Mater. Res. Soc. Symp. Proc. Vol. 1245 © 2010 Materials Research Society **1245-A03-04**

A New Approach to Light Scattering from Nanotextured Interfaces for Thin-Film Silicon Solar Cells

C. Battaglia[1], J. Escarré[1], K. Söderström[1], F.-J. Haug[1], D. Dominé[1,2], A . Feltrin[1,3] and C. Ballif[1]
[1]Ecole Polytechnique Fédérale de Lausanne (EPFL), Institute of Microengineering (IMT), Photovoltaics and Thin Film Electronics Laboratory, 2000 Neuchâtel, Switzerland
[2]now at Scuola Universitaria Professionale della Svizzera Italiana (SUPSI), Institute of Applied Sustainability to the Built Environment (ISAAC), 6952 Canobbio, Switzerland
[3] now at Kaneka Corporation, Imec, 3001 Leuven, Belgium

ABSTRACT

We investigate the influence of refractive index contrast on the light scattering properties of nanotextured interfaces, which serve as front contact for p-i-n thin-film silicon solar cells. We here focus on ZnO surfaces with randomly oriented pyramidal features, known for their excellent light trapping performance. Transparent replicas, with a different refractive index, but practically identical morphology compared to their ZnO masters, were fabricated via nanoimprinting. Within the theoretical framework we recently proposed, we show how the angular and spectral dependence of light scattered by nanostructures with identical morphology but different refractive index may be related to each other allowing direct comparison of their light trapping potential within the device.

INTRODUCTION

To further improve conversion efficiencies of thin-film silicon solar cells, efficient light management schemes are crucial, as the absorption coefficient of silicon is small in the near infrared region. The most common approach to improve optical performance is by means of light scattering at randomly textured interfaces.

In the p-i-n superstrate configuration, light trapping is traditionally achieved by scattering at the interface between the transparent front electrode and the absorbing silicon layers by exploiting either the natural, randomly-oriented pyramidal texture of ZnO grown via low-pressure chemical vapor deposition (LP-CVD) [1] or the crater-like texture of sputtered ZnO obtained by wet-etching in HCl [2].

We recently validated the use of replicated nanostructures fabricated via ultraviolet nanoimprinting as front contacts for the p-i-n configuration [3] by demonstrating short-circuit current densities as high as for state-of-the-art nanotextured ZnO master structures, known for their excellent light trapping performance [4, 5].

In this contribution we investigate, both experimentally and theoretically, the influence of refractive index contrast on the scattering behavior of nanotextured interfaces. As the replicated structures exhibit a refractive index n=1.5, which is lower than that of the corresponding ZnO master with n=2, a direct comparison of the measured optical scattering properties is not possible.

We here use a new methodology, we recently presented, to simulate the angular and spectral dependence of light diffusely scattered across nanotextured interfaces [6]. Our method allows us to relate the measured optical scattering data of the master and its corresponding replica via the experimentally determined surface profile and the refractive indices.

EXPERIMENT AND THEORY

Pyramidally-textured boron-doped ZnO layers of thickness 2 µm (called Z2 used for the development of amorphous silicon solar cells) and 5 µm (called Z5 used for the development of microcrystalline silicon solar cells) serving as a master for the replication process were grown via LP-CVD on 0.5 mm thick borosilicate glass. The 5 µm thick sample was subjected to a 20 min plasma treatment to adapt the surface morphology to the growth of microcrystalline silicon layers [4]. The UV nanoimprinting process is described in Ref. [7, 8].

Surface morphologies were measured using atomic force microscopy (AFM) probing an image size of 10x10 μm^2 with a resolution of 512x512 pixels. Angle-resolved scattering (ARS) of light was measured with a detector on a home-built goniometer under normal incidence onto the glass side using a laser at a wavelength of 543 nm. The spectral dependence of the ratio between diffuse to total optical transmittance, called haze, was measured with a photospectrometer equipped with an integrating sphere.

Our theoretical model makes use of a slightly modified Rayleigh-Sommerfeld diffraction integral proposed by Harvey [9] and requires only measured profile data and the refractive indices as input. In this model, light passing across the nanotextured interface, with a peak-to-valley roughness z_0, is assumed to acquire a phaseshift proportional to $z \cdot n_1 + (z_0 - z) \cdot n_2$, where z is the distance from the maximum peak height to the interface travelled in the first medium with refractive index n_1, and $(z_0 - z)$ the distance travelled after the interface in the second medium with refractive index n_2. Since the interface is textured, z depends on the morphology of the surface and is therefore a function of the lateral coordinates x and y. Thus the local phaseshift acquired by a plane wave after crossing the interface is $(n_1 - n_2) \cdot z(x, y) + n_2 \cdot z_0$. Summing up all plane waves exiting the roughness zone, taking into account this phaseshift, we obtain the radiance L in direction cosines space for a given wavelength λ.

$$L(\alpha_k, \beta_l, \lambda) \propto \left| \sum_{i=1}^{N} \sum_{j=1}^{M} e^{2\cdot\pi\cdot i\cdot\frac{n_2}{\lambda}\cdot(\alpha_k\cdot x_i+\beta_l\cdot y_j)} \cdot e^{2\cdot\pi\cdot i\cdot\frac{|n_1-n_2|}{\lambda} z(x_i, y_j)} \right|^2$$

Note that the term $n_2 \cdot z_0$ in the phaseshift leads to a position independent phaseshift and drops out when taking the absolute value for the radiance. The double sum can be handled efficiently using a fast Fourier transform approach. Here α_k and β_l are direction cosines in reciprocal space, related to the wavevector $\vec{k}$ via $\vec{k} = \frac{2\cdot\pi}{\lambda}(\alpha, \beta, \sqrt{1-\alpha^2-\beta^2})$.

For comparison of the angular dependence with experiment it is convenient to transform the radiance from cosine direction space (α, β) into spherical coordinates (ϑ, φ) using

$$\alpha = \sin\vartheta \cdot \cos\varphi$$

$$\beta = \sin\vartheta \cdot \sin\varphi$$

For light scattered at random interfaces, which does not show any azimuthal dependence, the experimental ARS curve for a given wavelength is obtained by integration over all azimuthal angles φ and by taking into account the projection factor $\cos\vartheta = \sqrt{1-\alpha^2-\beta^2}$ [9].

$$ARS(\vartheta,\lambda) = I(\lambda)\int_0^{2\cdot\pi} d\varphi \cdot L(\vartheta,\varphi,\lambda)\cdot\cos(\vartheta)$$

where $I(\lambda)$ is an angle independent factor taking into account the intensity of the incoming light. The spectral dependence of the scattered light as measured by the haze may directly be obtained from the radiance

$$Haze(\lambda) = \left(\sum_{k,l}^{\sqrt{\alpha_k^2+\beta_l^2}\le 1} L(\alpha_k,\beta_l,\lambda) - L(0,0,\lambda)\right) / \sum_{k,l}^{\sqrt{\alpha_k^2+\beta_l^2}\le 1} L(\alpha_k,\beta_l,\lambda)$$

As proposed by Harvey [9], the sum should only be carried out over real propagating modes characterized by $\sqrt{\alpha_k^2+\beta_l^2}\le 1$, as modes outside the unit circle are evanescent. The specular beam is represented by $L(0,0,\lambda)$, whereas experiment includes polar angles up to approximately 7° (for a discussion of this approximation see Ref [6]).

DISCUSSION

Fig. 1 shows AFM images of the master and replicated Z2 and Z5 ZnO surfaces with their pyramidal structure. In order to quantify the quality of the replication process, we compare the local height and angle histogram extracted from the AFM images. Both histograms are practically identical for master and replica indicating the high fidelity of the replication process.

We now focus on the angular dependence of the scattered light. In Fig. 2 the experimental and calculated ARS curves of the master and replicated structures are presented for scattering into air. As input for all calculations we used the AFM images of the masters in Fig. 1, adapting only the refractive index. The calculated curves nicely reproduce the experimental ARS curves. We note however that they deviate slightly at large scattering angles, presumably because we model the optical interface by a random phase-screen with zero thickness which neglects diffraction within the peak-to-valley depth of the rough interface [10]. The oscillations in the calculation are due to the finite AFM image size and may be reduced by either taking larger images or averaging over several images.

As can be seen the replicas generally scatter significantly less light into large angles compared to their masters. From the results of our calculations, we conclude that this is due to the reduced refractive index contrast when scattering from the replica (n=1.5) into air (n=1) compared to scattering from ZnO (n=2) into air. For sample Z2 the change in refractive index leads to a global reduction of the diffuse scattering. The shape of the ARS curve remains approximately the same. This is different for the Z5 sample. In this case, we observe enhanced scattering into smaller angles. This non-trivial behavior, observed in experiment, is well reproduced by the calculation.

In Fig. 3 we show the spectral dependence of the scattered light. The theoretical results for the Haze for scattering into air agree nicely with the experimental data. The reduction in Haze observed in experiment, when switching from the master to the replica, is well reproduced in the calculation and can therefore again be attributed to the change in refractive index.

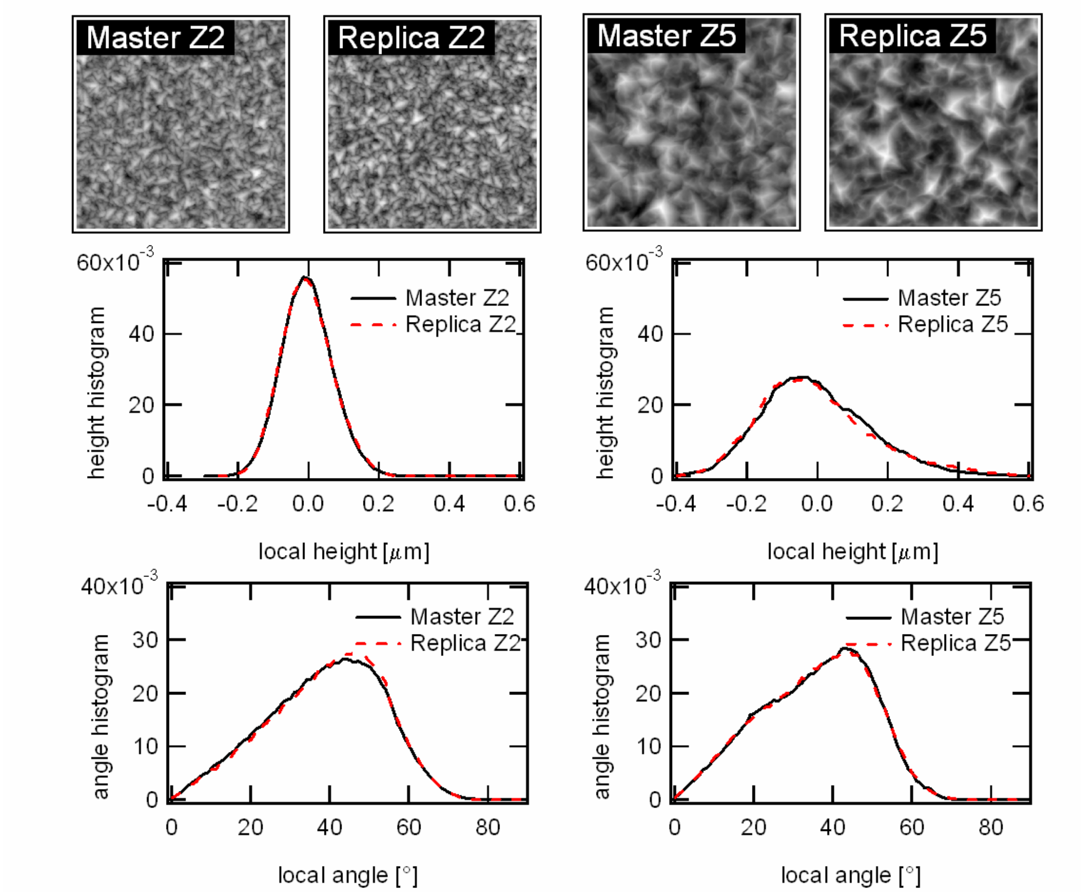

Figure 1: AFM topographies of ZnO masters and their transparent replicas, image size 10x10 μm^2, with the extracted local height and angle histograms.

However, note that once integrated into a solar cell device, scattering takes place into silicon (n=4), resulting in a higher refractive index contrast for scattering from the replica into silicon than from the master into silicon. Although not experimentally accessible, our theory allows determining the scattering behavior into silicon. As can be seen from Fig. 3, the higher refractive index contrast indeed leads to a higher haze value for the replica than the master.

It is important to note that the reflection behavior differs for scattering into air and into silicon. For the former case, total internal reflection may occur, which is not the case for scattering into silicon. In both situations however, the additional randomization produced by those reflections are not taken into account by our model.

At this point, it is interesting to come back to the theoretical expression for the radiance L. As the difference between replica and master only enters into the expression for the radiance through the terms $\frac{n_2}{\lambda}$ and $\frac{|n_1 - n_2|}{\lambda}$, we find that scattering from the replica ($n_1 = 1.5$) into air ($n_2 = 1$) at a certain wavelength λ, yields exactly the same value for the radiance for scattering from the ZnO master ($n_1 = 2$) into silicon ($n_2 = 4$) at the wavelength 4λ (exact for wavelength independent refractive indices). This scaling property of our model is useful in the sense that it allows to determine directly the scattering characteristics of the transmitted fraction of the light, such as ARS and haze at the interface between ZnO and silicon, by measuring the scattering of the replica into air at a quarter of the considered wavelength.

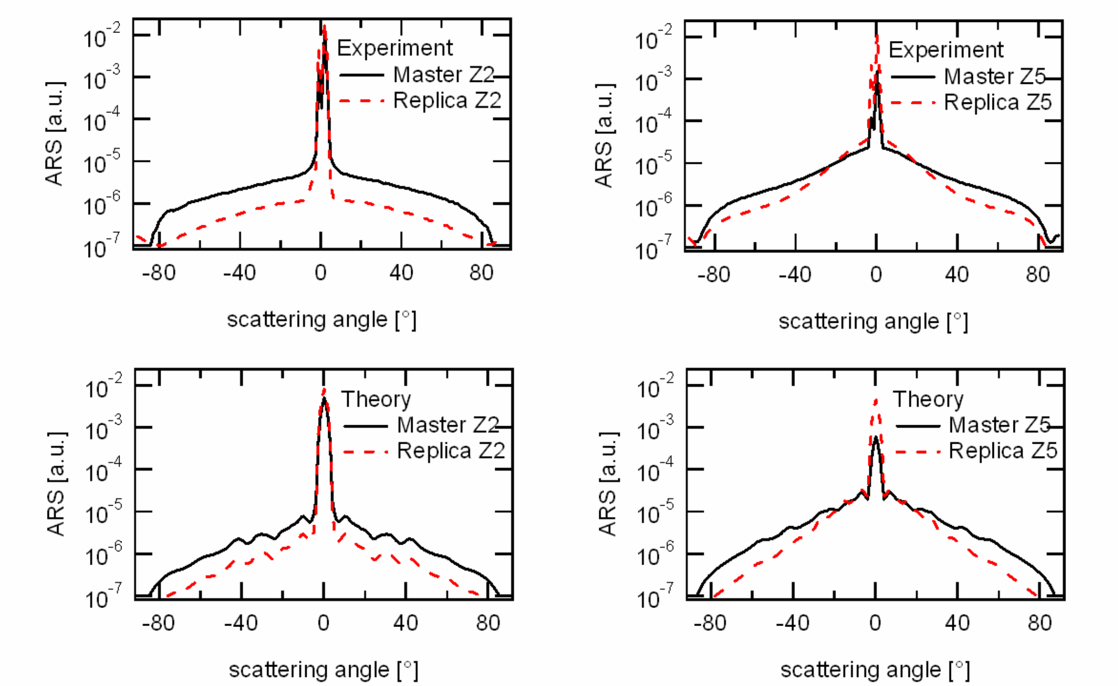

Figure 2: Experimental and calculated ARS curves for the ZnO masters and their corresponding replicas for a wavelength of 543 nm. For all calculations, only the AFM images of the masters in Fig. 1 were used, but the refractive index was adapted to account for scattering from either the ZnO (n=2) or the replica (n=1.5) into air (n=1).

CONCLUSIONS

We investigated the influence of refractive index contrast on the angular and spectral dependence of light diffusely scattered across nanotextured interfaces, which serve as front contact for p-i-n thin-film silicon solar cells. For this we compared the measured and simulated angle-resolved scattering intensity and its spectral dependence (haze) of pyramidally-textured ZnO surfaces and their replicas, exhibiting different refractive indices, but practically identical morphologies. We find a good match between theory and experiment for scattering into air. Our theoretical model also nicely reproduces the non-trivial change in scattering behavior when switching from the master to the replica, which may be attributed to the change in refractive index. Our theory furthermore allows to calculate scattering into silicon and offers us a scaling law, which allows to directly relate the scattering of transmitted light at the replica-air interface to the scattering of transmitted light at the master-silicon interface.

ACKNOWLEDGMENTS

Stimulating discussions with P. Cuony are gratefully acknowledged. The authors acknowledge support by the Swiss Federal Office for Energy (OFEN) under contract 101191 and the Swiss National Science Foundation under grant 200021-12577/1.

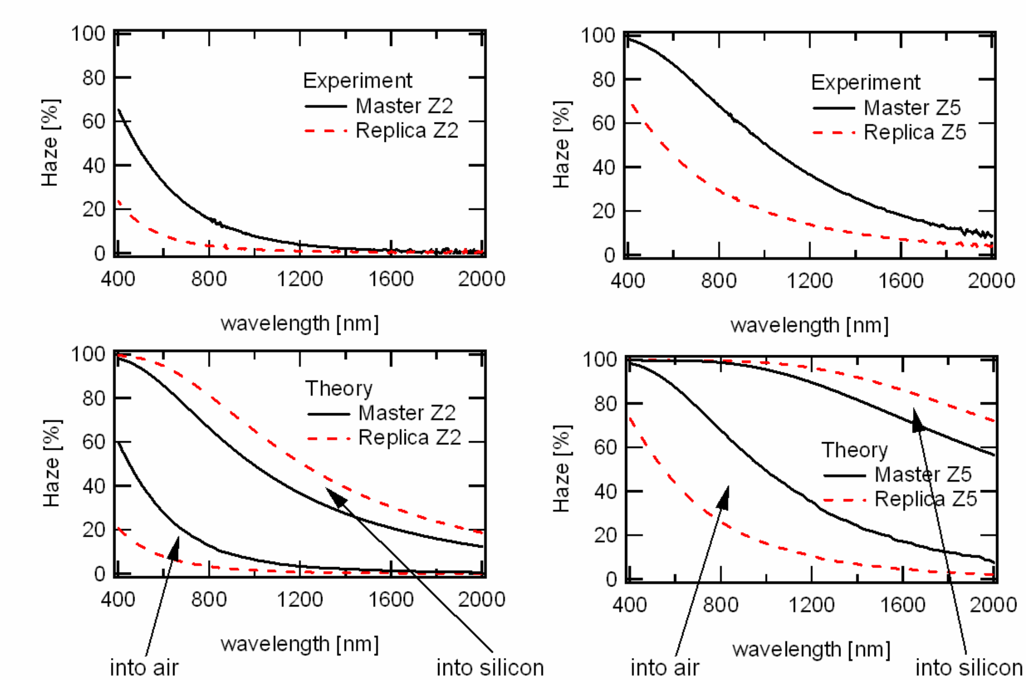

Figure 3: Experimental and calculated Haze curves for the ZnO masters and their corresponding replicas. Theoretical curves for scattering from the ZnO (n=2) and the replica (n=1.5) into air (n=1) and into silicon (n=4) are shown.

REFERENCES

1. S. Faÿ, J. Steinhauser, N. Oliveira, E. Vallat-Sauvain, and C. Ballif, Thin Solid Films 515, 8558 (2007)
2. M. Berginski, J. Hüpkes, W. Reetz, B. Rech, and M. Wuttig, Thin Solid Films 516, 5836 (2008)
3. C. Battaglia, K. Söderström, J. Escarré, F.-J. Haug, D. Dominé, P. Cuony, M. Boccard, G. Bugnon, C. Denizot, M. Despeisse, A. Feltrin, and C. Ballif, Appl. Phys. Lett. 96, 213504 (2010)
4. J. Bailat, D. Dominé, R. Schlüchter, J. Steinhauser, S. Faÿ, F. Freitas, C. Bucher, L. Feitknecht, X. Niquille, T. Tscharner, A. Shah, and C. Ballif, Proc. 4th Conf. Photovoltaic Energy Conversion, 1533 (2006)
5. S. Benagli, D. Borello, E. Vallat-Sauvain, J. Meier, U. Kroll, J. Hoetzel, J. Bailat, J. Steinhauser, M. Marmelo, G. Monteduro, and L. Castens, Proc. 24th European Photovoltaic Solar Energy Conference, 3BO.9.3 (2009)
6. D. Dominé, F.-J. Haug, C. Battaglia, and C. Ballif, J. Appl. Phys. 107, 044504 (2010)
7. J. Escarré, K. Söderström, O. Cubero, F.-J. Haug, and C. Ballif, in *Amorphous and Polycrystalline Thin-Film Silicon Science and Technology – 2010* (Mater. Res. Soc. Symp. Proc. **1245**, Warrendale, PA, 2010), A7.4
8. K. Söderström, J. Escarré, O. Cubero, F.-J. Haug, S. Perregaux, and C. Ballif, accepted for publication in Prog. Photovolt: Res. Appl.
9. J. E. Harvey, C. L. Vernold, A. Krywonos, and P. L. Thomson, Appl. Opt. 38, 6469 (1999)
10. D. Dominé, Ph.D. thesis, Université de Neuchâtel (2009)