P-type silicon as hole supplier for nitride-based UVC LEDs

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Abstract

The ineffective p-type doping of nitrides using magnesium (Mg), the best available dopant, has limited the development and performance of all III-nitride-based devices, including bipolar junction transistors and light emitting diodes (LEDs). For nitride-based ultraviolet (UV) LEDs, as the Al composition increases for achieving shorter wavelengths (e.g. <280 nm) into the UVC spectral range, the p-type doping issue, which causes very inefficient hole injection, becomes more severe than ever. In this work, we report the detailed study of using p-type Si as the hole supplier for high-Al composition UVC LEDs. We first describe the method of Si/GaN junction formation, where the lattice-mismatch challenge between Si and GaN is overcome by using a 0.5 nm thick Al₂O₃ layer at the interface. This serves as a physical separation layer between the two materials as well as a passivation, tunneling, and thermal buffer layer. High-resolution transmission electron microscope image illustrates the high-quality interface between Si and GaN. We further detail the hole transport mechanism of the p–p Si/GaN isotype junction through both simulations and experiments. The enhanced hole concentration in the AlGaN/AlN multiple quantum wells (MQWs) due to the use of p-type Si as the hole supplier is verified through comparison with conventional UVC LEDs. Finally, high-performance UVC LEDs made with AlN/AlGaN (Al:72%) MQWs employing p-type Si as their hole suppliers are demonstrated experimentally to serve as an example of the novel hole injector strategy.

1. Introduction

A fundamental limitation of III-nitride materials, particularly the nitrides with a high Al content, for junction type devices (e.g. light emitting diodes (LEDs) and bipolar junction transistors) is their ineffective p-type doping. Using magnesium (Mg), the best p-type dopant, to dope GaN, the highest available free hole concentration can reach the low 18th order due to the high ionization energy of Mg in GaN (0.2−0.15 eV for the Mg concentration between 1 × 10¹⁸ and 5 × 10¹⁹ cm⁻³ [1]). Such a hole concentration has enabled high-efficiency blue LEDs. However, the even higher ionization energy of Mg in AlGaN leads to a much lower hole concentration than the 18th order in p-type AlGaN. The ineffective p-type doping in AlGaN has severely hindered the development of AlGaN-based ultraviolet (UV) LEDs. A higher Al composition in AlGaN for shorter UV wavelengths would
result in a higher ionization energy of Mg (e.g. ~0.5 eV for AlN:Mg) [2–4]. At present, all AlGaN-based LEDs employ p-type GaN as their hole injector. The thickness of the p-type GaN is typically 100–200 nm thick as needed for a p-type contact layer. Due to the lattice mismatch between GaN and AlGaN, the quality of p-type GaN is often degraded, which further decreases the hole concentration in p-type GaN. On the other hand, n-type nitrides can reach a fairly high electron concentration level (close to 19th order), leading to a severe imbalance between the electron and hole concentrations within the active region [5]. The insufficient hole supply for III-nitrides contributes to the poor performance of LEDs made therefrom.

To improve the free hole concentration in high-Al content AlGaN materials, researchers have developed the approach of polarization doping [6–10] for the purpose of enhancing electrical conductivity. By linearly grading the Al composition of AlGaN, the polarization bound charge spreads to a 3D form and induces the formation of a mobile 3D carrier gas of the opposite charge, resulting in an increased carrier concentration [11–15] over impurity doping. However, to produce substantial effective doping, there must be a strong composition gradient (a large range of Al:Ga ratio). This capability diminishes as the Al composition in AlGaN quantum wells approaches the binary endpoint, AlN, which is necessary to generate high energy photons. Another method to improve the hole concentration requires the use of nanowires [16] where high strain can be accommodated in p-type GaN such that an improved fraction of Mg ionization in GaN is expected. Nevertheless, all these methods are incremental measures to improve the free hole concentrations in the quantum wells of UV LEDs.

In this paper, we describe a novel measure to substantially increase the hole concentration in nitride-based multiple quantum wells (MQWs). Instead of using Mg-doped nitride as the hole supplier, we used p-type Si for n-GaN MQWs. Since Si can be heavily doped (20th–21st order) into p-type Si using boron atoms, for which the ionization energy of boron is in the level of thermal energy, even if partial (e.g. ~10%) free holes in Si can be injected into the nitride-based MQWs, the imbalance between electron and hole concentrations in the MQWs could be greatly improved. However, the epitaxy of GaN on Si is challenging mainly due to a remarkable lattice mismatch. Si (111) plane is commonly chosen due to its trigonal symmetry facilitating a hexagonal Wurtzite structure. Due to lattice parameter differences, i.e. 0.3823 nm for Si (111) versus 0.3112 nm for AlN (0001) (or 0.3189 nm for GaN (0001)), the interface density of states can be very high with epitaxial growth [17]. The extensive practice of direct wafer bonding between two materials, which leads to poor interface quality with a very high density of interface states, has also proven to be infeasible for forming Si/GaN junctions of needed quality [18].

To solve the lattice mismatch challenge, we employed the ultrathin oxide interfaced heterostructure method [19, 20] to realize the Si/III-nitride isotype heterojunction. By inserting a layer of ultrathin oxide in between two semiconductors of any sized lattice constants, the two semiconductors are physically separated. As a result, interface defects due to lattice mismatch and/or crystal disorientation, as happens in wafer bonding, can be avoided. Without the concern of lattice mismatch, Si [100] was adopted based on the SOI material availability. The oxide type was selected such that it could effectively passivate the surfaces of the two semiconductors. If the oxide is sufficiently thin, it will serve as a quantum tunneling layer that allows the passage of both electrons and holes. The thickness of the oxide was determined by satisfying the requirements of both the surface passivation and efficient quantum tunneling. Practically, for a p–n diode fabricated using the heterostructure method, the influence of the oxide thickness is reflected on the diode ideality factor, rectification ratio, and reverse breakdown characteristics. With an optimum thickness of the oxide, ideal p–n diodes are readily achievable [20]. Since most oxides have plasticity at certain elevated temperatures, the oxides can serve as a thermal buffer layer that eases the bonding of the two semiconductors during chemical bonding to form the heterostructure.

The abovementioned heterostructure method is supposed to provide a universal scheme to form ideal abrupt junctions between dissimilar bulk (3D) semiconductors with minimum interface density of states. Nevertheless, application of the unique lattice-mismatched heterostructure method to solve the poor p-type doping challenges of III-nitrides has been very limited [21].

In this study, we formed a p-type Si/p-type GaN junction to enable the p-type Si as a hole injector. Heavily p-type doped Si nanomembrane (NM) was released from silicon-on-insulator (SOI), transferred and chemically bonded via rapid thermal anneal (RTA) to a p-type GaN layer that was first coated with a 0.5 nm thick Al2O3 layer using atomic layer deposition (ALD). Al2O3 is already well-known to be able to passivate both Si [22, 23] and GaN [24, 25] effectively. The 0.5 nm thickness of the ALD Al2O3 layer was decided based on the best measured current density–voltage characteristics of p-Si/Al2O3/p-GaN heterostructures. High-resolution transmission electron microscopy (HRTEM) was used to reveal the detailed lattice structure of the Si/Al2O3/GaN interface. The electrical conductivity and band alignment of the p-type Si/GaN isotype junction were experimentally examined via current–voltage (I–V) and capacitance–voltage (C–V) characterizations. The calculated band alignment information was then used to examine the hole injection condition into AlGaN/AiN MQWs via simulations. Finally, UVC LEDs sequentially consisting of 100 nm transferred p-type Si, 0.5 nm ALD Al2O3, and an epitaxially grown structure including 20 nm p-GaN, 2 nm Al0.75Ga0.25N/6 nm AlN x3 MQWs,
and 600 nm n-type Al$_{0.74}$Ga$_{0.26}$N on an AlN substrate were demonstrated with high performance to prove the practicality of using p-type Si as an efficient hole supplier.

2. Results and discussion

2.1. Si/GaN heterojunction formation

Figure 1 illustrates the Al$_2$O$_3$-interfaced Si/GaN junction formation method and fabrication process flow. In brief, a 0.5 nm thick Al$_2$O$_3$ layer was deposited using ALD on a 200 nm thick p-type GaN layer that was grown on an MQWs LED epitaxial structure. A heavily boron doped (5 × 10$^{19}$ cm$^{-3}$) SOI substrate was used to create single-crystalline Si NM of 100 nm thick by sacrificially etching away the buried oxide using hydrofluoric acid. The released p-type Si NM was transferred to the Al$_2$O$_3$-coated GaN and a thermal anneal was applied to form chemical bonding. Metal electrodes were formed on the Si and GaN surface via photolithography.

![Flowchart of Si/GaN heterojunction formation](image)

2.2. Band alignment of p-Si/Al$_2$O$_3$/p-GaN heterojunction

Despite the high-quality interface between Si and GaN, whether the band alignment between Si and GaN is suitable for hole transport is unknown. Thus, the electrical characteristics of the p-Si/Al$_2$O$_3$/p-GaN isotype heterojunction were experimentally examined. The I–V curves were measured from the p-Si/Al$_2$O$_3$/p-GaN isotype heterojunction and the data are plotted in linear scale, as shown in figure 2(b). The curves exhibit nearly Ohmic behavior with a very small ‘turn-on’ voltage. As the spacing between the metal contacts (shown in figure 1(b)) increased from 8 μm to 10 μm and 20 μm, the current decreased for a given voltage, as expected, due
to the larger lateral current spreading resistance in the p-GaN region. Furthermore, the \( C-V \) measurements of the p-Si/Al\(_2\)O\(_3\)/p-GaN heterojunction were carried out at a sweeping frequency of 1 MHz. The flat-band voltage was extracted to be \(-0.97\) V from the \( 1/C^2-V \) plot (figure 2(c)), which indicated a built-in potential \( \Phi \) of \(-0.97\) eV for the p-Si/Al\(_2\)O\(_3\)/p-GaN heterojunction. Since the free hole concentration of p-Si (\( 5 \times 10^{19} \) cm\(^{-3} \)) is more than one order of magnitude higher than that of p-GaN (\( \sim 1 \times 10^{18} \) cm\(^{-3} \)), it is reasonable to assume that the built-in potential mainly originated from the p-GaN (one-sided junction), which was manifested as a 0.97 eV downward band bending of the Ga-polar surface in the p-GaN, as shown in figure 2(d).

Additionally, x-ray photoelectron spectroscopy (XPS) was employed to examine the surface/interface of p-GaN with Al\(_2\)O\(_3\) ALD deposition (figure A1 in the supplementary information (SI) is available online at stacks.iop.org/NJP/21/023011/mmedia), which revealed a downward band bending value of 1.00 eV, consistent with the 0.97 eV band bending value obtained from the \( C-V \) analysis.

Based on the measurements of the built-in potential, the valence band offset between p-Si and p-GaN can be determined by the following expression [26]:

\[
\Phi = \Delta E_v - \delta_{p-Si} + \delta_{p-GaN},
\]

where \( \Phi \) is the built-in potential, extracted as \(-0.97\) eV. The energy differences, \( \delta_{p-Si} \) and \( \delta_{p-GaN} \), between Fermi levels and valence bands (\( E_F-E_v \)) for heavily doped p-Si and p-GaN (assuming an acceptor ionization ratio of 10% and no strain in GaN) were calculated to be \(-0.002\) eV and 0.12 eV, respectively. Therefore, the valence band offset \( |\Delta E_v| \) is 1.092 eV for the p-Si/Al\(_2\)O\(_3\)/p-GaN heterojunction. In comparison to the intrinsic \( |\Delta E_v| \) value of 2.32 eV, which can be obtained directly from the electron affinity rule, the energy barrier for hole transport from p-Si to p-GaN was substantially lowered. It is noted that the 20 nm GaN layer should have some strain due to the lattice mismatch between GaN and AlGaN, and the strain in GaN (the value is unknown) can change the bandgap of GaN and the band offset values. However, even if a fully strained GaN is considered, the valence band offset value (estimated to be \(-0.09\) eV) is still too small to undermine the hole injection mechanism. As a result, a much larger concentration of holes can be injected from Si into the MQWs.
transport across the heterojunction via intra-band tunneling and thermal emission, respectively, leading to quasi-Ohmic $I$–$V$ characteristics, as supported by the electrical measurements (figure 2(b)).

### 2.3. Effects of $p$-$Si/Al_{2}O_{3}/p$-GaN heterojunction on hole injection in UVC LEDs

To examine the effects of $p$-$Si/Al_{2}O_{3}/p$-GaN heterojunction on hole injection in UVC LEDs, two LED structures, shown in figure 3(a), were simulated using Silvaco®. Intra-band tunneling model was employed, and a polarization strength factor (a constant scale factor multiplied by the calculated spontaneous and piezoelectric polarization charges) of 0.4 was used in the simulations. There can be different types of screening effects to the field induced by polarization charge, which makes the scale factor smaller than 1 in most cases, such as ionization of dopant [27, 28]. The only difference between the two LED structures is their $p$-type sides: LED A has the $p$-$Si/Al_{2}O_{3}/p$-GaN heterojunction and the thickness of the $p$-type GaN layer is 20 nm, while LED B only has a 100 nm thick $p$-type GaN layer. After applying the band alignment of $p$-$Si/Al_{2}O_{3}/p$-GaN heterojunction as shown in figure 2(d), the band structures of LED A under zero bias (equilibrium) is plotted in figure 3(b)(i). For comparison, the band structure of LED B is plotted in figure 3(b)(ii). Figure 3(c) plots the band structures of the two LEDs under forward bias. As can be seen from figure 3(c)(i), the 20 nm $p$-type GaN in LED A is fully depleted and becomes a drift region for holes under forward bias. The strong internal electric field in the $p$-GaN layer, which caused the steep bending, is a combined effect of polarization and applied voltage. As a result, the 20 nm $p$-type GaN does not function as a hole injector in LED A, but rather a hole transport layer. In contrast, the 100 nm $p$-type GaN in LED B is only partially depleted and therefore is a hole supplier layer (figure 3(c)(ii)).

For LED A, under forward bias, the hole transport from the $p$-type Si into the $p$-type GaN became easier, as the triangular-shape energy barrier between the $p$-type Si and $p$-type GaN ($0.97$ eV) became narrower ($\sim 1$ nm) than its equilibrium state (figure 3(b)(i)), as shown in figure 3(c)(i), to facilitate hole tunneling. The favorable overall energy band alignment enabled efficient hole transport from the $p$-type Si to the drift region, and further to the MQWs region.
The ample hole supply from the p-type Si and the efficient hole transport path led to a much higher hole density in the MQWs of LED A than in LED B and thus a more balanced electron/hole density in LED A (the blue line in figure 3(d)(i)) than LED B (figure 3(d)(ii)), even though the p-type Si was only doped to the level of $5 \times 10^{19} \text{ cm}^{-3}$. As the doping concentration of the p-type Si was increased to $1 \times 10^{20} \text{ cm}^{-3}$ (without changing any other LED parameters), the hole density in the MQWs of LED A further increased (the green line in figure 3(d)(i)). The increase in hole density in the MQWs following the increase in the hole concentration in the p-type Si furthermore verified that the p-type Si functioned as a hole supplier.

Figure 3(c) plots the direct comparison of hole densities between the two LEDs in linear scale with the inset showing a logarithmic scale plot. It is observed that the hole density in LED A, especially within the first two QWs, is significantly enhanced in comparison to LED B. The enhanced density of holes in the MQW is expected to enhance the electron–hole radiative recombination and therefore, the performance of UVC LEDs. The more holes that are present in the MQWs, the higher the recombination rate. Figure B1 shows the higher recombination rate when the hole concentration in the p-type Si is higher. It is noted that the p-type Si hole injector approach used in this work is readily applicable to UV LEDs covering a broad range of wavelengths.

### 2.4. Experimental application of p-type Si hole injector to UVC LEDs

The epitaxial layers of LED A (figure 3(a)) were grown on an AlN substrate by low pressure organometallic vapor phase epitaxy (LP-OEVPE) in a custom high-temperature reactor (see the SI for details of epitaxial growth). Two different LED samples have been grown that differ only by the thickness of the p-type GaN layer. The 20 nm Mg-doped p-type GaN layer here was grown, and also serves as a barrier layer to circumvent the rapid oxidation of the AlN surface (see figure C1 in the SI). The procedure to form the p-type Si hole injector via Si NM transfer and bonding is identical to that shown in figure 1. A detailed LED fabrication process flow is shown in figure D1 in the SI. The completed three-dimensional UVC LED structure is illustrated in figure 4(a). Figure 4(b) shows a scanning electron microscope (SEM) image of a piece of Si NM that was bonded to the epitaxial LED layer. Figure 4(c) shows an SEM image of a part of a fabricated UVC LED and the inset shows an optical image of a top view of the LED.

It is crucial to maintain strain-free bonding during the bonding process, since the strain in the Si NM can create unwanted band bending or even surface states at the interface. In figure 4(d), the x-ray diffraction (XRD) 2-theta-omega scan range from 30° to 80° at $\sim0.05^\circ$ steps taken from the completed p-Si/AlO$_3$/p-GaN/i-AlN/i-Al$_{0.75}$Ga$_{0.25}$N/AlN MQWs/n-Al$_{0.75}$Ga$_{0.25}$N/AlN structure shows three peaks corresponding to the AlGaN (002) direction, Al$_2$O$_3$ (002), and Si (001) directions, respectively. FWHM values of
0.075° and 0.1° from AlGaN and Si indicate that both the AlGaN epi-layer and transferred Si NM layer maintained good crystallinity during the transfer/bonding processes. The appearance of the weak Al2O3 peak at 42.1° in the XRD spectrum (figure 4(d)) indicates the existence of crystalline Al2O3, which may be attributed to the re-crystallization of the ALD interfacial Al2O3 during the annealing process. The weak intensity and the wide FWHM is attributed to the small thickness and may also imply that the ALD deposited Al2O3 was only partially crystallized, which is consistent with the HRTEM image shown in figure 2(a). To evaluate the strain of the Si/AlN/AlGaN heterostructure, Raman spectroscopy was carried out using a Horiba LabRAM ARAMIS Raman confocal microscope equipped with a 50× objective lens and a monochromator that has a 1000 mm focal length with an f/8 aperture. The spectrometer resolution is 0.045 cm⁻¹ and the laser spot size is about 1 μm. The power of the 532 nm green excitation line (lexc) is 18.5 mW. As shown in figure 4(e), the Raman characteristic peak for Si nanomembrane [29] that is taken from the Si/AIN/AlGaN heterostructure appeared at 521 cm⁻¹, indicating that the transfer-printed Si nanomembrane layer did not experience any strain due to the bonding process. Also, the two phonon modes which appeared around 660 cm⁻¹ (E₂(high) mode) and 900 cm⁻¹ (A₁(LO) mode) implies that the top AlN electron barrier layer did not suffer from the built-in strain due to the lattice mismatch or different thermal expansion coefficient between the AlGaN [30] epi-layer and the substrate [31]. Overall, both the XRD results and Raman peak indicate that an extremely low strain and high crystal quality AlₓGa₁₋ₓN epi-layer was grown by using an AlN substrate. In addition, the transfer-printed Si nanomembrane layer does not create strain in AlGaN/AlN epi-wafer.

The measured current density–voltage (J–V) plot for the UVC LED (figure 5(a)) shows its rectifying characteristics. Figure 5(b) shows an optical image of an LED under forward bias with weak blue emission, most likely from the p-type GaN and deep levels elsewhere in the device, which is commonly reported in UVC LED literature [32, 33]. The linear scale of the electroluminescence (EL) spectra for various current densities from 15.3 to 244.5 A cm⁻² are depicted in figure 5(c), where the dominant emission peak appeared at 237 nm, which is from the Al₀.72Ga₀.28N/AIN MQWs. With the current densities ranging from 15.3 to 244.5 A cm⁻², the intensity of the 237 nm emission peak monotonically increases with increasing drive. Alongside the main peak emission, there is another rather weak parasitic peak detected in the near-UV range. The parasitic peak, likely from excitation by 237 nm photons of the top p-type GaN combined with deep levels in AlGaN, is much weaker than that of the recently reported 232 nm UVC LED [7]. The measured light output power and applied voltage...
versus the current density ($L-I-V$) of the LED is plotted in figure 5(d). It can be seen that the light output power increases linearly with current density up to 245 $A \text{ cm}^{-2}$, which is equivalent to a current of 320 mA. This linear behavior implies an absence of efficiency droop in this range, which directly results from the use of p-type Si as the hole injector. The fairly balanced electron–hole density improves quantum efficiency in comparison to the conventional p-type GaN-based hole injectors [34]. Besides the efficiency droop-free behavior, 265 $\mu W$ output power was measured from the UVC LED at a current density of 245 $A \text{ cm}^{-2}$ with an external voltage bias of 27.3 V, corresponding to an external quantum efficiency of 0.016% and a wall-plug efficiency of 0.003%. The efficiency is presently limited by the use of the relatively thick 20 nm p-type GaN layer, which absorbs UVC light, and thick AlN layer (28 nm), both inducing unnecessary series resistance. The relatively large turn-on voltage is a result of the large resistance of the near intrinsic AlN electron blocking layer and the non-ohmic metal contact made on the n-type Al$_{0.74}$Ga$_{0.26}$N layer. Further optimizations of the LED structures are expected to lead to better performances. While this is not the highest reported efficiency [33, 35], the results clearly indicate the advantages of using p-type Si as a hole injector given that our LED provides the high UVC output power under constant current mode, which includes self-heating effects. It is further noticed that the ultrathin oxide interfaced lattice-mismatched heterojunction approach is applicable to other combinations of materials for various applications [20].

3. Conclusion

In summary, using transferred p-type Si as the hole injector via the ultrathin oxide-interfaced large lattice-mismatched heterojunction approach proves to be a viable route toward overcoming the poor hole injection challenge of deep-UV LEDs. The wide applicability of the approach could enable many new III-nitride-based junction devices. Applying the approach to LEDs of shorter wavelengths and large LED arrays may be one of the directions of future research.

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Competing interests

The authors declare no competing financial interests.

Author contributions

SJC, DL, and J-HS contributed to the work equally. All performed the research. DL, BM, RD, JDA, SJC, KK, ZM, WZ, and J-HS interpreted the data and wrote the manuscript. DZ contributed to manuscript writing; JDA managed research progress. ZM designed the research.

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