VI-C Ultrafast Dynamics of Molecules in Intense Laser Fields

The behavior of molecules in an intense laser field (10^{12}–10^{18} W/cm^2) has been an attractive target of research for a deeper understanding of the light-matter interaction. In the present study, the characteristic nuclear dynamics occurring in intense laser fields, such as structural deformation and multiple breaking of chemical bonds, has been studied by a newly developed experimental method, called *coincidence momentum imaging* (CMI), which allows us to determine the momentum vectors of all the fragment ions ejected from a single parent molecule. By using the CMI technique combined with the pump-and-probe scheme, the real-time probing of the nuclear wavepacket evolution in the three-dimensional internal coordinate space is demonstrated. Based on this novel technique, it has been found that CS_2^{2+} formed in intense laser fields (~1.3×10^{14} W/cm^2) undergoes ultrafast dissociation along the symmetric stretching coordinate leading to simultaneous breaking of the two C–S bonds. A high temporal resolution pump-probe CMI measurement is under progress with sub-10 fs intense laser pulses.

VI-C-1 Probing the Ultrafast Nuclear Motion in CS_2^{2+} in Intense Laser Fields

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The nuclear dynamics of CS_2 exposed to an intense laser field (60 fs, 1.3×10^{14} W/cm^2) is studied by the pump-probe coincidence momentum imaging of the Coulomb explosion process, $CS_2^{3+} \to S^+ + C^+ + S^+$. From the dependence of momentum correlations among the fragment ions on the time delay between pump and probe pulses, the existence of the dissociation pathway along the symmetric stretching coordinate leading to concerted breaking of the two C–S bonds is identified in addition to the dissociation along the anti-symmetric stretching leading to $S^+ + CS^+$. It is also shown that the S–C–S bending motion is largely excited when both of the two C–S bonds stretch by the coupling of two different light-dressed states.

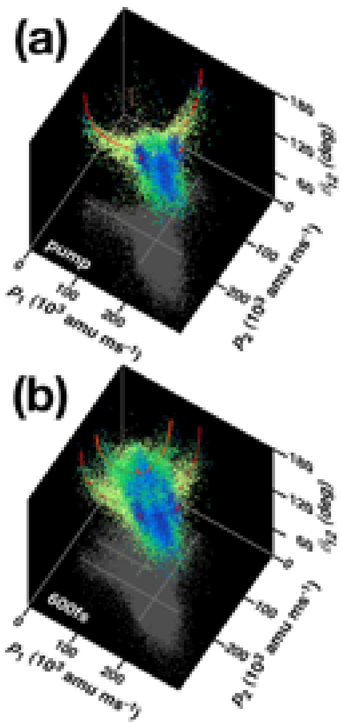

Figure 1. (a) Three-dimensional momentum correlation map for the Coulomb explosion process, $CS_2^{3+} \to S^+ + C^+ + S^+$, obtained only with a pump pulse (1.3×10^{14} W/cm^2, 60 fs, 800 nm), where θ_{12} represents the angle between the two momentum vectors $\boldsymbol{p}_1(S^+)$ and $\boldsymbol{p}_2(S^+)$ of the resultant S^+ ions, and $p_1 = |\boldsymbol{p}_1(S^+)|$ and $p_2 = |\boldsymbol{p}_2(S^+)|$. Solid lines represent the result of the simulation based on the classical free-rotor model. (b) Momentum correlation map at a time delay of Δt = 600 fs, exhibiting new features with a pair of wings in the low momentum region (p_1, $p_2 < 150 \times 10^3$ amu m/s).

VI-C-2 Concerted and Sequential Coulomb Explosion Processes of N_2O in Intense Laser Fields by Coincidence Momentum Imaging

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The Coulomb explosion dynamics of N_2O in intense laser fields (800 nm, 60 fs, ~1.6×10^{14} W/cm^2) is studied by the coincidence momentum imaging method. From the momentum correlation maps obtained for the three-body fragmentation pathway, $N_2O^{3+} \to N^+ + N^+ + O^+$, the ultrafast structural deformation dynamics of N_2O prior to the Coulomb explosion is extracted. It is revealed that the internuclear N–N and N–O distances stretch simultaneously as the bond angle ∠N–N–O decreases. In addition, two curved thin distributions are identified in the momentum correlation maps, and are interpreted well as those originating from the sequential dissociation pathway, $N_2O^{3+} \to N^+ + NO^{2+} \to N^+ + N^+ + O^+$.

VI-C-3 Development of an Intense Sub-10fs Laser Source with a Hollow Fiber/Chirped Mirror Compressor

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An intense ultrashort laser source was designed and developed to study the Coulomb explosion dynamics of molecules in a sub-10fs intense laser field. The output from a Ti:Sapphire laser system (800 nm, <40 fs, 1 kHz) was introduced to a hollow fiber placed on a V-shape block in a cell, filled with Ar at a pressure of ~0.1 MPa. During the propagation of the laser through the fiber, the spectral bandwidth was increased up to by the self-phase modulation effect (Figure 1(a)). The output from the hollow fiber was then collimated by a concave mirror and compressed by a pair of chirped mirrors. The pulse duration after the compression was measured to be 9.0 fs from the interferometric autocorrelation trace (Figure 1(b)). The output energy from the pulse compression system exceeds 0.4 mJ/pulse, which is sufficient to generate a field intensity of ~10^{16} W/cm^2 with an F/5 focusing optics.

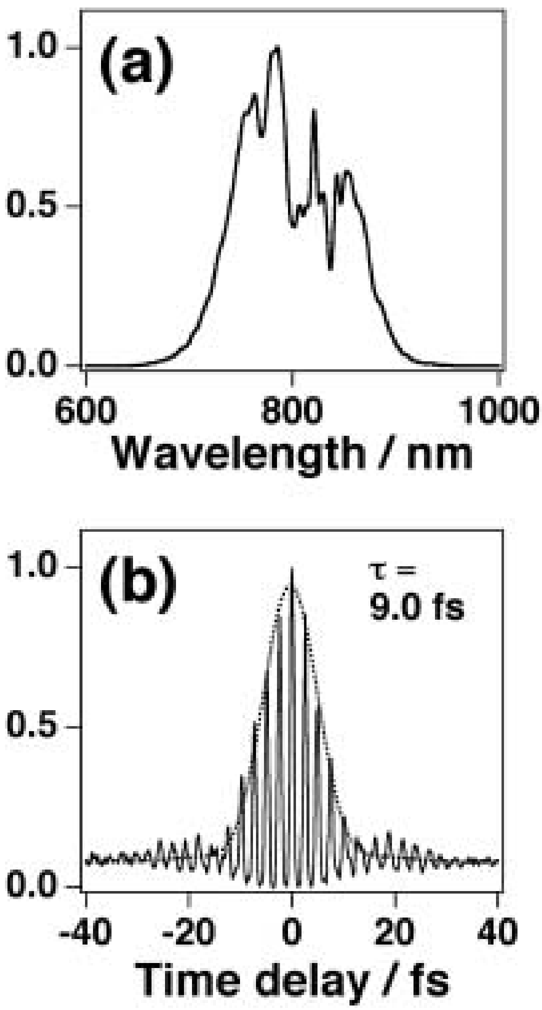

Figure 1. (a) The laser spectrum after the propagation through the hollow fiber. (b) The autocorrelation trace of the laser pulse after the compression by the chirped mirrors. The autocorrelation intensity profile obtained by Fourier transform of the spectrum (a) is plotted with a dotted curve. The pulse duration is determined to be 9.0 fs.