InGaN-based Flexible Light Emitting Diodes

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ABSTRACT

Novel layer release and transfer technology of single-crystalline GaN semiconductors is attractive for enabling many novel applications including flexible photonics and hybrid device integration. To date, light emitting diode (LED) research has been primarily focused on rigid devices due to the thick growth substrate. This prevented fundamental research in flexible inorganic LEDs, and limited the applications of LEDs in the solid state lighting (due to the substrate cost) and in biophotonics (i.e. optogenetics) (due to LED rigidity). In the literature, a number of methods to achieve layer transfer have been reported including the laser lift-off, chemical lift-off, and Smartcut. However, the release of films of LED layers (i.e. GaN semiconductors) has been challenging since their elastic moduli and chemical resistivity are much higher than most conventional semiconductors. In this talk, we are going to review the existing technologies and new mechanical release techniques (i.e. spalling) to overcome these problems.

Keywords: flexible, light emitting diode, Gallium Nitride, laser lift-off, chemical lift-off, Smartcut, spalling

1. INTRODUCTION

The invention of semiconductor based transistor in 1947 (which was later recognized with the 1956 Nobel Prize in Physics) led to the replacement of vacuum-tube based circuit elements with the semiconductor based ones. This semiconductor revolution in electronics has enabled mass-production of compact, efficient, reliable, and cheap electronic components leading the way to consumer electronics (computers and smart phones), defense and national security technology (radar), health and medicine (medical instruments), advanced manufacturing (machining), communications and information technology (internet), and entertainment tools (Xbox).

In the 21st century, a new semiconductor revolution is in progress, this time in photonics, with the light emitting diode (LED). Since the invention of first practical light emitting diode in 1962, developments in the physics, materials, chemistry, and epitaxial technologies led to the precise control over the LED emission wavelengths. Particularly, visible spectrum (blue, green, red) LEDs have been of interest with the hopes of enabling better lighting alternatives to existing vacuum-based lighting technologies (e.g. incandescence and florescence). This need is mainly driven by the facts that (1) Total annual energy consumption in the United States for lighting is ~ 800 Terawatt-hours costing > $80 billion to the public, and (2) Energy consumed for lighting throughout the world entails to greenhouse gas emission equivalent to 70% of the emissions from all the cars in the world. Thus, a semiconductor revolution in lighting can enable compact, efficient, reliable, and cheap lighting sources, leading to significant societal and environmental advantages. The recent awarding of the 2014 Nobel Prize in Physics to GaN-based “blue” LEDs for white lighting applications is an indicative of this promise.

LEDs are semiconductors in which the light emission comes from a crystalline layer called active layer, sandwiched between an n-type and a p-type layer. When a voltage is applied between the layers, electrons and holes injected into the LED layers recombine and the energy is released in the form of light. The amount of released light energy is strongly dependent on the design of LED layers as well as the design of LED packaging. Today, light-emitting diodes based on InXGa1-X:N alloy are the most promising candidates for solid state lighting (i.e. 2014 Nobel Prize in Physics). InGaN is a direct wide bandgap semiconductor with an emission that can span the useful visible spectrum from 400 nm (violet) up...
to 700 nm via increasing the indium content (x) of the In\textsubscript{x}Ga\textsubscript{1-x}N alloy. Basically, the alloy is precision engineered for light emission at a target wavelength and incorporated into the LED structure as the active layer.

<table>
<thead>
<tr>
<th>LED TECHNOLOGY</th>
<th>ADVANTAGES</th>
<th>DISADVANTAGES</th>
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<tbody>
<tr>
<td>LATERAL</td>
<td>Physical protection from environment after bonding to a heatsink or a die through substrate</td>
<td>High junction temperature</td>
</tr>
<tr>
<td></td>
<td>Easier integration with flip-chip technology (both contacts on one side)</td>
<td>Current crowding</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Low electrical- and thermal-conductive substrate</td>
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<tr>
<td>VERTICAL</td>
<td>Excellent current spreading</td>
<td>Expensive native (i.e. GaN) substrates</td>
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<td></td>
<td>Improved heat dissipation</td>
<td>Extra fabrication steps</td>
</tr>
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<td></td>
<td>High light extraction</td>
<td>Small area devices only</td>
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Table 1 | Due to lack of a cheap vertical LED generation technology, currently ~98% of all LEDs in the market are in lateral configuration that limits the LED performance (output power and efficiency). Schematic illustration and comparison of existing lateral and vertical LED technologies.

LEDs have two mainstream configuration options: Lateral vs. Vertical [1-4]. Lateral (conventional) light emitting diodes (LLEDs) have both contacts on one side as the typical substrate choices, such as sapphire (>98% of market share), is nonconductive. Under high current injection, LLEDs suffer from non-uniform current injection and local hot spots limiting the performance. Recently, vertical LEDs (VLEDs) have been developed through either releasing the substrate or employing conductive substrate, SiC. VLEDs outperform LLEDs in terms of output power and efficiency; however, are expensive (see Table 1 for a comparison) [1-4]. Moreover, available conductive substrates such as SiC and GaN are costly and not widely-available limiting the market penetration of VLEDs. Thus, novel LED layer separation schemes such as laser lift-off (LLO) and chemical lift-off have been developed to release LED layer for VLED fabrication. Table 1 summarizes and compares these two LED configurations.

Current Approaches to Thin-film Light Emitting Diodes

Today, there are a few conventional means to enable thin-film GaN-based vertical LEDs. The first is laser lift-off (LLO) [5-8]. Laser lift-off process employs an ultraviolet (UV) laser to illuminate LED layers from the UV-transparent (i.e. sapphire) substrate side. The LED buffer layer, that is typically GaN, absorbs the UV laser, heats up, and dissociates into Ga and N\textsubscript{2}, releasing the substrate. Main disadvantages of this technique are limit on the LED area (~0.25 mm\textsuperscript{2}) (due to laser spot size) and requirement for expensive tooling (due to laser scanning) [5-8]. This approach is the current means of industrial VLED production from sapphire substrates but has significant disadvantages such as yield (see Table 2) bottlenecking solid state lighting adaption. The second means to enable thin-film GaN-based VLED is chemical lift-off (CLO) [9-13]. Chemical lift-off process requires additional deposition step of an epitaxial layer such as Ga\textsubscript{2}O\textsubscript{3}, GaON, and BN on substrate prior to LED layer deposition. This approach increases the cost and scalability. In addition, chemical lift-off of LED layers takes a long time due to wet etch process and becomes more time-consuming and challenging for larger area substrates (≥4-inch) whereas LED epitaxial growth process desires financially otherwise (i.e. switching larger diameter substrates) [9-13]. In summary, chemical lift-off process is not suitable for industrial adaptation. Both of these existing technologies have advantages and disadvantages, as summarized in Table 2. Both of these existing technologies have not been enough to enable VLED technology commercialization yet a lone a thin-film (<5µm) flexible LED.

In this work, we review a novel mechanical means of release technique, called “controlled spalling” [14-18]. Controlled spalling is a novel thin-film enabling technology proved useful for circuits and solar cells [14-16]. Here we review this know-how for releasing LED layers from any substrate and investigate the opto-electro-thermal characteristics of this new category of VLEDs: Spalled LEDs (SLEDs). Controlled spalling is a room-temperature low-cost process requiring minimal space and tooling. A stressor layer such as Ni (second cheapest metal) is employed to control fracture mode and depth, and a flexible (such as a polyimide) tape is used as the handle layer. Main advantages of this technique over existing ones (i.e. chemical and laser lift-off) aside from reduced cost and easier implementation are.

Table 2 | Characteristics of this new category of VLEDs: Spalled LEDs (SLEDs).
(1) Large area film creation (demonstrated up-to 300-mm diameter Si films), (2) Substrate reuse possibility (demonstrated Ge substrate reuse via solar cell spalling), and (3) Applicability to any LED layers (pre- or post-fabrication) on any substrates. Moreover, unique array structures for LED lamps are possible on such large area LED layers as well as employing such large area intact LED layers into flexible transparent inorganic display arrays.

In summary, controlled spalling is a couple-year-old thin-film creation technology proven itself in circuits and solar cells. It offers a cheaper and faster means of generating VLEDs and opens up new possibilities for high efficiency high power LED lamps. Expertise in LEDs combined with experience in controlled spalling technology will enable LED lamps leap into general lighting and will open up new venues for unconventional LED lighting technologies from health and biomedicine to wearable consumer devices.

<table>
<thead>
<tr>
<th>Laser Lift-Off (LLO)</th>
<th>Chemical Lift-Off (CLO)</th>
<th>Controlled Spalling (CS)</th>
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<tr>
<td><strong>ADV.</strong></td>
<td><strong>ADV.</strong></td>
<td><strong>ADV.</strong></td>
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<tr>
<td>• Industry standard</td>
<td>• Laboratory demonstrations</td>
<td>• Large area devices allows more power</td>
</tr>
<tr>
<td>• Relatively reliable process</td>
<td>• Substrate reuse possible</td>
<td>• No loss of epitaxial area (flexible design)</td>
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<tr>
<td>• Mature wafer handling</td>
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<td>• Substrate reuse possibility (reduced cost)</td>
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<td></td>
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<td>• Flexible &amp; transparent LED market penetration</td>
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<tr>
<td><strong>DISADV.</strong></td>
<td><strong>DISADV.</strong></td>
<td><strong>DISADV.</strong></td>
</tr>
<tr>
<td>• Limited area per chip/device</td>
<td>• Time extensive (particularly for whole substrate releases)</td>
<td>• Immature Understanding</td>
</tr>
<tr>
<td>• Loss of epitaxial area</td>
<td>• Limited LED quality due to “etch layer” insertion in-between</td>
<td>• Thin film handling</td>
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<tr>
<td>• Extra steps for cleaning after lift-off</td>
<td></td>
<td>• Packaging immature</td>
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<td>• No chance of substrate reuse</td>
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**Table 2**: Comparison of conventional (i.e. Laser and Chemical Lift-Off) and proposed (i.e. Controlled Spalling) thin-film LED technologies shows the promise of controlled spalling for large area, cheap, scalable LED release

**Current Barriers in Thin-film Light Emitting Diodes**

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). However, such crystalline LED layers need to be grown epitaxially on crystalline host substrate for practical devices. Conventional choice of such substrates are sapphire (Al2O3), silicon carbide (SiC), and silicon (Si) that have ~13%, ~3%, and ~17% lattice-mismatch with LED layers. This leads to high defect densities (> 5x10^6/cm^2) in the active layer material. Defects act as non-radiative recombination centers, and limit the output power, efficiency, and spectral quality of LED lamps. Thus, it is essential to minimize defectivity through employment of lattice-matched substrates to improve LED efficiency from its current value of ~50% to its theoretical value of ~88%. 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 Table 1).

Recently, GaN freestanding (FS) substrates as large as 2-inch diameter become available. GaN FS substrates enable lattice-matched growth for LEDs reducing defectivity at least two orders of magnitude (< 10^6 cm^-2). This reduced defectivity promises the 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.

Another challenge stems from the fact that general illumination applications require high lighting power (~ 100 W) that demands high current injection (~ 50 A/cm^2) into the LED such that power density in a LED is 500W/cm^2 ~ x5 of that in a CPU. [19] However, as the LED lamp efficiency is ~50%, about only half of the injected energy produces light whereas the other half is lost as heat. Thus, under general illumination operating conditions, LED lamps’ lighting
characteristics (i.e. output power, efficiency, and spectral quality) is dominated and limited by the thermal effects. Under such high injection, 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. Conventional thermal management of LEDs require employment of high thermal conductivity packaging materials such as ceramics (i.e. BeO or AlN) as submounts and metals (i.e. Cu) as heatsinks. As such, the majority of the LED lamp cost (40% to 60% of all cost) is currently associated with the packaging. This necessitates reducing the packaging costs of a LED without reducing the thermal budget of the lamp. It is important to note that all these problems have to be addressed while maintaining the financial advantages of LEDs.

One way to benefit from GaN FS substrates and reduce the (substrate and operational) cost is through enabling substrate reuse. Recently, we have introduced a new thin-film generation technique called "controlled spalling" into literature (Table 2), and shown that thin-film devices generated by this means perform as good as their bulk counterparts. Controlled spalling is a process where a strain material such as Nickel (Ni) is deposited on top of the semiconductor epilayer leading to a tensile strain enabling the release of the semiconductor from the substrate (on which it was grown) through fracture mode control. The material characteristics along with stressor design control the thickness of the released thin film. We have recently applied this technology to a various materials such as silicon, germanium, and gallium arsenide substrates as well as to several of devices such as silicon integrated circuits [15] and III-V solar cells [14, 16, 17, 18] However, this technology will be most interesting for GaN FS substrates and LEDs as theoretically it is possible to engineer the stress levels of Ni to release such LED layers and enable thin-film LEDs and substrate reuse – solving the current issues in solid state lighting.

In summary, it is important to overcome these critical technological and financial bottlenecks via (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, (3) Engineered spalling layer release technique to generate the thinnest LEDs for improving thermal budget, and (4) Investigating multiple times of substrate reuse.

![Figure 1](https://example.com/figure1.png)

**Figure 1** | Our results suggest novel layer release technique “spalling” can enable world’s thinnest (< 5 µm) inorganic LEDs. (a) Numerical program flow chart and (b) Resulting spall depth as a function of Ni stressor stress, achievable via electroplating. Error bars correspond to variations in material constants.

2. EXPERIMENT

Our approach to form thin-film semiconductor layers is to spall semiconductors from the host substrate by tensile stress. This technique relies on the natural phenomenon that has been observed for decades that can be summarized as follows: If a layer possessing tensile stress is deposited on the surface of a brittle substrate, often the layer would peel away from the surface and, in doing so, remove a portion of the substrate. This mode of fracture is referred to as substrate spalling. Recently, it was demonstrated [20] that substrate spalling could be used as a means for fabricating thin Si substrates by depositing thick screen-printed metal (Al and Ag) pastes and annealing at 900 ºC. This idea is based on the
fact that the tensile stress in the metal layers increased upon cooling due to the coefficient of thermal expansion (CTE) mismatch between the metal and the Si, leading to spontaneous exfoliation of the surface. It follows from this fact that if enough tensile stress can be built up in a semiconductor, it will release itself by spalling. However, the high temperature steps, required for creating the necessary stress, and the occurrence of spontaneous fracture upon cooling, severely limit the usefulness of the above approach. Overall, the high temperature prohibits layer spalling of prefabricated devices, and spontaneous fracture almost always leads to other competing modes of fracture, such as film cracking. The first step in addressing this important need is to understand spalling mechanisms.

Our investigation relies on an analytical model of spalling that was developed in the late 1980s [21-23], which offers a direct and simple means of predicting critical loading conditions (film stress and thickness values) for which spalling fracture is possible as well as computing the equilibrium crack depth in the substrate. This unique mode of fracture results from the edge load created by the tensile stressor layer giving rise to a mixture of type I stress ($K_I$) (opening mode) and type II stress ($K_{II}$) (shear) which guides the crack to an equilibrium depth below the interface. The layer release is based on the fact that $K_{II}$ shear stress is corrective: (under the condition that opening stress is larger than the fracture strength of the material) Compressive stress drives the crack tip deeper whereas tensile stress drives the crack tip shallower. When the crack tip shear stress is zero, the steady state crack is achieved.

Here, we develop the theory and compute the critical loading conditions for GaN LED wafers. The general framework that will be followed is based on the model developed by Suo and Hutchinson [21-23] where a method of calculating the mode I (crack opening) and mode II (crack propagation) stress intensity factors (namely $K_I$ and $K_{II}$) for a three-layer beam structure with a pre-existing crack are given as:

$$K_I = \frac{P}{\sqrt{2\pi h}} \cos(w) + \frac{M}{\sqrt{2\pi h^3}} \sin(w + \gamma) \quad \text{(Equation 1)}$$

$$K_{II} = \frac{P}{\sqrt{2\pi h}} \sin(w) - \frac{M}{\sqrt{2\pi h^3}} \cos(w + \gamma) \quad \text{(Equation 2)}$$

where $P$ is the edge load and $M$ is the moment induced by the stressor layer, $U$, $V$ and $\gamma$ are dimensionless constants originating from the calculation of elastic energy stored in the beam structure far behind the crack tip, $h$ is the stressor layer thickness and $w$ is a dimensionless number that depends on the elastic dissimilarity of the stressor and the substrate and also depends on crack depth. Accordingly, the depth at which $K_{II}$ is zero determines the fracture depth and therefore the spalled layer thickness. To determine if spalling is possible, the value of $K_I$ at the fracture depth (when $K_{II}$ is zero) is compared to the fracture toughness of the substrate.

It is important to (1) Consider the anisotropy of the materials and take into account that the difficulty of crack to propagate in different directions are to be different, (2) Investigate the effects of crack initiation and crack initiation means, and (3) Effects of handle layer (so as to propagate the crack controllably). Hence, the theory and the model needs to be improved with input from the experimental results.

### 3. RESULTS AND DISCUSSION

The understanding of fundamental spalling behavior in layer release and investigating spalled thin-film InGaN-based LED layers will enable a more accurate prediction of thin-film photonic devices, a necessary requirement for enabling high efficiency high power light-weight, flexible, and conformal vertical devices.

Almost without exception, GaN LEDs (or any kind of GaN devices), grown on homo- or hetero-substrate, include either the substrate or a kind of a thick support layer in the final device. GaN LEDs including such thick substrate or support structures are rigid making them non-flexible (meaning that LED devices cannot be bent) restricting them to non-conformal designs. In bio applications (e.g. pain relief and therapeutic) such rigid devices cannot be used in-vivo as these implants lead to lesion and inflammation. Moreover, such rigid LEDs are almost exclusively are in lateral LED configuration – meaning that under the required injection levels (~200 mW/cm^2), the devices become unreliable and have limited lifetime. On the other hand, thin-film, flexible, and conformal vertical LEDs might prevent tissue lesions during insertions and prevent persistent irritation, which opens up in-vivo biomedical semiconductor photonics instrumentation research. Other advantages of vertical LEDs include higher efficiency (meaning less heating) and flexibility in LED design masks. Yet, thin-film, flexible, and conformal vertical LED is one of least studied technologies due to lack of a large-area layer release technology. Even today, there are no scalable technologies that can enable large area single crystalline thin-film GaN LEDs. Theoretical studies suggest bending stiffness is related to the material...
thickness with the third power suggesting thinner materials (even semiconductors) will become more flexible and less prone to mechanical failure against external forces due to deformation capability (which also means less irritation to body in-vivo applications). As such, the investigation of the **optical, structural, and mechanical** characteristics of thin-film GaN LEDs and their dependence on the thickness will be a valuable contribution to the field and may solve the long-standing debate on the nature of the flexibility of thin-film GaN semiconductors.

As described earlier, spalling, a recently engineered thin film release technique, will be most interesting for GaN FS substrates and LEDs as theoretically it is possible to engineer the stress levels of Ni to release such LED layers and enable thin-film LEDs and substrate reuse. We have recently demonstrated first of its kind vertical LEDs enabled through spalling. [28] The LED device was grown on sapphire substrate and the release of LED layer enabled a vertical LED formation. Most recently, we have shown that a cleave layer (such as epitaxial graphene on SiC) can be used for precise spalling of GaN-based LEDs from the LED-substrate interface. We have demonstrated the first (1) flexible lightweight blue LEDs and (2) substrate reuse. [29] **Vertical LEDs** offer excellent current spreading, improved heat dissipation, and high light extraction with respect to lateral LEDs. Compared to conventional LED layer release techniques used for forming vertical LEDs (such as laser-liftoff and chemical lift-off techniques), this process distinguishes itself with being wafer-scalable (large area devices are possible) and substrate reuse opportunity.

![Image](image-url)

**Figure 2 | Our technique demonstrating world’s largest area as well as world’s thinnest light emitting diodes.** (a) Schematic illustration of spalling layer release process that was re-engineered in 2013 for releasing the light emitting diode epitaxial device layers from rigid sapphire host substrate. (b) TEM study of world’s first fully-operational large area (2-inch diameter) thin-film LED devices on flexible substrates. (c) Electroluminescence spectrum of our light emitting diodes as-grown on rigid substrates (labeled as “bulk”) and released from the rigid substrates via spalling (labeled as “SLED”) show similar spectral output. [28] [Copyright (2013) The Japan Society of Applied Physics]

4. CONCLUSION

With the development of high performance single crystalline III-V devices, it becomes imperative to achieve similar high performance and function in light-weight and flexible devices. Novel layer release process of spalling, as we have recently demonstrated with highest specific power solar cells and thinnest light emitting diodes demonstrations, is a promising layer release technique suitable for enabling light-weight, flexible, and functional single crystalline III-V devices. High efficiency multi-junction solar cells, conventionally limited by substrate cost and substrate effects (thermal management and reliability), would be an ideal research area with substantial innovation opportunity. Light emitting diodes, conventionally suffering from lattice-mismatched substrates, promise a similar advantage thanks to recent availability of large area freestanding GaN substrates. Overall, III-V materials (As/P & N), photonics combined with spalling layer release technology suit a unique opportunity to innovate thin-film, flexible, and functional solar cells and light emitting diodes with world record performance. These research thrust areas are keys in creating a sustainable world for all of us.

This work was carried out in the Micro and Nanotechnology Laboratory, University of Illinois at Urbana-Champaign, IL, USA.

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Figure 3 | GaN-on-Graphene platform has been shown to be a useful means for enabling thin-film InGaN-based LEDs. [29]

REFERENCES