

# Efficient and stabilized molecular doping of hole-transporting materials driven by cyclic-anion strategy for perovskite solar cells



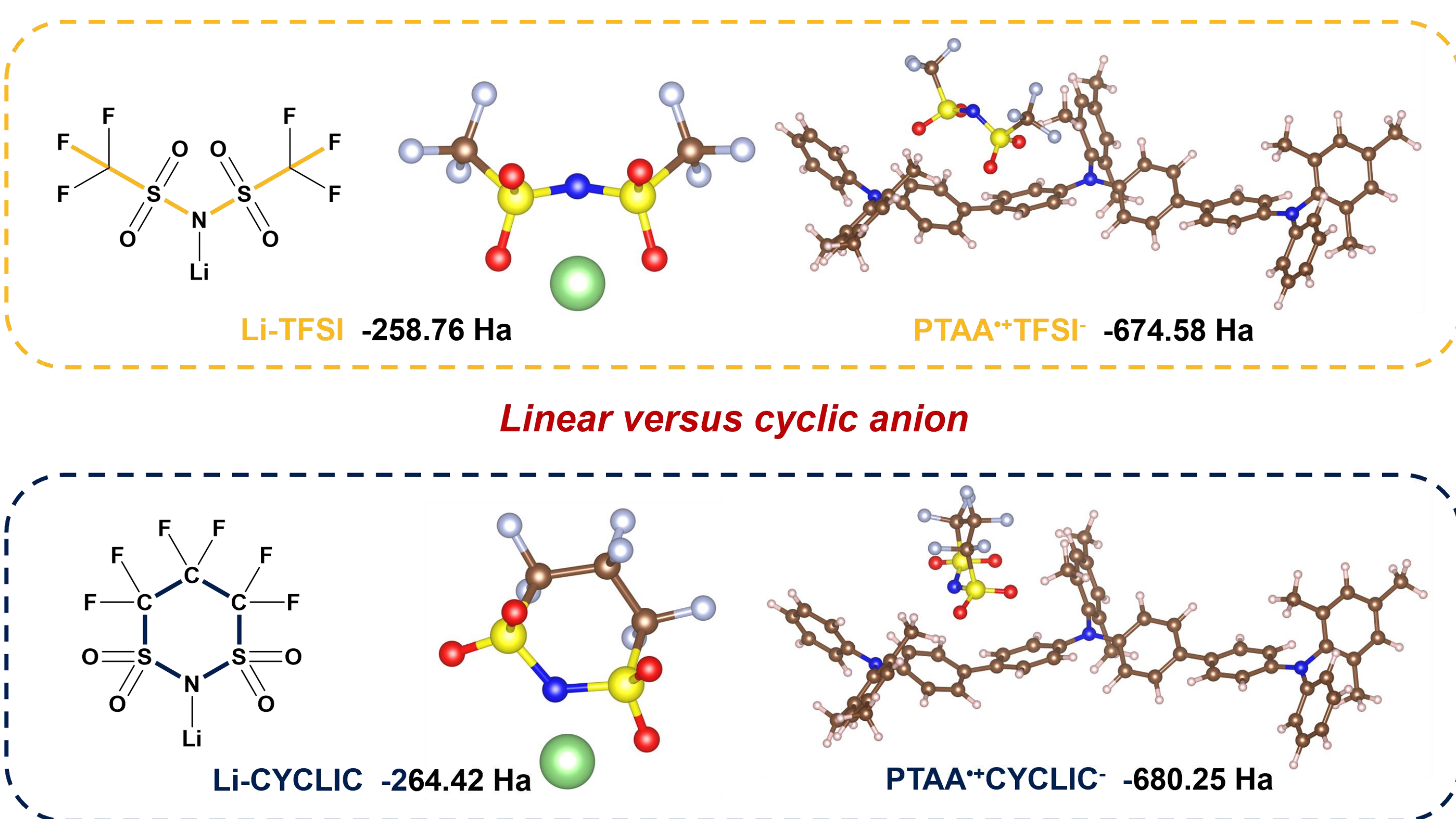
Huaibiao Zeng, Junsheng Luo\*

National Key Laboratory of Electronic Thin Films and Integrated Devices, School of Integrated Circuit Science and Engineering, University of Electronic Science and Technology of China, Chengdu 610054, P. R. China.

## Introduction

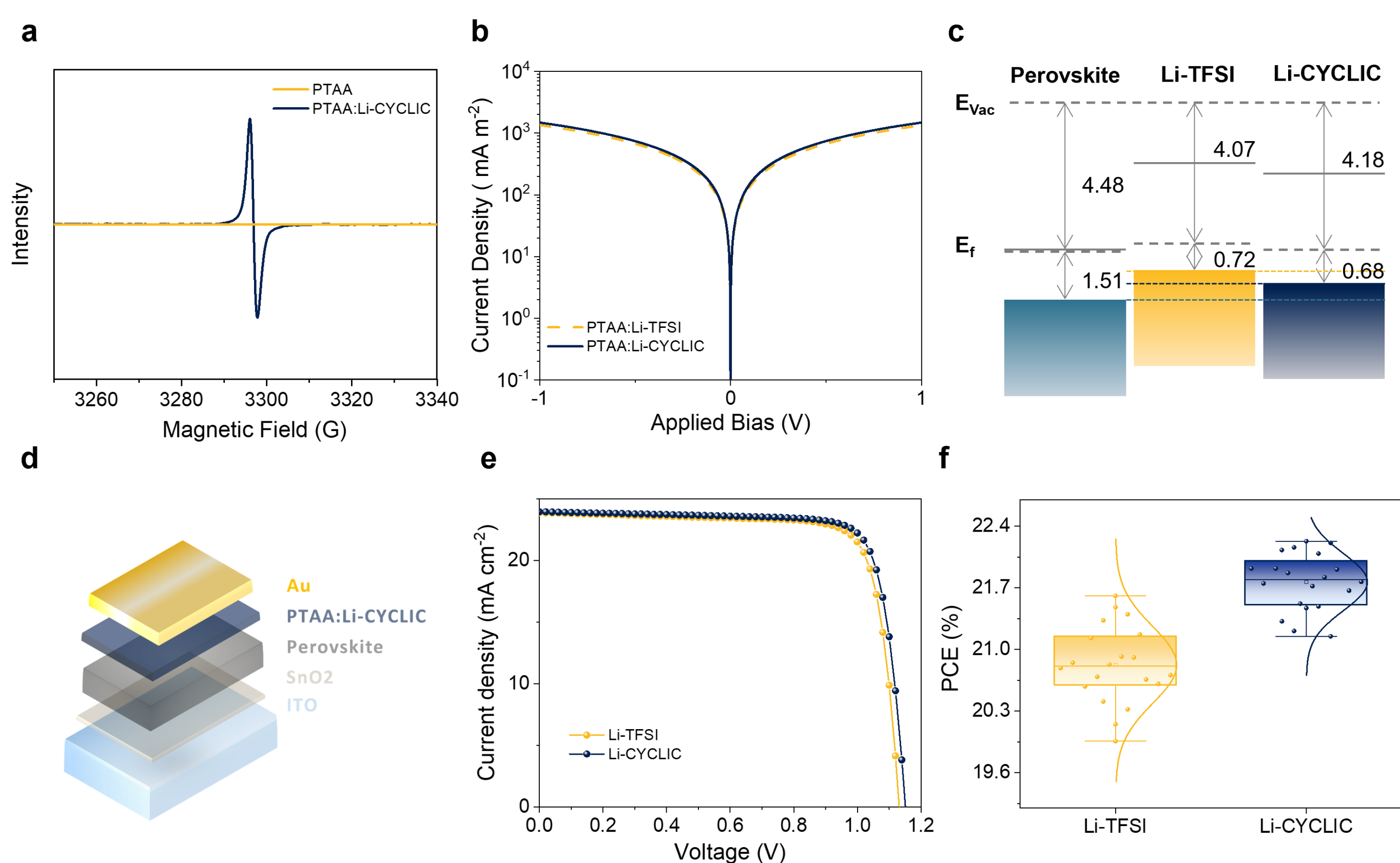
The introduction of Li-TFSI affects the long-term stability of PSCs, which is the most challenging issue limiting their commercialization. Ultra-hygroscopic Li-TFSI tends to absorb moisture when devices are exposed to the atmosphere, thus initiating moisture-induced degradation of the hole-transporting layer (HTL) and underlying perovskite layer. In addition, lithium ions with small van der Waals radius easily migrate from the HTL to the perovskite layer and even the electron-transporting layer (ETL) under the photo-thermal aging process, leading to decreased conductivity of HTM, thereby accelerating degradation of PSCs. We argue that enhancing the interaction between lithium ion and anion unit of dopant is an effective strategy to suppress the moisture absorption and migration of lithium ions, thus enhancing the long-term stability of PSCs.

Herein, a cyclic anion strategy was adopted on the molecular structure of traditional Li-TFSI, and a lithium 1,1,2,2,3,3-hexafluoropropane-1,3-disulfonimide dopant (Li-CYCLIC) is developed as an alternative to the Li-TFSI for PTAA in PSCs. Our design enables the Li-CYCLIC molecule to tackle the challenges of Li<sup>+</sup> hygroscopicity and migration and improve the thermodynamic stability of PTAA<sup>+</sup>CYCLIC<sup>-</sup> system that contributes to enhancing efficiency (22.23%) as well as improving extrinsic-environmental and intrinsic-operational stability of PSCs.



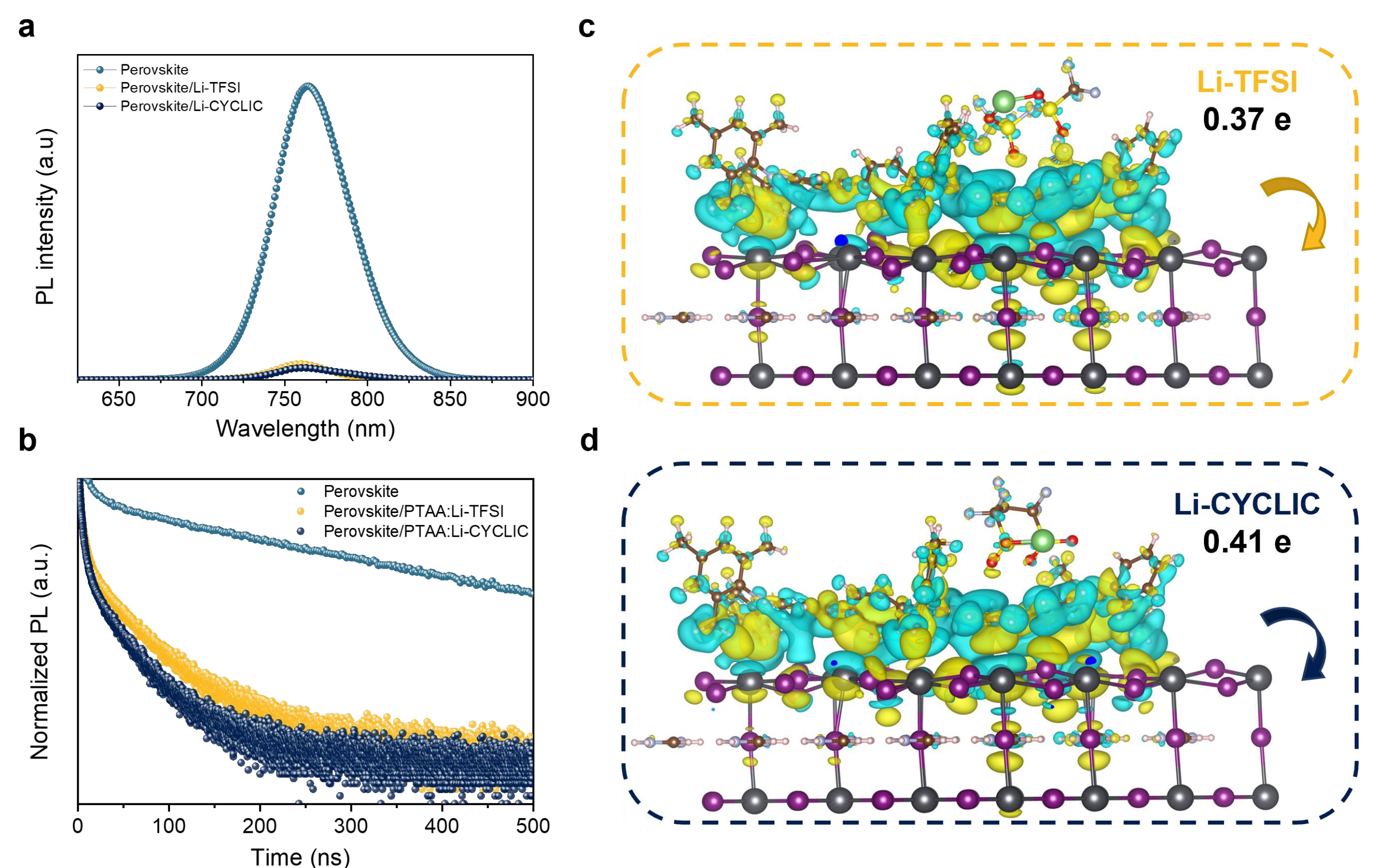
## Results and Discussions

### 1. Doping mechanism and performances



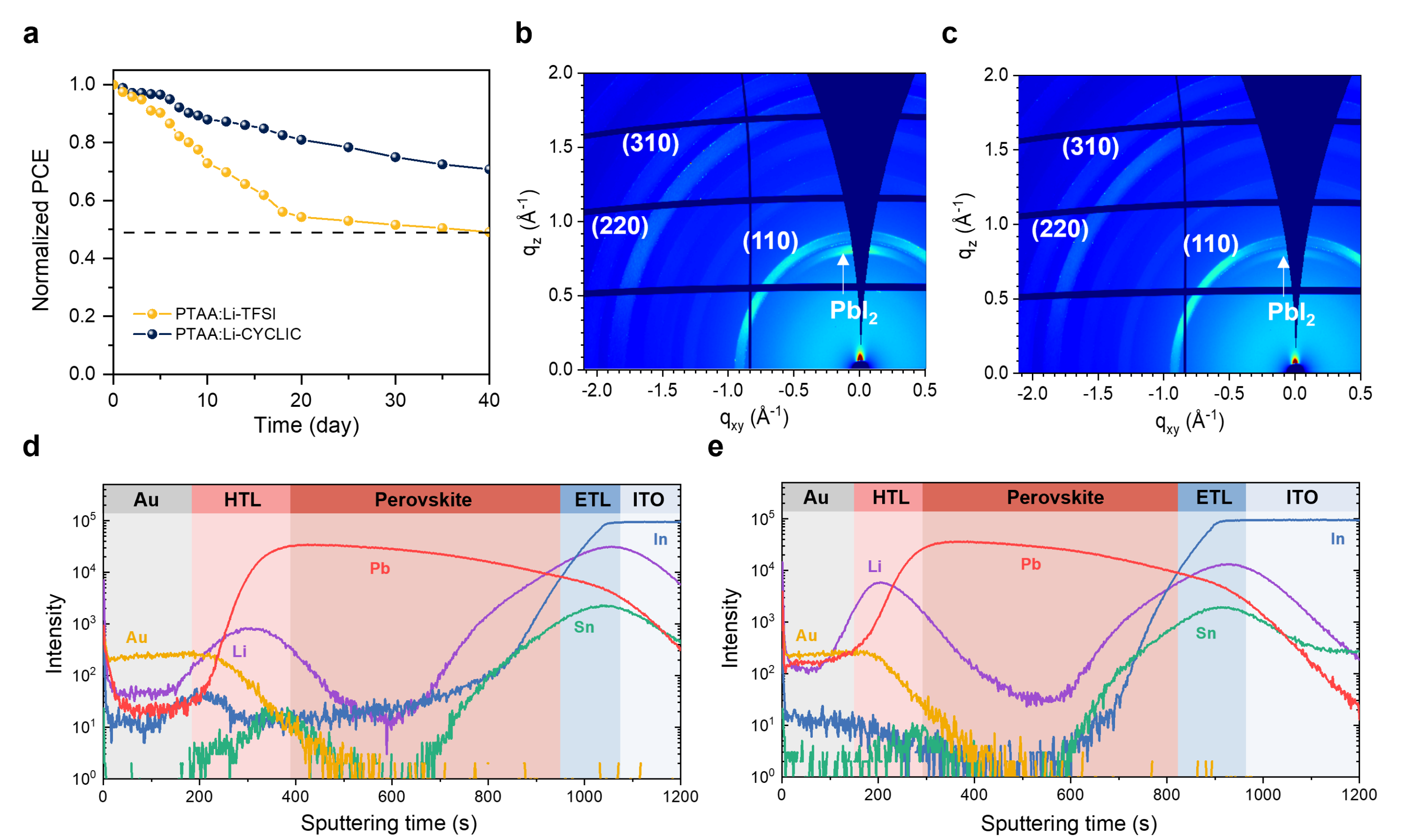
**Figure 1.** (a) ESR spectra based on PTAA without or with Li-CYCLIC. (b) Conductivity of PTAA films based on Li-TFSI and Li-CYCLIC. (c) Energy level alignments of perovskite and PTAA with Li-TFSI or Li-CYCLIC. (d) Structure of the PSCs. (e) Best J-V curves of PSCs based on Li-TFSI and Li-CYCLIC. (f) PCE distribution data for PSCs based on Li-TFSI and Li-CYCLIC.

### 2. Interface charge-transporting properties and defect passivation



**Figure 2.** (a) PL and (b) TRPL of the perovskite films without or with Li-TFSI and Li-CYCLIC doped PTAA. Electron transfer between perovskite and (c) PTAA: Li-TFSI and (d) PTAA: Li-CYCLIC (yellow means gaining electron and cyan means electron deficit).

### 3. Long-term stability



**Figure 3.** (a) Normalized efficiency of Li-TFSI and Li-CYCLIC doped devices in ambient air. 2D-GIXD image of aged device based on (b) Li-TFSI and (c) Li-CYCLIC. TOF-SIMS results of the aged devices based on (d) Li-TFSI and (e) Li-CYCLIC.

## Conclusion

Overall, this work established relationships between the dopant structure and its intrinsic doping and thermodynamics properties, and high-performance PSCs were achieved via cyclic anion strategy, which thereby opens up a unique methodology for discovering new p-type dopants for organic semiconductors and optoelectronic devices.

### References

1. X. Yang *et al.*, *Science*, 2022, 377, 1227-1231.
2. H. Wang *et al.*, *Nano. Energy*, 2020, 69, 104412.
3. H. Zeng, J. Luo *et al.*, *Chem. Sci.*, 2024, SC-EDG-03-2024-002020.R1 (Accepted).