DOI: 10.1002/adma.200701015

Light-Extraction Enhancement of GaInN Light-Emitting Diodes by Graded-Refractive-Index Indium Tin Oxide Anti-Reflection Contact**

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In photonics and optics, the refractive index of a material, first introduced by Isaac Newton as the *optical density*, is the most fundamental material constant. Since the refractive index determines refraction and reflection occurring at the boundary between two media, it is a critical parameter for the design of optical components, such as distributed Bragg reflectors (DBRs),[1,2] omnidirectional reflectors,[3–6] antireflection (AR) coatings,[7–10] and optical resonators.[11] In many cases, however, the unavailability of materials with desired refractive indices, particularly materials with very low refractive indices, prevents the implementation of optical components with very high performance. In addition, the choice of a material with desired refractive index often forces a compromise in other materials properties such as optical transmittance and electrical conductivity that are also important for most optoelectronic applications. Here, we show that oblique-angle deposition can be used to *tailor* the refractive index of a thin-film material that is chosen for its desired material properties other than refractive index. The unique ability to control the refractive index of thin film materials allows one to eliminate Fresnel reflection, one of the fundamental limitations in light-extraction efficiency of light-emitting diodes (LEDs), by fabricating coatings whose refractive index gradually decreases from the refractive index of the active semiconductor layer to the refractive index of the surrounding medium. As an example of this concept, we present a six-layer graded-refractive-index (GRIN) AR coating made entirely of a single material, indium tin oxide (ITO), chosen for its high conductivity, high optical transmittance, and low contact resistance with GaN. Each layer has a refractive index that is individually tuned to form a stack with refractive index graded from its dense ITO value down to the value close to that of air for an optimum AR performance. It is shown that GaInN LEDs with a GRIN ITO AR contact achieve a light-extraction efficiency enhancement of 24.3 % compared to the LEDs with dense ITO coating due to a strongly reduced Fresnel reflection at the ITO–air interface.

Oblique-angle deposition is a method of growing porous thin films, and hence thin films with low-refractive index (low-*n*), enabled by surface diffusion and self-shadowing effects during the deposition process.[12–16] In oblique-angle deposition, a random growth fluctuation on the substrate produces a shadow region that the incident vapor flux cannot reach, and a non-shadow region where incident flux deposits preferentially, thereby creating an oriented rodlike structure with high porosity. Figure 1 shows the cross-sectional scanning-electron microscopy (SEM) image of low-*n* ITO, which is electrically conductive and optically transparent in visible wavelengths,

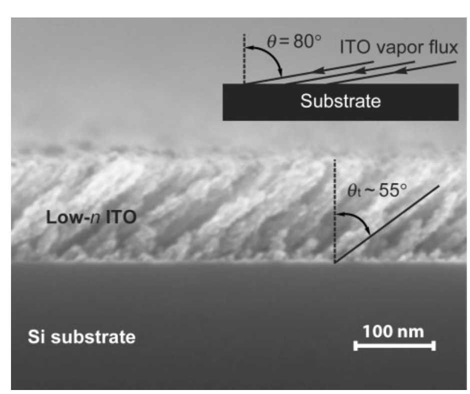

Figure 1. Scanning-electron micrograph (SEM) of low-refractive-index (low-*n*) ITO nanorod thin film on Si substrate, deposited using oblique-angle deposition with a deposition angle of 80°. The refractive index of the low-*n* ITO film is measured to be 1.29 using ellipsometry at λ=474 nm.

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[**] The authors gratefully acknowledge support from Samsung Electro-Mechanics Company, Sandia National Laboratories, Crystal IS Corporation, Troy Research Corporation, New York State Office of Science, Technology and Academic Research (NYSTAR), National Science Foundation (NSF), and the Department of Energy (DOE). Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

ADVANCED MATERIALS

grown by oblique-angle deposition. The deposition angle, defined as the angle between the normal to the sample surface and incident vapor flux, of $\theta = 80°$ resulted in a tilt angle, the angle between the normal to the sample surface and ITO nanorods, of $\theta_t \cong 55°$, which is consistent with the relationship, $\theta_t = \theta - \arcsin[(1-\cos\theta)/2]$, based on geometrical analysis.[17] The gaps between the ITO nanorods are much smaller than the wavelength of visible light, which implies that Mie and Rayleigh scattering can be neglected and the layer can be treated as a single homogeneous film with a uniform refractive index. Since the film was deposited by evaporation, the film thickness could be precisely controlled, which is a very important feature for optical components in which the thickness is less than the wavelength of light. Although the low-*n* ITO film shows a much lower refractive index (1.29) than bulk ITO (2.19), it still keeps its desirable properties, high electrical conductivity and optical transmittance, which was the reason for choosing ITO.

In oblique-angle deposition, the deposition angle determines the area of shadow region at the initial stage of the deposition. Figure 2 shows the top-view SEMs of an initial stage of ITO nanorods for various deposition angles. It is clearly shown that as the deposition angle increases, less area is covered by ITO because of the increase in the area of the shadow region where the incident vapor flux cannot reach. The subsequent incident vapor flux would deposit preferentially in the region already covered by ITO. Therefore, porosity, and hence the refractive index of the thin film made of any evaporable material can be precisely tuned from their bulk value to a value close to the refractive index of air by adjusting the deposition angle.

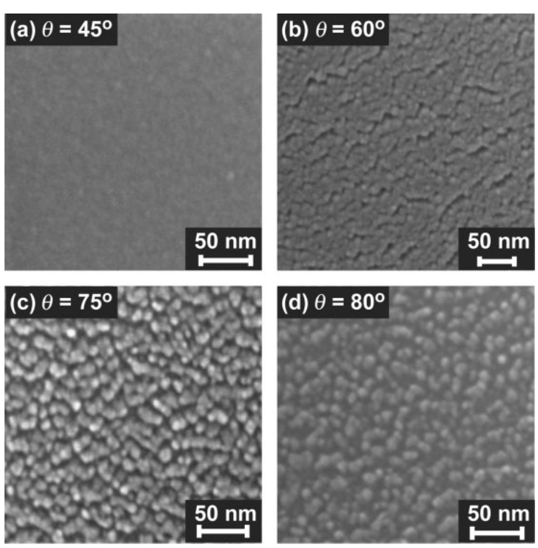

Figure 2. Top-view SEM images of low-*n* ITO thin film on Si substrate deposited using oblique-angle deposition technique with deposition angle of a) 45°, b) 60°, c) 75°, and d) 80°.

Low-*n* ITO films were deposited by oblique-angle deposition with various deposition angles. The refractive index of low-*n* ITO films measured by ellipsometry and fitted using the Cauchy model is shown in Figure 3. The Cauchy model expresses the refractive index as a function of wavelength as $n(\lambda) = A_n + B_n/\lambda^2 + C_n/\lambda^4$, where A_n, B_n, and C_n are constants.

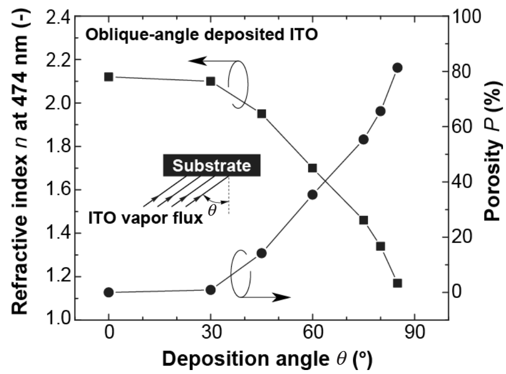

Figure 3. Measured refractive index and calculated porosity of ITO thin films as a function of the deposition angle during oblique-angle electron-beam evaporation. The Bruggemann effective medium approximation is used to calculate the porosity of the film.

The refractive index decreases with increasing deposition angle θ because of the increased porosity of the films, as expected from Figure 2. Note that the refractive index of the nanorod ITO layer with deposition angle of $\theta = 85°$ is $n = 1.17$, significantly lower than that of any conventional thin-film material. The refractive index of a porous material is determined by the porosity of the film and the refractive index of the dense material. The Bruggemann effective medium approximation gives effective refractive index of a low-*n* ITO film consisting of two components, air and dense ITO, with volume fractions V_{Air} and V_{ITO}, where $V_{Air}+V_{ITO}=1$, and refractive indices $n_{Air}=1$ and $n_{ITO}=2.19$.[18] Figure 3 also shows the porosity of low-*n* ITO films, which continuously increases with increasing deposition angle.

Single-layer AR coatings with quarter-wavelength thickness have been widely used, however, such conventional AR coatings only work at a single wavelength and at normal incidence. In contrast, it was reported that GRIN coatings yield broadband omnidirectional AR characteristics with transmittance $T = 100\ \%$, by complete elimination of Fresnel reflection, if the refractive index continuously varies from the substrate's index to the ambient's index.[19] The unique ability of oblique-angle deposition to tune the refractive index of virtually *any* thin-film material, and to attain refractive index values that are close to the refractive index of air, allows one to realize GRIN AR coatings.[10]

Figure 4a shows calculated reflectivity at the wavelength of 460 nm for GaN/air, GaN/dense ITO/air, and GaN/GRIN ITO/air, respectively. For the GRIN ITO, we assume that the refractive index of ITO varies from 2.19 to 1.17 based on our

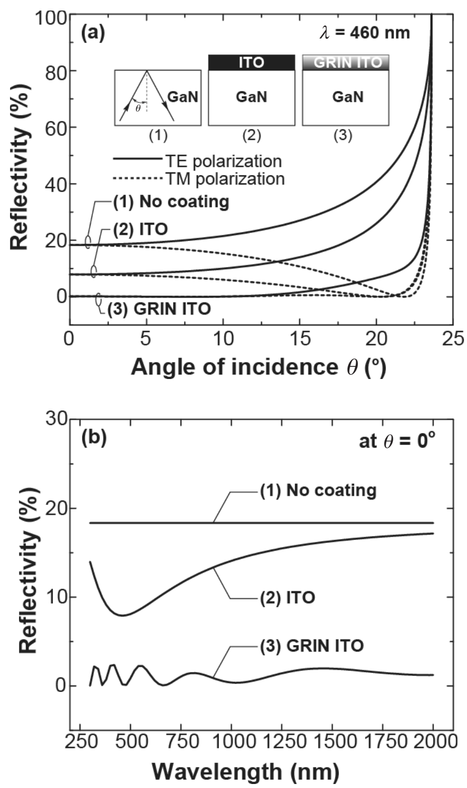

Figure 4. Comparison of calculated reflectivity versus a) angle of incidence and b) wavelength for a GaN surface 1) without coating, 2) with a conventional ITO AR coating and 3) GRIN ITO AR coating. The refractive index of GRIN ITO is varied from 2.19 to 1.17 based on our experimental results, and follows the modified quintic profile.

experimental results shown in Figure 3, and follows the modified-quintic-index profile.[7] The thickness of dense ITO AR coating is assumed to be quarter wavelength. GRIN ITO on GaN exhibited much lower reflectivity for both TE and TM polarizations than dense ITO on GaN and GaN with no AR coating over a large range of angles. The calculated reflectivity at normal incidence as a function of wavelength is also shown in Figure 4b. GRIN ITO on GaN exhibited much lower reflectivity than dense ITO on GaN and GaN with no AR coating. This is attributed to the fact that Fresnel reflection effectively vanishes for the GRIN AR with modified-quintic-index profile. The omnidirectionality advantage of the GRIN coating is particularly important for LED applications because of the isotropic emission from the active region. That is, light within the escape cone virtually experiences no Fresnel reflection.

In order to demonstrate the viability of this concept, blue GaInN LEDs (λ=474 nm) with a GRIN ITO AR coating grown by oblique-angle electron-beam deposition were fabricated. The GRIN AR coating, which also acts as a contact to p-type GaN, consists of six ITO layers in which the refractive index of each layer is tuned to a desired value to implement the modified-quintic-index profile. Figure 5 shows a cross-sectional SEM of the GRIN ITO AR contact, in which the stack

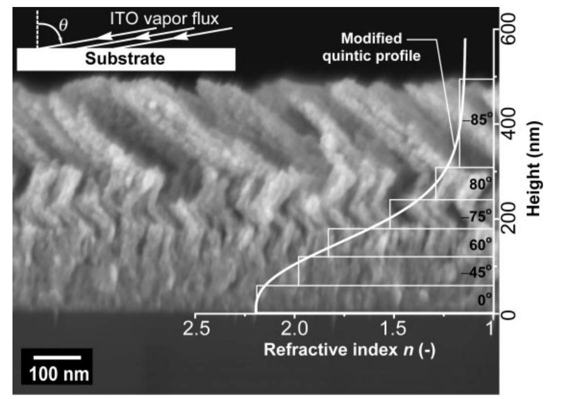

Figure 5. Cross-sectional SEM image of GRIN ITO AR coating with modified-quintic refractive-index profile. The GRIN ITO AR coating consists of 6 ITO layers with pre-determined refractive indices for optimum AR characteristics.

of ITO nanorod layers is clearly visible. All layers have well-defined interfaces. In order to prevent successive layers from filling into the space between nanorods of the previous deposition, negative deposition angles were used for every other layer. The bottom layer of the thin film structure has a refractive index 2.19, which closely matches the index of GaN. The top layer has a refractive index 1.17, close to the index of air. Therefore, the thin-film structure matches the refractive index of air and substrate and is expected to have the excellent AR characteristics that are shown in Figure 4.

Figure 6 shows the average light-output power of 30 representative top-emitting GaInN LED chips with ITO AR contact, and with GRIN ITO AR contact, as a function of forward current. The unencapsulated bare-chip output power of the reference LED structure in an integrating sphere was 4 mW at 20 mA injection current. In addition, a large-size

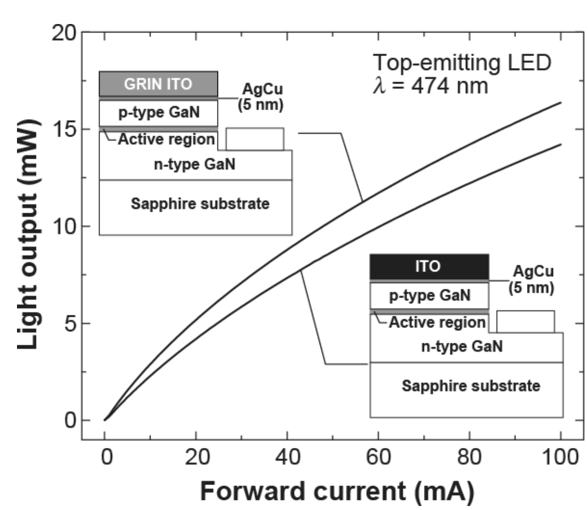

Figure 6. Average light-output power of 30 representative top-emitting GaInN LED chips with dense ITO coating and with GRIN ITO AR contact.

ADVANCED MATERIALS

blue-enhanced Si PIN photodetector (10 mm × 10 mm) is used for measuring the light-output power from the top of the LED chips. At an injection current of 20 mA, LEDs with GRIN ITO AR coating show 24.3% higher light output, 24.3 %, than LEDs with dense ITO AR coating at the injection current of 20 mA. The increased light output of the LEDs with GRIN ITO AR coating compared to LEDs with dense ITO coating is attributed to the virtual elimination of Fresnel reflection as expected from the theoretical calculation shown in Figure 4, and surface roughening of low-refractive-index ITO.

In conclusion, we showed that the refractive index of a thin-film material, which is chosen for its desirable properties other than refractive index, can be precisely tuned by using oblique-angle deposition. As a proof of the concept, we demonstrated a six-layer GRIN AR coating made entirely of ITO, chosen for its high electrical conductivity, high optical transmittance, and low contact resistance with GaN, with each layer having an individually tailored refractive index for optimum AR performance. It was shown experimentally that GaInN LEDs with the GRIN ITO AR contact achieved light-extraction efficiency enhancement of 24.3% over LEDs with conventional ITO contact by virtual elimination of Fresnel reflection, and surface roughening of low-refractive-index ITO.

Experimental

ITO nanorod thin films were grown by oblique-angle deposition using electron-beam evaporation. Pure ITO granules were used as evaporation source, and the deposition rate was well controlled at 2 Å s^{-1}. The apparatus used in the oblique-angle deposition had a sample stage, on which the substrate was loaded, with controllable polar-angle rotation. For each layer, the sample stage was fixed so that the substrate has a certain tilt angle with respect to the vapor flux direction, and hence a layer with desired refractive index was deposited.

The GRIN ITO AR coating was incorporated onto a GaInN LED emitting at a peak wavelength of 474 nm. The GaInN LED structure was grown by metal–organic chemical vapor deposition on *c*-plane sapphire substrate and consisted of a 3-µm-thick n-type GaN buffer layer, an n-type GaN lower cladding layer, a GaInN/GaN multiple-quantum-well active region, a p-type GaN upper cladding, and a highly doped p-type GaN contact layer. LED mesa structures were obtained by standard photolithographic patterning followed by chemically assisted ion-beam etching using Cl_2 and Ar, to expose the n-type cladding layer. Then, a 5-nm-thick AgCu alloy was deposited by electron-beam evaporation and annealed at 500 °C under O_2 ambient to form transparent ohmic contact with p-type GaN. A 500-nm-thick GRIN ITO AR contact consisting of six layers in which the refractive indices of the layers follow the modified-quintic-index profile, was deposited on the oxidized AgCu contact on p-type GaN by oblique-angle electron beam deposition with incident angles of 0°, –45°, 60°, –75°, 80°, and –85°. For comparison, LEDs with 500-nm-thick dense ITO, deposited with incident angle of 0° were fabricated on the same wafer piece. The inset of Figure 6 shows schematic cross-sectional views of the GaInN LEDs. The n-type contact for all samples was electron-beam evaporated Ti/Al/Ti/Au annealed at 650 °C for 1 min.

Received: April 27, 2007
Revised: September 14, 2007
Published online:

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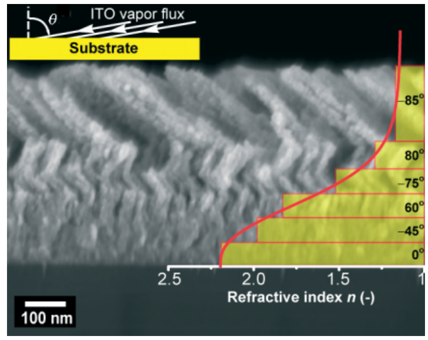

Light-Emitting Diodes

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