S E M I N A R

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PHY 2.1.29 (seminar room Huber), 10.15 a.m.

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High-harmonic spectroscopy using bi-elliptical fields

Over the recent years, high-harmonic generation has established itself as a promising spectroscopic technique. Notable examples include the tomographic imaging of molecular wave functions [1], the tracking of nuclear dynamics [2], and the reconstruction of the attosecond time-scale electron dynamics in molecules [3, 4]. All these applications, however, have been limited to laser fields with linear polarization. High harmonic generation has only recently been extended to circularly-polarized drivers by utilizing a technique known as bi-circular high-harmonic generation (BHHG) [5,6,7]. In this talk, I will demonstrate the spectroscopic applications of this technique to the study of structure and dynamics of gas-phase atoms and molecules in a self-probing manner. I will start with an analysis of the helicity asymmetry of BHHG in noble-gas atoms and then proceed by illustrating how BHHG can be applied to study dynamical symmetry-breaking in a time-dependent manner in the context of rotational and vibrational molecular motion.

Extension of high-harmonic generation to the regime of highly-elliptical fields opens up the way towards the study of chiral phenomena in high-harmonic generation. In this work, circular dichroism in the range of 3-8 % is observed on randomly oriented methyl oxirane (C₃H₆O) molecules in the gas phase. This chiral sensitivity is attributed to the sub-cycle chiral electron dynamics involving excited states of the cation that take place during the electron continuum propagation. Finally, I will present a study of time-dependent chirality based on following the temporal evolution of circular dichroism during the course of an ultrafast photodissociation reaction.