

I. GaN directly grown on the h-BN release layer

Figure S1 shows X-ray diffraction with the $2\theta/\omega$ configuration and an optical microscopy image for the GaN directly grown on the h-BN release layer. We supply the total amount of trimethylgallium to grow GaN film with the nominal thickness of 3 μm . X-ray diffraction from GaN (10 $\bar{1}$ 0), (0002), and (10 $\bar{1}$ 1) planes was observed, indicating the crystal structure of the GaN layer is polycrystalline. In addition, the GaN surface has isolated-GaN-island morphology, where the GaN islands cannot coalesce with each other.

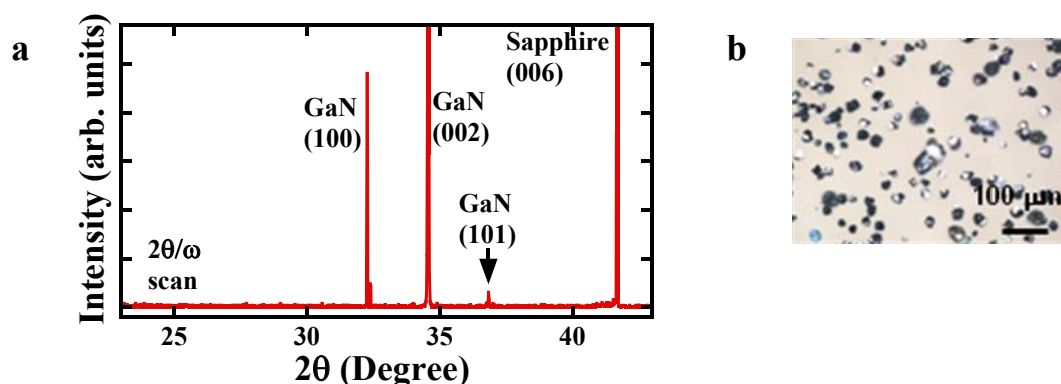


Figure S1 | **a**, X-ray diffraction with $2\theta/\omega$ configuration and, **b**, optical microscopy image for the GaN layer grown on the h-BN release layer.

II. Orientation relationship between GaN, AlN, h-BN and sapphire substrate

The crystallographic in-plane orientation relationship between the GaN film and sapphire substrate was investigated using the selected-area diffraction (SAD) patterns obtained by the TEM. The patterns for the substrate and the GaN film are shown in Fig. S2a and S2b, respectively. The incident electron beam azimuth was parallel to $[\bar{1}2\bar{1}0]_{\text{sapphire}}$ in Fig. S2a and the azimuth was $[1\bar{1}00]_{\text{GaN}}$ in Fig. S2b. From the arrangement of spots from the GaN and sapphire substrate, the $[11\bar{2}0]_{\text{GaN}}$ direction is parallel to $[\bar{3}030]_{\text{sapphire}}$ direction, that is, the $[11\bar{2}0]_{\text{GaN}}$ direction is rotated by 30° from the $[11\bar{2}0]_{\text{sapphire}}$ direction.

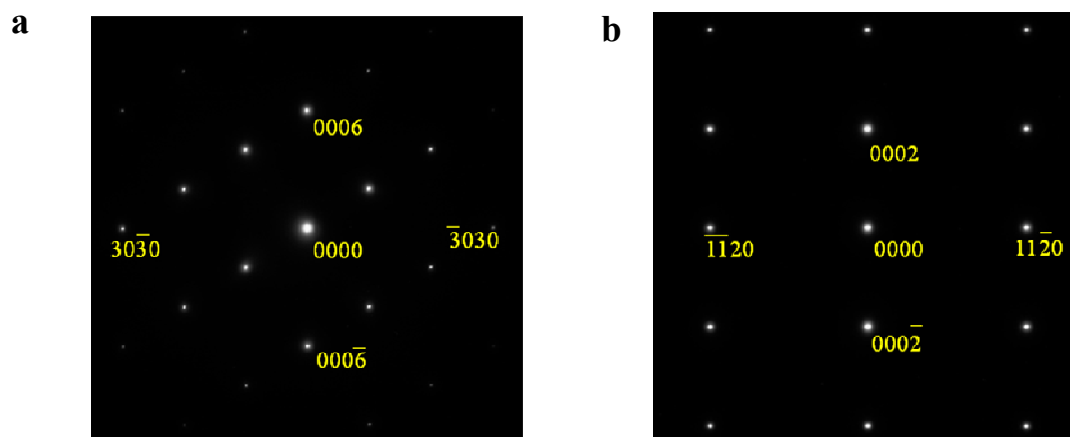


Figure S2 | SAD patterns of **a**, the sapphire substrate, **b**, the GaN film.

Figure S3a is a SAD of the h-BN/sapphire interface area. The incident electron beam azimuth was parallel to $[\bar{1}2\bar{1}0]_{\text{sapphire}}$. From the arrangement of spots from the h-BN and sapphire substrate, the orientation relationship between sapphire and h-BN can be identified as $(0001)_{\text{sapphire}} \parallel (0001)_{\text{h-BN}}$ and $[\bar{1}2\bar{1}0]_{\text{sapphire}} \parallel [1\bar{1}00]_{\text{h-BN}}$. Figure S3b is a SAD of the AlN/h-BN interface area. The incident electron beam azimuth was parallel to $[1\bar{1}00]_{\text{AlN}}$. From the arrangement of spots from the AlN and the h-BN, the orientation relationship between AlN and h-BN can be identified as $(0001)_{\text{AlN}} \parallel (0001)_{\text{h-BN}}$ and $[1\bar{1}00]_{\text{AlN}} \parallel [1\bar{1}00]_{\text{h-BN}}$. Therefore, the orientation relationship between GaN, AlN, h-BN, and sapphire can be also identified as $(0001)_{\text{sapphire}} \parallel (0001)_{\text{h-BN}} \parallel (0001)_{\text{AlN}} \parallel (0001)_{\text{GaN}}$ and $[\bar{1}2\bar{1}0]_{\text{sapphire}} \parallel [1\bar{1}00]_{\text{h-BN}} \parallel [1\bar{1}00]_{\text{AlN}} \parallel [1\bar{1}00]_{\text{GaN}}$.

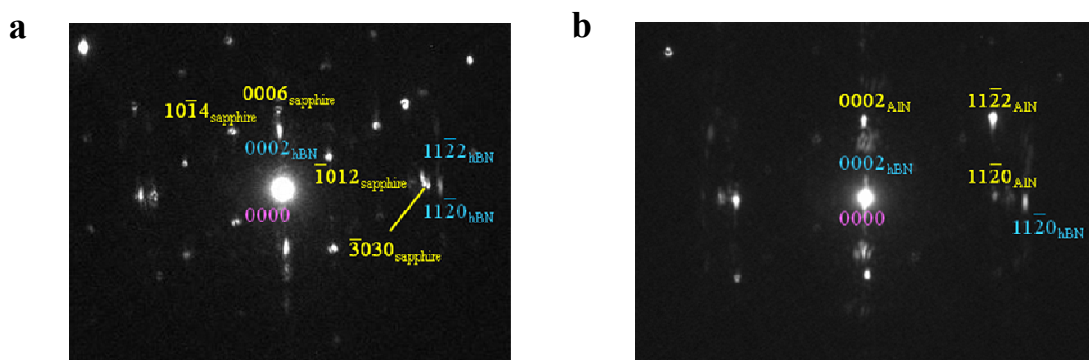


Figure S3 | SAD patterns of **a**, the h-BN/sapphire interface area, **b**, the AlN/h-BN interface area.

III. Dislocation distribution in the GaN film

From the cross-sectional TEM images in Fig. 2d and 2e, we conclude that the GaN film grown on the AlN contains three types of threading dislocations: edge, screw, and mixed. The edge and screw dislocations have Burgers vectors ($\mathbf{b}_e = \langle 11\bar{2}0 \rangle$ and $\mathbf{b}_s = [0001]$, respectively). In Fig. 2d, only the screw and mixed dislocations are visible and only edge and mixed dislocations are seen in Fig. 2e. We counted the number of dislocations and found that the mixed dislocations are the predominant ones in our sample ($8.6 \times 10^9 \text{ cm}^{-2}$). The edge and screw dislocation densities are $5.8 \times 10^9 \text{ cm}^{-2}$ and $2.2 \times 10^9 \text{ cm}^{-2}$, respectively. The total dislocation density is $1.6 \times 10^{10} \text{ cm}^{-2}$, which is two orders of magnitude higher than that in state-of-the-art GaN-based LEDs, which have typical dislocation densities in the low 10^8 cm^{-2} .

IV. GaN film grown on the AlGaIn layer on the h-BN release layer

Instead of the AlN on the h-BN release layer, we grew a wurtzite AlGaIn layer on the h-BN and then grew a GaN film on the AlGaIn (Fig. S3a). As seen for the GaN grown on the AlN on the h-BN release layer, the optical microscopy image of the GaN surface grown on the AlGaIn in Fig. S3b shows a flat and coalesced GaN surface and X-ray diffraction with the $2\theta/\omega$ configuration exhibits only GaN and AlGaIn (0002) diffraction peaks (Fig. S3c), indicating (0001) single-crystal GaN film grows on the (0001) AlGaIn on the h-BN layer. The Al content in the AlGaIn determined by the XRD is 16 %. Hence, the AlGaIn with the Al content of over 10 % grown on the h-BN release layer allows us to grow a flat single-crystal GaN layer. We performed X-ray diffraction pole figure measurement of the GaN (10 $\bar{1}$ 1) plane. Figure S3d shows GaN (10 $\bar{1}$ 1) diffraction peaks at $\Psi \sim 62^\circ$ with hexagonal symmetry, which is nearly equal to the angle between the GaN (10 $\bar{1}$ 1) and (0001) planes. The optical microscopy image, XRD, and XRD pole figure confirm that single-crystal wurtzite GaN also grows on the AlGaIn on the h-BN release layers.

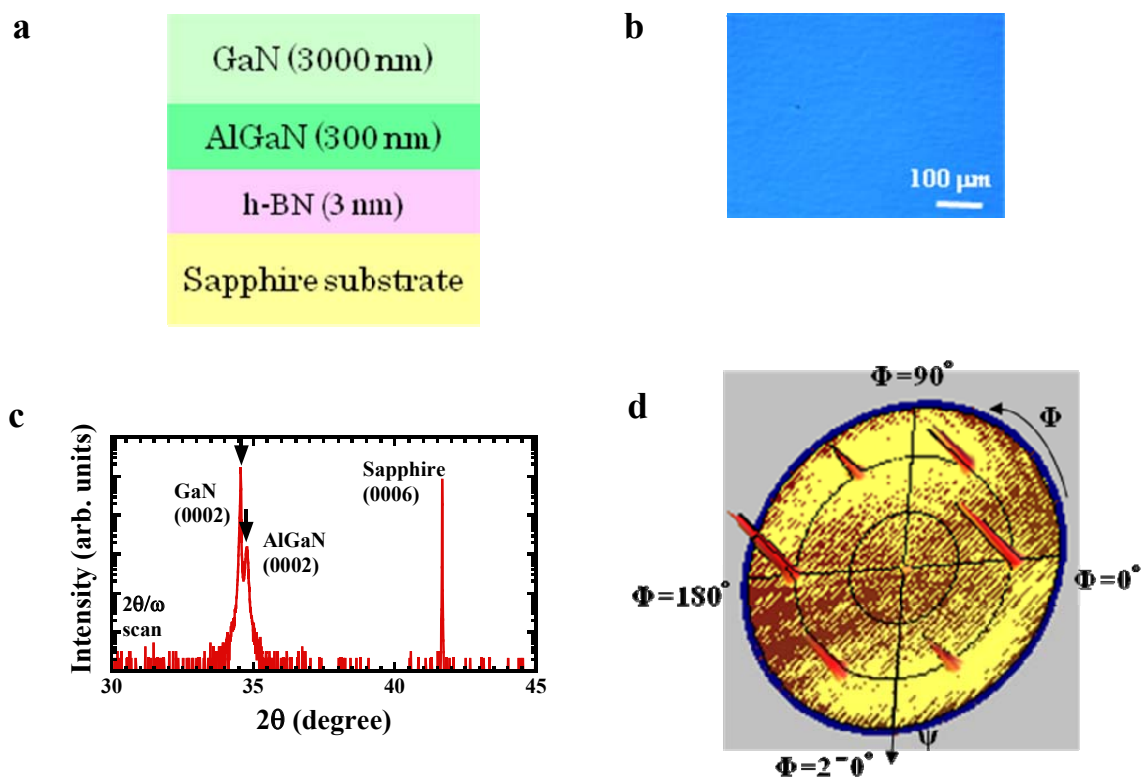


Figure S4 | Flat single-crystal wurtzite GaN is grown on the AlGaN on the h-BN release layer. **a**, Schematic illustration, **b**, optical microscopy image, **c**, X-ray diffraction with $2\theta/\omega$ configuration for the GaN film, **d**, X-ray diffraction pole figure for the GaN film.

V. The separated surfaces after the release

We performed X-ray photoelectron spectroscopy (XPS) measurement using Al K α (1486.6 eV) as the X-ray source (beam spot $\sim$ 2 mm). The escape depth of photoelectrons is estimated to be a few nanometers. Figures S5a and S5b show B 1s and N 1s XP spectra, respectively, obtained for the surface of the transferred film and the counter-surface of the sapphire substrate after the mechanical release. The position of the B 1s peak is same for both surfaces. The binding energy (BE) of 190.8 eV is close to the reported value for B-N bonds in h-BN. Identical results were obtained also in N 1s XPS: the N 1s peak position ($\sim$ 398.1 eV BE) is same for both surfaces. These results indicate that the separation of the film actually takes place at the h-BN release layer, as illustrated in Fig. 1d. The layered h-BN, in which honeycomb layers are weakly bound to each other via van der Waals forces like graphite, acts as a functional release for the mechanical transfer of the MQW film from the host sapphire substrate onto the foreign sapphire substrates.

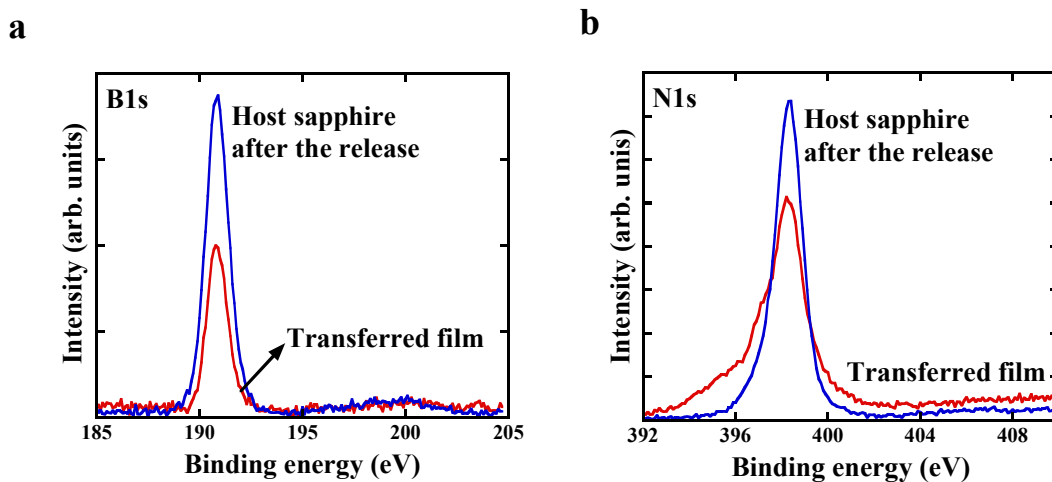


Figure S5 | **a**, B 1s XPS spectra for the surface of the transferred MQW film and the counter-surface of the sapphire substrate and, **b**, same for N 1s XPS spectra.

VI. Conventional and our LED structures

The LED structures, consisting of a 0.3- μm -thick Si-doped AlGaN layer, 3- μm -thick Si-doped GaN layer, ten-periods InGaN/GaN MQW structure, and 0.1- μm -thick Mg-doped GaN layer, were grown on a typical LT-AlN buffer layer and on the h-BN release layer. The electron and hole concentrations in the Si-doped and Mg-doped GaN layers were 2×10^{18} and $3 \times 10^{17} \text{ cm}^{-3}$, respectively. For the transferred MQW LED, after the standard fabrication process, we mechanically released the LED structure from the sapphire substrate without any supporting substrate. Then, we mounted it onto the indium sheet [Fig. S6a]. We fabricated the conventional MQW LED by the standard fabrication process without lift-off [Fig. S6b]. For the vertical-type transferred MQW LED, after the LED structure had been released, the Al/Au electrodes were deposited on the back side of the LED. Then, we mounted it onto the indium sheet [Fig. S6c].

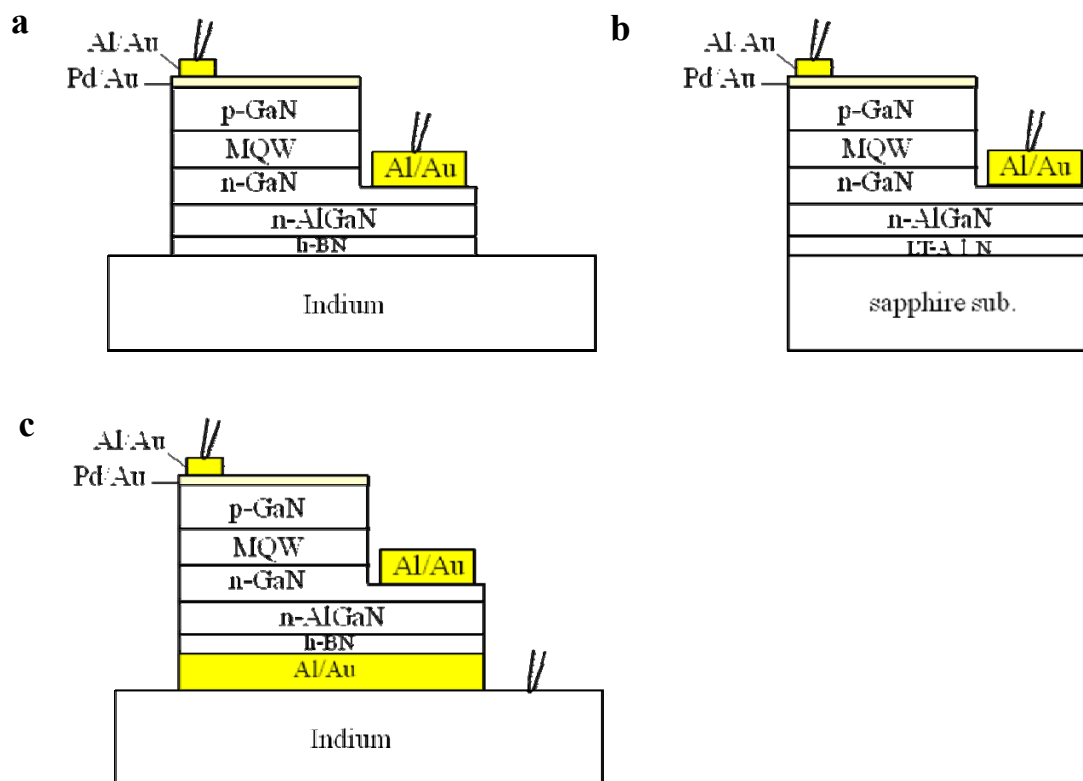


Figure S6 | Schematic illustrations of **a**, the transferred MQW LED, **b**, the conventional MQW LED without lift-off, and **c**, the vertical-type transferred MQW LED.