Metrological Optimization and Simulation of Spin Resonance Spectroscopy in Transmission Electron Microscopy

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Recent advances in Transmission Electron Microscopy (TEM), including aberration correction, cryogenic stabilization, and ultrafast electron pulses, have pushed spatial and temporal resolutions to unprecedented levels. However, extending this precision to the detection of single spins and their magnetic dynamics remains a significant challenge. We present a comprehensive theoretical and computational framework for Spin Resonance Spectroscopy (SRS) in TEM, such as nuclear (NMR) or electron spin resonance (ESR), enabling state-selective spin imaging at the nanoscale. By capturing both elastic and inelastic scattering processes between free-space electron probes and microwave-driven spin systems, we describe the resulting magnetic phase shifts and deflection angles imprinted on the electron wave function. We demonstrate through simulations that these phase shifts from individual spins are detectable in both image and diffraction modes.

To determine the precision limits of this technique, we quantify measurement sensitivity using Classical Fisher Information (CFI) and compare it against the Quantum Fisher Information (QFI) and the Helstrom bound. Our findings identify optimal imaging conditions, specifically the effects of defocus and beam width, to maximize the signal-to-noise ratio. We find that while standard diffraction and imaging measurements can approach quantum-limited performance for classical dipoles, orbital angular momentum (OAM)-resolved detection is required to restore sensitivity when electron backaction from a quantum spin becomes significant. This work lays the foundation for spin resonance spectroscopy with the atomic resolution of TEM, as well as for future experiments in quantum spin research and nanoscale material characterization.

References

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