Isolated, Circularly Polarized, Attosecond Pulse Generation

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Abstract: We experimentally demonstrate angularly and temporally isolated counter-rotating circularly polarized attosecond pulses which span from 30 to 45 eV.
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Circularly polarized extreme ultraviolet (EUV) and X-ray radiation has proven itself to be extremely useful for analyzing the structural, electronic and magnetic properties of materials. Using the extreme nonlinear-optical process of high harmonic generation (HHG), light from an ultrafast laser can be coherently upshifted to generate fully coherent beams in the EUV, with attosecond (*as*) pulse durations. However, until recently, it was not possible to directly generate or measure bright circularly polarized HHG pulses [1,2]. This is because for a linearly polarized driving field, the electron accelerates on a linear trajectory, and therefore easily re-collides with the parent ion. In contrast, for circularly-polarized driving lasers (or elliptically-polarized lasers with large ellipticity), the probability of recollision and the emission of high harmonics is much suppressed. Although many proposals have suggested indirect synthesis of circularly polarized *as* pulse trains, which are not well suited for the measurement of time dynamics, scaling to isolated circularly polarized *as* pulses has in all cases remained extremely difficult.

Recently, using collinear and non-collinear counter-rotating circularly polarized driving lasers, circularly polarized trains of *as* pulses were experimentally generated [1-3]. In the case of a non-collinear HHG geometry (NCP-HHG), numerical simulations suggest that isolated *as* bursts of circularly polarized light can be generated simply by reducing the driving laser pulse duration to the few-cycle regime [4]. Here we experimentally confirm this prediction by demonstrating a circularly polarized EUV HHG supercontinuum for the first time, spanning photon energies from 30-45 eV, and supporting 220 *as* transform-limited isolated pulses. Key to this advance was the use of few-cycle pulses synthesized from compressed multiple thin plate supercontinua (MPContinua) [5]. The unique scalability of the MPContinuum technique enables the realization of almost identical near-single-cycle pulses without using split mirrors or a second compressor setup.

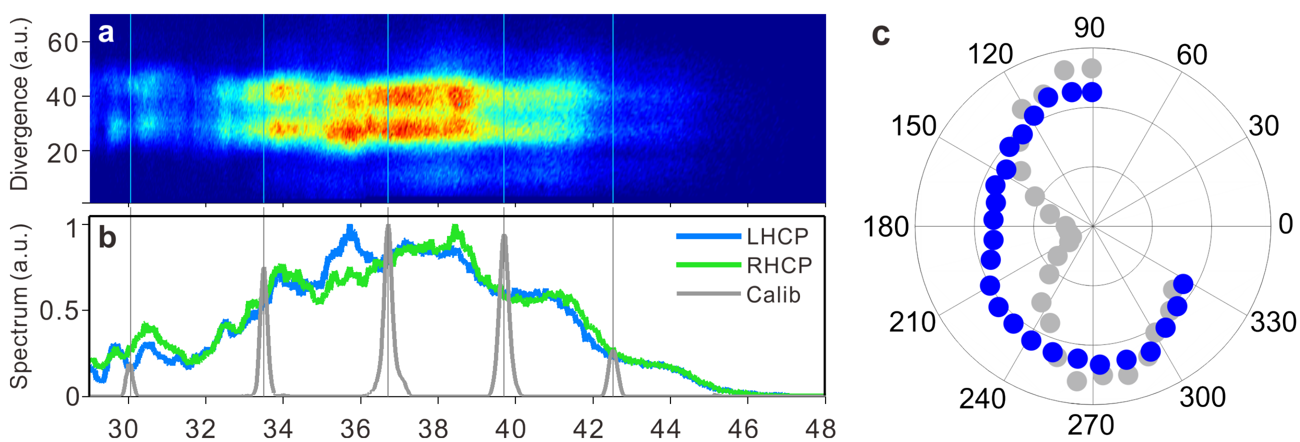

Fig. 1: (a) First experimental demonstration of an EUV circularly-polarized HHG supercontinuum. The upper and lower spectrum are purely right- or left- circularly polarized respectively. (b) Normalized integrated spectra of each circularly polarized supercontinuum. The black line shows discrete HHG driven by a 25 fs (10 cycles) pulse, as a calibration. (c) The intensity measurement as a function of EUV polarizer angle ranging from 90° to 330° for the HHG generated from crossing two counter-rotating circular polarized fundamentals (blue), together with the HHG driven by a linear fundamental (grey).

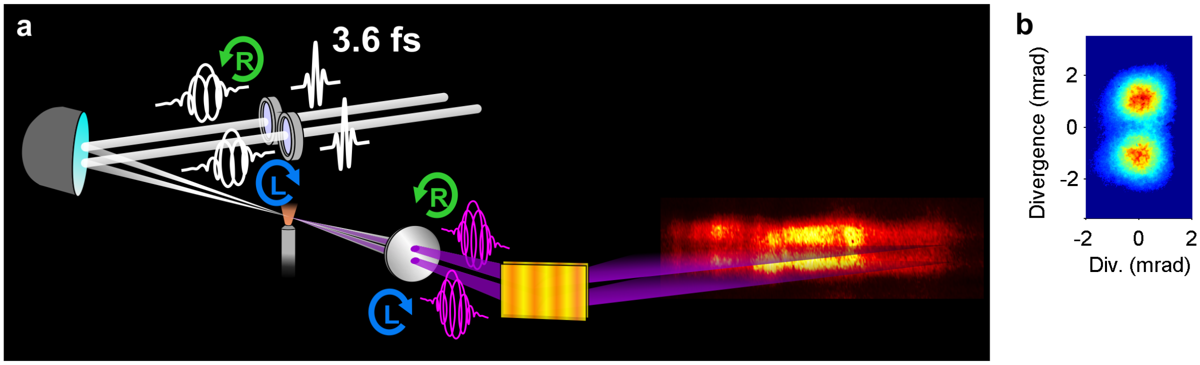

Fig. 2: (a) Isolated circularly polarized HHG scheme. Two counter-rotating circularly polarized 3.6 fs (1.4 optical cycles) pulses are focused into a gas target to produce a pair of left- and right- circularly polarized isolated *as* pulses via HHG. (b) Counter-rotating circularly polarized EUV beam profile.

The output of a commercial chirped pulse amplifier (25 fs, 1 kHz, 480 µJ) is split into two beams using a slightly misaligned Mach-Zender interferometer. Both linearly polarized beams are focused into an array of individual thin fused silica plates of 50 µm thickness [5]. The resulting MPContinua are simultaneously compressed using the same chirped mirrors to ~3.6 fs (1.4 optical cycles), which is confirmed by polarization gating cross-correlation frequency resolved optical gating. NCP-HHG is implemented by causing the two beams to have counter rotating polarizations using quarter and half wave plates and then focusing both pulses with a 20 cm focal length parabolic mirror in a crossed geometry into a vacuum chamber with a continuous Ar gas jet. The beam size is 3 mm and the full crossing angle is ~60 mrad. Both driving beams have a pulse energy of 60 µJ with 3 mm beam diameter just before the HHG chamber. After the Ar jet are two 200 nm Al filters followed by an EUV spectrometer which is calibrated to better than 0.3 eV using HHG from a 25 fs pulse.

When both driving pulses overlap in time and space, their polarizations are of opposite circularly polarization, and they are near single cycle (see Fig. 2); the continuous HH spectrum (integrated for 200 shots) shown in Fig. 1 is observed. A relative time delay between the two driving pulses provides narrowing of the polarization gating window which suppresses temporal small side lobes and results in a more continuous spectrum, in excellent agreement with our theoretical simulations (Fig. 3). The generated photon flux is estimated to be ~ $5{\times}10^6$ counts per shot over the whole EUV spectrum. Conservation of energy and spin angular moment ensures that this radiation is circularly polarized, which is further experimentally proven by a EUV polarization measurement, showing an almost equal HHG intensity distribution as a function of EUV polarizer angle (Fig. 1(c)). Moreover, the broadband supercontinuum nature of the right and left circularly polarized EUV spectrum is strong evidence that isolated *as* pulses are being generated.

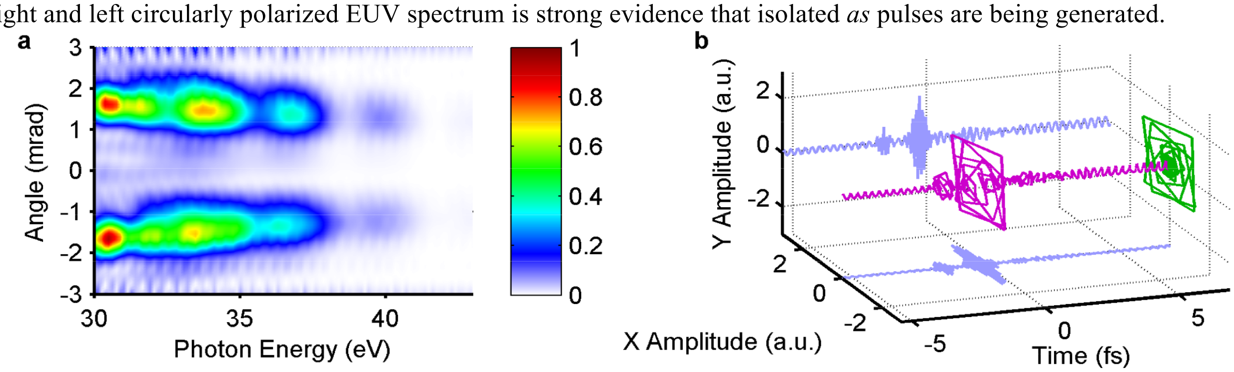

Fig. 3: Simulated circularly polarized spectrum (a) and *as* pulse (b) obtained through NCP-HHG in Ar driven by the counter rotating circularly polarized NIR pulses retrieved from the experiment, with a delay of 2.6 fs between them.

In summary, we demonstrate the first circularly-polarized isolated *as* pulses. In addition, the source is table-top based and produces two counter-rotating angularly isolated circularly polarized pulses. This work represents an advance towards capturing ultrafast dynamics of angular momentum transfer in atoms, chiral molecules, and magnetic materials with unprecedented temporal resolution.

References

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