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Probing Ultrafast Domain Fluctuations of the Order Parameter with Coherent X-Rays

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Nanoscale heterogeneities play an integral role in quantum materials where emergent phenomena can arise from cooperating and competing order parameters in the system. Coherent x-rays are particularly proficient in accessing these structural and electronic heterogeneities. For example, X-ray Photon Correlation Spectroscopy (XPCS) can directly measure the dynamic timescales of domain interactions in equilibrium by following how a scattered speckle pattern evolves in time [1]. However, the physical processes of nonequilibrium phenomena occur on ultrafast timescales, i.e., on the order or nanoseconds or less, and cannot be captured by standard techniques. Here, we developed a novel technique that employs a pump-probe-probe scheme to follow fluctuations of the order parameter that will allow us to independently resolve how various nanoscale domains evolve on ultrafast timescales. A pilot experiment performed at the LCLS X-ray Free Electron Laser to study how the charge, orbital and structural domains evolve in magnetite (Fe_3O_4) validates the ability of this experimental setup in measuring domain dynamics on picosecond timescales.

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