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Express Letter

Large Free-Standing GaN Substrates by Hydride Vapor Phase Epitaxy and Laser-Induced Liftoff

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Free-standing GaN, nearly equal in area to the original 2 inch wafer, was produced from 250–300 μm thick GaN films grown on sapphire by hydride vapor phase epitaxy (HVPE). The thick films were separated from the growth substrate by laser-induced liftoff, using a pulsed laser to thermally decompose a thin layer of GaN at the film-substrate interface. Sequentially scanned pulses were employed and the liftoff was performed at elevated temperature (>600°C) to relieve postgrowth bowing. After liftoff, the bow is only slight or absent in the resulting free GaN.

KEYWORDS: GaN, HVPE, liftoff, laser processing, free-standing, homoepitaxy

Due to the lack of native nitride substrates, films of GaN and related nitride compounds are commonly grown on sapphire wafers. Use of these foreign substrates is detrimental to the quality and function of the semiconductor. Numerous threading dislocations are generated in the epitaxial film to accommodate the large mismatch between the film and substrate. The thermal expansion coefficients (TEC) of the GaN and substrate materials differ, causing stress and bowing upon cooling from the growth temperature. The use of a sapphire substrate also complicates processing steps, such as formation of cleaved-edge facets and electrical backside contact. The extraction of heat from an operating device through the sapphire substrate is also hampered.

The ideal solution would to employ a homoepitaxial substrate. The growth of bulk GaN crystals is one approach to this solution, and advances have been made in obtaining crystal platelets.[1)] The availability and size of such crystals, however, still restricts their application. An alternative approach is the use of thick, epitaxially grown GaN films as substrates for further film growth. Sufficient thickness can allow the film surface to relax to the usual bulk lattice constant, alleviating the lattice mismatch problem. Crack-free GaN films on sapphire with film thickness as large as 300 μm have been synthesized using the high growth rate hydride vapor phase epitaxy (HVPE) technique.[2)] In order to solve problems like the thermal stress or facilitate thermal and electrical backside contact, however, it is still desirable to remove the original substrate. For example, Nakamura *et al.* attained improvements in the lifetime of laser diodes and cleaved facets after polishing the sapphire away.[3,4)] Another approach has been taken by Melnik *et al.*, who removed SiC growth substrates by reactive ion etching, producing free GaN with 30 mm diameter, and maximum crack-free areas of about 40 mm^2.[5,6)]

The determination that an intense pulsed laser of appropriate wavelength can be used to locally decompose and split the interface between the nitride film and sapphire substrate indicated a promising means of producing free-standing GaN films.[7,8)] The laser can induce thermal decomposition of the nitride materials.[9,10)] By transmitting the light through the sapphire to the GaN interface, the sapphire can be removed at once, without having to wear through it. This process has also been investigated by Wong and coworkers.[11)]

In this paper, we report the production of large free-standing GaN wafers, demonstrating successful liftoff of areas not limited to the laser beam size and overcoming TEC-induced strain. This involved growing the GaN films by HVPE to thicknesses in excess of 200 μm, for mechanical stability, and performing laser-induced liftoff at elevated temperatures to alleviate TEC-related bowing and cracking. We report stand-alone GaN wafers with approximately 10 cm^2 of crack-free area, which is to our knowledge, the largest free-standing GaN films yet produced.

The GaN films were first deposited on 2 inch diameter (0001) sapphire wafers by HVPE. GaN deposition was accomplished by reacting GaCl with ammonia (NH_3) at temperatures between 1000 and 1050°C. The ratio of NH_3 flow to HCl flow over the Ga was maintained at 35 throughout the deposition cycle. The high growth rate of the HVPE process affords the fabrication of extremely thick films in a convenient period of time. In this study, a growth rate of 100 μm/h was used to prepare a number of samples with total GaN thickness ranging from 200 to 300 μm. The dislocation density of GaN on sapphire films produced under similar growth conditions was less than 5×10^6 cm^2.[2)] Utilizing frontside and backside contacts after liftoff, capacitance-voltage measurements of the undoped GaN indicated a carrier concentration of approximately 3×10^{15} cm^{-3} at the upper portion of the film. Upon cooling the wafers after growth, the sapphire contracts more strongly than the GaN. Since the film thickness was comparable to that of the substrate, TEC differences resulted in substantial bowing of the wafer. Nevertheless, the wafers exhibited very little or even no near-surface cracking across the full 2 inch wafer.

The laser-induced liftoff was performed with the third harmonic from a Q-switched Nd:YAG laser, with 355 nm wavelength and photon energy slightly larger than the GaN bandgap. The pulses were of nominally 5 ns duration with a 7 mm beam diameter. The pulse energy at the wafer was adjusted with a polarizing beam attenuator and each region of the wafers was released by a single pulse, directed through the back of the sapphire substrate. Wafers with and without polished sapphire backsides were processed. Although the polished back is important for patterned liftoff,[7,8)] the unpolished surfaces function as a beam homogenizer which was found useful for processing large areas uniformly. To pro-

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tect the film during processing, the film side of the wafer was temporarily bonded to a metal support using a metal solder.

Incident pulse intensities of about 0.3 J/cm^2 were required for the interface splitting, using the Q-switched Nd:YAG laser, consistent (allowing for reflection and the diffusion of heat) with earlier estimates[8,9] of the threshold intensity for decomposition based on optical[12,13] and thermal effusion data.[14,15] This threshold intensity depends on the particular laser characteristics. Using a KrF excimer laser, Wong *et al.* reported a somewhat higher threshold intensity of 0.4–0.6 J/cm^2 for the 248 nm wavelength light[11] and tests with another KrF laser also indicated a threshold near 0.5 J/cm^2.[16] The higher optical extinction coefficient of GaN at 248 nm compared to 355 nm[12,13] should lower the threshold intensity, so the higher threshold is apparently due to the longer pulse duration (38 ns for ref. 11), which allows the heat to diffuse over a greater depth. The higher refractive indices of sapphire and GaN at 248 nm also result in somewhat higher reflectivity, but should only make a modest (2–3%) difference in terms of threshold.

Separating areas larger than the attainable beam size, at the required intensity, was achieved by sequentially scanning the pulses across the surface, as shown schematically in Fig. 1. The bowing of the wafers with thicker GaN films conflicted with this procedure, however, because the strain inhomogeneity at the boundary of the released and thus relaxed spots caused extensive fracturing. To reduce the bowing-induced fracturing, we then investigated heating of the GaN-sapphire system during the laser liftoff, which was found to reverse much of the strain caused by cooling from the growth temperature. The bowing was visibly reduced, and hardly noticeable at temperatures around 600°C, consistent with Leszczynski's study,[17] in which a temperature-dependent X-ray diffraction comparison of a bulk GaN crystal and a 2 μm-thick film on sapphire showed convergence of the lattice constant values at about 800 K and indicated the possibility of inelastic relaxation above this temperature. Indeed, GaN decomposes above about 800°C,[15] which also indicates an upper limit for the processing temperature. By performing the laser treatment at temperatures above 600°C, extended areas could then be released sequentially without the catastrophic fracturing.

In this way, free-standing GaN films were separated from 2 inch sapphire wafers, as shown in Fig. 2 for a film of 275 μm thickness. Except for some spots at the edges where thick deposits on the back of the substrate prevented illumination of the interface, the sapphire was removed, while leaving large crack-free areas of GaN. One crack, which completely broke this film into two pieces, developed after the sapphire was removed. The right-hand segment of GaN has approximately 10 cm^2 area, and is, to date, the highest quality wafer, in terms of material and crack-free area, that we have fabricated, but similar results in terms of size have been obtained several times. For instance, a complete self-supporting 2 inch diameter GaN wafer which contained several partial cracks has also been produced.

An important characteristic of the films after liftoff is that they exhibit little or no bowing, greatly facilitating their application in homoepitaxy or lithography. The use of such films as substrates for nitride film growth for devices is also likely to require polishing, which the bowing hinders, either to smooth the epitaxial HVPE surface or to remove roughness caused by the scanned laser beam on the back surface. As an illustration of the relaxation of the bowing, Fig. 3 shows X-ray diffraction rocking curves from the front surface of the 275 μm thick GaN film, before and after liftoff. A relatively narrow (002) FWHM of 153 arcsec was obtained before liftoff when the X-ray beam was 0.1 mm wide, which projects to a 0.3 mm spot on the sample. Increasing the beam width significantly increases the FWHM, as shown for the 0.5-mm slit measurement of 380 arcsec. This can be attributed to a bowing radius of approximately 0.8 m. The structure of the 0.5-mm measurement also indicates that the curvature is not homogeneous, but rather that individual grains (of some tenths of a millimeter size) are tilted with respect to each other.

To estimate the expected bowing radius, we take published values for the thermal linear expansion of Al_2O_3[18] and GaN.[19] For GaN, the a-axis expansion from room temperature to 600°C is about 0.3%, and to 1000°C, 0.5%. The corresponding numbers for sapphire are 0.45% and 0.8%, yielding a relative mismatch between the two materials of 0.15% and

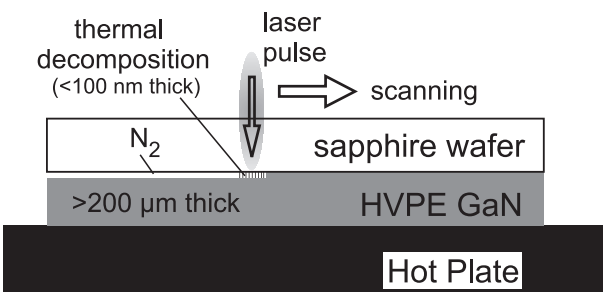

Fig. 1. Schematic diagram of the laser process for removal of the sapphire substrate from thick HVPE-grown GaN films.

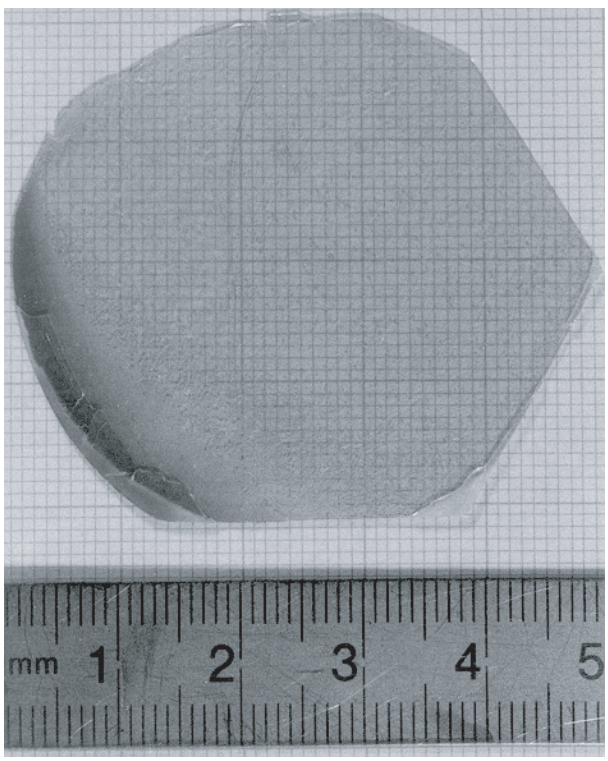

Fig. 2. Image of a 275 μm thick free-standing GaN film, after removal from the 2 in. sapphire substrate.

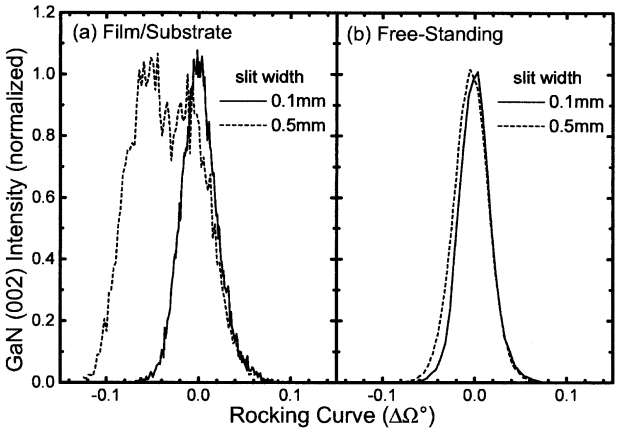

Fig. 3. X-ray diffraction rocking curves from the film shown in Fig. 2, before and after liftoff. The measured area is varied with the slit width.

0.3%. For the case of a wafer with 0.6 mm total thickness, where both faces are relaxed to the respective bulk lattice constants, the 0.15% mismatch would correspond to a 0.4 m bowing radius. This case is not unrealistic for the current films, where X-ray diffraction shows that the GaN film has essentially bulk lattice constants at the surface. The observed bowing of 0.8 m is thus weaker than could be expected for cooling from growth temperature or even from 600°C. This is consistent with the idea of some inelastic relaxation during the cooling for temperatures above 600 K,[17] from which point a 0.85 m radius could be expected. The estimate also confirms that the bowing can be alleviated, for the laser liftoff, below the decomposition temperature range.

After liftoff, the FWHM for measurements with 0.1 and 0.5 mm slit widths are 137 and 153 arcsec, showing much less dependence on the spot size. These measurements, which can be fit very well with Gaussian lineshapes, allow an estimate for the radius of residual bowing of about 4 m, or about 100 μm height difference from the wafer center to the edge. Inspection with a microscope also indicates such a value in the direction perpendicular to the crack, while bowing parallel to the crack was undetectable. For this film, it appears that the small amount of residual bowing is in the opposite direction from before liftoff. Other samples have shown differing degrees of initial and residual bowing, so it appears that this may also be controllable.

These results thus demonstrate that large area free-standing GaN wafers can be produced by combining the growth of very thick GaN films by HVPE with laser-induced liftoff. The HVPE technique enabled the deposition of thick GaN films on sapphire, which although considerably bowed, were predominantly uncracked at the free surface. The separation process is compatible with simultaneous wafer heating, which was found to relieve the TEC-induced strain in the heteroepitaxial system, permitting liftoff of crack-free GaN areas as large as 10 cm^2, which in turn permanently removed wafer bow.

Acknowledgements

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1) I. Grzegory, M. Bockowski, B. Lucznik, S. Krukowski, M. Wroblewski and S. Porowski: MRS Internet J. Nitride Semicond. Res. **1** (1996) 20.
2) R. P. Vaudo, V. M. Phanse, X. Wu, Y. Golan and J. S. Speck: *2nd Int. Conf. Nitride Semiconductors, Tokushima, 1997.*
3) S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, Y. Sugimoto, T. Koyaki, H. Umemoto, M. Sano and K. Chocho: Appl. Phys. Lett. **72** (1998) 2014.
4) S. Nakamura, M. Senoh, S. Nagahama, N. Iwasa, T. Yamada, T. Matsushita, H. Kiyoku, Y. Sugimoto, T. Koyaki, H. Umemoto, M. Sano and K. Chocho: Appl. Phys. Lett. **73** (1998) 832.
5) Yu. V. Melnik, K. V. Vassilevski, I. P. Nikitina, A. I. Babanin, V. Yu. Davydov and V. A. Dmitriev: MRS Internet J. Nitride Semicond. Res. **2** (1997) 39.
6) Yu. Melnik, A. Nikolaev, I. Nikitina, K. Vassilevski and V. Dimitriev: Mater. Res. Soc. Symp. Proc. **482** (1998) 269.
7) M. K. Kelly, O. Ambacher, R. Dimitrov, R. Handschuh and M. Stutzmann: phys. status solidi (a) **159** (1997) R3.
8) M. K. Kelly, O. Ambacher, R. Dimitrov, H. Angerer, R. Handschuh and M. Stutzmann: Mater. Res. Soc. Symp. Proc. **482** (1998) 973.
9) M. K. Kelly, O. Ambacher, B. Dahlheimer, G. Groos, R. Dimitrov, H. Angerer and M. Stutzmann: Appl. Phys. Lett. **69** (1996) 1749.
10) H. Chen, R. D. Vispute, V. Talyansky, R. Enck, S. B. Ogale, T. Dahmas, S. Choopun, R. P. Sharma, T. Venkatesan, A. A. Iliadis, L. G. Salamanca-Riba and K. A. Jones: Mater. Res. Soc. Symp. Proc. **482** (1998) 1015.
11) W. S. Wong, T. Sands and N. W. Cheung: Appl. Phys. Lett. **72** (1998) 599.
12) T. Kawashima, H. Yoshikawa, S. Adachi, S. Fuke and K. Ohtsuka: J. Appl. Phys. **82** (1997) 3528.
13) G. Yu, G. Wang, H. Ishikawa, M. Umeno, T. Soga, T. Egawa, J. Watanabe and T. Jimbo: Appl. Phys. Lett. **70** (1997) 3209.
14) Z. A. Munir and A. W. Searcy: J. Chem. Phys. **42** (1965) 4223.
15) O. Ambacher, M. S. Brandt, R. Dimitrov, T. Metzger, M. Stutzmann, R. A. Fischer, A. Miehr, A. Bergmaier and G. Dollinger: J. Vac. Sci. & Technol. B **14** (1996) 3532.
16) M. K. Kelly, M. Armbruster and A. Koch: unpublished.
17) M. Leszczynski, T. Suski, H. Teisseyre, P. Perlin, I. Grzegory, J. Jun, S. Porowski and T. D. Moustakas: J. Appl. Phys. **76** (1994) 4909.
18) *Thermal Expansion Nonmetallic Solids,* eds. Y. S. Touloukian, R. K. Kirby, R. E. Taylor and T. Y. R. Lee (IFI/Plenum, New York, 1977) Thermophysical Properties of Matter Vol. 13, p. 177.
19) K. Wang and R. R. Reeber: Mater. Res. Soc. Symp. Proc. **482** (1998) 863.