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Comprehensive Research on Stability of Amorphous Silicon and Alloy Materials and Devices

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Executive Summary

This report represents the progress achieved during the second year of our program to develop a-Si:H and a-(Si,Ge):H materials and devices with better stability by changing the chemistry of the growth technique. During this year, we have shifted our emphasis from cells made on tin oxide substrates(superstrate cells) to cells made on stainless steel substrates (substrate cells). The basic growth technique is to use a remote plasma beam of H or He, created by a low pressure ECR discharge, to create both growth and ion bombardment and/or etching during the growth of the films and devices. By inducing ion bombardment and etching, we can induce a more perfect lattice structure, and thereby improve the properties of the films and devices.

We find that the films produced using a He-beam discharge have very somewhat different properties from the films produced using a H-beam discharge. In particular, the He-beam produced films have a lower H content(about 4.5%) and a smaller Tauc bandgap (about 1.69 eV) than films produced using a H-beam discharge, whose H content is about 8%, and the Tauc bandgap correspondingly higher(about 1.72-1.73 eV). The electronic properties, except for stability, are similar, but the stability of the H-beam produced films is much better than that of the He-beam produced films, even though the He-produced films had a lower H content. This is a non-intuitive result, which contradicts some of the assumptions about the role of H in the film.

During this year, we have made excellent substrate devices using both H-ECR and He-ECR discharges. The fill factors of the devices are in the range of 70%, and the open circuit voltages are in the range of 0.85-0.86 V. We have studied the stability of these devices under prolonged illumination, and we find that the stability of the devices also depends significantly upon the growth technique. In agreement with the results of stability of films, we find that the devices produced using a H-beam are much more stable than the devices produced using a He beam. We have compared these stability data with the stability of similar devices made using glow discharge processes, and we find that the stability of the H-ECR devices is greater than that of glow discharge produced devices, whereas the stability of the He-ECR devices is comparable to the stability of glow-discharge produced devices.

From the results of the stability studies of both films and devices, we conclude that it is not the H content in the film that primarily affects stability, but rather, what the influence of H is on the growth chemistry. In particular, having an etching-type growth chemistry for both films and devices seems to be beneficial for improving stability without sacrificing electronic properties.

We have also initiated studies on the growth of a-(Si,Ge):H films and devices using the H-ECR growth technique. We have been able to make very good films and devices in a-(Si,Ge):H, using both H-ECR and He-ECR processes. We have also initiated work on fabricating substrate-type a-(Si,Ge):H devices, using the H-ECR process.

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Chapter I.

Studies on a-Si:H films and devices grown using two different plasma chemistries

I.1 Introduction

A. Growth of films and the influence of plasma chemistry.

During the previous years of this contract [1,2], we have shown that the use of a remote plasma beam of either He or Hydrogen, generated by an intense electron-cyclotron-resonance (ECR) discharge[3,4] to grow a-Si:H films is an attractive technique for controlling the chemistry of growth, and can lead to films and devices of acceptable quality. We had further shown that a-Si:H films of high stability could be made using a H-beam technique, and that the electronic properties of these films were comparable to the properties of the best glow-discharge deposited films[5]. We had also initiated work on studying the properties of the ECR plasma, and had shown that at low pressures, significant fluxes of H (or He) radicals and ions could be impinging on the substrate. At very low pressures and high H dilution, we could smoothly change the structure of the films from amorphous to microcrystalline to crystalline[6,7], all at the same temperature. Thus, the microstructure of the films depended critically upon the plasma chemistry, which could be changed by either changing the diluent gas, or by changing the pressure. A lower pressure discharge led to a greater flux of H ions and radicals arriving at the substrate, because the resonance zone of the reactor where the plasma generated most of the radicals and ions,was some distance(about 30 cm) away from the substrate. See Fig. 1.1 for a schematic diagram of the reactor. The silane was introduced near the substrate, and therefore, the leading reaction product being generated was likely to be SiH_3, produced by a reaction between H and SiH_4. Using the electron-impact and H-impact coefficients of silane dissociation, for example from [8], one can calculate that with a high dilution rate and the high H flux that we have in the reactor, the production rate of SiH_3 radicals is likely to be at least an order of magnitude greater than the rate of generation of other radicals such as SiH_2, SiH etc. This means that under such high H dilution, high H flux conditions, the growth takes place primarily from SiH_3 radical, and not from a zoo of other competing radicals. This selectivity of growth radicals, and the influence of etching-during-growth by H radicals, are both important distinguishing characteristics of a remote low pressure ECR growth technique, and we had emphasized in the previous reports that this technique had led to more stable films.

In previous work[2], we had shown that by using a He-ECR-discharge(henceforth called the He-ECR process), we could obtain films with a somewhat smaller bandgap, and lower H content, than when we used the H_2-ECR-discharge(henceforth called H-ECR process). The initial properties of the films were very comparable. In this work, we have systematically studied the device formation in these two different types of a-Si:H films.

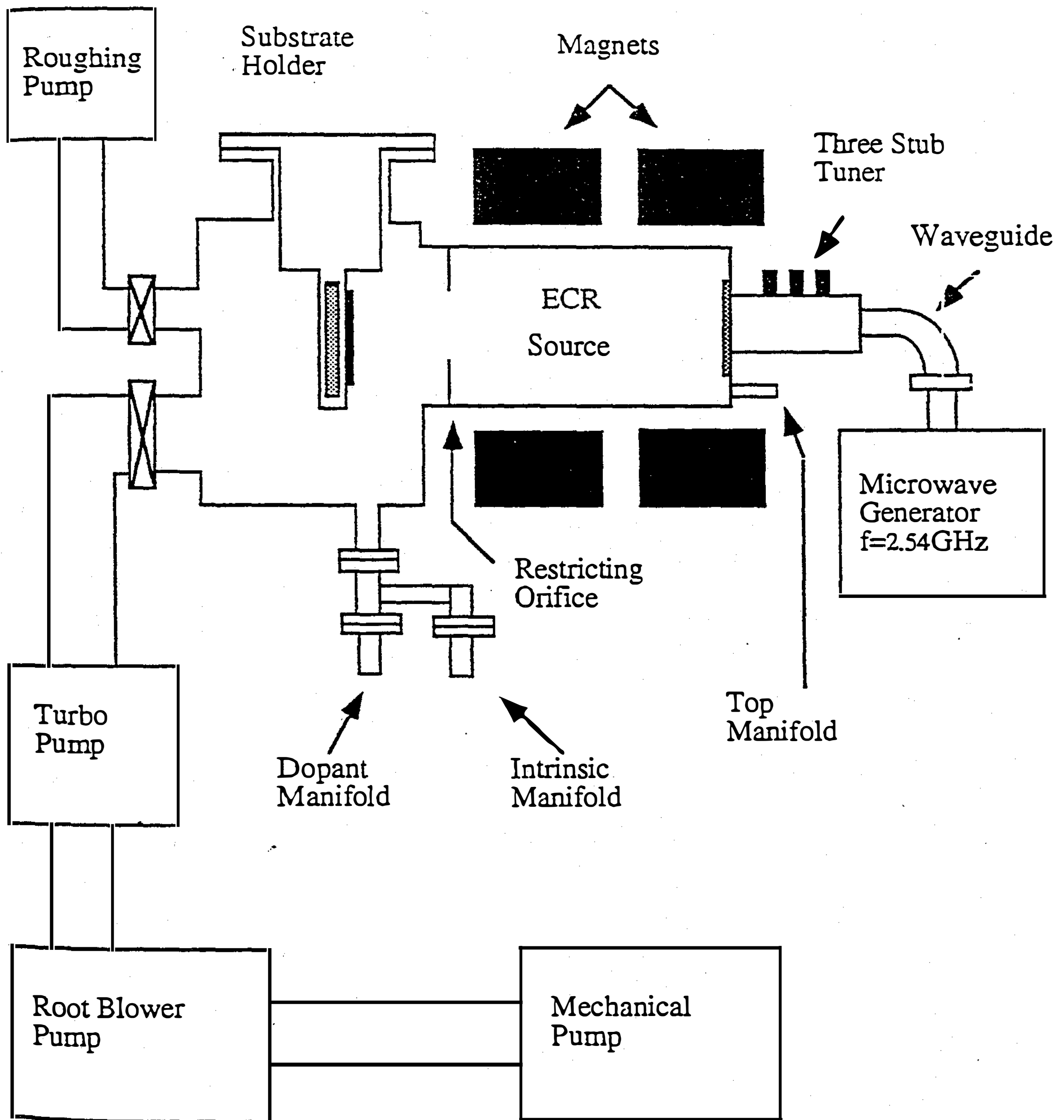


FIGURE 1.1 Schematic diagram of ECR reactor

B. Device fabrication

During the previous period, we had shown that one could make acceptable quality of devices on tin oxide substrates (superstrate devices). We had produced devices with reasonable fill factors (67%), but the voltages tended to be low (in the 0.78 V range). We had discussed that severe problems were encountered from the tendency of B to diffuse from the p into the i layer during a high temperature growth, and that while a new (Si,C) buffer layer, and a new doping profile in the p layer could reduce these problems[4], the diffusion problem remained severe and could not be easily eliminated. As soon as we changed the deposition chemistries and conditions, the problems came back.

I. 2. Fabrication of Substrate Devices.

Therefore, upon further reflection, and based on the advice of Dr. Guha of USSC, we decided to change the device structure to a substrate geometry. Our device structure is shown in Fig. 1.2. We start with a stainless steel(rough, only bright polished, not electro-polished) substrate, on which we grow successively n,i, a buffer, and p layers. We follow with a semi-transparent Cr(about 100 A thick) contact, which has a central Al bussbar on which one can put a probe down to measure the device.

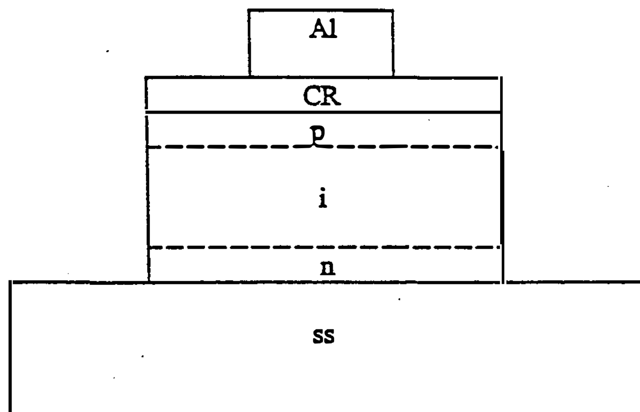


Figure 1.2. Substrate device geometry.

During the fabrication of these devices, we encountered several problems.

1. Contamination from residual phosphorus after doing the n layer.

The rough stainless steel substrate necessitates the deposition of about 500-1000 A of n layer to reduce shorting problems. We found that thicker n layers worked best. However, doing a long(15 minutes or so) deposition of n layers led to a coating of P on the walls of the rector, and the subsequent i layer ended up being contaminated by this residual P from the chamber. The reactive H plasma was etching the P off the walls and putting it back into the gas stream and thence into the layers. Quantum efficiency measurements confirmed this behavior [9]. As an example, we show in

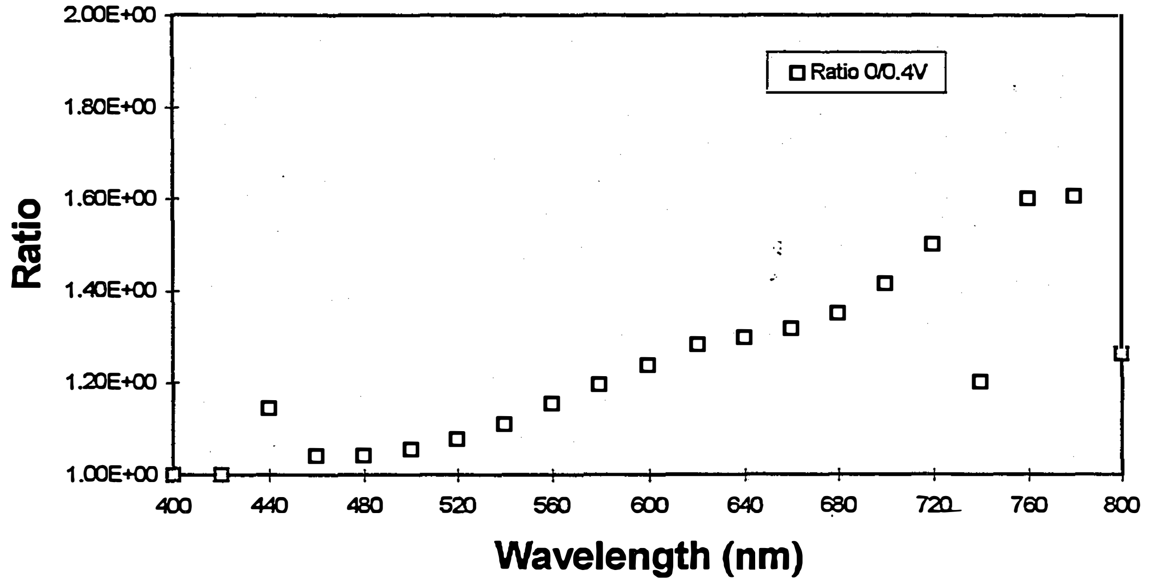


FIGURE 1.3 Ratio of Quantum Efficiency(QE) under zero bias to QE under forward bias, plotted as a function of wavelength. A high ratio at longer wavelengths indicates poor hole transport, a likely result of P contamination of the i layer.

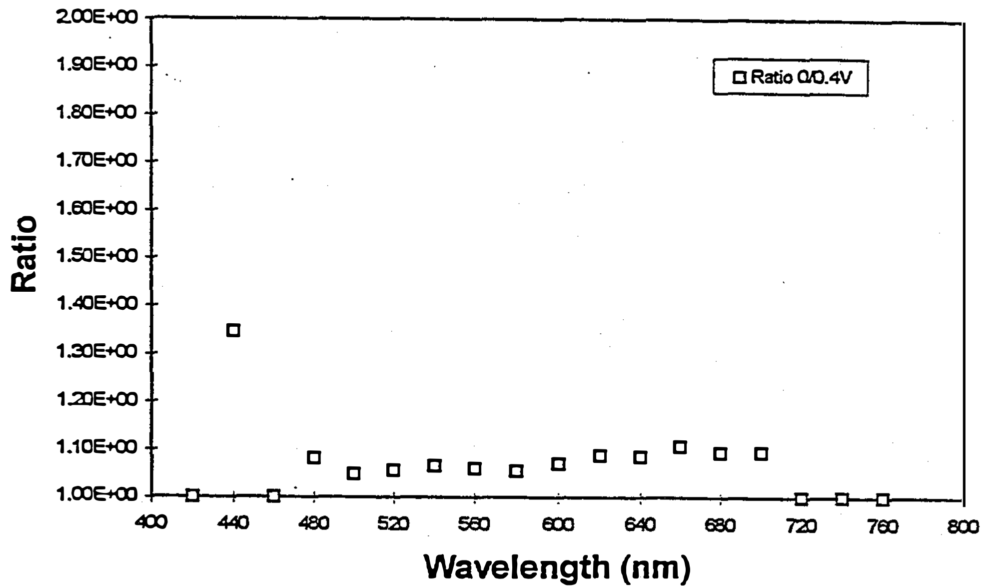


FIGURE 1.4 QE ratio for a good device where P contamination was reduced by using combination of purges and plasma cleaning of the chamber between n and i layers. A low QE ratio at long wavelengths is an indication of good hole transport properties.

Fig. 1.3 the ratio of QE measured under short-circuit and forward bias conditions for a sample where a thick n layer was done in the ECR reactor. The significant increase in QE ratio at long wavelengths is a signature of problems with hole collection.

Based on such data, we decided to use a double n layer deposition, where a 1000 A n layer was deposited in a glow-discharge reactor, and then, the sample was transferred to the ECR reactor, where a very thin(about 100 A) n layer was deposited. The QE ratio curve for this case is as shown in Fig. 1.4, where the QE ratio is low(below 1.1) across the entire wavelength range of interest, indicating little problem with hole collection under forward bias. As expected from the better QE ratios, the fill factors also improved when a double n layer was used. We have adopted this double n layer approach as a standard procedure for now.

Following the n layer, we do a thorough purge of the reactor chamber with nitrogen and silane-hydrogen mixtures to remove any residual phosphine gas, and then follow with a chamber etch using a H plasma, followed by the deposition of a dummy i layer on the chamber walls. The sample is shielded by a shutter during this procedure. The combination of plasma etch and dummy i layer deposition removes the last of the residual contaminants, and allows us to deposit a relatively pure i layer.

Since the sample is shielded during the etch and dummy layer deposition, there is the possibility that the sample holder, which is also shielded, can harbor some residual P. To remove this P, we do a 3 min. etch of the sample with H prior to deposition of the i layer.

2. Problems with poor electron collection at p-i interface.

While there is no B-diffusion problem for this device structure, since the p layer is deposited last and at low temperatures, there is the problem with electron back diffusion towards the p-i interface and subsequent recombination[9]. The p layer is a larger gap a-(Si,C):H layer, whereas the i layer is a smaller gap a-Si:H layer. Therefore, to minimize recombination, it is best to use a graded buffer layer which utilizes bandgap grading to enhance the electric field near the interface, which serves to drive away the electrons from the interface[9]. When we do an inappropriate buffer layer(not enough grading), the result is a poor collection of electrons at the p-i interface, as indicated by a QE ratio curve which is high for short wavelengths[9]. The results for a poor buffer layer for a substrate cell are shown in Fig. 1.5. By fine tuning the buffer layer, we were able to obtain good QE ratios as shown in Fig. 1.4, and good fill factors(Fig. 1.6).

1.3 Device Results

A) Cells grown using H-ECR growth process.

The I(V) curve for a cell grown using the H-ECR process is shown in Fig. 1.6. Please note that the current values are not meaningful, because of absorption in the semi-transparent Cr contact. However, both fill factor and voltages are meaningful. From Fig. 1.6, we see that the fill factor is very good. The i layer thickness was 4500 A, measured using reflection measurements in a spectro-photometer. The corresponding QE is shown in Fig. 1.7. The QE ratio curve was already shown

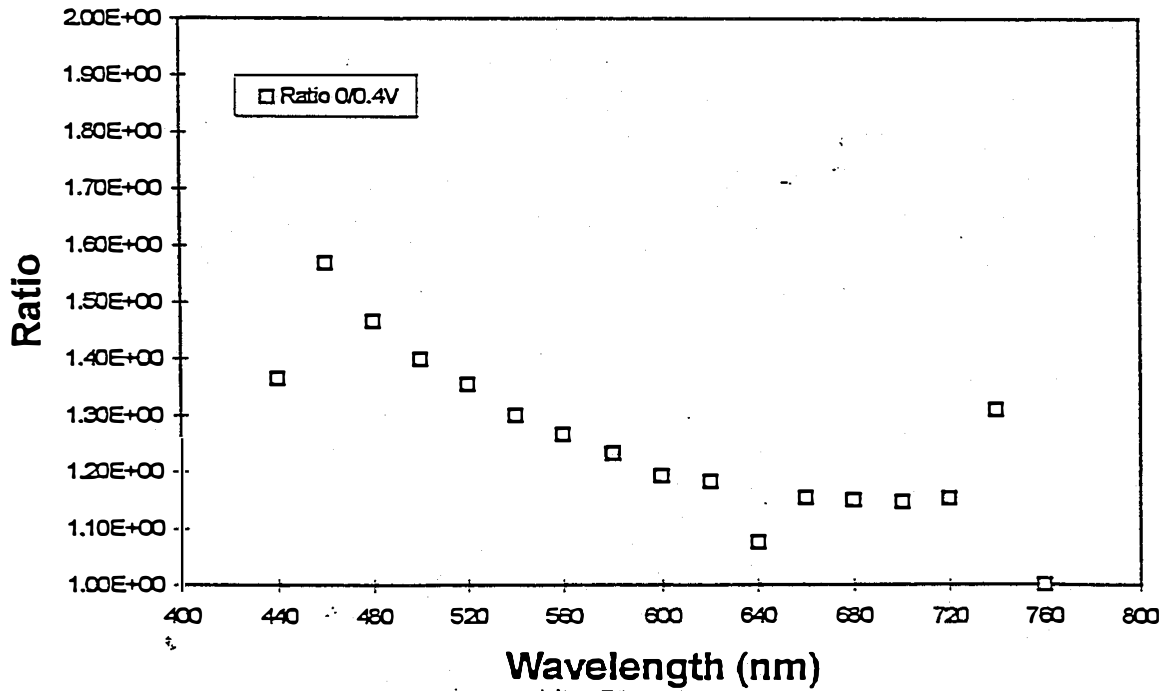


FIGURE 1.5 QE ratio curve for a device, showing problems at p/i interface because of differing bandgaps between the i and the p layers.

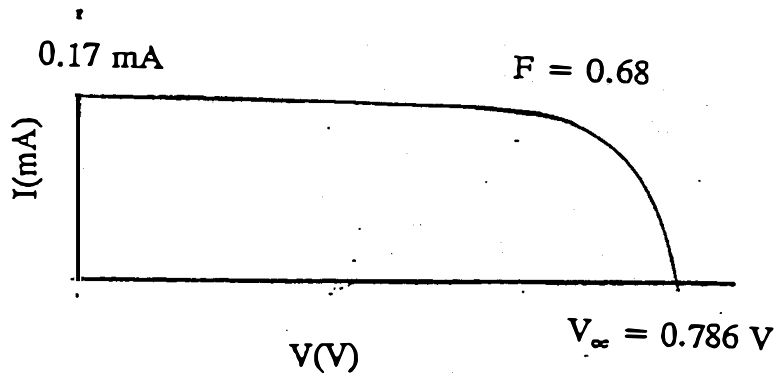
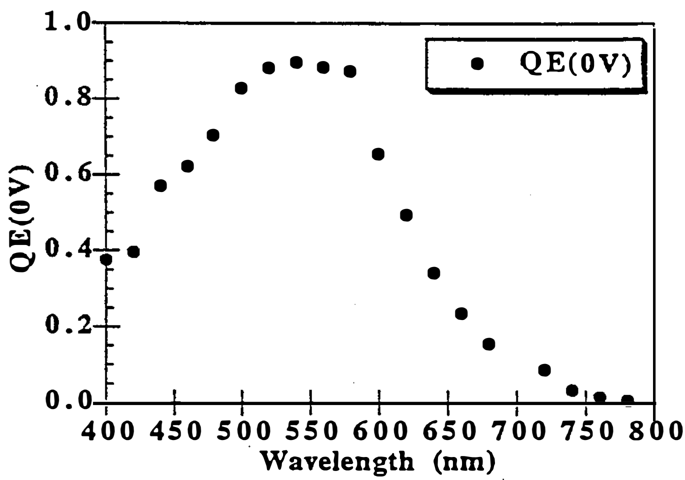


FIGURE 1.6 I(V) curve of a H-ECR cell under simulated AM 1.5 illumination. The voltage and fill factor are meaningful, but the current density is difficult to determine because of the Cr contact and the Al bussbar(See Fig. 1.1)



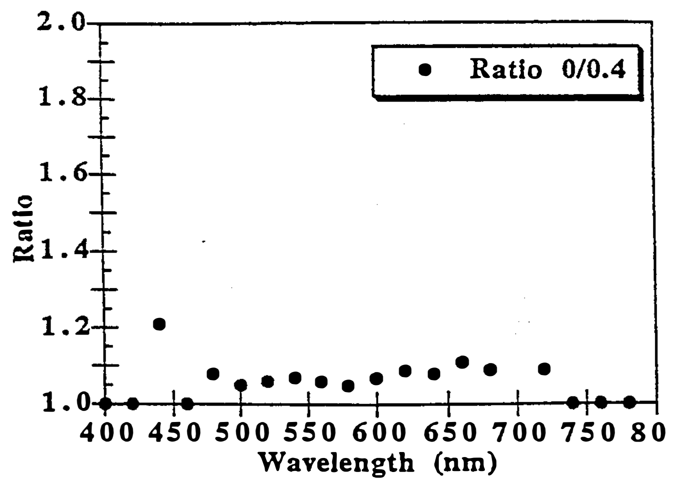


FIGURE 1.7 QE at zero and forward bias of the device whose I(V) curve was shown in Fig. 1.6. This is internal QE, arbitrarily normalized to 0.9 at maximum.

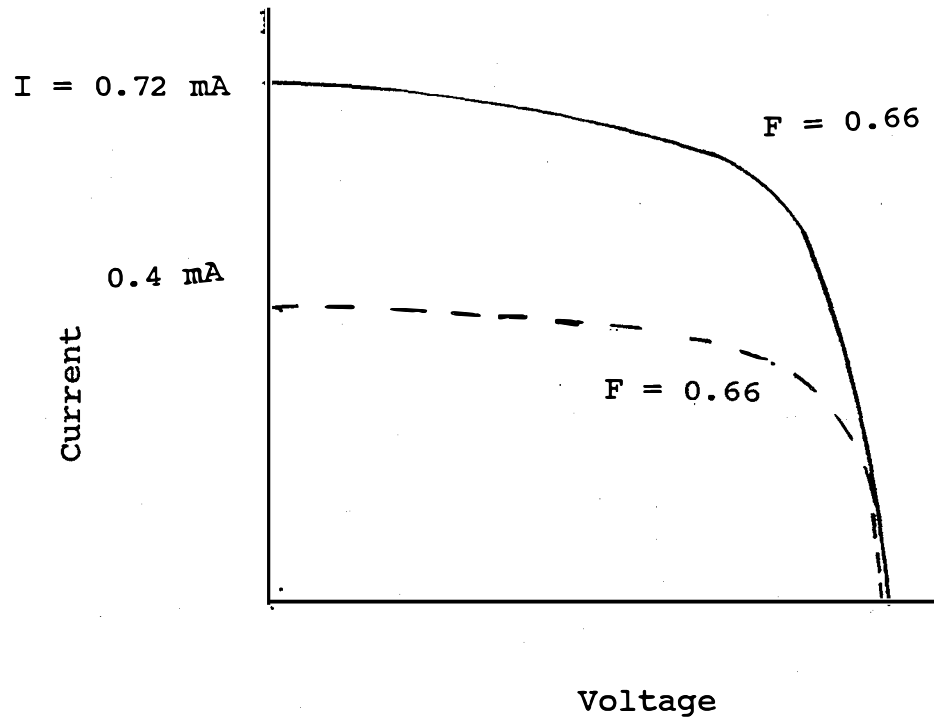


FIGURE 1.8 Two I(V) curves, taken at different light intensities, showing that the fill factor does not change upon increase in light intensity. This indicates an absence of significant field collapse due to hole trapping when the device is illuminated.

previously in Fig. 1.4.

Unlike the case for some reported work[10], we do not find any significant impact of light intensity on fill factor, implying little field collapse. This result is shown in Fig. 1.8, where we show the I(V) curves for an intensity corresponding to 1 sun, and 2 suns. The fill factors are essentially the same in each case.

We can estimate what the voltage would have been if the Cr contact had been replaced by an ITO contact, and the cell therefore, had full sun illumination shining on it.
To make this estimate, we measured the dependence of open circuit voltage and short circuit current on light illumination intensity. In Fig. 1.9, we show the behavior of open circuit voltage as a function of short circuit current. As expected, as the short circuit current increases, the voltage increases logarithmically, with a diode n factor of about 1.2. The full-sun voltage is about 0.85 V, comparable to the voltages in glow-discharge cells of similar bandgaps(1.72 eV).
Thus, by changing over to the substrate geometry, we have achieved one of our objectives, namely to increase the open circuit voltage in ECR cells.

B) Cells grown using He-ECR process

We also made cells on stainless substrates by using the He-ECR process. The I(V) curve of a cell is shown in Fig. 1.10, and the QE curve in Fig. 1.11. The fill factor is again very good.

We can compare the bandgaps of the i layer in the He-ECR cells with the bandgap of the i layer in the H-ECR cells by carefully measuring subgap QE for cells of similar thicknesses. Such a comparison is shown in Fig. 1.12, which shows that the i layer of the He-ECR cell, indeed, has a lower bandgap by about 35 meV, about the same range as the difference found in spectro-photometric measurements of Tauc gaps in films. The lower gap, of course,is a consequence of the lower H content in the He-ECR material when compared with the H-ECR material.

I.4 Comparison of stability of H-ECR and He-ECR devices.

A primary objective of the ECR deposition technique was to see if changes in plasma chemistry made changes in stability of materials and devices. In previous work, we had shown that it appeared that the ECR deposition, using the H-ECR process, did lead to substantial improvements in the stability of films and in superstrate devices. We now examine the stability of substrate type devices made using H-ECR and He-ECR process.

To do the experiment, we made 3 substrate cells, one by glow-discharge, one by H-ECR and one by He-ECR. All the cells had similar thicknesses of the i layer, and had similar voltages initial QE's and fill factors. The careful methodology for making such comparisons has been described previously in one of our reports[2].

Since fill factor is the parameter that changes most critically upon degradation, it is the parameter we focussed on. The cells were each illuminated under a focussed ELH lamp so that the illumination

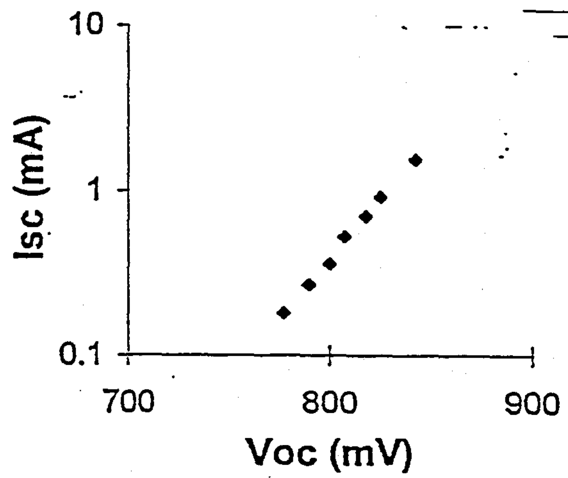


FIGURE 1.9 Open-circuit voltage vs. short-circuit current for a cell. The nearly logarithmic curve has a diode factor of 1.2.

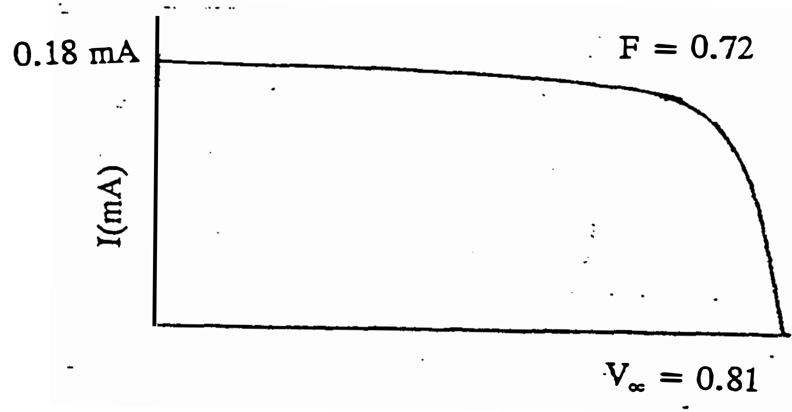


FIGURE 1.10 I(V) curve for a cell made using He-ECR process

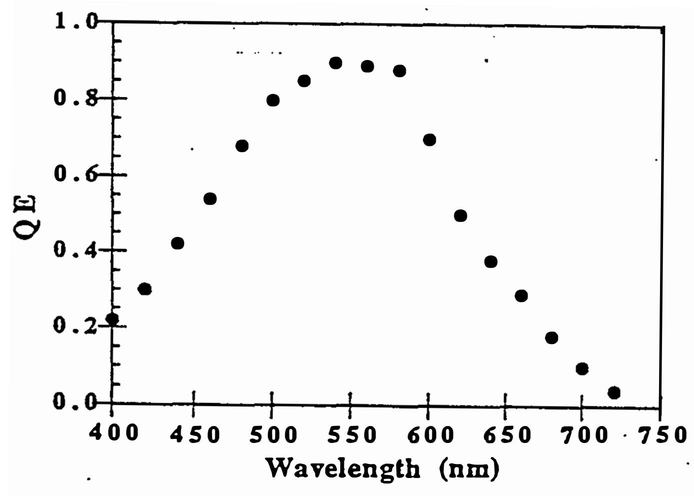


FIGURE 1.11 QE curve for a He-ECR cell

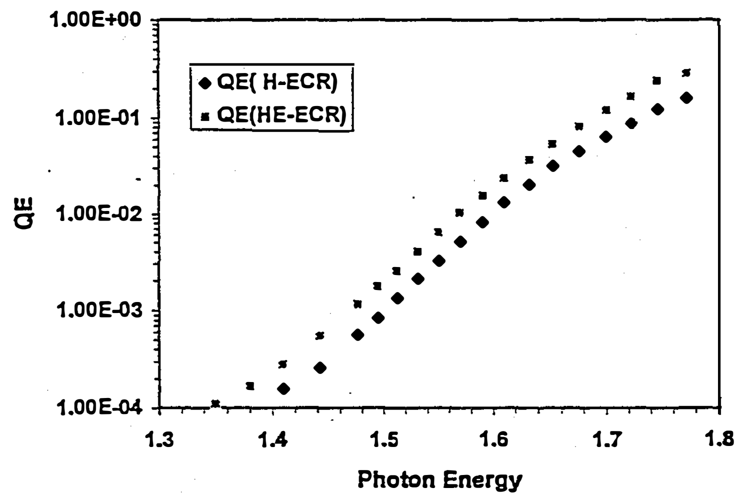


FIGURE 1.12 Comparison of subgap QE of two cells of nearly identical i layer thicknesses. One cell was made using H-ECR process, and the other, using He-ECR process. The lateral shift in QE to lower energies for the He-ECR cell is an indication of a lower bandgap of the i layer by about 35 meV.

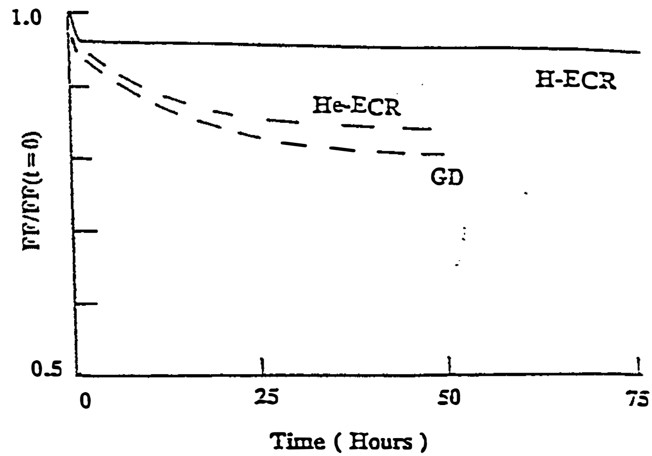


FIGURE 1.13 The degradation of fill factor for 3 cells under light soaking. All the cells had similar thicknesses of the i layer, and similar voltages and initial fill factors.

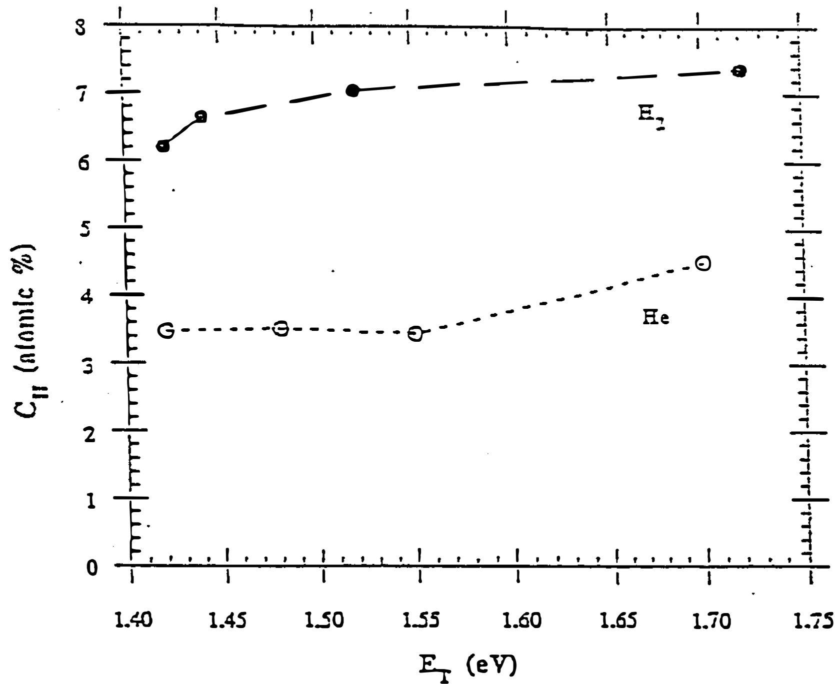


FIGURE 1.14 The H content vs. Tauc gap for H-ECR vs. He-ECR films of a-Si and a-(Si,Ge).

reaching the cell(after correcting for the absorption in Cr) was about 1.6 suns(160 mW/cm^2). The cells were mounted on a water cooled copper block kept at 20 C, with fans blowing air over the surface of the cell. In Fig. 1.13, we show the relative degradation in the fill factors of the 3 cells for different light soaking times. The result one gets is that the H-ECR cell is much more stable than the He-ECR cell, which is comparable in stability to the glow-discharge cell.

This result is unexpected, since the H-ECR cell had more bonded H(7-8%) than the He-ECR cell(4%). See Fig.1.14 . The only logical conclusion one can draw from this observation is that it is not the H content that matters, but the H chemistry during growth. During the growth of the device by H-ECR, H plays a role in both determining the growth chemistry, and in etching-during-growth. Both these phenomena lead to changes in microstructure during growth, and that may be the reason why the ECR cells are more stable than the He-ECR cells.

I.4 Summary of a-Si:H device work

In summary, we have developed a process for depositing high quality a-Si:H cells in a substrate geometry. The severe problem of P contamination in a single chamber reactor was solved using innovative n layers and a very thorough purge and plasma cleaning procedure.
Cells with high fill factors were obtained. When we measured the stability of these devices, we found that the cells deposited using the Hydrogen-ECR process were much more stabile than either glow discharge, or He-ECR cells. *This is an unexpected result which suggests that the role of H during the deposition needs to be carefully examined in order to further improve the stability of the material and the device.* It may also be fruitful to reexamine the role of other etchants such as F during growth, also in the ECR reactor, which serves to produce a plasma which can be carefully controlled for ion flux and energies.

Chapter II

Research on a-(Si,Ge):H materials and devices

II.1 Introduction

In previous work[2], we had shown that it was possible to make high quality a-(Si,Ge):H films with the ECR process. That work had concentrated on making films using He-ECR process, where He gas was used as the plasma gas. We had obtained high quality films, with low subgap α and low valence band tail Urbach energies, using that process. In Fig. 2.1, we show the data for Urbach energy as a function of Tauc gap, and in Fig. 2.2, we show the corresponding data for Subgap α, measured at the shoulder, also as a function of Tauc gap of the films.

II.2 Growth of a-(Si,Ge):H films using H-ECR technique.

Based on the discovery about the superior stability of H-ECR devices described in Chapter I above, it was decided to investigate the preparation of a-(Si,Ge):H films using the H-ECR process. The films were deposited using the following typical conditions:

Hydrogen/(Silane+Germane) ratio	20:1
Temperature of growth	300-325 C
Pressure	10 mT
Growth rate	0.8-1 A/sec

The Tauc gap was varied by varying the silane/germane ratios.

The films were characterized by measuring dark and photo-conductivity, activation energy, and subgap absorption using a two-beam photo-conductivity technique[11].

In Fig. 2.3, we show the photo/dark conductivity ratio of the films as a function of Tauc gap, and in Fig. 2.4, the activation energy. As expected, the activation energy decreases as the bandgap decreases.

In Fig. 2.5, we plot the Urbach energy of valence band tails as a function of Tauc gap, and compare it with the Urbach energies in films deposited using the He-ECR discharge[12], and RF triode glow discharge[1]. The values for the Urbach energies in H-ECR films are very comparable to the values obtained in He-ECR films, and the low values(below 50 mev) indicate good quality films.

In Fig. 2.6, we plot the values for subgap α as a function of Tauc gap, and compare it with values in He-ECR films, and in RF triode glow-discharge films reported earlier[1]. In agreement with Fig. 2.2 , this figure shows that the H-ECR films have initial properties very similar to the ones obtained in He-ECR films,and they appear to be superior to the properties in H-diluted glow discharge films.

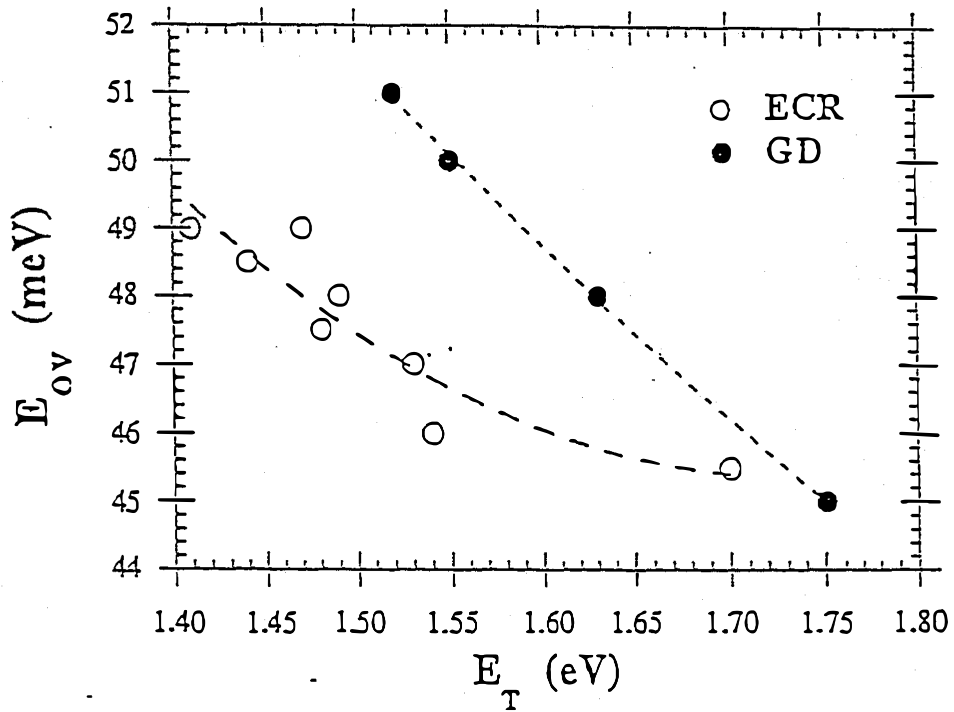


FIGURE 2.1 Urbach energy vs. Tauc gap for a-(Si,Ge):H Films prepared using either GD or He-ECR processes. A low value of Urbach energy indicates a higher quality material.

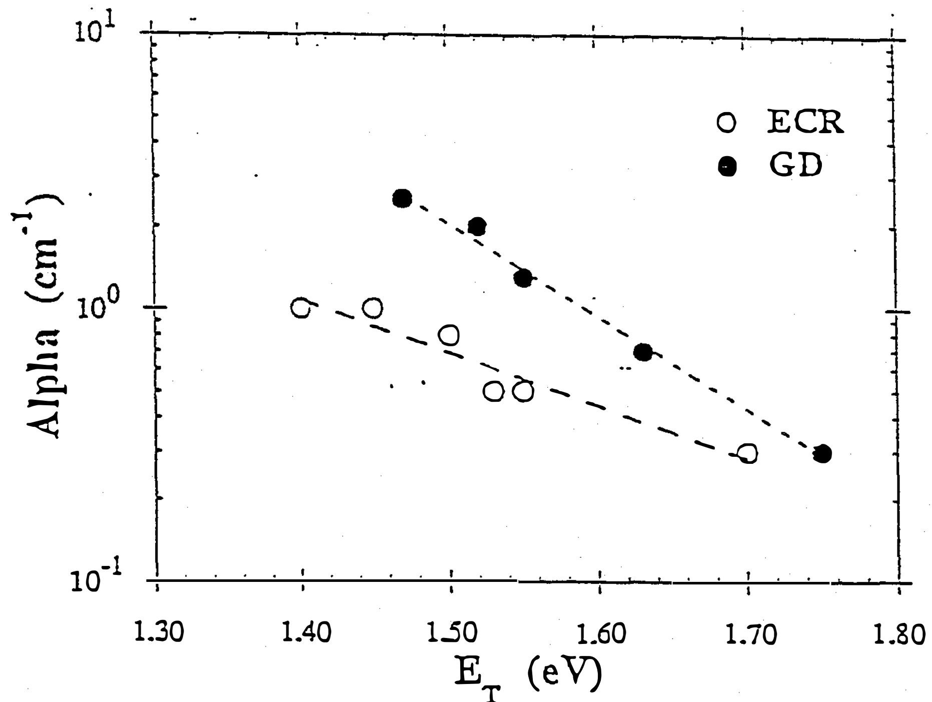


FIGURE 2.2 Subgap absorption coefficient, α, measured at the shoulder, as a function of Tauc gap for a-(Si,Ge):H films prepared using either the GD or He-ECR process.

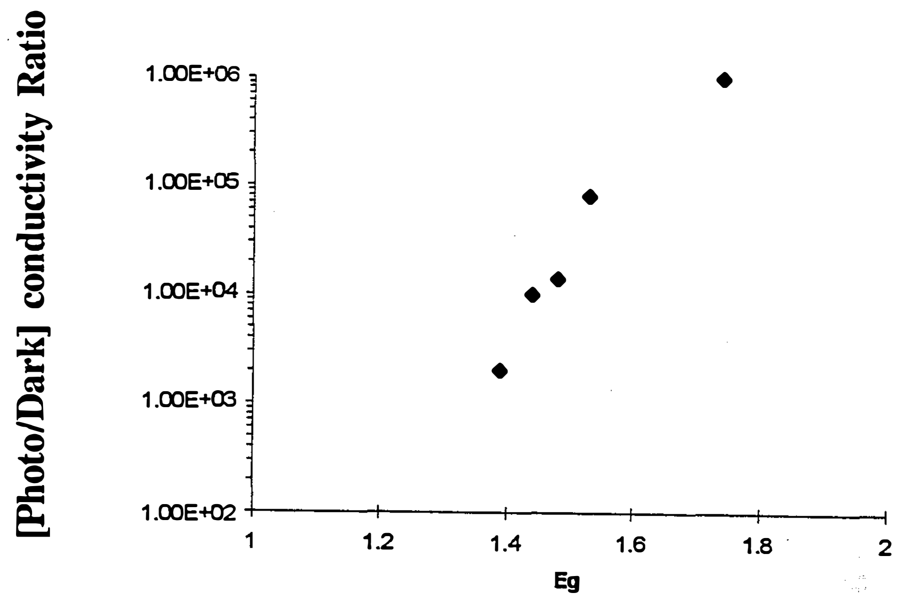


FIGURE 2.3 Dark and AM1.5 photo-conductivity of a-(Si,Ge):H films made using H-ECR process as a function of Tauc gap of the film

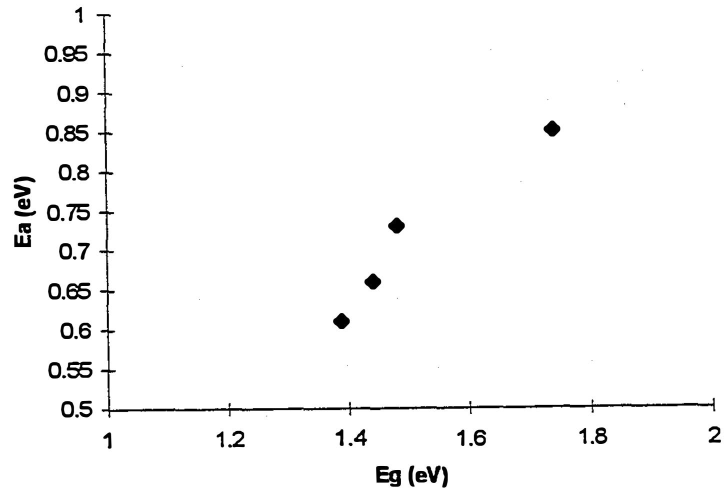


FIGURE 2.4 Activation energy of a-(Si,Ge):H films made using H-ECR process

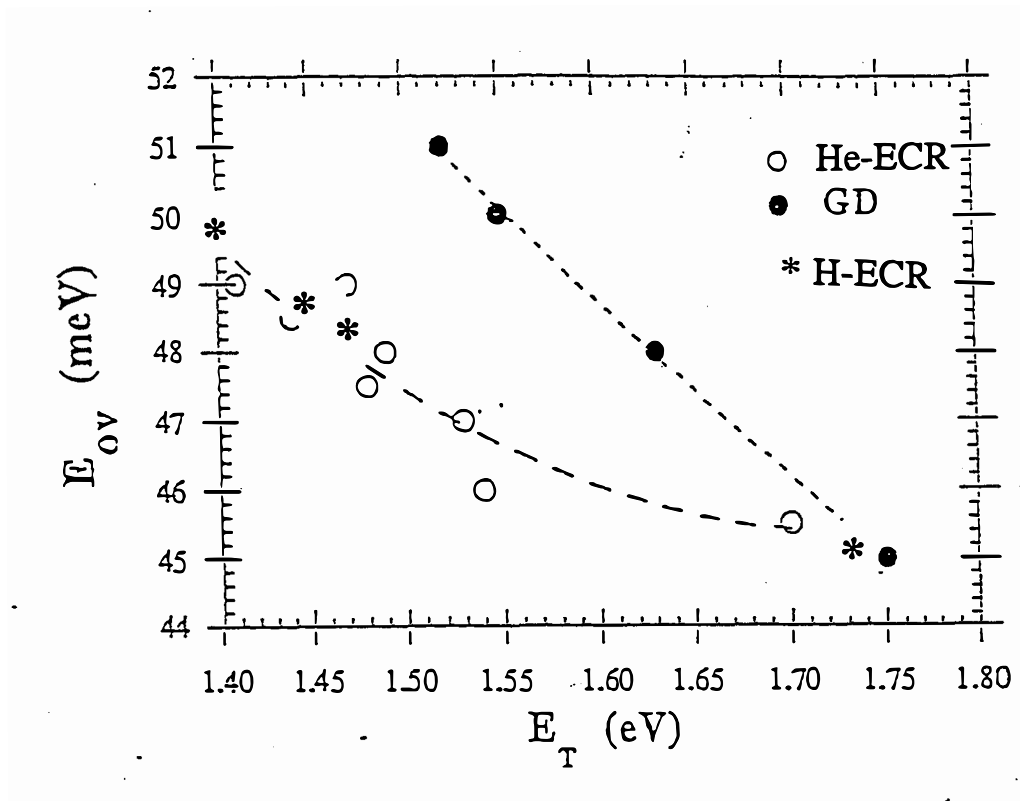


FIGURE 2.5 Urbach energy of a-(Si,Ge):H films made using H-ECR process, plotted vs. Tauc gap

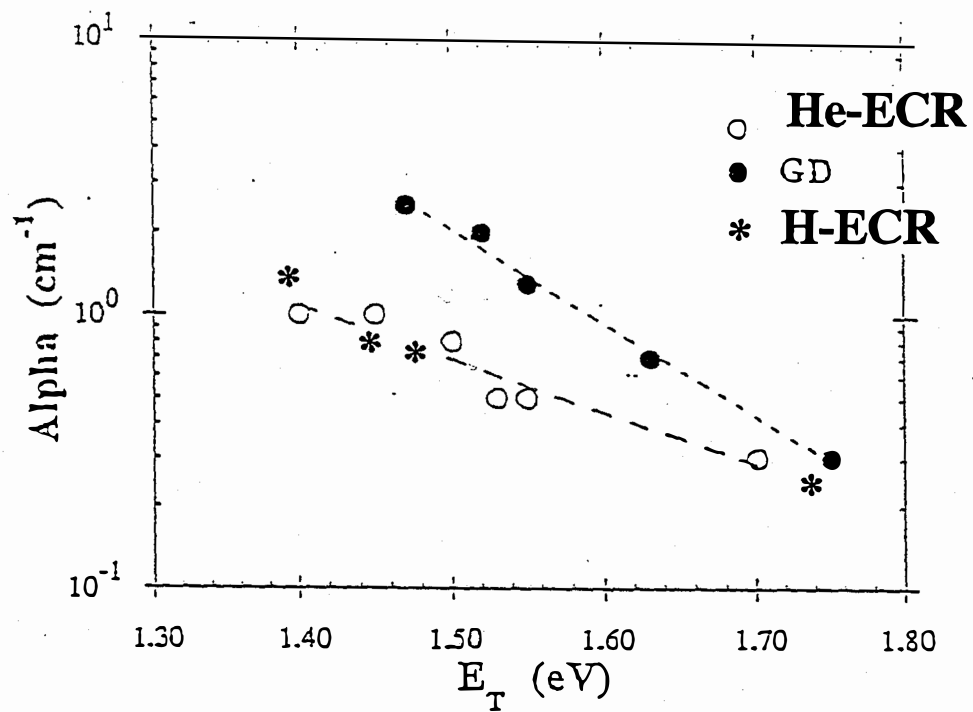


FIGURE 2.6 Subgap α of a-(Si,Ge):H films made using H-ECR process, plotted vs. Tauc gap

II.3 Devices in a-(Si,Ge):H

Based on these results, we decided to investigate some preliminary devices in H-ECR deposited a-(Si,Ge):H.

The basic device structure was a substrate geometry structure on stainless steel, as shown in Fig. 1. The n layer, which was deposited first, was an a-Si:H layer.After the n layer, we used the same procedure as used for fabricating good a-Si:H cells described in chapter I. A thorough purging and cleaning of the reactor was agin found to be necessary. Next, a short (100 A or so) a-(Si,Ge):H buffer layer was deposited on top of the n layer. The bandgap of this layer was varied smoothly from a-Si:H next to the n layer to the appropriate a-(Si,Ge):H bandgap characteristic of the I layer we were studying. Next, an a-(Si,Ge):H I layer was deposited. Next came a buffer layer, which matched the abdngap of the I layer to the bandgap of the p layer. Finally, the p-a-(Si,C):H layer was grown. A semi-transparent Cr dot and a central Al bussbar completed the device.

In Fig. 2.7, we show the I(V) curve for such a cell. The QE data is shown in Fig.2.8, and it shows the red response extending beyond the normal a-Si:H cell deposited on a non-reflecting stainless substrate(See Fig. 1.7 for comparison). The fill factor is reasonable, 58 %.

We next proceeded to deposit a graded gap a-(Si,Ge):H cell, also in the substrate geometry. In this cell, the bandgap was varied smoothly from a-(Si,Ge):H near the p layer to a-Si:H at the back n layer. The buffer layer at the p/i interface was varied smoothly from a-(Si,Ge):H to a-(Si,C):H.

The results for one of the cells are shown in Fig. 2.9. The fill factor is excellent, as expected because of the graded gap. Further devices of this kind will be made during the next year.

II.4 Summary of research on a-(Si,Ge):H

We have succeeded in depositing high quality a-(Si,Ge):H material using the H-ECR process. We have also deposited proof-of-concept a-(Si,Ge):H devices in t he substrate geometry using the process. When the bandgap was smoothly graded so as to enhance hole collection, a high fill factor was obtained. Future work will concentrate on using material with lower bandgaps. In the range of 1.4-1.45 eV, and in studying the stability of such cells.

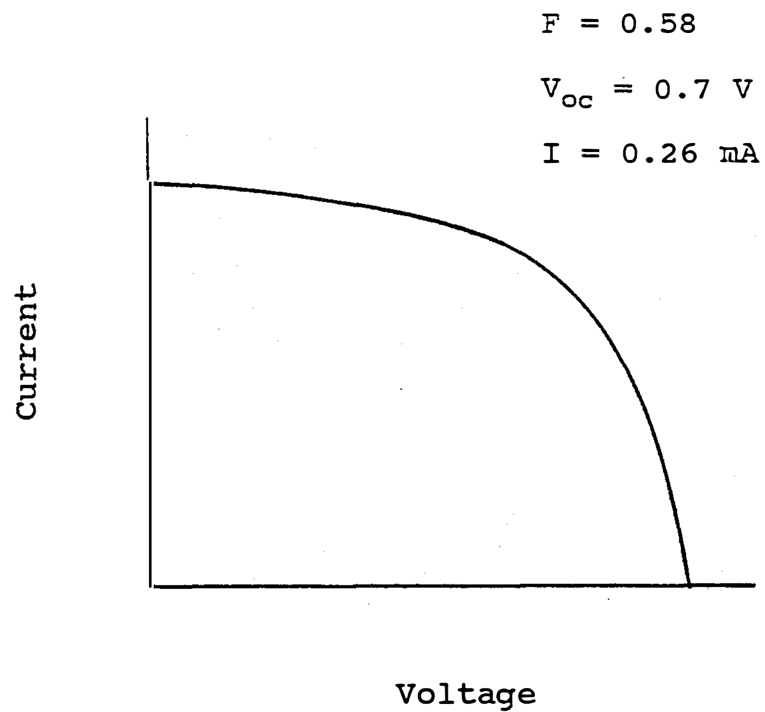


FIGURE 2.7 I(V) curve under illumination for an a-(Si,Ge):H cell of one bandgap.

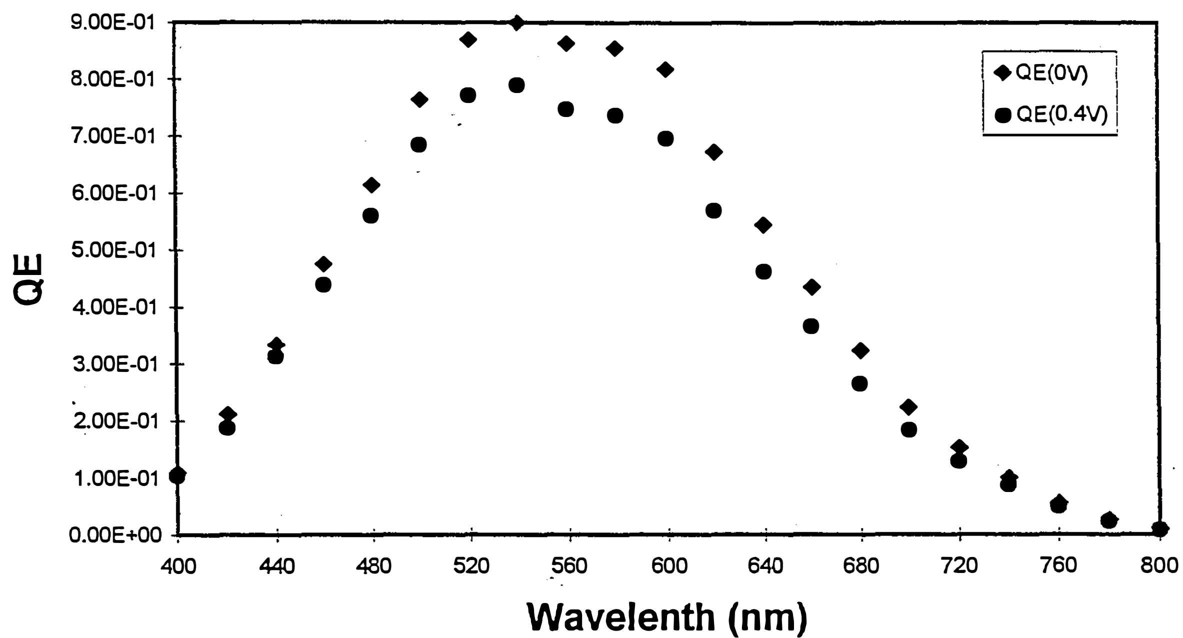


FIGURE 2.8 QE curve for the cell whose I(V) curve was shown in Fig. 2.7

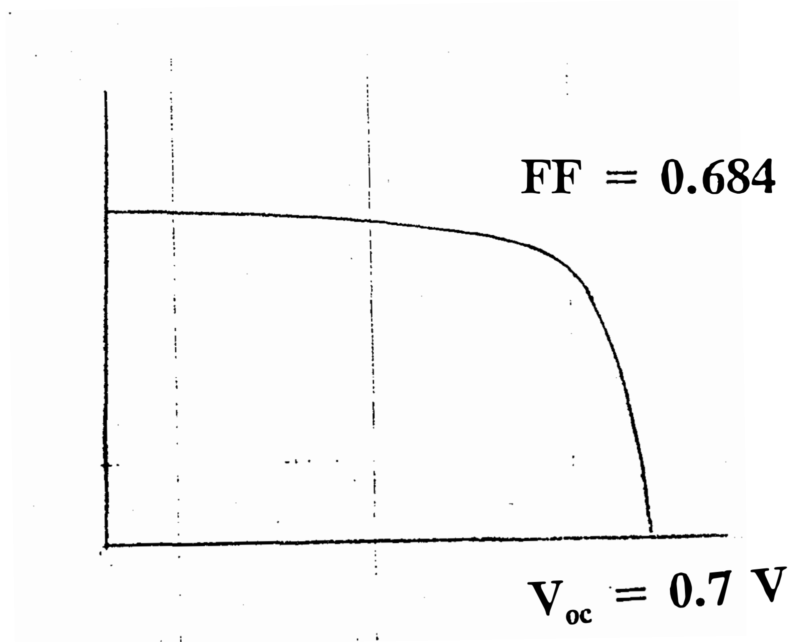


FIGURE 2.9 I(V) curve for a graded gap a-(Si,Ge):H p-i-n cell

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13. ABSTRACT *(Maximum 200 words)*
This report represents the progress achieved during the second year of Iowa State University's program to develop a-Si:H and a-SiGe:H materials and devices with better stability by changing the chemistry of the growth technique. Researchers shifted their emphasis from cells made on tin oxide substrates (superstrate cells) to cells made on stainless-steel substrates (substrate cells). By inducing ion bombardment and etching using both H-ECR (electron-cyclotron-resonance) and He-ECR discharges, researchers can induce a more perfect lattice structure and thereby improve the properties of the films and devices. Researchers found that He-beam-produced films have a lower H content (about 4.5%) and a smaller Tauc bandgap (about 1.69 eV) than films produced using a H-beam discharge, whose H content is about 8% and Tauc bandgap is about 1.72-1.73 eV. The stability of the H-beam-produced films is much better than that of the He-beam-produced films, even though the He-produced films had a lower H content. Researchers also made excellent substrate devices with fill factors in the range of 70% and open-circuit voltages in the range of 0.85-0.86 V. Researchers compared the stability data with the stability of similar devices made using glow-discharge processes, and they found that the stability of the H-ECR devices is greater than that of glow-discharge-produced devices, whereas the stability of the He-ECR devices is comparable to the stability of glow-discharge-produced devices. From the results of the stability studies of both films and devices, they conclude that it is not the H content in the film that primarily affects stability, but rather, the influence of H on the growth chemistry. In particular, having an etching-type growth chemistry for both films and devices seems to be beneficial for improving stability without sacrificing electronic properties.

14. SUBJECT TERMS

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