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Nippon Telegraph and Telephone (NTT)
Tokyo University of Science

Inner-shell electron motion monitored by ultrafast strobe light source

～Characterizing inner-shell electron using isolated attosecond pulse～

Nippon Telegraph and Telephone (NTT) corporation (Head office, Chiyoda-ku, Tokyo, Japan; Hiroo Unoura, President and CEO) and Tokyo University of Science(Kagurazaka Campus, Shinjuku-ku, Tokyo, Japan; Akira Fujishima, University President)successfully observed inner-shell electron motion using isolated ultrashort attosecond (as: 10^{-18} second) pulses of light[1]. An inner-shell[2] electron has ultrashort decay time from a few attoseconds to several ten femtoseconds (fs: 10^{-15} second); therefore, the dipole[3] response could not be characterized directly. We were able to characterize it by combining an isolated attosecond pulse (IAP) and an analytical method called SPIDER (spectral phase interferometry for direct electric-field reconstruction). The IAP has ultrashort duration and exists in extreme ultraviolet (XUV) region (wavelength: approximately 3-30 nm). It behaves as an instantaneous strobe light and can directly access the inner shell. In addition, the SPIDER method can fully characterize the dipole response (the decay time, dipole phase, and periodicity of dipole oscillation). The research on the inner shell is substantially importance for studies in quantum optics, chemistry, and material science in the future.
This achievement is reported in Nature Communications (online journal), December 16, 2014.
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1. Background

Current global society strongly depends on the information technology via high-speed telecommunications with large data capacity. Telecommunication systems are supported by various optical technologies for switching, detection, and transmission. The technologies are closely related to electron motion in fundamental physics. However, since an electron has an ultrafast time response, its properties have been hard to characterize. For example, as a camera needs a high-speed shutter to take stop-motion snap shots, an instantaneous strobe light is necessary in order to observe an electron with ultrafast motion.
In an atom, electrons are classified into outer shell (valance-shell) and inner shell (core-shell) types. In common devices, the outer shell with a low energy band gap (a few electronvolts) is used. However, since the inner shell has a larger band gap (a few orders magnitude higher than the outer shell), the electron motion is much faster (Fig. 1). For instance, the decay time of an excited inner-shell electron is on a scale from a few attoseconds to several hundred femtoseconds (that of an outer-shell electron is on the scale of nanosecond). Therefore, the inner-shell has not been well investigated, because the decay motion of the excited electron is too fast to compare to the previous laser light sources.

2. Achievements

We successfully observed electron motion with the dipole response in the inner shell using the combination of the IAP and the SPIDER[4] method. Using double optical gating (DOG) [5] technique, we generated an IAP with 192-as duration -one of the shortest in the world? and this IAP can approach the time scale of the electron motion. In addition, the SPIDER method can fully characterize the dipole response (the decay time, dipole phase, and periodicity of dipole oscillation) (Fig. 2). The achievement for inner shell with ultrafast motion may pave the way for the development of new types of optical devices and novel chemical reactions.

Experiments

(1) In order to observe the dipole response, the IAP is focused onto a target medium. Here, we selected a neon atom as the medium for the first demonstration. The dipole response induced by the IAP generates electromagnetic radiation, which is a coherent photoemission. The radiation and transmitted IAP from the neon atom propagate to a photon spectrometer in the XUV region and the result of spectral interferogram is monitored (Fig. 3). The information about the dipole response can be extracted from the interferogram after the IAP phase is substructed.

(2) In order to generate the IAP, we used the DOG technique [H. Mashiko et al., Phys. Rev. Lett. 100, 103906 (2008)] (Fig. 4). This technique combines two powerful optical gating methods: polarization gating and two-color gating. It can be used to generate an IAP using a multicycle driving laser. Consequently, the IAP can be temporally characterized by the attosecond

streak method [J. Itatani et al., Phys. Rev. Lett. 88, 173903 (2002)], which allows us to measure the pulse duration, spectral distribution, and phase (Fig. 5).

(3) The temporally characterized IAP induces a dipole response with 2s [inner-shell: $2s^2p^6$] and 3p [unoccupied outer-shell $2s^2p^6(2S_{1/2})3p$] in the neon atom, which produces electromagnetic radiation. The IAP and the radiation construct a spectral interferogram that can be measured by an XUV spectrometer (Fig. 6). With the SPIDER method (Fig. 7), the dipole response (the decay time, dipole phase, and periodicity of dipole oscillation) can be determined (Fig. 1).

3. Technical Features

(1) Generation of IAP using DOG (Fig. 4) [NTT]

The attosecond pulse can be generated from a nonlinear medium (e.g., rare gas, molecules, or solid) using an intense driving laser. This generation process is called high harmonic generation[6]. Generally, the attosecond pulse is generated at every half optical cycle of the driving laser field. Thus, the high harmonic generation commonly produces an attosecond pulse train in the temporal domain. To isolate the attosecond pulse from the trains, we used the DOG technique, which combines two optical gating methods for the driving laser: polarization gating and two-color gating. The polarization gating produces a linearly polarized field at the center of the driving laser and elliptically polarized fields at leading and trailing edges of the driving laser. The attosecond pulse is generated only in the linearly polarized field of the driving laser due to the process of high harmonic generation. Thus, the attosecond pulse can be isolated from the trains. In addition, the two-color fields (400- and 800-nm wavelengths in this experiment) can relax the depletion of the ground state population of the target medium in the leading edge of the driving laser, which increases the flux of the IAP and allows us to use a multicycle driving laser.

(2) Temporal characterization of IAP using attosecond streak camera (Fig. 5) [NTT]

Since there is no nonlinear crystal optics in the soft x-ray XUV region, the IAP is difficult to characterize directly: it has to be converted to a photoelectron (ionized electron) once using a target medium in the measurement. In this experiment, we selected a helium atom and detected its photoelectron using a regular time-of-flight system. We first excited a helium atom with a near-infrared (NIR) pulse, which is collinearly propagated with the IAP. The momentum energy of its photoelectron is shifted by the electric field of the NIR pulse. By scanning a relative delay time between the IAP and NIR pulse, an attosecond streak trace can be observed. Since the wave packet of the photoelectron is a replica of the IAP, the trace contains information about the pulse duration, spectral distribution, and phase of the IAP. Here, the duration of the measured pulse is 192 as.

(3) Determination of dipole response with SPIDER (Fig. 7) [NTT and Tokyo University of science]

The SPIDER (spectral phase interferometry for direct electric-field reconstruction) can characterize the dipole response: the pulse shape, spectral distribution, and phase of the electromagnetic radiation. (I) The measured interferogram (Fig. 6) in spectral domain is converted to temporal structure with Fourier transformation. (II) With temporal filtering (Fourier filtering), the IAP component is removed and the electromagnetic radiation component is extracted. (III) With the Fourier transformation, we extract the spectral distribution and the relative phase between the IAP and the radiation. Note that the SPIDER can only derive a relative phase, not an absolute phase. Thus, we have to subtract the phase of the IAP from the relative phase, which is already determined by the attosecond streak mentioned in (2). (IV) With the final Fourier transformation, the result parameters provide the decay time (35 fs), dipole phase, and periodicity of the dipole oscillation (90 as).

4. Future Plans

We successfully demonstrated the dipole response with the inner-shell electron using the combination of the IAP and the SPIDER method. The inner shell has both high energy and an ultrafast response time. To extend the application, we aim to observe the dipole response in solid-state dynamics. The present study may pave the way for the development of a new type of optical device with the inner shell. Furthermore, the inner shell will enable us to study novel chemical reactions, since the it is closer to the parent nucleus than the outer shell. In order to increase the temporal resolution, we will attempt to generate an IAP with less than 24-as duration (one atomic unit of time), which can be applied to characterize a deeper inner shell with shorter decay time.

Publication information

H. Mashiko, T. Yamaguchi, K. Oguri, A. Suda and H. Gotoh
“Characterizing inner-shell with spectral phase interferometry for direct electric-field reconstruction”
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Glossary

1. Attosecond pulse
   Attosecond corresponds to 1×10^{-18} second. The attosecond pulse is an optical light source with ultrashort pulse duration, which behaves as an instantaneous strobe light. Generally, the pulse exists in the XUV (3~30 nm) region.

2. Inner-shell
   An atom is constructed of a parent nucleus and electrons. Electrons are trapped by the nucleus by potential energy. An electron in the inner shell is located closer to the nucleus than the outer-shell. Thus, the potential energy of the inner shell is higher than that of the outer-shell.

3. Dipole

   The dipole is defined as the product of the magnitude of electric charges and the distance separating the charges. When an electron is excited by an input laser pulse, the electron and the ion with opposite polarities create the dipole. Then, the dipole oscillation induces charge oscillation and generates the electromagnetic radiation. Here, we studied the dipole response with the inner shell of a neon atom (a transition from 2s to 3p states).

4. SPIDER

   The SPIDER is analytical method of pulse characterization. With Fourier transformation, the temporal structure (the decay time), temporal phase, and periodicity of pulse oscillation can be extracted from the spectral interferogram (beat signal) constructing with two frequency components.

5. DOG

   The DOG technique combines two optical gating methods for the driving laser: polarization gating and two-color gating. Experimentally, the DOG can be constructed with three optics only (two quartz plates and a BBO crystal). Currently, the shortest attosecond pulse generated by the DOG technique has 67-as duration [K. Zhao et al., Opt. Lett. 37, 3891 (2012)].

6. High harmonic generation

   High harmonic generation is a highly nonlinear process. High harmonics are generated from a target medium using an intense driving laser. The principle can be explained well by the three-step model [P. B. Corkum Phys. Rev. Lett. 71, 1994 (1993)]: (i) An electron is initially trapped by the potential curve in the parent nucleus. Since the intense driving laser bends the potential curve, the electron is tunnel-ionized from the nucleus. (ii) The ionized electron is accelerated by the electric field of the driving laser. (iii) The propagation direction of electron is reversed due to the electric field of the driving laser after a half optical cycle. With recombination of the electron to the nucleus, the attosecond pulse can be generated.

Attachment·Reference

- Fig. 1: Inner-shell electron
- Fig. 2: Characterization of dipole response
- Fig. 3: Detection of dipole response
- Fig. 4: Generation of isolated attosecond pulse
- Fig. 5: Attosecond streak camera
- Fig. 6: Measurement of interferogram (Ne)
- Fig. 7: SPIDER

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