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Strain Effect on Photoluminescence from InGaN/GaN and InGaN/AlGaN MQWs*

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Abstract: Photoluminescence, HR-XRD, and Raman scattering spectra of InGaN/GaN MQWs and InGaN/AlGaN on sapphire and membranes with no substrate fabricated by laser lift-off are studied. In contrast to the emission peak from the membrane samples of InGaN/GaN MQWs, which blue-shifts after annealing at 700℃, a red-shift of the PL peak position in the InGaN/AlGaN MQW membrane sample is observed, showing different strain effects in these MQWs. In Raman scattering spectra, the InGaN/GaN MQW film without sapphire substrate has a lower E_2 mode frequency (567.5cm^{-1}) than that of the films with substrate (569.1cm^{-1}), which indicates that the compressive stress in the films is released partially when the sapphire substrate is taken off.

Key words: photoluminescence; InGaN; AlGaN; MQWs; strain
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1 Introduction

Ⅲ-Ⅴ nitride semiconductors have been widely applied in the fabrication of optoelectronic devices in a wide range, from green to ultraviolet. The active parts of these devices are usually InGaN/GaN and AlGaN/GaN multiple quantum wells (MQWs), which are epitaxially grown on sapphire, SiC, or Si substrate with MOCVD growth technology. Sapphire is the most commonly used substrate in the growth of GaN-based materials. However, the mismatches of thermal expansion coefficients and lattice constants between GaN and Al_2O_3, and the different lattice parameters of layers in quantum wells, result in strain in GaN, InGaN/GaN, and InGaN/AlGaN MQW films. It is well known that the quantum-confined Stark effect (QCSE) due to the strong piezoelectric field in GaN-based materials greatly influences the luminescence process in GaN-based quantum wells, and many researchers have studied the strain effects on structural and optical properties of GaN, InGaN/GaN MQWs, and InGaN/AlGaN MQWs[1~5]. Since it is difficult to identify clearly the mechanism of strain effect on the properties of the films of multiple layers, many efforts have been made to observe differences in properties by changing the stress in samples. Chen *et al*. studied InGaN/GaN Raman and PL spectra with different electric fields[5]. Johnson *et al*. reported the TAXRD and XTEM analysis of strain in InGaN/GaN MQWs on different GaN template layers[6]. Raman shifts of the E_2 and A_1 (LO) peaks of InGaN/GaN MQWs with different In contents were observed in Ref.[7].

In this work, we prepared InGaN/GaN MQWs and InGaN/AlGaN MQWs of similar indium content and well and barrier widths, in which the degree of strain should be different. Samples were grown on sapphire by MOCVD, and then they were transferred onto Si substrate by lift-off technology with a KrF pulsed excimer laser. With a comparative study of the photoluminescence (PL), Raman scattering spectra, and high resolution X-ray diffraction (HRXRD) of MQWs on sapphire and free-standing membranes, we discuss

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the strain effect in InGaN/GaN and InGaN/AlGaN MQWs structures.

2 Experiment

Five-period InGaN/GaN MQW and InGaN/AlGaN MQW structures over a 2μm Si-doped GaN layer were grown by LP-MOCVD on 300μm sapphire that was polished on one side. A 0.1μm Mg-doped GaN layer was grown on top of the MQWs. The structures were confirmed to be of good quality by high resolution transmission electron microscopy (HRTEM). After a Si (111) wafer was bonded to the top of the InGaN/GaN MQWs and InGaN/AlGaN MQWs, the film of epilayers was separated from the sapphire substrates by KrF excimer pulsed laser irradiation. The wavelength and pulse width of KrF laser were 248nm and 25ns, respectively, and the incident energy density of the laser beam was about 600mJ/cm^2. Following the laser processing, the film was transferred onto another Si wafer after removing the Si wafer on the top of the film, creating a membrane sample with p-GaN on top. The as-grown wafer and membranes were divided into pieces to go through thermal treatment processes at 700℃ in nitrogen ambient for 20min. Photoluminescence (excitation wavelength of 325nm from a HeCd laser, maximum power of 30mW) and Raman spectra in the backscattering configuration along the *c* axis (excited by 514.5nm from an Ar$^+$ laser, 4mW, Renishow 2MR-1000) were measured at room temperature. High resolution synchrotron XRD was carried out for characterizing the changes of structures in InGaN/GaN and InGaN/AlGaN and thus investigating the strain effects in the samples.

3 Results and discussion

Photoluminescence (PL) spectra from the InGaN/GaN and InGaN/AlGaN as-grown samples and membrane samples are shown in Figs. 1(a) and (b). The photoluminescence peak (412nm) of the InGaN/GaN MQW membrane sample was lifted off redshifts slightly from 410nm, which is the PL peak from the as-grown sample, but that of the membrane blueshifts obviously to 404nm after annealing at 700℃. Strain in the MQWs is reduced after removing the sapphire substrate or after thermal treatment, and the strong piezoelectric field in the InGaN/GaN MQWs decreases. Therefore, the photoluminescence peak should shift to a shorter wavelength[8]. Considering the 2 ~ 5nm variation in the luminescence peak that is caused by fluctuation of the indium content in the wafer, the small difference in luminescence peak position (2nm) in Fig. 1(a) may not just be a result of strain relaxation in the membrane samples since the obvious change of stress is not observed in this case. Our other InGaN/GaN MQWs with different In compositions (less than 20%) show blueshifts of the PL peak position when the sapphire is taken off, and the greater the indium content in InGaN, the larger the PL peak blueshifts are.

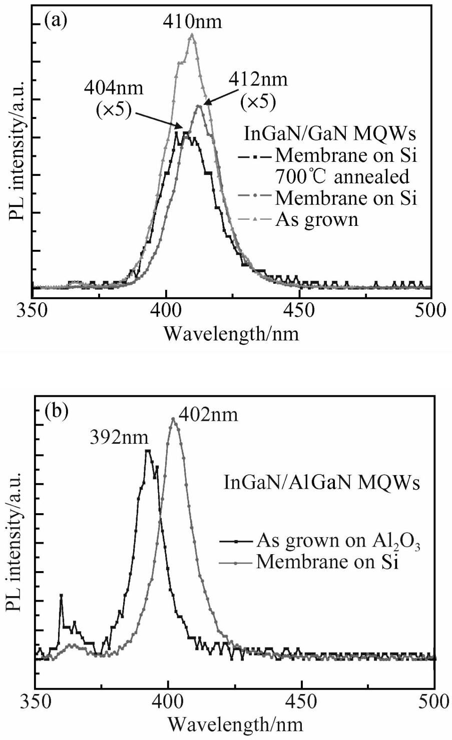

Fig. 1 (a) PL spectra of InGaN/GaN MQWs; (b) PL spectra of InGaN/AlGaN MQWs

Figure 1(b) shows quite different luminescence properties of InGaN/AlGaN MQWs. The peak position (402nm) of the membrane samples redshifts 10nm from that (392nm) of the as-grown samples, indicating different changes of the strain effect in InGaN/AlGaN MQWs and InGaN/GaN MQWs. The layers in both the InGaN/AlGaN and InGaN/GaN structures are under compressive stress after epitaxial growth on Al$_2$O$_3$ substrates. The stresses in the layers change differently when the substrate is taken off. The luminescence peak position shifts correspond to the variations of the piezoelectric field in the InGaN well layers. In contrast to the blueshift of the peak position in the photoluminescence spectra and less compressive strain in the InGaN/GaN MQW membrane, the obvious redshift in the InGaN/AlGaN MQWs implies that the piezoelectric field in the InGaN well layer becomes stronger after the laser lift-off process. This means that the changes of stresses in the InGaN well layers in InGaN/AlGaN MQWs and InGaN/GaN are different. With a small lattice parameter, AlGaN layers in InGaN/AlGaN MQWs compensate part of the compressive strain from the substrate during growth, but make InGaN well layers more compressive when the substrate is taken off. This is verified by the changes in the HRXRD profiles, which will be discussed later. Therefore, the effect of compressive strain on the InGaN layer can be observed more clearly in the membrane samples of InGaN/AlGaN MQWs than those of InGaN/GaN MQWs. In fact, the photoluminescence of GaN-based MQWs involves much more complicated processes than photo excitation and recombination, and interaction between the piezoelectric field, defect localization field, and carriers. Thus changes of photoluminescence result from the colligation of these effects when the substrate was taken off.

Raman scattering spectra were excited in the back-scattering geometry along [0001], with 514.5nm radiation of an Ar^+ laser. Figure 2 shows the Raman spectra of the wafer samples and membranes of InGaN/AlGaN MQWs. The E_2 mode and the A_1(LO) mode were detected in all of the samples. The spectra of the membrane samples are distinguished by the Si peak around $521cm^{-1}$, and no peaks of sapphire at $418cm^{-1}$ (A_{1g}) and $750cm^{-1}$ (E_g) were observed. Similar behavior of the E_2 mode was observed in the Raman spectra of the InGaN/GaN and InGaN/AlGaN samples. As shown in the inset of Fig. 2, the E_2 mode frequency of GaN shifts lower because the membrane samples have a lower value of $567.5cm^{-1}$ than $569.1cm^{-1}$ in films on sapphire substrate. The A_1 (LO) mode frequency is $735.0cm^{-1}$ in both the membranes and the wafers with Al_2O_3 of InGaN/AlGaN samples, but a $0.8cm^{-1}$ greater redshift occurs in the membrane samples than the wafer of InGaN/GaN. We believe that the decreases of the E_2 mode frequency and the A_1 mode frequency result from the residual stress change when the sapphire substrate is taken off. The E_2 mode phonon frequency is more sensitive than the A_1 mode to biaxial strain in the *c*-plane because the atomic displacement of the E_2 mode is in the *c*-plane, but that of the A_1 phonon mode is parallel to the *c*-axis[9]. According to Ref. [10], hydrostatic stress is the main part in our InGaN/GaN samples, and the stress decrease is about 0.5 ~ 0.6GPa during the laser lift-off process. Furthermore, the different behaviors of the A_1(LO) frequency imply that InGaN/AlGaN may have different kinds of strain from InGaN/GaN, which is in accordance with the PL results in Fig. 1.

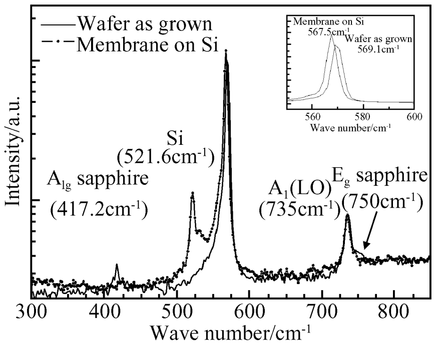

Fig. 2 Raman spectra of InGaN/AlGaN MQWs

Figure 3 shows HRXRD profiles of a 2θ scan in the (0002) of the AlGaN/InGaN MQW structure. Thin membrane samples may bend under different strains in the top and bottom parts, so measurements of the same membrane were performed by X-ray incidence from p-GaN and n-

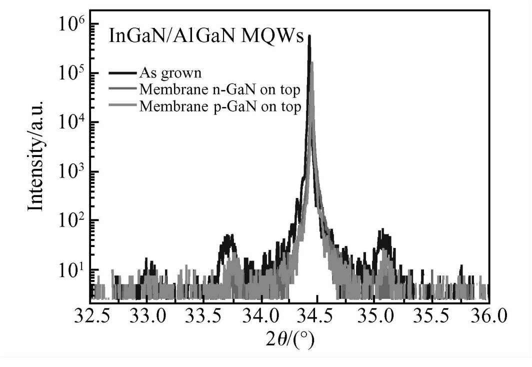

Fig. 3 HRXRD profiles of (0002) reflection of the InGaN/AlGaN MQWs

GaN, respectively. The refraction angles of GaN are about 0.02° greater in the membrane than the values of the wafer with sapphire. This means that the lattice along the *c* direction of GaN is compressed after the laser lift-off process, and the lattice in the plane stretched at the same time. This lattice relaxation of GaN layers can be observed when measuring both p-GaN top and n-GaN top. The very little difference (less than 0.001°) in the GaN diffraction angle means that the membrane sample of AlGaN/InGaN MQWs is nearly flat. On the other hand, XRD profiles of InGaN/GaN obviously shifted when measured by ways of p-GaN top and n-GaN top, which means that the membrane was bent under different strains in p-GaN and n-GaN after laser lift-off. As discussed above, AlGaN layers of small lattice parameters may compensate part of the compressive strain during growth on sapphire substrate and introduce more compressive stress in InGaN well layers. As a result, AlGaN barrier layers make InGaN/AlGaN MQWs more rigid than InGaN/GaN MQWs.

4 Conclusions

In summary, photoluminescence from InGaN/GaN and InGaN/AlGaN MQWs on sapphire substrate and membranes fabricated by laser lift-off have been observed. In contrast to the emission peak from the membrane samples of InGaN/GaN MQWs, which blueshifted after annealing at 700℃, a red-shift of the PL peak position in the InGaN/AlGaN MQW membrane sample was observed, showing different strain effects in these MQWs. AlGaN layers in InGaN/AlGaN MQWs compensate part of the compressive strain from the substrate during growth but make the layers more compressive when the substrate is taken off. The E_2 and A_1 mode frequencies in Raman scattering spectra decreased, and HRXRD analysis confirms that residual stress in the films of InGaN/AlGaN MQWs and InGaN/GaN MQWs are changed differently after laser lift-off.

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InGaN/GaN 和 InGaN/AlGaN 多量子阱中应变对光致发光特性的影响*

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摘要： 对蓝宝石衬底上的 InGaN/GaN 和 InGaN/AlGaN 多量子阱结构和经激光剥离去除衬底的 InGaN/GaN 和 InGaN/AlGaN 多量子阱结构薄膜样品，进行了光致发光谱、高分辨 XRD 和喇曼光谱测量. PL 测量结果表明，相对于带有蓝宝石衬底的样品，InGaN/GaN 多量子阱薄膜样品的 PL 谱峰值波长发生较小的蓝移，而 InGaN/AlGaN 多量子阱薄膜样品的 PL 谱峰值波长发生明显的红移；喇曼光谱的结果表明，激光剥离前后 E_2 模的峰值从 569.1 减少到 567.5cm^{-1}. 这说明激光剥离去除衬底使得外延层整体的压应力得到部分释放，但 InGaN/GaN 与 InGaN/AlGaN 多量子阱结构中阱层 InGaN 的应力发生了不同的变化. XRD 的结果证实了这一结论.

关键词： 光致发光谱；InGaN；AlGaN；多量子阱；应变

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