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主 論 文 の 要 旨

論文題目　**Study of high-quality InGaN growth by metalorganic vapor phase epitaxy**

(有機金属気相成長法による高品質のInGaN成長に関する研究)

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論 文 内 容 の 要 旨

InGaN alloy with advantages, such as direct band gap, good thermal stability, and the whole visible spectra coverage, is usually used as the active layer of optoelectronic devices. Metalorganic vapor phase epitaxy (MOVPE) as a main growth theory are widely applied in factories because of good growth rate and high crystal quality. For violet-blue emission, high internal quantum efficiency (IQE) has been achieved. However, for green, yellow, and even longer wavelength emission, the IQEs become much lower. Besides polarization and the quantum-confined Stark effect, the increased growth-related defects attribute to the reduction in IQE. The low growth temperature and high growth rate which are favorable for high InN mole fraction cause many types of defects, such as stacking faults and trench defects. These defects strongly increase non-radiative recombination. In this thesis, the InGaN layer morphology and the effect of gas phase temperature is systematically explored.

First, InGaN morphology is systematically studied in Chapter 3. All InGaN layers with 4-6 nm thickness were grown on c-plane GaN substrates in a horizontal MOVPE reactor. The InGaN morphology was studied by varying temperature or changing TEG (TMG) and TMI fluxes while keeping the other conditions constant. InGaN layers were also grown on GaN substrates with different miscut angle using the same growth condition. InGaN layer of these three series were also grown on GaN/sapphire template at the same time to keep the growth condition in gas phase consistent. Besides these samples, many additional samples with a greater variation of growth condition were used to compare the influence of GaN substrates versus GaN/sapphire template. The InGaN morphology change from step to 2D island and then to 3D dot with decreasing growth temperature or increasing growth rate. According to thermodynamic calculation, the vapor supersaturation of Ga and In increase with decreasing growth temperature or increasing growth rate. Moreover, the supersaturation of Ga dominates over that of In by several orders of magnitude, InGaN growth depends mainly on the Ga supersaturation. However, In is also needed since GaN growth at these temperatures does not lead to the observed morphologies. Decreasing growth temperature or increasing growth rate increases the supersaturation of Ga and In. Higher Ga supersaturation will increase the amount of metal atoms on the surface and when a critical amount is reached, then N atom has possibility to nucleate on the terrace. Therefore, with increasing the supersaturation, the morphology changes from steps to 2D islands. Moreover, after formation the island density increases and the size decreases with decreasing growth temperature and increasing growth rate (i.e. increasing supersaturation), corresponding to shorter diffusions length as initial nucleation. 3D dots form after the formation of 2D islands. A rougher surface with 2D islands will also cause the non-uniform strain distribution on the surface. The larger local strain on top of the islands (due to a thicker layer) will lead to an earlier 3D dot growth. Since InGaN dots form slightly earlier on areas with more strain and grow rapidly, the size distribution of these InGaN dots is not uniform. InGaN morphology is also depended on the miscut angle of substrate. According to BCF theory, the maximum of surface supersaturation is calculated to express the average of surface supersaturation on the terrace. The surface supersaturation decrease with increasing the miscut angle. Therefore, InGaN morphology change from 2D island to step and then to step bunching.

InGaN/GaN multiple quantum wells (MQWs) growth is investigated in Chapter 4. InGaN/GaN MQWs with different InN mole fraction were grown on GaN or sapphire. The well and barrier is 2.5nm and 5nm, respectively. A stepped surface is also desirable for abrupt interfaces in MQWs, since a rough interface would further accumulate strain and easily lead to lateral decomposition.

MQWs have a stepped morphology if an uncovered InGaN layer shows steps. However, if the InGaN layer has 2D islands, pits and trench defect easily occur in MQWs even with identical GaN barrier growth conditions. These trench defects are attributed to 2D island of InGaN layer and form at the border when strain relaxes. Five-period $In_{0.28}Ga_{0.72}N$/GaN MQWs were grown on GaN substrate and on GaN/sapphire template at the same growth condition. MQWs on GaN substrate have the stepped morphology without any pits and defects due to the step morphology of InGaN layer. However, MQWs on GaN/sapphire template have pits and trench defects which impact the IQE. Besides the high TD density, another important reason is the large supersaturation due to smaller miscut angle of the GaN/sapphire templates. For violet-blue emission, MQWs on GaN/sapphire template have high quantum efficiency because InGaN layer is grown by step-flow growth mode due to lower supersaturation. For long wavelength emission, high supersaturation is preferred to increase InN mole fraction. GaN substrate have better InGaN morphology not only due to lower TD density but also because a larger miscut angle decreases the surface supersaturation of individual terraces. Therefore, GaN substrates are expected to promote high-InN-mole-fraction InGaN growth.

The effect of gas phase temperature on InGaN grown by metalorganic vapor phase epitaxy is illustrated in Chapter 5. Four wafer trays with gaps of 150 μm, 500 μm, 1000 μm, and 1500 μm were used to control the difference between the wafer surface temperature and gas phase temperature. We define the gap as the distance between the top surface of the pocket and the bottom of the wafer. Five-period InGaN/GaN MQWs emitting wavelengths of 450 nm (series I), 500 nm (series II) and 550 nm (series III) were grown on 2-inch GaN/sapphire templates with each wafer tray. The simulation result of thermal distribution in this reactor by Virtual Reactor software (STR Group Inc.) shows that to maintain a constant surface temperature to grow MQWs with the same wavelength, we need to increase the heater temperature when using a larger-gap. At the same time, the gas phase temperature on the wafer tray before reaching the wafer surface increases from 647 °C to 713 °C, which is expected to increase the decomposition rate of NH_3. For series I, the peak intensity and FWHM of all samples are similar because of the high growth temperature (i.e. wafer surface temperature) which lead to high NH_3 decomposition rate on the surface. In this case, increasing the gap which improves NH_3 decomposition in the gas phase has no effect on the PL peak intensity and FWHM. For series II and III, the peak intensity decreases and the FWHM increases rapidly because at the lower growth temperatures the effective V/III ratio on the wafer surface is limited by the NH_3 decomposition. However, the increased gas phase temperature by a larger-gap wafer tray increased the peak intensity and reduce FWHM. The improvement of the PL intensity and FWHM at larger gap can be understood from the topography of the samples. For series I, all samples have a step morphology. However, for series II and III, the morphology of the MQWs includes many slits or In-rich clusters at narrow gap. Owing to the same barrier growth conditions as those for series I, such morphological deterioration is caused by the low growth temperature and high growth rate of the well layer. These growth conditions decrease the effective V/III ratio on the surface, which increase the roughness of the well layer surface. This rough well layer induces some defects such as slits and clusters during MQW growth, which are non-radiative centers. However, with a larger gap, the density of clusters was strongly decreased and the trenches became less and shallower.

According to the above discussion, the key contributions in this research could be summarized as follows.

Firstly, InGaN morphology is analyzed by supersaturation. Besides growth parameters the effect of miscut angle on InGaN growth is illustrated. And how to control InGaN morphology is suggested. Moreover, the effect of InGaN layer on MQW is illustrated.

Second, the effect of gas phase temperature on InGaN growth is studied for the first time. The PL improvement and surface flattening is observed and the possible reasons are suggested.