Spin Relaxation and Coherence in Low-Dimensional Lead Halide Perovskites and Nanocrystals

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The ability to control carrier spin and spin coherence through materials design offers exciting opportunities for spin-optoelectronic devices, quantum information technologies, and spin-selective photochemical processes. Lead halide perovskites have emerged as a versatile platform for exploring spin-dependent phenomena owing to their strong spin–orbit coupling, highly tunable electronic structures, and rich structural diversity spanning two-dimensional (2D) quantum wells and strongly confined nanocrystals. Despite rapid progress in understanding their optical and charge-transport properties, the fundamental mechanisms governing spin relaxation and coherence remain incompletely understood, limiting the rational development of perovskite-based spin technologies.

In this presentation, we highlight our recent efforts to elucidate and engineer spin dynamics in low-dimensional lead halide perovskites through the combined use of ultrafast nonlinear optical spectroscopy and rational chemical design. In 2D perovskite heterostructures, we show that interfacial charge transfer can suppress exciton-mediated spin relaxation by reducing electron–hole wavefunction overlap, leading to substantially prolonged spin lifetimes. In strongly quantum confined perovskite nanocrystals, we demonstrate that tailoring ligand–nanocrystal interactions through chiral charge-transfer interfaces can dramatically extend spin coherence times, while transition-metal doping enables the generation of coherent spin precession at room temperature without external magnetic fields.

Together, these studies reveal how quantum confinement, interfacial electronic coupling, chirality, and magnetic interactions can be leveraged to control spin relaxation and coherence across low-dimensional perovskite systems. By establishing general design principles that connect chemical structure with spin dynamics, this work provides a foundation for the development of next-generation spin-chiral optoelectronic and quantum materials.