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) All-optical orientation of linear molecules with combined linearly and elliptically polarized two-color laser fields [1]

We show that a combination of a fundamental pulse with linear polarization along the vertical direction and an elliptically polarized second harmonic pulse with both vertical and horizontal electric field components can be used to orient linear molecules efficiently, leading to higher degrees of orientation. Due to this specific combination of polarizations, the asymmetric hyperpolarizability interaction potential, which remains the same as that in a linearly polarized two-color laser field, is created along the vertical component of the elliptically polarized second harmonic pulse. On the other hand, the horizontal component suppresses the otherwise strong symmetric polarizability potential responsible for alignment, increasing the tunneling probability from the shallower potential well to the deeper one. As a result, the degree of orientation increases and can be controlled by changing the intensity of the horizontal component of the elliptically polarized second harmonic pulse. This study is the generalization of the all-optical molecular orientation technique based on the anisotropic hyperpolarizability interaction.

(2) Comparative studies of the degrees of orientation of CO molecules pumped by intense femtosecond two-color pulses based on high-order harmonic generation and Coulomb explosion imaging [2]

With an intense femtosecond two-color pulse optimized for the generation of even-order harmonics from CO molecules, we directly measure the actual degrees of orientation by utilizing the Coulomb explosion imaging technique with appropriate probe polarization. We find that the macroscopic orientation of CO molecules is negligible even when significant even-order harmonics are observed. This finding shows that the generation of even-order harmonics cannot be ascribed to the macroscopic orientation of CO molecules. The rotational wave packet of CO molecules created with an intense femtosecond two-color pulse is thought to be in an uninvestigated quantum state, which cannot be explained by the theoretical model based on the Born-Oppenheimer approximation, without inversion symmetry at any of the three steps of high-order harmonic generation, leading to the generation of even-order harmonics.

[1] Md. Maruf Hossain and Hirofumi Sakai, "All-optical orientation of linear molecules with combined linearly and elliptically polarized two-color laser fields," J. Chem. Phys. **153**, 104102(11 pages) (2020).

[2] Shinichirou Minemoto, Wataru Komatsubara, and Hirofumi Sakai, "Comparative studies of the degrees of orientation of CO molecules pumped by intense femtosecond two-color pulses based on high-order harmonic generation and Coulomb explosion imaging," J. Phys. B **53**, 235101(11 pages) (2020).