Impact of refractive index profile of nanostructured ITO films on light extraction efficiency

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Abstract. In this paper, we study composite nanostructured ITO films with different refractive index profiles, obtained by e-beam evaporation and magnetron sputtering, which can be used as a transparent conducting contacts in a wide variety of optoelectronic devices, e.g. AlInGaN light-emitting diodes. The composite ITO films that comprise of an underlying dense layer and a structured layer deposited on a glass substrate surface have been fabricated. Reflectance and transmittance spectra were measured. Numerical calculations have been utilized to obtain refractive index profiles of the studied films. Light extraction efficiencies of the studied films have been calculated.

1. Introduction

In the past decades transparent conducting oxides (TCO) have become an industrial standard in various fields of application (LCD and LED displays, sensor panels, solar cells, etc.) due to their reasonably high optical transmittance and electrical conductivity. The most common of TCO is indium tin oxide (ITO), which provides an optimal combination of transparency in the visible range of the optical spectrum and electrical conductivity.

One of the applications of ITO films is the use of them as contact pads in AlInGaN LED chips. The high value of the ITO work function (~4.6 eV) and, hence, reasonable values of the contact resistance to p-GaN layers in combination with good conductivity and high transparency in the visible range substantially increase the external quantum efficiency of LEDs with the contact based on ITO films. Nevertheless, an increase in the quantum efficiency can be obtained by means of a choice of the structure and deposition techniques of films due to the change in the refractive index [1,2].

The face-up AlInGaN LED chips, in which ITO films obtained by different deposition techniques are used as the upper transparent contact to p-GaN region, have been already considered in [3,4]. The effect of methods of obtaining thin ITO films on the film properties has been studied and it has been shown that the film deposition on the substrate heated up to a temperature above 400°C makes it possible to form structured films with a mean effective refractive index of about 1.2.

The method to form transparent conducting coatings based on ITO material with the controlled refractive index has been suggested in [5]. The method consists in the sequential deposition of the material by electron-beam evaporation and magnetron sputtering. Different density of the obtained films and, hence, their different refractive indices can be obtained by varying the ratio of mass fractions of the material deposited by different methods. Nevertheless, the measurements of transmission and reflection spectra of these films in combination with the numerical simulation show
that these films are characterized by the refractive index profile that varies along the axis perpendicular to the substrate plane.

In this work, we suggest using composite films formed by the dense ITO sublayer deposited on a substrate surface at room temperature in combination with the layers sequentially deposited by e-beam evaporation and magnetron sputtering. The dense sublayer both enhances current spreading on the contact surface and reduces the contrast of the refractive index at GaN interface; the upper composite layer is chosen taking into account the refractive index of the medium, into which the light is extracted. The combination of thicknesses of the sublayer and upper composite layer as well as the ratio of mass fractions of the material in the composite layer determine the interference conditions.

2. Experimental Results

In order to study the optical properties of the composite ITO films, several samples were prepared with the varying combinations and thicknesses of composing layers. To prepare the studied composite films, the combination of two deposition techniques, magnetron sputtering and e-beam evaporation, with annealing in nitrogen atmosphere were used. The experimental samples were multilayer coatings on glass substrates fabricated as follows. The first dense layer was deposited by magnetron sputtering on a cold (at room temperature) substrate, after deposition the layer was heated in vacuum up to a temperature of 450°C and then pure nitrogen was inlet to the chamber up to a pressure of ~800 mbar for 10 min. The second layer was formed by e-beam evaporation on the sample preliminarily heated up to 450°C with subsequent storage in nitrogen atmosphere at 800 mbar for 10 min. The last layer was deposited by magnetron sputtering at room temperature and then formed as in the first case. The thickness of separate layers was controlled with a quartz crystal sensor and varied from 50 to 170 nm.

Figure 1 shows cross-sectional scanning electron micrographs (SEM) of the obtained composite films marked, for convenience, as samples 1–4. Figures 1a and 1b show SEM images of the cross-sections of two double-layered composite films that comprise of one underlying dense layer deposited by magnetron sputtering on a cold glass substrate surface, and one structured layer formed by e-beam evaporation on the preliminarily heated sample. The clear interface between two layers can be seen in both figures. So, the thicknesses of dense layers can be determined from SEM cross-sections. However, thicknesses of the low-n structured layers are hard to be defined. As can be seen in figures 1a and 1b the upper surfaces of the structured layers are rough so instead of thickness of the structured layer below we will use mass equivalent of the dense film of deposited material, which corresponds to measured values. So, the phrase “equivalent thickness of a structured layer is 100 nm” means that the layer mass equals to the mass of a dense layer with thickness of 100 nm. Samples 1 and 2 have identical high-n 100 nm dense layer while the structured layers differ by the thicknesses of low-n structured layers. Equivalent thicknesses of structured layers in Sample 1 and Sample 2 are 50 nm and 120 nm, respectively. Figures 1c and 1d show cross-sectional SEM of two samples based on Sample 1. Sample 3 and Sample 4 are obtained by additional magnetron sputtering at room temperature of ITO films with an equivalent thickness of 70 nm and 170 nm above the low-n structured layer. A significant increase of density of the structured films can be seen in figures 1c and 1d. As a result, a set of samples with different density profiles was obtained. These non-uniform density profiles lead to non-uniform profiles of effective refractive index. Refractive index profiles determine optical properties of the films.

Sample 1 is considered as a reference. Sample 2 and Sample 3 can be obtained from the basic Sample 1 by deposition of additional structured and dense ITO layers with an equivalent thickness of 70 nm, respectively. Sample 4 can be obtained from Sample 3 by deposition of an additional dense ITO layer with equivalent thickness of 100 nm.

In this paper, we will consider the effect of use of the described four composite ITO films on the light-extraction from an AlInGaN LED chip.
Figure 1. Cross-sectional SEM images of the composite films
(a) Sample 1 – 100 nm high-n dense ITO / 50 nm equivalent low-n structured ITO
(b) Sample 2 – 100 nm high-n dense ITO / 120 nm equivalent low-n structured ITO
(c) Sample 3 – 100 nm high-n dense ITO / 50 nm equivalent low-n structured ITO / 70 nm equivalent high-n dense ITO
(d) Sample 4 – 100 nm high-n dense ITO / 50 nm equivalent low-n structured ITO / 170 nm equivalent high-n dense ITO

2.1. Optical properties of the studied samples

Optical transmittance and reflectance spectra were measured for the mentioned samples using Agilent Cary 430 spectroradiometer with normal incidence geometry. Then, by using numerical calculations we obtained the approximate refractive index profiles for each sample. To obtain profiles of effective refractive index profiles of composite films we used numerical calculations based on the transfer matrix approach for multilayered films [6]. The refractive index spectra for dense ITO films were obtained from [7] and modified according to the reflectance transmittance spectra of a single dense-layer ITO film on a glass substrate. The structured layers were approximated by series of porous sublayers with different constant fill factors (volume ratios). The effective refractive indices of each sublayer were determined by the Odelevsky rule for perpendicularly oriented pores in a dense material [8]. Theoretical fill factor and refractive index profiles were then chosen to fit the measured transmittance and reflectance spectra. The resulting profiles should describe films with given masses and thicknesses measured for each sample. Figure 2 shows the measured transmittance and reflectance...
spectra (solid line) and fitted spectra (dashed line). The fill factor profiles used for the calculations are also shown in the insets.

Figure 2. Measured (solid line) and fitted (dashed line) transmittance – reflectance spectra of four samples (a) sample 1 (b) sample 2 (c) sample 3 (d) sample 4.

2.2. Numerical calculation of the light extraction from AlInGaN LEDs

The obtained fill factor and refractive index profiles were used to calculate the relative extraction efficiency of transparent ITO composite p-contacts for face-up AlInGaN LED chips. The light extraction in a face-up AlInGaN LED chip is schematically shown in figure 3. Light is generated in the AlInGaN active region and then has to pass the transparent conductive p-contact – composite ITO film. It should be noted that most of the emitted light is reflected by the contact due to the total internal reflection (TIR). The TIR angle for the plane interfaces depends only on the refractive indices of GaN and ambient material and is relatively small (~ 24° for air and ~ 37° for commonly used silicones). Figure 4 shows the angle dependences of transmittance for four composite films similar to the films of samples 1 – 4 at 450 nm. As is seen, the first film significantly increases transmittance for the whole range of incident angles less than the TIR angle in comparison with other films. Though the exiting cone of light is relatively small in both situations, the resulting extraction is usually enhanced by utilization of the scattering substrates (e.g. profiled sapphire substrates).
To range the light extraction values of the studied composite films, we integrated the total transmittance of light with uniform angular distribution, which passes from GaN layer to the ambient air and optical silicone, within the TIR cone. Figures 5 and 6 show the spectral dependences of light extraction to the ambient air and optical silicone for four composite films similar to the films of samples 1 – 4. These dependences have different maxima and minima in the visible region of spectra depending on the film structure and ambient refractive index. One can see that the suggested composite films are spectrally sensitive, so the fine tuning of structured ITO film should be done in order to produce effective transparent conductive p-contacts for AlInGaN LED chips. As figures show, a reduction in the refractive index contrast leads to a decrease in the total reflection for the whole range of incident angles and an increase in the TIR angle, which results in a total increase in the light extraction.

It should also be noted that in this work we used dense ITO layers deposited by magnetron sputtering on a substrate at room temperature. However, when real ITO based contacts are fabricated, this method is not suitable because of the catastrophic degradation of thin p-GaN layer due to the action of ions of the magnetron plasma discharge. To eliminate this problem, the dense protective sublayer
deposited by e-beam evaporation on a substrate at room temperature should be used to protect p-GaN surface from the plasma effect.

3. Conclusion
To conclude, in this work composite multilayer ITO films with various structures deposited on glass substrates were studied. The measured normal incidence transmittance and reflectance spectra of the films were used to obtain the effective refractive index profiles by means of numerical calculations. The obtained profiles were used to calculate the angular dependences of transmittance of similar films deposited on GaN substrates. The calculated dependences were integrated to determine the total light extraction efficiency of the films for two ambient media – air and optical silicone. It was demonstrated that different film structures may be used to enhance light extraction for different wavelengths, so the fine tuning of structured ITO films is necessary in order to produce effective transparent conductive p-contacts for AlInGaN LEDs.

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