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Design and Performance of Nitride-based UV LEDs

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ABSTRACT

We overview several of the challenges in achieving high efficiency nitride-based UV (< 400 nm) LEDs. The issue of optical efficiency is presented through temperature-dependent photoluminescence studies of various UV active regions. These studies demonstrate enhanced optical efficiencies for active regions with In-containing alloys (InGaN, AlInGaN). We compare the performance of two distinct UV LED structures. GaN/AlGaN quantum well LEDs with $\lambda <$ 360 nm emission have demonstrated output powers > 0.1 mW, but present designs suffer from internal absorption effects. InGaN/AlInGaN quantum well LEDs with 370 nm< λ < 390 nm emission and > 1 mW output power are also presented.

INTRODUCTION

While much of the research in the nitride field has focused on the development of blue and green LEDs and laser diodes, UV (λ < 400 nm) LEDs based on GaN, AlGaN and/or AlGaInN active regions are also of great interest and are well suited to a number of applications. One of the most promising uses of a UV LED is as a high energy excitation source. In the biomedical and chemical sensing fields, UV LEDs can be used as compact and robust excitation sources of fluorescence. UV LEDs may also prove to be superior to blue LEDs in exciting phosphors for white lighting applications, due to improved color rendering. To date, there have been relatively few reports of UV LEDs based on the wide-bandgap nitride semiconductors. In particular, Akasaki et. al. [1] reported on a GaN/AlGaN double heterostructure (DH) LED with emission at 370 nm and up to 1.5% external quantum efficiency. Mukai, et. al. [2] have achieved an impressive 5 mW output at 371 nm from an InGaN/AlGaN DH LED with very low levels of indium (In) in the active region. That LED was reported to have up to 7.5% external quantum efficiency but the efficiency dropped by more than an order of magnitude as the emission wavelength shifted to 368 nm with the total elimination of In from the active region. Shorter wavelengths have been achieved by Han, et. al. [3] through a GaN/AlGaN multiquantum well (MQW) structure. These LEDs demonstrated a 354 nm emission peak with a narrow FWHM linewidth of 5.8 nm and relatively low output powers of 12 μW at 20 mA. Thus, while one group has demonstrated high (> 5%) efficiency LED performance for λ > 370 nm, many of the challenges inherent to the shorter wavelength emission regime still remain.

In this paper, we discuss challenges for achieving high performance UV LEDs and present the performance of two distinct UV LED structures. We first describe the materials growth and the general heterostructure designs for the UV LEDs that we have developed. In the next section, we review critical materials and design issues such as optical efficiency of UV active regions and internal absorption effects. We then report on the performance of GaN/AlGaN MQW LEDs with emission wavelengths < 360 nm as well as the performance of InGaN/AlInGaN MQW LEDs for 370 nm< λ < 390 nm.

MATERIALS GROWTH AND HETEROSTRUCTURE DESIGN

The nitride materials described in this paper were grown in a high speed (~ 1200 rpm) rotating disk MOCVD reactor on two inch sapphire substrates. Different growth conditions were employed for non-indium containing alloys (GaN, AlGaN) as compared to indium containing alloys (InGaN, AlInGaN). The GaN and AlGaN growths [3] were typically carried out at 1000-1080°C with hydrogen as the carrier gas. Ammonia (NH_3), Trimethylgallium (TMGa), Trimethylaluminum (TMAl) were used as the N, Ga, and Al precursors, respectively. The growth of InGaN and AlInGaN [4,5] was carried out at 750-800°C, with nitrogen as the carrier gas and Triethylaluminum (TEA) and Trimethylindium (TMI) as the Al and In sources, respectively. A standard two-step growth (550 and 1050 °C for the low and high temperatures, respectively) with a GaN low-temperature buffer layer (~250 Å) was used in this work.

We will focus on two general heterostructures for UV LEDs. The overall layer sequence is quite similar for the two structures, and is shown in Figure 1. The first design is an MQW LED structure with GaN quantum wells and $Al_xGa_{1-x}N$ barriers and emission in the $\lambda < 360$ nm region. The n-GaN buffer layer is typically 3 μm in thickness and Si-doped to a level of $2\text{-}5\times10^{18}$ cm^{-3}. The n-AlGaN cladding with x=0.15-0.20 is approximately 400 Å thick and doped to a similar level as the n-GaN layer. The multiquantum well region consists of 5 periods of 30 Å thick GaN quantum wells and 70 Å thick AlGaN (x=0.15-0.2) barriers. The p-AlGaN cladding is typically 400 Å thick and the contact layer is 0.05-0.1 μm thick p-GaN. The second type of UV LED structure utilizes InGaN QWs and AlInGaN barriers in the MQW active region. The MQW region consists of 47 Å thick $In_xGa_{1-x}N$ quantum wells with x=0.04 and 48 Å thick $Al_yIn_xGa_{1-x-y}N$ barriers with x=0.04 and y=0.14. The p-GaN cap layer is thicker for these structures, due to the fact that the longer wavelengths emitted from the active regions are not as strongly absorbed by the p-GaN layer. Thicknesses of 0.1 μm to 0.25 μm have been used.

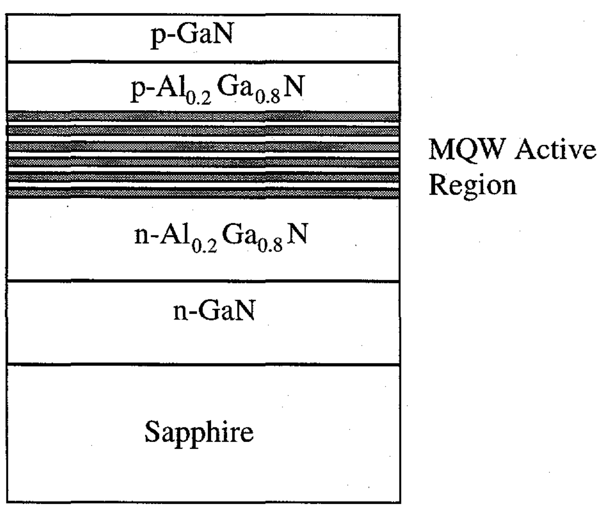

Figure 1: Schematic of UV LED MQW heterostructures

DISCLAIMER

OPTICAL EFFICIENCY OF UV ACTIVE REGIONS

Although relatively good performance has been reported in the near UV region of $\lambda >$ 370 nm, there exist a number of distinct challenges for achieving high efficiency at the UV wavelengths, and these challenges become particularly difficult for wavelengths shorter than 370 nm. One of the most intriguing issues in the nitride field concerns the high optical efficiency of nitride-based blue and green LEDs. Specifically, although a great deal of progress has been made in the development of InGaN-based light emitters, the role played by indium (In) in contributing to the optical efficiency is still quite controversial. A number of groups have proposed that the inhomogeneity of In incorporation results in carrier localization at In-rich regions and that this localization leads to enhanced optical efficiency [6-8]. Support of this hypothesis is found in cathodoluminescence experiments that demonstrate a variation of the PL emission energy on the microscale, suggesting that In composition variations on the order of several percent are possible [9]. Further insight is gained by the time-resolved spectroscopy experiments of InGaN quantum well structures performed by Narukawa, et. al. [10], which suggest that the density of non-radiative centers and possibly the non-radiative recombination mechanism itself is altered when In is included in the growth. A similar result was obtained by Kumano, et. al. [11] who suggest that increased optical efficiency is due to reduced non-radiative recombination centers with In incorporation. In contrast to these theories, other experiments [12] suggest that the majority of optical spectroscopy data on InGaN quantum wells can be explained entirely by piezoelectric field effects. Thus, it is clear that a strong consensus has not emerged as to how the presence of In in the QWs affects the optical efficiency and whether it is absolutely necessary for achieving high efficiency nitride LEDs.

This issue of whether In-containing QWs are needed for high efficiency is especially critical if we look at UV LED structures. Clearly the GaN MQW LEDs do not contain In in the active regions, and therefore would not benefit from the proposed improvements in optical efficiency seen in the blue and green LEDs. Furthermore, in order to achieve LED emission at λ < 390 nm from InGaN MQW structures, the In composition must be reduced to relatively small values ($x < 0.06$). Thus even in the InGaN/ AlInGaN MQW structures described in this report, the role played by In could be significantly reduced from that of blue and green LED structures with higher In compositions in the InGaN QWs.

In an effort to further elucidate these issues, we have performed a number of photoluminescence studies of InGaN, GaN and AlGaN MQW and bulk structures. In particular, we have performed temperature-dependent photoluminescence (PL) spectroscopy measurements on a number of MOVPE grown $In_xGa_{1-x}N$ epilayers in the low In composition regime ($x < 0.10$) [13]. This composition regime was chosen to examine whether a clear trend in optical efficiency and temperature dependent quenching of PL intensity can be found with the addition of just small amounts of In. Our work has also intentionally focused on relatively thick (0.2 μm) and doped bulk InGaN epilayers so that the role of piezoelectric field effects would be minimized [14]. This work is therefore distinct from the majority of the previously reported work that has focused on InGaN quantum wells with higher ($x \geq 0.1$) In composition.

The PL measurements were performed using a HeCd laser (325 nm) at a low power density of approximately 30 W/cm^2. A 0.3 meter spectrometer with an integrated UV enhanced CCD detector was used, with a spectral resolution of approximately 0.2 nm. The room temperature PL spectra for four InGaN epilayer samples is shown in Figure 2. A strong increase

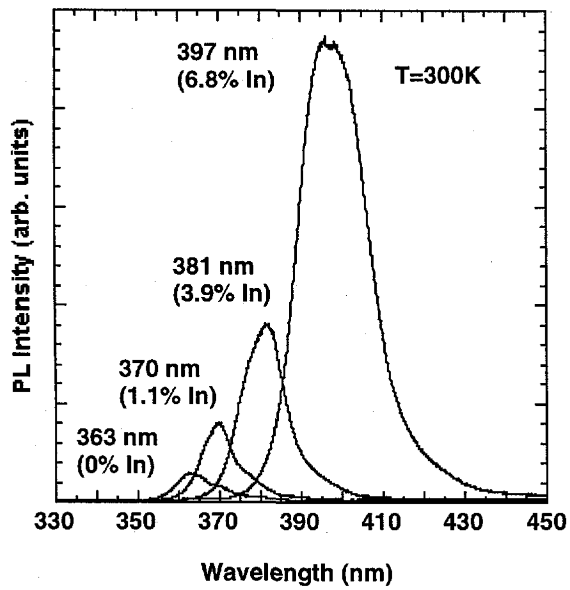

Figure 2 (a) Room temperature PL spectra of InGaN bulk epilayers.

in the integrated PL intensity is seen as In composition is increased, with more than a 25X increase as the peak wavelength shifts from 363-397 nm. The data suggest that the optical efficiency is highly dependent on the In composition. The full temperature dependence of the integrated PL intensity is plotted in an Arrhenius plot in Figure 3. The data is fit with the following formula [15]

$$I=I_0/[1+\alpha \exp(-E_a/kT)]. \qquad (1)$$

Here the non-radiative decay is assumed to be thermally activated such that the non-radiative

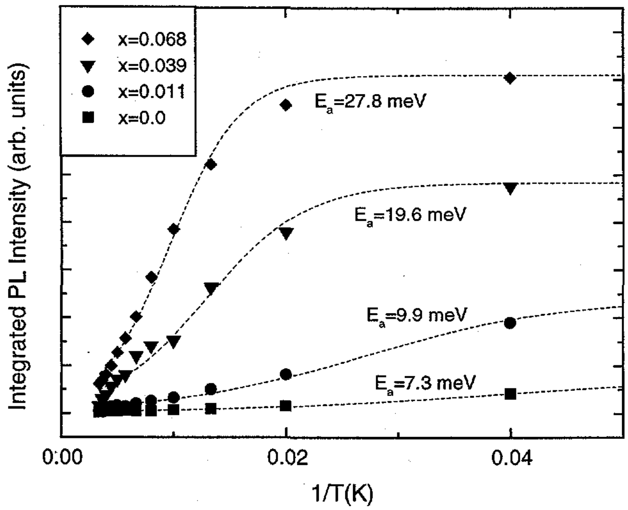

Figure 3: Temperature dependent integrated PL intensity of InGaN bulk eplilayers with In composition x. The E_a values are derived from fitting the data to equation 1 [13].

lifetime $\tau_{nr}=\tau_o\exp(E_a/kT)$ and E_a is the activation energy for PL quenching. The parameter α is equal to τ_r/τ_o where τ_r is the radiative lifetime. From the E_a values, we see a systematic increase in the activation energy as the indium composition is increased. The full temperature dependent data thus give further support of the hypothesis that increasing In composition improves the optical efficiency of the materials. Through the increasing E_a values with increasing In composition, we see that the In is reducing the effectiveness of non-radiative recombination mechanisms in quenching the PL intensity. Whether this effect is due to carrier localization or a modification of the nature of the non-radiative centers can not be determined from the data.

Additional temperature dependent PL measurements were performed on InGaN, GaN and AlGaN MQW structures. The InGaN QW structures were supplied by Meijo University and have 10 periods of 23 Å thick $In_{0.21}GaN_{0.79}N$ QWs with GaN barriers. The GaN MQW structure was grown at Sandia National Laboratories and consists of 4 periods of 30 Å thick GaN quantum wells with $Al_{0.20}Ga_{0.80}N$ barriers. It should be noted that this GaN MQW structure was grown on an AlGaN buffer so that the QW emission would not be confused with emission from a thick GaN buffer layer. The AlGaN MQW structure was identical to the GaN MQW structure except that approximately 5% aluminum was added to the QWs. In Figure 4, we show the low temperature (T=10K) photoluminescence spectra for the three MQW structures. One notable result is that at low temperature the peak PL intensity from the UV structures is quite comparable to that of the InGaN blue MQW structure. In Figure 5, we show the temperature dependence of the peak PL intensity for the three MQW structures. A strong distinction is seen in the total drop in peak PL intensity. In particular, the InGaN MQW structure experiences a relatively small (4X) loss in peak PL intensity from 10-300K, while the peak PL drop is 60X and 1000X for the GaN MQW and AlGaN MQW structure, respectively. In the quantitative comparison of the PL data for these three MQW structures, it is important to recognize that the growth conditions for the visible and UV MQW structures were quite different, and it is possible that optimization of the growth conditions for the UV MQW structures would improve the performance. Still, this data is a good representation of the performance of our current GaN and MQWs and high

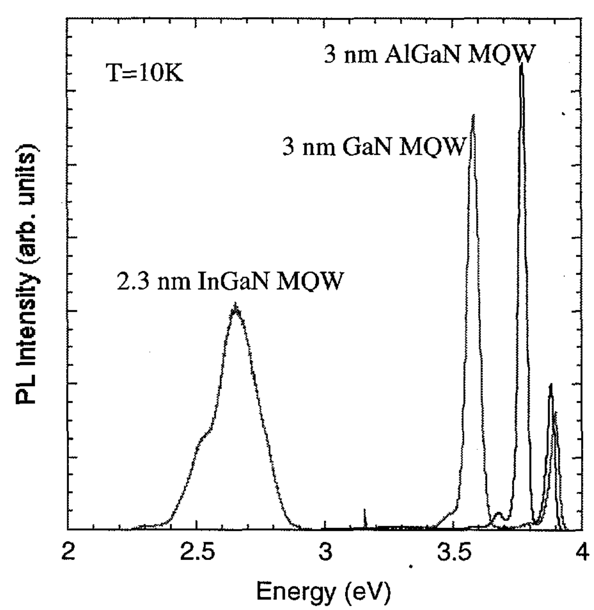

Figure 4: (a) T=10K PL of InGaN, GaN and AlGaN MQWs.

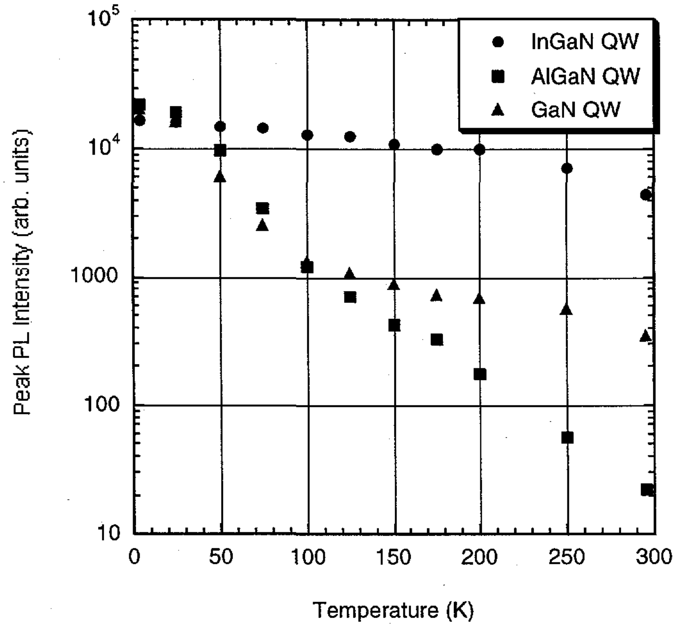

Figure 5: Temperature dependence of peak PL intensity for InGaN, GaN and AlGaN MQWs.

quality InGaN MQWs. In this data, we see evidence that the discrepancy in the optical efficiency between the visible and UV MQW structures is highly temperature dependent. In particular, the UV MQW structures are more susceptible to non-radiative recombination processes which serve to quench the PL intensities at room temperature. Furthermore, the severe 1000X drop in intensity from 10-300K for the AlGaN MQWs shows the increasing challenge of obtaining high optical efficiency at wavelengths in the 340 nm region and shorter.

As a final experiment, we performed temperature dependent PL studies of InGaN MQW structures where the In composition was kept to a low value (~ 4%) to enable room temperature emission at 380 nm in the UV. These structures are similar to the MQW region of the LED heterostructures described in section 2. The particular structures grown for PL studies consisted of 10 periods of 47 Å thick InGaN (x=0.04) QWs with AlInGaN barriers. A drop of ~5X in the integrated PL intensity from 10-300K is seen [16], which is similar to the performance of the blue-emitting InGaN MQW structure described in Figure 5.

In summary, the spectroscopic studies that we have performed on InGaN bulk films and InGaN, GaN and AlGaN MQWs have shown that samples with In-containing QWs can have significantly higher optical efficiency than samples with no In in the active region. This result might suggest that GaN/AlGaN MQWs may not be intrinsically capable of performing to the level of InGaN near UV and visible MQWs. To improve the performance of GaN/AlGaN MQWs, a further measure of growing the structures on epitaxially laterally overgrown GaN to significantly reduce dislocation densities may serve to largely improve the non-radiative recombination problem. Indeed, studies by Mukai et. al. have shown that GaN/AlGaN DH LEDs had up to 2X increased output powers when this technique was employed [17].

INTERNAL ABSORPTION EFFECTS FOR SHORTER WAVELENGTH (λ < 370 nm) UV LEDs

In consideration of the challenges in obtaining high efficiency UV LEDs, one must also seriously consider the detrimental role played by internal absorption for UV LEDs with $\lambda < 370$ nm. At the heart of this problem is the fact that most nitride-based LEDs rely on 3-4 μm thick n-GaN buffer layers and ~0.1 μm thick p-GaN capping layers for good current spreading and low contact resistance. While these GaN layers are rather transparent for blue LEDs operating at 450 nm, they become strongly absorptive as the QW emission wavelength reaches 370 nm and shorter wavelengths. This effect has been described by both Mayer et. al. [18] and Mukai et al. [19] as a significant contribution to the sudden loss of optical efficiency for $\lambda < 370$ nm.

We have most clearly seen this effect in the electroluminescence (EL) spectra of AlGaN MQW LEDs operating at 340 nm. These LEDs have similar AlGaN MQW active regions to those described in section 3.1. In Figure 6, we show the EL spectrum at 80 mA injected current. A clear delineation of the GaN absorption edge can be seen at approximately 365 nm. Fabry-Perot (F-P) oscillations exist in the spectrum for wavelengths below this absorption edge, signifying the transparency of the sample at those wavelengths. In contrast, the 340 nm peak from the AlGaN QWs is significantly reduced in intensity compared to what one would expect from the tail emission and shows no F-P oscillations. Thus, it is clear that if one needs high efficiency at these shorter wavelengths, a more transparent buffer layer, such as one consisting of higher bandgap AlGaN or AlInGaN must be employed. Progress in AlGaN buffer layers has been reported by Takeuchi et. al. [20], who have demonstrated growth of a blue laser structure on an $Al_{0.03}Ga_{0.97}N$ / $Al_{0.06}Ga_{0.94}N$ buffer layer.

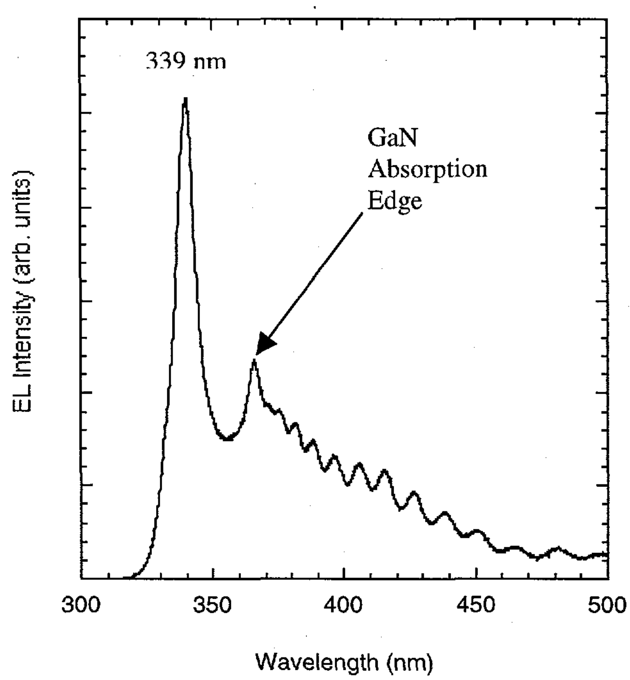

Figure 6: EL spectrum of an AlGaN MQW LED.

LED PERFORMANCE

A. PERFORMANCE OF GaN/AlGaN MQW LEDS for λ < 360 nm

We have fabricated GaN/AlGaN MQW LEDs using the heterostructure design shown described in section 2. 100 μm-250 μm mesas were defined by inductively coupled plasma (ICP) etching. Ti/Al/Ti/Au was used as the n-contact, and Ni/Au oxidized to form semi-transparent NiO [21] was used as the p-contact. Initial structures had a relatively narrow 30 Å QW, and electroluminescence peaked at 354 nm [3]. The L-I-V data was taken with a calibrated Si detector in close proximity to the sample, and demonstrated 12 μW of output power at a current of 20 mA (~ 180 A/cm^2 for these devices), and a turn-on voltage of approximately 4V. This performance results in an external quantum efficiency of below 0.1%, which is largely due to the internal absorption effects. The EL FWHM of 5.8 nm is significantly narrower than that reported for InGaN blue and green LEDs; a feature which is often desirable for spectroscopic applications.

We have further optimized the growth of the GaN QW regions as well as explored QW structures emitting at slightly longer wavelengths where the internal absorption effects would be reduced. In Figure 7, we show the performance of a GaN/AlGaN MQW LED with emission at 357.5 nm. From the EL spectrum, and one can see that the tail of the spectrum is enhanced due to the strongly reduced absorption of the GaN buffer layer at those wavelengths.

The L-I data shown in Figure 7b was taken from LEDs bonded to TO-headers (no encapsulation or lens), and using an integrating sphere coupled to a calibrated Si detector. These LEDs showed > 100 μW output at currents up to 50 mA (~ 100 A/cm^2 for these larger devices). While these powers are at least an order of magnitude less than that measured from commercially available blue and green LEDs, such powers are already sufficient for a number of fluorescence-based sensing applications.

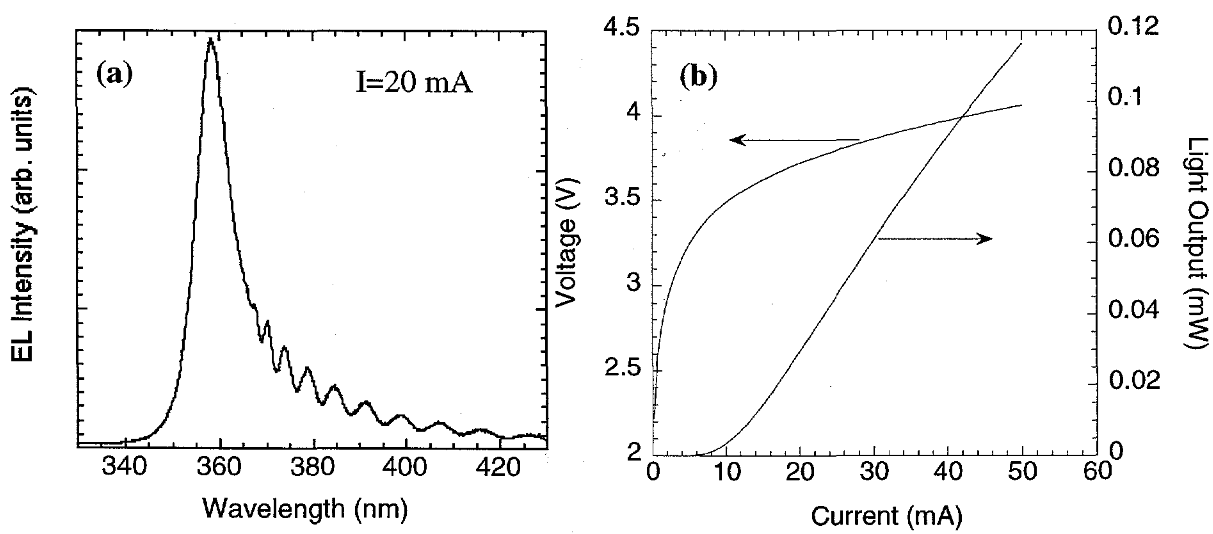

Figure 7: (a) Electroluminescence spectrum of a GaN/AlGaN MQW LED at 20 mA. (b) Light output-current-voltage data for this device.

B. PERFORMANCE OF InGaN/AlInGaN LEDS FOR 370 nm < λ < 390 nm

We have further explored UV LEDs employing AlInGaN quaternary alloys in the active region. These materials offer a great deal of flexibility in that they can be lattice matched to GaN and AlGaN buffer layers and thus critical thickness and cracking issues may be eliminated. Despite the obvious benefits of these quaternary alloys, the materials growth is quite challenging due partly to very dissimilar optimal temperatures for Al and In incorporation. As a result, very few reports have been made on the growth and optical properties of these materials [22,23]. The buffer and cladding layer designs are similar to GaN/AlGaN MQW structures, but the active region consists of 47 Å thick $In_xGa_{1-x}N$ quantum wells with x=0.04 and 48 Å thick $Al_yIn_xGa_{1-x-y}N$ barriers with x=0.04 and y=0.14. This quaternary alloy composition has been found to have a room temperature PL peak at ~357 nm.

In Figure 8, we show the electroluminescence spectrum and L-I-V data for a InGaN/AlInGaN MQW LED. The EL spectrum, taken at 20 mA, shows a peak at approximately 386 nm and a FWHM of 10 nm (84 meV). The Fabry-Perot oscillations seen throughout the spectrum indicate that these longer wavelengths experience relatively little absorption from the GaN layers in the structure. The L-I-V data, taken on devices bonded to a TO-header and using an integrating sphere and calibrated Si detector, show 0.5 mW at 50 mA (~100 A/cm^2) and greater than 1 mW powers at current levels of 100 mA (~ 200 A/cm^2). The peak external quantum efficiency of these devices is approximately 0.3%. Work is in progress to optimize growth and design of these structures to achieve higher operating efficiencies.

CONCLUSIONS

We have overviewed a number of critical issues that must be addressed to achieve high efficiency UV LEDs. The issue of optical efficiency of UV active regions was explored through photoluminescence studies of InGaN, GaN and AlGaN bulk epilayers and MQW structures. Our results suggest that improved optical efficiency is achieved for active regions with In-containing alloys in the QWs. We present performance data for GaN/AlGaN MQW LEDs with emission

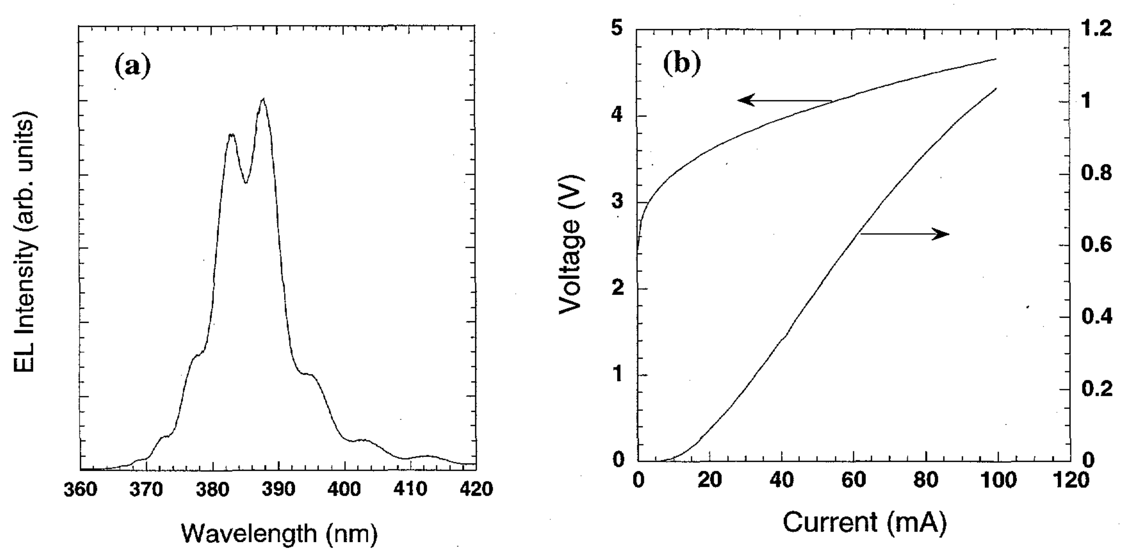

Figure 8: (a) Electroluminescence spectrum of InGaN/AlInGaN MQW LED at 20 mA. (b) Light output-current-voltage data for this device.

in the 354-358 nm region. Output powers > 100 μW have been achieved, which is sufficient for a number of fluorescence-based sensing applications. A new InGaN/AlInGaN MQW UV LED was described which demonstrated > 1 mW output powers at an emission wavelength of 386 nm. Further optimization of this structure may provide a high efficiency near-UV source suitable for phosphor excitation and white light generation.

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REFERENCES

[1] I. Akasaki and H. Amano, Jpn. J. Appl. Phys., **36**, 5395(1997).
[2] T. Mukai, D. Morita and S. Nakamura, J. Cryst. Growth, **189/190**, 778 (1998).
[3] J. Han, M. H. Crawford, R. J. Shul, J. J. Figiel, M. Banas, L. Zhang, Y. K. Song, H. Zhou and A. V. Nurmikko Appl. Phys. Lett., **73**, 1688 (1998).
[4] J. Han, T. B-. Ng, R. M. Biefeld, M. H. Crawford, and D. M. Follstaedt, Appl. Phys. Lett. 71, 3114 (1997)
[5] J. Han, J. J. Figiel, G. A. Petersen, S. M. Myers, M. H. Crawford, and M. A. Banas, to appear in Jpn. J. Appl. Phys. (2000)
[6] E. S. Jeon, V. Kozlov, Y.-K. Song, A. Vertikov, M. Kuball, A. V. Nurmikko, H. Liu, C. Chen, R. S. Kern, C. P. Kuo and M. G. Craford, Appl. Phys. Lett., **69**, 4194 (1996).
[7] S. Chichibu, T. Azuhata, T. Sota and S. Nakamura, Appl. Phys. Lett., **70**, 2822 (1997).
[8] T. Wang, D. Nakagawa, M. Lachab, T. Sugahara, S. Sakai, Appl. Phys. Lett., **74**, 3128 (1999) and references therein.
[9] S.F.Chichibu, A.Shikanai, T.Deguchi, A.Setoguchi, R.Nakai, H.Nakanishi, K.Wada, S.P.DenBaars, T.Sota, and S.Nakamura, Jpn. J. Appl. Phys. **39**, Part 1 (2000) (to be published)
[10] Y. Narukawa, S. Saijou, Y. Kawakami, S. Fugita, t. Mukai, S. Nakamura, Appl. Phys. Lett., **74**, 558 (1999).
[11] H. Kumano, K. Hoshi, S. Tanaka, I. Suemune, X. Q. Shen, P. Riblet, P. Ramvall and Y. Aoyagi, Appl. Phys. Lett., **75**, 2879 (1999).
[12] P. Riblet, H. Hirayama, A. Kinoshita, A. Hirata, T. Sugano, Y. Aoyagi, Appl. Phys. Lett., 75, 2241 (1999).
[13] M. H. Crawford, J. Han, M. A. Banas, S. M. Myers, G. A. Petersen and J. J. Figiel, to appear in MRS Internet J. of Nit. Semicond. Research (2000).
[14] V. Fiorentini, F. Bernadini, Phys. Rev. B, **60**, 8849 (1999).
[15] M. Leroux, N. Grandjean, B. Beaumont, G. Nataf, F. Semond, J. Massies, P. Gibart, J. Appl. Phys., **86**, 3721 (1999).
[16] M. H. Crawford, unpublished.
[17] T. Mukai and S. Nakamura, Jpn. J. Appl. Phys., **38**, 5735 (1999).

[18] M. Mayer, A. Pelzmann, C. Kirchner, M. Schauler, F. Eberhard, M. Kamp, P. Unger and K. J. Ebeling, J. Cryst. Growth, **189/190**, 782 (1998).
[19] T. Mukai, M. Yamada and S. Nakamura, Jpn. J. Appl. Phys.,**37**, L1358 (1998).
[20] T. Takeuchi, T. Detchprohm, M. Iwaya, N. Hayashi, K. Isomura, K. Kimura, M. Yamaguchi, H. Amano, I. Akasaki, Yw. Keneko, R. Shioda, S. Watanabe, T. Hidaka, Y. Yamaoka, Ys. Kaneko and N. Yamada, Appl. Phys. Lett., **75**, 2960 (1999).
[21] Jin-Kuo Ho; Charng-Shyang Jong; Chiu, C.C.; Chao-Nien Huang; Kwang-Kuo Shih; Li-Chien Chen; Fu-Rong Chen; Ji-Jung Kai, J. Appl. Phys., **86**, 4491 (1999).
[22] M. E. Aurner, S. F. LeBoeuf, F. G. McIntosh and S. M. Bedair, Appl. Phys. Lett., **75**, 3315 (1999).
[23] J. Han, J. J. Figiel, G. A. Petersen, S. M. Myers, M. H. Crawford, M. A. Banas and S. J. Hearne, to appear in MRS Internet J. Nitride Semicond. Research (2000).