

Nanostructured Antireflection Coatings for Optical Detection and Sensing Applications

Gopal G. Pethuraja^{1,2}, Roger E. Welser¹, John W. Zeller¹, Yash R. Puri¹, Ashok K. Sood¹, Harry Efstathiadis², Pradeep Halder², Nibir K. Dhar³ and Priyalal Wijewarnasuriya⁴

¹Magnolia Optical Technologies Inc., 52-B Cummings Park, Suite 314, Woburn, MA 01801

²Energy and Environmental Technology Applications Center (E2TAC), College of Nanoscale Science and Engineering, Albany, NY 12203

³DARPA/MTO, 675 North Randolph Street, Arlington, VA 22203

⁴U.S. Army Research Laboratory, 2800 Powder Mill Road, Adelphi, MD 20783

ABSTRACT

Optical components such as lenses, glass windows, and prisms are subject to Fresnel reflection due to the mismatch between the refractive indices of the air and glass. An optical interface layer, i.e., antireflection (AR) layer, is needed to eliminate this unwanted reflection at the air/glass interface. Nanostructured broadband and wide-angle AR structures have been developed using a scalable self-assembly process. Ultra-high performance of the nanostructured AR coatings has been demonstrated on various substrates such as quartz, sapphire, polymer, and other materials typically employed in optical lenses. AR coatings on polycarbonate lead to optical transmittance enhancement from approximately 90% to almost 100% for the entire visible, and part of the near-infrared (NIR), band. The AR coatings have also been demonstrated on curved surfaces. AR coatings on n-BK7 lenses enable ultra-high light transmittance for the entire visible, and most of the NIR, spectrum. Nanostructured oxide layers with step-graded index profiles, deposited onto the optical elements of an optical system, can significantly increase sensitivity, and hence improve the overall performance of the system.

INTRODUCTION

Incident light on optical windows and lenses will partially undergo Fresnel reflection due to the mismatch between the refractive indices of the air and glass. For normal light incidence, the Fresnel reflection at the interface of two mediums having refractive indices n_1 and n_2 is given by:

$$R = \left(\frac{n_1 - n_2}{n_1 + n_2} \right)^2$$

The reflection loss at the air/glass-lens interface is typically around 4% from the lens surface (~8% from both surface interfaces of the lens) at normal light incidence. This loss can be greater than 40% for off-angle light incidence. An optical interface layer with intermediate refractive indices at the air/glass interface can eliminate this unwanted reflection. Conventionally, a single-layer coating with optical thickness equal to one-quarter of the wavelength ($\lambda/4$) of interest has been used as an antireflection (AR) coating. Ideally, such single-layer AR coating should have a refractive index, $n_{\lambda/4}$, as given by:

$$n_{\lambda/4} = \sqrt{n_s \times n_a}$$

where n_s and n_a are the refractive indices of the substrate and air, respectively. Often, due to the unavailability of materials with desired precise refractive index values, the performance of such $\lambda/4$ AR coatings deviates from the optimum. This is especially the case for low refractive index substrates such as glass. An ideal single-layer $\lambda/4$ AR coating on a glass surface in air ambient would require a material with a refractive index of approximately 1.2. However, there is no conventional inorganic material that has such a low refractive index. Fundamentally, these single-layer $\lambda/4$ AR coatings can minimize reflection only for one specific wavelength at normal incidence and they are inherently unable to exhibit a spectrally broadband reduction in reflection over a wide range of incidence angles.

Recent developments have enabled nanostructured coatings to overcome this restraint and provide a new avenue for novel AR applications. The need for broadband and wider-angle AR structures has been significantly amplified in recent years due to the increasing practical utility of wider angle and higher sensitivity optical detection systems used in commercial and defense applications. While various approaches have been developed to create high-performance broadband AR structures, most of these have encountered difficulties due primarily to limitations in tuning the optical material refractive index and lack of controllability in achieving the desired thickness of this material. We have developed high-performance AR structures using a scalable self-assembly process to fabricate nanostructured SiO_2 multilayer structures [1]. The process has the ability to create ultra-low refractive index (down to 1.08) materials while offering controllability of the layer thickness and refractive index, thus overcoming these limitations [2].

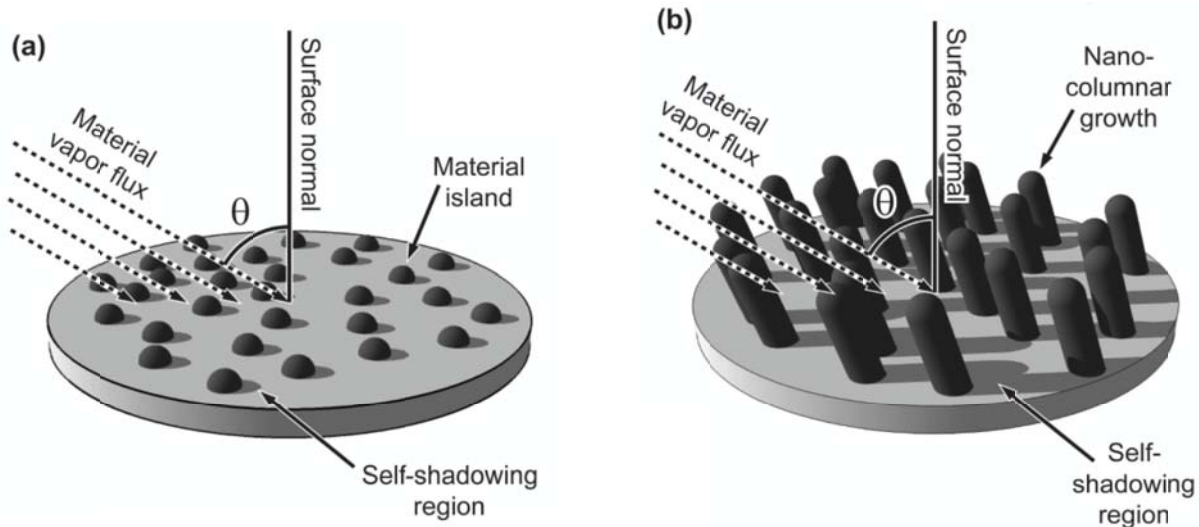


Figure 1: (a) Simplified schematic of the oblique-angle deposition process for synthesizing porous, nanostructured films, showing (a) the initial formation of material islands at random locations across the substrate, followed by (b) the formation of self-shadowed regions and nano-columnar growth when material vapor flux arrives at a non-normal deposition angle (θ) to the substrate.

Oblique-angle deposition has been used to tailor the refractive index of porous, nanostructured SiO_2 materials, and to build high-performance step-graded refractive index structures on glass and other relevant substrates [3]. Step-graded designs enable the formation of

antireflection structures that combine broadband and omnidirectional characteristics [4-7] . In this paper, we review the oblique-angle deposition process, summarize recent results from step-graded antireflection coatings on transparent polymer and lenses, and discuss our efforts to extend the nanostructured coating process to support larger area substrates.

EXPERIMENT

Oblique-angle deposition is a method of growing porous thin films [3], and hence thin films with low refractive indices, enabled by surface diffusion and self-shadowing effects during the deposition process. Random growth fluctuations on the substrate produce a shadow region that incident vapor flux cannot reach, and a non-shadow region where incident flux deposits preferentially, thereby creating oriented rod-like structures with high porosity as illustrated in Figure 1. The deposition angle, defined as the angle between the normal to the sample surface and the incident vapor flux, affects the tilt of the nanorod structures relative to the sample surface. Because the gaps between the nanorods can be much smaller than the wavelength of visible and infrared light, the nanostructured layers act as a single homogenous film with an intermediate refractive index between that of air and of the nanorod material, which decreases with increasing porosity.

SiO₂ coatings are well-known for their long-term stability and high transmittance over a wide spectral range. However, conventional dense SiO₂ has a refractive index of about 1.46, and thus is not an effective AR material for optical windows having a refractive index near 1.5. On the other hand, the refractive index of porous SiO₂ can be reduced to values of 1.1 or lower by increasing the porosity [2]. The use of porous nanomaterials fabricated by oblique-angle deposition offers unique advantages compared to other methods such as tunability of the refractive index, flexibility in material choice, simplicity of a physical vapor deposition process, and the ability to optimize the coating for any substrate-ambient material system. Figure 2 shows a cross-sectional scanning electron micrograph of typical porous nanomaterial thin films grown by oblique-angle deposition using silicon dioxide.

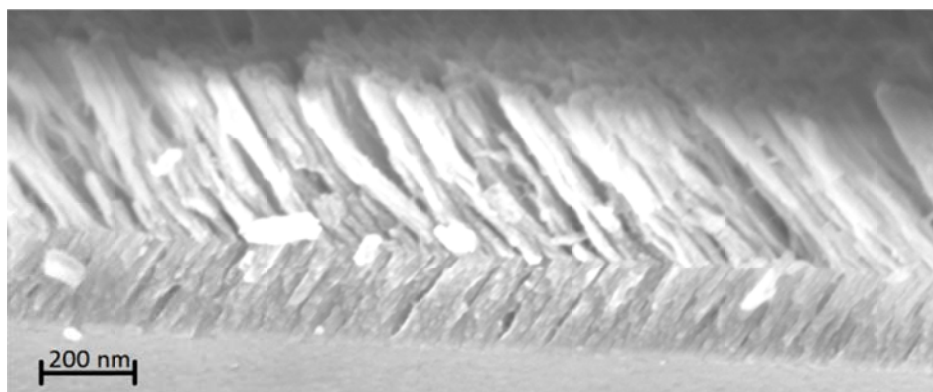


Figure 2. Cross-sectional scanning electron micrograph of a multi-layered nanostructured optical thin film synthesized by the oblique-angle deposition of silicon dioxide.

In this work, AR coatings with specular surfaces comprising multiple layers of porous SiO₂ have been deposited on optical lenses and UV stabilized polycarbonate substrates. The specific target thickness and refractive index values for the step-graded AR coating structure were chosen to minimize an unwanted dip in transmittance near the 550 nm wavelength due to interference effects observed in our earlier work [8].

RESULTS AND DISCUSSION

Broadband and high performance antireflection coatings have been demonstrated on polycarbonate substrates. Polycarbonate is an excellent plastic for display filter, plastic lenses, and face shields, and is commonly utilized in commercial and defense applications. It provides high impact resistance combined with an excellent flammability rating. Nanostructured SiO₂ multilayer AR coatings with an optimized step-graded index profile have been deposited on both sides of polycarbonate sheets. The AR coating eliminates nearly all the reflection loss and yields ultra-high optical transmittance. Figure 3 compares the optical transmittance spectra of AR-coated and uncoated polycarbonate sheets. The expanded transmittance spectrum over the visible band is plotted in the inset of Figure 3. As seen in the inset, the uncoated polycarbonate sheet shows approximately 90% transmittance over the visible spectrum (~400-800 nm). The AR coating on the polycarbonate sheet increases the transmittance to almost 100%, nearly a 10% enhancement in optical transmittance. The enhancement in the optical transmittance is observed for the entire visible and part of the near-infrared (NIR) band.

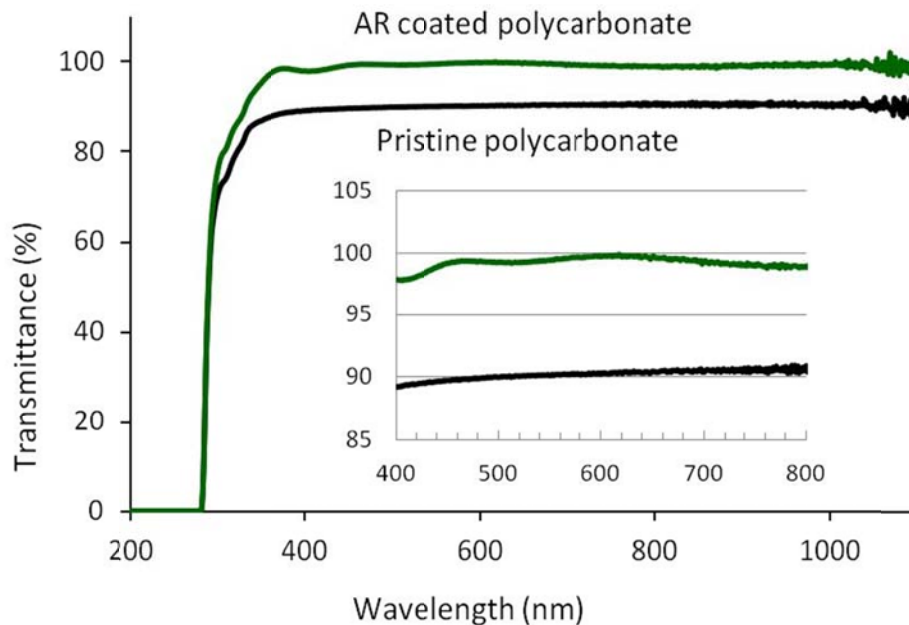


Figure 3: Optical transmittance spectrum of a transparent polycarbonate sheet (5 mil thickness) before and after application of the large-area AR coating. The AR coating yields nearly 100% transmittance.

Magnolia has designed and optimized step-graded index profiles for n-BK7 lenses and windows. Nanostructured SiO₂ layers of the desired refractive index were deposited on the surface of the optical components by an oblique angle deposition. Multilayer step-graded index profiles were created and optimized by controlling the refractive index and thickness of the individual layers. Figure 4 compares the transmittance of uncoated and nanostructured SiO₂ multilayer coated n-BK7 lenses. The nanostructured AR coating significantly improved optical transmittance through the lens from 94% to almost 100%. This optical transmittance enhancement has been preserved over the entire visible, and majority of the NIR, spectrum. Hence, the AR-coated lens can transmit the near-total optical signal to a sensor over a broader spectrum by eliminating the vast majority of unwanted reflections, enabling significantly higher responsivities in detector devices and thereby enhancing their effectiveness. This approach can be expanded to various infrared optical components and significantly improve imaging, sensing and detection capabilities of infrared systems.

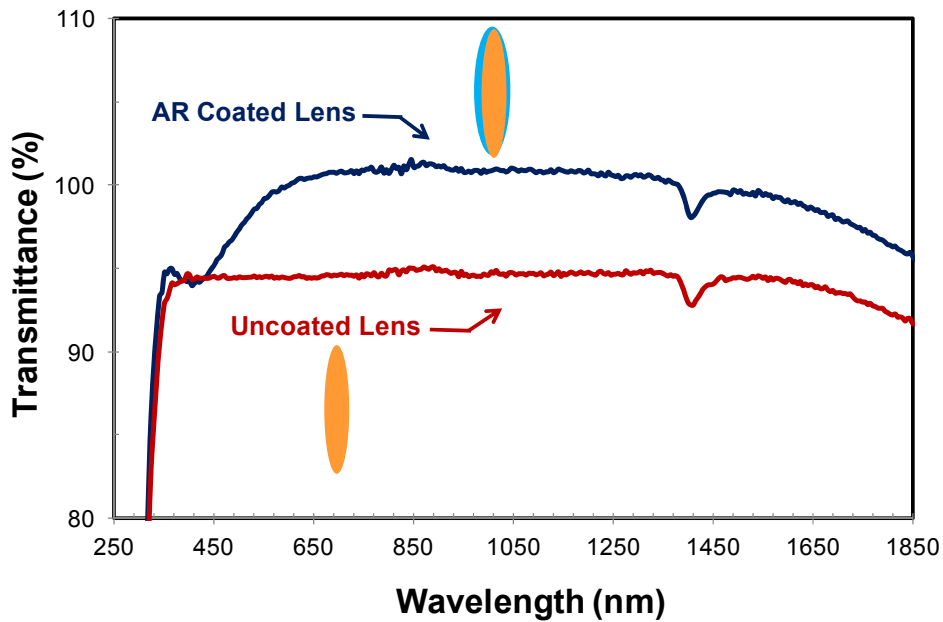


Figure 4: Measured wavelength-dependent transmittance of a nanostructured SiO₂ coated lens compared to an uncoated lens.

SUMMARY

This paper has shown that deposition of oblique-angle SiO₂ nanowires and nanorods offers an innovative approach for developing high-quality antireflection coatings for use on next-generation sensors and optical windows to minimize reflection losses for both defense and commercial applications. Step-graded nanostructured SiO₂ antireflection technology has been shown to be both broadband and omnidirectional in nature. Nanostructured broadband AR structures have been developed and demonstrated on various substrates and optical elements. AR coatings deposited on transparent polycarbonate substrates improve optical transmittance from 90% to almost 100% by eliminating unwanted refraction on both sides of the substrate. The broadband and wide-angle AR coatings have also been demonstrated on curved surfaces.

AR coatings deposited on n-BK7 lenses enhance their optical transmittance from 94% to almost 100% over the visible, and majority of the NIR, spectrum. Nanostructured oxide layers with step-graded index profiles, deposited onto the optical elements of an optical system, can significantly increase their sensitivity and hence improve the overall performance of the system. Continued efforts are underway to demonstrate nanowire-based AR coatings for spectral bands spanning from the ultraviolet into the infrared for next-generation sensors.

REFERENCES

- [1] A. K. Sood, A. W. Sood, R. E. Welser, G. G. Pethuraja, Y. R. Puri, X. Yan, D. J. Poxson, J. Cho, E. F. Schubert, N. K. Dhar, D. L. Polla, P. Haldar, and J. L. Harvey, "Development of Nanostructured Antireflection Coatings for EO/IR Sensor and Solar Cell Applications," *Mater. Sci. Appl. VO - 03*, no. 09, p. 633, 2012.
- [2] J.-Q. Xi, M. F. Schubert, J. K. Kim, E. F. Schubert, M. Chen, S.-Y. Lin, W. Liu, and J. A. Smart, "Optical thin-film materials with low refractive index for broadband elimination of Fresnel reflection," *Nat. Photonics*, vol. 1, no. 3, pp. 176–179, Mar. 2007.
- [3] D. J. Poxson, F. W. Mont, M. F. Schubert, J. K. Kim, and E. F. Schubert, "Quantification of porosity and deposition rate of nanoporous films grown by oblique-angle deposition," *Appl. Phys. Lett.*, vol. 93, no. 10, p. 101914, 2008.
- [4] R. E. Welser, A. W. Sood, A. K. Sood, D. J. Poxson, S. Chhajed, J. Cho, E. F. Schubert, D. L. Polla, and N. K. Dhar, "Ultra-high transmittance through nanostructure-coated glass for solar cell applications," in *Proc. of SPIE*, 2011, vol. 8035, p. 80350X.
- [5] R. E. Welser, A. W. Sood, G. G. Pethuraja, A. K. Sood, X. Yan, D. J. Poxson, J. Cho, E. Fred Schubert, and J. L. Harvey, "Broadband nanostructured antireflection coating on glass for photovoltaic applications," *Photovoltaic Specialists Conference (PVSC), 2012 38th IEEE*. pp. 3339–3342, 2012.
- [6] G. G. Pethuraja, A. Sood, R. Welser, A. K. Sood, H. Efstathiadis, P. Haldar, and J. L. Harvey, "Large-area nanostructured self-assembled antireflection coatings for photovoltaic devices," *Photovoltaic Specialists Conference (PVSC), 2013 IEEE 39th*. pp. 99–102, 2013.
- [7] A. K. Sood, G. Pethuraja, A. W. Sood, R. E. Welser, Y. R. Puri, J. Cho, E. F. Schubert, N. K. Dhar, P. Wijewarnasuriya, and M. B. Soprano, "Development of large area nanostructure antireflection coatings for EO/IR sensor applications," in *Proc. SPIE 8512, Infrared Sensors, Devices, and Applications II*, 2012, vol. 8512, p. 85120R.
- [8] S. Chhajed, D. J. Poxson, X. Yan, J. Cho, E. F. Schubert, R. E. Welser, A. K. Sood, and J. K. Kim, "Nanostructured multilayer tailored-refractive-index antireflection coating for glass with broadband and omnidirectional characteristics," *Applied Physics Express*, vol. 4, no. 5. p. 052503, 2011.