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OPTICAL AND ELECTRICAL PROPERTIES OF Tb–ZnO/SiO_2 STRUCTURE IN THE INFRARED SPECTRAL INTERVAL

Optical and electrophysical properties of terbium-doped zinc oxide films have been studied, by using the external reflection IR spectroscopy. The films were deposited onto silicon oxide substrates with the help of the magnetron sputtering method. A theoretical analysis of the reflection spectra of the ZnO/SiO_2 structure is carried out in the framework of a multioscillatory model in the spectral interval 50–1500 cm^{-1} and for the electrical field orientation perpendicular to the c-axis (E⊥c). The method of dispersion analysis is applied to determine the optical and electrical properties of ZnO films, as well as the oscillator strengths and damping coefficients in the ZnO film and the SiO_2 substrate. The influences of the phonon and plasmon-phonon subsystems in the ZnO film on the shape of IR reflection spectra registered from the Tb–ZnO/SiO_2 structure are elucidated.

K e y w o r d s: zinc oxide, SiO_2, IR reflection, thin film, dielectric substrate, phonon, plasmon, electron concentration.

1. Introduction

The development of modern opto- and nanoelectronics is inseparably linked with the miniaturization of certain functional elements in devices and their components. This became possible due to technological capabilities achieved in growing thin poly- and single-crystalline films. The properties of such systems will evidently be determined, first of all, by the quality of grown films and a capability to predict their optical and electrical characteristics [1–3].

Nowadays, there are a number of scientific works devoted to the study of the optical and electrophysical properties of thin semiconductor films on dielectric and semiconductor substrates [4–14]. At the same time, the influence of the film and substrate properties on the parameters of the film-substrate structure has not been analyzed enough. Moreover, the interaction of phonon and plasmon film excitations with the phonon subsystem of a substrate can substantially change the film properties in comparison with the properties of corresponding single crystals [3, 5, 6].

Among many studies dealing with the physical properties of thin films that stimulate their widespread application in various domains of science and engineering, the research of the optical and electrical properties of thin zinc oxide films deposited onto dielectric substrates is of considerable interest [4–12]. The choice of zinc oxide as an object to study is associated with its physical and chemical properties (optical, mechanical, piezoelectric, and others). It is one of the most promising materials that can become a basic one for a lot of innovations in photonics and spintronics in the next decades [15]. Furthermore, the unique optical properties of monocrystalline ZnO make this material suitable for the development of new opto- and nanoelectronic devices [1, 16]. Due to a wide band gap (3.37 eV at 300 K), zinc oxide is widely used as a material for short-wave light sources. It can be an alternative to GaN and SiC compounds, the cost price of which is a few orders of magnitude higher [17–19]. However, ZnO films have even more promising prospects in comparison with ZnO single crystals. ZnO films are used as transparent layers and, depending on the doping level, they can be either electrical insulators or conductors. As was shown in works [5, 20–22], the specific resistance ρ in thin ZnO films can be varied from 10^{-4} to 10^{10} $\Omega \times$ cm.

However, it should be noted that, although this topic is challenging and there is a large number of publications devoted to the researches of zinc oxide films, the available information in the literature concerning the study of the interaction between electromagnetic radiation and oscillations of various types (e.g., phonons, plasmons, and others) is not sufficient. Note that the interaction of dipole oscillations with free electrons in a zinc oxide film and with dipole oscillations in a substrate substantially affects the behavior of both the bulk and surface phonon and plasmon-phonon excitations, which significantly changes the optical properties of the structure as a whole. In this connection, there is an interest in the study of the optical and electrophysical properties of ZnO films on dielectric substrates. It is important to unveil the interaction of the phonon and plasmon-phonon subsystems of the film with the phonon subsystem of the substrate in a wide infrared spectral interval.

The aim of this work was to study terbium-doped ZnO films deposited on a SiO_2 substrate (Tb-ZnO/SiO_2 structures) in the spectral range of the ZnO plasmon-phonon resonance, by using the method of external reflection infrared (IR) spectroscopy in the geometry where the light incidence angles are close to the normal one. It is known that the Tb impurity is considered promising to obtain the green luminescence and to attain a high conductivity in the films. However, the literature data concerning the latter issue are rather controversial [23, 24].

Infrared spectroscopy is one of the most informative methods for studying the optical and electrophysical properties of thin films. It provides information not only about the physical and chemical properties of a film, but also about the parameters of the substrate and the quality of its surface treatment [5, 12].

A detailed analysis of the procedure aimed at obtaining the mutually consistent parameters for a single-oscillator model of ZnO was carried out in works [5, 25]. In work [25], we showed that ZnO is characterized by a significant anisotropy in the properties of its phonon subsystem and a weak anisotropy in the properties of its plasma subsystem. As a result, zinc oxide films are good model objects, which are convenient for the research of optical and electrophysical properties in the IR spectral interval in the presence of a coupling between long-wavelength optical vibrations in the lattice, film, and substrate, on the one hand, and the electron plasma in the zinc oxide film, on the other hand.

2. Experimental Technique

Films of ZnO doped with terbium were deposited onto SiO_2 substrates with the help of the radio-frequency magnetron sputtering in the plasma of argon ions. A zinc oxide target covered with calibrated Tb_4O_7 disks was used. Plates of synthetic amorphous silicon oxide (of the JGS1 type) with $1 \times 1 \times 0.2$ cm^3 dimensions and polished from both sides were used as substrates. Crystals of silicon oxide of this type are characterized by a high purity and the absence of pores and bubbles. They are transparent in the ultraviolet and visible spectral intervals. They do not absorb in a wavelength interval of 175–250 nm and intensively absorb in an interval of 2600–2800 nm (3550–3850 cm^{-1}), owing to the presence of OH groups.

The substrate temperature was 100°C. The power density at the target was 1.91 W/cm^2. The layer thickness was 632 ± 2 nm. The deposited films were found to be polycrystalline with their c-axis oriented perpendicularly to the substrate surface. The content of Tb^{3+} ions was about 3 at.%.

The infrared reflection spectra were measured at room temperature, by using a Bruker Vertex 70 V FTIR spectrometer. The incidence angle of excitation light was 13°. A golden mirror was used as a reference. The spectra were registered with a resolution of 1 cm^{-1}. The orientation of the electric field was selected to be perpendicular to the c-axis of the SiO_2 substrate. The experimental procedure was described in more details in works [7, 8].

3. Theoretical Part

The IR reflection spectra $R(\nu)$ of absorbing ZnO films deposited on a "semiinfinite" SiO_2 substrate were theoretically calculated in the spectral interval of "residual rays" for the film and the substrate. In so doing, the following mathematical expressions were applied [5, 7, 8]:

$$R(\nu) = \left\{\left(q_1^2 + h_1^2\right)\exp(\gamma_2) + \left(q_2^2 + h_2^2\right) \times \right.$$
$$\times \exp(-\gamma_2) + A\cos\delta_2 + B\sin\delta_2\Big\}/$$
$$/\Big\{\exp(\gamma_2) + \left(q_1^2 + h_1^2\right)\left(q_2^2 + h_2^2\right) \times$$
$$\left.\times \exp(-\gamma_2) + C\cos\delta_2 + D\sin\delta_2\right\}, \qquad (1)$$

where

$$A = 2\left(q_1 q_2 + h_1 h_2\right), \quad B = 2\left(q_1 h_2 - q_2 h_1\right),$$
$$C = 2\left(q_1 q_2 - h_1 h_2\right), \quad D = 2\left(q_1 h_2 + q_2 h_1\right),$$
$$q_1 = \frac{n_1^2 - n_2^2 - k_2^2}{\left(n_1 + n_2\right)^2 + k_2^2}, \quad h_1 = \frac{2 n_1 k_2}{\left(n_1 + n_2\right)^2 + k_2^2},$$
$$q_2 = \frac{n_2^2 - n_3^2 + k_2^2 - k_3^2}{\left(n_2 + n_3\right)^2 + \left(k_2 + k_3\right)^2},$$
$$h_2 = \frac{2\left(n_2 k_3 - n_3 k_2\right)}{\left(n_2 + n_3\right)^2 + \left(k_2 + k_3\right)^2},$$
$$\gamma_2 = \left(4\pi k_2 d\right)/\lambda, \quad \delta_2 = \left(4\pi n_2 d\right)/\lambda,$$

n_1, n_2, and n_3 are the refractive indices, and k_1, k_2, and k_3 the absorption indices of air, the ZnO film with thickness d_f, and SiO_2 substrate, respectively. The refractive indices n_2 and n_3 were calculated in the framework of the dielectric permittivity model with an additional contribution of active optical phonons ν_T and plasmons ν_p in the film and phonons in the substrate [9, 18, 19], by using the formula

$$\varepsilon_j\left(\nu\right) = \varepsilon_{1j}\left(\nu\right) + i\varepsilon_{2j}\left(\nu\right) = \\ = \varepsilon_{\infty j} + \frac{\varepsilon_{\infty j}\left(\nu_{Lj}^2 - \nu_{Tj}^2\right)}{\nu_{Tj}^2 - \nu^2 - i\nu\gamma_{fj}} - \frac{\nu_{pj}^2 \varepsilon_{\infty j}}{\nu\left(\nu + i\gamma_{pj}\right)}, \qquad (2)$$

where ν_{Tj} and ν_{Lj} are the frequencies of the transverse and longitudinal, respectively, optical phonons in the film and the substrate; γ_{fj} is the damping coefficient of optical phonons in the film and the substrate; and γ_{pj} and ν_{pj} are the damping coefficient and the plasma resonance frequency, respectively, in the ZnO film.

While calculating the IR reflection spectra from the surface of the ZnO/SiO_2 structure, the self-consistent parameters of zinc oxide in the geometry $E \perp c$ were used [25]. On the basis of the results obtained in works [26–28], the dielectric permittivity of SiO_2 is characterized by a manifestation of four oscillators in the IR spectral interval. According to work [5], in the case of a few elementary oscillators, the dielectric permittivity of the SiO_2 substrate can be written as follows (this is the so-called additive model of dielectric permittivity):

$$\varepsilon(\nu) = \varepsilon_1(\nu) + i\varepsilon_2(\nu) = \\ = \varepsilon_\infty\left(1 + \sum_{j=1}^{N} \frac{S_j}{\nu_j^2 - \nu^2 - i\gamma_j\nu}\right), \qquad (3)$$

where ν_j, γ_j, and S_j are the frequency, damping coefficient, and strength, respectively, of the j-th oscillator. The static dielectric permittivity is given by the formula

$$\varepsilon_0 = \varepsilon_\infty + \sum_{j=1}^{N} S_j.$$

The strength of the j-th oscillator, $\Delta\varepsilon_j$, and its damping coefficient γ_{fj} were determined by applying the Kramers–Kronig method in the framework of the dispersion analysis of the experimental and theoretically calculated IR reflection spectra from the SiO_2 substrate, when its surface is free from the ZnO film.

Parameters of the SiO_2 substrate used for the calculation of the IR reflection spectra $R(\nu)$ of the ZnO/SiO_2 structure

ν_{Tj}, cm^{-1}	$\Delta\varepsilon_j$	γ_{fj}/ν_{Tj}
457	0.95	0.015
810	0.05	0.1
1072	0.6	0.006
1160	0.15	0.04

4. Results and Their Discussion

4.1. Analysis of theoretical IR reflection spectra

4.2.1. Influence of the coefficient of phonon damping in the ZnO film

In Fig. 1, the IR reflection spectra calculated for ZnO films with various values of the damping coefficient for the phonon subsystem in the ZnO/SiO_2 structure are depicted. The calculations were performed in the framework of a multioscillatory mathematical model. The strength $\Delta\varepsilon_j$ of the j-th oscillator and its damping coefficient γ_{fj} were determined, by using the dispersion analysis. The corresponding obtained values are quoted in Table.

The electron concentration in the ZnO films was selected to equal $n_0 = 2.9 \times 10^{18}$ cm^{-3} for curves *1* to *5*. The phonon damping coefficient γ_f in the ZnO films was varies from 10 (curve *1*) to 50 cm^{-1} (curve *5*) with an increment of 10 cm^{-1}. The other parameters were selected to be constant, and their values are given in the figure caption.

The inset in Fig. 1 exhibits a scaled-up section of the spectrum from 300 to 550 cm^{-1}. As one can see, the influence of the damping coefficient in the

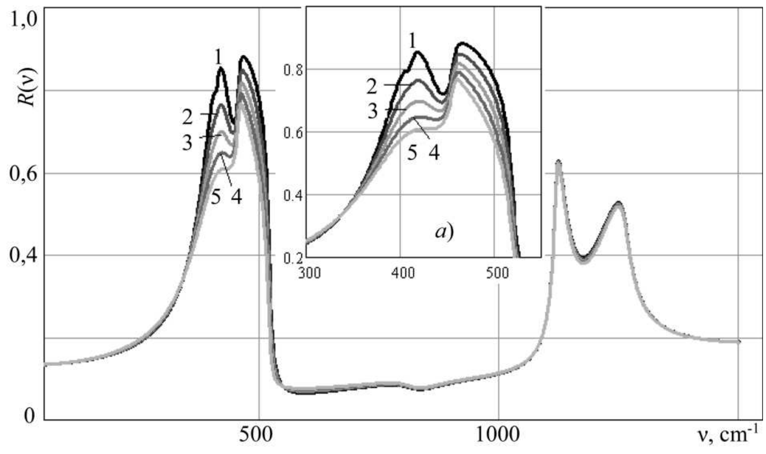

Fig. 1. Calculated spectra $R(\nu)$ of ZnO films on the SiO_2 substrate. The ZnO film parameters: d_f = 632 nm, ν_p = 500 cm^{-1}, γ_p = 2500 cm^{-1}, and γ_f = 10 (*1*), 20 (*2*), 30 (*3*), 40 (*4*), and 50 cm^{-1} (*5*). The scaled-up sections of the spectra $R(\nu)$ in the interval 300–550 cm^{-1} are shown in the inset

ZnO phonon subsystem on the IR reflection spectrum takes place only in this interval. The results obtained are in agreement with the data described in work [12]. Therefore, it can be argued that the interval between the frequencies of the transverse and longitudinal optical phonons in the ZnO film is the most sensitive to the changes in the damping coefficient of the phonon subsystem.

4.2.2. Influence of the ZnO film thickness on IR reflection spectra

The IR reflection spectra $R(\nu)$ calculated for ZnO/SiO_2 structures with various thicknesses of a zinc oxide film are exhibited Fig. 2. The film thickness acquired the values d_f = 50, 200, 400, 600, and 800 nm (curves *1* to *5*, respectively). The other parameters were chosen as follows: ν_p = 500 cm^{-1}, γ_p = 2500 cm^{-1}, and γ_f = 30 cm^{-1}.

It is evident from the figure that the growth of the ZnO layer thickness up to 800 nm, when other parameters of the phonon and plasma subsystems remain constant, results in a substantial distortion of the spectrum $R(\nu)$ in the intervals 200–400 cm^{-1} and 1000–1500 cm^{-1}. The corresponding changes take place owing to the interaction between the phonon and plasmon subsystems in the ZnO film, on the one hand, and the phonon subsystem in the SiO_2 substrate, on the other hand. A spectral maximum $R(\nu)$ = 0.85, which is invariant for all thicknesses of the zinc oxide film, is observed at a frequency of 475 cm^{-1}. The maximum sensitivity of the reflection coefficient $R(\nu)$ with respect to the film thickness variation is observed in the high-frequency section of the spectrum. The reflection coefficient grows by 40% in the spectral interval from 1100 to 1300 cm^{-1}.

4.2.3. Influence of the frequency of the plasmon-phonon resonance in ZnO on IR reflection spectra

Figure 3 illustrates the dependence of the IR reflection spectrum shape on the frequency of the plasmon-phonon resonance in a ZnO film with 632 nm thickness. Curves *1* to *5* are calculated for the parameters $\gamma_{p\perp} = \nu_{p\perp}$ varying from 1 to 1000 cm^{-1}, respectively. This interval corresponds to the concentration change from 10^{16} to 10^{19} cm^{-3}, the charge carrier mobility change from 10 to 200 cm^2/(V s), and the conductivity change from 100 to 410 Ω^{-1}cm^{-1}. From the figure, it is evident that the most significant modifications in the reflection spectra $R(\nu)$ are observed in the sections of 50–300 cm^{-1} and 520–1050 cm^{-1}, where the $R(\nu)$-values increase from 0.05 to 0.25, and in the interval 1200–1500 cm^{-1}, where $R(\nu)$ decreases from 0.7 to 0.03.

A characteristic feature of all spectra exhibited in Figs. 1 to 3 is the presence of peaks in the intervals 400–600 cm^{-2} and 1100–1300 cm^{-1}. Those peaks emerge owing to the influence of the phonon and plas-

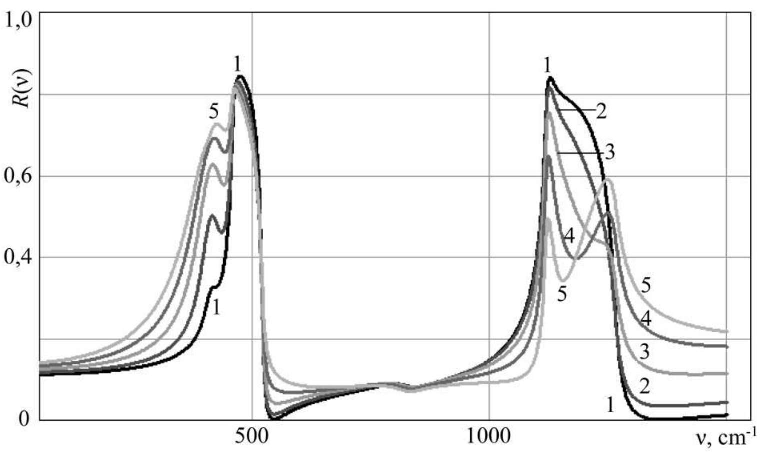

Fig. 2. Calculated spectra $R(\nu)$ of ZnO films on the SiO_2 substrate. The ZnO film parameters: $\nu_p = 500$ cm^{-1}, $\gamma_p = 2500$ cm^{-1}, $\gamma_f = 30$ cm^{-1}, and the film thickness $d_f = 50$, 200, 400, 600, and 800 nm (curves *1–5*, respectively)

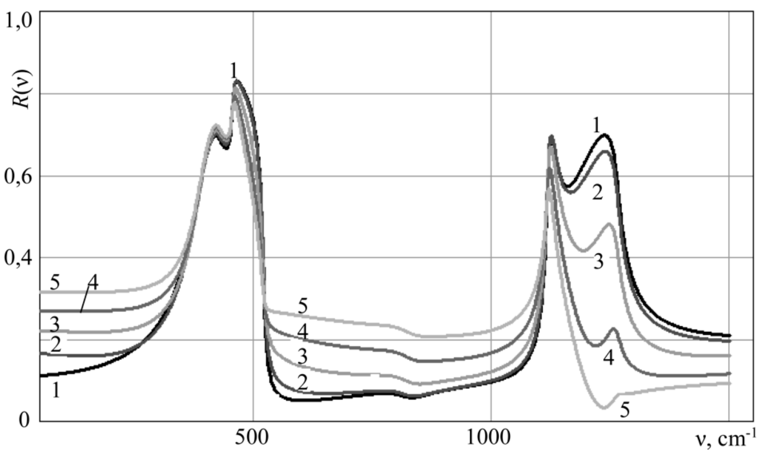

Fig. 3. Calculated spectra $R(\nu)$ of ZnO films on the SiO_2 substrate. The ZnO film parameters: $\gamma_f = 30$ cm^{-1}, the film thickness $d_f = 632$ nm, and $\nu_p = \gamma_p = 1$, 250, 500, 750, and 1000 cm^{-1} (curves *1–5*, respectively)

mon subsystems in the interval of "residual rays" in ZnO and the phonon subsystem in SiO_2. The growth of the charge carrier concentration in zinc oxide films from 10^{16} to 10^{19} cm^{-3} substantially distorts the reflection spectrum in a wide IR spectral range.

It should be noted that the IR reflection spectrum correlates with the frequencies of transverse and longitudinal optical phonons. The range of "residual rays" in the ZnO film occupies the interval 400–600 cm^{-1}, and the range of "residual rays" in the SiO_2 substrate is in the interval 350–1500 cm^{-1}. The frequencies of the inflection points on the low-frequency slope of the reflection bands of the film and the substrate turn out close to the frequency of transverse optical phonons, and those of the inflection points on the high-frequency slope of the spectra to the frequency of longitudinal optical phonons in SiO_2. It was found that, while simulating the dependence $R(\nu)$ in the range of "residual rays" using the Kramers–Kronig relations, the largest changes of the dielectric permittivity $\varepsilon(\nu)$ were observed near the frequency of the plasmon-phonon resonance in the zinc oxide film. If the frequency of the transverse optical phonon of the ZnO film changes, it insignificantly shifts the corre-

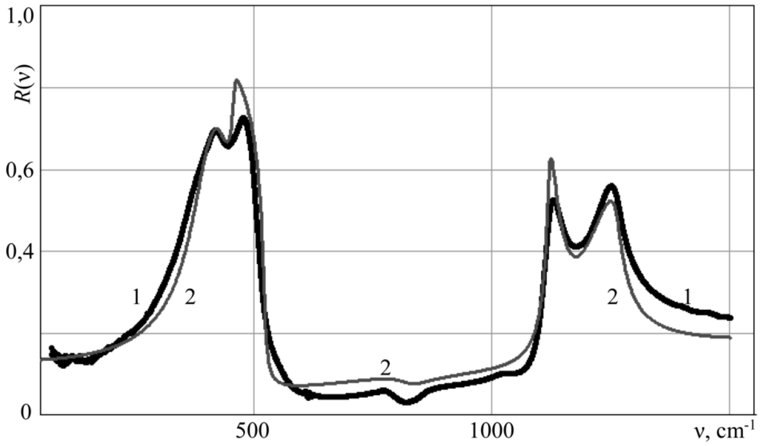

Fig. 4. Experimental (symbols *1*) and calculated (curve *2*) spectra $R(\nu)$ of ZnO films on the SiO_2 substrate. The ZnO film parameters: $d_f = 632$ nm, $\nu_p = 500$ cm^{-1}, $\gamma_p = 2500$ cm^{-1}, and $\gamma_f = 30$ cm^{-1}

sponding side of the reflection band and weakly affects the $R(\nu)$ spectrum in the high-frequency section.

4.2.4. Comparison of theoretical results and experimental data

The experimental spectra also demonstrate the presence of peaks in the intervals from 400 to 600 cm^{-1} and from 1100 to 1300 cm^{-1}. However, the theoretical values obtained for $R(\nu)$ in those intervals turned out overestimated in comparison with the experimental data. Taking into account that the ZnO film was doped with terbium, the change in the shape of the IR reflection spectrum can be explained by doping: not only the concentration of electrons, but also their mobility is relevant.

Figure 4 demonstrates the experimental IR reflection spectrum measured for a Tb-doped ZnO film in the ZnO/SiO_2 structure (curve *1*), as well as a theoretically calculated one (curve *2*). The latter was obtained for a film thickness of 632 nm. The substrate parameters are quoted in Table. The concentration n_0 of free electrons in the ZnO film was varied from 10^{16} to 10^{20} cm^{-3}. The parameters ν_p, γ_p, and γ_f were considered to be unknown for the ZnO film.

The calculated $R(\nu)$-curves were compared with the experimental ones, by using the least-squares method. The mean square deviation of the experimental reflection coefficient values from the calculated ones was determined, by following a procedure described in books [5, 13]. The coincidence of the theoretical reflection spectrum with the experimental one in the spectral interval 50–1500 cm^{-1} was $\delta = 10^{-2}$.

As one can see from Fig. 4, the best agreement between the theory and the experiment takes place at the frequency and the damping coefficient of plasmons and phonons $\nu_p = 500$ cm^{-1} and $\gamma_p = 2500$ cm^{-1}, respectively, and at $\gamma_f = 30$ cm^{-1}. The corresponding error does not exceed 3%. On the basis of the data obtained, the concentration of electrons, $n_0 = 2.9 \times 10^{18}$ cm^{-3}, and their mobility, $\mu = 90$ cm^2/(V s), were determined. This concentration is much lower than the concentration of Tb^{3+}, i.e. the doping does not increase the conductivity. This can be a result of the fact that the incorporation of terbium into the ZnO lattice leads to the formation of a compensating acceptor defect in the lattice [29, 30].

Note that the measurement of the dc conductivity of the film showed that it was about 10^{-7} Ω^{-1}cm^{-1}. Such values are much lower than those obtained from the IR reflection measurements, which testifies to the presence of high-ohmic regions in the film that are connected in series.

Rather a good agreement obtained for $R(\nu)$ in the range of "residual rays" in the film and the substrate (see Fig. 4) confirms the reliability of the mutually consistent values obtained by us for the bulk parameters of zinc oxide in work [25] and the validity of their application, while studying the textured polycrystalline ZnO films. Note that the relocation of doped zinc oxide films on SiO_2 substrates in the *xy*-

plane practically does not change the shape of the $R(\nu)$ spectrum, which testifies to the isotropic character of the optical and electrophysical properties of the examined system. Furthermore, it can be argued that the optical axis in the textured layers of zinc oxide and in SiO_2 is directed perpendicularly to the xy-plane ($c \perp xy$).

5. Conclusions

To summarize, from the results of complex researches carried out in this work, it follows that the IR reflection spectra of the air-ZnO film-SiO_2 substrate structure can be well simulated, if the self-consistent values of bulk parameters obtained in works [5, 25] for single crystals of zinc oxide in the geometry $E \perp c$ and the multioscillatory mathematical model with the parameters of the phonon subsystem taken from work [26] are used. The comparison between the optical constants for ZnO single crystals and the values obtained for ZnO films allows a conclusion to be drawn that the examined films had a good quality, so that the damping coefficients for the phonon and plasmon subsystems were practically identical. This result demonstrates that the method of IR reflection spectroscopy is convenient and informative for the determination of the lattice properties and the electrophysical properties of ZnO films. This non-destructive method was applied to determine the concentration of charge carriers and their mobility in the Tb-ZnO/SiO_2 structures.

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ОПТИЧНІ ТА ЕЛЕКТРИЧНІ
ВЛАСТИВОСТІ Tb–ZnO/SiO$_2$ В ІЧ-ОБЛАСТІ СПЕКТРА

Р е з ю м е

За допомогою методу ІЧ-спектроскопії зовнішнього відбивання досліджено оптичні та електрофізичні властивості плівок оксиду цинку, легованого тербієм. Плівки було нанесено на підкладки оксиду кремнію методом магнетронного напилення. Теоретичне моделювання спектрів для структури ZnO/SiO$_2$ проведено з використанням багатоосциляторної моделі в діапазоні 50–1500 см$^{-1}$ за орієнтації електричного поля перпендикулярно до c-осі ($E \perp c$). Методом дисперсійного аналізу визначено оптичні та електричні властивості плівки ZnO, а також силу осциляторів і значення їх коефіцієнта затухання для плівки та підкладки SiO$_2$. З'ясовано вплив фононної та плазмон-фононної підсистем плівки ZnO на форму спектра ІЧ-відбивання структури Tb–ZnO/SiO$_2$.