Vertical Thinking in Blue Light Emitting Diodes: GaN-on-Graphene Technology

C. Bayram*†

Innovative COmpound semiconductor Laboratory, Department of Electrical and Computer Engineering, University of Illinois at Urbana-Champaign, Urbana, IL USA 61801

IBM Research, T. J. Watson Research Center, Yorktown Heights, NY, USA 10598

H. Park,
Kyungpook National University, 80 Daehakro, Bukgu, Daegu, 702-701, Korea

C. Dimitrakopoulos,
Department of Chemical Engineering, University of Massachusetts Amherst, Amherst, MA USA 01003

ABSTRACT

In this work, we show that a 2D cleave layer (such as epitaxial graphene on SiC) can be used for precise release of GaN-based light emitting diodes (LEDs) from the LED-substrate interface. We demonstrate the thinnest GaN-based blue LED and report on the initial electrical and optical characteristics. Our LED device employs vertical architecture: promising excellent current spreading, improved heat dissipation, and high light extraction with respect to the lateral one. Compared to conventional LED layer release techniques used for forming vertical LEDs (such as laser-liftoff and chemical lift-off techniques), our process distinguishes itself with being wafer-scalable (large area devices are possible) and substrate reuse opportunity.

Keywords: light emitting diode, Gallium Nitride, Graphene

1. INTRODUCTION

Gallium Nitride semiconductor technology is unique as GaN visible light emitting diodes enable high efficiency deep ultraviolet to visible devices thanks to composition-independent direct-bandgap (see Figure 1). Among many applications of light emitting diodes, the most immediate impact is foreseen in solid state lighting.
Solid state lighting (SSL) holds the promise of a more energy-efficient, longer-lasting, more compact, and lower maintenance substitute for today's incandescent and fluorescent light sources. Since lighting currently represents about 22% of all electricity consumption, the adoption of SSL could significantly reduce greenhouse gas emissions. [1] Light-emitting diodes (LEDs) based on In,Ga,N alloys, are currently the most promising candidates for realizing efficient SSL. InGaN is a direct wide bandgap semiconductor with an emission which can span the entire visible spectrum via compositional tuning (see Figure 1). However, InGaN LED performance is highly wavelength-dependent. Indeed, ultra-bright and efficient blue InGaN-based LEDs are readily available [2] but the efficiency of InGaN-based green LEDs is still far from adequate for use in SSL. [3,4,5,6, 7]

![Figure 1 | Bandgap energy versus lattice constant plot of III-V materials. Direct bandgap nature of III-nitrides enables emission tunability from ultraviolet to infrared wavelengths, making them one of the most promising materials for photonics.](image)

One of the major problems of SSL technology is the lack of lattice-matched substrate. Visible light emitting diodes employ InGaN as the active layer material. By changing the indium content, LEDs can be tuned in the entire visible spectra (violet, blue, green, and red as seen in Figure 1). However, such crystalline LED layers need to be grown epitaxially on a crystalline host substrate for practical devices. Conventional choice of such substrates are sapphire (Al,O3), silicon carbide (SiC), and silicon (Si) that have ~13%, ~3%, and ~17% lattice-mismatch with LED layers. This leads to high defect densities (>10⁸/cm²). Defects act as nonradiative recombination centers, and limit the output power, efficiency, and spectral quality of LEDs. Moreover these conventional substrates are either non-conductive (such as sapphire) or non-transparent to visible light (such as SiC and Si) forcing “Lateral LED” formations (i.e. placement of two contacts of the LEDs on one side of the wafer, see Figure 2). However, general illumination applications require high lighting power (~ 100 W) that demands high current injection (~ 50 A/cm²) into the LED such that power density in a LED is 500W/cm² - x5 of that in a CPU. [8] Thus, under general illumination operating conditions, LED junction temperatures reach around 100ºC and this leads to reduction in lamp's lifetime, conversion efficiency, and spectral quality. Thus, it is essential to dissipate the heat generated in LEDs effectively and reduce the junction temperature. In order to enable high output power LEDs, it is important to employ (1) Vertical LED formation where the two contacts are placed on the different sides of the LED hence maximizing current spreading, (2) Lattice-matched freestanding GaN substrates to minimize defectivity and improve overall conversion efficiency, and (3) Engineered layer release techniques to generate the thinnest LEDs for improving thermal budget.
Recently, GaN freestanding (FS) substrates as large as 2-inch diameter become available. [9, 10] GaN FS substrates enable lattice-matched growth for LEDs reducing defectivity orders of magnitude ($<< 10^{6}$ cm$^{-2}$). This reduced defectivity promises LED lifetime to increase an order of magnitude and light power output to triple from what it is today. However, such lattice-matched substrates are expensive, thus conventional LED fabrication and packaging approach are not going to be economically feasible soon.

One way to benefit from GaN FS substrates and reduce the cost is through enabling substrate reuse. Indeed, we have recently demonstrated first of its kind vertical LEDs enabled through mechanical release. [11] The LED device was grown on sapphire substrate and the release of LED layer enabled a vertical LED formation.

Recently, visible light has become essential for bioapplications such as behaviour modification [12], drug delivery and activation [13], and biolaser pumping [14]. Even complex tissues such as brain are shown to be made “transparent” to visible light. [15] These work motivate for wavelength-tunable emitters and detectors that are flexible in nature for in-situ biomanagement. [16] Such thin vertical devices are quite promising for emerging biophotonics and will replace existing fiber-based light delivery techniques. As required visible light output power intensity is low, these bioLEDs might be powered through energy harvesting in-situ [17] or wirelessly [18].

A number of methods to achieve layer transfer have been reported earlier including the laser lift-off [19], chemical lift-off [20, 21, 22, 23], Smartcut [24], and mechanical lift-off [25, 26, 27]. However, the release of thick (several microns) films of III-nitrides (i.e. Al$_x$Ga$_{1-x}$In$_y$N), which has high elastic modulii is challenging since their modulii are x 3-5 higher than most conventional semiconductors. Laser lift-off technique has limited release area (~ mm$^2$) due to energy beam variations of source lasers. Even though successful release of high modulii layers has been demonstrated by the chemical lift-off technique (via selectively etching of underlying sacrificial buffer layers) its manufacturability potential

Figure 2 | Schematic view of lateral and vertical LEDs with table summarizing the major (dis)advantages of both. Vertical LEDs are better suited for high power applications due to higher thermal budget and current handling capabilities whereas lateral LEDs are much easily integrated with flip-chip bonding technology.
for large diameter substrate is limited because lateral etching time increases exponentially with increasing substrate diameter. Similarly, even though large area layer release of GaN is possible by the Smartcut process the maximum thickness of the released layer is limited to < 2 µm because the energy of a high volume commercial H+ ion implanter is limited to < 200 keV. Nonetheless typical GaN devices such as light emitting diodes (LEDs) require release of thicker layers than that. Thus, it is challenging to release high modulus semiconductors such as III-nitrides and silicon carbides while maintaining their original quality.

We have addressed these limitations of existing techniques by developing a cleave layer suitable for mechanical release and applied our technology for enabling light-weight, flexible, and functional blue light emitting diodes (See Figure 3 for the process flow).

**Figure 3 | Schematic of a method for growing/transferring single-crystalline thin films on/from epitaxial graphene. (a) Graphitization of a SiC substrate to form epitaxial graphene. (b) Epitaxial growth of GaN on graphene. (c) Deposition of a stressor layer (Ni). (d) Release of GaN from the substrate with a handling tape. (e) Transfer of the released GaN/Ni/tape stack on a host substrate. (f) Removal of the tape and Ni by thermal release and wet etching, leaving a GaN film on the host substrate. (adapted from [28])**

### 2. EXPERIMENT AND RESULT

Graphene forms weak van der Waals bonding to most materials [29]. Thus, graphene can be an ideal template for growth and transfer of single-crystalline semiconductor films if such films can be epitaxially grown. However, the growth of conventional semiconductors (covalent/ionic bonding) on two-dimensional layered compounds (van der Waals bonding) results in three-dimensional oriented polycrystalline clusters [30]. This is because a weak substrate-overlayer interaction suppresses adatom nucleation leading to a Volmer-Weber growth mode. To date, direct growth of single-crystalline semiconductor films on graphene has not been demonstrated yet although small scale GaN growth on a mechanically exfoliated graphene flake (tens of micron scale) was reported by using nanostructured ZnO nucleation seeds. [31]

Recently, epitaxial graphene formed on a single-crystalline SiC substrate is used as a template. The epitaxial graphene on SiC has a single orientation over the entire substrate. [32] More importantly, the vicinal macro steps of the SiC surface can provide periodic nucleation sites for the thin film. Single-crystalline GaN epitaxy with 3 Å root mean square (RMS) roughness and low defectivity (as low as 4 × 10⁸ cm⁻²) is reported on epitaxial graphene. [28] The entire single-crystalline GaN films were then released from the graphene and transferred on an arbitrary substrate. Further reuse of the
graphene/SiC substrate allowed growth and transfer cycles of GaN films for multiple times. InGaN/GaN epitaxial LED stacks were grown on a recycled graphene/SiC substrate (reused three times), and blue light emission was observed from the released LED stacks.

In this work, we report that graphene grown on SiC can be a template for the growth and transfer of single-crystalline films. We have grown visible LED structures on graphene/SiC substrates and obtained fully-functional vertical blue LED on plastic demonstrating versatility of our technique. The total thickness of the blue LED active layer is ~2.5 µm making it the thinnest inorganic GaN-based LED. This technique could potentially be applied for growing other single-crystalline 3D and 2D materials on graphene and transfer; leading to new class of inorganic thin-film devices.

3. CONCLUSION

Here we employed direct growth of high-quality single-crystalline GaN films on graphene. The GaN film was released and transferred onto arbitrary substrates. The post-released graphene/SiC substrate was reused for multiple growth and transfer cycles of GaN films. By using this technique, fully-functional flexible blue light emitting diodes (LEDs) were demonstrated (see Figure 4). InGaN/GaN epitaxial LED stacks were grown on a recycled graphene/SiC substrate (reused three times), and blue light emission was observed from the released LED stacks (see Figure 4 for research highlights).
REFERENCES


