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Conical intersection dynamics probed by homodyne high-harmonic spectroscopy

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High-harmonic spectroscopy is now established as a powerful method to probe the structure and dynamics of the valence shell of molecules. Recently, we have extended this technique to the time-resolved observation of chemical reactions. Exploiting the homodyne interference between the excited and unexcited molecules in a transient grating geometry, we were able to characterize the evolution of the electronic structure of Br_2 undergoing an adiabatic dissociation [1]. Here, we show that high-harmonic spectroscopy reveals electronic dynamics that occur when a photoexcited nitrogen dioxide molecule (NO_2) crosses a conical intersection. The electronic symmetry changes that occur as the molecule oscillates across the conical intersection appear as modulations in the coherently detected high-harmonic signal. Taking the measurement to longer delays, we observe the onset of the statistical dissociation dynamics leading to NO($^2\Pi$) and O(^{3}P).

[1] H. J. Wörner, J. B. Bertrand, D. V. Kartashov, P. B. Corkum and D. M. Villeneuve, Nature **466**, 604-607 (2010)