### 1 Sakai (Hirofumi) Group

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

#### Members: Hirofumi Sakai and Shinichirou Minemoto

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) Electron-wave-packet dynamics extracted from the ellipticity dependence of high-order harmonic generation in benzene molecules [4]

We measure the ellipticity dependence of high-order harmonic intensities generated from benzene ( $C_6H_6$ ) molecules with and without an yttrium aluminum garnet (YAG) laser field. We successfully extract the expansion dynamics of the electron wave packet by analyzing the ellipticity dependence based on the semiclassical electron trajectory. Without the YAG laser pulse, we find that the ellipticity dependence reflects the expansion dynamics of the electron wave packet and the difference in highest occupied molecular orbital compared to nitrogen (N<sub>2</sub>) molecules. We also measure the ellipticity dependence under the YAG laser field and show that the difference in the ellipticity dependence with and without the YAG laser field is qualitatively explained by the harmonic-order-dependent efficiency of the sum and difference frequency generation, whose efficiency is higher as the harmonic order becomes higher.

## (2) Improving molecular orientation by optimizing relative delay and intensities of two-color laser pulses [5]

We numerically explore molecular orientation dynamics with moderately intense nanosecond two-color laser pulses. It is believed that the nanosecond two-color pulse can adiabatically control the molecular orientation. However, in our simulation based on the time-dependent Schrödinger equation, which naturally includes nonadiabatic effects, the orientation dynamics shows clear deviation from the adiabatic approximation (AA) results, while the molecular alignment dynamics is in good agreement with the AA results. The nonadiabaticity is significantly influenced by three parameters, the intensities, and the relative delay of the two wavelengths. In this work, we clarify the reason behind the nonadiabaticity and provide the solution for achieving higher degrees of orientation.

- Hiroyuki Shimada, Kazma Komatsu, Wataru Komatsubara, Tomoya Mizuno, Soichiro Miyake, Shinichirou Minemoto, Hirofumi Sakai, Takuya Majima, Shigeki Owada, Tadashi Togashi, Makina Yabashi, and Akira Yagishita, "Two- and three-photon double ionization of helium by soft x-ray free-electron laser pulses," J. Phys. B 52, 065602 (2019) (8 pages).
- [2] Shinichirou Minemoto, Hiroyuki Shimada, Kazma Komatsu, Wataru Komatsubara, Takuya Majima, Soichiro Miyake, Tomoya Mizuno, Shigeki Owada, Hirofumi Sakai, Tadashi Togashi, Makina Yabashi, Piero Decleva, Mauro Stener, Shota Tsuru, and Akira Yagishita, "Time-resolved photoelectron angular distributions from nonadiabatically aligned CO<sub>2</sub> molecules with SX-FEL at SACLA," J. Phys. Commun. 2, 115015 (2018) (8 pages).
- [3] Hiroyuki Shimada, Shinichirou Minemoto, Kazma Komatsu, Wataru Komatsubara, Shintaro Yoshida, Takuya Majima, Tomoya Mizuno, Hirofumi Sakai, Shigeki Owada, Tadashi Togashi, Makina Yabashi, and Akira Yagishita, "Photoelectron spectroscopy of Rydberg excited states in singly charged molecular ions CS<sub>2</sub><sup>+</sup> by NIR laser pulses," J. Phys. B **51**, 225601 (2018) (8 pages).
- [4] Wataru Komatsubara, Shinichirou Minemoto, and Hirofumi Sakai, "Electron-wave-packet dynamics extracted from the ellipticity dependence of high-order harmonic generation in benzene molecules," Phys. Rev. A 98, 023416 (2018) (8 pages).
- [5] Je Hoi Mun and Hirofumi Sakai, "Improving molecular orientation by optimizing relative delay and intensities of two-color laser pulses," Phys. Rev. A 98, 013404 (2018) (8 pages).