Dynamics of carrier tunneling and recombination in asymmetric coupled InGaN multiple quantum wells

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Abstract: In this work, dynamics of carrier tunneling and recombination in InGaN-based asymmetric coupled multiple quantum wells (AC-MQWs) are systematically studied by excitation power-dependent and temperature-dependent photoluminescence (PL) measurements. With different pumping wavelengths of 405 and 325 nm, distinctly different PL spectral evolutions are observed, which could be well explained by the proposed anomalous carrier “reverse tunneling” based on the forbidden 1h→2e transitions in the AC-MQWs. The forbidden transitions are identified through the well agreement between the measured photo-modulated reflectance (PR) spectrum and the calculated interband transition energies. Our results indicate that, by ingeniously designing the MQW structure of the InGaN-based optoelectronic devices, it is possible to realize a specific interband optical transition which is even not allowed by the selection rule, and thereby effectively improve the carrier distribution across the QWs through the conventional and/or anomalous “reverse” carrier tunneling.

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1. Introduction

Semiconductor light-emitting diodes (LEDs) and laser diodes (LDs) based on InGaN/GaN multiple quantum well (MQW) structures have been unprecedentedly developed during the past three decades, and are already being commercially applied in many areas such as solid-state lighting, full-color display, and optical storage [1–4]. In particular, tuning the indium content of InGaN active mediums allows the output light from the devices to be easily tuned in a wide range from ultra-violet to green [5]. However, despite the tremendous improvements in crystal growth technique, several main challenges including “green gap” [6, 7] and “efficiency droop” [8, 9] in InGaN-based LEDs still hamper the further development of high-power and large-area LEDs for general illumination. The “efficiency droop” refers to the reduction in external quantum efficiency (EQE) of LEDs at high injection current densities. The physical mechanism causing the efficiency droop is still being debated and many theories have been proposed as the possible explanation, including carrier leakage [10–12], nonuniform carrier distribution [13], Auger recombination [12, 14], carrier delocalization [15], and junction heating [16], etc. Recently, several groups have reported that the efficiency droop can be effectively suppressed by intricately designing the InGaN/GaN MQW structures, such as the composition-graded barriers [17–21], and the thickness-graded barriers or wells [22–25]. The composition/thickness-graded coupled MQW structures can enable the enhancement of carrier injection and uniform carrier distribution for effectively increasing the recombination efficiency at a high current level [17–25]. This enhancement effect should be related to the carrier tunneling, capture, escape, and recombination processes among the novel asymmetric active regions. Up to now, although very few studies on the optical properties of InGaN-based asymmetric coupled MQWs (AC-MQWs) [26, 27] have been reported, detailed experimental investigations on the fundamental optical properties and the carrier dynamics of the InGaN-based AC-MQWs are still very lack, which are of significant interest in further improving the active-region structure and hence the device performance.

In this work, emission characteristics and physical dynamics of carrier tunneling and recombination of the InGaN AC-MQWs embedded in the depletion of a p-n junction are studied by excitation power-dependent and temperature-dependent photoluminescence (PL). The interband transition energies in the AC-MQWs are calculated using finite difference method and the results show a good agreement with the photo-modulated reflectance (PR) peaks. Then the forbidden transitions between the first quantized hole level and the second quantized electron level (1h→2e) are unambiguously identified. Here, the PL experiments were performed with different pumping wavelengths of 405 and 325 nm, respectively, the 405 nm excitation with a photon energy of 3.06 eV, is a below-barrier excitation, where carriers are excited and combined only in the quantum wells, while the 325 nm excitation with a photon energy of 3.82 eV, is an above-barrier excitation, where carriers are excited in both barriers and wells. By comparing the emission characteristics under different pumping conditions, carrier tunneling, capture, escape processes, and recombination mechanisms of the InGaN AC-MQWs are systematically investigated based on the forbidden 1h→2e transition.

2. Materials and methods

The InGaN AC-MQWs structure used in this study is the active region of a normal LED and was epitaxially grown on a (0001)-oriented sapphire by metalorganic chemical vapor
deposition (MOCVD) system. The epitaxial structure is schematically shown in Fig. 1, which consists of a 25-nm-thick GaN nucleation layer, a 1.8-µm-thick Si-doped $n$-type GaN layer, an asymmetric coupled InGaN/GaN MQWs active region, a 15-nm-thick $p$-type Al$_{0.2}$Ga$_{0.8}$N electron-blocking layer, followed by a 80-nm-thick Mg-doped $p$-type GaN upper cladding layer. The asymmetric coupled InGaN/GaN MQWs active region is constructed by 5-nm-thick GaN barriers and three In$_{0.2}$Ga$_{0.8}$N wells with different thicknesses of 3, 2.5, and 2 nm, respectively, with the last well near to the $p$-side.

The PL measurements were performed using a 405 nm continuous wave semiconductor laser diode and a 325 nm HeCd laser, respectively. To avoid the interference effect induced by multilayer films at the interfaces, photo-pumping and PL-signal-collection were masterly performed from the side of the sample, as shown in Fig. 1(a). The temperature-dependent PL experiments were performed on the sample held in a helium closed-circuit refrigerator with temperature range from 16 to 300 K. The PL signals were dispersed by a monochromator and detected by a cooled charge-coupled device (CCD).

3. Results and discussion

Figure 2(a) and 2(b) show the normalized excitation power ($P$) dependences of PL spectra at a temperature of 16 K with different pumping wavelengths of 405 and 325 nm, respectively. Three main emission peaks corresponding to the three well-thickness-varied QWs are readily identified, accompanied by a weak longitudinal optical phonon replica peak located at about 90 meV intervals to the low-energy side. At such a low temperature, radiative recombination is assumed to be the dominant recombination pathway and nonradiative recombination processes can be neglected. As it is shown, under low excitation power, only the emission peak of the widest well (3.0 nm well, with the lowest ground energy level) can be observed for both cases of 405 nm pumping (below-barrier excitation) and 325 nm pumping (above-barrier excitation). We attribute this to the conventional tunneling behavior between the adjacent QWs with different well widths: the electrons in the narrow well tunnels significantly to the wide one, hence, leading to the quenching of the luminescence for the narrow wells, as shown in Fig. 2(d). With increasing excitation power, the carrier distribution...
becomes progressively more uniform, and three emission peaks are gradually observed, despite the tunnel effect. However, different spectral evolutions are clearly revealed under the different pumping conditions at low temperature: (1) For the 325 nm pumping case, the emission from the mid well (2.5 nm well) grows much faster than the others with an excitation power range from 0.18 to 1.0 mW while the narrowest well (2.0 nm well) subsequently dominates the radiative recombination processes and exhibits the strongest luminescence as the excitation power is further increased ($P > 1.0$ mW). (2) for the 405 nm pumping case, however, the mid well shows the highest emission intensity and the narrowest well shows the weakest emission intensity when the excitation power is higher than 0.8 mW. Additionally, to reveal the nonradiative recombination processes of the investigated AC-MQWs, room temperature (RT, 300 K) excitation power-dependent PL measurements were also carried out with a pumping wavelength of 405 nm, as shown in Fig. 2(c). In contrast to the case at 16 K, the nonradiative centers are thermally activated and nonradiative recombination dominates the recombination processes. It is seen that under high excitation...
power, the narrow well exhibits a strong emission intensity, which is rather similar to that pumped by the 325 nm at low temperature.

To explain the above-mentioned spectral evolutions in detail and reveal the underlying physical mechanism, PR measurements, which have been proven to be a powerful experimental technique for investigating interband transitions in the QW [28, 29], were carried out at a cryogenic temperature of 80 K to clarify the quantized energy levels. As shown in Fig. 3(a), optical transitions related to the AC-QWs were distinctly observed. By fitting the experimental data with first-derivative Lorentzian (FDL) functions using the least-squares method, the optical transition energies were derived from the PR spectrum. Moreover, the quantized electron and hole energy levels and wave functions of the AC-MQWs were calculated using a finite-difference method based on the envelope-function approximation by taking the internal electric field into account. The electrostatic field in the $j_{th}$ layer of an arbitrary MQW or superlattice made of layers of material $k$ with a thickness of $l_k$ and dielectric constants of $\epsilon_k$ can be expressed as [30]:

$$E_j = \frac{\sum l_j (P_j - P_{j-1}) l_j / \epsilon_j}{\varepsilon_j \sum l_j / \epsilon_j}$$

with sums running on all layers (including the $j_{th}$). $P_j$ is the total (spontaneous plus piezoelectric) polarization in layer $j$. The electric field in the well was calculated to be about 2 MV/cm. The material parameters of InGaN/GaN AC-MQWs were taken from Ambacher et al. [31] and Vurgaftman et al. [32], and summarized in Table 1. The band offset ratio of conduction-to-valence band discontinuity is assumed to be 7:3 at the InGaN/GaN interfaces [33]. The experimental and theoretical transition energies derived from the PR spectrum and finite-difference calculations are listed in Table 2.

Table 1. Parameters of GaN and In$_{0.2}$Ga$_{0.8}$N used in the calculation.

|       | $\epsilon$ | $P_{ps}$ (C/m$^2$) | $P_{pz}$ (C/m$^2$) | $E_g$ (eV) @ 80 K | $m_e^*$ ($m_0$) | $m_p^*$ ($m_0$) |
|-------|------------|------------------|------------------|----------------|----------------|----------------|
| GaN   | 10.286     | -0.0339          | 0                | 3.505          | 0.200          | 1.600          |
| In$_{0.2}$Ga$_{0.8}$N | 11.146      | -0.0293          | 0.02282          | 2.726          | 0.182          | 1.606          |

Table 2. Calculated and measured transition energies of the In$_{0.2}$Ga$_{0.8}$N AC-MQWs.

| Well thickness (nm) | Optical transition | Measurement (eV) | Calculation (eV) |
|---------------------|--------------------|-----------------|-----------------|
| / GaN 3.47           | /                  | /               | /               |
| 3.0 1h→1e            | 2.66               | 2.66            | 2.66            |
| 2.5 1h→1e            | 2.76               | 2.75            | 2.75            |
| 2.0 1h→1e            | 2.83               | 2.85            | 2.85            |
| 3.0 1h→2e            | 2.99               | 3.00            | 3.00            |
| 2.5 1h→2e            | 3.11               | 3.12            | 3.12            |
| 2.0 1h→2e            | 3.20               | 3.26            | 3.26            |

Figure 3(b) shows the calculated first and second quantized energy levels and wave functions in the band diagram of the AC-MQWs structure. By comparing the transition energies between different levels with the PR spectrum, it is found that three lower-energy peaks in the PR spectrum are assigned to the interband transitions from the first quantized hole level to the first quantized electron level (1h→1e), and the other three higher-energy peaks correspond to the transitions between the first quantized hole level and the second quantized electron level (1h→2e) for each QW, as shown in Fig. 3(a). It is well known that in
zinc blend QW structure, e.g., InGaAs/GaAs system, which has a flat band profile, interband transitions must strictly obey the parity selection rule \((\Delta n = 0)\) due to the symmetry of the conduction and valence band structure. For III-nitride-based materials, however, the huge built-in piezoelectric field breaks the symmetry of electron and hole potential profile and wave function [34, 35], making the formally parity-forbidden transitions \((\Delta n \neq 0)\) possible. Actually, it is clearly seen from the shadow in Fig. 3(b) that the “1\(h\), 2\(e\)” states present a stronger overlap of electron and hole wave functions than those “1\(h\), 1\(e\)” states, meaning that the \(1h\rightarrow2e\) transitions with a large transition probability are more easily happened. Consequently, both the parity-allowed \((1h\rightarrow1e)\) and the parity-forbidden \((1h\rightarrow2e)\) transitions are able to be observed from the PR spectrum.

Note from Table 2 that the 405 nm pumping condition allows \(1h\rightarrow1e\) transitions for each QW but only \(1h\rightarrow2e\) transitions for the widest 3.0-nm well because the excitation photon energy of 3.06 eV is lower than the \(1h\rightarrow2e\) transition energies for both 2.0 and 2.5 nm QWs. In other words, only the first excited (2\(e\)) state of the 3.0 nm well and all ground states (1\(e\), 1\(h\)) of the three QWs are occupied by photo-generated carriers. Hence, the anomalous enhanced emission of the mid 2.5 nm well at 16 K shown in Fig. 2(a) can be well understood as follows. At low excitation power, most electrons tunnel to the widest 3.0 nm well and recombine. With increasing excitation power, the carriers distribution becomes increasingly uniform, and the quantized 2\(e\) energy level of the 3.0 nm well is gradually occupied by the excessive carriers due to the band-filling effect. It is known that the narrower QW has a higher recombination rate due to its stronger quantum confinement effect (QCE) and smaller quantum confined Stark effect (QCSE) [36]. For the 3.0 nm well, the small recombination rate limits its emission intensity, and thus, most excessive carriers on the 2\(e\) excited state reversely tunnel into the 1\(e\) ground state of the adjacent 2.5 nm well through the triangular potential barrier rather than relaxing to ground state in the 3.0 nm well. Besides, a considerable number of carriers in the 2.0 nm well are injected into the mid well via a conventional tunneling process. Despite the fact that the 2.0 nm well has the highest recombination rate, the recombination and tunneling losses of carriers cannot be effectively compensated, leading to a serious suppression of the radiative emission. On the contrary, in the mid 2.5 nm well, carriers consumed by radiative recombination can be rapidly compensated by both conventional and reverse tunneling processes, as discussed. Consequently, a majority of carriers radiate in the mid well with a relatively high recombination rate and eventually lead to the anomalous enhanced emission. For the 325 nm pumping case in Fig. 2(b), however, as the excitation power increases, large amounts of carriers are generated in not only the InGaN QWs but also the GaN barriers. The recombination losses of carriers on both ground and excited states can be effectively compensated via carrier tunneling and capture processes. Therefore, due to the high recombination rate, the narrow QW ultimately exhibits a stronger emission than the wider QWs at high excitation powers of \(P > 1.0\) mW.
At 300 K, the nonradiative centers are thermally activated and the nonradiative recombination dominates the recombination process. As shown in Fig. 2(c), with increasing excitation power under 405 nm pumping, the PL evolution exhibits an initially enhanced emission of the mid well, and subsequently evolves into a dominant emission of the narrower 2.0 nm well. Unlike the case at 16 K, a large quantity of nonradiative recombination centers formed by dislocations or defects lead to a serious deterioration in carrier radiation efficiency at RT [37]. In general, the narrower QW has the stronger QCE and the better crystal quality because of its smaller built-in electric field compared to the wider one [38]. As a consequence, it is convincing to consider that the radiation efficiency of carriers in narrower...
QWs is much higher than that in the wider ones due to the relatively strong carrier localization and the low dislocation density. Furthermore, the carriers on the 2e excited state are closer to the sharp triangular potential barrier and present a much smaller localized energy compared to those on the ground states. Hence, most carriers on the 2e excited state can easily escape from the well and be recaptured by the nonradiative centers, which suppresses the reverse tunneling behavior. Thus, carrier losses in the mid well could not be effectively compensated any more. Based on the above two mechanisms, the 2.0 nm well with high radiation efficiency eventually dominates the radiative emission at 300 K with an excitation power larger than 20 mW.

Fig. 4. Excitation power dependences of the PL intensity of each QW in the AC-MQWs structure.

To further understand the carrier tunneling and recombination processes, the PL intensity dependence on the excitation power for each QW at 16 K is summarized in Fig. 4. Generally, the collected PL intensity, $I$, is proportional to the injected carrier density with a power index of $F$ [39, 40]. By assuming that $P$ is approximately proportional to the injected carrier density for a fixed spot size of a pumping laser, the dependence could be characterized by $I \propto P^F$, where parameter $F$ physically reflects the various recombination processes. Here, noteworthy, index $F$ is intimately related to the carrier tunneling and escape processes among the AC-MQWs. If $F = 1$, it indicates that the radiative recombination dominates. On the contrary, if $F > 1$, the carrier tunneling and escape occur as nonradiative processes which provide shunt paths to the injected carriers at a low temperature. For the 405 nm pumping case shown in Fig. 4(a), the emission intensity of the 3.0 nm well varied approximately linearly with excitation power ($F = 1.1$), indicating that radiative recombination dominates the recombination process. As for the 2.5 and 2.0 nm wells, however, the conversion from the superlinear ($F = 1.9, 2$) to the linear ($F = 1.1$) dependence of $I$ on $P$ is presented with continuously increasing excitation power. This conversion means that the recombination mechanism changes from the dominant nonradiative tunneling processes to the dominant radiative processes, which can be explained by the gradual suppression of conventional tunneling and the appearance of reverse tunneling as the power increases. In the 325 nm pumping case, the excessive carriers escape from the well or diffuse though the QW region without being captured by the well when it is under strong excitation. The aggravated nonradiative carrier escape and diffusion in the 3.0 nm well hence result in an increase in $F$ from 1.1 to 1.6. For the narrower wells, however, the tunneling suppression effect plays a
more important role than the carrier escape/diffusion behavior. As a result, the values of $F$ respectively decrease from 2.9 and 3 to 1.4 and 1.7, indicating that a larger proportion of carriers recombine radiatively under strong excitation than weak excitation.

Fig. 5. Temperature-dependent PL spectra with pumping wavelengths of 405 nm (a) and 325 nm (b). The emission intensity of each QW as a function of temperature under the different pumping wavelengths of 405 nm (c) and 325 nm (d).

Figures 5(a) and 5(b) show the normalized temperature dependence of PL spectra over a temperature range from 16 to 300 K. The corresponding emission intensities versus temperature for each well are plotted on log-log scales in Fig. 5(c) and 5(d), respectively. Under 405 nm pumping, the narrow QW shows a slower decrease in emission intensity with increasing temperature for $T < 70$ K due to the stronger QCE and the intrinsically improved crystal quality, which can be identified from the slope value of the PL intensity curves. When $T > 70$ K, the nonradiative centers are thermally activated and defect-related nonradiative recombination starts to dominate the recombination process, particularly for the high-energy carriers on the $2e$ excited state. Then the serious carrier loss of the $2e$ excited state significantly suppresses the reverse tunneling process, leading to the rapidest decrease in emission intensity of the 2.5 nm well. As for the 325 nm pumping case in Fig. 5(d), a slightly “S-shaped” intensity evolution is clearly seen for the 2.0 nm QW with a temperature range from 50 to 180 K. This should be determined by the temperature-dependent carrier tunneling behavior [36]. That is, with increasing temperature from 16 to 70 K, the elevated thermal
energy progressively compensates the energy difference ($\Delta E$, shown in Fig. 3(b)) between the tunneling energy levels of the 2.0 and 2.5 nm wells, which increases the probability of carrier tunneling from the 2.0 nm well to the 2.5 nm well. The tunneling probability reaches to largest value at $T = 70$ K and then decreases as the temperature further increases. Such tunneling mechanism finally contributes to the S-shaped evolution of the emission intensity.

4. Conclusions

In summary, emission characteristics and physical dynamics of carrier tunneling and recombination of the InGaN/GaN AC-MQWs are systematically investigated by excitation power-dependent and temperature-dependent PL measurements. With different pumping wavelengths of 405 and 325 nm, distinctly different PL spectral evolutions are observed, which are well understood by the proposed anomalous carrier “reverse tunneling” based on the forbidden $1h\rightarrow2e$ transitions in the AC-MQWs. The forbidden transitions are identified through the well agreement between the measured PR spectrum and the calculated interband transition energies. The quantized energy-levels and associated wave functions of the AC-MQWs were calculated using a finite-difference method with taking into account of the internal electric field. Moreover, carrier tunneling, capture, escape processes and recombination mechanisms are further clarified by the excitation power and temperature dependences of the emission intensity. Conclusively, by ingeniously designing the MQW structure of the InGaN-based optoelectronic devices, it is possible to realize a specific interband optical transition which is even not allowed by the selection rule, and thereby effectively improve the carrier distribution across the QWs through the conventional and/or anomalous “reverse” carrier tunneling. These results will provide a useful guidance to fabricate novel high-performance devices.

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