Multicoincidence Studies of Ionization of Chiral Molecules in Strong Laser Fields

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Synopsis: We study the three-dimensional momentum distribution of the photoelectrons of a chiral molecule employing circularly polarized light (800nm). Dependencies of the spatial orientation of the molecule, as well as its fragmentation channel are examined.

The investigation of chiral systems is of great interest, especially the determination of the configuration is a keystone to stereochemistry. Chiral molecules exist as two enantiomers, which are mirror images of each other. The difference in the interaction between a chiral molecule and left and right handed circularly polarized light has been an important probe for the investigation of chiral molecules, termed circular dichroism (CD). Unfortunately, this effect is very weak which impedes deeper analysis and studies on single molecules.

The observation of the photoelectron circular dichroism (PECD, a helicity dependent forward / backward asymmetry of electron emission with respect to the light propagation) has proven PECD to be a comparatively strong effect for the investigation of chiral systems. Most recently it was shown that PECD is an universal effect in all ionization regimes (single photon, multiphoton, above-threshold and tunnel ionization), occurring as an inherent chiral feature.[1] However, employing velocity map imaging spectrometers, which measures 2D projections of the 3D photoelectron distributions, experiments have been limited to the observation of randomly oriented molecules, confining (when examining with circularly polarized light) the observation of an asymmetry in the forward/backward direction of the electron emission.

With the COLTRIMS technology, [2,3] however, one is now able to (partially) orient the molecules in space by post selection of fragment channels and fragment ion directions, which becomes possible as the 3D momenta of all ionic fragments are measured in coincidence.

For single photon absorption, it was reported that (partially) orienting the molecule in space enhances the PECD dramatically [4]. For these uniaxially oriented molecules the sign of the PECD remains unchanged upon flipping this molecular axis from the forward to the backward directions with respect to the light propagation axis.

At first we investigated the electron emission from enantiopure methyl-oxirane in the tunneling regime for partially oriented molecules. (800 nm, 40 fs, circularly polarized)

Additionally, we investigated the electron emission out of a racemic mixture of CHFBr. Previously, we already used laser induced Coulomb Explosion Imaging to determine the absolute configuration of a single molecule [5]. By means of post selection of the ionic fragments we determined both the 3D molecular orientation and the fixed-in-space electron emission pattern.

References

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