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Non-linear and Ultrafast Circular Dichroism Measurements at FELs

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The combination of polarization control with the unique properties of free-electron lasers in terms of high intensity and ultrashort pulse duration offer unique perspectives on non-linear and time-resolved insights in “handed” systems. Handedness, i.e. chirality, is of crucial interest for understanding the origination of life on earth but also in the spin properties of fundamental as well as complex systems.

A common technique to study chiral properties is to measure the absorption differences of opposing helicities of circularly polarized light, the so called circular dichroism (CD). For spin interactions with circularly polarized light, photoelectron spectroscopy can be used to determine dichroic yield differences in the dipole plane. For chiral molecular structures, the dichroism is also imprinted in the symmetry breaking of the electron emission beyond the dipole plane, i.e. the photoelectron circular dichroism (PECD). PECD is by far more sensitive to the molecular structure and therefore, a superior technique for chiral sensing.

Regarding the interaction between helical light and atomic systems, we present a highly non-linear study exploring electron yield and angular distribution imprinted CD in multi-photon two-color ionization in helium. This study reveals up to perfectly dichroic absorption and a helicity dependence on an optical laser induced Stark shift in ionic helium.

As first approach to establish FELs as tools for ultrafast, site-specific investigations of structurally chiral molecules, we furthermore present a variety of aspects from recent and present studies at LCLS, SSRL, FLASH and FERMI that are crucial to ultimately allow for time-resolved PECD studies on expanding molecular systems. Together with further studies of non-dichroic origins of symmetry breaking in multi-photon absorption, the complexity of PECD is discussed in the light of future, time-resolved FEL applications.

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