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Investigation of large Stark shifts in InGaN/GaN multiple quantum wells

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Photoluminescence (PL) spectra of InGaN/GaN multiple quantum wells excited by ultrafast laser pulses are investigated over broad ranges of excitation levels and temperatures. The PL peak energy undergoes blue, red, zero, and blue shifts with increasing the excitation fluence density. Such a peculiar behavior can be explained based on competing processes of screening of the built-in electric field by the photogenerated carriers, band-gap renormalization, and band-filling effect. We have also measured and analyzed the dependence of the PL energy and linewidth on the temperature. Due to the interplay between the band-gap renormalization and band-filling effect, the PL energy shifts to the highest value, whereas the PL linewidth reaches the minimum value at ≈60 K. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4775605]

I. INTRODUCTION

Wurtzite III-nitride-based quantum wells (QWs) have tremendous applications in light emitting devices[1] and laser diodes[2,3] in the spectral range from visible to ultraviolet. Conventional InGaN/GaN QWs were grown along the polar [0001] axis on a *c*-plane substrate. The charges induced by spontaneous polarization are accumulated at the interfaces of InGaN and GaN, resulting in large built-in electric fields across QWs. Since the lattice constants of well and barrier materials are mismatched, electric fields can be induced by strain through piezoelectric effect in QWs. Owing to the piezoelectric spontaneous polarization fields, the wave functions of electrons and holes in QWs are spatially separated due to the quantum-confined Stark effect (QCSE). Such spatial separation of the electrons and holes significantly affects the efficiency of light emission. Since the difference of spontaneous polarization between GaN and InN is very small, the built-in electric field induced by spontaneous polarization in InGaN-based QWs is much weaker than that induced by strain. For the InGaN-based QWs with the emission wavelengths in the range of from UV to blue, the In composition is relative low, and therefore, the lattice mismatch between well and barrier materials is small. As a result, the emission efficiency is not dramatically affected by QCSE. However, with increasing the In concentration in QWs, the emission efficiency can be dramatically reduced due to the increase in the piezoelectric polarization field.[4]

In order to improve the emission efficiency, researchers introduced three different approaches: (i) using InGaN as a barrier material[5] or substrate[6] resulting in the lower lattice mismatch, and therefore the reduced built-in electric field; (ii) employing the staggered,[7] graded,[8] or dip-shaped[9] QWs structures to increase the overlap between the wave functions of electrons and holes;[10] and (iii) growing the QWs along nonpolar $[11\bar{2}0]$ (a-plane),[11] $[1\bar{1}00]$ (m-plane),[12] and semipolar[13] directions to eliminate the built-in electric field. It is worth noting that in (i) and (ii) above, the strain-induced electric field is still present in QWs, whereas in the approach of (iii) above, the weak strain-induced electric field is present along the growth direction of QWs,[14] due to the second order strain tensor.

Due to the presence of the spontaneous and piezoelectric polarizations, the mechanism for carrier recombination in InGaN-based QWs is quite different from that of conventional III-V arsenide QWs. Indeed, in the past the carrier recombination took place at the band-tail state[15,16] and localized state.[17–19] The peak emission energy was shifted with the increase in either the excitation power or the temperature. Such shifts were attributed to band-filling effect,[15,16] screening of internal electric field by photogenerated carriers,[16] and inhomogeneity and carrier localization[18,19] in nitride QWs. However, InGaN-based QWs with high In concentrations have not been carefully investigated.

In this article, we investigate the time-integrated photoluminescence (PL) of InGaN-based multiple QWs with the light emission in the green region in a broad range of excitation levels and temperatures. Following our analysis, we have provided explanations to the shifts of the PL peak energy with increasing the excitation level and temperature.

II. SAMPLE DESCRIPTION AND EXPERIMENT CONDITIONS

The InGaN/GaN MQWs used in our experiments were grown by vertical-type metalorganic chemical vapor deposition (MOCVD) on 2.8-μm-thick n-doped GaN grown on c-plane double-side polished sapphire substrate. The active region of MQWs consists of four periods 4.5 nm thick $In_{0.22}Ga_{0.78}N$ QWs with 15 nm thick u-GaN barrier layer. In order to mitigate the thermal effect, and, comparing with the carrier lifetime in QWs, ensure that the sufficient carriers were generated in QWs within very short time, the frequency-doubled Ti:sapphire regenerative amplifier was employed as the excitation source in our measurement. The final ultrafast laser pulses with central wavelength, pulse width, and repetition rate of 393 nm, 180 fs, and 250 kHz,

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respectively, were focused on the MQWs sample through a $f \approx 20\,\mathrm{cm}$ convex lens. In our experiment, the sample was kept in cryostat and the temperature was changed from 4.5 K to 300 K. To eliminate the interference fringes over the PL spectra, for which originated from the Fabry-Pérot cavity formed by u-GaN/air interface and n-GaN/sapphire interface, the MQWs sample was placed in the parallel orientation, i.e., the sample surfaces are parallel to the direction of signal collecting setup.

III. RESULTS AND DISCUSSIONS

At different fluence densities, we have measured the PL spectra at 4.5 K, see Fig. 1(a). As the fluence density is increased, the PL peak energy undergoes a shift. Indeed, from 0.70 μJ/cm^2 to 1.11 mJ/cm^2, the peak energy is blue-shifted from 2.241 eV to 2.289 eV, i.e., an amount of the shift as large as 48 meV. Between 1.11 mJ/cm^2 and 4.45 mJ/cm^2, the peak energy is red-shifted by 14 meV. In the range of 4.45 mJ/cm^2–17.8 mJ/cm^2, the shift is negligible. Above 17.8 mJ/cm^2, however, the peak energy undergoes blue-shift again. Such a complicated behavior of PL peak energy vs. fluence density can be explained by us as follows. The blue shift in the range of 0.70 μJ/cm^2–1.11 mJ/cm^2 is due to the screening of the built-in electric field by the photogenerated carriers. As the fluence density is increased from 1.11 mJ/cm^2 and 4.45 mJ/cm^2, the conduction band-edge is renormalized, and therefore, the effective band-gap is reduced. However, as the fluence density is further increased from 4.45 mJ/cm^2 to 17.8 mJ/cm^2, the negligible shift is probably caused by the cancellation between the band-gap renormalization and band-filling effect. Above 17.8 mJ/cm^2, the blue shift is caused by the over-taking band-filling effect. According to Ref. 20, at a sufficiently high excitation fluence density, the density of the photogenerated electrons in QWs is large enough to significantly modify the conduction band-edge. In comparison, due to the large effective mass of holes, the valence band is not significantly modified by the holes. Following Ref. 20, we have illustrated the conduction band edges of the QWs at three different excitation levels of 13.9 μJ/cm^2, 0.28 mJ/cm^2, and 22.3 mJ/cm^2, see inset of Fig. 1, corresponding to the carrier densities of $3.47 \times 10^{12}\,\mathrm{cm}^{-2}$, $6.94 \times 10^{13}\,\mathrm{cm}^{-2}$, and $5.55 \times 10^{15}\,\mathrm{cm}^{-2}$, respectively. Obviously, the conduction band-edges are significantly shifted upward. According to the inset of Fig. 1(b), each modified conduction band-edge is characterized by two local potential minima located at Λ_a and Λ_b. Such a modified band-edge has caused the effective band-gap to increase, i.e., band-filling effect.

On the other hand, at room temperature, the behavior of the PL transition energy vs. the fluence density is quite different, see Fig. 2. Indeed, as the fluence density is increased from 6.94 μJ/cm^2 to 278 μJ/cm^2, the PL peak energy is blue-shifted from 2.275 eV to 2.371 eV, i.e., an amount of 96 meV. Such an amount of the blue shift is comparable to that observed previously.[16] It primarily originates from the screening of the built-in electric field by the photogenerated carriers. As the fluence density is increased from 278 μJ/cm^2 to 1.11 mJ/cm^2, there is a plateau. As the fluence density is further increased from 1.11 mJ/cm^2 to 31.1 mJ/cm^2, the transition energy is red-shifted from 2.371 eV to 2.355 eV, i.e., by an amount of 17 meV. This red shift is probably caused by band-gap renormalization. A much larger shift at room temperature could be explained by the fact that the band-gap renormalization takes place at much higher fluence densities. Recently, we measured the carrier lifetimes in InGaN/GaN QWs.[21] According to our analysis,[21] at sufficiently low temperatures, the carrier lifetimes can be extremely long, due to the carriers occupying the localized states. At room temperature, however, the carriers at the localized states are thermally ionized. Consequently, the carrier lifetimes at room temperature are much shorter. Obviously, the strength of band-gap renormalization depends on the quasi-steady-state density of the photogenerated carriers, which are determined

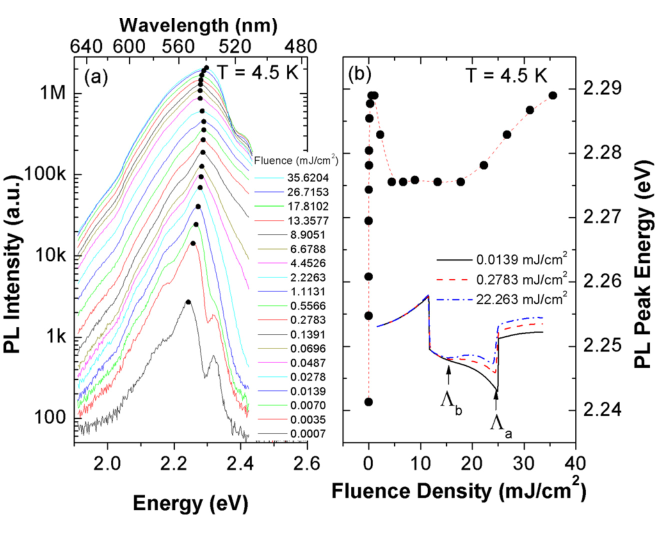

FIG. 1. (a) PL spectra of InGaN/GaN MQWs at different excitation fluences, and (b) PL peak energy as a function of excitation fluence at 4.5 K. Dots are experimental data, and dashed curve in (b) is for eye-guide purpose. The inset is simulated conduction band edges for QWs at three different excitation levels.

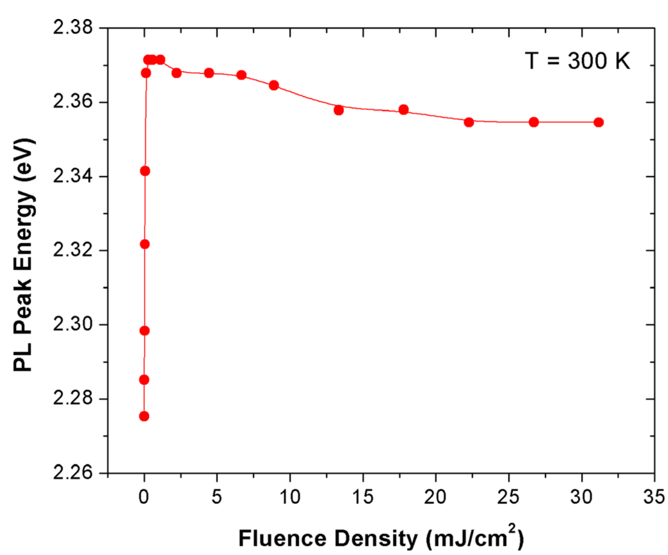

FIG. 2. PL peak energy vs. fluence density, measured at room temperature. Solid curve was obtained after smoothing the lines connecting adjacent data points to show major features of the dependence.

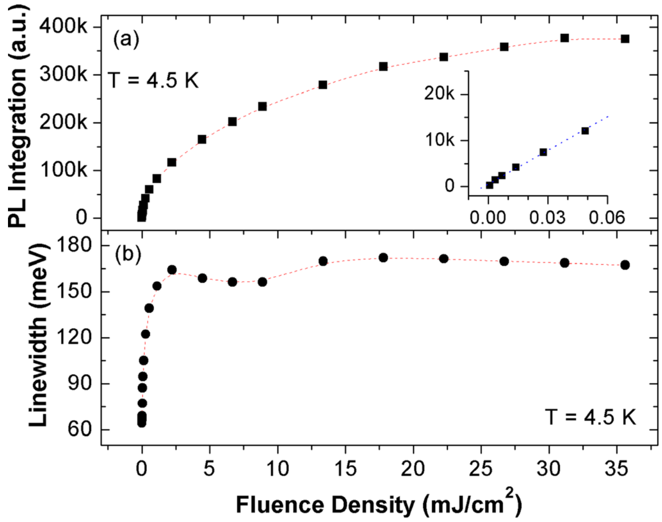

FIG. 3. (a) Time-integrated PL intensity and (b) linewidths of PL spectra as a function of excitation fluence density at the temperature of 4.5 K. Dots are experimental data, and dashed curves are for eye-guide purpose. The inset shows the PL integration under very low excitation fluence, and solid line is linear fitting for experimental data.

by the generation rate and recombination time constant. Therefore, at room temperature, much higher fluence densities are required to reach the dominant regime of the band-gap renormalization. Similarly, the band-filling effect also requires much higher fluence densities at room temperature. All these effects are quite different if the pump laser is CW or it has the pulse width close to the carrier recombination time.

According to the inset of Fig. 3(a), the PL intensity increases linearly with increasing the excitation fluence density, when the fluence density is below 48.7 μJ/cm^2. Such a behavior indicates that the overlap between the electrons and holes does not change significantly in such a range. On the other hand, the linewidth (i.e., FWHM) of the PL peak is increased from 64 meV at 0.70 μJ/cm^2 to 154 meV at 1.11 mJ/cm^2, see Fig. 3(b). Such an increase is probably due to the band-gap renormalization. Above 1.11 mJ/cm^2, the PL linewidth is more or less kept at a constant value, whereas the PL intensity versus fluence density starts to sharply deviate from the linear dependence, see Fig. 3. Such behaviors are probably the competition between the band-gap renormalization and band-filling effect. Finally, when the fluence density exceeds 22.26 mJ/cm^2, the slow increase the PL intensity with the fluence density is the indication of a strong and over-taking band-filling effect.

We have also measured temperature dependence of the PL spectra at three different excitation levels, i.e., 13.9 μJ/cm^2, 0.28 mJ/cm^2, and 22.3 mJ/cm^2. The decrease of the PL intensity with increasing the temperature can be clearly observed at all three excitation levels, see Figs. 4(a)–4(c). It is worth noting that at the fluence densities of 0.28 mJ/cm^2 and 22.26 mJ/cm^2, besides the primary PL peak located below 2.40 eV, a rather broad peak covering from 2.40 eV to 3.05 eV can be identified, see Fig. 4(d). In comparison, at the low fluence density of 13.9 μJ/cm^2, only a single PL peak is observed over the entire temperature range. Compared with the thermal quenching behaviors of the dominant PL peak, the PL intensity of this broad peak changes little from 10 K to 300 K. Such a broadband emission peak may originate from the recombination of the excitons inside the barrier layers.[15,17]

The activation energies can be extracted from an Arrhenius plot of the PL intensity. Consider a model based on two recombination channels.[22] The Arrhenius plot of the PL intensity can be fitted by using the following equation:

$$I_{\rm PL}(T) = \frac{I_0}{1 + C_1 \exp\left(-\frac{E_{\rm a}}{k_B T}\right) + C_2 \exp\left(-\frac{E_{\rm b}}{k_B T}\right)}, \quad (1)$$

where $I_{\rm PL}(T)$ is the temperature-dependent PL intensity, I_0 is the PL intensity at 4.5 K, k_B is Boltzmann constant, C_1 and C_2 are corresponding rate constant of two channels, and $E_{\rm a}$ and $E_{\rm b}$ are the activation energies for the two recombination

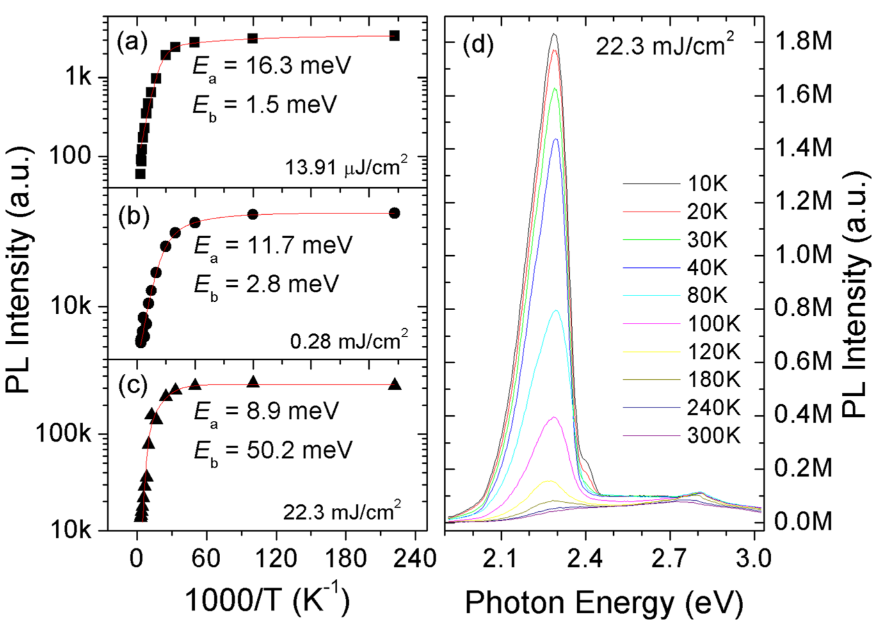

FIG. 4. Arrhenius plots of time-integrated PL intensity for excitation fluence of (a) 13.9 μJ/cm^2, (b) 0.28 mJ/cm^2, and (c) 22.3 mJ/cm^2. Dots are experimental data and solid curves are fitting results by using Eq. (1). And (d) temperature-dependent PL spectra at excitation fluence of 22.3 mJ/cm^2.

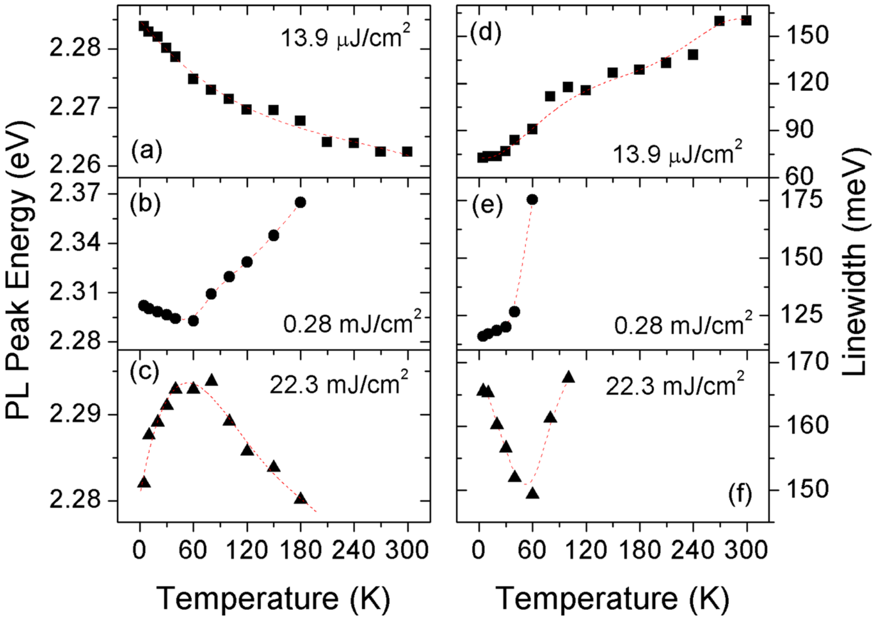

FIG. 5. PL peak energy (left column) and linwidth of PL spectra (right column) as a funtion of temperature for three excitation levels. The corresponding excitation fluence is indicated in figures. Dashed curves were obtained after smoothing the lines connecting adjacent data points to show major features of the dependence.

channels, respectively. By using Eq. (1), our data can be well fitted. As a result of the nonlinear least-square fit, we have obtained the activation energies at different excitation fluence densities, listed in Figs. 4(a)–4(c), respectively. One can see that by comparing Fig. 4(a) with Fig. 4(b) with increasing the fluence density from 13.9 μJ/cm^2 to 22.3 mJ/cm^2, the activation energy of the channel #1 is decreased from 16 meV to 8.9 meV. In contrast, the activation energy of the channel #2 is increased slightly from 1.5 meV to 2.8 meV with the increasing of excitation fluence from 13.9 μJ/cm^2 to 0.28 mJ/cm^2, and then dramatically increased to 50 meV at 22.3 mJ/cm^2.

Besides the thermal quenching of the PL intensity, we have deduced the PL peak energies and linewidths at the three excitation levels as a function of the temperature, see Fig. 5. Due to the presence of the broadband emission peaks mentioned earlier, the PL peak energies and linewidths cannot be accurately extracted from the measured PL spectra in relatively high temperatures for high excitation fluence densities. At the excitation fluence density of 13.9 μJ/cm^2, the photogenerated carriers are more or less localized in the vicinity labeled by Λ_a, see Fig. 1(b). As the temperature is increased from 4.5 K to 300 K, the PL peak energy is redshifted by 22 meV from, whereas the linewidth is increased from 73 meV to 160 meV. We believe these behaviors are caused by the reduction in the screening effect by the photogenerated carriers, as the carriers are thermally excited to a higher energy level. However, when the fluence density is increased to 0.28 mJ/cm^2, the band-gap renormalization becomes important. As the temperature increases, band-gap renormalization becomes weakened, and therefore, the PL peak is blue-shifted. The dramatic increase in the PL linewidth is due to the thermal excitation of the carriers. When the excitation level is set to 22.3 mJ/cm^2, band-gap renormalization is competing with band-filling effect. The blue- and red-shifts in Fig. 5(c) represent the weakening of the band-gap renormalization and band-filling effect, respectively. Due to the interplay of these two effects, the PL linewidth reaches a minimum value of 149 meV at ≈60 K, whereas the PL energy shifts to the highest value.

IV. CONCLUSION

In conclusion, we have studied PL spectra of InGaN/GaN multiple quantum wells excited by ultrafast laser pulses over broad ranges of temperatures and excitation levels. The PL peak energy vs. excitation fluence density exhibits unique behaviors. They can be explained based on the competition among the screening of the built-in electric field by the photogenerated carriers, band-gap renormalization, and band-filling effect. We have also measured and analyzed the dependence of the PL energy and linewidth on the temperature. Due to the interplay between the band-gap renormalization and band-filling effect, the PL energy shifts towards the highest value, whereas the PL linewidth reaches the minimum value at ≈60 K.

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