Applied Physics Express **4** (2011) 052503
DOI: 10.1143/APEX.4.052503

Nanostructured Multilayer Tailored-Refractive-Index Antireflection Coating for Glass with Broadband and Omnidirectional Characteristics

Sameer Chhajed, David J. Poxson[1], Xing Yan[1], Jaehee Cho, E. Fred Schubert*, Roger E. Welser[2], Ashok K. Sood[2], and Jong Kyu Kim[3]

Future Chips Constellation, Department of Electrical, Computer, and Systems Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180, U.S.A.
[1] *Future Chips Constellation, Department of Physics, Applied Physics, and Astronomy Rensselaer Polytechnic Institute, Troy, NY 12180, U.S.A.*
[2] *Magnolia Optical Technologies, Inc., Woburn, MA 01801, U.S.A.*
[3] *Deparment of Materials Science and Engineering, Pohang University of Science and Technology, Pohang 790-784, Korea*

Received March 3, 2011; accepted April 20, 2011; published online May 11, 2011

The design, fabrication, and characterization of a broadband, omnidirectional, graded-index anti-reflection (AR) coating on a glass substrate, fabricated by using nanostructured low-refractive-index ($n = 1.05$–1.40) silica, is reported. The AR coating is designed by using a genetic algorithm and fabricated by using oblique angle deposition. The AR coating is designed for the wavelength range of 400 to 2500 nm and 0 to 40° angle of incidence. The measured average optical transmittance between 1000 and 2000 nm is improved from 92.6 to 99.3% at normal incidence by using a two-layer AR coating deposited on both surfaces of the glass substrate.

Reducing optical reflection from surfaces is important to many applications in optics. This is commonly achieved through coating[1–5] or texturing[6–11] the surface of interest. Numerous applications involving dielectric or semiconducting materials use the light that is transmitted through the material's surface. Examples of such application are optical lenses, windows, photovoltaic devices, and photodetectors. Glass (amorphous SiO_2) is one such dielectric material widely used in a variety of optical applications, e.g., lenses, windows, and as a cover or encapsulation for semiconductor optoelectronic devices. Glass is completely transparent for wavelengths longer than 400 nm. However, due to Fresnel reflection, it reflects about 4% of the incident light from its surface (~8% from two surfaces). This reflection is undesirable in many applications as it can degrade the efficiency of the underlying device (e.g., the efficiency of a solar photovoltaic cell), reduce the signal-to-noise ratio (e.g., in a photodetector), and cause glare [e.g., from liquid crystal display (LCD) screens, computer monitors, and televisions]. For such applications, it is important not only to reduce reflectance but also to improve the transmittance through the surface. This requires that the coating material be nonabsorbing and the coating surface be specular.

Conventionally, a single-layer coating with optical thickness equal to one-quarter of the wavelength ($\lambda/4$) of interest has been used as an AR coating. Ideally, such single-layer $\lambda/4$ anti-reflection (AR) coating should have a refractive index, $n_{\lambda/4}$, as given by[12)]

$$n_{\lambda/4} = \sqrt{n_{\text{substrate}} \times n_{\text{air}}}. \quad (1)$$

Often due to the unavailability of materials with the desired, exact value of the refractive index, 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 $(1.46)^{1/2} \approx 1.2$. There is no conventional inorganic material that has such a low refractive index. Also, 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 spectrally broadband reduction in reflectance over a wide range of angles of incidence.

In 1880, Lord Rayleigh mathematically demonstrated that graded-refractive-index layers have broadband antireflection properties. Multilayer stacks of materials with different refractive indices have been used in order to achieve broadband reduction in reflection.[13,14] AR coatings with specular surface made of multiple discrete layers of nonabsorbing materials can exploit thin-film interference effects to reduce the reflectance while improving transmittance. Recently, it has been shown that discrete multilayer AR coatings can outperform continuously graded AR coatings, thereby offering powerful techniques to reduce reflectance.[15]

Optimization of multilayer AR coatings is a difficult challenge because of the extremely large and complex dimensional space of possible solutions. Analytical methods to optimize AR coatings are not feasible due to the complexity of the problem. Heuristic methods such as needle optimization,[16] jump elimination,[17] and genetic algorithm[18] are commonly used. Our approach utilizes a computational genetic algorithm method, the details of which have been recently discussed in the literature.[18]

Until recently, due to the unavailability of optical materials with very low and tunable refractive indices ($n < 1.4$), near-perfect graded-index AR coatings could not be realized for glass substrates. We previously demonstrated a refractive index of as low as 1.05 for nanoporous SiO_2 by using oblique-angle deposition.[19] Figure 1 shows the refractive index and calculated porosity of oblique-angle-deposited SiO_2 versus deposition angle following the formula developed by Poxson *et al.*[20] The figure shows the tunability of the refractive index of an optical material to virtually any value between its bulk value and a value close to 1.0. The scanning electron micrographs in Fig. 1 show the gradual increase in porosity of a SiO_2 film starting from a dense bulk film deposited at an angle of 0° to a highly nanoporous film deposited at 75°. Unlike any other method, the use of porous nanomaterials fabricated by oblique-angle deposition, as described in this paper, offers advantages such as tunability of refractive index, flexibility in choice of material, simplicity of a physical vapor deposition process, and the ability to optimize the coating for any substrate-ambient material system. For multilayer AR coatings

*E-mail address: EFSchubert@rpi.edu

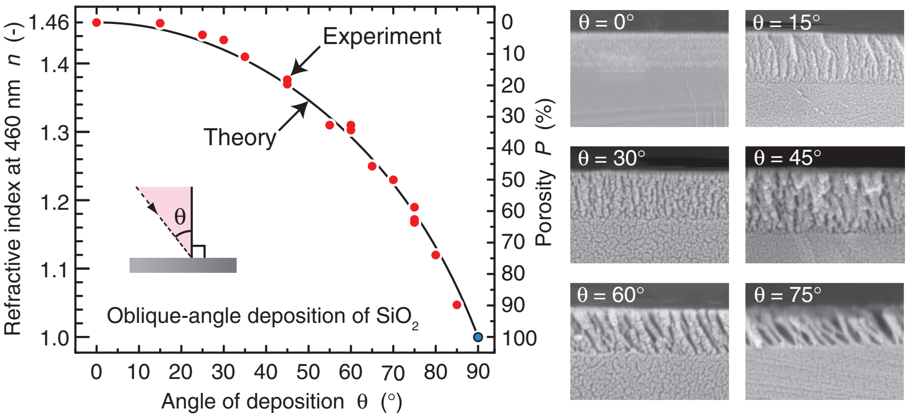

Fig. 1. Refractive index and porosity of oblique-angle-deposited SiO_2 versus deposition angle. The theoretical curve is after Poxson *et al.*[20] The scanning electron micrographs show a gradual increase in porosity of SiO_2 starting from a dense bulk film at 0° deposition angle and to a highly nanoporous film at 75° deposition angle.

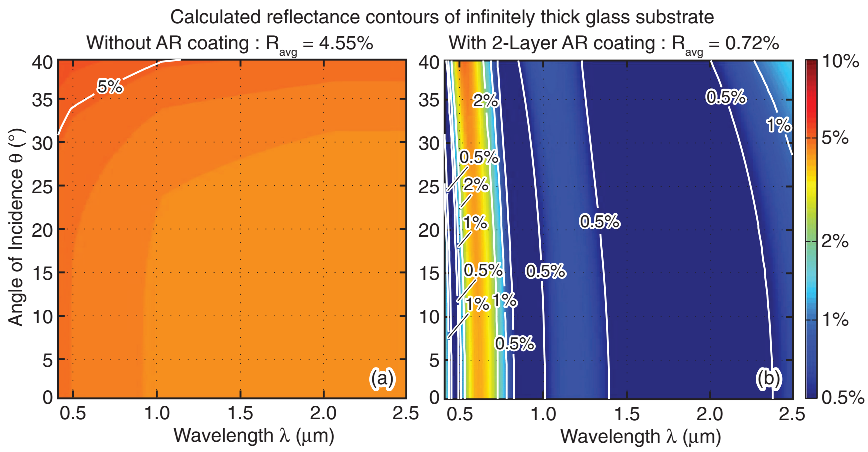

Fig. 2. Contour plots of calculated reflectance versus wavelength and angle of incidence for an infinitely thick glass substrate with (a) no AR coating and (b) single-side two-layer AR coating.

reported here, the refractive index of the layers is step-graded, i.e., decreased in discrete steps, from the substrate value of 1.46 to a value of 1.18.

A two-layer AR coating, optimized for glass substrate, is fabricated and characterized, as described below. During the theoretical design optimization process, the thickness t, as well as the porosity P of each layer in the multilayer graded-index AR coating, is allowed to vary. All the coatings are optimized in the wavelength range of 400 to 2500 nm and angle of incidence range of 0 to 40°. Figure 2 shows the contour plots of calculated reflectance versus wavelength and angle of incidence.

Our AR coating design is a two-layer AR coating with each layer having a tailored refractive index fabricated by using oblique angle electron-beam deposition. The details of this deposition technique are discussed elsewhere.[21–23] The tooling factor (thickness of film on substrate/thickness displayed on thickness monitor) depends on the deposition angle and was calculated for each angle using a recently developed formula.[20] The appropriate deposition angle was then chosen for the deposition of the actual AR coating layers based on Fig. 1 and the required value of the refractive index.[20] Three samples were prepared: (a) a reference glass substrate with no AR coating (called Sample A), (b) a glass substrate with a two-layer AR coating on one side (called Sample B), and (c) a glass substrate with a two-layer AR coating on both sides (called Sample C). The two-layer AR coating is composed of a first layer of porous SiO_2 ($P = 22\%$, $n = 1.36$ at 630 nm, $t = 197$ nm) deposited at a 54° substrate angle, and a second layer of porous SiO_2 ($P = 60\%$, $n = 1.18$ at 630 nm, $t = 289$ nm) deposited at a 78° substrate angle. The thickness and refractive index values of each coating are measured using variable angle spectroscopic ellipsometry. The thickness values were also confirmed using scanning electron microscopy. Note that all layers of the multilayer AR coating are made of a single material, porous silica (porous SiO_2). Silica is an excellent material for AR coating on a glass substrate as it is native, stable, and robust.

The optical transmittance of the multilayer AR samples was measured using (i) a normal incidence broadband spectrophotometer measurement setup and (ii) a variable angle and wavelength-dependent transmittance measurement setup. The thickness of the glass substrate used is 250 µm, much greater than the range of wavelengths under consideration; therefore, we do not expect optical interference effects from the substrate. Figure 3 shows the measured wavelength-dependent transmittance of a glass

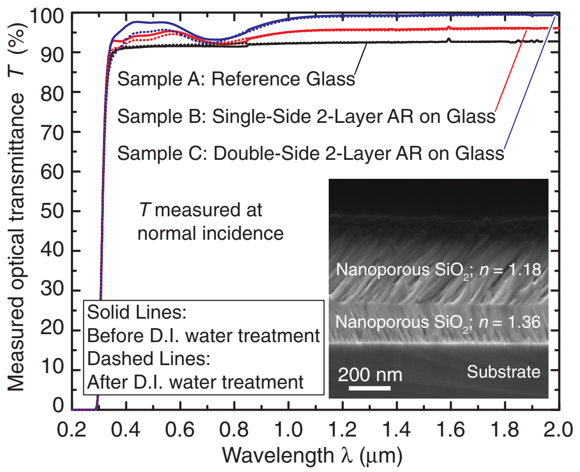

Fig. 3. Measured wavelength-dependent normal incidence transmittance of glass substrate with (a) no AR coating, (b) single-side two-layer AR coating, and (c) double-side two-layer AR coating. The inset shows the scanning electron micrograph of the two-layer AR coating (D.I.: deionized).

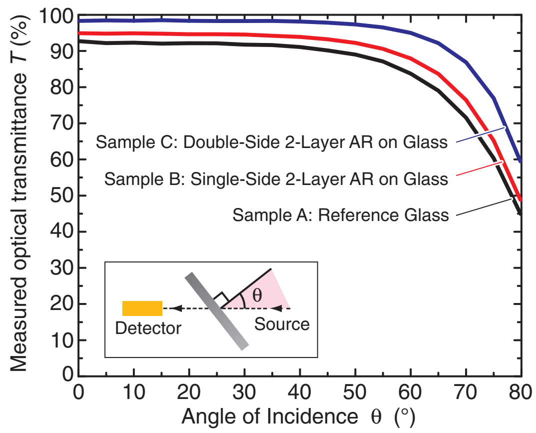

Fig. 4. Measured angle-dependent transmittance of glass substrate with (a) no AR coating, (b) single-side two-layer AR coating, and (c) double-side two-layer AR coating.

substrate with (a) no AR coating (Sample A), (b) single-side two-layer AR coating (Sample B), and (c) double-side two-layer AR coating (Sample C). The inset shows the scanning electron micrograph of the two-layer AR coating. The wavelength-dependent normal incidence transmittance of Samples A, B, and C is measured between 200 and 2000 nm using a JASCO V-570 spectrophotometer. The measurements reveal that the average transmittance between 400 and 2000 nm for an uncoated glass substrate is 92.2% (Sample A) and it increases to 95.1% for a glass substrate with a two-layer AR coating on one side (Sample B) and further increases to 98.1% for a glass substrate with a two-layer AR coating on both sides (Sample C). The local minima seen in the transmittance curves around 700 nm are due to the thin-film interference effects of the AR coating. The average transmittance between 1000 and 2000 nm for a glass substrate with a double-side AR coating is measured to be 99.3%.

Angle-dependent reflectance measurements between 350 and 1700 nm are performed using a custom-built laboratory setup. For each sample, the reflectance data is measured for angles of incidence ranging between 0 and 80° with 5° increments. Figure 4 shows the plots of the measured angle-dependent transmittance. It shows very high transmittance (very low reflectance) over a wide range of incident angles, demonstrating the predicted broadband and omnidirectional characteristics of the two-layer graded-index AR coating. The average transmittance, for wavelengths between 350 and 1700 nm and incident angles between 0 and 40°, for the uncoated glass substrate is 92% (Sample A) and it increases to 94.6% for a glass substrate with a two-layer AR coating on one side (Sample B) and further increases to 98.3% for a glass substrate with a two-layer AR coating on both sides (Sample C).

While the mechanical stability of porous films can be an area of concern, we have not observed any signs of disintegration, degradation of performance, or other adverse effects on these samples even after several months. As a test, we immersed the three samples in an ultrasonic water bath for 15 min. The samples were then blow-dried with N_2 and baked at 120 °C for 12 h in N_2 ambient. The transmittance spectra of all the three samples were then re-measured using the JASCO spectrophotometer; the transmittance showed no significant change at wavelengths longer than 800 nm. The adhesion of the multilayer AR coating made from nano-structured thin films was tested using a "scotch tape test" in which a piece of standard adhesive-backed packing tape was applied to the sample and then removed. The AR coating was not damaged during the test.

In conclusion, we have demonstrated a multilayer, broad-band, omnidirectional AR coating made of a single material with tailored-refractive-index layers on a glass substrate. The availability of the nanostructured low- and tunable-n materials deposited by using oblique-angle deposition has allowed the fabrication of highly effective AR coatings for low-refractive-index substrates such as glass. The measured average optical transmittance of an uncoated glass substrate between 1000 and 2000 nm is improved from 92.6 to 99.3% at normal incidence by using a two-layer AR coating deposited on both surfaces of the glass substrate.

Acknowledgments Support by the Magnolia Optical Technologies Inc. (USA), the National Science Foundation (USA), and Samsung LED Company (Korea) is gratefully acknowledged.

1) L. Holland and T. Putner: J. Sci. Instrum. **36** (1959) 81.
2) J. Zhao and M. A. Green: IEEE Trans. Electron Devices **38** (1991) 1925.
3) R. Schuhmann and M. Schulz-Grosser: Proc. SPIE **3133** (1997) 256.
4) D. J. Aiken: Sol. Energy Mater. Sol. Cells **64** (2000) 393.
5) B. Kumar *et al.*: Proc. 31st IEEE Photovoltaic Specialist Conf., 2005, p. 1205.
6) K.-S. Han *et al.*: Sol. Energy Mater. Sol. Cells **94** (2010) 583.
7) H. A. Miska: Sol. Energy Mater. **11** (1984) 231.
8) N. Yamaguchi *et al.*: J. Sol–Gel Sci. Technol. **33** (2005) 117.
9) S. W. Kim *et al.*: J. Appl. Phys. **96** (2004) 6766.
10) P. Papet *et al.*: Sol. Energy Mater. Sol. Cells **90** (2006) 2319.
11) C. H. Sun *et al.*: Appl. Phys. Lett. **92** (2008) 061112.
12) H. A. MacLeod: *Thin Film Optical Filters* (Elsevier, Amsterdam, 1968).
13) W. Wang and H. Hao: Chin. Opt. Lett. **8** (2010) 35.
14) S. D. Gupta and G. S. Agarwal: Opt. Express **15** (2007) 9614.
15) M. F. Schubert *et al.*: Appl. Phys. Express **3** (2010) 082502.
16) A. V. Tikhonravov *et al.*: Appl. Opt. **35** (1996) 5493.
17) Y.-Y. Liou *et al.*: Jpn. J. Appl. Phys. **46** (2007) 5143.
18) M. F. Schubert *et al.*: Opt. Express **16** (2008) 5290.
19) J.-Q. Xi *et al.*: Nat. Photonics **1** (2007) 176.
20) D. J. Poxson *et al.*: Appl. Phys. Lett. **93** (2008) 101914.
21) M. F. Schubert *et al.*: Appl. Phys. Lett. **90** (2007) 141115.
22) K. Robbie and M. J. Brett: J. Vac. Sci. Technol. A **15** (1997) 1460.
23) K. Robbie *et al.*: J. Vac. Sci. Technol. B **16** (1998) 1115.