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EFFECTS OF THE BUFFER LAYER IN METALORGANIC VAPOUR PHASE EPITAXY OF GaN ON SAPPHIRE SUBSTRATE*

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High-quality GaN film with a smooth surface, free from cracks, can be grown on a sapphire substrate by metalorganic vapour phase epitaxy (MOVPE) using a thin AlN buffer layer. The most essential role of the buffer layer is found to be (i) the supply of the nucleation sites with the same crystal orientation as the substrate, and (ii) the promotion of the lateral growth of GaN due to the decrease in interfacial free energy between the substrate and the epitaxial GaN film.

1. INTRODUCTION

GaN is one of the most promising materials for the application of blue light emitting devices because it has a direct energy band gap of 3.39 eV at room temperature. For the growth of GaN, heteroepitaxial growth on the sapphire substrate has commonly been used But it has been very difficult to grow high-quality GaN film with a smooth surface free from cracks, because of the large lattice and thermal mismatches between GaN and sapphire. Recently, we succeeded in improving the quality of epitaxial GaN films by predepositing a thin AlN buffer layer before the growth of GaN[1], and we have also developed a high-efficiency blue light emitting device[2] by using this method.

In this paper, the role of the AlN buffer layer is investigated, and the difference in the mode of growth with and without the buffer layer is discussed.

2. PROCESS FOR THE DEPOSITION OF AlN LAYER AND THE GROWTH OF GaN FILM

Epitaxial growth was performed at atmospheric pressure by horizontal-type MOVPE reactor. Sapphire (0001)$\leqslant \pm 1°$ plane (hereafter called (0001) just-substrate) and (0001) tilted toward [10$\bar{1}$0] and [11$\bar{2}$0] by 3° (hereafter called off-angle substrate) were used for the substrate. For the deposition of AlN, trimethylaluminium (TMA) (+15 °C, 30 standard $cm^3\ min^{-1}$ H_2) and NH_3 (1 standard $l\ min^{-1}$) were used as source gases and H_2 (1.5 standard $l\ min^{-1}$) was used as carrier gas. For

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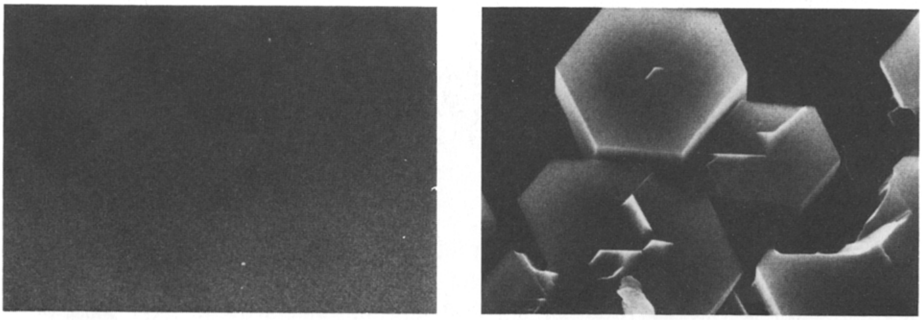
the growth of GaN, trimethylgallium (TMG)(− 15 °C, 40 standard cm^3 min^{-1} H_2) was used as the Ga source. Deposition temperature was 600 °C for AlN, and 1040 °C for GaN.

3 RESULTS AND DISCUSSION

3.1 Effects of AlN buffer layer

Figure 1(a) shows the surface morphology of GaN film grown on sapphire (0001) just-substrate with an AlN buffer layer. (The thickness of the AlN buffer layer is about 500Å.) As can be seen, GaN film with a flat surface free from cracks is obtained. However, when GaN is grown on sapphire (0001) just-substrate without an AlN buffer layer as in Fig. 1(b), a three-dimensional (3D) island growth with faceted structure appears which does not cover the whole area of the substrate, even if the growth time exceeds 30 min.



(a) (b)

Fig. 1 SEM photograph of GaN films grown on sapphire (001) just-substrate (a) with and (b) without AlN buffer layer.

Crystal quality of GaN grown on sapphire was characterized by the double crystal X-ray rocking curve (XRC) method. The full width at half maximum (FWHM) for GaN grown with AlN buffer layer was about 1.9 min, which is much narrower than the FWHM of GaN grown without AlN buffer layer (about 8.2 min). In this case, the broadening of FWHM of XRC represents the fluctuation of crystal orientation of GaN, which is reduced by using AlN buffer layer.

Photoluminescence of GaN films was measured at 4.2 K. Nondoped GaN grown with AlN buffer layer showed sharp I_2 line emission (excitons bound to neutral donor, FWHM, 1.1 meV). Its intensity was very strong compared with GaN grown without AlN buffer layer (about two orders of magnitude). Broad band emission due to deep level impurities dominated the photoluminescence spectrum of GaN grown without AlN buffer layer. From these PL measurements, it is indicated that GaN grown with AlN buffer layer has scarcely any deep level impurities and/or lattice defects compared with GaN grown without AlN buffer layer.

Hall measurement was performed by the van der Pauw method, and the result is shown in Fig. 2. All GaN layers showed n-type conduction. Carrier concentration of GaN grown with AlN buffer layer was $(2–5) \times 10^{17}$ cm^{-3} at room temperature, which is two orders of magnitude lower than that of the film grown without AlN buffer layer, and the mobility was 350–430 cm^2 V^{-1} s^{-1} at room temperature, which is about one order of magnitude higher than that of the film grown without AlN buffer layer.

All these results show that by using AlN buffer layer, defects that originate in the heteroepitaxial growth with large lattice mismatch are much reduced, and GaN films of flat surface with high crystal quality can be obtained.

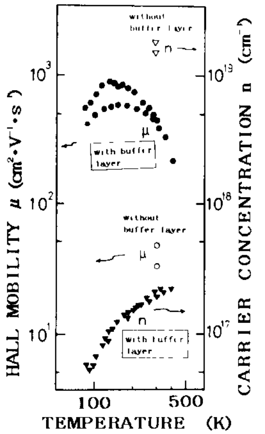


Fig 2 Electrical property of nondoped GaN films with and without AlN buffer layer

3 2 DISCUSSION

Let us consider the reason for the effect of the buffer layer on the growth process of GaN. The surface morphology of an AlN buffer layer deposited at 600 °C is shown in Fig. 3(a)(1), and its RHEED pattern in Fig. 3(b)(1). No special structure can be seen under low magnification (1000 ×), but under high magnification (50 000 ×), a fine particle-like structure can be seen. The RHEED pattern (Fig. 3(b)(1)) shows a halo pattern with diffuse spot, indicating that this film has both amorphous-like structure and crystal-like structure. GaN grown on this AlN buffer layer at 1040 °C is also shown in Fig 3, with growth times of 5 min, 10 min, 20 min and 60 min. Clearly hexagonal mesa-island growth can be seen at the initial stage of GaN on this AlN film (Fig 3(a)(2)). It can also be observed that lateral growth proceeds favourably after a certain period of time (Figs. 3(a)(3) and 3(a)(4)), that quasi two-dimensional (2D) growth then dominates (Fig. 3(a)(5)), and that finally the whole area of the substrate is fully covered and the GaN surface is flat. The corresponding RHEED pattern is shown in Fig. 3(b). Although Fig. 3(b)(2) shows a spotty pattern indicating that the GaN is three dimensional, Figs. 3(b)(3)–3(b)(5) show a streaky pattern indicating that the GaN surface is flat.

From the SEM observation, the process for the growth of GaN on sapphire using AlN buffer layer is found to consist of three stages, which are (i) nucleation of fine AlN crystallites and the island growth of GaN around the nucleation site, (ii) lateral growth of GaN islands and their coalescence, and (iii) quasi 2D growth of GaN.

As has been stated, when GaN is grown without a buffer layer, a rather longer time is necessary to cover the whole area of the substrate than when it is grown with a buffer layer. Figure 4 shows the horizontal and vertical growth rates of GaN islands.

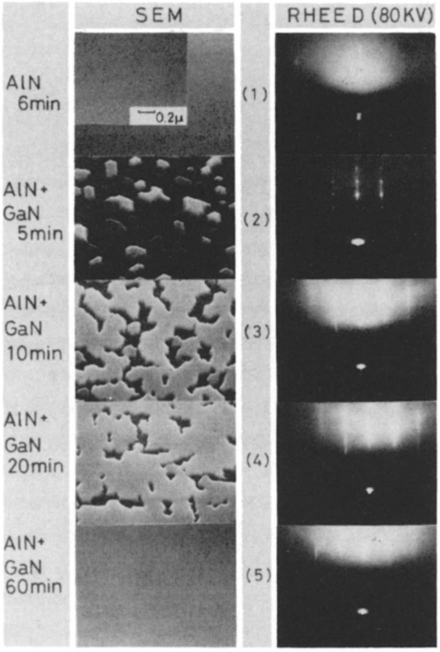


Fig 3 (a) SEM photograph and (b) corresponding RHEED pattern, of (1) AlN layer and (2–5) GaN film with different growth times

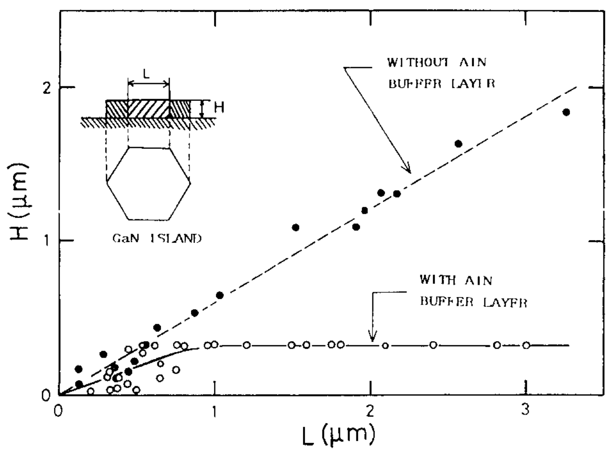


Fig 4 Horizontal *vs* vertical growth rates of GaN islands with and without AlN buffer layer

When GaN is grown with an AlN buffer layer, lateral growth clearly dominates at a certain thickness (in this case it is roughly 3000 Å.) When GaN is grown without an AlN buffer layer, the ratio of horizontal to vertical growth does not change in the whole region, indicating 3D growth

In order to clarify the effect of the density of nucleation sites, off-angle substrate (which is thought to have many nucleation sites) is used for comparison Figure 5 shows the differential interference contrast (DIC) micrograph of GaN grown without AlN buffer layer on (0001) just-substrate and 3° off-angle substrate tilted toward [10$\bar{1}$0] and [11$\bar{2}$0] sapphire. Contrary to how it grows on just-substrate, GaN covers the whole area in a short period when it is grown on off-angle substrate. This film is characterized by photoluminescence measured at 4.2 K. The FWHM of near band edge emission is much broader (13 meV) and its intensity is about two orders of magnitude weaker than that of the film grown with AlN buffer layer. It is also suggested that the quality of GaN film grown without an AlN buffer layer on the off-angle substrate is inferior to that of GaN grown with an AlN buffer layer. These results indicate that the effect of the AlN buffer layer does not originate in the supply of nucleation sites.

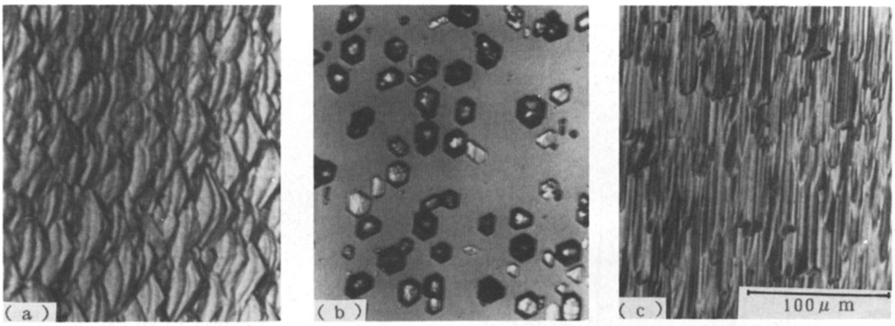


Fig 5 DIC photomicrograph of GaN grown without AlN buffer layer on sapphire substrate (a) (0001) 3° off-angle substrate tilted toward [10$\bar{1}$0], (b) (0001) just-substrate, (c) (0001) 3° off-angle substrate tilted toward [11$\bar{2}$0]

From these results, the following mechanism is suggested. The fluctuation of the orientation of nucleation sites at the initial stage of the growth of GaN using an AlN buffer layer is relatively small, and the interfacial free energy between the epitaxial film and the substrate is small, GaN is then grown laterally from the substrate Macroscopic lattice defects are not introduced even after the coalescence. Rather, the orientational fluctuation of nucleation sites grown without AlN buffer layer is thought to be rather large, containing many lattice defects. Moreover, macroscopic lattice defects are also introduced after the coalescence, and then poor crystalline quality GaN is obtained. Therefore the role of the AlN buffer layer is thought to be (i) the supply of the nucleation sites with low orientational fluctuation, and (ii) the promotion of lateral growth of GaN.

Finally, the effect on the GaN film, of the conditions under which the AlN layer is deposited, is studied. The optimum thickness of AlN is about 500 Å. If it is too thick (*i e.* 2500 Å), GaN film becomes polycrystal. It is thought that the buffer layer is too thick to transmit the crystalline information of the substrate such as the orientation When the deposition temperature of AlN exceeds or is close to the temperature at which single crystal AlN can be grown, surface morphology of GaN shows many hexagonal hillocks, and fluctuation of crystal orientation also increases Note the rough surface of the AlN layers in the SEM photograph in Fig. 6. So it is thought that lateral growth of GaN film is obstructed on these AlN layers.

4. SUMMARY

The growth mechanism of GaN grown on sapphire with and without AlN

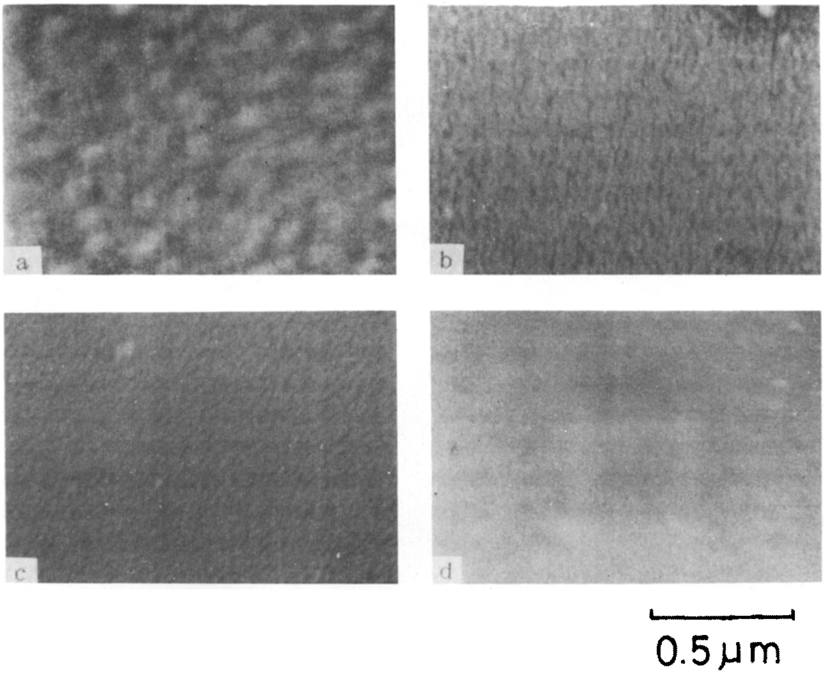


Fig 6 SEM photograph of AlN buffer layer deposited at different temperature ((a) 1100 C, (b) 1000 C, (c) 900 C, (d) 600 C)

buffer layer is investigated in this study. Two essential roles for the AlN buffer layer are found to be (i) the supply of nucleation sites of low orientational fluctuation and (ii) the promotion of lateral growth of GaN

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