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Three-dimensional simulation of the femto-second pulsed laser interacting with a nitrogen molecule

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ABSTRACT

Interaction of femto-second pulsed laser with a nitrogen molecule at various angles is investigated numerically in the present study. We propose a three-dimensional time-dependent Schrödinger equation (TDSE) solver using finite volume method (FVM) with single-active-electron (SAE) assumption. We construct the model potential by combining the soft-Coulomb potential and Yukawa potential and fitting both the first ionization energy and orbital type of highest-occupied molecular orbital (HOMO) of a N_2 molecule. The ratio of simulated maximum ($\chi = 0°$) to minimum ($\chi = 90°$) ionization yield is 1.9 and it agrees more favorably with the converted experimental data (2.3–3.3) than the MO-ADK model does (10.0). This shows that the proposed model potential in the 3D TDSE solver can faithfully predict the ionization of a nitrogen molecule under strong pulsed laser incidence and is a very useful tool for future application in similar analysis of more complicated molecules.

1. Introduction

As the experimental technique has improved for laser–molecule interaction, the frontier applications were carried out such as molecule imagining and light source of short wavelength. Diatomic molecule is the simplest molecule and has been usually taken as a prototype for the laser–molecule interaction study. Recently, the experiments of laser induced ionization for diatomic molecule have been carried out [1]. We choose N_2 molecule as the target molecule for our studies, because of its relative simpler electronic structure and available experimental data for comparison. Fundamentally, these physical systems are three-dimensional problems, and thus the three-dimensional calculation for the laser molecule interaction is required. Thus, in the present study we would like to employ a previously developed parallel 3D TDSE solver [5] with a newly proposed model potential for nitrogen to predict the ionization yield of nitrogen and compare with available experimental data.

2. Theoretical approaches

2.1. Simplified TDSE

In the present study, we have assumed Born–Oppenheimer approximation (BOA) to neglect the effect of nuclear movement and employed single active electron (SAE) approximation [2] coupled with a newly proposed model potential to simplify multi-electron TDSE. The laser field is represented by a time varying electric field.

The key issue of SAE assumption is how to make a good effective potential. The soft-Coulomb potential method has been shown to perform reasonably well in the past. The basic idea of a soft-Coulomb potential is to remove the singularity of Coulomb potential and to represent the screened electrons of core electron. The soft-Coulomb potential can be written as:

$$\frac{-Z_{eff}}{\sqrt{|\vec{r}-\vec{R}|^2+\alpha}} \tag{1}$$

where $\vec{R}$ is the position vector of nucleus, Z_{eff} and α are tunable parameters that are usually fitted to match the ground state energy of atomic or molecular system and the asymptotic behavior of Coulomb potential at $r \to \infty$. But sometimes the two tunable parameters cannot satisfy both the ground state energy and the asymptotic behavior of Coulomb potential at the same time. To overcome this problem, we combine the soft Coulomb potential

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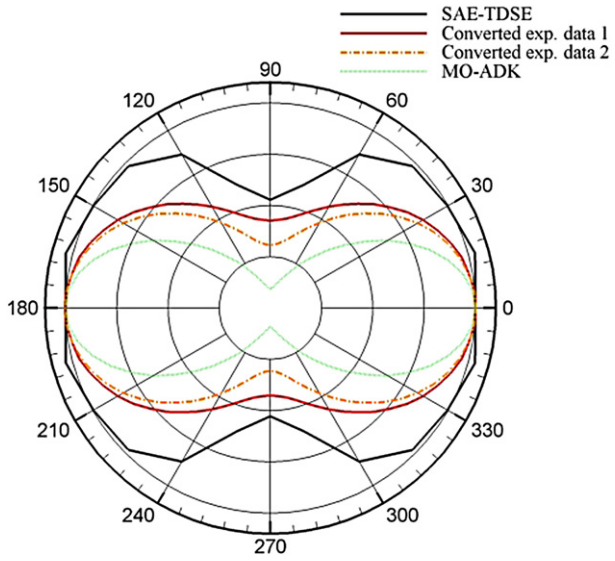

Fig. 1. The dependence of ionization yield on laser incident angle χ. SAE-TDSE is our present calculation (black solid line). Converted experimental data 1 (red solid line), data 2 (orange dash–dot line) and MO-ADK (green dotted line) are from Ref. [1]. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

with the Yukawa potential [3]. This leads to the new Yukawa-like soft-Coulomb potential which can be written as follows:

$$V_{eff} = \sum_i -Z_{eff}\left(1 + Z_{core} \cdot e^{-\beta|\vec{r}-\vec{R}_i|}\right)/\sqrt{|\vec{r}-\vec{R}_i|^2 + \alpha} \qquad (2)$$

where $\vec{R}_i$ is the position vector of nucleus i. With the four parameters (Z_{eff}, Z_{core}, α, and β), we can easily fit both the ground state energy and the asymptotic behavior of Coulomb potential at $r \to \infty$.

We determine these parameters by solving the 3D one-electron stationary Schrödinger equation ($H\psi = E\psi$) using the finite-element method with the following strategies:

(1) To fit the asymptotic behavior of Coulomb potential at $r \to \infty$ and equal to $-1/r$.
(2) To match both the first ionization energy and orbital type of highest-occupied molecular orbital (HOMO) of N_2 molecule.

3. Numerical method

We have previously developed a parallel 3D TDSE solver using finite volume method on Cartesian grid with non-uniform hexahedral cells. This code employs a stagger time marching scheme [4] and spatial domain decomposition for parallel computing. Details can be found in Ref. [5].

4. Results and discussion

In this study, we perform the TDSE simulation with the laser illuminating on a N_2 molecule with different incident angles (χ = 0°–90°) with respect to the molecular axis. The laser wavelength is 820 nm, intensity is 1.5×10^{14} W/cm^2, and total time duration is 10 optical cycles (~27.36 fs), which is modulated by an envelop function. Note this envelop function is the same as that defined by Eq. (10) in [6], but with only half duration. The envelop function is constant (= 1) between 2.5 and 7.5 cycles. The effective potential parameters for the N_2 molecule at equivalent bond length (2.075 au) are selected as: $Z_{eff} = 0.5$, $Z_{core} = 7$, $\alpha = 1.35$, and $\beta = 0.51$. The 5th molecular orbital calculated by solving stationary Schrödinger equation is σ_g orbital that is the same as

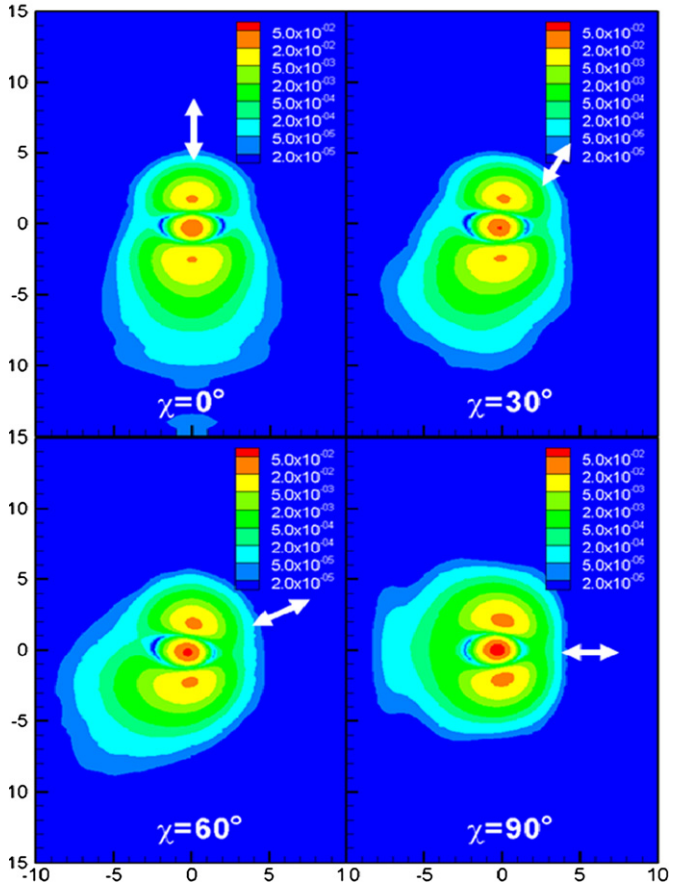

Fig. 2. Electron probability distribution slice at $x = 0$ plane at $t = 5$ optical cycle.

HOMO of N_2 molecule, and the orbital energy is −0.5748 eV that is nearly the same as the experimental first ionization energy of N_2 molecule. Note all the simulations were conducted using nearly 4 million cells, the time step size 0.004 au and 282 716 time steps.

4.1. Ionization yield of nitrogen molecule

Fig. 1 shows the dependence of the simulated ionization yield on the incident angle of laser along with the converted experimental data and theoretical data based on the MO-ADK model [1]. We have found that the simulations show minor fluctuation at small incidence angles. However, the simulated ratio (1.9) of maximum ($\chi = 0°$) to minimum ($\chi = 90°$) ionization yield compare more favorably with the converted experimental data (2.3–3.3) than the calculated ratio (10.0) by the MO-ADK model. This shows that direct 3D simulations are necessary to reproduce the measured ionization yield of a nitrogen molecule under laser illumination.

4.2. Electron probability distribution

Fig. 2 demonstrates the electron probability distribution which be sliced at $x = 0$ plane at $t = 5$ optical cycle (mid of total time propagation) for different laser incident angles. The direction of arrow indicates the direction of incident laser electric field oscillation. The electron probability distribution is clearly deformed by the laser illumination, and the electron tends to oscillating in the direction of incident laser.

5. Conclusion

Interaction of femto-second pulsed laser with a nitrogen molecule at various angles is investigated numerically in the present

study. We propose a three-dimensional time-dependent Schrödinger equation (TDSE) solver using finite volume method (FVM) with single-active-electron (SAE) assumption. We construct the model potential by combining the soft-Coulomb potential and the Yukawa potential. Predicted angular ionization yields are closer to the experimental data than the data of the MO-ADK model are. The method we have proposed could be a good tool for theoretical analysis for laser–molecule interaction.

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