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Defects in perovskite crystals

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Perovskite solar cell (PSC) is now a shining star in photovoltaics field^[1]. Benefiting from excellent optoelectronic properties of perovskite materials, the certified power conversion efficiency (PCE) of PSCs has reached 26.0%^[2], showing great potential for commercialization. In essence, the efficiency of solar cells is determined by the radiative and nonradiative recombination of photogenerated charge carriers. The unfavorable nonradiative recombination mainly assisted by the trap states leads to severe charge carrier loss and thus unsatisfactory efficiency.

Organic-inorganic hybrid perovskite materials are ionic crystals with typical ABX_3 perovskite structure^[3], where A is monovalent cations like CH_3NH_3^+ (MA^+), $\text{NH}_2\text{CHNH}_2^+$ (FA^+) and Cs^+ , B is divalent cations (Pb^{2+} or Sn^{2+}), and X is halogen anions (I^- , Br^- or Cl^-) (Fig. 1(a)). Owing to the diversity of compositions, various defects like vacancies, interstitial ions, and anti-sites can be generated in the solution-processed polycrystalline films. These defects mainly exist at grain boundaries and interfaces, which significantly influence the efficiency and stability of PSCs. Therefore, most works have focused on passivating defects to improve the device performance.

Considering the structural feature and ionic nature of perovskite materials, most trap states are point defects and inevitably charged. The positively charged defects include uncoordinated Pb^{2+} ions, halogen vacancies, interstitial cations, A_X , B_X and B_A anti-sites, while negatively charged defects are uncoordinated I^- ions, cation vacancies, interstitial halogen ions, X_A , X_B and A_B anti-sites (Fig. 1(b))^[4]. For anti-site defects, A_X means A cation occupies X site. In addition to charged defects, metal Pb clusters can also form in the perovskite films due to the reduction of uncoordinated Pb^{2+} . The generation of a defect is directly determined by its formation energy. Once a defect forms, it will introduce additional transition energy level (Fig. 1(c))^[5]. The transition levels near the conduction band minimum (CBM) or valence band maximum (VBM) edge are called shallow-level trap states, like vacancies and interstitial ions, whose formation energy is relatively low. The trapped carriers in shallow-level defects have high de-trapping probability to fall back into the CBM or VBM, thereby contributing little to nonradiative recombination. But defect migration can lead to undesirable local band bending, phase segregation, and current–voltage (J – V) hysteresis, as well as degra-

tion of perovskite films due to interfacial reactions^[6]. Transition energy levels located within the middle third of forbidden band are identified as deep-level trap states, like uncoordinated ions, anti-sites and Pb clusters, which have higher formation energy. Different from shallow-level trap state, once a deep-level trap state is formed, it will serve as charge recombination centre and induce nonradiative recombination, thus decreasing the PCE. In order to diminish negative impact of various defects, the passivators should have following functions: (1) providing chemical bonding to deactivate the defects; (2) forming wide-bandgap phases or type-I heterojunction at grain boundaries and interfaces to suppress nonradiative recombination; (3) optimizing crystal orientation to reduce structural defects.

Lewis acid/base coordination chemistry is widely employed to passivate uncoordinated defects. Lewis acid and Lewis base can accept and donate electrons, which means that they can passivate electron-rich and hole-rich defects via forming Lewis adducts, respectively. Fullerene (C_{60}) and its derivatives PCBM are the most popular Lewis acids due to their excellent electron-accepting ability, making them powerful to passivate Lewis base type defects. Combining theoretical calculation and experimental characterizations, Xu *et al.* revealed that PCBM can effectively passivate I_{Pb} antisite and uncoordinated I^- to suppress nonradiative recombination and promote electron extraction (Fig. 2(a))^[7]. In addition, a PCBM passivation layer can also inhibit anion migration that may result in J – V hysteresis^[8]. Compared with Lewis acid, Lewis base type passivators are more widely employed. Most Lewis bases base on nitrogen, oxygen, and sulphur atoms containing lone pair of electrons. Noel *et al.* first reported that organic molecules like thiophene and pyridine can serve as Lewis base to passivate uncoordinated Pb^{2+} (Fig. 2(b))^[9]. Thiophene and pyridine can donate electrons to Pb^{2+} and form a coordinate covalent bond, neutralizing the excess positive charge of uncoordinated Pb^{2+} . As a result, the treated perovskite films demonstrated suppressed nonradiative recombination and enhanced carrier lifetimes. Later, Lewis bases bearing amines, hydroxyl and carbonyl groups show effective passivation on uncoordinated Pb^{2+} defects^[10–13]. Moreover, some conjugated small molecules or polymers also show excellent passivation on Lewis acid traps. These molecules usually contain functional groups like thiophene, pyridine, amine, thiocyanato and so on, thus providing multiple passivation sites^[14–17].

Introducing hydrogen bonding is also an effective method to passivate defects of perovskite films^[18–21]. Li *et al.*

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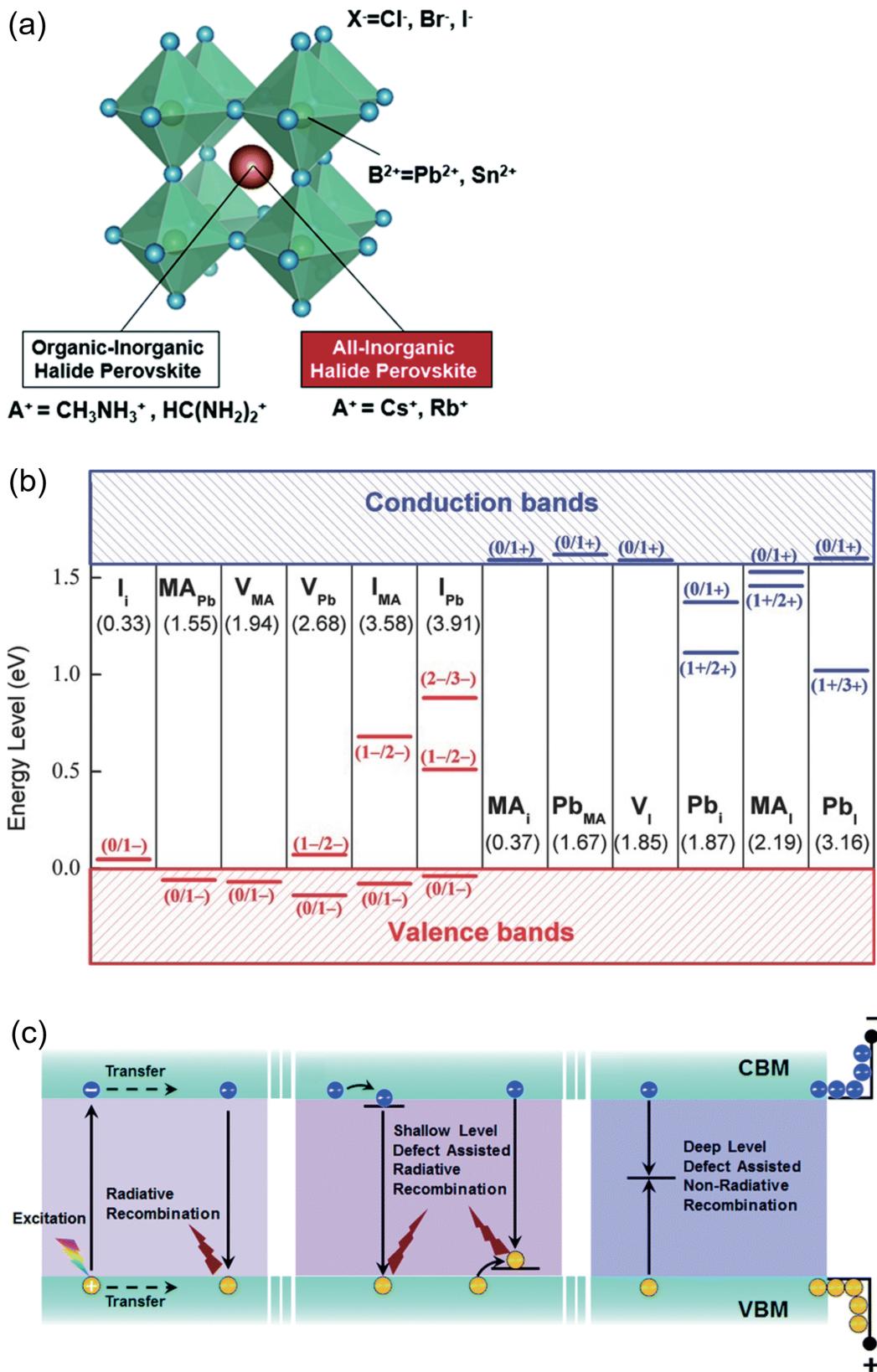


Fig. 1. (Color online) (a) Crystal structure for perovskite materials. Reproduced with permission^[3], Copyright 2019, the Royal Society of Chemistry. (b) Calculated transition energy levels of different defects in MAPbI₃. The corresponding formation energies are shown in parentheses. Reproduced with permission^[4], Copyright 2014, Wiley-VCH. (c) Charge recombination pathways in perovskite films. Reproduced with permission^[5], Copyright 2018, the Royal Society of Chemistry.

demonstrated that (4-aminobutyl) phosphonic acid hydrochloride (4-ABPACl) can heal the grain boundaries through strong hydrogen bondings between $\text{PO}(\text{OH})_2$ and NH_3^+ terminals and the perovskite ($\text{O}-\text{H}\cdots\text{I}$ and $\text{N}-\text{H}\cdots\text{I}$) (Fig. 2(c))^[18]. The modi-

fied perovskite gave higher PCEs. In addition to these molecules with hydroxyl and amino groups, fluorides are also used as passivators by forming hydrogen bonding between fluorine atoms and MA⁺ or FA⁺^[21]. These interactions can not

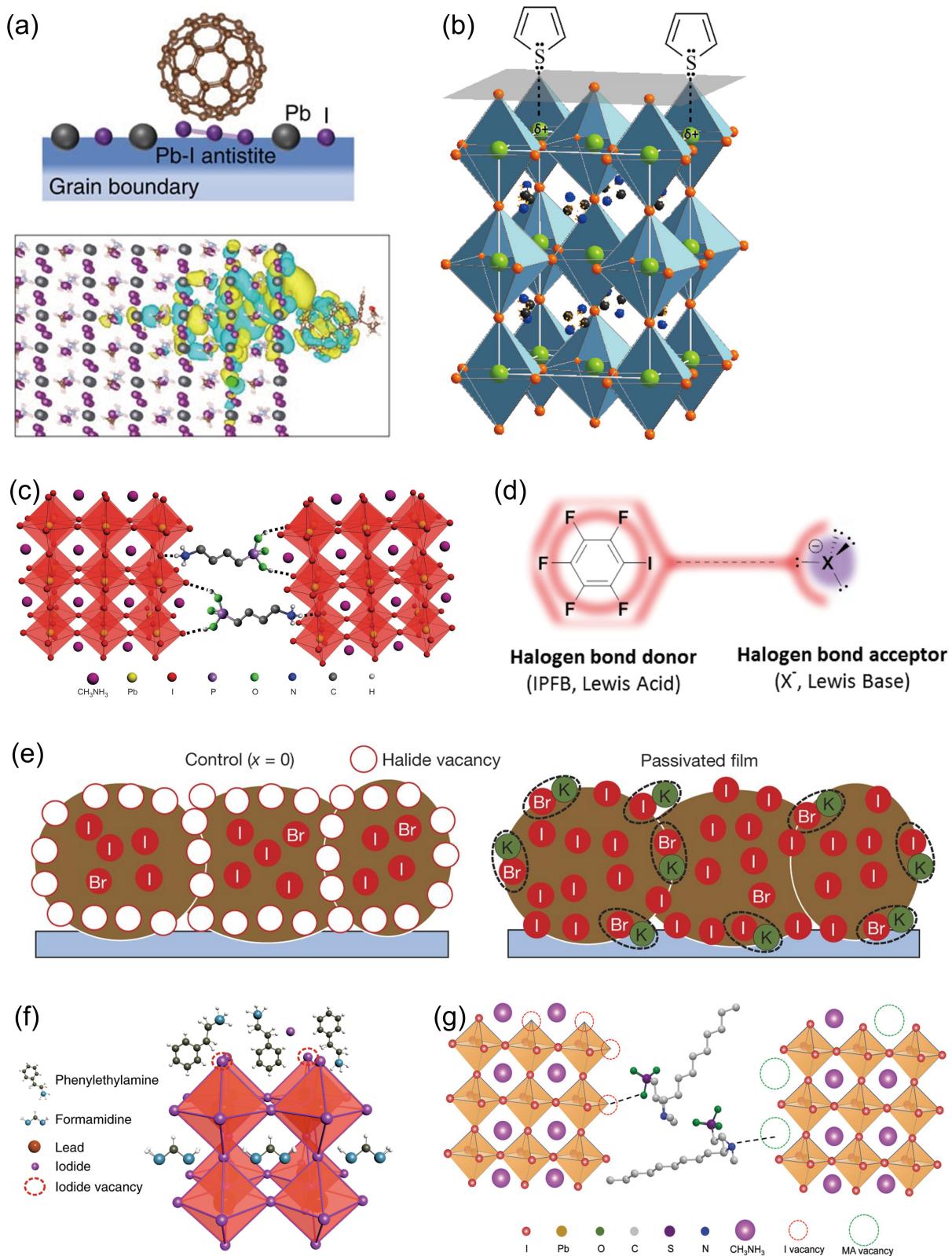


Fig. 2. (Color online) (a) Pb-I antisite defects passivated by PCBM. Reproduced with permission^[7], Copyright 2015, Nature Publishing Group. (b) Uncoordinated Pb^{2+} passivated by thiophene. Reproduced with permission^[9], Copyright 2014, American Chemical Society. (c) Hydrogen bondings (O-H...I and N-H...I) between 4-ABPACl and perovskite film. Reproduced with permission^[18], Copyright 2015, Nature Publishing Group. (d) Halogen bonding between IPFB and halogen anion. Reproduced with permission^[21], Copyright 2014, American Chemical Society. (e) Halide vacancy passivated by K^+ . Reproduced with permission^[28], Copyright 2018, Nature Publishing Group. (f) PEAI post-treatment on perovskite film. Reproduced with permission^[29], Copyright 2019, Nature Publishing Group. (g) MA and I vacancies passivated by 3-(decyldimethylammonio)-propane-sulfonate zwitterion. Reproduced with permission^[31], Copyright 2018, Wiley-VCH.

only modulate crystallization to enhance crystallinity but also improve structural stability of perovskite films.

Recently, halogen bonding has been used to enhance the efficiency of PSCs^[22–25]. Snaith *et al.* identified that the

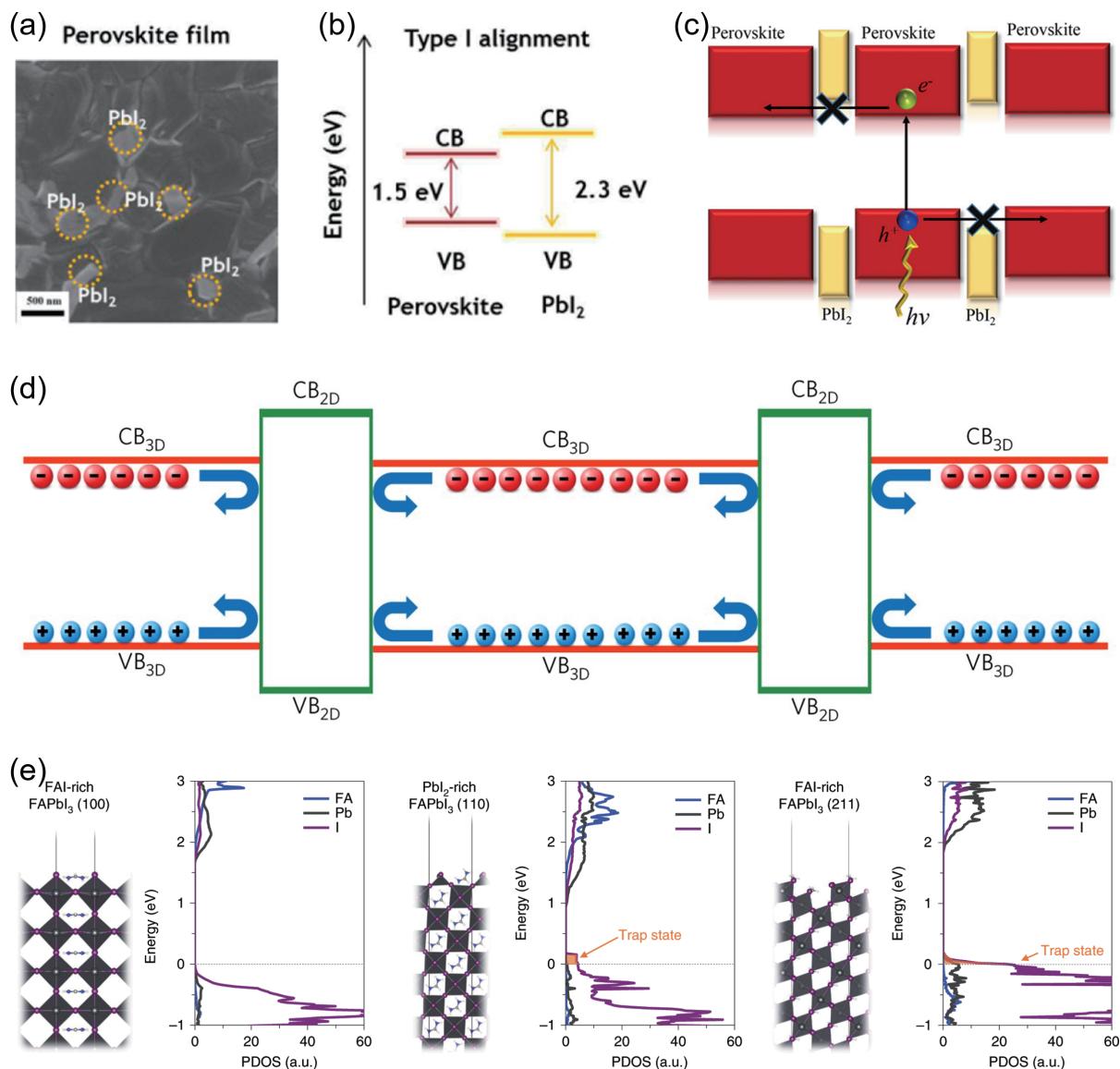


Fig. 3. (Color online) (a) SEM image for perovskite film containing PbI_2 phases. (b) Type-I energy level alignment. Reproduced with permission^[34], Copyright 2014, American Chemical Society. (c) PbI_2 as charge-transport barrier at grain boundaries. Reproduced with permission^[35], Copyright 2016, American Chemical Society. (d) 2D perovskites establish type-I heterojunction. Reproduced with permission^[36], Copyright 2017, Nature Publishing Group. (e) DFT calculation for trap states on the surface of different crystallographic facets. Reproduced with permission^[40], Copyright 2020, Nature Publishing Group.

uncoordinated I^- at perovskite surface are responsible for the charge accumulation and consequent recombination losses. They modified perovskite surface by using iodopentafluorobenzene (IPFB), which can form halogen bonding with uncoordinated I^- to passivate these defects^[22]. In IPFB, the fluorine atoms withdraw electrons out of the benzene ring, then the benzene ring in turn withdraws electrons from I^- , thereby leaving a partially positive charge around iodine atom (Fig. 2(d)). This electropositive region would interact with halogen anions to form strong halogen bonding. Similar results were achieved by using 4-iodo-2,3,5,6-tetrafluorobenzoic acid (l-TFBA)^[25].

Ionic passivation bases on electrostatic interaction between passivators and the charged defects in perovskite films. In earlier studies, Na^+ and K^+ cations were selected to passivate defects like MA vacancies and uncoordinated halogen ions via ionic bonding^[26]. Owing to small ionic radius, Na^+ and K^+ cations are unfavorable to occupy A-sites, but

they can occupy the interstitial sites in perovskite lattice to suppress ion migration and eliminate hysteresis^[27]. Some research suggested that Na^+ and K^+ cations can also assemble at grain boundaries and surface, forming KI or KBr to passivate defects and improve carrier lifetimes (Fig. 2(e))^[28]. Since You *et al.* reported highly efficient PSCs through phenylethy-lammonium iodide (PEAI) post-treatment (Fig. 2(f))^[29], organic ammonium halides have been intensively used as effective passivators. The halogen ions can fill in halogen vacancies, while $-\text{NH}_3^+$ terminal can interact with negatively charged defects in perovskite films, suppressing nonradiative recombination^[30]. Beyond these materials, zwitterions can simultaneously passivate positively and negatively charged defects (Fig. 2(g))^[31]. Usually, zwitterions contain positively charged ammonium group ($-\text{NH}_3^+$) and negatively charged carboxylate ($-\text{COO}^-$), phosphate ($-\text{PO}_4^{2-}$) or sulfonic ($-\text{SO}_3^-$) groups^[31–33].

Introducing wide-bandgap phases to form a type-I hetero-

junction at grain boundaries and interfaces can effectively suppress nonradiative recombination. The most common approach is to introduce excess PbI_2 to perovskite films (Fig. 3(a))^[34]. The type-I energy alignment (Fig. 3(b)) between PbI_2 and perovskite can hold back charge transfer at grain boundaries (Fig. 3(c)), thereby suppressing nonradiative recombination^[35]. Excess PbI_2 might also induce film degradation. In view of this, converting excess PbI_2 to two-dimensional (2D) perovskites by using long-chain ammoniums becomes a frontier technique. The 2D perovskites can establish type-I heterojunction to suppress nonradiative recombination (Fig. 3(d))^[36]. Moreover, the strong hydrophobicity of 2D perovskites would protect 3D perovskite from water erosion. Based on this technique, scientists have made highly efficient and stable PSCs. It should be noted that the issues for 2D or quasi-2D perovskites are crystal orientation and layer-layer distance. The undesirable crystal orientation and long layer-layer distance would definitely block charge transport in the film and at interfaces, which might sacrifice J_{sc} and FF^[37]. Therefore, optimizing crystal orientation and layer-layer distance of 2D or quasi-2D perovskites is critical.

Though using passivators is promising to deactivate various defects, the initial generation of defects should not be ignored. Perovskite layers made from solution are polycrystalline films that inevitably present grain boundaries. Since grain boundaries contain various defects, increasing grain size to reduce grain boundaries is vital for high-performance PSCs. Moreover, the optoelectronic anisotropy of perovskite films is quite important. Different crystal facets might present different trap states^[38, 39]. Single-crystal perovskites show less defects. Zheng *et al.* investigated the dependence of defect density on crystallographic facets *via* density functional theory (DFT) calculations based on a cubic FAPbI_3 model^[40]. They revealed that there are no defects on (100) facet, while the (110), (111) and (211) facets present defects (Fig. 3(e)). By incorporating a trace amount of surface-anchoring alkylamine ligands (AALs) into perovskite precursor, they achieved (100)-oriented crystal growth with lower defect density and enhanced carrier mobility. Similar phenomenon was observed in Pb-Sn mixed perovskites^[41, 42]. Optimizing crystal orientation and exploring facet-dependent defect variation have attracted interests^[43–45].

In short, different defects in perovskite layer are discussed, and typical passivation strategies are highlighted. More investigation on the generation, location and types of defects would be helpful for making highly efficient and stable PSCs.

Acknowledgments

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Jie Sun got her BS from Minzu University of China in 2021. Now she is a PhD student at University of Chinese Academy of Sciences under the supervision of Prof. Liming Ding. Her research focuses on perovskite devices.



Shengzhong (Frank) Liu received his PhD from Northwestern University in 1992. After his postdoctoral research at Argonne National Lab, he joined high-tech companies in US for research including nanoscale materials, thin-film solar cells, laser processing, diamond thin films, etc. His invention at BP Solar on semi-transparent photovoltaic module won R & D 100 Awards in 2002. In 2011, he was selected into China talent program, and now he is a professor at Shaanxi Normal University.



Liming Ding got his PhD from University of Science and Technology of China (was a joint student at Changchun Institute of Applied Chemistry, CAS). He started his research on OSCs and PLEDs in Olle Inganäs Lab in 1998. Later on, he worked at National Center for Polymer Research, Wright-Patterson Air Force Base and Argonne National Lab (USA). He joined Konarka as a Senior Scientist in 2008. In 2010, he joined National Center for Nanoscience and Technology as a full professor. His research focuses on innovative materials and devices. He is RSC Fellow, and the Associate Editor for *Journal of Semiconductors*.