Opportunities for chiral discrimination using high harmonic generation in tailored laser fields

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Abstract

We discuss future perspectives for chiral discrimination with HHG, the ways of increasing chiral dichroism using tailored laser pulses, new detection schemes involving high harmonic phase measurements, and reconstruction of chiral response in pump-robe schemes.

In high harmonic generation, the electron liberated from the molecule by a strong laser field is first accelerated by the field and then brought back to recombine with the hole left in the parent molecule. Strong dependence of the returning electron energy on the phase of the oscillating laser field ensures that different harmonics are emitted at different electron return times [1]. As a result, each harmonic takes a snapshot of the recombining system at a specific moment [2], giving raise to high harmonic spectroscopy as a tool to study electron dynamics in molecules. The dynamics of the recombining system is recorded in the properties of harmonic light, with attosecond temporal resolution. The time resolution is determined by the difference in the emission times between the consecutive harmonics.

Chiral discrimination with high harmonic generation (cHHG method) has been introduced in recent work [3]. In its original implementation, the cHHG method works by detecting high harmonic emission from randomly oriented ensemble of chiral molecules driven by elliptically polarized field, as a function of ellipticity. Ref [3] also used the high harmonic spectra to resolve the electronic chiral response with 0.1 femtosecond temporal resolution.

We will discuss future perspectives in the development of this novel method, the ways of increasing chiral dichroism using tailored laser pulses, new detection schemes involving high harmonic phase measurements, and concentration-independent approaches to chiral discrimination. Specifically, we consider the application of elliptic counter-rotating two-color laser fields [4,5], to induce, enhance and manipulate chiral response of HHG from randomly oriented gas of chiral molecules.

Using the example of epoxypropane molecule, we show theoretically that application of two-color counter-rotating elliptically polarized laser fields yields an order of magnitude enhancement of chiral dichroism (Fig. 1) compared to single color elliptical fields.

We also describe how one can introduce a new functionality to cHHG: concentration-independent measurement of the enatiomeric excess in a mixture of randomly oriented left and right molecules. We show the importance of harmonic phase measurement and how it can be used in this context.

Finally, for arbitrary configurations of laser fields, we connect the observables of the cHHG method to the amplitude and phase of chiral response, providing a basis for reconstructing wide range of chiral dynamics from cHHG measurements, with femtosecond to sub-femtosecond temporal resolution.

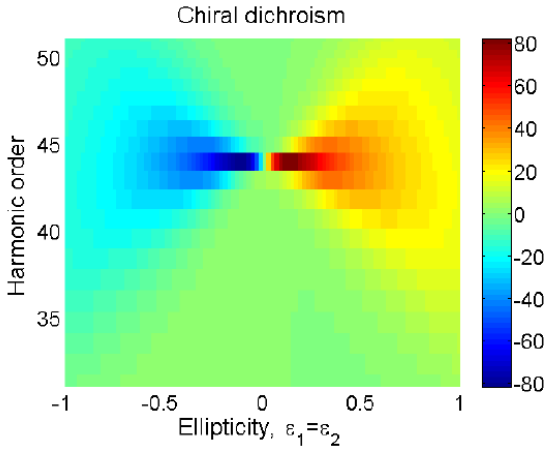

Fig 1. HHG in epoxypropane in two-color counter-rotating elliptically polarized fields. Left panel: Estimated chiral dichroism $Q(N,\varepsilon)= 2(Y_S(N,\varepsilon)-Y_R(N,\varepsilon))/(Y_S(N,\varepsilon)+Y_R(N,\varepsilon))$, where $Y_{S,R}$ is the harmonic yield for left (S) and right (R) molecules.

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