VI-C Ultrafast Dynamics of Molecules in Intense Laser Fields

The interaction between molecules and an intense laser field (10^{12}–10^{18} W/cm^2), whose electric field component comparable with the intramolecular Coulombic field, has been an attractive target of research to elucidate fundamental physical properties underlying in the unique features that do not appear in the weak interaction regime. In the present study, the characteristic dynamics occurring in intense laser fields, such as structural deformation and multiple breaking of chemical bonds, is studied by a newly developed experimental method, called *coincidence momentum imaging*, which allows us to determine the momentum vectors of all the fragment ions ejected from a single parent molecule in an intense laser field. Based on the correlation among the fragment momentum vectors, the evolution of nuclear motion on the light dressed potential energy surfaces and the hydrogen migration process have been clarified. A new coincidence momentum imaging system is constructed to detect the ions and electrons produced from a molecule placed in an intense laser field.

VI-C-1 Nuclear Dynamics on the Light-Dressed Potential Energy Surface of CS_2 by Coincidence Momentum Imaging

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The non-sequential three-body Coulomb explosion, $CS_2^{3+} \rightarrow S^+ + C^+ + S^+$, in an intense laser field (0.2 PW/cm^2, 60 fs) is studied by the coincidence momentum imaging of the fragment ions. The observed angle distribution of the momentum vectors of the two S^+ ions, $\boldsymbol{p}_1(S^+)$ and $\boldsymbol{p}_2(S^+)$, exhibited a peak at the small angle $\theta_{12} \sim 140°$, showing that the nuclear motion is induced along the bending coordinate to a large extent prior to the explosion. On the other hand, the difference between their absolute values, $\Delta p_{12} = |\boldsymbol{p}_1(S^+)| - |\boldsymbol{p}_2(S^+)|$, has a sharp distribution peaked at $\Delta p_{12} = 0$, suggesting that the symmetric stretching motion dominates over the anti-symmetric stretching motion in the laser field. Based on the energy dependence of the momentum vector correlation, the characteristic nuclear dynamics of CS_2 on the light-dressed potential energy surfaces in the intense laser field is discussed.

VI-C-2 Hydrogen Migration in Acetonitrile in Intense Laser Fields Studied by Coincidence Momentum Imaging

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The two-body Coulomb explosion of acetonitrile, $CH_3CN^{2+} \rightarrow CH_{3-n}^+ + H_nCN^+$ (n = 0–2), in intense laser fields (0.15 PW/cm^2, 70 fs) is studied by the coincidence momentum imaging technique. It is found that the fragment ions for n = 0 are ejected mostly to the direction of the laser polarization vector with $\langle\cos^2\theta\rangle$ = 0.68, where θ is the angle between the fragment recoil direction and the laser polarization vector, while the angle distribution becomes more isotropic as n increases, *i.e.*, $\langle\cos^2\theta\rangle$ = 0.49 for n = 1 and $\langle\cos^2\theta\rangle$ = 0.37 for n = 2. From this characteristic correlation between the anisotropy in the fragment ejection and the hydrogen migration, the Coulomb explosion dynamics competing with the hydrogen atom transfer from the methyl group to the nitrile group is investigated.

VI-C-3 Hydrogen Migration in Acetonitrile in Intense Laser Fields in Competition with Two-Body Coulomb Explosion

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Two-body Coulomb explosion processes of acetonitrile (CH_3CN) and deuterated acetonitrile (CD_3CN), $CH_3CN^{2+} \rightarrow CH_{3-n}^+ + H_nCN^+$ and $CD_3CN^{2+} \rightarrow CD_{3-n}^+ + D_nCN^+$ (n = 0, 1, 2), in an intense laser field (0.15 PW/cm^2, 70 fs) are investigated by the coincidence momentum imaging method. The comparable yields derived for the three pathways (n = 0, 1, 2) shows that the hydrogen atom migration proceeds in competition with the Coulomb explosion. The angular distributions of the fragment ions for n = 0 exhibits a sharp peak along the laser polarization direction, while the angular distribution becomes more isotropic as n increases. Based on a least-squares analysis of the fragment anisotropy, the dissociation lifetimes of the doubly charged acetonitrile were determined, from which the time scale of the hydrogen migration as well as the deformation of the C–C–N skeleton prior to the explosion were discussed.

VI-C-4 Design and Development of an Ion-Electron Coincidence Momentum Imaging System

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A new coincidence momentum imaging system has been developed to study the behavior of molecules exposed to an intense laser field ($\sim 10^{15}$ W/cm^2) based on the momentum vectors of fragment ions and electrons ejected from a single parent molecule. The electrons and ions are guided by electrodes in the velocity mapping configuration and detected by a pair of position sensitive detectors with delay line anodes,

which are placed face by face in an ultrahigh vacuum chamber with a base pressure < 10^{-8} Pa. The performance of the electron detection has been studied with Xe and CS_2 in an intense laser field (~ 10^{13} W/cm^2, 35 fs, 800 nm). The concentric ring patterns visible in the electron image (Figure 1), separated by the photon energy (~ 1.5 eV) in the energy scale, represent the above-threshold ionization (ATI) process.

Figure 1. Electron image obtained for CS_2 in an intense laser field (1.5 × 10^{13} W/cm^2, 35 fs, 800 nm). In order to to emphasize weak features, a logarithmic intensity scale is adopted. The concentric ring patterns represent the ATI process.