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What happens after a molecule absorbs a photon

Light induced processes fundamentally rely on using electronic motion enabled by photon absorption. Whether photoisomerisation, proton or electron transfer, all cases are not driven by the initial absorbed photon; instead the molecule efficiently converts the obtained energy into the desired dynamics. Responsible are the underlying potential energy surfaces and crossings between them such as conical intersections, a paradigm for the breakdown of the Born-Oppenheimer approximation. Although they are well-understood theoretically, they have remained experimentally largely inaccessible. I will show how the observation of atomic motion in real-time can provide unique information on molecular dynamics and the associated potential energy surfaces. By launching vibrational wavepackets in one electronic state and investigating the effect of electronic surface crossings on their dynamics, we can study how nuclear degrees of freedom couple electronic states. We observe drastically different signatures for unreactive vs reactive internal conversion that point to the involvement of few critical coordinates and provide evidence for multi-molecular conical intersections mediating singlet fission.