Solution of the time-dependent Schrödinger equation describing H_2 in strong laser fields including nuclear motion and non-adiabatic radial couplings

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The accurate quantum treatment of atoms and molecules in strong laser fields is a prerequisite to understand and predict new and surprising experimental findings and is especially of interest to ”produce a movie” of the dynamics of electrons and nuclei exposed to laser fields. The CI-based spectral approach developed earlier [1, 2, 3] allows for solving the TDSE equation describing H_2 in strong laser fields within the fixed-nuclei approximation in full dimensionality, including an arbitrary alignment of the molecule (6-dimensional electronic problem). We present a fully quantum mechanical extension of this method that includes nuclear vibrational motion, incorporating even the so far ignored non-adiabatic couplings. The newly implemented approach differs in three aspects from a previously implemented method for solving the TDSE of H_2 including vibrational motion [4, 5]. First, prolate-spheroidal coordinates are used for the electronic part instead of a one-center expansion. Second, the nuclear motion is not implemented using an expansion in field-free Born-Oppenheimer eigenstates, but in a more efficient way. Third, the previously ignored non-adiabatic radial couplings are included.

As a first application of our approach, we have reconsidered the dramatic effect of nuclear motion compared to the fixed-nuclei approximation (FNA) at a single fixed internuclear, i. e. the equilibrium distance found for few-photon ionization in [4]. A more detailed analysis of this previously reported dramatic breakdown the Born-Oppenheimer approximation indicates that it is crucial how the FNA is defined. No nuclear motion during the pulse would suggest that results obtained within the fixed-nuclei approximation for different internuclear distances R should be weighted with the probability density of the initial nuclear wavefunction (termed ”frozen-nuclei approximation” in [6]). Figure 1 shows that this weighting leads to significantly different results compared to using only the equilibrium distance R=1.4 a.u., explaining the dramatic effect already within this approximation. It is additionally investigated how the results further change when (i) including the nuclear motion during the pulse as in [4, 5] and (ii) also including non-adiabatic radial couplings.

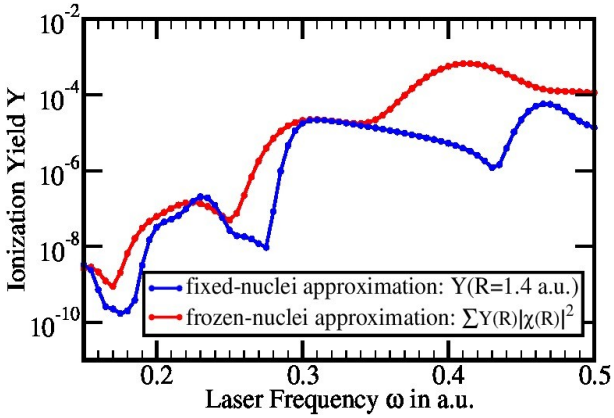

Fig. 1: Comparison of different FNA versions for a 10fs cos²-envelope pulse with peak intensity 10^{12} W/cm².

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