Windowless Observation of the Photodissociation Dynamics of CS_2

A.D. Smith[1], E. Warne [1], D. A. Horke [2], D. Bellshaw [4], C.M. Cacho [3], A.J.H. Jones [3], E. Springate[3*], A. Kirrander [4], R. T. Chapman [3], R.S. Minns [3]

[1] *Chemistry, University of Southampton, Highfield, Southampton SO17 1BJ, UK*

[2] *Center for Free-Electron Laser Science, DESY, Notkestrasse 85, 22607 Hamburg, Germany and The Hamburg Centre for Ultrafast Imaging, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany*

[3] *Central Laser Facility, STFC Rutherford Appleton Laboratory, Didcot, UK*

[4] *EaSTCHEM, School of Chemistry, University of Edinburgh, David Brewster Road, Edinburgh, EH9 3FJ, UK*

Time resolved photoelectron spectroscopy with high harmonic generation (HHG) sources is a windowless technique as all states can in principle be ionized, allowing observation of the important reaction intermediates and products. We present measurements with a 6 eV pump and 21 eV HHG probe that allow for the unambiguous assignment of the full reaction pathway in a polyatomic molecule. Our measurements follow the dissociation dynamics of CS_2, including ground state depletion, population of multiple excited electronic states, dynamics in the initially excited singlet state, subsequent conversion into the triplet manifold of states (Figure 1 A) as well as production of the final dissociation products. The HHG probe allows us to monitor the entire reaction coordinate, in comparison with previous studies which only measured the singlet state, even when 6 eV [1] or 7.8 eV [2] probe photons were used.

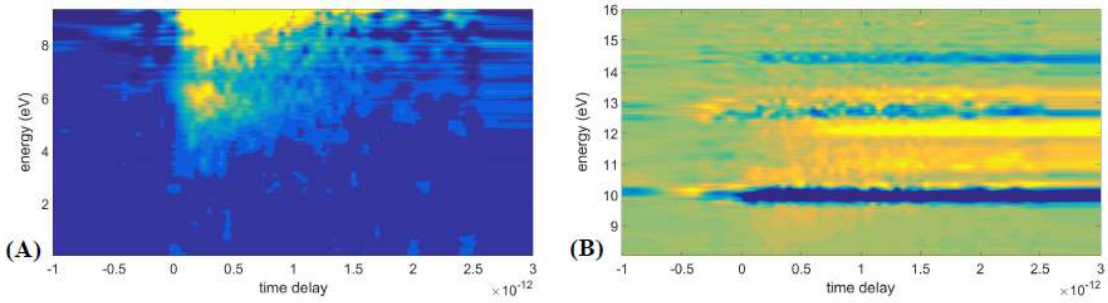

Figure 1. Time-resolved electron energy spectra of the binding energy ranges covered by the excited single and triplet states (A) and by the ground state depletion and final dissociation products (B).

References:

[1] D. Bellshaw, D.A. Horke, A.D. Smith, H.M. Watts, E. Jager, E. Springate, O. Alexander, C. Cacho, R.T. Chapman, A. Kirrander, R.S. Minns, Chem. Phys. Lett, online (2017).

[2] R. Spesyvtsev, T. Horio, Y. -I. Suzuki, T. Suzuki, J. Chem. Phys **142**, 074308 (2015).