

# Large-Area Nanostructured Self-Assembled Antireflection Coatings for Photovoltaic Devices

Gopal G. Pethuraja<sup>1,2</sup>, Adam Sood<sup>1</sup>, Roger Welser<sup>1</sup>, Ashok K. Sood<sup>1</sup>, Harry Efstathiadis<sup>2</sup>, Pradeep Haldar<sup>2</sup>, and Jennifer L. Harvey<sup>3</sup>

<sup>1</sup> Magnolia Solar, Inc., 54 Cummings Park, Woburn, MA 01801; and 251 Fuller Road, Albany, NY 12203, USA

<sup>2</sup>Energy and Environmental Applications Center (E2TAC), College of Nanoscale Science and Engineering, University at Albany, State University of New York, Albany, NY 12203, USA

<sup>3</sup>NYSERDA, 17 Columbia Circle, Albany, NY 12203, USA

## ABSTRACT

The scalability of nanostructured, self-assembled antireflection (AR) coatings has been demonstrated on 6-inch glass and silicon wafers. Ultra-high transmittance through these large-area coatings has been confirmed by measuring the short circuit current of a CIGS-based thin film photovoltaic (PV) device placed below the large-area AR-coated glass wafer. At normal light incidence, the light transmitted through the AR coated glass wafer yields 5% more short-circuit current compared to the uncoated glass wafer. At off-angle incidence, the light transmitted through the AR-coated wafer yields nearly 20% higher short-circuit current compared to light transmitted through an uncoated glass wafer. The large-area AR coating preserves ultra-high transmittance over a wide range of incident angles and has the potential to enhance PV device performance from dawn to dusk.

*Index terms* — antireflection coating, nanostructures, silicon dioxide, broadband, omni-directional.

## I. INTRODUCTION

A variety of transparent protective sheets such as glass and polymers have been used for photovoltaic (PV) modules. The light propagating from the air into the protective sheet undergoes unwanted Fresnel reflection at the air/sheet interface due to the dissimilar optical properties of the media. Although Fresnel reflection losses are relatively low at normal incidence, they can become quite substantial for off-angle light incidence. For example, Fresnel reflection at an uncoated glass/air interface generally varies from over 4% at normal incidence to more than 40% at a 75° incident angle. The Fresnel reflection limits the amount of light transmitted to the underlying energy harvesting device. Minimizing the reflection losses increases the density of photo-generated

carriers, resulting in an enhancement of module efficiency.

It has been known for some time that Fresnel reflection losses theoretically can be minimized between two media by grading the refractive index across the interface. However, the unavailability of materials with the desired refractive indices, particularly materials with very low refractive indices, has prevented the implementation of graded and step-graded refractive index designs. Recently, however, a new class of optical thin-film materials consisting of porous nanorods has enabled the realization of ultra-low refractive index materials [1]. In particular, oblique-angle deposition is a simple physical vapor deposition process that can be used to tailor the refractive index of a wide variety of thin-film materials [2].

Oblique-angle deposition has been used to build high-performance step-graded refractive index structures on glass substrates [3-4]. Step-grade designs enable the formation of antireflection (AR) structures that combine broadband and omnidirectional characteristics. Recently, ultra-high transmittance of step-graded nanostructured AR coatings has been demonstrated on laboratory scale glass substrates [5]. Practical implementation of these self-assembled, nanostructured optical coatings for various applications depends, in part, on their scalability. In this work, we report on the results of nanostructured AR coatings that have been scaled up to a large area glass substrate. Light transmitted through the AR-coated large-area glass wafer yields 5% higher short-circuit current at normal light incidence and more than 20% higher short-circuit current at off-angle light incidence on a PV device, compared to light transmitted through an uncoated glass wafer.

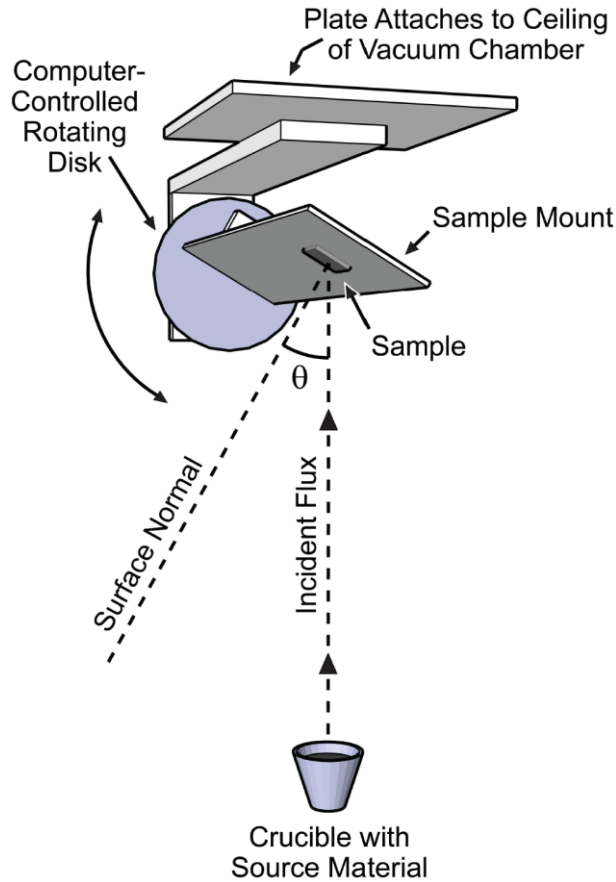


Fig. 1. Illustration of the experimental setup used in the oblique-angle deposition technique for porous nanostructured materials. The incident physical vapor flux strikes the sample at an angle relative to the surface normal.

## II. EXPERIMENTAL

Oblique-angle deposition is a method of growing porous thin films, and hence thin films with low-refractive index enabled by surface diffusion and self-shadowing effects during the deposition process. Figure 1 shows the experimental setup used in the oblique-angle deposition technique for growing porous nanostructured materials. 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 an oriented rod-like structure with high porosity as illustrated in Figure 2. The deposition angle, defined as the angle between the normal to the sample surface and the incident vapor flux, results in the formation of nanorod structures that are tilted 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 a refractive index intermediate between air and the nanorod material, decreasing in refractive index with increasing porosity.

SiO<sub>2</sub> coatings are well-known for their long-term stability and high transmittance over a wide spectral range. However, conventional dense SiO<sub>2</sub> has a refractive index around 1.46, and thus is not an effective AR material for optical windows with a refractive index near 1.5. On the other hand, the refractive index of porous

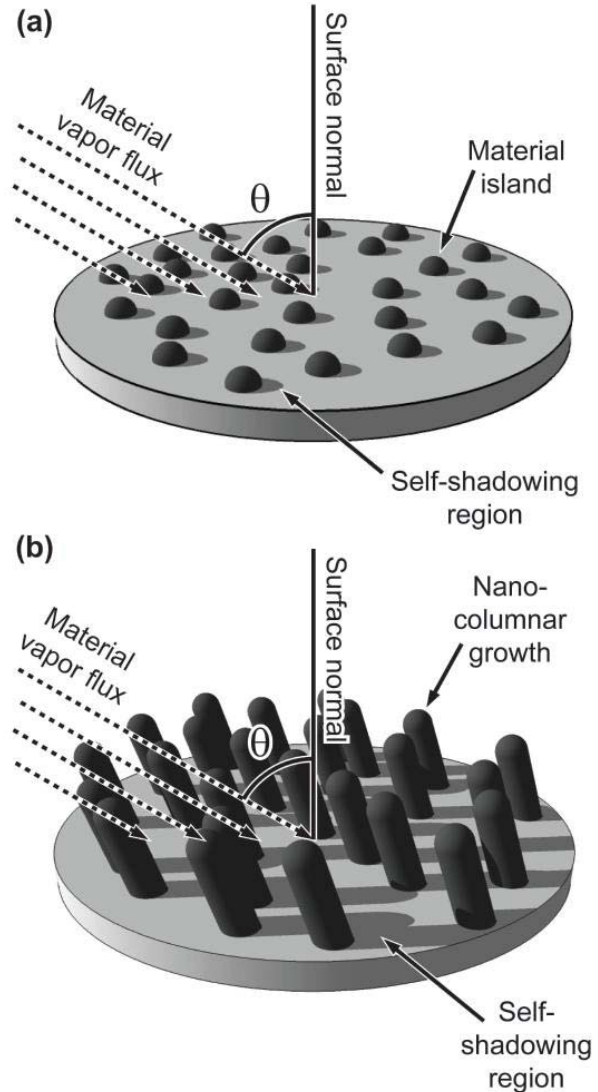


Fig. 2. (a) Simplified schematic of the oblique-angle deposition process for synthesizing porous, nanostructured films, showing (a) the initial formation of material islands at random locations across the substrate, followed by (b) the formation of self-shadowed regions and nano-columnar growth when material vapor flux arrives at a non-normal deposition angle ( $\theta$ ) to the substrate.

SiO<sub>2</sub> can be reduced to values of 1.1 or lower by increasing the porosity [1]. Unlike any other method, the use of porous nano-materials fabricated by oblique-angle deposition offers advantages 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 3 shows a cross-sectional scanning electron micrograph of typical porous nano-material thin-films grown by oblique-angle deposition using silicon dioxide.

In this work, AR coatings with specular surfaces comprised of multiple layers of porous SiO<sub>2</sub> layers have been deposited on 6-inch glass wafers. The specific target thickness and refractive index values for the step-graded structure were chosen to minimize an unwanted dip in transmittance near 550 nm due to interference effects observed in our earlier work [4]. Double-sided structures were prepared in an electron-beam evaporator using a multi-step process that included two or three different deposition angles and two-step deposition at each angle. The two-step deposition enhances the thickness

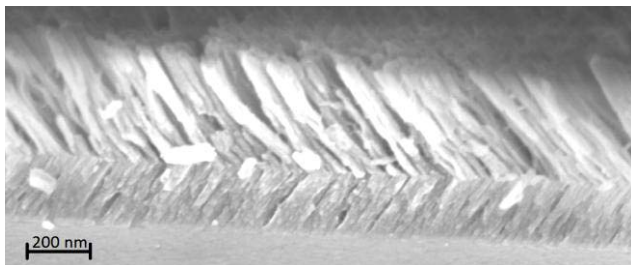


Fig. 3. Cross-sectional scanning electron micrograph of a multi-layered nanostructured optical thin film synthesized by the oblique angle deposition of silicon dioxide.

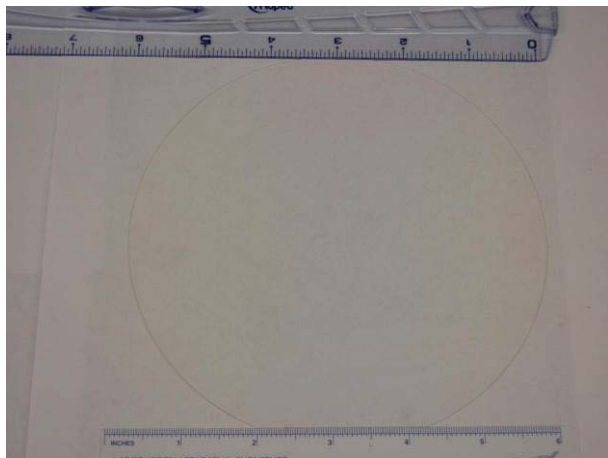


Fig. 4. Photograph of the large-area AR-coated glass wafer that was fabricated and tested during this study.

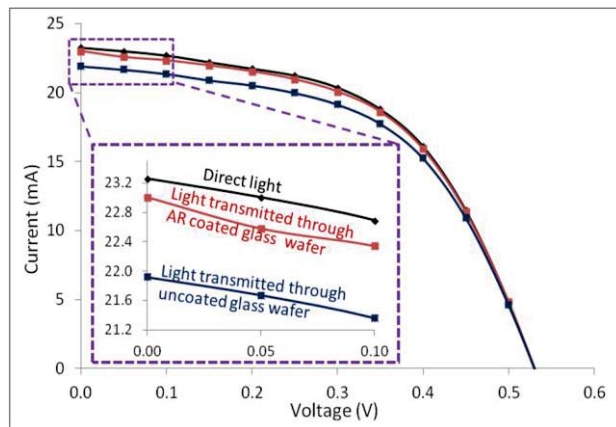


Fig. 5. Current-voltage characteristics of a CIGS photovoltaic device under illumination of direct light, as well as light transmitted through an AR-coated glass wafer and light transmitted through an uncoated glass wafer.

uniformity of each layer, as the deposited layer typically shows a linear non-uniformity in refractive index. A slight off-set of the deposition angle was employed to achieve graded refractive index of the layers for the entire area of the wafer. Figure 4 shows an optical image of the large-area AR-coated glass wafer with a 6" diameter.

To confirm the antireflective characteristics of the large area AR coating, light from a solar simulator was transmitted through both an AR-coated and an uncoated glass wafer, and analyzed using a CIGS thin-film-based PV device. The short circuit current of the PV device is used to gauge the transmittance functionality of the AR-coated glass wafer. The AR-coated and uncoated glass wafers were placed in the path of the light at the midpoint between the solar simulator and the PV device. The incident angle of the light on the glass wafer was changed by tilting the wafer at predefined angles with a tilt axis perpendicular to the direction of light transmittance. The light transmitted through the AR-coated and uncoated glass wafers has been analyzed by measuring current-voltage characteristics of the underlying CIGS device.

### III. RESULTS AND DISCUSSION

Light transmitted through the AR-coated glass wafer yields 5% more short circuit current compared to light transmitted through the uncoated glass wafer at normal light incidence. This is illustrated in Figure 5. It shows the current-voltage characteristics of the test PV device at various illumination conditions. In all cases, there is no change in the open circuit voltage. On the other hand, the short-circuit current is significantly influenced by the

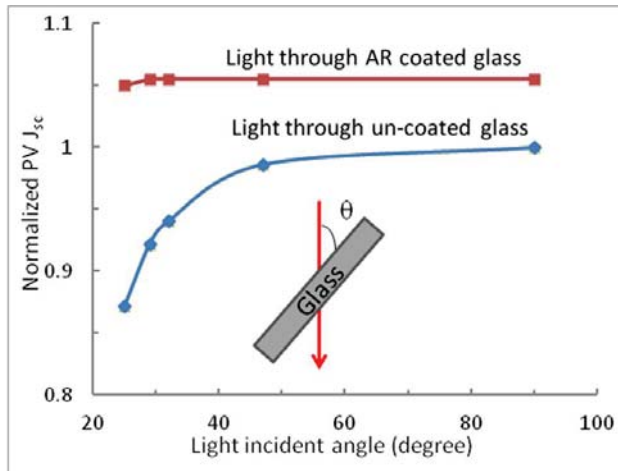


Fig. 6. Comparison of normalized short circuit current generated by a solar cell for light transmitted through an AR coated and an uncoated glass wafer for a range of light incident angle. At normal incidence, light transmitted through AR coated glass yields 5% more current than light transmitted through uncoated glass. At off-angles, AR coated glass yields approximately 20% more current than light transmitted through uncoated glass.

illumination conditions. Light transmitted through an uncoated glass wafer shows 6% lower short-circuit current compared to the direct light. This lower current is attributed to the Fresnel reflection at the air/glass interfaces. On the other hand, the light transmitted through the AR-coated glass wafer shows 5% higher short-circuit current than the light transmitted through the uncoated glass wafer. The observed increase in current is due to a reduction in the reflection losses and resultant increase in transmittance through the AR-coated glass.

The short circuit current analysis indicates that the light transmitted through the AR-coated glass wafer yields 20% higher short-circuit current compared to the light transmitted through the uncoated glass wafer at off-angle light incidence. Figure 6 compares the short circuit current observed under various illumination conditions and light incident angles. In the case of light transmitted through the uncoated glass wafer, the normalized short-circuit current varies from 1 to 0.87 as the light incident angle changes from 90 to 25 degrees. In the case of light transmitted through AR-coated glass wafer, the short-circuit current varies from 1.06 to 1.05 as the light incident angle changes from 90 to 25 degrees. The observed short-circuit current when illuminated through the AR-coated glass wafer is within 2% of the short-circuit current observed for direct light over the entire range of light incident angles used in this study. These results confirm the ultra-high transmittance of the AR-coated glass wafer. It is clear that the nanostructured AR

coatings have the potential to transmit sunlight into the underlying energy-harvesting device from dawn to dusk.

#### IV. SUMMARY

In summary, the deposition of AR coatings comprised of step-graded, nanostructured  $\text{SiO}_2$  has been successfully scaled up to a 6-inch glass wafer, and its ultra-high transmittance functionality has been confirmed. Light transmitted through an AR-coated glass wafer generates higher photocurrent in a PV device compared to light transmitted through an uncoated glass wafer. Light transmitted through an AR-coated glass wafer is within 2% of the short-circuit current observed for direct illumination over a wide range of incident angles. Light transmitted through an uncoated glass wafer provides 6 to 20% lower short-circuit current compared to direct illumination. The nanostructured self-assembled AR coating has the potential to preserve high performance of PV modules from dawn to dusk.

#### ACKNOWLEDGEMENT

The authors thank the New York State Energy Research and Development Authority (NYSERDA) for supporting this work via contract # ERDA1-0000017026.

#### REFERENCES

- [1] J.-Q. Xi, M. F. Schubert, J. K. Kim, E. F. Schubert, M. Chen, S.-Y. Lin, W. Liu, and J. A. Smart, "Thin film with low refractive index for broadband elimination of Fresnel reflection," *Nature Photonics*, vol. 1, p. 176-179, March 2007.
- [2] 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. 101914, September 2008.
- [3] S. Chhajed, D. J. Poxson, X. Yan, J. Cho, E. F. Schubert, R. E. Welsler, 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. 052503, May 2011.
- [4] R. E. Welsler, 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 Nanostructured-Coated Glass for Solar Cell Applications," *Proc. of SPIE*, vol. 8035, no. 80350X, April 2011.
- [5] R. E. Welsler, A. W. Sood, G. G. Pethuraja, A. K. Sood, X. Yan, D. J. Poxson, J. Cho, E. F. Schubert and J. L. Harvey, "Broadband Nanostructured Antireflection Coating on Glass for Photovoltaic Applications," *Photovoltaic Specialists Conference (PVSC), 2012 38<sup>th</sup> IEEE*, pp.003339-003342, 3-8 June 2012.