1 Sakai (Hirofumi) Group

Research Subjects: Experimental studies of atomic, molecular, and optical physics

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Our research interests are as follows: (1) Manipulation of neutral molecules based on the interaction between a strong nonresonant laser field and induced dipole moments of the molecules. (2) High-intensity laser physics typified by high-order nonlinear processes (ex. multiphoton ionization and high-order harmonic generation). (3) Ultrafast phenomena in atoms and molecules in the attosecond time scale. (4) Controlling quantum processes in atoms and molecules using shaped ultrafast laser fields. A part of our recent research activities is as follows:

(1) All-optical control of pendular qubit states with nonresonant two-color laser pulses [1]

Practical methodologies for quantum qubit controls are established by two prerequisites, i.e., preparation of a well-defined initial quantum state and coherent control of that quantum state. Here we propose a new type of quantum control method, realized by irradiating nonresonant nanosecond two-color (ω and 2ω) laser pulses to molecules in the pendular (field-dressed) ground state. The two-color field nonadiabatically splits the initial pendular ground state $|\tilde{0},\tilde{0}\rangle$ to a superposition state of $|\tilde{0},\tilde{0}\rangle$ and $|\tilde{1},\tilde{0}\rangle$, whose relative probability amplitudes can be controlled by the peak intensity of one wavelength component (ω) while the peak intensity of the other component (2ω) is fixed. The splitting of the quantum paths is evidenced by observing degrees of orientation of ground-state selected OCS molecules by the velocity map imaging technique. This quantum control method is highly advantageous in that any type of polar molecules can be controlled regardless of the molecular parameters, such as rotational energy, permanent dipole moment, polarizability, hyperpolarizability, and hyperfine energy structures.

(2) Ultrafast X-ray photoelectron diffraction from free molecules: Simulations of diffraction profiles from transient intermediates in the elimination reaction of $C_2H_4I_2$ [2]

We have performed the simulations of C1s X-ray photoelectron diffraction (XPD) profiles from $C_2H_4I_2$, bridged and classical anti-forms of C_2H_4I intermediates and C_2H_4 products to capture structures of transient intermediates in the elimination reaction of $C_2H_4I_2$, under our ultrafast X-ray photoelectron diffraction (UXPD) scheme for free molecules using soft X-ray free-electron laser (SXFEL). In the UXPD scheme, the sample molecules are aligned in advance by near-infrared (NIR) laser with ns pulse duration before applying a pump-probe method. Then, we have considered alignment control of $C_2H_4I_2$ by using the elliptically polarized NIR laser to realize the UXPD experiments for the free molecules. As the results of simulations of XPD profiles from the laser-aligned $C_2H_4I_2$ molecules, we have demonstrated the two-dimensional (2D) color maps of the C1s XPD profiles from $C_2H_4I_2$, C_2H_4I, and C_2H_4. The 2D color maps have revealed that the transient C1s XPD profiles from the bridged-form and classical anti-form C_2H_4I intermediates exhibit remarkable differences, reflecting different intra-molecular scattering pathways of C1s photoelectrons within the intermediates. Thus, the present result has proved that UXPD for the free molecules has an advantage, compared with other traditional diffraction methods.

[1] Je Hoi Mun, Shinichirou Minemoto, Dong Eon Kim, and Hirofumi Sakai, "All-optical control of pendular qubit states with nonresonant two-color laser pulses," Commun. Phys. **5**, 226(7 pages) (2022).

[2] S. Minemoto, J. H. Mun, T. Teramoto, A. Yagishita, and S. Tsuru, "Ultrafast X-ray photoelectron diffraction from free molecules: Simulations of diffraction profiles from transient intermediates in the elimination reaction of $C_2H_4I_2$," J. Electron Spectrosc. Relat. Phenom. **258**, 147221(7 pages) (2022).

[3] T. Teramoto, S. Minemoto, T. Majima, T. Mizuno, J. H. Mun, A. Yagishita, P. Decleva, and S. Tsuru, "Basic studies toward ultrafast soft x-ray photoelectron diffraction; its application to probing local structure in iodobenzene molecules," Struct. Dyn. **9**, 024303(12 pages) (2022).