Polarization Control of Isolated Attosecond Pulses

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Abstract: By adjusting the ellipticity of counter-rotating polarized few-cycle pulses for non-collinear high order harmonic generation, we obtain full control of polarization states of isolated attosecond pulses for the first time.

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Ultrafast Extreme ultraviolet (EUV) beams carrying spin angular momentum are currently thoroughly studied for their applications for investigating the structural, electronic, and the magnetic properties of materials, discriminating between enantiomers as well as working out how chiral molecules interact. However, such light sources are still limited to only a few free-electron laser facilities [1]. In SACLA, a linearly polarized X-ray FEL beam is going through a phase retarder and then converted into circular polarization. In FERMI and LCLS, researchers have demonstrated the generation of X-ray and EUV pulses with polarization control using special undulator, containing strong magnets which precisely control the wiggling motion of the electrons, therefore, the polarization of the produced radiation. Although powerfully, the pulse duration of FEL is typically tens of femtoseconds. Furthermore, the large scale facilities and high costs result in a limited number of beamlines.

High harmonic generation driven by femtosecond lasers makes it possible to capture the fastest dynamics in molecules and materials. However, to date the shortest subfemtosecond (attosecond, 10^{-18} s) pulses have been produced with only the linear polarization, with limits the range of physics that can be explored. In 2016, we have experimentally demonstrated the generation of isolated circularly polarized attosecond pulses by non-collinear high harmonic generation (HHG) with two few-cycle circularly polarized counter-rotating pulses (~3.6 fs, 1.4 optical cycles, generated with the MPContinua method [2]). This produces a pair of HHG supercontinua beams, one with left-circular and one with right-circular polarization, and spanning photon energies from 25 to 40 eV with a transform limit of 190 *as* [3].

In this work, we further experimentally demonstrated full polarization control of isolated attosecond pulses ϵ_{atto}, by adjusting the ellipticity of two counter-rotating driving pulses, ϵ_{fund}. Importantly, the polarization state of the attosecond pulses were fully analyzed with an EUV polarimeter, composed of two rotatable sets of triple-reflection polarizers, which unambiguously determines the ellipticity and helicity of attosecond pulses. Fig.1 shows the polarization states, Stokes parameters (S_1/S_0, S_2/S_0, S_3/S_0), of isolated attosecond pulses, which have been produced.

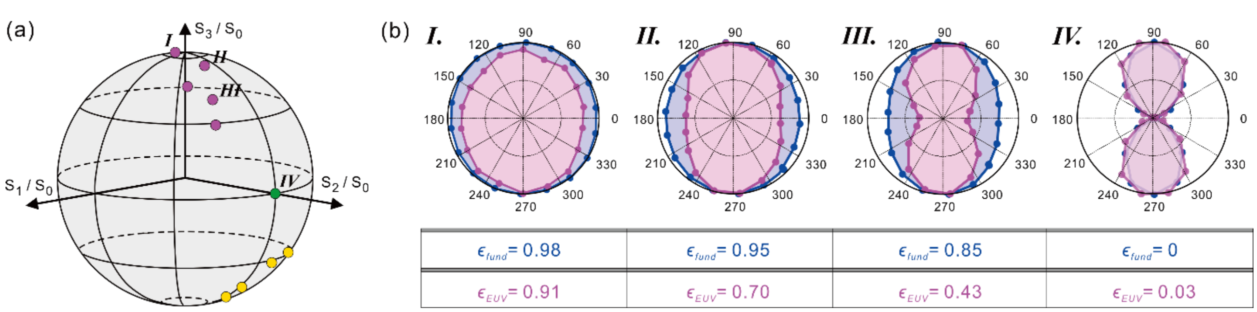

Fig 1: (a) The polarization states, Stokes parameters (S_1/S_0, S_2/S_0, S_3/S_0), of isolated attosecond pulses have been generated, characterized, and marked on the Poincaré surface (purple and yellow dots represent positive and negative helicity, respectively), while one HHG pulse driven by linearly polarized fundamental was also measured (green dot and inset IV). (b) Blue (and purple) lines representing fundamental (and attosecond) pulse intensity varied with the axis of a polarization analyzer, together with one table giving ellipticity relations between them.

Experimentally, to control the ellipticity of attosecond pulses, two elliptically polarized few-cycle fundamental beams with the same ellipticity ϵ_{fund} but opposite helicity are prepared and focused into a gas jet non-collinearly (see Fig. 2). These two incident electric fields can be written as

$$E_{fund.R} = [\epsilon_{fund} \cdot e^{i(+k\cdot x\cdot sin\theta+k\cdot z\cdot cos\theta)} \cdot \hat{x} + e^{i(+k\cdot x\cdot sin\theta+k\cdot z\cdot cos\theta+\pi/2)} \cdot \hat{y}] \cdot e^{-2.77\cdot(t/\tau)^2-i\omega t}$$
$$E_{fund.L} = [\epsilon_{fund} \cdot e^{i(-k\cdot x\cdot sin\theta+k\cdot z\cdot cos\theta)} \cdot \hat{x} + e^{i(-k\cdot x\cdot sin\theta+k\cdot z\cdot cos\theta-\pi/2)} \cdot \hat{y}] \cdot e^{-2.77\cdot(t/\tau)^2-i\omega t},$$

where $E_{fund.R}$ and $E_{fund.L}$ represent right-handed and left-handed elliptically polarized light, ϵ_{fund} is input ellipticity of two fundamentals, θ is half crossing angle between two input beams, x and z are transverse and longitudinal distance away from the center of the focal plane, ω is the angular frequency of the driving laser and τ is the full width at half-maximum (FWHM) pulse duration. At the focal plane, z=0, the electric field E_{focus} becomes

$$E_{focus}(x) = E_{fund.R} + E_{fund.L} = [2\cdot \epsilon_{fund} \cdot \cos(k \cdot x \cdot sin\theta) \cdot \hat{x} - 2\sin(k \cdot x \cdot sin\theta) \cdot \hat{y}] \cdot e^{-2.77\cdot(t/\tau)^2-i\omega t}.$$

Two fundamentals interferingly form local E-field vectors, rotating crossing the transverse direction $\hat{x}$ with a period of $2\pi/(ksin\theta)$. For one specific position *x*, local E-field oscillates linearly, which is excellent for high order harmonic generation. Each location with linear polarization acts as a high-order harmonics local emitter and superposes elliptically polarized EUV beam in the far field. As a result, the ellipticity ϵ_{atto} of attosecond pulses can be well controlled by fundamental ellipticity ϵ_{fund} as presented in Fig. 1. Moreover, this non-collinear scheme also support generation of isolated attosecond pulse, when driven by few-cycle pulses. This has been theoretically and experimentally demonstrated [2,4].

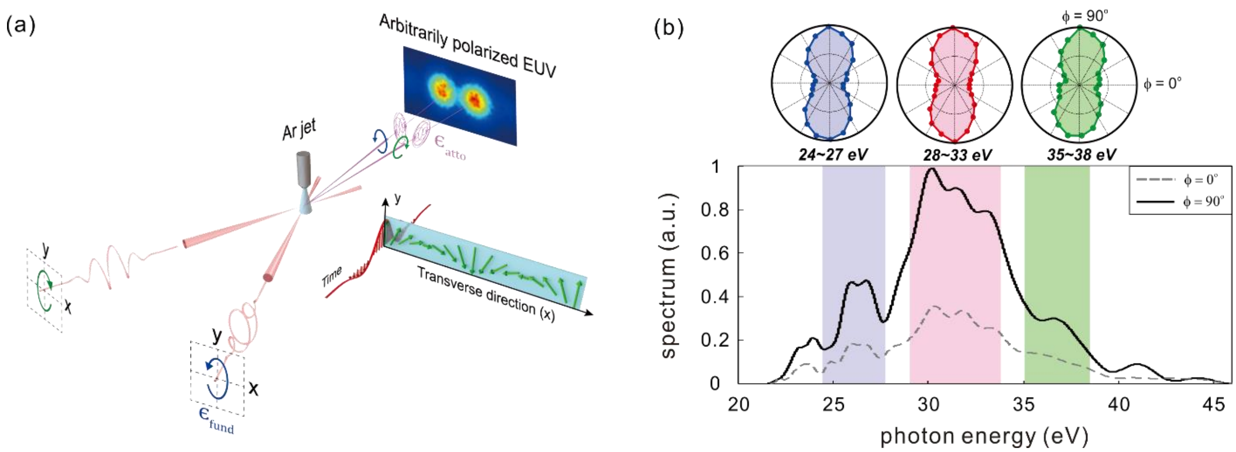

Fig 2: (a) Schematic of the experimental setup. Two elliptically polarized fundamental beams with opposite helicity are non-collinearly focused into a gas jet to generate arbitrary polarized isolated attosecond pulse. The inset shows the electric field distribution on the focal plane – local E-field vectors rotating crossing the transverse direction $\hat{x}$. In addition, temporally the driving pulses are as short as one-cycle, generating isolated attosecond pulses. (b) Continuous spectra taken when an analyzer selects its polarization along the semi-major (black line) and the semi-minor (gray dashed line) axis of elliptical attosecond pulses, while the average ϵ_{atto} = 0.43. Ellipticity in three different photon energy ranges are analyzed separately. They are almost identical and shown on top.

In summary, by adjusting the ellipticity ϵ_{fund} of two counter-rotating few-cycle incident pulses, we have experimentally demonstrated the full control of polarization states of isolated attosecond pulses for the first time. The generation of isolated, arbitrarily polarized attosecond EUV pulses makes major breakthroughs for the next attosecond frontiers, resolving the basic process associated to energy and angular momentum transfer between the electron/spin system, and symmetry-dependent characteristics of chiral biomolecules, dynamics of structural, electronic, and magnetic properties of materials with unprecedented temporal resolution.

Reference

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