

Dynamics of Atoms & Molecules in Intense Laser Fields

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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 directions:

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

1. Development of Novel Photoelectron–Photoelectron Coincidence Spectrometer

Molecules in intense laser fields show characteristic reaction processes, such as above-threshold dissociation (ATD), bond softening/hardening and deformation of the geometrical structures. Since these interesting phenomena are all associated with rearrangement of the electron distribution by the intense laser fields, photoelectrons should provide direct information on how these reactions are induced. Here we developed a novel electron–electron–ion coincidence spectrometer to detect a pair of photoelectrons emitted simultaneously from a single molecule to understand the electron correlation dynamics in intense laser fields.

The spectrometer is based on a magnetic bottle type photoelectron spectrometer (see Figure 1). A strong permanent magnet located close to the FEL focus point provides an inhomogeneous magnetic field, which serves as a magnetic mirror for photoelectrons. The reflected photoelectrons are hence guided by the weak magnetic field of a long solenoid

toward a microchannel plate (MCP) detector placed at the end of a 1.5 m flight pass. The high collection efficiency with a 4π -sr detection angle and the long light pass enables us to detect pairs of electrons emitted from a single atom. In order to securely identify the counter part ion, a pulsed high voltage is applied to the permanent magnet and ion collection electrodes to guide the ions to the same MCP detector used for electrons. The spectrometer was applied to double ionization of CS_2 in IR (1.030 μm) intense laser fields, to reveal the ejection of the two electrons mostly proceeds sequentially at a field intensity of $2.8 \times 10^{13} \text{ W/cm}^2$.

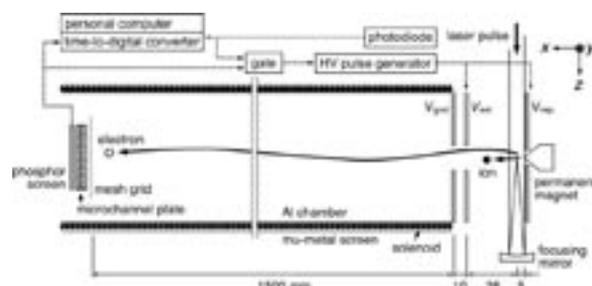


Figure 1. Schematic of photoelectron coincidence spectrometer equipped with collection electrodes for ion detection.

2. Visualization of Ultrafast Molecular Isomerization by Laser Coulomb Explosion Imaging

Novel features of molecules in intense laser fields can be utilized for new applications. Laser Coulomb explosion imaging is one such application, which provide a unique opportunity for real-time probing of large-scale structural changes in ultrafast chemical reactions.

Figure 2 shows the Newton diagram of three-body Coulomb explosion pathway of formaldehyde, $\text{H}_2\text{CO}^{3+} \rightarrow \text{H}^+ + \text{H}^+ + \text{CO}^+$. The diagram observed for the 7 fs case shows a clear and dense distribution of the H^+ ion, located mostly in the first and fourth quadrant. The corresponding distribution for

the 35 fs case exhibits a significant broadening extending towards smaller momentum angle $\theta_{\text{H-H}}$. This finding shows that, when few-cycle intense laser pulses (7 fs) are used, the geometrical structure of the molecule is almost frozen along the H-C-H bending coordinate during the interaction with the laser pulse. On the other hand, significant deformation of the geometrical structure both along the C-H stretching and H-C-H bending coordinates is observed for the 35 fs laser pulses. The origin of the structural change can be understood in terms of the nuclear dynamics in the dication states populated in the rising edge of the laser pulse.

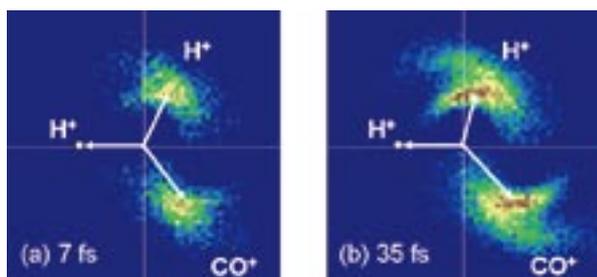


Figure 2. The Newton diagrams of $\text{H}_2\text{CO}_3^+ \rightarrow \text{H}^+ + \text{H}^+ + \text{CO}^+$ in (a) 7 fs and (b) 35 fs intense laser fields, respectively. The amplitude of the linear momentum of the first H^+ is normalized to unity and the normalized vector is placed on the negative x -axis.

3. Multiphoton Double Ionization of Ar in Intense Extreme Ultraviolet Laser Fields Studied by Shot-by-Shot Photoelectron Spectroscopy¹⁾

Non-linear optical processes in extreme ultraviolet (EUV) and X-ray laser fields have attracted increasing attention after the advent of intense ultrashort free electron lasers (FEL), for their importance in a variety of applications, such as the creation of warm dense matter, optical switching and imaging of single bio-molecules. In addition to experiments on bulk materials, a number of experimental studies are devoted to clarify non-linear responses of isolated atoms, as they serve as an ideal benchmark for basic understanding on the non-linear EUV process in which both valence and core electrons can participate. We demonstrate that the shot-by-shot spectral monitoring of SASE FEL pulses and photoelectrons provides a clear understanding on the atomic responses to the intense EUV field. We have applied this technique to the multi-photon multiple ionization of Ar irradiated with SASE FEL pulses of mean photon energies around 21 eV. The sequential two-step double ionization due to three-photon absorption, (i) $\text{Ar} + h\nu \rightarrow \text{Ar}^+ + e^-$ and (ii) $\text{Ar}^+ + 2h\nu \rightarrow \text{Ar}^{2+} + e^-$, is unambiguously identified as the dominant multiphoton pathway in a laser field of $\sim 5 \text{ TW/cm}^2$. Moreover, the shot-by-shot monitoring of the mean photon energies of the fluctuating FEL spectra clearly uncovered the effect of resonances in the sequential double ionization.

The $\text{Ar}^{2+} 3p^{-2}$ structures shown in Figure 3 clearly show that both the peak intensities and profiles sensitively depend on the photon energy. This observation manifests the presence

of intermediate resonances in the two-photon ionization of Ar^{2+} . In fact, excited Ar^+ states of the $[3p^{-2}(^1D)]3d$ configuration lie close to the photoabsorption from $\text{Ar}^+ 3p^{-1}$ as shown in Figure 3, and these Ar^{+*} states can generate resonances in the two-photon ionization of Ar^+ . The intensity evolutions are dependent on the final Ar^{2+} levels, which implies that Ar^{+*} resonances are not common in the formations of these different Ar^{2+} levels.

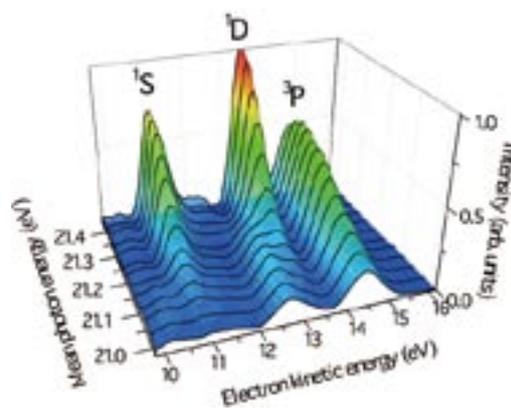


Figure 3. Photoelectron spectra in the range of the $\text{Ar}^{2+} 3p^{-2}$ peaks, derived from a single-shot-basis analysis. In the analysis, the single-shot photoelectron spectra obtained at three different nominal FEL photon energies of 21.0, 21.2 and 21.4 eV were sorted by the mean photon energies determined from the $\text{Ar}^+ 3p^{-1}$ peak energies, and then averaged within narrow photon energy ranges that divide the whole photon energy range into 13 segments.

4. Generation of Laser High-Order Harmonics and Application for Ultrafast Reaction Probing

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. Preliminary pump-probe experiments on the photodissociation of Br_2 show a steep rise (~ 80 fs) of the $\text{Br}(^2P_{3/2})$ yields, reflecting a ultrashort pulse duration (< 20 fs) of the 27th harmonic pulses used to probe the $\text{Br}(^2P_{3/2})$ product. Full characterization of the temporal profile of the harmonic pulses and application to the time-resolved photoelectron imaging are in progress.

Reference

- 1) Y. Hikosaka, M. Fushitani, A. Matsuda, C.-M. Tseng, A. Hishikawa, E. Shigemasa, M. Nagasono, K. Tono, T. Togashi, H. Ohashi, H. Kimura, Y. Senba, M. Yabashi and T. Ishikawa, *Phys. Rev. Lett.* **105**, 133001 (2010).