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ZnO - WIDE BANDGAP SEMICONDUCTOR AND POSSIBILITIES OF ITS APPLICATION IN OPTICAL WAVEGUIDE STRUCTURES

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Abstract

The paper presents the results of investigations concerning the application of zinc oxide - a wideband gap semiconductor in optical planar waveguide structures. ZnO is a promising semiconducting material thanks to its attractive optical properties. The investigations were focused on the determination of the technology of depositions and the annealing of ZnO layers concerning their optical properties. Special attention was paid to the determination of characteristics of the refractive index of ZnO layers and their coefficients of spectral transmission within the UV-VIS-NIR range. Besides that, also the mode characteristics and the attenuation coefficients of light in the obtained waveguide structures have been investigated. In the case of planar waveguides, in which the ZnO layers have not been annealed after their deposition, the values of the attenuation coefficient of light modes amount to $\alpha \approx 30$ *dB/cm*. The ZnO layers deposited on the heated substrate and annealed by rapid thermal annealing in an N_2 and O_2 atmosphere, are characterized by much lower values of the attenuation coefficients: $\alpha \approx 3$ *dB/cm* (TE0 and TM0 modes). The ZnO optical waveguides obtained according to our technology are characterized by the lowest values of the attenuation coefficients α encountered in world literature concerning the problem of optical waveguides based on ZnO. Studies have shown that ZnO layers elaborated by us can be used in integrated optic systems, waveguides, optical modulators and light sources.

Keywords: Wide band gap oxide semiconductors: ZnO, integrated optics structures: planar waveguides.

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1. Introduction

Elements and systems of integrated optics are widely applied, among others as optical waveguides, light sources, light detectors, optical modulators, optical filters as well as optical sensor structures [1–4]. Of special interest are optical sensor structures sensitive to some selected gaseous environments [1,5,6,7]. The range of application of gas sensors based on systems of integrated optics comprises many domains, from industry to medicine and protection of the environment [8–9]. An advantage of optical waveguide sensors, including photonic sensors is, among others, a wide group of gaseous substances which can be determined by means of them [8,10]. Most important, however, is the fact that optical and photonic sensors are resistant to disturbances of electromagnetic fields and can operate in explosive environments [11–12]. The structure of a photonic gas sensor comprises optic planar or strip waveguides, input and output couplers (for the input and output light) and a sensor layer deposited on the waveguides, sensitive to the given gaseous environment. All of them are located on the same substrate. Of essential attraction is getting such a material which simultaneously plays both the role of a waveguide layer and the sensor layer. Due to its optical and chemical properties zinc oxide ZnO seems to be just such a material [13–17].

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The physical properties, including optical ones, of ZnO layers depend substantially on their internal structure, which are conditioned by the method and parameters of their manufacture and their thermal treatment [18]. Zinc oxide is characterized by a considerable value of refractive index $n \approx 2$ [19–21]. This material is transparent in the visible range – its optical absorption edge is on the level of about $\lambda \approx 380\ nm$, and the width of the energy band gap is about $E_g \approx 3.3\ eV$ [22–23]. The aim of this work was to present the results of investigations concerning the optical properties of ZnO layers deposited by means of reactive cathode sputtering. The ZnO structures are planned to be applied as photonic gas sensors, including sensors of NH_3 and NO_x in atmospheric air [24–26].

2. Experimental Details

Investigations concerning the properties of ZnO layers were divided into several stages. These investigations were taken up in order to gain extensive knowledge about the properties of zinc oxide ZnO layers and the ways of their applications in systems of integrated optics (including optical sensor structures sensitive to some given gaseous mediums).

For these reasons, the optical properties of the ZnO layers ought to be determined. The values of the refractive index were determined ellipsometrically, applying the spectral ellipsometer (SENTECH SE850). The spectral transmissions were tested by means of a measurement system consisting of a spectrometer (HR 2000+ES), the light source (DT-mini-2-GS), and a system adjusting the investigated structures. The characteristics of spectral transmission were measured in the optical range from ultraviolet to near infrared. Next, the mode properties of the ZnO optical planar waveguides were investigated, first of all - the light attenuation of the respective modes. The waveguide properties of the ZnO layers (modal characteristics) were investigated making use of a measurement system comprising a semiconductor laser with a wavelength of $\lambda = 677\ nm$, a goniometer, the system polarizing the beams of light, a lock-in amplifier (and a computer). The test stand for the determination of the mode characteristics has been presented schematically in Fig. 1a.

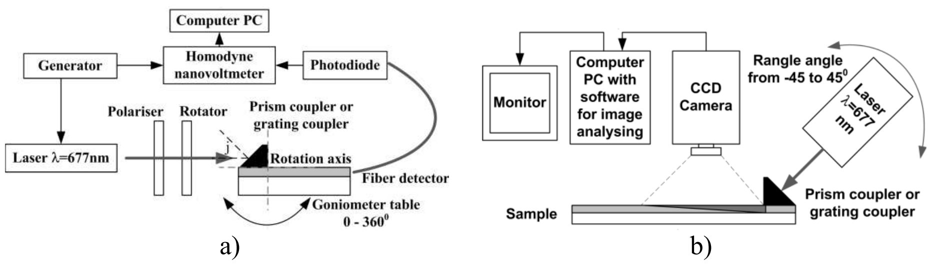

Fig. 1. Scheme of test stands for determination of the mode characteristics a) and the attenuation coefficients of the optical mode in the optical planar structure b).

The test stand for the determination of the attenuation coefficient α comprises a semiconductor laser ($\lambda = 677\ nm$), the optical system polarizing the light beam and a CCD camera recording the propagating optical beam (modes). The diagram of the test stand for the determination of the attenuation coefficient of light is to be seen in Fig. 1b.

During the transmission of light in the optical planar waveguide the propagating modes are attenuated and scattered. Assuming that changes of the intensity of the scattered light are proportional to the changes of optical power propagated in the given mode, we can determine the attenuation coefficient of light α in the planar waveguide concerning the given mode [19,21]. Recording the scattered light by means of a camera permits to determine the

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coefficient of attenuation α [19]. The input of light into a waveguide may be realized by means of a grating coupler [24]. In the presented investigations , light was introduced into the waveguide structure by means of a prism coupler, made of BGO crystal (with a refractive index of $n = 2.54$), which permitted to excite the waveguide modes selectively.
It is much easier to use a prism to excite optical modes in a planar waveguide than to apply a Bragg's grating coupler. Prism couplers are often used to study the transmission properties of planar waveguides. The solution using a prism coupler causes that the waveguide structure is not a planar one but a three-dimensional structure. The value of the effective refractive index Neff in the system with a prism coupler is determined basing on the measured mode characteristics, using the equation [25]:

$$N_{eff}(\Theta) = \sin\delta\sqrt{n_p^2 - n_c^2 \cdot \sin^2\Theta} + n_c \cdot \cos\delta \cdot \sin\Theta . \tag{1}$$

where: Θ - angle of incidence of the beam of light onto the prism, determined in relation to the normal direction; δ - breaking angle of the prism; n_p – refractive index of the prism; n_c – refractive index of the environment (medium surrounding a waveguide).

The ZnO layers were investigated also applying a scanning electron microscope with field emission (NEON 40, produced by Carl Zeiss Ltd.).

3. The technology of manufacturing ZnO layers

The optical properties of ZnO waveguides, and especially the attenuation of optical modes in them depend on the used technology. The ZnO layers were manufactured at the Institute of Electron Technology in Warsaw by means of reactive cathode magnetron sputtering. The waveguide layers were deposited from a ceramic ZnO target (99.99% purity) in an atmosphere which was a mixture of oxygen (30%) and argon (70%). The sputtering processes were run in the RF mode in the following conditions: the current of the cathode amounting to $I_c = 140\ mA$, the total pressure to $P_{Ar+O2} = 1x10^{-2}\ mBar$, and the partial pressure of the oxygen to $P_{O2} = 3x10^{-3}\ mBar$. The rate of depositing the layers was $V_d \approx 20\ nm/min$. In the case of these layers, the substrates were not being heated in the course of their deposition. Besides that, the layers were also deposited in analogical conditions from the ceramic ZnO target on the heated substrates up to a temperature of $T = 500\ ^oC$. The ZnO layers were then annealed using the RTA technique. The ZnO layers of planar waveguides were deposited on BK7 glass or quartz substrates made in the form of thin plates.

4. Results of experimental investigations

Investigations of the optical properties of the waveguide structures were started by determining the refractive index for ZnO layers (Fig. 2). At the wavelength $\lambda = 632.8\ nm$ the refractive index was $n = 1.9785$. Spectral investigations of the refractive index were also carried out in the range from near ultraviolet ($\lambda = 370\ nm$), when $n \approx 2.3$, over the visible range when $n \approx 2.0$, to infrared ($\lambda = 1800\ nm$) when $n \approx 1.9$. The spectral characteristics of the coefficients of optical absorption were also determined. The relation of the refractive index and the absorption coefficient as a function of the waveguides have been presented in Fig. 2. Ellipsometric investigations permitted also to determine the thickness of the ZnO layers, which amounted to $d_{ZnO} = 310\ nm$. (As quoted earlier, in these investigations the spectral ellipsometer SENTECH SE 850 was applied).

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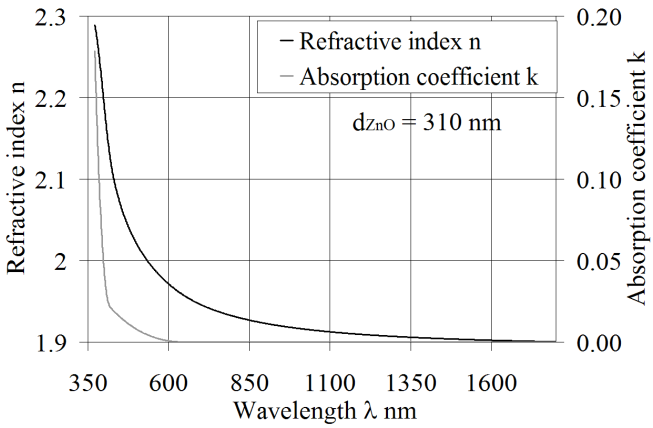

Fig. 2. Refractive index of a ZnO waveguide layer as a function of wavelength.

Next, the spectral transmission of a ZnO layer with a thickness of $d_w = 790\ nm$ on a substrate of BK7 glass plate was investigated. The analysis of the achieved spectral transmission of light across the ZnO layer proved that this material is transparent in the tested range of wavelengths from $\lambda = 400\ nm$ to $\lambda = 1000\ nm$. The diagrams on Fig. 3 present also the characteristics of the spectral transmission across the substrate of BK7 glass. In the characteristics Fresnel reflections are observed.

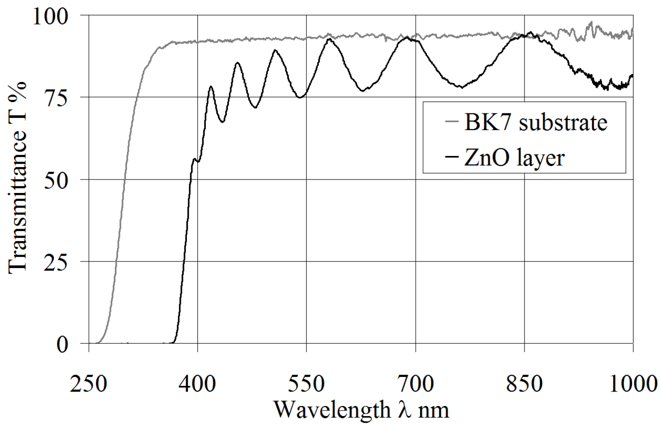

Fig. 3. Spectral transmission characteristics of a ZnO layer deposited on a BK7 substrate.

Investigations concerning the refractive index of ZnO layers permitted to determine the theoretical dependences of the effective refractive index N_{eff} in the function of the thickness of the waveguide ZnO layer (Fig. 4). In our numerical analyses the system Optiwave Mode 2D Solver was used. The numerical analyses concerned two kinds of substrates, viz. BK7 glass and quartz, assuming that: $n_{ZnO} = 1.98$, $n_{quartz} = 1.45$, $n_{BK7} = 1.51$ at the wavelength $\lambda = 677\ nm$.

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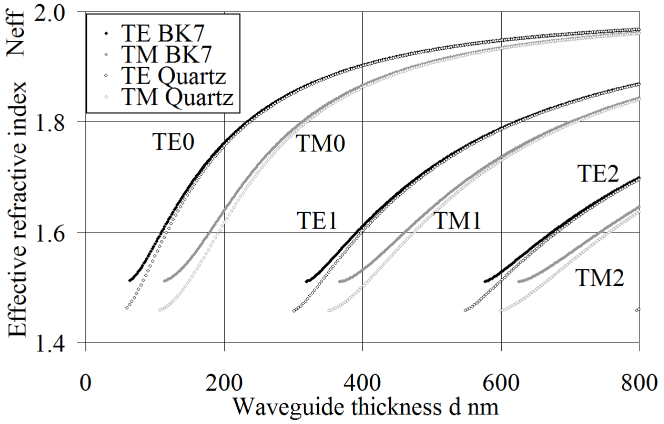
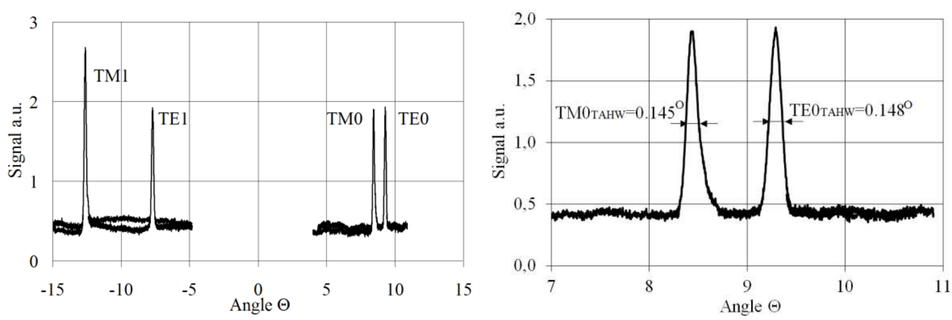


Fig. 4. Theoretical dependence of effective refractive indices as a function of the thickness of the ZnO waveguide layer.

The theoretical analyses permitted to determine the thickness dw of the ZnO layer deposited on the BK7 substrate in order to get for $\lambda = 677\ nm$ a single-mode or multi-mode structures of planar waveguides. Single-mode structures (TE0 and TM0 modes) are obtained when the thicknesses of the layers exceed the value $d_w \approx 100\ nm$ (exceeding the thickness of the cut-off for the modes TE0 and TM0) up to $d_w < 300\ nm$. If the waveguide layer is thicker than $d_w > 300\ nm$, consecutive orders of TE1 and TM1 modes do appear. The values of the effective refractive indices at the given thickness of the ZnO layers depend also on the applied substrate. Waveguide structures deposited on quartz substrates are characterized by somewhat lower values of the effective refractive index than structures deposited on a substrate of BK7 glass with the same thickness of ZnO waveguides. Figs 5a and 6a present the experimental mode spectra of two ZnO waveguide structures with thicknesses of about $d_w \approx 500\ nm$, deposited without previous annealing a), and after annealing by means of the RTA technique b). In the case b) the structure was annealed sequentially, at first for $t = 10\ min.$ in a nitrogen atmosphere at $T = 400\ ^oC$ in order to get rid of stresses and then for $t = 10\ min.$ at $T = 500\ ^oC$ and $T = 600\ ^oC$ in oxygen O_2. The annealing of the layer influences the value of angular half-width of the recorded signals. The performed investigations indicate that in the case of layers obtained without annealing the angular half-width is larger than in the case of layers subjected on annealed substrates. A precise analysis has been presented in Fig. 5b and Fig. 7b and in Table 1.



a) b)

Fig. 5. Mode characteristics of structures with ZnO waveguides without annealing in the function of angle *Θ* of the light incidence: a) all mode characteristics; b) mode characteristic with mark the angular half-width (TAHW) for TE0 and TM0 modes.

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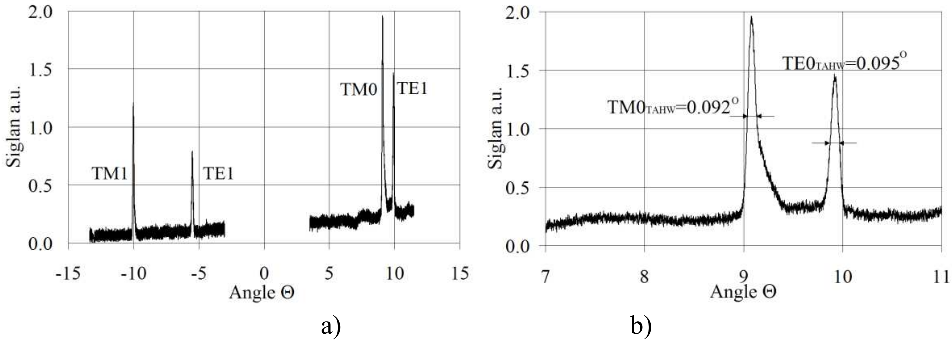

Fig. 6. Mode characteristics of structures with ZnO waveguides after annealing at: $T = 400\ ^oC$ in N_2 and next $T = 500\ ^oC$ in O_2 and at $T = 600\ ^oC$ in O_2 in the function of angle Θ of incidence of light a) all mode characteristic; b) mode characteristics with mark the angular half-width (TAHW) for TE0 and TM0 modes.

Table 1. – Angular half-widths of the propagated modes.

Without annealing of the ZnO waveguide layer: a)		After annealing of the ZnO waveguide layer: b)	
Modes	The angular half-width (TAHW) angles ^{o}	Modes	The angular half-width (TAHW) angles ^{o}
TE0	0.148	TE0	0.095
TE1	0.180	TE1	0.114
TM0	0.145	TM0	0.092
TM1	0.164	TM1	0,090

The mode characteristics of the investigated planar waveguides (Fig. 7) permitted to determine the values of effective refractive indices of the modes.

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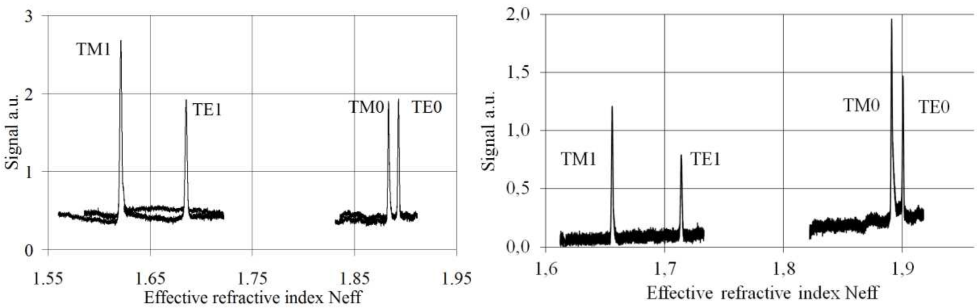
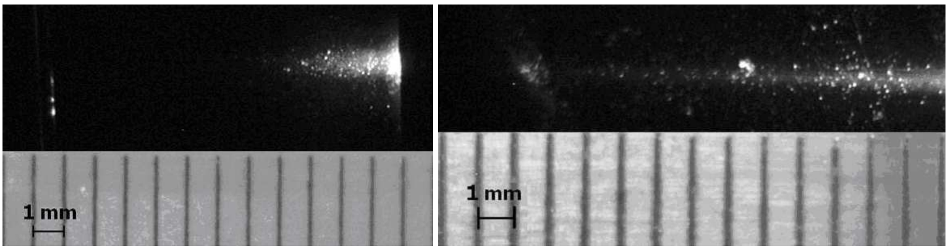


a) b)

Fig. 7. Mode characteristic of the structure with a ZnO waveguide: a) without annealing; b) after annealing at *T = 400 ºC* in N_2 and next at *T = 500 ºC* in O_2 and at *T = 600 ºC* in O_2.



a) b)

Fig. 8. Photos of the light propagation in ZnO waveguide layers: a) without annealing; b) after annealing at *T = 400 ºC* in N_2 and next at *T = 500 ºC* in O_2 and at *T = 600 ºC* in O_2.

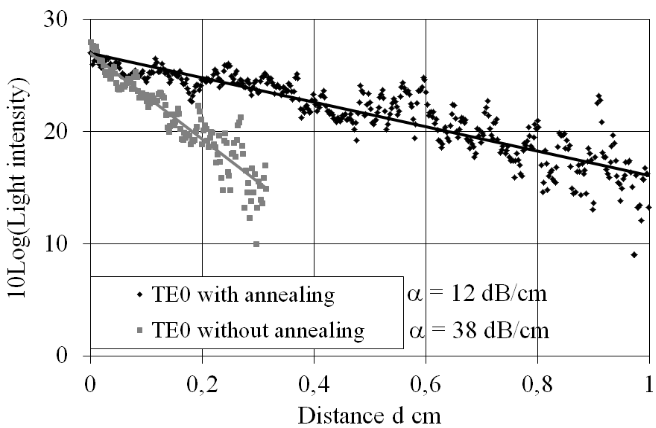


Fig. 9. Propagation of TE0 modes in ZnO waveguide layers without annealing and after annealing at *T = 400 ºC* in N_2 and next at *T = 500 ºC* in O_2 and at *T = 600 ºC* in O_2.

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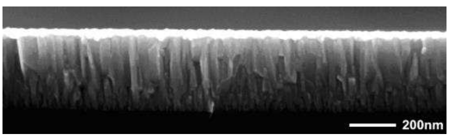
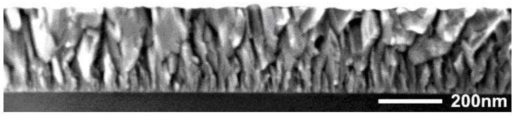
The analysis of the intensity of scattered light as a function of the distance of mode propagation in the waveguide permits to determine the coefficient of attenuation concerning the given mode. Figures 8 and 9. illustrate the images of the trails of light during the propagation in two ZnO waveguides. The annealing of the structures with ZnO waveguide leads to a substantial increase of the path of propagation. In the case of unannealed ZnO waveguides the path of propagation amounts to about $l \approx 3$ *mm*, and for ZnO waveguides annealed at $T = 500\,^oC$ and $T = 600\,^oC$ (in O_2 atmosphere) it increases to $l \approx 10$ *mm*. Fig. 9 presents the dependence of the intensity of light as a function of the distance of propagation concerning the polarization TE0 of the modes. Thus, the attenuation coefficient α, decreases in the case of layers annealed at $T = 500\,^oC$ and $T = 600\,^oC$ to only $\alpha = 12$ *dB/cm* (value determined by means of the method presented in Fig. 1b.). This annealing changes the considerable attenuation ($\alpha = 12$ *dB/cm*) in comparison with the value $\alpha = 38$ *dB/cm* observed in the case of waveguide layers which had not been subjected to thermal treatment.

The observed improvement of the waveguide properties may be explained by analyzing the fractures of ZnO layers, making use of the scanning electron microscope SEM with a field emission, type NEON 40 (produced by Carl Zeiss Ltd.). Fig. 10. Shows SEM images of fractures of the investigated structures with ZnO layers. As is to be seen, the internal structure of the layer changes due to annealing. Layers which have not been subjected to thermal treatment display distinctly an oriented fine-grained polycrystalline structure (Fig. 10a at the boundary: the waveguide layer and the substrate, ZnO grains of a smaller size are to be observed, which indicates the existence of a nucleation layer. The nucleation layer decreases the contrast of the refractive indices between the substrate and the waveguide ZnO layer, which – among others – resulted in a larger angular width of the mode peaks in the unannealed waveguides (Fig. 10a). Annealing leads evidently to a growth of the grains, depending on the temperature applied in this process (Fig. 10b). Annealing restricts the layers adjacent to the substrate (increasing the contrast refractive indices between the substrate and the waveguide layer). Annealing of the structure diminishes locally the mechanical stresses in the waveguide layer. Larger grains restrict the Rayleigh scattering of the propagating modes. These investigations doubtlessly prove the favorable influence of annealing on the optical properties of the waveguides.



a)



b)

Fig. 10. SEM images of the fracture in the ZnO layer: a) without annealing; b) after annealing at $T = 500\,^oC$ and next at $T = 600\,^oC$ in O_2 atmosphere.

Moreover, the technology of planar waveguides was modified by the deposition of the ZnO layer (from the ZnO target) on a heated quartz substrate at a temperature of $T_s = 500\,^oC$. After their deposition the ZnO layers were annealed for $t = 10$ minutes at $T = 400\,^oC$ in O_2 atmosphere and for $t = 20$ *minutes* at $T = 500\,^oC$ in N_2 atmosphere. The mode characteristics are to be seen in Fig. 11.

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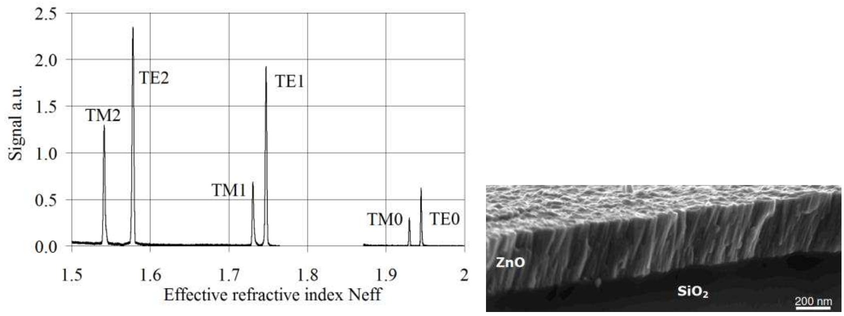
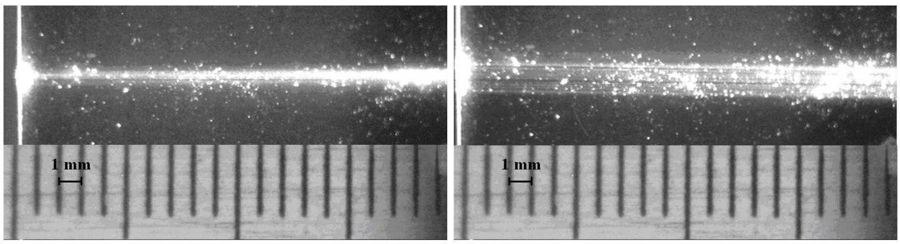


a) b)

Fig. 11. Mode characteristic of ZnO structures: a); SEM images of the fracture in the ZnO layer: b).



a) b)

Fig. 12. Propagation of light with a wavelength of $\lambda = 677\ nm$ in the ZnO waveguide: a) mode TE0; b) mode TM0, after annealing the structure at $T = 600\ ^oC$ in O_2.

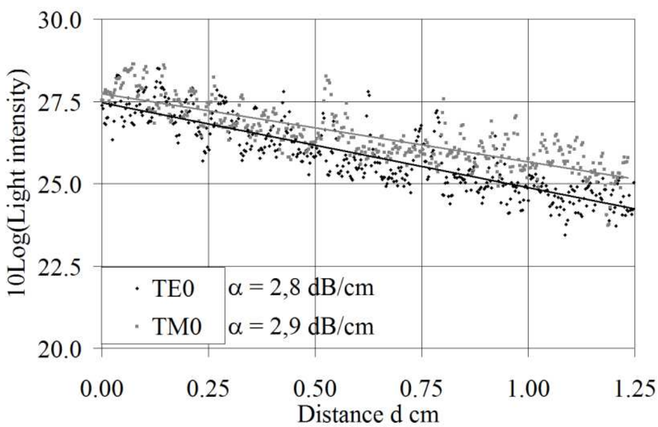


Fig. 13. Distribution of the intensity of light in the function of the distance of propagation.

In such a structure the propagation of light extends to a distance of more than $l = 20\ mm$, both in the case of the TE0 and TM0 modes. The image of the propagation trail of light concerning the modes TE0 and TM0 is to be seen in Fig. 13. Our analysis of the obtained images permitted to determine the attenuation coefficient in both these modes on the level $\alpha \approx 2.8\ dB/cm$ and $\alpha \approx 2.9\ dB/cm$, respectively. The attenuation coefficients concerning modes of higher orders are somewhat larger, amounting to $\alpha \approx 4.8\ dB/cm$ for the mode TE1 and

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$\alpha \approx 6.1\ dB/cm$ for the mode TM1. Waveguide layers with attenuation of a few dB/cm may be used in planar optical systems and in integrated optics [1,15,18].

5. Conclusions

The performed investigations concerning the optical transmission properties proved that ZnO is a transparent material within the range from near ultraviolet ($\lambda_t > 370\ nm$) to infrared. ZnO is also characterized by a high value of the refractive index in the range: $n = 2.3$ in near ultraviolet, $n = 2.0$ in the visible spectrum of light and $n = 1.9$ in the range near infrared. This material displays a relatively high optical dispersion.

Investigations of planar waveguide structures on the basis of ZnO have shown to what extent the process of deposition and thermal treatment affects their optical properties. In the case of ZnO structures not subjected to annealing the values of attenuation amount to $\alpha \approx 30\ dB/cm$. A considerable decrease of the attenuation coefficients was observed in the case of layers annealed after their deposition on the substrate, viz. $\alpha \approx 12\ dB/cm$. (The annealing of the ZnO waveguide layer affects also the angular half-width of the waveguide modes.) The smallest attenuation coefficient of waveguides characterizes ZnO layers deposited on a heated substrate, applying a ceramic ZnO target, and next annealed applying the RTA technique: at $T = 400\ ^oC$ for $t = 10\ min.$ in O_2 atmosphere and at $T = 500\ ^oC$ for $t = 20\ min.$ in N_2 atmosphere and again at $T = 500\ ^oC$ for $t = 20\ min.$ in O_2 atmosphere. In the case of such planar waveguide structures the values of the attenuation coefficient dropped to $\alpha < 3\ dB/cm$. Optical waveguides with such attenuation levels can already be practically used in systems of integrated optics.

The authors of this paper assess that in the case of proposed ZnO waveguides technology the attenuation of optical modes ($\alpha < 3\ dB/cm$) is the lowest in optical waveguides based on ZnO semiconductor layers, concerning the visible range, ever quoted in the specialist world literature.

Zinc oxide is a material with interesting optical properties. The authors of this paper have a well-founded hope that waveguide layers based on ZnO with a relatively low attenuation of the optical modes will find applications in integrated optic systems [24–25] and in optical gas sensors [26].

Acknowledgements

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