

Solution-Printed Perovskite Tandem Solar Cells

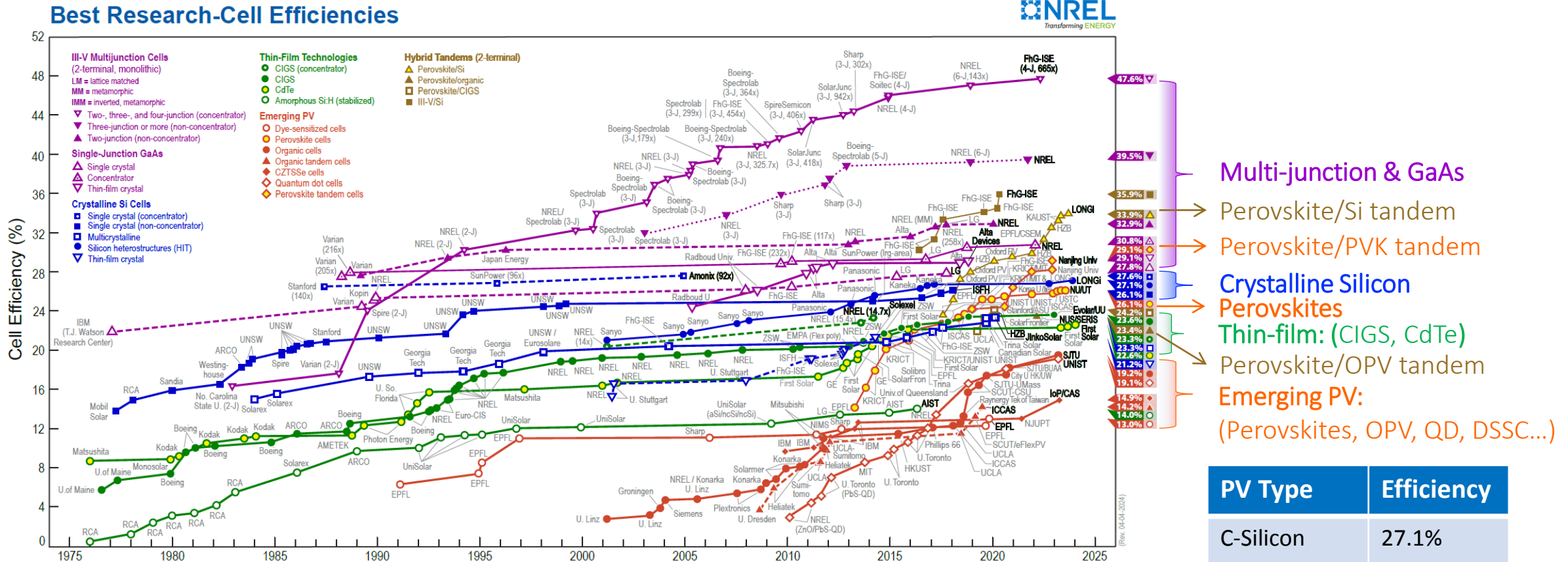
Fei Guo
Jinan University
21.05.2024



OUTLINE

- ❑ Introduction to scalable crystallization of perovskite films
- ❑ Our old / recent results
 - Vacuum quenching / Perovskite-based tandems
- ❑ Summary
- ❑ Acknowledgement

Efficiency Evolution of Photovoltaics

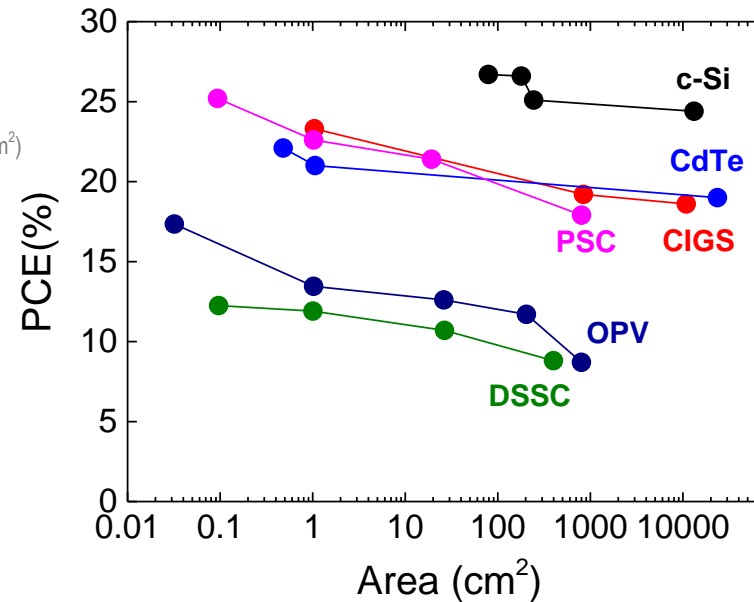
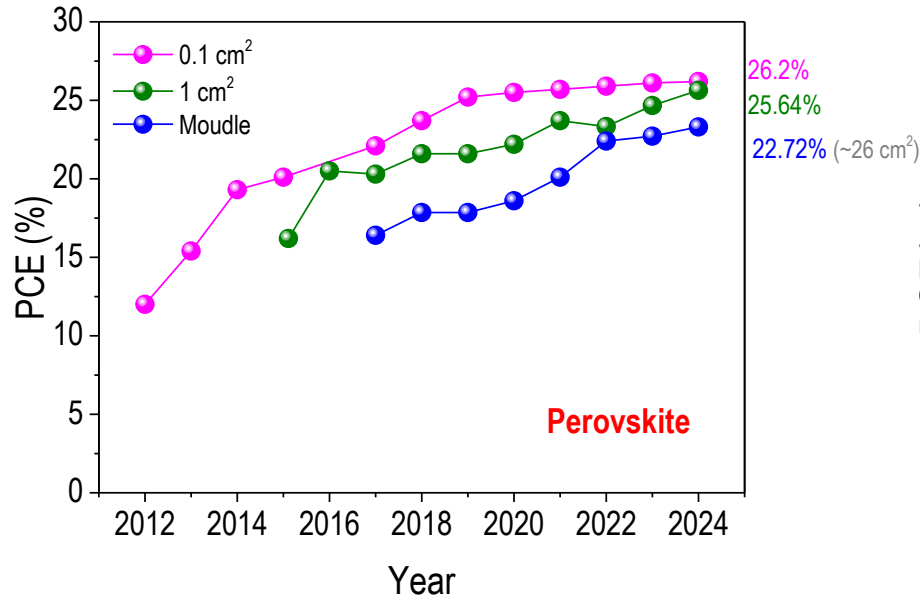


Emerging PV = Third generation PV = Solution Processable PV

PV Type	Efficiency
C-Silicon	27.1%
Perovskite	26.1%
CIGS	23.6%
CdTe	22.6%
OPV	19.2%

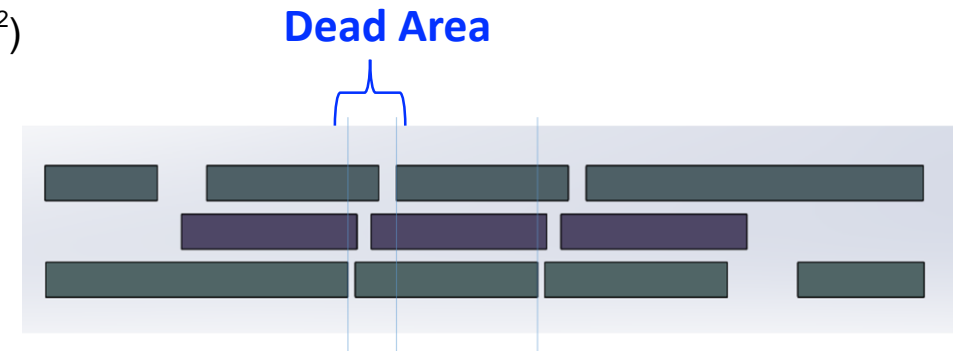
Efficiency Losses During Upscaling

Large efficiency gap between small-size devices and large-area modules



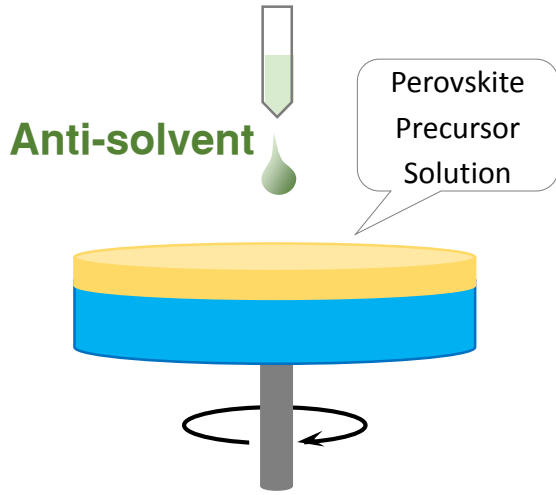
Solutions:

- Scalable coating of high-quality perovskite films
- Advanced module design and fabrication technology



Efficiency Losses During Upscaling

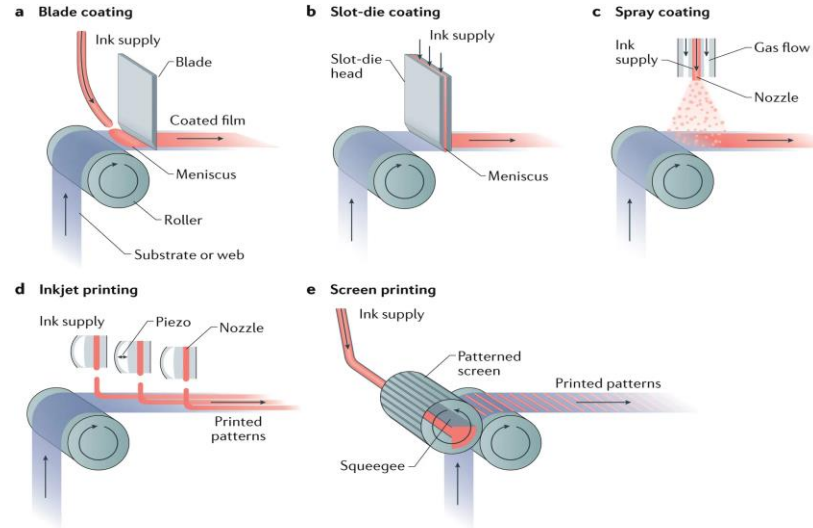
Spin coating



Pros & Cons:

- ✓ Easy to handle in lab
- ✓ Small amount of solution
- ✓ Good at producing uniform film
- ✗ High material waste (> 90%)
- ✗ Non scalable (< 5×5 cm²)
- ✗ Unable to translate to scalable lines

Scalable coating



Crystallization protocols are difficult to be transferred to scalable coating lines

Challenges:

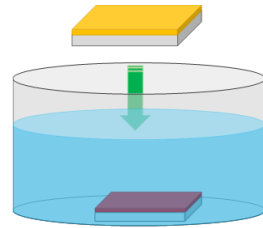
- ❑ **Film homogeneity (macro)**
Solvent evaporation and solute migration/aggregation take place simultaneously
- ❑ **Crystal morphology (micro)**
Control crystallization dynamics
- ❑ **Crystal structure**
Phase purity, crystal orientation, phase segregation, defects ...

It is of essential importance to **control crystallization dynamics** to achieve large scale fabrication of perovskite thin-films

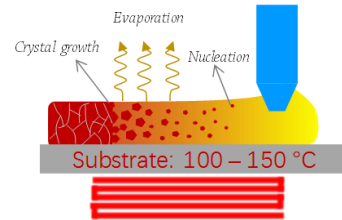
Nature Reviews Materials, 2018, 3, 1-20.

Established Crystallization Protocols

Nat Energy, 2017, 2, 17038.
ACS Energy Lett, 2018, 3, 322.
Sustain Energy Fuels, 2018, 2, 2442.
 ...



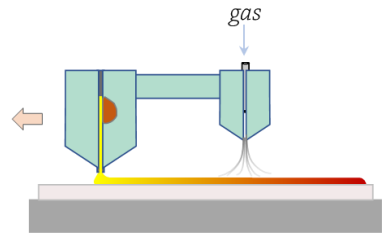
Antisolvent quenching



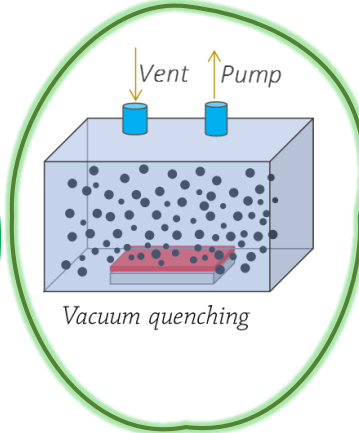
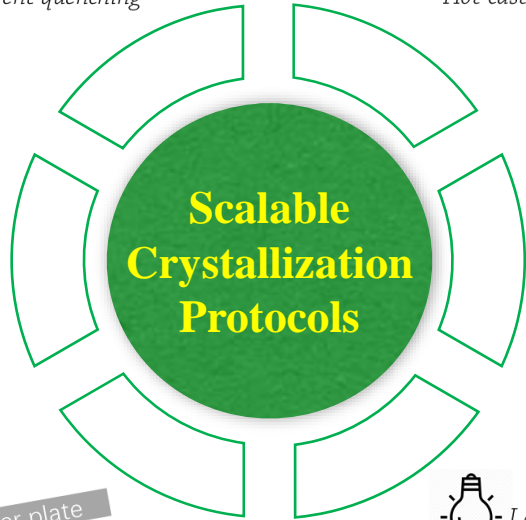
Hot-casting

Energ Environ Sci, 2015, 8, 1544.
Adv Energy Mater, 2017, 7, 1700302.
Sci Adv, 2019, 5, eaav8925.
Adv Mater, 2020, 32, 2000995.
Nat Energy, 2018, 3, 560-566.
 ...

J Power Sources, 2015, 277, 286.
Joule, 2019, 3, 402.
Sci Adv, 2019, 5, eaax7537.
Adv Energy Mater, 2020, 10, 1903108.
ACS Energy Lett, 2019, 4, 2393.
 ...



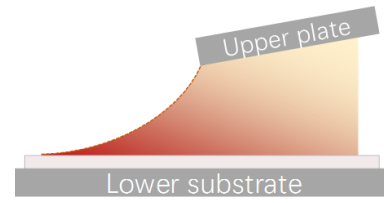
Gas quenching



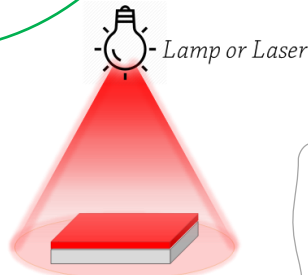
Vacuum quenching

Adv. Funct. Mater. **2019**, 29, 1900964.
Adv. Sci. **2019**, 6, 1901067.
Nano Energy, **2019**, 66, 104099.
Adv. Energy Mater., **2020**, 10, 2000173.
Adv. Funct. Mater., **2020**, 30, 2001240.
ACS Energy Lett., **2020**, 5, 1386-1395.
Adv. Funct. Mater., **2021**, 30, 2107644.
Energ Environ. Sci., **2020**, 13, 4666.
Adv. Funct. Mater., **2022**, 2112146.
ACS Energy Lett. **2023**, 8, 502.

Nat Commun, 2017, 8, 16045.
Energ Environ Sci, 2016, 9, 2295.
Adv Mater, 2017, 29, 1701440.
Nature, 2017, 550, 92..
 ...



Meniscus assisted crystallization



Radiative annealing

J Mater Chem A, 2015, 3, 9123
Adv Energy Mater, 2020, 10, 1902898.
Energ Environ Sci, 2020, 13, 1187.
Adv Energy Mater, 2018, 8, 1702915
 ...

Our Printed Perovskites

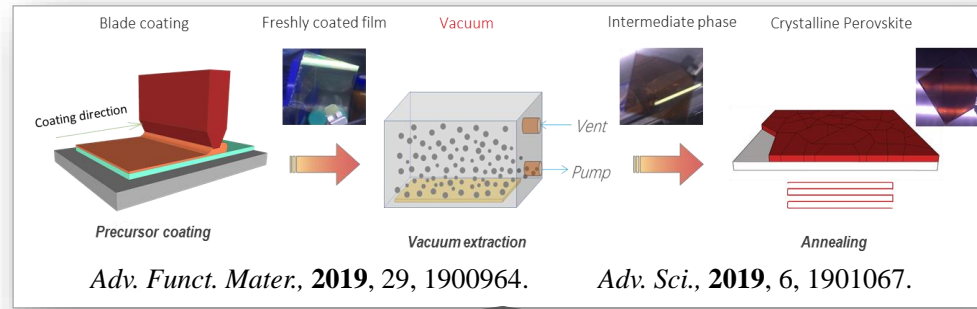
Since 2017

Jinan Uni

Scalable crystallization technique

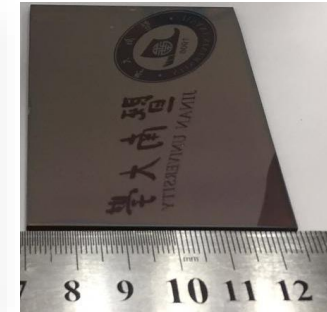


Precursor ink



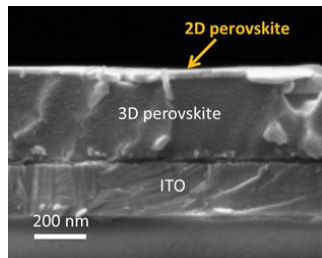
Adv. Funct. Mater., **2019**, 29, 1900964.

Adv. Sci., **2019**, 6, 1901067.



Large-area film

Dimensionality

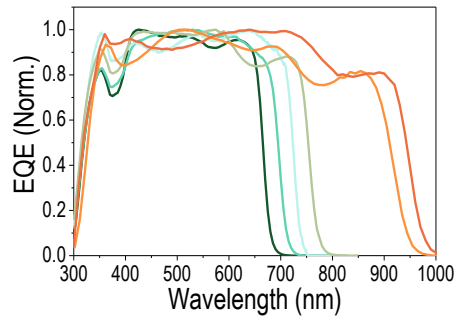


Energy Environ. Sci., **2020**, 13, 4666.

Nano Energy, **2022**, 91, 106658.

Adv. Funct. Mater., **2022**, 2112146

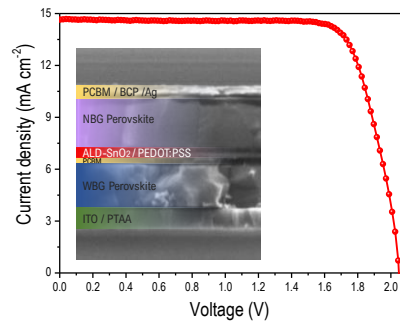
Wide/narrow bandgap



ACS Energy Lett., **2020**, 5, 1386-1395.

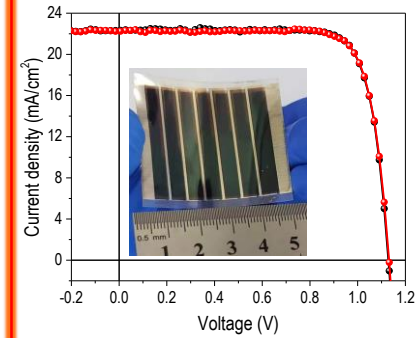
Nano Energy, **2019**, 66, 104099.

Tandems



ACS Energy Lett. **2023**, 8, 502-512

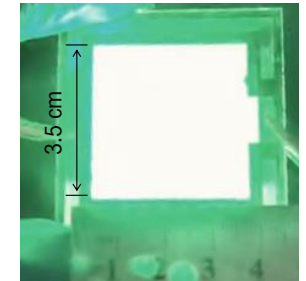
Flexible device



Adv. Funct. Mater., **2020**, 30, 2001240.

Adv. Sci., **2021**, 8, 2101856.

Perovskite LED



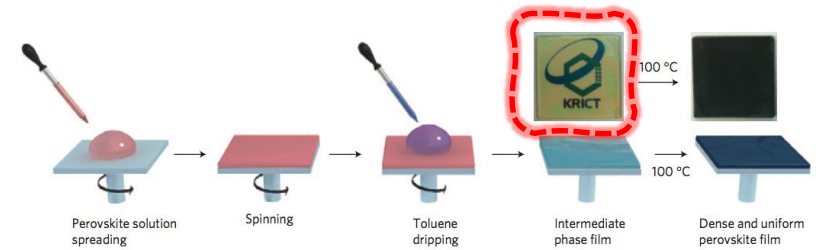
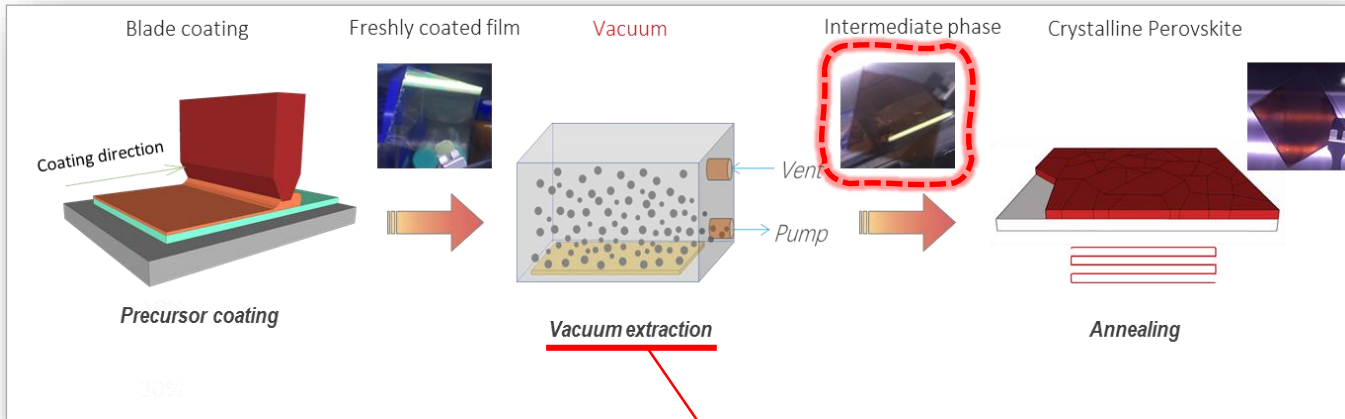
Adv. Funct. Mater., **2021**, 32, 2107644.

ACS Nano, **2023**, 17, 4483

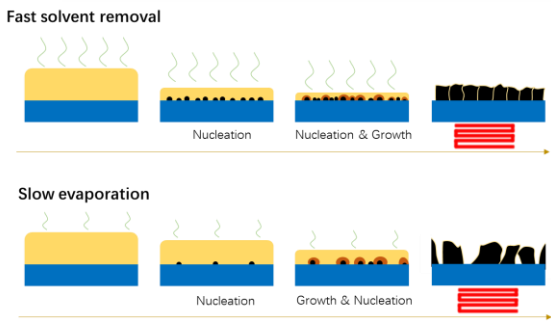
报告提纲

- Introduction to scalable crystallization of perovskite films
- **Our old / recent results**
 - Vacuum quenching / Perovskite-based tandems**
- Summary
- Acknowledgement

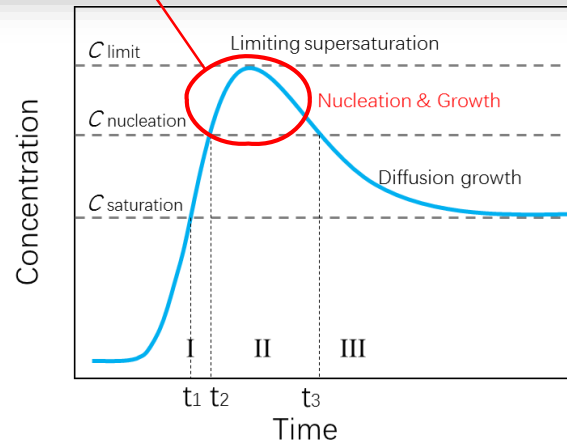
Our Method - Vacuum Quenching



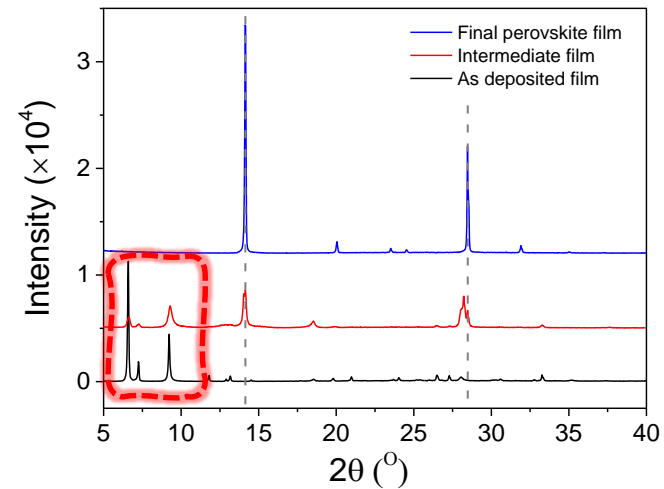
Nat. Mater. **2014**, *13*, 897.



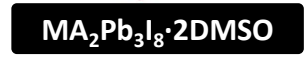
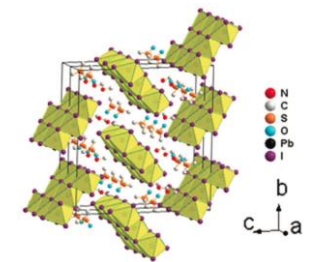
Energy Environ. Sci., **2020**, *13*, 4666.



Vacuum: Fast nucleation and prevent solute migration/aggregation by fast solvent removal



Adv. Sci., **2019**, *6*, 1901067.



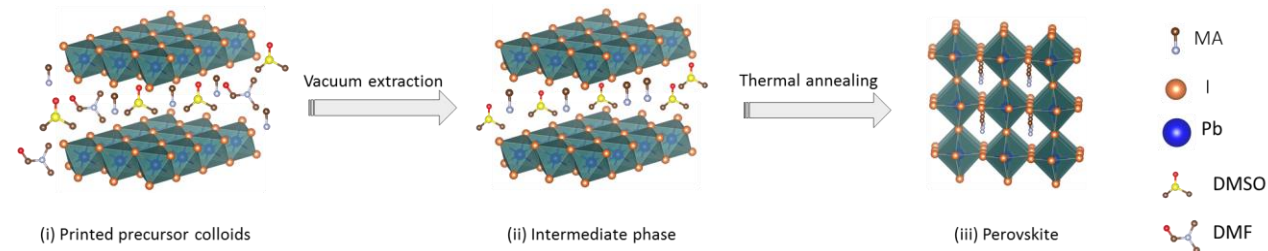
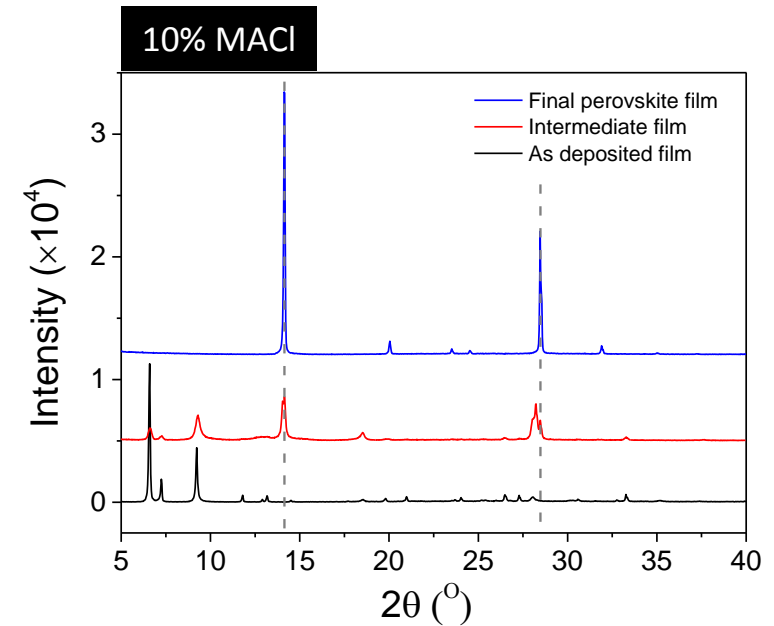
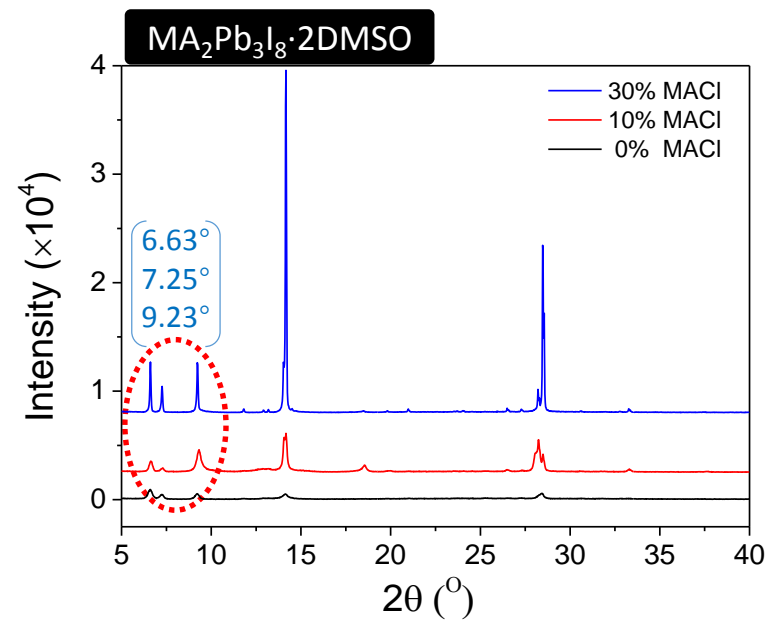
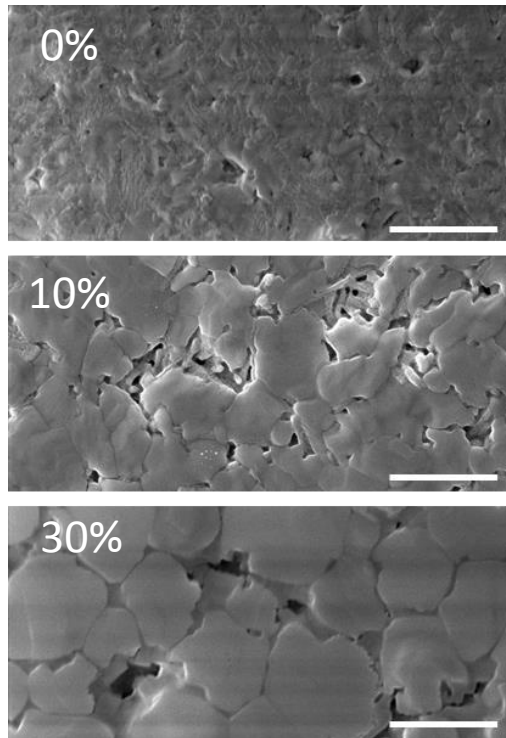
J. Am. Chem. Soc. **2016**, *138*, 9919.

Our Method - Vacuum Quenching

Crystallization dynamics

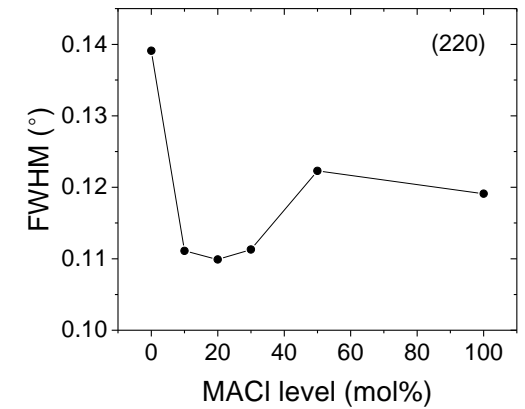
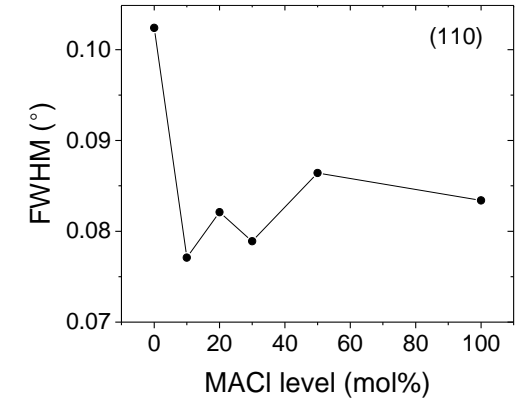
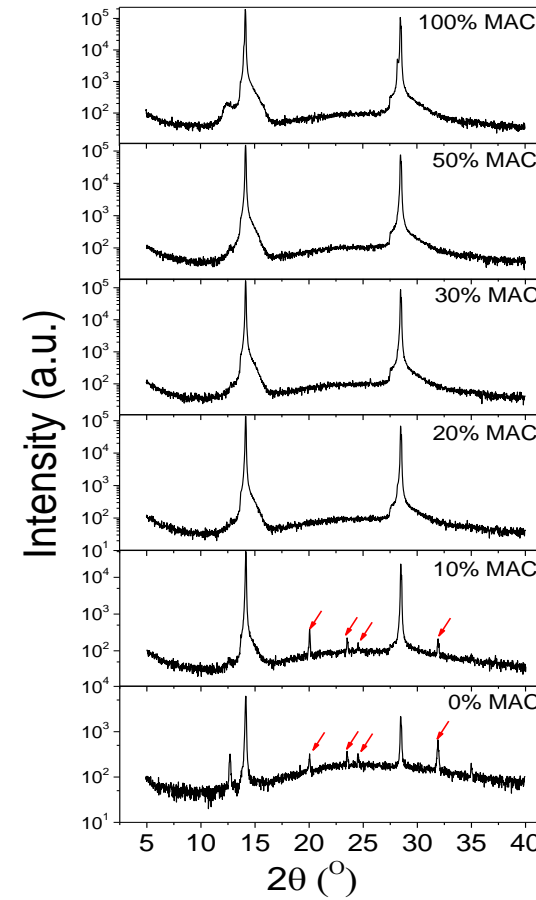
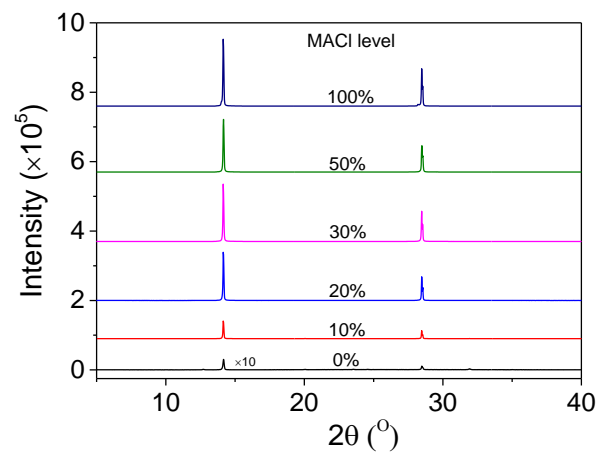
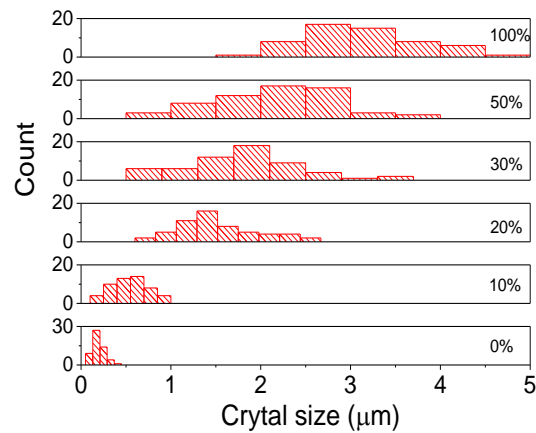
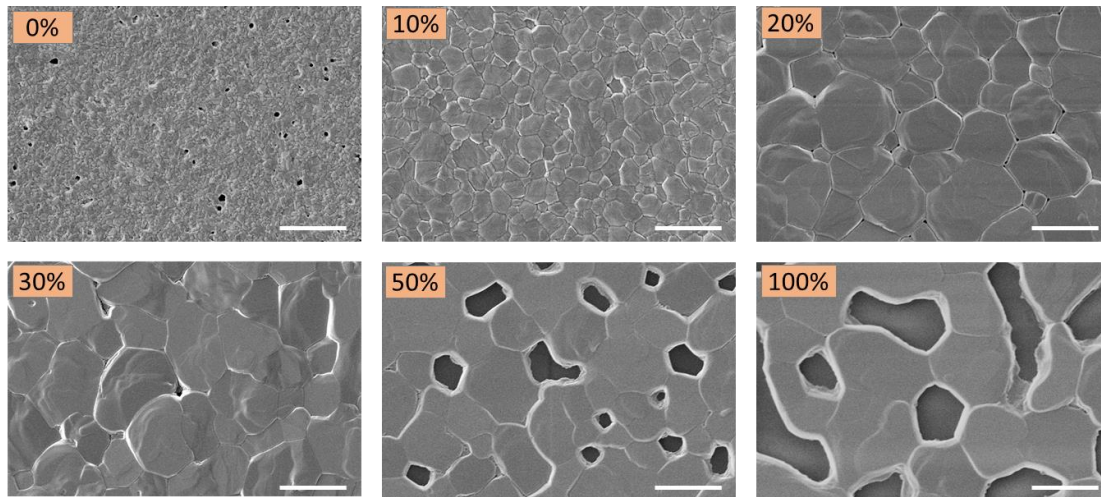
Additive MACI: Modulate crystal morphology and stabilize phases

Intermediate film



Our Method - Vacuum Quenching

MAPbI₃ crystal quality

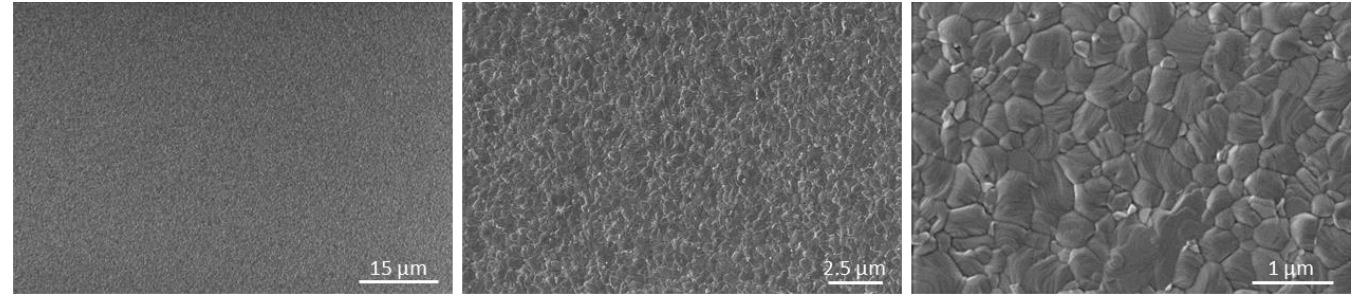


Our Method - Vacuum Quenching

Importance of vacuum

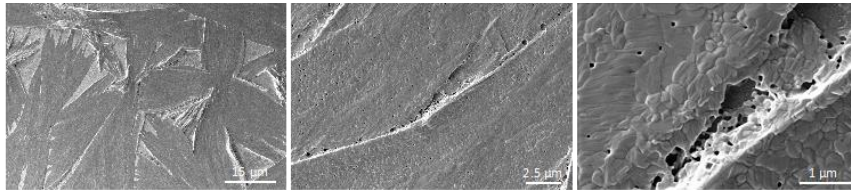
With vacuum

10% MACI

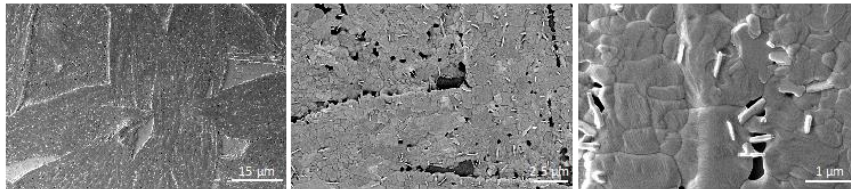


Without vacuum

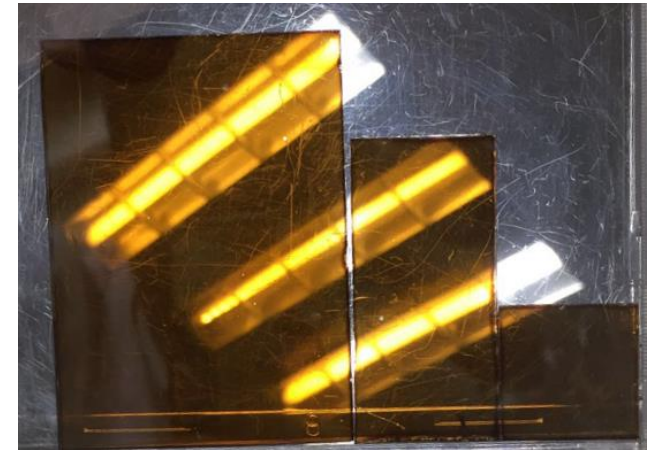
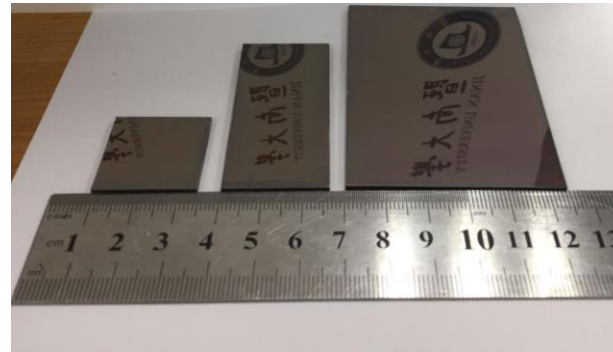
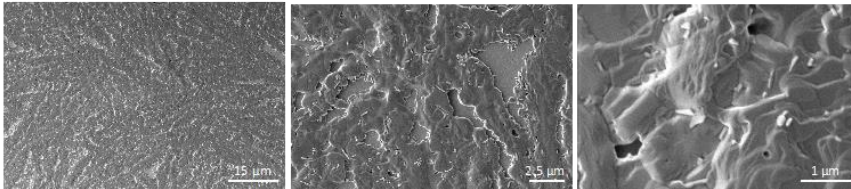
0% MACI



10% MACI



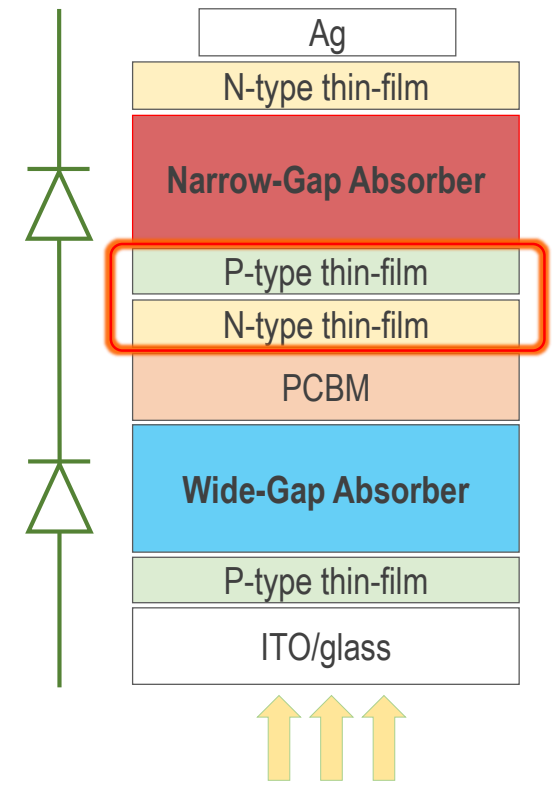
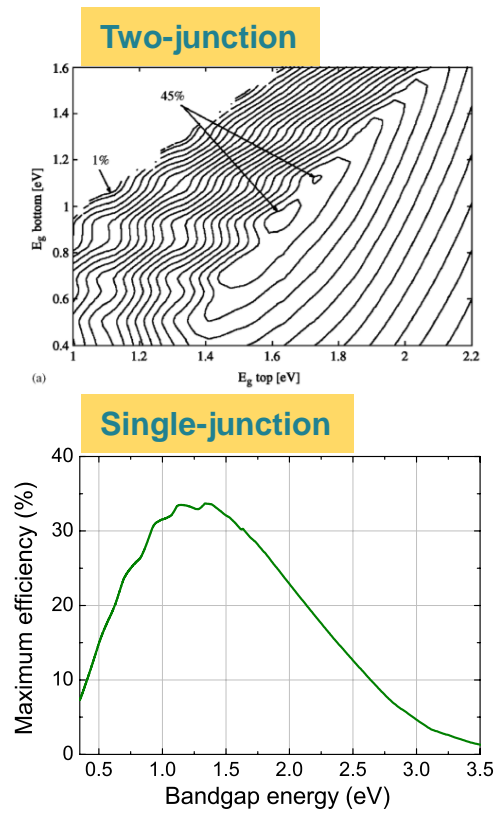
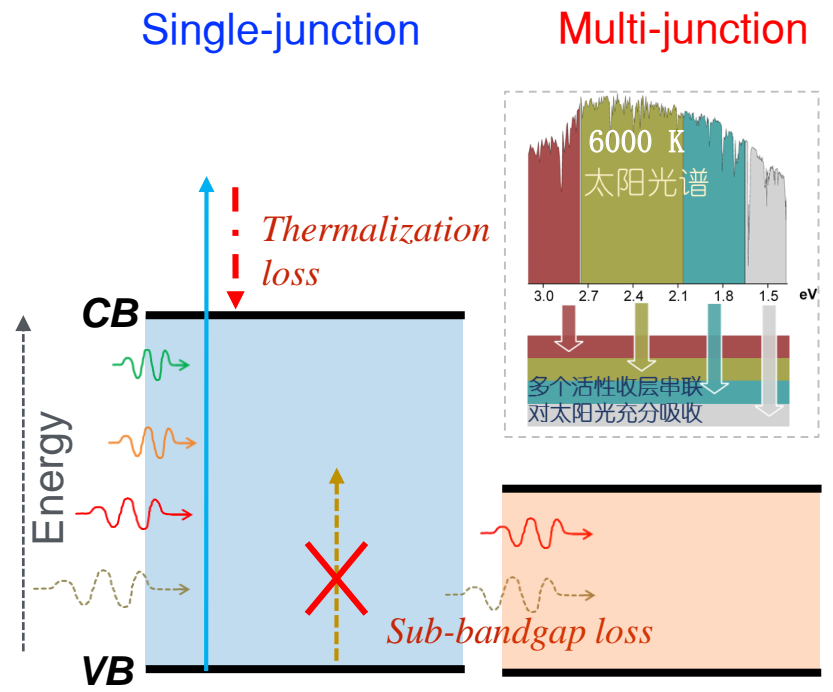
30% MACI



报告提纲

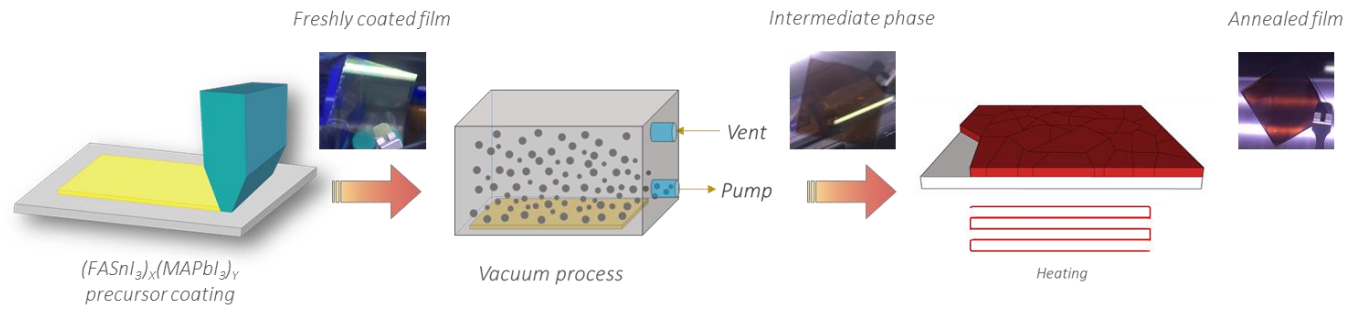
- Introduction to scalable crystallization of perovskite films
- Our old / recent results
 - Vacuum quenching / Perovskite-based tandems
- Summary
- Acknowledgement

Why Tandem Architecture ?

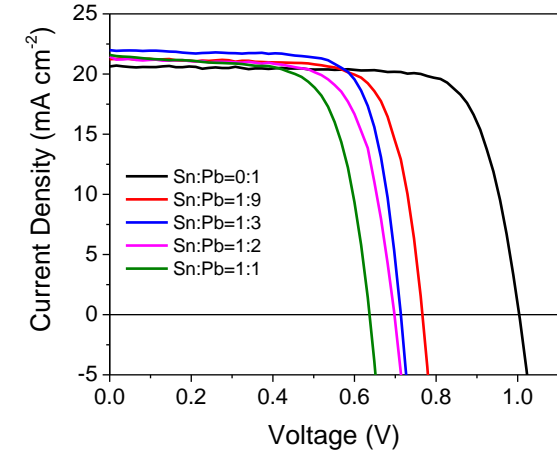
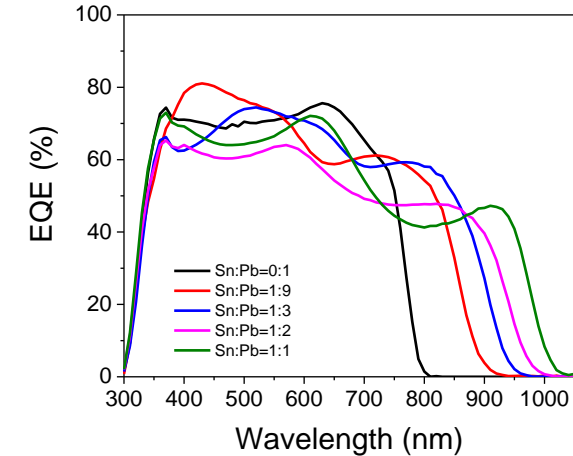
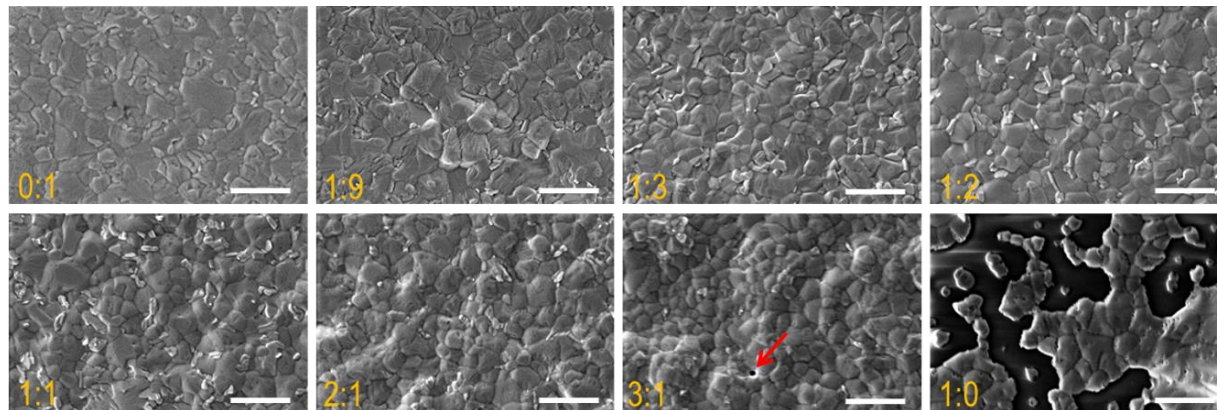


- Narrow gap**
- Thick enough to ...
- Charge recombination**
- High transmittance
 - Low ohmic resistance
 - Prevent solvent penetration
- Wide gap**
- Low voltage loss

Printing Low-bandgap perovskites



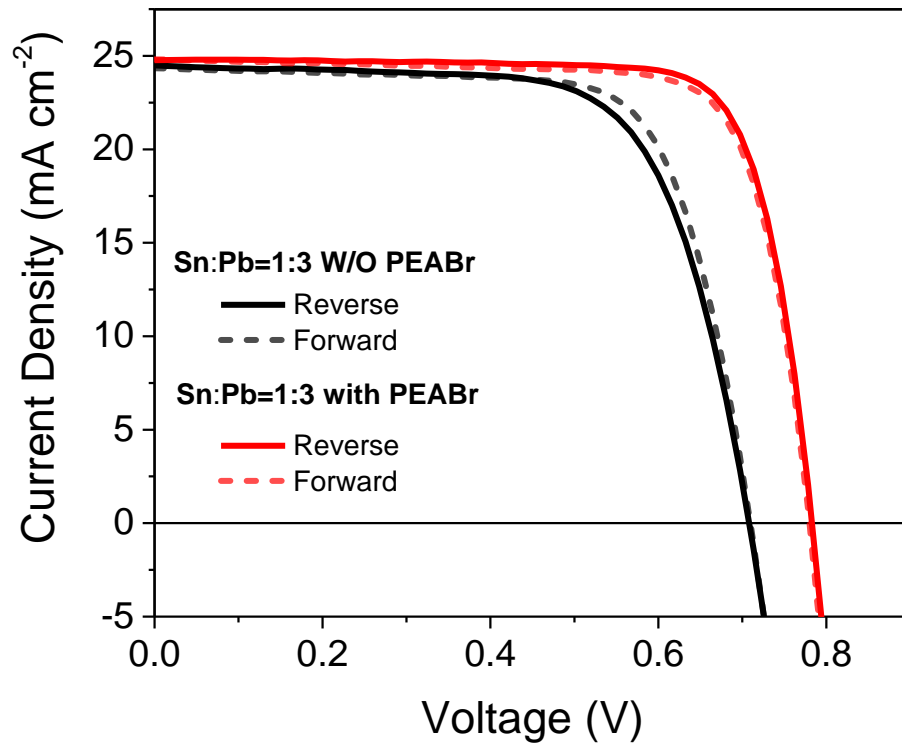
Different Sn-Pb ratios



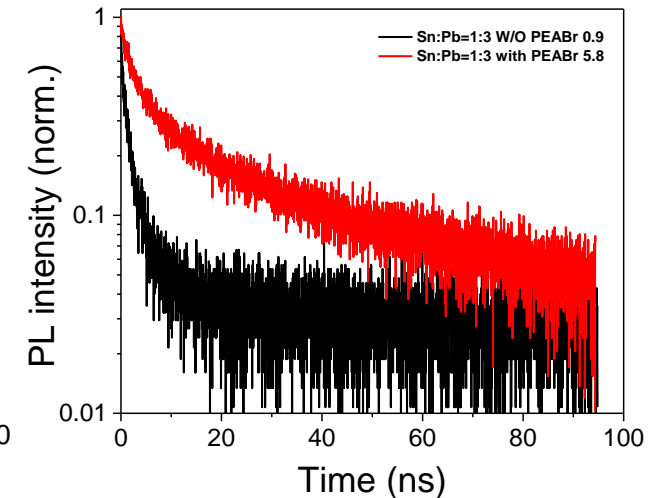
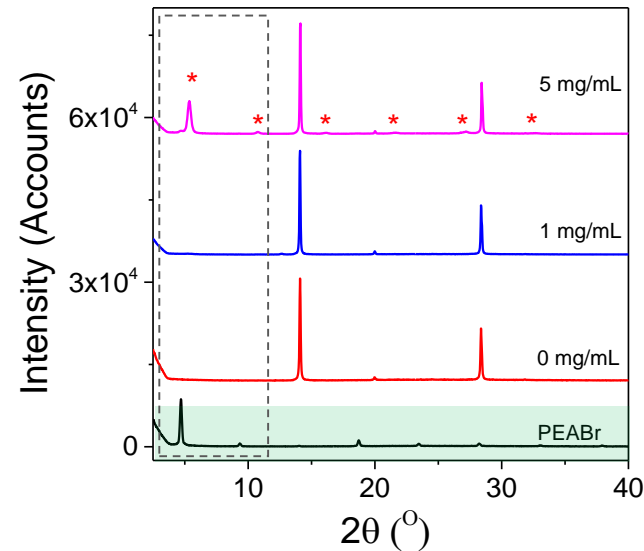
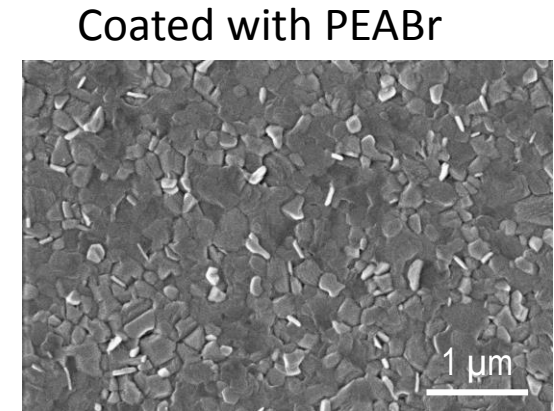
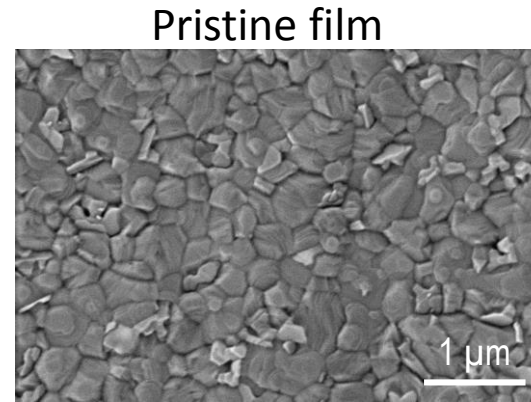
High FFs but high Voc losses!

Passivation?

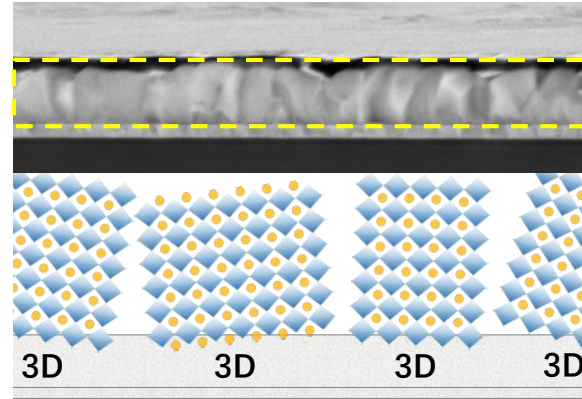
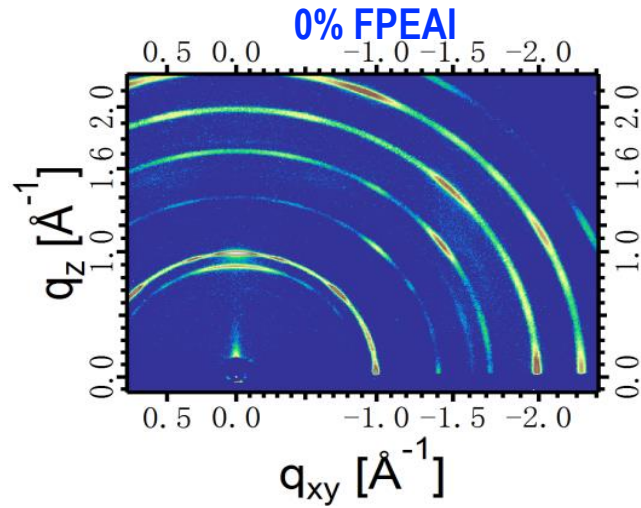
Passivation with PEABr (Two-step)



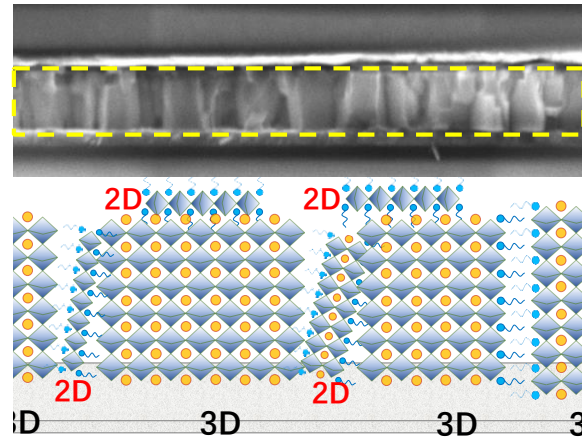
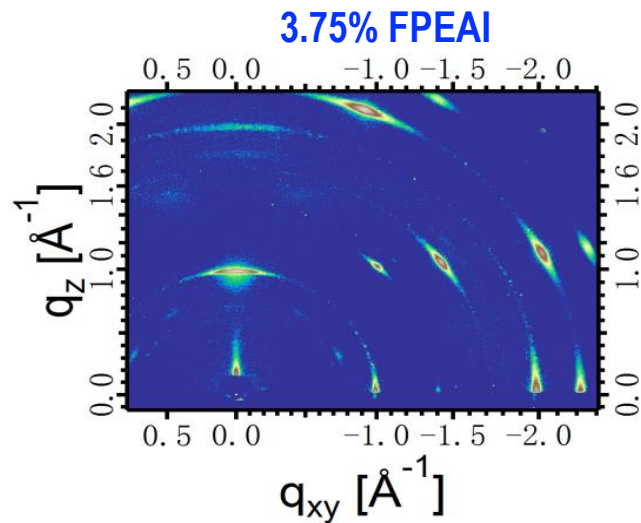
$J_{sc}=24.9$ $V_{oc}=0.78$ $FF=0.78$ $PCE=15.15\%$



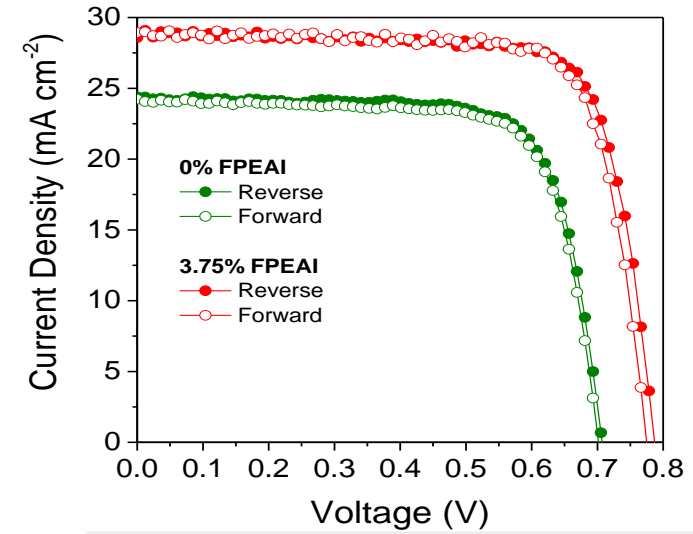
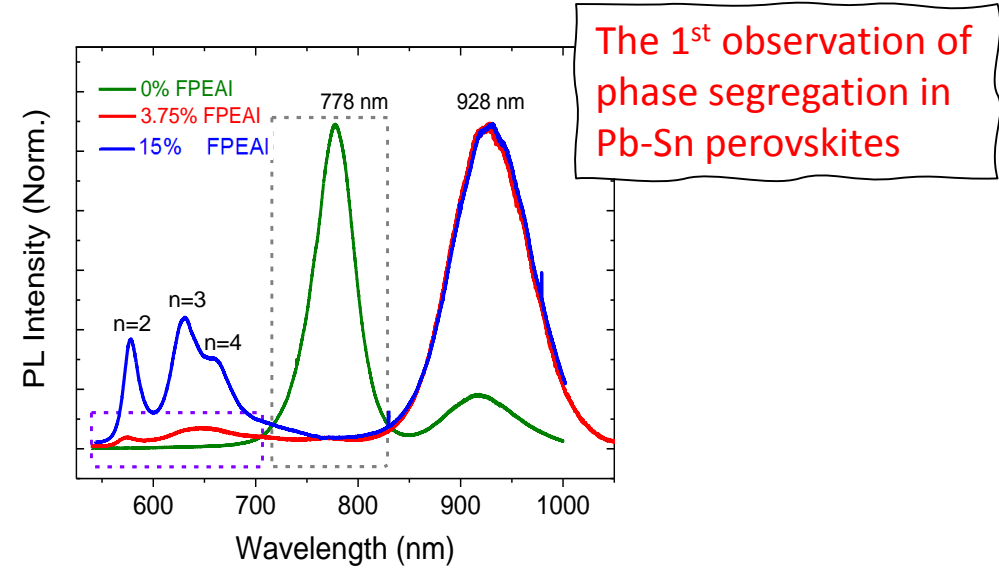
2D/3D Mixed Pb-Sn Perovskites



Random 3D

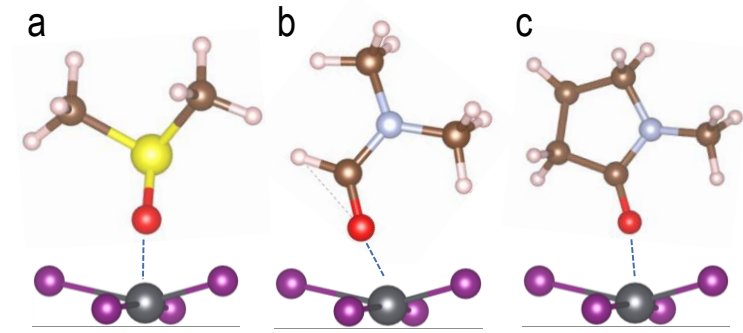
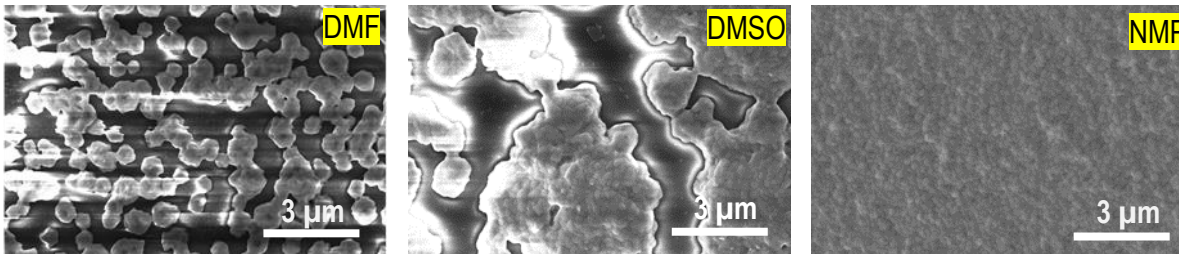
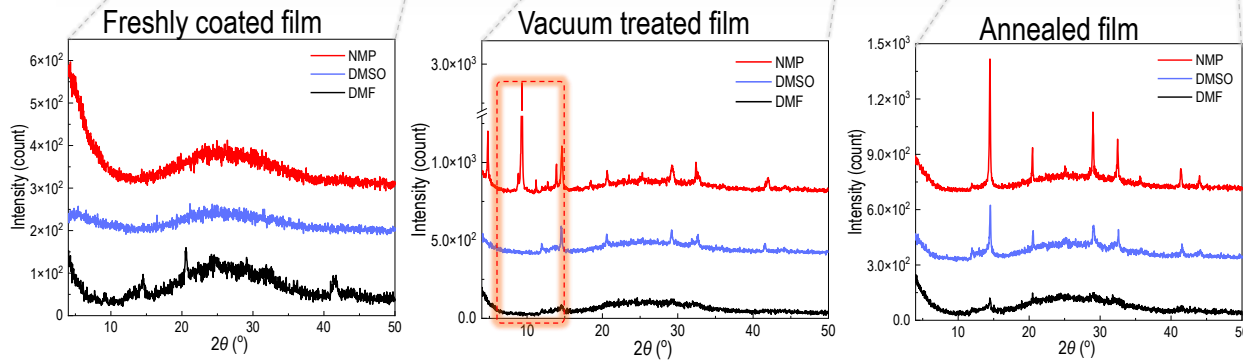
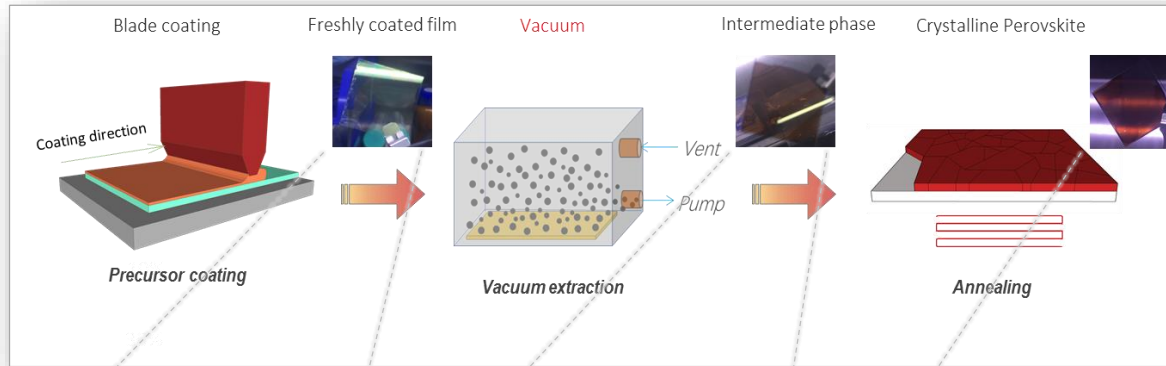


Vertical 2D/3D



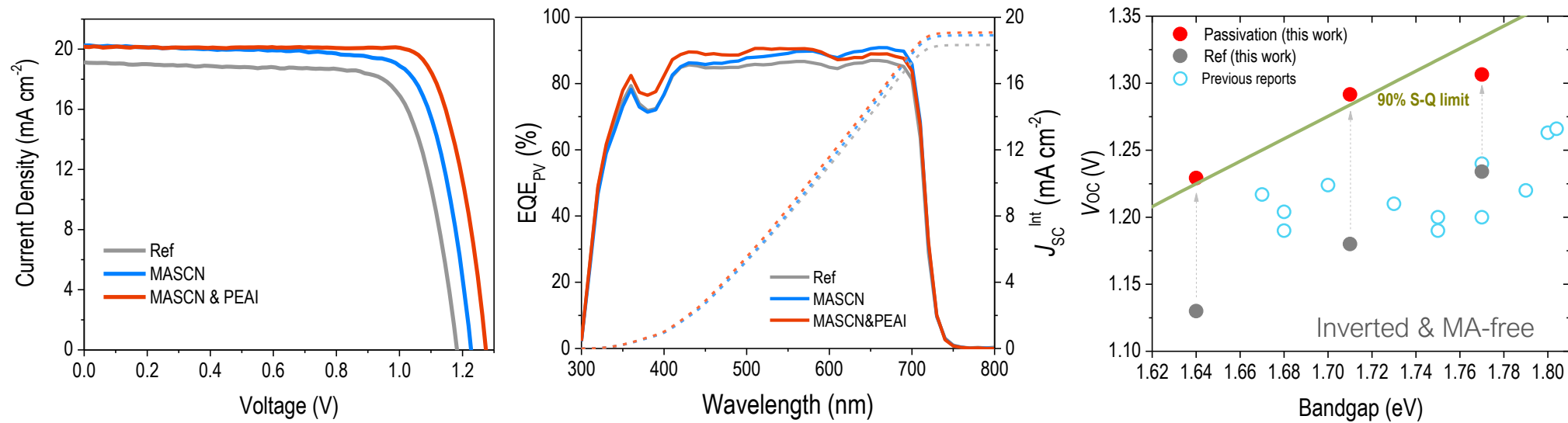
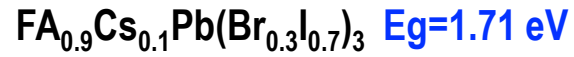
$J_{sc}=28.4$ $V_{oc}=0.79$ $FF=0.78$ $PCE=17.51\%$

Printing Wide-bandgap Perovskites



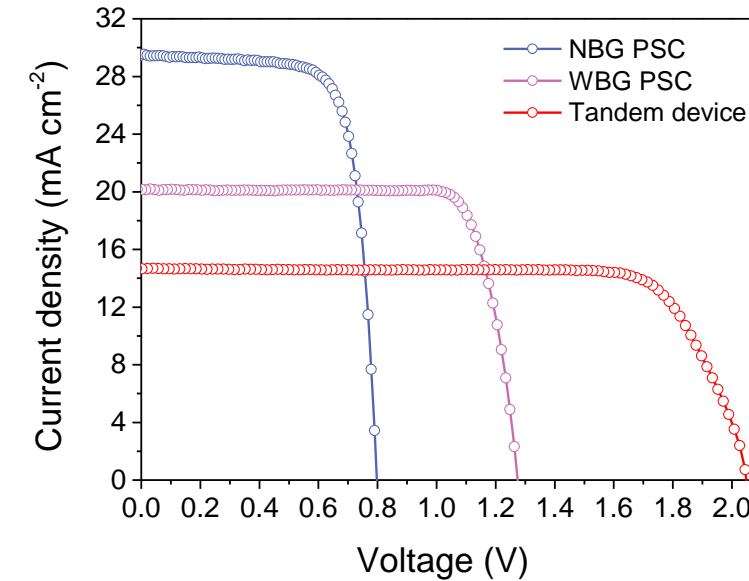
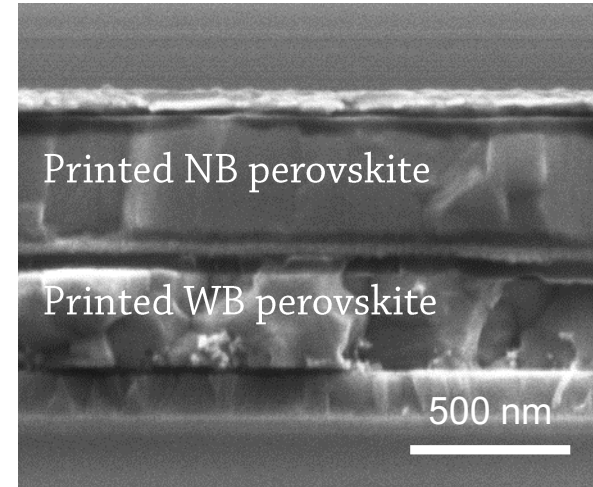
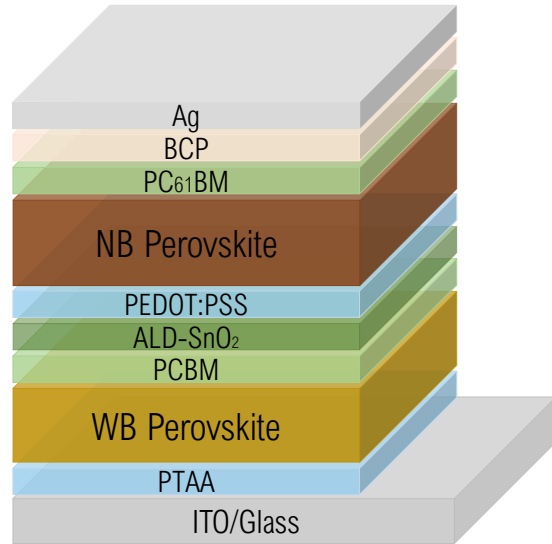
E_{PbI_2}	E_{DMF}	E_{DMSO}	E_{NMP}	E_{Total}	ΔE
-7.735	-68.034	-	-	-76.65	-0.881
-7.735	-	-50.421	-	-59.094	-0.938
-7.735	-	-	-93.98	-102.798	-1.083

The **strongest binding strength** between NMP and PbI_2 , enables to form an **intermediate adduct more stable** than DMSO and DMF, finally giving rise to **controllable crystallization kinetics**.



Wide-bandgap Perovskite	V_{oc} [V]	J_{sc} [mA cm ⁻²]	$J_{sc, EQE}$ [mA cm ⁻²]	FF [%]	PCE [%]
Ref	1.18	19.53	18.34	71.15	16.40
MASCN	1.23	20.24	18.92	76.04	18.99
MASCN&PEAI	1.27	20.32	19.09	81.22	20.84

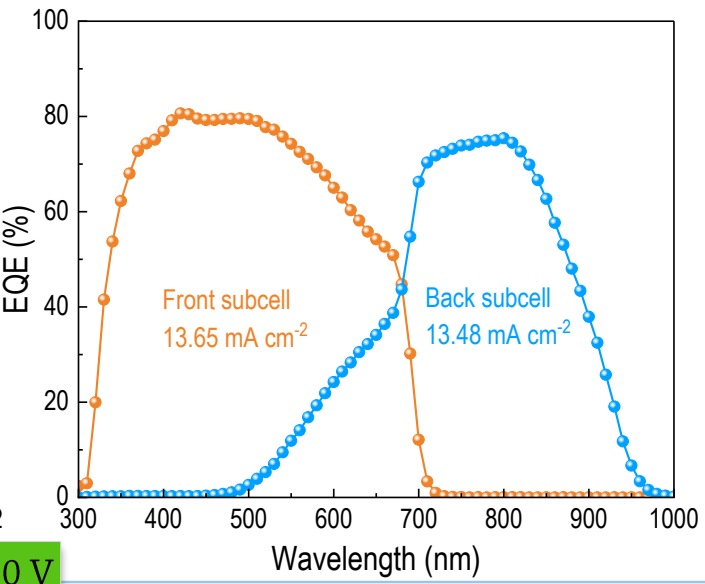
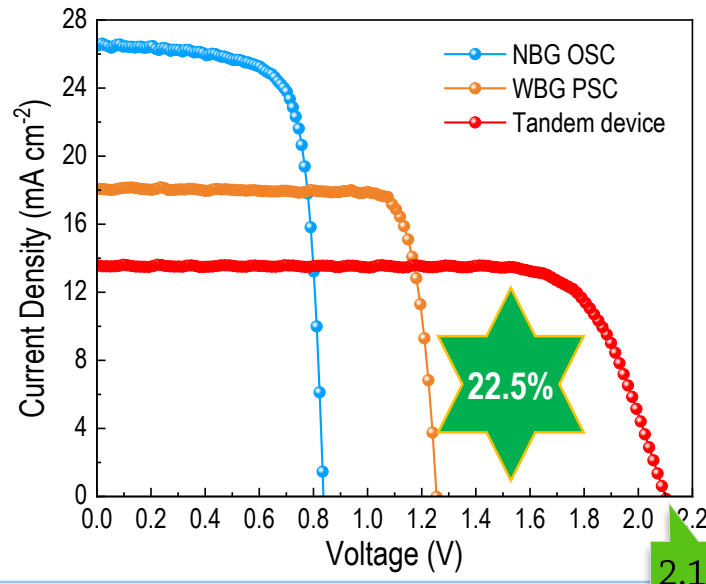
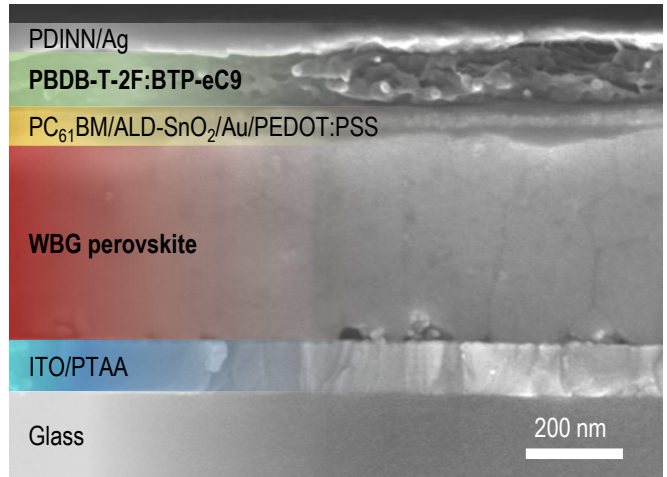
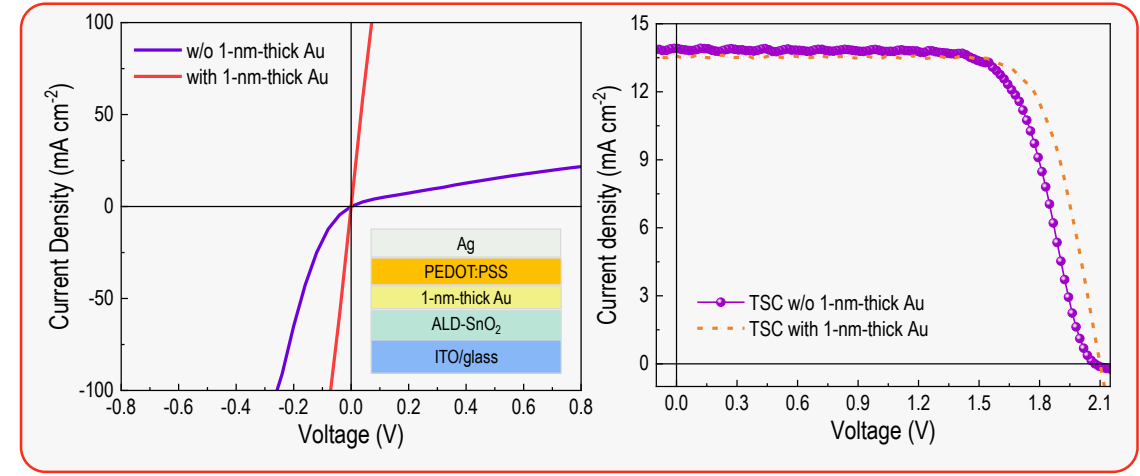
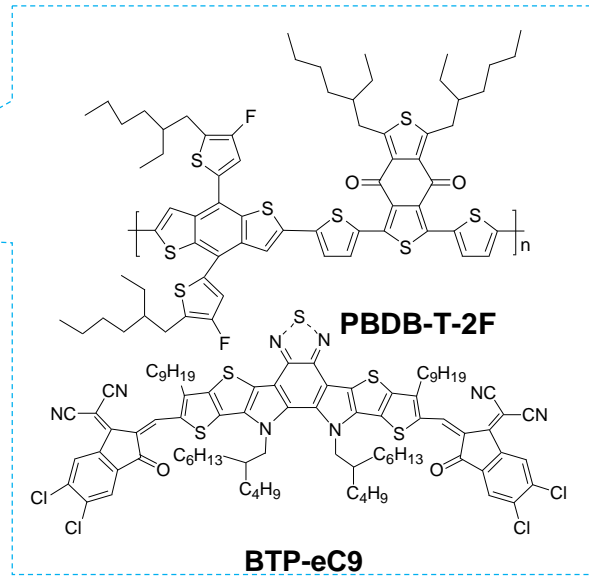
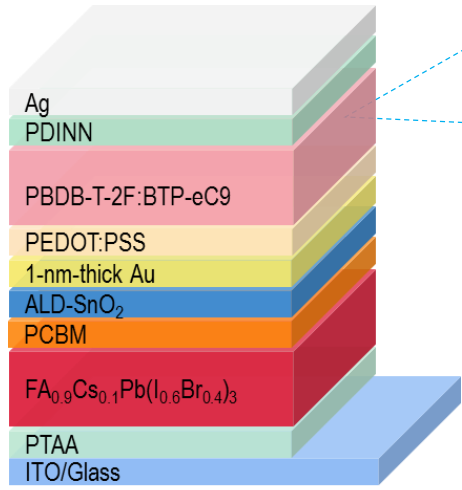
Perovskite - Perovskite



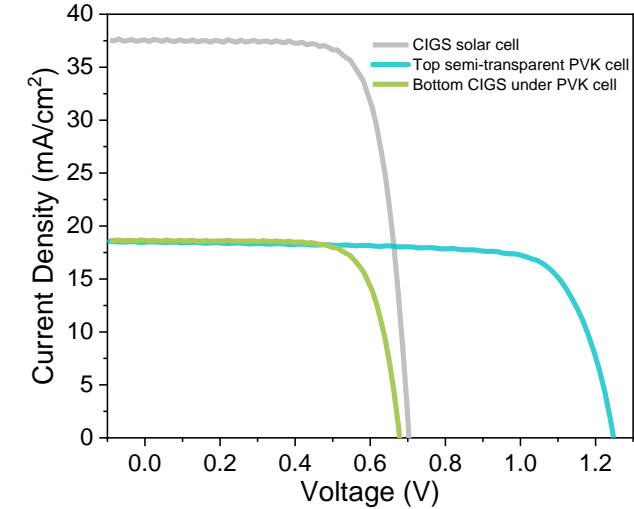
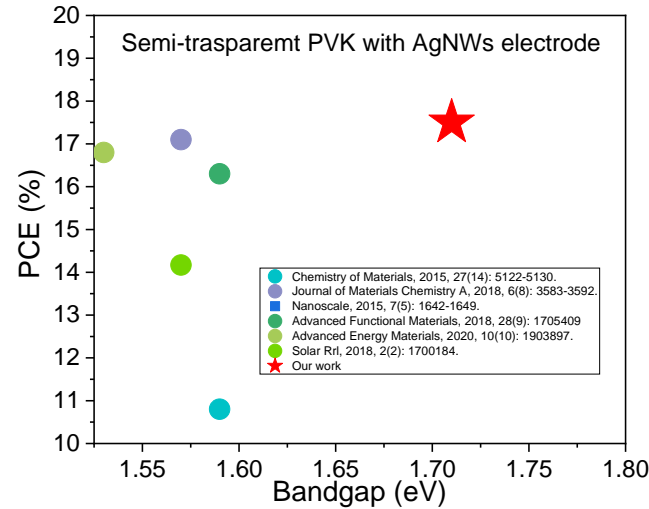
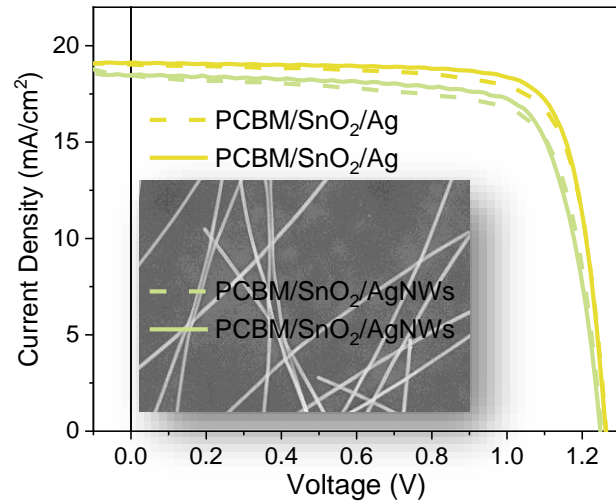
Wide-bandgap Perovskite	V_{OC} [V]	J_{SC} [mA cm^{-2}]	FF [%]	PCE [%]
Wide-bandgap	1.27	20.32	81.22	20.84
Narrow-bandgap	0.80	29.47	75	17.57
All-perovskite tandem	2.05	14.66	78.7	23.65

Printed Perovskite Tandems

Perovskite - Organic



Perovskite - CIGS



	PCE	V _{oc} (V)	I _{sc} (mA/cm ²)	FF
CIGS	19.61	0.702	37.52	74.38
Semi-transparent PVK cell	17.50	1.255	18.44	0.756
Bottom CIGS under PVK cell	9.35	0.678	18.58	74.24
4T tandem solar cell	26.85	-	-	-

报告提纲

- Introduction to scalable crystallization of perovskite films
- Our old / recent results
 - Vacuum quenching / Perovskite-based tandems
- Summary
- Acknowledgement

Summary

- Developed a generic **VACUUM-QUENCHING** crystallization method, which enables to scalable deposit high-quality large-area perovskite thin films of different bandgaps for versatile applications.
- Identified that **NMP is an ideal solvent** in controlling crystallization kinetics of scalable process wide-bandgap perovskite thin films.
- All-perovskite and perovskite-organic tandem devices are fabricated based impermeable **ALD-deposited SnO₂** charge recombination layers.
- **Novel Cr-based** charge recombination layers without ALD procedure are developed to build perovskite-perovskite and perovskite-organic tandem devices.

Acknowledgement

Dean of iNET: Prof. Yaohua Mai

Chief Contributors:

Hongbing Li, Chaoran Chen, Ting Huang, Hongwei Lai, Xinming Zhou, Hongwei Lai, Shudi Qiu, Chaohui Li

Collaborator:

FAU: Dr. Karen Forberich, Prof. Christoph J. Brabec

EPFL: Prof. Mohammad K. Nazeeruddin

Zhengzhou Uni: Prof. Xianhu Liu

Henan Uni: Prof. Shi Chen

Wuhan Uni: Prof. Jianmin Li

**Thank you for your
kind attention**

国家自然科学基金
基金

广东省自然科
学基金

暨南大学启动
经费

i-NET
团队&平台



郑州大学
Zhengzhou University



华南理工大学
South China University of Technology



陕西师范大学
SHAANXI NORMAL UNIVERSITY



南方科技大学