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**Time-Resolved Observation of Molecular Dissociation Using High Harmonic Generation** JULIEN B. BERTRAND, University of Ottawa, HANS JAKOB WÖRNER, DANIIL KARTASHOV, DAVID M. VILLENEUVE, PAUL B. CORKUM — We have realized the first experiments towards dynamic orbital tomography by observing high harmonics generation from dissociating molecules. A pump pulse centered at 400 nm launches a dissociative wave packet in an electronically excited state and a delayed 800 nm pulse generates high harmonics from the excited sample. In a complementary experiment, the ion yield of the two-color experiment has been measured and the range of intensities corresponding to dominant single-photon excitation of the molecules has been identified. In both systems, excitation enhances ionization at short pump-probe delays but decreases the harmonic yield. The results obtained for the dissociation of Br2 and NO2 are compared and found to reveal characteristic details of two fundamentally different dissociation mechanisms. The requirements for dynamical orbital tomography are discussed.

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