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Cubic aluminum nitride and gallium nitride thin films prepared by pulsed laser deposition

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Abstract

The growth of cubic aluminum nitride (AlN) and cubic gallium nitride (GaN) is studied. The effects of ambient pressure and substrate temperature on the structure of the AlN and GaN films are systematically investigated. It is shown that the films are amorphous when the temperature and the pressure are too low. Cubic AlN is obtained at a temperature of 800°C and a pressure of 0.2 Torr. Cubic GaN can be obtained at 600°C with a cubic AlN buffer layer.

Keywords: GaN; AlN; Crystallinity

1. Introduction

Aluminum nitride (AlN) and gallium nitride (GaN) are attractive materials for high temperature microelectronic and optoelectronic device applications. In the case of GaN, much success has been gained recently in making light emitting devices [1]. For both AlN and GaN, applications in high temperature microelectronics are emerging [2].

Both AlN and GaN crystallize either in the stable hexagonal (wurtzite, α-phase) or metastable cubic (zinc-blende, β-phase) polytypes, depending on the growth conditions. The vast majority of research on AlN and GaN thin film has been focused on the hexagonal structure. There have also been a few reports on the cubic AlN [2–6] and cubic GaN thin film [7–10].

The study of the cubic phases of AlN and GaN is important as there are significant differences in the physical and electronic properties, between the cubic and hexagonal phases [7]. For example, cubic GaN has the advantages of easier doping, easier cleaving for laser facets, and easier contacting. It also appears that β-GaN has higher electron and hole mobilities than α-GaN. Preparation of cubic GaN has been reported for a number of substrates including Si, cubic SiC, GaP, and GaAs [7–10].

In this study, we used the technique of pulsed laser deposition (PLD) to prepare β-AlN and β-GaN. PLD is now an established method for thin film deposition [11]. Its advantages have been amply demonstrated in many instances where PLD films compare favorably with films made using other methods. In this paper, we report a systematic study of the growth of cubic AlN and cubic GaN. Specifically, the effects of substrate temperature and ambient pressure on the structures of AlN and GaN thin

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films were investigated. We shall show that it is possible to obtain reasonable quality cubic AlN thin films. We shall also show that using cubic AlN as the buffer layer, it is possible to prepare cubic GaN with reasonable quality.

2. Experimental

The experiment was performed in a standard vacuum deposition chamber with a base pressure of 10^{-5} Torr. At this pressure, there is a certain degree of oxygen incorporation into the thin films. However, it is believed that the percentage will be very small especially when a solid target is used in the deposition. An ArF excimer laser at 193 nm was used. In the case of AlN, the target was a cold pressed and sintered pellet of AlN. In the case of GaN films, a liquid Ga target was used, which is also our standard technique [12]. In both cases, the substrate was sapphire (0001). The target–substrate distance was 4 cm. The deposition laser fluence was about 6 J/cm^2 at 10 Hz.

There are a few parameters which can be varied in PLD: the substrate temperature T_s, the ambient chamber pressure P, the gas flow rate, the target–substrate distance D and the laser fluence J. Among them, the substrate temperature T_s and the ambient pressure P are the most important parameters in the control of structure and quality of PLD-produced films. In this study, the deposition pressure and substrate temperature were varied systematically. For AlN films, the experimental pressure and temperature values are shown in Table 1.

All the films were deposited with 36,000 laser pulses. The film thickness obtained was about 400 nm. The structures of all the films were analyzed by XRD. For AlN, both N_2 and NH_3 are used as the reactive gas. For GaN, NH_3 was used exclusively.

Table 1
Ambient pressure and substrate temperature used in PLD of AlN

	Temperature (°C)	Pressure (Torr)	N_2 flow rate	Pump (sccm)
1	800	1×10^{-5}	0	turbo
2	800	2×10^{-4}	1.5	turbo
3	800	2×10^{-3}	15	turbo
4	800	2×10^{-2}	1.5	mechanical
5	800	2×10^{-1}	15	mechanical
6	600	2×10^{-1}	15	mechanical
7	400	2×10^{-1}	15	mechanical

Table 2
Lattice constants of relevant materials

	a (Å)	c (Å)
α-AlN	3.1114	4.9792
β-AlN	–	7.913
α-GaN	3.186	5.178
β-GaN	–	4.5
α-Al_2O_3 (sapphire)	4.7588	12.992

3. Results

3.1. Cubic AlN

The lattice constants of α-AlN, β-AlN, α-GaN, β-GaN and sapphire are listed in Table 2. It can be seen that the lattice matching condition is not satisfied in all cases. Hence, under normal conditions, the

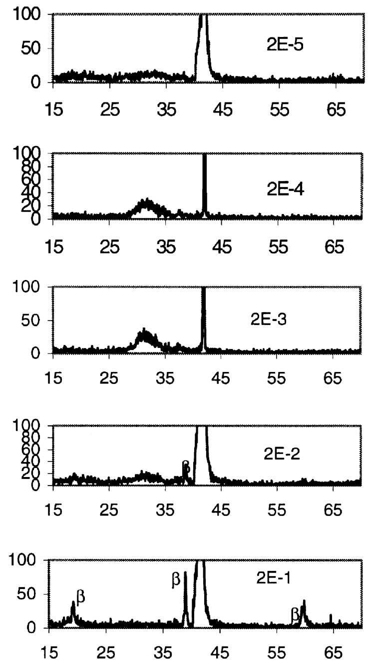

Fig. 1. XRD plots of the AlN films deposited at different ambient nitrogen pressures.

films obtained should be polycrystalline. Fig. 1 shows the XRD results of the effect of deposition pressure on the structure of the AlN films. It can be seen that the films are mostly amorphous when the pressure is too low. The quality of the film is better at higher N_2 pressures. At a pressure of 0.2 Torr, the film obtained consists mostly of β-AlN. Experimentally, it is also observed that when the ambient pressure is too high, poor films are obtained. This is presumably due to the collisional cooling effect of the laser plume during PLD [11].

The XRD data obtained is consistent with previous measurements of PLD films of AlN [12]. In Ref [12], the AlN(0002) peak is at 36°. The most prominent β-AlN peak in Fig. 1 is at 39°. This indicates that here, indeed, the cubic phase has been obtained.

Fig. 2 shows the XRD results of the effect of substrate temperature on the structure of the AlN films. It can seen that if the substrate temperature is high, the crystalline structure of the film is better. The film consists mostly of cubic β-AlN when the substrate temperature is 800°C. On the other hand, the film consists mostly of amorphous AlN when the temperature is below 400°C.

The results above show that cubic AlN film cannot be obtained when the temperature and pressure are too low. This is consistent with the fact that cubic AlN is more difficult to grow than the hexagonal phase. High ambient pressure and high substrate temperature are hence needed to activate the surface sufficiently to grow the cubic films.

Table 3
Conditions for depositing GaN

Targets	liquid Ga
Laser fluence	6 J/cm^2
Substrate	sapphire (0001)
Substrate temperature	600°C
Target–substrate distance	4 cm
Base pressure	1×10^{-5} Torr
Flow rate of NH_3 gas	15 sccm
Pressure of deposition	0.2 Torr

3.2. Cubic GaN

For the case of GaN, we have previously shown that a liquid gallium target can be used to grow good quality hexagonal GaN films [13]. It is also well known that cubic GaN can be obtained using MOCVD and other growth techniques at low substrate temperatures [9,10], but the quality of the film tends to be poor. High temperature is generally needed to improve the quality of the films, but at high temperature, the α-phase dominates.

In order to grow β-GaN at high substrate temperature, we used β-AlN as the buffer layer. The same idea has been applied in the growth of α-GaN,

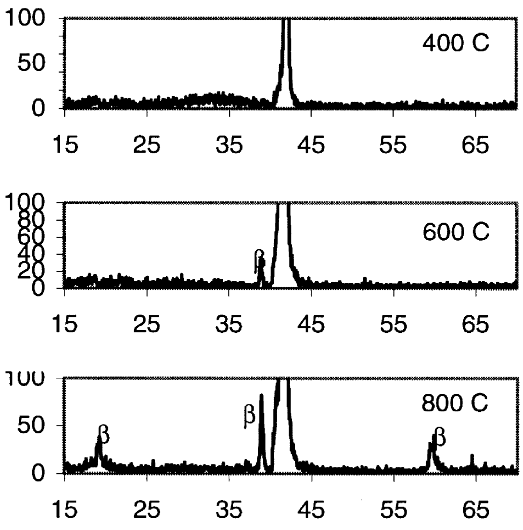

Fig. 2. XRD plots of the AlN films deposited at different substrate temperatures using N_2.

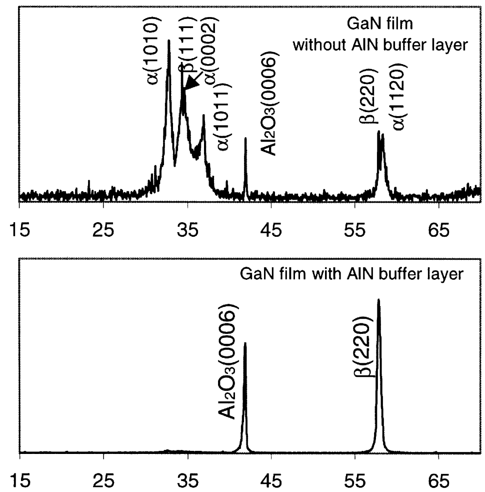

Fig. 3. XRD plots of GaN films with and without the AlN buffer layer.

where it was shown that α-AlN can be used as a buffer layer to grow α-GaN.

The β-AlN was deposited using the technique described in Section 2. Sapphire (0001) was used as the substrate. The β-AlN buffer layer was prepared by 18,000 pulses of lasers using the same conditions as mentioned above for the cubic AlN film. GaN thin films with and without the AlN buffer layer were fabricated with 10^5 pulses of lasers using the conditions shown in Table 3. The thickness of the GaN films with and without buffer layer were 1540 and 1347 nm, respectively.

Fig. 3 shows XRD results for GaN thin films with and without AlN buffer layer. In the XRD spectra of the GaN thin film without the AlN buffer layer, peaks of both hexagonal and cubic GaN coexisted at $2\theta = 34.5°$ and $58.0°$. There are also hexagonal GaN peaks at $2\theta = 34.7°$, α(0002) and 58.3°, α(1120). Cubic GaN peaks are observed at $2\theta = 34.45°$, β(111) and 57.90°, β(220). In the XRD spectra of GaN thin film with AlN buffer layer, there is only one clear peak which is the peak of cubic GaN at $2\theta = 57.90°$, β(220). Comparing the XRD plots of the GaN films with and without the AlN buffer layer, it can be seen that the former signal is much stronger and cleaner, indicating better crystallinity.

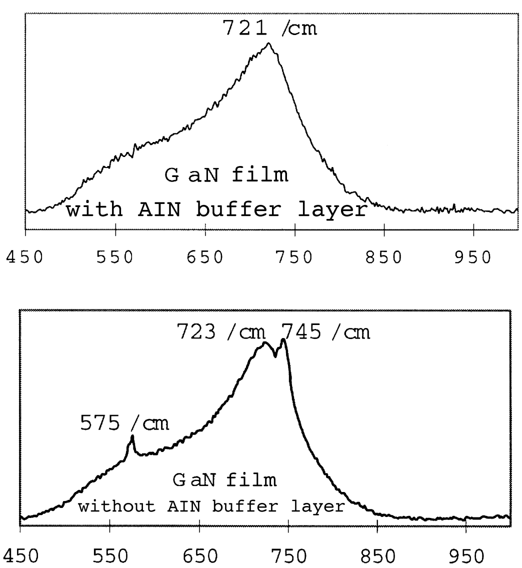

Fig. 4. Micro-Raman plots of GaN films with and without the AlN buffer layer.

In order to confirm the XRD results, a further investigation was carried out using room temperature micro-Raman spectroscopy. Fig. 4 shows the result of Raman spectra for GaN thin film with and without the AlN buffer layer. In the Raman spectra of GaN film without the AlN buffer, the peaks of both hexagonal and cubic GaN were found. Hexagonal GaN peaks are observed at 745 and 575 cm^{-1}, and also the peaks of the cubic GaN at 723 cm^{-1}. In the Raman spectra of GaN thin film with the AlN buffer layer, there is only one peak which is due to β-GaN at 721 cm^{-1}.

Clearly, the above results of XRD and Raman spectra show that GaN thin film without AlN buffer layer consists of a mixture of hexagonal and cubic GaN structures. GaN thin film with the AlN buffer layer consists predominantly of cubic GaN. The presence of only one strong diffraction peak in the XRD data also implies that the crystallinity of the β-GaN obtained is good.

4. Conclusions

In conclusion, it is shown that cubic AlN films can be obtained using PLD when the ambient pressure is 0.2 Torr and the substrate temperature is 800°C. The cubic structure is obtained even though the substrate of sapphire has a hexagonal symmetry. The conditions observed for growing cubic AlN are rather broad. However, at low pressures and/or low temperatures, the deposited films consist mostly of amorphous AlN.

In the case of PLD deposited GaN using liquid gallium target, our previous results indicated that good quality films can be produced. However, the present result indicates that those peaks are a mixture of cubic and hexagonal structures. However, using cubic AlN as a buffer, pure cubic GaN can be obtained. Further study is needed to characterize also the optoelectronic and electronic properties of the cubic GaN films obtained at 600°C using AlN buffer.

Acknowledgements

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