



بررسی خواص اپتیکی و ترکیبی ایندیوم آلومینیوم نیتريد به عنوان یک ماده جدید در گسیلنده های نوری

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چکیده- ایندیوم آلومینیوم نیتريد (InAlN) یک ماده نیمه هادی مهم و البته چالشی از دسته نیمه هادی های نیتريدی بوده که اخیراً مورد توجه دانشمندان قرار گرفته است. یکی از چالش های اساسی فرآوری این ماده، مشخص کردن پهنای باند در غلظت های پایین ایندیوم است و هنوز تحقیقات و داده های قابل اتکایی برای آن وجود ندارد. در اینجا، چاه های کوانتومی به روش اپیتکسیال بر روی زیرلایه از جنس آلومینا رشد داده شده و مشاهده شد که در طول موج های بین ۳۰۰ الی ۳۵۰ نانومتر نور گسیل می کنند. همچنین لایه های با ضخامت حدود ۱۰۰ نانومتر رشد داده شده و پهنای نواری InAlN بوسیله فتولومینسانس اندازه گیری شد. مشاهده شد که با افزایش دمای رشد از غلظت ایندیوم کاسته شده و همچنین با افزایش غلظت ایندیوم پهنای باند به سرعت و بصورت غیر خطی کاهش می یابد.

کلید واژه- ایندیوم آلومینیوم نیتريد، اپیتکسی، گسیلنده نور، باند انرژی

Optical and Compositional properties of Indium Aluminium Nitride as a New Active Material for Light Emitting Applications

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Abstract- Among all III-Nitride materials Indium Aluminium Nitride (InAlN) is an important but challenging material system used in many applications including InAlN/GaN high electron mobility transistors, InAlN/GaN Bragg reflectors and InAlN-based photodetectors. Despite this, a significant discrepancy (± 0.3 eV) exists in the band-gaps reported for even the most studied and technologically relevant compositions, around the lattice matched condition to GaN. For accurate estimation of band-gap across the whole composition range, a detailed data set for the lowest indium content region is needed, but has not been available. Here InAlN/AlGaN multiple quantum wells (MQWs) emitting between 300 and 350 nm have been prepared by metalorganic organic chemical vapor deposition (MOCVD) on planar AlN templates. Also we studied InAlN epitaxial layers grown on AlN by MOVPE at different temperatures and determined their band-gap E_g by means of photoluminescence excitation spectroscopy.

Keywords: Indium Aluminium Nitride, InAlN, Band-Gap, Luminescence, Epitaxy, MOCVD

1. Introduction

III-nitride-based light emitting devices have reached high power conversion efficiency in the blue-violet spectral range. Many efforts are being made to expand this success into both the green/yellow region where InGaAlP based emitters lose their efficiency due to the transition from direct to indirect band-gap [2], and into the ultraviolet (UV) region where currently there is no alternative to III-nitrides. For the UV emitters, AlGaIn is the most widely employed as the active region quantum well (QW) material [3]. AlGaIn is a relatively well understood alloy, both in terms of the physics and growth conditions used [4]. In contrast, InAlN is less well understood and requires radically different growth conditions that are a compromise between those for the binary components [5]. Furthermore no work has been reported on its use as a light emitting material, though Hirayama et al. [6] and a few other groups [7], [8] and [9] have reported the inclusion of small (<6%) mole fraction of In and, more recently, up to 10% [10] into quaternary InAlGaIn alloy QWs. Despite this little, if any, attention has been paid to InAlN as a material for the active region of light emitting devices, though it is extensively used in Bragg reflectors [11], high electron mobility transistors [12], and as barrier material in lattice matched GaN/InAlN MQWs [13]. In this paper, we report on the use of InAlN as a promising alternative active region for 300–350 nm emitters.

Also band-gap and bowing parameter of this alloy is reported in whole range of composition.

2. Experimental

All samples studied here were grown on c-Al₂O₃ substrates by MOCVD in a 3×2" reactor using TMGa, TMIn, TMAI and ammonia as precursors. The choice of substrate was determined by the need to have UV transparency in practical light emitting devices, and to ensure the III-nitride growth was also c-oriented, which gives the most reliable

structural and morphological quality in this material system. For optical studies the InAlN/AlGaIn 5QW stacks were grown on 2 μm AlGaIn templates, which were deposited on 2.5 μm AlN on c-Al₂O₃. All samples were grown at nominally identical conditions except for QW growth temperature T_{QW}. Growth temperature was varied in the range of 710-790°C. Samples were characterized by X-ray diffraction (XRD) system. Photoluminescence (PL) was done using a 244 nm laser or monochromator coupled Xe-lamp at the room temperature. PL and PL excitation (PLE) spectra were detected using imaging spectrometer equipped with a CCD camera and a photomultiplier.

For determination of band-gap and bowing parameter, 80-100 nm thick In_xAl_{1-x}N epilayers were grown on 2.5 μm AlN-templates on c-Al₂O₃. The In composition, x, was varied from below 0.01 to 0.224 using growth temperatures T_g in the range of 940°C to 730°C. A high V/III ratio of 5400, TMIn/TMAI ratio of 1, pressure of 70 mbar and nitrogen ambient were used for InAlN; the growth rate at such conditions was about 0.05 nm/s, decreasing slightly with increasing temperature.

3. Results and Discussions

3.1. Photo-Luminescence

As expected the indium composition x(In) increases with T_{QW} lowering, Figure 1. We have found that the indium content increase is linear in T_{QW} range from 790°C down to 730°C. Further drop of the T_{QW} down to 710°C leads to a significantly smaller increase of indium composition (cf. the experimental data for 710, 720°C and the linear fit of data on 730-790°C). We derived these values assuming fully strained QWs in our XRD model, which might not be the case for the highest indium content QWs in this series.

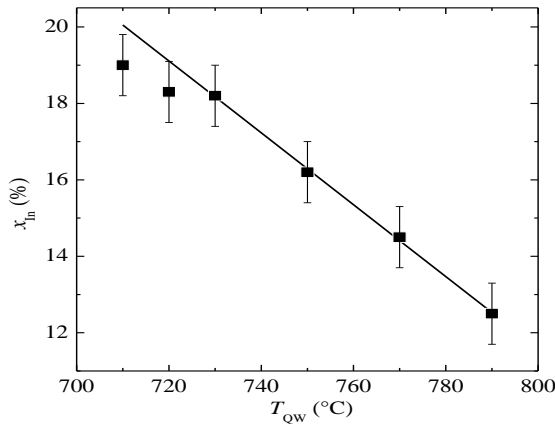


Figure 1: QW indium content as a function of TQW determined from XRD data.

The room-temperature (RT) PL spectra of the samples consist of an AlGa_xN band at 265 nm, a wide deep-level-defect band located at 410 nm and a QW-related band, for which peak position depends on indium composition, Figure 2.

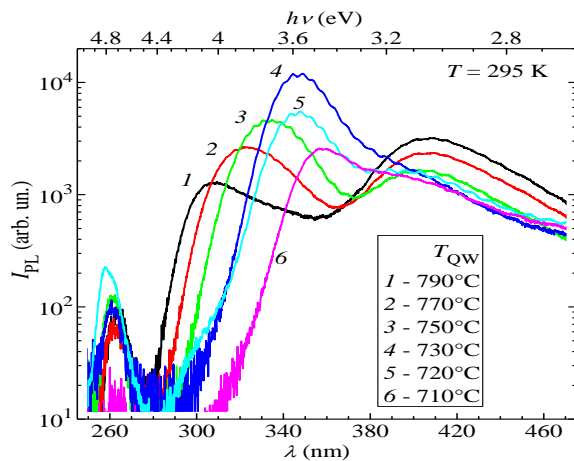


Figure 2: RT PL spectra of InAlN/AlGa_xN MQWs grown at different TQW.

3.2 Band-gap and bowing parameter

By extracting bowing parameter $b(x)$ from $E_g(x)$ using standard expression (1) and endpoints $E_{g,InN}$ and $E_{g,AlN}$ for pure AlN and InN specified in Figure 3, and combining our data to values from literature [14-18] we managed to obtain expression for InAlN band-gap bowing parameter:

$$b(x) = \frac{25.5}{\left(1 + \left(\frac{x}{0.0075}\right)^4\right)^{0.161}}$$

and thus for band-gap itself describing well data in the whole range of compositions.

$$E_g(x) = x E_{g,InN} - b(x) x (1 - x) + (1 - x) E_{g,AlN} \quad (1)$$

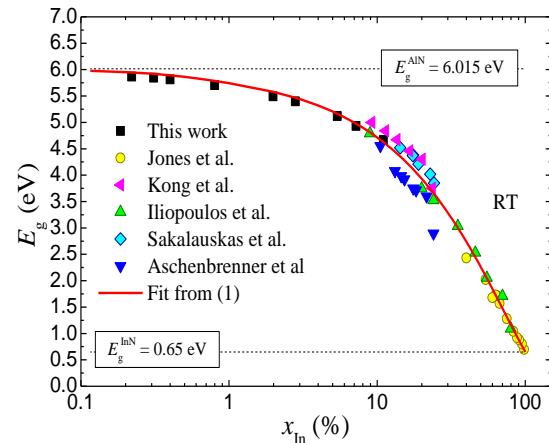


Figure 3: InAlN band-gap as a function of In content.

4. Summary and Conclusions

In summary, we reported on the optical properties of 80-120 nm thick $In_xAl_{1-x}N$ epitaxial layers prepared on AlN templates with low In content. Using the band as a probe for photoluminescence excitation spectroscopy a detailed determination of the variation of the $In_xAl_{1-x}N$ effective band-gap with indium content in the range $0.0022 < x < 0.11$. The band-gap was found to decrease rapidly with indium content resulting in $In_xAl_{1-x}N$ band-gap bowing parameter of above 25 eV in the $x \rightarrow 0$ limit. By introducing bowing parameter formula, we are able to calculate band-gap of InAlN alloy at the full range of composition. Also we were able to get the near UV emission which proposing new material for light emitting applications like LEDs and LDs.

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References

- [1] V. Z. Zubialevich, T. C. Sadler, D. V. Dinh, Shahab. N. Alam, H. Li, P. Pampili, P. J. Parbrook, "Enhanced UV luminescence from InAlN quantum well structures using two temperature growth", *J. Lumin.*, vol. 155, pp. 108–111 (2014).
- [2] M.R. Krames, O.B. Shchekin, R. Mueller-Mach, G.O. Mueller, L. Zhou, G. Harbers, M.G. Craford, *J. Disp. Technol.* 3 (2007) 160.
- [3] T. Gessmann, E.F. Schubert, *J. Appl. Phys.* 95 (2004) 2203.
- [4] M. Kneissl, T. Kolbe, C. Chua, V. Kueller, N. Lobo, J. Stellmach, A. Knauer, H. Rodriguez, S. Einfeldt, Z. Yang, N.M. Johnson, M. Weyers, *Semicond. Sci. Technol.* 26 (2011) 014036.
- [5] M. Matloubian, M. Gershenson, *J. Electron. Mater.* 14 (1985) 633.
- [6] S. Keller, S.P. DenBaars, *J. Cryst. Growth* 248 (2002) 479.
- [7] H. Hirayama, Y. Enomoto, A. Kinoshita, A. Hirata, Y. Aoyagi, *Appl. Phys. Lett.* 80 (2002) 1589.
- [8] T. Wang, Y.H. Liu, Y.B. Lee, J.P. Ao, J. Bai, S. Sakai, *Appl. Phys. Lett.* 81 (2002) 2508.
- [9] M. Kneissl, Z. Yang, M. Teepe, C. Knollenberg, N.M. Johnson, A. Usikov, V. Dmitriev, *Jpn. J. Appl. Phys.* 45 (2006) 3905.
- [10] Y. Sakai, T. Egawa, *Jpn. J. Appl. Phys.* 48 (2009) 071001.
- [11] S. Chichibu, T. Azuhata, T. Sota, S. Nakamura, *Appl. Phys. Lett.* 69 (1996) 4188;
- [12] S. Chichibu, T. Azuhata, T. Sota, S. Nakamura, *Appl. Phys. Lett.* 70 (1997) 2822.
- [13] J.-F. Carlin, M. Ilegems, *Appl. Phys. Lett.* 83 (2003) 668.
- [14] A. Dadgar, F. Schulze, J. Bläsing, A. Diez, A. Krost, M. Neuburger, E. Kohn, I. Daumiller, M. Kunze, *Appl. Phys. Lett.* 85 (2004) 5400.
- [15] R. E. Jones, R. Broesler, K. M. Yu, J. W. Ager, E. E. Haller, W. Walukiewicz, X. Chen, and W. J. Schaff, "Band gap bowing parameter of $\text{In}_{1-x}\text{Al}_x\text{N}$ ", *J. Appl. Phys.*, vol. 104, art. 123501 (2008);
- [16] E. Iliopoulos, A. Adikimenakis, C. Giesen, M. Heuken, and A. Georgakilas, "Energy bandgap bowing of InAlN alloys studied by spectroscopic ellipsometry", *Appl. Phys. Lett.*, vol. 92, art. 191907 (2008);
- [17] E. Sakalauskas, H. Behmenburg, C. Hums, P. Schley, G. Rossbach, C. Giesen, M. Heuken, H. Kalisch, R. H. Jansen, J. Bläsing, A. Dadgar, A. Krost and R. Goldhahn, "Dielectric function and optical properties of Al-rich AlInN alloys pseudomorphically grown on GaN", *J. Phys. D: Appl. Phys.*, vol. 43, art. 365102 (2010);
- [18] T. Aschenbrenner, H. Dartsch, C. Kruse, M. Anastasescu, M. Stoica, M. Gartner, A. Pretorius, A. Rosenauer, T. Wagner, and D. Hommel, "Optical and structural characterization of AlInN layers for optoelectronic applications", *J. Appl. Phys.*, vol. 108, art. 063533 (2010);
- [19] W. Kong, A. Mohanta, A. T. Roberts, W. Y. Jiao, J. Fournelle, T. H. Kim, M. Losurdo, H. O. Everitt, and A. S. Brown, "Room temperature photoluminescence from $\text{In}_x\text{Al}_{1-x}\text{N}$ films deposited by plasma-assisted molecular beam epitaxy", *Appl. Phys. Lett.*, vol. 105, art. 132101 (2014).