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Modelling laser-matter interactions for simple molecules

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Synopsis We consider the interaction of a molecule with an ultrashort laser pulse within the mean field approximation. A model has been generalized to treat the interaction of atomic hydrogen with an electromagnetic pulse. The kernel of the Coulomb potential in the momentum space is replaced by a sum of products of separable potentials supporting a bound state of the atom and built from the corresponding bound state wave function. The molecular orbitals are expressed in terms of Gaussians and generated by means of a quantum chemistry software. We analyze the ionization yield as a function of the size of the Gaussian basis for H₂ and H₂O molecules and the results are compared to those already existing in the literature.

Treating the interaction of a multi-electron system with an oscillating field is never a simple task. Either one solves numerically a time-dependent Schrödinger equation (TDSE) or one uses the time-dependent density functional theory. Using either of these methods is always time consuming and computationally very expensive. Here, we develop an alternative approach that requires reasonable computer resources. It uses modern quantum chemistry codes to generate molecular orbitals expressed in terms of Gaussian type orbitals (GTO). This simplifies the calculations but the basis size becomes crucial for spatially extended system since GTO's are strongly localized near the nuclei.

Our approach is based on a simple model, first introduced for atomic hydrogen [1, 2] and generalized, within the mean field approximation to the treatment of the interaction of a simple molecule with an attosecond laser pulse.

The complicated molecular potential is approximated with a separable potential that describes only one molecular orbital:

$$V(\vec{p}, \vec{u}) = v(\vec{p})v^*(\vec{u}), \quad (1)$$

with

$$v(\vec{p}) = -\frac{1}{a} \left[\epsilon_0 - \frac{p^2}{2} \right] \Phi_0^{var}(\vec{p}), \quad (2)$$

where a is a constant. The molecular orbital $\Phi_0^{var}(\vec{p})$ is precalculated variationally in a GTO basis set using the well established quantum chemistry software package GAMESS-US. Assuming the external field is linearly polarized and described in terms of the vector potential $A(t)\vec{e}$ where \vec{e} is a unit vector along the polarization axis, we solve the following one-

electron TDSE in momentum space:

$$\left[i \frac{\partial}{\partial t} - \frac{p^2}{2} + \frac{1}{c} A(t)(\vec{e} \cdot \vec{p}) \right] \Phi(\vec{p}, t) + v_n(\vec{p}) \int \frac{d\vec{u}}{(2\pi)^3} v_n^*(\vec{u}) \Phi(\vec{u}, t) = 0, \quad (3)$$

the solution of which can be written analytically in terms of a function $F(t)$ which in turn is the solution of a one dimensional Volterra integral equation of the second kind, depending only on time:

$$F(t) = F_0(t) + \int_0^t K(t, \xi) F(\xi) d\xi \quad (4)$$

(see [1, 2] for the details of the calculations).

As a first step, this model is applied to atomic hydrogen with the ground state expressed in terms of GTO's. The results are compared to those obtained (i) by using the exact ground state wave function [2] and (ii) by solving the TDSE in the case of the exact Coulomb potential. This allows one to study the adequacy of different GTO basis sets (namely, 6-31G and 6-311++G**) and the applicability of the model in general.

A similar study is performed for molecular hydrogen H₂ and water H₂O. For the latter one, we can consider different molecular orbitals, but it is only for the HOMO orbital that we have an ionization process that keeps the same geometry of the molecule without exciting any vibrational states. Results are compared to those given in the literature.

References

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