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Ultrafast ptychographic imaging of order-disorder correlations in hybrid nanoparticle topologies

We investigate functional properties and collective responses in self-assembled ligand-coated nanoparticle (NP) supracrystals triggered by external light excitation [1]. Focusing on determining how changes in core-ligand, core-core, and ligand-ligand interactions influence the mesoscale photoinduced responses, our study aims to understand the role of heterogeneity and order/disorder correlations out-of-equilibrium [2, 3]. Imaging these systems is challenging, because of the coexistence of elements with different atomic numbers, across multiple length and time scales, each with different cross-sections and radiation damage thresholds [4, 5]. To address these challenges, we developed an ultrafast tabletop microscope relying on EUV Ptychographic Coherent Diffractive Imaging (CDI) [6] from High-Harmonic Generation (HHG). In ptychographic CDI, a coherent illumination function is scanned across the sample in many adjacent and overlapping positions to obtain full-field images with high spatial resolution and sensitivity to both quantitative material composition (amplitude) and height (phase) contrast [6, 7]. In the ultrafast regime, the obtained quantitative material and dynamic structural information are provided with high spatiotemporal resolutions [8]. We demonstrate that it is possible to visualize, simultaneously, ligand shells, core morphology, and their overall 2D superlattice distribution. Moreover, we show that applying unsupervised deep learning in ptychographic imaging speeds up algorithm convergence and increases the fidelity of the reconstruction. We do so by comparing randomly initialized ("Cold Start") or using an approximate probe guess ("Warm Start") approaches. As a practical example, we provide the study of the out-of-equilibrium response of ligand-coated CsPbBr₃ NP supracrystals, where we address the problem of core-size dependence and supracrystalline heterogeneity.

References

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