A Dry Lift-off Method for Patterning Perovskites

Cheng Chang, Chen Zhou, Mark Odendahl and Lih Y. Lin
Department of Electrical and Computer Engineering, University of Washington, Seattle, WA 98195, USA
Author e-mail addresses: cchang36@uw.edu, chenzou@uw.edu, merko@uw.edu, lylin@uw.edu

Abstract: In this paper, we demonstrate a new method to pattern perovskites using a dry lift-off process. By utilizing parylene-C as a sacrificial layer, patterns with <12 um features and multi-color patterns can be achieved.

1. Introduction

In recent years, organometal halide perovskites (OHP) have aroused great interests for optoelectronics applications due to their excellent optical, electrical and chemical properties. However, perovskites dissolve in most polar solvents, like water and lithographic developers, which makes perovskites not compatible with widely-used photolithographic processes which are necessary for device fabrications [1]. To overcome this limit, orthogonal lithography has been proposed to pattern perovskites [2]. In this method, fluorinated polymer resist, which is orthogonal to perovskites quantum dots (both the resist and its developer will not react with perovskite quantum dots), is used as a sacrificial layer. The polymer is then lift off using a fluorinated solvent that does not dissolve the perovskite layer. However, this method applies well to perovskite quantum dots but not quasi-2D or 3D perovskites. This is due to the wettability difference between solvents for quantum dots (surface tension for heptane is 19.3) and quasi-2D/3D perovskites (surface tension for DMSO is 43.7). In this paper, we report a new method utilizing parylene-C and a dry lift-off process [3]. The method is compatible with quantum dot, 2D, quasi-2D and 3D perovskites. The perovskite materials are isolated from the solvent during the fabrication process, thus their properties and performance will not be affected. In our prior work utilizing parylene-C as part of micro-fluidic channels, we achieved parylene-C patterns with <10 um resolution [4]. By repeating the process, different perovskite materials can be patterned on the same substrate, and we demonstrate the utility of the approach through patterning a dual-color perovskite pixel array. The method can be applied to the fabrication of perovskite micro-LED arrays.

2. Fabrication Process

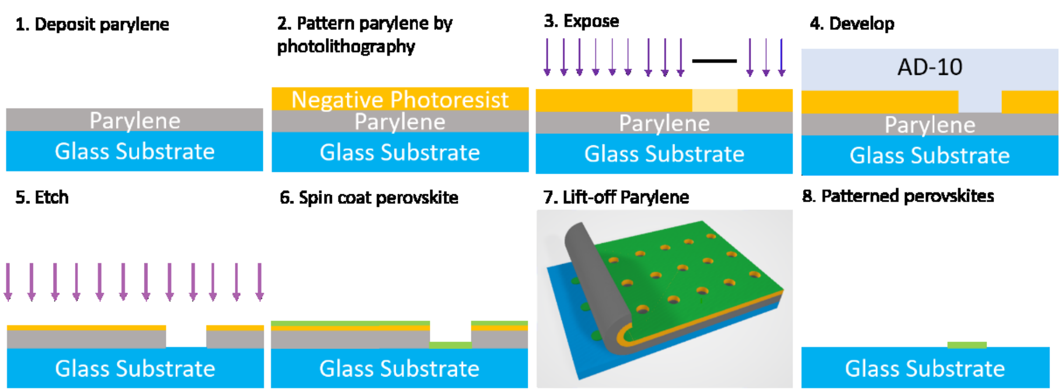

Fig. 1. Fabrication process of the proposed method to achieve high-resolution patterning of perovskites. The process utilizes patterning parylene-C which is lift off later to achieve perovskite patterns, avoiding any solvents contacting the perovskite layer.

The fabrication process of the proposed method is shown in Fig. 1. A layer of parylene-C is first deposited on the substrate. A glass substrate is used as an example and in our proof-of-concept demonstration. But this method is compatible with other substrates as well. Photolithography is used to pattern the parylene-C. Negative photoresist is used for the illustration. Afterwards, a parylene-C layer is etched through to the glass substrate. A thin layer (30 nm) of perovskite is spun cast on the surface. Since parylene-C is hydrophobic with high structural integrity and strength, it's easy to peel off the entire layer through mechanical means, as illustrated in Step 7. During this lift-off process, the perovskite layer does not come into contact with any chemicals or solvents, so their electrical and optical properties will be well preserved.

In addition to single layer patterns, this dry lift-off method also works for patterning of multi-color perovskites by simply repeat the fabrication process. After the first perovskite layer is patterned on the substrate, another layer of parylene-C can be directly deposited on the patterned perovskites. We have verified that parylene-C does not interact with perovskites. The existing perovskite patterns are therefore covered and protected by the parylene-C during the subsequent photolithography processes.

3. Fabrication and Characterization Results

The resolution of this dry lift-off method is only limited by the resolution of photolithography on the parylene-C layer, so the process can potentially achieve very small patterns. In Fig. 2(a), we demonstrate an array of quasi-2D perovskite dot patterns with a diameter of <12 um using the proposed method. As discussed earlier, it's straightforward to apply this method to pattern multi-color perovskite layers. Here we demonstrate a dual-color pixel array with green-emitting ($CsPbBr_3$+PEABr) and red-emitting ($CsPbI_3$+PEABr) perovskites. The materials were synthesized following a protocol similar to what's reported in [5]. Figure 2(b) to (e) show the dual-color patterns under a fluorescence microscope, imaged with bright field (Fig. 2(b)), fluorescence mode with green and red emission filter (Fig. 2(c)), green-only filter (Fig. 2(d)) and red-only filter (Fig. 2(e)). The incomplete green perovskite patterns resulted from misalignment during the 2nd parylene-C photolithography process which partially expose the green perovskite layer underneath. This can be mitigated by designing the patterns and photomasks properly. In order to determine if the lift-off process has any negative effects on the perovskites, we compare the photoluminescence (PL) of the materials before and after patterning. Figure 2(f) shows that the linewidth and the intensity of the emission peaks are mostly unaffected by the patterning process.

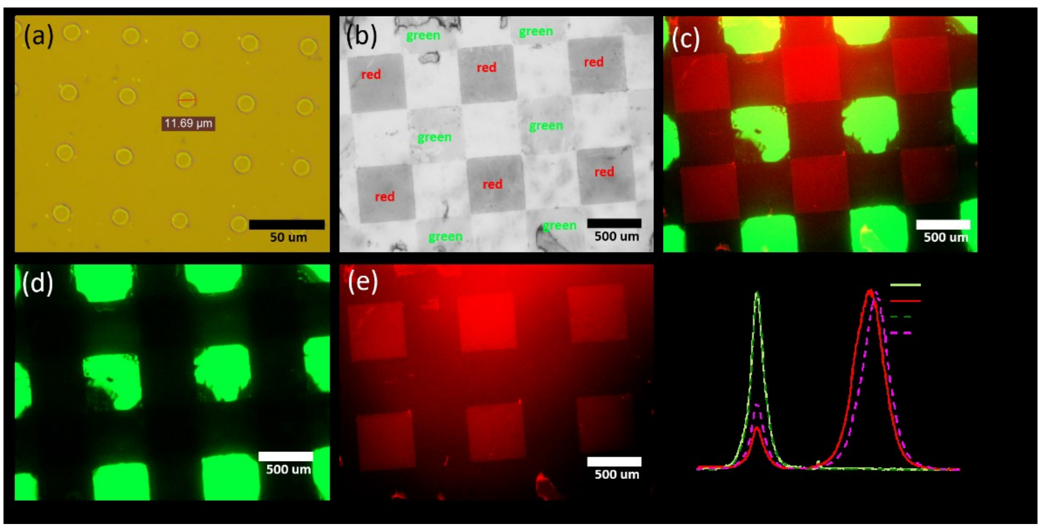

Fig. 2. Patterned perovskites and PL spectra. (a) An array of perovskite dots. (b) Bright-field and (c)-(e) fluorescence images of a dual-color perovskite pixel array. (f) PL spectra before and after patterning.

4. Conclusion and Future Work

In summary, we developed a dry lift-off fabrication method to pattern perovskites. With this method, high-resolution as well as multi-color perovskite patterns can be achieved. The quality of the perovskites is not compromised by the process, and further optimization of the method can improve the fabrication results. Besides simple patterns, more complicated devices such as micro-LED arrays and photodetectors are also feasible using a similar process.

5. References

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