STUDY OF PLD GROWN ZnO FILMS FOR LIGHT EMITTING APPLICATIONS

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By

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SYNOPSIS

In the past few decades, the energy demand is growing at a rapid pace due to a surge in population and economic growth [1]. Searching for an efficient, light and un-extinguishable energy source is prime hunt for human from centuries and if the source is green, then it has a little impact on nature and the environment. So, the energy issue should be addressed scientifically. In 1970s, we faced a serious energy crisis due to substantial petroleum shortage and our whole economy collapsed. The crisis led to stagnant economic growth in many countries as oil prices climbed. The energy crisis in the future can be avoided by searching for new sources or by exploiting untouched resources. Moreover, other alternate may be the exploitation of more efficient gadgets, turning off gadgets when they are not in use, utilizing public transport and by much more energy-saving things.

In the profile of energy resources in India, coal has a dominant position; it constitutes about 51% of India’s primary energy resources followed by oil (36%), natural gas (9%), nuclear (2%) and hydro (2%). The utility electricity sector in India had an installed capacity of 278.734 GW, which consumes about 72% of the coal produced in the country [2-3]. Still, in India, there are around 300 million people having no access to electricity. Currently, in India, lighting accounts for approximately 30 percent of total residential electric power use, followed by refrigerators, fans, electric powered water heaters, and TVs. Electric lighting burns up to 25-30% of the average home energy budget. Light-emitting diode (LED) and compact fluorescent lights (CFL) bulbs have revolutionized energy-efficient lighting. LEDs are solid light bulbs, which are a more superior source of light due to its lower energy consumption (~100 lumens/watt), longer lifetime, smaller size, faster switching, etc. [4-6]. A lot of research is going on solid-state lighting from 1990s [7]. In 2014, Nobel prize in physics was awarded in the field of LEDs to “Isamu Akasaki” (Meijo University, Nagoya, Japan and Nagoya University, Japan), “Hiroshi Amano” (Nagoya University, Japan), and “Shuji Nakamura” (University of California, Santa Barbara, CA, USA) for searching the possibility of fabrication of blue LED. LED is a centre of research for communities and industries due to its commercial advantage and large possibilities. The vendors at the Lagpat Rai market in Delhi, India, which was popular for their electronic goods, and spares have fully upgraded their shops to LEDs because of broad opportunity, great margin, and future scope.

The first solid state LED was fabricated on SiC material system, which is an indirect band gap semiconductor [8]. The efficiency of SiC based LED is quite small (0.005%.). After that, different colored LEDs were fabricated from GaAs and GaN materials by doping [9-12]. Bright blue, white and green LEDs were fabricated from GaN [13-15]. GaN is a wide and direct band gap semiconductor having high thermal conductivity, high peak electron velocity, high saturation electron velocity, intense breakdown field, good electron and hole mobility, very hard, mechanically stability, high-heat capacity for heterojunction etc., which makes it an excellent candidate among all the semiconductors for fabricating high power and efficient optical devices [16]. One of the interesting properties of GaN and its compound semiconductors is that it covers the whole visible spectrum. The band gap of GaN can be varied from 0.8 eV to 6.2 eV by In and Al compounding. The GaN is low sensitive to ionizing radiation, which makes it suitable for solar cell arrays for satellites. All the properties discussed above, make GaN a convenient candidate for fabrication of LED [17]. The internal quantum efficiency of GaN LEDs rises with an increase of current density up to a certain limit, above that, the efficiency decreases, which is called "Efficiency Droop". This "efficiency droop" is a major problem in GaN LED chips, which inherits its operation at high-current [18]. The origin of efficiency droop is still notorious. Auger recombination, piezoelectric polarization, electron overflow, non-radiative recombination, dislocation density, defects, etc. are considered as major causes responsible for efficiency droop [19-20]. Further, the higher cost, maintenance, sophisticated/expensive growth, toxic nature are other challenges faced by researchers and industrialist.

In the present work, the problem of efficiency droop in GaN LEDs was investigated by TCAD. The problem of efficiency droop in GaN LEDs cannot be eliminated completely, but only reduced. The efficiency droop was reduced by 25% by polarization doping in quantum wells [21-23]. To overcome this problem completely in GaN LEDs, an alternative scheme is required. Simulation results proved that pure ZnO and hybrid GaN/ZnO based LED with Cd and Al doped ZnO shows no efficiency droop. So, ZnO can be used as an alternate material for fabrication of LED. ZnO has similar hexagonal wurtzite crystal structure and lattice constants as of GaN material. ZnO is a low cost direct band gap (3.34 eV) material, which is easily available and environmental friendly. The fundamental absorption spectra in ZnO have higher excitonic binding energy (~ 60 meV) then GaN (~25 meV) at room temperature [24]. These properties of ZnO made it an attractive material for the optoelectronic devices and applications like laser diode, LED, quantum well devices, etc. [25-26]. The optimized simulated hybrid LED structure is shown in figure 1.

p-Contact
p-GaN (500 nm)
p-AlGaN (30 nm)
p-AlGaN (10 nm)
CdZnO (5 nm)
CdZnO (7 nm)
n-ZnO (200 nm)
n-Contact

Figure. 1. Hybrid LED structure

ZnO based thin films can be deposited by molecular-beam epitaxy, metal-organic chemical vapor deposition, RF sputtering, pulsed laser deposition (PLD), sol-gel etc. [27-28]. MOCVD and MBE techniques are most commonly used for the fabrication of optoelectronic and spintronic devices from ZnO. A cost-effective PLD technique is used here to check the possibility of fabricating ZnO based LED. PLD got popularized in 1980, as a quick and reproducible oxide thin film deposition technique. It is a physical vapor deposition technique, in which excimer laser like F2 (λ = 157 nm); XeF (λ = 352 nm); KrF (λ = 248 nm); KrCl (λ =222 nm); ArF (λ= 193 nm) and XeCl (λ = 308 nm) can be used to deposit material on a substrate at a low pressure. KrF (λ = 248 nm) excimer laser at ~ 2 J/cm^2 is used in the present work, as it creates the highest gain system in ZnO [29].

The main aim of the thesis is to reduce the problem of efficiency droop in InGaN/GaN LED. For eliminating the 'droop', a ZnO/GaN based hybrid system is studied. Finally, the possibility of fabricating the hybrid ZnO based LEDs from PLD technique was deliberated. Brief sketch of our proposed thesis is as follows:

Chapter I: This provides a brief introduction to the current energy scenario. In India, approximately 30 percent of total residential electricity is used for lighting. Incandescent bulbs, CFL lamps are commonly used as a lighting source in India. Due to government initiatives, from the last few years, solid state lighting devices (LEDs) were introduced in India to reduce energy budgets at residential and commercial buildings, by cutting the demand by 20%. These LEDs are superior source of light due to longer lifetime, lower energy consumption and smaller size. GaN shows a promising material for solid state lighting with an efficacy greater than 150 Lumens/Watt. In this chapter, material properties, band structure and applications of GaN have been discussed. In addition to this, an alternate candidate, ZnO and its material properties, band structure and growth has also been included.

Chapter II: In the second chapter, a brief overview of experimental techniques is given. ZnO based thin films can be deposited by MOCVD, MBE, PLD, DC sputtering, ion beam irradiation technique and even by sol gel method. Out of that, MOCVD and MBE techniques are most commonly used for fabrication of optoelectronic device. This chapter also introduces various characterization techniques used in the present research work viz., X-ray diffraction (XRD), Raman spectroscopy, photoluminescence spectroscopy, atomic force microscopy (AFM), UV-Visible, photoluminescence, X-ray photoelectron spectroscopy (XPS), valence band spectroscopy (VBS), soft X-ray absorption spectroscopy (XAS) and Hall measurement.

Chapter III: In this chapter, the basic Physics of quantum well LEDs are discussed. Different types of recombination models like radiative recombination, Auger recombination, Shockley-Read-Hall recombination etc. are discussed here. 'Efficiency droop' is a major problem faced by researcher in InGaN/GaN LEDs. Auger recombination, piezoelectric polarization or field, non-uniform distribution of holes in quantum wells, dislocations, junction heating and electron overflows over active layers are some of the major causes responsible for the problem of efficiency droop. ATLAS from M/s Silvaco has been used to study the droop problem in GaN LEDs. By polarization doping, the quantum wells and electron blocking layers, an 25% improvement in internal quantum efficiency have been observed. GaN is an expensive material and is very difficult to grow and LEDs fabricated from this material system show efficiency droop. ZnO can be used as an alternate option with similar lattice parameters, hexagonal wurtzite structure, direct and wide band gap (3.37 eV). From the analysis, it has been concluded that ZnO/GaN hybrid LED system show no efficiency droop. The results of this investigation play lead role in designing a droop free LED system for solid state lighting.

Chapter IV: In this chapter, Mg doped ZnO (MZO) thin films have been studied. $Zn_{0.85}Mg_{0.15}O$ thin films were deposited on quartz substrate by pulsed laser deposition at different oxygen partial pressures and substrate temperatures. XRD results revealed that deposited films were textured along c-axis and maintain wurtzite crystalline symmetry. Increase in grain size, optical band gap energy and emission intensity were noticed with an increase in oxygen partial pressure and substrate temperature up to 1mTorr and 500 °C, respectively. Above these values, the grain size, bandgap and emission intensity were decreased due to the segregation of oxygen and accumulation of defects at grain boundaries.

Chapter V: In this chapter, Cd doped ZnO films were studied to see the possibility of fabrication of quantum wells for LED structure. Thin films of $Zn_{1-x}Cd_xO$ and $Zn_{1-x}Cd_xO/ZnO$ hetero-junctions were deposited on glass substrate by pulsed laser deposition technique. XRD results confirm that deposited films were oriented along c-axis and contain wurtzite crystalline symmetry. The grain size decreases with increasing Cd concentration. The valence and conduction band offsets of hetero-structures were estimated by X-ray photoelectron, valence band, and UV-visible spectroscopy. Type-II band alignment (staggered gap) with ratios of conduction band to valence band offsets ($\Delta E_C/\Delta E_V$) was found to be 0.77 and 0.59 for $Zn_{0.95}Cd_{0.05}O/ZnO$ and $Zn_{0.9}Cd_{0.1}O/ZnO$ hetero-structures, respectively, which can be used in longer wavelength regime optoelectronic devices. The higher value of valence band offset as compared to conduction band offset suggests that the transport at interface is mainly due to electrons.

Chapter VI: Group V elements like N, P, As, Sb and Bi are usual p-type dopants for ZnO. Among these, Sb is considered as one of the most important and active materials revealing venerable properties towards the achievement of efficient p-type doping. So, in the **present chapter**, Sb doped ZnO thin films have been studied. PLD grown un-doped ZnO films show n-type behavior, due to large oxygen vacancies and defects, so, it is very hard to get p-type doping with low Sb content in ZnO. 3% and 6% Sb doped ZnO and ZnSbO/ZnO were deposited on glass substrate by PLD technique. Hall effect measurement show p-type behavior of 3% Sb doped films, whereas 6% Sb doped films shows n-type behavior. The valence band offsets at $Zn_{1-x}Sb_xO$/ZnO hetero-interface were measured by photoelectron spectroscopy. The change in band alignment has been observed with the dopant (Sb) concentration. Ratios of conduction band offset to valence band offset were estimated to be 1.67 and 0.04 for x = 0.03 and 0.06, respectively, for Sb doped films. A Type-II band alignment was observed at the $Zn_{0.97}Sb_{0.03}O$/ZnO interface, whereas the Type-I band alignment took place at the $Zn_{0.94}Sb_{0.06}O$/ZnO interface.

Chapter VII: In this chapter, the structural, optical and electronic structure of n-type (Al doped) ZnO thin films grown by PLD were investigated. X-ray diffraction measurements reveal that Al doped thin films are textured along the c-axis and have wurtzite structure. Al doping in ZnO films leads to increase in grain size due to relaxation in compressive stress. Enhancement in band gap of ZnO films with the Al doping is also noticed which can be ascribed to the Brustein–Moss shift. The changes in the electronic structure caused by Al in the doped thin film samples are understood through X-ray absorption measurements. Hall effect analysis show, n-type behavior of the Al doped ZnO films. The carrier concentration in Al doped films increases with Al doping, whereas, mobility decreases with Al doping.

Chapter VIII: Conclusions and future scope of the study are presented in this chapter. Simulation results show, (i) polarization doping can be used to reduce the problem of 'efficiency droop' in GaN LEDs and (ii) in ZnO alone and ZnO/GaN hybrid LED material systems, this problem can be eliminated completely. Consequently, in next step experimental technique is used to check the possibility of ZnO based LED structure by pulsed laser deposition (PLD) technique.

It was concluded that, pure ZnO and hybrid ZnO LED material systems show no efficiency droop, but it is very difficult to grow p-type ZnO and Type I CdZnO films by PLD due to which it was not possible to fabricate a visible LED from ZnO system alone.

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