The varied nature and roles of nanoscale surface defects in perovskite thin films

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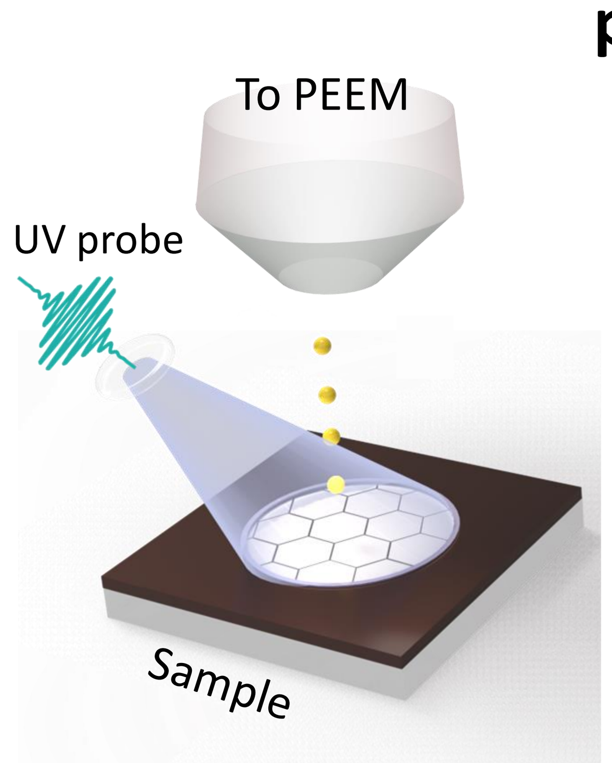
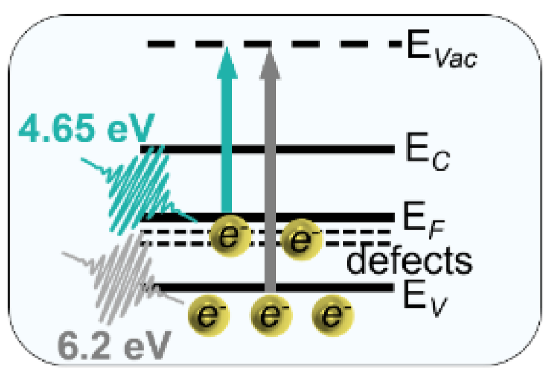
Abstract: we identified multiple types of nanoscale defects in hybrid perovskite thin films and found that they play different roles in charge trapping: from highly detrimental to relatively benign.

1. What are hybrid halide perovskites?

- Semiconducting materials that are used to fabricate state-of-the-art solar cells.
- Efficiency of perovskite solar cells reached 25.7 % in single junction configuration, which is comparable to efficiencies of single crystal silicon solar cells (26.1%) **[1]**.
- Performance of perovskite thin films is impacted by the presence of defects **[2]**.
- Defects in perovskites have been reported to induce non-radiative losses that occur on microscale **[3]**.

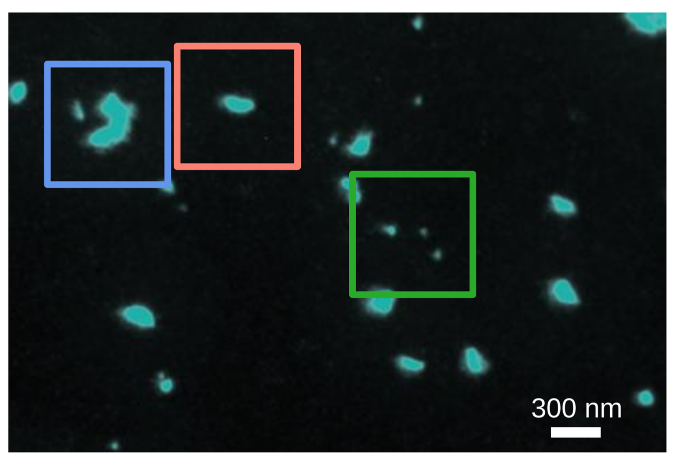
2. Imaging defects and grains in hybrid perovskites



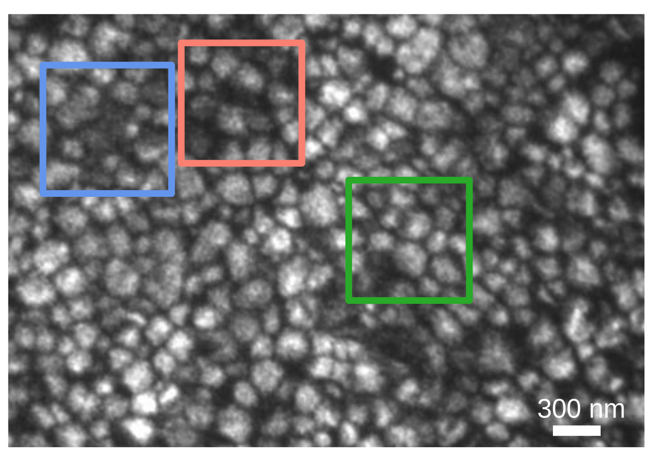


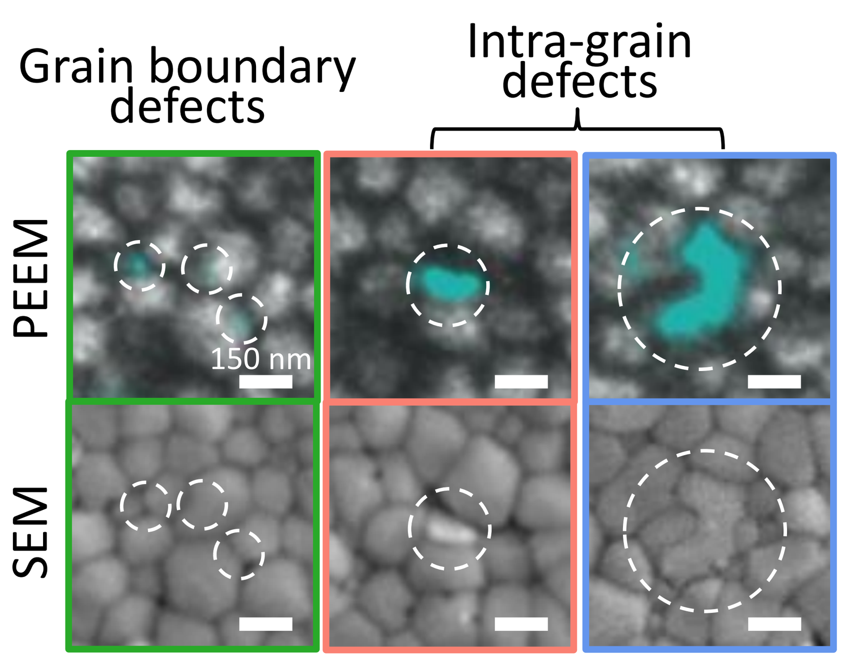
- With 4.65 eV probe photons we imaged occupied mid gap (defect) states in perovskite thin films **[4]**.
- With 6.2 eV probe photons, we imaged surface morphology of polycrystalline perovskite films.

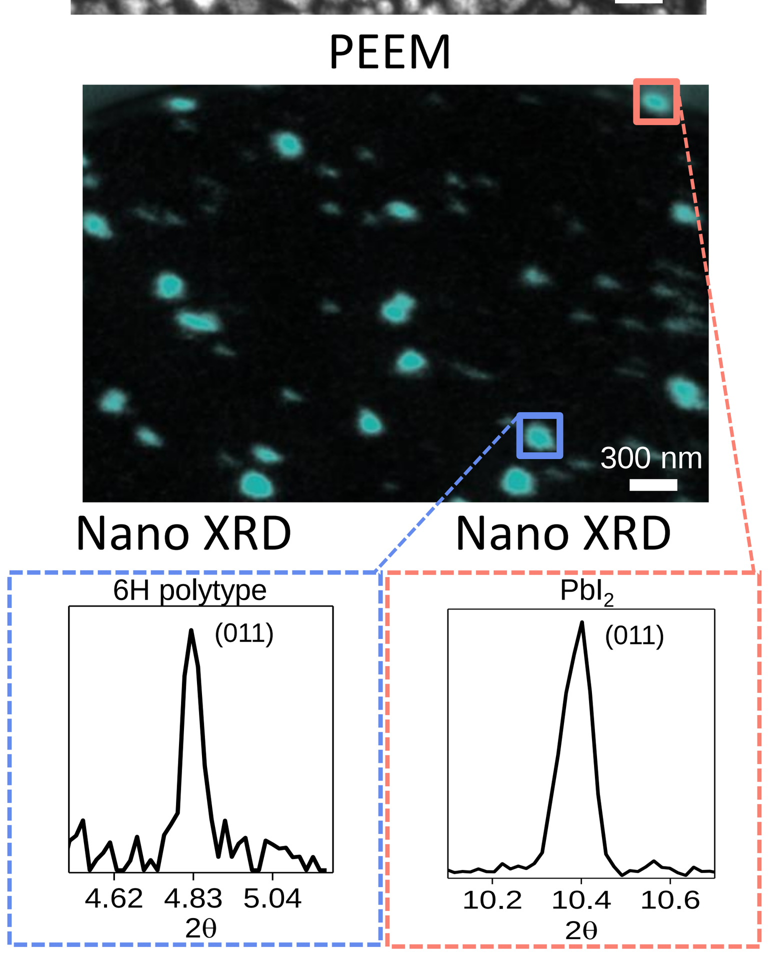
PEEM image of nanoscale defects



PEEM image of surface morphology

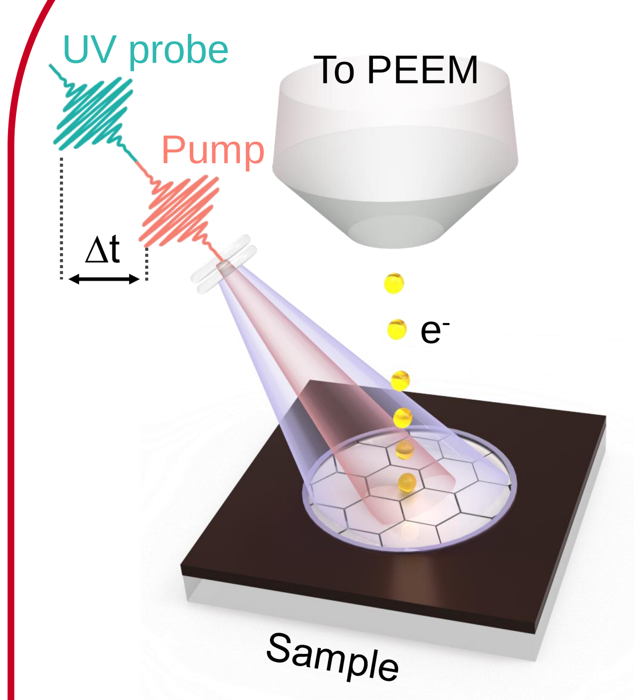


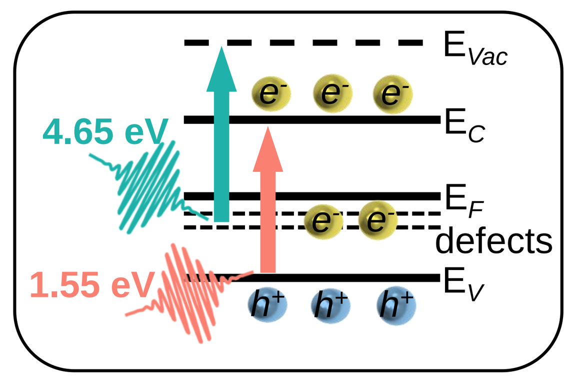




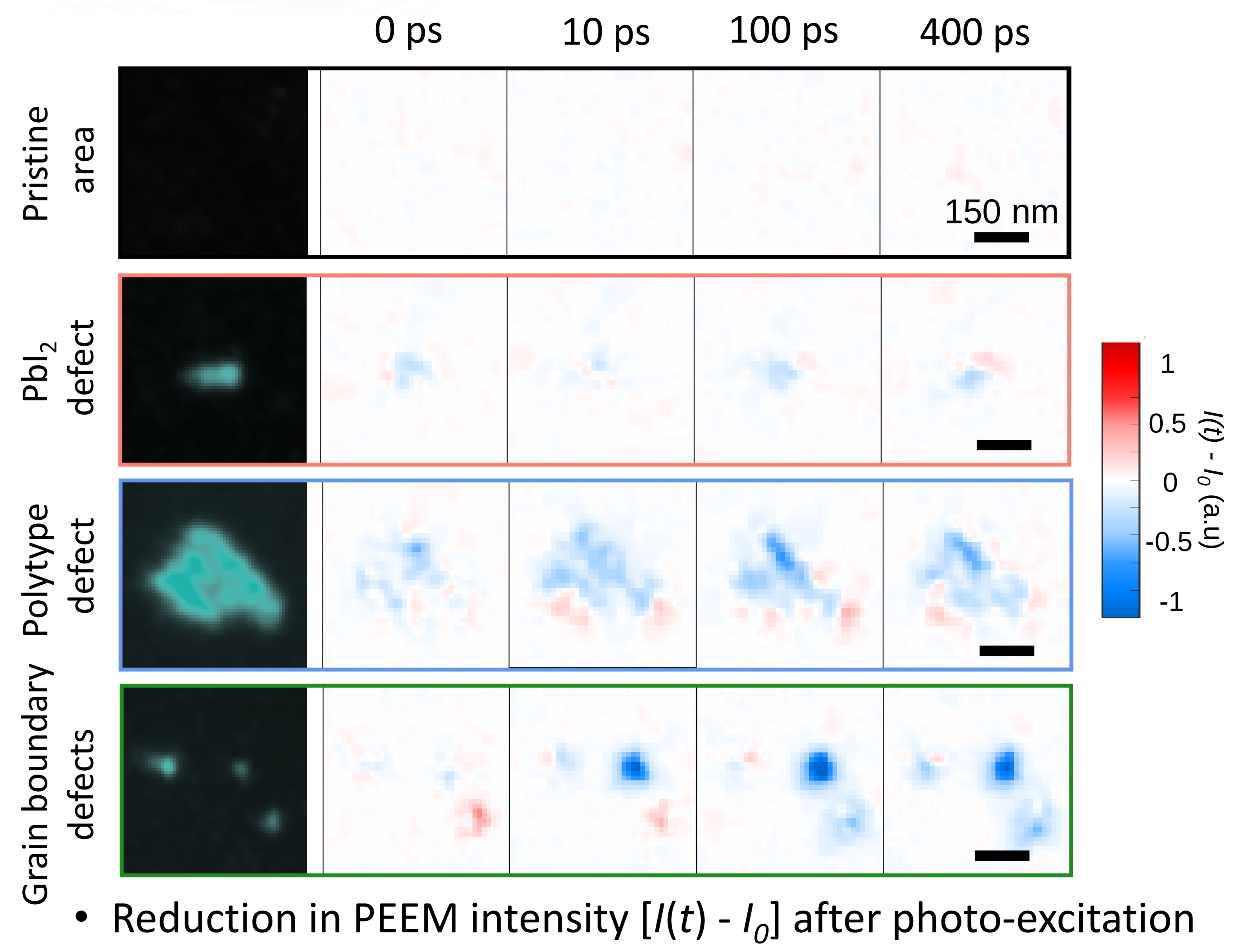
- Nanoscale defects in perovskite into were categorized into grain boundary and intra-grain defects.
- Intra-grain defects were associated with PbI_2 and 6H hexagonal polytype perovskite phases **[5]**.

3. Trapping dynamics of different defect types





- With pump photons, we photo-excite electrons from valence band to conduction band of perovskite.
- With UV probe photons, we monitor the response of defects to photo-excitation.



- Reduction in PEEM intensity [$I(t)$ - I_0] after photo-excitation indicates reduction of population of defect states, or trapping of photo-excited holes.
- Defects associated with PbI_2 phases, were not observed to participate in trapping of charges from perovskite.
- Defects associated with polytype phase and grain boundary defects, showed hole trapping signal. This hole trapping results in non-radiative recombination and reduces performance of perovskites **[5]**.

4. Conclusion

- $Cs_{0.05}MA_{0.17}FA_{0.78}(I_{0.83}Br_{0.17})_3$ perovskite thin films contain multiple types of nanoscale surface defects.
- Intra-grain defects are associated with lead iodide and hexagonal polytype phase impurities.
- Defects associated with hexagonal polytype phase and grain boundary defects, participate in trapping of photo-excited holes.
- Defects related to PbI_2 were not observed to trap charges when photo-exciting perovskite.
- Defective phase impurities should be eliminated to improve performance of perovskite thin films.

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References:
[1] NREL, Best Research-Cell Efficiency Chart (2022), https://www.nrel.gov/pv/cell-efficiency.html. **[2]** Ball J. M. & Petrozza A. *Nat. Energy* 1, 16149 (2016). **[3]** deQuilettes D. W. *et al. Nat Commun*, 7, 11683 (2016). **[4]** Doherty T. A. S., Winchester A. J. *et al. Nature* 580, 360 (2020). **[5]** Kosar S. *et al. Energy Environ Sci*., 14, 6320 (2021).