

Stimulated x-ray Raman Scattering at x-ray Free-Electron Laser Sources

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The invention of x-ray free-electron lasers (XFELs) opens the pathway to transfer powerful spectroscopic techniques from the optical to the x-ray region, to ultimately study the interplay of coherent electronic and vibrational dynamics on the timescale of chemical reactions. Resonant Inelastic X-ray Scattering (RIXS) is a viable spectroscopic tool to study the valence-electronic environment of a target atom, which can be selectively addressed by the resonantly tuned x-ray beam. The cross section for this process is, however, small, so that even with powerful XFEL pulses the recording of single-shot RIXS spectra of dilute samples or molecules in the gas phase are challenging. This hampers the application of RIXS in optical pump x-ray probe experiments, which would, for example, be one method of choice to study light-induced chemical reactions. Here we demonstrate how this bottleneck can be overcome by stimulating the RIXS process. We present results of a recent experiment on stimulated RIXS at the Linac Coherent Light Source XFEL in a gas sample of atomic neon, thereby amplifying the RIXS signal by 8 orders of magnitude. Using broadband XFEL pulses, which are characterized by a stochastic, spiky substructure of their spectrum, both “pump” and “dump” photons can be provided in a single pulse, thereby stimulating the scattering process and driving an exponential amplification of the signal. Despite the overall broad bandwidth, high-resolution RIXS spectra can be achieved by statistical analysis of a series of stochastic single-shot spectra. The energy resolution is determined by the spectral coherence of the XFEL source, and in principle allows for vibrational resolution. These findings open up a new class of experiments at XFEL sources, with a colossal increase of the RIXS signal. Results of our recent experiment will be presented, along with a perspective and theoretical feasibility study of stimulated RIXS measurements in small molecules.