1-1-13. Attosecond Science

Overview

When reaching attosecond (as, 1 as = 10^{-18} seconds) time-resolution, an electron wavepacket motion can be probed in the time scale faster than molecular motion or chemical reaction dynamics. Attosecond science has two approaches. One method uses an attosecond laser pulse in the extreme ultra-violet or the soft X-ray regions, known as "high-harmonic generation". The other method is to use the electron re-collision process in which the "re-colliding electron pulse" is extracted from an atom or a molecule to be measured. Both approaches are employed to measure electron dynamics of molecules with attosecond time-resolution.

Current Status and Frontiers

A breakthrough from femtosecond to attosecond science had prevailed in the early 21st century. The basic model for attosecond science is the tunnel ionization and the re-collision model. When an intense, infrared laser pulse, referred to as a "fundamental" pulse", is focused into atoms or molecules, tunnel ionization occurs to eject an electron wavepacket in the ionization continuum. The electron pulse re-collides with the ion core, and thus it is referred to as a "re-colliding electron". The re-collision process leads to double ionization (reaction), elastic scattering and high-harmonic generation (or an attosecond laser pulse) processes.

1. The approach using high-harmonic generation

When the pulse duration of the fundamental pulse is longer, attosecond pulses can be produced at every half optical period. The adjacent attosecond bursts have opposite phases. This laser pulse is referred to as an "attosecond pulse train".

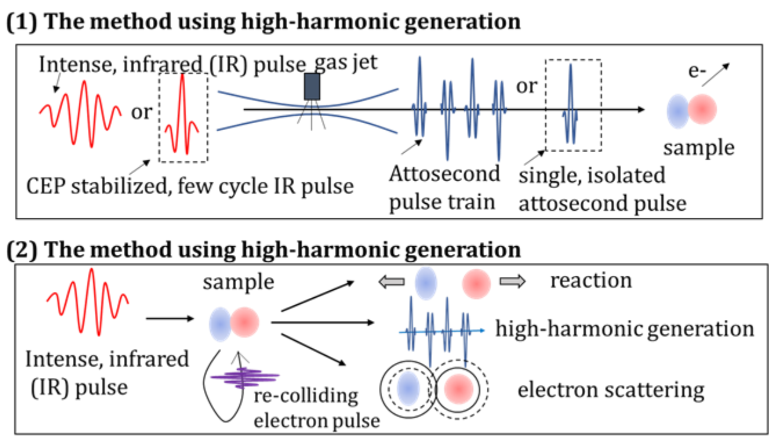

Figure 1. Two approaches for attosecond science.

The spectrum of the attosecond pulse train includes harmonics whose photon energies are odd multiplied by that of the fundamental field. When the pulse duration of the fundamental laser field is so short that only few-cycle is included (few-cycle laser pulse) and the carrier-envelope phase (CEP) is stabilized, a condition arises in which the electron re-collides with the ion only once. It leads to the generation of a single, isolated attosecond pulse.[1,2] By combining the attosecond pulse with another laser pulse, a time-resolved photoelectron spectroscopy and a transient absorption spectroscopy were performed to measure the dynamics of atoms and molecules, such as photoionization delay.[3]

2. Re-collision approach

By irradiating an intense laser pulse to a sample atom or molecule itself, the "re-colliding electron pulse" is extracted from the atom or the molecule itself. The re-colliding electron can probe electron and molecular dynamics with an attosecond resolution.[4] In case an elastic scattering process is used for probing, the method is refereed to as "laser induced electron diffraction" or "LIED". The structure and dynamics of the atom or the molecule can be also obtained from the spectrum of high-harmonics emitted from the atom or molecule. This method is referred to as "high-harmonic spectroscopy". Using the high-harmonic spectroscopy, chemical reaction dynamics has been studied[5] and information in the band gap structure of a solid state material is obtained.[6]

References

1. M. Hentschel, et al., *Nature*, **414**, 509–513 (2001).
2. A. Baltuška, et al., *Nature*, **421**, 611–615 (2003).
3. P. B. Corkum, F. Krausz, *Nature Phys.*, **3**, 381–387 (2007).
4. H. Niikura, et al., *Nature*, **417**, 917–922 (2002); *Nature*, **421**, 826–829 (2003).
5. H. J. Wörner, et al., *Nature*, **466**, 604–607 (2010); *Science*, **334**, 208–212 (2011).
6. G. Vampa, et al., *Nature*, **522**, 462–464 (2015); *Phys. Rev. Lett.*, **115**, 193603 (2015).

Future Perspectives and Directions

To fully understand chemical reaction dynamics, it is necessary to evaluate the changes in both molecular structure and electronic states or molecular orbitals. Especially, visualization of temporal changes in the molecular orbital during reaction dynamics is one of the dreams for chemists. Attosecond science provides the approaches using the ultra-short laser pulses and the re-colliding electron pulse. With these approaches, developing new theoretical proposals and experimental methodologies for the investigation of chemical reaction dynamics will be the next challenge.

(Author: Hiromichi Niikura)