

A Review On Applications Of Zinc Oxide Nanostructures

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Abstract

The present paper is a critical review of structural and optical, electrical properties along with nanostructure applications based on Zinc oxide. ZnO is an attractive material for applications in electronics, photonics, acoustics, and sensing. The XRD patterns of the nanostructured ZnO thin films showed the hexagonal wurtzite crystal structure that were preferably grown along the (002) or (101) planes. The optical properties, Band gaps, transmittance of films deposited on glass substrates were also studied with the various optical transmittance spectra. The band gap energy of the deposited ZnO thin films found varying from 3.18 to 3.85eV. Annealing of electrodeposited films decreased the band gap considerably. Depending on the crystal size, the transmittance of the ZnO thin film was found to be about 70 to 90%. Among the other promising areas of application for ZnO are acoustic wave devices, due to large electromechanical coupling in ZnO, and devices utilizing nanostructured such as biosensors and gas sensors and solar cells, since it is relatively easy to produce such forms of ZnO nanostructures, which have good charge carrier transport properties and high crystalline quality. Despite the significant progress made, there are still a number of important issues that need to be resolved before ZnO can be transitioned to commercial use.

Keywords: Zinc Oxide, Thin film, Nanostructure, Review.

1. INTRODUCTION

Zinc oxide is an inorganic compound which is a wide band gap semiconductor of the II-VI semiconductor group. This semiconductor is known for its several favorable properties, such as good transparency in the visible and high infrared spectrum, high electron mobility; wide and the direct band gap, large exciton binding energy, high thermal conductivity and strong room temperature luminescence. These properties are used in many applications such as transparent electrodes of thin film solar cells for light transmission and the extraction of photocurrent, heat-protecting windows, transparent oxide thin-film transistors, light-emitting diodes, varistors, piezoelectric devices, etc. Crystalline zinc oxide is thermochromic in nature

i.e. changing from white to yellow colour when heated and return to the earlier state on cooling [1].

Zinc oxide is a technologically important inorganic compound with the formula ZnO. Zinc and oxygen are the elements of group II and VI with atomic numbers 30 and 08 and atomic weights 65.382 and 15.999 respectively. In the stable form, zinc oxide takes a hexagonal wurtzite crystal structure with lattice parameters $a = 3.2458\text{\AA}$ and $c = 5.2006\text{\AA}$ ($c/a = 1.6022$). The other two forms of ZnO are zinblende and rock-salt structure. The zinblende structure can be stabilized only by the growth of ZnO on the cubic lattice substrates and the rock salt structure may be obtained under very high pressure about 10GPa and return to the former state of hexagonal wurtzite structure on decompression [2]. ZnO has wide direct bandgap energy of 3.40 eV at room temperature and large exciton binding energy ($\sim 60\text{meV}$). This high binding energy ($\sim 60\text{meV}$) assures efficient exciton emission at room temperature under very low excitation energy. ZnO materials show an n-type electrical conductivity due to its native or intrinsic defects such as oxygen vacancies and interstitial zinc atoms. Intrinsic zinc oxide thin films are highly resistive in nature, but when commonly doped with Group III elements such as Ga, In or Al, they become conducting [3].

In many respects, ZnO is considered to be an alternate to GaN for device applications owing to its relatively low production cost and superior optical properties. However, reproducible and stable p-type doping remains to be the most daunting obstacle to producing bipolar ZnO-based devices. It is a given that GaN device technology is much more mature as GaN-based very high-performance electronic and optical devices have already been commercialized. GaN-based power field effect transistors (FETs) are capable of producing over 800 W of continuous-wave (CW) power in the communication band, light-emitting diodes (LEDs) have created a large market with emphasis in performance being on the efficiency although they are already more efficient than even the fluorescent bulbs, and blue lasers are already used in game consoles and high definition video players. The same, however, cannot yet be said about ZnO particularly when its potential applications overlap considerably with those of GaN. On the electronic side, the relatively low mobility of ZnO as compared to GaN and nearly four times stronger electron–phonon coupling together with relatively low thermal conductivity are serious shortcomings for ZnO. However, transparent thin film transistors (TFTs) built from poly-ZnO appears to hold some potential. Furthermore, the potential worldwide shortage of indium in the face of growing demand for indium tin

oxide (ITO) seems to be stimulating the exploration of ZnO-based transparent oxides, which if successful, could become a huge application area. [4-6].

On the optical device front, ZnO needs to show high p-type conductivity along with high quality heterojunctions for realization of competitive light emitters. One major advantage of ZnO is its high 60-meV+ exciton binding energy compared to 25 meV of GaN. If lasers utilizing excitonic transitions were to be built, ZnO would have an advantage over GaN provided that p-type conductivity is obtained and other necessary processing capabilities are developed for ZnO. Another promising application area of ZnO is acoustic wave devices owing to its large electromechanical coupling, particularly along the c-direction. Further yet, ZnO appears to be well suited for producing nanostructures, which may be used for devices. A significant part of the recent research in the field of ZnO-based devices and applications deals with ZnO nanostructures (nanowires, nanobelts, etc.) and their integration with the mainstream semiconductor materials V such as Si, GaN, and organic semiconductors. ZnO nanowires have attracted a lot of attention due to their good charge carrier transport properties and high crystalline quality. Such 1-D systems have unique properties that make them potentially attractive for nanoscale devices [LEDs, lasers, photodetectors, chemical/biosensors, and surface acoustic wave (SAW) devices], which have been intensively investigated [7-9].

ZnO thin films are deposited by the several conventional growth techniques such as DC or RF magnetron sputtering [10], Metalorganic chemical vapor deposition [11], pulsed laser deposition, spray pyrolysis [12], chemical bath deposition, molecular beam epitaxy and cathodic electrodeposition method [13]. The cathodic electrodeposition method is emerging as an efficient nanotechnology for the production of ZnO thin films and nanostructures because of its simplicity, low cost equipments, and suitable for large area substrate [13].

The present chapter deals with the critical review on current status and future prospects about ZnO devices and their different applications

2. STRUCTURAL PROPERTIES

Various researchers up to date have used X-ray diffraction technique to examine the structural characteristics of the ZnO thin film electrodeposited on the several conducting substrates. M. Fahoume et al. [14] studied the structure of ZnO thin films by using the X-ray diffractometry (XRD) and transmission electron microscopy (TEM). They have reported that

the ZnO thin films deposited on the ITO coated glass substrate were polycrystalline in nature with a hexagonal crystal structure (zincite) at pH = 4. At a temperature of 70°C, the film had no preferred orientation and the most intense peak of XRD pattern was along the (101) plane. When the temperature of the bath was increased to 80°C, the most intense peak of XRD pattern was changed and along the (002) plane. They have also reported that when the ZnO thin films were deposited on the copper substrate then it exhibits only one peak along the (002) plane and the degree of orientation along the (002) direction was increased with increasing the bath temperature. The grain size of the film deposited on the copper substrate was larger than the film deposited on the ITO coated glass substrate. Overall, they found that the preferred orientation of the grains was dependent on the type of substrates and bath temperature.

N. H. Al-Hardan *et al.* [15] studied the XRD pattern of deposit ZnO thin film on the ITO coated glass substrate and reported that the film exhibited the polycrystalline nature and the resulted XRD pattern showed that ZnO thin film had hexagonal wurtzite crystal structure. They have reported that the most intense peak in the XRD pattern was along the (002) plane and the ZnO thin film was growing parallel to the c-axis of the hexagonal crystal structure. J.S. Wellings *et al.* [16] studied the various XRD patterns of the as deposited ZnO thin film on the FTO coated glass substrates at different potential from -0.9V to -1.025V. They have reported that ZnO thin films were polycrystalline in nature with hexagonal wurtzite crystal structure and (002) preferred orientation except a sample which was deposited at -0.925V vs. Ag/AgCl. They have also reported that improved crystallinity was observed at -0.975V as compared to the other potentials. They have also observed the effect of annealing on the XRD pattern of the ZnO thin film deposited at -0.975V and concluded that the XRD spectra of both the as deposited and annealed samples had no detectable or noticeable changes [17,18].

C. Gu *et al.* [19] studied the effect of different cathodic potentials and deposition time on the XRD patterns of the ZnO thin films electrodeposited on the ITO coated glass substrates. They have reported that the preferred orientation was changed with the cathodic potential and deposition time. At the potential of -1.3V for 300s, only the (002) preferred orientation was recognized. As the cathodic potentials were decreased from -1.3V to -2.0V, the (002) preferred orientation was weakened and at the -2.0V, the ZnO thin films had random orientations. They were also reported that the (002) preferred orientation of ZnO thin film was also retained at -1.3V after a long deposition time of 1200s. They have also reported

that the initial deposited ZnO film on ITO coated glass substrate had (002) preferred orientation and it was independent of the applied cathodic potential. They have also studied the various XRD patterns of ZnO thin films electrodeposited on ITO coated glass substrates at -1.7V for different deposition times. As the deposition time was increased from 120s to 1200s, the (002) preferred orientation was weakened and after 1200s, the deposited films also had random orientation. C. Coskun et al. [20] studied the XRD patterns of annealed and non annealed ZnO thin films deposited on the ITO coated glass substrate. They have reported that the preferred orientation of the grains was along the (101) direction for both the annealed and non annealed ZnO thin films. They have also reported that the crystallite sizes of around 31nm and 63nm were obtained from the Scherrer equation for non annealed and annealed ZnO thin films respectively. Q. Wang et al. [21] also studied the effect of different deposition potentials and annealing on the XRD pattern of ZnO thin films deposited on the Zn substrate and reported that the films had hexagonal wurtzite crystal structure and the most intense peaks in all the patterns were corresponding to the (002) plane. They have also studied the annealing effect on the XRD pattern of ZnO thin film deposited at -1.0V and observed that the peak corresponding to (002) plane becomes broadened and debased after annealing at 380°C in the horizontal tube furnace. LI Junwei et al. [22] studied the effect of potential and temperature on the XRD patterns of porous ZnO thin films. They have reported that the porous ZnO thin film prepared at the potential of -0.7V had a preferred orientation along the (002) direction, but at the lower potential of -0.9V, the (002) preferred orientation was weakened and the most intense peak corresponding to (101) plane was observed. At the potential of -1.1V and -1.4V, the other peaks corresponding to (100) plane also became stronger and films had random orientations. From the XRD patterns of the ZnO thin films, they have reported that the ZnO films showed the hexagonal wurtzite crystal structure. L. Zhang et al. [23] studied the XRD spectra of as prepared ZnO thin film deposited on the fluorine-doped SnO₂ coated glass substrate. The XRD pattern of the prepared film revealed that the ZnO film had a hexagonal wurtzite crystal structure with preferred orientation along the (002) direction. J. Cembrero et al. [25] studied the XRD pattern of ZnO film deposited on the FTO coated substrate and reported that the film had a hexagonal wurtzite crystal structure with the preferred orientation along the (002) direction.

A. Goux et al. [26] studied the effect of different deposition temperature on the XRD patterns of ZnO thin films deposited on FTO coated glass substrate. At the temperature of 22°C, the XRD pattern of film exhibited the peaks of substrate only and no peak of ZnO was

observed. At the temperature of 34°C, the XRD pattern exhibited the peaks of ZnO {(100) and (101)} along with the peaks of substrate and showed that the ZnO film had hexagonal wurtzite crystal structure. When the temperature was increased, then the peaks become narrower and (002) reflection started to emerge and becomes the preferred orientation at the higher temperature. T. Mahalingam et al. [27] studied the XRD patterns of ZnO thin films deposited at various potentials from -0.8V to -1.2V and reported that the films were polycrystalline in nature and showed the hexagonal wurtzite crystal structure. For all the films deposited at various potentials, the most intense peak in the XRD patterns was along the (002) plane. The intensity of the (002) plane was the maximum for the film deposited at -1.0V vs. SCE whereas the XRD patterns of other films deposited at -0.8V and -1.2V showed that the intensity of (002) plane decreased in comparison to the film deposited at the potential of -1.0V.

G. Zou et al. [28] studied the XRD patterns of the as deposited Zn thin film and the Zn thin films annealed at various temperatures from 100 to 500°C. The XRD pattern of the as deposited Zn thin film revealed that the film had only one strong orientation along the (002) direction. When the Zn thin film was annealed at 300°C for 6 hours, then the film was completely transformed to ZnO thin film and a weak peak {ZnO (100)} along with the two strong peaks {ZnO (002), (101)} was observed, which showed that the film possessed a hexagonal wurtzite crystal structure. When the Zn film was annealed at 500°C for 6 hours, then the ZnO thin film exhibited polycrystalline hexagonal crystal structure with (002) preferred orientation. X. Qin et al. [29] studied the XRD patterns of the ZnO thin films deposited on the ITO substrates with 0.5% op-10 surfactant and without surfactant. They have reported that both the ZnO thin films showed the polycrystalline nature with the hexagonal wurtzite structure. The grain size of the ZnO thin film prepared without the surfactant and with the surfactant was calculated as 54.5nm and 38.8nm respectively. B. E. Prasad et al. [31] studied the effect of Zn concentration, deposition temperature, and time on the XRD patterns of the ZnO thin films deposited on the stainless steel flag. They have reported that at high concentrations, the crystals were growing along the (101) direction, but at the high deposition temperatures and at high deposition times, the crystals were growing along the (102) and (103) direction [32].

3. OPTICAL PROPERTIES

Reiterating, ZnO is considered as a potentially attractive material for light-emitting devices emitting in the ultraviolet (UV) region, because of its large exciton binding energy (60 meV). Since reproducible p-type ZnO has not yet been developed widespread fabrication of ZnO p–n homo-junctionbased LEDs has not been possible. In the interim, n-type ZnO on other available and comparable p-type materials was used for at least investigating the particulars of junctions participated by ZnO. This subject has seen a great deal of activity, and hetero-junctions have been realized using various p-type materials such as Si, GaN, AlGaIn, SrCu₂O₂, NiO, ZnTe, Cu₂O, CdTe, SiC, diamond, ZnRh₂O₄, and ZnO/GaAs. Some details of these hetero-structures concerning their growth and properties can be found in [33] and [34]. In general, electroluminescence (EL) under forward bias was observed to be weak, while in some cases the p–n heterojunctions were considered for uses as ultraviolet photodetectors (PDs). If studies on radiation hardness of ZnO hold true [35], ZnO based PDs in and of themselves could have superior resistance to ionizing radiation and high-energy particles. In the absence of p–n junctions, EL properties of ZnO have also been exploited by fabricating metal–insulator–semiconductor (MIS) structures which do not require p-type ZnO [36].

Schottky diodes based on ZnO have also been implemented as high-efficiency UV photodetectors. In one particular study of Nakano *et al.* [37], a spin coated transparent and conducting polymer [poly (3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT : PSS)] has been used as a Schottky contact on hydrothermally grown Zn-polar single crystal ZnO (Tokyo Denpa). Schottky diodes of 500 X 500 μm² in size exhibited current densities of ~ 4 X 10⁻⁹ A/cm² at -10 V and ~10 A/cm² at +2 V (ideality factor n = 1.02 and Schottky barrier height $\phi_b = 1.1$ eV). Under illumination and at zero bias, the photodiodes attained a significantly large responsivity of 0.3 A/W at 370 nm corresponding to unity quantum efficiency, and a UV/visible rejection ratio of about 10³. Such structures that do not involve p-type ZnO are promising for application in UV detection. ZnO is also extremely attractive as a transparent contact layer due to its high conductivity and transparency in addition to its low cost (needs to be lowered further), nontoxicity, and relatively low deposition temperature. In general, PDs based on other semiconductors with ZnO transparent window layers would have a significant advantage over the ITO window varieties because the transparency of ZnO at 450 nm is more than that of ITO (~90%), and ZnO is also expected to offer lower resistance.

3.1. LIGHT-EMITTING DIODES

One of the first if not the first ZnO-based hybrid heterostructure LEDs was fabricated by Drapak [38] in 1968 who used Cu₂O as a p-type layer. In all the other hybrid structures that followed, the emission observed under forward bias either originated in the p-type layer, or showed a very weak contribution at the ZnO band edge corresponding to extremely small external quantum efficiencies. When p-AlGa_N was used to favor hole injection into ZnO thereby promoting emission in that material, strong EL peaking at ~390 nm due to excitonic recombination within ZnO has been observed [39], [40]. Although these results show that p-AlGa_N is a good candidate for fabricating efficient hybrid heterostructure LEDs with ZnO active layers, all ZnO-based LEDs incorporating stable p-ZnO are needed for light emitters to compete with those based on nitrides.

Reports on all ZnO-based light-emitting devices have also started to appear [41], [42] as p-type ZnO began to become available. However, production of stable and device-quality p-type ZnO has not been realized despite a large number of publications reporting successful demonstration of p-type as discussed in detail in [43]. Incorrect interpretation of the van der Pauw–Hall measurements has been suggested [44] as one of the reasons for the controversial reports of p-type conduction in ZnO. In wide bandgap semiconductors doped with a high density of acceptor-type impurities, localization is an endemic problem: carriers are trapped and cannot follow the Lorentz force and the electric force induced by the low fields employed in Hall measurements. High carrier concentrations in p-type ZnO, which are often reported in the literature, are caused by very low Hall voltages. Corresponding low-mobility values measured are usually indicative of strong localization in the material, which, if present, brings the applicability of conventional Hall measurements under question, particularly at low temperatures. Therefore, the interpretation of Hall effect measurements should be made very carefully.

For reliable p-type behavior, first high density of donor type defects should be minimized and almost intrinsic nearly perfect single crystal ZnO films should be obtained, which is still considered a major challenge. Even when such an approach was adopted, to the extent possible nitrogen doping resulted in very low p-doping concentration ($\sim 10^{16} \text{ cm}^{-3}$) [42], though recent first principles calculations suggest that nitrogen is a deep acceptor and

cannot lead to hole conductivity [45]. Under such conditions, injection from the highly doped n-type ZnO to the lightly doped p-type ZnO is dominant and the recombination takes place primarily in the p-type ZnO. This degrades the recombination efficiency as the quality of the p-type material is inferior. The EL emission coincides with the PL band at 440 nm which is thought to represent the donor–acceptor pair (DAP) recombination in p-type ZnO. To circumvent this problem, the p-doping level must be increased ($> 10^{18} \text{ cm}^{-3}$).

Near-bandedge EL has also been observed from ZnO homojunction LED structures fabricated using phosphorous doped p-ZnO on gallium-doped n-ZnO grown by radio frequency (RF) sputtering on c-sapphire [46]. The p-ZnO layer was reported to have a questionably high hole concentration of $1.0 \times 10^{19} \text{ cm}^{-3}$ but a mobility of only $1 \text{ cm}^2 / \text{V-s}$ after rapid thermal annealing at $800 \text{ }^\circ\text{C}$ in N_2 ambient for 5 min. The UV emission was evident at 380 nm at room temperature. By inserting 40-nm-thick $\text{Mg}_{0.1}\text{Zn}_{0.9}\text{O}$ barrier layers with a 40-nm-thick n-ZnO layer in between near the p–n junction, the carrier recombination was confined in the n-type ZnO layer, which resulted in the suppression of the defect-related broad band centered at $\sim 650 \text{ nm}$ and enhancement of the band edge emission by 55%.

ZnO-based LEDs were also fabricated using p-type arsenic-doped BeZnO and ZnO layers grown by hybrid beam deposition [47]. The hole concentrations in the p-type layers were reported to range from high 10^{16} cm^{-3} for BeZnO to mid- 10^{17} cm^{-3} for ZnO. The active region was composed of seven undoped $\text{Be}_{0.2}\text{Zn}_{0.8}\text{O}$ (7 nm)/ZnO (4 nm) quantum wells (QWs). The turn on voltage was above 10 V, and UV EL peaks were observed at 363 and 388 nm at room temperature, which were attributed to localized excitons in the QWs and donor or acceptor bound excitons, respectively. The green band centered at $\sim 550 \text{ nm}$ blue shifted with increasing current injection. The peak at 363 nm was the dominant spectral feature at current injection levels above 20 mA. Similar active regions were used to fabricate laser structures with $\text{Be}_{0.3}\text{Zn}_{0.7}\text{O}$ cladding layers [48]. Under pulsed current injection, sharp Fabry–Pérot oscillations were observed over the 3.21-eV emission peak, above a threshold current density of $\sim 420 \text{ A/cm}^2$. However, no further analysis of the cavity resonance mode separation was reported.

In terms of nanostructure devices, ZnO nanotips have been used to increase radiation outcoupling in the form of an improved passive light extraction layer in GaN light emitters [49]. The Ga-doped ZnO (GZO) layer, which has the dual role of contact to p-GaN and optically transparent window, was deposited on top of conventional InGaN/GaN LED

structures. Then, ZnO nanotips were grown on GZO coated GaN as a light extraction layer. In comparison with a conventional Ni/Au p-electrode GaN LED, the light emission efficiency was enhanced by a factor of ~ 1.7 . Enhancement in light extraction efficiency was explained by the interaction between the spontaneous emission from the GaN LED and ZnO nanostructures. The results represent one of the low-cost and large-scale fabrication methods for the integration of ZnO nanotips with GaN-based optoelectronic devices using epitaxial growth technologies without the need for e-beam lithography or etching. Even without the nanotips, GZO or Al-doped ZnO (AZO) coated GaN in place of ITO, which is used in all GaN-based LEDs, might turn out to be very lucrative in part fuelled by worldwide shortage of In.

3.2 OPTICALLY PUMPED LASERS

There are plenty of reports on UV lasing from ZnO based materials and devices under optical pumping, for example, lasing from ZnO photonic crystals [50], [51] and random lasing from ZnO powder under certain pumping conditions [52]. ZnO-based microcavities promising for low-threshold surface-emitting polariton lasers are discussed in [53].

Yet another possibility for the realization of a surface emitting laser is the use of an intracavity diffraction element. If the period of such a diffracting structure is carefully adapted to the emission wavelength and the effective refractive index of the device, distributed feedback (DFB) will occur. In the case of a second-order diffraction grating for optical feedback, the surface emission is at exactly 90° to the epitaxial layer plane [54]. In two recently demonstrated devices based on ZnO material, a third-order grating provided feedback. In the latter case, the optical output takes place at angles slightly different from 90° , but nevertheless the device can be regarded as a surface emitter. In an optically pumped laser, the diffraction grating for DFB is typically etched directly into the active semiconductor material [55], [56]. Experiments have shown, however, that direct etching of a diffraction structure into a ZnO thin film is quite problematic. In order to circumvent difficulties with surface roughness, back reflections, and the etching process itself, a Si_3N_4 host layer for the grating needs to be deposited on the ZnO film. The thickness of this additional nitride layer has to be adapted to both the waveguide properties of the epitaxial layer stack and the desired coupling constant of the DFB laser.

One of the main purposes of optically pumped ZnO based DFB laser is to understand their spectral characteristics by investigation of their wavelength tuning behavior. In addition, the well-defined, line-shaped far-field distribution with precisely measurable emission angles allows an unambiguous confirmation of lasing action. Finally, the relatively large pumped volume of such a horizontal cavity surface emitter generates optical output powers which are large enough to be measured with standard photodetectors. This again is a considerable advantage for assessing the most important basic characteristics of the device [57]. All characteristics of optically pumped ZnO-based DFB lasers follow the expected behavior known since 1972 when Kogelnik and Shank published their seminal work on the coupled mode theory [58].

ZnO-based DFB lasers with thin film active regions have recently been implemented [59]. The active region consisted of a thin (5–10 nm) MgO nucleation layer followed by a nominally 200-nm-thick ZnO active region. A 120-nm-thick Si₃N₄ host layer for the grating deposited by plasma-enhanced chemical vapor deposition completed the waveguide structure. A schematic of the waveguide refractive index profile along with the vertical mode profile cleared that an optical overlap on the order of 7% can be achieved for the grating layer. By using a full 120-nm corrugation depth, this geometry results theoretically in a coupling strength of 400 cm⁻¹ for a third-order diffraction grating.

In an attempt to reduce the pumping threshold of optically pumped ZnO-based DFB lasers, multi-QW active regions have been employed [60]. The basic fabrication concept resembles that used for the thin film devices described above, however, the exact realization is nevertheless subject to several important changes. Because of the lower refractive index of Mg-containing Mg ZnO compounds, the thicknesses and compositions of a waveguide using a QW active region need to be adapted. The epitaxial growth was initiated with a 20-nm-thick ZnO buffer, followed by a 400-nm Mg_{0.3}Zn_{0.7}O lower cladding, a 30-nm Mg_{0.2}Zn_{0.8}O lower waveguide layer, a nominally 93-nm-thick active region, and another 30-nm Mg_{0.2}Zn_{0.8}O upper waveguide layer. The stack was then covered with a 50-nm-thick Mg_{0.3}Zn_{0.7}O upper cladding layer, and capped with 150-nm Si₃N₄ for the grating. The active region consists of ten 3-nm-thick ZnO QWs with 7-nm-thick Mg_{0.2}Zn_{0.8}O barrier layers in between. Similar waveguide simulations as in the sample with thin film active region together with a grating depth of 100 nm resulted in a simulated coupling strength of 550 cm⁻¹. Owing to the QWconfinement shift, the emission wavelength of this sample was 360 nm, considerably

shorter than that of the thin film device. However, the smaller effective index of this Mg-containing waveguide nevertheless resulted in a longer grating period of 276 nm.

As an epilogue, it should again be mentioned that unavailability of reliable p-type conductivity prevents ZnO from competing with GaN for optoelectronic applications to a first extent. There are of course continuing developments in the GaN industry to satisfy the increasing demands for larger data storage capacity by developing short wavelength emitters of high efficiency, such as high coherence and low-threshold UV/violet lasers. ZnO-based low-threshold or nearly threshold less lasers, especially those based on polariton lasing in ZnO [53], will also have applications in ever increasing telecommunications and optical storage, in addition to being possible strong light pump sources for white light generation. Therefore, this too underlines the need to develop reliable p-type ZnO that will allow demonstration of high-efficiency ZnO light sources.

3.3 TRANSPARENT CONDUCTING OXIDES AND TFTs

Although Si has shown its supremacy in the field of TFT technology with amorphous Si (a-Si) and polycrystalline- Si (poly-Si), an interest is developing for the optically transparent analog, the transparent thin film transistor (TTFT) technology [61]. Unlike conventional FET or TFT structures, TTFTs require all of the device components (channel, gate, electrodes, and substrate) to be transparent. Such invisible TTFTs can be used in wide range of commercial and military applications where transparency is required. The interest is in part because it is expected that the characteristics of TTFT will not degrade on exposure to visible light due to the wide bandgap of its active channel layer, whereas the characteristics of amorphous or poly-Si TFT degrade. Therefore, measures would not be required in this case to shield the active channel layer from visible light, unlike the commercial amorphous silicon transistors in which visible light generates excess carriers, and therefore, must be shielded. Also it should be noted that such invisible TFTs using ZnO as an active channel achieve much higher field effect mobilities than amorphous silicon transistors ($0.5 \text{ cm}^2/\text{V}\cdot\text{s}$), the major material of today's FET technology. Besides, high-quality crystalline ZnO films can be grown at relatively low deposition temperatures on various substrates including amorphous glasses.

Before delving into the discussion of transistor structures, a brief review of the transparent oxides based on ZnO in their own merits is warranted. In most optoelectronic devices such as flat panel displays, LEDs, and solar cells, it is essential to use a transparent electrode such as a thin film transparent conducting oxide (TCO). Although tin-doped ITO thin films deposited by magnetron sputtering are widely used today for most transparent electrode applications, there are many reports on other TCO semiconductors such as aluminumdoped zinc oxide (AZO), gallium-doped zinc oxide (GZO), indium-doped zinc oxide (IZO), tin-doped zinc oxide (TZO) in large part due to the expected worldwide shortage of In. The growing demand for ITO thin-film transparent electrodes is the impetus for looking at alternative TCO materials since a stable supply of ITO may be difficult to achieve for the expanding market because of the cost and, as mentioned, limited amount of indium available. ZnO-based TCOs have attracted significant attention due to their good conductivity, high optical transparency (in the 90% range), excellent surface smoothness, low deposition temperature, good etchability for patterning, and good reproducibility [62–64]. Candidates for transparent amorphous oxides having large electron mobilities must be constituted of heavy posttransition metal cations with an electronic configuration $(n-1)d^{10}ns^0$, where $n \geq 5$ [65]. Oxide semiconductor XZO films, where $X = Al, Ga, In, Sn$, are prepared from a target made of homogeneous $Al_2O_3(ZnO)_n$, $Ga_2O_3(ZnO)_n$, $In_2O_3(ZnO)_n$, or $SnO_2(ZnO)_n$ compounds. Although various dopants have been used to achieve ZnO TCO films, low resistivity (below $2 \times 10^{-4} \Omega \text{ cm}$) and high carrier concentration (above 10^{21} cm^{-3}) have only been obtained in AZO and GZO films [66]. For example, resistivities as low as $0.85 \times 10^{-4} \Omega \text{ cm}$ and $0.81 \times 10^{-4} \Omega \text{ cm}$ have been obtained for ZnO : Al [67] and ZnO : Ga [68] thin films grown by pulsed laser deposition, comparable to values obtained for ITO (e.g., $0.72 \times 10^{-4} \Omega \text{ cm}$ [69]). In general, the electrical properties of the TCO films strongly depend on the deposition methods and conditions. AZO and GZO films with resistivities on the order of $10^{-5} \Omega \text{ cm}$ have been achieved by PLD, but preparing films on large substrates with high deposition rates is still challenging.

A typical schematic of a ZnO-based bottom-gate-type TTFT structure reported to have an optical transmission (including substrate) of ~75% for visible light [70]. A glass substrate was blanket coated with a 200-nm-thick layer of sputtered ITO to serve as the gate and a 220-nm-thick layer of aluminum–titanium oxide (ATO) deposited by atomic layer deposition to serve as the gate insulator. After deposition of the ZnO layer by ion beam sputtering, a rapid thermal anneal, typically at 600 °C–800 °C in O_2 , was employed to increase

the ZnO channel resistivity in order to improve the electrical quality of the ATO/ZnO interface, and to enhance the crystallinity of the ZnO layer. The direct current (dc) electrical characteristics involves the drain current I_D , gate current I_G , gate voltage V_{GS} , and the drain voltage V_{DS} , for a ZnO channel TFT having a channel length and width-to-length ratio of 1500 μm and 10 : 1, respectively. These curves indicate an n-channel, enhancement-mode TFT operation with excellent drain current saturation and a maximum drain current on-to-off ratio of $\sim 10^7$. The effective channel mobility and the turn on gate voltage were found to be 0.35–0.45 $\text{cm}^2/\text{V}\cdot\text{s}$ and -12 V, respectively.

Song et al. [63] reported on TFTs using amorphous ZnO : In (IZO) for the active channel layer and gate– source–drain electrodes fabricated by RF magnetron sputtering on glass substrates at room temperature while amorphous AlO_x served as the gate dielectric. The devices exhibited low threshold voltages of 1.1 V, on/off ratios of $\sim 10^6$, saturation currents of 1.41 μA at 5 V, and optical transmittance of 80% (including the substrate) in the visible range. Nomura et al. [71] proposed a novel semiconducting material, amorphous oxide semiconductor In–Ga–Zn–O (a-IGZO), for the active channel in TFTs. They deposited a-IGZO on polyethylene terephthalate (PET) at room temperature that exhibited a Hall effect mobility above 10 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$. TFTs fabricated on such PET sheets showed saturation mobilities of 6–9 $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$, and device characteristics were stable during repetitive bending of the TFT sheet. The transfer characteristics showed a low off-current, of the order of 10^{-7} A, and a $\sim 10^3$ on-to-off current ratio. Studies on the effect of bending on the TFT characteristics showed that the performance of the TFT after repetitive bending remained largely unaffected. Only a slight decrease in the saturation current was observed. The TFT was stable at temperatures up to 120 $^\circ\text{C}$, but inoperative at higher temperatures, probably owing to the softening of the PET substrate. Another key issue for ZnO TFTs is the selection of the gate insulator. Among the various gate dielectric materials tested, such as SiO_2 , HfO_2 , $(\text{Pb,Zr})\text{TiO}_3$, ZnMgO , and Y_2O_3 , high dielectric constant $\text{Bi}_{1.5}\text{Zn}_{1.0}\text{Nb}_{1.5}\text{O}_7$ (BZN) has been shown to result in very low operation voltages (< 4 V) in TFTs with ZnO serving as the electron channel [72]. At an operating voltage of 4 V the devices exhibited a field effect mobility (drift mobility) and a current on/off ratio of 0.024 $\text{cm}^2/\text{V}\cdot\text{s}$ and 2×10^4 , respectively, and the threshold voltage was 2 V. High optical transparency ($> 80\%$ for wavelengths above 400 nm), low-temperature processing, and low operation voltage of ZnO-based TFTs with integrated BZN dielectric are promising for transparent device technology.

Lowering the carrier concentration in a TFT channel is crucial for reducing the leakage current. This can be achieved by compensating the n-type carriers by acceptor doping. High-performance thin TFTs with nitrogendoped ZnO as the channel grown by atomic layer deposition on glass substrates have been demonstrated by Lim et al. [73]. The entire TFT fabrication process was carried out below 150 °C including the Al₂O₃ gate insulator deposition before the ZnO channel growth. By nitrogen doping, the electron concentration in ZnO was lowered to $6 \times 10^{14} \text{ cm}^{-3}$. Consequently, the enhancement-mode TFT devices (40 μm channel width, 20 μm channel length) so produced exhibited off-currents as low as 2 pA, with a threshold (turn-on) voltage of 4.7 V, an on/off drain current ratio of 9.5×10^7 , and a saturation mobility of 6.7 cm²/V-s at 35-V drain bias. The devices with undoped ALD-grown ZnO channels did not show any pinch-off and depletion as the electron concentrations in the channel were up to $5 \times 10^{19} \text{ cm}^{-3}$.

Enhancement-mode MgZnO-based field-effect transistors utilizing HfO₂ as the gate dielectric and ITO-coated glass substrates have also been reported [74]. A polycrystalline ZnO channel doped with Mg (10%) and P (2%, assuming that it serves as p-type compensating center) was used to decrease the electron carrier concentration. Mg doping increases the band gap, and therefore, increases the activation energy for defect-related donors, helping the compensation of n-type carriers together with P doping. In these devices, an on/off ratio of 10³ and channel mobility on the order of 5 cm²/V-s were obtained. Another report [75] compared ZnO thin film metal–semiconductor field effect transistors (MESFETs) and metal–oxide– semiconductor (MOS) FETs with either P-doped (2%) or undoped ZnO channels and gate lengths as small as 1.5 μm. Growth of a ~ 0.8–0.9 μm-thick ZnO buffer on sapphire or glass substrates was deemed necessary prior to depositing the active layers in order to reduce the gate leakage current. As would be expected due to the relatively low barrier height of metals on n-type ZnO (0.6–0.8 eV), the MOS structure with 50-nm-thick (Ce,Tb) MgAl₁₁O₁₉ gate dielectric showed an order of magnitude lower gate leakage current than the MESFET. The MOS gate structures using P-doped ZnO channels showed good depletion mode conductance modulation with a field-effect mobility of 5.32 cm²/V-s, as compared to the very poor modulation by the metal structures.

Transparent nature of ZnO, potential achievement of low-cost polycrystalline ZnO, and fabrication technologies that are amenable to TFT production appears to suggest that

ZnO may find a niche application in transparent thin film technologies with added features over the Si-based varieties.

4. ELECTRICAL PROPERTIES

Due to its moderately high (very high for a semiconductor) electromechanical coupling coefficients, ZnO has been successfully used in thin film piezoelectric devices such as bulk acoustic wave and SAW resonators, filters, sensors, and microelectromechanical systems (MEMS), the most common application being the SAW filter, which has been an important component in mass consumer items such as TV filters and wireless communications systems. Successful development of sputtering of high-quality ZnO thin films on Si has been a breakthrough in the SAW device field as it allowed integration of ultrasonics with Si electronics for the first time, liberating device design from bulk piezoelectric substrates. For implementing acousticwave devices piezoelectric ZnO thin films grown on nonpiezoelectric substrates having high acoustic velocity and low propagation loss (diamond, sapphire), weakly piezoelectric materials such as quartz, amorphous substrates such as glass, and other semiconductors (Si, GaAs, InP, etc.) have been used [76].

SAW devices have been implemented with ZnO and $Mg_xZn_{1-x}O$ thin films grown on various substrates such as Si, GaAs, quartz, sapphire, SiC, diamond, GaN, and AlN [76], [77]. The central frequency is at ~ 420 MHz (Rayleigh wave), which gives a sound velocity of 4200 m/s using $\lambda_0=10 \mu\text{m}$. A large effective electromechanical coupling coefficient K_{eff}^2 of 6% was obtained. To reduce the conductivity and achieve piezoelectricity, the as-grown n-type ZnO films were compensated by Li doping during MOCVD growth [78].

To explore higher frequency capability, ZnO thin films have been prepared on high acoustic velocity materials such as SiC and AlN. SAW devices fabricated using 400-nm-thick ZnO thin films deposited on commercial SiC substrates have shown generalized SAW modes with acoustic velocities of 7000 m/s and pseudo-SAW modes with acoustic velocities of 12 500 m/s [79]. ZnO thin films deposited on diamond, which has the highest acoustic velocities among all materials, have shown to exhibit SAW velocities as large as 11 600 m/s with coupling coefficients up to 1.1% [80]. Using 0.65- μm wide electrode devices with a center frequency of 10 GHz, a coupling coefficient of 1.5% was also demonstrated for the fifth harmonic of the second Sezawa mode [81]. All the aforementioned results show that ZnO

thin films on various substrates can be successfully implemented as efficient SAW devices. Even though piezoelectric materials such as LiNbO_3 and LiTaO_3 have larger electromechanical coupling coefficients, as high as 38% and 14%, respectively, ZnO is promising for applications that require electroacoustic modulation and monolithic integration with other semiconductor technologies.

ZnO-based SAW devices have been used for UV photodetection sensing. The SAW UV photodetector is based on the acoustoelectric interaction. The photogenerated carriers increase the conductivity in the material which in turn increases the attenuation and decreases the SAW velocity, and therefore, the SAW frequency. SAW UV detectors with ZnO piezoelectric layers (highly compensated by Li doping with a resistivity above $10^7 \mu\text{cm}$) have also been demonstrated on r-sapphire using a thin $\text{Mg}_{0.2}\text{Zn}_{0.8}\text{O}$ buffer layer in between for isolation and as a diffusion barrier for Li. The IDTs were placed on the piezoelectric layer with the ZnO UV absorbing layer in between the transmitter and receiver IDTs. A frequency shift up to 11MHz and an insertion loss increase of ~ 23 dB were observed in the central Sezawa mode frequency (711 MHz) when the device was illuminated with 365-nm light of intensity 2.32 mW/cm^2 . The corresponding maximum phase shift was measured at 365 nm as 107° . ZnO SAW UV detectors with such performance were proposed for use as passive zero-power remote wireless sensors [82]. These ZnO-based SAW devices can also be used for gas [83] and biochemical [84] sensing.

5. NANOSTRUCTURE APPLICATIONS

ZnO nanorod/nanowire FET sensors may create opportunities for highly sensitive and selective real-time detection of a wide variety of gas molecules and biomolecules. The principle of gas sensor operation depends on the nature of gas molecules and is based on the modification of nanowire FET channel conductivity. Oxygen vacancies in ZnO function as n-type donors on oxide surfaces and are electrically and chemically active. Upon adsorption of charge accepting gas molecules, for example, NO_2 and O_2 , electrons are depleted from the conduction band resulting in a reduced conductivity of the n-type oxide. On the other hand, molecules that chemically react with surface oxygen (for example, CO and H_2) interact with surface adsorbed oxygen on ZnO and remove it leading to an increase in conductivity. ZnO exhibits strong adsorption of molecules on the surface, which affects the electrical characteristics of ZnO-based devices, dependent on surface-mediated phenomena. Thanks to the large surface-to-volume ratio of the nanostructures, the detection sensitivity of FET

biosensors may be increased to a single-molecular detection level by measuring the small conductance changes caused by binding of biomolecular species on a nanorod conduction channel [85].

5.1. GAS SENSORS

There is a strong interest in the development of lightweight gas sensors capable of parts per million (ppm) range sensitivity and extended operation at low-power levels. All experimental results demonstrate that ZnO nanowires, owing to the large surface area, have a potential for detecting NO₂ [85], NH₃ [86], NH₄ [87], CO [87], H₂ [88], H₂O [89], O₃ [90], H₂S [91], and C₂H₅OH [92]. Detection of gas molecules is usually achieved by measuring the resistivity change of either the nanocrystalline ZnO films or the nanowire channel of an FET.

Cho et al. [85] reported a 1.8 fold decrease in resistance of well-dispersed ZnO nanorods at 1-ppm NO₂, while there was no significant change in resistance at 50-ppm CO. Sputtered nano-crystallite Cu-doped ZnO films of columnar structure with average grain size of 5 nm exhibited very high sensitivity (2.7–20 ppm) to CO at 350 °C [87]. A substantial change in resistance of the ZnO films was also observed at a low operating temperature of 150 °C when the sensor was exposed to 6-ppm CO. The sensitivity for room temperature detection of hydrogen by ZnO nanorods was shown to be greatly enhanced by sputter-depositing clusters of Pd on the surface [63]. An increase in the resistivity by a factor of 5 was observed upon exposure to hydrogen concentrations of 10– 500 ppm compared to that without Pd. Pd-coated ZnO nanorods showed sensitivities lower than 10 ppm, with 95% recovery of the initial conductance after 20 s.

ZnO nanorods are also promising candidates for detecting extremely low concentrations of H₂S. High response and good selectivity of ZnO nanorods to low concentrations of H₂S was observed by Wang et al. [91]. The ratio of the electrical resistance in air to that in 0.05-ppm H₂S was measured to be 1.7 at room temperature. One of the most important parameters of gas sensors is their selectivity. The selectivity is achieved by applying different voltages to the gate of a nanowire FET or by performing measurements at different temperatures since different gas molecules have different activation energies. Such selectivity for NH₃ and NO₂ gas molecules, where the refresh (erase) voltages, negative gate voltages required for electrical desorption of adsorbed gas molecules, for the two gas

molecules are significantly different making it possible to distinguish different gas species [93].

5.2. BIOSENSORS

The potential of ZnO nanostructures as nanosized biosensors has also been explored for detecting different biological molecules. Development of 1-D ZnO nanostructures as biosensors is in the embryonic stage and only a limited number of reports are available [94]–[94]. The 1-D ZnO biosensors have advantages such as stability in air, nontoxicity, chemical stability, electrochemical activity, ease of synthesis, and bio-safe characteristics. As in the case of gas sensors, the principle of operation is that the conductance of ZnO nanorod FETs drastically changes when biomolecules are adsorbed.

The key factor in most biological processes is the need for a small change of the pH concentration created by the release of H^+ ions during biochemical reactions. Therefore, determination of pH is a prerequisite for many processes. The sensing mechanism for pH is the polarization- induced bound surface charge by interaction with the polar molecules in the liquids. Application of ZnO nanorods as pH sensors for intracellular chemical sensing is under development and a room temperature sensitivity (change in surface potential) as high as ~59 mV per decade change in the pH value has been reported [99].

5.3 SOLAR CELLS

Solar cells represent a very promising renewable energy technology because they provide clean (beyond manufacturing) and renewable energy reducing our dependence on fossil oil and our impact on the environment. Dye-sensitized solar cells (DSSCs), using inorganic semiconductors, are being studied for very efficient, inexpensive, large-scale solar energy conversion. DSSCs have been explored as possible substitutes for conventional silicon cells, but they suffer from possible dye agglomeration or electrolyte leakage [100]–[104].

During its traversal to the photoelectrode, an electron in a polycrystalline solar cell usually crosses about 10^3 to 10^6 nanoparticles, and disorder structure of the nanoparticle film leads to enhanced scattering of free electrons, thus reducing the electron mobility and

increasing detrimental electron recombination rate. Replacement of the nanoparticle film with an array of oriented single crystalline nanorods would result in the rapid collection of carriers generated throughout the device as the nanorods provide a direct path from the point of photogeneration to the conducting substrate, thus effectively reducing the electron recombination losses. A typical DSSC structure, which has three main components: 1) a thick (~10 μm) film of wide bandgap semiconductor nanostructures (TiO_2 , SnO_2 , or ZnO), 2) a monolayer of organic dye molecules absorbed onto the semiconductor nanostructures, and 3) a liquid electrolyte containing the redox (reduction-oxidation reaction) couple I^-/I_3^- that penetrates in between the dye-coated nanostructures. Electron transport in the single crystalline rod is expected to be several orders of magnitude faster than that in a random polycrystalline network. However, increased surface area by nanorods, as compared to thin films, also increases the surface recombination causing reduction in efficiency [105].

CONCLUSION

The present review clears that, ZnO offers some potential in providing electronic, photonic, and spin-based devices, and encouraging progress has been made in the research phase. Despite this progress, there are still a number of important issues that are in need of further investigation before this material can be transitioned to commercial use for the stated applications. The task is made more difficult by the highly successful GaN which competes for similar applications. However, there is some niche applications of ZnO which are not addressed by GaN which if explored fully might pave the way for ZnO . TTFT, ZnO -based transparent oxides, laser structures exploiting the large exciton binding energy of ZnO are among these applications.

The review suggest that the crystal structure of films was independent of the initial conditions and the most of the films showed polycrystalline in nature. From the XRD patterns, it is concluded that the nanostructured ZnO thin film had hexagonal wurtzite crystal structure and the crystals were preferably grown along the (002) or (101) planes. The grains or crystallites size of different films was varied from 10nm to 10 μm and concluded that the electrodeposited ZnO films had nanostructured particles and the grain size of the films increases after annealing process at different temperature. In most cases, the band gap energy of the deposited films decreased after the annealing process, due to the decomposition of Zn(OH)_2 into ZnO and H_2O . Because the Zn(OH)_2 is a wider band gap material as compared to the ZnO and is normally coexisted with the ZnO in the as-deposited films.

In the case of TFTs, the critical issues are the mobility in noncrystalline or polycrystalline ZnO, and the choice of gate oxide, substrate, and the methods to deal with high electron concentrations encountered in ZnO channel layers. For electrical characteristics, the on-to-off ratio as well as the transconductance is important to home in on. ZnO also lends itself to applications in sensing, in part due to ease with which ZnO can be produced in the form of nanostructures. There is still much to be understood in terms of the mechanism of ZnO gas sensors and biosensors. Although a number of ZnO sensors have been reported on detecting different gas molecules and biomolecules, the selectivity (not unique to ZnO) remains to be the main issue since identification of the nature of the absorbed molecules is very important. Although several approaches have been proposed, which employ different activation energies of different gas molecules, this field is still in a state of infancy and much more effort needs to be expended to pave the way for improved selectivity of ZnO sensors and increase the lifetime of the devices.

Regarding solar cells, which received some attention although the competition is very stiff, further studies are required to improve the current density and efficiency. This could be achieved by application of various electrolytes [106] and doping the ZnO films to improve its conductivity. There is also control of the properties of individual building blocks which at this point is inadequate and device-to-device reproducibility is low.

As for the nanostructures, ZnO nanostructures (nanowires, nanorods, etc.) provide a path to a new generation of devices, but a deliberate effort has to be expended for ZnO nanostructures to be taken seriously for large-scale device applications, and to achieving high device density with accessibility to individual nanodevices. Reliable methods for assembling and integrating building blocks into circuits need to be developed.

Finally, ZnO as a semiconductor is facing a very stiff competition from GaN which is much more mature in terms of devices (also has acceptable p-type dopant unlike the case in ZnO). Lack of a credible p-type doping hampers widespread optical emitters in ZnO. Furthermore, highly ionic nature of ZnO with large electron phonon coupling and low thermal conductivity does not bode well for ZnO based electronic devices. Nanostructures seem a little easier to produce with ZnO, but it remains to be seen whether nanostructures in general as hyped would really make inroads in the area of devices.

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