Quantum Dynamics in Tailored Intense Fields

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Chiral dichroism in high-order harmonic generation

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High-harmonic generation is a nonlinear process that converts intense infrared radiation into high-frequency light. It can be understood as a sequence of three steps: tunnel ionization, laser-driven acceleration of the electron in the continuum, and recombination with the core resulting in the emission of harmonic light. Since there is a well-defined relationship between the duration of the electron round-trip and the energy released during recombination, the harmonic spectrum provides snapshots of the dynamics in the ion. The recent application of elliptically polarized fields to the generation of high-order harmonics in chiral molecules has allowed to probe chiral electron dynamics with sub-femtosecond time resolution [1], opening new directions in high-harmonic spectroscopy [2]. In general, the chiral response of a system increases with the ellipticity (chirality) of the driving field, usually maximizing for circularly polarized light. However, the harmonic signal quickly drops with ellipticity as the liberated electron misses recollision with the core. In this context, the use of two-color counter-rotating bi-circular fields [3] constitutes a promising tool for exploring time-resolved chiral dynamics as they allow recombination while maximizing chiral responses.

We have calculated the high-order harmonic spectra of the chiral molecule propylene oxide interacting with different configurations of laser fields. The chiral response of the system arises due to the interplay between electric and magnetic effects. Our calculations have been performed using the method described in [4], including accurate photorecomination matrix elements evaluated in the framework of density functional theory [5,6]. The resulting harmonic dipole has been averaged coherently on a Lebedev grid in order to account for the experimental condition of randomly oriented molecules. In this presentation, I will discuss the most relevant features arising in the high-order harmonic spectra of propylene oxide, showing that high-harmonic spectroscopy constitutes a promising tool for probing chiral electron dynamics with sub-femtosecond time resolution.

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