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A table-top ultrashort light source in the extreme ultraviolet for circular dichroism experiments

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Circular dichroism in the extreme ultraviolet range is broadly used as a sensitive structural probe of matter, from the molecular photoionization of chiral species[1–3] to the magnetic properties of solids[4]. Extending such techniques to the dynamical regime has been a long-standing quest of solid-state physics and physical chemistry, and was only achieved very recently[5] thanks to the development of femtosecond circular extreme ultraviolet sources. Only a few large facilities, such as femtosliced synchrotrons[6,7] or free-electron lasers[8], are currently able to produce such pulses. Here, we propose a new compact and accessible alternative solution: resonant high-order harmonic generation of an elliptical laser pulse. We show that this process, based on a simple optical set-up, delivers bright, coherent, ultrashort, quasi-circular pulses in the extreme ultraviolet. We use this source to measure photoelectron circular dichroism on chiral molecules, opening the route to table-top time-resolved femtosecond and attosecond chiroptical experiments.

High-order harmonic generation (HHG) is an extremely nonlinear optical process that occurs when an intense femtosecond light pulse interacts with a gaseous target. Many characteristics of the high-harmonic radiation make it a unique and ideal source for time-resolved studies, including small size, low cost, good spatial coherence[9], brightness[10], tunability[11] and ultrashort pulse duration (femtosecond[12] to attosecond[13]). HHG is thus being increasingly used as a light source for atomic[14], molecular[15,16], surface and solid-state[17] time-resolved spectroscopy.

HHG is generally achieved in rare gases using linearly polarized laser fields, yielding linearly polarized extreme ultraviolet (XUV) radiation, parallel to the fundamental polarization. When interacting with matter, such a radiation simply defines an axis. By contrast, circular polarized light (CPL) defines an orientation of space and is thus a unique probe of chiral matter. As such, its production is becoming a requested figure of merit of short-wavelength sources, especially in the 10–30 eV range, close to the ionization thresholds of most molecular systems.

Converting a linearly polarized harmonic beam to circular polarization can be achieved using multiple reflections on surfaces, which have a different complex reflectivity of S and P polarization and induce a phase shift between these two components[18]. Such a set-up reduces the photon flux by two orders of magnitude. The direct generation of circular harmonics with high efficiency is thus preferable for practical applications. Although the most natural way to generate CPL from high harmonics would seem at first sight to be to use a circularly polarized fundamental laser pulse, the harmonic emission decreases exponentially with laser ellipticity[19], which imposes severe restrictions on the range of usable ellipticities. The ellipticity of the harmonic radiation in rare gases being generally lower than that of the driving laser, an efficient generation of XUV pulses with ellipticities above 20% is precluded[20]. The only reported exception to this rule is harmonic 17 from neon[20], which is characterized by higher ellipticity but a very low generation efficiency.

Using additional electric or magnetic fields can counteract the decay of the harmonic signal with ellipticity, enabling HHG from quasi-circular laser pulses[21,22]. Fleischer *et al.* recently demonstrated the ellipticity control of harmonics using a combination of circularly polarized 800 nm and 410 nm pulses[23]. They were able to produce a spectrum consisting of double peaks with alternately left and right circular polarization. This method is interesting because it provides a universal way of generating circular XUV photons with good efficiency. It is particularly appropriate for XUV magnetic circular dichroism experiments, which rely on the measurement of absorption spectra. Nevertheless, this source is not the most appropriate for photoionization of polyatomic molecules, in which each spectral peak produces a broad photoelectron spectrum associated with different cationic states with different ionization dynamics. The overlap of photoelectron spectra from spectrally shifted left and right CPL would make analysis quite complex. Furthermore, on the attosecond timescale this source produces trains of linearly polarized attosecond pulses with alternating polarization direction, and cannot be used for attosecond circular dichroism.

An alternative solution for the generation of elliptical harmonics emerged a few years ago in the form of molecular HHG[24–26]. It was shown that the ellipticity of high harmonics may reach 40% in nitrogen molecules aligned at 60° from the polarization direction of linear generating laser pulses[25,26]. Although this method may provide brighter sources, it is quite cumbersome to implement because of the equipment required to efficiently align molecular samples.

In addition to their rather low efficiency and/or relative complexity, the elliptical HHG experiments reported to date have been unable to unambiguously prove the existence of circular harmonic emission, as they could not distinguish unpolarized radiation from circular radiation[20]. This questions the actual value of the ellipticity and the possibility of using such elliptical sources in practical applications.

In this Letter, we show that resonant HHG in elliptical laser fields provides a unique source of quasi-circular XUV radiation within a simple optical set-up. We demonstrate the influence of resonances on the high-harmonic ellipticity by studying the simple case of argon. Using an XUV polarizer, we find that harmonic 5 from a

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