Nonlinear X-Ray Absorption Observed Below Sample Damage Fluence

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We present X-ray-intensity-dependent X-ray absorption spectroscopy over the Co L_3 edge of a Co/Pd multilayer sample. Using moderate fluence, monochromatic pulses with 0.2 eV bandwidth, and 25 fs duration, we find significant deviations (~10% or greater) of the resonant absorption—even below the sample damage threshold of ~45 mJ/cm^2. The resonant absorption decreases at the peak of the resonance and increases at the low photon energy side of the resonance. Such changes are most naturally attributed to changes in the occupations of low energy states through the secondary electron cascade. Using the X-ray absorption sum rules, we find that these changes correspond to changes of more than a hundred holes per absorbed X-ray photon. When measuring X-ray absorption on the same sample, but now with higher intensity and wider bandwidth pulses (3 eV bandwidth pulses of ~5 fs duration and ~1 J/cm^2), similar features are detected. There is additionally, however, a decrease in absorption at photon energies well below the resonance energy. We attribute this additional feature to stimulated inelastic scattering. Our results set a surprisingly low fluence limit on when resonant experiments at XFELs can linearly probe sample properties. They also show how pulses from XFELs may be chosen or designed to preferentially observe certain nonlinear effects, such as stimulated inelastic scattering, over others.