

# 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) Characteristics of high-order harmonics generated from atoms and aligned molecules with carrier-envelope-phase-stabilized 25-fs pulses [1]**

With carrier-envelope phase stabilized pulses, the self-referencing technique is applied to evaluate the relative phase of high-order harmonics generated in atoms and aligned molecules. The Fourier transform analysis from the frequency domain to the time domain shows that the *effective* duration of the driving pulse, during which the specific orders of harmonics are efficiently generated, is decreased as the harmonic order is increased. In the case of aligned molecules, the interference fringes between the two adjacent odd-order harmonics from N<sub>2</sub> are more distinctive than those from CO<sub>2</sub>, which may be explained by the difference in the complexity associated with the symmetry of highest occupied molecular orbitals between N<sub>2</sub> and CO<sub>2</sub>.

**(2) High-order harmonics generated from aligned molecules with carrier-envelope-phase-stabilized 10-fs pulses**

With carrier-envelope-phase-stabilized 10-fs pulses, harmonic spectra from aligned molecules are observed and analyzed. The control of the carrier-envelope phase allows us to observe spectral interferences caused by nonadiabatic change in the intensity of the driving pulse. The Fourier transform analysis reveals that there are clear contributions from the spectrally broadened short and long trajectory components in addition to the contribution from the usual odd-order harmonic components. We further examine the possibility of evaluating the phase change accompanied by the destructive interference in harmonic spectra observed in aligned CO<sub>2</sub> and N<sub>2</sub> molecules.

**(3) Laser-field-free orientation of state-selected asymmetric top molecules**

With combined electrostatic and shaped laser fields with a slow turn on and rapid turn off, laser-field-free orientation of asymmetric top iodobenzene molecules with higher degrees of orientation has been achieved for the first time. In order to further increase the degrees of orientation, state-selected molecules are used as a sample. It is confirmed that higher degrees of orientation are maintained in the laser-field-free condition for about 10 ps, which is long enough to study femtosecond-attosecond dynamics in molecules, after the rapid truncation of the laser pulse. This accomplishment means not only that a unique molecular sample has become available in various applications but also that the present technique can be used as a new approach to investigate rotational dynamics of molecules.

- [1] Yusuke Sakemi, Kosaku, Kato, Shinichirou Minemoto, and Hirofumi Sakai, "Characteristics of high-order harmonics generated from atoms and aligned molecules with carrier-envelope-phase-stabilized 25-fs pulses," *Physical Review A* **85**, 051801(R) (4 pages) (2012). Selected for *Virtual Journal of Ultrafast Science* Vol. 11, Iss. 6 (2012).