Finite element DVR method for molecular single and double ionization by strong laser pulses

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Synopsis We are developing an *ab initio* numerical method to compute molecular single and double ionization processes induced by intense laser pulses. We present our first time-dependent simulations in the H_2^+ molecular ion including both electronic and nuclear degrees of freedom. We compare different methods for the extraction of excitation and ionization probabilities from the numerical time-dependent wave function, which will be applied to more complex targets in the future.

We solve the time-dependent Schrödinger equation (TDSE) for the H_2^+ molecule in full dimensionality, with three spatial degrees of freedom for the electron, and nuclear motion restricted to one dimension (neglecting its angular dependence). The method is closely related to previous approaches employed in atoms [1, 2], employing a single-center expansion for the electronic degrees of freedom, with spherical harmonics describing the angular dependence. The radial component of both the electronic and nuclear motion is described using a finite-element discrete variable representation (FEDVR) basis.

In order to avoid unphysical reflections and to impose pure outgoing boundary conditions in the driven equations (see below), we apply exterior complex scaling (ECS) for both the electronic and internuclear radial coordinates [3].

As an initial benchmark result, we compute the (pure) dissociation and ionization cross sections for single-photon absorption, using three distinct methods. First, we obtain the total absorption (dissociation+ionization) from the optical theorem, $\sigma = \mathrm{Im}\langle\psi_0|\hat{\mu}\frac{1}{\hat{H}-\omega}\hat{\mu}|\psi_0\rangle$, where $|\psi_0\rangle$ is the ground state, $\hat{\mu}$ is the dipole operator (along the internuclear axis), $\hat{H}$ is the Hamiltonian, and ω is the driving frequency. Second, we propagate the system under the influence of an ultrashort laser pulse (which also gives access to multiphoton processes). We follow the formalism developed in [1] to implicitly integrate to infinite times for extracting the final observables. The dissocation and dissociative ionization cross sections are obtained by either integrating the probability flux through properly chosen surfaces, or by extracting the energy- and angle-resolved cross sections using surface integrals over testing functions [3, 1].

Figure 1 shows the cross sections for single-photon absorption using the latter implementation, extracted from the time-dependent wave packets obtained after interaction with laser fields of varying central frequency and duration. We obtain perfect agreement with ref. [4].

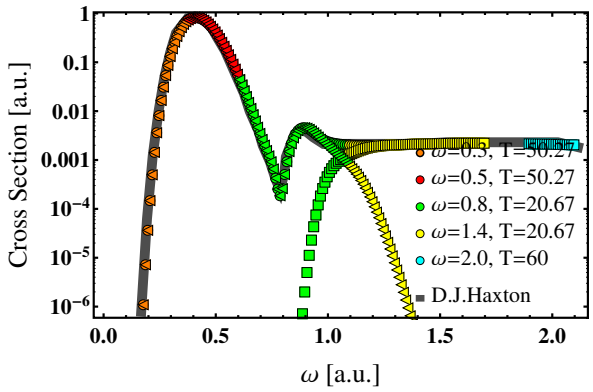

Figure 1. Absorption cross section for molecular ion for polarization parallel to the bond axis as a function of the photon energy. Full line: Optical theorem. Rectangles: dissociative ionization. Triangles: dissociative excitation. Circles: total cross section. Each color stands for a different field use in the solution of the TDSE; the central frequency and pulse duration in atomic units are indicated.

References

[1] A. Palacios, T. N. Rescigno and S. W. McCurdy 2007 *Phys. Rev. A* **76** 043420

[2] J. Feist *et al.* 2008 *Phys. Rev. A* **77** 043420

[3] T. N. Rescigno and C. W. McCurdy 2000 *Phys. Rev. A* **62** 032706

[4] D. J. Haxton 2013 *Phys. Rev. A* **88** 013415

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