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Attosecond Electron Dynamics

Zhi-Heng Loh*
Division of Chemistry and Biological Chemistry, and Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

Abstract

Advances in strong-field laser physics and ultrafast optics allow the study of ultrafast dynamics on electronic time scales. Here, recent developments from the past year in the field of attosecond electron dynamics are highlighted.

High-intensity femtosecond laser pulses with electric field strengths comparable to those experienced by valence electrons in matter enable the light-field manipulation of electron dynamics on the attosecond time scale [1]. The semiclassical model by Corkum [2] provides the conceptual framework for the analysis of such experiments conducted in the strong-field regime. Within this model, the interaction of intense laser fields with matter is decomposed into three steps – 1) ionization, 2) acceleration of the ejected electron by the laser field, and 3) recollision of the electron with the ion core (Fig. 1). Each of these processes is temporally confined to within a fraction of a single laser-field cycle and is also phase-locked to the laser field. Strong-field laser physics therefore provides access to the study of electron dynamics that occur on attosecond time scales. Here, we highlight some of the recent experimental efforts from the past year that have enhanced our understanding of attosecond electron dynamics in atoms, molecules, and condensed matter.

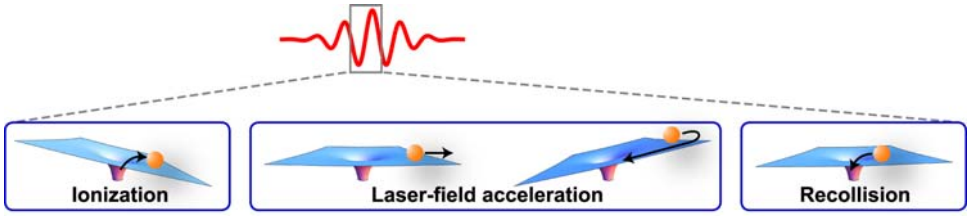

Fig. 1. Steering of attosecond valence electron dynamics synchronized on the sub-optical cycle time scale to an intense femtosecond laser field.

Tunnel ionization is the primary step in all strong-field phenomena. The time it takes for an electron to traverse the barrier is of fundamental interest, and is recently determined by the attosecond angular streaking method to be vanishingly small (<100 as) [3]. In a series of follow-up measurements employing the same experimental technique, the tunneling delay time for argon atoms is found to decrease as the laser intensity peak increases, i.e., as the Keldysh parameter decreases [4]. On the other hand, the tunneling delay time for helium atoms is found, to within experimental error, to be independent of the laser peak intensity. Along with the solution to the Schrödinger equation within the adiabatic approximation, the intensity dependence observed for argon highlights the importance of the Stark shift and the laser-induced dipole moment in determining the tunnel ionization rate. The smaller polarizability of helium makes the above effects negligible, thereby resulting in tunneling delay times that are independent of laser peak intensity.

* E-mail address: zhiheng@ntu.edu.sg

In addition to tunneling delay times, the moments at which electrons are born and at which they subsequently recombine with the ion to produce high-order harmonics have also been clocked [5]. This is accomplished by measuring the high-order harmonic generation (HHG) efficiency as a function of the phase offset between an orthogonally polarized weak $2\omega_L$ field and the intense ω_L fundamental field. The weak $2\omega_L$ field serves to steer the electron in the continuum away from the ion so as to suppress radiative recombination. At the same time, it breaks the mirror symmetry between adjacent half-cycles of the ω_L field, which results in the emission of even-order harmonics; tuning the phase offset allows the selection of electron trajectories with maximum contrast between adjacent odd- and even-order harmonics. Such an experiment therefore measures the time between ionization and recombination for the various harmonic orders. It is found that the ionization times measured for helium present a marked departure from those calculated based on the semiclassical model of HHG [2], even though the recombination times are in good agreement with theoretical prediction. The observed discrepancy can be explained in terms of the non-zero velocity of the electron as it emerges from the tunneling barrier. Beyond atomic systems, the ionization times of energetically similar orbitals in molecules can be almost indistinguishable. It is found that two-color HHG can successfully discriminate between the ionization times of the highest occupied molecular orbital (HOMO) and the HOMO–2 orbital of the CO_2 molecule, therefore demonstrating the remarkable sensitivity of this technique. These experiments permit the accurate determination of ionization and recombination times, which is essential to the study of attosecond electron dynamics in strong-field phenomena.

The strong-field ionization of multiple electronic states in a phase-coherent fashion yields electronic nonstationary states that drive electron motion [6]. Whereas the ionization of atoms [6] and small molecules (diatomics [7] and triatomics [8]) have previously been observed to yield multiple ion states as products, the participation of multiple electronic states in the strong-field ionization of complex molecules was established only recently [9]. In the case of large molecules, the dense manifold of ion states that can be accessed, along with their low molecular symmetry, in many cases potentially complicate measurements by soft X-ray absorption spectroscopy [6] and high-order harmonic interferometry [8], both of which have been successfully applied to observe phase-coherent ionization in atoms and small molecules. A new approach – channel-resolved above-threshold ionization (CRATI) – employs covariance mapping to correlate a particular photofragment mass with an above-threshold ionization (ATI) channel [9]. Since ATI electron spectra identify the ionization potentials of the electronic states, and that the dissociation of parent ions in electronically excited states yields photofragments, CRATI provides direct evidence for the involvement of multiple electronic states in strong-field ionization. The study employed butane and 1,3-butadiene, of which the former is found to exhibit a response that is dominated by the first electronically excited state of the ion. The experimental results are supported by theoretical calculations that solve the coupled-channel time-dependent Schrödinger equation.

Aside from investigating the ionization step, much new information can also be obtained from probing the recolliding electron wave packet. For sufficiently energetic wave packets, laser-induced electron diffraction (LIED) [10] is akin to conventional electron diffraction and can be employed to retrieve molecular structure. In ref. [11], mid-infrared laser pulses at wavelengths of 1.7, 2.0, and 2.3 μm are used to produce highly energetic electron wave packets via the λ^2–scaling of the ponderomotive energy. Rescattering energies exceeding 200 eV are accessed as a result. Analysis of the momentum distribution of the rescattered electrons by quantitative

rescattering theory [12] yields the rescattering differential cross-section. LIED yields the bond lengths of N_2 and O_2 upon strong-field ionization by the same mid-infrared laser pulses that subsequently steer recollision. Varying the laser wavelength allows different time delays between ionization and recollision to be accessed, thereby enabling the freeze-frame capture of ultrafast molecular dynamics. The range of wavelengths employed in ref. [11] corresponds to time delays of 4 – 6 fs. The experimental results reveal an elongation upon ionization of the N—N bond length to 1.14 Å (from 1.10 Å in neutral N_2), whereas the O—O distance is reduced from 1.21 Å in O_2 to 1.10 Å in O_2^+. These changes are in accord with molecular orbital theory – the removal of an electron from a bonding (anti-bonding) orbital in the case of N_2 (O_2) elongates (contracts) the bond. This study demonstrates the LIED can be used to retrieve molecular structure with sub-ångström spatial resolution and femtosecond time resolution.

The availability of quasi-monocycle laser pulses allows inelastic scattering of the recolliding electron wave packet – a process known as nonsequential double ionization (NSDI) – to be probed with unprecedented detail [13]. Employing sub-4-fs laser pulses in the near-infrared confines the primary ionization event to a single laser cycle, which in turn circumvents the multiple ionization-recollision cycles that potentially complicate data analysis. Reaction microscope measurements made with such short laser pulses reveal that the recolliding electron does not directly eject the secondary electron via impact ionization (this is the well-known (*e*,2*e*) mechanism [2]). Instead, the recolliding electron excites the ion core, which then undergoes subsequent ionization by the laser field. The highly differential nature of these measurements, which also include carrier-envelope phase (CEP) scans of the laser pulse, will allow them to serve as an important benchmark for theoretical models of electron correlation.

There is increasing interest in the study of strong-field phenomena in nanoscale metallic structures due to the strong plasmonic enhancement of the local field [14]. Until recently, however, the effect of the spatial confinement of the local field on electron emission remains unknown. In a new study, gold nanotips are illuminated with laser pulses that vary in wavelength from 0.8 – 8.0 μm [15]. Saturation of the maximum photoelectron kinetic energy is observed at long wavelengths and is attributed to the near-field decay of the local field enhancement. By introducing a spatial adiabaticity parameter, which is determined by the relative length scales of the near-field decay and the electron quiver motion, it is found that the quiver amplitude at longer wavelengths exceeds the near-field decay length. Under such circumstances, the electron no longer undergoes quiver motion, but instead escapes irreversibly from the local field. The reduced ponderomotive acceleration results in lower photoelectron kinetic energies. This study shows that both spatial and temporal effects must be considered to furnish a complete description of strong-field phenomena in nanoscale systems.

As evidenced by the studies highlighted here, progress in attosecond science is driven by the simultaneous development of novel experimental capabilities and theoretical models. The recent emergence of new light sources, such as attosecond kiloelectronvolt X-ray pulses [16] and sub-ångström-wavelength X-ray free-electron lasers [17], promises further insights into electronic phenomena that occur on attosecond time scales.

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