Ultrafast time-resolved probes of chirality during photochemical reactions

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In this talk, we will focus on the first time-resolved PECD experiments with circularly polarized harmonics, thereby introducing a general experimental approach for chiral femtochemistry. These experiments include several novel aspects: Firstly, we have introduced a simple approach to generate circularly polarized VUV radiation based on low-order-harmonic generation. Secondly, we have performed a complete polarization measurement of the generated VUV harmonics. Thirdly, we have combined this light source with an angle-resolved photoelectron detection to measured time-resolved photoelectron circular dichroism (PECD).

Selected chiral molecules are ionized in a 1 + 1' pump-probe scheme involving linearlypolarized 266 nm and circularly-polarized 133 nm pulses with sub-100 fs temporal resolution. One of the structurally simplest chiral molecules, CHFBrI, will be discussed in terms of its molecular dynamics and time-dependent chirality associated with the photoinduced C-I bond breaking. The experimental results are supported by high-level *ab initio* electron-molecule scattering calculations. The broad applicability of the time-resolved PECD measurement scheme is demonstrated on another chiral molecule, 2-iodobutane. Whereas CHFBrI displays a non-vanishing PECD at long pump-probe delays, the PECD decays to zero after the photo-dissociation of 2-iodobutane, reflecting the effective chirality of the product radicals on long timescales.