Supporting Information

Direct Growth of Etch Pit-Free GaN Crystals on

Few-Layer Graphene

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Figure S1 is Raman spectra of ozone-treated graphene and pristine graphene. Small D/G intensity ratio in the pristine graphene indicates high quality of the grown graphene. After ozone treatment in atomic layer deposition (ALD) chamber, D-band was increased significantly. Oxygen-functional groups such as epoxy and/or hydroxyl groups are likely formed due to abundant defective sites which are further functionalized under ambient conditions.



Figure S1. Raman spectra of raw graphene (black) and ozone treated graphene (red). The D/G ratio is indicated in the figure.

Figure S2 is AFM images of pristine graphene (a) and deposited GaNs with various nucleation times (b)-(d). Small agglomerates of GaN with sizes less than 100 nm were formed at early low temperature regime. The number of density of GaN nucleation seeds relied on the nucleation times. The wrinkles and bumpy structures on graphene were also used as nucleation seeds.



Figure S2. GaN nucleation density as indicated by a AFM mode for various nucleation times: (a) raw graphene after ozone treatment, (b) 100 sec. Circle marks the wrinkle, (c) 210 sec, (d) 300 sec. The insets show the 3D morphology.

Figure S3 is the crystal growth orientation and d-spacing value of GaN crystals on SiO₂, Gr/SiO₂, sapphire and Gr/sapphire were measured by x-ray diffraction (XRD). The 2 θ peaks each of the GaN crystals was confirmed at 34.8°, 34.6°, 34.6° and 33.9°. The d-spacing values of each case show 2.57 Å, 2.59 Å, 2.59 Å and 2.64 Å at (0002) diffraction peak. The difference of d-spacing value of 0.02 and 0.05 Å for GaN grown on graphene/SiO₂ and graphene/sapphire compared to GaN grown on SiO₂ and sapphire could be related to strain relaxation. These correspond to the (0002) and (0004) diffraction peaks of wurtzite GaN.



Figure S3. XRD θ - 2 θ scan of GaN on (a) SiO₂, (b) Gr/SiO₂, (c) sapphire and (d) Gr/sapphire substrate.

Although the grown GaN was microcrystalline, light emitting diodes(LED) structure was fabricated to demonstrate efficient light emission in microcrystal, as shown in Fig. S4. The GaN-based LED structures were grown on graphene/sapphire substrate by MOCVD. LED structure consists of a Si-doped n-type GaN layer, five-periods of InGaN/GaN multi-quantum wells (MQWs), and a 150 nm thick Mg-doped p-type GaN layer. After cooling down from the thermal cleaning step, a GaN nucleation layer of around 25 nm was deposited at 530 °C on graphene/sapphire substrate. A 2 μ m thick undoped GaN epilayer were grown subsequently at 1040 °C. Thereafter, the temperature was gradually decreased to 750 °C to grow InGaN/GaN MQWs in N₂ ambient. The MQWs consist of five periods of 3 nm thick InGaN well layers and 12 nm thick GaN barrier layers. Finally, the Mg-doped p-type GaN epi layer was grown in H₂ ambient at 950 °C. The local spot of LED was demonstrated in Figure S4.



Figure S4. (a) Schematic diagram of LED structure grown on graphene/sapphire substrate. (b) Optical images of light emission from the as-fabricated LED.