Exciton spin dynamics in lead-halide perovskites crystals and nanocrystals

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Variety of lead halide perovskites serve as a versatile platform for exploring spin-related phenomena and the influence of crystal symmetry and the spatial confinement on spin dynamics [1].

In this study, we examine exciton spin polarization at cryogenic temperatures in bulk perovskite crystals with about cubic ($FA_{0.9}Cs_{0.1}PbI_{2.8}Br_{0.2}$ and $FAPbBr_3$) and orthorhombic ($MAPbI_3$ and $CsPbBr_3$) symmetries. A remarkably high optical orientation of 85% is observed demonstrating strong spin selectivity [2,3]. The optical orientation remains stable with excitation energy detuning up to 0.3 eV from the exciton resonance and declines beyond 1 eV. The absence of spin relaxation acceleration in both structural phases suggests the suppression of the Dyakonov-Perel mechanism and the absence of the Dresselhaus-Rashba splittings, indicating preserved spatial inversion symmetry [3]. Coherent spin quantum beats, and electron-hole spin correlations provide further confirmation of these effects. Additional symmetry reduction due to spatial confinement in the $CsPbI_3$ perovskite nanocrystals does not activate spin relaxation via mechanisms associated with the loss of inversion symmetry. On the other hand, the optical orientation of exciton spins is governed by exchange interaction effects within the exciton.

The highly polarized exciton emission and extended spin relaxation times [2-4] in lead halide perovskites make them promising candidates for quantum information and spintronic technologies, enabling efficient optical spin manipulation and long-lived spin coherence.

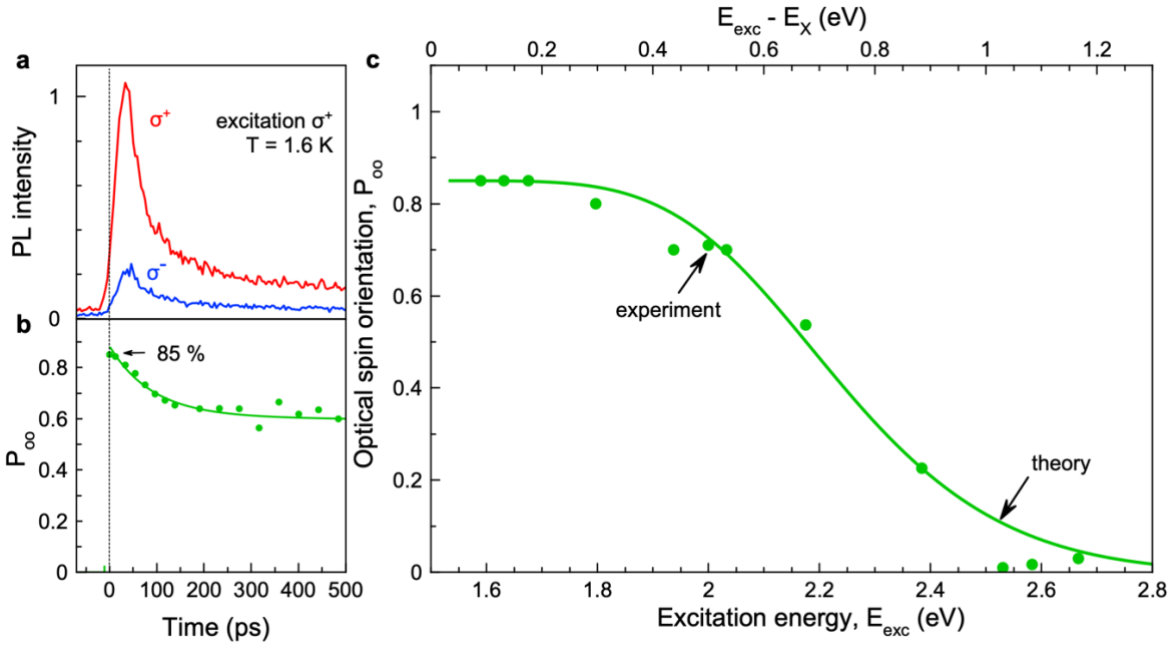

Figure 1: (a) Photoluminescence dynamics recorded in σ^+ (red line) and σ^- (blue line) polarization under σ^+ excitation at T = 1.6 K in $FA_{0.9}Cs_{0.1}PbI_{2.8}Br_{0.2}$ crystal. (b) Time evolution of the optical orientation degree $P_{oo}(t)$. The solid line represents an exponential fit. (c) Dependence of the initial optical orientation $P_{oo}(0)$ on the excitation energy (symbols). The upper axis indicates the detuning relative to the exciton resonance E_{exc} - E_X. The green line shows the theoretical curve accounting for Elliott-Yafet spin relaxation due to interaction with longitudinal optical.

References

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