GaN based blue-LED

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***Abstract*— In this study we shortly introduced importance of the invention of GaN based blue-LED, which will forever change the idea of solid state lighting. Moreover, we discussed about the theoretical aspects regarding the devices. The bottleneck in this technology is the growing of high quality and cost effective GaN wafers. We thoroughly described traditional hetero-epitaxial GaN fabrication techniques and finally discussed on more recent homo-epitaxial methods to grow bulk GaN wafers. Furthermore, we introduced some different applications related to blue-LED's and finally provided some aspects related to the future trends and perspectives of the technology.**

I. INTRODUCTION

First LEDs date from the late 50's to the early 60's. The first LEDs were near-infrared emitting GaAs type of tunnel diodes, discovered by James R. Biard and Gary E. Pittman. The problem was that they didn't emit visible light. The first properly bright LEDs were invented in the 1970's, and they were designed for fiber communications. The first commercial LEDs were expensive, costing around 200 dollars a piece, but already in the 70's the price was down to about five cents each. Modern blue LEDs can be bought from amazon for around 40 cents a piece.

Since LEDs emit light in certain wavelengths, depending on the doped material, higher energy gaps were harder to create than ones with lower energy. Red and green LEDs were invented decades before the blue LED, since it was harder to create the gap needed for a blue light, and the technology required for manufacturing them wasn't there yet. Different material combinations have been used, but modern blue LEDs are manufactured using indium, gallium and nitride. By controlling the amount of indium and gallium, different kinds of efficiencies and wavelengths of blue can be achieved. By adding aluminium, it is even possible to manufacture UV-LEDs.

The inventors of the blue LED won the 2014 Nobel prize in physics for their work, which tells us of how great of an importance they are. White light can be produced using blue, red and green LEDs. Since LED's can produce about 300 lumens per watt, compared to a fluorescent light which produces 70, we could save vast amounts of energy by replacing all lighting with LEDs.

II. THEORETICAL BACKGROUND

The operation principle of LEDs is based on the semiconductor PN-junction. A PN-junction in a semiconductor is interface between two semiconductor regions of different doping. p-side is doped with acceptor impurities such as Boron which have one less valence electron compared to the semiconductor valence shell and N-side is doped with donor impurities such as Phosphorus which have one extra electron on valence shell compared to the semiconductor. P-type semiconductor regions have a deficiency of electrons on valenece band and acceptors leave an electron void or hole in the lattice that can be considered as a positive charge carrier. n-type semiconduntors have a surplus of electrons on conduction band and donors leave an extra electron to move around in the lattice. A PN-junction can be formed when p- and n-type semiconductors come into contact for example by ion implating n-type region into p-type substrate. [1]

When electrons and holes diffuse across the junction and recombine, a positively charged region is left on n-side as the impurities there lose the electrons and a negatively charged region is formed on the p-side as electrons recombine to holes near formerly neutral atoms. This generates an electric field that opposes the diffusion and equilibrium is formed. The charged regions together form a depletion region that has (nearly) no free charge carriers. The bending of energy bands also leads to formation of a potential barrier called built-in potential, leaving n-type side at higher potential. Band diagram of a PN-junction can be seen in Figure 1.[1]

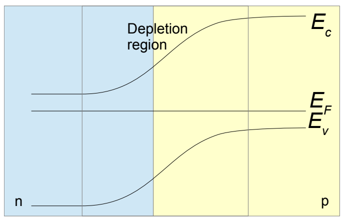

Fig. 1: PN-junction band diagram describing conduction band energy, fermi level and valence band energy.

PN-junction is considered forward biased when higher potential is applied to p-side of the junction than n-side, and reverse biased when n-side is at higher potential. In general, current may flow only when the junction is forward biased and only a small drift current exists while in reverse bias, unless the reverse voltage exceeds the junction's breakdown voltage. [1]

LED is essentially a forward biased PN-junction. The wavelength emitted by the LED depends on the bandgap of the semiconductor. Carriers are provided by applying a forward bias over the PN-junction allowing current to flow and injection electrons and holes into the junction where they may recombine and release a photon. LEDs in general require a direct bandgap semiconductor. In direct bandgap semiconductor, the conduction band and valence band energies (which are not just flat lines but curved) attain the minimum and maximum value at the same value of wavevector, respectively. Due to this, the radiative recombination of an electron-hole pair is more likely to occur than in indirect bandgap semiconductor where minimum and maximum values are not attained at the same value of wavevector and where interaction with phonon is also required. [1]

A usual PN-junction described earlier is homojunction because both sides are of the same material. A way to improve the efficiency of a LED is making a double heterojunction. In double heterostructure junction a semiconductor layer either intrinsic or possibly doped, is surrounded by two layers of another semiconductor material with a different bandgap than middle region, with one layer p-type and the other n-type so that two interfaces of different materials are formed. The structure of heterojunction is described in Figure 2. In general the middle layer (called active area) has a lower bandgap than the surrounding semiconductor. In this way the charge carriers injected into active layer can be confined by the potential barrier. If the active layer is very thin, it may be called quantum well. The configuration also reduces reabsorption because the active layer has lower bandgap and photons will have lower energy than the bandgap of surrounding semiconductor and acts as a window for light emitted due to recombination in active area, i.e. is transparent at that wavelength. [2]

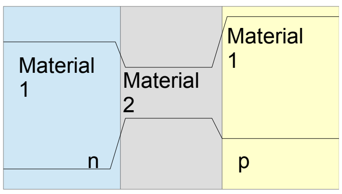

Fig. 2: Heterojunction structure in which material 2 has a lower bandgap than material 1.

Typically or often LEDs are manufactured from heterojunctions of compound semiconductors, i.e. semiconductors that contain at least 2 different elemental semiconductors. Binary compound contains 2 elements and ternary compounds contain three. Important materials are gallium arsenide GaAs based ternary compounds, gallium phosphite GaP and indium. Often the compound semiconductors consist of group III and V elements. An important aspect of compound semiconductors is that the bandgap can be manipulated by changing the relative amount of some elements in the compound. Usually in ternary compounds the ratio of two elements is varied. [1] [2]

Blue LED can be manufactured from silicon carbide (SiC) which has 2.86 eV bandgap. However, the bandgap is indirect and the external quantum efficiency in addition to luminuous efficiency are low for SiC blue LEDs. An alternative material used in construction of blue LEDs is binary compound semiconductor gallium nitride, GaN, which has a direct bandgap and ternary compounds indium gallium nitride InGaN and aluminum gallium nitride AlGaN. GaN does not match substrate materials well. Sapphire has (one of) the closest and is used as substrate to grow GaN, but mismatch is still significant. Lattice mismatch produces defects in the crystal and reduces LED efficiency. A blue LED fabricated by the Nobel prize winner Shuji Nakamura extensively used GaN compounds and used InGaN as active layer. The bandgap of InGaN is manipulated by changing the relative amounts of indium and gallium. Nakamura's LED contains double-heterostructure. Active layer was surrounded by n- and p-type AlGaN layers. N-type and p-type GaN layers had been used for contacts before AlGaN layers. This LED also used sapphire as substrate and also a buffer layer of GaN before other layers. [1] [2] [3]

III. Manufacturing

Manufacturing of blue-LED's is quite straight forward task since it can be manufactured using quite standard semiconductor fabrication techniques. However, the substrate material used in these devices, i.e. gallium nitride (GaN) was difficult task to grow. Basically it took several thousand trials to even grow a single cystalline GaN (c-GaN), which had no cracks and didn't have too high dislocation densities. These difficulties arises from the fact that compound semiconductors, e.g. especially III-V semiconductors cannot be grown using typical Czochralski process used in Silicon growth. Therefore, more demanding processes, i.e. epitaxial methods have been developed [4]. Moreover, doping of GaN was the most difficult task in fabricating a PN-junction based blue-LED. However, nowadays the doping is not an issue, GaN can be doped with Mg for p-type GaN and with Si for n-type [5].
In this section we are going to introduce few common hetero-epitaxial methods used in today's industry of blue-LED's for lateral LED structures. Furthermore, we will discuss on Bulk GaN fabrication since it provides superior quality, thus, better performance over hetero-epitaxially fabricated counterparts.

Hetero-epitaxial methods have been around for several decades to fabricate compound semiconductor wafers on top of a foreign substrates, such as silicon, silicon carbide and sapphire. Today most of the LED's are fabricated on top of sapphire, since it is the most cost effective substrate to fabricate GaN on, however a lattice mismatch between foreign substrates and GaN will cause difficulties due to dislocations, therefore, novel methods for fabrication of GaN have been developed extensively.

Metal-Organic Vapour Phase Epitaxy (MOVPE) or Metal Organic Chemical Vapour Deposition (MOCVD) is the mostly used hetero-epitaxial method to fabricate GaN typically on top of either sapphire or silicon substrate. In MOVPE, source materials are metal-organic materials in which a weak bond between metal and organics is broken at temperatures above 400 °C. Therefore, normal growth temperatures are in range from 500 to 800 °C, causing MOVPE to be diffusion limited. Growth rate is approximately $1\,\mu m/h$, which makes it reasonable for growing of quantum structures. MOVPE is also cold wall reactor technique decreasing contamination by parasitic reactions on reactor walls. In MOVPE growth can be monitored in real-time, which provides a cutting edge against many other techniques. [2]
However, MOVPE is expensive and hazardous due to the toxic gases used in this method. Moreover, MOVPE can provide poor quality crystals due to lattice mismatch between foreign substrates and imperfections especially on the surface of sapphire substrates. High dislocation density is one of the key reasons why higher lumenous flux is difficult to achieve using LED's. In order to improve lumenous flux it is definitely important to improve manufacturing of GaN wafers. Recent studies [6] have shown varying dislocation densities as high as 10^9 cm^{-2} for this method.

Molecular beam epitaxy (MBE) is slower than MOVPE and it requires ultra high vacuum, that takes days to pump when maintenance is required. Of course, MBE reactors have pressurised interlock so that when loading a sample into the chamber the whole reactor is still in ultra high vacuum. In MBE the substrate is heated up to enhance atom diffusion on the surface. However, the walls of the chamber are cooled to trap excess reactants. MBE has also usually mainly solid source materials, that are quite difficult to handle. The growth can be interrupted abruptly by closing shutters front of the effusion cells. Despite many difficulties, MBE has very good thickness control, resulting in sharp mono-atomic interfaces. Growth is limited by the flux of group-III compound, provided that the group-V part is in excess. [2]
MBE suffers from similar issues as its competing counterpart MOVPE when it comes to the dislocation density and finally lumenous flux. However, its advantage is a bit lower dislocation density of varying from 10^6 cm^{-2} and 10^5 cm^{-2}. A short comparison between MBE and MOVPE is collected into Tab I.

TABLE I: Comparison of MOVPE and MBE in GaN growth.

	Pros	**Cons**
MBE	Control of doping	Slower growth
	Good crystalline quality	Expensive
	Composition at the monolayer level	Ultra high vacuum required
MOVPE	Control of doping	Poisonous precursor gases
	Faster growth	C and H fromp precursors are incorporated into layers
	Easier maintenance	
	Widely used in industry	

There is a growing demand of higher quality and more cost effective GaN substrates, however, it seems that the MOVPE and MBE are slow growth rate methods, leading into a higher price for thick wafers. Therefore, those methods cannot be used to improve the LED structures by using vertical structures, such as $100\,\mu m$ thick LED structures. In order to improve the efficiency and luminous flux of the devices it is important to use vertical structures [7]. Due to this fact MOVPE and MBE can only be used to grow lateral structures.

Many research groups and companies have started to improve fabrication techniques for growing bulk GaN wafers either on top of sapphire or even on a GaN seed. These bulk wafers could be used to fabricate superior vertical LED structures, which would eventually increase the power output.

The so called bulk wafer fabrication methods may overcome these issues, one of which is the known as Hydride Vapour Phase Epitaxy or Halogen Vapour Phase Epitaxy (HVPE), which is the mostly used method to grow Bulk GaN. This most promising method has big advantage over MOVPE and MBE, since it can produce high-quality material at high growth rates due to a high surface migration of the halide species, motivating the versatility of HVPE as a growth method for both device applications and substrate application. [8]
The most important growth reaction in HVPE is the reaction between CaCl and ammonia:

$$CaCl + NH_3 \rightarrow GaN + H_2 + HCl, \qquad (1)$$

where GaCl is provided into the system using a solid Ga placed inside the tube and heated to 1000 °C while sulphuric acid HCl is fed to the system as can be seen in Fig. 3, the result is of course GaCl which will then react with ammonia NH_3 as was shown in React. 1.

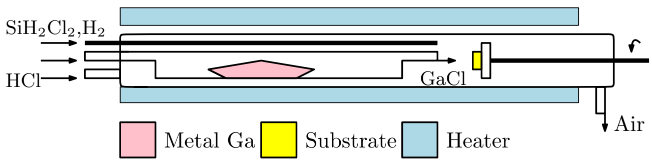

Fig. 3: Basic process of HVPE requires mainly HCl and metallic Ga as source materials. *Reproduced from [7].*

In HVPE one may grow the GaN film either hetero-epitaxially, i.e. on a foreign substrate or homo-epitaxially on a GaN seed. However, in other methods HVPE is usually used to grow high quality seed. The GaN wafers without foreign substrate are called as free standing wafers, which provides superior emission power when compared to their counterparts on sapphire [9]. Growth rate of this method can be as high as $300\,\mu$m/h and the dislocation densities of 10^4 cm^{-2} can be achieved[7]. However, it is known that the so called Threading Dislocation Density (TDD) increases as a function of thickness, therefore the average of TDD is mostly below 5×10^6 cm^{-2} [7]. However, the problem with this method is that it tends to crack if the growth rate is increased. [9] Currently the record of thickest GaN wafer has a thickness of roughly $500\,\mu$m [8], which was manufactured using a certain crystalline quality improvement technique known as Epitaxial lateral overgrowth (ELOG) technique. However, this sample was grown on GaAs substrate, which is expensive when compared to sapphire.
Another huge problem is related to the wafer bending or bowing discussed in [7], [9], [8], [10]. Wafer bowing is known to be a problem when aligning the mask in lithography process of especially sub-micron linewidth structures [11] but may become a problem in even more conservative linewidths. Moreover, the when the diameter of the wafer is increased the wafer bending becomes evident. [7] It must also be denoted that a Japanese company Sumitomo Electric has stated that they have produced even 6-in wafers in diameter using HVPE.

Ammonothermal method is similar to solvothermal synthesis used to synthesize quartz and it can be used to grow high quality true bulk based on a supercritical ammonia NH_3 as solvent for Ga. Additionally some mineralizing agents may be used with ammonia. [12] In ammonothermal method polycrystalline GaN powder is heated roughly to 400 °C degrees in a supercritical ammonia atmosphere of 2000-3500 atm [13], [7]. Due to this supercritical nature of ammonia it is evidently hazardous to control this type of method safely. This method provides lower TDD of even 10^3 cm^{-2} [7] and 10^4 cm^{-2} [14] have been reported. The ammonothermal growth is conducted by transporting the mass from high- to low- temperature zones and has been reported to possess several advantages, including lower dissolution density due to solubilizing in the higher temperature zone using a baffle between the zones. In Fig. 4 one may see the poly-cystal GaN area as the low temperature zone and the area behind the baffle the high temperature of roughly 500 °C zone. Ammonothermal method is in principle a low growth rate (ca. $20\,\mu$m/h) method but can produce high quality bulk GaN with no wafer bow issues or bending issues except some issues if HVPE seed is used [12].

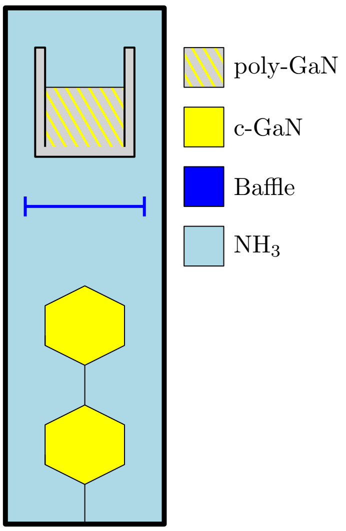

Fig. 4: In ammonothermal growth poly crystalline GaN powder is heated to roughly 400 °C in a pressurized super critical ammonia solution, from which the single crystalline GaN (c-GaN) is grown. *Reproduced from [7].*

Apart from these methods there is a new arising fabrication method for high quality GaN known as sodium (Na) flux method where Ga-Na melt is placed on top of HVPE or MOVPE grown seed layer on sapphire. This crucible containing the melt and the seed wafer is placed into a reactor where temperatures are elevated to roughly 800 °C degrees. Nitrogen gas is vented into this reactor in order to obtain GaN synthesis [15]. This method have been improved using so called necking technique, which has been used in Czochralski process for decades to diminish grain boundary dislocations. In this technique the growth is constrained to a small aperture on top of the GaN seed [16]. This technique has decreased the TDD to 10^3 cm^{-2} while the seed had TDD of 10^9 cm^{-2} [17]. Reason behind the drastic decrease of TDD in Na-flux method is not yet known . The high quality GaN crystals produced by this method seem promising. However, Na-flux method has not yet been used to grow large diameter wafers but rather small crystals of the size 10 mm x 10 mm x 10 mm [7]. Nevertheless, the method seems promising for the future of low cost and high quality wafers.

IV. Applications

A. *LED vs. Other Illumination Methods*

The main application of LEDs which are a solid-state light source is, of course, illumination. Compared to the first massively produced incandescent lamp or later fluorescent lamps, LED light source has a lot of benefits.

Incandescent lamp uses a spiral, heated by electric current. Its radiation is close to one of a black body, so there is a theoretical limit for their efficiency. Given a commonly used Tungsten filament heated up to 3000 K, using black body illumination expressions, it is possible to calculate theoretical limit of efficiency as 17 lm/W whereas LED sources do not have such theoretical limit and by 2005 their efficiency already reached 425 lm/W.[18].

Fluorescent lamp is more convenient compared to incandescent lamp, but still less efficient than a LED. Due to energy loss during conversion of a 250 nm UV photon from Mercury arc discharge to visible light, these lamps' efficiency is limited to only 90 lm/W.

Moreover, in [19] environmental issues are emphasized. Usage of solid-state light sources will allow to decrease worldwide illumination power consumption by 50%. Because of higher efficiency, LEDs can reduce greenhouse gases emission, fuel consumption and mercury pollution coming from recycling of fluorescent lamps.

B. From Blue LED to White Light

Obviously, blue wavelengths are not useful for everyday illumination purposes, hence ways of conversion blue to daylight are suggested. For each of possible solutions, there are two main parameters – efficiency and color rendering index (CRI) which is the the ability of a light source to show the true colors of an object. Different methods of achieving white light are a compromise between efficiency and CRI.

Applications in everyday illumination (in homes, offices, displays etc.) do not require high CRI, but being the most common, require highest possible efficiency. For these cases it is possible to use blue LEDs with phosphors. The most common phosphor for this purpose is Ce-doped YAG: $Y_3Al_5O_{12} : Ce^{3+}$. Ce-doped YAG absorbs blue radiation from the diode by allowed $4f^1 \rightarrow 5d^1$ transition and emits yellow light via the reverse $5d^1 \rightarrow 4f^1$. The emission from the lowest excited level is spin–orbit split $4f^1$ ground states, leading to an extremely broad emission band with a FWHM > 100 nm. The yellow emission from YAG:Ce phosphor is afterwards combined wirh transmitted blue radiation originating from the diode itself. Such a mixture results in relatively white light (CCT > 4000 K) and yields reasonable color rendering index (CRI of 70 – 80) and efficiency of up to 425 lm/W [18], enabling this technology to be used in many applications where color rendering requirement is not as important as efficiency[20].

Such a solution, despite its high efficiency and robustness, lacks red component in the light, which makes it appearing less natural for human eye. There are other phosphors suggested based on *Nitride, Oxynitride, Oxide, Oxyhalide*, and *Halide* phosphors. Some of these have a possibility to replace Ce:YAG in future.

Another way of creating white light are tri- and tetrachromic approaches. These provide a better CRI suitable for any applications, including even artistic lights. Composition of these solutions are shown in Fig. 5.

This method implies placement of different colored LEDs in close spatial proximity, so that naked eye cannot distinguish them. Such composition allows to modify not only the brightness of light source, but also its spectral compounds, i.e. color. This gives a possibility of artistic lights and even fully LED displays where single pixel is represented by a trichromatic light source discussed above. For example, color changing is widely used by Philips® in their *Ambi-Light* technology.

White LED sources based on energy-efficient blue LEDs are now commonly used in street illumination, automotive industry (headlights and interior illumination) and other illumination purposes. The greatest application benefit of LEDs is, apart from high energy efficiency, large variety of shapes for different solutions. Such sources can be combined in a stripe to make a long source, be placed in any light bulb package to ensure easy integration.

V. Future Perspectives

Main purpose of GaN LEDs is to replace older and less energy efficient light sources like incandescent or fluorescent lights. However, unlike its predecessors, LED technology has a great capability of high-frequency modulation. This

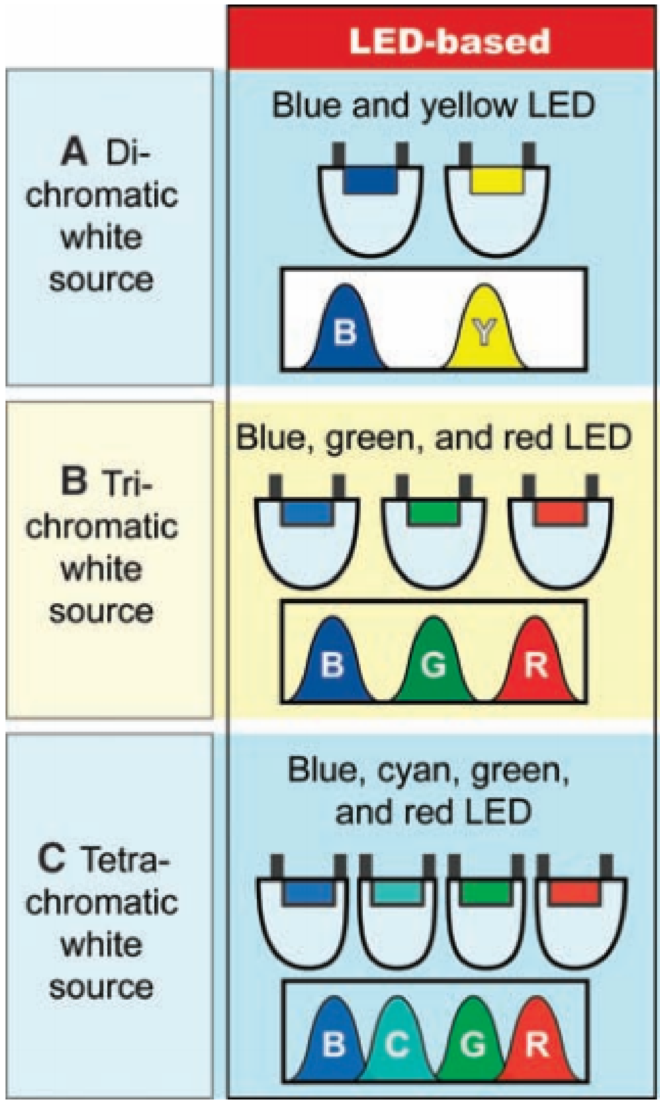

Fig. 5: LED-based approaches for white light sources implemented as di-, tri-, and tetrachromatic sources. Highest luminous source efficiency and best color rendering are obtained with dichromatic and tetrachromatic approaches, respectively. Trichromatic approaches can provide very good color rendering and luminous source efficiency. *Reproduced from [18].*

allows the diodes in future to be utilized not merely as light sources, but also implement high-frequency signalling unrecognisable by human eye. This technology can provide area for device communication with environment in a "smart house", communication of cars with each other and road objects (traffic lights, road signs etc.) and various application in artistic light.

VI. Conclusions

In this report we described the need for blue LEDs in a modern world. Moreover, shortly described theoretical background behind blue-LEDs especially the main principle, PN-junction, and its meaning in the operation of LEDs. It is concluded that currently GaN based compound semiconductors are very important and probably the best alternative in blue LED manufacture. It is important to denote that there at the moment MOVPE based GaN on sapphire blue LED's are too expensive and their luminous flux is not that good, therefore ammonothermal or HVPE or even Na-flux method will overcome these issues in the near future to provide cost effective and high quality freestanding bulk GaN wafers for LED fabrication. Illumination solutions based on discussed LEDs are predicted to reduce power consumption and open large amount of possibilities in future.

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