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Research Highlight

Epitaxial lift-off for controllable single-crystalline perovskites

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The intriguing attributes of organic–inorganic hybrid perovskites, such as excellent carrier dynamics, outstanding optical tunability, and cost-effective solution processability, have put them on par with or even to be partially superior over the conventional semiconductors, enabling a myriad of applications including solar cells, photodetectors, light-emitting diodes, lasers, and transistors [1–4]. Compared to the polycrystalline perovskites, the single-crystalline ones perform ameliorative performances and enhanced stability due to the eliminative grain boundaries and less defects, will profoundly advance the application potential and are therefore highly desired [5]. Various strategies such as temperature-lowering method, antisolvent diffusion, inverse temperature crystallization, geometry-confinement growth, and vapor-phase epitaxy have been applied to grow the single crystalline perovskites [6–8], however, either the deficient control in geometry, size, and component of the as-synthesized perovskites or the substrate restrictions assisted with these methods leaves the difficulty of integrating perovskites into high-performance devices.

Recently, Prof. Sheng Xu and colleagues [9] demonstrate an innovative approach for fabricating single-crystalline organic–inorganic hybrid perovskite films with precise control in the thickness, area, and composition gradient on arbitrary substrates. In this approach, a bulky single-crystalline perovskite serves as the mother substrates for the epitaxial growth of the single-crystalline perovskite films, and then the films can be peeled off from the mother substrates and transferred to the targeted substrates with the intact crystal quality (Fig. 1a).

A layer of patterned mask made of parylene or polyimide or copper foil is deposited or attached on the surface of the mother substrates prior to the epitaxial growth. The mask thicknesses and patterns along with the solution growth parameters such as temperature, time, precursor concentration and component render the growth of the films in a controllable manner, with the thickness ranging from 600 nm to 100 μm, the area up to 5.5 cm × 5.5 cm, and the composition gradient from $MAPbI_3$ to $MAPb_{0.5}Sn_{0.5}I_3$ in the thickness direction. A high-resolution transmission electron microscope image indicates the epitaxial relationship between the epitaxial films and the mother substrates. In addition, X-ray diffraction and photoluminescence characterizations demonstrate the consistent crystal structure and comparable crystal quality between the epitaxial films and the mother substrates. The epitaxial films are ready to be transferred to the targeted substrates after a combination of in-plane rotation and lift-off with the mask layer as the holder. The in-plane rotation breaks the connection between the epitaxial films and the mother substrates, contributing to the integrity of the detached films. A pretreatment process of the targeted substrates using either diethyl ether or the corresponding supersaturated perovskite growth solution is introduced to enhance the bonding of the transferred films with the targeted substrates. Finally, the mask is removed by the dry etching or direct peeling off, following by a washing process of the transferred perovskite films with the saturated GBL solution to remove the residuals and smooth the surfaces. The scanning electron microscope (SEM) images of a transferred film onto a curved general substrate is presented in Fig. 1b, showing the intactness and grain-boundary-free feature of the transferred films enabled by this epitaxial lift-off approach.

An optimal thickness of ~2 μm of the epitaxial film is identified by both thickness-dependent external quantum efficiency spectra and current–voltage measurements, endowing a balance between maximal optical absorption and efficient charge transport for the photovoltaic devices. In addition, the thin profile of the films permits the mechanical flexibility, allowing a minimum bending radius of ~2.5 mm assisted with the neutral mechanical plane design. The flexible single-crystalline perovskite solar cell, for the first time, is achieved, performing a small performance degradation after 300 cycles of bending measurements under a radius of ~5 mm, as well as a power conversion efficiency (PCE) of 15%–17% with an area of 0.5 cm × 0.5 cm (Fig. 1c). The PCE can be further increased to 17%–19% utilizing the composition engineering, where the single-crystalline films of $MAPb_{0.5+x}Sn_{0.5-x}I_3$ ($0 < x < 0.5$) with the designed gradient are achieved by dynamically exchanging the precursor solutions in a programmable manner during the perovskite growth. The generated grade bandgap within the $MAPb_{0.5+x}Sn_{0.5-x}I_3$ film enables an improved charge carrier dynamics such as the longer carrier lifetime, endowing the solar cell a higher open-circuit voltage than that based on the pure

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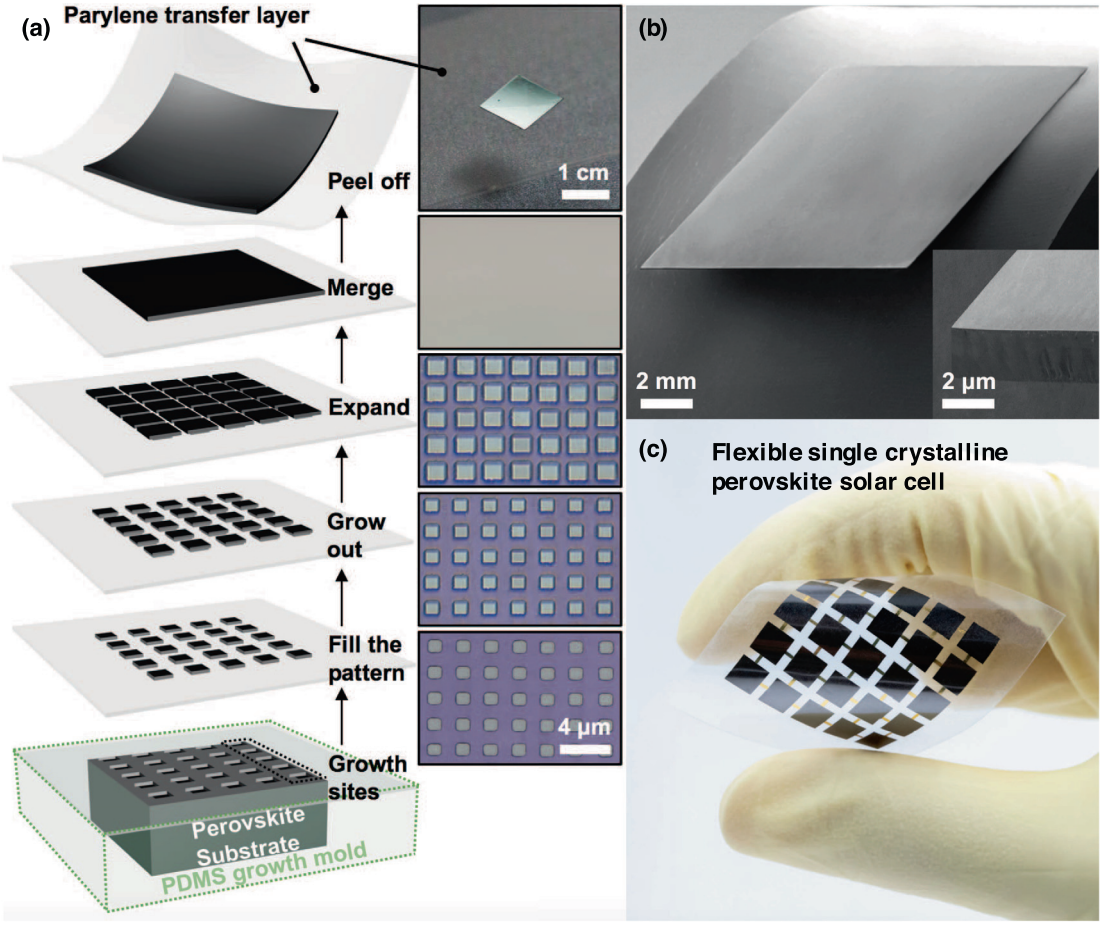

Fig. 1. (Color online) Epitaxial lift-off for the flexible single crystalline perovskite devices. (a) The illustrated process and the corresponding optical images for the flexible single crystalline perovskites. (b) SEM images of a representative flexible single crystalline perovskite film. (c) The optical image of the flexible single crystalline perovskite solar cell [9]. Copyright © 2020, Springer Nature.

$MAPbI_3$. The solar cell also shows much better environmental stability compared with the polycrystalline counterpart due to the absence of grain boundaries.

Challenges remain about the authors' achievements could be the time-consuming and mass-production of this approach. The growth of the mother substrates, in other words, the preparation of the bulky single-crystalline perovskites usually takes a long time, especially for those requiring big size and high quality. And the current fabrication process is complex, needing to be further simplified to improve the yield. This new approach holds great promise in realizing the commercial applications of perovskites after addressing these issues.

In summary, a new protocol has been proposed for fabricating flexible single-crystalline perovskites with controllable dimensions and compositions on arbitrary substrates. The concept of this protocol is expected to extend to other emerging materials to boost the relative applications.

Conflict of interest

The authors declare that they have no conflict of interest.

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