Stimulated X-ray emission using hard X-rays

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Current XFEL spectroscopy experiments are using primarily the linear interaction of matter with the X-ray pulses taking advantage of their very short duration. However, given the ultrahigh intensities that can be obtained when focusing these pulses, nonlinear X-ray matter interaction can also occur. Here, an exciting future perspective is the transfer of nonlinear spectroscopy from the optical to the X-ray spectral region, which is known for its sensitivity to the electronic structure of e.g. 3d transition metal ion complexes. In this study, the application and interpretation of stimulated emission using hard X-rays is discussed for a variation of Mn complexes.

A highly focused X-ray beam tuned to above the Mn 1s ionization energy creates a population inversion along its beam path, allowing for either amplified spontaneous emission as for K-alpha1, or amplified stimulated emission using a second seed color as for K-alpha2 or K-beta emission in the forward direction. The resulting spectra are resolved by a flat analyzer crystal and recorded by a 2d-detector.

We find spectra at amplification levels extending over four orders of magnitude until saturation, and observe bandwidths below the Mn 1s core-hole lifetime broadening in the onset of the stimulated emission. In the exponential amplification regime the measured spectral width is constant over three orders of magnitude, pointing to the build-up of transform-limited pulses of ~1 fs duration. Driving the amplification into saturation leads to broadening of the line. Importantly, the chemical sensitivity of the stimulated X-ray emission to the Mn oxidation state appears to be preserved. Differences in signal sensitivity and spectral information compared to conventional (spontaneous) X-ray emission spectroscopy are discussed.

In this presentation the experimental results will be discussed in light of theoretical simulations using simplified models that are able to reproduce the observed experimental findings.