Supplementary Information

A Universal High-Resolution Micro-Patterning Technique for Solution-Processed Materials

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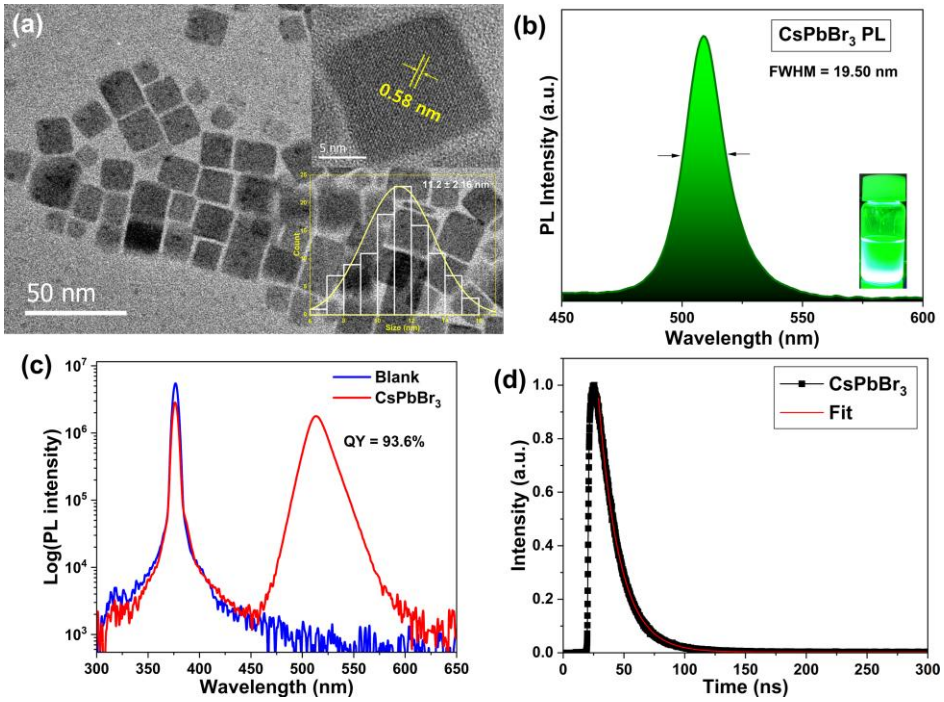
Supplementary Note 1. Synthesis of $CsPbBr_3$ perovskite quantum dots (PQDs)

Chemicals. Reagent grade precursors, $PbBr_2$ (99.9%), CsBr (99.9%), octanoic acid (OTAc) (≥ 99%), didodecyldimethylam- monium bromide (DDABr) (98%), tetraoctylammonium bromide (TOABr) (98%) were purchased from Sigma-Aldrich.

To synthesize green $CsPbBr_3$ PQDs, Cs_2CO_3 (0.5 mmol) was dissolved in 10 mL of OTAc to form the "Cs_2CO_3-in-OTAc solution." $PbBr_2$ (0.5 mmol) and 1 mmol of TOABr were added to 5 mL of toluene to create the "$PbBr_2$-and-TOAB solution." DDABr was dissolved in toluene at a concentration of 0.05 mol/L to prepare the "DDABr solution." Sequentially, 1 mL of the Cs_2CO_3-in-OTAc solution was injected into the $PbBr_2$-and-TOAB solution, followed by adding 1.3 mL of the DDAB solution. After 2 minutes, 11 mL of ethyl acetate was added. The resulting mixture was centrifuged at 10 000 rpm for 5 minutes, and the precipitates were dispersed in toluene. This centrifugation-redispersion cycle was repeated, and the final PQD precipitates were dispersed in toluene for further characterization.

$CsPbBr_3$ PQD sample characterization. The morphology of the $CsPbBr_3$ PQDs was investigated using specific instrument models such as transmission electron microscopy (TEM) and high-resolution TEM (HR-TEM) images acquired on a FEI Tecnai G2 F20 S-TWIN HR(S)TEM operated at a 200 kV accelerating voltage. The $CsPbBr_3$ PQDs were drop-casted on a 300 mesh copper Formvar/carbon grid and dried in a vacuum overnight. The PL spectra of perovskite films and patterns were measured by our home-made micro-PL system, which includes a continuous wave (CW) laser (λ = 405 nm) as the excitation source, an objective lens for focusing the laser beam onto the sample, a CCD camera (Chameleon 3, FLIR) for observing the laser beam location, and an optical spectrometer (OSM 100, Newport) for collecting the PL. The PLQY of the PQD-dispersed toluene solutions was measured using an integrated sphere unit, with a CCD beam profiler (Thorlabs) for determining the focused beam size, and a silicon photodiode (Newport 818-UV) for measuring the power. The PLQY was calculated as the number of emitted photons divided by the number of absorbed photons. Time-resolved PL decays were measured using a time-correlated single photon counting system (FluoTime 100, PicoQuant) with a pulse laser excitation source (470 nm, 60 ps, 40 MHz).



Supplementary Fig 1. (a) TEM image of the $CsPbBr_3$ PQDs (inset image: corresponding HR-TEM and particle size distribution images), (b) PL spectrum of the PQD film (inset image: $CsPbBr_3$ PQDs in solution under UV light), (c) PLQY of the $CsPbBr_3$ PQDs in solution, and (d) TRPL decay curve of $CsPbBr_3$ PQDs.

Supplementary Note 2. Mechanical dry lift-off micro-patterning method

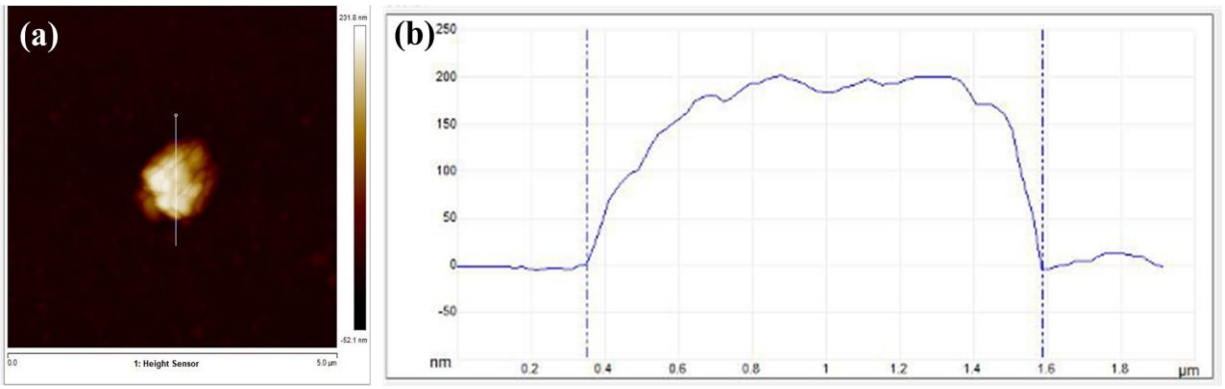
A Glass substrate is first cleaned for 5 min using sonication in acetone, then isopropyl alcohol (IPA), and finally rinsed in de-ionized (DI) water using a spin rinse dryer (SRD). Alignment marks are put down on the wafer using a typical photolithography method. Next, a parylene-C dimer of 6.3 gm is used for deposition of a 3-µm-thick parylene layer using CVD (Specialty Coating Systems Model PDS2010 Labcoter).

For the photolithography, positive photoresist is spin-coated on the parylene layer and baked at 100C for 2 minutes. A mercury arc lamp UV exposure with a dosage of around 11.44 mW/cm^2 at 400 nm for 9 seconds is applied with an ABM SemiAuto Aligner. The resist is developed in AD-10 developer for 70 seconds. The parylene layer is etched using a reactive ion etch (RIE) system, transferring the pattern trenches onto the parylene. The remaining photoresist is etched away using O_2 plasma RIE. Finally, the QD solution is drop-casted on the substrate with a pipette. After a few minutes, the parylene is peeled off the substrate using tweezers once the solution is dried. To achieve a multi-color pattern, the process is repeated, starting with the parylene layer deposition. The alignment marks on the wafer ensure alignment between layers of different colors.

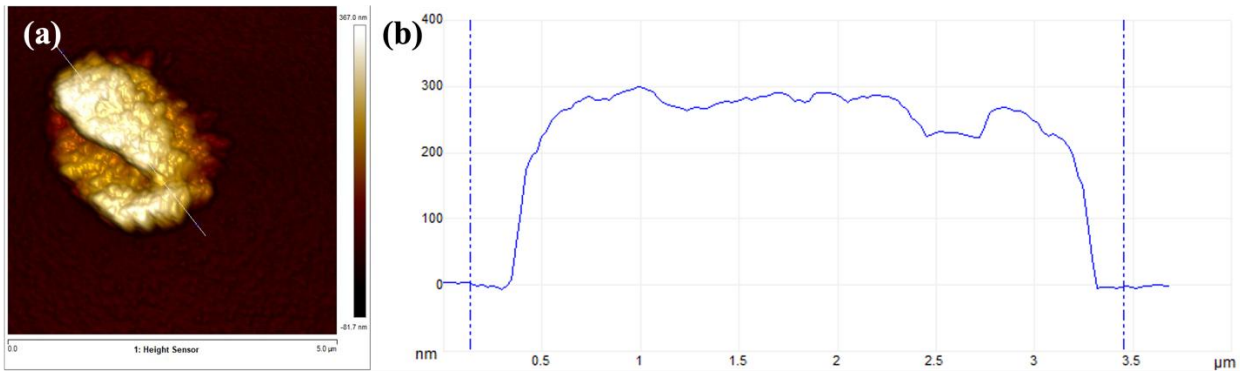
Additionally, for patterning the 3-µm thick parylene layer, we used a photoresist of approximately the same thickness (~3 µm). In subsequent iterations of the fabrication process, to address challenges associated with using a thick photoresist layer, we introduced an additional aluminum (Al) layer on top of the parylene layer before photolithography. This Al layer was then patterned using photolithography and wet etching, effectively transferring the mask pattern onto the Al layer. The substrate was then subjected to dry etching of the parylene, with the top Al layer serving as the etching mask.

Supplementary Note 3. Surface topography of single feature

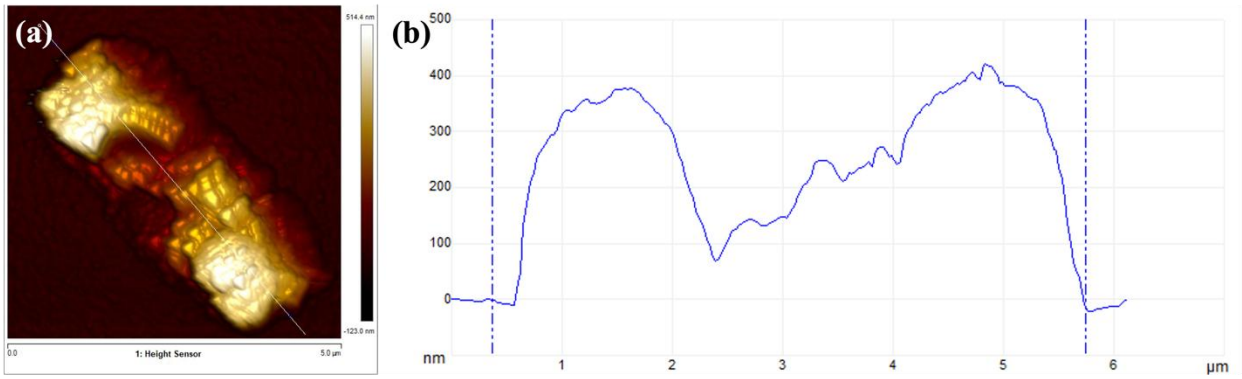
AFM measurements are taken on features of varying sizes to analyze their topography. Supplementary Figures 2, 3, and 4 show the topography of 1.2 µm, 3 µm, and 5.3 µm size features. Thicker edges are observed for 5.3 µm size feature, indicating pronounced coffee ring effect. This effect became less noticeable as the feature size is decreased. For comparison, in Supplementary Figure 5(a) and (b), the surface morphologies for 50 µm size features of spin-coated perovskites, previously measured by our research group, show a more pronounced effect.[1] Also, vacuum deposited perovskites (Supplementary Figure 5(c), (d)) show uniform surface compared to spin-coated or drop-casted perovskites. Using dispensing methods like the vacuum deposition, slot-dye coating can improve the surface profile of the patterned material.



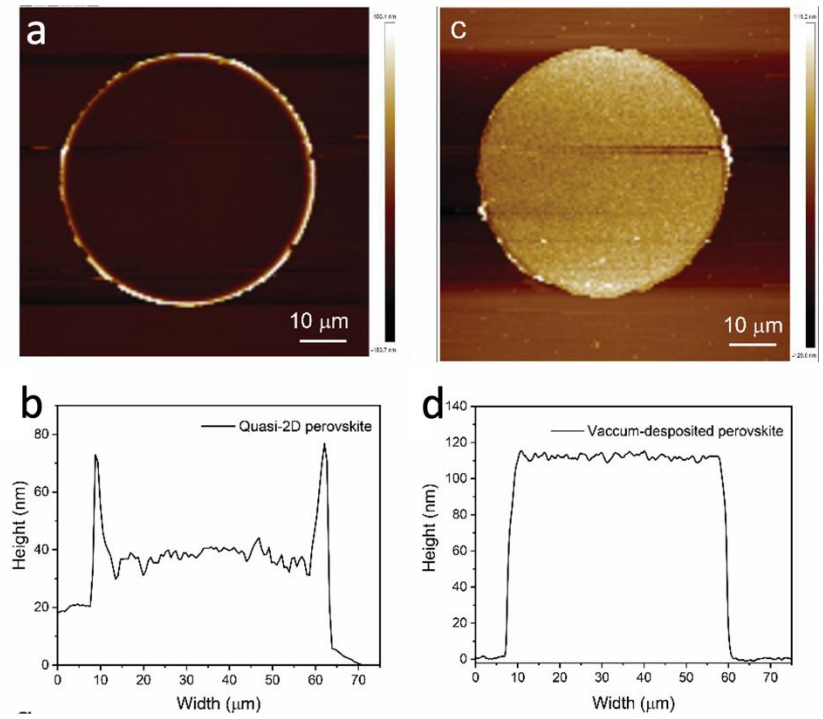
Supplementary Fig 2. (a) 2D height profile from AFM of a single patterned QD feature, and (b) cross-sectional height profile of the QD feature showing a diameter of ~1.2 µm and height of ~180 nm. The profile is taken along the vertical arrow in (a).



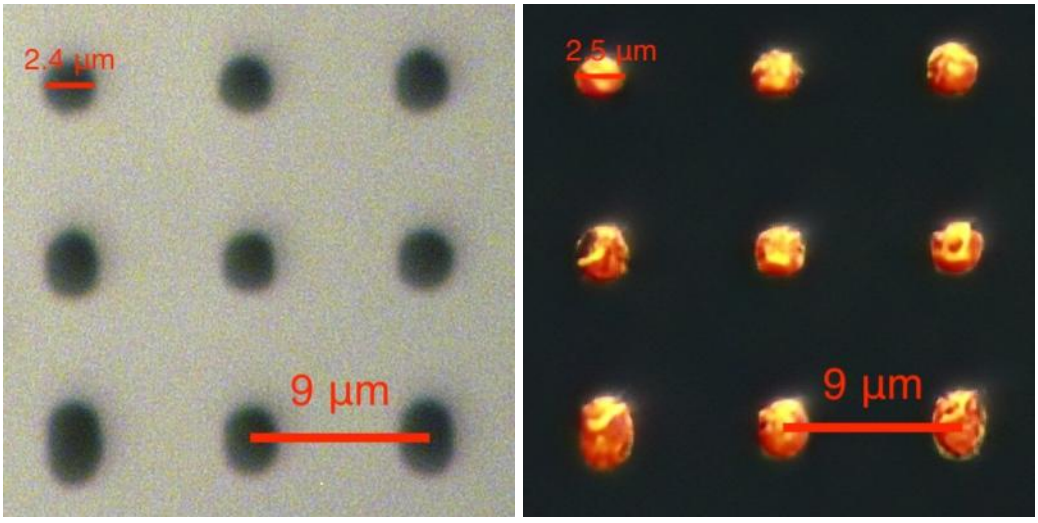
Supplementary Fig 3. (a) 2D height profile from AFM of a 3 μm QD feature, and (b) cross-sectional height profile of the QD feature showing a height of ~280 nm. The profile is taken along the diagonal line in (a).



Supplementary Fig 4. (a) 2D height profile from AFM of an elongated 5.3 μm QD feature showing peaks at the edges, and (b) cross-sectional height profile of the QD feature showing ~420 nm thickness at the peak. The profile is taken along the diagonal line in (a).



Supplementary Fig 5. (a)-(b) AFM and height profile images of 50 μm circles for spin-coated quasi-2D perovskites, and (c)-(d) AFM and height profile images of 50 μm circles for vacuum-deposited perovskites. Reprinted (adapted) with permission.[1] Copyright {2020} American Chemical Society.



Supplementary Fig 6. [left] The features after development of parylene. [right] The QD pattern observed under fluorescence microscope for the parylene pattern on the left.

References

1. Zou, C. et al. Photolithographic Patterning of Perovskite Thin Films for Multicolor Display Applications. *Nano Letters* **20**, 3710-3717 (2020).