Electronic supplementary information for

Spatially Shaped Femtosecond-Laser-Assisted 100 nm Ultrafine Patterning of Quantum Dots for High-Resolution Micro-LED Displays

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Preparation of the SU-8 photoresist layer

The SU-8 photoresist was fabricated on a GF33 glass substrate using a standard photolithographic process with careful substrate preparation and process control. Before coating, the GF33 glass substrate was ultrasonically cleaned in absolute ethanol, thoroughly rinsed with deionized water, and dried under a nitrogen stream. The substrate was then baked on a hot plate at 150 °C for 10 min to remove residual moisture. After cooling to room temperature, 2 μm SU-8 2002 negative photoresist was deposited onto the substrate by spin coating using a two-step program to ensure uniform film formation. The photoresist was first spread at a low rotational speed of 600 rpm for 15 s, allowing the SU-8 to uniformly cover the substrate surface. This step was followed by a high-speed spin at 3200 rpm for 35 s to define the final film thickness and improve thickness uniformity across the substrate. Following spin coating, the samples were left to rest under ambient conditions for approximately 3 min to relieve internal stress and allow partial solvent evaporation. The coated substrates were then soft-baked on a hot plate at 95 °C for 1 min to further remove residual solvent. Ultraviolet (UV) exposure was subsequently carried out with an exposure dose of 80 mJ cm^{-2} for 10 s without the mask. After exposure, a post-exposure bake was performed at 95 °C for 1 min to promote cross-linking in the exposed regions, resulting in a well-defined 2 μm SU-8 photoresist layer on the GF33 glass substrate.

Experimental setup of laser drilling

The fabrication of a homogeneous micropore mould in SU-8 photoresist is performed using femtosecond (fs) laser pulses shaped into both Gaussian and Bessel beams via the experimental setup shown in **Fig. 1**. The laser system employed was a Spectra-Physics Solstice Ace system capable of delivering 35 fs ultrafast pulses at a central wavelength of 800 nm with a tunable repetition rate ranging from 1 Hz to 1000 Hz **(Fig. S1).** The average output power could be varied up to 7 W. A motorized filter wheel combined with a Neutral density (ND) filter was used to modulate the pulse energy and initial beam diameter. The laser beam was directed through a series of high-reflectivity mirrors (M1–M2), and the number of pulses was controlled using a gated mode in the laser source. The initial Gaussian beam had a full diameter of approximately 8 mm. For Bessel-beam generation, the expanded Gaussian beam was directed through a fused-silica axicon lens (base angle = 2°, refractive index n = 1.45), converting it into a conical wavefront. A plano-convex optical lens with a focal length (f = 100 mm) was used to form the intermediate Bessel region. The beam was passed through a dichroic mirror (DM) and subsequently demagnified and focused onto the specimen using a 50 × objective lens with a numerical aperture (NA) of 0.65. For comparison, Gaussian-beam drilling was also performed by removing the axicon and convex lens, allowing the original Gaussian beam to be directly focused onto the SU-8 layer using the same objective. The samples consisted of 500 μm-thick glass substrates uniformly coated with a 2 μm-thick layer of SU-8 2002 photoresist. All SU-8 samples were mounted on a six-axis precision translation stage (Physik Instrument M-840.5DG or equivalent), offering sub-micrometer resolution in motion control. During the drilling process, the axial focal zone of the beam was precisely aligned within the SU-8 layer using a live charge-coupled device (CCD)-assisted automated Z-axis focal plane calibration method to avoid optical or thermal damage to the underlying glass substrate. The frequency of the beam and scanning speed are utilized to control the space between the holes. Hole spacing was maintained between 2 μm and 15 μm to minimise thermal accumulation and stress interactions. Real-time monitoring of beam alignment and process control was conducted using CCD cameras in a top-view configuration. A digital power meter is used to check the power during the experiment, and the initial spot diameter of the beam is fixed to use the laser alignment paper and spot measuring equipment. In the initial stage for lower focus, the optical microscope is used to analyze the different microstructures fabricated by controlled parameters like Pulse energy, frequency, number of pulses, and ± focal depth.

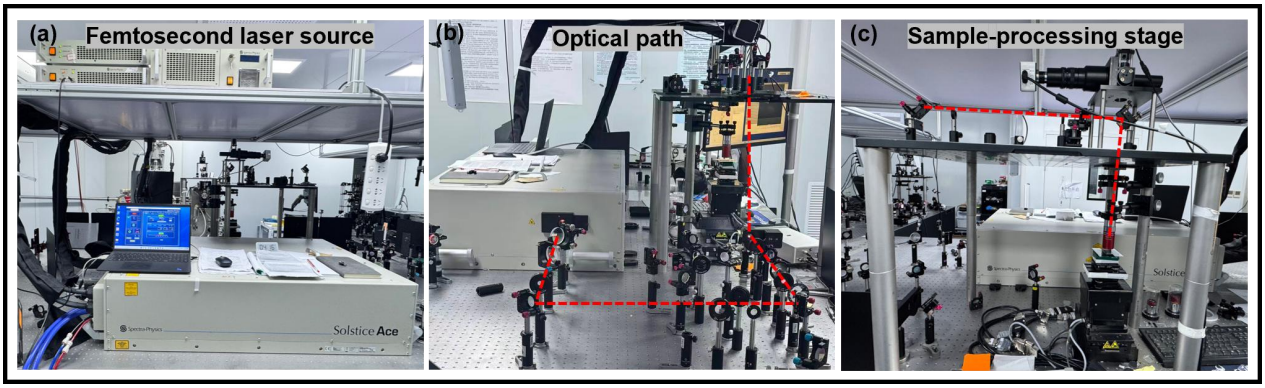

Fig. S1 Experimental setup of the fs-laser Bessel-beam micro-drilling system, including (a) the fs-laser source, (b) the optical beam path and beam-shaping configuration, and (c) the micro-drilling and sample-processing platform.

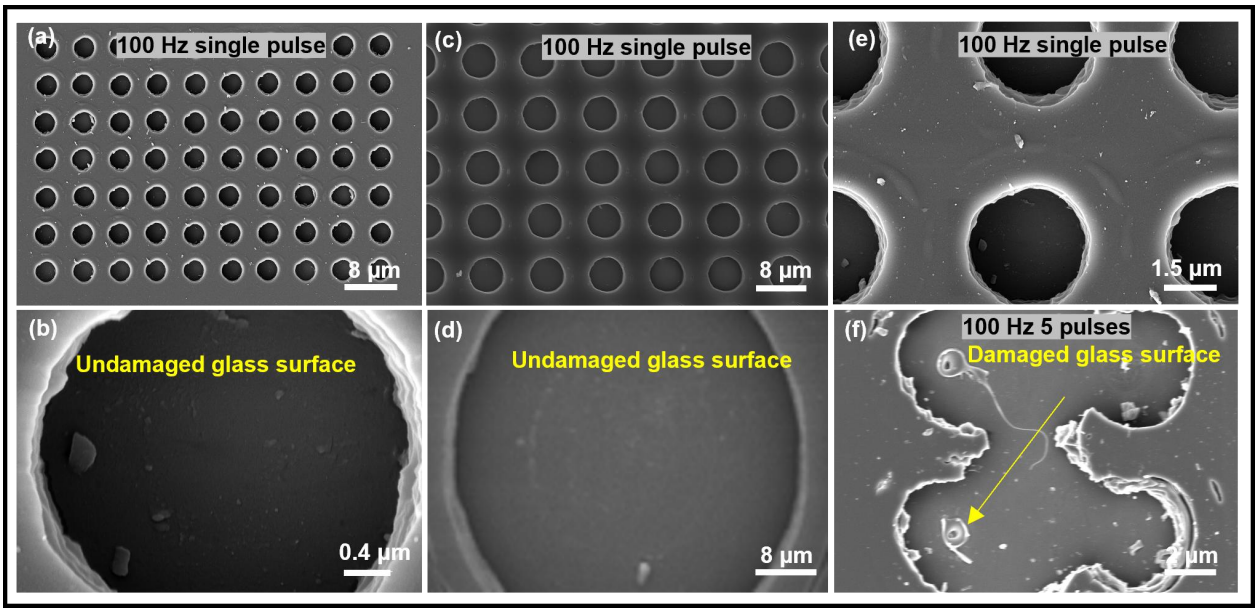

Fig. S2 Scanning electron microscopy (SEM) images of fs-laser-drilled microholes in SU-8 at different pulse energies with a Bessel beam. Arrays fabricated at 100 Hz with E = 19 μJ (a–b) and at 100 Hz with E = 25 μJ (c) exhibit uniform morphology, sharp edges, and undamaged glass substrates. A zoomed-in view at 100 Hz (d-e) confirms the absence of peripheral defects, whereas drilling at 100 Hz (f) with multiple pulses (E = 19 μJ, 5 pulses) leads to glass damage and irregular hole boundaries.

Energy-dependent size tunability

The hole diameter is tuned step-by-step by reducing the pulse energy while keeping the repetition rate and optical alignment constant, using single-pulse fs Bessel-beam irradiation. First, the pulse energy is set to a relatively high level to generate large, well-defined micropores: 46 μJ, 40 μJ, 33 μJ, and 27 μJ produce holes with mean diameters of approximately 37 μm, 27 μm, 19 μm, and 11 μm, respectively **(Figs. 3a–d)**. Next, the pulse energy is reduced in smaller

increments to access the mid-microscale regime; at 25 μJ and 23 μJ**,** the hole diameters decrease to approximately 8 μm and 5.5 μm (**Figs. 3e** and **3f)**. The pulse energy is then lowered further to approach few-micrometer apertures: at 19 μJ and 14 μJ, the resulting hole diameters are approximately 3.5 μm and 1.8 μm, respectively (**Figs. 3g** and **3h).** Finally, to reach the submicron regime, the pulse energy is decreased to 10 μJ, 8 μJ, 6.5 μJ, 5 μJ, and 4 μJ, yielding holes with diameters of 1 μm, 500 nm, 350 nm, 250 nm, and 100 nm (**Figs. 3i** and **3j).** Enlarged views of the smallest features are provided in **Figs. 3k** and **3l**. Throughout this stepwise energy reduction, the holes maintain good circularity and uniformity with minimal edge debris, indicating stable and reproducible ablation under single-pulse Bessel-beam exposure. This controlled, energy-dependent size scaling enables the fabrication of micro- to subpixel apertures relevant to micro-LED CC-layer design.

Luminous uniformity analysis method

A CCD camera was used to record the fluorescence images of the Cadmium selenide (CdSe) quantum dot colour conversion (QDCC) layer under 360 nm UV excitation. After converting the images into 8-bit grayscale intensity maps, ImageJ was used to extract the photoluminescence (PL) intensity of individual QD pixels. According to the "Measure methods of the Chinese electronic industry standard for Light-Emitting Diode (LED) Displays, the luminous uniformity I_{RJ} of a display pixel array is calculated using the luminous intensity distribution of randomly selected pixels. For each QDCC layer sample, 30 pixels were chosen from the array to compute the arithmetic mean intensity $\bar{I}$.

The luminous uniformity I_{RJ} is defined as:

$$\boldsymbol{I_{RJ} = 1 - \frac{|I_i - \bar{I}|_{max}}{\bar{I}} \times 100\%}$$

where, $\boldsymbol{I_i}$ is the fluorescence intensity of any selected pixel.

$\boldsymbol{\bar{I} = \frac{1}{30}\sum_{i=1}^{30} I_i}$ is the mean intensity of the 30 selected pixels.

$\boldsymbol{|Ii - \bar{I}|_{max}}$ represents the maximum deviation from the mean intensity among the selected pixels. Using this method, the 10 μm red CdSe QD array achieved a luminous uniformity of 90%, while the 20 μm green CdSe QD array reached 97%, both exceeding the minimum requirement specified in the LED display standard.

Single-colour QDCC layer preparation

Single-colour CdSe QDCC layers were fabricated by the fs-laser drilling and micropore filling steps. First, an SU-8 2002 film with a 2 μm thick film was spin-coated onto a glass substrate. Micropore arrays were fabricated in the SU-8 layer by single-pulse fs-laser drilling, after which the patterned SU-8 film functioned as a rigid and well-defined micropore mould for subsequent QD filling. For single-colour QDCC layer fabrication, a UV-curable CdSe QD gel, either green or red, was dispensed onto the SU-8 micropore mould using a micropipette. The QD gel was then spread repeatedly across the surface with a soft squeegee to promote uniform infiltration, allowing the QD material to fully penetrate and fill each micropore, while excess gel on the surface was gradually removed. After filling, the sample was exposed to low-intensity UV light for 2 minutes to cure and stabilize the QD gel within the SU-8 cavities. A gentle mechanical polishing step was subsequently applied to remove residual material from the surface; this step was performed with particular care, as the SU-8 layer thickness is only 2 μm, and excessive force can damage the polymer layer or distort the micropore structure. Finally, a thin transparent photoresist layer was spin-coated over the entire sample to encapsulate the QD-filled micropores, providing protection against moisture and oxygen and improving long-term optical stability. Through this controlled filling, curing, and careful cleaning process, a uniform single-colour CdSe QDCC layer with well-defined pixel geometry and stable photoluminescence was reproducibly fabricated, forming a reliable platform for optical characterisation and micro-LED QDCC layer applications.

Optical and Photoluminescence Characterisation of QDCC layers

In the initial stage, optical microscopy was used to examine microstructures fabricated under different processing conditions, including pulse energy, repetition rate, number of pulses, and ± focal depth. SEM was then used to examine the microstructures obtained after the fabrication and determine the uniformity of hole depths, morphology of sidewalls, and structural fidelity on the nanometer scale. The patterned and fabricated QDs were then systematically analyzed according to Zeiss confocal, two-photon, and super-resolution fluorescence microscopy to compare the pixel morphology, pore filling quality, emission uniformity, and photoluminescence distribution with the SU-8 micropore arrays. A blue light excitation system was used to provide fluorescence excitation to measure the steady-state emission spectra of the QDCC layer. Based on the measured spectra, important optical parameters, such as peak emission wavelength and

full width at half maximum, were derived in order to measure spectral characteristics and purity of colour. Chromaticity coordinates (x, y) were then obtained using the emission spectra to ascertain the coverage of the National Television System Committee (NTSC) colour gamut by the fabricated QDCC layers. Origin software was used to create PLC charts in which the calculated coordinates of (x, y) were plotted, and the NTSC reference triangle was added to understand and measure the colour gamut coverage. For quantitative analysis of emission uniformity, fluorescence images were processed using ImageJ software. During this step, RGB channels were separated, and individual micropores were selected as regions of interest to extract single-pixel PL intensity values. PL intensity histograms were then generated to evaluate pixel-to-pixel brightness variation across the micropore arrays. Origin software was further applied for plotting emission spectra, PL intensity distributions, chromaticity diagrams, and related statistical analysis.

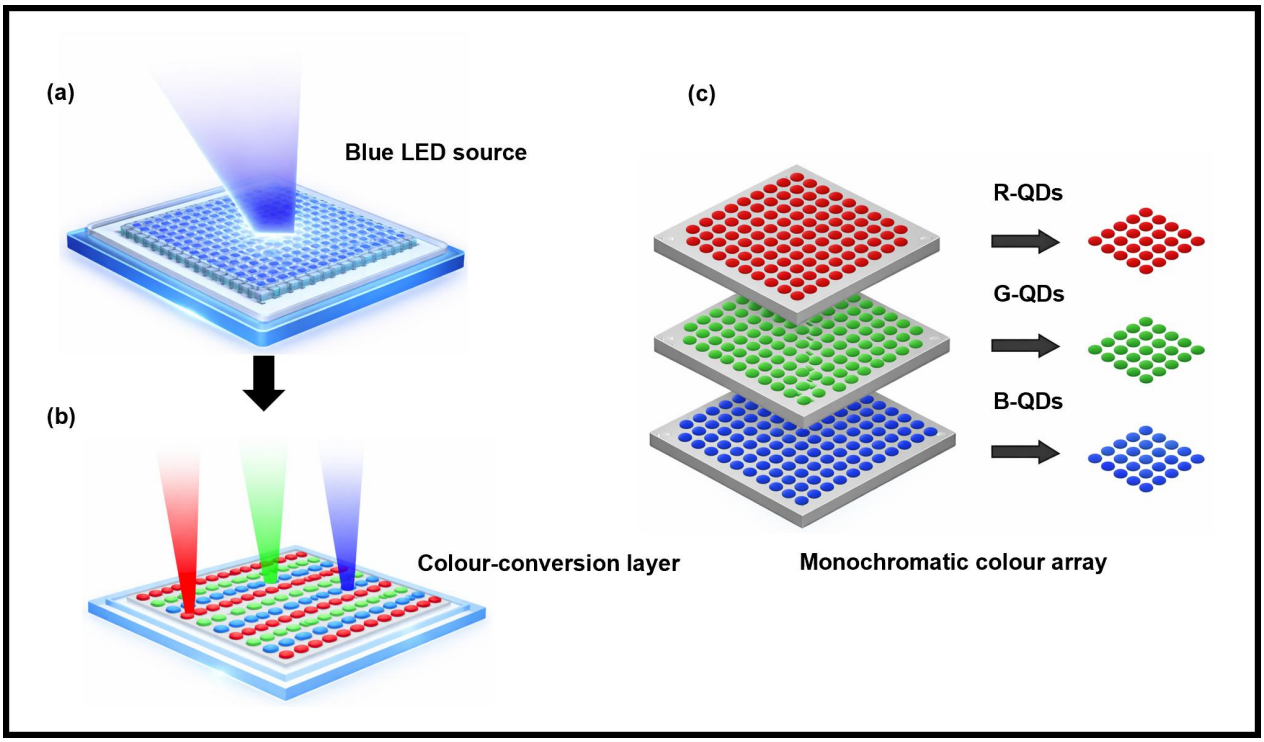

Fig. S3 Working principles of QDCC layer in micro-LED displays. (a) The array of blue micro-LEDs is displayed, which serves as the excitation source. (b) Patterned layers of red and green QD convert blue light to red and green in order to produce all colours. (c) Monochromatic QD patterns.

Dual-colour QDCC layer preparation

A dual-colour CdSe QDCC layer was fabricated by extending the single-colour micropore filling process through repeated fs-laser drilling and QD filling steps. After forming the first colour QD layer, either red or green, inside the SU-8 micropore mould, the operator repositioned the sample on a computer-controlled six-axis translation stage for the second drilling step a careful alignment mattered at this stage. The new micropores needed accurate registration with the predefined array layout. The new pores also needed safe spacing from the previously filled QD pixels. Poor alignment or tight spacing between adjacent pores disturbed the planned red and green pixel layout. Poor alignment also increased the risk of damage, contamination, or material carryover into the first filled QD sites. After confirming alignment, the operator drilled a second micropore array into the SU-8 layer using the same optimized single-pulse fs-laser settings. The operator then filled the new micropores with the opposite colour CdSe QD gel using repeated squeegee coating to drive uniform infiltration and complete pore filling. The operator removed excess surface material through gentle mechanical polishing. The operator then stabilized the filled QDs using low-intensity UV curing. This workflow produced well-defined red and green QD pixels with clear spatial separation and low cross-contamination. After finishing the red and green QD layers, the operator used the fs-laser a third time to drill an additional micropore array and left this array unfilled. These QD-free micropores serve as optical transmission windows for blue excitation light, enabling direct blue emission from the underlying micro-LED (**Fig. 6a**) without interaction with the QDs, while adjacent red- and green-QD-filled micropores provide wavelength-converted emission, thereby completing the RGB architecture. Finally, a thin transparent polymer encapsulation layer was spin-coated over the entire structure to protect the QD-filled micropores from moisture and environmental exposure, ensuring long-term optical stability and mechanical robustness of the dual-colour QDCC layer.

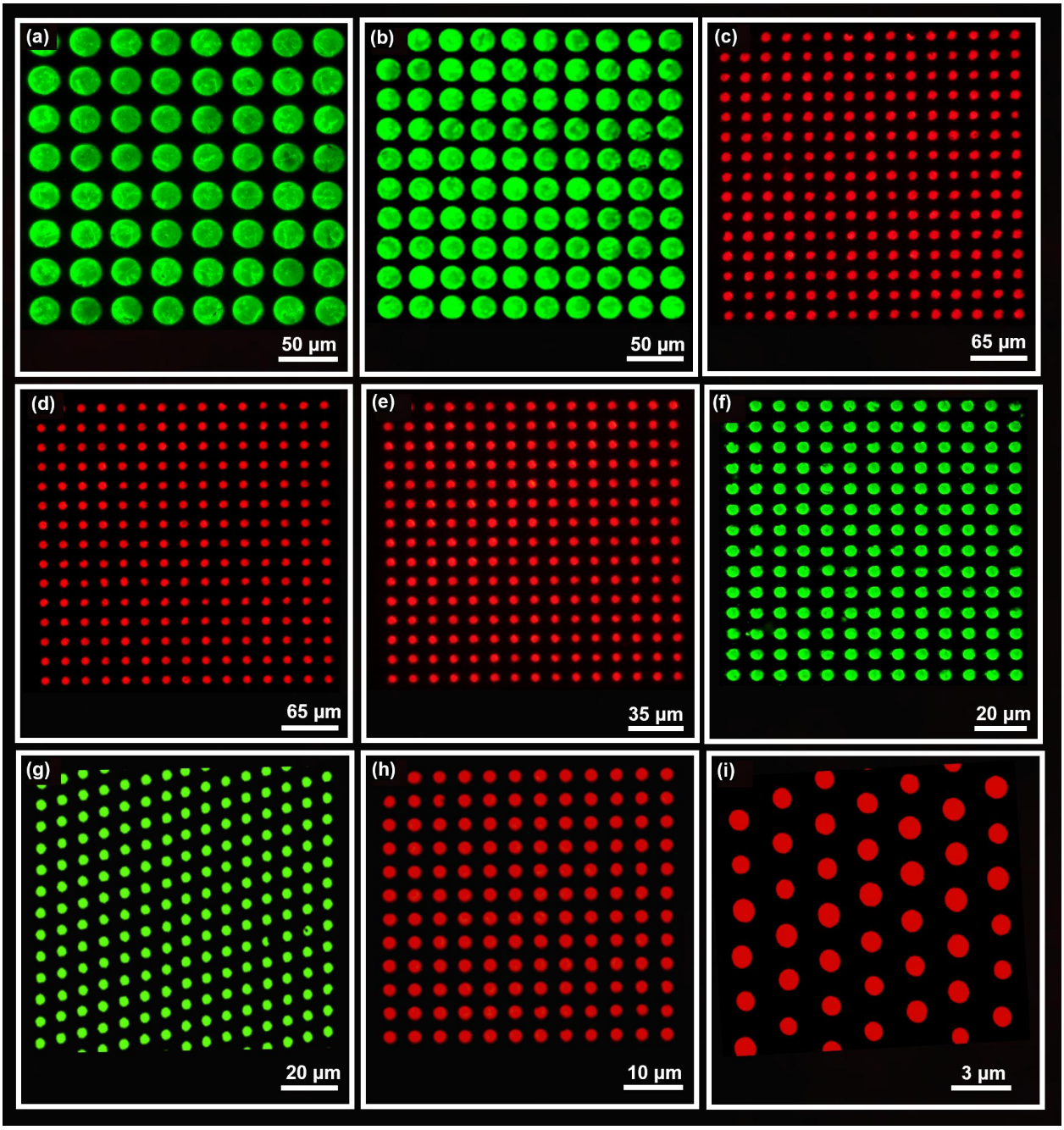

Fig. S4 Fluorescence micrographs of CdSe QD–filled SU-8 micropore arrays with tunable pixel diameters, demonstrating high-resolution pattern formation over a wide size range. (a-e) QD arrays with pixel diameters of 27 µm, 21 µm, 10 µm, 8 µm, 6.5 µm. (f-i) QD arrays with pixel diameters of 5 µm, 3.5 µm, 2 µm, and 700 nm exhibit uniform emission, circular pore geometry, and consistent spacing across large areas.

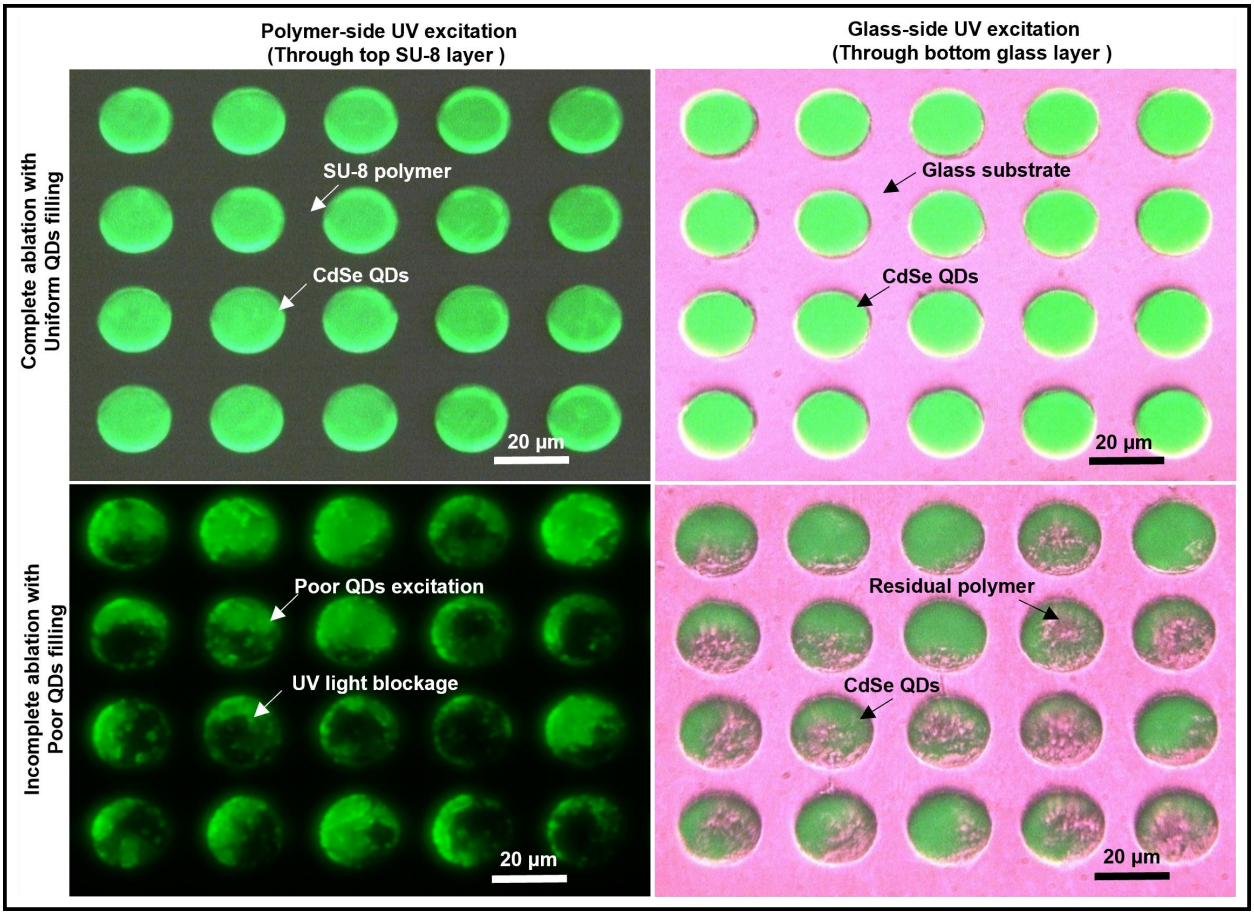

Fig. S5 Cross-sectional optical images of CdSe QD–filled SU-8 micropores recorded using (left) standard optical microscopy and (right) UV illumination. In the optical image, the SU-8 layer and the QD-filled cavities can be distinguished by their contrast against the glass substrate. Under UV light, the confined CdSe QDs emit bright fluorescence along the micropore locations, confirming successful filling and stable retention of the QDs within the SU-8 cavities after curing. The clear emission pattern further demonstrates that the QDs remain uniformly embedded and well-aligned with the micropore geometry.

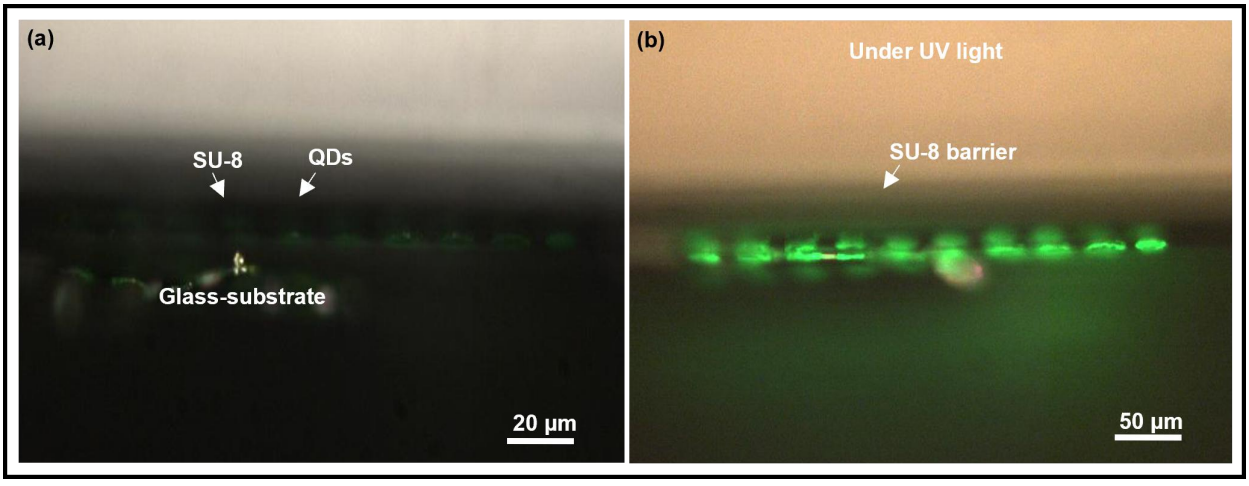

Fig. S6 Cross-sectional optical images of CdSe QD–filled SU-8 micropores. (a) The panel shows the structure under normal illumination, where the SU-8 layer and the embedded QDs are visible above the glass substrate. (b) The panel under UV excitation shows the confined QD fluorescence clearly outlines the filled micropores, confirming that the SU-8 layer effectively defines the cavity boundaries and maintains the vertical confinement of the QDs during and after curing.

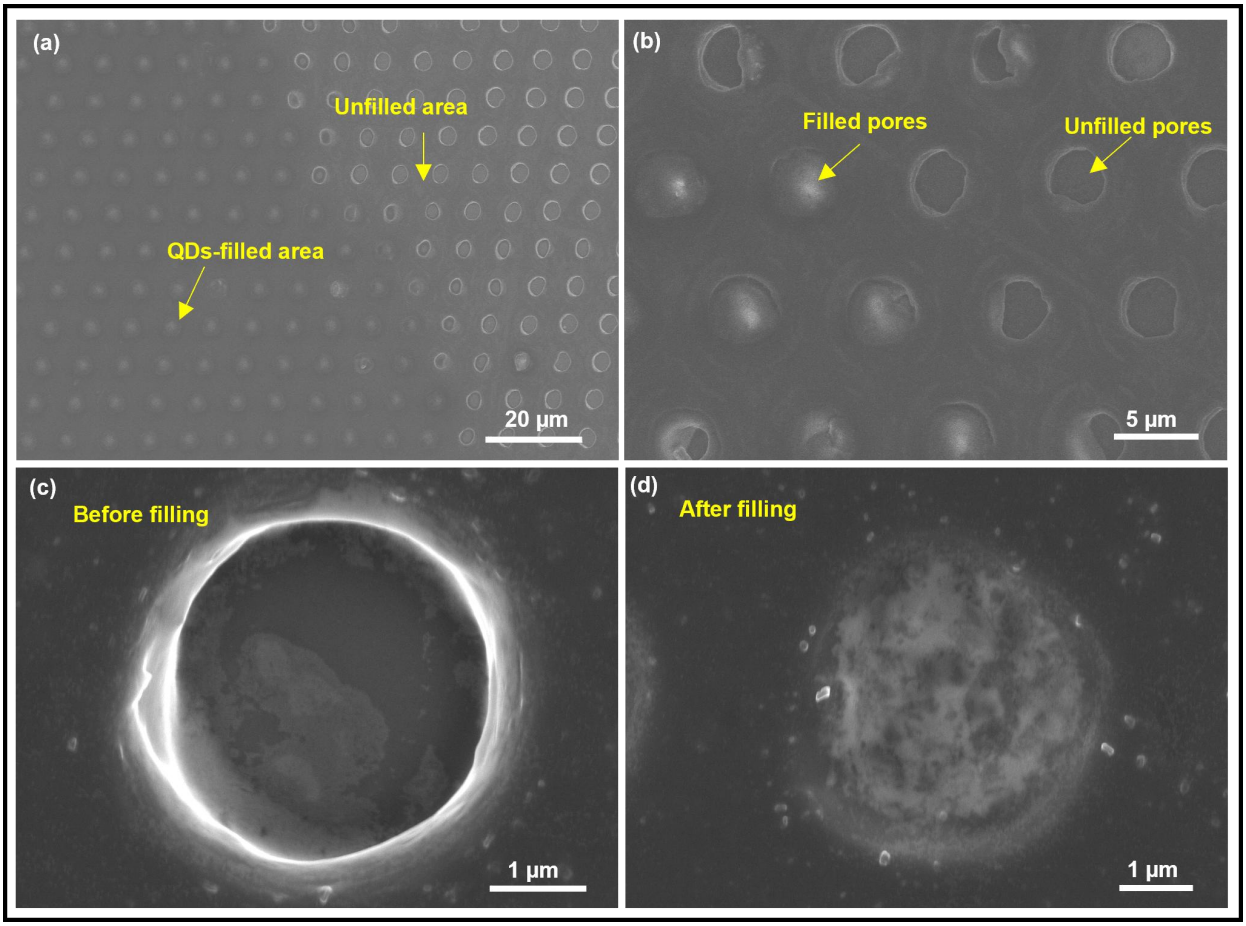

Fig. S7 (a–b) SEM images showing a patterned array of micropores where some are filled with QDs and others remain unfilled, highlighting the selective deposition. (c) Image of a single micropore before filling, and (d) the same pore after successful QDs deposition, confirming material presence and distribution within the pore cavity.

Emphasizing micro pore filling

Fig. S7 compares SU-8 micropores before and after QD filling and shows how the filling procedure affects pore occupancy. **Figs. S7a** and **S7b** present SEM images of a patterned micropore array after QD deposition. The same region contains both filled and unfilled micropores. This outcome shows that filling uniformity depends on the deposition method and the processing sequence, and the process does not produce uniform filling by default. At the

single pore level, **Fig. S7c** shows a representative micropore before QD deposition with a clean, well-defined cavity. After filling, **Fig. S7d** shows the same micropore with QD material present inside the cavity. This confirms deposition and retention of the emissive material inside the pore. The array level and single pore results show a clear requirement for process control during QD filling. The workflow needs controlled dispensing and strict adherence to the intended step order. If the workflow drifts from the planned sequence, incomplete filling and non-uniform filling occur, and reduce pixel uniformity in the micro-LED CC layer.

Emphasizing RGB architecture

Fig. S8 presents the stepwise fabrication and optical response of single-colour and dual-colour QDCC layers based on laser-drilled micropore arrays. **Figs. S8a, S8c,** and **S8e** show fluorescence images recorded under UV excitation, highlighting the emission patterns of the QD-filled micropores, whereas **Figs. S8b, S8d,** and **S8f** present the corresponding optical images without UV excitation, revealing the underlying micropore layouts and filling configurations. In the first configuration (**Figs. S8a** and **S8b)**, a single-colour QDCC layer is fabricated by drilling one set of micropores and filling them with red CdSe QD**,** resulting in a uniform single-colour emission under UV excitation. To realize multi-colour integration, a dual-colour QDCC layer is fabricated through sequential micropore drilling and selective filling. As shown in **Figs. S8c** and **S8d**, two distinct micropore lines are filled with red and green CdSe QDs, respectively, producing alternating red and green emission under UV excitation. In the third configuration (**Figs. S8e** and **S8f)**, an additional micropore line is introduced but intentionally left unfilled. In this architecture, two micropore lines are filled with red and green QDs, while the third, empty micropore line functions as a transmission channel for blue light emitted from the underlying blue micro-LED**.** This design enables dual-colour QD conversion combined with direct blue emission, completing the RGB pixel configuration within a single layer without requiring blue colour-conversion materials.

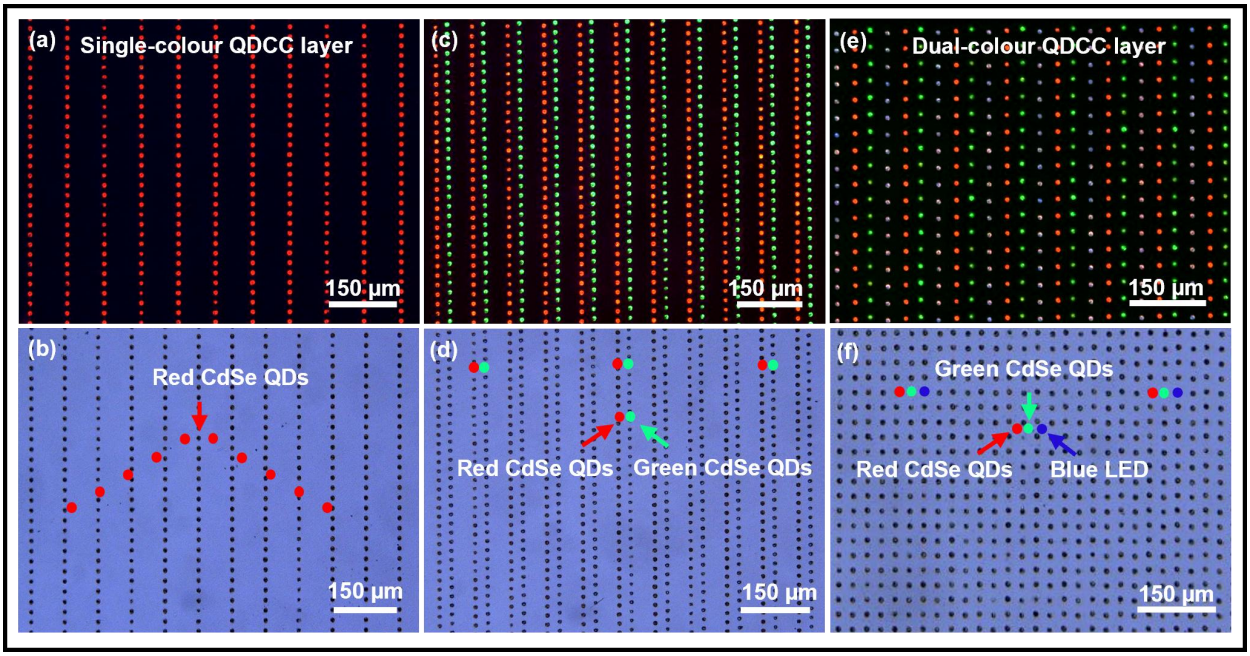

Fig. S8 Configuration of single- and dual-colour QDCC layer with a micropore diameter of 11 μm. (a-b) Single-colour QDCC layer micropore patterns without QD filling and after QD filling. (c-d) Two micropore arrays containing red and green CdSe QDs, together with corresponding arrays without QD filling. (e-f) Dual-colour QDCC layer consisting of red and green QD-filled micropores and a third QD-free micropore for blue-light transmission, along with an array without QD filling, enabling RGB operation.