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The Design, Properties and Future Applications of Perovskite Thin films with Selective Organic Molecules

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14. ABSTRACT

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The design, properties and future applications of perovskite thin films with selective organic molecules

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Summary

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Achievements:

Lead halide perovskite solar cells have realized unprecedentedly rapid progress in power conversion efficiency (PCE) since its solid state development in 2012, now exceeding an impressive 23%. This rapid progress has promoted tremendous research efforts to investigate the fundamental physical and chemical properties of the perovskite materials. Of particular interest is whether or not the optoelectronic (e.g. band structure, absorption, carrier lifetime, mobility) and physical properties (e.g. crystal growth, ion migration, environmental stability) of the perovskite crystal can be tailored via the organic molecules. In this project, we performed comprehensive studies from the investigation of intrinsic properties to applications to a variety of optoelectronic properties depending on organic components in perovskite materials.

1. Investigation of organic component dependent intrinsic properties of PEROVSKITE materials

Among the family of perovskite materials, methylammonium lead triiodide ($CH_3NH_3PbI_3$, $MAPbI_3$) has been most commonly for which optoelectronic properties have been well-investigated. Later, formamidinium lead iodide ($HC(NH_2)_2PbI_3$, $FAPbI_3$) attracted remarkable attention due to its potential for improvements in optoelectronic properties and stability. Despite the many studies utilizing FA-based perovskite materials for high efficiency solar cells, the intrinsic electrical, optical, and structural properties of $FAPbI_3$ have not been studied. We demonstrated for the first time the growth of a 5 mm sized $FAPbI_3$ single crystal to study its intrinsic properties.[1] Thermal, optical and electrical properties including mobility, conductivity, carrier concentrations and defect density were investigated. We have rationalized the superior performance of $FAPbI_3$ based solar cells over $MAPbI_3$, and demonstrated efficient photodetectors based on the single crystals.

Inspired by the superior optoelectronic properties and stability of the FA-based perovskite materials, we have demonstrated for the first time FA-based light emitting diodes (LED).[2] Our LED devices showed superior efficiency and stability over conventional LED devices based on MA-based perovskite materials. Later, we also adopted FA-based perovskite materials in solar cell devices where PCEs exceeding 20% were demonstrated.[3] In the study, we investigated a self-passivation effect by volatilization of the organic FA cation in $FAPbI_3$ perovskite thin films. We unraveled a close relationship between the stoichiometry and defect density in the perovskite films. The direct correlation between the defect density and characteristic current-voltage hysteresis of perovskite solar cells was demonstrated.

2. Design of functional organic molecules to tune perovskite material and device properties

Taking advantage of our expertise in organic materials, we also developed a variety of functional organic molecules to tune the properties of perovskite materials and devices. We have synthesized organic molecules that can function as both charge transporting and interfacial modification layers.[4-6] We found that the energy level alignment and functional groups of the organic molecules play crucial roles in performance and stability of the perovskite solar cells. The application of functional small molecules in the perovskite solar cells enabled significant enhancement in PCE as well as stability of the devices.

Based on the investigation of fundamental properties of the perovskite materials, we learned the importance of crystal growth for reducing defect density of the perovskite materials. For crystal growth engineering, we designed organic small molecule additives for tuning molecular interactions within the perovskite precursor solution. We successfully demonstrated crystal growth engineering, where the grain size of the perovskite films was enhanced from around 100 nm to larger than 1 μm.[7] Our small molecule additives: 1) interact with perovskite precursors to retard nucleation and thus enhance the crystallinity, and 2) precipitate at grain boundaries to passivate defects. We further developed our idea to understand the general rules governing molecular interactions and proposed possible rules for selection of additive molecules: 1) hydrogen bonding ability, 2) steric accessibility, and 3) hardness-softness matching.[8] We exemplified the application of the rules to to formulate the composition of precursor solutions for fabricating high quality $FAPbI_3$ perovskite films. Owed to the high quality perovskite films, we achieved PCEs exceeding 20 % with high reproducibility. Recently, we further applied our understanding of the interactions of organic materials with perovskite materials to develop a new perovskite/polymer cross-linked system. We found that the polymeric cross-links with perovskite grains to passivate defects and enhance the environmental stability (manuscript under revision in *Nature Communications*).

We also demonstrated novel white emitting LED devices based on perovskite/organic hybrid systems.[9] Blue emitting perovskite LED devices were fabricated using Cs-based perovskite nanocrystals. To generate white light emission, we incorporated yellow color emitting polymeric materials, poly[2-methoxy-5-(2-ethylhexyloxy)-1,4-phenylenevinylene] (MEH:PPV). We successfully fabricated for the first time the white emitting LED devices based on the perovskite/organic hybrid system where efficient energy transfer from the perovskite nanocrystals to the MEH:PPV was confirmed from photo-physical studies.

3. Design of new functional perovskite materials with select organic A cations

Based upon the understanding on the relationship between the intrinsic properties of perovskite materials and organic molecules, we designed new hetero-dimensional perovskite materials. We have established the solution composition-dependent phase formation of 2D/3D hybrid perovskite materials and related kinetic models for crystal growth. Based on the study, we were able to fabricate hetero-dimensional perovskite thin films where the bulk is composed of 3D perovskite while the surface or grain boundaries are composed of 2D perovskite.[10] The 2D perovskite at the surface with hydrophobic organic molecules protects the 3D perovskite from the environmental degradation caused by moisture, heat and light to enhance the phase stability of the bulk 3D perovskite. Our hetero-dimensional perovskite films did not show degradation after exposure to relative humidity of 80% for 24 h whereas the bare perovskite film was completely degraded. Furthermore, the 2D perovskite passivates defects at the grain boundaries, and consequently the photoluminescence lifetime of the perovskite films were elongated more than 10 times from around 40 ns to 500 ns. Owing to superior optoelectronic properties, we demonstrated the perovskite solar cells with PCE exceeding 21%. Notably, one of our devices was certified by Newport, and showed a *stabilized* PCE of 19.77%, which is the highest *stabilized* certified PCE among all the perovskite solar cells. Also, the perovskite solar cell with 2D perovskite showed superior operational stability, where T80 lifetime (time at which PCE of the device decays to 80% of initial PCE) of the solar cells was improved from 592 h to 1362 h. We noticed that the 2D perovskite at grain boundaries can suppress ion migration, which is a major factor causing the degradation of perovskite materials under a built-in or applied electric field.

Inspired by the suppressed ion migration, we recently fabricated LED devices based on hetero-dimensional perovskite films. The superior optoelectronic properties of the hetero-dimensional perovskites over conventional 3D perovskite materials enabled significant enhancement of electroluminescence efficiency from 0.46 ph/el% to 7.70 ph/el%. Furthermore, our LED device showed superior operational T50 lifetime exceeding 200 h compared to the devices based on conventional 3D or quasi-2D (< 0.2 h, manuscript in preparation).

4. Summary

In summary, we have investigated the effects of organic molecules on intrinsic optoelectronic and physical properties of perovskite materials, where important properties were unraveled to provide a basis for the development of a variety of perovskite optoelectronic devices. We also developed functional organic materials to enhance the properties of perovskite materials and devices. The understanding of molecular interactions between the organic molecules and perovskite materials facilitated the development of organic materials that can be used as a charge transporting layer, interfacial modification layer, crystal growth and defect passivation agent. A novel perovskite/polymer cross-linked system was demonstrated for the first time. Finally, we developed a new hetero-dimensional perovskite system of which optoelectronic properties and stability is superior to conventional pure 3D perovskite or quasi-2D perovskite materials. Our new hetero-dimensional perovskite materials enabled highly efficient and stable perovskite solar cells and light emitting diodes. We believe that the fundamental understanding on the molecular interactions between the perovskite materials and organic molecules as well as new materials concepts (perovskite-polymer cross-linking, hetero-dimensional perovskite system) will provide important insights towards the development of commercially available perovskite optoelectronic devices.

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Abstract

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New discoveries, inventions, or patent disclosures:

Do you have any discoveries, inventions, or patent disclosures to report for this period?

No

Please describe and include any notable dates

Do you plan to pursue a claim for personal or organizational intellectual property?

Changes in research objectives (if any):

Change in AFOSR Program Officer, if any:

Extensions granted or milestones slipped, if any:

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

Report Document - Text Analysis

Report Document - Text Analysis

Appendix Documents

2. Thank You

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