Ultrafast chiroptical interaction with strutured light

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The goal of my thesis is to study chiral light-matter interaction on the molecular scale and in particular to develop new forms of chiroptical spectroscopy finding new way in which we can distinguish enantiomers. A key tool to investigate and distinguish chiral molecules in the past has been the Spin angular momentum (SAM) of light during photoelectron angular distributions (PAD) spectroscopies [1]. Only more recently the influence of the Orbital angular momentum (OAM) of light during the photoionization process has been taken into account as well. Recently, a two-color photoionization experiment, using a circularly polarized extreme ultraviolet beam and a circularly polarized infrared (IR) beam carrying OAM, revealed a helical dichroism (HD) around 10% in Helium[2]. In this work we investigate the influence of both OAM and SAM in PAD in different ionization regimes, using two-color multiphoton ionization by time-overlapping femtosecond laser pulses, with a 515 nm beam carrying SAM $\sigma_{515} = \pm 1$ and a 1030 nm beam with SAM $\sigma_{1030} = \pm 1$ and OAM $m_{1030} = \pm 1$. First, we used 4 green photons to excite Rydberg states of Ethanol, and then from 1 to 5 IR photons to ionize the excited molecules. In this case we observed a Helical Dichroism, defined as:

$$HD = PAD(\sigma_{515}, \sigma_{1030}, m_{1030}) - PAD(\sigma_{515}, \sigma_{1030}, -m_{1030}) \quad (1)$$

on the order of 10 %, which vanishes if the Rydberg wavepacket is created by a linearly polarized beam σ_{515}=0. Second, we increased the 515 nm intensity to generate Above-threshold-ionization peaks and reduce the 1030 nm intensity to produce perturbative continuum-continuum transitions. The resulting sidebands show a HD around 3%, which persists with σ_{515}=0. The helical dichroisms we measured result from light-matter interaction beyond the electric dipole approximation. Non-dipole terms are generally weak, but the specific configurations we used – using delocalized Rydberg electrons, or delocalized continuum electrons – increases their contributions. We thus expect that this geometry will be favorable to probe molecular chirality, which is the next step of the project.

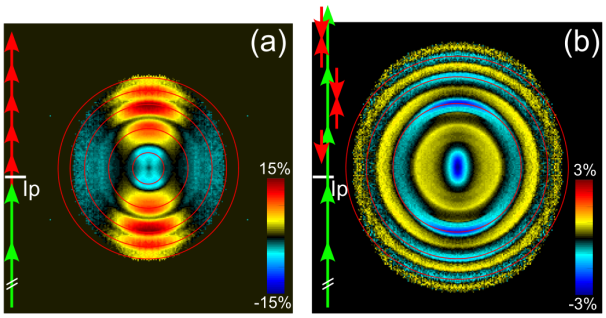

Figure 1: Normalized HDs measured with 1030nm and 515nm circular pulses in ethanol for the Ryberg excitation a) and ATI regime b) using a velocity map imaging spectrometer.

References

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[2] G. De Ninno *et al.* Nat. Phot. 14, (2020).