Molecules in Few-Cycle Intense Laser Fields

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When exposed to intense laser fields, molecules exhibit a variety of exotic features as the magnitude of the electric field component is comparable with that of the Coulomb field within a molecule. For a deeper understanding of the light-matter interaction, we investigate how atoms and molecules behave in strong and ultrashort laser fields, which carry only a few optical cycles within the pulse.

1. Electronic and Nuclear Responses of Fixed-in-Space H_2S to Ultrashort Intense Laser Fields[1)]

The Coulomb explosion dynamics in non-resonant, ultrashort intense laser fields (12 fs, ~10^{14} W/cm^2) is studied for H_2S with its orientation fixed in space, to clarify how the electronic and nuclear responses change by the direction of laser polarization direction (ε) in the molecular frame (Figure 1). The momenta of the respective fragment ions, $p_1(H^+)$, $p_2(H^+)$ and $p_3(S^+)$ produced in the Coulomb explosion process, $H_2S^{3+} \rightarrow H^+ + S^+ + H^+$, were determined as three-dimensional vectors in the laboratory frame. The kinetic energy release (E_{kin}) and momentum angle (θ_{12}) distributions obtained for the respective directions revealed that the geometrical structure is almost frozen during the interaction with the laser fields for x//ε, while it becomes elongated along the laser polarization vector when ε is parallel to the y- or z- axis, demonstrating that the Coulomb explosion dynamics of H_2S in intense laser fields can be manipulated by the polarization direction in the molecular frame.

The elongation in the molecular structure can be interpreted in terms of the induced dipole potential, $U_{id} = -\alpha\varepsilon^2/2$. As the molecular structure stretches along ε, the corresponding component of polarizability tensor α becomes larger, which lowers the potential energy to induce the nuclear dynamics towards the elongated structures. Alternatively, the observed structural deformation can be explained by the population transfer to charge transfer (CT) states characterized with the ionic charge distribution and the large transition moments from the ground state. Because of the ionic character, CT states can be stabilized in intense laser fields against the covalent ground state when the direction of the charge separation is parallel to the electric field ε. The large transition moments then facilitates the laser induced non-adiabatic population transfer from the ground state to the CT state, which leads to the enhanced ionization and dissociation in intense laser fields by the localized charge distribution.

In the case of H_2S, CT states with the H^-–S–H^+ (and H^+–S–H^-) type configuration and those with the H_2^+–S^- or H_2^-–S^+ character are expected to contribute to the dynamical processes for the y//ε and z//ε directions, respectively. Possible candidates of such CT (ionic) states are the electronically excited 3^1B_2 and 5^1A_1 states located at 13.9 eV and 13.7 eV above the ground state at the equilibrium geometry. The transition moments from the ground state, X^1A_1 , are $|\mu|$ = 1.57 and 1.34 a.u., along the y- and z-axis, respectively.

On the other hand, no such CT states can be coupled with the ground state when ε is perpendicular to the molecular plane. The ionization process for x//ε should be dominated by the tunneling ionization from the HOMO $2b_1$ extending perpendicular to the plane. The ionized electron is then rescattered by the ion core to form highly charged parent ions within a few optical cycles, which minimizes the structural deformation prior to the Coulomb explosion.

In conclusion, we have demonstrated that the Coulomb explosion dynamics of H_2S in intense laser fields can be manipulated by the polarization direction ε in the molecular frame. The observed dependence of the electronic and nuclear responses to the non-resonant laser fields can be explained in terms of the character of the CT states, which serve as the “doorway states” to the structural deformation of small polyatomic molecules in intense laser fields as well as to the ionization and fragmentation processes discussed previously. The understanding of the electronic and nuclear response to the laser fields polarized in the molecular frame presented here will provide new prospects for efficient quantum control in the intense field regime.

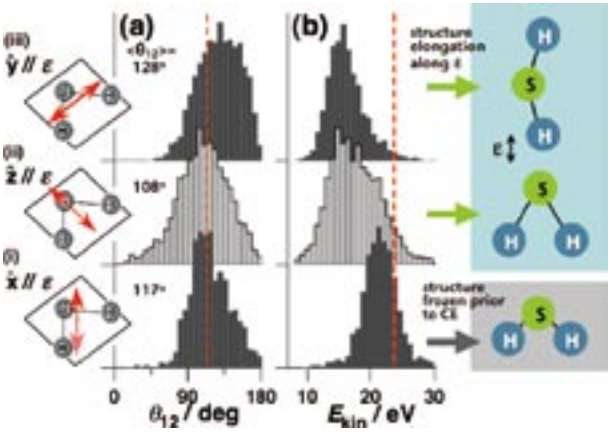

Figure 1. (a) Momentum angle θ_{12} distribution and (b) total kinetic energy E_{kin} distribution obtained for three different directions of ε, in the molecular frame, (i) *x*//ε, (ii) *y*//ε and (iii) *z*//ε.

2. Dalitz Plot Analysis of Coulomb Exploding O_3 in Ultrashort Intense Laser Fields[2)]

The three-body Coulomb explosion of ozone, $O_3^{3+} \rightarrow O^+ + O^+ + O^+$, in ultrashort intense laser fields (2×10^{15} W/cm^2) is studied with two different pulse durations (9 and 40 fs) by the coincidence momentum imaging method. The nuclear dynamics in the laser fields is discussed using the Dalitz plot,[3)] developed originally to describe the three-body reactions in elementary particle physics, which provides a compact two-dimensional representation of the relative momentum sharing among the three fragments produced through the dissociation of small polyatomic molecules.

In a Dalitz plot, normalized kinetic energy parameters of the three fragments are plotted in the Cartesian coordinate system (*x*,*y*),

$$x = (\varepsilon_1 - \varepsilon_2)/(\sqrt{3}E_{kin}), \quad (1)$$

$$y = \varepsilon_3/E_{kin} - 1/3, \quad (2)$$

where ε_i is the kinetic energy of the *i*-th fragment and $E_{kin} = \varepsilon_1 + \varepsilon_2 + \varepsilon_3$. A set of three fragment momenta that fulfills the momentum conservation condition falls within a circle of 1/3 in radius in this plot, and form a uniform distribution if there is no correlation among the three fragment momenta.

The Dalitz plot obtained for the 9-fs intense laser fields exhibits a sharp peak centered at the origin as shown in Figure 2, showing that all the three fragment ions tend to have the same momentum values, by the strong Coulombic interactions during the explosion process. When the pulse duration is increased from 9 to 40 fs, a broadening of the Dalitz plot distribution is identified, in addition to a decrease in the total kinetic energy release. The analysis based on a simple Coulomb explosion model shows that the geometrical structure of ozone remains almost unchanged during the interaction with the few-cycle intense laser fields, while a significant structural deformation along all the three vibrational coordinates, including the anti-symmetric stretching coordinate, is identified in the 40 fs laser fields. The observed nuclear dynamics are discussed in terms of the population transfer to the excited states of O_3.

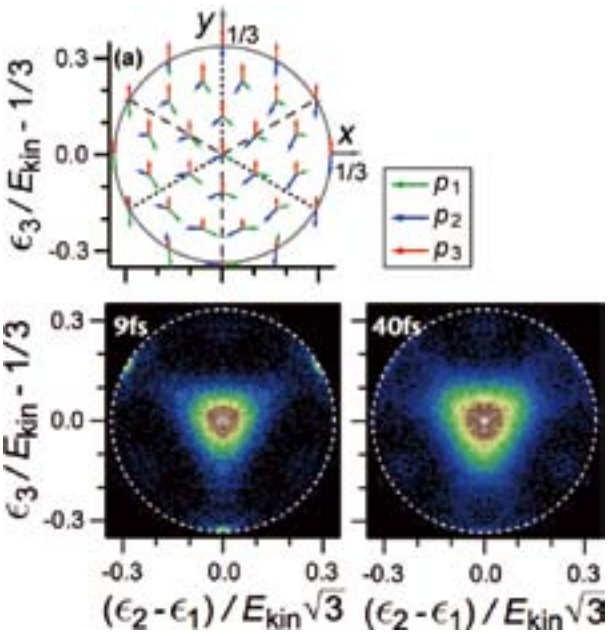

Figure 2. Dalitz plots obtained for the Coulomb explosion process of ozone, $O_3^{3+} \rightarrow O^+ + O^+ + O^+$, in 9 and 40 fs intense laser fields (2×10^{15} W/cm^2).

References

1) A. Hishikawa. E. J. Takahashi and A. Matsuda, *Phys. Rev. Lett*. **96**, 243002 (2006).
2) A. Matsuda, E. J. Takahashi and A. Hishikawa, *J. Chem. Phys*. **127**, 114318 (2007).
3) R. H. Dalitz, *Philos. Mag*. **44**, 1068–1080 (1953).

Award

HISHIKAWA, Akiyoshi; Morino Fundation for Molecular Science.