Multiphoton PECD in the Ionization of Fenchone and Camphor by Short Intense Laser Pulses

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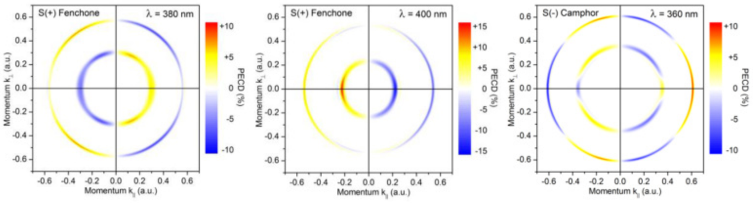

Figure 1: Three- and four-photon PECD for fenchone and camphor. On the left: the multiphoton PECD of fenchone computed for the photon wavelength of 380 nm. In the middle: the PECD of fenchone computed for the photon wavelength of 400 nm. On the right: the PECD of camphor computed for the photon wavelength of 360 nm. The photoelectrons emitted due to the three-photon ionization form the inner ring, while the photoelectrons emitted due to the four-photon process the outer ring. The PECD signal is obtained as the relative difference between the electron spectra computed for left and right circularly polarized pulses. The signal is normalized to the maximal intensities of the respective spectrum. The pulse propagates along the laboratory z-axis, which coincides with k_{||}.

Introduction

Chiral molecules cannot be brought into coincidence with their mirror images. Therefore, a molecule and its mirror image have different properties when interacting with circularly polarized light, such that the angular distribution of the emitted photoelectrons depend on whether the light was left or right circularly polarized. The difference of two distributions is called photoelectron circular dichroism (PECD). The PECD depends on the chosen molecule and experimental parameters of the laser pulse. In the literature, the dependencies of the three- and four-photon PECD of camphor and fenchone on the wavelength and intensity of the pulse were studied experimentally. A reliable quantitative theoretical interpretation of these experimental results is missing.

Methods

In this project, the time-dependent single center (TDSC) method is used to study the multiphoton PECD of fenchone and camphor theoretically. The method consists in the propagation of a one electron wave packet in a chiral potential while interacting with a short circularly polarized laser pulse. To simulate the photoionization of randomly oriented molecules in the gas phase, it is necessary to perform calculations with a large number of different molecular orientations in space. This justifies the usage of a high performance computer cluster. In order to simulate the available experiments, the calculations were performed for several wavelengths and peak intensities of the laser pulse for each of the considered molecules.

Results

Figure 1 depicts the presently computed three- and four-photon PECD (in percent) of fenchone and camphor for different wavelengths of the pulse. It shows the difference between the spectra computed for the left and right circular polarized light normalized to the maxima of the intensity in each spectrum. The PECD signal appears as a ring at a given electron momentum k which obeys the energy conservation. One can see, that the computed PECD depends strongly on the chosen wavelength of the pulse (for 400 and 380 nm it has an opposite sign for fenchone), as well as on the molecule (it exhibits different nodal structures for fenchone and camphor).

Discussion

The theoretical results obtained with the TDSC method are in a good agreement with the available experimental data. This demonstrates the capability of the used theoretical method to predict multiphoton PECD for different real molecules. Further theoretical studies of PECD require calculations for tailored laser pulses, such as: Lissajous type electric field configuration, broad band coherent excitation, and tunnel ionization by infrared pulses.

Publications

Müller, A.D. et al. Photoelectron circular dichroism in the multiphoton ionization by short laser pulses. II. Three- and four-photon ionization of fenchone and camphor. J. Chem. Phys. 148 (2018) 214307
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https://doi.org/10.17170/kobra-2018121761

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