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) Stronger orientation of state-selected OCS molecules with relative-delay-adjusted nanosecond two-color laser pulses [1]

Using the all-optical molecular orientation technique with intense nonresonant two-color laser pulses, stronger molecular orientation $|\langle \cos\theta \rangle| \sim 0.34$ is achieved by employing the following two strategies: (1) carbonyl sulfide molecules lying in the lower rotational states are selected using a home-built molecular deflector and (2) the rising parts of the two wavelengths of the pump pulse are adjusted by introducing a Michelson-type delay line in the optical path. The achieved degree of molecular orientation is higher than that observed in the proof-of-principle experiment [Oda *et al.*, Phys. Rev. Lett. **104**, 213901 (2010)] by about an order of magnitude and the highest ever characterized directly by Coulomb explosion imaging with appropriate probe polarization.

(2) All-optical control of pendular qubit states with nonresonant two-color laser pulses [2]

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.

[1] Md. Maruf Hossain, Xiang Zhang, Shinichirou Minemoto, and Hirofumi Sakai, "Stronger orientation of state-selected OCS molecules with relative-delay-adjusted nanosecond two-color laser pulses," J. Chem. Phys. **156**, 041101(7 pages) (2022).

[2] Je Hoi Mun, Shinichirou Minemoto, Dong Eon Kim, and Hirofumi Sakai, "All-optical control of pendular qubit states with nonresonant two-color laser pulses," to appear in Commun. Phys. (8 pages) (2022).