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Reorientation-Driven Degradation in Oriented Perovskite Films: Shifting Facet Engineering to Thermodynamic Stability

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Hybrid perovskite solar cells (PSCs) suffer from underexplored links between crystallographic orientation and thermal stability, especially in narrow-bandgap devices. We fabricate highly oriented mixed Sn-Pb perovskite films via an additive-free two-step method. Accelerated ageing studies at 120 °C reveal that high orientation paradoxically compromises stability, and PSCs built from highly oriented perovskite films retain only 73% of their initial PCE versus 89% in less-oriented devices. Operando grazing incidence wide-angle X-ray scattering of the PSCs shows that thermal stress induces significant reorientation and lattice distortion in the oriented crystallites, accumulating pronounced microstrain that accelerates the PSC degradation. Structural analyses confirm progressive crystallographic transitions, including grain reconfiguration, shifts toward isotropy, and systematic diffraction migrations. Critically, we demonstrate that metastability is an intrinsic consequence of high crystallographic order, which is why the very high alignment strategies that enhance performance induce thermodynamic vulnerability. This necessitates redesigning crystal engineering priorities where suppressing instability requires engineering thermodynamic equilibrium states over maximising alignment for stable perovskite photovoltaics.

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