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Research Article

Optical Properties of Zno + 12% ITO Thin Films on Substrates of Anode Aluminum Oxide

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Abstract

ZnO thin films of zinc oxide doped with 12 % ITO (indium tin oxide) on anodic aluminum oxide substrates are formed in vacuum during high-frequency repetitively pulsed laser deposition. The morphology of films on porous and non-porous surfaces of substrates was studied by atomic force microscopy. The optical properties of the films in the visible, near, and middle IR regions of the electromagnetic radiation spectrum, the Raman spectra, and also the features of the photoluminescence characteristics have been experimentally investigated. Zinc oxide films can be used in optoelectronic transducers, as luminescent material, in the form of transparent electrodes, sensitive layers of gas and biological sensors, catalysts, X-ray and gamma-radiation detectors.

الملخص.

تتشكل الأغشية الرقيقة من أكسيد الزنك *(ZnO)* ( ) (أكسيد الإنديوم) على ركائز أكسيد الألمنيوم المؤكسد *ITO* المخدر مع 12٪ في الفراغ أثناء ترسيب الليزر النبضي عالي التردد المتكرر. تمت دراسة مورفولوجيا الأفلام على الأسطح المسامية وغير المسامية للركائز بواسطة الفحص المجهري للقوة الذرية. الخواص البصرية للأفلام في مناطق الأشعة تحت الحمراء المرئية والقريبة والمتوسطة من طيف الإشعاع الكهرومغناطيسي، أطياف رامان، وكذلك ميزات خصائص التلألؤ الضوئي تم اختبارها تجريبياً. يمكن استخدام أفلام أكسيد الزنك في محولات الطاقة الضوئية الإلكترونية، مثل مادة الإنارة، في شكل أقطاب شفافة، وطبقات حساسة من الغاز وأجهزة الاستشعار البيولوجية، والمحفزات، وأجهزة الكشف عن الأشعة السينية وأشعة جاما.

Keywords: *ZnO thin films, high-frequency pulsed laser deposition, anodic alumina substrates.*

Introduction

Thin transparent conducting oxide films are successfully used both in science and in technology (Yu X *et al*, 2006; Stadler A, 2012; Semikina T.V *et al*, 2010; Wagner J.F. *et al*, 2008). Zinc oxide is a wide-gap semiconductor with unique electrophysical and optical properties (Wöll C, 2007; Janotti A *et al*, 2009; Ü. Özgür *et al*, 2005; Ellmer K, 2008; Djurišić A.B *et al*, 2010, N. Qin, 2011). Transparent conductive films of zinc oxide, belonging to the group of transparent conductive oxides (TCO, transparent conducting oxide), combine relatively high values of optical transmission and electrical conductivity and are of great interest for the creation of various optoelectronic devices and devices (M. Caglar, 2009; G.V. Lashkaryov, 2017; L.K. Krastev, 2013; A.A. Bobkov, 2015; N.A. Lashkova, 2016).

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Zinc oxide can be used as a material in optoelectronic converters as a luminescent material, in the form of transparent electrodes, sensitive layers of gas and biological sensors, catalysts, ultraviolet, X-ray, and gamma radiation detectors (D. Tainoff, 2008; D. Tainoff, 2014).

To obtain such films, vacuum deposition is used

Thermal, electron-beam, ion-plasma, magnetron, pulsed-laser, chemical vapor-phase (Zima V.N *et al*, 2013; N.P. Klochko, 2013; Semikina T.V. *et al*, 2016). Each of these methods has certain advantages and disadvantages. Pulsed laser deposition of thin oxide films has certain advantages (Kashkul I.N.K., 2017; Chumakov A.N, 2012; A.N. Chumakov, 2012; A.N. Chumakov, 2016; Chumakov A.N *et al*, 2017) [22–26]. ZnO + 12% ITO films were deposited on anodic alumina substrates by multipulse high-frequency laser evaporation in vacuum. Substrates of anodic aluminum oxide were obtained by electrochemical oxidation

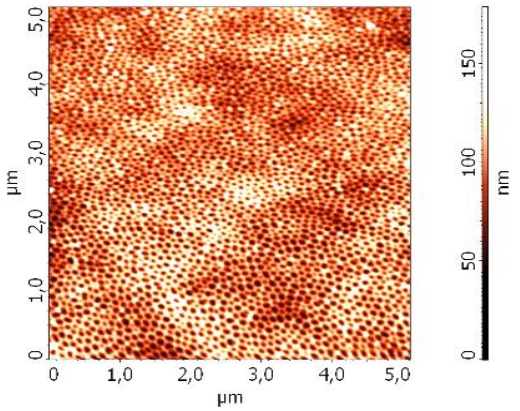
(anodization) of aluminum, followed by conversion to γ-oxide by annealing at a temperature of 850 ° C. The results of the study include the morphology of the formed ZnO + 12% ITO thin films on γ-Al2O3 substrates, their transmission and Raman scattering spectra, and also photoluminescence spectra.

Methodology for the preparation and study of thin films

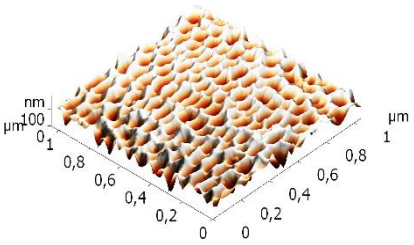
The experimental laser setup contained: a laser source with an adjustable laser pulse repetition rate from 5 to 50 kHz, an optical system for directing laser radiation at an appropriate target composition, a vacuum chamber and a measurement and diagnostic module. The source of radiation in the setup was a neodymium glass laser (operating wavelength λ = 1.06 μm). To obtain a multi-pulse mode of laser generation with a high pulse repetition rate inside the cavity, a passive optical shutter of crystalline lithium fluoride LiF with $F2^-$ color centers previously irradiated was installed near the blind mirror. By varying the laser pump level and the optical density of the shutter, a change in the laser pulse repetition rate was achieved, and the laser pulse duration at half maximum was about 85 ns. The vacuum system of the installation provided experiments at a pressure reduced to 2.7 Pa. The operating mode of effective erosive plasma formation was realized due to the multi-pulse high-frequency laser action on the surface of the sprayed target. The deposition of macroscopically homogeneous thin films was achieved at a laser power density of q = 32 MW / cm2 and a pulse repetition rate of f ~ 20–30 kHz. The surface topography of the samples was studied using a Solver P47-Pro scanning probe microscope (NT-MDT, Russia) in the semi-contact mode. The dependence of the transmission of optical radiation by thin films in a wide spectral range (from ultraviolet (UV) to near infrared (IR) was measured on a Carry 500 Scan spectrophotometer, which is a multi-purpose, automated system that provides measurement, processing of output information and its registration. IR transmission spectra thin films were also measured using a NEXUS infrared Fourier spectrometer (Thermo Nicolet). The spectra were recorded in the spectral range 500–4000 cm-1 with a resolution of 2 cm-1 after 128 s canals using a DTGS detector. Raman spectra were recorded in the spectral range 320–2500 cm-1 on a SpectroPro 500i spectrometer after 30 scans. The scattering was excited by radiation with a wavelength of 532 nm, the power of which on the sample did not exceed 25 mW. Collection of scattered radiation - "backscattering." Photoluminescence spectra were recorded on an automated spectrofluorimeter SM 2203 (firm SOLAR).

The results obtained and their discussion

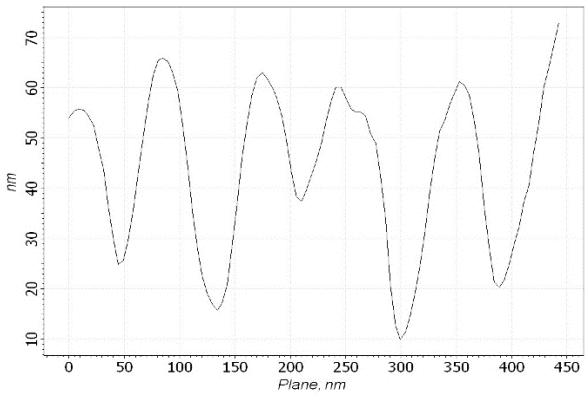
Using atomic force microscopy, it was found that the initial substrate of □-alumina has a porous (Fig. 1, a, b) and non-porous surface (Fig. 1, d, e). As can be seen from fig. 1a, d, the pores are arranged orderly over the entire surface of the substrate. An analysis of the cross-sectional profile along the selected lines in Fig. 1c, f showed that the average pore diameter is 50 nm, while the distance between the pores is 40-60 nm.



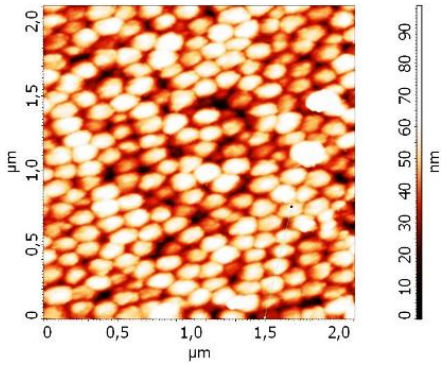
(a)



(b)



(c)



(d)

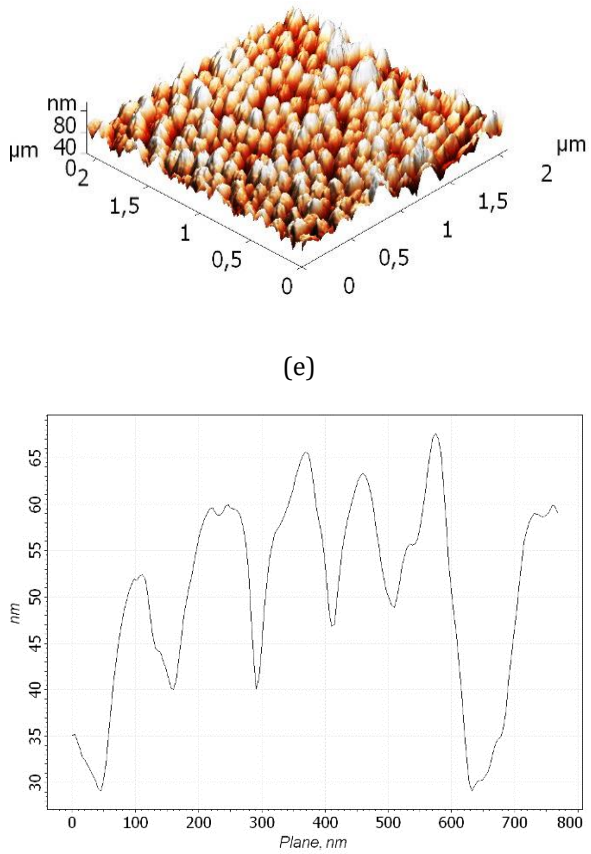
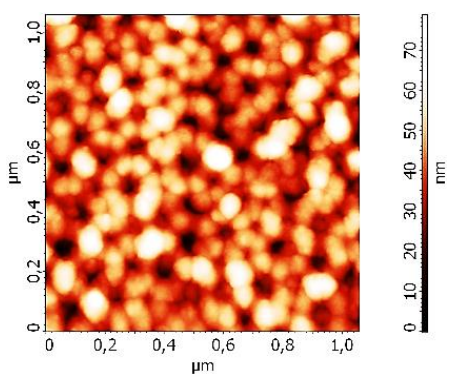


Fig. 1. Surface topographies of the initial substrate of alumina: on the porous side:
a - two-dimensional image, b - three-dimensional image, c - sectional profile; from the non-porous side: d - two-dimensional image, e - three-dimensional image, f - section profile along the selected line.

When a ZnO + 10% ITO film is deposited on a из-alumina substrate by the method of multipulse high-frequency laser evaporation in vacuum, a nanoscale structure is formed. As can be seen from fig. 2, the film repeats the structure of the substrate, while the pores of the substrate are filled with partially or completely deposited material. The average size of the structural elements of the film is 50 nm; however, particles with a lateral size of 80–100 nm are found on the surface, but their fraction is insignificant.



(a)

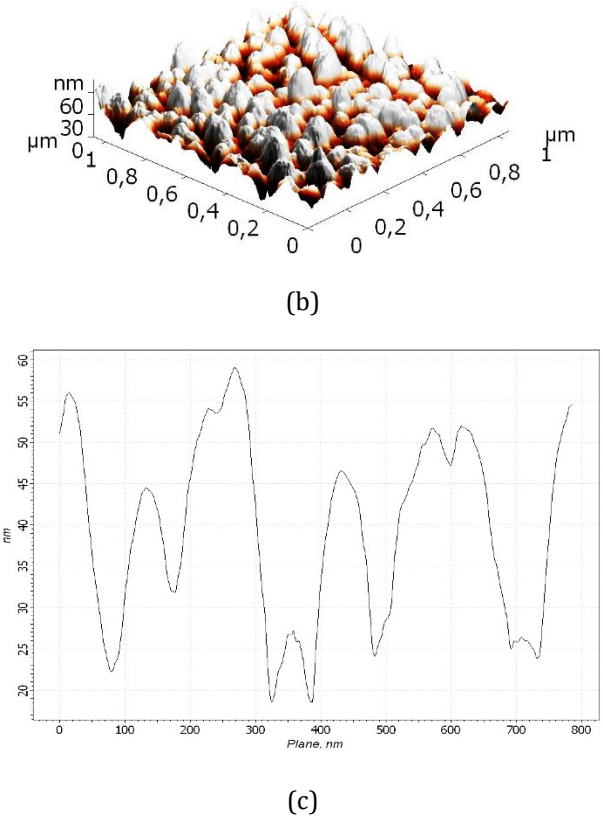
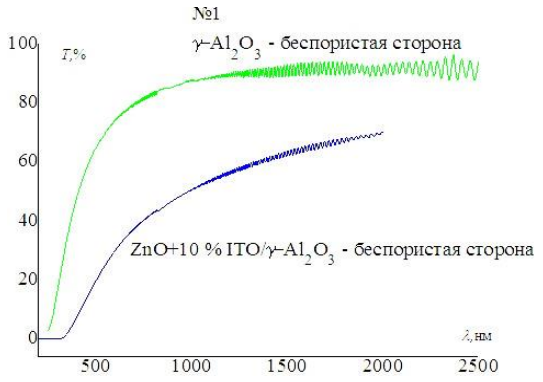


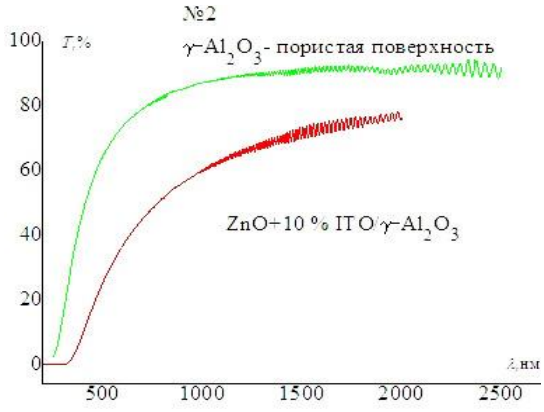
Fig. 2. Surface topography of a laser-deposited thin ZnO + 12% ITO film on a substrate
□-alumina from the porous side: a, c - two-dimensional image; b - three-dimensional image; d - sectional profile along the selected line.

The transmission of the alumina substrate from the non-porous side and the ZnO zinc oxide films deposited on it doped with 12% ITO (indium-tin oxide) (sample no. 1) are shown in Fig. 3 a. The transmission spectra of the □-alumina substrate from the side of the porous surface and the zinc oxide films deposited on it (sample No. 2), shown in Fig. 3b were recorded on a Carry 500 Scan spectrophotometer in the UV, visible and near-IR spectral ranges. The transmission of the alumina substrate on the non-porous side sharply increases to 600 nm, then slows down and reaches 90% in the wavelength region above 1100 nm, while the transmission for the zinc oxide film on the ?-alumina substrate on the non-porous side is characterized by smooth growth, moreover in both cases, transmission oscillations with a characteristic period of 30 nm are noticeable in the region of wavelengths above 1100 nm.

The transmission of the alumina substrate from the porous surface increases sharply to 600 nm, then slows down and reaches 90% in the wavelength region above 1200 nm, while the transmission for the zinc oxide film on the □-alumina substrate of the porous surface is characterized by a noticeable increase, in both cases, transmission oscillations are noticeable in the wavelength region above 1200 nm.



(a)



(b)

Fig 3. Transmission spectra in the visible and near infrared region of the alumina substrate on the non-porous side and zinc oxide film with ITO additives on it (No. 1, a) and transmission spectra of a alumina substrate on the porous surface side and a zinc oxide film with ITO additives on it (No. 2, b)

In fig. 4b and 5b, the transmission band is visible in the region of 1260 cm – 1, and the absorption intensity of the OH group in the IR spectrum (3500 cm – 1) has significantly decreased. When recording Raman spectra, the luminescence maximum is observed for the initial sample at 1726 cm-1 (Fig. 6a), and for the coated sample at 1892 cm-1 (Fig. 6b).

From the condition of interference intensity maxima, we can estimate the thickness of the deposited film of zinc oxide h:

$$2hn = m\lambda. \quad (1)$$

Taking into account the first order of interference m = 1, the wavelength λ = 1.5 μm and the refractive index of zinc oxide n = 2.02, we obtain the approximate value of the deposited zinc oxide film thickness h 0.4 μm.

The luminescence spectra of samples No. 1 and No. 2 practically coincide in shape (Fig. 7) and are independent of the excitation wavelength. The luminescence excitation spectra of samples No. 1 and No. 2 also almost coincide in shape and are independent of the recording wavelength (Fig. 8). These experimental facts suggest that the glow belongs to the emission centers of the same type. However, the luminescence intensity of sample No. 2 is more than 2 times higher than the luminescence intensity of sample No. 1.

The substantiation of the use of nanoporous alumina substrates with a high level of pore regularity filled with sensitive layers has been carried out.

Designs of an ionizing radiation detector are proposed that are created by filling particles of a luminescent material based on compounds of ZnO salts that respond to radioactive and ultraviolet radiation, porous from Al2O3.

A technique has been developed for the formation of sensitive ZnO layers using synthesis in aqueous and non-aqueous solutions, which made it possible to obtain precipitation without cadmium and zinc oxides.

1. Experimental substantiation of heat treatment modes for modifying porous amorphous alumina based on its placement between flat ceramic plates with a load of 0.02 kg per 1 cm2, which allowed the formation of a high-temperature metastable γ-phase Al2O3 with a shrink coefficient of up to 0.6% s surface irregularity ± 15 nm.

2. The technique for filling through pores in Al oxide with particles of a luminescent material, based on the vacuum method for the conditions of movement of colloidal solutions in microholes of nanoporous Al2O3, the introduction of additional ultrasonic treatment, which allowed the formation of ZnO sediments in microholes from aqueous and non-aqueous solutions.

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