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High harmonic spectroscopy of hole dynamics: where is the hole created after tunnel ionization of a molecule?¹ OLGA SMIRNOVA, ALEX HARVEY, FELIPE MORALES, MARIA RICHTER, MISHA IVANOV, SERGUEI PATCHKOVSKII, Max-Born Institut, Max-Born-Str 2A, 12489 Berlin, Germany, YANN MAIRESSE, CELIA, Universite de Bordeaux - CNRS - CEA, F33405 Talence, France — Ultrafast ionization of a molecule creates a multi-electron wavepacket in the ion. Its dynamics can be viewed as a motion of a hole. One of the most interesting questions, and one of the key experimental challenges, is to identify the initial shape, location and momentum of the created hole. If the hole is created via tunnel ionization, should it always be aligned with the direction of the laser electric field? Should it be initially located on the 'down-stream side' of the molecule, adjacent to the potential barrier through which the electron was removed by tunneling? We use high harmonic spectroscopy to address this question, combining accurate theoretical modelling with detailed experimental measurements. We find that the location of the hole does not follow the simplistic picture of adiabatic tunneling: the hole is not created adjacent to the potential barrier through which the electron tunnels out. Our results are obtained in the regime where several electronic states of the cation are strongly coupled by both the ionizing field and via the electron-electron correlation. The displacement of the hole might be indicative of correlation-induced time-delays during multi-photon ionization, similar to correlation-induced delays known for one-photon ionization.

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