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OBTAINING TECHNOLOGIES AND OPTICAL AND PHOTOELECTRIC PROPERTIES IN THE ZnO - $Mg_xZn_{1-x}O$ SYSTEM FOR OPTOELECTRONIC APPLICATIONS

134.01 - Physics and materials technology

Abstract of the PhD thesis

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SUMMARY

CONCEPTUAL GUIDELINES OF RESEARCH

Actuality and importance of the topic

Wide bandgap oxides, especially those in nanostructured form, have a wide range of applications, such as gas sensors, fuel cells, advanced ceramics, chemical sensors, biosensors, batteries, solar cells, pyroelectrics, super-capacitors, catalysts and anti-corrosion coatings [1]. Wide bandgap oxides have been increasingly used in the last 5 years in obtaining oxide film based transistors for flat panel displays, due to their high mobility and transparency [2]. A key strategy in these developments was based on the use of heterojunction [3]. ZnO is an important semiconductor and piezoelectric material among oxide compounds, which has a high potential for many applications such as transparent conductive films, field emission devices, varistors, piezoelectric transducers, resonators and sensors [4]. The last decade has seen an explosion in the study of ZnO nanostructures for gas sensors and sensitized solar cells [5-7]. There is also a significant interest in ZnO-based UV radiation sensors [8,9], for the development of which there is a stringent need in obtaining solid solutions for the control the material's band gap and spectral sensitivity range [10]. The $Mg_xZn_{1-x}O$ alloy system covers a wide spectral range of ultraviolet (UV) radiation between the direct band gap of 3.36 eV for ZnO to 7.8 eV for MgO at room temperature [10-12].

The issue of developing optoelectronic devices for the ultraviolet (UV) range of the optical spectrum is of great current interest. The most common optoelectronic devices are radiation detectors and emitters, such as light emitting diodes (LEDs) and lasers. Currently, such devices for the UV spectral region are fabricated on the basis of diamond [13] or gallium nitride (GaN)-based materials [14,15].

However, diamond is a too expensive material, while for GaN-based devices there is a lack of technologies for producing bulk GaN wafers to be used as a basis for the fabrication of devices. Typically, GaN layers are grown on substrates from other materials, such as sapphire, silicon carbide etc., leading to stresses and defects in the GaN layers. A viable alternative for such devices is zinc oxide (ZnO), since it does not contain expensive metals such as Ga. Apart from that, large single-crystal substrates of sufficient purity for the development of ZnO-based optoelectronic devices are obtained by the hydrothermal method and they are delivered commercially. Photodetectors based on ternary oxides cover a large part of the visible, UV-A, UV-B, and UV-C spectral ranges, they having high and well-established parameters, which are of major importance in optical radiation detection or dosimetry, including antibacterial treatment.

Description of the situation in the research area and identification of problems

It was found that the characteristics of ZnO films can be radically changed by doping with different elements of groups II-III-VIII of the periodic table, such as Mg, Al, Ga, In, Ni, Pd. In order to change the band gap of the material and, respectively, to shift the spectral range of sensitivity (in the case of radiation detectors) towards shorter wavelengths from 365 nm to less than 200 nm, doping with Mg would be efficient. This element is the most widely used one to modify the properties of films and produce solid MgZnO solutions. The ionic radii of magnesium (0.57 Å) and zinc (0.60 Å) are quite close to each other, so that Zn^{2+} ions are easily substituted by Mg^{2+} ones. Therefore, ternary oxides can provide possibilities for modelling optical, luminescent and photoelectric properties over a fairly wide range by extending the energy bandgaps of the films, through the composition tuning in the system (the *x* parameter value). However, the crystal structure of zinc oxide is hexagonal wurtzite-type with tetrahedral coordination, whereas magnesium oxide has a cubic (rocksalt-type) structure with octahedral coordination. This difference of crystal structures limits possibilities of obtaining a single-phase material. A single-phase material was reliably obtained only for the value of $x = 0 - 0.35$ and 0.65 - 1, while a mixed phase material was formed with intermediate compositions [11,16]. Widening of the range of solid solutions compositions with singe phase structure is still an actual issue.

MOCVD, ALD, PLD, and MBE technological methods have been basically user for obtaining single phase MgZnO solutions. However, in spite of some advantages, these technologies are expensive and they have a series of other disadvantages, such as a high temperature of deposition, a need of high vacuum, toxic precursors etc. Spin coating and aerosol deposition method have the advantages of ensuring the control of solid solutions stoichiometry, possibility of doping, and easy preparation of homogeneous films with excellent photoelectrical and optical properties.

Injection photodiodes have been previously demonstrated on the basis of n-CdS/p-CdTe heterostructures, multilayer n+-CdS–n-CdS_xTe_{1-x}–p-$Zn_xCd_{1-x}Te$–Mo structures, and other heterostructures with photosensibility in the spectral range of 500–800 nm wavelengths. At the same time, possibilities of developing injection photodiodes based on MgZnO solid solutions remained practically unexplored.

The aim of the research

The aim of this work was to elaborate and develop technologies for obtaining wide bandgap oxides by cost-effective methods, including ZnO, $Mg_xZn_{1-x}O$, and $(Ga_xIn_{1-x})_2O_3$ oxide films with controlled composition and morphologically, and to explore their optical, luminescent and photoelectric properties for applications in optoelectronic devices such as UV radiation photodetectors.

The main objectives of the work

- Development of technologies for obtaining ZnO films, oxide $(Ga_xIn_{1-x})_2O_3$ compounds and $Mg_xZn_{1-x}O$ solid solutions with controlled composition and morphology by means of aerosol deposition and spin coating method.
- Study of the composition of obtained materials by means of energy dispersive X-ray (EDX) spectroscopy, as well as the identification of the range of compositions of MgZnO solid solutions with hexagonal (wurtzite), mixed and cubic (rock salt) structure, by means of X-ray diffraction (XRD) and Raman spectroscopy methods.
- Study of the optical properties of materials as a function of composition and crystal structure by using absorption and luminescence spectroscopy.
- Determination of the dependence of the morphology and roughness of the obtained films on the technological conditions of production for each deposition method, using scanning electron microscopy (SEM) and atomic force microscopy (AFM).
- Study of the electrical and photoelectrical characteristics of the produced structures as a function of composition, crystal structure and film morphology for further device development.
- Elaboration of different structures designs on Si substrates, such as metal-semiconductor-metal (MSM) structures and heterostructures for UV optical radiation detection based on ternary MgZnO oxides or $(Ga_xIn_{1-x})_2O_3$ oxide compounds.
- Study of optical radiation photodetector parameters (responsivity and detectivity) by electrical (current-voltage) and photoelectrical characterization, depending on photodetector design and component films properties.
- Carrying out a comparative study of wide bandgap oxide materials such as $(Ga_xIn_{1-x})_2O_3$ through determining the dependence of the morphology and crystal structure of the obtained films on the technological conditions, by means of scanning electron microscopy (SEM) and X-ray diffraction (XRD), as well as performing a study of the dependence of the optical absorption edge on the composition of films and of photoelectrical properties of structures based on them.

Scientific research methodology

In order to achieve the aim and objectives of the research, the literature was studied and the following technological methods of deposition and film characterization were used:

- For obtaining the oxide films of ZnO, $Mg_xZn_{1-x}O$, and $(Ga_xIn_{1-x})_2O_3$, spin coating and aerosol deposition methods were used.

- The developed films were characterized using the following techniques: the morphology of films was investigated by scanning electron microscope (SEM); their quantitative chemical composition was determined by energy dispersive X-ray (EDX) detector; the topographic and roughness analysis of the films was investigated by atomic force microscope (AFM); structural properties as well as crystallographic phase content in the oxide films were studied by X-ray diffraction (XRD); determination of vibrational modes was performed using Raman scattering spectroscopy; radiative electronic transitions were investigated by photoluminescence spectroscopy.
- The optical properties of the oxide films were measured at room temperature (300 K) by means of absorption spectroscopy using a Jasco V-670 spectrometer.
- MSM photodiodes based on oxide films have been developed using photolithography.
- The design of different photodetector devices obtained on Si substrates was carried out by developing the following heterostructures: n-$Mg_xZn_{1-x}O$/p-Si with different Mg concentrations and multilayer devices such as Ag/n-$Zn_{0.60}M_{0.40}O$/n-$Zn_{0.90}Mg_{0.10}O$/p-Si/Al and Ag/n-$Zn_{0.65}Mg_{0.35}O$/n-$Zn_{0.85}Mg_{0.15}O$/p-Si/Al.
- Photosensitivity parameters and charge transfer mechanisms in these structures were investigated by measuring current-voltage characteristics in the dark and under illumination with radiation of different wavelengths.

The scientific novelty and originality of the research

- The influence of technological parameters for obtaining ZnO, $Mg_xZn_{1-x}O$ and $(Ga_xIn_{1-x})_2O_3$ oxide films by cost-effective methods on their morphology, chemical composition, crystal structure and vibrational properties was established.
- *For the first time,* a selective ultraviolet (UV) photoreceptor has been developed by depositing from chemical solutions (spin coating deposition or aerosol deposition) a $Mg_xZn_{1-x}O$ absorption film with the value of x = 0 - 0.8 on a Si substrate, and a transparent $Mg_xZn_{1-x}O$ film with a value of x being deposited on top of the absorption film, providing a bandgap at least 0.1 eV higher than that of the absorption film (*Patent No. 4618).*
- Photodetectors have been developed that operate as injection photodiodes at forward bias, their responsivity (R) being about 460 $mA \cdot W^{-1}$, while classical diodes operate at reverse bias.

The solved scientific problem is related to the development and optimization of technologies for obtaining wide bandgap oxides with controlled composition and morphology by means of cost-effective methods, and the study of their optical, luminescent and photoelectrical properties for applications in electronic devices such as ultraviolet radiation photodetectors.

The theoretical significance and the applied value of the work

- The dependence of the bandgap and crystal structure of the ternary compounds on the Mg concentration was established from the study of optical and structural properties of materials with different compositions.
- The slow photoconductivity relaxation component of ternary oxide MgZnO as a function of film deposition method was determined.
- The compositional range of MgZnO solid solutions with hexagonal (wurtzite), with cubic (rock salt), and with mixed phase structures was identified by applying Raman spectroscopy and X-ray diffraction (XRD).
- The applied value of the given work consists in the development of selective photodetectors based on oxide films for UV radiation detection. Multilayer devices based on $Zn_{0.85}Mg_{0.15}O$/n-$Zn_{0.65}Mg_{0.35}O$ oxide films with a responsivity of 460 $mA \cdot W^{-1}$ have been demonstrated, while single layer $Mg_xZn_{1-x}O$ structures showed a much lower responsivity of only 3.0 $mA \cdot W^{-1}$. The given structures have been shown to work as injection photodiodes at forward bias, while classical diodes work at reverse bias.

Main scientific results submitted for defense

1. The optimal technological conditions for obtaining $Mg_xZn_{1-x}O$ films by aerosol deposition are as follows: deposition temperature of about 500 °C, chemical solution injection rate of 1 ml/min, molar mass of each precursor of 0.35 M and the use of a carrier gas flow (O_2 + Ar), while for obtaining $Mg_xZn_{1-x}O$ films by spin coating method the optimal technological conditions are as follows: deposition in cycles of 20 seconds each at a rotational speed of 2000 rpm, followed by heat treatment at 500 °C for one hour in an combined (O_2 + Ar) atmosphere.
2. The crystallite sizes in $Mg_xZn_{1-x}O$ films prepared by spin coating method decrease from 140 nm to 30 nm with increasing Mg content, while in $Mg_xZn_{1-x}O$ films obtained by aerosol deposition the morphology changes from 100 nm crystallites to 250 nm conglomerate crystallites. The morphology of the films deteriorates with increasing *x-value* above 0.8 for both technological methods.
3. The wurtzite structure predominates in films of the ternary $Mg_xZn_{1-x}O$ compound obtained by aerosol deposition and by spin coating method up to 60 % Mg content, while films with higher Mg content contain inclusions with cubic phase of MgO (rock-salt). The incorporation of Mg into

the wurtzite structure is demonstrated by shifting the XRD (222) reflex to larger angles 2Θ and the Raman $E_2^{(low)}$ mode to higher wavenumbers.

4. The introduction of Mg up to a 60 % content is efficient for tuning the bandgap of $Mg_xZn_{1-x}O$ solid solutions and for shifting the spectral range of sensitivity from 3.3 eV to 5.1 eV, while the spectral range of sensitivity can be changed from 3.6 eV for In_2O_3 to 4.9 eV for Ga_2O_3 by introducing Ga into the composite $(Ga_xIn_{1-x})_2O_3$ films.
5. The component of slow photoconductivity relaxation in the ternary MgZnO compound obtained by the spin coating method is due to local fluctuations in the solid solution composition, which leads to increased response time of photodetectors to UV light irradiation, while samples obtained by aerosol deposition method show a faster photoresponse.
6. Elaboration of heterostructures on p-Si substrates with two layers of $Mg_xZn_{1-x}O$ improves the selectivity and photosensitivity parameters of the devices compared to single-layer devices. These devices operate in the injection photodiode mode at forward bias, demonstrating a responsivity of 460 $mA\cdot W^{-1}$ and a detectivity of $1x10^{10}$ cm $Hz^{1/2}W^{-1}$.

Approval of the scientific results

The main results of the research performed within the thesis have been presented at the following national and international conferences and exhibitions:

- International Conference ,,**Technical and Scientific** Conference of Students, Master and PhD Students", Technical University of Moldova, Chisinau, (2019, 2021, 2022);
- International Conference "**Advanced Topics in Optoelectronics, Microelectronics and Nanotechnologies ATOM N-2022**", X and XI editions, Constanta, Romania (2020 and 2022)**;**
- International Conference "**Nanotechnologies and Biomedical Engineering**", Technical University of Moldova, 4th and 5th editions, Chisinau, Republic of Moldova, (2019 and 2021);
- **Applied Nanotechnology and Nanoscience** International Conference **- ANNIC 2021**, Paris, France, March 24-26, Online, (2021);
- International Conference ,,**The 12th International Conference on Intrinsic Josephson effect and horizons of superconducting spintronic", (SPINTECH-NANO-2021)**", 22-25 September, Chisinau, Republic of Moldova, (2021);
- Bristol Center for Functional Nanomaterials **Annual Conference**, September 17 - 18, Bristol, UK, (2020);

- International Conference "**Contemporary Trends in the Development of Science: Visions of Young Researchers**", editions VII, VIII and IX, USDC, Chisinau, (2018, 2019, 2020);
- International Conference ,,**Telecommunications, Electronics and Informatics**", 6th edition, Technical University of Moldova, 24-27 May, Chisinau, Moldova, (2018);
- International Conference ,,**Materials Science and Condensed Matter Physics**", 9th edition, 25-28 September, Chisinau, Moldova, (2018);
- International Conference ,,**Microelectronics and Computer Science**", 9th edition, Technical University of Moldova, Chisinau, October 19 - 21, (2017);
- National Conference ,,**The future belongs to us**", VI edition, State University of Moldova, Chisinau, Moldova, 6-7 October, (2016);
- National Conference ,,**Scientific Annals of USM. Natural and Exact Sciences. Economic Sciences**", State University of Moldova, Chisinau, Moldova, September 26, (2016).

Publications on the thesis topic

The main results of the research related to the thesis topic have been published in **18** scientific papers, including **3** articles in journals in the Web of Science and SCOPUS databases with impact factor, **2** articles in national journals, **4** articles in the proceedings of scientific events included in the Web of Science and SCOPUS databases, **9** publications at national and international conferences, **4** single-authored articles, the list of which is presented at the end of this Abstract.

The volume and structure of the thesis

The thesis consists of an introduction, five chapters, general conclusions and bibliographical references consisting of **159** titles, and is set out on **123** pages of basic text, containing **70** figures and **21** tables.

Keywords: nanotechnologies, UV radiation photodetectors, optoelectronics, band gap engineering, MgZnO, thin films, spin coating deposition, aerosol deposition method.

THESIS CONTENT

In the **introduction,** the actuality and the importance of the research topics justified, the aim and objectives of the thesis, the methodology of the scientific research, the scientific novelty of the obtained results, the scientific problem solved, the theoretical and applied significance of the work, the main scientific results submitted for defense, the approval of the scientific results, as well as the volume and structure of the work are presented.

Chapter 1 includes a review of studies previously performed and reported in the scientific literature on fabrication methods, crystal structure, vibrational, optical, photoelectrical properties and current-voltage (I-U) characteristics of ternary MgZnO oxide. The description of oxide films such as $(Ga_xIn_{1-x})_2O_3$ is also given. The literature data on optical radiation photodetectors based on $Mg_xZn_{1-x}O$, $Al_xGa_{1-x}N$ and $(Ga_xIn_{1-x})_2O_3$ films are also reviewed.

Chapter 2 presents the technological methods for the preparation of oxide films and the experimental technique. The methods of aerosol deposition and spin coating are described, including their main components, the technological procedures applied for obtaining the films, and how each chemical solution was obtained. The methods of characterization and measurements of the obtained samples are also described and discussed, such as scanning electron microscopy, identification of the elemental composition of materials by means of energy dispersive X-ray spectroscopy, atomic force microscopy, study of crystal structure by X-ray diffraction, Raman scattering spectroscopy and measurement of photoluminescence spectra. The procedures for measuring the optical, photoelectrical properties and current-voltage (I-U) characteristics of the obtained films are also presented in this chapter, as well as the description of the elaboration of structures for photodetectors based on oxide compounds.

In **Chapter 3** the oxide films of MgZnO obtained by the spin coating method are characterized. Figure 1 illustrates the morphology evolution of ternary $Mg_xZn_{1-x}O$ oxides as a function of the Mg content of films (the *x* value) obtained by the spin coating method and subjected to thermal treatment at 500 °C for 1 h in a combined atmosphere of $(O_2 + Ar)$. It can be seen that the hexagonally shaped crystallites decrease in size from 140 nm ($x = 0.0$) to 30 nm ($x = 0.6$), and at a concentration of 0.8 the crystallites become shapeless and conglomerated with each other with sizes of about 20-150 nm. The roughness of the films was deduced from the AFM profiles, and the RMS and RSkew values were found to be of 12 nm, and 0.272, respectively. The crystallite height was 63 nm at the scan size of (5x5 μm^2) for the $Zn_{0.8}Mg_{0.2}O$ films obtained by the spin coating method. At the same time, the EDX analysis results showed that the compositions of the $Mg_xZn_{1-x}O$ films are stoichiometric within the instrument accuracy of ± 5 %.

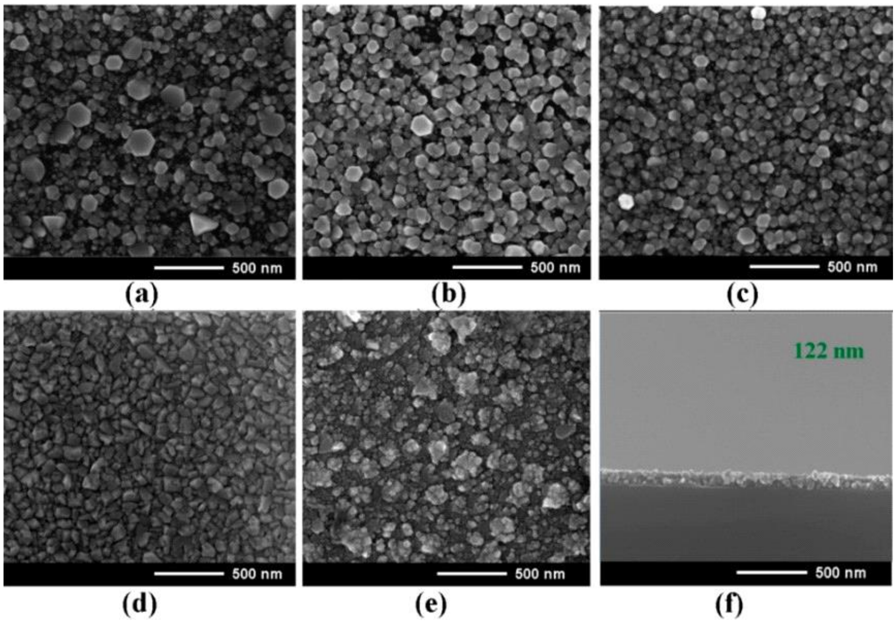

Fig. 1. SEM images of $Mg_xZn_{1-x}O$ films deposited on Si substrate by the spin coating method and thermal treated at 500 °C for 1 h in a combined atmosphere of (O_2 +Ar). The *x-values* of the films are as follows: 0 (a); 0.2 (b); 0.4 (c); 0.6 (d); 0.8 (e). The SEM image in cross-section of a film with *x* = 0.4 is shown in (f)

The XRD picture of the $Mg_xZn_{1-x}O$ films ($0 < x < 0.8$) obtained by the spin coating method is shown in Figure 2 a. The XRD investigations demonstrate that the wurtzite phase is present in the films up to the *x* value of 0.8. At the same time, reflections (002) and (202) of the cubic (rock salt) phase appear at *x* values greater than 0.4. The position of the (002) reflection approaches the position of a band, which represents an overlap of the (101) reflection from the hexagonal (wurtzite) phase and the (111) reflection from the cubic (rock salt) phase. The XRD data are also confirmed by Raman scattering spectroscopy results (Figure 2 b), which similarly demonstrate an efficient incorporation of Mg atoms into the wurtzite lattice of $Mg_xZn_{1-x}O$ films by shifting the $E_2^{(low)}$ mode towards higher wavenumbers. The peaks at about 100 cm^{-1} and 438 cm^{-1} are assigned to the ZnO non-polar optical phonon $E_2^{(low)}$ (low-frequency) and $E_2^{(high)}$ (high-frequency) modes, respectively. The band at 583 cm^{-1} comes from a combination of A_1 (LO) and E_1 (LO) modes. In this chapter, the dependence of morphology and crystal structure on the thermal treatment of $Mg_xZn_{1-x}O$ oxide films is also described. It was determined that the critical and most important parameter in obtaining uniform and stoichiometric oxide films is their treatment temperature after the deposition process. Figure 3 shows the morphology of $Zn_{0.5}Mg_{0.5}O$ films with a thickness of about 100 nm subjected to thermal treated in a combined atmosphere of (O_2+Ar) at 500 °C for different process duration ranging from 15 to 60 min. It can be seen that the films are formed of crystallites with a nearly uniform distribution on the film surface, while the average crystallite size increases from about 20 nm to 70 nm with increasing treatment duration.

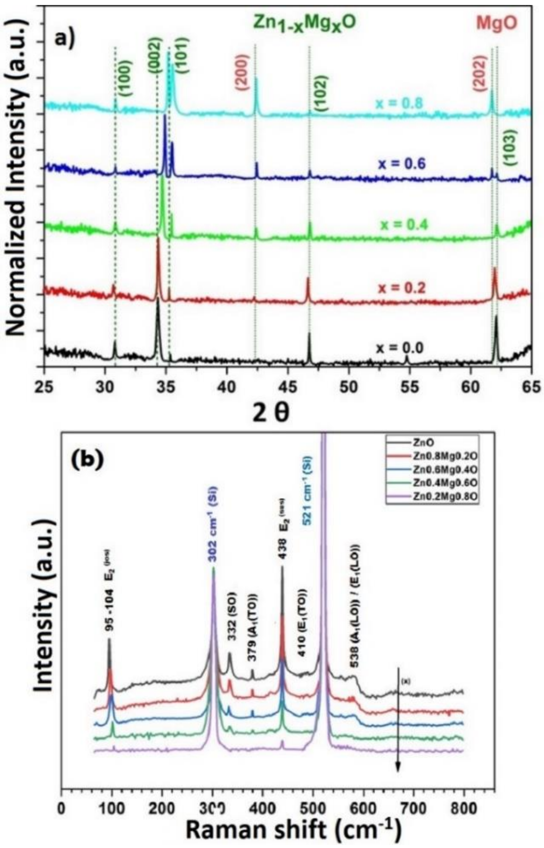

Fig. 2. XRD pattern (a) and Raman spectra (b) of $Mg_xZn_{1-x}O$ films deposited on Si substrate by the spin coating method

It was found that the films quality increased with increasing the thermal treatment temperature up to 500 °C, while the further increase of the annealing temperature resulted in the degradation of the film morphology, their cracking, and deviation of stoichiometry towards the oxygen excess.

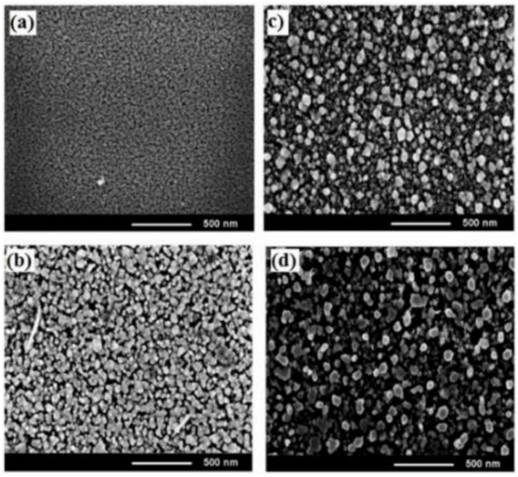

Fig. 3. SEM images (top view) of $Zn_{0.5}Mg_{0.5}O$ films deposited on Si substrate by the spin coating method and subjected to thermal treatment at 500 °C in a combined atmosphere of (O_2 +Ar) for 15 min (a), 30 min (b), 45 min (c), 60 min (d)

The XRD analysis shown in Figure 4 proves that the crystallite size increases and the crystalline quality improves with increasing thermal treatment duration up to 60 min, as indicated by the increase in XRD reflections intensity of the hexagonal (wurtzite) phase indexed according to PDF file no. 01-078-3032. At the same time, the (200) reflex from the cubic (rock salt) phase increases in intensity at thermal treatment duration longer than 30 min.

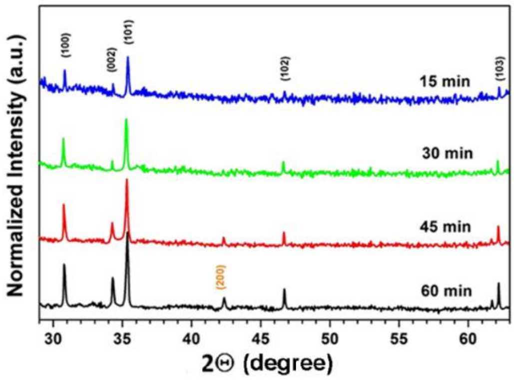

Fig. 4. XRD pattern of $Zn_{0.5}Mg_{0.5}O$ films deposited on Si substrate by spin coating method and subjected to thermal treatment in a combined atmosphere of (O_2 +Ar) at 500 °C for different process duration.

It has also been shown that the evolution of film morphology is also highly dependent on thermal treatment in different atmospheres. Figure 5 shows the morphology of oxide films of $Zn_{0.8}Mg_{0.2}O$/Si obtained by the spin coating method with no heat treatment, with heat treatment at 500 °C in air, or in a combined atmosphere of oxygen and argon ($O_{80\%}$+$Ar_{20\%}$).

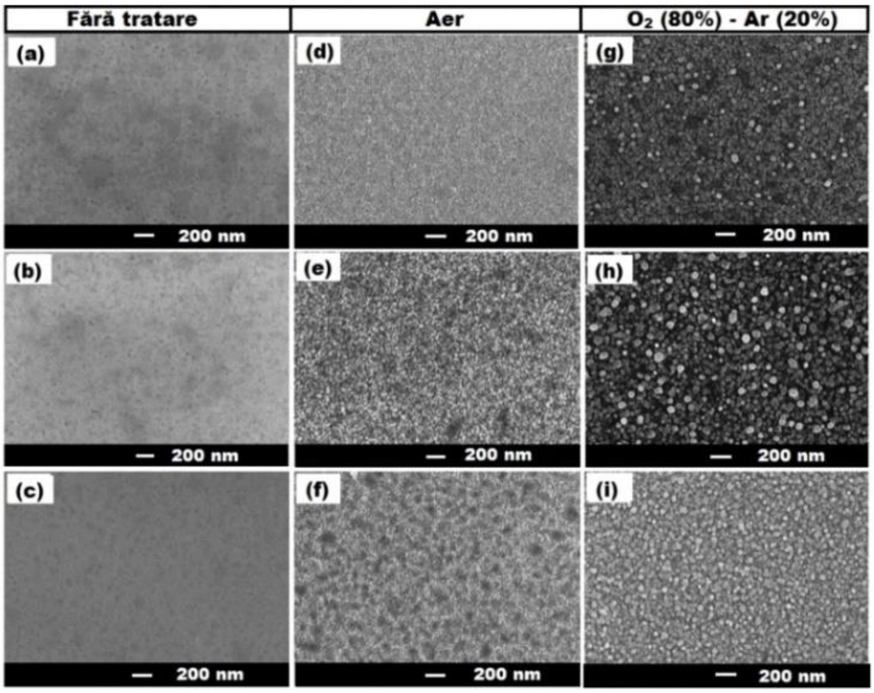

Fig. 5. SEM images (top view) of $Zn_{0.8}Mg_{0.2}O$/Si oxide films obtained by the spin coating method and thermally treated in different atmospheres: I column - samples without heat treatment with 15, 10, 5 layers (a, b, c); II column - samples thermally treated in air with 15, 10, 5 layers (d, e, f); III column - samples thermally treated in atmosphere of (O_2 +Ar) with 15, 10, 5 layers (g, h, i).

It can be seen from SEM images that the films without thermal treatment are amorphous, without any nanocrystallites, regardless of their thickness (Figure 5. a, b, c). Once the thermal treatment is done in air, nanocrystallites are already evident (Figure 5. d, e, f). The most pronounced nanocrystallites were formed in the samples with 10 deposited layers (Figure 5. e, h), reaching sizes of about 10 - 70 nm. XRD analysis revealed a single-phase wurtzite structure of the $Zn_{0.8}Mg_{0.2}O$ film after thermal treatment in air, while the highest crystalline quality and nanocrystallites size is obtained after thermal treatment in combined atmosphere of (O_2 +Ar) (Figure 5. g, h, i), indicated by the higher intensity of XRD reflexes.

Another very important study comes from photoluminescence (PL) spectroscopy investigations. As can be seen from Figure 6, the PL spectra of thermally treated films at 500 °C consist of a broad emission band at both room temperature (300 K) and low temperatures (20 K), which shifts towards higher photon energies with increasing Mg content in the alloy. The higher the value of x, the larger the difference between the bandgap and the PL band maximum. Moreover, the luminescence is excited by photon energy of 3.81 eV, which is much smaller than the bandgap for thin film with x value of 0.40 (4.28 eV).

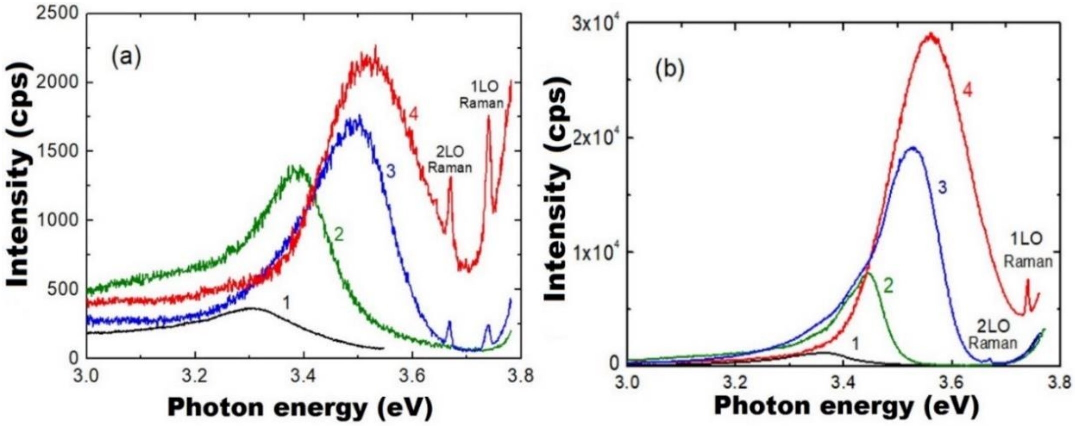

Fig. 6. PL spectra of $Mg_xZn_{1-x}O$ films deposited on Si substrates by the spin coating method with *x* values of 0.00 (1); 0.05 (2); 0.15 (3); and 0.40 (4), heat treated at 500 °C and measured at a) 300 K and b) 20 K.

It was determined from the photoluminescence spectra that the slow component of photoconductivity relaxation in spin coated films is due to local fluctuations in the solid solution composition. Local potential fluctuations induced by composition fluctuations lead to the formation of deep band tails in the bandgap with a width from 70 meV for $x = 0.0$ to 400 meV for $x = 0.4$, which make it possible to excite photoluminescence at sub-band gap photon energies. A model for the band tails distribution is proposed in Figure 7 for MgZnO films. Because the luminescence intensity is higher at lower temperatures and, respectively, the position of the luminescence band maximum can be determined more accurately, this model was developed for 20 K, however, the trends of increasing "band tails" with increasing x are also valid at room temperature (300 K).

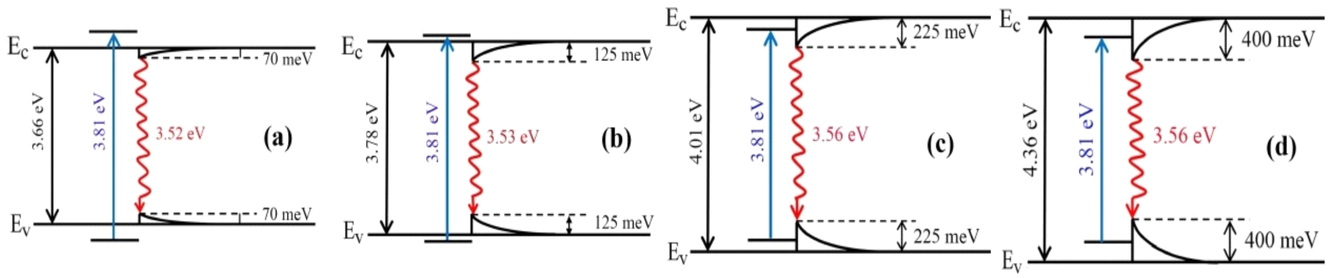

Fig. 7. A model for the band tails distribution at 20 K in $Mg_xZn_{1-x}O$ films with the *x* value composition of a) 0.10; b) 0.15; c) 0.25 and d) 0.40.

In Chapter 4, oxide films of $Mg_xZn_{1-x}O$ and $(Ga_xIn_{1-x})_2O_3$ with thicknesses between 80 - 150 nm and uniform morphology obtained by aerosol deposition at 500 °C on p-Si (100) substrates are characterized. SEM images of $Mg_xZn_{1-x}O$ oxide films with composition range of *x* = 0.00 - 0.80 are shown in Figure 8. Hexagonal structures with sizes around 100 - 250 nm are formed in films with *x* value up to about 0.30, while the shape of the crystallites becomes structureless with further increase of Mg content, but retaining to some extent the crystallite size. This behavior is explained by the increasing concentration of MgO inclusions with cubic structure in the hexagonal $Mg_xZn_{1-x}O$ wurtzite phase. The roughness parameters of the films were determined from AFM image analysis, so that the RMS values were found to be 5.2 nm and RSkew 0.1163. The crystallite height was around 35 nm at a scan size of (5x5 μm^2) for $Zn_{0.8}Mg_{0.2}O$ films obtained by the aerosol deposition method.

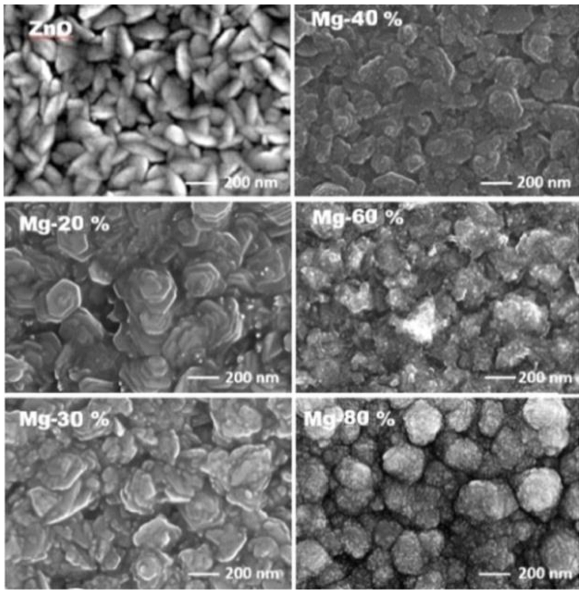

Fig. 8. SEM images (top view) of oxide films of $Mg_xZn_{1-x}O$ with the composition range of *x* = 0.00 - 0.80 deposited on Si substrates by aerosol deposition method.

XRD data and RAMAN analysis for $Mg_xZn_{1-x}O$ films obtained by aerosol deposition show the same tendency as observed for films deposited by the spin coating method (Figure 9 a, b). These investigations demonstrate that the wurtzite structure is present in $Mg_xZn_{1-x}O$ films even at 80 % Mg content with some inclusions from the MgO cubic phase. Mg incorporation into the wurtzite structure is demonstrated by shifting the XRD (222) reflection to larger 2Θ angles and the Raman $E_2^{(low)}$ mode to higher wavenumbers.

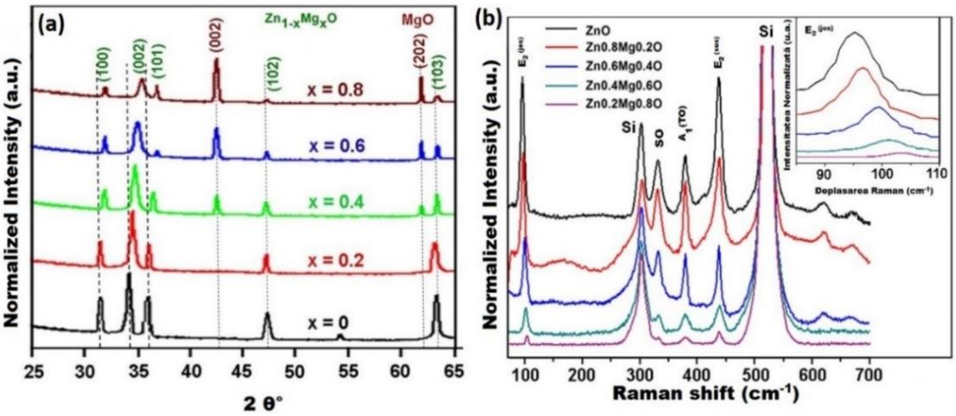

Fig. 9. XRD pattern (a) and RAMAN spectra (b) of $Mg_xZn_{1-x}O$ films deposited on Si substrate by the aerosol deposition method.

Oxide $(Ga_xIn_{1-x})_2O_3$ films were also produced by aerosol deposition, and their morphology is shown in Figure 10. From the SEM images one can see that the films consist of nanocristalites which sizes increase with increasing Ga content from 70 nm to 180 nm, while the thickness of the films is almost constant around 150 - 170 nm. EDX analysis results showed that the chemical composition of the obtained films is nearly stoichiometric within the experimental instrument errors of ±5%.

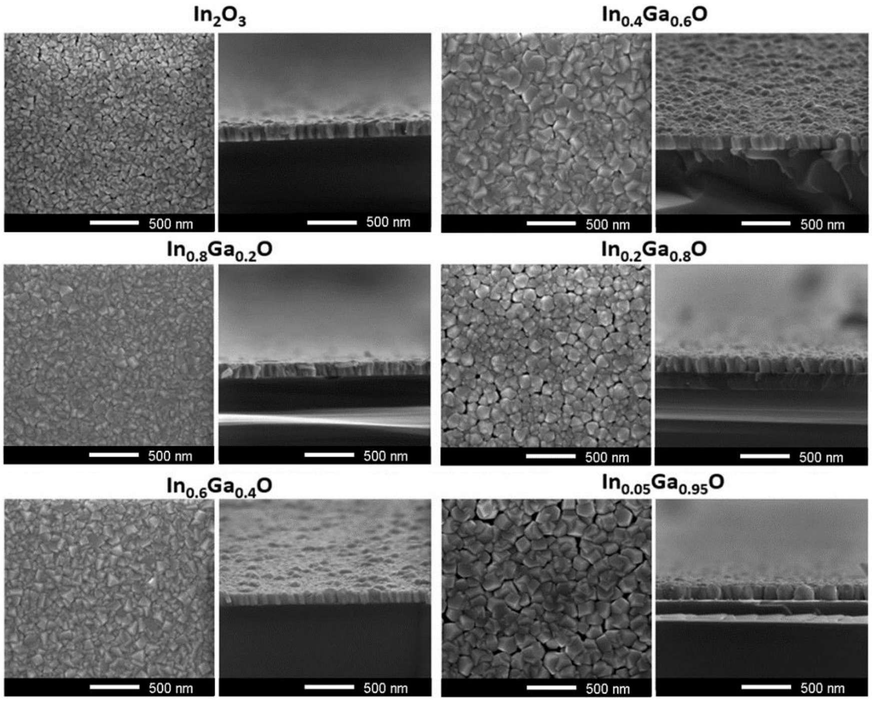

Fig. 10. SEM images (top view and cross section) of oxide films of $(Ga_xIn_{1-x})_2O_3$ grown on Si substrates by aerosol deposition method.

XRD investigations prove the formation of a solid solution of $(Ga_xIn_{1-x})_2O_3$ with a cubic structure, characteristic of In_2O_3 crystals. The efficient incorporation of Ga atoms into this structure is indicated by the shift of the (222) reflections of the cubic phase towards higher values 2Θ with increasing Ga content (Figure 11 a). At values of *x* greater than 0.6 a biphasic composite is formed, consisting of the $(Ga_xIn_{1-x})_2O_3$ solid solution with cubic structure and crystallites of the β-Ga_2O_3 monoclinic phase. The position of the reflex (111) from the β-Ga_2O_3 phase does not change with increasing Ga content in the film. The Raman spectrum of a sample with In_2O_3 composition (Figure 11 b) confirms the volume-centered cubic phase (bcc) with (Ia-3) space group, while the Raman modes of the cubic phase decrease in intensity with increasing the Ga content in films, new peaks appearing in the spectrum at 198 cm^{-1} and 415 cm^{-1} related to the A_g (3) and A_g (6) symmetry modes of the β-Ga_2O_3 phase.

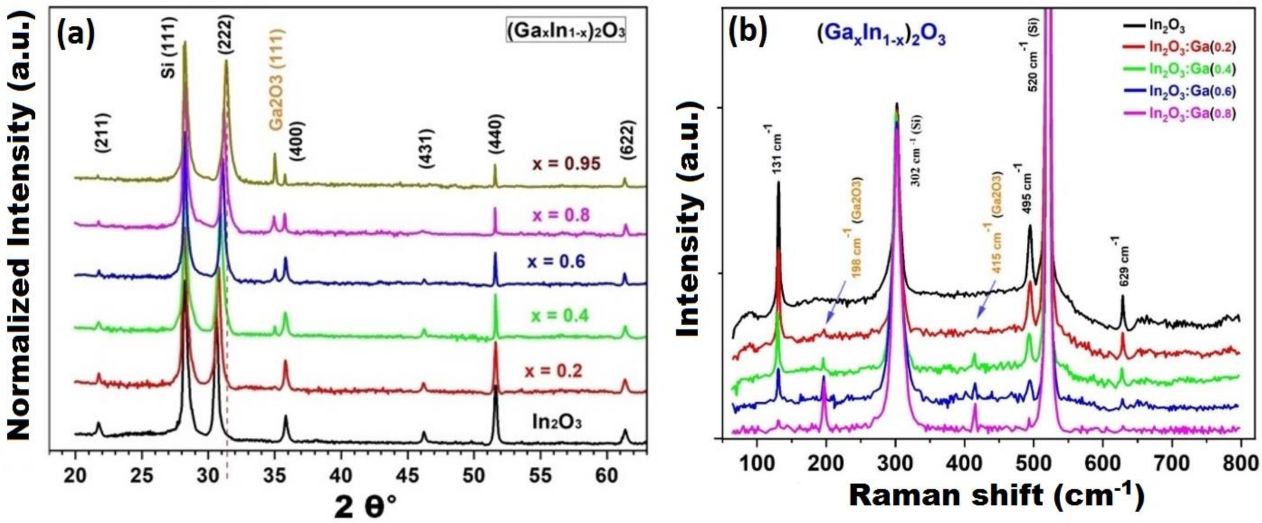

Fig. 11. XRD pattern (a) and RAMAN spectra (b) of $(Ga_xIn_{1-x})_2O_3$ films deposited on Si substrate by the aerosol deposition method.

In Chapter 5, photodetectors devices obtained from oxide films are described and analyzed. The obtained MgZnO oxide films were tested for photodetection applications in metal-semiconductor-metal (*MSM*) design configuration with Pd interdigital metal contacts. The films showed sensitivity to UV light irradiation, where the photosensitivity was much higher for samples obtained by spin coating compared to those obtained by aerosol deposition. The current-voltage (I-U) characteristic curves are symmetrical for both polarization directions, and the characteristic measured in the dark is linear for both growth methods demonstrating the formation of ohmic contacts (Figure 12). The characteristic deviates from linear under UV light irradiation (200-400 nm, 2.2 mW/cm^2), and the sample shows photosensitivity, which increases with increasing polarization. From the current-voltage characteristics it was shown that the devices under illumination work like Schottky diodes, both at reverse and forward bias.

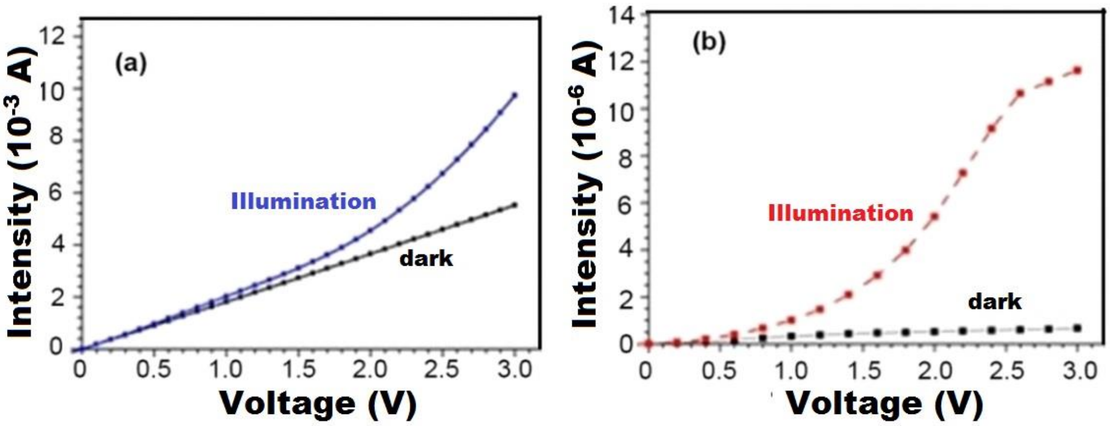

Fig. 12. Current-voltage characteristics in the dark and under UV light irradiation for $Zn_{0.8}Mg_{0.2}O$ films deposited on Si Substrate by aerosol deposition (a) and by spin coating (b).

The investigation of photoconductivity excited under irradiation with different wavelengths for oxide films obtained by both methods demonstrated photosensitivity in a fairly broad spectral range from ultraviolet (UV) to infrared (IR).

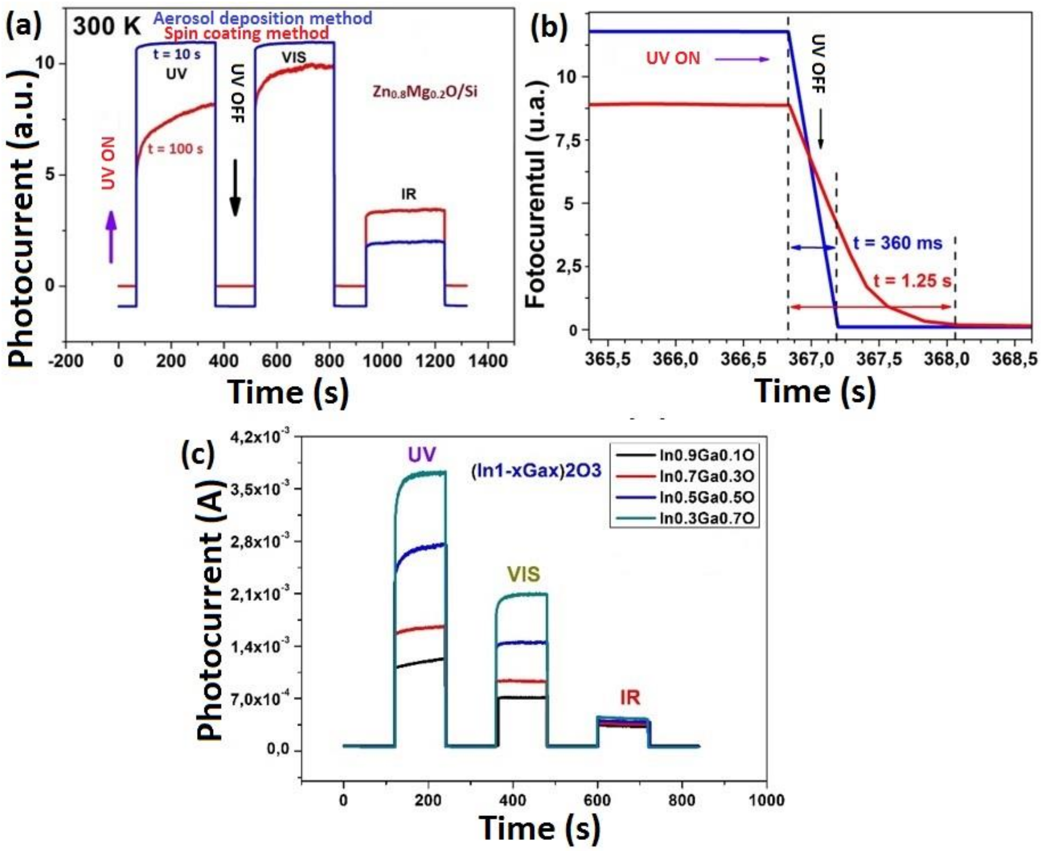

Fig. 13. Photocurrent relaxation measured at 300 K in vacuum under irradiation with different wavelengths for oxide films of $Zn_{0.8}Mg_{0.2}O$ (a, b) and $(Ga_xIn_{1-x})_2O_3$ (c).

Photocurrent relaxation in samples obtained by aerosol deposition and by spin coating method (Figure 13 a, b) demonstrates two components: a fast relaxation component in the range of seconds and a slow relaxation component with a relaxation time of hundreds of seconds. The response time when the optical radiation is switched on is around 250 ms for the aerosol deposition method, and it is around 360 ms when the optical radiation is switched off. For the spin coating method, the response time is around 260 ms when optical radiation is switched on, and it is around 1.25 s when the radiation is switched off, as illustrated in Figure 13 b. A long relaxation component is also observed in the $(Ga_xIn_{1-x})_2O_3$ films, which showed a response time of 370 ms when optical radiation is switched on and off (Figure 13 c).

The performance of UV photodetectors in MSM configuration with interdigital Pd metal contacts on the surface of $Zn_{0.8}Mg_{0.2}O$/p-Si films was also characterized by responsivity (R) and detectivity (D*) as compared to the $(Ga_xIn_{1-x})_2O_3$ oxide films with different Ga content, and their parameters are presented in Table 1.

Table 1. Parameters of photodetectors based on films of $Zn_{0.8}Mg_{0.2}O$ and $(Ga_xIn_{1-x})_2O_3$ for the UV domain.

Photodetector structure	U (V)	(R) $mA·W^{-1}$ (ASP/SC)	(D*) $cm·Hz^{1/2}·W^{-1}$ (ASP/SC)	Photodetector structure	U (V)	(R) $mA·W^{-1}$	(D*) $cm·Hz^{1/2}·W^{-1}$
$Zn_{0.8}Mg_{0.2}O$/Si	1	10 / 21	$6.7×10^8/1.6×10^9$	$In_{0.9}Ga_{0.1}O$/Si	5	18	$1.3 × 10^7$
	2	33 / 60	$1.5×10^9$ / $4×10^9$	$In_{0.7}Ga_{0.3}O$/Si		24	$1.9 × 10^7$
	5	79 / 150	$3.1×10^9$ /$6.3×10^9$	$In_{0.5}Ga_{0.5}O$/Si		42	$3.4 × 10^7$
	10	264 / 380	$9.7×10^9$ / $2×10^{10}$	$In_{0.3}Ga_{0.7}O$/Si		58	$4.8 × 10^7$

Injection photodiodes based on wide bandgap oxide films deposited on Si substrates have also been studied in this chapter. The current-voltage characteristics represented in linear voltage coordinates and logarithmic current coordinates do not fit the classical formula for a p-n junction, but the characteristics fit a straight line in double-logarithmic coordinates, which means that the characteristic corresponds to a power function $I \propto U^n$, according to the Lampert's theory. Figure 14 shows the current-voltage characteristics of films obtained by the spin coating method plotted for direct bias in double-logarithmic scale for *x* values of 0.1, 0.2 and 0.4. The value of *n* for the dark current characteristics decreases from about 4 for $Mg_xZn_{1-x}O$ films with *x* value of 0.1 to about 3 for *x* value of 0.2, and to about 2 for *x* value of 0.4. The ratio of the photocurrent to the dark current at 1 V bias is about 3 for all values of *x*. However, at 0.1 V bias, this ratio decreases from about 100 for *x* value of 0.1, to about 10 for *x* value of 0.2, and to about 3 for *x* value of 0.4.

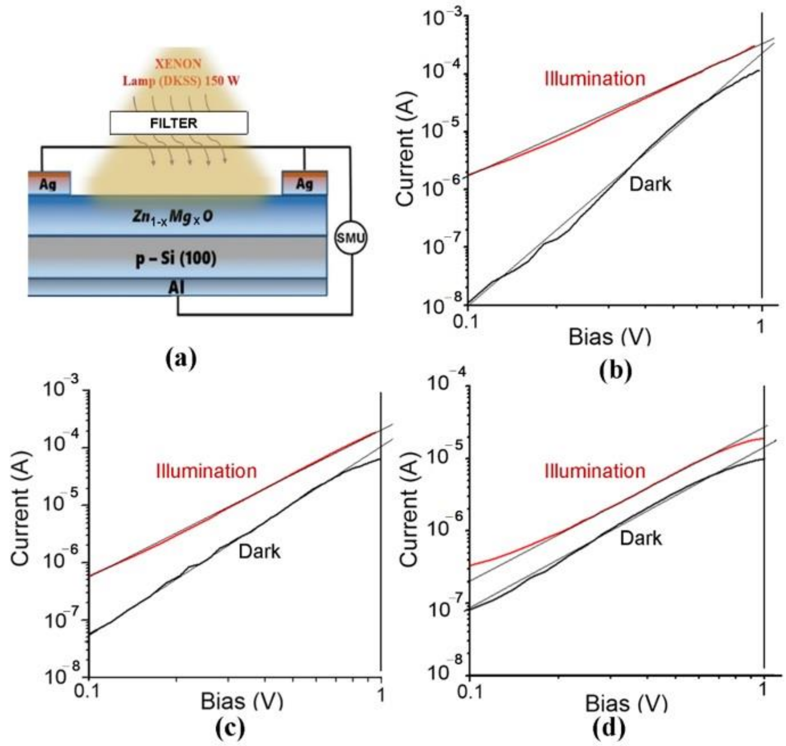

Fig. 14. Design of an n-$Mg_xZn_{1-x}O$/p-Si photodiode (a) and current-voltage characteristics of the devices plotted for double-logarithmic scale direct bias for *x* values of 0.1 (b), 0.2 (c) and 0.4 (d).

At the same time, the responsivity (*R*) is quite low at 0.1 V bias for all values of *x*, it being around (10-30) $\mu A \cdot W^{-1}$. The responsivity is higher at 1 V bias, but decreases from about 3 $mA \cdot W^{-1}$ for *x* value of 0.1, to about 2 $mA \cdot W^{-1}$ for *x* value of 0.2, and to about 0.1 $mA \cdot W^{-1}$ for *x* value of 0.4. The parameters of these devices are generally low, because the ratio of photocurrent to the dark current decreases with increasing bias voltage, and the device operation at voltages higher than 1 V becomes inefficient.

To overcome this disadvantage, a device structure with two layers of $Mg_xZn_{1-x}O$ with different values of *x* has been developed, as shown in Figure 15 a, where the top layer of $Zn_{0.65}Mg_{0.35}O$ with higher band gap plays the role of a window, it is a transparent layer for UV-B radiation, which protects the base (absorption) layer with $Zn_{0.85}Mg_{0.15}O$ composition and is expected to reduce the density of surface states. The charge carriers generated by the incident light are separated by the internal electric field of the p-n junction. The band gap gradient formed in the Ag/n-$Zn_{0.65}Mg_{0.35}O$/n-$Zn_{0.85}Mg_{0.15}O$/p-Si/Al favors the movement of charge carriers due to the internal electric field resulting from the gradient of the ternary oxide composition, and the photodetector sensitivity maximum is determined by the composition of the absorption layer.

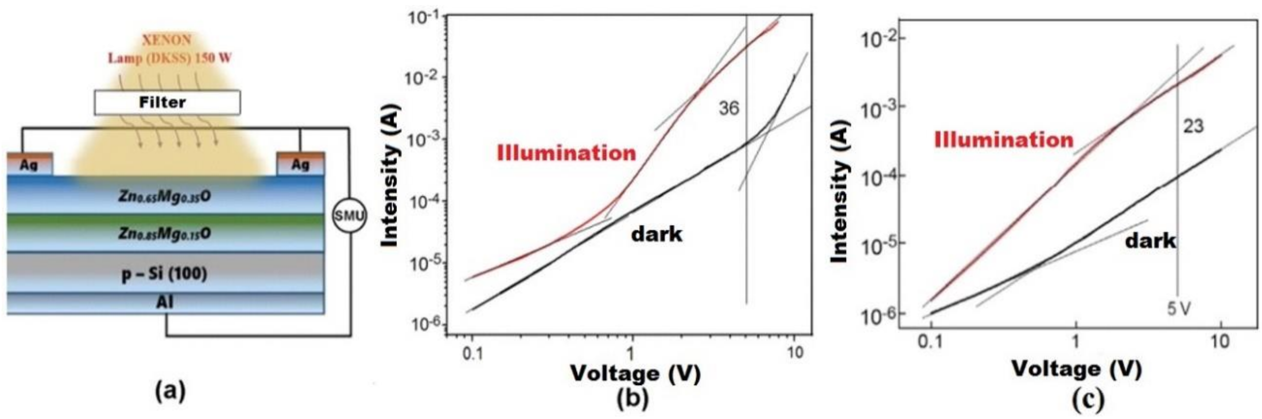

Fig. 15. (a) Design of an Al/p-Si/n-$Zn_{85}Mg_{15}O$/n-$Zn_{65}Mg_{35}O$/Ag photodiode. (b) Current-voltage characteristics of the device fabricated by aerosol deposition method, plotted for double-logarithmic scale direct bias at forward (a) and reverse (b) bias voltages.

The thickness of the absorption film was 300 nm and that of the transparent window approximately 160 nm. Moreover, unlike a classical p-n junction, which functions as a photodetector only at reverse bias, the investigated heterojunction functions as a photodetector at both forward and reverse bias voltages. The value of *n* is about 2-3 at forward bias, while at reverse bias voltages the value of the parameter *n* varies from 1 to 2 both in the dark and under UV light irradiation. The ratio of photocurrent to the dark current increases from 2 to 36 with increasing forward bias from 0.6 V to 5 V, while this ratio is equal to 23 for reverse bias of -5 V, i.e. the sensitivity of the photodetector is 1.5 times higher at forward bias compared to reverse bias (Figure 15 b, c). The responsivity of such a device at forward bias (5 V) is of 460 $mA \cdot W^{-1}$, while at reverse bias (-5 V) it is only of 35 $mA \cdot W^{-1}$.

Consequently, it can be seen that devices made of two junctions are not sensitive to infrared (IR) radiation, unlike devices with a single junction of $Mg_xZn_{1-x}O$ (Figure 16). The responsivity of a single-layer device decreases with increasing the Mg concentration down to 0.1 $mA \cdot W^{-1}$ for *x* value of 0.4 at direct bias of 1 V. On the other hand, the device based on n-$Zn_{0.85}Mg_{0.15}O$/n-$Zn_{0.65}Mg_{0.35}O$ multilayer structure is 8 times more sensitive to UV radiation than to visible radiation, while this ratio is 4 for the n-$Zn_{0.90}Mg_{0.10}O$/n-$Zn_{0.60}Mg_{0.40}O$ film device and it is about 2.5 for the $Zn_{0.80}Mg_{0.20}O$ single film device.

The parameters of the developed photodetectors based on oxide films of $Mg_xZn_{1-x}O$ are summarized in Table 2.

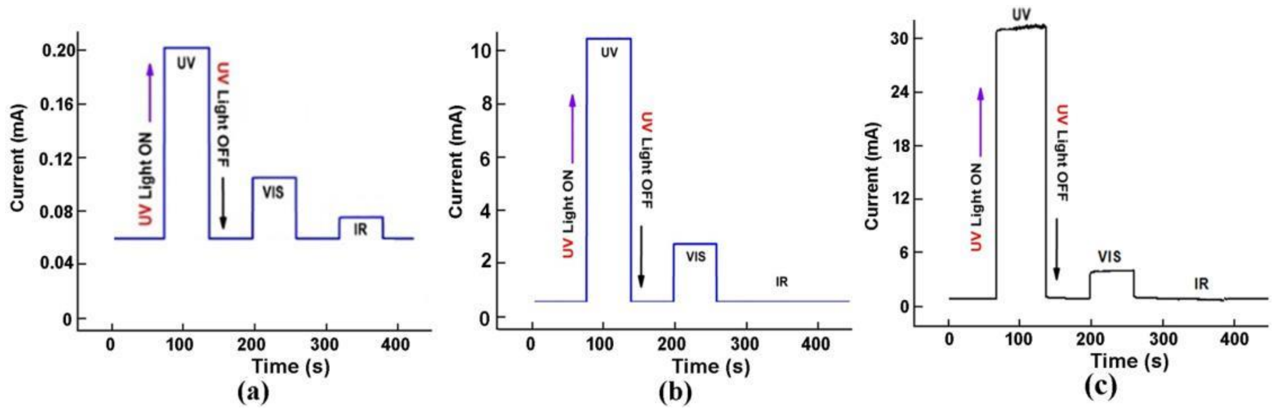

Fig. 16. Photocurrent relaxation measured at 300 K with irradiation at different wavelengths for a photodetector with a $Zn_{0.80}Mg_{0.20}O$ film (a), a photodetector with n-$Zn_{0.90}Mg_{0.10}O$/n-$Zn_{0.60}Mg_{0.40}O$ films (b) and a photodetector with n-$Zn_{0.85}Mg_{0.15}O$ /n-$Zn_{0.65}Mg_{0.35}O$ films (c).

Table 2. Parameters of photodetectors based on $Mg_xZn_{1-x}O$ films for the UV range.

Photodetector structure	Responsivity (R), $mA \cdot W^{-1}$	Detectivity (D*), $cm \cdot Hz^{1/2} \cdot W^{-1}$
$Zn_{0.9}Mg_{0.1}O/Si$	3.0	2.0×10^8
$Zn_{0.8}Mg_{0.2}O/Si$	2.2	1.8×10^8
$Zn_{0.6}Mg_{0.4}O/Si$	0.1	2.0×10^7
$Zn_{0.9}Mg_{0.1}O/Zn_{0.6}Mg_{0.4}O/Si$	150	3.5×10^9
$Zn_{0.85}Mg_{0.15}O/Zn_{0.65}Mg_{0.35}O/Si$	460	1.0×10^{10}

The energy band of MgZnO films increases with increasing Mg concentration. The optical bandgap of films was determined from the point of intersection of the linear segment of the function $(\alpha h\nu)^2$, presented in Figure 17, with the photon energy axis, according to the Tauc formula, and the mathematical relationship is written as follows (1) [17]:

$$(ah\upsilon)^2 = B(h\upsilon-Eg) \quad (1),$$

where, α is the absorption coefficient, hυ corresponds to the photon energy, B is a constant, and the exponential factor 2 corresponds to the allowed direct optical transitions, which can be attributed to the semiconductors of the ZnO family.

A direct bandgap of 3.37 eV results from Figure 17 for the ZnO (curve 1), while bandgaps of 3.75 eV, 4.02 eV and 4.35 eV are estimated for *x* values of 0.2 (curve 2), 0.3 (curve 3) and 0.4 Mg (curve 4), respectively. The bandgap increases up to 5.2 eV with increasing *x* value up to 0.6 (curve 5) for samples prepared by both technological methods (spin coating and aerosol deposition). Therefore, one can see that the sensitivity range of the films can be adjusted from UV - A to UV - C by changing the value of *x* from 0.00 to 0.60.

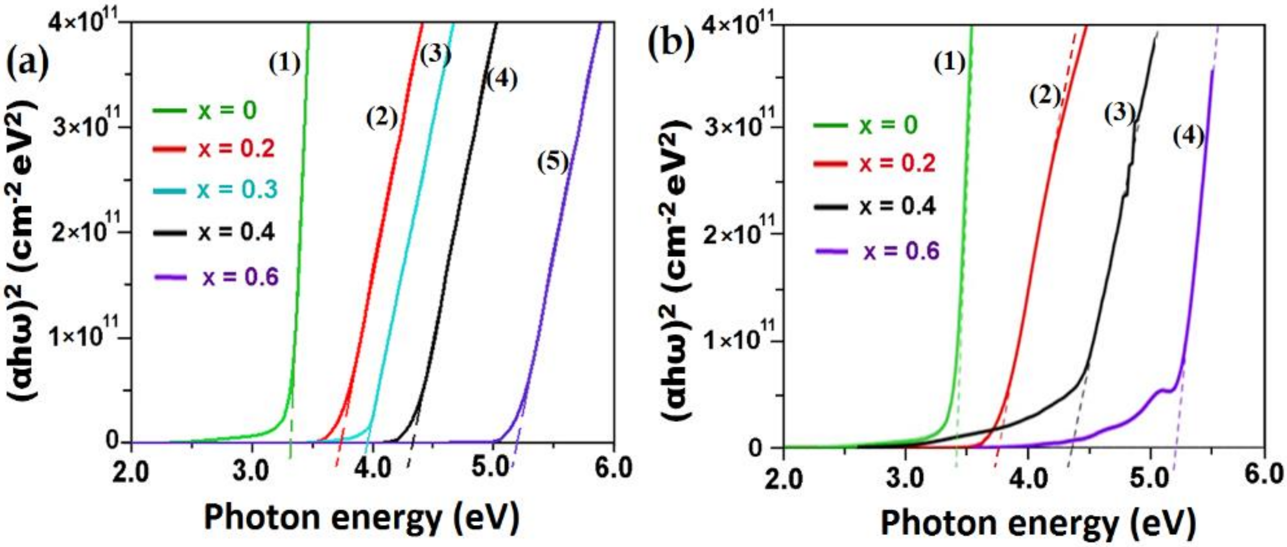

Fig. 17. Tauc plot of optical absorption spectra measured at room temperature for $Mg_xZn_{1-x}O$ films deposited on quartz substrates by spin coating (a) and aerosol deposition method (b).

Figure 18 shows the optical band gap deduced from the Tauc plot as a function of the Mg content in films (*x*). The obtained data are compared with data previously reported in the literature for films prepared by PLD. The experimental data are also fitted to the standard bowing equation, corresponding to the cubic (rock salt) phase (upper curve) and to the wurtzite phase (lower curve). According to the reference [18], the bandgap fits the wurtzite phase up to the value of *x* of 0.27, while it fits the cubic phase for the value of *x* higher than 0.4. On the other hand, our results show that the band gap of films prepared by both the spin coating and aerosol deposition methods fit the curve corresponding to the wurtzite phase up to the value of *x* of 0.6. These data demonstrate that, despite the fact that the films represent a mixture of wurtzite and cubic phases at x-values higher than 0.4, as indicated by the XRD data, the wurtzite phase is predominant and determines the optical bandgap of films with x-values up to 0.6. This observation proves the possibility of extending the band gap of wurtzite-type MgZnO films obtained by spin coating or aerosol deposition method towards shorter wavelengths. So far, a 4.55 eV bandgap has been obtained for a wurtzite-type $Mg_xZn_{1-x}O$ film with an x-value of 0.55 prepared by the MBE growth method on an Al_2O_3 substrate using a quasi-homo $Mg_{0.17}Zn_{0.83}O$ buffer layer. This buffer layer was applied to accommodate the host structural discrepancies and therefore to avoid phase separation in a high Mg content film [21, 22].

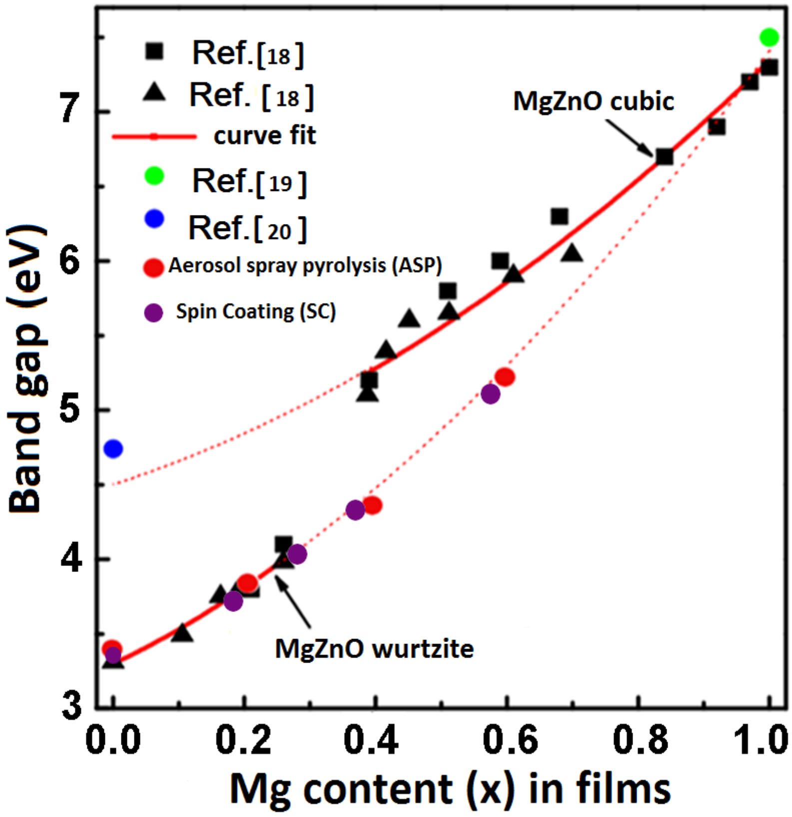

Fig. 18. Dependence of the band gap of $Mg_xZn_{1-x}O$ films on the Mg content in films deposited by spin coating method (SC) (purple dots) and by aerosol spray pyrolysis (ASP) (red dots).

GENERAL CONCLUSIONS AND RECOMMENDATIONS

1. Technological conditions for obtaining $Mg_xZn_{1-x}O$ and $(Ga_xIn_{1-x})_2O_3$ oxides with broad energy band directed composition and morphology have been developed and optimized by cost-effective methods such as spray pyrolysis and spin coating deposition with reproducible morphological, optical and photoelectric properties. (*Subchapter 3.1 and 4.1)*
2. The optimal conditions for obtaining uniform, stoichiometric and highly crystalline MgZnO films by the spin coating method is their thermal treatment in a combined atmosphere (O_2 + Ar) at 500 °C within 60 minutes after the deposition process. (*Subchapter 3.2)*
3. It has been shown that increasing the Mg content leads to shapeless crystallites in the aerosol method, while the spin coating method causes the crystallites to decrease in size from 140 nm to 30 nm. The morphology of the films $(Ga_xIn_{1-x})_2O_3$ also changes, with crystallites increasing in size from 70 nm to 180 nm. (*Subchapter 3.1, 4.1 and 4.3)*
4. XRD and Raman scattering investigations demonstrate the efficient incorporation of Mg atoms into the hexagonal wurtzite structure, on the grounds that the (100), (002), (101) reflexes are gradually shifted towards higher values 2Θ, and the $E_2^{(low)}$ symmetry mode shifts towards higher wavenumbers. These investigations offer the possibility of extending the band gap of wurtzite-type MgZnO films with *x-values* up to 0.8. (*Subchapter 3.1 and 3.2)*
5. It has been shown that the band gap of all the oxide compounds investigated increases with increasing Mg or Ga concentration in the films, so that for the ternary oxide $Mg_xZn_{1-x}O$ with x = 0.6, the band gap increases to 5.1 eV, and for $(Ga_xIn_{1-x})_2O_3$ it changes from 3.62 eV (x = 0.2) to 4.85 eV (x = 0.95). (*Subchapter 5.3 and 5.4)*
6. Based on $Mg_xZn_{1-x}O$ films, photodetectors were developed in MSM configuration, so that by the spin coating method the values of the characterization parameters are R = 380 $mA\cdot W^{-1}$ and D* = 2×10^{10} $cm\times Hz^{1/2}W^{-1}$, while by the aerosol deposition method the parameters are R = 260 $mA\cdot W^{-1}$ and D* = 1×10^{10} $cm\times Hz^{1/2}W^{-1}$. (*Subchapter 5.1)*
7. Heterojunctions operating at direct bias in the injection photodiode regime have been demonstrated based on $Mg_xZn_{1-x}O$ films grown on Si substrates. To improve the photodetector parameters, the $Mg_xZn_{1-x}O$ multilayer design was developed and in this way the responsivity of the devices increased from 3 to 460 $mA\cdot W^{-1}$, and the ratio of illumination current to the dark current increased from 2 to 36. (*Subchapter 5.2)*

RECOMMENDATIONS

1. To obtain MgZnO films with composition and morphology directed by the spin coating method, it is recommended to subject them to heat treatment at 500 °C in a combined atmosphere (O_2 + Ar) and the treatment time after the deposition process should not exceed 60 minutes.
2. When preparing oxide films by the spin coating method, in order to obtain a uniform film thickness similar to that obtained by aerosol deposition for 10 minutes, 10 deposition cycles are recommended.
3. For energy band changing of $Mg_xZn_{1-x}O$ films and obtaining the band gap value around 5 eV, Mg content around 60 atomic % is recommended. In the case of ternary oxide $(Ga_xIn_{1-x})_2O_3$, to raise the band gap to 4.85 eV, it is recommended to set the *x* parameter in the precursor solution to 0.95.
4. For the production of UV radiation photodetectors in the MSM configuration with fast reaction time, it is recommended to deposit the films of oxide compounds by the aerosol deposition method, and for the development of photodetectors with higher photosensitivity, it is recommended to apply deposition by spin coating method.
5. To improve the responsivity, detectivity, UV selectivity and response time parameters of heterojunction photodetectors based on $Mg_xZn_{1-x}O$ films on p-Si substrates, it is recommended to use the two-film design, one of which plays the role of a window and the second is the absorption layer.

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Articles in journals from Web of Science and SCOPUS databases with impact factor:

1. **MORARI, V.**, URSAKI, V. V., RUSU, E. V., ZALAMAI, V. V., COLPO, P., TIGINYANU, I. M. Spin-Coating and Aerosol Spray Pyrolysis Processed MgxZn1-xO Films for UV Detector Applications. În: *Nanomaterials*, 2022, Vol. 12, Nr. 18, 3209, (10pp). https://doi.org/10.3390/nano12183209. **IF = 5.570.**
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3. **MORARI, V.,** PANTAZI, A., CURMEI, N., POSTOLACHE, V., RUSU, E. V., ENACHESCU, M., TIGHINEANU, I. M., URSAKI, V. V. Band tail state related photoluminescence and photoresponse of MgZnO solid solutions nanostructured films. În: *Beilstein Journal of Nanotechnology,* 2020, Vol. 11, pp. 899–910. https://doi.org/10.3762/bjnano.11.75. **IF = 2.612.**

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ADNOTARE

la teza cu titlul **„Tehnologii de obținere și proprietățile optice și fotoelectrice în sistemul ZnO - $Mg_xZn_{1-x}O$ pentru aplicații optoelectronice”**, înaintată de candidatul Morari Vadim, pentru conferirea titlului științific de doctor în științe fizice, la specialitatea **134.01 - Fizica și Tehnologia Materialelor**.

Structura tezei: Teza este compusă din introducere, 5 capitole, concluzii generale și recomandări bibliografie din 159 de titluri, 123 pagini text de bază, 70 de figuri și 21 de tabele. Rezultatele obținute au fost publicate în 18 lucrări științifice, dintre care 3 articole în reviste cu factor de impact, 2 articole în reviste naționale și 9 lucrări la conferințe naționale și internaționale.
Cuvinte cheie: filme oxidice semiconductoare, soluții solide $Mg_xZn_{1-x}O$, depunere prin centrifugare, depunere din aerosoli, microscopie electronică, difracție de raze X, banda interzisă, fotodetectori.
Scopul lucrării: Elaborarea tehnologiilor de obținere a filmelor oxidice cu banda interzisă largă prin metode cost-efective, inclusiv a filmelor nanostructurate în sistemul ZnO-$Mg_xZn_{1-x}O$ cu compoziție și morfologie dirijată și explorarea proprietăților lor optice, luminescente și fotoelectrice pentru aplicații în dispozitive optice și optoelectronice, precum filtre de transmisie și fotodetectoare de radiație ultravioletă.
Obiectivele cercetării: Elaborarea tehnologiilor de obținere a soluțiilor solide $Mg_xZn_{1-x}O$, a compușilor oxidici $(Ga_xIn_{1-x})_2O_3$ cu compoziție și morfologie dirijată, prin metoda de depunere din aerosoli și depunere prin centrifugare. Studiul proprietăților morfologice, structurale, vibraționale, optice și fotoelectrice ale filmelor obținute în funcție de compoziția lor și parametrii tehnologici aplicați. Elaborarea și caracterizarea fotoreceptoarelor de radiație optică în baza filmelor obținute. Efectuarea studiului comparativ a materialelor oxidice de $(Ga_xIn_{1-x})_2O_3$ cu banda energetică largă.
Noutatea și originalitatea științifică: A fost stabilită influența parametrilor tehnologici de obținere a filmelor oxidice prin metode cost-efective asupra morfologiei, compoziției chimice, structurii cristaline și proprietăților vibraționale, optice și fotoelectrice. În premieră au fost elaborate fotodetectoare de radiație UV în baza soluțiilor solide $Mg_xZn_{1-x}O$, care funcționează în regim de fotodiode cu injecție la polarizare directă, iar în baza comparației parametrilor lor cu cei ai fotodetectoarelor MSM a fost identificat design-ul optimal din punct de vedere al responsivității, detectivității, selectivității și timpului de reacție al dispozitivului.
Problema științifică principală soluționată: Elaborarea și optimizarea tehnologiilor de obținere a filmelor oxidice cu banda energetică largă cu proprietăți fizice dirijate prin metode cost-efective pentru aplicații în dispozitive optoelectronice.
Semnificația teoretică și aplicativă a lucrării: În baza analizei spectrelor de absorbție optică a fost stabilită dependența benzii interzise a filmelor oxidice de compoziția chimică, iar analiza spectrelor de fotoluminescență a scos în evidență influența tehnologiilor aplicate asupra distribuției stărilor energetice în banda interzisă, care au efecte asupra caracteristicilor fotoelectrice. Tehnologiile elaborate au fost aplicate pentru elaborarea fotodetectoarelor de radiație UV în diferite configurații, care acoperă o parte din diapazonul vizibil, UV-A, UV-B și UV-C.
Implementarea rezultatelor științifice: Fotodetectoarele în baza filmelor oxidice de Ag/n-$Zn_{0.65}Mg_{0.35}O$/n-$Zn_{0.85}Mg_{0.15}O$/p-Si/Al sunt aplicate pentru detectarea radiației optice, mai ales a radiației UV de la UV-A spre UV-C (*brevet de invenție Nr. 4618*).

ABSTRACT

of the thesis entitled **„Technologies of obtaining and optical and photoelectric properties of the ZnO-$Mg_xZn_{1-x}O$ system for optoelectronic applications"**, presented by the candidate Morari Vadim, for obtaining the degree of Doctor in Physical Sciences with specialty **134.01 - Physics and technology of materials**.

Thesis structure: The thesis consists of an introduction, 5 chapters, general conclusions and recommendations, bibliography from 159 titles, 123 pages of basic text, 70 figures and 21 tables. The results were published in 18 scientific papers, including 3 articles in journals with impact factor, 2 articles in national journals and 9 papers at national and international conferences.

Keywords: Oxide semiconductor films, $Mg_xZn_{1-x}O$ solid solutions, spin coating, aerosol deposition, electron microscopy, X-ray diffraction, bandgap, UV photodetectors.

The aim of the work: Development of technologies for obtaining wide-bandgap oxide films by cost effective methods, including nanostructured films of the ZnO-$Mg_xZn_{1-x}O$ system with controlled composition and morphology, and exploring their optical, luminescent, and photoelectric properties for applications in optical and optoelectronic devices, such as transmission filters and photodetectors for UV radiation.

Research objectives: Development of technologies for obtaining films of $Mg_xZn_{1-x}O$ solid solutions, $(Ga_xIn_{1-x})_2O_3$ oxide compounds with controlled composition and morphology, by means of spin coating and aerosol deposition. Study of morphological structural, vibration, mechanic, optical and photoelectrical properties of the produced films depending on their composition and technological parameters. Development and characterization of optical radiation photodetectors based on the produced films. Conducting a comparative study of $(Ga_xIn_{1-x})_2O_3$ oxide materials with wide band gap with the basic material researched in the thesis.

Scientific novelty and originality of the results: The influence of technological parameters for obtaining the oxide films by cost-effective methods on their morphology, chemical composition, crystallographic structure, and vibration, optical and photoelectric properties was established. UV photodetectors working as injection photodiodes at direct bias have been developed for the first time on the basis $Mg_xZn_{1-x}O$ solid solutions films, and their optimal design from the point of view of responsivity, detectivity, selectivity and response time of the device was identified through a comparison of their parameters with those of MSM photodetectors.

The main scientific problem solved: Development and optimization of technologies for obtaining wide-bandgap oxide films with controlled physical properties by means of cost-effective methods for applications in optoelectronic devices.

Theoretical significance and practical value of the work: The dependence of the bandgap of oxide films on their chemical composition was established as a result of the analysis of the optical absorption spectra, while the analysis of the luminescence spectra highlighted the influence of the applied technologies on the distribution of energy states in the bandgap, which impact the photoelectric characteristics. The developed technologies have been applied to the development of UV radiation photodetectors in various configurations, covering a part of the visible range, UV-A, UV-B and UV-C wavelengths.

Implementation of scientific results: Photodetectors based on Ag/n-$Zn_{0.65}Mg_{0.35}O$/n-$Zn_{0.85}Mg_{0.15}O$/p-Si/Al oxide films are applied to detect optical radiation, especially UV radiation from UV-A to UV-C (*invention patent No. 4618*).

АННОТАЦИЯ

Диссертация **„Технологии получения и оптические и фотоэлектрические свойства в системе ZnO-$Mg_xZn_{1-x}O$ для оптоэлектронных применений"**, представленной Вадимом Морарь на соискание степени доктора физических наук по специальности **134.01 - Физика и технология материалов.**

Структура диссертации: Диссертация состоит из введения, 5 глав, общих выводов и рекомендаций, библиографии из 159 наименований, 123 страниц основного текста, 70 рисунков и 21 таблиц. Результаты работы опубликованы в 18 научных работах, в том числе 3 статьи в журналах с импакт-фактором, 2 статьи в отечественных журналах и 9 докладов на всероссийских и международных конференциях.

Ключевые слова: оксидные полупроводниковые пленки, твердые растворы $Mg_xZn_{1-x}O$, осаждение путем центрифугирования и аэрозолями, запрещенная зона, фотоприемники.

Цель работы: Разработка технологий для получения широкозонных оксидных пленок, в том числе наноструктурированных пленок в системе ZnO-$Mg_xZn_{1-x}O$ с управляемым составом и морфологией и исследование их оптических, люминесцентных и фотоэлектрических свойств для применения в оптических и оптоэлектронных устройствах, таких как УФ фотоприемники.

Задачи работы: Разработка технологий для получения пленок твердых растворов $Mg_xZn_{1-x}O$, оксидных соединений $(Ga_xIn_{1-x})_2O_3$ с управляемым составом и морфологией, методом ценрифугирования и осаждения из аэрозолей. Исследование морфологии, структурных, колебательных, механических, оптических и фотоэлектрических свойств в зависимости от их состава и технологических параметров их осаждения. Разработка и характеризация фотоприемников оптического излучения на основе оксидных пленок $Mg_xZn_{1-x}O$ и $(Ga_xIn_{1-x})_2O_3$.

Новизна и оригинальность: Установлено влияние технологических параметров осаждения оксидных пленок экономичными методами на морфологию, химический состав, кристаллографической структуры, колебательных, оптических и фотоэлектрических свойств. Впервые разработаны фотоприемники УФ излучения на основе пленок твердых растворов MgxZn1-xO, работающие как инжекционные фотодиоды при прямом смещении, а путем сравнения их параметров с фотоприемниками полупроводник-метал-полупроводник найдена оптимальная конструкция прибора с точки зрения чувствительности, обнаружительной способности, избирательности и времени отклика.

Решенная научная проблема: Разработка и оптимизация технологий получения широкозонных оксидных пленок с управляемыми физическими свойствами экономичными методами для применения в оптоэлектронных приборах.

Теоретическая значимость и прикладная ценность работы: На основе анализа спектров оптического поглощения, была установлена зависимость *запрещённой* зоны оксидных пленок от химического состава, а анализ спектров фотолюминесценции выявил влияние применяемых технологий на распределение энергетических состояний в запрещенной зоне, которые сказываются на фотоэлектрические характеристики. Разработанные технологии применены для разработки фотоприемников УФ-излучения в различных конфигурациях, охватывающих часть видимого диапазона, УФ-А, УФ-В и УФ-С.

Реализация научных результатов: Фотоприемники на основе оксидных пленок Ag/n-$Zn_{0.65}Mg_{0.35}O$/n-$Zn_{0.85}Mg_{0.15}O$/p-Si/Al применяются для обнаружения оптического излучения, в частности, ультрафиолетового излучения от УФ-А до УФ-С (*патент на изобретение № 4618*).

MORARI VADIM

OPTICAL AND PHOTOELECTRIC PRODUCTION TECHNOLOGIES AND PROPERTIES IN THE ZnO - $Mg_xZn_{1-x}O$ SYSTEM FOR OPTOELECTRONIC APPLICATIONS

134.01 - Physics and technology of materials

Abstract of the PhD thesis

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