

# Novel Process for Direct Bonding of GaN onto Glass Substrates using Sacrificial ZnO Template Layers to Chemically Lift-off GaN from c-sapphire

D. J. Rogers<sup>(a)\*</sup>, A. Ougazzaden<sup>(b,c)</sup>, V. E. Sandana<sup>(a,d,e)</sup>, T. Moudakir<sup>(b)</sup>, A. Ahaitouf<sup>(c)</sup>, F. Hosseini Teherani<sup>(a)</sup>, S. Gautier<sup>(f,c)</sup>, L. Goubert<sup>(a,d,e)</sup>, I. A. Davidson<sup>(g)</sup>, K. A. Prior<sup>(g)</sup>, R. P. McClintock<sup>(h)</sup>, P. Bove<sup>(a)</sup>, H.-J. Drouhin<sup>(d)</sup> and M. Razeghi<sup>(e)</sup>.

<sup>(a)</sup>Nanovation, 8 Route de Chevreuse, Châteaufort, 78117, France

<sup>(b)</sup>Georgia Institute of Technology/GT-Lorraine, 2-3 rue Marconi, 57070 Metz, France

<sup>(c)</sup>UMI 2958 Georgia Tech-CNRS, 2-3 rue Marconi, 57070 Metz, France

<sup>(d)</sup>Department of Irradiated Solids, Ecole Polytechnique, 91128 Palaiseau, France.

<sup>(e)</sup>Center for Quantum Devices, ECE Department, Northwestern University, Evanston, Illinois 60208, USA

<sup>(f)</sup>LMOPS-EA 4423, Univ. de Lorraine et SUPELEC, 2 rue Edouard Belin, 57070 METZ, France

<sup>(g)</sup>Institute of Photonics and Quantum Sciences, SUPA, School of Engineering and Physical sciences, Heriot-Watt University, Edinburgh, EH14 4AS, Scotland.

<sup>(h)</sup>MP Technologies LLC, 1801 Maple Ave., SUITE 5110/20, Evanston, IL 60201-3135, USA

## ABSTRACT

GaN was grown on ZnO-buffered c-sapphire (c-Al<sub>2</sub>O<sub>3</sub>) substrates by Metal Organic Vapor Phase Epitaxy. The ZnO then served as a sacrificial release layer, allowing chemical lift-off of the GaN from the c-Al<sub>2</sub>O<sub>3</sub> substrate via selective wet etching of the ZnO. The GaN was subsequently direct-wafer-bonded onto a glass substrate. X-Ray Diffraction, Scanning Electron Microscopy, Energy Dispersive X-ray microanalysis, Room Temperature Photoluminescence & optical microscopy confirmed bonding of several mm<sup>2</sup> of crack-free wurtzite GaN films onto a soda lime glass microscope slide with no obvious deterioration of the GaN morphology. Using such an approach, InGaN based devices can be lifted-off expensive single crystal substrates and bonded onto supports with a better cost-performance profile. Moreover, the approach offers the possibility of reclaiming and reusing the substrate.

**Keywords:** (In)GaN, LED, Multijunction Photovoltaic, ZnO, Selective Wet Etching, Chemical Lift-off, Direct Wafer Bonding, Glass Substrate

## 1. INTRODUCTION

Since the development of p-type doping of GaN in the early 1990s, there has been rapid industrial development and an exponential market expansion for (In)GaN-based Light Emitting Diodes [1]. Recently, promising novel (In)GaN-based devices such as high efficiency multijunction photovoltaics (PV) are emerging [2]. Currently such devices employ expensive single crystal substrates because high quality GaN layers cannot be obtained directly on cheaper substrates. Moreover, most GaN is grown on c-sapphire (c-Al<sub>2</sub>O<sub>3</sub>) because native GaN crystals with an appropriate size/cost/performance profile are not available in industrial quantities. c-

$\text{Al}_2\text{O}_3$  substrates bring additional constraints such as poor thermal/electrical conductivity, limited size and relatively high cost levels. Thus it is desirable transfer the (In)GaN from the single crystal substrate to a better alternative. Recently [3-8], the authors showed that ZnO thin films could be used as sacrificial templates for the chemical lift-off of (In)GaN thin films from single crystal substrates in an alternative to the laser lift-off process [9] currently used by industry. In the conventional industrial process, the GaN-based layers are transferred and bonded to an alternative substrate using an adhesive interlayer. This paper gives an overview of a novel process developed for the direct wafer bonding of GaN onto glass (or other) substrates. In such direct bonding, there is no adhesive interlayer. This offers many advantages including the possibility for subsequent thermal processing, the possibility for Ohmic contacts and the potential for good thermal conduction and optical coupling [10].

## 2. EXPERIMENT

### 2.1 Thin Film Growth

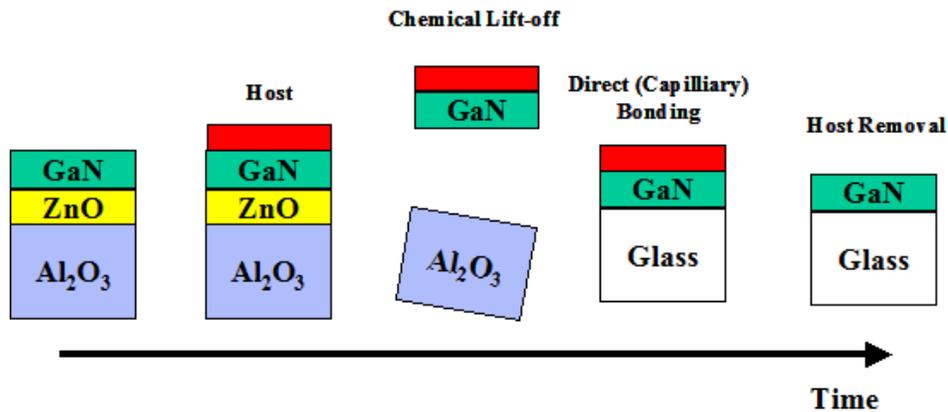
ZnO thin films were grown on *c*- $\text{Al}_2\text{O}_3$  substrates using pulsed laser deposition (PLD) from a 99.99% pure, sintered, ZnO target in molecular oxygen with a coherent KrF ( $\lambda = 248$  nm) excimer laser, as described elsewhere [3]. GaN overlayers were grown using metal-organic vapor phase epitaxy (MOVPE). The dissociation of ZnO observed during conventional MOVPE growth of InGaN/GaN was combated through the use of a low pressure/temperature MOVPE approach with  $\text{N}_2$  as a carrier gas and dimethylhydrazine added to the ammonia (nitrogen precursor) in order to enhance the concentration of atomic nitrogen. This approach allowed growth of well-crystallized wurtzite GaN at a reduced substrate temperature.

### 2.2 Characterisation

Sample morphology was examined at the optical scale using a Leitz Wetzlar microscope with digital image acquisition and in the sub-optical range using a Hitachi S4800 field emission gun-Scanning Electron Microscope (FEG-SEM) equipped with an Energy Dispersive X-ray (EDX) spectroscopy system for compositional microanalysis. The crystal structure of the samples was investigated using high resolution X-Ray Diffraction (XRD) performed in a Panalytical MRD Pro system using Cu  $K\alpha$  radiation. Optical properties were studied via Room Temperature (RT) Photoluminescence (PL) with a Coherent Innova frequency-doubled Ar-ion laser (244 nm) and an iHR 550 Horiba Jobin-Yvon spectrometer.

### 2.3 Chemical Lift-off & Wafer Bonding

GaN films were transferred onto optically-flat Corning microscope slides (soda-lime glass) via a process of chemical lift-off and direct fusion bonding modeled on that developed for the chemical lift-off of ZnSe from GaAs substrates and bonding onto glass/silica using MgS release layers [10-15], as shown in Figure 1.



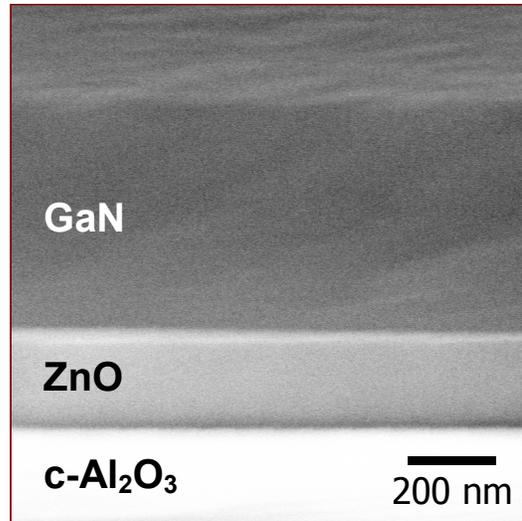
**Figure 1** Process flow for chemical lift-off and direct fusion bonding of GaN onto glass.

In this process a ZnO interlayer acts first as a crystallographic template for GaN and then as a sacrificial release layer, which is selectively etched away in 0.1M HCl in order to separate the GaN from the sapphire substrate. A temporary “host” substrate of Apiezon “W” wax is used to maintain the integrity of the GaN during the transfer process. After removing the ZnO layer, the GaN is transferred onto a glass substrate by applying a small compressive force so as to promote capillary action between the GaN and the glass. After drying, the wax is removed by dissolution in xylene. Yablonovich et al. investigated the nature of the chemical bonding in such a process and concluded that it is probably via the van der Waals interaction [10].

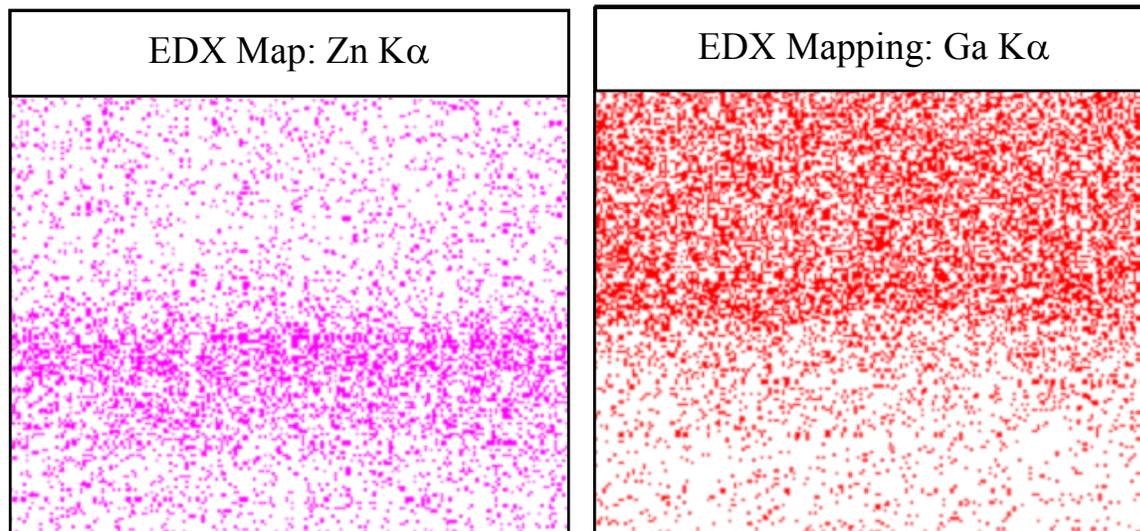
### 3. RESULTS & DISCUSSION

#### 3.1 Growth of GaN on ZnO/c-Al<sub>2</sub>O<sub>3</sub>

Figure 2 shows a FEG-SEM image of a fracture cross-section of the MOVPE GaN / PLD ZnO grown on a c-Al<sub>2</sub>O<sub>3</sub> substrate and Figure 3 shows corresponding EDX K $\alpha_1$  line intensity mappings of Zn and Ga.

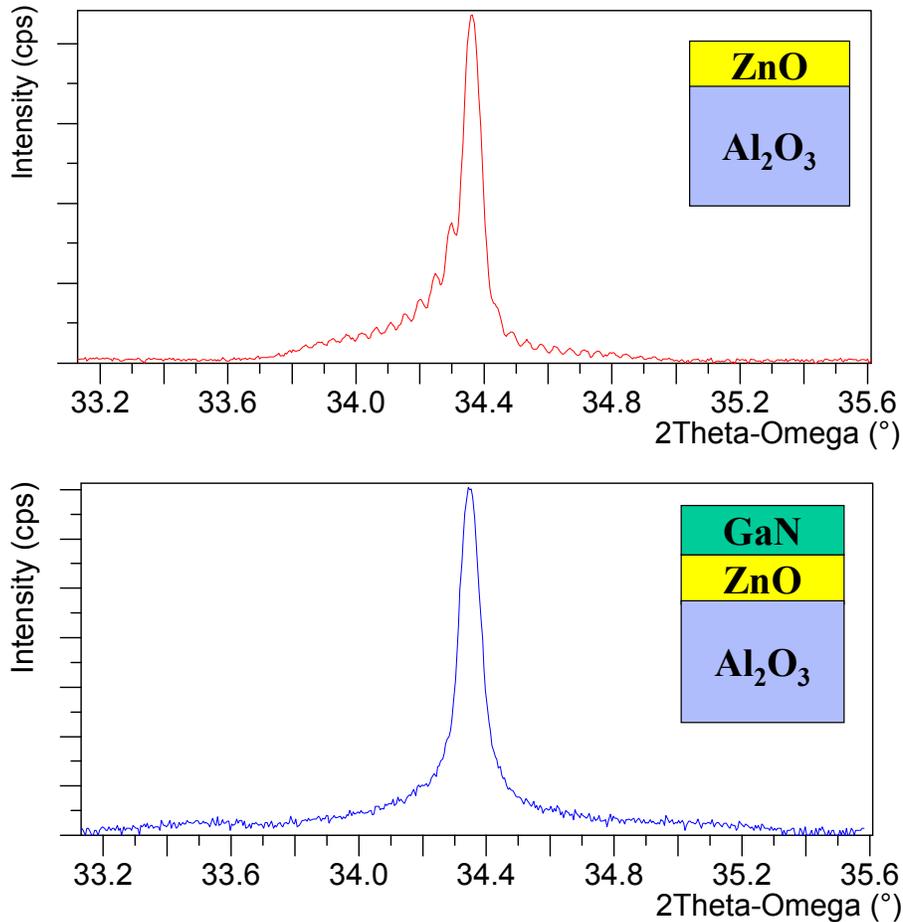


**Figure 2** FEG-SEM image of a fracture cross-section of the GaN/ZnO/c-Al<sub>2</sub>O<sub>3</sub> heterostructure.



**Figure 3** EDX maps for the GaN/ZnO/c-Al<sub>2</sub>O<sub>3</sub> cross-section shown in Figure 2 (contrast is proportional to  $K\alpha_1$  line intensity). The EDX maps confirm the identification of the GaN and ZnO layers.

The sample showed continuous GaN and ZnO layers with a smooth interface and surface. Film thickness was estimated to be about 210 nm for the ZnO layer and 550 nm for the GaN layer. Figure 4 shows XRD  $2\theta/\Omega$  scans of the (0002) peak for the ZnO/sapphire and GaN/ZnO/sapphire prior to chemical lift-off.

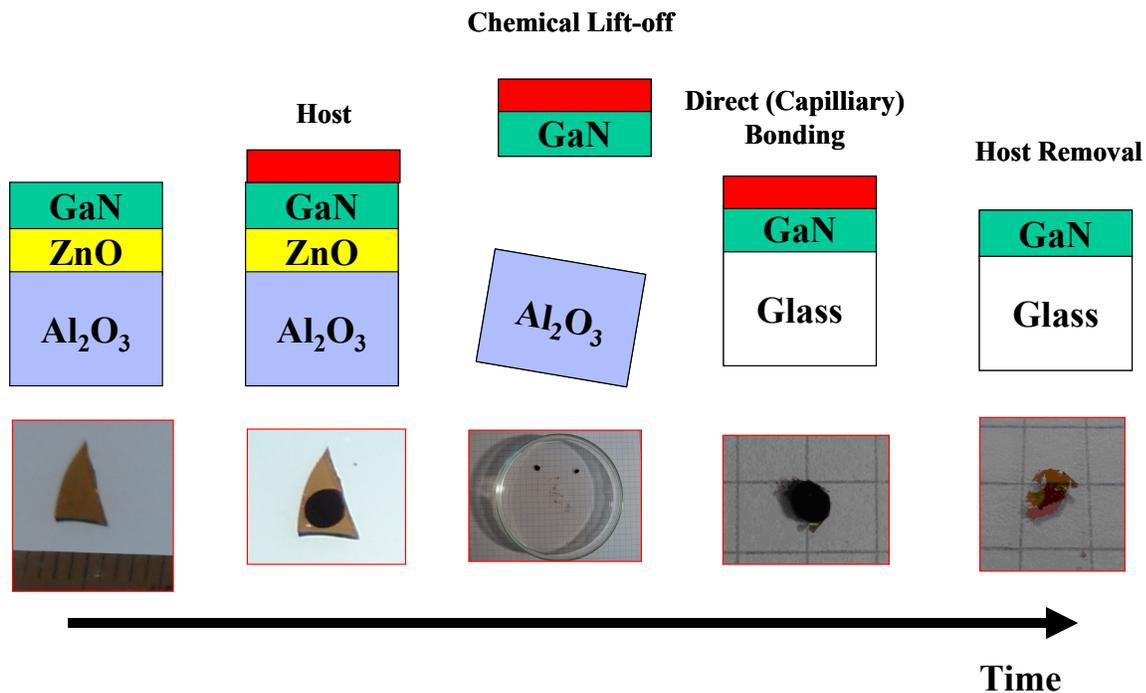


**Figure 4** XRD  $2\theta/\Omega$  and  $\Omega$  scans of the (0002) peak for the ZnO/sapphire (upper) and GaN/ZnO/sapphire (lower) prior to chemical lift-off.

Both scans are indicative of wurtzite ZnO with strong c-axis orientation. The ZnO peak has a Full Wave Half-Maximum (FWHM) of  $0.05^\circ$  and a peak position corresponding to a c lattice parameter of  $5.215\text{\AA}$  (compared to a value of  $5.206\text{\AA}$  for equilibrium wurtzite ZnO). This suggests that the ZnO was under slight compressive strain in the film plane. The peak also shows Pendellösung fringes, which indicate that the film had a relatively smooth surface over the scale of the XRD beam spot. The fringe spacing corresponds to a film thickness of  $\sim 200\text{ nm}$ , which is in agreement with the SEM cross-section shown in Figure 2. After GaN growth, a single peak is visible with a FWHM of  $0.06^\circ$  and a peak position corresponding to a c lattice parameter of  $5.218\text{\AA}$ . This is very similar to that for the ZnO prior to GaN growth and significantly higher than the value of  $5.186\text{\AA}$ , which would be expected for equilibrium wurtzite GaN. Thus the GaN was also under slight compressive strain, which could well be due to epitaxy on the ZnO template.

### 3.2 Chemical Lift-off and Wafer Bonding onto Glass

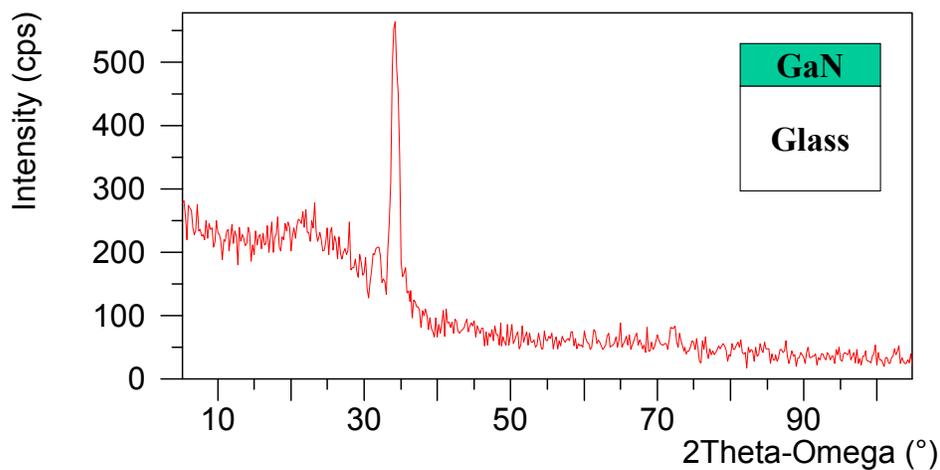
Figure 5 shows photographs of a sample during the chemical lift-off and direct fusion bonding process.

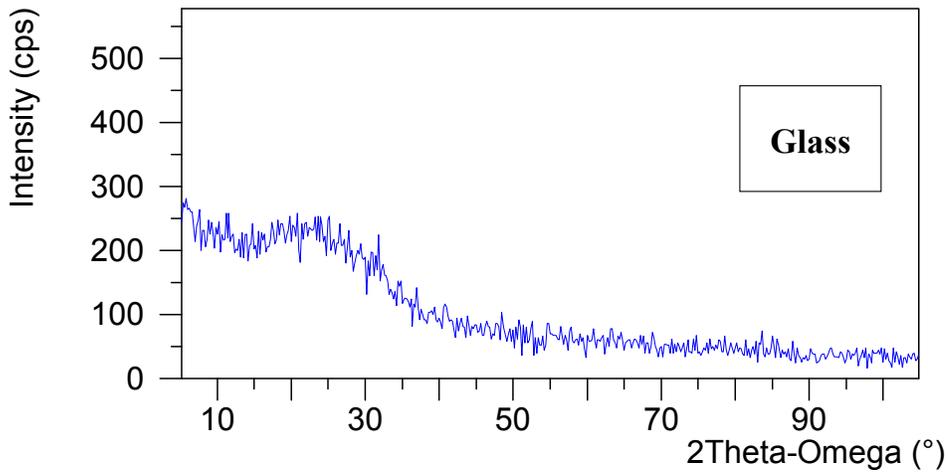


**Figure 5** Process flow and photographs of the chemical lift-off and wafer-bonding (scale of bonded pieces is given by 5mm grid on paper).

In the final image of the figure it can be seen that after host removal several mm<sup>2</sup> were bonded onto the glass.

Figure 6 shows XRD 2θ/Ω scans for the glass substrate before and after the GaN wafer bonding.

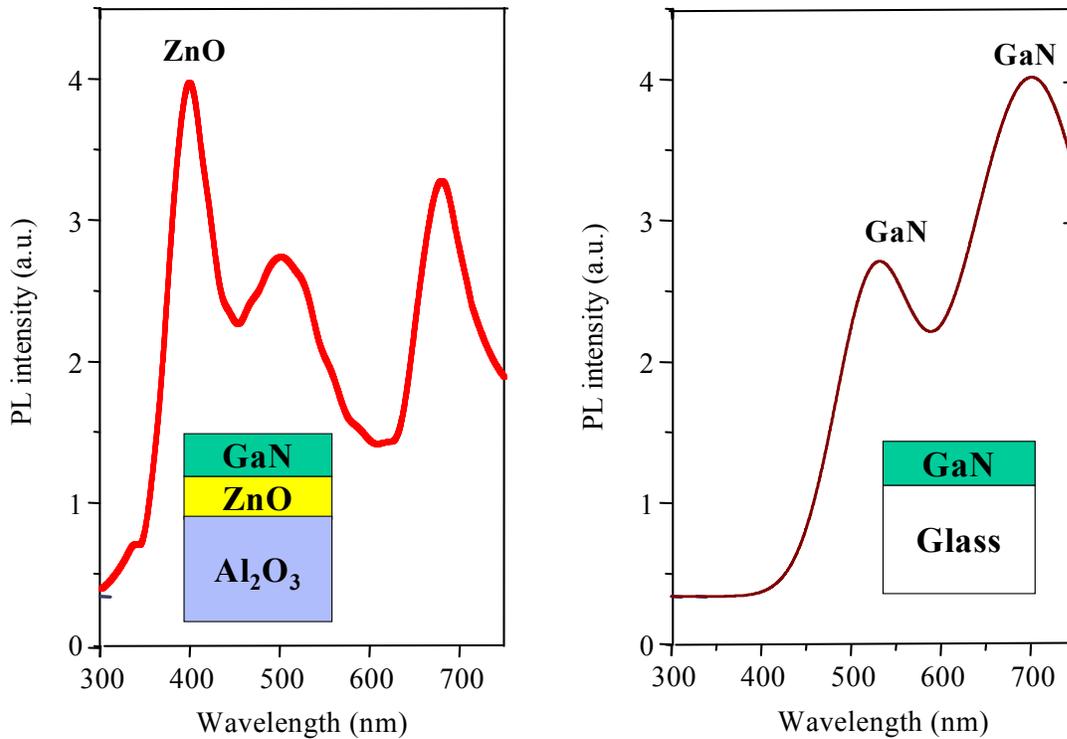




**Figure 6** XRD  $2\theta/\Omega$  scans for the GaN/glass (upper) and glass (lower).

After bonding, a peak appears at a position corresponding to a wurtzite GaN (0002) peak with a  $c$  lattice parameter of about  $5.23 \text{ \AA}$ . This is comparable to the value observed prior to lift-off in view of the increased FWHM of  $\sim 0.6^\circ$ . The origin of this peak broadening is not clear but it could be related to strain relaxation.

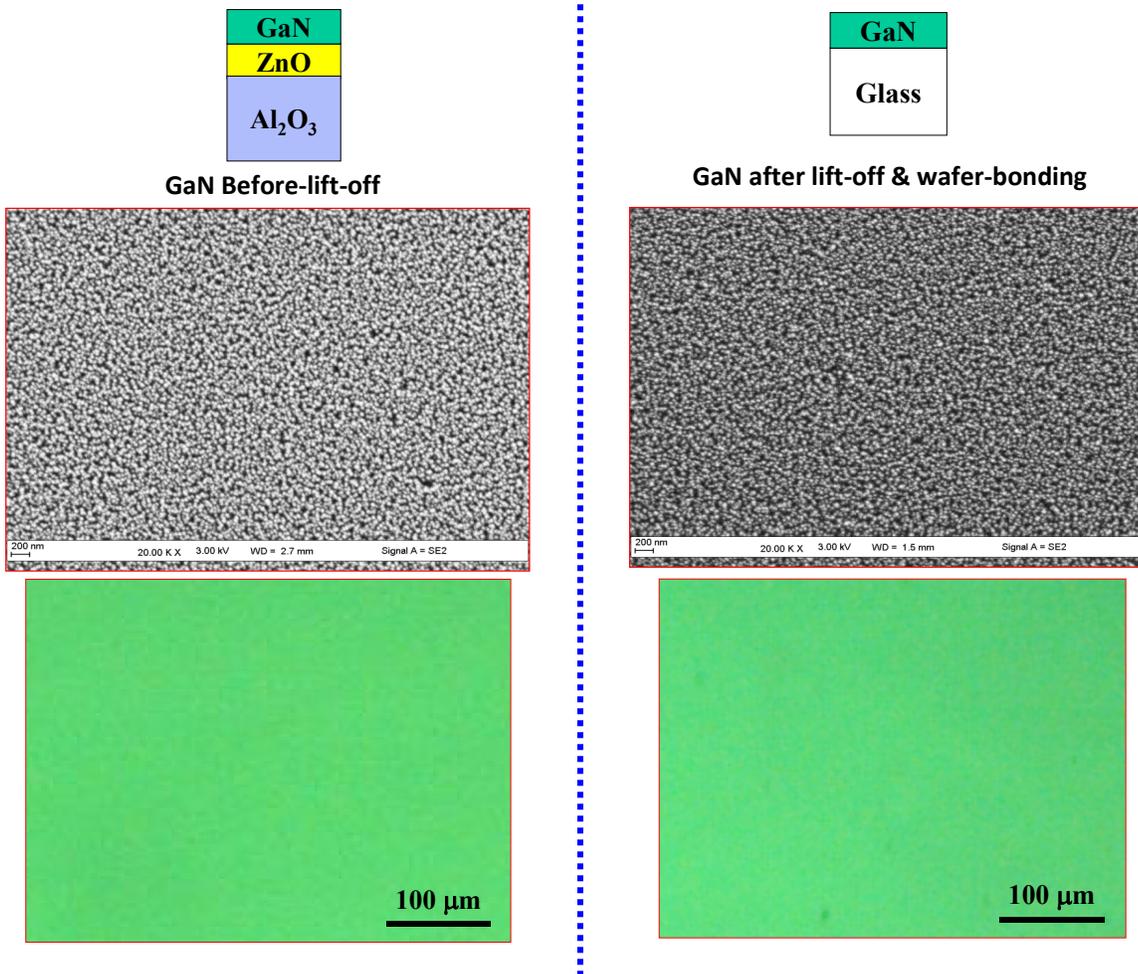
Figure 7 shows RT PL spectra for the sample before/after lift-off and wafer bonding.



**Figure 7** RT PL spectra for the sample before and after lift-off & wafer-bonding.

Prior to lift-off there is a peak centred just under 400 nm, which is typical for wurtzite ZnO near band edge (NBE) emission, and two peaks centred at about 500 and 680 nm. After the chemical etching the ZnO NBE peak has disappeared and the two other peaks remain, which suggests that they originate from the GaN. Both of these peaks appear slightly red-shifted after the bonding, which may be related to strain relaxation inferred from the XRD.

Figure 8 shows SEM and optical microscope image of the GaN surface before and after the lift-off and bonding.



**Figure 8** SEM (upper) and optical microscope (lower) images of the GaN surface before and after the lift-off and wafer bonding.

Both the optical/and SEM images are very similar before/after the lift-off/bonding. In all cases, the surfaces appear to be free of obvious cracking, pit, particle/droplet or defect problems.

## 4. CONCLUSION

~550 nm thick GaN thin films were grown by MOVPE on ~200 nm thick ZnO-buffered c-sapphire substrates. SEM, XRD, EDX, RT PL and optical microscopy confirmed highly c-axis oriented wurtzite ZnO and GaN layers with similar c-axis, lattice parameters, a smooth interface, low surface roughness and no obvious cracking, pit, particle/droplet, pinhole, or defect problems. The GaN was then chemically lifted off the c-sapphire and direct fusion bonded onto soda lime glass using the ZnO template as a sacrificial release layer. XRD, SEM, EDX, RT PL and optical microscopy confirmed bonding of several mm<sup>2</sup> of crack-free wurtzite GaN films with no obvious deterioration of the GaN morphology. Using such an approach (In)GaN based devices can be lifted-off relatively expensive single crystal substrates and bonded onto a support with a better cost-performance profile. It has been demonstrated elsewhere [16] that a similar process has the potential to be scaled up to 2 inch diameter wafers and beyond. Moreover, the approach offers the possibility of reclaiming and reusing the substrate.

## ACKNOWLEDGEMENTS

The authors would like to thank the French Agence National de la Recherche for funding this research via the “New PV on Glass” programme.

## REFERENCES

- [1] R. Haitz & J. Y. Tsao Phys. Status Solidi A 208, 1 (2011) 17
- [2] E. Matioli et al. Appl. Phys. Lett. **98**, (2011) 021102
- [3] D. J. Rogers et al. Proc. of SPIE 5732 (2005) 412
- [4] D. J. Rogers et al. Appl. Phys. Lett. 91 7 (2007) 071120
- [5] A. Ougazzaden et al., Journal of Crystal Growth 310 (2008) 944–947
- [6] D. J. Rogers et al. J. Vac. Sci. Technol. B 27, 3 (2009) 1655
- [7] A. Ougazzaden et al., Proc. of SPIE 7603 (2010) 76031D-1
- [8] S. Gautier et al., Proc. of SPIE 7940 (2011) 79400X-1
- [9] W. S. Wong et al. Appl. Phys. Lett. 75 (1999) 1360
- [10] E. Yablonovich et al. Appl. Phys. Lett. **56**, 2419 (1990).
- [11] M. Konagai et al. Journal of Crystal Growth 45 (1978) 277
- [12] E. Yablonovich et al. Appl. Phys. Lett. 51 26 (1987) 2222
- [13] A. Balocchi et al. Appl. Phys. Lett. **86**, (2005) 011915
- [14] C. Bradford et al. Journal of Crystal Growth 278 (2005) 325–328
- [15] A. Curran et al. Semicond. Sci. Technol. **22** (2007) 1189–1192
- [16] J. J. Schermer et al. Phys. Stat. Sol. (a) **202**, No. 4, (2005) 501–508

[\\*rogers@nanovation.com](mailto:*rogers@nanovation.com)