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Nonlinear X-Ray and Auger Spectroscopy at X-Ray Free-Electron Laser Sources

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X-ray free-electron lasers (XFELs) open the pathway to transfer non-linear spectroscopic techniques to the x-ray domain. A promising all x-ray pump probe technique is based on coherent stimulated electronic x-ray Raman scattering, which was recently demonstrated in atomic neon. By tuning the XFEL pulse to core-excited resonances, a few seed photons in the spectral tail of the XFEL pulse drive an avalanche of resonant inelastic x-ray scattering events, resulting in exponential amplification of the scattering signal by of 6-7 orders of magnitude. Analysis of the line profile of the emitted radiation permits to demonstrate the cross over from amplified spontaneous emission to coherent stimulated resonance scattering. In combination with statistical covariance mapping, a high-resolution spectrum of the resonant inelastic scattering process can be obtained, opening the path to coherent stimulated x-ray Raman spectroscopy. An extension of these ideas to molecules and a realistic feasibility study of stimulated electronic x-ray Raman scattering in CO will be presented. Challenges to realizing stimulated electronic x-ray Raman scattering at present-day XFEL sources will be discussed, corroborated by results of a recent experiment at the LCLS XFEL. Due to the small gain cross section in molecular targets, other nonlinear spectroscopic techniques such as nonlinear Auger spectroscopy could become a powerful alternative. Theory predictions of a novel pump probe technique based on resonant nonlinear Auger spectroscopic will be discussed and the method will be compared to stimulated x-ray Raman spectroscopy.