Generation and Characterization of Isolated, Circularly Polarized, Attosecond Pulses

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Abstract: We experimentally demonstrate angularly and temporally isolated counter-rotating circularly polarized attosecond pulses at 33 eV. The pulse contrast and circular polarization of the beams are fully characterized by a field autocorrelator and an EUV polarimeter.

Circularly polarized extreme ultraviolet (EUV) and X-ray radiation is a useful tool to analyze the structural, electronic and magnetic properties of matter. Recently, circularly polarized trains of attosecond (*as*) pulses were experimentally generated through high harmonic generation (HHG) using collinear and non-collinear counter-rotating circularly polarized driving lasers [1]. 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 by reducing the driving laser pulse duration to the few-cycle regime [2]. We demonstrate isolated, circularly polarized *as* pulses experimentally by combining two simultaneously compressed pulses (~3.6 fs, 1.4 optical cycles, generated with the MPContinua method [3]) with counter rotating polarizations in a crossed geometry (Fig. 1a). 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 [4].

To characterize these isolated *as* pulses we implement a Fourier transform field autocorrelation because it is a straightforward way to distinguish an isolated *as* EUV pulse from a train of pulses [5]. One of the circularly polarized EUV beams is sent to a split mirror, which separates the beam into two halves. The relative time delay is controlled by a motorized translation stage with sub-nm resolution. The resulting interference pattern is measured at different delays between with an x-ray CCD, which provides the autocorrelation of the HHG beam. The HHG autocorrelation data (Fig. 1b) shows a single isolated pulse with a width of 380 *as*, and a spectrum that supports a transform-limited 190-*as* pulse. The main EUV pulse has a high amplitude contrast ($\approx 16x$) compared to adjacent pulses, demonstrating that it is an isolated pulse.

Most importantly, the polarization state of the *as* pulses is fully analyzed with an EUV polarimeter composed of two rotatable sets of triple-reflection polarizers [6]. The Stokes parameters (Fig. 1c; S_1/S_0 , S_2/S_0 , S_3/S_0) of the left- and right-circularly polarized beams are measured to be (-0.13, 0.19, 0.97) and (0.26, 0.01, -0.96) respectively, which unambiguously shows that the two *as* pulses are circularly polarized with opposite helicity.

In summary, we have generated and characterized circularly polarized isolated *as* pulses for the first time. This work paves the way towards *as* metrology of circular dichroism, e.g., capturing ultrafast dynamics of angular momentum transfer in atoms, chiral molecules, and 2D and magnetic materials with unprecedented temporal resolution.



Fig. 1: (a) Schematic of isolated circularly polarized HHG, together with the beam profiles. Two counter-rotating circularly polarized short fundamentals are focused into a Ar gas jet to produce a pair of left- and right- circularly polarized isolated *as* pulses. (b) Experimental field autocorrelation trace of circularly-polarized HHG. The inset shows excellent agreement between Fourier-transform spectrum (blue) and grating-dispersed spectrum (red). Note that the bandwidth-limited pulse duration is half of this coherence time (c) The polarization state, Stokes parameters (S_1/S_0 , S_2/S_0 , S_3/S_0), of this *as* pulse pair was measured by an EUV polarimeter separately and marked on the Poincaré surface (blue and green dots), while one HHG pulse driven by linearly polarized fundamental was also measured (grey dot).

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