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X-ray Emission Spectroscopy at X-ray Free Electron Lasers: Limits to Observation of Unperturbed Electronic Structures.

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Modern free electron lasers provide intense X-ray pulses with ˜1012 photons within ˜10-100 femtoseconds. Such pulses enable new experimental techniques and provide unique opportunities for investigation of electronic and nuclear dynamics on their intrinsic time-scales. Interaction of ultra-bright, ultra-short X-ray pulses with matter can induce a multitude of nonlinear excitation processes which must be carefully considered when planning spectroscopic measurements and interpreting data recorded at XFELs. In most cases correct interpretation of the spectroscopic response and analysis of the electronic structure hinges on the assumption of single photon excitations. Here we attempted to answer the fundamental question on the limits to probing the ground (or native) electronic structure of a 3d transition metal ion at XEFL sources. Ions of the 3d transition metal (Mn2+) in a lighter element (O, C, H) environment were used as a model system. X-ray emission spectroscopy recorded from Mn2+ at different pulse conditions demonstrate spectral changes as a function of increased pulse intensity and pulse duration. To explain these changes, we develop a rate equation based on sequential ionization and relaxation events forming multiply ionized states during a single pulse which agree with observed spectroscopic trends. The percentage of Mn Kβ emission recorded after the 1st, 2nd and 3rd 1s ionization events is calculated from the developed rate equation model and validated by experimental measurements. A method for estimating shifts in atomic X-ray emission lines from sequential ionization during a single XFEL pulse is given. From our data we infer that, in addition to multiple ionization, the impact of electron cascades is more significant for longer pulses. We note that while use of shorter X-ray pulses will help to counteract additional effects of electron cascades it will not help to overcome the spectral shifts due to sequential ionization. Presented data and associated analysis will help with experimental designs at current and upcoming XFELs where even higher intensities and shorter pulses are expected. 3d elements have a variety of important applications such as in bio-inorganic catalysis, chemical catalysis and energy storage / conversion making robust protocols for their XFEL analysis of general importance.

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