Imaging orbitals from the attosecond emission of aligned molecules

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A strong laser field interacting with molecules drives ultrafast intra-molecular electron wavepackets, resulting in the emission of attosecond XUV bursts. In linear molecules, the interaction of the laser-driven electron wavepacket with the core leads to quantum interferences in the recombination step. We characterized the attosecond emission from transiently-aligned CO2 molecules and demonstrated that these interferences can be finely controlled by changing the electron recollision angle. Our control of the interference results in an attosecond pulse shaping [Boutu et al., Nature Physics 2008]. Moreover our measurements give direct access to the transition dipole matrix elements between the continuum states and the molecular orbitals involved in the emission process. When the HOMO gives the dominant contribution, a tomographic reconstruction is possible within a plane wave approximation for the recolliding electron. We performed a fully experimental reconstruction of the N2 HOMO, where the polarization-resolved harmonic emission was characterized in amplitude and phase. This reconstruction exhibits Angström spatial resolution, mainly limited by the accessible spectral range.