The Effects of Atomic Hydrogen on the III-Nitride Growth Dynamics in RF-Molecular Beam Epitaxy

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1 Introduction

Presently, the group III-nitrides are the most promising material systems for photonic devices in the green-blue-violet region. To date, most studies in metal organic chemical vapor deposition (MOCVD) have either neglected the effects of hydrogen on the growth, or have only considered its effect in compensating *p*-type dopants such as Mg.[1,2] On the other, recent studies in molecular beam epitaxy (MBE) have shown that the use of radical hydrogen and atomic hydrogen (H) are effective in the thermal cleaning[3] and increasing the growth rate of GaN[4] and AlN.[5] Myers *et al* reported that the effect of atomic H on increasing the growth rate only depends on the presence of atomic H. However, the detailed mechanism of increased GaN growth rate is not clear. We have shown that introducing atomic H during GaN growth not only improves the crystal quality of the films[6], but also increases the In composition during InGaN growth by RF-MBE.[7] In this paper, we investigated the effects of atomic H on the III-nitride growth dynamics by RF-MBE.

2 Experiment

The growth of GaN was carried out in a conventional MBE system with a Ga effusion cell and RF plasma source (SVT. Association 2.75") as the nitrogen source. The atomic H was generated by using a simple cracking cell with a tungsten filament heated by direct current. In this study, the filament temperature was fixed at 1500°C and hydrogen flow rate was changed from 0.15 to 1.50 SCCM. The cracking efficiency into atomic H was approximately 1.0 % in our operating conditions. To investigate the basic GaN growth dynamics in the conventional MBE mode, GaN layers were grown at different substrate temperatures, nitrogen flow rates and RF powers. And to investigate the effect of atomic H on the GaN growth dynamics, GaN layers were grown at different substrate temperatures and hydrogen flow rates, while the nitrogen flow rate and RF power were kept constant. The growth dynamics was studied by *in situ* reflection high-energy electron diffraction (RHEED).

3 Results and Discussion

Several works have reported on the characterization of GaN growth surface stoichiometory by monitoring the surface RHEED reconstructions or patterns.[8] It is reported that high quality GaN is obtained under slightly III-rich condition.[4] In our system, the RHEED pattern during GaN growth changes from a spotty pattern to a sharp then to a dim streak with changing III/V ratio from a nitrogen-rich, to III/V=1 (or slightly III-rich), then to III-rich, respectively. Therefore, the group V conditions (nitrogen flow rate and RF power) were fixed so that we can estimate the relative growth rate change with atomic H by measuring the Ga flux at which the RHEED pattern changes from spotty to sharp streak pattern.

Firstly, we investigated the conventional MBE mode of GaN from the RHEED pattern transition. Figure 1 (a) shows the substrate temperature dependence of Ga flux at the transition. The nitrogen flow rates and RF powers were 2.0, 3.0, 4.0 SCCM and 330, 340, 350W, respectively. The transition curve indicates that when the substrate temperature is increased, the so-defined Ga flux increases exponentially. And the curves are displaced upward with increasing nitrogen flow rate. These results are in good agreement with the previous report.[8] The form of the transition curve can be interpreted as a sum of the constant and exponential terms,

$$\Phi_{Ga} = \alpha\Phi_{N*} + A\exp\left(-\frac{Ea}{kTs}\right) \tag{1}$$

where Φ_{Ga}, Φ_{N*}, α, are the Ga flux, reactive nitrogen flux, and the participating factor of the nitrogen flux in the growth, respectively. The Ga desorption from the surface at a temperature Ts is given by the second term.

Secondly, we investigated the effect of atomic H on the GaN growth dynamics in the same manner as above. Fig. 1 (b) shows the substrate temperature dependence of Ga flux at the RHEED pattern transition at a nitrogen flow rate of 3.0 SCCM, and RF power of 340 W. The hydrogen flow rates were 0.15,0.45, 0.90, and 1.50 SCCM, respectively. The transition curves are displaced upward with increasing hydrogen flow rate for a fixed nitrogen flow rate. This result indicates that the nitrogen species participating in the growth increase with hydrogen passing through the cracking cell. The transition curve at a hydrogen flow rate of 1.5 SCCM and nitrogen flow rate of 3.0 SCCM in Fig. 1 (b) shows a higher GaN growth rate than for without hydrogen at a N_2 flow rate of 4.0 SCCM in Fig. 1(a). In addition, the results are different from the results of Myers *et al*, who showed that the effect of atomic H on the growth rate increase depends only on the presence of atomic H.

Finally, Fig. 1 (c) shows the hydrogen flow rate dependence of Ga flux at each substrate temperature. The hydrogen flow rate 0.00 SCCM in Fig. 1(c) corresponded to conventional MBE mode (without atomic H). The Ga flux (growth rate) at each substrate temperature linearly increased with hydrogen flow rate from 0.00 to 0.45 SCCM, and saturated at about 0.50 SCCM and above. From the result, there are two different observed regions showing the effect of atomic H on growth. One is a linearly dependent region, and the other is a saturated region. To our knowledge, this is the first work that clarifies the hydrogen flow rate dependence (the effect of atomic H) of the GaN growth rate. It is possible that Myers *et al* only observed the saturated region in their work. It is suggested that atomic H during GaN growth produces additional nitrogen species that participate in the growth ($\Phi_N(H)$), and, this effect can be increased to Eq.(1) as follows:

$$\Phi_{Ga} = \mathrm{a}\Phi_{N*} + A\exp\left(-\frac{Ea}{kTs}\right) + \Phi_N(H) \qquad (2)$$

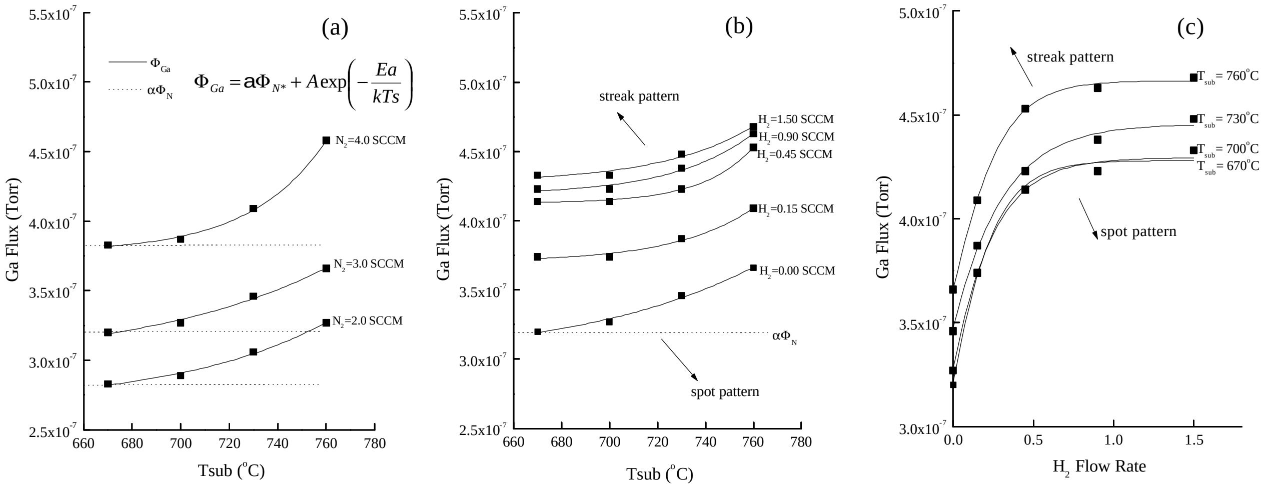

Figure 1 Substrate temperature dependence of Ga flux at the RHEED pattern transition point in (a) conventional MBE mode, (b) atomic H introduced MBE mode, and (c) hydrogen flow rate dependence of Ga flux, respectively.

4 Conclusions

We investigated the effects of atomic H on the GaN growth dynamics by RF-MBE. Firstly, the transition curves of the RHEED pattern show that the Ga flux increases exponentially with increasing substrate temperature, and that the curve is lifted upward with increasing nitrogen flow rate. This result is consistent with the previous report. Secondly, we investigated the effect of atomic H on the GaN growth. In this case, the curve is shifted upward with increasing hydrogen flow rate without changing the condition of nitrogen. This result suggests that nitrogen species participating to the growth increase when hydrogen flow rate increases. Finally, we investigated hydrogen flow rate dependence of Ga flux. We observed two regions, which are a linear and a saturation region.

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