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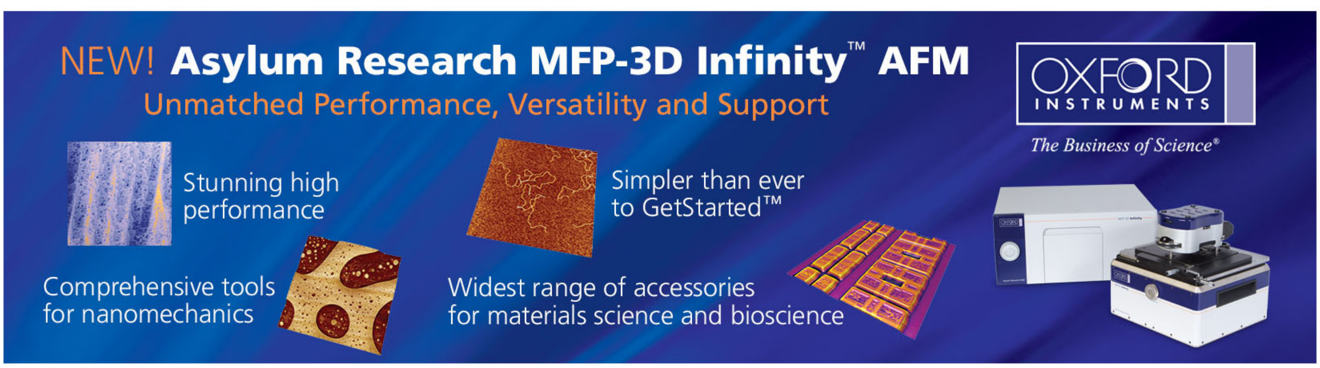

Enhanced internal quantum efficiency in graphene/InGaN multiple-quantum-well hybrid structures

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The enhanced internal quantum efficiency of InGaN/GaN multiple-quantum-wells (MQWs) structure is demonstrated by paving the graphene layers on the MQWs surface. Compared to the conventional MQWs, the internal quantum efficiency of the graphene/InGaN MQWs hybrid structure exhibits a remarkable 2-fold increase. The high charge carrier density in graphene layer is accounted for the enhanced internal quantum efficiency. Moreover, the negligible photoluminescence emission peak shift with increasing the excitation power as well as the decrease of radiative recombination lifetime are attributed to the reduced quantum-confined Stark effect, which correlates to the screening of the polarization field in the *c*-plane nitride-based quantum well structure. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4745211]

GaN-based light-emitting diodes (LEDs) attract more attention in the application for solid-state lighting because of their various advantages such as the long lifetime, small size, high efficiency, and low energy consumption. Their wide spectral range can cover from near ultraviolet to near infrared. As the emission wavelength of InGaN multiple-quantum-wells (MQWs) varies from blue to green, the significant reduction of internal quantum efficiency (IQE) is due to the high dislocation density resulting from the lattice mismatch between the sapphire substrate and active region and the more Indium fluctuations in the structure, leading to large non-radiative recombination rate.[1] On the other hand, the MQWs structure grown along the c-direction suffers from the quantum confinement Stark effect (QCSE) as a result of the existence of the strong polarization filed, leading to the reduction of the electron-hole wave-function overlap and radiative recombination rate.[2] In order to maximize the external quantum efficiency of nitride LEDs, the optimization of IQE in InGaN/GaN MQWs is one of the important issues. Recently, several approaches have been proposed to suppress the charge separation to improve the radiative recombination rate. For the improvement of IQE in InGaN/GaN MQW LEDs, the polarization-induced effects may be a key factor. So far, there are some investigations devoted to study the Si-doping effects on InGaN/GaN MQWs structure, and the partial screening polarization fields is proved to bring a smaller stoke-shift, reduced charge separation, and the increase of radiative recombination.[3–5] Besides, growth along the non- and semi-polar oriented directions have been explored to avoid such polarization effects. But the existence of high threading dislocation (TD) density acts as the nonradiative recombination centers and charge scattering centers, which are responsible for poor internal quantum efficiency and low carrier mobilities.[6–8] The various modification methods on quantum well structures, such as the polarization-matched AlGaInN or InGaN barriers, have demonstrated that polarization effects can be effectively eliminated and the radiative recombination rate is improved.[9–12] However, it is difficult to grow AlInGaN layers with high crystalline quality due to the differences between optimal incorporation conditions for growth of AlN and InN. Also, the crystalline quality of active layers could be worse as more InGaN barriers and wells are deposited repeatedly due to the accumulation of large compressive strain.[13]

In this work, we presented an effective approach to enhance the IQE in the *c*-plane InGaN/GaN MQWs covered with graphene layers on the surface, and the influences of graphene layers on optical properties in nitride quantum well structure were discussed in detail. By using the power-dependent photoluminescence measurements, the internal quantum efficiency of the InGaN/GaN MQWs covered with graphene layers was demonstrated to have a 2-fold enhancement compared to the conventional MQWs, due to the stronger charge screening effect in the interface between InGaN/GaN MQWs and graphene layers. The increase of radiative recombination rate obtained in the time-resolved PL measurement and no shift in the emission energy were observed, indicating the absence of polarization effects in the InGaN/GaN MQWs.

The samples in this study were grown on (0001) *c*-plane sapphire substrates by a low-pressure metal organic chemical vapor deposition (MOCVD) system. The ten periods InGaN/GaN MQWs consisting of 12 nm-thick GaN barrier layers and 2 nm-thick InGaN well layers in each period were grown on a 2-μm-thick n-type GaN template on *c*-plane sapphire substrates. The first five periods of InGaN/GaN quantum wells near the n-type GaN template were doped with Si at concentration of about $5 \times 10^{17}\,\mathrm{cm}^{-3}$. On the other hand, the last five InGaN/GaN MQWs near the surface remained un-doped. After growth, the graphene layers grown on the copper substrates by chemical vapor deposition (CVD) were

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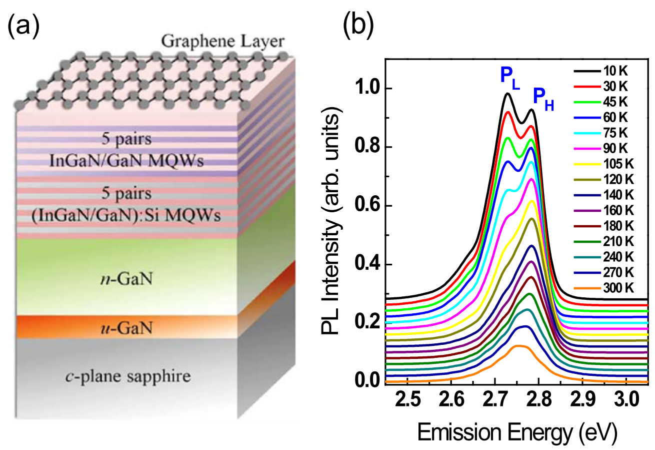

FIG. 1. Schematic diagram of InGaN/GaN MQWs with graphene layers. (b) The temperature-dependent photoluminescence spectra of the as-grown InGaN/GaN MQWs structure.

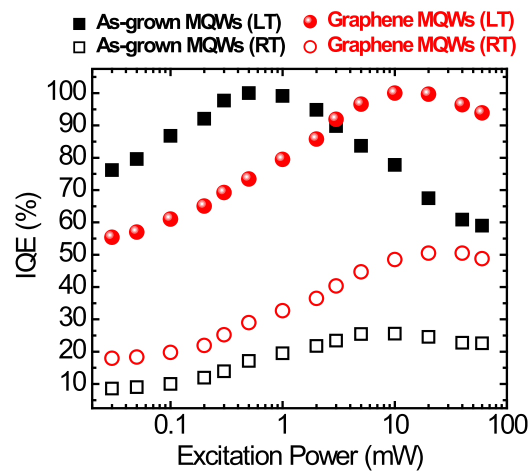

FIG. 2. Power-dependent relative internal quantum efficiency curves of as-grown and graphene InGaN/GaN MQWs structures.

transferred to the above InGaN/GaN MQWs structure, and this structure was labeled as graphene MQWs (GMQWs). A schematic drawing of InGaN/GaN GMQWs structure is shown in Fig. 1(a). The excitation power-dependent and time-resolved PL measurements were carried out to analyze the internal quantum efficiency as well as optical properties for as-grown MQWs and GMQWs. The 266 nm pulse laser generated by a frequency tripled mode-locked Ti: sapphire laser (Mira 900) with the pulse width of 200 fs and repetition rate of 76 MHz was used for the pumping source.

Temperature-dependent PL spectra ranging from 10 to 300 K of InGaN/GaN as-grown MQWs structure is shown in Fig. 1(b). Two obvious emission peaks measured from the spectra of as-grown InGaN/GaN MQWs at 10 K lie at about 2.73 eV (P_L) and 2.79 eV (P_H) with the separation of about 60 meV, respectively. With increasing the temperature, the PL intensity of P_L emission peak revealed a quicker thermal quenching than that of P_H emission peak, and then P_L emission peak gradually degenerated to be a shoulder as the temperature raised to 105 K. According to the thermal quenching behavior of temperature-dependent PL, the P_L emission peak can be identified as the donor-bound excitons recombination originated from the partial Si doping in the first five periods of InGaN/GaN MQWs.[4] P_H emission peak is due to InGaN quantum well energy level transition.

To evaluate the internal quantum efficiency accurately, we performed the power- and temperature-dependent PL measurements on the as-grown MQWs and GMQWs structures. Figure 2 shows the measured efficiency as a function of excitation power at 10 and 300 K for as-grown MQWs and GMQWs, respectively. The efficiency is defined as the collected photon numbers divided by the excitation photon numbers; and all normalized to the maximum efficiency at 10 K. According to the experimental results, the internal quantum efficiency increases with the excitation power and then reach its maximum value. It is attributed to a fact that the radiative recombination rate gradually becomes dominant while excitation power increases. The maximum IQE value at 10 K was defined assuming that the nonradiative recombination could be neglected and droop behavior was about to start. In terms of the peak efficiency at 300 K, the internal quantum efficiency (~50%) of GMQWs was significantly enhanced in comparison to that of as-grown MQWs (25%), corresponding to a 2-fold enhancement of efficiency. It indicated a rare behavior that the photon emission efficiency within the InGaN/GaN MQWs active region had changed as the surface was covered with the graphene layers. However, the IQE enhancement generally required complicated methods to modify the epitaxial structure and/or to improve the crystal quality. Therefore, the characteristics of GMQWs shall be worth further studying.

The power-dependent PL spectra for as-grown MQWs and GMQWs at 10 K were performed as shown in Fig. 3. The PL intensities increased with the excitation power. The emission peak energy as a function of excitation power was plotted in Fig. 3(c). In the as-grown MQWs, the blue-shift of the PL emission peak energy occurred with increasing the excitation power, primarily due to the screening of QCSE and band-filling effect. The energy shift of P_L and P_H emission peaks were estimated to be as 15 meV and 23 meV, respectively. The smaller blue-shift value of P_L emission peak was attributed to the partial screening effect of QCSE due to the Si doping in the first five periods of InGaN/GaN MQWs. On the other hand, the P_L and P_H emission peaks in InGaN/GaN GMQWs showed an extremely small energy shift with the excitation power ranging from 0.02 to 20 mW. It was interesting to note that the negligible peak energy shift of the PL emission was very similar to the observation of reduced or absent QCSE in the non-polar GaN-based MQWs, due to the polarization-free behavior.[14] Since both of the P_L and P_H emission peaks in InGaN/GaN GMQWs showed negligible energy shift, the influence of the graphene layers was not only on the top InGaN MQWs but also on the Si-doped InGaN MQWs at least 70 nm away from the surface.

Furthermore, the time-resolved PL spectra for as-grown MQWs and GMQWs have been performed at 10 K to investigate the effects of graphene layers on the carrier recombination mechanism while the influence of the nonradiative recombination process could be excluded at low temperature, as shown in Fig. 4. Here, the decay traces recorded the P_H emission originated from the un-doped InGaN/GaN MQWs energy level. The experimental results can be fitted by the

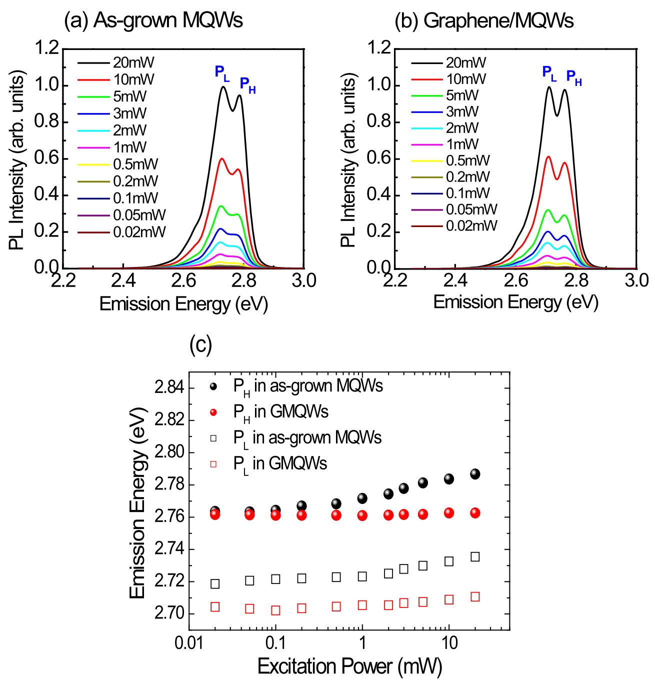

FIG. 3. Power-dependent photoluminescence spectra of (a) as-grown and (b) graphene InGaN/GaN MQWs at 10 K. (c) The emission peak positions versus the excitation power.

stretched exponential decay shape $I(t) = I_1(0)\exp(-t/\tau_1) + I_2(0)\exp[-(t/\tau_2)^\beta]$. The parameter $I(t)$ means the PL intensity at time t, β is the dimensionality of the localizing centers, and τ_1 and τ_2 are the initial lifetimes of carriers. Normally, the fast carrier lifetime represents the radiative recombination of QWs excitons and determines the PL intensity and carrier radiative emission efficiency. The fitted values of the short lifetime were also labeled in Fig. 4. It showed that the GMQWs had a shorter lifetime of 8.95 ns than that of 17.3 ns for the as-grown MQWs. Despite the short lifetime should exhibit stronger emission intensity in GMQWs samples, the comparable PL emission intensities in the as-grown MQWs and GMQWs samples shown in Fig. 3 could be due to the absorption of pumping power and the emission power passing through the top graphene layers.[15,16] It is worth noting that these related results could be dependent on the layer number of graphene. The shorter carrier lifetime in GMQWs sample also indicated the increased radiative recombination rate, which was consistent with the

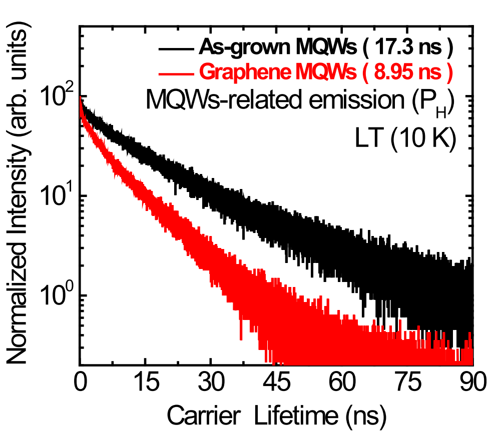

FIG. 4. Time-resolved photoluminescence spectra for as-grown and graphene InGaN/GaN MQWs at 10 K.

experimental result of internal quantum efficiency as shown in Fig. 2. As compared with the as-grown InGaN/GaN MQWs, the GMQWs showed the enhanced quantum efficiency, reduced carrier lifetime, and negligible energy shift with increasing the excitation power, where such results were similar to the screening effect of QCSE resembling the polarization-free behavior in the non-polar MQWs. The above results indicated that the graphene layers on the top of InGaN/GaN MQWs could induce the reduction of polarization field and enhanced electron-hole wave-function overlap, which in turn enhanced the IQE. As we knew that graphene electronic properties that arise from the carbon atom has four electrons and three of which are tied up in bonding with its neighbors. One interesting consequence of this unique band structure is that the electrons in graphene are sort of free. Originally, electrons and holes were spatially separated by the polarization field in the InGaN/GaN MQWs, but the free-carrier-induced field provided by the graphene layer was opposite to the polarization field. The two fields tended to cancel each other out for high sheet densities, thus re-establishing the conditions for electron-hole recombination emission.[17,18] Therefore, due to the strong screening effect on the polarization field, the GMQWs exhibited the enhanced recombination rate and higher internal quantum efficiency in comparison to the as-grown InGaN/GaN MQWs.

In summary, the optical properties of graphene/GaN-based MQWs hybrid structure have been investigated. The 2-fold enhanced internal quantum efficiency from graphene/GaN-based MQWs was attributed to the large free carriers in the interface between GaN-based MQWs and graphene layers, leading to the screening of polarization field. The polarization-free-like behavior, enhanced radiative recombination rate, and the significant efficiency improvement had been demonstrated. Since the graphene layer could be a good candidate for the transparent current spreading layer on the p-GaN, these results suggested that applying graphene layers on top of InGaN LEDs was promising to achieve high efficiency light emitting devices.

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