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Local Strain Heterogeneity Influences Non-Radiative Recombination in Perovskite Films

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Photovoltaics based on organometal halide perovskite semiconductors have obtained remarkable power conversion efficiencies (PCE).[1] In just 6 years of research, perovskites have become the most efficient thin-film technology, with a current record PCE of 24.2%. Despite this outstanding progress so far, substantial scope exists for further improvements. Early progress in device PCE largely leveraged the development of new deposition chemistries for smoother, more uniform films. As such, fundamental understanding of loss mechanisms has arguably trailed progress in PCE. Further PCE improvements will be harder won, and built upon successful identification of limiting loss mechanisms and their subsequent minimisation through realisation of successful crystal growth strategies.

Despite being an inherently low-loss system, a substantial grain-scale heterogeneity in radiative efficiency still exists in state-of-the-art polycrystalline films.[2] Amelioration of the low-quality grains is required to raise the open-circuit voltage to its thermodynamic limit. However, the origin of this spatial distribution of these losses is not yet understood. Chemical surface passivation improves overall photoluminescence quantum efficiency; yet heterogeneity remains,[2] suggesting a prominent loss mechanism within the bulk material itself.

We perform correlative confocal time-resolved photoluminescence mapping and synchrotron-based scanning X-ray microdiffraction to uncover the structural origin of non-radiative pathways. We observe a consistent statistical anticorrelation between carrier lifetime and compressive lattice stain of the perovskite films[3] (see Figure 1). That is, strained regions possess shorter photoluminescence lifetimes. Density functional theory (DFT) predicts an increased formation of halide vacancies under higher compressive strain, which are known to form shallow trap states which facilitate non-radiative decay. Furthermore, our detailed structural mapping and characterisation reveal surprising new layers of heterogeneity in perovskite films. New features at the scale of many local grains (20–100 μm^2), as well as sub-grain features are presented. By identifying strain as a cause of non-radiative recombination, our work guides researchers to design fabrication pathways that minimise strain and provide opportunities to further reduce losses in perovskite devices.

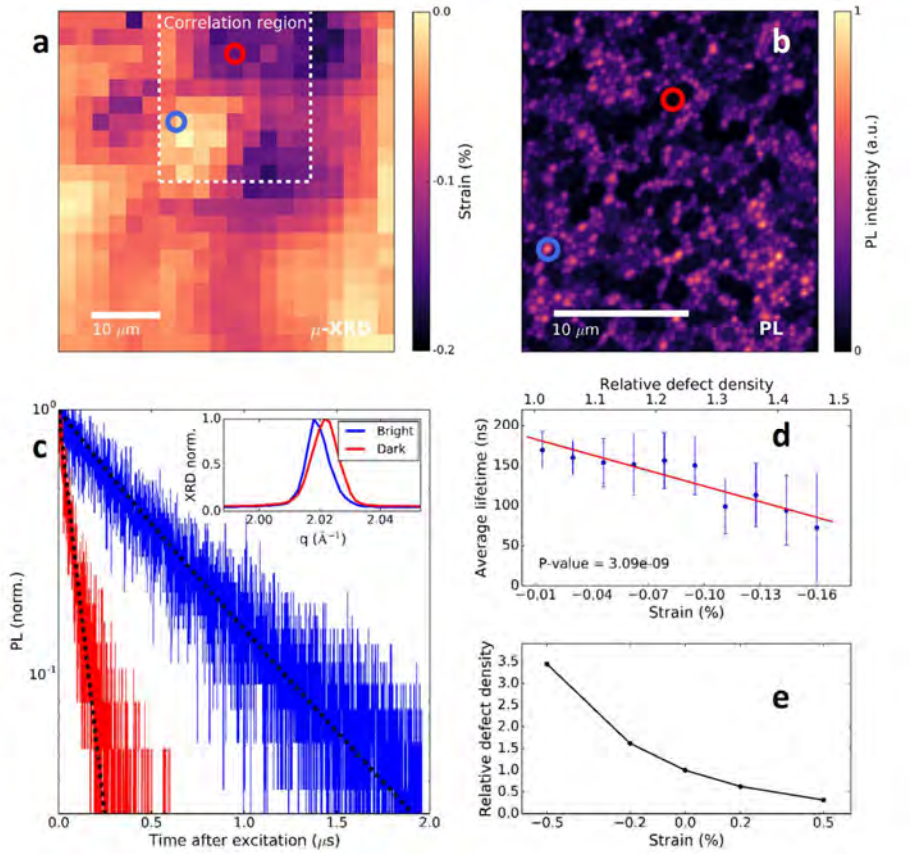

Figure 1. Correlation of local structural properties to local photoluminescence lifetime of a $CH_3NH_3PbI_3$ perovskite film. (a) Strain map from scanning X-ray microdiffraction indicating correlation region to confocal TRPL. (b) Corresponding PL intensity map. (c) Local region correlations between bright grains (blue markers) and dim grains (red markers); and inset: corresponding (220) diffraction peaks. (d) statistical correlation of local PL lifetime and local strain across the correlation region. (e) Relative iodide vacancy defect levels as a function strain level obtained via DFT calculation. Figure taken from reference [3].

References

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