Coherence and Structural Information in Intense X-Ray–Driven Systems

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Coherent X-ray imaging with intense X-ray free-electron laser (XFEL) pulses operates in a regime where ultrafast electronic dynamics, rather than static structure, can govern the coherence and information content of measured signals. Rapid ionization and electronic relaxation during the pulse create transient nonequilibrium states [1] that modify both scattering and emission processes. Understanding how these dynamics influence coherence is therefore essential for interpreting XFEL imaging experiments. In this presentation, I discuss two complementary manifestations of coherence in strong-field XFEL interactions. First, transient electronic resonances that emerge during ultrafast ionization dynamics can strongly modify resonant X-ray scattering pathways [2–5]. These transient states enable interference between elastic scattering and resonance fluorescence channels, revealing how electronic dynamics shape the coherence properties of scattered X-rays.

Second, I examine fluorescence intensity correlation [6], sometimes referred to as incoherent diffractive imaging [7], which extracts structural information from second-order correlations of X-ray fluorescence, closely related to Hanbury Brown–Twiss intensity interferometry [8]. Even when conventional scattering signals are degraded by radiation-induced ionization and the presence of delocalized electrons, fluorescence correlations can encode structural and elemental information and may provide an alternate route to imaging under intense XFEL conditions.

These results highlight how ultrafast electronic dynamics shape both scattering and fluorescence coherence in XFEL experiments and suggest new opportunities for exploiting emission-based correlations alongside conventional scattering for imaging structures and dynamics of nanoscale systems.

References

[1] E P. Kanter, B. Krässig, Y Li, *et al.*, Phys. Rev. Lett. **107**, 233001 (2011)

[2] P J Ho, B J Daurer, M F Hantke, *et al.*, Nat. Commun. **11**, 167 (2020)

[3] S Kuschel, P J Ho, A Al Haddad, *et al.*, Nat. Commun. **16**, 847 (2025)

[4] A Venkatesh and P J Ho, Phys. Rev. A **111**, L021101 (2025)

[5] A Venkatesh and P J Ho, arXiv:2506.06585 (2025)

[6] P J Ho, C Knight, L Young, *et al.*, Struct. Dyn. **8**, 044101 (2021)

[7] A Classen, K Ayyer, H N Chapman, *et al.*, Phys. Rev. Lett. **119**, 053401 (2017)

[8] R Hanbury Brown and R Q Twiss, Nature **177**, 27 (1956)