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Laser Control of Molecular Dynamics:

Enhanced Ionization and Optical Centrifuge

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We discuss two topics on theoretical studies of laser-molecule interactions.

[A] Mechanism of enhanced ionization of H_2 and H_3^+

The effects of spatial symmetry regulated by the molecular spin states on the ionization dynamics are investigated in two-electron molecular systems. As typical two-electron molecules, we focus on the ionization dynamics of H_2 and H_3^+ in an ultrashort, intense laser pulse ($I \geq 10^{14}$ W/cm^2, wave length λ = 800 nm) by solving exactly the time-dependent Schrödinger equation for a one-dimensional model. Enhanced ionization is observed at a critical internuclear distance *Rc* in both singlet and triplet states of H_2 and H_3^+. Comparing the dynamics of H_2 and that of H_3^+, it is clarified that not only the ionization potential of the molecule but also the spatial symmetry of the wave function has an important effect on the ionization dynamics. In the case of H_3^+, Rc～8 a.u. for both singlet and triplet states, whereas Rc of the singlet state (Rc～6 a.u.) becomes much different from that of triplet state (Rc～2 a.u.) for H_2. These results imply that the ionization of the even-electron molecules is strongly influenced by the spatial symmetry which is governed by both spin states and the configuration of nuclei.

[B] Molecular rotation in an optical centrifuge

Cold molecules can be produced either by photoassociation of cold atoms or using buffer gas cooling. While cooling simplifies control over translational motion, complete control of rotations still requires deep angular traps. In this study, a method that accelerates molecular rotations and breaks the anisotropic molecular bonding by using strong nonresonant and chirped laser pulses is presented. If the two beams are linearly chirped with respect to each other, the polarization rotates with constant acceleration, creating a steadily accelerating angular “trap” for the molecule. We demonstrate the capability of the effective rotation and selective dissociation of diatomic isotopes based on a time-dependent quantum approach. We also focus that the rotational-transition through a “pendular state", namely the mechanism of adiabatic passage, is completely achieved in linear molecular systems, meaning that the efficient adiabatic excitation is attained in the optical centrifuge.