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Characteristics of aluminum oxide deposition on an array of ITO nanowhiskers

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In this work, we studied the process of deposition of aluminum oxide onto films formed by ITO nanowhiskers, as well as the effect of the protective layer on the optical characteristics of the coating. At the initial stages, a thin Al_2O_3 layer is formed on the surface of individual nanowhiskers, maintaining the gradient nature of the refractive index of the composite coating. As the Al_2O_3 layer thickness increases, due to the variation of ITO nanowhiskers in height, a relief is formed by dome-shaped convexities on the surface of the resulting film with a characteristic scale of up to 1 mkm, which makes it possible to create a structure that effectively scatters light.

Keywords: nanostructured films, ITO, magnetron sputtering, aluminum oxide, light scattering.

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Coatings based on transparent conducting indium-tin oxide (ITO) have become widely popular in optical and electronic instruments. These coatings are deposited by various vacuum methods, such as magnetron sputtering [1], electron-beam evaporation [2] or chemical vapor-phase deposition [3].

A change in the sputtering process parameters may result in a modification of the produced coatings. For example, if the magnetron sputtering process is carried out in oxygen-free plasma, and the glass substrate is heated to the temperature above the temperature of indium and tin melting, ITO film growth is carried out using vapor-liquid-crystal mechanism, which causes formation of filamentary ITO nanocrystals on the substrate surface, and their diameter directly depends on the size of the molten metal drops [4,5]. The density of the coating formed from filamentary ITO nanocrystals monotonously increases in the film from the minimum values on its outer interface to the density of the solid ITO film near the interface with the substrate. Appearance of the coating density gradient, in its turn, causes appearance of the gradient of the effective refractive index in the direction perpendicular to the substrate surface, which provides for the suppression of Fresnel reflection at the interface of the media and, accordingly, the translucence of optical elements with coating from ITO [6]. One of the disadvantages of such modification of the indium-tin oxide coating surface is the possible degradation of optical characteristics of the material as a result of interaction with the environment due to the developed surface of filamentary nanocrystals. The solution to this problem may be application of an optically transparent coating Al_2O_3 on top of nanocrystals. But due to deposition of the coating on the very developed surface (according to calculations in [7] the specific surface increases by more than 20 times in process of structuring) in the process of protective coating growth the final thickness of the formed layer Al_2O_3 may change, if the chemical methods of film deposition are used. The thicknesses of coatings Al_2O_3 specified in the paper are obtained from the estimation of the thickness of films deposited on the reference samples with the even ITO film. This paper studied the process of aluminum oxide deposition on the films formed by filamentary ITO nanocrystals and the effect of the protective layer on the optical characteristics of the coating.

The coatings were obtained, which consist of the array of filamentary ITO nanocrystals with the applied protective coating with thickness from 10 to 300 nm on top of them. ITO films were applied on magnetron sputtering installation Izovac Astra-S. The films were sputtered on the heated substrate with the oxygen absence in the working chamber. Protective coatings Al_2O_3 were grown by the method of molecular layering on installation Picosun P-300B. Using Radiant installation IS-LI, directional patterns were studied for the direction of diffuse transmission of a directed laser beam incident normally on the surface (radiation wavelength 488 nm). To obtain SEM images of the films, scanning electron microscope JEOL JSM-7001F was used. The transmission and reflection spectra of the samples were examined using an Optronic Laboratories OL 770 spectroradiometer.

Figure 1 shows SEM images of chip of sample with formed layers of nanostructured ITO and Al_2O_3 layers of

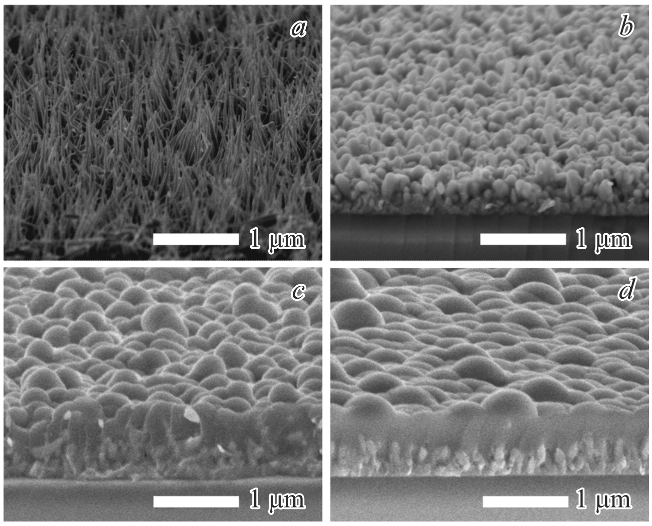

Figure 1. SEM-image of the film formed by an array of filamentary ITO nanocrystals without the aluminum oxide layer (*a*) and with the deposited layer of Al_2O_3 with the following thickness: *b* — 10 nm, *c* — 150 nm, *d* — 300 nm.

various thicknesses, the number of deposition cycles of which corresponds to 10, 150, 300 nm film deposited on smooth substrate.

At initial stages the film Al_2O_3 covers individual filamentary nanocrystals, not forming solid coating. Isotropism of the process of aluminum oxide layer growth on the structured ITO film causes considerable modification of the coating surface with occurrence of micron size relief on the final stage. Due to spread of ITO nanocrystals by height during deposition Al_2O_3 with layer thickness of 300 nm the relief is formed from dome-shaped convexities with specific scale of around 0.5 mkm and more.

Figure 2 shows transmission spectra of the produced structures. Spectra of specimens with thicknesses of aluminum oxide 10 and 150 nm show no interference maxima and minima. This is matched with data of SEM-images of specimens: thinly applied layer Al_2O_3 coats certain ITO nanocrystals, maintaining the gradient of film density in direction from the medium to the substrate and the related gradient of the material refractive index, there is no clear interface of the medium and the film at the same time. At thickness Al_2O_3 225 nm the interference maxima and minima appear, as a result of the change in the profile of the effective refractive index of the coating. With the further coverage of the surface up to 300 nm Al_2O_3 the specific size of individual irregularities at the ends of the longest ITO nanocrystals starts exceeding the radiation wavelength, which results in increased scattering of the light incident on the surface, in this case the Fresnel reflection is suppressed.

Figure 3 presents the results of measurement of angular light scattering diagrams with specimens normalized to maximum value. As the thickness of the deposited layer of aluminum oxide increases, light scattering increases. Light scattering effect is not found on a specimen with thickness of Al_2O_3 10 nm. The scale of heterogeneity on its surface (around 100 nm) is less than the wavelength of laser radiation (488 nm), therefore, the effective medium approximation works, and the surface roughness is treated by the wave as the averaged medium. At thickness Al_2O_3 150 nm a relief is formed on the surface with the scale of around 250–300 nm, which also enables using effective medium approximation. Due to the increase of relief scale and appearance of certain convexities of large size, the scattering increases compared to the specimen with 10 nm Al_2O_3. It is interesting how scattering decreases in a specimen with thickness of Al_2O_3 225 nm compared to thickness of 150 nm and increases in a specimen with thickness of Al_2O_3 300 nm. Apparently, decrease in

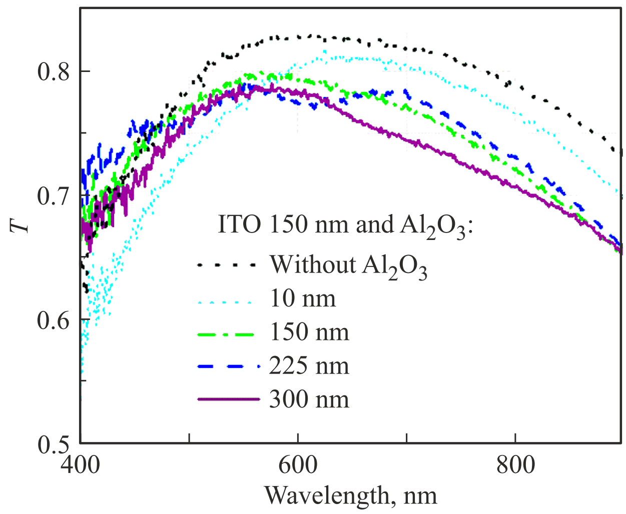

Figure 2. Transmission spectra of structured ITO with the deposited layer of aluminum oxide of various thickness: 10, 150, 225, 300 nm.

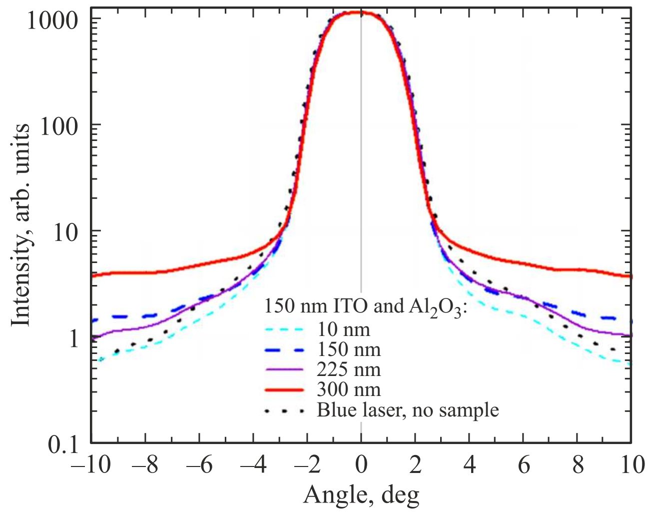

Figure 3. Scattering of structured ITO with the deposited layer of aluminum oxide of various thickness: 10, 150, 225, 300 nm.

scattering is related to flattening of the relief under surface coverage, and subsequent increase in scattering is related to formation of irregularities on the surface with the size of more than 0.5 mkm, which is more than the radiation wavelength, therefore, radiation will no longer perceive the surface as an averaged medium. These data correlate well with the transmission spectra of specimens, where the specimen with layer thickness Al_2O_3 225 nm has the marked interference maxima and minima.

The paper shows that at the initial stages the film Al_2O_3 coats the surface of separate filamentary ITO nanocrystals with a thin layer, preserving the gradient nature of the coating. As the thickness Al_2O_3 increases, the relief is formed by dome-shaped convexities on the surface of the produced film with the specific scale to 1 mkm, which makes it possible to create the structure efficiently scattering light, which may be used to increase the efficiency of light output in various optoelectronic instruments.

Conflict of interest

The authors declare that they have no conflict of interest.

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