REAL-TIME MONITORING OF CHIRALITY LOSS IN MOLECULAR PHOTODISSOCIATION BY TRANSIENT X-RAY CIRCULAR DICHROISM

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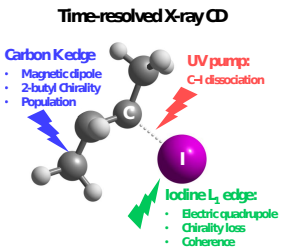

Newly developed X-ray sources offer valuable insight on molecular dynamics with unprecedented spatiotemporal resolutions and element sensitivity. Significant advances made in the control of the polarization of X-ray beams enables real-time monitoring of molecular chirality, which is an indispensable subject for understanding and controlling biological process. We theoretically apply time- and frequency-resolved X-ray Circular dichroism (TRXCD) spectroscopy to discern the time evolution of molecular chirality at different element windows during the photodissociation of 2-iodobutane. Following an optical excitation, the iodine atom dissociates from the chiral center, which we capture by quantum nuclear dynamics simulations. A resonant X-ray pulse then probes the iodine or carbon atom through an element-specific core-to-valence transition. The TRXCD signal at the iodine L_1-edge captures the timing of chirality loss, c.a 70 fs. The strong electric dipole–electric quadrupole (ED−EQ) response at this high X-ray regime makes this signal sensitive to vibronic coherences. At the carbon K-edges, the signals re-capture the chirality of 2-butyl radical and the spin state of the iodine atom. The stronger electric-magnetic dipole response make the signals more intuitive for the electronic population than coherence. Overall, the element-specific TRXCD signal offers a unique window into the time-dependent chirality of molecules.

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