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Time and Space Resolved High Harmonic Imaging of Electron Tunnelling from Molecules

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High harmonic generation in intense laser fields carries the promise of combining sub-Angstrom spatial and attosecond temporal resolution of electronic structures and dynamics in molecules, see e.g. [1-3]. High harmonic emission occurs when an electron detached from a molecule by an intense laser field recombines with the parent ion [4]. Similar to Young's double-slit experiment, recombination to several "lobes" of the same molecular orbital can produce interference minima and maxima in harmonic intensities [1]. These minima (maxima) carry structural information – they occur when the de-Broglie wavelength of the recombining electron matches distances between the centers. We demonstrate both theoretically and experimentally that amplitude minima (maxima) in the harmonic spectra can also have dynamical origin, reflecting multi-electron dynamics in the molecule. We use high harmonic spectra to record this dynamics and reconstruct the position of the hole left in the molecule after ionization. Experimental data are consistent with the hole starting in different places as the ionization dynamics changes from tunnelling to the multi-photon regime. Importantly, hole localization and subsequent attosecond dynamics are induced even in the tunnelling limit. Thus, even "static" tunnelling induced by a tip of a tunnelling microscope will generate similar attosecond dynamics in a sample. We anticipate that our approach will become standard in disentangling spatial and temporal information from high harmonic spectra of molecules.

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