Supporting Information

Theoretical modeling: Performance of micro-LEDs versus device size

In micro-LEDs, the carrier recombination rate R as a function of the carrier density n can be expressed using the ABC model:

$$R = An + Bn^2 + Cn^3 \tag{1}$$

where the coefficients *A*, *B* and *C* represent the Shockley-Read-Hall (SRH) recombination, the radiative recombination, and the Auger recombination, respectively. For most III-V semiconductors, the Auger recombination coefficient (*C*) is approximately 10^{-30} cm⁶ s⁻¹.^{1, 2} At low injection currents ($n < 10^{18}$ cm⁻³) for display purposes, the Auger recombination (*Cn*³) can be neglected. At equilibrium, the carrier generation (*G*) and recombination (*R*) are balanced. Therefore, the carrier density *n* is determined by

$$An + Bn^2 = \frac{j}{ed}$$
(2)

where *j* is the injected current density (assumed to be 36 A/cm² in the following calculations), *e* is electron charge and *d* is the thickness of LED active region (*j*/*ed* is the carrier generation rate *G* in the LED). And the internal quantum efficiency η_{int} for radiative emission is simplified as:

$$\eta_{\rm int} = \frac{Bn^2}{An + Bn^2} \tag{3}$$

The SRH recombination is determined by the defects in the bulk material and on the sidewall surface.

$$A = \frac{1}{\tau_{\text{bulk}}} + v_{\text{s}} \frac{1}{L} \tag{4}$$

 $1/\tau_{\text{bulk}}$ is the bulk recombination rate, v_s is the surface recombination velocity, and *L* is the equivalent lateral dimension of the micro-LED (to be specific, the ratio between the volume and sidewall surface area of the LED MQW active region). Combining Eqs. (3) and (4),

$$\eta_{\text{int}} = \frac{Bn^2}{\left(\frac{1}{\tau_{\text{bulk}}} + v_{\text{s}}\frac{1}{L}\right)n + Bn^2} = \frac{Bn}{\frac{L + v_{\text{s}}\tau_{\text{bulk}}}{L\tau_{\text{bulk}}} + Bn} = \frac{BnL\tau_{\text{bulk}}}{L + v_{\text{s}}\tau_{\text{bulk}} + BnL\tau_{\text{bulk}}}$$
(5)

where *n* is a constant under a fixed current density, and *B*, τ_{bulk} , v_{s} are also constants for specific materials. Therefore, η_{int} decreases when the LED size *L* increases.

For InGaP red LED, *B* is assumed to be 2×10^{-10} s⁻¹ cm³,³ τ_{bulk} is approximately 100 ns⁴ and v_{s} is 4×10^4 cm/s.⁵ For InGaN blue and green LED, radiative emission parameter *B* of 1×10^{-8} cm³ s⁻¹ was measured,⁶ τ_{bulk} is 10 ns⁷ and v_{s} is 10^4 cm/s.⁴ Fig. S1 plots theoretically predicted η_{int} as a function of device sizes for red and blue LEDs. The results are consistent with experimental observations shown in previous studies.^{8,9}

In addition, the tolerance for alignment errors during LED fabrication and transfer assembly also decreases with the device size, as shown in Fig. S2. Here we assume that the tolerance is within $\pm -5\%$ of the LED size.¹⁰

tolerance = LED size
$$\times 5\%$$

Individual device fabrication

The layout of the stacked RGB micro-LED structure includes (from bottom to top): a polyimide substrate (thickness: 75 µm), an indium gallium phosphide (InGaP) based red LED (size: 140 µm × 195 µm × 7 µm), a SU-8 layer (thickness: 5 µm), an optical filter layer based on multilayer titanium dioxide (TiO₂) and silicon dioxide (SiO₂) (size: 150 µm × 230 µm × 8 µm), a SU-8 layer (thickness: 5 µm), an indium gallium nitride (InGaN) based green LED (size: 125 µm × 180 µm × 7 µm), a SU-8 layer (thickness: 5 µm), an InGaN based blue LED (size: 125 µm × 180 µm × 7 µm). Contacts of each individual LED are metallized with sputtered metal layers (Cr/Au/Cu/Au = 10/100/500/100 nm).

Fabrication of red LEDs

The red LED device structure is grown on a GaAs substrate by metal-organic chemical vapor deposition (MOCVD). The detailed structure is listed in Table S1, which involves (from bottom to top): the GaAs substrate, an $Al_{0.95}Ga_{0.05}As$ sacrificial layer and an $In_{0.5}Ga_{0.5}P$ red LED. Active areas of the InGaP LED (area: 140 µm × 195 µm) are defined by photolithographic process and acid based wet etching. Sputtered metal layers (Ge/Ni/Au for n-GaAs and Cr/Au for p-GaAs) serve as cathode and anode, respectively. After removing the $Al_{0.95}Ga_{0.05}As$ sacrificial layer in a hydrofluoric acid (HF) based solution (HF:water = 1:10 by volume), patterned photoresist works as anchors to tether free-standing thin-film devices on the GaAs substrates. Using poly (dimethylsiloxane) (PDMS) stamps, released devices can be picked up and transferred onto another substrate.

A detailed description of the process to fabricate the freestanding microscale red LED is listed below (also see Fig. S3 and Ref. 11)¹¹:

- 1. Deposit 500 nm thick SiO_2 by PECVD.
- 2. Clean the wafer with acetone, isopropyl alcohol (IPA), and deinoized (DI) water.
- 3. Dehydrate at 110 °C for 10 min.
- 4. Spin coat positive photoresist (PR) (SPR220-v3.0, Microchem, 500 rpm / 5 s, 3000 rpm / 45 s) and soft-bake at 110 °C for 1.5 min.
- Expose PR with UV lithography tools (URE-2000/25, IOE CAS) with a dose of 300 mJ/cm² through a chrome mask and post-bake at 110 °C for 1.5 min.
- Develop PR in aqueous base developer (AZ300 MIF), rinse with DI water and hard-bake at 110 °C for 20 min.
- 7. Etch SiO₂ with buffered oxide etchant (BOE 6:1) for \sim 100 s and rinse with DI water.
- 8. Clean the PR in processed wafer using acetone, IPA, DI water.
- Etch GaP in a mixture of KOH / K₃[Fe(CN)₆] / H₂O (1:4:15, by weight) at 80 °C (hot water bath) for 60 s with gently shaking and rinse with DI water.
- 10. Etch GaP in a mixture of HCl / H₂O (1:5, by volume) for about 2 mins to remove roughness
- 11. Clean the processed wafer (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 12. Pattern PR SPR220-3.0.
- 13. Etch InAlP / MQWs / InAlP / DBR in a mixture of HCl / H₃PO₄ (1:1, by volume) for 10 s with vigorous shaking, repeat (3~4 times) until the surface is clean and shiny, and then rinse with DI water.

- 14. Etch n-GaAs in a mixture of H₃PO₄ / H₂O₂ / H₂O (3:1:25, by volume) for 5 min and rinse with DI water.
- 15. Remove PR (SPR220-v3.0) in processed wafer using acetone, IPA, DI water.
- 16. Remove the SiO₂ with BOE 6:1 solution for \sim 2 mins and rinse with DI water.
- Clean the processed wafer in step 15 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 18. Spin-coat negative photoresist (AZ nLOF 2070, 500 rpm / 5 s, 3000 rpm/ 45 s) and soft-bake at 110 °C for 2 min.
- 19. Expose with 365 nm optical lithography with for a dose of 45 mJ/cm² through a chrome mask and post-exposure bake at 110 °C for 35 s.
- 20. Develop PR in aqueous base developer (AZ300 MIF) and rinse with DI water.
- 21. Sputter 20/20/200 nm of Ge / Ni / Au.
- 22. Lift-off PR in acetone.
- 23. Rapid temperature annealing (RTA) at 220 °C for 35 s.
- 24. Clean the processed wafer in step 23 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 25. Spin-coat negative photoresist (AZ nLOF 2070, 500 rpm / 5 s, 3000 rpm / 45 s) and soft-bake at 110 °C for 2 min.
- 26. Expose with 365 nm optical lithography with for a dose of 45 mJ/cm² through a chrome mask and post-exposure bake at 110 °C for 35 s.
- 27. Develop PR in aqueous base developer (AZ300 MIF) and rinse with DI water.
- 28. Sputter 20/200 nm of Cr / Au.
- 29. Lift-off PR in acetone.

- 30. Clean the processed wafer in step 29 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 31. Pattern PR SPR220-3.0.
- 32. Etch GaAs in a mixture of $H_3PO_4/H_2O_2/H_2O$ (3:1:25, by volume) for 9 min and rinse with DI water.
- 33. Clean the processed sample with acetone, IPA, and DI water.
- 34. Pattern PR SPR220-3.0.
- 35. Etch in diluted HF (49% HF : DI water = 1:10, by volume) to release the devices from growth substrates for 1.2 hour and rinse with DI water.
- 36. Then the free-standing red LED arrays are formed and ready for picking up.

Fabrication of green and blue micro-LEDs

The InGaN based green and blue LED structures are grown on 2-inch or 4-inch conventional planar sapphire substrates using MOCVD. The main LED structure (from bottom to top) includes: the sapphire substrate, a GaN buffer layer, an n-GaN, an InGaN / GaN multiple-quantum-well (MQW) layer, a p-GaN and an indium tin oxide layer for top contacts. The detailed structures of blue and green LEDs are listed in Tables S2 and S3, respectively. The thickness of the entire epitaxial structure is about 7 μ m. LED devices are lithographically fabricated, with Cr/Au layers serving as ohmic contacts. Inductively couple plasma reactive ion etching (ICP-RIE) is used to define the LED mesa (lateral dimension 180 μ m × 125 μ m). After bonding the fully fabricated LED arrays onto a thermal release tape (TRT) (Nitto Denko Corp.), laser lift-off (LLO) is applied to separate the thin-film LEDs from sapphire substrates by thermally decompose GaN into gallium (Ga) metal and nitrogen gas at the interface. A krypton

fluoride (KrF) excimer laser at 248 nm (Coherent, Inc., CompexPro110) serves as the light source, with a uniform irradiation area of 5 mm \times 15 mm. The power density during LLO is optimized to be around 0.6 J/cm² for LEDs. After laser irradiation, the micro LEDs are released from sapphire by mild mechanical force at 70 °C (melting point of Ga is 29.7 °C). The Ga residual is removed by immersing the samples into dilute ammonia (1:5 in water) at room temperature for about 30 mins. By heating up to 120 °C, the LEDs are detached from the TRT (the critical release temperature is about 110 °C) and ready for transfer printing by PDMS. The procedures are shown in Fig. S4 and Ref. 12.

Modeling and Fabrication of thin-film filters

Optical filter design is performed based on the transfer matrix method. The designed TiO₂ / SiO₂ filter structure (19 nm TiO₂ / 15 periods of 89 nm SiO₂, 52 nm TiO₂ / 63 nm SiO₂ / 66 nm TiO₂ / 22 periods of 73 nm SiO₂ + 42.5 nm TiO₂ / 154 nm SiO₂, total thickness ~6.6 μ m, see Table S4) is deposited on GaAs wafers using ion beam-assisted sputter deposition, with substrates heated up to 300 °C (HB-Optical, Shenyang, China). After laser milling (Nd:YVO₄ laser, 1064 nm) the filter to form small rectangles (size: 160 μ m × 230 μ m), the GaAs substrate is fully removed by wet etching (NH₄OH : H₂O₂ : H₂O = 1:1:2). The released, freestanding micro-filter can be picked up and transferred by PDMS. The procedures are shown in Fig. S5 and Ref. 13.

Preparation of the adhesive solution

The adhesive solution comprises: bisphenol A glycerolate (1 glycerol / phenol) diacrylate, 3-(Trimethoxysilyl) propyl methacrylate, spin-on-glass (SOG 500F, Filmtronics Inc.), 2-Benzyl-2-(dimethylamino)-4'-morpholinobutyrophenone, and anhydrous ethanol. The weight ratio is 200:100:100:9:2000. Stir at room temperature until full mixing. Store the mixed solution in refrigerator (4 °C) for future use. See Ref. 14.

Fabrication of tandem RGB LEDs

A detailed description of the process to fabrication is listed below:

Substrate preparation

- 1. Cut the polyimide substrate (thickness: 75 μ m) into small pieces (size: 22 mm \times 22 mm).
- 2. Laminate the polyimide film onto a PDMS coated glass slide
 - a. Degas PDMS (Sylgard 184) 10 : 1 (base: curing agent, by weight).
 - b. Spin cast PDMS to a glass slide (cleaned by NH₄OH : H₂O₂ : H₂O = 1:1:5, 80 °C, 10 min) at 500 rpm/ 5 s, 3000 rpm/ 45 s to form a PDMS film (thickness: 40~60 μm).
 - c. Bake the glass for \sim 3.5 min at 110 °C.
 - d. Laminate the polyimide film onto the PDMS coated glass. Make sure that the polyimide attaches to PDMS tightly without any bubbles).
 - e. Bake the sample for 30 mins at 110 °C until PDMS is fully cured.

Transfer red LEDs

- 3. Clean the polyimide sample by acetone, isopropyl alcohol (IPA) and deionized (DI) water.
- Spin coat negative photoresist (PR) (AZ nLOF 2070, 500 rpm / 5 s, 3000 rpm / 30 s) and soft-bake at 110 °C for 2 min.
- Expose with 365 nm optical lithography with a dose of 45 mJ/cm² (URE-2000/25, IOE CAS) through a chrome mask and post-exposure bake at 110 °C for 90 s.
- 6. Develop PR in aqueous base developer (AZ300 MIF) and rinse with DI water.

- 7. Deposit 30 nm of Cr as the markers by sputtering.
- 8. Lift-off PR in acetone.
- 9. Clean the substrate and dehydrate at 110 °C for 10 min.
- 10. Spin coat the prepared adhesive liquid (3000 rpm, 45 s) on the substrate and soft-bake at 110 °C for 4 min.
- 11. Transfer print red LEDs from the source wafer onto the processed substrate with PDMS stamps.
- 12. Cure under to UV for 1 h and bake at 110 °C for 1 h.
- Photoresist (SPR220-v3.0 on the device) clean by reactive ion etching with reactive ion etching (O₂, 100 sccm, 90 mTorr, 150 W) for 12 min.

red LED encapsulation

- 14. Clean the processed wafer in step 13 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 15. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 16. Spin coat SU8-2002 epoxy (500 rpm / 5 s, 3000 rpm / 30 s).
- 17. Soft-bake at 65 °C for 1 min and 95 °C for 1 min.
- 18. Pattern SU-8 to expose contact pads of LED by exposing with UV lithography with a dose of 100 mJ/cm².
- 19. Post-bake at 65 °C for 1 min and 95 °C for 2 min.
- 20. Develop in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.
- 21. Hard bake at 110 °C for 20 min.

red LED metallization

- 22. Clean the processed wafer in step 23 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 23. Pattern PR AZ nLOF 2070.
- 24. Deposit 10 nm / 600 nm / 200 nm of Cr / Cu / Au by sputter coater.
- 25. Lift-off PR in acetone.

Transfer filters

- 26. Clean the processed sample in step 25 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 27. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 28. Spin coat SU8-3005 epoxy (500 rpm / 5 s, 3000 rpm / 30 s).
- 29. Soft-bake at 65 °C for 30 s.
- 30. Transfer print filters on top of red LEDs with PDMS stamps.
- 31. Pattern epoxy by exposing with UV lithography tools with a dose of 150 mJ/cm^2 .
- 32. Post-bake at 65 °C for 1 min and 95 °C for 3 min.
- 33. Develop the exposed area in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.
- 34. Hard-bake at 110 °C for 20 min.

Transfer green LEDs

- 35. Clean the processed sample in step 34 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 36. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 37. Spin coat SU8-3005 epoxy (500 rpm / 5 s, 3000 rpm / 30 s).
- 38. Soft-bake at 65 °C for 30 s.
- 39. Transfer print green LEDs on top of red LEDs with PDMS stamps.
- 40. Pattern epoxy by exposing with UV lithography tools with a dose of 150 mJ/cm².
- 41. Post-bake at 65 °C for 1 min and 95 °C for 3 min.
- 42. Develop the exposed area in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.
- 43. Hard-bake at 110 °C for 20 min.

Green LED metallization

- 44. Clean the processed sample in step 43 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 45. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 46. Spin coat SU8-3005 (500 rpm / 5 s, 3000 rpm / 30 s).
- 47. Soft-bake at 65 °C for 1 min and 95 °C for 3 min.
- 48. Pattern SU-8 to expose contact pads of LEDs by exposing with UV lithography with a dose of 150 mJ/cm².
- 49. Post-bake at 65 °C for 1 min and 95 °C for 3 min.
- 50. Develop in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.

- 51. Hard bake at 110 °C for 20 min.
- Clean the processed wafer in step 51 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 53. Pattern PR AZ nLOF 2070.
- 54. Roughen surface by reactive ion etching (O₂, 100 sccm, 90 mTorr, 150 W) for 10s.
- 55. Deposit 10 nm / 600 nm / 200 nm of Cr / Cu / Au by sputtering.
- 56. Lift-off PR in acetone.

Transfer blue LEDs

- 57. Clean the processed sample in step 56 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 58. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 59. Spin coat SU8-3005 epoxy (500 rpm / 5 s, 3000 rpm / 30 s).
- 60. Soft-bake at 65 °C for 30 s.
- 61. Transfer printing blue LEDs on top of green LEDs with PDMS stamps.
- 62. Pattern epoxy by exposing with UV lithography tools with a dose of 150 mJ/cm².
- 63. Post-bake at 65 °C for 1 min and 95 °C for 3 min.
- 64. Develop the exposed area in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.
- 65. Hard-bake at 110 °C for 20 min.

Blue LED metallization

66. Clean the processed sample in step 65 (acetone, IPA, DI water) and dehydrate at 110 °C

for 10 min.

- 67. Expose to ultraviolet induced ozone (UV Ozone) for 10 min.
- 68. Spin coat SU8-3005 (500 rpm / 5 s, 2000 rpm / 30 s).
- 69. Soft-bake at 65 °C for 1 min and 95 °C for 3 min.
- 70. Pattern SU-8 to expose contact pads of LEDs by UV lithography with a dose of 150 mJ/cm².
- 71. Post-bake at 65 °C for 1 min and 95 °C for 3 min.
- 72. Develop in propylene glycol monomethyl ether acetate (PGMEA) for 1 min and rinse with IPA.
- 73. Hard bake at 110 °C for 20 min.
- 74. Clean the processed sample in step 73 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 75. Pattern PR AZ nLOF 2070.
- 76. Roughen surface by reactive ion etching (O₂, 100 sccm, 90 mTorr, 150 W) for 10s.
- 77. Deposit 10 nm / 600 nm / 200 nm of Cr / Cu / Au by sputtering.
- 78. Lift-off PR in acetone.

Device encapsulation and laser milling

- 79. Clean the processed sample in step 78 (acetone, IPA, DI water) and dehydrate at 110 °C for 10 min.
- 80. Expose to ultraviolet induced ozone (UVO) for 10 min.
- 81. Pattern SU8-3005 epoxy (500 rpm / 5 s, 3000 rpm / 30 s), cure under to UV for 150 mJ / cm² and bake at 100 °C for 30 min.

Device characterization

Current-voltage characteristics of micro-LEDs are recorded using a computer-controlled Keithley 2400 source meter. LED electroluminescence is measured with a spectrometer (HR2000+, Ocean Optics). EQE is measured using a spectroradiometer system with integrating sphere (LabSphere). For frequency response measurement, LEDs are modulated at different frequencies (0.001–100 MHz) by a function generator. The light is detected with a silicon detector (Thorlabs, PAD 10), and the signals are transferred to an oscilloscope (Tektronix, DSA 71254C). The FIB-SEM image is captured by with an electron microscope (ZEISS Auriga SEM / FIB Crossbeam System, Germany). The optical microscopy images are taken by a optical microscope MC-D800U(C). The transmittance spectra of the optical filters are measured using a UV-vis-IR spectrophotometer (Cary 5000, Varian). Typical scans are performed from 400 nm to 800 nm with a 2 nm resolution at every 5° from 0° to 70°. For angular dependent emissive profiles, devices are mounted onto a goniometer scanning from -90° to 90° at a step of 5° , at an injection current of 3 mA. The emission intensity is captured by a standard Si photodetector (DET36A, Thorlabs). Fluorescence images of the stacked RGB micro-LED array are captured by an Olympus IX53 microscope equipped with a Xenon arc lamp, in which the excitation light and the emission light pass through a set of fluorescence filter combinations (red: EX AT540/25×, BS AT565DC, EM AT605/55m; green: EX AT480/30×, BS AT505DC, EM AT535/40m; blue: EX AT375/28×, BS AT415DC, EM AT460/50m; Chroma Tech. Corp.).

LED luminance and current efficiency

The luminous flux of micro-LEDs, Φ_{lum} , is obtained from the radiometric light power using the equation

$$\Phi_{\rm lum} = 683 \frac{\rm lm}{\rm W} \int_{\lambda} V(\lambda) P(\lambda) d\lambda$$
(6)

where $P(\lambda)$ is the power spectral density measured by the integrating sphere (LabSphere), i.e. the light power emitted per unit wavelength, and the refactor 683 lm/W is a normalization factor. $V(\lambda)$ is the conversion between radiometric and photometric units, which is provided by the luminous efficiency function or eye sensitivity function. This function is referred to the *CIE 1931 V(\lambda) function*.

Luminance is the luminous flux per steradian (sr) per chip (here we assume our micro-LEDs have an ideal Lambertian emission profile):

Luminance =
$$\frac{\Phi_{\text{lum}}}{sr \times area} = 683 \times \pi \times \frac{\text{lm}}{\text{W} \times \text{area}} \int_{\lambda} V(\lambda) P(\lambda) d\lambda$$
 (7)

$$CE = \frac{\Phi_{\text{lum}}}{sr \times I} = 683 \times \pi \times \frac{\text{lm}}{\text{W} \times \text{I}} \int_{\lambda} V(\lambda) P(\lambda) d\lambda$$
(8)

For a given power-spectral density $P(\lambda)$, the degree of stimulation required to match the color of $P(\lambda)$ is given by

$$X = \int_{\lambda} \bar{x}(\lambda) P(\lambda) d\lambda \tag{9}$$

$$Y = \int_{\lambda} \overline{y}(\lambda) P(\lambda) d\lambda$$
 (10)

$$Z = \int_{\lambda} \bar{z}(\lambda) P(\lambda) d\lambda \tag{11}$$

The chromaticity coordinates x and y are calculated from the tristimulus values according to

$$x = \frac{X}{X + Y + Z} \tag{12}$$

$$y = \frac{Y}{X + Y + Z} \tag{13}$$

Thus, the value of a chromaticity coordinate is the stimulation of each primary light (or of each type of retinal cone) divided by the entire stimulation $(X + Y + Z)^{15}$.

References

1. Ferrini R, Guizzetti G, Patrini M, Parisini A, Tarricone L, Valenti B. Optical functions of InGaP/GaAs epitaxial layers from 0.01 to 5.5 eV. The European Physical Journal B - Condensed Matter 2002, 27(4): 449-458.

2. Zhang M, Bhattacharya P, Singh J, Hinckley J. Direct measurement of auger recombination in $In_{0.1}Ga_{0.9}N/GaN$ quantum wells and its impact on the efficiency of $In_{0.1}Ga_{0.9}N/GaN$ multiple quantum well light emitting diodes. Applied Physics Letters 2009, 95(20).

3. Walker AW, Schon J, Dimroth F. Extracting Nonradiative Parameters in III–V Semiconductors Using Double Heterostructures on Active p-n Junctions. IEEE Journal of Photovoltaics 2018, 8(2): 633-639.

4. Boroditsky M, Gontijo I, Jackson M, Vrijen R, Yablonovitch E, Krauss TF, et al. Surface Recombination Measurements on III-V Candidate Materials for Nanostructure Light-Emitting Diodes. Journal of Applied Physics 2000, 87(7): 3497-3504.

5. Pearton SJ. Comparison of surface recombination velocities in InGaP and AlGaAs mesa diodes. Journal of Vacuum Science & Technology B 1994, 12(1).

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6. Muth JF, Lee JH, Shmagin IK, Kolbas RM, Casey HC, Keller BP, et al. Absorption coefficient, energy gap, exciton binding energy, and recombination lifetime of GaN obtained from transmission measurements. Applied Physics Letters 1997, 71(18): 2572-2574.

7. Hu Z, Nomoto K, Song B, Zhu M, Qi M, Pan M, et al. Near unity ideality factor and Shockley-Read-Hall lifetime in GaN-on-GaN p-n diodes with avalanche breakdown. Applied Physics Letters 2015, 107(24): 243501.

8. Oh J, Lee S, Moon Y, Moon JH, Park S, Hong KY, et al. Light output performance of red AlGaInP-based light emitting diodes with different chip geometries and structures. Optics Express 2018, 26(9): 11194-11200.

9. Olivier F, Tirano S, Dupre L, Aventurier B, Largeron C, Templier F. Influence of sizereduction on the performances of GaN-based micro-LEDs for display application. Journal of Luminescence 2017, 191: 112-116.

10. Geum D-M, Kim SK, Kang C-M, Moon S-H, Kyhm J, Han J, et al. Strategy toward the fabrication of ultrahigh-resolution micro-LED displays by bonding-interface-engineered vertical stacking and surface passivation. Nanoscale 2019, 11(48): 23139-23148.

11. Ding H, Lu L, Shi Z, Wang D, Li L, Li X, et al. Microscale optoelectronic infrared-tovisible upconversion devices and their use as injectable light sources. Proceedings of the National Academy of Sciences 2018, 115(26): 6632-6637.

12. Li L, Liu C, Su Y, Bai J, Wu J, Han Y, et al. Heterogeneous Integration of Microscale GaN Light-Emitting Diodes and Their Electrical, Optical, and Thermal Characteristics on Flexible Substrates. Advanced Materials Technologies 2018, 3(1): 1700239.

13. Liu C, Zhang Q, Wang D, Zhao G, Cai X, Li L, et al. High Performance, Biocompatible Dielectric Thin-Film Optical Filters Integrated with Flexible Substrates and Microscale Optoelectronic Devices. Advanced Optical Materials 2018, 6(15).

14. Kim T-i, Kim MJ, Jung YH, Jang H, Dagdeviren C, Pao HA, et al. Thin Film Receiver
Materials for Deterministic Assembly by Transfer Printing. Chemistry of Materials 2014, 26(11):
3502-3507.

15. Schubert, E. Fred, Thomas Gessmann, and Jong Kyu Kim. Light emitting diodes. Kirk-Othmer Encyclopedia of Chemical Technology (2000).

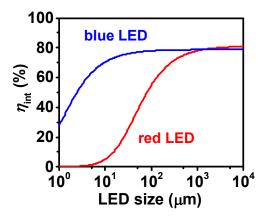


Figure S1. Theoretically modeled performance (internal quantum efficiencies) for an InGaN based blue LED and an InGaP based red LED, as a function of the LED size. Here we assume both LEDs have a square shape.

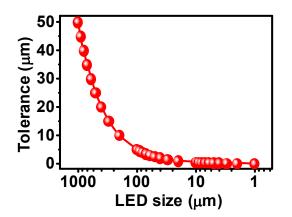


Figure S2. Estimated alignment tolerance for errors during LED microfabrication and transfer printing, as a function of the LED size. Here the tolerance is assumed to be within $\pm -5\%$ of the LED size.

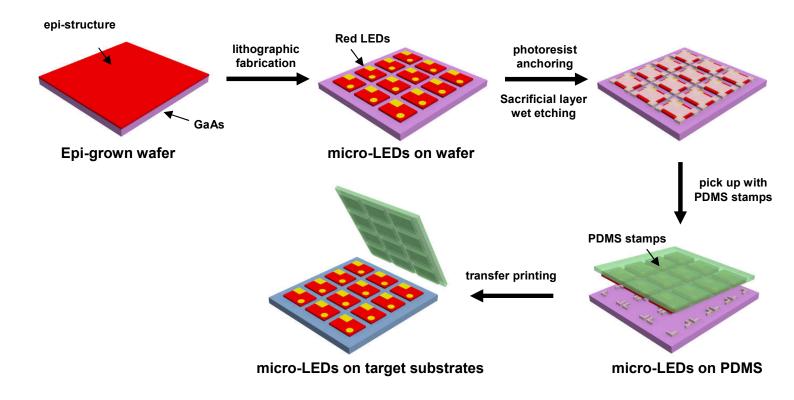
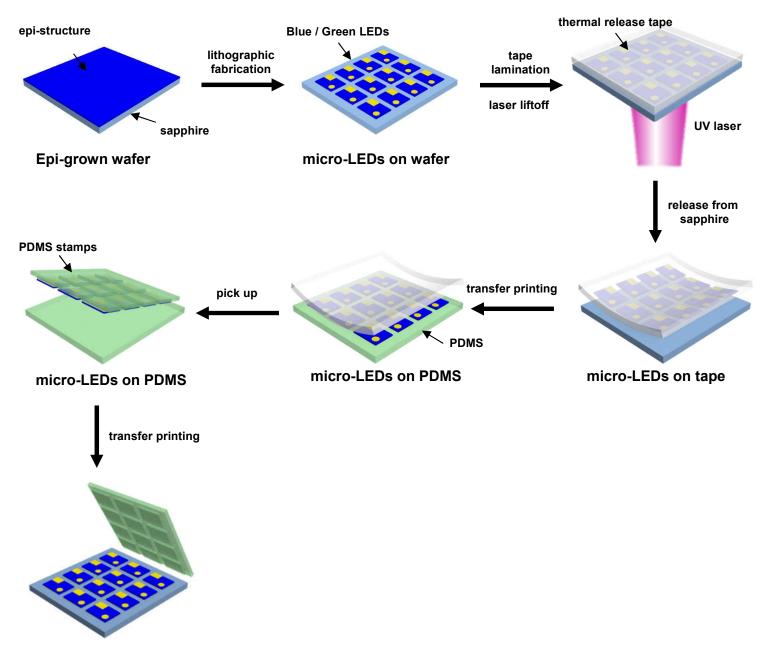


Figure S3. Schematic illustration of the process flow for fabricating and transfer printing InGaP based red LEDs, including epitaxial growth, lithographic fabrication, sacrificial layer etching, PDMS pick up and transfer printing.



micro-LEDs on target substrates

Figure S4. Schematic illustration of the process flow for fabricating and transfer printing InGaN based blue and green LEDs, including epitaxial growth, lithographic fabrication, laser lift-off, thermal release tape release, PDMS pick up and transfer printing.

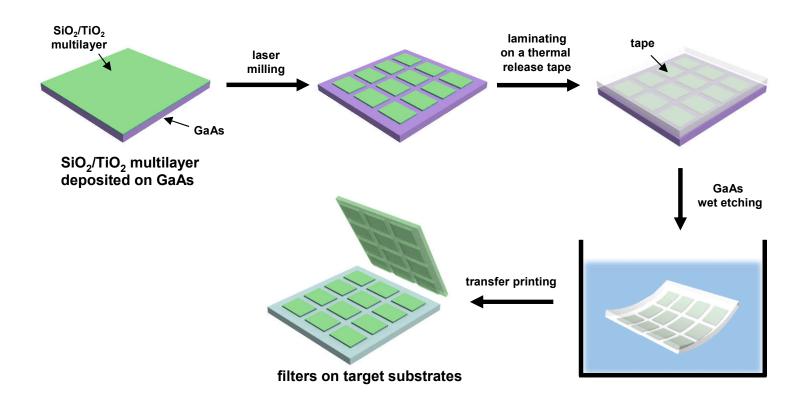


Figure S5. Schematic illustration of the process flow for fabricating and transfer printing SiO_2/TiO_2 based multilayered optical filters, including oxide deposition, laser milling, GaAs substrate etching and transfer printing.

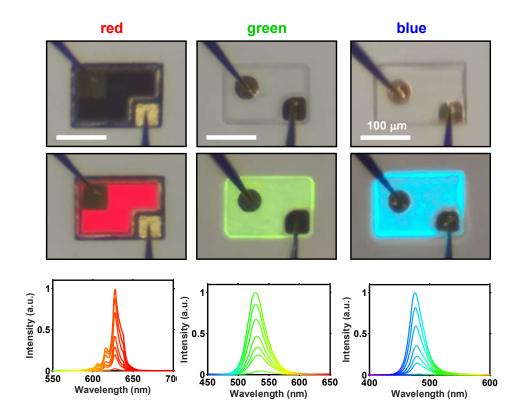


Figure S6. Microscope images of individual red (right), green (middle) and blue (right) micro-LEDs with (top) and without (middle) electroluminescence, and their corresponding electroluminescence spectra with varied injected currents.

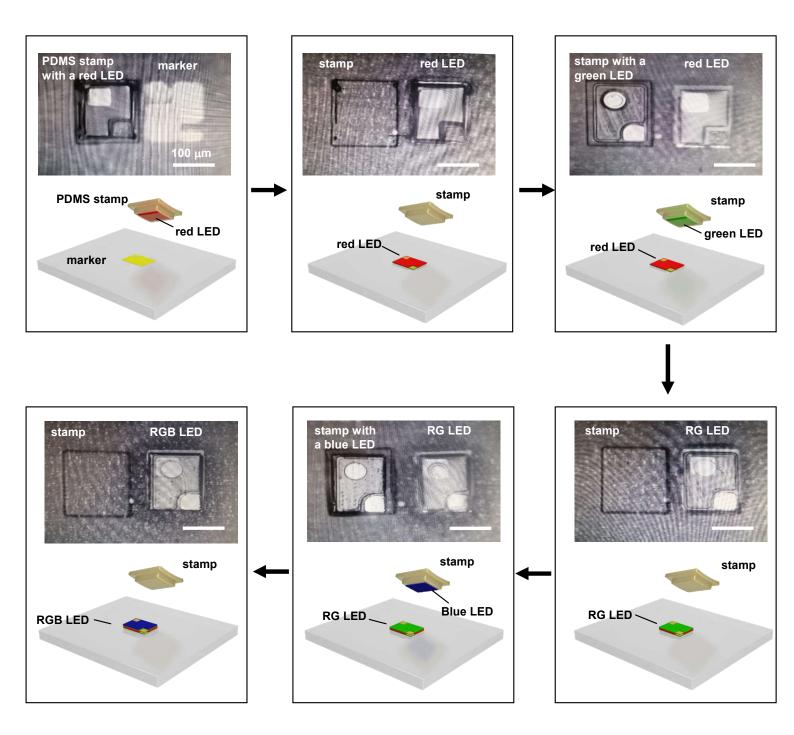


Figure S7. Microscope images and corresponding schematic illustration of the process flow for fabricating a tandem RGB micro-LEDs structure by transfer printing, here the transfer process of the thin-film filter layer is not included, in order to display the LED structures.

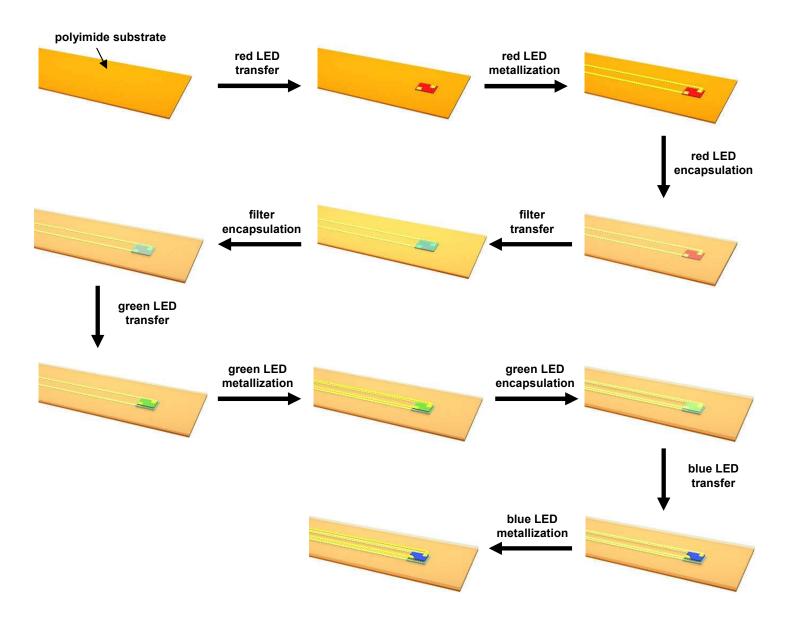


Figure S8. Schematic illustration of the process flow for fabricating the tandem RGB micro-LEDs.

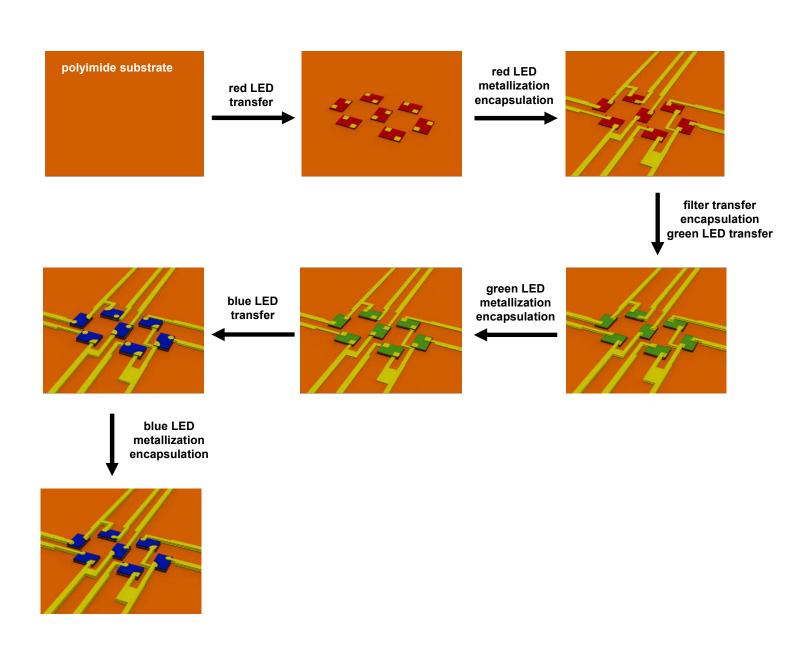


Figure S9. Schematic illustration of the process flow for fabricating an array of tandem RGB micro-LEDs.

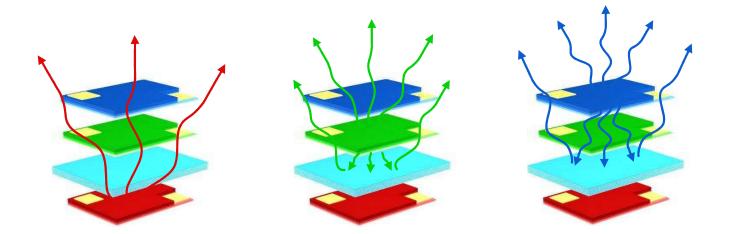


Figure S10. Schematic illustration of the irradiation from the red, green and blue micro-LEDs in a tandem stack with a thin-film filter.

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Figure S11. Images of various decimal numerals shown by an array of tandem RGB micro-LEDs forming a seven-segmented display.

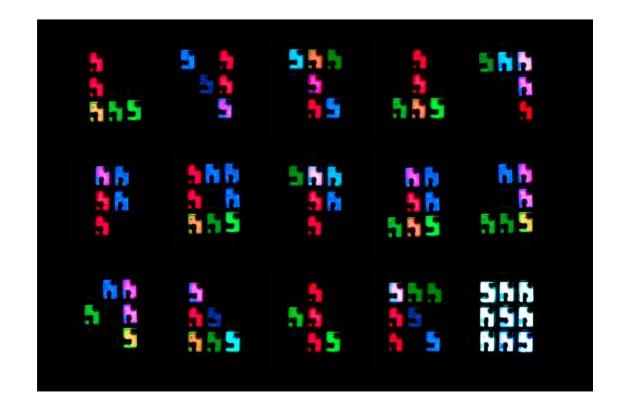


Figure S12. Images of different patterns displayed with a 3*3 array of tandem RGB micro-LEDs.



materials	thickness (nm)	doping (cm ⁻³)	dopant
p++ GaP contact	200	1e20	С
p+ GaP window	2000	5e18	Mg
p InAIP	800	1e18	Mg
InAIP / InGaP MQWs	200	-	-
n InAIP	200	8e17	Si
In _{0.5} Al _{0.5} P / In _{0.5} Al _{0.25} Ga _{0.25} P reflector 12 loops	1200	3e18	Si
n+ GaAs contact	1000	6e18	Si
Al _{0.95} Ga _{0.05} As sacrificial	500	-	-
GaAs substrate	-	-	-

Table S1. Epitaxial structure of the InGaP red LED wafer. The arrow indicates the light emission direction for the red LED, which is unidirectional because of the $In_{0.5}Al_{0.5}P / In_{0.5}Al_{0.25}Ga_{0.25}P$ multilayered reflector underneath the InAlP / InGaP MQWs.



materials	thickness (nm)	
ΙΤΟ	230	
p+ GaN	120	
Electron blocking layer	20	
MQWs	130	
Strained layers	300	
n+ GaN	3000	
u GaN	3500	
sapphire substrate	-	



Table S2. Epitaxial structure of the InGaN blue LED wafer. The arrow indicates the light emission direction for the blue LED, which is bidirectional.



materials	thickness (nm)
ΙΤΟ	230
p+ GaN + MQWs	800
n+ GaN	2100
u GaN	4000
sapphire substrate	-

Table S3. Epitaxial structure of the InGaN green LED wafer. The arrow indicates the light emission direction for the green LED, which is bidirectional.

materials	thickness (nm)	
TiO ₂	19	
SiO ₂	89	} 15 periods
TiO ₂	52	for the periods
SiO ₂	63	
TiO ₂	66	
SiO ₂	73	<pre> 22 periods</pre>
TiO ₂	42.5	\int 22 periods
SiO ₂	154	
GaAs substrate	-	

Table S4. Structure of the multilayered dielectric optical filter deposited on GaAs.

Movie S1



Movie S1. Video for a single pixel of tandem RGB micro-LEDs assembled on a polyimide substrate, displaying different colors under current injection.

Movie S2



Movie S2. Video for an array of tandem RGB micro-LEDs forming a seven-segmented display, showing various decimal numerals.

Movie S3



Movie S3. Video for a 3×3 array of tandem RGB micro-LEDs, displaying various colored patterns.