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Non-linear effects in x-ray emission of liquid water with ultra-high fluence at LCLS

The oxygen K-edge x-ray emission (XE) spectrum of liquid water at ultra-high x-ray fluences was studied at the SXR Instrument of the x-ray free electron laser (FEL) LCLS. XE spectra were measured from a liquid micro-jet in vacuum for a wide range of x-ray fluences by varying x-ray spot size, flux and pulse duration. The threshold fluence for reproducing reference data measured at BESSY II could be determined to be approximately 0.1 J/cm^2 for 100 fs long LCLS x-ray pulses. For higher x-ray fluences, strong deviations from the reference spectrum were discovered. A non-linear behavior of the emission intensity with increasing incident x-ray fluence and a distortion of the spectral shape were observed in particular. These can be modeled by taking into account the reabsorption of x-ray emission by valence-excited states. We propose a mechanism by which x-ray emission photons from core-hole decay are reabsorbed in the sample by water molecules in valence-excited states formed via auger decay. This novel effect of valence-excited state absorption competes with other non-linear effects such as stimulated x-ray emission. Due to the much longer valence-hole lifetime compared to the core-hole lifetime it becomes important already for comparably low fluences.
The data and the model will be presented and implications as well as possible applications for future XE studies at FELs will be discussed. More than that, experimental parameters and geometries for minimizing valence-excited state absorption and maximizing stimulated x-ray emission will be presented.

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