Imaging orbitals with attosecond and Angström resolutions

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An intense short (few-femtosecond) laser pulse interacting with a molecule in the gas phase may liberate by tunnel ionization an attosecond electron wave packet (EWP). This EWP is then accelerated by the laser field and made to recollide with the core one laser cycle later. The attosecond XUV emission resulting from the recombination encodes rich information on the possibly transient electronic [1,2] and nuclear [3,4] configuration of the core. The recolliding EWP may be seen as a probe of the core in the direction of the laser field. By characterizing the attosecond emission (in intensity, phase and polarization) for different molecular alignment angles, one can access the recombination dipole moment in the molecular frame. The *spectral phase* of this transition dipole is a unique quantity that cannot be accessed through other means, like photoionization experiments. It encodes the structure of the radiating molecular orbital and allows the reconstruction of the orbital amplitude and phase using a tomographic procedure [1,5,6]. It thereby becomes possible to image orbitals with a spatial resolution in the ångström range, and a temporal resolution in the attosecond range [5]. This paves the way to monitoring deformations of these orbitals during chemical reactions [7].

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