Atoms & Molecules in Intense Laser Fields

Department of Photo-Molecular Science
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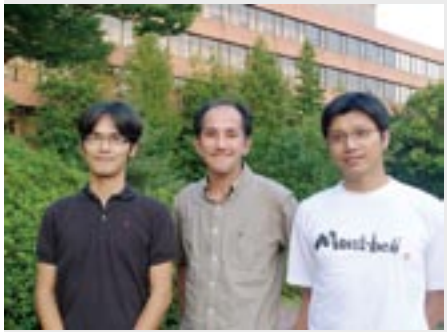

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Intense laser fields, comparable in magnitude with the Coulomb field within atoms and molecules, can be generated by focussing high-energy and ultrashort laser pulses. When exposed to such a strongly perturbing field, molecules exhibit various exotic features that cannot be observed in weak laser fields. We are seeking a deeper understanding of the behavior of molecules in intense laser fields, to elucidate how molecules interact with light, as well as to apply the new features they exhibit to the real-time visualization of ultrafast chemical reactions and their control. In particular, we focus on the following research themes:

(1) Understanding of atomic and molecular dynamics in intense laser fields and the control
(2) Ultrafast reaction imaging by laser Coulomb explosion imaging
(3) Generation and application of ultrashort soft-X-ray pulses by laser high-order harmonics.

1. Electron-Ion Coincidence Momentum Imaging of Molecular Dissociative Ionization in Intense Laser Fields: Application to CS_2[1)]

Molecules in intense laser fields show characteristic reaction processes, such as above-threshold dissociation (ATD), bond softening/hardening and deformation of the geometrical structures, associated with rearrangement of the electron distribution. Here, we performed photoelectron spectroscopy of the dissociative ionization of CS_2 to clarify the electron dynamics in intense laser fields. We developed a new electron–ion coincidence imaging system to label each photoelectron with the counterpart ion, so that the electron dynamics for different pathways can be discussed separately from the corresponding photoelectron images.

Photoelectron images of CS_2 in 35 fs intense laser fields (2 $\times 10^{13}$ W/cm^2, 800 nm, linearly polarized) recorded in coincidence with the parent ion, CS_2^+, show clear concentric ring patterns due to the above-threshold ionization (ATI) process. On the other hand, broad structure-less distributions elongated along the direction of the laser polarization are observed in the coincidence electron images for the CS^+ and S^+ fragment ions. The difference in the electron images indicates that the dissociative ionization does not proceed sequentially by the formation and photodissociation of CS_2^+ in intense laser fields. The origin of the difference in the spectra was discussed in terms of the direct ionization to the dissociative state and of the (multiple) electron recollision process.

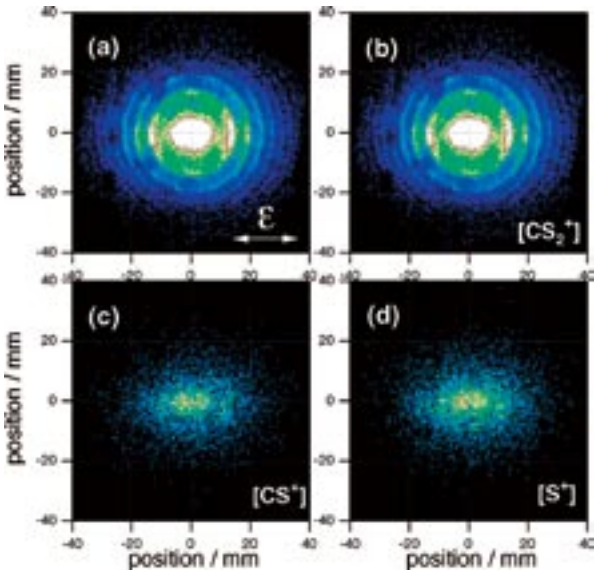

Figure 1. Photoelectron images of CS_2 recorded at a field intensity of 2.1×10^{13} W/cm^2 (800 nm, 35 fs) (a) and those recorded in coincidence with CS_2^+ (b), CS^+ (c) and S^+ (d). The concentric ring patterns appearing in (a) are the ATI peaks. The laser pulses are linearly polarized. The total electron-ion coincidence count rate is less than 0.03 per laser shot.

2. Multiple Explosion Pathways of the Deuterated Benzene Trication in 9-fs Intense Laser Fields[2)]

The formation and fragmentation of multiply charged molecular ions are commonly observed as the responses of molecules exposed to intense laser fields. The fragmentation dynamics in these interactions have been extensively studied in recent years as they provide insight into the interplay between the electron and nuclear dynamics with large degrees of nuclear freedom in the presence of strong alternating electric fields. Here we employ the coincidence momentum imaging technique to clarify the fragmentation process of benzene in 9 fs intense laser fields (1×10^{15} W/cm^2). By using this technique we can unambiguously identify the Coulomb explosion pathways from one particular charge state of interest and determine the momenta of the fragment ions in the laboratory frame.

Five two-body and eight three-body Coulomb explosion pathways from the trication ($C_6D_6^{3+}$), associated with the deprotonation and ring-opening reactions, are identified. It is found from the fragment momentum correlation that all the observed three-body explosion processes proceed sequentially via the two-body Coulomb explosion forming molecular dications, $C_mD_n^{2+}$, with (m,n) =(6,5), (5,5), (5,4), (4,4), (4,3) and (3,3), which further dissociate into pairs of monocations (see Figure 2). The branching ratios of the observed Coulomb explosion pathways, estimated from the number of the corresponding coincidence events, largely deviate from the previous RRKM prediction, showing that the dissociation processes are not fully statistical.

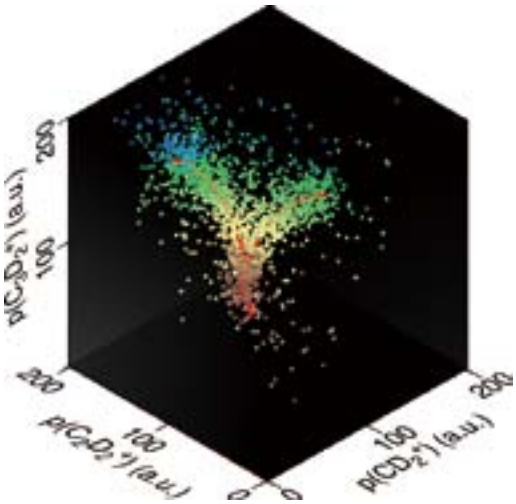

Figure 2. Three-dimensional momentum correlation map, plotting all the coincidence events identified for the three-body Coulomb explosion pathway, $C_6D_6^{3+} \rightarrow C_3D_2^+ + C_2D_2^+ + CD_2^+$, in intense laser fields (9 fs, 1×10^{15} W/cm^2). The results of simulations based on the free-rotor model for the sequential three-body explosion are shown by the red solid lines.

3. Development of Time-Resolved Reaction Imaging System with Laser High-Order Harmonics[3)]

Laser high-order harmonics have novel features such as i) photon energy higher than several 100 eV, ii) extremely short pulse duration in the sub-fs regime, iii) high-quality spatio-temporal coherence, iv) high photon flux comparable to synchrotron radiation and v) simple and precise synchronization with other laser light sources. Because of these aspects, the laser high-order harmonic pulses are of great interest as a potential light source for time-resolved spectroscopy of ultrafast dynamics that could not be elucidated in real time by conventional techniques.

Figure 3 shows the schematic of new time-resolved reaction imaging system developed in our group. It consists of a laser high-order harmonics source, a soft X-ray spectrograph, a beam line equipped with a time delay line as well as a pair of dielectric mirrors to select one of the harmonic orders, and an electron-ion coincidence imaging chamber. The photoelectron spectra recorded for Xe with the 59 th harmonics (~92 eV) show the 4d inner-core photoelectron and the associated Auger electron peaks. Real-time probing of chemical reaction dynamics using this system is in progress.

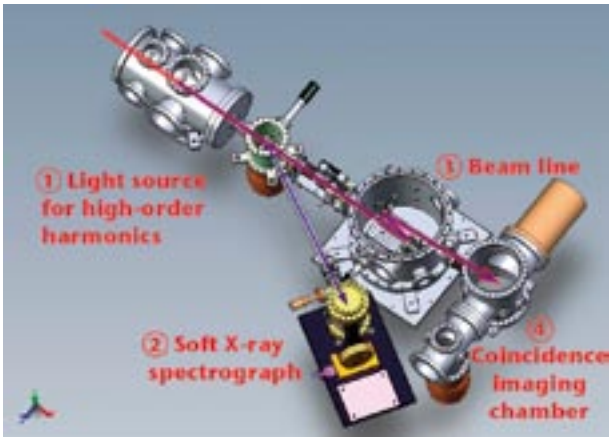

Figure 3. Schematic of time-resolved reaction imaging system developed at IMS, consisting of (i) a high-order harmonics source, (ii) a soft X-ray spectrograph, (iii) a beam line and (iv) a coincidence momentum imaging chamber.

References

1) A. Matsuda, M. Fushitani and A. Hishikawa, *J. Electron Spectrosc. Relat. Phenom.* **169**, 97–101 (2009).
2) A. Matsuda, M. Fushitani, R. D. Thomas, V. Zhaunerchyk and A. Hishikawa, *J. Phys. Chem. A* **113**, 2254–2260 (2009).
3) M. Fushitani, A. Matsuda, E. J. Takanashi and A. Hishikawa, *J. Phys: Conf. Series* **185**, 012009 (4 pages) (2009).