Extended Fluorescence Emission Time from Clusters in Intense X-ray Free-electron Laser Pulses

PHAY HO, CHRIS KNIGHT, LINDA YOUNG, Argonne National Laboratory — Motivated by the recent work by Classen and coworkers [Phys. Rev. Lett. 119, 053401 (2017)] of exploiting the Hanbury Brown and Twiss effect for molecular imaging with x-ray fluorescence, we theoretically examined the fluorescence spectrum of a nanocluster from XFEL pulses with our MC/MD method. Using Ar clusters as a prototype, we focused on fluorescence processes in intense x-ray fields and found that non-linear x-ray absorption leads to a high-degree of ionization and creates a dense electron environment within the sample on the femtosecond timescale. These ultrafast processes produce x-ray emission profiles in an extended sample that are very different from the atomic profile. In addition to the direct photoionization pathways, electron-ion recombination processes provide additional pathways in clusters to reach the same fluorescence channels and give rise to higher yields in K\(_{\alpha}\) and its hypersatellite from double-core-hole state (K\(_{\alpha}^H\)). The presence of the recombination pathway leads to extended fluorescence emission time beyond the lifetime of the core-excited states. We show that the K\(_{\alpha}^H\) emission line can be a good candidate for fluorescence imaging as it has relatively short emission time compared to the x-ray induced distortion time.

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