}|} + \vec{E}\cdot\vec{x}$. The semi-classical Hamiltonian of the model in the case of GWP is

$$\mathcal{H} = \langle\varphi_G|\hat{H}|\varphi_G\rangle = \frac{1}{2}\vec{p}^2 + \frac{3}{\omega}\gamma^2 + \frac{\omega^2}{4} - \frac{1}{r}\mathrm{erf}(\sqrt{\omega}r) + \vec{E}\cdot\vec{r}. \tag{4}$$

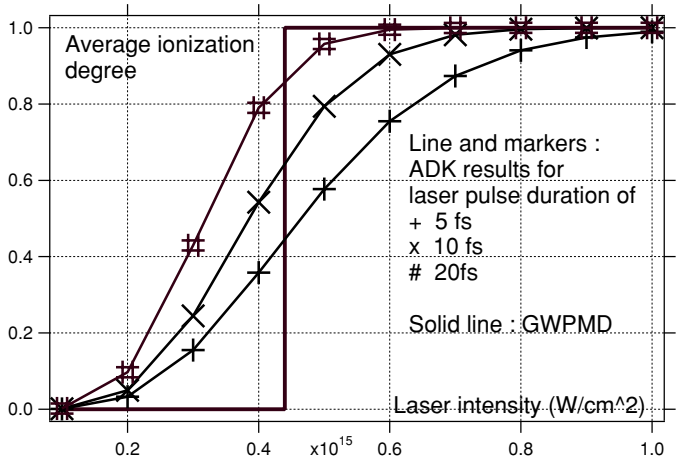

Figure 1. Average ionization degree after interaction with the pulse versus laser intensity given by the ADK model for different values of $\tau = 5$, 10, 20 fs, respectively, 2, 4 and 8 laser periods. GWPMD results are presented for $\tau = 10$ fs.

Minimization of $\mathcal{H}$ when $\vec{E} = \vec{0}$ yields the ground state energy value of $E_{1s} = -0.424$ au which is not far from the exact value of -0.5 au. The Lagrangian given by equation (1) is $\mathcal{L} = \dot{\vec{r}} \cdot \vec{p} + \frac{3}{2\omega}\dot{\gamma} - \mathcal{H}$, and equation (2) leads to

$$\dot{\vec{r}} = \frac{\partial \mathcal{H}}{\partial \vec{p}}, \qquad \dot{\vec{p}} = -\frac{\partial \mathcal{H}}{\partial \vec{r}}, \qquad \frac{\partial}{\partial \omega}\frac{3}{2\omega}\dot{\gamma} = \frac{\partial \mathcal{H}}{\partial \omega}, \qquad \frac{\mathrm{d}}{\mathrm{d}t}\frac{3}{2\omega} = -\frac{\partial \mathcal{H}}{\partial \gamma}. \tag{5}$$

The equations for $(\vec{r}, \vec{p})$ are similar to those given by classical Newton's laws for a particle evolving in the semi-classical potential $-\frac{1}{r}\mathrm{erf}(\sqrt{\omega}r) + \vec{E} \cdot \vec{r}$. Moreover, equations for (γ, ω) can also be derived from probability current conservation $\frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot \vec{j} = 0$.

We test the TDVP model behaviour in the case of OFI of hydrogen and compare it to the ADK model [3], commonly used for OFI problems. We have considered a linearly polarized laser field, the amplitude of which is $\vec{E} = E_0 \vec{u_x} \cos(2\pi ct/\lambda) \sin^2(\pi t/\tau)$, with a laser wave length value of $\lambda = 800$ nm and a laser pulse duration $\tau = n\lambda/c$ ($n = 2, 4, \ldots$). Results of the ADK model are reported in figure 1 for the values of $\tau = 5$, 10 and 20 fs.

We can see in figure 1 that the ionization threshold of GWPMD is close to the quantum result, but as in the classical case, the average ionization degree after interaction with the laser pulse could only be either 0 or +1. We can also remark that the GWPMD yields an ionization probability that is less dependent on the laser duration than the ADK results.

3.2. N-electron wavefunction applications

Now we study a heavy atom as a nucleus of charge Z surrounded by N electrons, which are described by a N-electron wavefunction $\varphi(\vec{x_1}, \ldots, \vec{x_N})$, interacting with a time-dependent laser field. The Hamiltonian is $H = \sum_{i=1}^{N} \left\{ -\frac{1}{2}\frac{\partial^2}{\partial \vec{x}_i^2} - \frac{Z}{|\vec{x}_i|} + \vec{E}(t) \cdot \vec{x}_i \right\} + \sum_{i,j} \frac{1}{|\vec{x}_i - \vec{x}_j|}$. The N-electron wavefunction is built with a Slater sum of one-electron GWP $\varphi_G(\vec{q}_i; \vec{x}_i)$. As shown in [4], the calculation of the semi-classical $\mathcal{H}$ leads us to

$$\mathcal{H} = \sum_{i=1}^{N} \mathcal{H}_i^{(1)} + \sum_{i,j} \frac{\mathrm{erf}(\sqrt{\omega_{i,j}} r_{i,j})}{r_{i,j}} + \sum_{i=1}^{N} \vec{E}(t) \cdot \vec{r}_i + \mathcal{H}^{\mathrm{exc}}, \tag{6}$$

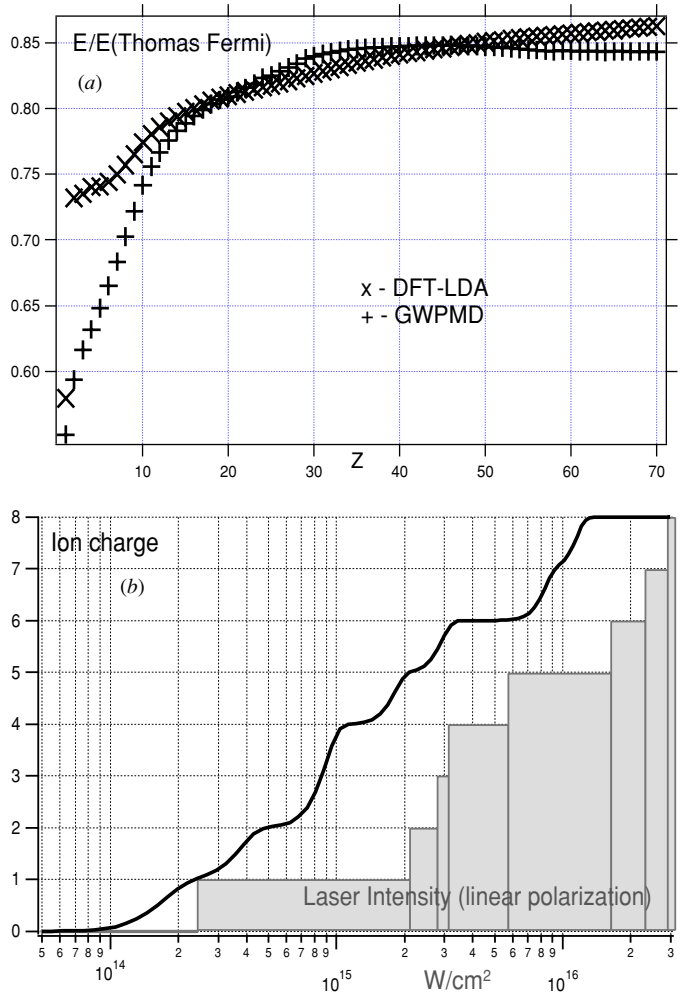

Figure 2. (*a*) Ground state energy of atoms versus *Z*. Comparison with DFT-LDA calculations. (*b*) Study of the OFI of a xenon atom, ion charge versus laser intensity. The curve represents ADK model results, bars are for GWPMD results.

where $\mathcal{H}_i^{(1)}$ is the one-electron semi-classical Hamiltonian,

$$\mathcal{H}_i^{(1)} = \frac{\vec{p}_i^{\,2}}{2} + \frac{3}{\omega_i}\gamma_i^2 + \frac{\omega_i^2}{4} - \frac{Z}{r_i}\mathrm{erf}(\sqrt{\omega_i} r_i), \tag{7}$$

$\frac{1}{\omega_{i,j}} = \frac{1}{\omega_i} + \frac{1}{\omega_j}$, and where the exchange-correlation term $\mathcal{H}^{\mathrm{exc}}$ comes from the part of the wavefunction due to anti-symmetrization. To calculate this term, a two-body approximation was made, as suggested in [4] which takes the form

$$\mathcal{H}^{\mathrm{exc}} = \sum_{i,j} \mathcal{T}_{i,j}^{\mathrm{exc}}, \qquad \mathcal{T}_{i,j}^{\mathrm{exc}} = f\left(\omega_i, \omega_j, \Delta_{i,j}^2\right) \mathrm{e}^{-\Delta_{i,j}^2} \tag{8}$$

where $\Delta_{i,j}^2 = \frac{\omega_i \omega_j}{\omega_i + \omega_j} r_{i,j}^2 + \frac{1}{\omega_i + \omega_j} p_{i,j}^2$. We can see that this term acts as a repulsion term in phase space for two electrons having the same spin.

Let us consider the ground state configuration. Within the GWPMD model, the ground state configuration can be determined by minimizing the semi-classical Hamiltonian $\mathcal{H} = \langle \hat{H} \rangle$ given by (6) with $\vec{E} = 0$. In figure 2(*a*) we report results of $\mathcal{H}$ minimization considering two Gaussian wave packets (spins up and down) for each shell (only exchange correlations between shells are considered here) together with those obtained with the DFT with local density approximation (LDA) [7]. We can observe in figure 2(*a*) that although very crude, the GWPMD model yields results which are in much better agreement with those of the DFT than with the Thomas–Fermi model.

As an example of GWPMD for the dynamical problem, using (6)–(8), we have studied the OFI of a xenon atom. We have considered the eight (5s–5p) electrons, whereas the electrons in the inner shells are described by fixed GWPs. In figure 2(*b*), we have reported the GWPMD and ADK results for the evolution of the average ionization degree of the xenon atom after interacting with a 40 fs laser pulse, in terms of the laser intensity. We can see in figure 2(*b*) that, again despite the very crude approximation used for the one-electron wavefunction, the good order of magnitude of ionization threshold is recovered by the GWPMD.

4. Conclusion

We have shown that the TDVP based on a Gaussian wavefunction can provide a valuable framework to study, using an *ab initio* method, the dynamics of atomic strongly coupled Coulomb systems. Using the TDVP together with the simplest shape of a Gaussian wave packet, one can already obtain a realistic description of the SCCS dynamics. Work is in progress to improve the accuracy of the model by introducing linear combinations of Hermite Gaussian wave packets.

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