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Interfacial dipole engineering in all-inorganic perovskite solar cells†

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Severe nonradiative recombination and energy level mismatch in perovskite solar cells (PSCs) are key factors affecting efficiency. Here, we report an effective strategy for surface passivation and interfacial dipole engineering of perovskite films. By precisely introducing electron-withdrawing and electron-donating groups on 7-azaindole, we have effectively controlled the passivation ability of N atoms and the polarity of the interfacial dipole, thereby regulating the perovskite surface's work function and obtaining the optimal energy level matching. This strategy yields an impressive efficiency of 10.76% for the CsPbBr₃ PSC and exceptional stability.

Organic–inorganic halide perovskite solar cells (PSCs) have garnered significant attention, achieving a record-breaking power conversion efficiency (PCE) of 26.1%.^{1–3} However, their stability falls below commercial requirements when exposed to moisture, heat, and light, primarily due to the degradation of organic groups like MA⁺ and FA⁺.⁴ To overcome these challenges, all-inorganic CsPbX₃ (X[−] = I[−], Br[−], Cl[−], or mixed) perovskites have emerged as a promising solution.^{5,6} While iodide-containing perovskites offer more suitable bandgaps, their black-phased structure easily transforms into non-perovskite yellow phases, hampering progress in iodide-containing all-inorganic PSC research.⁷ On the other hand, iodide-free CsPbBr₃ perovskite exhibits excellent weatherability and a wide bandgap of 2.3 eV, making it highly promising for next-generation photovoltaic technologies, tandem solar cells, and high-voltage electronics.⁸ However, CsPbBr₃ PSCs have achieved a reported PCE of 11.21%, which is still significantly below its theoretical efficiency limit.⁹

Solution-processed perovskite films are prone to generating defects at surfaces and grain boundaries, which act as nonradiative

recombination centers, leading to decreased open circuit voltage (V_{oc}) and fill factor (FF) in CsPbBr₃-based PSCs. This leads to undesired energy loss.^{10,11} Furthermore, proper energy level alignment at the interface is crucial for minimizing energy losses.¹² Therefore, an effective strategy to mitigate energy loss and enhance device PCE involves repairing surface defects and promoting energy level alignment.

Surface defects are addressed using Lewis base molecules and polymers. These passivators, containing N, O, or S atoms, reduce uncoordinated Pb²⁺ ions and defects at the film surface and grain boundaries.^{13,14} Recent investigations have revealed that the introduction of electron-donating methoxyl (−OCH₃) and electron-withdrawing nitryl (−NO₂) on the benzene ring can effectively regulate the passivation ability of molecules.^{11,15} Directly regulating the electron density of passivation sites, independent of the benzene ring, is a critical focus for future research. While passivators effectively reduce defects, limited charge extraction efficiency at interfaces remains a barrier to enhancing device performance.

Asymmetric molecules possess a dipole moment created by positive and negative charge centers. This distribution of charges alters the work functions (WFs) and strongly influences charge carrier transport at the interface.¹⁶ For instance, Zhang *et al.* introduced donor– π –acceptor (D– π –A) dipoles at the perovskite/electron transport layer (ETL) interface, enhancing the built-in electric field and lowering the WF.¹⁷ Wu *et al.* incorporated two new naphthalimide-based molecules (PX, X = F, I) into the perovskite layer, creating an interfacial electrical dipole layer that enhances hole extraction and reduces the WF.¹⁸ Because the direction and intensity of surface dipoles depend on specific molecule-to-surface interactions, which cause an increase and decrease of the WF, precisely controlled dipoles will determine the energy level matching, which can help influence carrier-oriented extraction transfer kinetics.

In this work, we developed a strategy for surface passivation and cooperative construction of interfacial dipoles. By investigating three different 7-azaindole (7-AI) derivatives, we

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