HIGH-RESOLUTION X-RAY STIMULATED RAMAN SPECTROSCOPY USING STOCHASTIC PULSES

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X-ray free-electron lasers (XFELs) generate high-intensity x-ray pulses which enable x-ray nonlinear spectro-scopies. The extension of nonlinear spectroscopies to the x-ray domain promises the observation of electronic dynamics on their natural timescales with atomic spatial resolution. Stimulated x-ray Raman spectroscopy is an especially powerful tool, which in a propagation geometry combines large signal enhancement through stimulated emission with ultrahigh energy resolution that overcomes core-hole lifetime broadening. We present high-resolution stimulated Raman spectroscopy realized using stochastic XFEL pulses and correlation techniques. A covariance map between the transmitted SASE pulse and the stimulated Raman scattering produces a high-resolution x-ray Raman spectrum. This promising tool could be applied to study ultrafast electronic and molecular dynamics such as charge transfer in complex systems.