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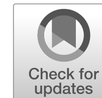

Direct high-temperature growth of GaN on Si using trimethylaluminum preflow enabling vertically-conducting heterostructures

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In this work, we demonstrate that GaN can be directly grown at high temperature on Si(111) substrates by metalorganic CVD without using any intentional AlN buffer, by simply employing a trimethylaluminum (TMAl) preflow. We found that n-GaN layers directly grown on n-Si with a TMAl preflow not only present a better crystalline quality compared to the use of thin AlN buffers, but also exhibit orders-of-magnitude improvement in vertical current conduction between GaN and Si, thanks to the absence of highly resistive AlN layers. Our proposed technique opens a new pathway for the effective realization of fully-vertical GaN-on-Si devices. © 2024 The Author(s). Published on behalf of The Japan Society of Applied Physics by IOP Publishing Ltd

Vertical GaN-on-Si diodes[1–3] and transistors[4,5] are excellent candidates for power electronics, combining the exceptional physical properties of GaN (high critical electric field, high electron mobility and high saturation velocity) with the low-cost and large diameter of silicon substrates.[6] For GaN-on-Si devices, a fully-vertical structure (where current flows vertically through electrodes located on opposite sides of the wafer) is preferable, to avoid current crowding and achieve low on-state resistance.[7,8] Fully-vertical GaN-on-Si diodes[7,8] as well as transistors[9] have been demonstrated, however, complex fabrication steps to remove the silicon substrate and resistive buffer layers are necessary, which might hinder their adoption at an industrial level.

In principle, fully-vertical structures could be easily realized simply through the n-GaN/n-Si heterostructure (the conduction band offset b`etween GaN and Si is 0.3 eV)[10] which theoretically enables vertical current conduction with minimal interface resistance from the n-doped GaN layer to the n-doped Si substrate. The challenge however is that GaN is grown on Si using an AlN buffer layer, to prevent, among others, catastrophic meltback etching of the Si substrate during GaN growth (caused by reactions between Si and Ga).[11] This highly resistive AlN interlayer undermines current conduction from GaN to the Si substrate. To improve current transport, the use of ultra-thin AlN buffers (down to 3 nm) has been proposed;[12–14] however, this still resulted in a significant barrier to current conduction and high values of on-resistance.[15,16]

Recently, our group demonstrated that GaN could be directly grown at high temperature on $ScAlMgO_4$ substrates by metalorganic CVD (MOCVD) using a trimethylaluminum (TMAl) preflow.[17] Here we show for the first time that GaN can be successfully grown directly at high temperature also on Si substrates, using a TMAl preflow, without any intentional AlN buffer layer. Our approach not only leads to a better crystalline quality, but also to a considerable enhancement of several orders of magnitude in vertical current conduction for n-GaN on n-Si structures, compared to the use of thin AlN buffers. Our method opens a new, effective and practical way for the realization of high-efficiency fully-vertical GaN-on-Si devices, in which the silicon substrate does not need to be removed and can instead be a functional part of the device. Moreover, this approach may also be applied to the growth of arbitrarily thin GaN layers directly on Si (like e.g. for buffer-free HEMTs).

To study the direct growth of GaN on Si with TMAl preflow and compare its properties to GaN layers grown using conventional AlN buffers, a set of six samples were grown: three GaN-on-Si samples using an AlN buffer layer, with AlN buffer thicknesses of 100 nm, 10 nm and 3 nm, and three GaN samples directly grown on Si with TMAl preflow, with preflow times of 5 s, 15 s, and 30 s.

All samples were grown by MOCVD using a horizontal Aixtron AIX 200/4 RF-S reactor on 2-inch Si(111) n-type phosphorus-doped wafers, with thickness of 500 μm and resistivity of ~10 Ωcm. For the GaN samples directly grown on Si with TMAl preflow, after an initial thermal cleaning step of the Si substrate in H_2 at 1050 °C (wafer surface temperature) and 100 mbar, the temperature and pressure were ramped down to 925 °C and 50 mbar for the TMAl preflow, which was performed by opening exclusively the TMAl line with a flow of 46 μmol min^{-1}, using H_2 as carrier gas. At the end of the preflow step, the TMAl line was closed, while NH_3 and trimethylgallium (TMGa) were simultaneously opened to start the GaN growth, using H_2 as carrier gas, temperature of 925 °C, pressure of 50 mbar and with TMGa and NH_3 flows of 121 μmol min^{-1} and 201 mmol min^{-1}, respectively. For the samples grown with an AlN buffer, after the initial thermal cleaning step of the Si wafer at 1050 °C, the reactor temperature and pressure were changed to 1000 °C and 50 mbar for the AlN growth, with TMAl and NH_3 flows of 46 μmol min^{-1} and 40 mmol min^{-1}, respectively; then, the temperature was ramped down to 925 °C for the GaN growth, which was performed using the same conditions as the GaN layers directly grown with TMAl preflow. All GaN layers were Si-doped, with an electron concentration of ~4 × 10^{18} cm^{-3}. To avoid cracking, all GaN layers were grown with a thickness of 110 nm, which is just below the critical thickness for crack generation determined in Ref. 18 for the case of GaN on Si with a 100 nm-thick AlN buffer layer.

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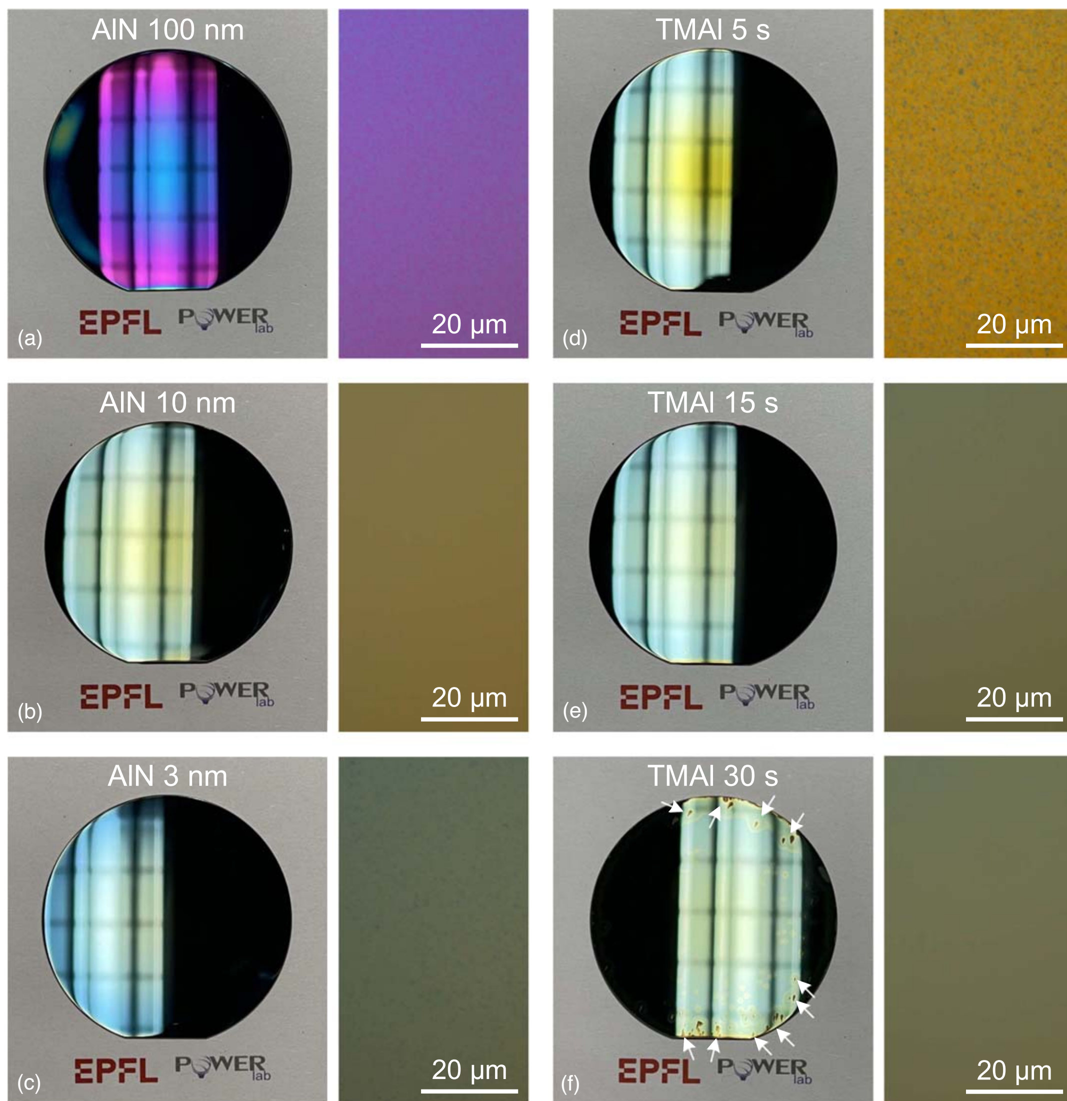

Fig. 1. Photographs and optical microscope (OM) images at the center of the GaN-on-Si wafers grown using an AlN buffer with thickness of: (a) 100 nm, (b) 10 nm, (c) 3 nm, and of the GaN-on-Si samples directly grown with a TMAl preflow of: (d) 5 s, (e) 15 s, and (f) 30 s. The GaN layer directly grown with a TMAl preflow of 15 s (e) exhibited an excellent surface morphology. The white arrows in (f) indicate meltback etched regions.

Figures 1(a)–1(c) show photographs and optical microscope (OM) images of the GaN-on-Si wafers grown with conventional AlN buffers. As expected, these growths were successful and resulted in single-crystalline GaN layers. The samples with AlN thickness of 100 nm and 10 nm [Figs. 1(a) and 1(b), respectively] presented an optically smooth, mirror-like surface, while the sample with 3 nm-thick AlN buffer appeared rougher from the OM image [right side in Fig. 1(c)], despite still presenting a mirror-like surface to the naked eye.

We then evaluated the direct growth of GaN on Si using a TMAl preflow without any intentional AlN buffer. As a first experiment, a short preflow time of 5 s was used [Fig. 1(d)], resulting in the successful growth of a crack-free, mirror-like GaN layer. Nevertheless, while the GaN surface was smooth by visual inspection, it appeared rough under OM with the highest magnification [Fig. 1(d)]. The surface morphology was greatly improved by a longer preflow time of 15 s, which resulted in a flat, mirror-like GaN surface [Fig. 1(e)]. This demonstrates that GaN can be directly grown at high temperature on Si without any intentional AlN buffer, simply using a TMAl preflow. A longer TMAl preflow of 30 s however led to a degradation of the GaN surface close to the wafer edges [Fig. 1(f)], with a rough texture typical of a meltback etched Si substrate,[11] while the wafer center remained smooth as in the 15 s preflow case. This degradation is attributed to an excessive accumulation of Al on the wafer surface due to the longer preflow, leading to meltback etching of the Si wafer, initiated by Si–Al reactions.[19] This accumulation occurs at the wafer periphery due to the horizontal reactor design (single 2-inch wafer carrier with rotation) and the low gas velocity during preflow.

The surface morphology at the center of each wafer was further investigated by atomic force microscopy (AFM). The GaN sample with a 100 nm-thick AlN buffer presented a fairly smooth surface (RMS = 1.5 nm), with some holes [Fig. 2(a)]. A similar roughness of the GaN surface was obtained for the 10 nm-thick AlN buffer, although the hole density increased [Fig. 2(b)]. Finally, the sample with a 3 nm-thick AlN buffer presented a larger hole density, and deeper than in the previous conditions [Fig. 2(c)], resulting in a much rougher surface (RMS = 3 nm), as expected from the OM images.

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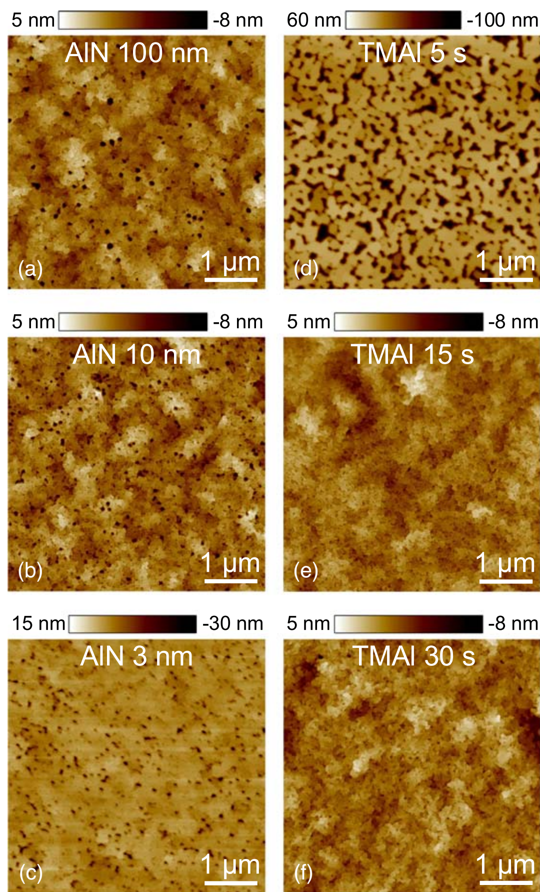

Fig. 2. AFM measurements of the GaN surface at the wafer center for the samples grown with AlN buffer thickness of: (a) 100 nm, (b) 10 nm, (c) 3 nm, and for the samples directly grown with a TMAl preflow time of: (d) 5 s, (e) 15 s, and (f) 30 s. The best surface morphology was obtained for the samples directly grown with TMAl preflows of 15 s and 30 s, with RMS roughness of 1.0 nm and no holes observed.

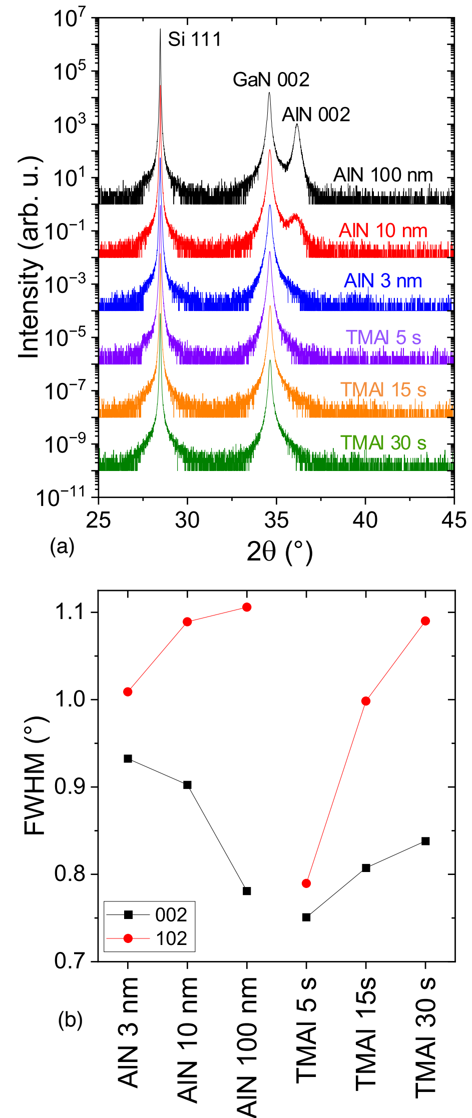

Fig. 3. (a) Symmetric 2θ-ω XRD scans and (b) FWHM values of the X-ray rocking curves (XRC) for the GaN 002 and GaN 102 diffraction peaks of the GaN-on-Si samples grown using an AlN buffer and of the GaN-on-Si samples directly grown with a TMAl preflow. Narrower XRC peaks were obtained in GaN samples directly grown with the TMAl preflow, indicating an improved crystalline quality compared to the use of conventional AlN buffers.

In the case of GaN samples directly grown with TMAl preflow, the GaN layer clearly did not coalesce using a 5 s preflow [Fig. 2(d)], leading to a very rough surface (RMS = 23 nm). On the other hand, with a preflow of 15 s [Fig. 2(e)] and 30 s [Fig. 2(f)], the GaN surface was very smooth, presenting a much better morphology than the best GaN layer obtained using an AlN buffer [Fig. 2(a)], with RMS roughness of 1.0 nm and no holes observed.

The polarity of the GaN layers directly grown with TMAl preflow and of the reference samples with AlN buffer was checked by immersion in KOH solution (7 mol l^{-1}) at 80 °C for 5 min. In all cases, the GaN layers were not etched by the KOH solution, confirming that all samples were Ga-polar.

Next, the crystalline structure of all samples was characterized by 2θ-ω X-ray diffraction (XRD) measurements [Fig. 3(a)]. For the GaN samples with AlN buffer thickness of 100 and 10 nm, the Si 111, GaN 002 and AlN 002 diffraction peaks were visible. The intensity of the AlN peak decreased for thinner buffer layers, and could no longer be detected in the 3 nm-thick buffer sample. The XRD scans of samples grown with TMAl preflow were essentially identical and indistinguishable from the case with 3 nm-thick AlN buffer, presenting only the Si 111 and GaN 002 peaks. This implies that any Al-containing layer that could be present at the GaN/Si interface is too thin to be detected.

The full width half maximum (FWHM) values of the X-ray rocking curves (XRC) for the GaN 002 and GaN 102 reflections for all GaN samples are shown in Fig. 3(b). For the GaN-on-Si samples grown using AlN buffer layers, the GaN 002 peak (which is sensitive to screw and mixed dislocations)[20] became broader for thinner AlN layers, going from 0.78° with 100 nm-thick AlN to 0.93° with 3 nm-thick AlN, as fewer of these dislocations could recombine in the buffer itself, thereby increasing their density in the GaN layer. At the same time, the GaN 102 peak, whose FWHM depends on the edge dislocation density,[20] became narrower with thinner AlN buffer layers (from 1.11° to 1.01°) due to a reduced coalescence of grains. This resulted in a rougher surface with larger density of holes (as seen in Fig. 2) and hence in a lower density of edge dislocations (which arise upon coalescence of grain boundaries).[21]

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For the samples directly grown with TMAl preflow, the GaN 002 peak had only a small dependence on the preflow time, varying from 0.75° for 5 s to 0.83° for 30 s, while the GaN 102 peak became broader with longer preflows (from 0.79° to 1.09°) due to the different grain distribution and coalescence dynamics: a TMAl preflow of 5 s was too short and led to an uncoalesced GaN layer [Fig. 2(d)], resulting in the narrowest GaN 102 peak. Increasing the TMAl preflow time to 15 s and 30 s resulted in fully coalesced samples, which consequently exhibited broader GaN 102 peaks. The larger FWHM for the 30 s TMAl sample could be due to a larger density of grains, and hence grain boundaries, induced by the longer preflow.

More importantly, the sample grown with a 15 s preflow time presented smaller XRC FWHM values compared to the ones grown with thin AlN buffers, reflecting its better crystalline quality. Therefore, by optimizing the preflow time, the direct high-temperature growth of GaN on Si with a TMAl pre-treatment not only successfully results in a fully coalesced surface without traces of meltback etching, but it also greatly improves the GaN surface morphology and its crystal quality compared to using AlN buffers.

To explain such substantial improvement in crystalline quality, we propose a growth model shown in Fig. 4. During the preflow phase, TMAl is injected into the reactor [Fig. 4(a)], and dissociates creating layers (at the nano- or subnano-scale) of Al and C or C-containing compounds (such as Al_4C_3) on the Si surface [Figs. 4(b)–4(c)]. Such decomposition process occurs for TMAl on Si(100),[22] thus here we assume a similar process happens for Si(111). Nevertheless, if this TMAl preflow step is too long, the excessive accumulation of Al may provoke meltback etching (as observed with 30 s TMAl preflow). At the end of the preflow step, the TMAl line is closed, and the NH_3 and TMGa lines are simultaneously opened [Fig. 4(d)]. In the initial moments, the Al deposited on the Si surface is quickly nitridized by NH_3 into ultra-thin AlN domains, which then act as wetting layers for the subsequent GaN growth [Fig. 4(e)]. Initially, the growth of GaN starts in the 3D mode, due to the presence of the C-containing layers acting as nanomasks [Fig. 4(f)], favoring lateral growth, as well as dislocation bending and annihilation (analogously to epitaxial lateral overgrowth).[23] As a result, this process improves the crystal quality compared to the use of a conventional AlN buffer, which lacks such beneficial nanomasking effect. This initial 3D growth occurs at the nano-scale, and therefore could not be detected by in-situ reflectance measurements. Finally, GaN islands quickly coalesce and growth continues in 2D mode [Figs. 4(g)–4(h)]. This growth mode is analogous to GaN growth on sapphire when SiH_4 is introduced with NH_3 just before growing the low-temperature GaN buffer:[24] in that case, a SiN_x nanomask is formed on the sapphire surface, leading to a similar reduction in dislocation density.

So far, we have shown that growing GaN on Si directly at high temperature with a TMAl preflow not only is possible, but also leads to an improved morphology and crystal quality. Another important aspect is that this growth technique leads to a substantially improved vertical current conduction through the GaN–Si heterostructure, compared to the use of AlN buffer layers. For the electrical

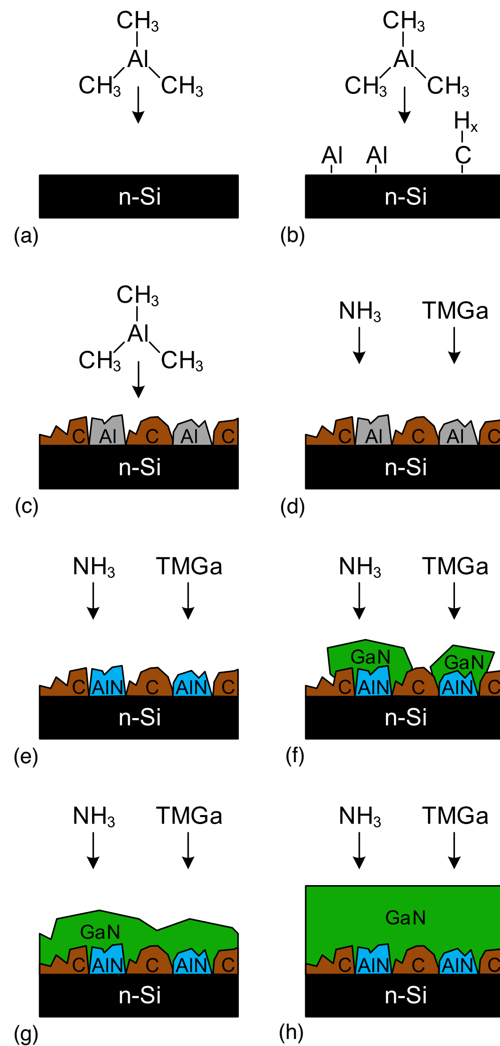

Fig. 4. Proposed mechanism for the direct growth of GaN on Si with TMAl preflow. (a) At the beginning of the preflow step, the TMAl line is opened. (b) The TMAl decomposes on the Si surface into Al and C (or C-containing compounds). (c) During the preflow step, layers of Al and C (or C-containing compounds) are formed on the Si surface, at the nanometer scale (undetectable by XRD). (d) At the end of the preflow step, the TMAl line is closed, and NH_3 and TMGa lines are simultaneously opened. (e) The Al layers on the Si surface are quickly nitridized by NH_3 into AlN. (f) GaN growth starts from the AlN islands, with the C layers acting as nanomasks, resulting in an initial 3D growth which promotes lateral overgrowth and dislocation reduction. (g) The GaN islands quickly coalesce and (h) growth continues in 2D mode.

characterization, we fabricated test structures on the Si-doped GaN layers (with electron concentration of $4 \times 10^{18}\ cm^{-3}$), by depositing Cr/Au metal stack (50 nm/250 nm) as ohmic contact to the n-GaN and In as ohmic contact to the backside of the n-Si wafers. The devices were isolated via mesa structures around the n-GaN ohmic contacts by dry etching. Among the samples grown with TMAl preflow, we considered only the case with a 15 s preflow, which presented the best surface and crystal structure (the sample with 5 s preflow was not fully coalesced, while the one with 30 s preflow presented traces of meltback etching, and therefore were excluded from this analysis). The current–voltage (I–V) characteristics for the n-GaN on n-Si sample with 15 s TMAl preflow and for the reference samples with AlN buffers are shown in Fig. 5. We note that although both contacts are ohmic in these experiments, an asymmetric I–V

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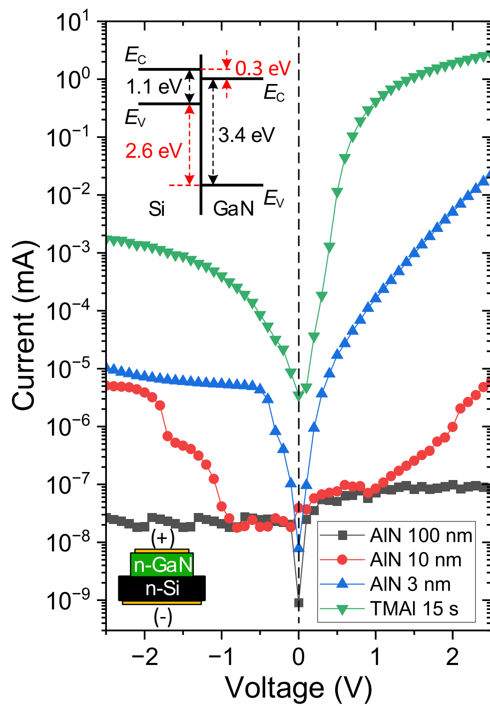

Fig. 5. Current–voltage (*I–V*) measurements of the n-GaN/n-Si heterostructures, directly grown with a 15 s TMAl preflow (green), and grown with a conventional AlN buffer, with thicknesses of 100 nm (black), 10 nm (red) and 3 nm (blue). The vertical current conduction is significantly improved by directly growing the GaN epi-layer on Si with a TMAl preflow. Here, a forward bias refers to a positive voltage applied to the n-GaN (bottom-left inset). The inset in the top-left schematically represents the band alignment of GaN and Si, showing a small energy barrier of 0.3 eV in the conduction band for a current flowing from Si to GaN, and is responsible for the asymmetric *I–V* curves.

curve is expected due to the small conduction band misalignment between GaN and Si of 0.3 eV (inset of Fig. 5). Negligible current conduction was measured on the sample with a 100 nm-thick AlN buffer, while only a very small current could flow in the sample with a 10 nm-thick AlN buffer, due to leakage through defects in the AlN layer. The 3 nm-thick AlN buffer was eventually thin enough to enable a rather limited current flow (Fig. 5). On the other hand, a considerable improvement in vertical conduction was obtained for the n-GaN sample grown with a 15 s TMAl preflow, resulting in over a 3 orders-of-magnitude higher forward current compared to the 3 nm-thick AlN buffer (Fig. 5). We attribute this substantial increase to the ultra-thin AlN layers or islands induced by the TMAl preflow (as speculated in our proposed growth mechanism), which are thin enough to allow current conduction. The same trend also occurred for reverse *I–V* curves (Fig. 5), with the sample grown with TMAl preflow exhibiting a much larger current compared to the case with a 3 nm AlN buffer. These results demonstrate the superior advantages of directly growing GaN-on-Si heterostructures with a TMAl preflow, which not only delivers better morphology and crystal quality but also ensures exceptional vertical current transport, which surpasses that of conventional AlN buffers.

In summary, in this work we demonstrated that GaN can be directly grown at high temperature on Si(111) substrates simply by using a TMAl preflow, without any intentional AlN buffer. We found that growing n-GaN layers directly on n-Si with a TMAl preflow leads to a superior surface morphology and crystalline quality compared to use of conventional AlN buffers. Furthermore, the use of TMAl preflow led to a substantial improvement of over three-order-of-magnitude in vertical current conduction through the GaN-on-Si heterostructures, enabling the realization of more efficient fully-vertical GaN-on-Si devices, in which the silicon substrate may become a functional part of the device, as well as novel devices that require effective current conduction to the substrate.

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