Stimulated electronic x-ray Raman scattering at x-ray free-electron laser sources

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CFEL

X-ray free-electron lasers (XFELs) open the pathway to transfer non-linear spectroscopic techniques to the x-ray domain, to study the interplay of electronic and vibrational degrees of freedom by time-domain spectroscopy. A promising all x-ray pump probe technique is based on coherent electronic x-ray Raman scattering. The realization of these ideas is hampered by the extremely small inelastic scattering cross sections of x-rays with matter. A way to solve this problem is to stimulate the scattering process.

I will present the first experimental demonstration of nearly saturated stimulated electronic x-ray Raman scattering using the LCLS XFEL [1]. By tuning the relatively broad XFEL pulses to the core-excited Rydberg resonances in the pre K-edge region of neon, resonance scattered photons drive an avalanche of resonant inelastic x-ray scattering events. The scattering process is seeded by the spectral tail of the XFEL pulse, resulting in exponential amplification of the scattering signal with an enhancement of 6-7 orders of magnitude compared to the number of seed photons, similarly to the recently demonstrated photo-ionization pumped x-ray laser [2]. Analysis of the line profile of the emitted radiation permits to demonstrate the cross over from amplified fluorescence to coherent resonance scattering: In case of coherent scattering, the scattered x-ray radiation shows a pulse-to-pulse fluctuation of the line shape and the spectral peak position, resulting from a stochastic detuning of individual spectral spikes of the XFEL from resonance. In combination with statistical covariance mapping, a high-resolution spectrum of the resonant inelastic x-ray scattering process can be obtained, opening the path to coherent stimulated x-ray Raman spectroscopy. An extension of these ideas to molecules will be discussed [3].

[1] C. Weninger, M. Purvis, D. Ryan, R. A. London, J. D. Bozek, C. Bostedt, A. Graf, G. Brown, J. J. Rocca & N. Rohringer, submitted (2013)
[2] N. Rohringer, D. Ryan, R. A. London, M. Purvis, F. Albert, J. Dunn, J. D. Bozek, C. Bostedt, A. Graf, R. Hill, S. P. Hau-Riege, & J. J. Rocca , Nature, 481, 488 (2012).
[3] V. Kimberg & N. Rohringer, Phys. Rev. Lett. 110, 043901 (2013).