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位相制御FELを用いたアト秒スケールの光イオン化遅延時間計測

[1] 東北大学，[2] 理科科学研究所，[3] 東京大，[4] Moscow State Uni,
[5] Uni Freiburg，[6] TU Wien，[7] ELI-ALPS，[8] European XFEL,
[9] Uni Trieste，[10] Elettra，[11] ENEA，[12] Uni Nova Gorica，[13] Swinburne Uni
○You Daehyun[1,2]，Iablonskyi Denys[1]，上田潔[1,2]，石川顕一[3]，Oyunbileg Tugs[3]，織茂悠貴[3]，佐藤健[3]，Gryzlova Elena V[4]，Staroselskaya Ekaterina I[4]，Grum-Grzhimailo Alexei N[4]，Sansone Giuseppe[5]，Kumar Pravinm M[5]，Carpeggiani Paolo A[6]，Füle Miklós[7]，Csizmadia Tamás[7]，Meyer Michael[8]，Mazza Tommaso[8]，D'Elia Alessandro[9]，Callegari Carlo[10]，Di Fraia Michele[10]，Plekan Oksana[10]，Giannessi Luca[10,11]，Allaria Enrico M[10]，De Ninno Giovanni[10,12]，Trovò Mauro[10]，Badano Laura[10]，Diviacco Bruno[10]，Gauthier David[10]，Mirian Najmeh S[10]，Penco Giuseppe M[10]，Rebernik Primoz R[10]，Spampinati Simone[10]，Spezzani Carlo[10]，Gaio Giulio[10]，Prince Kevin C[10,13]

Attosecond delays in photoionization studied with seeded FEL

○Daehyun You[1,2], Denys Iablonskyi[1], Kiyoshi Ueda[1,2], Kenichi L Ishikawa[3], Tugs Oyunbileg[3], Yuki Orimo[3], Takeshi Sato[3], Elena V Gryzlova[4], Ekaterina I Staroselskaya[4], Alexei N Grum-Grzhimailo[4], Giuseppe Sansone[5], Pravinm M Kumar[5], Paolo A Carpeggiani[6], Miklós Füle[7], Tamás Csizmadia[7], Michael Meyer[8], Tommaso Mazza[8], Alessandro D'Elia[9], Carlo Callegari[10], Michele Di Fraia[10], Oksana Plekan[10], Luca Giannessi[10,11], Enrico M Allaria[10], Giovanni De Ninno[10,12], Mauro Trovò[10], Laura Badano[10], Bruno Diviacco[10], David Gauthier[10], Najmeh S Mirian[10], Giuseppe M Penco[10], Primoz R Rebernik[10], Simone Spampinati[10], Carlo Spezzani[10], Giulio Gaio[10], Kevin C Prince[10,13]
[1] Institute of Multidisciplinary Research for Advanced Materials, Tohoku University, Japan
[2] RIKEN SPring-8 Center, RIKEN, Japan
[3] School of Engineering, the University of Tokyo, Japan
[4] Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Russia
[5] Institute of Physics, Albert-Ludwigs-Universität Freiburg, Germany
[6] Technische Universität Wien, Austria
[7] ELI-ALPS, Hungary
[8] European XFEL, Germany
[9] Department of Physics, University of Trieste, Italy
[10] Elettra-Sincrotrone Trieste, Italy
[11] ENEA Centro Ricerche Frascati, Italy
[12] Laboratory of Quantum Optics, University of Nova Gorica, Slovenia
[13] Department of Chemistry and Biotechnology, Swinburne University of Technology, Australia

【Abstract】 When an electron is ejected from an atom after absorption of a photon, there is an extremely short delay between the photon absorption and the electron emission. This interval, called the Eisenbud-Wigner-Smith (EWS) delay, is on the order of a few attoseconds (10^{-18} seconds). Currently, there are two primary methods to measure the EWS delay. Both methods are based on optical laser pulses including infrared (IR) pulses as part of the technique. The IR field, however, perturbs the Coulomb field in atomic systems. Here we present a new method

to measure the EWS delay, using coherent-controlled free-electron laser (FEL) pulses; no IR pulse is required. Therefore, no corrections regarding the measurement of the EWS delay are necessary.

【序】The age of attosecond physics was ushered in by the invention of methods for probing samples on a time scale less than femtoseconds [1], and currently, many such ultrafast phenomena are being investigated. Photoemission is one such process, which has attracted much interest, particularly regarding the time delay between the absorption of a photon and the emission of a photoelectron, which is called the Eisenbud-Wigner-Smith (EWS) time delay [2]. There are currently two main methods of measuring the EWS delay: attosecond streaking [3] and RABBITT [4]. Both methods are based on optical laser pulses including infrared (IR) pulses as part of the technique. The IR field, however, perturbs the Coulomb field in the atomic system; thus, it is necessary to apply corrections during analysis and maintain a sufficiently low IR intensity.

Here we present a new method to measure the EWS delay. In this method, we use short wavelength XUV light, consisting of phase-locked first (ω) and second harmonic (2ω) pulses. This method eliminates the perturbation of the IR field present in the other approaches. Furthermore, being interferometric, our technique does not require extremely short, few-cycle pulses, but it depends instead on extremely accurate phase control (few attoseconds). Such fine control is available from the Italian free-electron laser, FERMI [5]. We report the measurement of EWS delay difference between electrons emitted by single-photon and two-photon processes of Ne, as a demonstration of the new method.

【実験方法】We carried out the measurements at LDM beam-line, FERMI. The photon energies were set to 14.3 eV, 15.9 eV, and 19.1 eV for ω, which is below the Ne ionization threshold (21.6 eV), while the second harmonic 2ω is above the threshold. The dichromatic light beam with fixed relative phase crosses the atomic jet with He and Ne mixture. A velocity map imaging spectrometer measures angular distribution of ejected electrons.

【結果・考察】Our target process is the photoionization of Ne by 1 or 2 photons:

single-photon process: $\mathrm{Ne} + \mathrm{photon}(2\omega) \rightarrow \mathrm{Ne}^{+}(2p^{-1}) + e(s\text{- and } d\text{-waves})$,

two-photon process: $\mathrm{Ne} + 2\mathrm{photon}(\omega) \rightarrow \mathrm{Ne}^{+}(2p^{-1}) + e(p\text{- and } f\text{-waves})$.

Because these four partial waves of an electron emitted by the two different pathways interfere with each other, the electron angular distribution is correlated to the unique phase shifts of each partial wave. The phase shifts can be extracted, by scanning the optical phase difference between ω and 2ω. On the other hand, the EWS delay $\tau(E)$ is defined as the derivative of phase η with respect to the photoelectron kinetic energy E: $\hbar\, \partial\eta/\partial E$ [2], where η is the phase shift of each partial wave. Hence, we have measured the phase shift differences at various photon energies and took their derivative to estimate the EWS delay. We find the EWS delay differences $\tau_{\mathrm{two}}-\tau_{\mathrm{single}}$ of −50±15 attoseconds at 14—16 eV and −7±5 attoseconds at 16—19 eV, which are in good agreement with theory.

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