Maximum statistical increase of optical absorption in textured semiconductor films

H. W. Deckman, C. B. Roxlo, and E. Yablonovitch

Exxon Research and Engineering Company, Linden, New Jersey 07036

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Complete statistical randomization of the direction of propagation of light trapped in semiconductor films can result in a large absorption enhancement. We have employed a calorimetric technique, photothermal deflection spectroscopy, to monitor the absorption of a-SiH_x films textured by the natural lithography process. The observed enhancement factors, as high as 11.5, are consistent with full internal phase-space randomization of the incoming light.

The idea of trapping light in a semiconductor device by total internal reflection had been proposed[1] as early as 1968. Recently, in a statistical-mechanical analysis[2] of the light-trapping problem, it was shown that the effective absorption of a textured semiconductor film could be enhanced as much as a factor $4n^2$ over that of a plane-parallel sheet, where n is the semiconductor index of refraction. This enhancement factor, which is 50 for crystalline silicon, is a much larger effect than had previously been suspected. This result has had practical implications in the design of a-SiH_x solar cells, in which increased absorption leads to improved efficiency.[3,4]

For the full increase, $4n^2$, to be obtained in an optically thin film, the texture must randomize the direction of light within the medium so as to fill the internal optical phase space. Complete statistical randomization of the internal light is never absolutely ensured, and the $4n^2$ factor must be regarded as an upper limit. In this Letter, we report on a broad class of surface textures that have the property of actually approaching the full measure of light trapping as allowed by statistical mechanics. We have employed a calorimetric technique, photothermal deflection spectroscopy (PDS), to monitor the absorption of a-SiH_x films. These films were microtextured by a new process, which we call natural lithography.[5] By varying the texture size, we experimentally defined the range of texture geometries that permit full statistical light trapping.

It is convenient to use semiconductor films in these studies because their large refractive index n permits a large enhancement effect $4n^2 \approx 50$. In the present study amorphous silicon hydride (a-SiH_x) films were deposited on fused-quartz substrates. The substrates had previously been textured with identical, randomly packed microcolumnar posts, which were produced by the natural lithography process. Therefore the substrate imposed its texture on the film grown upon it. Each sample contained a flat region, which was used as a reference to determine the unenhanced absorption. Texture on the samples was varied by changing the diameter, height, and profile of the post structures.[4,5] Post heights and diameters varied from 0.03 to 0.5 μm and 0.05 to 2 μm, respectively, and the standard deviation of individual post dimensions was less than 10%. The semiconductor films were not conformal with the post structures but were deposited with a step coverage of 0.6–0.8 for 0.7–1.5-μm-thick films. The mean thickness of textured and flat films differed by no more than 10%.

Absorption measurements were performed calorimetrically using PDS.[6] Samples were immersed in CCl_4 and illuminated through the quartz substrate by monochromatic chopped light. The CCl_4 index matches to the fused-quartz substrate. Heat generated in the sample by absorption deflected a He–Ne-laser beam grazing the sample surface. The spectra obtained by measuring this deflection were normalized to 100% absorption at high photon energies, where the sample is totally absorbing. This normalization technique also removes any uncertainty resulting from changes in reflectivity from sample to sample. Reflectivity at the semiconductor–dielectric interface is only 17%; thus wavelength-dependent reflectivity corrections are expected to be small compared with the large enhancement factors.

Figure 1 is a comparison between flat and textured a-SiH_x samples showing that the absorption probability is significantly greater in the textured sample, especially at low photon energies. It should be noted that the spectrum for the flat sample has been corrected to remove interference fringes. This is equivalent[7] to measuring the absorption of the flat sample with a light source that is incoherent. To show that full statistical behavior has been achieved, absorption in the textured sample is compared with a theory that assumes complete internal randomization of light. The absorption expected in textured samples can be calculated by considering multiple reflections of a light ray inside the semiconductor. The enhanced absorption probability F_{en} is then an infinite geometric progression. The Lambertian-angle-averaged path length of a randomized ray is twice the mean thickness l of the film. The sum of the geometric progression is then modeled by

$$F_{en} = \frac{(1 - e^{-2\alpha l})T}{1 - e^{-2\alpha l} + (n_2^2/n_1^2)Te^{-2\alpha l}}. \quad (1)$$

Here l is the mean thickness of the sample, α is the op-

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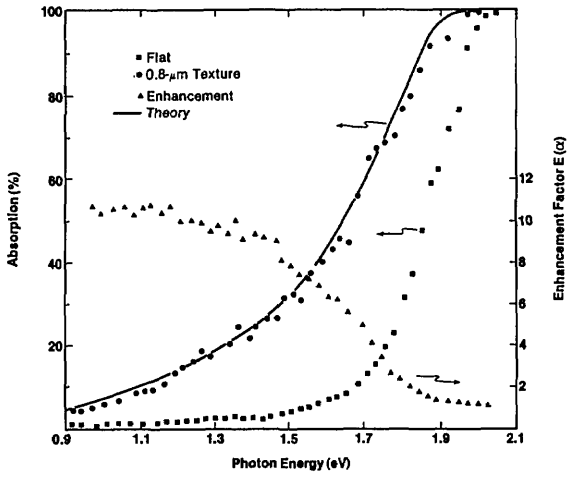

Fig. 1. Comparison of normalized absorption probability for flat and textured 0.85-μm-thick a-SiH$_x$ films. The texture was produced by depositing the film upon a quartz substrate patterned with a densely packed random array of 0.8-μm-diameter, 0.24-μm-high microcolumnar posts. Solid line shows behavior expected if light is completely internally randomized by the texture. The enhancement factor $E(\alpha)$ for the textured films is also shown.

tical absorption coefficient, n_1 and n_2 are the refractive indices of the textured medium and the surrounding dielectric, respectively, and T is the Fresnel transmission coefficient of the semiconductor–dielectric interface. Because fused silica (n = 1.45) and CCl_4 (n = 1.46) have nearly the same index relative[7] to a-SiH$_x$ (n_1 = 3.5) in the spectral region of interest, we can make the approximation n_2 = 1.46 on both sides. The solid line in Fig. 1 shows the enhanced absorption expected from Eq. (1) for complete randomization. Agreement with experiment indicates that essentially complete randomization was achieved.

To compare enhancement in different films, we define an enhancement factor

$$E(\alpha) = F_{\text{exp}}/F_{\text{flat}}, \tag{2}$$

where F_{exp} is the experimentally measured absorption probability in the textured film and

$$F_{\text{flat}} = \frac{T(1 - e^{-\alpha l})}{1 - (1 - T)e^{-\alpha l}} \tag{3}$$

is the absorption probability in an equivalent flat sample.

When the light is completely randomized by the texture, the maximum attainable enhancement factor that applies in the limit of a low absorption coefficient is $2(n_1/n_2)^2$ rather than the $4n^2$ factor described in Ref. 2. This occurs because in our geometry there is no rear reflector, which doubles the escape probability and thus reduces the enhancement factor by 2. In addition, since there is no provision for light trapping in the CCl_4, the index of refraction n_2 of the semiconductor must be referred to the n_1 of CCl_4 rather than to the index 1 of air. The net effect of these two changes is that the maximum enhancement factor in the weak-absorption limit is $2(n_1/n_2)^2$ rather than the full $4n^2$ factor, which would require a rear reflector as well as light trapping in the low-index surrounding medium.

Films with differing optical constants can be compared by plotting the measured enhancement factor [Eq. (2)] as a function of single-pass absorption. Figure 2 shows such a universal plot for 0.8- and 1.1-μm-thick a-SiH$_x$ films textured with 0.8- and 0.5-μm-diameter microcolumnar posts, respectively. The solid line shows the behavior expected if complete internal randomization ($F_{\text{exp}} = F_{\text{en}}$) is achieved. Close agreement of the experimental points with theory indicates that full statistical behavior has been achieved for light trapped in semiconductor films. This result was not achieved with all microstructures, however.

When the texture does not efficiently scatter light, then only a fraction β of the incoming light will be randomized. The randomization fraction β is determined by modeling the experimental absorption F_{exp} as the weighted sum of a randomized portion and a specular portion:

$$F_{\text{exp}} = \beta F_{\text{en}} + (1 - \beta)F_{\text{flat}}. \tag{4}$$

This model [Eq. (4)] is valid as long as the fraction of light scattered by the texture is completely randomized. Statistical ray optics[8] as well as Monte Carlo simulations have shown that there is an overriding tendency for light initially scattered out of the loss cone to be completely randomized. In the low-absorption limit ($\alpha l \ll 1$), Eq. (4) can be rewritten as

$$\beta = \frac{E(0) - 1}{2(n_1^2/n_2^2) - 1}, \tag{5}$$

where $E(0)$ is the enhancement factor for weakly absorbed light. Hence a measurement of the enhancement factor can be directly related to the randomization fraction β. If the randomization is total (β = 1), then $E(0) = 2n_1^2/n_2^2$, as predicted by statistical ray optics.

Equation (5) has been used to define the randomization fraction β for a range of textures in which the post height was held constant. Figure 3 shows the ran-

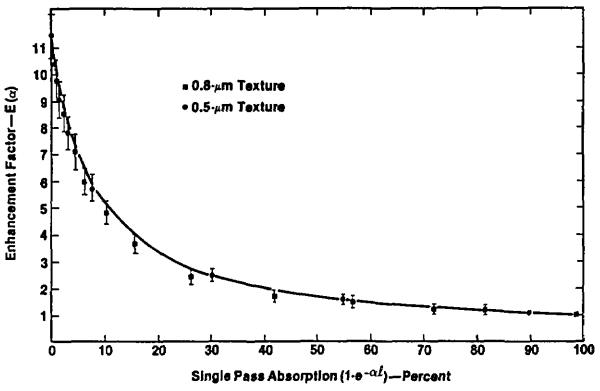

Fig. 2. A plot of enhancement factor $E(\alpha)$ versus single-pass absorption for a-SiH$_x$ films textured with 0.5- and 0.8-μm-diameter microcolumnar posts that were 0.24 μm high. Solid line, which intercepts the ordinate at $2(n_1/n_2)^2$, shows behavior expected if light is completely randomized. This type of graph is a universal curve, which is independent of the material used.

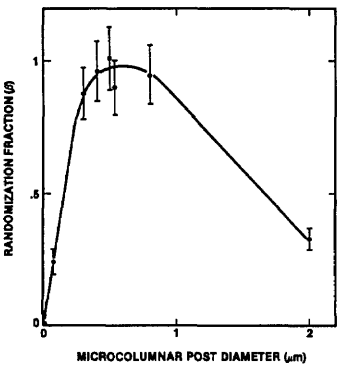

Fig. 3. Dependence of randomization fraction β on microcolumnar post diameter, holding post height constant in the narrow range of 1400–2200 Å. The solid line is simply a smooth curve to guide the eye.

domization fraction β obtained for structure diameters from 0.05 to 2 μm and post heights in the narrow range of 1400–2200 Å. The maximum enhancement factor measured for the textures shown in Fig. 3 was 11.5. Nearly total randomization is obtained when the structure diameter is comparable to a wavelength of light (λ/n_2 = 0.2–0.4 μm), but β decreases for either larger or smaller features. Although we know of no theory that predicts the scattering efficiency from a particular microstructure, this behavior can be understood qualitatively: larger posts have large flat regions on top of and between them, which reflect specularly. On the other hand, features much smaller than a wavelength also happen to be poor scatterers. Dependence of randomization fraction β on etch depth has not been systematically studied, although etch depths less than 1400 Å generally resulted in significantly lower randomization fractions.

In this Letter we have shown that the extent to which light is internally randomized in thin films can be experimentally determined by using PDS. Textures with feature sizes roughly equal to a wavelength of light can be fabricated that completely internally randomize incident light. Absorption enhancement factors up to 11.5 have been observed, in agreement with the predictions of statistical ray optics. These results are important for the optimization of solar cells and other devices for which high levels of absorption are desired in thin films.

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