Quantum mechanical investigation of electronic and nuclear dynamics of molecules in intense laser fields

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We have accurately solved the time-dependent Schrödinger equations of H_2^+ and H_2 by using an efficient grid-point wave packet method based on the dual transformation technique [1]. The calculated electronic and nuclear dynamics is analyzed by using "field-following" time-dependent adiabatic states $\{|n\rangle\}$ defined as eigenfunctions of the "instantaneous" electronic Hamiltonian $H_{el}(t)$ including the interaction with a laser electric field $\varepsilon(t)$. The analysis has shown that nuclear dynamics (such as chemical reactions) in a near-infrared field can be described in terms of wave packet propagation on time-dependent adiabatic potentials and nonadiabatic transitions due to temporal change in $\varepsilon(t)$ [2] .

The above method of describing dynamics in terms of time-dependent adiabatic states is also useful for analyzing and predicting the electronic dynamics of molecules in intense laser fields. A time-dependent adiabatic state can be regarded as a doorway state to ionization. To clarify the mechanisms of ionization of *multi-electron* molecules (such as enhanced ionization), we developed a theoretical framework to calculate the ionization probability of a *time-evolving* electronic state as a function of internuclear distances; the present adiabatic state approach or a multi-configurational time-dependent Hartree-Fock method [3] is combined with the so-called intense-field many-body S-matrix theory [4].

The following three effects that are crucial for ionization in intense laser fields are examined.

i) Deformation of the electronic wave function of a time-dependent adiabatic state due to field-induced intramolecular electron transfer (e.g., formation of an ionic state such as H^+H^-).
ii) Role of electron correlation.
iii) Field-induced nonadiabatic transitions to higher time-dependent adiabatic states.

We present the results for theoretical calculations of the ionization probability and the photoelectron angular distribution.

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[2] H. Kono *et al.*, *Laser Phys.* **13,** 883 (2003). H.Kono et al. *Chem. Phys.* in press (2004).
[3] T. Kato and H. Kono, *Chem. Phys. Lett.* **392**, 533 (2004).
[4] See, e.g., A. Becker and F.H.M. Faisal, *Optics Express* **8**, 383(2001).