Monte Carlo wave packet theory of dissociative double ionization

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Synopsis Nuclear dynamics in strong-field double ionization processes are predicted using a stochastic Monte Carlo wave packet technique. The model takes input from electronic structure calculations and strong-field electron dynamics, and allows for field-dressed dynamics within a given molecule as well as several jumps between different charge states. The method is computationally efficient, applicable to a wide range of systems and relies on no empirical parameters. Nuclear kinetic energy release spectra for H_2 (D_2) in strong near-infrared laser pulses of 40 fs duration are compared to experiments and excellent agreement is obtained, showing the importance of accounting consistently for both neutrals and ions during the pulse.

When a molecule is exposed to a strong laser pulse the system is excited and ionizes and the nuclei start to dissociate. For pulses of current interest, e.g., 800 nm, 10^{14} W/cm^2 and durations in the femtosecond regime, all these processes are in principle accurately described by the time-dependent Schrödinger equation. The nonlinearity introduced by the external field is, however, so strong that the solution of this equation is impossible even for H_2, and modeling has focused separately either on ionization or dissociation dynamics. The purpose of the present poster is to provide a consistent theoretical framework that treats ionization and dissociation on an equal footing, and is applicable to any molecular system where electronic structure, dipole moment functions, and ionization dynamics can be determined. We illustrate the methodology using H_2 (D_2) since this is the simplest system, and experiment and theory exist. Our work shows, that the feeding of the molecular ion from the neutrals throughout the pulse, not accounted for in previous theory, is essential to explain details in the experiments.

Starting from $H_2((1s\sigma_g)^2)$, we study the nuclear dynamics as the population is transferred to the $1s\sigma_g$ and $2p\sigma_u$ states of H_2^+ and subsequently ionized leading to Coulomb explosion. This is done by treating ionization as a decay process, incorporated in a master equation for the molecular density matrix. The so-called Monte Carlo wave packet technique involves wave functions instead of density matrices and is computationally easier for this problem.

An outline of the physical processes accounted for by our treatment are given in Fig. 1. We assume the Born-Oppenheimer approximation and an initial nuclear wave function consistent with the electronic ground state [Fig. 1(a)]. The laser field excites the electronic state and with a given probability, the molecule is ionized and jumps to the lowest state in H_2^+[Fig. 1(b) → Fig. 1(c)]. The dynamics proceeds in the coupled $1s\sigma_g$-$2p\sigma_u$ system [Fig. 1(c)] until further ionization (a second jump) leaves the bare nuclei in the $1/R$ Coulomb potential [Fig. 1(d)]. To obtain the kinetic energy release (KER), a projection onto Coulomb scattering state is made after the second ionization.

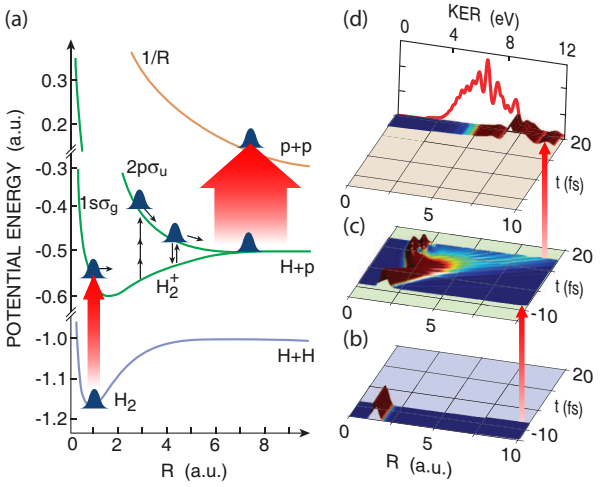

Fig. 1. Left panel: Sketch of the double ionization process for H_2 using field free BO curves. Right panel: One realization of the wave function dynamics in neutral, singly ionized and doubly ionized H_2.

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