Quantum Frontiers

Molecular science in intense laser fields, Attosecond science, and Quantum computing

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Quantum Frontiers Laboratory was established in April 2023 to take over the activities of the Quantum Chemistry Laboratory until March 2023, and to further expand the frontier of molecular science. Professor Yamanouchi, the laboratory's group leader, is promoting the research activities in "Next Generation Laser (ATTO Division)," which was started in 2017 as one of the 10-year term MEXT "Quantum leap flagship program," as the division head, to explore frontiers in attosecond science by developing attosecond light sources and advanced measurement systems in Japan.

We have been making an effort to establish "Attosecond Laser Facility," an international user facility for frontier research in attosecond science, in cooperation with the Institute for Attosecond Laser Facility, established in 2022 under the Committee for Presidential Initiative of the University of Tokyo, and the Center for Attosecond Laser Science, School of Science.

Furthermore, in addition to promoting ultrafast intense laser field science [1,2] and attosecond science [2,3], we have been conducting a research project on quantum computing, "Qudit-boson quantum-operation realization by trapped ions (2023–)," to develop quantum algorithms and construct a trapped-ion-type quantum computer in collaboration with RIKEN, Institute of Science Tokyo, Tokyo Metropolitan University, DIC Corporation, and Hamamatsu Photonics K.K. under the CREST Quantum Frontiers Program.

Ultrafast molecular dynamics in intense laser fields

Recent advances in laser technology made it possible to generate an ultrashort-pulsed intense laser field whose field intensity is as high as 10^{15}-10^{16} W/cm^2, which is comparable with or even larger than the Coulombic field within atoms and molecules. In such an intense laser field, electrons in atoms and molecules are influenced largely by the external electric field of light, and consequently, the subsequent dynamics of atoms and molecules are sensitively dependent on the characteristics of the light pulse. From the investigation of atoms and molecules interacting with an intense laser field, we can learn fundamental aspects of a light-matter interaction (Fig. 1).

In the past few decades, our group has been investigating the following research subjects; (1) "ultrafast hydrogen migration" in hydrocarbon molecular ions by the "coincidence momentum imaging" method, which records momentum vector distributions of fragment ions generated through the Coulomb explosion in an intense laser field, (2) strong-field ultra-high resolution Fourier-transform spectroscopy, (3) "laser-assisted elastic electron scattering" to investigate ultrafast electron dynamics within atoms and molecules in an intense laser field and "laser-assisted electron diffraction" to probe ultrafast variation of the geometrical structure of molecules with the femtosecond temporal resolution, (4) attosecond spectroscopy in which photoelectrons and photo-ions are detected after molecules are ionized by high-order harmonics generated using few-cycle laser pulses, (5) excitation processes of atoms and molecules in laser-induced filaments, which are formed by focusing intense laser pulses into gaseous media, (6) the mechanism of population inversion in N_2^+ and lasing, (7) theory for describing atoms and molecules and their dynamics in an intense laser field, (8) application of quantum computing to atomic and molecular problems, and (9) material processing by the extreme ultraviolet (EUV) femtosecond laser pulses.

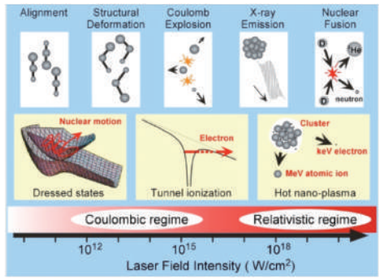

Figure 1. Dynamics of molecules in intense laser fields [1, 2].

Real-time measurements of ultrafast molecular dynamics and SURF spectroscopy

When molecules are irradiated with extreme ultraviolet (EUV) light, molecular ions are generated not only in the electronic ground state but also in the electronically highly excited states upon the ionization. To probe in real time the vibrational and dissociation processes of molecular ions in the electronically highly excited states, we have constructed a pump-probe beamline that can irradiate a sample gas with few-cycle near-IR laser pulses and EUV light pulses generated as high-order harmonics of the few-cycle laser pulses and developed a velocity map imaging spectrometer that detects fragment ions with a position-sensitive detector. Using this experimental setup, we have performed pump-probe measurements with an O_2 sample gas and measured the delay-time dependence of the kinetic energy distribution of O^+ generated via the dissociation of O_2^+ (Fig. 2). By numerical simulations, we have shown that O_2^+ oscillates in the $a^4\Pi_u$ state with a period of about 40 fs and dissociates in the $3^2\Pi_u$ state over a period of several tens of femtoseconds [4].

By pump-probe coincidence momentum imaging measurements of methanol, CH_3OH, using few-cycle intense laser pulses, we revealed that the yield of the H_3^+ periodically oscillates as a function of the delay time and that the period of the recorded oscillation (~38 fs) obtained by the Fourier transform corresponds to the period of the C-O stretching vibration of methanol cation [5]. By extending the delay time range of the Fourier-transform spectroscopy, we determined the vibrational frequency of D_2^+ to a precision of 10^{-4} cm^{-1} (Fig. 3) [6]. By the method of strong-field ultra-high-resolution Fourier-transform (SURF) spectroscopy with an extended delay time range of 500 ps, we determined the spin-orbit splitting energies of rare gas atoms to a relative precision of 10^{-7}. Furthermore, we have extended the delay time range up to 13 ns by introducing a long-arm interferometer and determined the hyperfine structure in the spin-orbit levels of $^{83}Kr^+$ with a precision of 10^{-5} cm^{-1} [8].

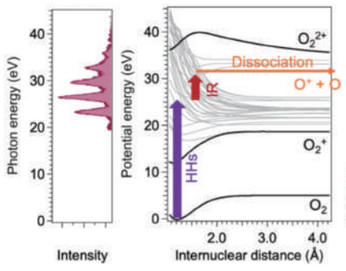

Figure 2. Pump (EUV) — probe (near IR) measurements of ultrafast dissociation dynamics of electronically highly excited states of O_2^+.

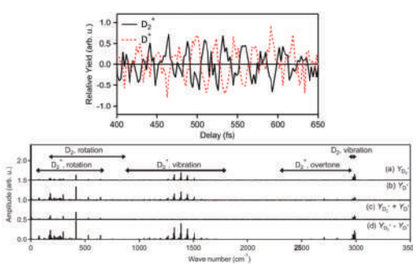

Figure 3. The results of SURF spectroscopic measurements of D^+ and D_2^+: The ion yields as a function of the delay time between the pump and probe pulse and the Fourier transform spectra [6].

EUV femtosecond laser processing

By focusing femtosecond laser pulses into a rare-gas medium, high-order harmonics of femtosecond laser pulses in the EUV wavelength region are generated. We focused the EUV laser pulses into a submicron spot on a surface of metal and semi-conductor materials using a focusing mirror with high surface accuracy and achieved submicron-size laser processing (Fig. 4) [9, 10].

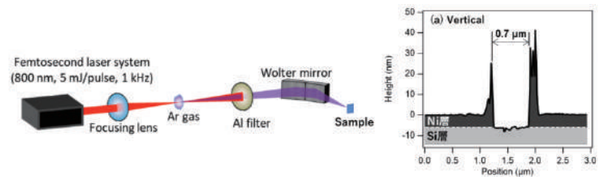

Figure 4. An experimental setup for extreme-ultraviolet femtosecond laser processing (left) and the recorded profile of the ablation spot on the Ni thin-layer-coated Si substrate (right) [10].

Development of theories to describe molecular dynamics in intense laser fields

By extending the first-principle quantal theory called multi-configuration time-dependent Hartree-Fock (MCTDHF) method, by which multi-electron dynamics in atoms and molecules interacting with an intense laser field can be calculated, we developed the extended MCTDHF method to calculate electro-protonic wave functions [10, 11]. When molecules are irradiated with an intense laser pulse, various dynamical processes such as electronic, vibrational, and rotational excitations and ionization proceed simultaneously. By the MCTDHF method, we have performed a numerical simulation of H_2^+ interacting with a 400-nm few-cycle intense laser pulse and showed that the electronic, vibrational, and rotational excitations are properly described [13].

Application of quantum computing to molecular science and development of a trapped-ion-type quantum computer

In recent years, there has been remarkable progress in the field of quantum computing both in hardware and in software and quantum algorithms. We have applied quantum computing to a variety of problems such as calculations of vibrational wave functions of CO_2 [14], Hückel molecular orbitals [15], and spin-spin interaction [16]. We have calculated the dynamics of an interacting spin chain using two currently available quantum computers, i.e., IBM's superconducting circuit type quantum computer and Quantinuum's trapped-ion type quantum computer, and revealed that both types of quantum computers can be used for simulations of model quantum systems with high fidelity (Fig. 5) [17].

We have also performed simulations of the time evolution of a three-level system interacting with an intense laser field (Fig. 6) [18] and of a spin-boson system representing electronic energy transfer in photosynthesis [19], demonstrating the importance of selecting appropriate algorithms and error mitigation methods.

On the other hand, in the recent development of trapped-ion type quantum computers, an approach using the continuous variables has attracted attention because it can facilitate highly efficient quantum error correction. Using the continuous variables, we have theoretically proposed a method to control entanglement with higher speed and accuracy than conventional methods and have undertaken an experimental demonstration [20].

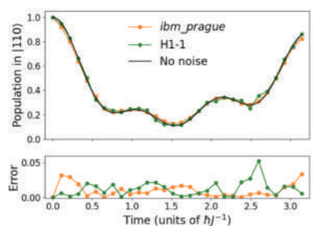

Figure 5. Upper panel: Time-dependent population in the qubit state $|110\rangle$ calculated using IBM's quantum computer *ibm_prague* (see ibm.com/quantum) and Quantinuum's quantum computer H1-1 (see quantinuum.com/). The solid black curve denotes the ideal noise-free result. Lower panel: The absolute value of the deviation of the population from the ideal noise-free case [17].

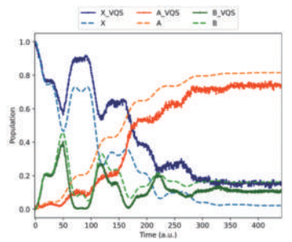

Figure 6. Results of the simulation of air-lasing using a NISQ device [18].

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