Chapter 9

ZnO Thin Films: Fabrication Routes, and Applications

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Abstract

Thin films have become a hot topic in the field of nanotechnology. Due to their optical and electrical characteristics, thin-film semiconductor oxides are among the semiconductor oxides with the greatest range of applications. The most popular semiconductor component is zinc oxide (ZnO). ZnO, a wide bandgap semiconductor (Eg = 3.37 eV at room temperature), have been widely used in electronic, optoelectronic, and information technology device platforms. Nano-ZnO thin films have a wide range of applications due to their remarkable properties. There are several methods for developing a thin layer of the ZnO nanomaterial. Sputtering, chemical vapor deposition (CVD), molecular beam epitaxy, pulsed laser deposition (PLD), and spray pyrolysis are among these methods. Although sputtering is the most commonly used method for high-quality applications, chemical vapor deposition and spray pyrolysis are also popular due to their low cost and ease of use. This chapter provides a brief overview of the various fabrication routes, characterization
techniques, and applications of ZnO thin films, allowing us to investigate the chemical, structural, optical, and electrical properties of ZnO thin films, as well as their various applications.

Keywords
Zinc Oxide (ZnO), Thin Film, Fabrication Techniques, Semiconductor, Routes

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1. Introduction
A 2D nanomaterial technology known as "thin film technology" include "plate-like thin film" has the potential to create efficient and adaptable equipment at the micro- or
nanoscale. Microelectronic devices, high energy density harvesters, high sensitivity low power sensors, anti-reflective and protective coatings on modern equipment, transparent electrodes, etc. are just a few of the applications and advantages that thin films have. As a consequence, various methodologies are being developed to generate thin films of good quality [1, 2].

A class of substances known as metal oxides is crucial to several disciplines, including physics, chemistry, and materials science [1]. ZnO, one of the metal oxides, has the potential to be a semiconductor oxide. Additionally, many types of ZnO nanostructures, including nano-ZnO thin films, have also been the focus of numerous research investigations and are being used in business and technology.

High demand in research is because of its high and direct energy gap (3.37 eV) at room temperature, large excitation binding energy (60 meV), high electron mobility, high power stability, inherent piezoelectric properties, environmental compatibility, and relatively easy construction technology [1-3]. The II -VI compound semiconductor exhibits significant electrical, optical, and magnetic properties. Accordingly, this II - VI semiconductor has been widely applied in various fields including electronics, spintronics, sensors, solar cells, piezoelectric nanogenerators, light-emitting diodes, lasers, memory devices, acousto-optic devices, UV detectors, gas sensors, and transparent conductive oxides [4, 5].

ZnO thin film synthesis has gained prominence as a study topic in recent years. Control of the layer morphology must be maintained from a microscopically uniform and flat surface to a nanoscale tailored surface in order to successfully complete these varied applications [6].

1.1 ZnO thin films

The technique of coating a surface with an exceptionally thin layer of material is known as thin film deposition (a few nanometers to 100 micrometres thick, or on the scale of a few atoms thick). However, a significant stage in many applications is the controlled fabrication of thin films. Using thin films, a wide range of innovative and adaptable devices can be generated. Thin films have shown to be important for the development of microelectronic devices, reliable sensors, protective layers on a variety of goods, and antireflective coatings on solar cells, among other uses. Consequently, as the demand for cutting-edge technology grows, efficient methods for thin-film deposition are being developed [7, 8].

ZnO is a versatile oxide that exhibits a unique combination of multifunctional qualities in addition to being a wide bandgap II-VI compound semiconductor and a piezoelectric material [9]. ZnO can be found in a variety of various crystal forms. The three crystal structures of ZnO identified in the literature are wurtzite, rocksalt, and zincblende [2-4]. The latter is hexagonal, while the first two are cubic. The thermodynamically stable form of ZnO is in the Wurtzite crystal structure at room temperature. The lattice constants are a=b=3.25, c=5.2, and c/a=1.60, which is not far from the ideal value for a hexagonal cell, c/a=1.633 [7, 10, 11]. Due to its electro-optical features, high electrochemical stability, big band gap, natural abundance, and low toxicity, ZnO is becoming one of the most widely
utilised transparent conductive oxides. The properties of ZnO thin films are dependent on the non-stoichiometry of the films, which is caused by oxygen vacancies and interstitial zinc.

1.2 Historical background

Without a question, semiconductor materials have altered the world in ways that no one could have predicted. Alessandro Volta coined the word "semiconducting" in early 1782.[1]. Since then, the semiconductor industry has grown rapidly around the world. When sensitive patches on ZnO crystals were tested with thin copper wires as a detector for radio sets, researchers started looking into ZnO materials for semiconductor capabilities in the early 1920s [12]. In 1935, Fritsch et al. synthesized polycrystalline ZnO films by evaporation method, which observed the semiconductor behaviour of undoped ZnO films [13].

It has been shown that a number of deposition processes may deposit thin films of device quality as a result of recent significant efforts in research and development dedicated to the generation and characterization of thin ZnO films. When high-quality single-crystal and epitaxial layers were made accessible, this material underwent a new trend that made it possible to fabricate electro-optical and optical equipment.

ZnO thin films are gaining significance for applications in many disciplines due to its broad energy gap, substantial piezoelectric, electro-optical, and nonlinear optics coefficients, high electrical conductivity, optical transparency, and structural defects. Furthermore, the material has a variety of applications in the field of nanotechnology because it is simultaneously biosafe and biocompatible. A variety of nanostructures can now be generated employing innovative construction methodologies. Additionally, due to its neutral chemical behaviour, it can be used in a wide range of scientific and industrial applications.[2, 4, 14]. However, discovering methods to produce ZnO thin films at scales that are both affordable and efficient, as well as developing these films with enhanced characteristics, remain substantial difficulties in the fields of production and research and development (R&D).[7].

Following that, in 1950, the bulk ZnO crystal was investigated as a typical II-VI compound semiconductor. A significant portion of research at this time was devoted to the study of the electrical and optical properties of ZnO (such as n-type conductivity, absorption spectrum, and electroluminescence decay parameters) [15, 16].

It is important to note that thin films have grabbed the bulk of the scientific community's attention since 1960, initially due to the utilization of ZnO's piezoelectric potential in surface acoustic wave devices [17, 18]. Moreover, there was a considerable amount of growth in the ZnO research in the mid 1960s, with a significant amount of work and research being carried out in both theory and practice. Laudise et al. reported that large macroscopically sound ZnO crystals suitable for preliminary transducer and other experiments have been grown hydrothermally [19].
Significant work on the creation of ZnO devices with a lower complexity was accomplished in the 1970s [20, 21]. Many techniques have been examined for fabrication of high-quality ZnO thin films in the 1980s, including metal-organic chemical vapor deposition (MOCVD), spray pyrolysis, and radio-frequency (RF) magnetron sputtering [22].

In the 1990s, researchers analyzed further into fabrication of high-purity bulk ZnO wafers, and in 1998 they succeeded adopting the transition vapor phase technique. Researchers tended to concentrate more on the mechanisms of ZnO development and the factors that influence its applicability for smart technologies in the early 2000s.

The development of ZnO thin films for acoustical and optical devices has thus been the subject of several experiments on a range of substrates (glass, sapphire, and diamond). In the early studies, thin ZnO films were formed using techniques including chemical vapor deposition and magnetron sputtering. However, the nature of the ZnO thin films was primarily polycrystalline. By using radio frequency magnetron sputtering and other growth techniques that allowed for refined control of the deposition process, such as molecular beam epitaxy (MBE), pulsed laser deposition (PLD), metal-organic chemical vapor deposition (MOCVD), and hydride or halide vapor-phase epitaxy, subsequent research tends to result in the production of high quality ZnO single crystal films (HVPE)[2, 4].

1.3 Scope of this chapter

We will present a good rundown of ZnO thin films in this chapter. The special characteristics, various fabrication techniques, and uses of ZnO thin films will all be covered in the section that follows.

2. Structural features

According to our discussion, a thin layer consisting of this material has attracted a lot of attention in recent years, and researchers have conducted extensive research in this area. Generally, in crystal form, atoms or ions follow a periodic order. The crystal structures found in oxide compounds are numerous and, in some cases, complex and are found as single microstructures in compounds that contain two and three oxides. The general structures of monoxide (Mo), where M represents the metal cation, have been observed in the form of wurtzite and sodium chloride (NaCl) [23]. In general, Most of the group II–VI binary compound semiconductors crystallize in either cubic zinc blende or hexagonal wurtzite structure where each anion is surrounded by four cations at the corners of a tetrahedron, and vice versa. As a compound II-VI, ZnO exists as three forms: wurtzite, zinc bland, and rock salt. The wurtzite lattice of ZnO has been shown in Figure 1 [2, 4].
Only by emerging on cubic substrates can the structure of zinc blende ZnO be established. A hexagonal wurtzite structure with the lattice constants $a = b = 3.249$, $c = 5.204$, and $a/b = 1.633$ is thermodynamically stable at normal temperature, in contrast to the rocksalt structure, which is found under exceptionally high pressures [10 GPa]. Two sub-lattices ($\text{Zn}^{2+}$ and $\text{O}^{2-}$) that are layered underneath of one another were taken into consideration to characterize this structure. This signifies that each $\text{O}$ ion surrounds a $\text{Zn}$ ion in a tetrahedral arrangement, and vice versa. The chosen structure for optoelectronic applications is a wurtzite structure [2, 24]. Table 1 indicates the properties of wurtzite lattice structure of ZnO.

Figure 1: The wurtzite crystal lattice of ZnO: small circles represent zinc atoms, whereas large circles illustrate oxygen atoms [4].
Table 1. the properties of bulk wurtzite lattice structure of ZnO [4].

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lattice parameter</td>
<td>a= b=3.250 Å</td>
</tr>
<tr>
<td></td>
<td>c= 5.206 Å</td>
</tr>
<tr>
<td></td>
<td>u= 0.348</td>
</tr>
<tr>
<td></td>
<td>c/a= 1.593-1.6035</td>
</tr>
<tr>
<td>Density</td>
<td>5.606 gm/cm³</td>
</tr>
<tr>
<td>Melting point</td>
<td>2248 K</td>
</tr>
<tr>
<td>Stable crystal structure</td>
<td>wurtzite</td>
</tr>
<tr>
<td>Dielectric constant</td>
<td>8.66</td>
</tr>
<tr>
<td>Refractive index</td>
<td>2.008, 2.029</td>
</tr>
<tr>
<td>Band gap (Eₘ)</td>
<td>3.37 eV (direct)</td>
</tr>
<tr>
<td>Exciton binding energy</td>
<td>60 meV</td>
</tr>
<tr>
<td>Electron/ Hole effective mass</td>
<td>0.24 m₀ /0.59 m₀</td>
</tr>
<tr>
<td>Hole mobility (300) K</td>
<td>5-50 cm³/Vs</td>
</tr>
<tr>
<td>Electron mobility (300) K</td>
<td>100-200 cm²/Vs</td>
</tr>
<tr>
<td>Heat capacity</td>
<td>9.6 cal/molK</td>
</tr>
<tr>
<td>Born effective charge</td>
<td>2.10</td>
</tr>
<tr>
<td>Heat of crystallization</td>
<td>62 KJ/mol</td>
</tr>
<tr>
<td>Interinsic carrier concentration</td>
<td>&lt;106 cm⁻³</td>
</tr>
<tr>
<td>Bond length</td>
<td>1.977 um</td>
</tr>
<tr>
<td>Thermal conductivity</td>
<td>0.6, 1.2</td>
</tr>
<tr>
<td>Breakdown voltage (10⁶ V cm⁻¹)</td>
<td>5.0</td>
</tr>
<tr>
<td>Saturation velocity (10⁷ cms⁻¹)</td>
<td>3.0</td>
</tr>
<tr>
<td>Ionicity</td>
<td>62%</td>
</tr>
</tbody>
</table>

3. Synthesis methods

Thin film technology played a crucial role in controlling the properties of ZnO thin films, since the same material deposited using different techniques usually had different physical characteristics. The reason for this is that the electrical and optical properties of the films depended substantially on the structure, morphology, and type of impurities within them. In this sense, films grown using any particular technique may have different properties due to the variation in various deposition parameters, and therefore, the properties can be
tailored by varying the deposition parameters. The development of ZnO films with improved optical, electronic, or structural properties is expected to lead to improved conversion efficiencies in the future, while the development of processes for large areas at high growth rates are critical to meeting production cost targets [25].

Various methods have been used to grow ZnO thin films, including chemical vapor deposition (CVD), sputtering, pulsed laser deposition (PLD), molecular beam epitaxy (MBE), spray pyrolysis and so forth, which have improved its electrical and optical properties; These applications are discussed in this section. The first three methods, CVD, sputtering and PLD, lead to the best ZnO films when it comes to high conductivity and transparency. Both of these methods, CVD and particularly the sputtering technique, can also be applied to large areas, making them the most advanced methods for depositing ZnO in the industry [25, 26].

3.1 Commonly used methods for synthesis

The two primary categories for producing ZnO thin films are physical methods and chemical approaches, as was already mentioned. Each of these two strategies has advantages and disadvantages depending on which is picked. Depending on the different uses, a strategic goal might be set.

3.1.1 Sputtering

The sputtering technique is one of the adaptable ways to deposit ZnO thin films. It is seen that the samples have greater uniformity and regulated composition when this method (magnetron sputtering) is compared to chemical methods (such as sol-gel and chemical-vapor deposition methods). Additionally, we can better regulate the film's thickness with this technique. This method's simplicity, affordability, and low operating temperature are further benefits [17, 27]. This technology operates by applying voltage between the cathode and the anode, where the anode operates as the substrate holder and the cathode as the target holder, to establish a gas plasma. It experiences severe ion bombardment before having particles expelled from the cathode. Particles are deposited on a substrate after diffusing away from the cathode surface. Sputtering often takes place between $10^{-2}$ and $10^{-3}$ Torr of pressure. A high-frequency generator is linked between the electrodes for non-conducting samples and insulators, and a DC voltage is delivered between the anode and cathode for conducting targets (DC sputtering) (RF Sputtering), as shown in Figure 2 of the schematic [28, 29]. Magnetron sputtering is suitable for applications where a high deposition rate or a low substrate temperature are needed. Magnetron sputtering uses a magnetic field to enhance the sputtering rate, which increases the rate of deposition as well [28] at each optimum conditions.
At a specified substrate temperature, these films are synthesized by magnetron sputtering from high purity ZnO targets. In growth medium with O$_2$/Ar+O$_2$ gas ratios ranging from 0 to 1 at pressures of $10^{-3}$ to $10^{-2}$ Torr, growth occurs predominantly.

O$_2$ is used as the reactive gas and Ar is employed as a gas to speed up the sputtering process in the approach that has been stated. Another option is to generate ZnO by Zn target in an Ar+O$_2$ combination using a DC sputtering method. Additionally, the amount of sputtering yield rate from the ZnO target may be controlled by adjusting the magnetron sputtering power applied to the plasma. In order to stabilise the system and eliminate impurities from the target surface, this experiment requires the target to be presputtered for 5 to 15 minutes before deposition begins. The structural and optical characteristics of ZnO films produced by RF MSD in an O$_2$/Ar gas mixture are only marginally affected by the growing temperature in the diapason range of 200 to 300 °C. It has been demonstrated that using pure argon plasma produces non-stoichiometric porosity ZnO sheets with a reflectivity index of approximately 1.65. The ZnO films produced by RF MSD in an oxygen-rich gas combination are stoichiometric solid polycrystals with reflectivity indices and energy band gaps of around 2.0–2.1 eV and 3.12–3.2 eV, respectively [17].

Direct current (DC), radio frequency (RF), and high-power impulse magnetron sputtering (HiPIMS) are only a few of the different types of magnetron sputtering techniques. Radio frequency magnetron sputtering (RF MS)(13.56 MHz, 27 MHz) enables the sputtering of both conductive and nonconductive materials. When non-conducting ZnO is being deposited, a metallic Zn target is typically sputtered in an oxygen environment [30]. Partial pressure is a crucial process parameter in each scenario, and the desired power density is...
typically no greater than 10 W/cm$^2$. To manage the stoichiometry of the deposited films, one can regulate the amount of oxygen doping used during the RF process. By sputtering the conductive target, Zn oxide coatings can also be produced using DC MS. This typically necessitates using targets composed of metallic Zn material. Similar to RF MS, the intended power density is limited to 10 W/cm$^2$. Recently, high-power impulse magnetron sputtering has received a lot of attention. With this technique, the target is sputtered with current pulses that are extremely high and brief—their length is on the order of a dozen or more microseconds. The modulation frequency for current waveforms is several kHz. Though the sputtering efficiency is somewhat lower, it is important to note that the average power released in the target is comparable to that of conventional DC and RF MS procedures. Conversely, due to phenomena associated with the start and pulse suppression of the HiPIMS approach, mechanical properties of deposited films, such as hardness or optical qualities, can be improved [31]. In a recent report, Sm doped ZnO thin films were fabricated by RF sputtering on glass and silicon substrate [32]. AFM technique was utilized to measure the surface roughness and grain size distribution of the films. In the work, the surface roughness was found to improve with the Sm intercalation. Figure 3 shows the 3D AFM images, grain size distribution, and surface roughness images of the Sm:ZnO thin films. The grain size distribution was found to increase from 73.20 nm to 82.57 nm, while the surface roughness was found to decrease from 6 nm to 2.35 nm with Sm inclusion.

![AFM images](image)

Figure 3: 3D AFM images, grain size distribution and surface roughness of Sm:ZnO thin films [32].
3.1.2 Chemical vapor deposition (CVD)

CVD, one of several techniques for fabricating thin films of semiconductor materials, is particularly alluring due to both its high quality and its capacity for mass manufacturing. CVD or spray pyrolysis preparation processes are comparable to those used for chemical bath deposition (CBD). It is necessary to dissolve and evaporate the zinc precursor in a solution. Precursor evaporation can be accomplished in a number of methods. The zinc precursor first needs to be simple to evaporate or to put it another way, it must be a volatile precursor. The zinc ingot used as a precursor can then evaporate at high temperatures. The final step involves employing equipment like an atomizer or an ultrasonic transducer to atomize the precursor or turn it into a mist. The zinc precursor solution has been transformed into a mist by an ultrasonic device and transported by inert gas into the heat furnace chamber, where a substrate has been positioned as the target for creating a thin layer of ZnO. Some of the zinc precursor particles react with an oxygen-containing molecule in the heater chamber after the precursor mist or vapor has been delivered there and arrived, growing the molecule. The substrate is then covered with this molecule. This molecule breaks down into zinc as a result of a high temperature or sufficient energy, which subsequently interacts with oxygen to create ZnO. The solvent leaves the compartment after evaporating. Likewise, a thin film of ZnO is formed in conjunction with the carrier gas. When zinc precursor enters the chamber, it may adhere to the substrate and then engage in a reaction or bonding with other materials. This alternative reaction process another oxygen-containing molecule. The zinc precursor molecule and oxygen-containing molecule react with the ZnO due to the high temperature and sufficient energy to decompose, releasing some degraded solvents that are carried away by gas before the ZnO molecule finally transforms into ZnO thin film.

Several CVD-type methods are available [29]:
1. APCVD: (Atmospheric Pressure Chemical Vapor Deposition) deposition under atmospheric pressure;
2. LPCVD: (Low-Pressure Chemical Vapor Deposition) low-pressure deposition;
3. MOCVD: (Metal Organic CVD) the use of organometallic precursors;
4. PACVD: (Plasma Assisted Chemical Vapor Deposition) with the assistance of a plasma. As shown in Figure 4, in this method, one or more gaseous species react with a solid substrate (surface). The metallic oxides are commonly created in this process by vaporising the organo-metallic compounds. Condensate from a vapor is transmitted to a substrate surface, where a heterogeneous process typically breaks it down. Different types of breakdown occur depending on the species that transport the volatiles. Decomposition conditions should be such that the reaction only occurs on the substrate surface or close to the surface, and not in the gaseous state, to avoid the creation of powdery deposits that could cause haziness in the films during deposition [2, 4]. An important aspect of the CVD technique is that the deposited ZnO is the result of chemical reactions between substrates and vapor phase precursors. The vapor is subsequently transported to the growth zone by
the carrier gas. The reactions take place in a reactor where the necessary temperature profile is generated in the direction of the gas flow. Another variation of APCVD is AACVD which is known as aerosol assisted CVD technique. AACVD has some advantages (non-volatile precursors, easily handled, wide range) over APCVD and is in current trend for the fabrication of ZnO thin films [33]. Recently, Al doped ZnO thin films were fabricated using AACVD. Different optoelectric properties were optimized by merely varying the precursors. Table 2 summarizes the modified optoelectric properties for different precursors.

![Figure 4. The main steps of deposition by the CVD method.](image)

Table 2: Modified optoelectronic properties of Al: ZnO thin films fabricated using aerosol assisted CVD technique [33].

<table>
<thead>
<tr>
<th>Solvent</th>
<th>(T_{Aa00-700}/%)</th>
<th>(E_g/\text{eV})</th>
<th>(\rho \times 10^{-2}/\Omega \text{ cm})</th>
<th>(n \times 10^{19}/\text{cm}^3)</th>
<th>(\mu \text{cm}^2 \text{ V}^{-1} \text{ s}^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>MeOH</td>
<td>83</td>
<td>3.25</td>
<td>0.5</td>
<td>14.0</td>
<td>9.0</td>
</tr>
<tr>
<td>MeOH and toluene</td>
<td>78</td>
<td>3.25</td>
<td>27.2</td>
<td>7.1</td>
<td>0.3</td>
</tr>
<tr>
<td>MeOH and THF</td>
<td>80</td>
<td>3.25</td>
<td>44.2</td>
<td>5.3</td>
<td>0.3</td>
</tr>
<tr>
<td>MeOH and (n)-hexane</td>
<td>77</td>
<td>3.25</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>MeOH and cyclohexane</td>
<td>75</td>
<td>3.26</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>MeOH and ethyl acetate</td>
<td>67</td>
<td>3.24</td>
<td>0.2</td>
<td>9.7</td>
<td>13.3</td>
</tr>
</tbody>
</table>
3.1.3 Pulsed laser deposition (PLD)

Three stages compensate PLD, and each is repeated thousands of times all throughout the deposition process.

1. The vaporization of the target material,
2. The transport of the vapor plume;
3. The growth of a film on a substrate.

Laser pulses are seldom effective in removing material layer-by-layer and in a clean manner. Instead, the target surfaces are chemically and physically deformed [34].

By using this technique, the substance to be deposited, i.e. the target, is ablated by means of laser pulses leading to the formation of plasma. When the plasma is exposed to the background gases supplied into the vacuum chamber, it interacts with the gases. Eventually, it condenses on the surface of the object to be coated with the material, i.e., the substrate, and nucleates to get deposited as a thin film. PLD provides the advantage of being simpler, faster, and cheaper than other technologies; high quality films with the desired crystalline structure can be produced with this technology. However, there are a number of factors involved which determine the quality of the film, such as pressure optimization, target composition and shape, substrate temperature and laser pulse characteristics [7].

A schematic of the typical PLD system is shown in Figure 5. This method has a number of advantages, including [4, 7, 34, 35]:

1. The ability to create particles using high energy sources,
2. Possibility of controlling the film thickness down to even one atomic layer by adjusting the number of pulses,
3. The resulting ZnO films exhibit an extremely smooth surface, compared with films formed by other physical vapor deposition processes,
4. The growth of high-quality films at low substrate temperatures (200 to 800 °C) and creating a thin film with the desired crystalline structure, which occurs because of the high kinetic energies of ejected atoms and ionized particles (>1 eV),
5. The similarity between the target film and the films grown,
6. A simple experiment setup,
7. Easy to produce multilayer films from different materials by successive erosion of different targets,
8. The possibility of growing ZnO films at low temperatures on flexible substrates, in order to produce flexible flat-panel displays [36].

Generally, for the growth of ZnO thin films by PLD method, a combination of UV excimer lasers KrF: =248 nm and ArF: =193 nm along with a pulsed Nd: yttrium aluminum garnet YAG laser at =355 nm is employed for the ablation of the ZnO target in an oxygen medium. In some cases, Cu-vapor laser emitting at =510–578 nm was also used for the same purpose. The common target is a cylindrical ZnO tablet made from compressed ZnO.
powder that is made from compressed ZnO powder. It has been demonstrated recently that ZnO single crystals can be used to grow high quality ZnO thin films. In general, the properties of the grown ZnO films are affected by the substrate temperature, the ambient oxygen pressure, and the laser intensity [4].

![Figure 5: Schematic diagram of a PLD system [2, 4].](image)

**3.1.3 Spray pyrolysis (SP)**

The SP method, which is one of the chemical methods for the growth of thin films, is widely used for the deposition of a wide variety of thin films. It involves spraying a (usually) aqueous solution containing soluble salts of the desired compound onto a heated substrate. The SP process utilizes endothermic (pyrolytic) decomposition of a metallic compound which is dissolved in a liquid mixture and applied to a heated substrate. By
using this method, the doping process is relatively simple and the percentage of doping in the sample is only affected by changing the concentration of the dopant in the solution. This method has the following advantages:

1. The suitability for the deposition of thin films with large areas and good reproducibility at high speeds,
2. The low consumption of materials,
3. The low cost of the material used,
4. There is no need of great quality target and vacuum at various stages.

Also, there are some disadvantages to this approach [29, 37]:

1. It is not suitable for the deposition of thin films of very small thickness,
2. Restrictions on the choice of substrates due to high process temperatures (the effects of substrate temperature on the structural, electrical, and optical properties of ZnO thin films prepared using this method have been investigated),
3. The material used to coat the surface should be soluble,
4. Edge effects (a greater thickness around the edges of the substrate),
5. The difficulty of cleaning the laborious device.

Number of researchers have utilized SP method for the deposition of pure and doped ZnO thin films [38-40]. In the recent reported work, Srivastha et. al. have fabricated ZnO thin films on glass substrate and studied the effect of substrate temperature on the morphology, defect chemistry and photoconductivity of the films [41]. The results revealed that maximum photosensitivity was observed for thin films deposited at 450 °C.

3.1.4 Molecular beam epitaxy (MBE)

The MBE technology facilitates the creation of thin films with exceptional crystalline quality and extremely low roughness at low temperatures and under extremely high vacuum (10^{-10} Torr). The reaction of atomic or molecule fluxes on a single crystal substrate that has attained the proper temperature is the basis for the method's functioning mechanism [29]. In order to generate beams of atoms or molecules with an angular dispersion, the solid source materials are put into evaporation cells. To increase growth homogeneity, the substrate is heated to the necessary temperature and continually rotated as needed.

The formation of high-quality transparent and conductive ZnO thin films was carried out by controlling the growth of the atomic layer at different temperatures on various substrates (including glass, sapphire, and polyethylene terephthalate) [42]. The source materials typically used in the growth of ZnO thin films by MBE are Zn metal and O₂. A high purity Zn metal is evaporated from an effusion cell, where the cell temperature can be changed to investigate the effect of the Zn flux on the growth rate and material properties. Moreover,
by directing an oxygen radical beam on to the film surface, a high oxidation efficiency can be achieved [4].

In general, molecular beam epitaxy has the following advantages [4, 29]:
1. Precise control of the deposition parameters,
2. In situ diagnostic capabilities, including possible controls and in situ analysis
3. A low growth rate, which allows doping at the atomic level,
4. Very precise control of the thicknesses of the thin layers,
5. The ability to control every step of the process in an automated manner,
6. No boundary layer.

4. Properties
The following section provides an overview of the structural, optical, and electrical properties of ZnO thin films, including some of the following:
1. High transparency in the visible and near infrared spectral regions,
2. Generating heavily doped sheets with a high free electron density but a low resistance
3. Good contact with the active semiconductors (absorber layers),
4. The possibility of preparing the transparent conductive oxide (TCO) layers on large areas (>1 m²) using deposition methods such as magnetron sputtering or metal-organic chemical vapor deposition (MOCVD),
5. A possibility for preparing high-quality ZnO films at low substrate temperatures,
6. Ability to prepare tailored surfaces with the desired light scattering properties for light trapping, which is particularly important for thin film solar cells made of Si.
7. Low material costs, nontoxicity, and abundance in the earth's crust.

4.1 Physical properties
The structural attributes of these thin films will be thoroughly explained in this section. Some of the primary characteristics of ZnO are shown in Table 3, as can be seen. Physical parameters may vary with the roughness of the films. Roughness of the films is an critical issue which hinders the utilization of films in device applications. In the last few decades, roughness of the ZnO films have been minimized by number of routes like varying the substrate, substrate temperature, precursors and many other experimental conditions. In general, all thin films fabricated under vacuum suffer from stress (thermal + intrinsic). Where thermal stress originates form the different thermal extinction coefficient (α) of substrate and coating materials. For example, Si substrate exhibit α of $2.5 \times 10^{-6}/°C$. Hexagonal ZnO crystal exhibits $a_{11}$ and $a_{33}$ of 6.05 and $3.53 \times 10^{-6}/°C$ respectively which are quite greater than as reported for Si [43]. This lead to the creation of tensile stress in
ZnO thin films when substrate comes to temperature from higher temperature. Secondly, intrinsic stress results from the crystallographic flaws created during thin film fabrication. Another reason for the comprehensive stress is due to reflection of oxygen and argon atoms from target. The developed stress is responsible for the change in elastic constants and thereby can change many other physical and other properties of ZnO thin films.

Table 3. Physical Parameters of ZnO [44]

<table>
<thead>
<tr>
<th>Properties</th>
<th>Parameters (Value) at 300 K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Crystalline structure</td>
<td>Wurtzite</td>
</tr>
<tr>
<td>Lattice parameters</td>
<td>a=b=3.249 Å, c=5.2042 Å</td>
</tr>
<tr>
<td>Refractive index</td>
<td>2.008-2.029</td>
</tr>
<tr>
<td>Band gap energy</td>
<td>3.37 eV, direct</td>
</tr>
<tr>
<td>Melting point</td>
<td>1975 °C</td>
</tr>
<tr>
<td>Density</td>
<td>5.606 g/cm³</td>
</tr>
<tr>
<td>Electron effective mass</td>
<td>0.28 m₀</td>
</tr>
<tr>
<td>Exciton binding energy</td>
<td>60meV</td>
</tr>
</tbody>
</table>

4.2 Electrical properties

It is challenging to test the electrical characteristics of these oxides due to the fact that the quality of the many ZnO samples available varies greatly. The concentration of charge carriers varies depending on the layer quality, but is typically approximately $10^{16}$ cm$^{-3}$. P-type atoms have a maximum of $10^{19}$ cm$^{-3}$ holes and an upper limit of $10^{20}$ cm$^{-3}$ electrons for N-type atoms. This high p-type conductivity value, nevertheless, is debatable and hasn't been demonstrated in real-world applications.

Additionally, at 300 K, the bond excitation energy of ZnO, an II-VI semiconductor, is equal to 60 meV, making ZnO an ideal material for opto-electric devices. In other words, a significant exciton binding energy and a wide and direct energy gap (3.44 eV at 4 K and 3.37 eV at normal temperature) show that effective excitonic emission in ZnO can continue at room temperature. ZnO is a suitable material for optoelectronic devices based on excitonic phenomena because excitons often have stronger oscillators than direct electron transfers to holes in direct gap semiconductors [5, 45]. Undoped ZnO is typically regarded as an n-type semiconductor. It is significant to note that ZnO's electrical resistivity can be enhanced through doping, the introduction of more zinc atoms into the interstitial space, or the establishment of oxygen vacancies. The material's electrical resistance is decreased by the presence of zinc interstitials and oxygen vacancies, which function as electron donors. A shallow donor defect called interstitial hydrogen may also be the source of the free
electrons [29]. It is crucial to remember that the preparation conditions, particularly heat treatment, have an impact on the electrical resistivity of ZnO thin films. As previously mentioned, it is simple to produce n-type ZnO by replacing zinc atoms with Group III elements (Al, Ga, In, and B) and Group IV elements (Si, Ge, and Zr), or by replacing oxygen atoms with a Group VII element (F, Cl) for p-type ZnO doping with Group I elements (Li, Na and K). It should be noted, nevertheless, that type p is more challenging to obtain than type n.[29, 46].

4.3 Optical properties

In recent times, there has been a significant amount of research conducted on the optical processes and properties of ZnO, including its transmission, absorption, optical gap, and photoluminescence. This property is a direct consequence of the material's wide band gap (about 3.37 eV at room temperature), ultraviolet luminescence (around 380 nm), and good radiative recombination. Additionally, this material's high exciton radiation at room temperature and above, which is 2.4 times as powerful as the thermal energy generated (K \( \text{B} T=25 \text{ meV} \)), is facilitated by the exciton binding energy (60 meV) of this substance [14]. These thin films can be utilised as a barrier against ultraviolet radiation as well as transparent in the visible and near ultraviolet-visible wavelength ranges because of their direct and wide band gaps and low permeability in the ultraviolet region. But since excitonic recombination is caused by the high exciton binding energy at ambient temperature, ZnO thin films could one day compete with other materials to produce UV and blue light generating devices. Consequently, epitaxial and polycrystalline ZnO thin films have shown optically pumped lasing. [29, 47, 48]. Several ZnO characteristics are listed in Table 4.

Table 4. Some optical properties of ZnO[4, 49, 50]

<table>
<thead>
<tr>
<th>Properties</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>The average transmittance (%)</td>
<td>86-92</td>
</tr>
<tr>
<td>Refractive index</td>
<td>2.008–2.029</td>
</tr>
<tr>
<td>Absorption coefficient (cm(^{-1}))</td>
<td>( \sim 10^4 )</td>
</tr>
</tbody>
</table>

Optical properties of ZnO thin films are influenced by the following factors [29]:
1. The thickness
2. The optical gap
3. The surface roughness
4. The doping
5. The crystalline quality of the film
6. The deposition method

5. Potential applications of ZnO thin films

During the last decade, ZnO material has been studied extensively and employed in micro-scale and nano-scale devices. As stated previously, ZnO is a very versatile substance which can be applied in many micro and nano systems. A combination of its wide band gap, piezoelectric, electrical, and optical properties make ZnO thin films unique and crucial for a wide range of technological applications today and in the future. With its various properties, ZnO is a promising candidate for applications in spintronics, medical applications such as sun screen lotions and ointments, solar cells, UV light emitting diodes, sensors, surface acoustic devices, transparent electronics such as transparent thin film transistors and transparent conductive electrodes [10].

5.1 Future potential applications for thin films

ZnO thin films are the potential source of diverse application in numerous areas of technology. Gas sensors are one of the most widespread applications for ZnO thin films. Under temperature-controlled conditions, this thin film can detect a broad array of gases. The fabrication of ZnO gas sensors utilizes a wide range of materials, including thin films and thick films. Some metal ions that can be doped with ZnO are indeed being considered in order to enhance the performance and gas sensing capabilities of the thin film of ZnO. Some metal ions that can be doped with ZnO are being taken into account in order to enhance the efficiency of the thin film of ZnO and its gas sensor technologies. In this regard, ZnO thin films doped with aluminum oxide are effective at detecting triethanolamine, and ZnO thin films doped with indium, copper, and iron are effective at detecting ethanol [51].

Also, transparent semiconducting ZnO thin films have attracted considerable interest due to their excellent properties, such as high infrared reflectance, excellent chemical stability, UV absorption, and high transparency in the visible region, which makes ZnO suitably suitable for many optics and electronics applications, including solar cells, flat displays, and chemical sensors. Further, ZnO is non-toxic and exhibits a strong tendency to grow in a c-axis direction. The versatile nature of ZnO makes it suitable for using in a verity of solid state electronic devices [52]. Prakash et al. Prepared ZnO:Tb\(^{3+}\) embedded polystyrene nanocomposite thin film. ZnO:Tb\(^{3+}\) composite films were found to have a homogeneous distribution of nanophosphor nanoparticles, and the photoluminance characteristics improved when the nanophosphor content was raised. In addition, the photoluminance investigation of the ZnO:Tb\(^{3+}/PS\) films that was dependent on temperature indicated that the photoluminance intensity dropped with rising temperature owing to thermal quenching. These results showed that the ZnO:Tb\(^{3+}\) nanophosphor inserted in the ZnO:Tb\(^{3+}/PS\)
nanocomposite film kept their distinctive luminous capabilities without a noticeable spectrum shift, emphasizing their useful applications in solid-state lighting [53].

Use of ZnO's piezoelectric properties in mass-sensitive sensors that focus primarily on the acoustic wave phenomenon is another application. They function similarly to bulk acoustic wave devices and can be used for chemical and biological monitoring. The device resonates at its resonance frequency, which is regulated by the amount of external mass associated to the resonator. This signifies that the frequency changes and the mass that is loaded on the device can be accurately determined when there is a mass-loading effect on the bulk acoustic wave resonator [54]. Furthermore, ZnO is also considered a good choice for UV detectors due to its high on/off current ratio, fast response and recovery, large photo-response current, and wavelength selectivity [55]. Aside from sensor applications, ZnO is also highly sought after for thin film transistors, light emitting diodes, gas sensing, solar cells, surface acoustic wave receivers, and energy harvesting devices. There are several applications associated with each of the properties of ZnO thin films.

ZnO thin film gas sensing behaviour is heavily influenced by O\textsubscript{2}/Ar ratios, with higher values indicating greater sensitivity to diverse gases due to reduced carrier density. Without an external bias, ZnO films modify their resistance in two distinct ways depending on the oxygen partial pressure of the surrounding gas and the film's temperature. Bulk dispersion cannot be ignored, even at the reduced temperatures at which these thin film sensors operate. p-ZnO thin films doped with AlN and AlAs are synthesized using RF magnetron sputtering for the purpose of gas sensing. By applying a relatively low potential of 1V, the manufactured p-ZnO films were subjected to various ppm of NH\textsubscript{3} at room temperature, 100 °C, and 150 °C. Compared to Al mono-doped n-ZnO film, both films demonstrated superior responsiveness to NH\textsubscript{3} at ambient temperature. The AlAs co-doped p-ZnO thin film outperforms both films in terms of responsiveness and response/recovery time by a wide margin throughout a wide temperature range. This highlights the fact that AlAs co-doped p-ZnO thin films may be employed in gas sensing applications with performance improvement [56]. Chang et al. investigated the influence of film thickness on sensor response of ZTF with thicknesses ranging from 65 nm to 390 nm produced by RF reactive sputtering on a SiO\textsubscript{2}/Si wafer. The film thickness of 65 nm yielded the highest sensitivity and the quickest reaction [57, 58]. Batra et al. [59] fabricated Al:ZnO thin film for the development of effective cholesterol biosensor. The produced ChOx/Al:ZnO/Pt/glass bioelectrode has a good sensitivity (173 μAmM\textsuperscript{−1} cm\textsuperscript{−2}) and linear performance in a range of cholesterol concentrations (0.6-12.9 mM) with sufficient certainty. Immobilized ChOx has strong attraction for cholesterol, as evidenced by a reduced Michaelis Menten constant (2.53 mM). These findings provide strong evidence that RF-sputtered Al:ZnO thin films can serve as an efficient substrate for the immobilization of biomolecules such as ChOx in the detection of cholesterol. Bott et al. [60] used a steady voltage source to measure the conductivity shift of polycrystalline ZnO films and compacted discs in air mixes of CO, CH\textsubscript{4}, and H\textsubscript{2} from 300 to 500 degrees Celsius. The conductance of polycrystalline oxide films was shown to be more sensitive to CH\textsubscript{4} than that of single crystals [61].
Photovoltaic cells, often known as solar cells, are used to transform sunlight into electricity. The most well-known solar cells contain a massive area p-n junction as their bare minimum component. Whenever exposed to light, the electron-poor p-type semiconductor layer of a p-n junction, such as ZTF, is attracted to the electron-rich n-type semiconductor layer. The highest optical transmittances of 93% can be found in ZnO:Al films, making them ideal for use in solar cells [62, 63]. Ali et al synthesized ZnO thin film by thermal evaporation technique. There is research into the (current-voltage) properties of a ZnO/p-Si heterostructure. There was found to be a 50mA short circuit current, 0.45 V open circuit voltage, 36% effectiveness, and a fill factor of 6%. Findings indicate that ZnO/p-Si heterostructures may be useful in solar cells applications [64]. Using RFMS, coatings of fluorine- and aluminum-co-doped ZnO (FAZO) were formed on glass substrates. The optical transmittance spectra of FAZO films deposited a are displayed in Figure 6(a) in the range between 300 and 1200 nm. Figure 6(b) shows the optical property of prepared thin film and with increase the temperature absorption edges are changes. FAZO films have a lower transmission edge than ITO and FTO films, but their greater transmittance from 400 to 1200 nm makes them appropriate as transparent electrodes for high-efficiency solar cells. Figure 6(c) is a diagram of the structure of the perovskite solar cells with FAZO, FTO, and ITO electrodes. Apart from the clear electrode, the devices were made in the same way, and the J–V curves for each one is presented in Figure 6(b). Perovskite solar cells made with the FAZO film were more efficient at turning light into electricity than reference cells made with F-doped SnO2 and Sn-doped In2O3, showing that it can be used in high-efficiency solar cells [65]. Agrawal et al. employed responsive DC magnetron sputter with variable oxygen flow rates to produce a ZnO thin film of 600 nm thickness on a glass surface, and then annealed the film subsequently. Film was deposited in 21% oxygen environments are transparent at 85% and exhibit a resistance of 15 x 10^3 cm after being annealed at 300 degree Celsius [66]. Aranovich et al. [67], studied the electrical & photovoltaic characteristics of a hetero-junction formed by spray pyrolyzing ZnO sheets over single crystal p-type CdTe. When the cells are exposed to sunlight, the open-circuit voltage is 0.54 V and the short-circuit current is 19.5 mA/cm^2. This means that the cells are 8.8% efficient. Effectiveness of a p-n heterojunction CuInS2/ZnO solar cell is affected by its deposition parameters, resistivity, and annealing temperature.
ZnO-based LEDs are divided into two categories homojunction LEDs or heterojunction LEDs. Using the PLD method, Rogers et al. [68] grew a thin layer of ZnO on front of a p-type GaN layer in an oxygen environment, which allowed them to examine nonlinear rectification performance at a ZnO band gap equivalent forward bias voltage of 3.3 V. On a ZnO wafer, Guo et al. [69] produced a ZnO homostructural diode with a p-ZnO: N/n-ZnO junction. Created by combining an N-Al co-doped p-type ZnO film with an Al-doped n-type ZnO layer, ZnO p-n homo-junctions were shown by Lu et al. [70]. Lim et al. [71] show how to make UV-emitting ZnO homo-junction LEDs by developing P-doped p-type ZnO atop Ga-doped n-type ZnO. Das et al., n-type SnO$_2$:F (U-type Asahi) thin films and widely obtainable ZnO:Ga. ZnO:Ga film smooth surface and few spikes imply good surface structure. In order to be most effective in OLED applications, 170 nm ZnO:Ga films need to have an electrical resistivity of $9.6 \times 10^{-5}$ Ωcm, a sheet resistance of less than 5.6 Ω/cm, an optical transparent of better than 90%, a smooth surface, and fewer spikes [72]. These applications are shown in the table below (as shown in Table 4).
## Table 4. The applications related to the properties of ZnO thin film

<table>
<thead>
<tr>
<th>Property</th>
<th>Advantages associated with this properties</th>
<th>Application</th>
<th>Ref.</th>
</tr>
</thead>
</table>
| 1. Direct and wide band gap (3.44 eV at low temperature and 3.37 eV at room temperature) | • higher breakdown voltages  
• ability to sustain large electric fields  
• lower electronic noise  
• high-temperature and high-power operation | optoelectronics field (in the blue / UV region) light emitting diodes, laser diodes and photo detectors | [4, 8, 73] |
| 2. Large exciton binding energy (60 meV) | • efficient excitonic emission in ZnO can persist at room temperature and higher  
• the stability against thermal dissociation of excitons | optical devices based on excitonic effects | [5, 14] |
| 3. Large piezoelectric constants | • Piezoelectric/ pyroelectric properties created by combining two properties (1. low symmetry of the wurtzite crystal structure, 2. Large electro-mechanical coupling) | sensors, transducers and actuators | [74] |
| 4. Strong luminescence (In the green–white region of the spectrum) | • The broad peak of the emission spectrum =495 nm [half-width of 0.4 eV] | fluorescent displays and field emission displays | [75] |
| 5. Strong sensitivity of surface conductivity to the presence of adsorbed species | • The high surface sensitivity of the ZnO thin films exposed the various gases due to the conductivity of ZnO thin films | cheap smell sensor | [76] |
| 6. High thermal conductivity | • in order to increase the thermal conductivity | additive a substrate for homoepitaxy or heteroepitaxy | [77] |
5.2 **Nanotechnology's thin-film revolution**

Nano-ZnO can display the effects of quantum confinement, which include size reduction and an increase in surface area or the surface to volume ratio. This opens up the possibility for the development of new physical, optical, chemical, and chemical characteristics. This encouraged the researchers to fabricate ZnO as a nanostructure material using a variety of techniques and in a diverse range of shapes, comprising nanoparticles, nanowires, nanorods, and nanotubes, nanoplates, etc. [78].

ZnO nanostructures, in particular ZnO thin films, have been significantly used in a broad range of industries and applications as nanotechnology has advanced. It has been shown that this material's nanostructures can be fabricated efficiently at low temperatures utilizing low-cost and low-temperature methodologies [24]. ZnO has a high excitation binding energy and a significant band gap. Additionally, because of its advantageous piezoelectric properties, it has the potential to generate nanoscale electromechanical coupling devices, such as sensors and actuators [55].

For instance, due to its biocompatibility and lack of toxicity to living tissues, ZnO is of tremendous interest in biological applications. Numerous studies have been conducted on this material's nanostructures in a variety of medical specialties, from diagnosis to treatment. Because of their stability, high electrochemical activity, quick electron transfer, biocompatibility, and simplicity of manufacture, ZnO-based biosensors are extensively used [79]. In light of this, ZnO nanostructures are currently the subject of intensive research for the creation of future spintronic, potho-electronic, and medicinal devices.

**Conclusion**

ZnO thin films have already been briefly covered in this chapter. ZnO and its thin film have been the focus of several research studies due to their significance in industry and technology. Additionally, piezoelectric, semiconducting, and photoconducting qualities can be found in ZnO thin films. They have a high dielectric constant, a wide forbidden energy gap, and a high nonlinear optical coefficient in addition to having high electrical and optical transparency. High piezoelectric, acoustic-optical, and electro-optical coefficients are provided by ZnO films. ZnO films are employed in the creation of surface acoustic wave devices, numerous electro-optical devices, integrated optics, gas and biological sensors, pressure sensors, and photovoltaic solar cells due to the aforementioned qualities as well as their inexpensive cost. ZnO thin films, however, can be synthesized using a variety of deposition techniques. Sputtering, CVD, and SP processes, on the other hand, have been exploited more extensively due to their simplicity and low cost for high-quality applications. Additionally, some of the structural, electrical, and optical characteristics of ZnO films were investigated.
Aknowlgement

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Reference


