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Charge Carrier and Spin Diffusion in a Polycrystalline and Single Crystal Lead Halide Perovskite Semiconductor

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Light-induced transient gratings (LITG) spectroscopy is a powerful technique for probing carrier transport dynamics in semiconductors. Despite its complexity, the non-destructive and contactless nature, along with simultaneous multi-parameter acquisition renders the technique an attractive alternative to others, where magnetic fields, high spatial resolution, or cryogenic conditions may be needed. In this work, we employ the user-friendly HARPIA-TG spectrometer (*Light Conversion*) to investigate both carrier and spin transport in the halide perovskite semiconductor $FA_{0.95}Cs_{0.05}PbI_3$ with varying degrees of crystallinity. Population gratings generated with parallel-polarized excitation pulses probe carrier diffusion and recombination, while cross-polarized excitation produces spin gratings that enable direct observation of spin transport and relaxation dynamics.

Carrier dynamics measurements yield ambipolar diffusion coefficients on the order of 1 cm^2/s (Fig. 1(a)) and diffusion lengths approaching 1.5 µm in single crystals, with strong attenuation in polycrystalline films due to grain boundary barriers (Fig. 1(b)). In contrast, spin grating measurements reveal diffusion coefficients exceeding 100 cm^2/s, indicating spin transport occurring on timescales nearly two orders of magnitude faster than population diffusion at room temperature. The substantial enhancement in spin transport suggests a mechanism involving exchange-mediated interactions and doping-assisted effects that facilitate ultrafast spin propagation prior to picosecond-order relaxation. These results demonstrate the capability of a single experimental setup to probe both carrier and spin dynamics, offering new insight into spin motion in metal–halide perovskites and highlighting their potential for spintronic applications.

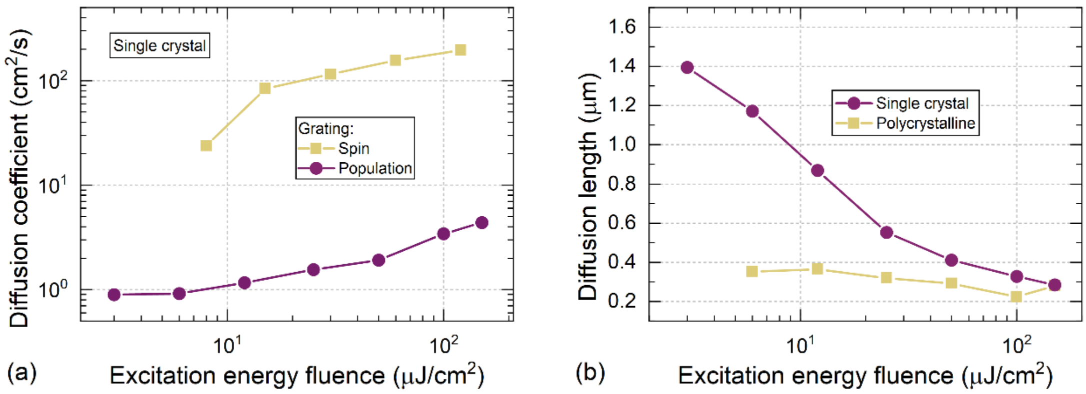

Fig. 1 Diffusion coefficient in a single crystal acquired by probing spin and population gratings (a). Carrier diffusion length in a single crystal and a polycrystalline film (b).