Ruddlesden-Popper-type (RP) quasi-two-dimensional (2D) perovskite solar cells (PSCs) have attracted extensive attention due to their unique photovoltaic properties and excellent device stability. Due to disordered crystallization, the film quality of RP perovskite remains unsatisfactory.

To achieve better photovoltaic performance, various strategies focusing on crystallization regulation have been applied. At present, related research focuses on the photovoltaic properties of perovskite films after crystallization regulation, while the understanding of the crystallization process is lacking.

Recently, Prof. Zhou Huiqiong's group from National Center for Nanoscience and Technology (NCSNT) of the Chinese Academy of Sciences (CAS) has developed a sulfonium cations-assisted intermediate engineering strategy to study the evolution of intermediates and the film properties of quasi-2D perovskites. The study was published online in Advanced Materials.

The researchers developed a facile strategy for intermediate engineering by employing sulfonium cations to regulate the transformation of intermediates during the crystallization process and improve the film quality of quasi-2D perovskites.

The intermediates were characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM) to reveal the composition and transformation process of the intermediates. The introduction of sulfonium cations inhibited the formation of unfavorable solvated lead iodide and promoted the formation of favorable perovskite intermediates with fiber-like morphology, which is conducive to the formation of high-quality perovskite crystals. The above effects have been confirmed in quasi-2D perovskite with different n values and 3D perovskites.

They then carried out in-situ photoluminescence spectroscopy (PL) to study the crystallization during the film formation process. It was found that the introduction of sulfonium cations could not only accelerate but also optimize the crystallization process, leading to high-quality perovskite films.

Based on the above results, regulating the properties of intermediates is an effective way to obtain high-quality perovskite films. With improved film quality, the device achieved a power conversion efficiency of 19.08% at room temperature and 20.52% at low temperature (180 K). In addition, the corresponding devices showed improved operational stability, with 84% of the initial efficiency maintained after 1000 h under maximum power point (MPP) tracking at 40 °C.

"The comprehensive understanding of the evolution of intermediates allows us to clarify the mechanism of the improved film quality via sulfonium cations
treatment, and the findings of this study will benefit the future research on the crystallization regulation of perovskite solar cells," said Prof. Zhou.