Van der Waals Epitaxy of High Quality AlN towards Deep Ultraviolet Light Emitting Diodes on Monolayer Graphene

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Van der Waals epitaxy of III-nitrides on two-dimensional (2D) materials has attracted significant attention, as it promises significantly reduced dislocation densities compared to conventional heteroepitaxy on lattice mismatched substrates.[1] The resulting epilayers can also be readily exfoliated and transferred to arbitrary substrates for subsequent device processing and integration.[2] To date, however, controlled nucleation of adatoms on the sp2-hybridized 2D surfaces has remained difficult, due to the extremely low surface energy, which limits the formation of large-area single-crystalline epilayers.

Here, we have investigated the epitaxy and characterization of AlN and AlGaN-based heterostructures on monolayer graphene by using plasma-assisted molecular beam epitaxy (MBE). Commercial one monolayer graphene was transferred on sapphire and AlN template substrates to explore the effects of crystallographic information underneath graphene on the epitaxial relationship. The initial adatoms stacking manner and nucleation orientation can follow the epitaxial registry of the AlN template beneath graphene, whereas it is more challenging for graphene covered sapphire to maintain the epitaxial relationship. Without using any pretreatment and buffer layer, we have demonstrated that single-crystalline AlN can be achieved on monolayer graphene covered AlN template (Figure 1). Such AlN has comparable crystal quality and band edge emission with the commercial AlN template. We have further investigated the epitaxy and fabrication of AlGaN-based deep ultraviolet light emitting diodes (DUV-LED) on monolayer graphene. The as-fabricated devices exhibit excellent current-voltage characteristics and strong electroluminescence emission at ~260 nm with a maximum external quantum efficiency (EQE) of ~4%. The device performance is comparable to identical structures grown directly on commercial AlN template.

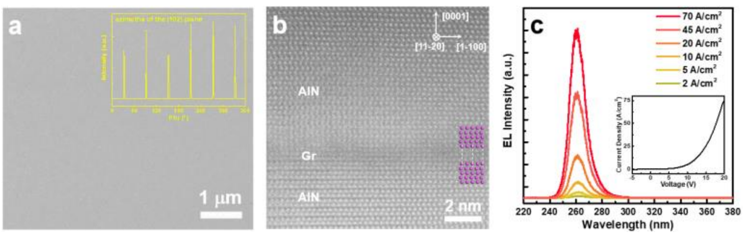

Figure 1. (a) SEM image of AlN grown on monolayer graphene, (b) HAADF-STEM image of AlN/graphene interface, (c) EL spectra of as-fabricated DUV-LED.

[1] Y. Qi et al., J. Am. Chem. Soc. 2018, 140, 11935-11941.
[2] J. kim et al., Nat. Commun. 2014, 5, 4836.

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Supplementary Information

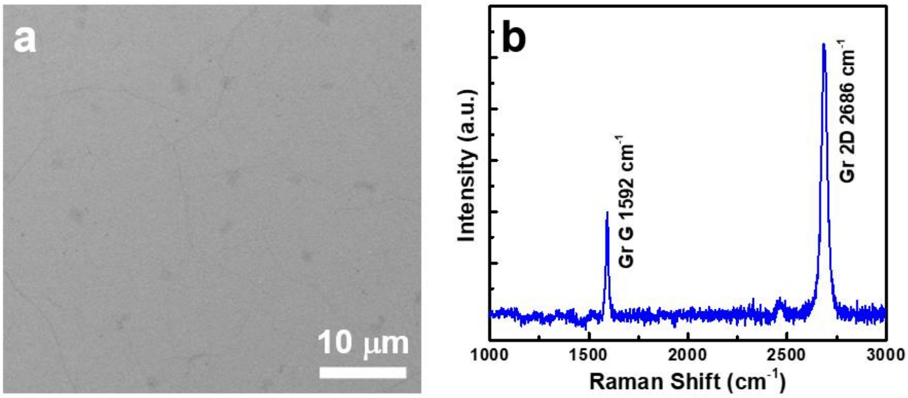

Figure S1. (a) SEM image of transferred graphene from copper foils onto MBE grown AlN/sapphire template, (b) the corresponding Raman spectrum.

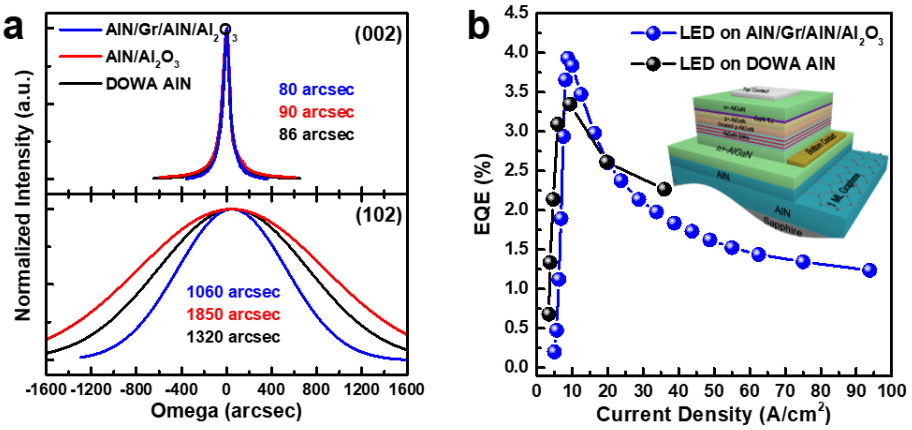

Figure S2. (a) XRD ω-scan rocking curve of (002) and (102) planes for 500 nm AlN/graphene/AlN template, AlN/sapphire grown by MBE, and commercial DOWA 1 μm AlN template. (b) EQE versus current density measured at room temperature for DUV-LED on AlN/Gr/AlN template and commercial DOWA AlN. The inset shows the schematic diagram of AlGaN DUV-LED structure grown on AlN/graphene/AlN template.