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Reaction dynamics simulation: From XFEL-induced Coulomb explosion to strand breaks in DNAs

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We have theoretically investigated the reaction dynamics of molecules induced by heat or by radiation fields. The methods used are the conventional classical reaction dynamics based on density functional theory (DFT) or density-functional based tight-binding (DFTB) approaches. We have also developed quantum wavepacket methods for excited state dynamics (e.g., nonadiabatic transitions) and electronic dynamics. In this talk, we present the results obtained by applying those methods to X-ray free-electron laser (XFEL) induced Coulomb explosions of molecules and strand breaks in DNAs.

XFEL-induced Coulomb explosion X-ray free-electron lasers (XFELs) [1] provide intense femtosecond x-ray pulses that promise direct observations of moving atoms during chemical reactions. Among various approaches proposed for probing atoms in motion is the 3D momentum correlation measurement for the atomic ions created via Coulomb explosion of molecules, called Coulomb Explosion Momentum Imaging (CEMI). XFEL pulses create extremely highly charged ions in the femtosecond timescale by multiple inner-shell ionization and thus are also expected to realize the femtosecond time-resolved CEMI for monitoring the dynamics of intermediate states. We developed a semiempirical DFTB approach to simulate the experimentally observed momentum correlation between fragment ions and kinetic energy distributions of ions [3]. We applied it to iodine-containing molecules such as 5-iodouracil. The present study demonstrates that the DFTB approach developed can be used for reproducing the experimental results and revealing the mechanisms of XFEL-induced Coulomb explosion [1], which enables the reconstruction of the positions of atoms in motion, referring to the momentum correlation measurement of the atomic ions created via XFEL-induced Coulomb explosion of complex molecules.

DNA strand break Damage to robust DNA polymers upon exposure to radiation occurs when single or double DNA strands break. Radiation damage in DNA is induced by direct and indirect effects of radiation. In the direct mechanism, the sugar-phosphate backbone is ionized upon exposure to high-energy radiation, which leads to strands breaks. The sources of indirect effects on strand breaks are heat and OH radicals etc. Recently, Mathur et al. investigated in detail the indirect effects on the strand breaks of plasmid DNAs [2]. They probed femtosecond laser induced damage to aqueous DNA that results from the strong-field interaction with water wherein heat and free radicals are generated in situ. These produce nicks in DNA under physiological conditions. The experimental results indicate that exposure to intense femtosecond pulses of 1350 and 2200 nm light induces single strand breaks (SSBs) and double strand breaks (DSBs) in DNAs. They proposed the scenario that DSBs are induced mostly by the action of two or more OH radicals, while the attack of an OH radical triggers only a SSB; use of OH scavengers establishes that the probability of a two-hit event reduces much faster than a one-hit event with increasing scavenger concentration. They also concluded that thermal effects induce SSBs but do not induce DSBs.

The detailed mechanisms of strand break however remain unclear at a molecular level. We have performed reaction dynamics simulations for short strand DNAs using the DFTB method. The roles of heat and OH radicals on SSB and DSB are examined to clarify the mechanisms of strand breaks.

[1] K. Nagaya et al, Faraday Discuss. **194**, 537 (2016).
[2] A. K. Dharmadhikari et al., Phys. Rev. Lett. **112**, 138105 (2014).