JUSTC
Research Highlight
http://justc.ustc.edu.cn
Received: September 27, 2022; Accepted: October 20, 2022

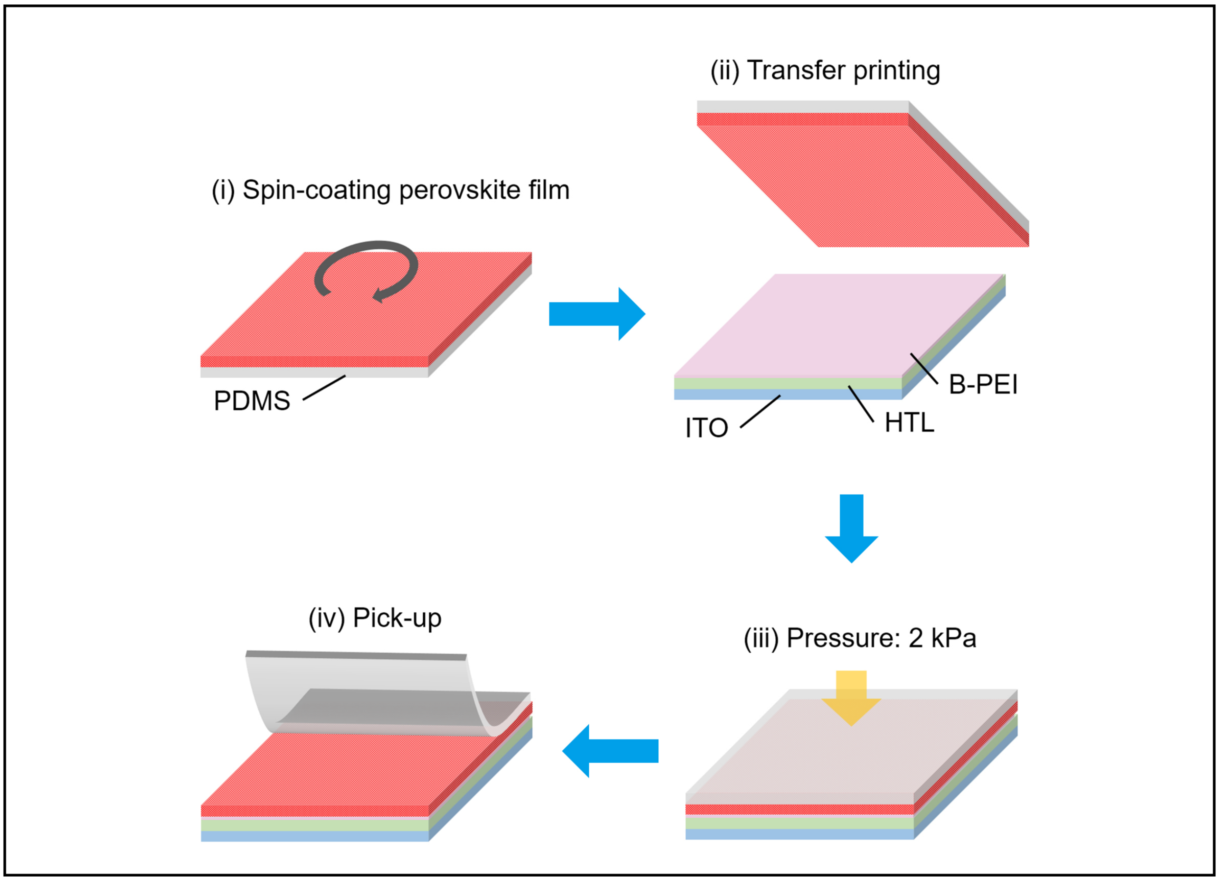
Robust transfer-printing method for perovskite films and nano-structures

Peiyuan Pang, and Guichuan Xing ✉

Joint Key Laboratory of the Ministry of Education, Institute of Applied Physics and Materials Engineering, University of Macau, Macau 999078, China

✉Correspondence: Guichuan Xing, E-mail: gcxing@um.edu.mo

Graphical abstract



Schematic illustration of the modified transfer printing process.

Public summary

■ This research highlight summarizes a robust mass transfer printing method for perovskite films fabrication with nano-structures reported by Xiao and colleagues.

■ This transfer printing method enables the fabrication of large-area perovskite nanostructures with high resolution.

■ Using this method, white PeLEDs with red and sky-blue emission perovskite micro-stripes has been achieved.

Citation: Pang P Y, Xing G C. Robust transfer-printing method for perovskite films and nanostructures. *JUSTC*, **2022**, 52(11): 1. DOI: 10.52396/JUSTC-2022-0139

JUSTC

Research Highlight

http://justc.ustc.edu.cn

Received: September 27, 2022; Accepted: October 20, 2022

Robust transfer-printing method for perovskite films and nanostructures

Peiyuan Pang, and Guichuan Xing ✉

Joint Key Laboratory of the Ministry of Education, Institute of Applied Physics and Materials Engineering, University of Macau, Macau 999078, China

✉Correspondence: Guichuan Xing, E-mail: gcxing@um.edu.mo

Cite This: *JUSTC*, **2022**, 52(11): 1 (3pp)   **Read Online**

CLC number: TB383; TN312⁺.8   **Document code:** A

Metal halide perovskites, as a promising semiconductor material, have been successfully used in electroluminescent devices because of their desirable characteristics, such as good conductivity, high color purity, tunable bandgap, low cost and solution process ability. In the past few years, significant progress has been made in the development of high-efficiency perovskite light-emitting diodes (PeLEDs). These efficient PeLEDs are mainly achieved by sophisticated spin-coating methods, which can easily control the perovskite's composition, film thickness, morphology and crystallinity. However, with the continuous development of PeLEDs, commercial production problems have to be solved, such as large-area production, high-resolution patterning and substrate diversity, which are difficult for the current spin-coating process.

In addition to the commonly used spin-coating method, several alternative methods, such as photolithography[1,2], inkjet printing[3,4], thermal evaporation[5,6] and transfer printing[7,8], have been developed. For now, however, those methods still have issues that limit their application. For example, for the photolithography technique, the morphology and optoelectronic properties of the perovskite film were easily damaged during the developing or etching processes[2]. The inkjet printing method also has many challenges, such as low pattern resolution and the coffee ring effect in the film forming process[4]. The explorations of thermal evaporation have been very limited due to the large vapor pressure difference between organic and inorganic components[5,6]. Recently, Zhengguo Xiao and colleagues reported a robust mass transfer printing method for perovskite film fabrication with nanostructures[9]. The PeLEDs fabricated by such a method show external quantum efficiencies (EQEs) similar to those fabricated by spin-coating. This robust transfer printing method also enables the fabrication of large-area perovskite nanostructures with a high resolution up to 1270 pixels per inch (ppi). Based on this technology, white PeLEDs can be fabricated by laterally aligning red- and sky-blue-emitting perovskite microstripes.

The common transfer printing method has the advantages of low cost and high pattern resolution. However, it requires a high pressure up to hundreds of kPa when picking up the films by using poly(dimethylsiloxane) (PDMS) with nanostructures[10]. In addition, when the film thickness is lower than 20 nm or higher than 200 nm, the pick-up yield seriously decreases[11]. Inserting a soluble polymer layer as a sacrificial layer is a feasible strategy to improve the pick-up yield; however, the dissolving process may cause damage to the film.

Xiao and colleagues first spin-coated perovskite films onto a PDSM substrate instead of using PDSM as transfer media, which significantly improved the integrity of the perovskite films. To successfully transfer-print the perovskite films, the separation energy between the perovskite/PDMS interface, $G^{\text{Perovskite/PDMS}}(v) = G_0^{\text{Perovskite/PDMS}}[1+\varnothing(v)]$, should be less than that between the perovskite/substrate interface $G^{\text{Perovskite/Substrate}}$, where $\varnothing$ is an increasing function of velocity[12]. Due to the extremely low surface energy of PDMS (19.8 $\text{mJ}\cdot\text{m}^{-2}$) compared to most other organic films and glass (> 200 $\text{mJ}\cdot\text{m}^{-2}$)[13] and the gel state of the perovskite films[14], the $G_0^{\text{Perovskite/Substrate}}$ value is much higher than $G_0^{\text{Perovskite/PDMS}}$. Therefore, a small pressure of only approximately 2 kPa is needed, and the peeling speed can reach 1 $\text{cm}\cdot\text{s}^{-1}$, one order of magnitude higher than the reported values for the transfer printing process. An ultrathin amine functional polymer B-PEI is spin-coated on the receiver substrate to enhance the contact with perovskite and passivate the interface (Fig. 1).

Using the modified transfer-print method, red and sky-blue PeLEDs are fabricated. The transfer-printed PeLEDs show the highest EQEs of 10.5% (red) and 6.7% (sky-blue), which are comparable with those fabricated by the spin-coating method. Moreover, the transfer-printed perovskite films show almost the same morphology, chemical composition and optoelectronic properties as the spin-coated films, consistent with the similar performances of PeLEDs fabricated by both types of methods.

Combined with the intaglio method, the modified transfer-print method can transfer not only whole films but also perovskite nanostructures. First, the PDMS substrate with a perovskite film is pressed on a silicon wafer with concave patterns and then peeled off. The remaining perovskite film on the PDMS is transferred to the target substrate to achieve perovskite patterns. The perovskite patterns have very sharp

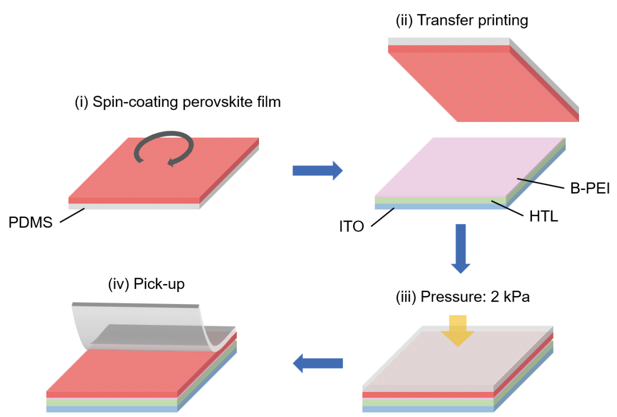


Fig. 1. Schematic illustration of the mass robust transfer printing process.

edges and uniform strong photoluminescence (PL) emission, which benefits from the high transfer yield approaching 100%. In addition, the resolution of the perovskite patterns can reach 1270 ppi.

High-resolution perovskite patterns make it possible to fabricate pixelated white PeLEDs. The emission layer of white PeLEDs can be obtained by horizontally alternating red and sky-blue perovskite patterns. The device structures of red and sky-blue PeLEDs are ITO/Poly-TPD/B-PEI/red perovskite/PMMA/TPBi/LiF/Al, and ITO/Poly-TPD/PVK/sky-blue perovskite/TPBi/LiF/Al, respectively. Xiao and colleagues easily prepared perovskite films over 1.5 cm × 1.5 cm with a microstrip width of 100 μm. Since the current density of sky-blue PeLEDs is lower than that of red PeLEDs at constant voltage, leading to lower brightness and EQE, to ensure white light emission, the microstrip width of blue PeLEDs is adjusted to 200 microns, while the microstrip width of red PeLEDs is kept to 100 microns.

Since the band gap of red perovskite is narrower than that of sky-blue perovskite, the turn-on voltage of red PeLEDs is lower. By increasing the PMMA layer thickness in the red PeLEDs, the turn-on voltage can be easily increased; however, the EQE decreases significantly. The optimized white PeLEDs show higher EQEs of 0.75% at approximately 4 V. The relatively low EQE value should result from the leakage current from the gaps between microstrips and the performance loss of the red PeLEDs. Fabricating heterogeneous multitasked structures is one of the solutions[11]. Changing the parallel microstrips into a multitasked structure can effectively avoid the leakage current. However, the multitasked structure also needs to solve the problem of band gap matching between different luminescent materials to avoid single photoluminescence resulting from carrier transfer.

To promote the commercial production of PeLEDs, we need to prepare uniform thin films with large areas, high-resolution patterning and great tolerance to different substrates, which are currently difficult to achieve by spin-coating processes. As a low-cost and efficient method, the transfer printing method has great potential in realizing the commercial production of PeLEDs. A key feature of the transfer printing method is that it makes the selection of materials for perovskite device structure design more extensive, allowing the use of substrate materials that do not match the growth of perovskite in the spin-coating process to achieve more demanding applications[7]. Another key feature is that this method can achieve high-resolution patterning of perovskite materials, which makes it possible to fabricate pixelated devices.

In this transfer-printing method reported by Xiao and colleagues, the original films are first spin-coated on PDMS substrates, which still cannot avoid the spin-coating process in device fabrication. Because the original films should be uniform, large-area and high-quality films prepared on PDMS substrates, other methods suitable for large-area preparation can be used, such as blade coating, slot-die coating, spray coating, inkjet printing and roll-to-roll printing technology. After obtaining large-area original films, high-resolution perovskite patterns can be prepared by combining the transfer-printing and intaglio methods. The proposed transfer-printing method can complement and adapt to the existing large-area film forming process, and create the possibility for the commercial production of large-area and high-resolution patterned perovskite devices.

To summarize, Xiao and colleagues improved the traditional PDSM-based transfer printing technology and proposed a robust transfer printing method with a transfer yield close to 100%. A small pressure of only approximately 2 kPa is needed, and the peeling speed can reach 1 $cm \cdot s^{-1}$, one order of magnitude higher than the reported values for the transfer printing process. The morphology, composition and performance of the transferred perovskite films are almost the same as those of the original films. This robust transfer printing method also enables the fabrication of white PeLEDs with micropatterns. This achievement not only plays an important role in promoting the realization of PeLED industrial production, but also provides a new reference for the preparation of other luminescent and photovoltaic devices.

Acknowledgements

This work was supported by the Science and Technology Development Fund, Macao SAR (FDCT-0044/2020/A1, 0082/2021/A2), UM's research fund (MYRG2020-00151-IAPME), the National Natural Science Foundation of China (61935017, 62175268) and Shenzhen-Hong Kong-Macao Science and Technology Innovation Project (Category C) (SGDX2020110309360100).

Conflict of interest

The authors declare that they have no conflict of interest.

Biographies

Peiyuan Pang received his master's degree from the South China University of Technology in 2021. He is currently a doctoral candidate at the Institute of Applied Physics and Materials Engineering, University of Macau. His research mainly focuses on perovskite electroluminescent devices.

Guichuan Xing received his Ph.D. degree in Physics from the National University of Singapore in 2011. He is currently a Professor at the Institute of Applied Physics and Materials Engineering, University of Macau. His major research interests include ultrafast laser spectroscopy, nano optoelectronics, and perovskite for light harvesting and light emission.

DOI: 10.52396/JUSTC-2022-0139
JUSTC, **2022**, 52(11): 1

References

[1] Xia B, Tu M, Pradhan B, et al. Flexible metal halide perovskite photodetector arrays via photolithography and dry lift-off patterning. *Advanced Engineering Materials,* **2022**, *24*: 2100930.

[2] Zou C, Chang C, Sun D, et al. Photolithographic patterning of perovskite thin films for multicolor display applications. *Nano Letters,* **2020**, *20*: 3710–3717.

[3] Wei C, Su W, Li J, et al. A universal ternary-solvent-ink strategy toward efficient inkjet-printed perovskite quantum dot light-emitting diodes. *Advanced Materials,* **2022**, *34*: 2107798.

[4] Minemawari H, Yamada T, Matsui H, et al. Inkjet printing of single-crystal films. *Nature,* **2011**, *475*: 364–367.

[5] Du P, Li J, Wang L, et al. Efficient and large-area all vacuum-deposited perovskite light-emitting diodes via spatial confinement. *Nature Communications,* **2021**, *12*: 4751.

[6] Ávila J, Momblona C, Boix P P, et al. Vapor-deposited perovskites: The route to high-performance solar cell production. *Joule,* **2017**, *1*: 431–442.

[7] Carlson A, Bowen A M, Huang Y, et al. Transfer printing techniques for materials assembly and micro/nanodevice fabrication. *Advanced Materials,* **2012**, *24*: 5284–5318.

[8] Linghu C, Zhang S, Wang C, et al. Transfer printing techniques for flexible and stretchable inorganic electronics. *npj Flexible Electronics,* **2018**, *2*: 26.

[9] Li Z, Chu S, Zhang Y, et al. Mass transfer printing of metal-halide perovskite films and nanostructures. *Advanced Materials,* **2022**, *34*: 2203529.

[10] Kim T H, Cho K S, Lee E K, et al. Full-colour quantum dot displays fabricated by transfer printing. *Nature Photonics,* **2011**, *5*: 176–182.

[11] Kim T H, Chung D Y, Ku J, et al. Heterogeneous stacking of nanodot monolayers by dry pick-and-place transfer and its applications in quantum dot light-emitting diodes. *Nature Communications,* **2013**, *4*: 2637.

[12] Meitl M A, Zhu Z T, Kumar V, et al. Transfer printing by kinetic control of adhesion to an elastomeric stamp. *Nature Materials,* **2006**, *5*: 33–38.

[13] Choi M K, Yang J, Kang K, et al. Wearable red–green–blue quantum dot light-emitting diode array using high-resolution intaglio transfer printing. *Nature Communications,* **2015**, *6*: 7149.

[14] Jeong J W, Yang S R, Hur Y H, et al. High-resolution nanotransfer printing applicable to diverse surfaces via interface-targeted adhesion switching. *Nature Communications,* **2014**, *5*: 5387.

DOI: 10.52396/JUSTC-2022-0139
JUSTC, **2022**, 52(11): 1