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Ultra-fast nonlinear optical response of chiral molecules with a focus on conformer sensitivity

Tuesday, June 24, 2025 10:00 AM (20 minutes)

Chirality is a fundamental geometric property, present from molecular to macroscopic scales. Traditional chiroptical methods rely on weak magnetic interactions, limiting their efficiency. We aim to develop chiral recognition methods based solely on electric-dipole interactions, offering enhanced enantiosensitivity [1].

We investigate the carrier-envelope phase (CEP) dependence of enantio-sensitive observables within the electric dipole approximation by numerically analyzing the nonlinear response of randomly oriented chiral molecules in the gas phase. Using time-dependent density functional theory (TDDFT), we model their interaction with few-cycle, tightly focused, CEP-controlled linearly polarized laser pulses. Tight focusing induces a longitudinal field component, creating a forward-elliptically polarized field [2]. This drives a chiral response perpendicular to the polarization plane, leading to the emission of even-order chiral harmonics in addition to the odd-order achiral harmonics. Their CEP-dependent interference results in enantiosensitive non-linear optical rotation [2]. Here we explore the sensitivity of the CEP-dependent signal to chiral molecular conformations and the uniqueness of the CEP molecular markers.

We focus on the chiral dynamics of essential amino acids, using serine as a prototypical case. We analyze its three dominant conformers in the gas phase, with relative populations of 43.7%, 18.8%, and 14.8% [3]. By placing a polarizer before the detector, one can convert enantio-sensitive polarization properties into an enantio-sensitive intensity distribution, which can be considered as chiral “QR codes” mapping the chiral dichroism of emitted harmonics as a function of the CEP of the incident light. We show that chiral dichroism (CD) vs. CEP has different patterns for different conformers (note that for the same harmonic order, the CD maximizes at different CEP values in serine I and serine II, see Fig. 1), reflecting different molecular phase accumulation due to ultrafast electron dynamics in two conformers and making chiral QR codes suitable for molecular fingerprinting.

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<img src=”https://www.dropbox.com/scl/fi/xysdk4xjdbfvet3f1ksjf/QR$_{C}ODE_{S}erine.png?rlkey = g3606vfcn1hqnrltw9m61lli7st = tebqlhx4dl = 1”width = ”300” >$

Figure 1: Chiral “QR codes”: Chiral Dichroism (CD) of the emitted harmonics as a function of the CEP of incident light for (a) serine I and (b) serine II conformers.

References:
[1] D. Ayuso *et al.*, PCCP **24**, 26962 (2022). DOI: 10.1039/D2CP01009G
[2] D. Ayuso *et al.*, Optica **8**, 1243 (2021). DOI: 10.1364/NLO.2021.NW2A.2
[3] K. He and W. D. Allen, J. Chem. Theory Comput. **12**, 3571 (2016). DOI: 10.1021/acs.jctc.6b00314

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