Formation of a light-scattering microrelief during atomic-layer deposition of a dielectric layer on a nanostructured film of indium-tin oxide

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This paper presents a method for creating a light-scattering relief at the stage of formation of the lower dielectric layer of electroluminescent displays. To study the process of formation of a light-scattering relief, films with different thicknesses of the Al_2O_3 layer were fabricated, SEM images of cross-sections of the formed films were taken, and the luminous intensity angular distribution of scattered light were measured. It is demonstrated that the relief obtained at the dielectric boundary creates a light-scattering structure that improves the light extraction efficiency from the active layer of an electroluminescent display.

Keywords: nanostructured ITO, light scattering relief, light extraction efficiency.

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1. Introduction

In the manufacture of film light sources (LED, display structures), one of the frequently occurred problems is the low efficiency of light output due to the effect of total internal reflection at the interface between the light-emitting layers and adjacent transparent materials. Light emitted at angles greater than the angle of total internal reflection is absorbed in the active layer, reducing the efficiency of the device. To increase the proportion of light output, a light-scattering relief is created at the boundaries of the active layer, increasing the proportion of light output outside. Such a relief can be pre-formed on the substrate [1–3], created during the process of growing active layers, or obtained at the post-growth stage by etching or deposition of structured layers [4]. This paper solves the problem of light extraction from the phosphor layer of electroluminescent displays, located between two dielectric layers. Most of the phosphor layer materials used have a relatively high refraction index ($n(\mathrm{ZnS}) = 2.3-2.4$, $n(\mathrm{CdS}) = 2.5$), while the refraction index of the dielectric layers has lower values ($n(\mathrm{Al_2O_3}) = 1.76$, $n(\mathrm{SiO_2}) = 1.45$). The use of dielectric layers with higher refraction index leads to the problem of light output to surrounding layers with lower index. Thus, part of the light is locked in the active layer and is not removed from it in the plane-parallel geometry of the layers.

The paper presents the results of creating a light-scattering microrelief during atomic layer deposition of the dielectric (Al_2O_3) onto films of nanostructured indium-tin oxide.

2. Experimental procedure

The nanostructured layer of indium tin oxide (ITO), which consists of filamentary nanocrystals located predominantly perpendicular to the substrate, was used as the lower sublayer. The ITO layer was deposited by electron beam evaporation from ITO granules ($90\,\mathrm{wt.\%}\ In_2O_3 + 10\,\mathrm{wt.\%}\ SnO_2$) onto a BK-7 glass substrate preheated to 550°C. The process was carried out in a vacuum chamber evacuated to $5 \cdot 10^{-7}$ mbar, the material deposition rate was monitored by a quartz sensor and was 10 nm/min. The distance between the material source and the substrate holder with the sample was 250 mm. Next, the sample was annealed in a sputtering chamber at the same temperature in a high purity nitrogen atmosphere at a pressure of 800 mbar. Next, Al_2O_3 layer was deposited using the atomic layer deposition method in Picosun P-300B unit. Application by atomic layer deposition method occurs cyclically with alternate injection of trimethylaluminum (TMA) and water. The reagent injection time was 0.1 s, the pumping out time was — 3 s for TMA and 5 s for water. In this case, the formation of Al_2O_3 film occurs layer by layer over the entire surface of the nanocrystals.

To obtain SEM images of the films, JEOL JSM-7001F scanning electron microscope manufactured by JEOL Ltd., Japan was used. The directional patterns of scattered radiation were studied using a measuring system for monitoring the spatial distribution of luminous intensity IS-LI ™Luminous Intensity Measurement System (Radiant Imaging).

3. Experimental results and discussion

When forming Al_2O_3 films by atomic layer deposition on smooth substrate, the film thickness is determined by the number of reagent injection-pumping out cycles. When films are deposited onto the developed surface of a nanostructured ITO film at the initial stage, due to the high

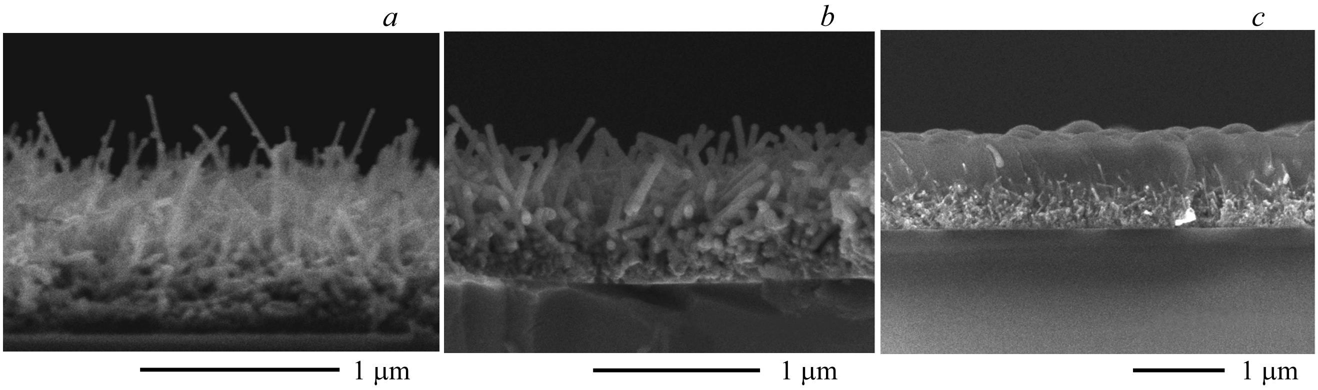

Figure 1. SEM image of film Al_2O_3 with thickness *a* — 10 nm, *b* — 20 nm and *c* — 300 nm deposited on top of a nanostructured layer ITO.

specific surface area of filamentary nanocrystals, the deposition rate of the material increases and the final thickness of the formed layer Al_2O_3 can differ significantly from the thickness of smooth film with the same number of injection-pumping out cycles. Since the height of nanocrystals has a scattering, during eposition of Al_2O_3 at late stages a relief is formed, consiting of dome-shaped convexities on the surface of the resulting film with a characteristic scale of about 0.5 mkm, which is slightly longer than the wavelength of visible radiation in active layers of electroluminescent displays. Figure 1 shows SEM images of chip of sample with formed layers of nanostructured ITO and Al_2O_3 layers of various thicknesses, the number of deposition cycles of which corresponds to 10, 20 and 300 nm film deposited on smooth substrate. At initial stages (Figure 1, *a, b*) the film Al_2O_3 covers individual filamentary nanocrystals, not forming solid coating. At the final stage (Figure 1, *c*) a continuous film with a characteristic relief on the surface is formed. On the above chip of the formed coating, three regions can be distinguished — ITO sublayer, a layer of filamentary ITO nanocrystals filled with Al_2O_3, as well as continuous layer Al_2O_3.

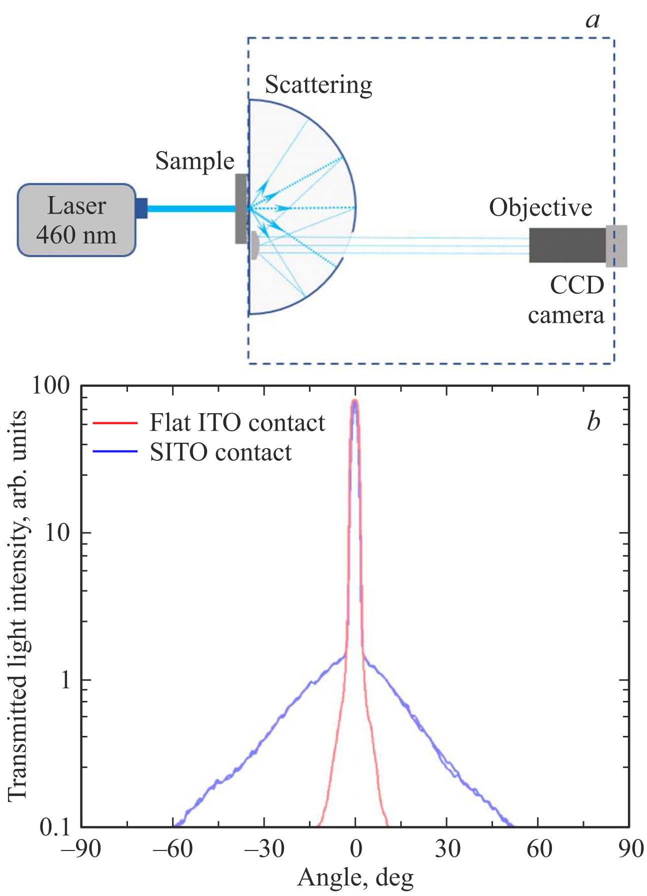

Figure 2. *a* — measurement scheme; *b* — directional patterns of light scattering on samples.

To compare the scattering ability of the manufactured samples, the luminous intensity directional patterns of optical laser radiation incident perpendicular to the surface of the samples were measured. Figure 2 shows a diagram for measuring directional patterns for normal light incidence on the sample from the substrate side, as well as measurement results for two samples — Al_2O_3 layer 300 nm thick deposited on smooth ITO layer, as well as Al_2O_3 layer formed in the same deposition process on nanostructured ITO sublayer. The main part of the radiation at normal incidence propagates without scattering for both samples. At the same time, the intensity of the scattered light integrated over the hemisphere differs by 5 times for two samples. In the case of light incidence at angles greater than the angle of total internal reflection when light is transmitted into optically less dense medium, part of the radiation, that was not scattered, is reflected from the surface, remaining in the active region. In plane-parallel display structures, the significant portion of the light generated in the active layer, due to the effect of total internal reflection, experiences multiple reflections from the interfaces with the surrounding layers and is absorbed. The presence of the light-scattering relief leads to radiation energy redistribution and increase in the efficiency of light output from the active layer.

4. Conclusion

In the presented paper, a method is proposed for the formation of dielectric layers Al_2O_3, deposited by atomic layer deposition, having microrelief with characteristic scale of 0.5 mkm. As a sublayer for such a film a conductive nanostructured layer of ITO is used, using electron beam evaporation from ITO granules onto preheated substrate. The relief formation on the surface of the dielectric is associated with the uniform growth of the film at the ends of the most elongated filamentary nanocrystals. The resulting relief makes it possible to create effective light-scattering structure that improves the efficiency of light output from the active layer of the electroluminescent display. The proposed method for creating light-scattering relief surface of the dielectric differs from the existing technological process only in the mode of ITO layer deposition and does not introduce additional technological operations into the process of manufacturing light-emitting structures, which makes it possible to use it in the production of electroluminescent displays and high-brightness indicators.

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Conflict of interest

The authors declare that they have no conflict of interest.

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