Toward Femtosecond Time-Resolved Inner-Shell Transient Absorption Spectroscopy of Ultrafast Dynamics in Sulfur-Containing Molecules

MING-FU LIN, DANIEL NEUMARK, Department of Chemistry, UC Berkeley, STEPHEN LEONE, Department of Chemistry and Physics, UC Berkeley, OLIVER GESSNER, USXL, CSD, Lawrence Berkeley National Laboratory — Sulfur-containing compounds play an important role in many applications such as polythiophene-based organic solar cells or the removal of sulfur compounds by hydrodesulfurization in the petroleum industry. Ultrafast relaxation dynamics (e.g. dissociation, internal conversion and intersystem crossing) of sulfur-containing molecules after photoexcitation have attracted considerable attention as a pathway to a better understanding of the fundamental chemistry of these systems. The novel technique of extreme ultraviolet (XUV) femtosecond transient absorption spectroscopy provides real-time access to the time-dependent structure and transient electronic states of molecules in the vicinity of a specific atom. The usable photon energy range of a high-order harmonic based XUV transient absorption setup has been extended up to 180 eV, enabling measurements in the vicinity of the sulfur 2p edge (165 eV). This new capability opens the route to monitor ultrafast intramolecular dynamics from the unique perspective of well-localized sulfur atoms. Preliminary results will be presented on the photoinduced ultrafast ring-opening, dissociation and vibrational relaxation dynamics in gaseous thiophene (C₄H₄S) and carbon disulfide (CS₂).

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