Aluminum incorporation efficiencies in *A*- and *C*-plane AlGaN grown by MOVPE*

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The aluminum incorporation efficiencies in nonpolar *A*-plane and polar *C*-plane AlGaN films grown by metalorganic vapour phase epitaxy (MOVPE) are investigated. It is found that the aluminum content in *A*-plane AlGaN film is obviously higher than that in the *C*-plane sample when the growth temperature is above 1070 °C. The high aluminum incorporation efficiency is beneficial to fabricating deep ultraviolet optoelectronic devices. Moreover, the influences of the gas inlet ratio, the V/III ratio, and the chamber pressure on the aluminum content are studied. The results are important for growing the AlGaN films, especially nonpolar AlGaN epilayers.

Keywords: metalorganic chemical vapor deposition, nitrides, semiconducting III–V materials, semiconducting ternary compounds

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1. Introduction

AlGaN alloys are important for optoelectronic devices such as light emitting diodes (LEDs),[1–3] laser diodes (LDs)[4] and photodetectors (PDs)[5] in the UV spectral region from 200 nm to 365 nm, which can be tuned by changing the Al content in the AlGaN ternary alloy. These devices can be applied to important areas such as disinfection/sterilization,[2] homeland security,[2] and secure space-to-space communication.[6] However, the quantum efficiencies of these devices are still poor due to the high defect densities, high contact resistances, and low light extraction efficiencies, etc.[1,2] Moreover, the device fabricated on traditional *C*-plane AlGaN suffers the large quantum confined Stark effect (QCSE), which is caused by the strong polarization effect in the [0001] growth direction.[7] The QCSE would lead to the spatial separation of electron and hole wave functions, reducing the optical transition probability and the device efficiency.[7] Therefore, without a polarization field in the growth direction, the AlGaN optoelectronic device may achieve a high device efficiency.

As is well known, high Al content AlGaN film for a deep UV device is difficult to grow due to the low Al incorporation efficiency in AlGaN. A lot of work has been done on studying the mechanism of governing the Al content in the as-grown *C*-plane AlGaN and it has been found that many growth parameters are relevant.[8–10] However, there are few reports on the growth condition for the nonpolar AlGaN film, especially *A*-plane AlGaN.

In this paper, a comparison of the Al content between nonpolar *A*-plane and polar *C*-plane AlGaN films grown by MOCVD is made. The influences of the growth parameters, such as the trimethylaluminum (TMAl) and ammonia flow rates, the reactor pressure, and the growth temperature on the aluminum content are investigated.

2. Experimental procedure

The AlGaN samples were grown in a homemade metalorganic vapour phase epitaxy (MOVPE), which has been described in our previous paper.[11] The *A*- and *C*-plane AlGaN films were grown on the ∼ 1 μm *A*- and *C*-plane GaN templates, respectively. The methods of fabricating high-quality *A*- and *C*-plane GaN templates can be found elsewhere.[12,13] In all the experiments, the total flow rate of TMAl and trimethylgallium (TMGa) was kept constant (50 μmol/min), while the flow rate ratio between TMAl and (TMAl + TMGa) was varied from 5% to 75%. Hydrogen was used as the carrier gas. The V/III ratio which is the flow ratio between elements of groups V and III, was changed from 900 to 3600 by adjusting the ammonia flow rate. When the ammonia flow rate was changed, the hydrogen flow rate was also adjusted in order to maintain a constant total flow rate of the inlet gas. The chamber pressure was varied between 30 Torr to 70 Torr (1 Torr = 1.33322×10^2 Pa), while the growth temperature was in a range of 1010 °C–1100 °C. The growth time was fixed

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at 15 min. The compositions of the AlGaN epilayer are measured by the x-ray diffractometry (XRD: Philips X' Pert Pro diffractometer).

3. Results and discussion

3.1. Al content versus TMAl flow rate

Figure 1 shows the plots of measured Al content versus TMAl flow rate respectively for *A*- and *C*-plane AlGaN films, which are grown at a substrate temperature of 1100 °C, chamber pressure of 50 Torr and V/III ratio of 900. As expected, the Al content values in both nonpolar and polar samples increase with the TMAl flow rate since the total flow rate of group-III precursors is kept constant. The dashed line corresponds to the mass-transport limit which represents the relation of mole fraction of Al atoms to the total mole fraction of Al and Ga atoms at the chamber inlet.[9] We can see that each of all the samples has a larger Al content than the transport limit. Our results are quite different from many previous research results, in which the Al content in AlGaN is smaller than the mass transport limit.[8,9] According to the previous studies,[8–10] the deviation of Al content in AlGaN from the gas phase inlet ratio is mainly due to the gas phase parasitic reactions and the desorption of the group-III elements from the AlGaN surface. Since the parasitic reaction between TMAl and ammonia is much more serious than that between TMGa and ammonia,[6] the Al content in the layer would decrease. On the contrary, at about 1100 °C, the absorbed Al atoms are stuck to the lattice site, while the absorbed Ga atoms are not stable enough.[14] As a result, the Al content would be increased by the desorption of the Ga atoms. If the parasitic reaction dominates, the Al content would be less than the gas inlet ratio. And if the Ga desorption is a major factor, the Al content would be larger than that. In this group of experiments, the pressure and the V/III ratio are low, so the gas phase parasitic reaction would be less important than the Ga desorption. As a result, all the samples each have a larger Al content than the transport limit.

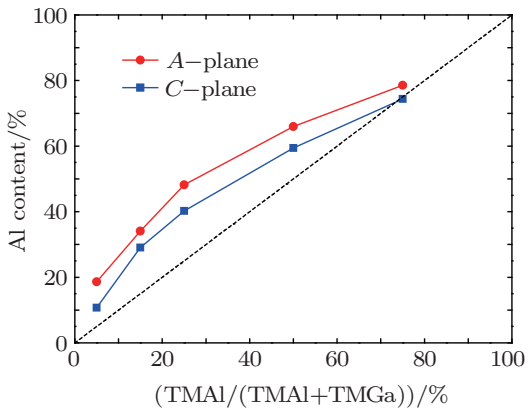


Fig. 1. (color online) Plots of Al content versus TMAl gas inlet ratio for *A*- and *C*-plane AlGaN films, where the dashed line corresponds to the mass-transport limit.

Moreover, the Al content of the *A*-plane AlGaN sample is higher than that of the *C*-plane one, which might be due to the different atomic bond configurations on *C*- and *A*-planes. At a fixed temperature, the Ga desorption rate depends on the number of surface nitrogen atom dangling bonds and the density of nitrogen atoms. As shown in Fig. 2, each surface nitrogen (N) atom has three dangling bonds that point upward in the *C*-plane surface, while a nitrogen has single dangling bonds that point upward in the *A*-plane surface. What is more, the atomic density of N atoms in the *C*-plane surface is 1.1×10^{15}/cm^2, while that in the *A*-plane surface is 0.7×10^{15}/cm^2.[15] Therefore, the Ga desorption rate of *A*-plane AlGaN is higher than that of the *C*-plane AlGaN in the present growth condition, leading to a lower efficiency of Ga incorporation into *A*-plane AlGaN. The higher Al content in *A*-plane AlGaN makes it more beneficial to fabricating the deeper-UV optoelectronic devices.

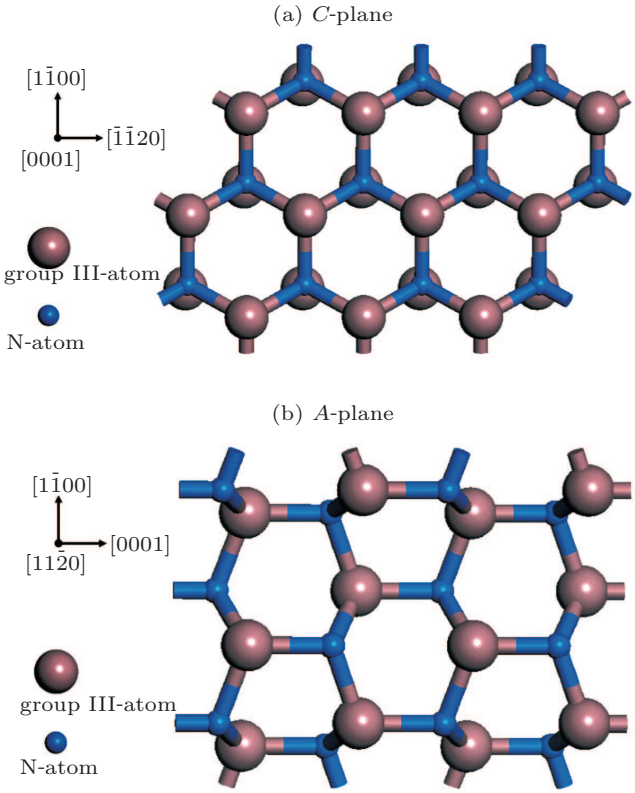


Fig. 2. (color online) Atomic configurations of the (a) *C*-plane and (b) *A*-plane AlGaN surface.

3.2. Al content versus chamber pressure

The dependencies of Al content values in *A*- and *C*-plane AlGaN on chamber pressure are shown in Fig. 3. It is obvious that the Al content decreases with pressure, which can be attributed to the enhanced parasitic reaction. At higher chamber pressure, both the TMAl and ammonia pressures are increased, so the gas phase parasitic reactions that have negative effects on the film growth become more serious. This phenomenon seems to be more severe when the TMAl flow rate is higher. We can see that when the pressure is 70 Torr and the

TMAl inlet ratio is above 0.5, the Al content in AlGaN film becomes lower than the mass-transport limit. We should note that the Ga desorption rate would be slightly increased at high pressure due to the increased H_2 partial pressure, which was demonstrated by earlier studies.[16,17] The increased Ga desorption rate can increase the Al content in AlGaN, however, it is not as important as the reduced TMAl precursor caused by the increased parasitic reactions at higher pressure, as indicated by our experiments. Moreover, we find that the Al incorporation efficiency in the *C*-plane AlGaN is much lower than that in the *A*-plane sample, especially when the TMAl flow rate is increased, which might be due to the different atomic bond configurations on *C*- and *A*-planes.

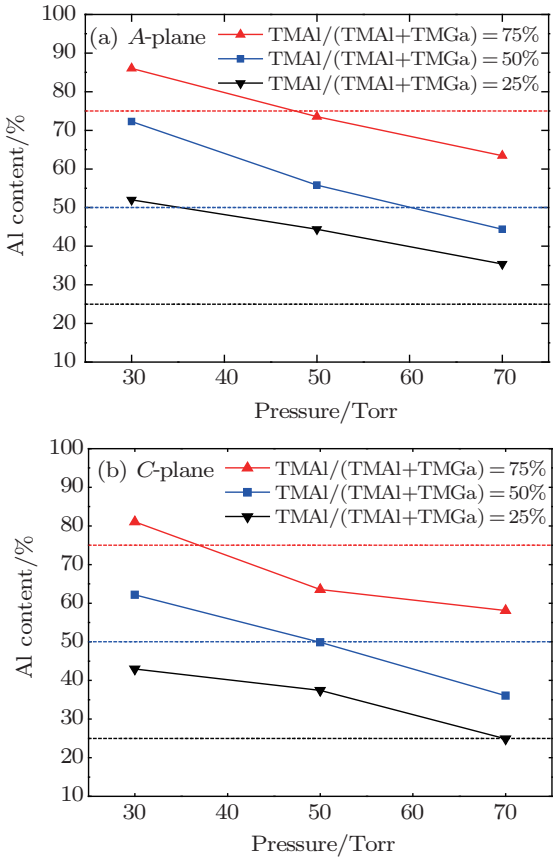


Fig. 3. (color online) Plots of Al content versus chamber pressure in (a) *A*- and (b) *C*-plane AlGaN films, where the dashed lines correspond to the mass-transport limit.

3.3. Al content versus V/III ratio

When the V/III ratio increases, the NH_3 partial pressure increases and the H_2 partial pressure decreases since the total flow rate is kept unchanged. The increased NH_3 partial pressure will enhance the parasitic reaction between TMAl and NH_3, thus reducing the Al incorporation efficiency. Meanwhile, the reduced H_2 partial pressure reduces the Ga desorption rate,[16,17] which also is harmful for Al incorporation in AlGaN. Both of the above two factors result in the reduction of Al content in AlGaN as the V/III ratio increases as shown in Fig. 4.

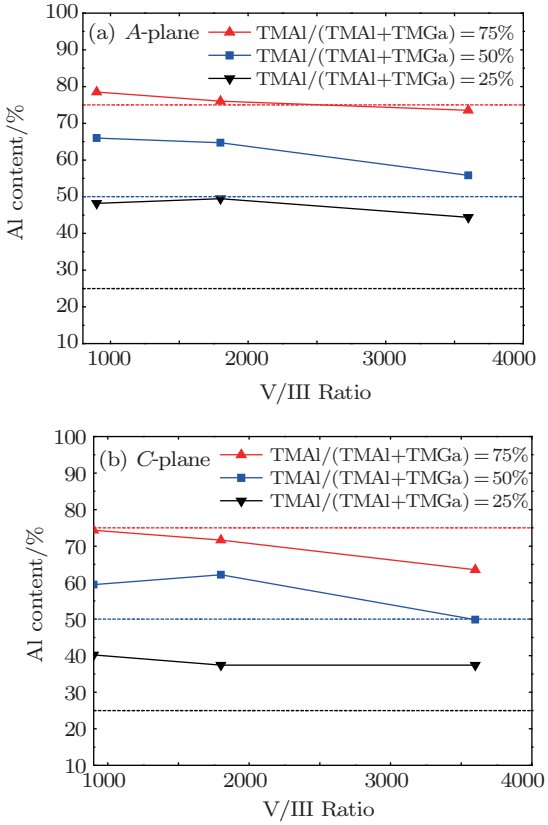


Fig. 4. (color online) Plots of Al content versus V/III ratio in (a) *A*- and (b) *C*-plane AlGaN films, where the dashed lines correspond to the mass-transport limit.

3.4. Al content versus the growth temperature

In previous studies, it was found that the parasitic reactions between TMAl and NH_3 increase at an elevated growth temperature,[6] which is not beneficial to increasing the Al content in AlGaN film. On the contrary, the desorption of Ga atoms becomes more rapid when the temperature is increased, which is beneficial to increasing that. These two factors make the relation between the Al content and the growth temperature unclear. In Fig. 5, we show the plots of Al content in *A*- and *C*-plane AlGaN films versus growth temperature. As for the *A*-plane sample, the Al content is lower than the transport limit and changes little when the temperature is below 1040 °C. When the temperature is further increased, the Al content increases rapidly and becomes higher than the transport limit. A similar trend is also found in the *C*-plane samples. It seems that when the temperature is low, the effect of Ga desorption is less important than the parasitic reactions. However, when the temperature is high, the Ga desorption becomes much more important than the parasitic reaction. Moreover, we can see that at a lower temperature the Al content is almost the same in the *A*-plane sample as that in the *C*-plane sample, while at a higher temperature, the Al content in the *A*-plane sample becomes higher than that in the *C*-plane sample. The reason may be that the Ga desorption rate of the *A*-plane AlGaN is higher than that of the *C*-plane AlGaN at higher temperature, leading to a high efficiency of Al incorporation into *A*-plane AlGaN.

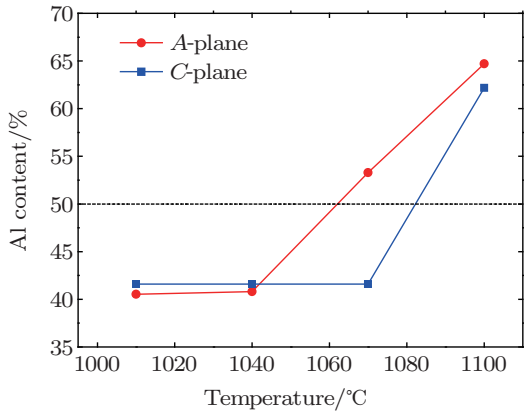


Fig. 5. (color online) Variations of Al content with growth temperature in *A*- and *C*-plane AlGaN films. The dashed line represents the mass-transport limit.

4. Conclusions

In this paper, we carry out the experimental study of the Al content values in nonpolar *A*-plane and polar *C*-plane AlGaN films grown by MOVPE. The deviation of Al content from the transport limit is mainly due to the parasitic reaction and the Ga atom desorption. Low chamber pressure, low V/III ratio, and high growth temperature are beneficial to the Al incorporation. Moreover, it is found that the Al content in the *A*-plane AlGaN sample is larger than that in the *C*-plane sample when the growth temperature is above 1070 °C, which might be due to the difference in atomic bond configuration between on *C*-plane and on *A*-plane. Our results indicate that the *A*-plane AlGaN films are more suitable for fabricating deep-UV optoelectronic devices, in which a high Al content AlGaN is necessary.

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