VI-C Molecules in Few-Cycle Intense Laser Fields

When the intensity of a laser field reaches as large as ~10^{15} W/cm^2, the magnitude of the electric field component becomes comparable with that of the Coulomb field within a molecule. Thus, the electrons bound in a molecule become heavily perturbed by the external electric field to form a new class of states, often referred to as light-dressed states. Since an internuclear potential within a molecule is deformed in response to the temporal variation of the amplitude of the light field, dynamical processes of molecules such as vibration and chemical bond breaking should be controlled if the light field is properly designed. In the present study, we employed extremely short intense laser pulses with the duration less than 10 fs, to clarify the behavior of molecules through the three-dimensional momentum measurements of the fragment ions produced from a single parent ion.

VI-C-1 Electronic and Nuclear Responses of Fixed-in-Space H_2S in Ultrashort Intense Laser Fields

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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. The momenta of the respective fragment ions, $\boldsymbol{p}_1(H^+)$, $\boldsymbol{p}_2(H^+)$ and $\boldsymbol{p}_3(S^+)$ 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.

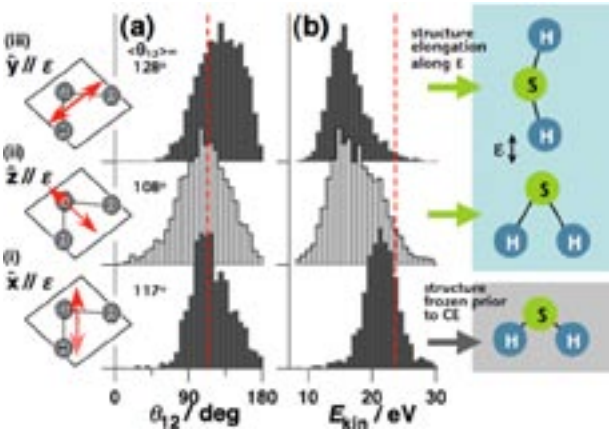

Figure 1. (a) Momentum angle θ_{12} distribution and (b) total kinetic energy E_{kin} distribution obtained for three different directions of ε in the molecular frame, (i) *x*//ε, (ii) *z*//ε and (iii) *y*//ε. Each distribution is normalized at the peak. The dotted lines indicate the θ_{12} and E_{kin} values obtained by a classical simulation of the Coulomb explosion on the *ab initio* PES, θ_{12} = 113.6° and E_{kin} = 23.6 eV, which are smaller than those expected from the corresponding Coulombic PES, θ_{12} = 124.1° and E_{kin} = 29.2 eV.

VI-C-2 Coulomb Explosion Imaging of Molecular Structures with Ultrashort Intense Laser Pulses

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The Coulomb explosion process of CS_2 in ultrashort (<10 fs) intense laser field (>10^{15} W/cm^2) is investigated. At the field intensity of 5 × 10^{15} W/cm^2, six different symmetric pathways are identified in the Coulomb explosion of CS_2, $CS_2^{z+} \rightarrow S^{p+} + C^{q+} + S^{r+}$; (*p*,*q*,*r*) = (1,1,1), (1,2,1), (2,1,2), (2,2,2), (3,2,3), (4,2,4) by the coincidence momentum imaging. A good agreement between the results obtained for the (4,2,4) pathway and the momentum distribution expected from the geometry of neutral CS_2 was obtained, indicating that the molecular structure is directly reflected in the momentum distribution when fragment ions from highly charged parent ions are monitored.