Spectator-Nuclei Reveal Nonlinear Light–Matter Interaction Driven by an X-Ray Free-Electron Laser

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Time-resolved nuclear forward scattering (NFS) from Mössbauer nuclei has long served as a textbook example of coherent, linear X-ray optics: at synchrotron sources, delayed quantum beats are faithfully described by low-excitation nuclear resonant propagation theory. In this talk, I will show that this paradigm breaks down when the 14.4 keV resonance of ^{57}Fe is driven by a hard X-ray free-electron laser.

Using the European XFEL, we observe several anomalies that have no counterpart in conventional NFS, including a loss of sensitivity to sample thickness and a surprising non-commutativity between sample and attenuator. A systematic survey shows that all observables collapse onto a single control parameter — the effective energy load deposited in the sample — with a clear onset threshold and approximately logarithmic growth above it.

Because the nuclei themselves remain far from saturation, we interpret this regime as spectator-nucleus nonlinear optics: XFEL-driven electronic, structural, and magnetic dynamics in the host induce transient disorder that dephases and redistributes the coherent nuclear propagation. Mössbauer nuclei thus emerge as ultra-sensitive interferometers for nonlinear X-ray–matter dynamics at XFEL intensities.