Investigation of Optical Properties of Zinc-Oxide Thin Films Deposited on Various Substrates: A Simulation Study

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Abstract: ZnO is an important II-VI semiconductor material for devices with possible applications such as piezo-electric transducers, spin functional devices, gas sensors, transparent electronics in solar cell, UV light emitters, surface acoustic waves and smart windows. In this study, the optical properties of ZnO thin films deposited on various substrates (quartz, sapphire, KDP, fused silica, BK7 glass, BaF₂, GeO₂ and diamond) have been investigated. In order to investigate the optical properties, a simulation program was developed. Famous computing language “MATLAB” was chosen as the coding language because of its high ability to generate ideal graph. The optical properties (refractive index, transmittance, extinction coefficient, absorption coefficient etc.) were studied as a function of wavelength in the range from 330nm to 1110nm. The refractive indices, extinction coefficients, absorption coefficients, dielectric constants were also studied. The transmittance spectra of ZnO thin films deposited on various substrates were investigated for different thicknesses (150nm, 400nm, 800nm) of the ZnO films. The effects of interference on transmission spectra were also investigated. Fused silica and BaF₂ are the best substrates for the deposition of the ZnO thin film and give good reason for its applications in opto-electronic devices.

Keywords: Optoelectronics, Thin Film, ZnO, Simulation, Matlab

1. Introduction

Zinc Oxide (ZnO) is an important II-VI semiconductor material, wide direct band gap semiconductor of around 3.4eV with high exciton binding energy (60 MeV) even at room temperature [1, 2]. It also has several favorable properties including good transparency, high electron mobility, wide band gap and strong room temperature luminescence. ZnO adopts the four-time coordinated wurtzite phase in its ground state that belongs to the hexagonal h₆ symmetry group and thus plays an important role in many fundamental and technological applications [3]. One main attractive feature of ZnO is the ability to band gap tuning via divalent substitution on the cation side heterostructures [4]. Zinc Oxide (ZnO) is one of transparent conducting oxide (TCO) materials whose thin films attract much interest because of typical properties such as high chemical and mechanical in hydrogen plasma, high optical transparency in the visible and near infrared region [5–7]. Due to this properties ZnO is a promising material for electronic or optoelectronic applications such as solar cells (anti-reflecting coating and transparent conducting materials), gas sensors, liquid crystal displays, heat mirrors, surface acoustic devices etc [8–10]. In addition to the traditional applications ZnO thin films could also be used in integrated optics [11]. To cram these optoelectronic devices it is obligatory to identify the optical properties. In designing modern optoelectronic and optical devices it is important to know the thickness, refractive index and absorption coefficient as a function of wavelength to predict the photoelectric behavior of a device [12]. Besides, in order to develop ZnO thin films for optoelectronic devices with superior performance it is also
indispensable to expose the character and consequences of substrates.

In 2013, N. Bouchenak Khelladi and N. E. Chabane Sari studied the optical transmission properties of ZnO thin film deposited on BK7 glass and Sapphire.

In this work the optical properties of ZnO thin film deposited on about eight various substrates for different thicknesses (150nm, 400nm, 800nm) were scrutinized using a simulation program “Matlab”.

2. Materials & Methods

The brief model of the thin absorbent films on a transparent thick substrate is shown in Figure 1. Here d and n is the thickness and refractive index of the thin films respectively. The substrate has a thickness of the several orders of magnitude larger than ‘d’ and the refractive index is ‘S’. The index of surrounding air is defined as \( n_o = 1 \). \( R_1 \) is the intensity of the reflected light on the interface between air and film, and \( R_2 \) is the reflection on the interface between the film and substrate in middle. Reflection at the interface between the substrate and air under substrate is not considered here [13]. In this work quartz, sapphire, KDP, fused silica, BK7 glass, BaF\(_2\), GeO\(_2\) and diamond were used as substrates.

The Sellmeier equation for the refractive index, n, of ZnO thin film as a function of wavelength is given by,

\[
n(\lambda) = A + \frac{B\lambda^2}{\lambda^2 - C_1} + \frac{D\lambda^2}{\lambda^2 - C_2} + \frac{E\lambda^2}{\lambda^2 - C_3}
\]  

(1)

where A, B, C, D and E are fitting parameters, \( \lambda \) is the wavelength of light (nm). Fitting parameters calculated at different powers for different thickness, as deposit varies significantly [1]. The Sellmeier coefficients of ZnO are given in table 1.

From these fitting parameters the refractive index of ZnO has been studied using the MATLAB programming software. The refractive index of the substrate,

\[
S^2(\lambda) = 1 + \frac{A_1\lambda^2}{\lambda^2 - \lambda_1^2} + \frac{A_2\lambda^2}{\lambda^2 - \lambda_2^2} + \frac{A_3\lambda^2}{\lambda^2 - \lambda_3^2}
\]  

(2)

The Sellmeier coefficients of BK7-glass and other substrates are given below in table 2 and 3.

### Table 1. Fitting parameter by the method VASE of Sellmeier model for Zinc Oxide [14].

<table>
<thead>
<tr>
<th>A</th>
<th>B</th>
<th>C (nm)</th>
<th>D</th>
<th>E (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.0065</td>
<td>1.5748 ×10(^6)</td>
<td>1 ×10(^6)</td>
<td>1.5868</td>
<td>260.63</td>
</tr>
</tbody>
</table>

The Sellmeier equation for the refractive index of ZnO thin film as a function of wavelength is given by,

\[
n(\lambda) = A + B\lambda^2 + \frac{D\lambda^2}{\lambda^2 - E^2}
\]  

(1)

where A, B, C, D and E are fitting parameters, \( \lambda \) is the wavelength of light (nm). Fitting parameters calculated at different powers for different thickness, as deposit varies significantly [1]. The Sellmeier coefficients of ZnO are given in table 1.

From these fitting parameters the refractive index of ZnO has been studied using the MATLAB programming software. The refractive index of the substrate,

\[
S^2(\lambda) = 1 + \frac{B_1\lambda^2}{\lambda^2 - C_1} + \frac{B_2\lambda^2}{\lambda^2 - C_2} + \frac{B_3\lambda^2}{\lambda^2 - C_3}
\]  

(2)

The Sellmeier coefficients of BK7-glass and other substrates are given below in table 2 and 3.

### Table 2. Sellmeier coefficients for BK7 glass [14].

<table>
<thead>
<tr>
<th>Glass</th>
<th>( B_1 )</th>
<th>( B_2 \times10^{-1} )</th>
<th>( B_3 )</th>
<th>( C_1 \times10^{12} )</th>
<th>( C_2 \times10^{14} )</th>
<th>( C_3 \times10^{16} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>BK7</td>
<td>1.0396</td>
<td>2.3179×10(^{-1})</td>
<td>1.0104</td>
<td>6.0069×10(^{-2})</td>
<td>2.0017×10(^{4})</td>
<td>1.0356×10(^{8})</td>
</tr>
</tbody>
</table>

### Table 3. The Sellmeier coefficients of various substrates were given below [15].

<table>
<thead>
<tr>
<th>Material</th>
<th>( A_1 )</th>
<th>( A_2 )</th>
<th>( A_3 )</th>
<th>( \lambda_1 ) (nm)</th>
<th>( \lambda_2 ) (nm)</th>
<th>( \lambda_3 ) (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz, n(_o)</td>
<td>1.35400</td>
<td>0.010</td>
<td>0.9994</td>
<td>92.612</td>
<td>1070</td>
<td>9850</td>
</tr>
<tr>
<td>Sapphire</td>
<td>1.023798</td>
<td>1.058264</td>
<td>5.280792</td>
<td>61.4482</td>
<td>110.7</td>
<td>17926.56</td>
</tr>
<tr>
<td>KDP n(_o)</td>
<td>1.25400</td>
<td>0.0100</td>
<td>0.0992</td>
<td>96.46</td>
<td>6977.7</td>
<td>5984.8</td>
</tr>
<tr>
<td>Fused Silica</td>
<td>0.696749</td>
<td>0.408218</td>
<td>0.890815</td>
<td>69.066</td>
<td>115.662</td>
<td>46386.42</td>
</tr>
<tr>
<td>BaF(_2)</td>
<td>0.63356</td>
<td>0.506762</td>
<td>3.8261</td>
<td>57.789</td>
<td>109.681</td>
<td>11841.931</td>
</tr>
<tr>
<td>GeO(_2)</td>
<td>0.80686642</td>
<td>0.71815848</td>
<td>0.85416831</td>
<td>68.972606</td>
<td>153.96605</td>
<td>11841.931</td>
</tr>
<tr>
<td>Diamond</td>
<td>0.3306</td>
<td>4.3356</td>
<td></td>
<td>175</td>
<td>106</td>
<td></td>
</tr>
</tbody>
</table>
From these coefficients with the Matlab the refractive indices of the substrates have been calculated.

The extinction coefficient of ZnO is given by the following relation.

\[ k(\lambda) = F_k \lambda e^{-G_k \left( \frac{1}{n_k} - \frac{1}{x} \right)} \]  

(4)

The Cauchy parameters of ZnO are given below in table 4.

<table>
<thead>
<tr>
<th>Fk (nm(^{-2}))</th>
<th>Gk (nm)</th>
<th>Hk (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0178</td>
<td>7327.1</td>
<td>337.87</td>
</tr>
</tbody>
</table>

With the help of equation (2.4) and Cauchy’s parameter, the extinction co-efficient of ZnO thin film has been calculated.

After that the value of \( R_1 \) and \( R_2 \) has been calculated by the following equation

\[ R_1 = \frac{(n-1) + k^2}{(n+1) + k^2} \]  

(5)

\[ R_2 = \frac{(s-n)^2}{(s-n)^2} \]  

(6)

The absorption coefficient of ZnO thin film is given by the following equation [14]

\[ \alpha(\lambda) = \frac{4\pi k}{\lambda} \]  

(7)

The expression of the transmission of ZnO thin films deposited on different substrates is given by [13],

\[ T(\lambda) = T_0(\lambda) - 2\sqrt{R_1 R_2} \cos[\delta(\lambda)] \]  

(8)

\[ \delta(\lambda) = 2\pi \frac{2nd}{\lambda} + \pi \]  

(9)

In equation (8), \( T_0(\lambda) \) is considered to be the term of transmission with no interference effect. The thicknesses of the films were taken 150nm, 400nm, 800nm. For different thicknesses of ZnO thin films deposited on various substrates, the transmittances have been determined by Matlab program. On the other hand, the transmission spectrum can be divided into two terms. They are (1) non-interference term (2) interference effect term.

The transmission of all ZnO thin film deposited on different substrates have been determined from the following formula [14],

\[ \alpha(\lambda) = \frac{1}{d} \ln \left( \frac{A}{B T_0} \right) \]  

(10)

where, \( A = 16n^2 s \) & \( B = (1 + n)^{3/2}(n + s^2) \)

From the equation (x), transmittance (without interference effect), \( T_0 \) for all ZnO thin films have been determined by Matlab program for different thicknesses (150nm, 400nm, 800nm) of the film.

The transmittance (with interference effect) can be expressed as [13],

\[ T(\lambda)_i = T(\lambda) - T_0(\lambda) \]  

(11)

From the equation (xi), transmittance (with interference effect), \( T(\lambda) \), for all ZnO thin films have been determined by Matlab program for different thickness (150nm, 400nm, 800nm) of the film.

3. Results and Discussion

The optical properties of ZnO thin films were investigated as a function of wavelength in the region of 330-1110nm.

3.1. Refractive Index

The variation of refractive indices with wavelength for ZnO and different substrates are shown in Figure 2(a) to 2(i).
Figure 2. Variation of refractive index as a function of wavelength for (a) ZnO, (b) quartz, (c) sapphire, (d) KDP, (e) fused silica, (f) BK7-glass, (g) BaF₂, (h) GeO₂, and (i) diamond.
It was observed that the refractive indices decreased with the increase in wavelength in the range from 330nm to 1110nm for all the substrates and ZnO thin films. It was evident from graph that the value of refractive index went in a single series of stages from one point to another point in the higher wavelength region. At 330nm the refractive index is 1.57 for quartz, 1.80 for sapphire, 1.54 for KDP and BK7 glass, 1.48 for fused silica, 1.49 for BaF$_2$, 1.66 for GeO$_2$ and 2.5 for diamond. By comparing among all substrates it was observed that diamond exhibits highest refractive index and fused silica exhibits lowest refractive index.

### 3.2. Absorption Coefficient and Extinction Coefficient

It was observed that absorption coefficient decreased with the increase in wavelength as shown in Figure 3(a). From 330nm to 400nm wavelength region the absorption coefficient decreased sharply and after that the value of absorption coefficient was almost constant in the higher wavelength region.

![Figure 3. Variation of (a) absorption coefficient and (b) extinction coefficient as a function of wavelength for ZnO thin film.](image)

From the Figure 3(b) it was clear that the extinction coefficient decreased with the increase in wavelength. The rise and fall in the extinction coefficient is related to the absorption of light. The value of extinction coefficient was high at UV region and moved towards zero above 410nm which indicated that there was tiny absorption in the visible region.

### 3.3. Transmission

Figure 4(a) to 4(h) shows the transmission spectra of ZnO thin films of different thicknesses deposited on various substrates.
It was observed from Figure 4(a) to 4(h) that the films exhibit transmittance between 80% and 95% for wavelengths greater than 550nm. The films were highly transparent in the visible region and near infrared region but low in the ultraviolet region. According to the value of transmittance the observed transmission spectra can be divided into four regions.

(i) Transparent region (500-1110) nm: In the transparent region the value of absorption was near to zero and the transmittance was about 80% to 95% which was controlled by the refractive indices of ZnO and substrates.

(ii) Weak absorption region (430-500) nm: The value of absorption was small and the transmission started to decrease in this region.

(iii) Medium absorption region (370-430) nm: In the medium absorption region the value of absorption was large and hence the transmission was low compared to absorption.

(iv) Strong absorption region (below 370 nm): For the effect of absorption the transmission decreased severely in this spectral region.

The rise and fall or peaks in the transmission spectra occurred due to the effect of reflectance and interference of the incident light. At the wavelength 610nm, quartz showed transmittance about 92%, sapphire about 92%, KDP about 93%, BK7 glass about 92%, fused silica and BaF$_2$ about 94%, GeO$_2$ about 90% and diamond showed about 79%. By comparing among these substrates it was found that fused silica and BaF$_2$ exhibited highest transmittance of about 94% and diamond exhibited lowest transmittance of about 79% in the visible region. It can be concluded that fused silica and BaF$_2$ are the best substrates for the deposition of ZnO thin film.

3.4. Transmission Without Interference Effect

The optical transmission spectra without interference effect of the ZnO thin films of different thicknesses deposited on various substrates as a function of wavelength ranging between 330nm to 1110nm were represented from Figure 5(a) to 5(h). These figures showed excellent transmittance across the visible and near infrared regions. The films showed transmittance between 70% to 85% for wavelengths greater than 450nm but low for wavelengths smaller than 400nm. In the spectra there were no peaks which were the evidence of optical transmission spectra without interference effect.
Figure 5. Transmission spectrum without interference effect of ZnO thin film for different thicknesses deposited on (a) quartz substrate, (b) sapphire substrate, (c) KDP substrate, (d) fused silica substrate, (e) BK7 glass substrate, (f) BaF$_2$ substrate, (g) GeO$_2$ substrate, and (h) diamond substrate.
At wavelength greater than 590nm, the transmittance was about 83% for quartz, 81% for sapphire, 83% for KDP and BK7 glass, 84% for fused silica and BaF$_2$, 82% for GeO$_2$ and 73% for diamond. Fused silica and BaF$_2$ exhibited highest transmittance and diamond exhibited lowest transmittance in the visible region among all these substrates.

3.5. Transmission with Interference Effect

The optical transmission spectra with interference effect of the ZnO thin films of different thicknesses (150nm, 400nm, 800nm) deposited on various substrates as a function of wavelength ranging between 330nm to 1110nm were represented from Figure 6(a) to 6(h).
It was observed that there were interference peaks with maxima and minima. These interference fringes were produced due to multiple reflection of light between the top surface of the film which was in contact with air and the bottom surface of the film that was in contact with the substrate. From figures it was also seen that the number of peaks increased with the increase in thickness. These occurred due to interference effect. So it can be said that the effect of interference increases with the increase in thickness.

4. Conclusions

In this work the optical properties of the ZnO thin films have been investigated including the refractive indices, absorption coefficient, extinction coefficient and transmission spectra of ZnO thin film for different thicknesses (150nm, 400nm, 800nm) deposited on about eight different substrates. The refractive indices, extinction coefficient and absorption coefficient decrease with the increase in wavelength and are almost consistent in the higher wavelength region. By comparing among various substrates it is observed that fused silica exhibits lowest refractive index of 1.48 and diamond exhibits highest refractive index of 2.51 at 330 nm. This indicates that light travels faster through fused silica than other substrates.

The films were found to exhibit high transmittance (75-95)% in the visible and near infrared region. The optical transmission spectra showed four major regions: strong absorption region (below 370nm), medium absorption region (370-430) nm, weak absorption region (430-500) nm, and the transparent region (500-1110) nm. Among various substrates fused silica and BaF$_2$ exhibit highest transmittance of about 94% at 610 nm.

Moreover, the non-interference and interference effect were observed in transmission spectra. In the non-interference term the films showed transmittance about (70-90)% in the range (450-1110) nm. There were no peaks in the transmission spectra because here the effect of reflectance and interference is totally neglected. In the interference effect term there exist maxima and minima which are the evidence of interference effect. This interference is due to the multiple reflections at the interface of films/substrate and at the interfaces of film/air. Moreover, the number of peaks increases with the increase of thickness. These also happen due to interference effect. Finally, it can be concluded that fused silica and BaF$_2$ are the best substrates for the deposition of ZnO thin film in respect of highest transmittance and low refractive indices.

The low absorbent and high transparent materials are very important for optoelectronic applications. Thus, these studies are very much expected to aid in the design and selection of proper substrate of ZnO thin film for the potential application of optoelectronic devices.

References


