Growth of GaN on epitaxial graphene using intercalated GaN interlayer

Seung Hoon Lee[1], Chengye Dong[2], Brian M. Bersch[3], Joshua A. Robinson[1,2], and Joan M. Redwing[1,2]

[1]Department of Materials Science and Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802, USA
[2]2-D Crystal Consortium Materials Research Institute, The Pennsylvania State University, University Park, Pennsylvania 16802, USA
[3]Northrop Grumman Mission Systems, Linthicum Heights, Maryland 21090, USA

Remote epitaxy of GaN on 2D materials such as graphene opens up new opportunities for producing free-standing and transferable films. Epitaxial graphene formed on SiC is a promising substrate for remote epitaxy due to its pristine, high-quality surface. However, the inherently low surface energy of graphene and lack of dangling bonds pose challenges for GaN nucleation, often resulting in widely spaced nuclei that impede the formation of fully coalesced GaN films. Plasma treatment can be used to introduce defects into the graphene, increasing nucleation density. However, this approach often leads to randomly oriented nuclei, degrading the structural quality of the resulting GaN films. An alternative strategy is to modify the surface energetics of graphene through intercalation into the van der Waals gap at the graphene/SiC interface. Previously we demonstrated the formation of a two-dimensional form of GaN at the epitaxial graphene/SiC interface via Ga intercalation and nitridation. In this study, we extend this work to demonstrate the use of such a 2D GaN interlayer to promote oriented GaN nucleation and film formation on epitaxial graphene.

Metalorganic chemical vapor deposition (MOCVD) was used for 2D GaN formation and subsequent epitaxial growth of GaN, using epitaxial buffer graphene grown by sublimation from SiC. Trimethylgallium (TMGa) and UHP ammonia gas (NH_3) were used as Ga and N sources, with hydrogen (H_2) as the carrier gas. The 2D GaN layer was initially formed by heating the epitaxial graphene to 675 °C in a H_2 ambient at 100 Torr. Gallium intercalation was carried out by pulsing the TMGa source (2 secs on/3 secs off) for 100 cycles. The sample was then annealed in NH_3 for 30 minutes to convert the intercalated Ga to 2D GaN. Following intercalation, the temperature was increased to 800 °C under a flow of NH_3 for the GaN epilayer growth. GaN growth was carried out in two stages: a low-temperature (LT) stage at 800 °C and a high-temperature (HT) stage at 1000 °C, both at 50 Torr and V/III=12267.1. Field emission scanning electron microscopy (FE-SEM) and atomic force microscopy (AFM) were used to analyze the morphology and nucleation behavior, while Raman spectroscopy and X-ray photoelectron spectroscopy (XPS) confirmed intercalation through C 1s and Si 2p core levels and an I(2D)/I(G) ratio. Crystallinity was assessed using X-ray diffraction (XRD) rocking curve and asymmetric phi scan.

Initial samples were prepared in which the process was stopped immediately after the 2D GaN formation step to confirm intercalation. FESEM and AFM revealed the presence of raised structures, approximately 2-4 nm in height, beneath the graphene, which are consistent with 2D GaN formation reported previously. Raman spectra and XPS provided further evidence of intercalation, including peak shifts of C 1s and Si 2p core levels in XPS and an increase in the I(2D)/I(G) ratio Raman peaks. Nucleation studies, during which the GaN growth was stopped after the 800 °C step, revealed a significant increase in GaN nucleation on 2D GaN intercalated regions compared to non-intercalated regions. The addition of the high temperature 1000 °C step resulted in the formation of well-coalesced regions of GaN aligned along step terraces on the epitaxial graphene surface. XRD measurements indicated that GaN grown on intercalated GaN was epitaxially aligned to the SiC substrate with symmetric and asymmetric XRD rocking curve full width at half-maximum (FWHM) of 420 and 1000 arcsec, respectively. These findings demonstrate the influence of intercalated GaN on the nucleation and crystallinity of GaN grown on graphene. Furthermore, they provide a pathway to address the challenges of epitaxial growth on 2D surfaces such as graphene.

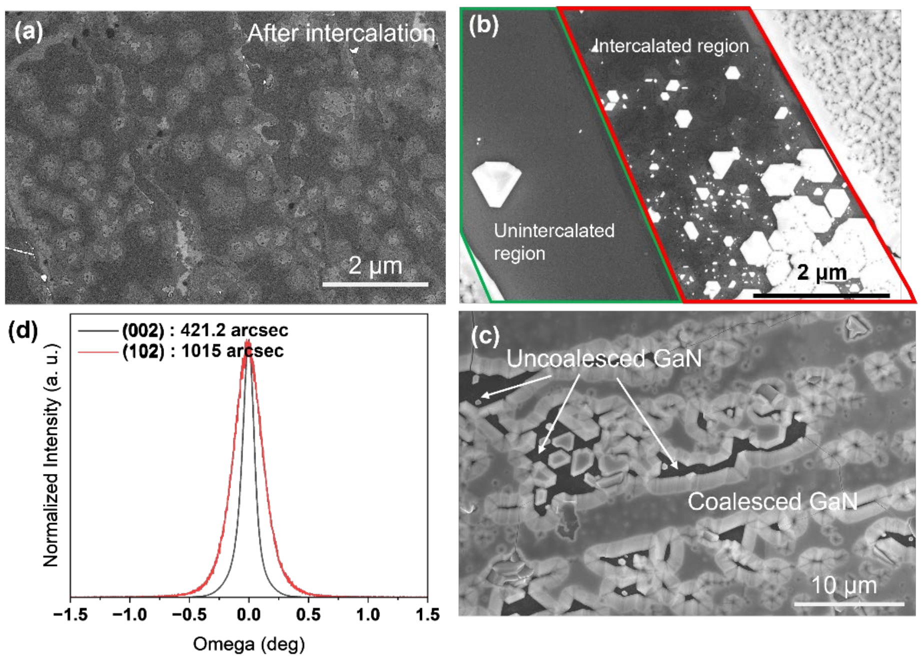

Fig. 1. SEM images of (a) intercalated GaN, (b) low-temperature GaN, and (c) high-temperature GaN on it with (d) symmetric and asymmetric XRD rocking curve of GaN on intercalated GaN. GaN nucleation preferentially occurred on the intercalated region (red) and selective nucleation led to uncoalesced region. Aligned growth of coalesced GaN to a specific direction, which corresponds to the terrace of graphene, was shown and that may be due to the multi-layers of graphene along the step edge, which suppressed nucleation. XRD rocking curve showed the epitaxially grown GaN on intercalated GaN.