Elementary excitation in Hybrid Lead-Halide Perovskites

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Hybrid perovskites represent a new, disruptive, technology in the field of optoelectronics. They have the potential to overcome the performance limits of current technologies and achieving low cost and high integrability. Hybrid halide perovskite, e.g. CH3NH3PbX3 [X = Cl, Br, or I], are usually deposited as polycrystalline thin-films with variable mesoscale morphology depending on the growth conditions. The obtained grain size ranges from tens to thousands of nm. Over the last two years the impressive improvement of photovoltaic performance has been driven by radical empirical evolution of the device architecture and processing methodologies. However, there is a considerable lack of understanding of material properties, both as pristine films and their embodiment in a device. Here we demonstrate that the electron-hole interaction is sensitive to the microstructure of the material. We find that by control of the material processing during fabrication both free carrier and Wannier excitonic regimes are accessible, with strong implications for optoelectronic devices. The long-range order of the organic cation dipole field is disrupted by polycrystalline disorder introducing domain walls where dipole twinning breaks down. The variations in electrostatic potential found for smaller crystallites suppress exciton formation, while larger crystals of the same composition demonstrate an unambiguous excitonic state. In addition, we demonstrate that it is also possible to design the emissive properties for a single material composition by designing the processing routs. By simply tuning the average crystallite dimension in the film from tens of nanometers to a few micrometers, it is possible to tune the optical band gap of the material along with its photoluminescence lifetime. We demonstrate that larger crystallites present smaller bandgap and longer lifetime which correlates to a smaller rediative bimolecular recombination coefficient. We also show that they present a higher optical gain, becoming preferred candidates for the realization of lasing devices.