Investigation of Photoelectron Circular Dichroism induced by Strong Field Ionization

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Synopsis: We studied the three-dimensional momentum distribution of photoelectrons released by circularly polarized light (800 nm) from chiral molecules. Strong dependencies of the spatial orientation of the molecule, as well as its fragmentation channel were observed.

Chiral molecules exist as two enantiomers, which are mirror images of each other, a left- and a right-handed version. The difference in the interaction between a chiral molecule and chiral probe, as circularly polarized light, has been an important tool for chiral discrimination. Unfortunately, this circular dichroism (CD) is usually very weak. Instead of investigating the total CD in the absorption probability, a much stronger signal can be obtained by measuring emitted electrons. This so-called photoelectron circular dichroism (PECD) is on the percent level. It results in a forward/backward asymmetry in the electron emission with respect to the light's propagation direction. Most recently it was shown that PECD is an universal effect in all ionization regimes (single photon, multiphoton, tunnel ionization and above-threshold ionization), occurring as an inherent chiral feature [1]. For laser-induced ionization PECD has so far been investigated with velocity map imaging, which limits to randomly oriented samples. However, for single photon absorption it was reported that (partially) orienting the molecule in space enhances the PECD signal [2].

Therefore we employ the COLTRIMS reaction microscope technology [3,4] for a more detailed investigation. A gas jet containing the sample molecules is intersected with a strongly focused laser pulse (40 fs, 800 nm). The created electrons and ions are projected with a rather strong electric field (100 V/cm) on position and time-sensitive micro channel plate (MCP) detectors with delayline anode. From the position of impact and flight the momenta of all charged particles can be derived. Using a rather high electric field has two advantages: the fragments hitting the MCP detector have enough energy to directly create a signal instead of the prerequisite of post acceleration. This makes the use of meshes obsolete and enhances the five particle detection efficiency by a factor 3. The high field also results a better fragment mass separation. Upon ionization (single or multiple) the molecule can dissociate or undergo Coulomb explosion. Measuring the ionic fragments allows post orienting the molecule in space or at least fixing on axis. Also the fragment mass gives additional information, which molecular states were ionized.

Here we present data for enantio-pure methyloxirane in the regime of tunnel ionization. The intensity was chosen to be rather low, such that the HOMO-1 is singly ionized and dissociates in a C_2H_3O + CH_3. In addition, a reduced amount of double and triple ionization was observed. The strength of PECD and how it depends on the (partly) orientation will be discussed. In a second experiment, we investigated the electron emission from a racemic mixture of CHFClBr. Therefore, we first determined the absolute configuration of Coulomb exploding molecules after 4- and 5-fold ionization for each molecule [5] and then post oriented the molecule in space. While for randomly oriented molecules, we couldn't observe any PECD, for certain orientations quite strong effects were observed.

References

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