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Growth of GaN films with controlled out-of-plane texture on Si wafers

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ABSTRACT

GaN films were deposited on Si (400) wafers by a pulsed laser deposition technique, and it was shown that out-of-plane texture of the film is controllable although the film and the substrate do not have any interface epitaxy. The texture of the film can be set either in *c*-axis or *a*-axis direction, thereby achieving polar or nonpolar film surfaces as desired. The GaN film and Si substrate were found to be separated by a thin amorphous interface layer consisting of Si, Ga, and O atoms, that can enhance the bonding between GaN and Si. This study shows the possibility of depositing GaN films on Si wafers at low cost and the potential of integrating Si based electronics with GaN based optoelectronics.

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1. Introduction

GaN and related materials such as InGaN or AlGaN with wide direct band gaps have been under intense attention due to their technological importance for use in optoelectronic devices [1,2 for review]. Their current uses are primarily focused on light-emitting diode based optical devices, but their potential for other applications such as spintronics [3] and high power/frequency electronics [4,5] are beginning to be recognized. More recently, GaN was synthesized in the form of nanowires [6–8] and their applications are being explored along with nanowires of ZnO, which shares the same crystalline wurtzite structure as well as similar electrical properties as wide band gap semiconductors [9,10].

For practical applications, GaN thin films have usually been deposited on various single crystal substrates with consideration of lattice and symmetry matches. Successful depositions have been primarily on sapphire and SiC where good epitaxial relationships exist [11,12]. On the other hand, deposition on a Si wafer is known to be difficult to achieve because of the large mismatch between Si and GaN crystal structures [13]. There have been reports on the growth of GaN on Si (111) surfaces [14,15]. Successful GaN deposition on a Si (400) surface is rarely found in the literature [16]. Yet, the deposition of GaN films on a Si wafer might allow the use of current Si technology for the fabrication of GaN based devices and the integration of Si based electronics with GaN based optoelectronics, which can largely reduce the cost.

The capability of controlling the crystalline texture of the deposited film would enable the integration of anisotropic properties of GaN to the GaN based structures/devices. In particular, the polarity difference among different crystallographic planes of GaN [17] generated a lot of interest in controlling the texture of the GaN film because the internal polarization field along the film growth direction affects the performance of GaN based devices [18–22]. There have been very few reports on the growth of GaN films along the nonpolar directions on a few different substrates [15–23] and almost none on a Si (400) surface.

In the present work, we demonstrate that GaN films can be grown with their *c*-axis or *a*-axis perpendicular to the Si substrate surface by tuning the deposition conditions, which allows the growth of textured GaN films with either polar (c-plane) or nonpolar (m-plane) surfaces. Two approaches were explored in the present work; (I) using a textured ZnO film as a scaffold to set the texture of the subsequent GaN layer, and (II) controlling the energy of the deposited Ga and N atoms in the pulsed laser deposition (PLD) process to grow textured GaN films. Texture controlled growth of ZnO films has previously been reported [24]. It was shown that the energy of the ablated atoms that were to be deposited on the substrate was the deciding factor in determining the resulting texture of the film. Considering that ZnO and GaN are of the same family of structures, similar effects could be realized for GaN deposition, which motivated our approach (II). The use of ZnO as a buffer layer to deposit GaN on a Si wafer [25,26] has previously been considered to increase the bonding between GaN and Si, but the controlled texture of the ZnO layer can be an additional advantage, that motivated the approach (I) in the present study. The texture controlled ZnO layer is expected to serve not only as a buffer layer but also as a template to set the texture of GaN films deposited on top of a ZnO layer.

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2. Experimental details

A Si (400) wafer of ~2×2 cm^2 with native oxides on the surface was mounted on the sample stage in the PLD chamber, which was then pumped down to the base pressure in the 10^{-7} Torr range. The GaN target was prepared by pressing the commercial material (99.99% purity purchased from Alfa Aesar Inc.) into a disk with 2.5 cm diameter and 0.5 cm in thickness, and a bulk ZnO sputtering target (99.99% purity purchased from LTS Inc.) was used for PLD of ZnO films. Both ZnO and GaN targets were mounted simultaneously in the multiple target stage so they can be ablated in sequence without breaking the vacuum. Two sets of samples were made for the two routes mentioned previously. For the first set, a ZnO film was deposited first at room temperature from the ZnO target ablated with UV laser pulses ($\lambda = 248$ nm, Coherenet Compex 205) in vacuum to a desired texture by varying the laser fluence from 1 to 40 J/cm^2 as described in Ref. [24]. The substrate was then heated up to approximately 900 °C in Ar atmosphere of approximately 0.27 Pa. The flow rate of Ar gas was kept at 20 sccm. Ar pressure was increased up to ~267 Pa by decreasing the pumping rate, and ammonia (NH_3) gas was introduced into the chamber at a rate of 10 sccm during the PLD deposition of GaN. Therefore, the reaction of NH_3 molecules with the ZnO layer at high temperature [27] and the growth of GaN film occur simultaneously. Laser pulses were shot on the target at 5 Hz and the total pressure during the deposition was maintained at ~933 Pa. For the second set, very thin Au films (~2 nm thickness) were deposited on the Si wafer instead of textured ZnO films, and GaN was deposited with various laser pulse energies. The thin Au layers were deposited first on the Si wafer because GaN does not stick to the Si surface and the GaN deposition rate on the bare Si substrate is negligibly small. The texture of the GaN layer was examined by X-ray powder diffraction (XRD) scans measured in Bragg–Brentano geometry using a Panalytical X'Pert Pro system equipped with Cu X-ray source. Only the peaks corresponding to the crystal planes parallel to the substrate surface were measured in the scan. The surface morphology of the film was examined with a field emission scanning electron microscope (LEO 1550 FESEM) operated at 15 kV, and the interface structure between substrate and film was examined by cross sectional transmission electron microscopy using JEOL 4000EX TEM operated at 400 kV.

3. Results and discussion

With a high pressure maintained during the deposition, the surface of the film was found to be rough as seen in Fig. 1(a) with the grain size approximately 100–300 nm, although the film surface appears optically smooth when observed with bare eyes. Crystalline facets were clearly observed. The energy dispersive X-ray spectrum (EDS) obtained from the area in Fig. 1(a) is shown in Fig. 1(b).

Fig. 2(a) and (b) show XRD patterns of two samples with ($10\bar{1}0$) and (0001) textures of GaN, respectively. These GaN films were deposited on top of the textured ZnO layers that had been deposited with laser pulse energies of 40 and 1 J/cm^2, respectively, yielding ZnO textures [24] of ($10\bar{1}0$) and (0001) with a thickness of ~100 nm. However, it is noteworthy that XRD peaks corresponding to the ZnO under-layer were severely suppressed or completely missing in Fig. 2(a) and (b), indicating that ZnO layers' removal by NH_3 gas was achieved before GaN completely covered the ZnO film. When the ZnO under-layer is too thick to be removed completely before the GaN layer covered the top surface of the ZnO, the remaining ZnO under the GaN exhibits ZnO peaks as shown in Fig. 2(c) and (d), where ZnO peak intensity is proportional to the amount of residual ZnO layer. Therefore, the thickness of the ZnO, speed of the reaction between ZnO and NH_3, and the deposition speed of GaN should be in dynamic balance to achieve a pure GaN film on a Si wafer surface. In order to confirm that all ZnO were removed for the sample shown in Fig. 2(b), EDS were obtained from multiple spots on the film surface, one of

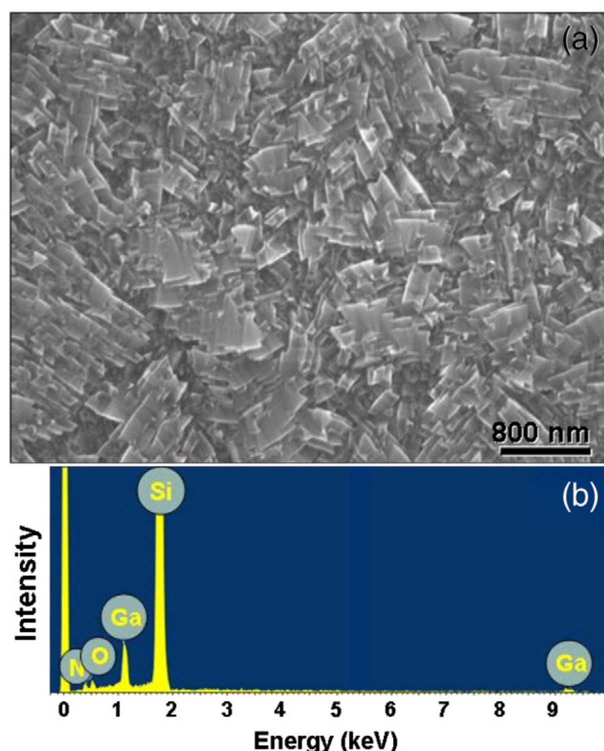

Fig. 1. (a) Surface morphology of GaN film examined with a scanning electron microscope. (b) EDS taken from the sample of GaN film deposited on top of <100 nm thick ZnO layer.

which is shown in Fig. 1(b). No Zn peak was detected at any area of the film surface, confirming that ZnO was removed completely.

As the removal of ZnO and the deposition of GaN occur simultaneously, the interface structure between the GaN film and the Si substrate is not sharp as revealed by cross-sectional transmission electron microscopy (TEM) images shown in Fig. 3(a). With the high resolution TEM (HRTEM) (Fig. 3(b) and (c)), an ~50 nm thick amorphous layer with occasional pores was observed at the interface between GaN and the Si substrate. With the EDS results in consideration, the amorphous interface layer is thought to be SiO_2 or amorphous Ga oxide, or a mixture of both. On the other hand, the crystalline nature of the GaN film could be confirmed with the high resolution lattice fringes of the GaN grains (Fig. 3(c)). Considering that

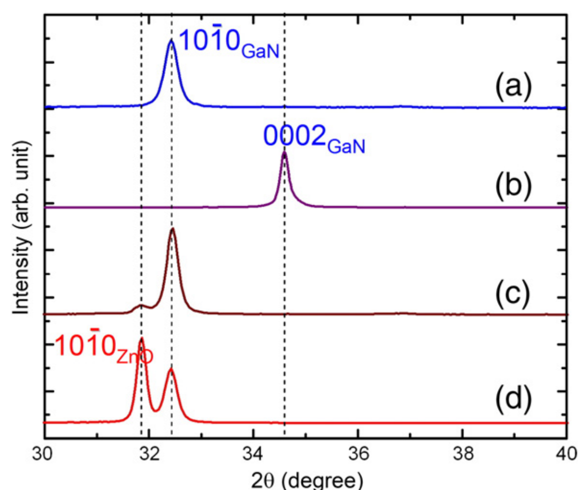

Fig. 2. X-ray diffraction patterns of GaN films on ZnO with various textures. (a) and (b) are GaN film textured in the $10\bar{1}0$ and 0002 directions, respectively. (c) and (d) are GaN textured films deposited on thicker ZnO films.

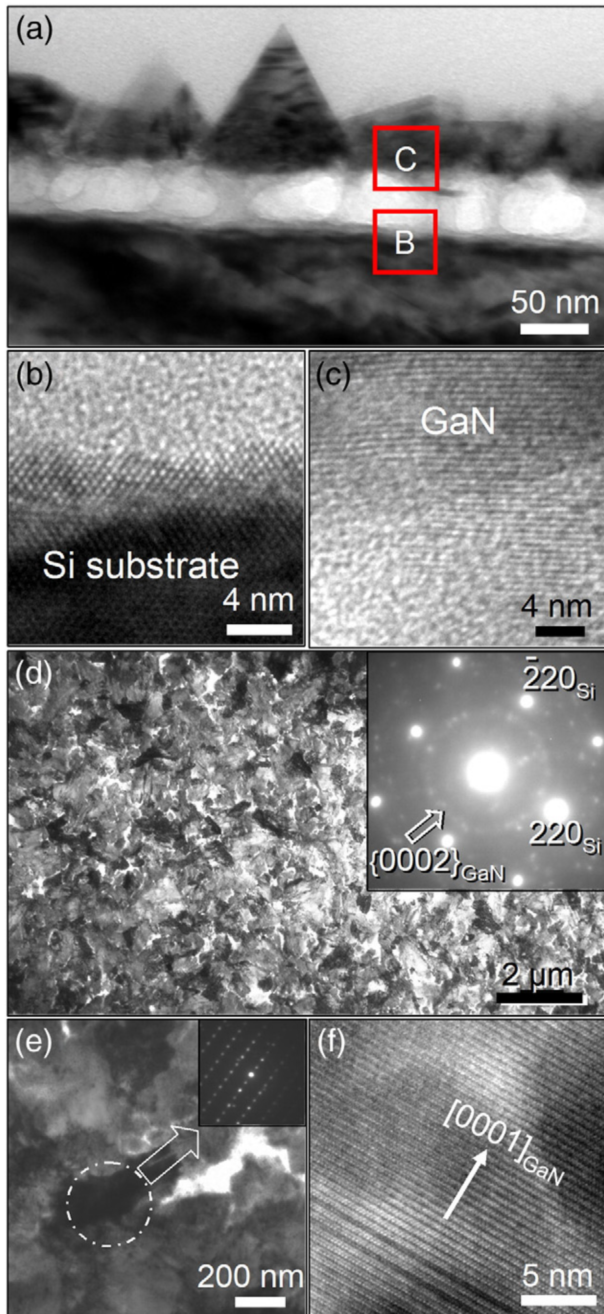

Fig. 3. (a) Cross-sectional TEM images of GaN films on Si wafer after ZnO was removed from the interface between the substrate and GaN film. (b) and (c) are HRTEM images at the interface area showing Si substrate, amorphous interface layer, and GaN film. Lattice images of Si and GaN can be observed. (d) Plan view and electron diffraction pattern (inset) of [10$\bar{1}$0] textured GaN film. One of the grain and its SAED pattern and HRTEM image are shown in (e) and (f), respectively.

a main problem with GaN on a Si wafer is the thermal mismatch between them [13], the presence of a porous and amorphous interface layer is expected to increase the robustness of the deposited film. Poor bonding of GaN to the Si wafer surface was also examined in the present study. Attempts to grow a GaN film directly on the Si surface were made, but very little or no GaN film could be formed on the bare Si surface. Plane-view TEM of the GaN film with (10$\bar{1}$0) texture (corresponding XRD pattern in Fig. 2(a)) is shown in Fig. 3(d)–(f). The electron diffraction pattern inserted in Fig. 3(d) confirms the (10$\bar{1}$0) texture of the film. Compared to the strong intensity of {0002} diffraction ring, {10$\bar{1}$0} and {10$\bar{1}$1} rings are negligible. This indicates that *c*-axis of the GaN grains is mostly within the film surface, although the grains are distributed randomly within the film plane. The selected area electron diffraction (SAED) pattern from a grain marked in the middle of Fig. 3(e) was recorded and displayed as an inset. HRTEM image from the same area was also put in Fig. 3(f). Both of them clearly indicate that the *c*-axis lies in parallel to the film plane.

The second set of samples made without ZnO layers were also examined with XRD. In analogy to the ZnO texture change with the ablating laser power [24], the film is expected to be textured with the low surface energy crystal plane when the laser fluence is minimal. Using the film deposited with laser pulses of low fluence (~1–2 J/cm^2), the preferred texture of GaN was measured to be (10$\bar{1}$0) as shown in Fig. 4(a). On the other hand, deposition with high fluence (~35 J/cm^2) laser pulses yielded the film with preferred texture of (0001) (Fig. 4(b)). Therefore, in contrast to ZnO, the (10$\bar{1}$0) surface (m-plane) of GaN has a smaller surface energy than the (0001) surface. The observed self-textures according to the surface energies of the crystal planes have been revealed because the films were deposited without an epitaxial relationship to the substrates. In the case of epitaxial growth on a substrate such as sapphire or ZnO, the film grows in the crystalline direction of the substrate regardless of the surface energy. In Fig. 4, it is also noted that the texture is not as complete as in the case of ZnO films shown in Ref. [24] probably because the differences between the surface energies of the corresponding crystal planes of GaN are not as significant as the surface energy differences between ZnO crystal planes, which is the case for GaN (10$\bar{1}$0) and (11$\bar{2}$0) surfaces (118 and 123 meV/Å^2, respectively) [28]. The high surface energy of the (0001) plane is also reflected in the shape of the GaN nanowire tips made without catalyst particles although the theoretical value was not available in the literature: Most of ZnO nanowires grown along the *c*-axis direction are terminated with flat (0001) planes at the tip ends, but the GaN nanowires were commonly observed to grow along other crystallographic directions [6,29,30]. Therefore, it is understood that the self texture of GaN film develops in balance with the energy of the deposited Ga and N atoms ablated by the pulsed laser.

It is noted that all films were polycrystalline with the thicknesses of 100–200 nm. Although the electrical quality of the films was not explored in the present study, texturing of the film would introduce the crystallographic anisotropy to the electrical as well as the structural characteristics of the film Additional layer growth on top of the textured thin films would be necessary to enhance the quality of the films for the practical applications.

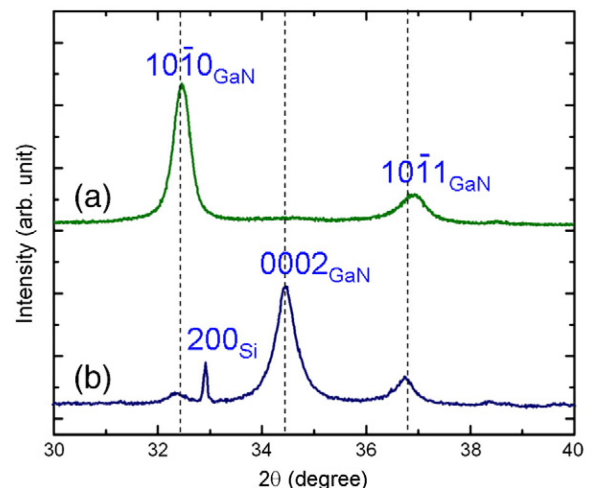

Fig. 4. Texture of GaN film was controlled with the ablating laser power. Films used in (a) and (b) were deposited with the laser fluences of 2 and 35 J/cm^2, respectively. The sharp peak at 2θ≈33° in (b) is from Si substrate.

4. Conclusion

In conclusion, GaN polycrystalline films with controlled *out-of-plane* texture either in *c*-axis or *a*-axis were grown on Si wafers, respectively, by controlling the deposition conditions. Although it was demonstrated that the self-texture of GaN can be controlled by laser fluence during the PLD procedure, a superior control was achieved by utilizing a thin temporary ZnO layer with controlled texture. The texture of the temporary layer was transferred to the GaN layer before it was eventually replaced by an amorphous Ga–Si–O layer that can enhance the bonding of GaN to the Si wafer surface. With the control of *c*-axis orientation, polarity of the film surface is controlled as well. This study shows the possibility of depositing GaN films on Si wafers at low cost and the potential of integrating Si based electronics with GaN based optoelectronics.

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