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Towards understanding dynamics of atoms and molecules in intense laser fields

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Abstract. Isolated atoms and molecules subjected to intense, long-wavelength laser fields undergo a number of processes, including ionization, fragmentation, and emission of high harmonics of the incident light. These processes can be used for a number of technological purposes (such as table-top coherent XUV and soft X-ray sources), and contain a rich body of information on the structure and dynamics of the parent molecules and their ions. I will describe the physical origin of these processes, and give some examples of their applications for strong-field spectroscopies. I will also briefly discuss some of the numerical techniques used for numerical simulations of electron dynamics in strong fields.

Keywords: Tunnelling ionization, High-harmonics generation, Femtosecond nuclear dynamics, Strong-field spectroscopies

PACS: 32.80.Rm, 32.80.Wr, 33.20.Xx

Intense ($\geq 10^{13}$ W cm $^{-2}$) infrared laser (IR) fields readily ionize neutral atoms and molecules in the gas phase, even though overcoming the ionization potential (IP) may require absorption of tens of photons within a few femtoseconds. The ponderomotive energy U_p ($U_p = E^2/2\omega^2$) of free electrons in such fields can reach hundreds of electron-volts.

Strong-field ionization in turn triggers a rich variety of processes, both electronic and nuclear. Most of the ionized electrons will depart directly to the detector, forming the above-threshold ionization (ATI) spectrum. Depending on the phase of the laser field at the time of electron's birth and laser polarization, fraction of the ionized electrons may return to the ion core. Elastic scattering from the ion core produces energetic photoelectrons up to $10 U_p$ in energy (high above-threshold ionization, HATI); inelastic collisions lead to ion-core excitation and secondary ionization events (non-sequential double ionization, NSDI); finally radiative recombination leads to emission of coherent radiation up to $3.2U_p$ in energy (high-harmonics generation, HHG).

Each of these processes gives access to structural and dynamical information for the target molecule. The ATI spectrum contains information on molecular electronic structure[1]. The HATI spectrum encodes the internuclear distances[2], similar to electron-diffraction images. By adding a perturbative XUV field at a controlled delay, the HATI spectrum can also be utilized to image the continuum electron wavepacket[3].

The recolliding electron wavepacket contains components from a few eV to potentially hundreds of eVs in energy, allowing a single-shot measurement of photoionization cross-section up to the HHG cutoff[4]. Under favourable conditions, valence electron correlation processes could also be resolved[5].

The ion formed by the strong-field ionization process will in general be in a coherent superposition of the electronic states, and will undergo femtosecond-time evolution. The signatures of this dynamics are imprinted in the amplitude, phase,[6] and ellipticity[7, 9] of the emitted HHG radiation, potentially with attosecond time and sub-Ångström spatial resolution. The dynamics can in turn be controlled by manipulating molecular alignment relative to the field polarization direction[8]. Generally, the molecular ion is also not at the equilibrium nuclear configuration, and will undergo femtosecond nuclear dynamics. Due to the coherent nature of the HHG emission, phase of the nuclear wavepacket is imprinted on the harmonics spectrum[10], allowing direct experimental imaging of nuclear wavepackets near the equilibrium geometry[11] and at conical intersections[12].

Theoretical modelling of strong-field processes is challenging due to the strongly non-perturbative nature of the interactions, variety of the possible physical processes, and presence of several important time scales (attoseconds to picoseconds). As the result, fully ab initio treatment is so far only possible for one- and some two-electron systems. For the promise of the new field of strong-field spectroscopy and control to be fully realized, new computational techniques are urgently needed. I will briefly discuss an approach we have been developing[13], which could allow rigorous treatment of molecules with tens of atoms in intense laser fields.

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