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Solar Energy Materials & Solar Cells 57 (1999) 1–7

Solar Energy Materials & Solar Cells

Absorption enhancement in ultra-thin textured AlGaAs films

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Abstract

We have studied light randomization and the absorption enhancement in textured ultra-thin $Al_xGa_{1-x}As$ films, with a thickness corresponding to a few optical wavelengths. A correlation between the degree of light randomization and trapping, with the scale length of the texturization geometry was found. The observed absorption enhancement corresponds to 90% of the best possible theoretical value, or 90% light randomization. A modified photon gas model is proposed to calculate the light trapping and absorption at the band edge in the textured thin films.

Keywords: Ultra-thin textured AlGaAs films; Light randomization; Photon gas model

It was proposed in late 1970s and early 1980s that light trapping by total internal reflection could be used to increase light absorption in semiconductor wafers. Several techniques were developed, such as natural lithography [1], metal islands [2] and anodical etch of the porous silicon [3] to texturize thin silicon sheets for light trapping. Yablonovitch [4] showed, that in the low absorption limit, total randomization of the light leads to the enhancement of absorption by the factor of $2n_f^2$, where n_f is film's refractive index. This results were confirmed experimentally by Deckman et al. [5] by applying the natural lithography technique to amorphous silicon films.

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0927-0248/99/$ – See front matter
PII: S0927-0248(98)00160-3

Table 1
Structure of the GaAs/AlGaAs quantum well wafer

GaAlAs	0.32 μm	Window layer
GaAs	0.20 μm	Active layer
GaAlAs	0.44 μm	Window layer
AlAs	0.05 μm	Sacrificial layer
GaAs	>100 μm	Substrate

However, at the moment there is no theory for angular dependence of light scattering from surfaces produced by this process because perturbation methods require that the ratio of roughness height to the wavelength be small [6] while the quasi-classical small slope approximation [7] require a relatively smooth surface.

In this paper we report results of our study of light randomization and absorption enhancement in ultra-thin GaAs/AlGaAs films with a thickness of only a few optical wavelength. The ultra-thin films were textured using the natural lithography while varying the density of cylindrical surface structures. A modified photon gas model, which successfully describes the absorption at the band edge will be presented.

The sample preparation method is by natural lithography: A GaAs/AlGaAs double hetero-structure wafer (see Table 1) is patterned with commercially available carboxylate modified 0.95 μm polystyrene spheres. The sphere solution is first diluted with methanol to 1% concentration by weight and then surface deposited by dropping a small quantity of a solution on the wafer and spinning the wafer at 1500–2000 rpm. The wafer is spun to distribute the spheres across the surface, and to allow the methanol to evaporate. The sphere solution concentration and the revolution speed were varied until the conditions were found such that approximately 50% of the wafer area is covered by spheres. From Fig. 1a–d, the effect of varying the angular velocity (while holding constant the sphere concentration in solution) on sphere areal density can be seen. The resultant sphere density distribution does not vary significantly across the surface of a given sample. The key point is to avoid building up multiple layers of spheres, which totally coat the surface and provide no patterning.

When the desired sphere distribution is obtained, the sample is etched using the chemically assisted ion beam etching process to transfer a pattern, using the spheres as a lithographic mask. In our work the transferred pattern consists of 0.25 μm high mesas, which is approximately $\frac{3}{4}$ of the thickness of the top AlGaAs layer. Following etching, the top three epitaxial layers (~ 1 μm) of the wafer, containing the active layer of the device, are removed from the substrate using the epitaxial lift-off procedure (ELO) [8]. The sample is bonded to a glass slide with the untextured side against the glass by using a UV curable polyurethane adhesive. The spectral reflectance, $R(\lambda)$, of the textured and untextured film was measured over a white surface using a standard integrating sphere setup. Then, absorbance $A(\lambda)$of the sample simply becomes

$$A(\lambda) = 1 - R(\lambda), \tag{1}$$

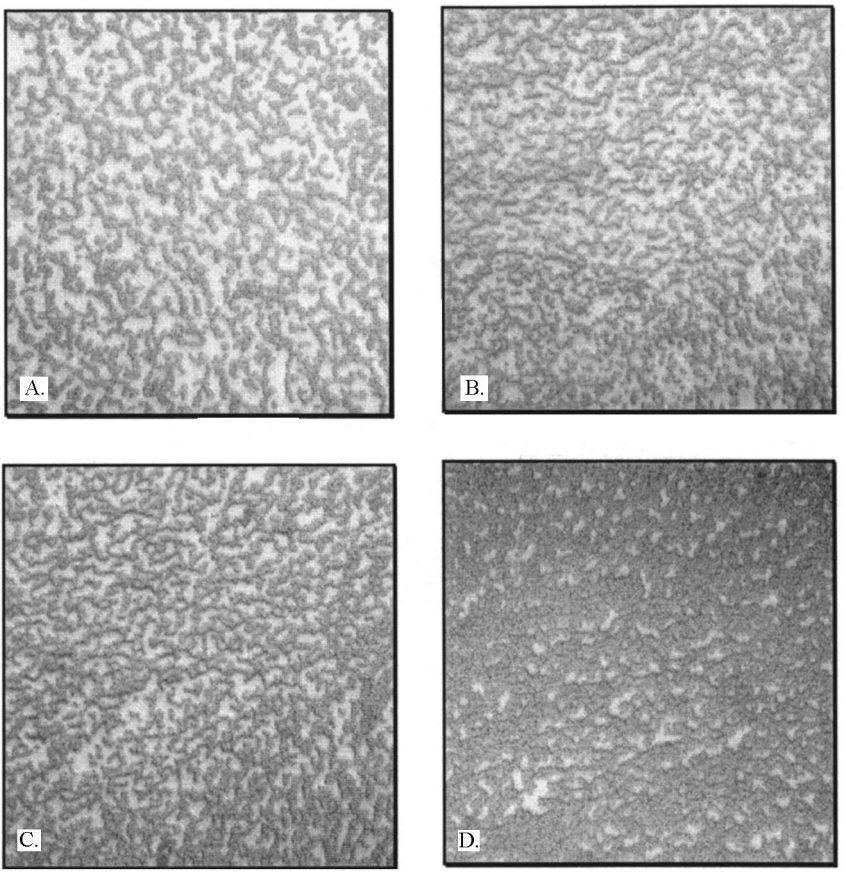

Fig. 1. Variation of sphere density with angular velocity. The concentration is 30 drop of sphere solution in 1.5 ml of methanol. The darker area represents spheres. The spheres tend to cluster at higher surface concentrations: (a) sphere distribution for 1800 rpm; (b) sphere distribution for 1700 rpm; (c) sphere distribution for 1600 rpm; (d) sphere distribution for 1550 rpm.

since there is no transmission outside of the sphere. Samples were held horizontally by gravity so that no optically absorbing adhesive materials were necessary inside the sphere. In all our measurements, the probe beam was incident on the glass side with the semiconductor film on the rear, as shown in Fig. 2. In this configuration specular reflections from the glass–air and the semiconductor/glass interfaces were identical for the textured and untextured samples. Thus, changes in reflectivity at the front surface were not a concern, since the texturing was at the rear surface.

In all the textured samples, an increase in absorption was measured in comparison with the untextured films (see Figs. 3 and 4). The maximum theoretical absorption, which can be attained is $\approx 80\%$ due to incident beam reflectivity from the glass/air (4%) and semiconductor/glass (16%) interfaces. The best results were attained for

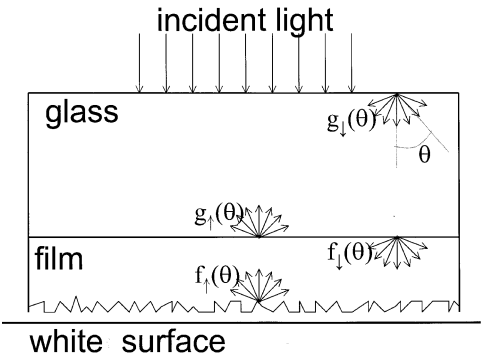

Fig. 2. Experimental configuration and definition of terms used in the modeling.

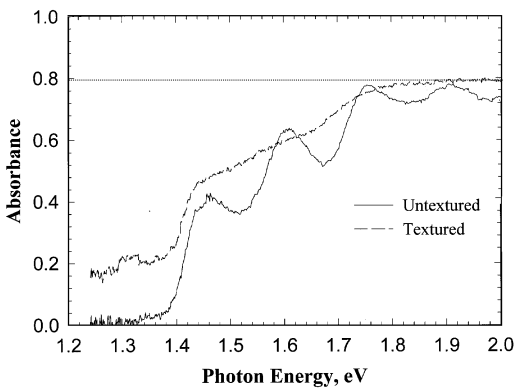

Fig. 3. Sample with the sparse distribution of spheres. The absorption enhancement is minimal.

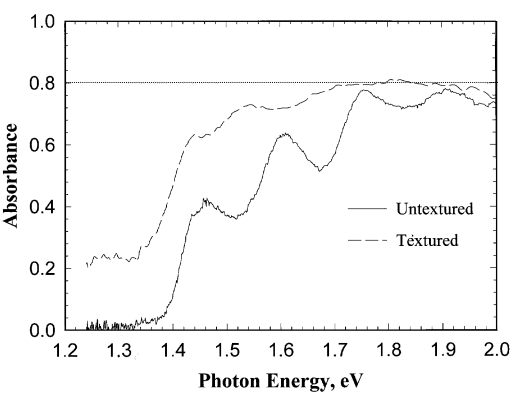

Fig. 4. Sample with 50% of the surface area covered with spheres. A large absorption enhancement is obtained near the band edge.

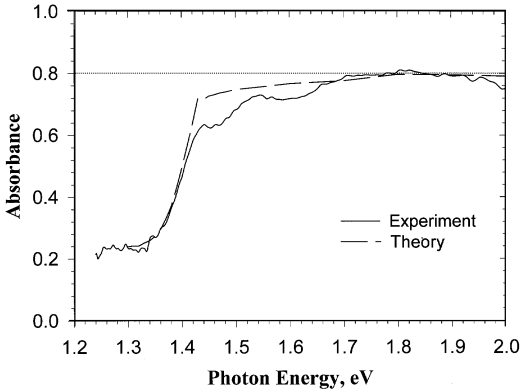

Fig. 5. Comparison between the maximum theoretical absorption for a textured film and the values obtained experimentally.

samples, which were coated by approximately 50% area coverage of polystyrene spheres and the corresponding 0.25 μm high mesas. An absorbance increase from 45% up to $\approx 75\%$ of a maximally achievable result occurs for the sample in Fig. 4 near the band edge. In our best samples, the experimental value nearly reaches the maximum absorbance predicted by theory, as can be seen in Fig. 5. The absorbance oscillations, which occur for photon energies above the bandgap, are due to Fabry–Perot fringes. The damping of these Fabry–Perot oscillations in the textured film is an additional evidence of light randomization produced through surface texturization. At energies below the band gap, free carrier absorption is enhanced from $< 1\%$ up to the level of $\approx 20\%$. Unfortunately, this produces heat rather then electron–hole pairs.

We now describe the photon gas model:

Consider a system consisting of the semiconductor film of thickness d with absorption coefficient α and refractive index n_f attached to a glass slide with the index of refraction n_g (as in Fig. 2). One surface of the film (opposite to the glass) is textured and lies adjacent to the white reflecting surface so that incident monochromatic light enters the film after passing through the glass slide. We assume that all photons which reach the textured surface and white backing are scattered in all upward directions with distribution function $S(\theta)$, where θ is the polar angle. If scattering is perfectly Lambertian, $S(\theta) \propto \cos\theta$. Further, let us call $f_\uparrow(\theta)$ the flux density of photons in the film near the textured surface, which are traveling up at angle θ. Due to symmetry, $f_\uparrow$ depends only on θ. Also, let $f_\downarrow(\theta)$ be a flux of photons in the film near the film/glass interface, which are traveling down at angle θ. In the same manner, $g_\uparrow(\chi)$ and $g_\downarrow(\chi)$ are defined as the photon flux densities in the glass at the film/glass interface going up and at the glass/air interface going down, respectively (Fig. 2). Reflection at the glass/air boundary gives a relation between $g_\uparrow(\chi)$ and $g_\downarrow(\chi)$:

$$g_\downarrow(\chi) = R_{ag}(\chi) g_\uparrow(\chi), \tag{2}$$

where $R_{ag}(\chi)$ is the reflection coefficient at the glass/air boundary given by the Fresnel formulae. Detailed balance at the glass/film interfaces results in the following relation:

$$g_{\uparrow}(\chi) = f_{\uparrow}(\theta)\exp(-\alpha d/\cos\theta)T_{fg}(\theta) + g_{\downarrow}(\chi)R_{fg}(\theta), \tag{3}$$

where χ and θ are related by Snell's law. This formula simply states that flux aimed upward in the glass is formed by reflection of photons in the glass from the film/glass boundary and by transmission of the film photon flux, attenuated in the absorbing film, through the same boundary. The same reasoning provides another relation for $f_{\downarrow}(\theta)$ in the semiconductor film:

$$f_{\downarrow}(\theta) = f_{\uparrow}(\theta)\exp(-\alpha d/\cos\theta)R_{fg}(\theta) + g_{\downarrow}(\chi)T_{fg}(\theta). \tag{4}$$

Also, the upward photon flux at the bottom surface is formed by the scattering of incoming flux and the downward flux of photons on the textured surface. If a monochromatic photon flux of intensity I is incident on the glass surface, to a good approximation $IT_{ag}(0)T_{fg}(0)(1 + R_{fg}(0)R_{ag}(0))$ is transmitted into the semiconductor, and photon flux that reaches the textured surface has intensity

$$I_{tx} = IT_{ag}(0)T_{fg}(0)(1 + R_{fg}(0)R_{ag}(0))\exp(-\alpha d/\cos\theta). \tag{5}$$

Since there is no absorption on the bottom surface, the incoming flux must totally balance the outgoing flux. This gives the final equation for the system:

$$\int f_{\uparrow}(\theta)\cos\theta \mathrm{d}\Omega = I_{tx} + \int f_{\downarrow}(\theta)\exp(-\alpha d/\cos\theta)\cos\theta \mathrm{d}\Omega, \tag{6}$$

where integration is performed over 2π steradian. These equations can be solved for any assumed angular distribution of light scattering $f_{\uparrow}(\theta) = S(\theta)$ by integration of Eq. (6) over the solid angles. In the photon gas model of Deckman et al. [5] an average Lambertian length for absorption is used. This is not valid for the strongly nonlinear dependence of absorption on the propagation angle. Our theory, being more general, explicitly takes into account absorption of photons scattered at different angles. Instead of assumption of totally randomized photons, the model can deal with any angular dependence of scattering, such as Lambertian in this paper. It can also accommodate a dependence on incoming beam incidence angle. Instead of treating the freestanding film, the sample is treated as bonded on the glass or sapphire slide, thus taking into account a practical problem of the light trapping in the supporting slide.

Given this photon gas model, the amounts of reflected and absorbed light can be calculated. Knowing the absorption spectrum of the film and its dispersion, one can obtain the full spectral dependence of absorbance of the textured semiconductor film bonded to a glass slide. Since the samples we studied contained various $Al_xGa_{1-x}As$ alloy layers, the absorption coefficient α and the refractive index n_f in the above formulae are average values weighted by the thickness of the corresponding layers. The calculated absorbance dependence for perfect Lambertian texturing is presented in Fig. 5 along with the experimental curve. Two curves show good agreement leading us to believe that in the samples which have approximately 50% of the surface area

covered by 0.25 μm high mesas at least 90% perfect light randomization has been achieved.

We have studied light randomization and absorption in AlGaAs films textured by means of natural lithography. A reproducible thin film fabrication process was developed that provides 90% of ideally predicted band-edge absorption relative to our theoretical model. Since the 1 μm diameter spheres are visible in the optical microscope, natural lithography with this sphere size is easy to monitor and optimize. Our technique is applicable for fabrication of thin film solar cells and LED's based on III–V compounds. Epitaxial lift-off technology allows us to fabricate very thin AlGaAs solar cells [9], making them lighter and cheaper and providing higher operating point voltages.

References

[1] Deckman, J.N. Dunsmuir, Appl. Phys. Lett. 41 (1982) 377.
[2] H. Stewart, D. Hall, Appl. Phys. Lett. 69 (1996) 2327.
[3] A. Krotcus, V. Pacebutas, J. Kavaliauskas, I. Subacius, K. Grigoras, I. Simkiene, Appl. Phys. A 64 (1997) 357.
[4] Eli Yablonovitch, J. Opt. Soc. Am. 72 (1982) 899.
[5] H.W. Deckman, C.R. Wronsky, H. Witzke, E. Yablonovitch, Appl. Phys. Lett. 42 (1982) 968.
[6] P.M. van den Berg, T.J. Fokkema, J. Opt. Soc. Am. 69 (1979) 27.
[7] A.G. Voronovitch, Sov. Phys. JETP 62 (1985) 65.
[8] E. Yablonovitch, R. Gmitter, J.P. Haribson, R. Bhat, Appl. Phys. Lett. 51 (1987) 2222.
[9] X.Y. Lee, A.K. Verma, C.Q. Wu, M. Goertemiller, E. Yablonovitch, Thin film GaAs solar cells on glass substrates by epitaxial lift-off, Proc. 25th PVSC, 13–17 May 1996, Washington DC, pp. 53–55.