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ABSTRACT

Large negative thermal quenching (NTQ) of the yellow luminescence (YL) for a temperature increase from 5 to 300 K is observed in non-polar InGaN/GaN quantum well (QW) samples due to the thermal migration of carriers from the InGaN QW layers to the GaN barrier layers for the first time. Such an unusual phenomenon happens only when the carriers are optically excited inside the QW layers, providing solid evidence for the occurrence of thermal transfer of photoexcited carriers from the QW layers to the GaN barrier layers. A simple model considering the thermal transfer of carriers is proposed to interpret the observed NTQ phenomenon. The thermal activation energy of the carriers is determined by fitting the reciprocal temperature dependence of the YL intensity in the Arrhenius plot with the model.

I. INTRODUCTION

Thermal quenching is a common phenomenon for photoluminescence (PL) widely observed in many materials.[1–3] Thermal quenching of defect related PL can be explained by multicenter and one-center mechanisms.[4–6] In a multicenter mechanism, the carriers bound at donors or acceptors are thermally excited to the conduction or valence band and then recombine through a non-radiative channel. In a one-center mechanism, PL related carriers can recombine through the non-radiative channel, which is induced by the crossover between the excited and ground states of a PL related defect in the configuration-coordinate diagram.[6,7] For the near-band-edge emissions, the thermal quenching of PL of bound and free excitons can be caused by their detrapping and debinding.[8] In addition, the randomly distributed non-radiative centers in materials can reduce overall carrier density and contribute to the thermal quenching of all kinds of PL.[9]

On the other hand, negative thermal quenching (NTQ) has been observed for PL in several materials including ZnO,[10] $InP_{1-x}Bi_x$,[11] Mn^{4+}-activated fluoride phosphor,[12] and $FASnI_3$.[13] The NTQ of PL manifests as a substantial increase of PL intensity with increasing the temperature in a certain range. It is usually explained by thermal transmitting of electrons from other nearby energy states to the initial states of PL transitions.[4,11] In fact, NTQ of a specific PL was also observed in GaN, which is explained by quenching of other kinds of PL sharing minority carriers.[14] However, to the best of our knowledge, such a NTQ phenomenon has not yet been reported in InGaN/GaN quantum well (QW) samples.

Even in QW samples, thermal quenching of PL is commonly observed, which degrades the performance of light-emitting devices based on QW structures.[15–19] On one hand, thermal quenching of PL in QW structures can be caused by both thermal emission and non-radiative recombination of carriers.[20,21] These two mechanisms are indistinguishable in the thermal quenching of PL in QW. An effective method capable of identifying thermal emission is thus needed. On the other hand, the thermal emission of carriers from QW had been proposed for a long time and was claimed to be

experimentally observed in the literature studies.[15–17] However, the above-barrier-bandgap optical excitations were usually adopted in these experiments. Such circumstances make the cases more complicated since the carriers can be optically excited in both barrier layers and QWs. For example, the thermal increase of relative PL intensity between barrier and QW was proposed as an evidence for the thermal emission of carriers from QW.[15] However, the possible fast thermal decline of the QW PL intensity caused by the thermal activation of non-radiative centers within the QW could be an alternative explanation. Therefore, to our personal viewpoints, a thermally induced substantial increase of PL signals in barrier layers would be solid evidence if the carriers were only optically excited within QWs, which has not yet been demonstrated.

The broad band yellow luminescence (YL) was a deep-level associated luminescence in GaN samples, and it may be caused by carrier transition from conduction bands or shallow donors to deep acceptors.[22–25] The characteristic temperature for the YL thermal quenching can be greater than 450 K, and below this temperature, the YL intensity would keep nearly constant.[24,26] Therefore, in a temperature range $\leqq$ 450 K, the change of YL intensity with temperature should be proportional to the change of electron density in the conduction band; thus, YL can act as an indicator of a carrier density change in GaN.

Herein, we report an observation of large NTQ in YL luminescence in non-polar InGaN/GaN QW samples under the optical excitation only in the QW region for the rise of temperature from 5 to 300 K. Over a four-time increase in YL luminescence indicates efficient thermal transfer of photoexcited carriers from the non-polar QWs to the GaN barrier layers where the YL is produced. By fitting the reverse temperature dependence of the YL intensity with the standard Arrhenius model, the thermal activation energy of carrier transfer is deduced.

II. EXPERIMENTAL DETAILS

In this section, the experimental details are introduced. The non-polar InGaN/GaN QW samples studied in the present study were grown on GaN epilayers with patterned pyramid shaped holes on the plane with a metal organic chemical vapor deposition technique.[27,28] The flat top plane is a non-polar (11−20) facet, and exposed faces on each hole are various semi-polar facets. Three periods of InGaN/GaN QWs were grown on such substrates and ended with a 100 nm GaN cap layer. Figure 1(a) shows a schematic structure diagram of the as-grown samples with three periods of polar and semi-polar QWs, while Fig. 1(b) presents a scanning electron microscope (SEM) image of the used GaN substrate. Three samples labeled as A, B, and C were optically investigated in the experiment, and the area ratio between non-polar and semi-polar faces increases for them with an order of A, B, and C.[27] During the PL experiments, the samples were attached on the cold finger of a closed cycle cryostat, and the temperature of samples can be changed from 5 to 300 K. For each sample, its variable-temperature PL spectra were measured, excited by lasers with wavelengths of 325, 370, and 405 nm, respectively, corresponding to the optical excitations within the top GaN cap layer, mainly within non-polar QWs and within semi-polar QWs, respectively. As shown and argued below, such selective optical excitations within different layers can make the interesting YL NTQ phenomenon observable.

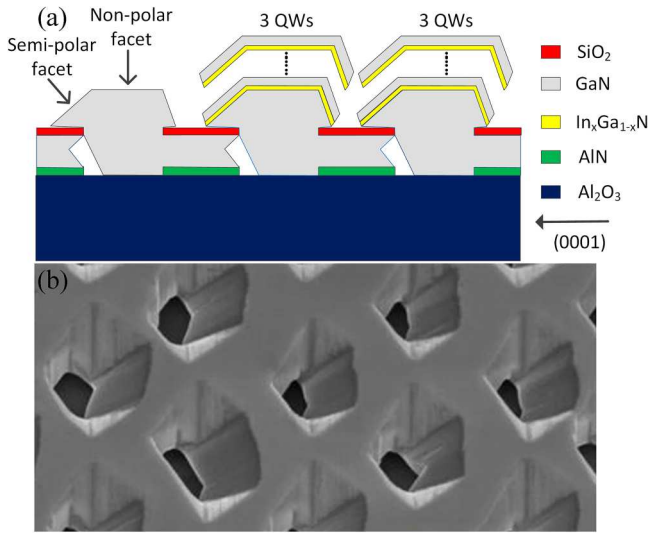

FIG. 1. Structure of the QW sample. (a) A schematic diagram (not in scale) for QW grown on non- and semi-polar GaN facets. (b) SEM image of the GaN substrate with non-polar top plane and semi-polar holes.

III. RESULTS

Figure 2 shows measured variable-temperature PL spectra of sample B. The excitation laser was a 370 nm picosecond laser with an average output power of 2.2 mW and a repetition rate of 80 MHz for the PL spectra in Fig. 2(a) and the continuous-wave 325 nm laser with an output power of 30 mW for the PL spectra in Fig. 2(b). In Fig. 2(c), the 405 nm excitation laser with 250 mW output power was used. Multiple PL peaks can be found in the spectra because of the different indium incorporation efficiency in non- and semi-polar facets.[27–29] The peak labeled as P0 includes two wavelength components of 356.9 and 362.6 nm, and they are emissions of the free excitons and basal-plane-stacking fault related excitons, respectively, in the GaN layer.[30] The peak at 383.1 nm labeled as P1B was the PL peak from the non-polar QWs of sample B at 5 K. The peaks labeled as P3 and P4 were the PL signals from the QWs with the GaN substrate in semi-polar facets of (10−11/01−11) and (10−1−1/01−1−1), respectively.[27] As will be argued later, these semi-polar QWs unlikely make significant contributions to the observed YL NTQ. The resolved broad blue band peaking at 419 nm at 300 K could be a C-related donor–acceptor transition in GaN.[31,32] The broad band YL in the GaN layer with a peak wavelength of 563 nm is labeled as P5. The sharp peak at 650 nm in Fig. 2(b) was due to the higher order reflection of the Rayleigh scattering line of the 325 nm laser by the reflecting grating in a monochromator. In Fig. 2(a), the sharp and strong signal at 370 nm was the Rayleigh scattering line of the 370 nm laser. The corresponding photon energy of a 370 nm laser is smaller than those of the two wavelengths of P0. Thus, the 370 nm laser may not excite carriers

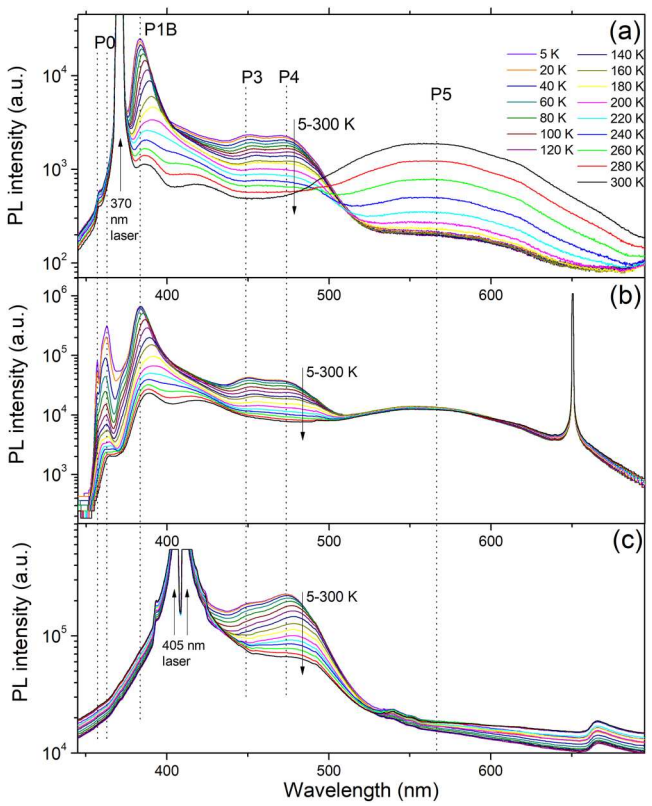

FIG. 2. Measured variable-temperature PL spectra of sample B. (a) Excited by a 370 nm laser. (b) By a 325 nm laser. (c) By a 405 nm laser.

in GaN layers effectively, whereas carriers can be efficiently excited in non-polar QWs.

For the PL spectra in Fig. 2(b), the 325 nm excitation laser can effectively excite carriers in the top GaN cap layer since the penetration length of a 325 nm UV laser was 85–100 nm within GaN.[1,2] From Fig. 2(b), the PL intensity of P0 decreases with increasing the temperature from 5 to 300 K. The PL intensities of P1B, P3, and P4 are also an overall decrease with the rise of temperature. These PL components thus show normal thermal quenching behavior caused by thermal activation of non-radiative carrier recombination channels in barriers or QWs. In sharp contrast to these peaks, the intensity of the YL band (P5) keeps nearly constant from 5 to 300 K, and the reason is that the characteristic temperature of YL thermal quenching can be greater than 450 K.[25] The nearly constant YL intensity suggests that there is no significant thermal transfer of photoexcited carriers from the QW region to the GaN barrier layers when the samples were excited with the 325 nm laser. Moreover, under this excitation condition, carriers participating in YL are dominantly from direct optical excitation in the GaN cap layer, and the YL signal was too strong to make sensitive to the contribution from the minor QW thermal emission of carriers. However, the situation was largely changed when the sample was optically excited by the 370 nm laser because the YL signal in the GaN cap layer may be mainly from the contribution of the QW thermal emission of carriers. As shown in Fig. 2(a), significant enhancement of the YL luminescence with temperature indeed indicates that the efficient thermal transfer of carriers from the QW to the barrier layers since the 370 nm laser may mainly excite carriers within the non-polar InGaN well layers. The integrated intensity of the YL band at 300 K is about 7.08 times of the integrated intensity at 5 K.

Note that the peaks of P1B, P3, and P4 overall show a similar thermal quenching property with those in Fig. 2(b). Therefore, it may be safe to claim that the YL intensity change could be a good indicator of the carrier density change in the GaN barrier layers in the interested temperature range, especially in the case of no efficient optical excitation of carriers in the GaN barrier layers. It is also noted that the P0 peak of the GaN layers is relatively much weaker in Fig. 2(a), suggesting scarce direct optical excitation of carriers in the GaN barrier layers.

Theoretically speaking, all types of QWs producing PL signals of P1B, P3, and P4 can act as the sources for carrier thermal emission. To determine which one is dominated for the thermal emission, variable-temperature PL spectra of sample B were measured with a 405 nm laser as the excitation light source. The 405 nm light may only excite carriers within the semi-polar QWs for producing P3 and P4 peaks. Interestingly, no YL signal was found at all temperatures as can be seen in Fig. 2(c); thus, the semi-polar QWs can be excluded as the dominated sources for carrier thermal emission. The shallow QW giving the PL of P1B is thus deduced to be the main source for carrier thermal emission to the GaN barrier layer. The statement sounds reasonable. As the temperature is increased, the number of carriers occupying the higher energy levels in QW increases exponentially because of thermal populating governed by the Boltzmann distribution law. As a result, the thermal emission probability of carriers from the QW to the barrier layers shall

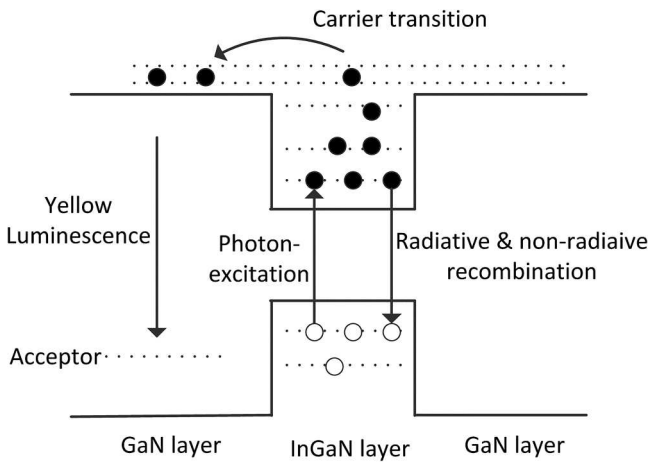

FIG. 3. A schematic diagram for the QW thermal emission and the induced YL negative thermal quenching.

significantly enhance with the rise of temperature. Figure 3 shows a schematic diagram of the photoexcitation and thermal emission of carriers in QW. The YL luminescence in the barrier layers is also indicated by a downward arrow.

Similar results were also observed in samples A and C. Figure 4 shows the variable-temperature PL spectra of samples A and C excited by a 370 nm laser. The peaks labeled as P2 are the PL from the QW with a semi-polar facet of (11−22). For reference, the PL spectrum for each sample at 5 K under the 325 nm laser excitation is also shown by a dotted line. The peak labeled as P1A in Fig. 4(a) was the PL from QW with a non-polar facet for sample A, and the PL peak labeled as P1C in Fig. 4(b) stems from the non-polar QW in sample C. The PL peak positions of P1A and P1C are on resonance with a 370 nm laser; thus, these QWs can be effectively excited by a 370 nm laser. It is these non-polar QWs that provide the source for carrier emission, resulting in large YL NTQ in samples A and C, as seen in Fig. 4. The integrated intensity of the YL band at 300 K is about 4.98 and 10.7 times of the integrated intensity at 5 K for Figs. 4(a) and 4(b).

IV. DISCUSSIONS

In this section, the thermal activation energy of thermal emission of carriers from QWs is deduced through fitting YL NTQ curves in Arrhenius plots, as shown in Fig. 5. The measured YL intensity (solid squares) vs reciprocal of temperature is depicted in the figure for the three samples, and the fitting curves (solid lines) are also shown. For simplicity, a simple Boltzmann distribution was adopted for the thermal occupation of photoexcited carriers at energy levels in QW. By assuming that the YL intensity increase is proportional to the density of thermally transferred carriers, the total YL intensity may be expressed as

$$I = I_0 + I_1 \exp(-E_0/k_B T), \tag{1}$$

where I_0 is a constant YL background intensity, I_1 a proportional constant, E_0 the thermal activation energy, k_B the Boltzmann constant, and T the absolute temperature. From Fig. 5, the Arrhenius curves of the experimental YL intensity can be well fitted using Eq. (1) for all three samples. Fitting values of I_0 were 1.4×10^4, 1.1×10^5, and 1.2×10^5, respectively, for samples A, B, and C. Adopted values of I_1 were 2.6×10^7, 1.0×10^9, and 2.0×10^8, and values of E_0 159.7, 189.1, and 135.3 meV, respectively, for samples A, B, and C. The obtained values of E_0 sound reasonable. For the

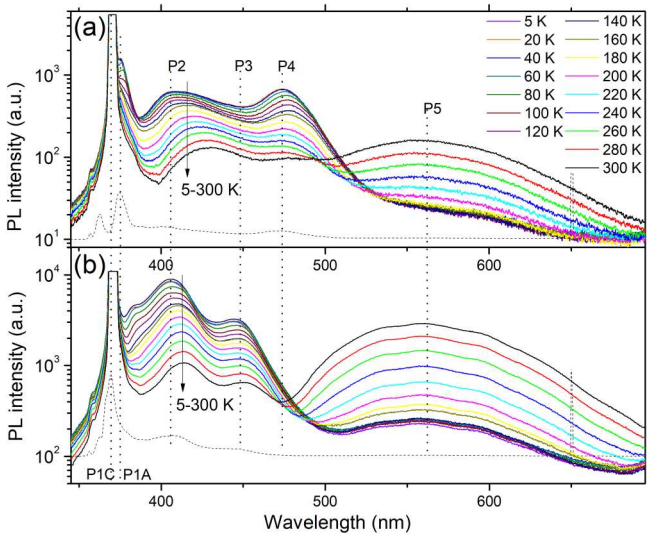

FIG. 4. Variable-temperature PL spectra with 370 nm laser excitation. (a) For sample A. (b) For sample C. The dotted line in each sub-figure corresponds to the PL spectral profile excited by a 325 nm laser at 5 K.

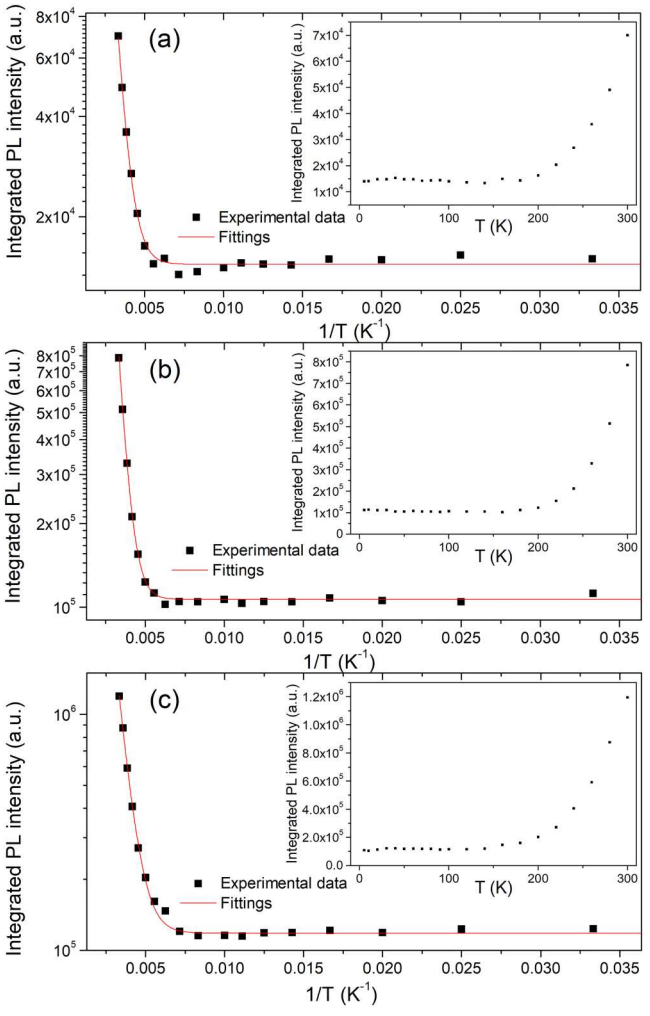

FIG. 5. Experimental integrated YL intensity (solid squares) vs reciprocal of temperature and fitting curves (solid lines) using Eq. (1). (a) For sample A. (b) Sample B. (c) Sample C. Inset figures are the integrated YL intensity vs temperature.

non-polar QWs in samples A, B, and C, their main PL peak positions are 375.0, 383.1, and 369.5 nm at 5 K. The redshift of the PL peak position may reflect the indium concentration increase for non-polar QW samples C, A, and B. For samples C, A, and B, their corresponding non-polar QW depths increase in the order of C, A, and B, which is consistent with the increased tendency of the thermal activation energy of E_0. The PL peak position differences of P1B with respect to P1A and P1C are 71.01 and 119.76 meV, respectively. The E_0 differences of sample B relative to samples A and C are 29.4 and 53.8 meV, respectively. The PL peak differences are about two times of E_0 differences, which is reasonable for supposing that the conduction band offset is equal to the valence band offset.[33]

It is worth noting that although the observed large NTQ phenomenon was explained by the efficient QW thermal emission of photoexcited carriers in the present study, other possible mechanisms may exist. In a recent article, a sharp structure at ~366 nm was experimentally confirmed by Hubáček *et al.* in the photoluminescence excitation (PLE) spectra of GaN yellow emission.[34] As the temperature is increased, the PLE peak of YL may undergo a red shift since the bandgap shrinks with the rise of temperature. Therefore, this absorption peak may shift toward the 370 nm excitation light with increasing the temperature. As a result, the absorption coefficient of 370 nm may increase, possibly giving rise to thermal enhancement of YL. We have conducted variable-temperature PL measurements on several Si-doped and intentionally undoped GaN epilayers (without QWs) grown on sapphire substrates and confirmed the NTQ phenomenon for the YL band under the subbandgap optical excitations. Activation energies of 1100.1 and 283.3 meV were obtained for undoped and Si-doped GaN, which might be related to the mediated role of V_{Ga} and Si_N for carrier transition between the GaN valence and conduction band. Figure 6 shows the example NTQ spectra of undoped GaN. The results experimentally indicate that the smaller NTQ activation energies of 135.3–189.1 meV in the present study are specific to QW-contained GaN samples, relating the observed NTQ to the presence of QW layers, supporting that the NTQ in the present study is caused dominantly by QW carrier thermal emission. The results may not be in favor of a PLE redshift mechanism. As the inner GaN mechanism, it may not be affected significantly by the presence of QW. With the presence of QW, the QW levels can be a dominant mediating level over native defects in GaN because of its high energy level density and low activation energy. Finally, it is noted that although the PLE redshift in principle can thermally enhance YL, the actual law that the photon-absorption thermal increase obeys caused by this mechanism was seldom discussed in literature studies, which means that there may be no sufficient evidence in literature studies supporting that the PLE redshift would induce exponential-like YL enhancement as Eq. (1). The detailed results about NTQ in undoped and Si-doped GaN for probing deep acceptors would be summarized as a separated paper.

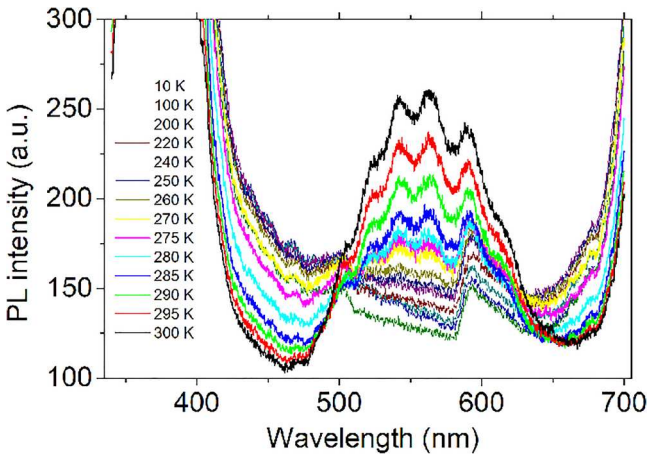

FIG. 6. The NTQ spectra of undoped GaN with the activation energy of 1100.1 meV. The NTQ occurs from 270 K in contrast to 180 K of QW samples.

V. CONCLUSIONS

A large NTQ phenomenon was observed for the YL luminescence band from the GaN barrier (buffer) layers in the non-polar InGaN/GaN QW samples only when the carriers were optically excited within the QWs. It is found that such a large NTQ phenomenon is unambiguously ascribed to the efficient thermal transfer of photoexcited carriers in QW to the GaN barrier layers. The thermal activation energy was thus determined by analyzing the temperature dependence of the YL intensity in the Arrhenius plot. The obtained thermal activation energy increases with the QW depth increase, which is consistent with the attribution of observed NTQ to QW thermal emission. The findings could be of technological significance in the field of GaN-based materials and optoelectronic devices.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data that support the findings of this study are available within the article.

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