Abstract Submitted
for the DAMOP20 Meeting of
The American Physical Society

Resonant nonlinear X-ray spectroscopy of acetyl fluoride for probing electronic dynamics with space selectivity[1] STEFANO M. CAVALETTO, SHAUL MUKAMEL, Department of Chemistry, University of California, Irvine — Ultrashort, coherent X-ray pulses from X-ray free-electron lasers (FELs) enable direct access to core excited states in ultrafast time scales. They can be used to extend multidimensional nonlinear spectroscopy into the X-ray regime, and probe the dynamics of a large number of valence and core excited states thanks to their broad bandwidths. Here, we present resonant X-ray sum-frequency-generation (XSFG) signals in order to access novel information on molecular dynamics. In XSFG, the dynamics of a valence electronic wave packet, initiated by an optical or ultraviolet pulse, are subsequently probed by a coherent resonant X-ray pulse with a properly set delay. By resonantly exciting different element-specific core states, the X-ray probe can be employed to monitor the molecular dynamics with high spatial selectivity. Due to their short wavelengths, hard-X-ray FEL pulses are also sensitive to spatial variations within the size of the molecule. Using the minimal-coupling Hamiltonian, we show how XSFG can reveal effects going beyond a description of the light-matter interaction based on the multipole expansion.

[1]S.M.C. was supported by the Alexander von Humboldt foundation through the Feodor Lynen program, and by the Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy through Awards No. DE-SC0019484.

Stefano Cavaletto
Department of Chemistry, University of California, Irvine

Date submitted: 30 Jan 2020 Electronic form version 1.4