## Lead Halide Perovskite Pixel Arrays Fabricated by Ultrathin Reusable Metal Mask

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Lead halide perovskites are a class of materials that have attracted significant attention due to their exceptional optoelectronic properties. However, one of the challenges in working with perovskite arrays is achieving high-resolution patterning, as they are inherently unstable when exposed to conventional photolithography solvents. In this study, we propose a novel patterning process for perovskite arrays using a high-resolution, large-scale metal mask and spin-coating.

Our approach involves using a reusable micrometer-thick lithographically-defined metal mask with minimum openings as small as 500 nm in aperture size. By adopting this method, we successfully fabricated highly crystalline multi-color perovskite micro-disk arrays. One of the unique advantages of this approach is that it can combine both solution-based and vapor-deposition processing techniques. This allows us to engineer sandwich-structured perovskite photodiode stack arrays with a diameter of 50 um, which has profound applications in the field of micro-LEDs and micro-nano devices.

Fig. 1a schematically depicts the process of creating high-resolution multi-pattern perovskite arrays using an ultrafine metal mask. The metal mask was developed by implementing interference lithography to generate photoresist patterns on the conductive substrate and then transferring the pattern to a metal mask through electroplating. The perovskite patterning process involves preparing a substrate with a tightly attached thin metal mask, depositing the perovskite precursor solution on the substrate by spin-coating, and filling the arrays defined by the metal mask. After baking at 100 °C for 1 min, crystalline perovskite arrays are obtained, and the reusable metal mesh template can be mechanically detached from the substrate without causing any fractures in the patterned perovskite arrays. Finally, the sample is immersed in a toluene solvent, which dissolves the adhesion layer in the perovskite arrays.

Fig. 1b shows the photo of a fabricated ultrathin and ultrafine metal mask and its tilted top surface and cross-sectional scanning electron microscopy (SEM) analyses further confirmed the uniformity and ultrathin thickness (1um) of the metal mask (Fig. 1c and Fig. 1d). Furthermore, the successful demonstration of a large-area, high-resolution metal mask (with a period of 1um and hole size of 500 nm) is presented in Fig. 1e. Illustratively, Fig. 1f shows a 1.5 cm wide square perovskite array patterned onto a 3 cm conductive transparent substrate. The detailed morphology of the perovskite array was characterized using scanning electron microscopy (SEM), as shown in Fig. 1g. Fig. 1h reveals the smooth surface and well-defined edges of a single-pixel SEM image, illustrating the high

quality and precise arrangement of perovskite structures with a diameter of 30 um and a period of 120 um on the substrate. To further verify the quality and precise arrangement of the patterned perovskite arrays achieved using the metal mask, fluorescence microscopy was employed, yielding uniform and brightly illuminated images of the perovskite arrays (Fig. 1i).

This method enables the patterning of perovskite micro-arrays on versatile substrates with reusable metal templates. Furthermore, it can be applied to other materials to facilitate the development of micro-structured optoelectronic devices.



**Fig. 1. Schematic illustrations: (a)** Patterning perovskite micro-disk arrays using a free-standing metal mask. **(b)** Image of a metal mask image with 30 um diameter hole arrays, the central patterned area covering 2.25cm<sup>2</sup>. **(c)** SEM image of metal mask film. **(d)** Thickness characterization of the metal film. **(e)** Metal mask image with 500-nm-diameter and 1-um-period hole arrays, the central patterned area covering 2.25cm<sup>2</sup>. **(f)** the image of patterned perovskite arrays on the ITO substrate. **(g)** SEM image of patterned perovskite arrays with 30 um diameter. **(h)** SEM image of magnified single perovskite pixel. **(i)** Fluorescence image of green perovskite arrays with 5 um diameter.

## References

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