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Atomic Layer Epitaxy of AlAs Using Trimethylamine-Alane and Tris-Dimethylamino-Arsenic

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Atomic layer epitaxy (ALE) of AlAs using trimethylamine-alane (TMAAl) and tris-dimethylamino-arsenic (TDMAAs) was studied. The one- and two-monolayer self-limiting growths of AlAs were observed depending on TMAAl pressure and substrate temperature. The self-limiting mechanism of the AlAs ALE was discussed with the quadrupole mass spectrometric (QMS) studies.

1. Introduction

Atomic layer epitaxy (ALE) of AlAs has been extensively studied in recent years because of its importance for fabrication of ultrathin quantum-effect devices, and both one- and two-monolayer self-limiting growths have been reported by using various growth methods and Al source materials[1-5]. However, the details of the self-limiting mechanism have not been understood sufficiently. Especially in the ALE of AlAs, how to reduce the carbon incorporation is a problem because of the stable Al-C bond. In MOCVD or MOMBE growth of AlAs, trimethylamine-alane (TMAAl) is known to have an excellent property to reduce the carbon incorporation in the grown films.

In this paper, we report the one- and two-monolayer self-limiting growths of AlAs using TMAAl and tris-dimethylamino-arsenic (TDMAAs). TDMAAs is preferable from the viewpoint of the safety due to the very low vapor pressure in comparison to arsine, and it also has the possibility to be free from the carbon incorporation because of the indirect bonding of As to carbon atoms[6]. The self-limiting mechanism of the AlAs ALE is discussed with the quadrupole mass spectrometric (QMS) studies.

2. ALE of AlAs

TMAAl and TDMAAs were alternately supplied to a (001) GaAs surface in an ultra-high vacuum chamber without thermal cracking. The cleaning of the GaAs surface was performed with a hydrogen-plasma beam excited by electron-cyclotron resonance. A clear (4x2) Ga-stabilized (001) GaAs surface was observed with the reflection high-energy electron diffraction (RHEED) after cleaning. The one cycle of the alternate gas supply in this experiment was set to be TMAAl (1s), purging (5s), TDMAAs (1s), and purging (18s). The number of the growth cycle was 500 which gives 141.5- or 283.0-nm thick films when the growth rate is one-monolayer or two-monolayer-per-cycle, respectively. After the ALE growth of AlAs, 30.0-nm thick GaAs cap layer was grown at 460°C by ALE.

Figure 1 shows the growth rate per cycle measured at 350°C against the TMAAl pressure. The TDMAAs pressure during the supply was fixed to 1.0×10^{-5} Torr. The one- and two-monolayer self-limiting growths were observed for the TMAAl pressure range from 2.0×10^{-6} to 4.0×10^{-6} Torr, and above 6.0×10^{-6} Torr, respectively.

Figure 2 shows the substrate temperature dependence of the growth rate per cycle measured for the two kinds of the TMAAl pressures. The TDMAAs pressure during the supply was fixed to 1.0x10^{-5} Torr. At the fixed TMAAl pressure of 2.0x10^{-6} Torr, the one-monolayer self-limiting growth was observed in the temperature range from 310 to 400°C. At the fixed TMAAl pressure of 1.0x10^{-5} Torr, the two-monolayer self-limiting growth was obserbed in the temperature range from 350 to 450°C.

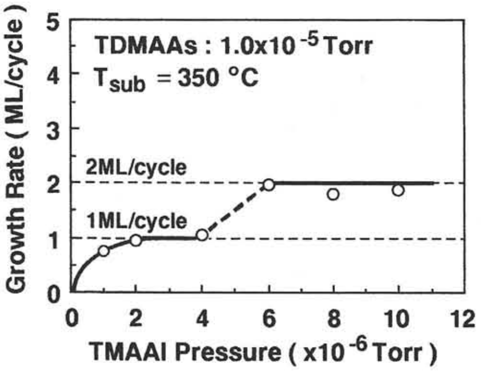

Fig. 1. TMAAl pressure dependence of the growth rate per cycle. The one- and two-monolayer self-limiting growths were observed for the TMAAl pressure range from 2.0x10^{-6} to 4.0x10^{-6} Torr, and above 6.0x10^{-6} Torr, respectively.

3. Self-limiting mechanism

For the purpose of studying the self-limiting mechanism of AlAs ALE, temperature programmed desorption (TPD) was measured. After 30 cycles of the alternate gas supply at 330°C, which satisfies the one-monolayer self-limiting condition, the AlAs surface was terminated with the TMAAl supply. Then the substrate temperature was increased with the rate of 5°C/min. Clear desorption of 28 amu (NCH_2) was obserbed above 400°C as shown in Fig. 3. The TPD response for 15 amu (CH_3), 29 amu (NCH_3), and 44 amu ($N(CH_3)_2$) were very weak. The same TPD responses were also observed after the alternate supplies at the lower substrate temperature of 271°C.

By changing the purging time, t, after the TDMAAs supply as shown in Fig. 4, the peak intensity of the 28 amu response shown in Fig. 3 was measured. The purging time after the TMAAl supply was fixed to 5s. The TMAAl and TDMAAs pressures during the respective supplies were 2.0x10^{-6} Torr and 1.0x10^{-5} Torr, which satisfy the one-monolayer ALE condition at 330°C as shown in Fig. 2. Figure 5 shows the 28 amu response of TPD vs purging time after the TDMAAs supply. The desorption time

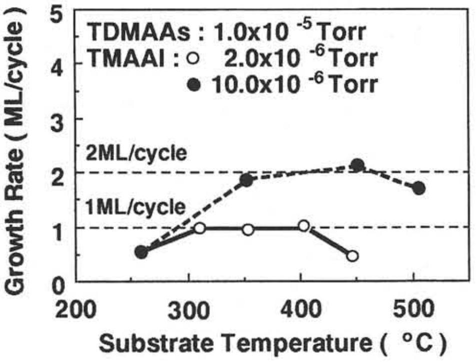

Fig. 2. Substrate temperature dependence of the growth rate per cycle. At the fixed TMAAl pressure of 2.0x10^{-6} Torr, the one-monolayer self-limiting growth was observed in the temperature range from 310 to 400°C. At the fixed TMAAl pressure of 1.0x10^{-5} Torr, the two-monolayer self-limiting growth was observed in the temperature range from 350 to 450°C.

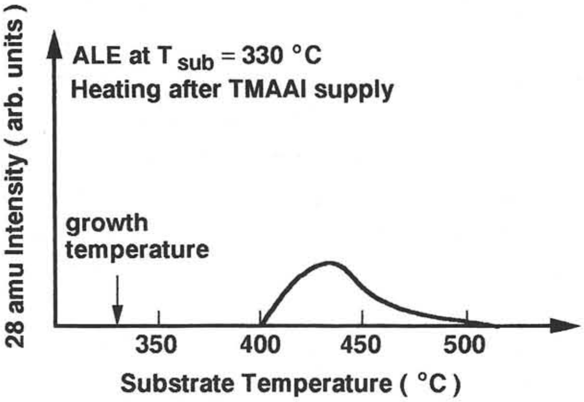

Fig. 3. Temperature programmed desorption of 28 amu (NCH_2) for substrate heating. After 30 cycles of the alternate gas supply at 330°C, which satisfies the one-monolayer self-limiting condition, the AlAs surface was terminated with the TMAAl supply. Clear desorption of 28 amu was observed above 400°C.

constant of 37s was estimated with the exponential fit of Exp(-t/τ) shown by the solid line.

The study on decomposition mechanism of TDMAAs with RHEED together with a rate-equation analysis indicates that As-stabilized surface after the TDMAAs supply is covered with the monomethyl-amine (NCH_3, MMA) below 500°C[7]. These results indicate that the MMA partially desorbing during the purging time after the TDMAAs supply is transferred to the Al surface during the TMAAl supply. This gives the one-monolayer self-limiting mechanism where the excess adsorption of TMAAl is prevented by the MMA on the Al surface, which is the same mechanism as that of the GaAs ALE using TEG and TDMAAs[8].

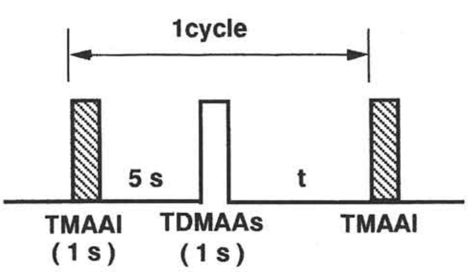

Fig. 4. Schematic to measure desorption of 28 amu from the AlAs surface. The purging time after TMAAl was fixed to 5s.

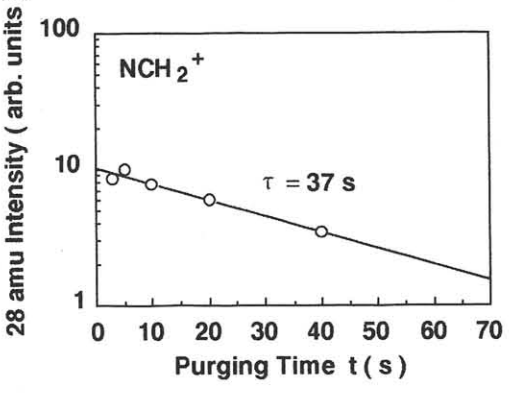

Fig. 5. The 28 amu response of the TPD vs purging time after the TDMAAs supply. The desorption time constant of MMA on the As surface of 37s was estimated with the exponential fit to Exp(-t/τ) shown by the solid line.

One the other hand, when TPD was measured after 30 cycles of the alternate gas supplies which satisfy the two-monolayer self-limiting condition, The response of 28 amu was drastically reduced. This result shows that Al atoms deposit on the As surface to form an atomic layer were followed by the further deposition of Al atoms to form the second atomic layer in the two-monolayer self-limiting regime[4].

4. Conclusions

We have studied the ALE of AlAs using TMAAl and TDMAAs. The one- and two-monolayer self-limiting growths of AlAs were achieved in the temperature range from 310 to 400°C, and 350 to 450°C, respectively. The self-limiting mechanism of the AlAs ALE was discussed with the QMS studies. It was shown that the MMA covering the Al surface plays an important role in the one-monolayer self-limiting mechanism, and the two-monolayer self-limitation results from the two-monolayer adsorption of Al atoms on the As surface.

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