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Optical joint density of states in InGaN/GaN-based multiple-quantum-well light-emitting diodes

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ABSTRACT

The optical joint densities of states of three InGaN/GaN-based light-emitting diodes with different emission wavelengths (violet, blue and green) operated at various currents were investigated. The results indicate that the blueshift of the emission with increasing current is related to the variation in optical joint density of states. Thus, the blueshift is ascribed to the screening of the piezoelectric field by carriers. A tail at the low-energy end of the density of states, corresponding to localized states, was found, and the presence of these tails broadens the spectra of the devices.

1. Introduction

Because of the wide range of energy gaps in group-III nitride-based materials, InGaN/GaN-based multiple quantum wells (MQWs) have been used in a broad range of emission wavelengths, such as blue, green and yellow light-emitting diodes (LEDs) [1]. Although the number of applications of these devices has increased in the past decade, their special features still attract considerable attention. For example, despite their high dislocation density (typically in the range 10^8–10^{10} cm^{-2}), their luminescence efficiency is peculiarly higher than expected [2]. Also, a large Stokes-like shift occurs between the emission peak and the absorption edge, and this shift correlates with indium content [3]. Many studies have confirmed that indium atoms are important to these phenomena [4]. Since localized states that result from self-organized In-rich regions are regarded as quantum dots [5] or quantum disks [6], electron–hole pairs are likely associated with the localized states rather than being transferred to threading dislocations that act as nonradiative centers [7,8]. Additionally, the quantum-confined Stark effect (QCSE) that is caused by the piezoelectric field has been found to have a large influence on the emission [9]. Because of a large lattice mismatch between GaN and InGaN, a strong strain-induced piezoelectric field tends to shift the quantum-confined level to lower energy [10]. However, the reasons for the blueshift of the emission peak with increasing current and the anomalous broadening of the full width at half maximum (FWHM) are still under debate. Since both the photoluminescence (PL) and the electroluminescence (EL) peaks are at the absorption tail, some researchers have suggested that the blueshift results mainly from the band filling effect of the localized states that correspond to the formed In-rich regions [11]. Others have argued that the blueshift is related to the screening of the piezoelectric field by carriers [12]. Chichibu et al. [13] argued that the blueshift is a combined effect of band filling of localized states and screening of the QCSE. Notably, most related studies have used PL excitation (PLE) to obtain the absorption spectrum. However, the electrically injected carriers and the corresponding emission prevent the absorption spectrum from being obtained under large forward bias. Therefore, the band filling effect of localized states and the piezoelectric effect with a forward current are difficult to determine, and definitive explanations of the blueshift and anomalous increase in the FWHM are still unavailable. Since research has left many questions unanswered, more work must be done.

2. Experiments

This paper presents a method for determining the optical joint density of states of nitride-based LEDs using an EL spectrum instead of a PLE. The proposed method of extracting the optical joint density of states is to divide the EL spectrum by the Boltzmann distribution function, to determine the effect of current on the blueshift and the increase in the FWHM [14].

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Similar methods, in the form of relations among spontaneous emission, absorption and gain spectra, have been extensively used in studying the details of InGaAsP, AlGaAs, InGaAs and GaAs laser diodes in various aspects [15–22]. Besides, the authors have successfully explained the carrier dynamics, the redshift of the edge emission, the junction temperature and the broadening of the emission spectrum of AlGaInP LEDs using this method [23,24]. All of these successful precedents justify the method used in this paper. The samples investigated herein are commercial products that were grown on the *c*-plane sapphire substrates by metal–organic chemical vapor deposition. Three InGaN/GaN-based light-emitting diodes with different emission wavelengths (violet, blue and green) were adopted. To obtain their spectra, the devices were placed on a heat sink and operated in pulsed mode with a frequency of 1000 Hz and a duty cycle of 10% to diminish any possible effect of Joule heating. Thus, the junction temperatures of the measured devices were determined only by the environment, and the corresponding Boltzmann distribution functions were obtained from the ambient temperatures. Furthermore, since the obtained raw spectra are the relative wavelength-dependent energy spectral density functions $P(\lambda)$, the relative frequency-dependent energy spectra $P(h\upsilon)$ were derived using the transformation $P(h\upsilon)=(\lambda^2 h/C_0)P(\lambda)$, where C_0 is the speed of light in a vacuum and h is Planck's constant; the relative frequency-dependent photon spectra of interest, $N(h\upsilon)$, described in the following paragraphs, were then obtained by $N(h\upsilon)=P(h\upsilon)/h\upsilon$.

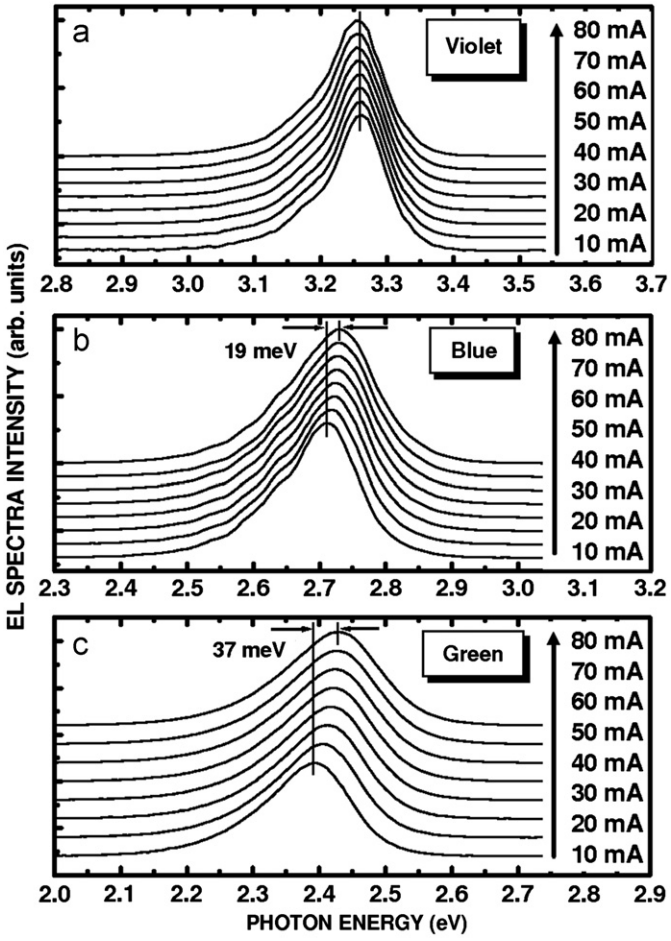

Fig. 1. EL spectra of (a) violet, (b) blue and (c) green InGaN/GaN-based LEDs at various currents from 10 mA to 80 mA at 300 K. Emission peak shifts in the three spectra are 0 meV, 19 meV and 37 meV, respectively, (for interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

Fig. 1 presents the normalized EL spectra of the samples under various currents at 300 K. The peaks shifted to higher energy as the current increased, and the degree of shift was correlated with the indium content. From 10 mA to 80 mA, the blueshifts for the violet, blue and green LEDs were 0 meV, 19 meV and 37 meV, respectively. However, the FWHMs for the violet, blue and green LEDs were 89 meV, 140 meV and 160 meV, respectively, and were almost independent of the driving current, indicating that the FWHM was strongly correlated with the indium content but weakly correlated with current density. Similar results concerning the blueshift and the increase in the FWHM have been observed in many studies, including those mentioned above.

3. Results and discussion

To determine how localized states and piezoelectric field affect the spectrum under various currents, the optical joint densities of states of these devices are determined under various currents from the measured spectra. The spectra presented herein are the relative frequency-dependent photon spectra; they should be proportional to the spontaneous emission rates arising from direct band-to-band transitions (photons/second/frequency/volume) [14,23,24]:

$$r(h\upsilon) \propto \rho(h\upsilon)e^{(-h\upsilon/KT)} \tag{1}$$

where $\rho(h\upsilon)$ is the optical joint density of states, which is the density of allowable transition states that satisfy the *k*-selection rule, or equivalently, the density of states for electron–hole pairs, and $e^{-h\upsilon/KT}$ is the Boltzmann distribution function, which describes the probability of occupation of each state by an electron–hole pair. Generally, the optical joint density of states has the same mathematical form as the electron (hole) density of states, except in that the effective mass of electron (hole) should be replaced by the reduced mass of electron–hole pairs [14]. Thus, when the optical joint density of states is obtained, the characteristics of the electron and hole densities of states are also obtained, and the relative optical joint density of states can easily be deduced from the spontaneous emission spectrum by merely dividing the emission spectrum by the Boltzmann distribution function. Notably, such a method is preferable to PLE, since PLE obtains the optical joint density of states via an absorption spectrum, which cannot be obtained at large forward bias. Fig. 2 shows the relative optical joint densities of states obtained from the EL spectra. All of these curves show two distinct

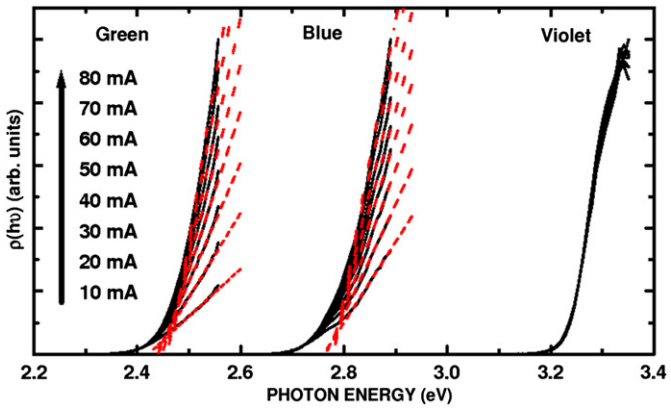

Fig. 2. Relative optical joint density of states in violet, blue and green LEDs under currents from 10 mA to 80 mA, calculated by dividing EL spectra by thermal distribution (for interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

regions—a low-energy tail and a high-energy region, where the density of states increases steeply with increasing energy. Since the low-energy tails of all of these samples are independent of current, the tails are attributed to the formation of localized states. In contrast, the steep increase of the optical joint density of states in the high-energy region indicates that these states correspond to the unlocalized states of the two-dimensional quantum structure [25], and its characteristics depend strongly on the samples. The violet LED yields the largest slope among these samples, and its density is independent of current; for blue and green LEDs, the curves of densities strongly depend on current, and their slopes increase with the current, as revealed by the linear fitted dashed lines. Since optical joint density of states of a well is highly correlated with the distributions of electric potential and the corresponding electric field around the well, the variation of optical joint density of states with current indicates that the electric field around the well was also modified by the current. Such an exotic current dependence has not been observed in non-polar materials, such as AlGaInP [23], and should be related to the strong piezoelectric field in nitride material and the screening of this field by the injected carriers. For an ideal square quantum well, the optical joint density of states should be a step function that switches from zero to a constant at the energy of effective band gap (material band gap plus quantum confined energies of electron and hole). However, in InGaN/GaN quantum well, the strong piezoelectric field can tilt the potential of the quantum well, as shown in Fig. 3(a), and electrons and holes in the same well should be spatially separated on opposite sides of the well. Thus, the lowest transition energy in the well is decreased due to QCSE [26] and the function of optical joint density of states should extend into the low energy region and gradually diminish to zero when approaching the lowest transition energy. Accordingly, the slope of increase of optical joint density of states with energy is negatively correlated with the degree of QCSE and the strength of piezoelectric field around the wells. This is the reason why the violet LED has the largest slope among these samples, since the wells of it has the lowest indium content, and the piezoelectric field around the wells should be the smallest. Furthermore, as the operating current was increased, the number of carriers in the wells increased, and the piezoelectric field drove the carriers to screen the field itself. Thus, the QCSE was eased, and the lowest transition energy was increased, as shown in Fig. 3(b). This is the reason why the slopes of blue and green LEDs are increased with the operating current. Chichibu et al. [27] and Choi et al. [28] reported similar results. Their results reveal that the slope of the absorption edge is determined by the piezoelectric field and can be screened by increasing the Si-doping concentrations in the barriers.

Since the densities of states in the high-energy region depend on current, the blueshift of the spectra may be attributed to this phenomenon. To quantitatively examine this possibility, the densities of states of blue and green LEDs at various currents were approximated by the linear fitted dashed lines plotted in Fig. 2; then, the theoretical emission spectra were obtained by multiplying these fitted curves by the Boltzmann distribution function. Fig. 4 presents the blueshifts that were obtained from these theoretical spectra and from the experiment. The agreement between these two sets of data clearly demonstrates the proposed cause of the blueshift, and provides evidence of the screening of the piezoelectric field by carriers. The underlying meaning of the covariant change of the optical joint density of states and the value of the blueshift can be further revealed by the following analytical procedure. If the linear relation $\rho(h\upsilon)=m(h\upsilon-E_0)$ is used to denote the optical joint density of states, where m is the slope and E_0 is the intercept of the $h\upsilon$-axis, then the spectrum should be $m(h\upsilon-E_0)e^{-h\upsilon/KT}$ and the peak should be located at E_0+KT. Thus, the shift of the peak is a consequence of the shift of E_0. Recalling that the optical joint density of states in high-energy region corresponds to the unlocalized states of the two-dimensional quantum structure, this E_0 should be the aforementioned lowest transition energy of the unlocalized states, and its value should strongly depend on the strength of electric field around the well. This result justifies the conclusion that the blueshift is a consequence of screening of the piezoelectric field by the injected carriers. However, the FWHMs of the numerically simulated spectra are all around 63 meV, which are consistent with the FWHM of $2.45KT$ obtained from the analytical spectrum $m(h\upsilon-E_0)e^{-h\upsilon/KT}$, but are lower than those obtained experimentally. This discrepancy results from the linear approximations of the densities of states that wholly ignored the existence of localized states in the low-energy tails [5]. Although these localized states are known to be independent of current, as mentioned above, they still contribute appreciably to the emission spectra. In fact, the low-energy sides of the EL spectra are determined by the energy distribution of these localized states. Thus, as the indium content is increased, the low-energy tail should be further extended, and the corresponding FWHM of the emission spectrum should increase.

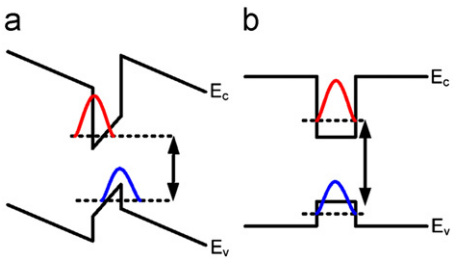

Fig. 3. Band diagrams and wave functions of elections and holes under (a) piezoelectric field and (b) screened piezoelectric field.

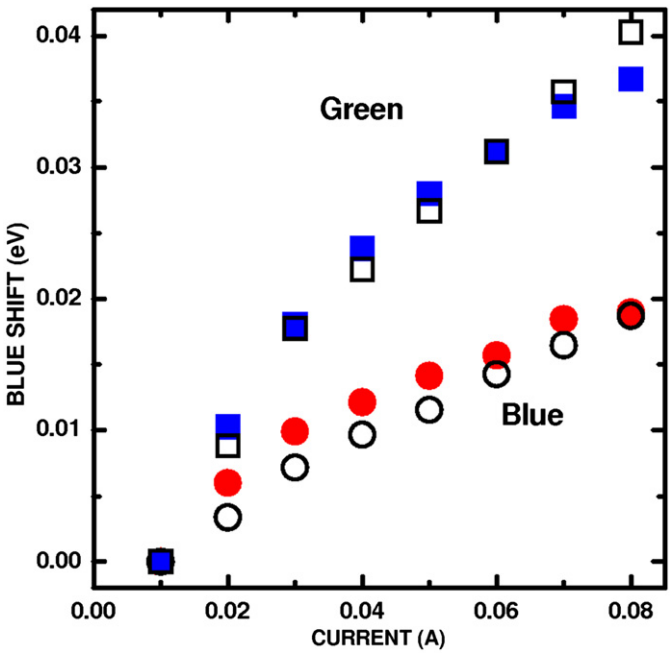

Fig. 4. Experimental and theoretical EL peak shifts with current in blue (green) LED are denoted by solid circles (solid squares) and open circles (open squares), respectively, (for interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).

4. Conclusion

In summary, the optical joint densities of states of three InGaN/GaN-based LEDs with different emission wavelengths were determined at various currents. The optical joint density of states has two distinct regions, a high-energy region, which corresponds to the unlocalized quantum well states and a low-energy tail, which corresponds to the localized states. As the operating current is increased, the density of states in high-energy region changes with the screening of the piezoelectric field. This fact causes the peak of the EL spectrum to blueshift with increasing current. The low-energy tail of the optical joint density of states determines the extension of the low-energy side of the EL spectrum, and this fact explains the anomalous increase in the FWHM of the emission of nitride-based LEDs. Furthermore, both of these effects are enhanced by increasing indium content, which is responsible for the increase in blueshift and FWHM with the emission wavelength of the device.

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