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Correlation of Light-induced Enhancement of Open-Circuit Voltage and Structural Change of Heterogeneous Silicon Solar Cells

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ABSTRACT

We observe a significant light-induced increase in the open-circuit voltage, V_{oc}, of thin-film silicon solar cells whose intrinsic (*i*) layer consists of an amorphous and microcrystalline mixed phase. The increase depends on the *i*-layer thickness, the *i*-layer deposition temperature, the initial V_{oc} values, and the light-soaking intensity. An increase of as large as 150 mV is observed. The original V_{oc} is restored after subsequent thermal annealing. *In-situ* photoluminescence (PL) spectroscopy is used to investigate this metastable phenomenon. We find that the PL intensity and peak-energy position associated with the amorphous component of the heterogeneous material increase upon light soaking, suggesting a structural change. We propose that a reduction of microcrystalline volume fraction or size is responsible for the V_{oc} enhancement.

INTRODUCTION

Hydrogenated amorphous silicon (a-Si:H) alloy materials and solar cells near the onset of microcrystallinity have received a great deal of attention due to their superior film properties and solar cell performance [1]. The effect of hydrogen dilution during film growth was first reported by Guha et al. in 1981 [2]. The technique has now been widely used to reach the amorphous-to-microcrystalline transition [3-10]. We have previously shown that the onset of the transition depends on deposition conditions such as hydrogen-dilution ratio and film thickness [7]. The transition region is easily identified by monitoring the open-circuit voltage (V_{oc}) of the solar cell. As one approaches the threshold of transition from the amorphous region, V_{oc} increases slightly, reaches a maximum, then drops down sharply into the transition region. The sudden drop in V_{oc} is also accompanied by a large dispersion in the V_{oc} values. As the dilution ratio or film thickness is further increased, V_{oc} eventually converges to a typical microcrystalline silicon value of ~0.5 V.

In this study, we report results on a series of light-soaking experiments on solar cells prepared in the amorphous-to-microcrystalline transition region. The solar cells exhibit initial V_{oc}'s ranging from greater than 1 V to less than 0.5 V, encompassing the amorphous, the mixed-phase, and the microcrystalline silicon intrinsic (*i*) layers. We find that after one-sun light soaking, cells in the amorphous and microcrystalline regions show a small reduction in V_{oc}, while those in the mixed phase display significant V_{oc} enhancement [11]. An increase of nearly 140 mV after one-sun illumination at 50 °C is observed for the most heterogeneous cells. We also find that the change in V_{oc} (ΔV_{oc}) depends on the *i*-layer thickness, the *i*-layer deposition temperature, the initial V_{oc}, and the light-soaking intensity. Subsequent thermal annealing at 150 °C for 2 hours substantially restored the original V_{oc} values.

We have previously demonstrated that photoluminescence (PL) spectroscopy is a sensitive tool for studying structural and electronic properties of solar cells near the amorphous-to-microcrystalline transition [12]. In this study, we again employ PL to investigate this metastable phenomenon. We carry out *in-situ* light soaking on heterogeneous cells and analyze the accompanying PL spectra. We find that both the PL intensity and the peak-energy position associated with the amorphous component increase after light soaking. This indicates that there exists a correlation between the enhancement in V_{oc} and a structural change. We suggest that the size or volume fraction of the microcrystallites in the heterogeneous cells shrinks during light soaking, resulting in a larger ratio of amorphous-to-microcrystalline volume fraction.

EXPERIMENTAL RESULTS AND DISCUSSION

a-Si:H *n i p* solar cells were deposited on stainless steel substrates using a radio-frequency glow-discharge technique. Indium-tin-oxide (ITO) dots of 0.05 cm^2 and 0.25 cm^2 were deposited on the *p* layer as the top contact. Each substrate typically contains over 30 ITO dots on a 4 cm x 4 cm area. The *i* layer was deposited using various hydrogen-dilution ratios to produce solar cells of amorphous, heterogeneous, and microcrystalline phases. Current density versus voltage (*J-V*) characteristics were measured at 25 °C under AM1.5 illumination.

Figure 1 plots the dependence of initial V_{oc} on hydrogen dilution for cells with an *i*-layer thickness of ~5000 Å, where R refers to the standard dilution we use in our laboratory for obtaining the highest efficiency solar cells [13]. As the dilution is increased and approaches R = 1, V_{oc} shows a small increase, due to a slight widening of the optical bandgap and improvement of intermediate-range order [7]. As the dilution is further increased to R > 1, V_{oc} drops sharply and shows a large degree of dispersion, reflecting the heterogeneous nature of the material. The lowering of V_{oc} can be correlated to an increase of volume fraction and size of low-bandgap microcrystallites that are embedded in a high-bandgap amorphous matrix. As the dilution approaches R = 2, the dispersion diminishes and converges to a V_{oc} value of ~0.45 V, indicating a substantial microcrystalline inclusion.

It is interesting to point out that in the transition region, the dispersion in V_{oc} is so large for a given dilution that it is possible to obtain solar cells containing all three phases on the same substrate. In Fig. 2 (a), we plot the initial V_{oc} value versus solar cell location on the 4 cm x 4 cm substrate for such a case. One can see that at the center of the

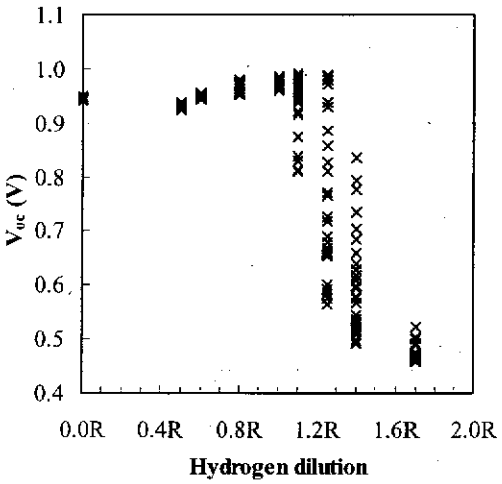

Figure 1. Dependence of initial V_{oc} on hydrogen dilution ratios for *i*-layer thickness of ~5000Å.

substrate, V_{oc} is the highest and exceeds 1 V, indicating the amorphous phase. At the four corners, V_{oc} is the lowest and is around 0.5 V, indicating the microcrystalline phase. Most of the cells between the center and the corners have V_{oc} values between 0.5 V and 1 V, reflecting the heterogeneous phase. We then light soaked the entire sample under one-sun intensity at 50 °C for 20 hours and measured the V_{oc} again. The results are plotted in Fig. 2(b). One can see that the V_{oc} for the cells at the center and the four corners decreases by a small amount, while that of the heterogeneous cells increases significantly. The light-induced effect is displayed in Figs. 3 and 4, where one can easily see the dependence of ΔV_{oc} on the initial V_{oc} values.

From Fig. 4, one can make the following observations. For the cells in the substantially microcrystalline ($V_{oc} \leq 0.55$ V) or amorphous ($V_{oc} \geq 1$ V) regions, light soaking causes a small reduction in V_{oc}. For the cells with initial V_{oc} slightly higher than 0.5 V or slightly lower than 1 V, the increase of V_{oc} is small. For the heterogeneous cells, light soaking gives rise to an increase in V_{oc} of different magnitudes. The largest increase has a magnitude of ~140 mV and occurs around 0.8 V, nearly halfway between 0.55 V and 1 V. All of the V_{oc} changes due to light soaking are substantially restored upon subsequent annealing at 150 °C for 2 hours, also shown in Fig. 4. Annealing of the cells prior to light

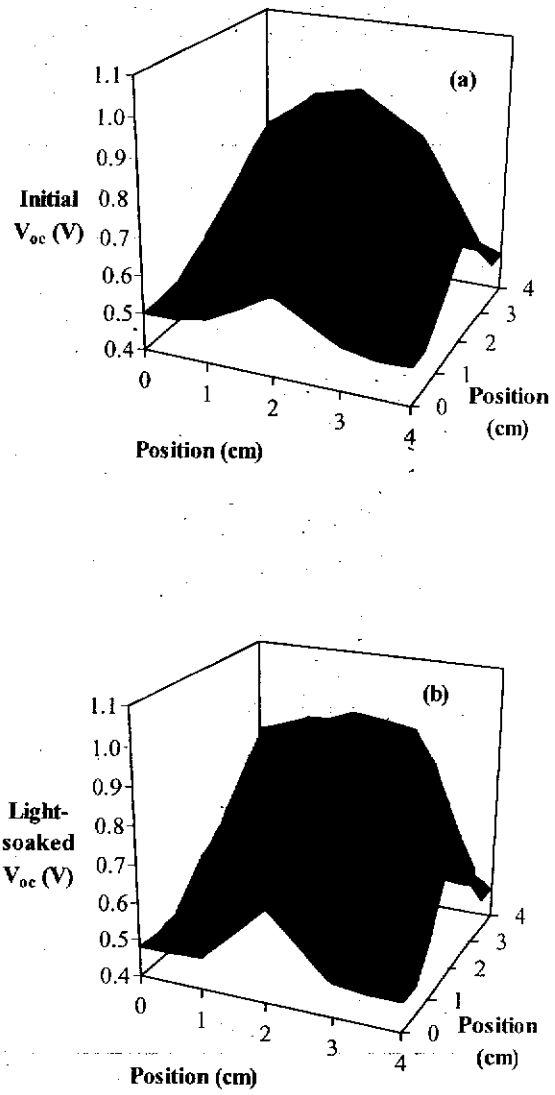

Figure 2. (a) Initial and (b) light-soaked V_{oc} as a function of solar cell position.

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soaking has little effect on the initial V_{oc}.

We have also studied the *i*-layer thickness dependence of the light-induced effect. In Fig. 5 we plot ΔV_{oc} versus initial V_{oc} for cells with *i*-layer thicknesses of 5000 Å and 1 μm. The light soaking was carried out at 50 °C for 150 hours, but the majority of the ΔV_{oc} occurs in the first 20 hours. It is clear that the V_{oc} increase is more pronounced for the thicker cell. In addition, we have light soaked the 5000 Å cell with an intense 30-sun light for 2 hours, and a maximum increase of 150 mV of the original V_{oc} is observed near V_{oc} = 0.7 V. The higher intensity apparently produces a larger ΔV_{oc}, as shown in Fig. 5.

Because the initial V_{oc} of a solar cell depends on the bandgap of the *i* layer, we have also investigated the effect of the *i*-layer deposition temperature on ΔV_{oc}. We used the same hydrogen-dilution ratio but different *i*-layer temperatures of 175 °C, 200 °C, 250 °C, 300 °C, and 325 °C to deposit cells with the same *i*-layer thickness of 5000 Å. The dependence of ΔV_{oc} on the initial V_{oc} after one-sun light soaking for 20 hours is shown in Fig. 6. We notice that the initial V_{oc} values of the lower-temperature solar cells are higher than those of the higher-temperature ones because of the wider bandgap of the *i* layer. In fact, for the 325 °C cells, the highest initial V_{oc} is only 0.833 V. Furthermore, the maximum ΔV_{oc} values for the 175 °C and 325 °C

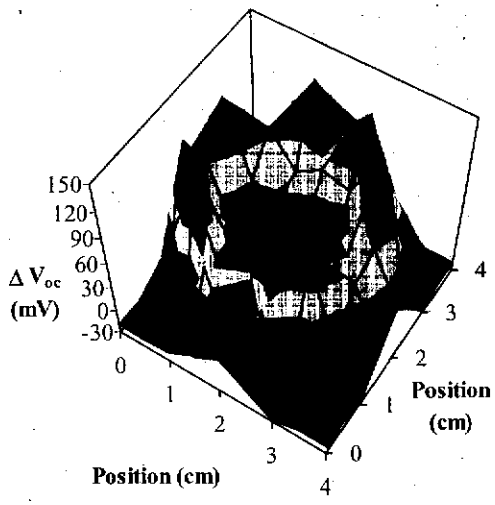

Figure 3. ΔV_{oc} as a function of solar cell position.

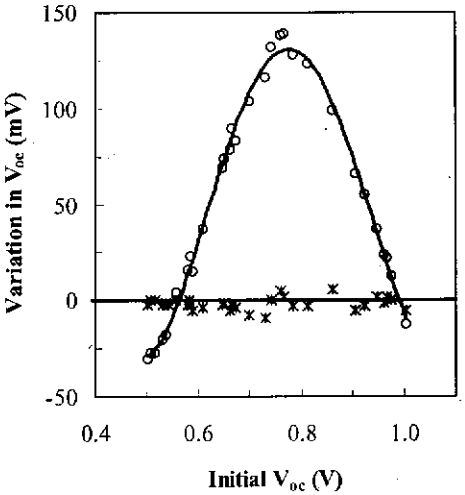

Figure 4. ΔV_{oc} as a function of the initial V_{oc} for ~5000 Å thick cells after one-sun light soaking for 20 hrs. (O). The line is a fit to the data and serves as a guide to the eye. Subsequent annealing at 150 °C for 2 hrs. (∗) substantially restored the original V_{oc} values.

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cells are less than that of the 250 °C cells. In fact, as shown in Fig. 7, the 250 °C sample appears to be the most susceptible to change and gives rise to the largest ΔV_{oc} for the hydrogen-dilution ratio used.

In addition, we have studied the dark *J-V* characteristics of the cells with different phases before and after light soaking. Figure 8 (a) shows the dark *J-V* for a cell in the amorphous region with an initial V_{oc} = 1.030 V. A straight line representing a typical diode characterized by $J = J_o exp(qV/nkT)$ is observed. After light soaking, the quality factor n is increased from 1.80 to 1.84 and the saturated current density J_o from 7.5×10^{-13} A/cm^2 to 2.8×10^{-12} A/cm^2, consistent with an increase in the defect density and recombination events due to the Staebler-Wronski effect (SWE) [14]. For cells in the microcrystalline region, the dark *J-V* characteristics (not shown) exhibits high currents throughout the voltage range measured and simply shifts to even higher currents after light soaking. For cells in the mixed phase that show a large increase of V_{oc} after light soaking, the initial dark *J-V* exhibits a two-segment feature as shown in Fig. 8 (b) for a cell with an initial V_{oc} = 0.843 V. After light soaking, the dark current in the low-voltage (V < 0.4 V) segment increases, similar to the amorphous case of Fig. 8 (a), while the current in the high-voltage (V > 0.4 V) segment is suppressed, as if the resistance of the material has increased or the barrier height of the diode has become larger [15].

Based on the above experimental data, we now discuss possible causes

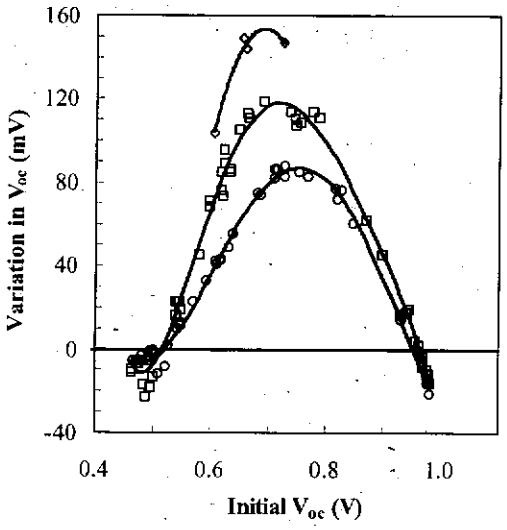

Figure 5. ΔV_{oc} after one-sun light soaking for 150 hrs. as a function of the initial V_{oc} for cells with an *i*-layer thickness of ~5000 Å (○) and 1 μm (□). ◇ denotes ΔV_{oc} produced by a 30-sun light soaking for 2 hrs. on a ~5000 Å cell.

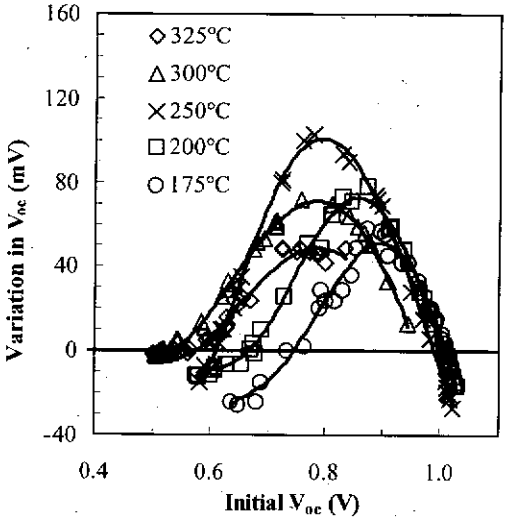

Figure 6. ΔV_{oc} as a function of the initial V_{oc} for ~5000 Å thick cells deposited at five substrate temperatures.

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for the V_{oc} increase. We propose that the increase is associated with a reduction in the microcrystalline volume fraction or size. We suggest that the material in the mixed phase is more susceptible to change; the volume fraction or the size of the microcrystallites in this material could change easily as is apparent from the scatter in the V_{oc} values, even in solar cells on the same 4 cm x 4 cm substrate area. When the volume fraction or size of the microcrystallites is very small ($V_{oc} \geq$ 1 V) or very large ($V_{oc} \leq 0.5$ V), the carrier transport and cell performance are dominated by their respective amorphous or microcrystalline properties. Only when the microcrystallinity is in the transition region does the change in the microstructure affect the V_{oc} significantly. This explains the observed dependence of the V_{oc} increase with respect to the initial V_{oc}.

We now discuss the results of PL spectroscopy. We have previously shown that PL is a sensitive tool capable of revealing important information on structural and electronic properties of solar cells near the transition region [12]. Here we employ PL spectroscopy to further investigate whether the light induced ΔV_{oc} is correlated with any structural change. PL was excited by using a 100 mW/cm^2 632.8 nm laser beam, and the sample was mounted on a cold stage. The PL spectra were obtained using a grating monochromator equipped with a liquid-nitrogen cooled Ge detector.

We first measure PL spectra at various locations of a heterogeneous sample at 80 K. Figure 9 plots

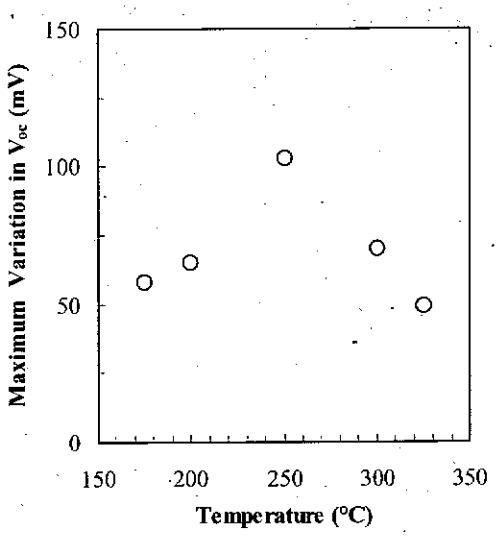


Figure 7. Maximum ΔV_{oc} in figure 6 as a function of the *i*-layer deposition temperature.

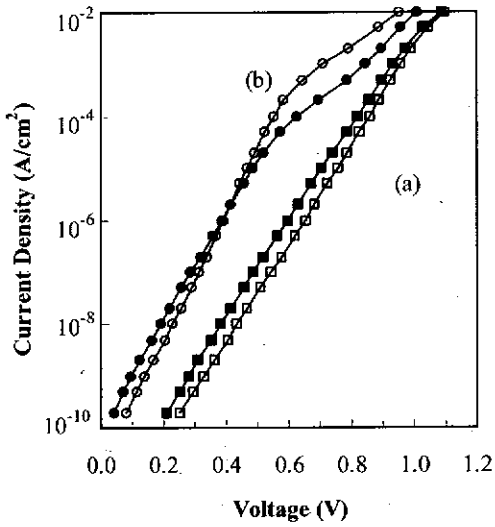


Figure 8. Dark *J-V* characteristic of solar cells in the (a) annealed (□)and light-soaked (■) states in the amorphous phase, and (b) annealed (○) and light-soaked (●) states in the mixed phase.

relative PL intensity versus photon energy for five different locations corresponding to five different initial V_{oc} values. It is readily observed that for the area with $V_{oc} = 0.506$ V where the material contains more microcrystallites, the PL peak occurs at a lower photon energy and has a lower intensity, consistent with our earlier findings [12]. As the V_{oc} increases, both the intensity and the energy of the PL peak associated with the amorphous component increase. In fact, for the $V_{oc} = 0.939$ V material, which is nearly entirely amorphous, the PL signal exhibits the highest intensity and peaks at the highest energy. The PL intensity and peak position for the five measurements are shown in Fig. 10. This establishes the correlation between the initial V_{oc} value, which reflects the heterogeneity, and the PL signal, which reflects the structural and electronic properties of the material.

We next investigate the effect of light soaking on the PL spectrum. In order to avoid any ambiguity that might be introduced by removing and replacing the sample, we carried out an *in-situ* experiment in that the sample and the optical system were fixed in place, and the V_{oc} was monitored as well. We first measured the PL spectrum at an area having $V_{oc} = 0.860$ V. The sample was then allowed to warm up to room temperature for light soaking without any movement. We next light soaked the sample with a 1 W / cm^2 red light for 0.5 hour and measured the PL spectrum again at 80 K. Indeed, the PL spectrum showed an enhanced intensity and shifted to a higher energy, as shown in Fig. 11. An additional 2.5-hour light soaking produced further enhancement, also shown in Fig. 11.

From Fig. 9, we have established that higher PL intensity and peak-energy position correlate with higher V_{oc}, which is associated with a more amorphous structure. The shift in the PL spectrum towards higher intensity and peak energy is consistent with the observed increase in V_{oc} after light soaking. We therefore conclude that the light-induced enhancement in V_{oc} is associated with a structural change based on PL spectroscopy.

Light exposure has been shown to cause structural changes in the material, and the reduction in the volume fraction or size of the microcrystallites could result from this structural change. Moreover, recent studies on the light-induced volume expansion [16, 17] show a larger effect on materials near the amorphous-to-microcrystalline transition region than in the regular amorphous region. This also supports the view that the material in the transition region is less stable. In addition, Baugh and Han [18] have recently proposed a model of metastable changes in a two-domain

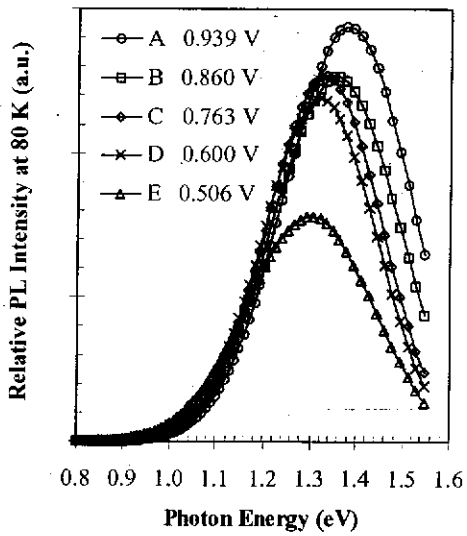


Figure 9. Relative PL intensity at 80 K as function of the initial V_{oc} for five V_{oc} values.

amorphous-silicon network; they calculated that a small movement of boundaries between high- and low-ordered domains could cause a large volume expansion. These studies are consistent with our observations. The notion that the change in V_{oc} is caused by the changes in the microstructure is also consistent with the dark *J-V* characteristics of Fig. 8 (b). The fact that the high-voltage segment shows a reduced current after light soaking can be explained as resulting from a reduction in the microcrystalline volume fraction.

Although it is reasonable to argue that a change in the volume fraction or size of the microcrystallites can cause the observed V_{oc} variation, the process of the microstructure change is not well understood. Kaiser et al. [19] found that a hydrogen-plasma treatment induces crystallization of a-Si:H but reduces the crystalline volume fractions for microcrystalline silicon. Grain boundaries can produce a significant effect in the hydrogen-induced conversion of crystallites to amorphous silicon, a process that reduces the strained Si-Si bonds. Nuclear magnetic resonance and infrared spectroscopy confirm that in an amorphous matrix containing crystallites, most hydrogen clusters in the grain boundaries [20, 21]. It is reasonable to assume that the hydrogen content in the crystallites is much lower than the grain boundaries. There exist many strained (or weak) Si-Si bonds in crystallites near the grain boundaries. During light soaking, some strained Si-Si bonds are broken; hydrogen moves in and

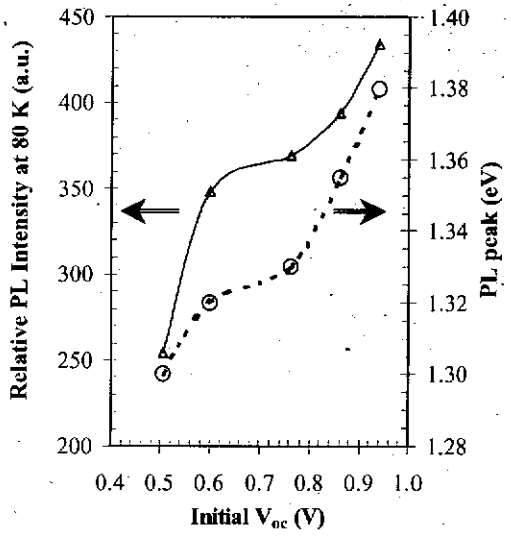


Figure 10. Relative PL intensity and peak energy position at 80 K as function of the initial V_{oc}.

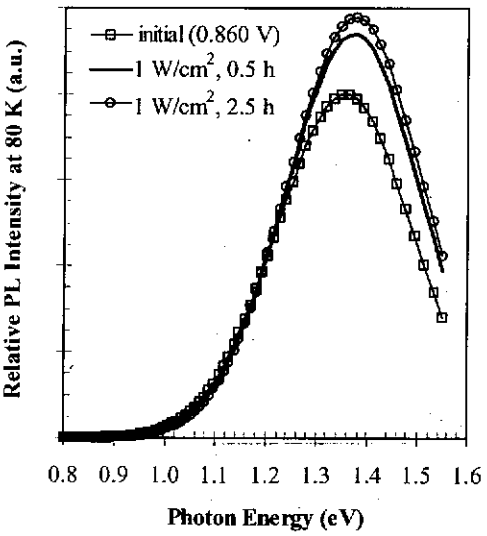
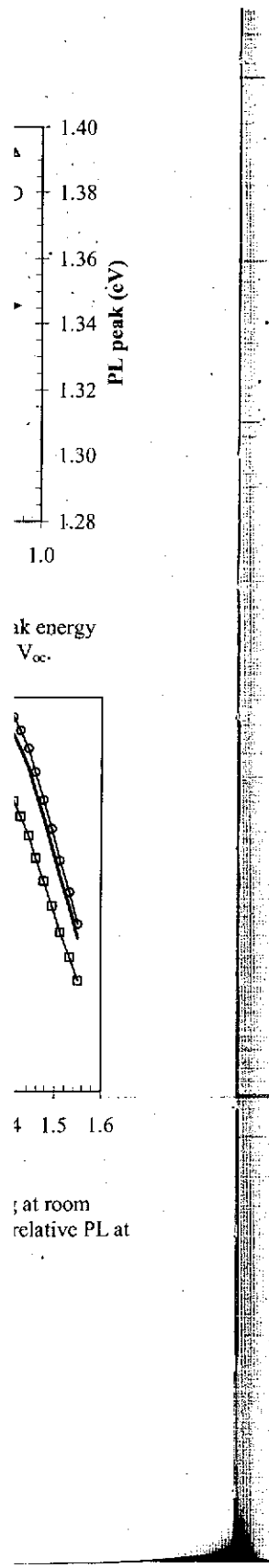


Figure 11. Effect of *in-situ* light soaking at room temperature by 1 W/cm^2 red light on the relative PL at 80 K.



terminates some of the broken bonds. This effectively causes the shell layer of the grains to become more disordered. Although the microscopic picture is complicated, we can expect two consequences: the defect density increases after light soaking, and the region near the grain boundaries becomes more disordered or changes to amorphous. The latter reduces the size of the grains and the microcrystalline volume fraction, thus the enhanced V_{oc}.

In amorphous and mixed-phase silicon materials, light soaking causes both the electronic-defect creation (SWE) [14] and volume expansion (structural change) [16, 17]. In regular a-Si:H materials, the SWE dominates; one observes a decrease in both the PL efficiency and V_{oc} upon light soaking. In the mixed-phase materials, the structural change dominates; one observes an increase in PL efficiency and V_{oc}. In general, the grain boundary regions contain the most unstable sites that show reversible structural change upon light soaking and annealing [18]. A large volume fraction of the grain boundary in the mixed-phase materials results in structural instability.

We should point out that previous studies have reported observations of small (< 50 mV) V_{oc} increase after light soaking [15, 22-25]. However, the cause of the increase was attributed to a light-induced activation of boron in the *p* layer [25], a voltage shift in the dark *J-V* characteristics [15], or not given at all [22-24]. In this study, we observe a much larger effect that depends on the bulk properties of the solar cells such as the *i*-layer thickness and its deposition temperature, and the heterogeneity of the material. The phenomenon is observed only for cells in the mixed phase with the maximum effect occurring in the middle of the amorphous-to-microcrystalline transition region. Photoluminescence spectroscopy further establishes a correlation of the V_{oc} enhancement with a structural change. The most heterogeneous material is the most susceptible to a light-induced structural change.

We should also point out that we have investigated the light-induced effect using X-ray diffraction [26] and Raman spectroscopy. However, no detectable change was observed after light soaking. Further investigation is obviously needed. In addition, since the V_{oc} enhancement and the Staebler-Wronski effect [14] are both light induced and reversible, possible correlation of the two phenomena is being investigated and will be reported separately [27].

CONCLUSIONS

The light-induced enhancement of V_{oc} for heterogeneous-silicon solar cells has been investigated. We find that an increase in V_{oc} depends on the *i*-layer thickness, the *i*-layer deposition temperature, the initial V_{oc} value, and the light-soaking intensity. *In-situ* PL spectroscopy is used to establish the correlation of ΔV_{oc} and a structural change. We suggest that a reduction in the volume fraction or size of the microcrystallites is responsible for the enhancement in V_{oc}.

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Amorphous and

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ABSTRACT

The Pulse
in the kHz range.
the 'OFF' cycle n
we report the incr
Si p-i-n solar cells
i-layer growth rat
efficiency of 8.3%
layer growth rate
Pulsed PECVD te
(220) orientation
orientation, grain
are investigated.
achieved for the i

INTRODUCTIO

For amorpho
photovoltaic appl
deposition rate of
at the rate of <3
chamber system,
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PECVD, which c
however, not eas
Pulsed PECV
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MVSystems Inc.
film transistor m