Supporting Information

Link between Photoelectron Circular Dichroism and Fragmentation Channel in Strong Field Ionization

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Error discussion in the calculation of the PECD

Assuming that reversal of the helicity of the ionizing light is equivalent to mirroring the enantiomer for the PECD, it should make no difference whether the two enantiomers or the two light helicities are included in the normalized difference. Experimentally, however, there are obvious differences: The two polarizations of the light are not perfectly mirror-symmetrical, but with a motorized stage, they can easily be swapped on a short timescale (here every three minutes). This compensates for the long-term laser drifts.

Although the two enantiomers are perfectly mirror-symmetrical, they cannot be swapped rapidly. With an often quite low vapor pressure, the chiral molecules remain in the gas lines for a long time, contaminating the sample measured second upon a rapid change. In the present experiment, R-methyloxirane was changed to S-methyloxirane only once and each sample was measured for 2-3 days. Thus, in the comparison of the enantiomers, the long-term drifts of the laser play a role. In Fig. S1, the PECD is calculated by the normalized difference of the enantiomers or the helicities of the light, respectively. Although qualitatively equal (sign of the PECD, magnitude and sign change at approximately p_{et} = 0.7 a.u.), the two PECD maps differ in their details.

Which of the two graphs is closer to the exact PECD does not only depend on the stability of the laser or the circularity of the circularly polarized light. Relevant quantities are also the dependence of the PECD on the ellipticity of the light and the dependence of the PECD on the transversal momentum.

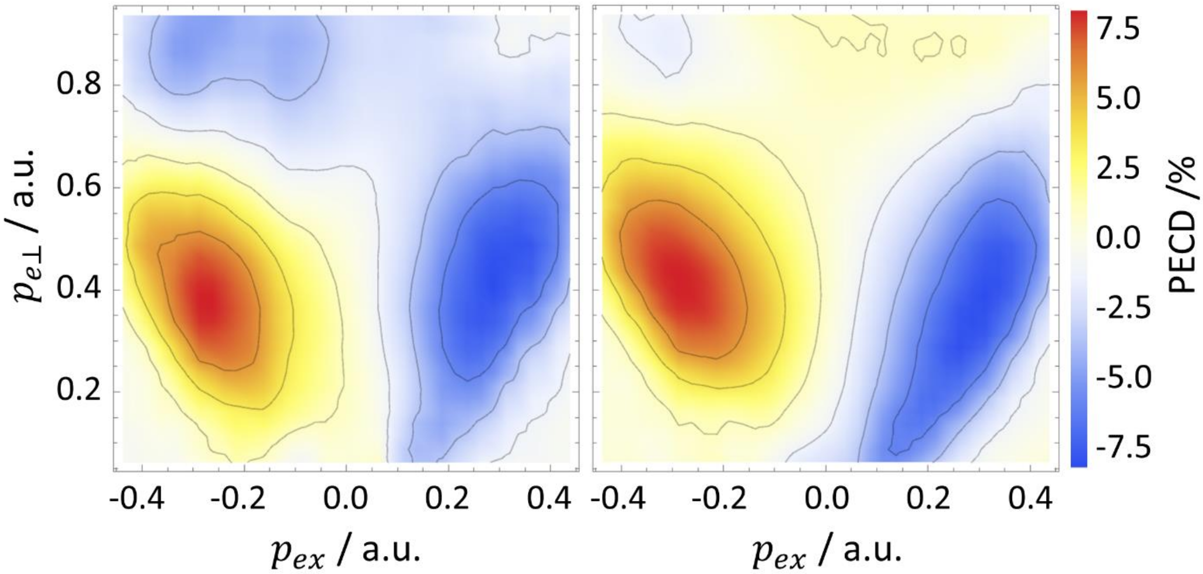

Fig. S1 Comparison between the normalized difference between the enantiomers or the helicities of light, gating on the parent ion. Left: PECD, calculated from the normalized difference between the enantiomers. $PECD(p_{ex}, p_{et}) = \frac{R_{LCP}(p_{ex}, p_{et}) - S_{LCP}(p_{ex}, p_{et})}{R_{LCP}(p_{ex}, p_{et}) + S_{LCP}(p_{ex}, p_{et})}$. Right: Normalized difference between the helicities of the light. $PECD(p_{ex}, p_{et}) = \frac{R_{LCP}(p_{ex}, p_{et}) - R_{RCP}(p_{ex}, p_{et})}{R_{LCP}(p_{ex}, p_{et}) + R_{RCP}(p_{ex}, p_{et})}$.