行政院國家科學委員會專題研究計畫　成果報告

氮化鋁鎵六角錐與自聚性量子點之顯微光學量測

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一、中英文摘要

本計劃主要針對金屬有機化學氣相磊晶系統所成長的氮化鋁鎵薄膜做一系列的微螢光光譜和微拉曼光譜之研究。在顯微鏡下，我們可以在樣品表面觀察到幾種不同形狀的六角丘狀結構，它們的大小分佈約是 2~16 μm。因為應力對螢光光譜的影響很小，而且 Hillock 內外的鋁組成濃度，從 EDX 所量測出來的結果與微螢光光譜所推算出來的一致，所以將使用微螢光光譜去決定 Hillock 的鋁組成濃度。

從微螢光光譜的量測中，發現 Hillock 結構內部會出現額外的發光譜峰。根據微螢光光譜可以推導出 Hillock 內外的鋁組成濃度分別約是 4%和 11%。接著使用微拉曼光譜去針對 Hillock 結構進行實際量測，實驗結果顯示，Hillock 結構內部 E_2 模態位置約在 570 cm^{-1}，其外部位置約在 573 cm^{-1}。比較微螢光光譜和微拉曼光譜之計算和量測結果，發現 E_2 模態位置在 Hillock 結構內外分別有約 1.5 cm^{-1} 和約 3 cm^{-1} 的偏差，而此實驗結果顯示，Hillock 結構受到了壓縮的應力影響。

我們亦使用微拉曼光譜去分析 Hillock 結構之形成機制。其結果顯示，Hillock 結構內部之 E_2 模態位置約在 570 cm^{-1} 而不會隨著聚焦深度變深而改變；聚焦深度越深，隨之出現的是 sapphire E_g 位於 577 cm^{-1} 的訊號。由微拉曼光譜的縱深分析結果，其證明了 Hillock 結構形成是從 AlN 緩衝層開始長成。

關鍵詞：氮化鋁鎵、拉曼光譜、螢光光譜、六角錐結構、

Abstract

In this report, we analyzed $Al_xGa_{1-x}N$ epilayer which was grown by MOCVD system, with the aid of the micro-photoluminescence (μ-PL) and micro-Raman (μ-Raman) systems. Under the microscope, we observed several types of hexagonal hillocks on the epilayer with sizes from 2 to 16 μm.

Because the PL results are relatively insensitive to the strain and the Al fraction from Energy Dispersion X-ray Spectrometer (EDX) measurements agrees with that deduced from μ-PL whether inside or outside the hillock, we used μ-PL spectra to determine the Al fraction of hillock.

From the μ-PL spectra, we found that an additional emission peak inside the hillock structure, the calculated Al fraction is about 4% and 11% inside and outside the hillock. However, the experimental results of μ-Raman spectra show that E_2 mode frequency is ~570 cm^{-1} and ~573 cm^{-1} inside and outside the hillock, respectively. These are blue shifted by ~1.5 cm^{-1} and ~3 cm^{-1} so that hillocks bear compressive stress.

We also used μ-Raman scattering to investigate how deep hillocks are formed. The results showed that the E_2 mode frequency remains at ~570 cm^{-1} inside hillock, it dose not shift while the focus depth increases. However, the sapphire E_g mode frequency at 577 cm^{-1} grows obviously with the increasing focus depth. According to the depth analysis, it is evident that the formation of hillock is from the AlN buffer layer.

Keywords: AlGaN, Hillock, Raman, Photoluminescence

二、緣由及目的

The pyramid-like structures on Ⅲ-Ⅴ compound semiconductors surfaces have been widely discussed. These structures are formed spontaneously[1-6] or artificially[7-12], and they all have particular optical and electrical properties. For example, some researches have studied the emission characteristics on artificial pyramid-like structures[7-11] with the aid of high spatial resolution, and results showed that the emissions from apex and sidewall are quite different. Besides the artificial pyramid-like structures, other studies focused on the spontaneously-formed pyramid-like structures, including the formation mechanism[13-16], the optical properties[17-19], and the electrical properties[20].

The purpose of this report is to study the stress influence on hillocks and how deep hillocks are formed. We examined spontaneously formed hexagonal hillocks on $Al_xGa_{1-x}N$ thin film. By using a microscope to demarcate the hillock position, we can study the same hillock on this sample and analyze the stress influence on various sizes of hillock, combining the μ-PL and μ-Raman measurements.

三、結果與討論

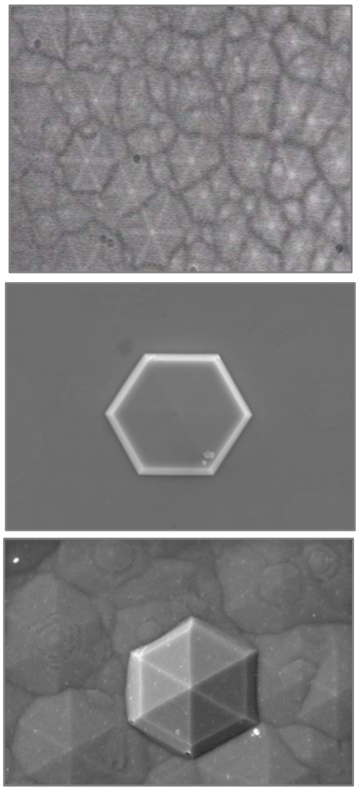

Figure 1: SEM picture of three different type hillocks

As shown in SEM pictures of Fig.1, there are three types of hillocks on AlGaN sample. The first one called pyramid-like hillock, is distributed extensively on the sample and the angle between the sidewall and the plane region is about 3°; the second one called mesa-like hillock, has a quasi-flat top and the angle between the sidewall and the plane region is about 55°; the third one called tent-like hillock, has two-step sidewall, and the angle between plane region and bottom sidewall is about 22° while the upper sidewall is about 13°. The size distribution is about 3~5μm and 6 ~11μm for mesa and tent-like hillocks, respectively. In this paper, we will look into the stress influence on the optical and phonon properties of different hillock type and size.

As shown in Fig. 2, the μ-PL spectra of the tent-like hillock show the position dependence along the horizontal line. The results show the I_{NBE} appearance at 342nm (3.625eV) outside the hillock. Besides, we also observe another emission peak at 353nm (3.512eV) and its intensity is stronger than the I_{NBE} from the outside region. In the previous study, we concluded that dissimilar peaks inside and outside the hillock are due to the Al variation and the theoretical calculation of Al fraction through the results of μ-PL is very close to the EDX measurements.

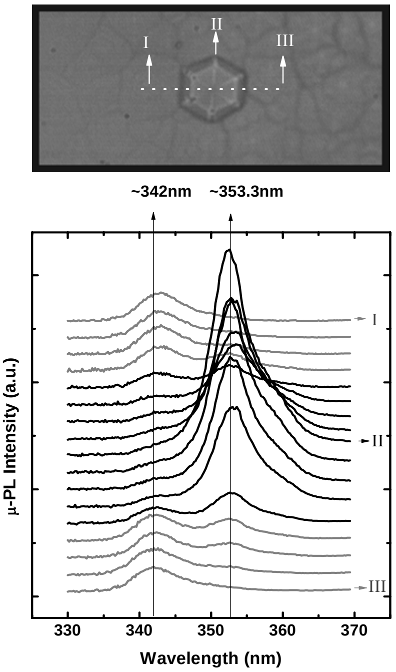

Figure 2: Optical image and spatially resolved μ-PL spectra of tent-type hillock.

Since the optical properties of hillocks are dominated by the Al fraction fluctuation, the above spectra show the corresponding Al fraction to be about 4% and 11% inside and outside the hillock, respectively. Then, we adopted the Al fraction and applied the formula list below to calculate the strain free E_2 mode frequency of this 10μm hillock which is ~568.5 cm^{-1} inside the hillock and ~570 cm^{-1} outside the hillock as shown by the dashed lines in Fig. 3.

$$E_2^{AlxGa_{1-x}N}(High) = E_2^{GaN}(1-x) + E_2^{AlN}x - b_{E_2(High)}x(1-x)$$

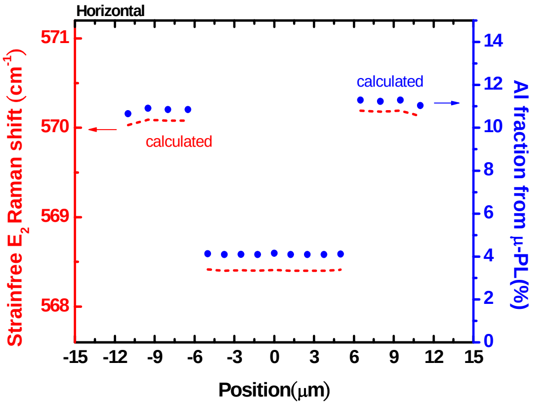

Figure 3: The calculated strain-free Raman E_2 mode of tent-like hillocks.

In order to expose the influence of stress on tent-like hillocks, we compare the strain free E_2 mode frequency obtained from Al fraction with the experimental Raman results. As shown in Fig. 4, the Raman E_2 mode shift deviation is ~1.5 cm^{-1} and ~3 cm^{-1} inside and outside the tent-like hillock respectively. They are blue-shifted implying that the outside of hillock bears much larger compressive stress than the hillock itself, with the apex bearing little larger stress than its surroundings. The compressive stress is estimated to be 0.3 GPa and 0.7 GPa inside and outside the tent-like hillock, respectively.

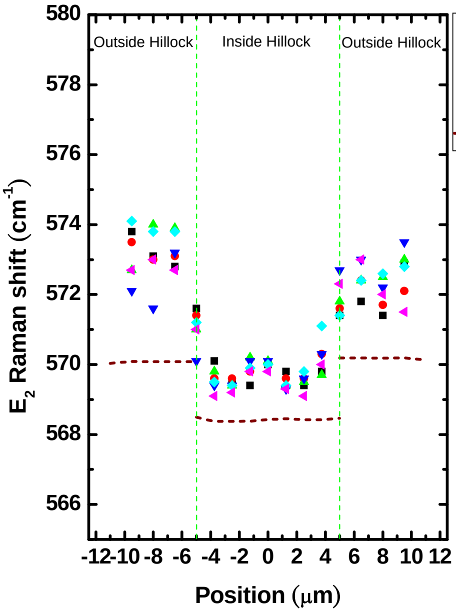

Figure 4: Raman results between inside and outside hillocks

On the other hand, the full widths at half maximum (FWHM) of μ-PL and μ-Raman scattering of this tent-like hillock are shown in Fig. 5. It is obvious that the FWHM inside the hillock is narrower than that outside the hillock and it decreases gradually to the hillock center by 25meV and 3.5 cm^{-1}. In Fig. 6, the μ-PL and μ-Raman scattering show much stronger intensity inside the hillock than that outside the hillock by a factor of 5-6. From our results, hillock appears to be a good light emission structure.

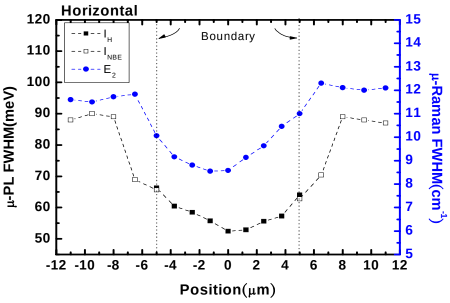

Figure 5: FWHM of μ-PL and μ-Raman

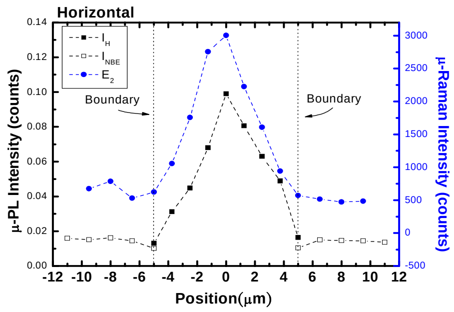

Figure 6: Intensity of μ-PL and μ-Raman

On the tent-like hillock center, its Raman spectra at different depths are shown in Fig. 7. The E_2 mode frequency remains at ~570 cm^{-1}, and it does not shift while the focus depth increases. This result also match the model (fig.8) which assume the hillock formation from AlN buffer layer.

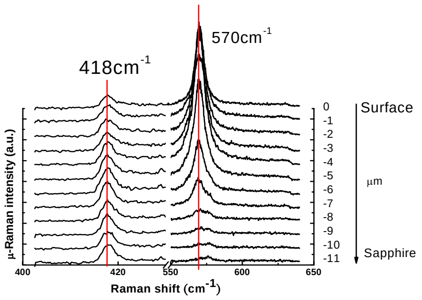

Figure 7: Raman depth profile of hillock

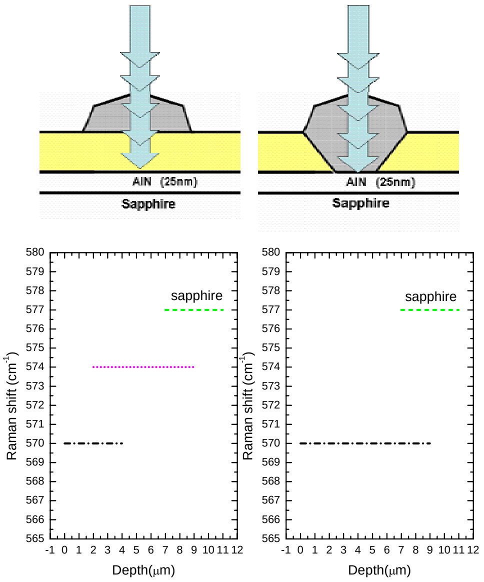

Figure 8: Assumption model of depth profile.

According to the depth analysis of tent-like hillock, it is evident that the formation of hillock is from the AlN buffer layer, in consistent with the assumption. It is likely that hillock position is determined by the AlN buffer grown on sapphire. When $Al_XGa_{1-X}N$ is deposited on the AlN buffer, hillocks are then formed spontaneously by balancing and reducing the internal strain of the sample film.

四、結論

The μ-PL results showed that an additional peak I_H appears in tent-like hillock besides the I_{NBE} emission. This phenomenon is attributed to the Al concentration and stress variations. Moreover, the PL results are relatively insensitive to the strain, and the Al fraction deduced from μ-PL spectra. For this Al fraction, we calculated the strain free E_2 mode frequency to be ~568.5 cm^{-1} and ~ 570.2 cm^{-1} inside and outside the hillock. The experimental results showed that the E_2 mode frequency are deviated by ~1.5 cm^{-1} and ~3 cm^{-1} from the strain free E_2 mode frequency.

We also used μ-Raman scattering to investigate how deep hillocks are formed. The results showed that the E_2 mode frequency remains at ~570 cm^{-1} inside hillock, and it dose not shift while the focus depth increases. However, the sapphire E_2 mode frequency at 577 cm^{-1} grows obviously with the increasing focus depth. According to the depth analysis, it is evident that the formation of hillock is from the AlN buffer layer.

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