Electrical Interface Characterization of Ultrathin Amorphous Silicon Layers on Crystalline Silicon

Patrick Thoma,* Evelyn Tina Breyer, Oana-Maria Thoma, Georgeta Salvan, and Dietrich R. T. Zahn

Heterojunctions of amorphous (a-Si) and crystalline (c-Si) silicon combine the favorable absorption characteristics of a-Si for the solar spectrum, high energy conversion efficiencies, low processing temperatures, potential for low production costs, and a reduced amount of silicon used due to thin a-Si films. To investigate a-Si/c-Si heterojunctions, amorphous p-type silicon (a-Si) with a thickness of 5 nm is deposited via radio frequency-pulsed magnetron sputtering on p-doped, (100)-oriented, crystalline silicon (c-Si) wafers. During deposition, the crystalline silicon wafers are kept at 430 °C and the hydrogen flow rate is varied from 0 to 50 sccm (standard cubic centimeters per minute). Temperature-dependent current–voltage measurements are carried out to investigate the dominant transport mechanisms of electrical conduction. Moreover, the influence of hydrogen on physical properties such as barrier height, activation energy, and electrical conductivity is analyzed. A current–voltage dependence as predicted by the thermionic emission model for the low forward-bias region below 0.25 V is observed. Temperature regimes for nearest neighbor hopping and band conduction are revealed by the Arrhenius plot and are found to depend on the hydrogen flow rate.

1. Introduction

As research and industry constantly try to find cheaper and more efficient solar cells for energy harvesting, attention was recently drawn toward so-called heterojunction solar cells consisting of different semiconducting layers.[1–8] Notably, the heterojunction of amorphous (a-Si) and crystalline (c-Si) silicon combines the favorable absorption characteristics of a-Si for the solar spectrum,[9] high energy conversion efficiencies,[10–12] low processing temperatures,[11] potential for low production costs,[13] and a reduced amount of silicon used due to thin a-Si films.[14] a-Si-based solar cells often have an intrinsic silicon layer between a-Si and c-Si. By omitting this intrinsic layer, we intend to study candidates for simpler and cheaper solar cell designs.

However, obtaining film properties suitable for photovoltaic applications is challenging due to high defect densities,[15] low interface quality,[16] leading to interface recombination,[17] and the fact that electrical transport models present in a-Si/c-Si heterojunctions are still under debate.[18–22] To tune the desired physical properties, it is essential to first understand the physical mechanisms that govern these systems.

Earlier work on a-Si/c-Si systems often focused on the importance of annealing temperatures during or after deposition,[1,3,23–25] which affect structural and electrical properties of the samples.[1,17,24–27] The most common technique for the sample and large-scale preparation of solar cells composed of amorphous and crystalline silicon is plasma-enhanced chemical vapor deposition (PECVD).[22,28,29] which allows large-area deposition at low temperatures and high deposition rates.[30] However, when using PECVD, complex plasma chemistry has to be considered as a mixture of multiple gases is used.[31] Moreover, this technique often requires the use of toxic gases such as SiH₄.[12]

An important aspect concerning silicon thin-film preparation is the presence of hydrogen, which plays a determinant role in influencing the Fermi-level position,[27] band offset,[33] surface state density,[2] and passivation of dangling bonds.[34] This has been mostly studied using PECVD,[13,31,35–37] and hot-wire chemical vapor deposition.[16,38] However, how the amount of hydrogen affects important properties of radio frequency (RF)-pulsed magnetron sputter-deposited films is rarely studied.

Magnetron sputtering deposition is a thin-film preparation method which does not involve the use of toxic gas[39] and is suitable for wafer-scale manufacturing.[40] In addition, sputtering offers the advantage of high deposition rates at low temperatures[41] and the possibility to control the amount of hydrogen incorporated during deposition. By changing the hydrogen flow rate during the RF magnetron sputtering process and applying
electrical measurements over a wide temperature range, we were able to observe the change of electrical conductivity and conduction mechanisms due to increasing hydrogen incorporation. Moreover, changes in thermal activation energies and barrier heights were determined. Typically, temperature regimes used for silicon sample preparation are in the range of 250 °C. According to a study by Breyer,[42] however, showed that higher substrate temperatures in the range of 420–450 °C lead to thin films with thermally activated band conduction. This result hints toward interesting physical effects which we wanted to investigate in more detail.

An extensive study on the hydrogen content present in the films and the related influence on optical properties using infrared spectroscopy, Raman spectroscopy, spectroscopic ellipsometry, and charge carrier transient spectroscopy can be found in earlier work.[43]

2. Experimental Section

p-doped, (100)-oriented, crystalline silicon substrates with a resistivity of 9–19 Ω cm and a native oxide layer were cleaned with acetone, ethanol, and deionized (DI) water for 10 min each in an ultrasonic bath. To remove the native oxide layer, as well as passivate the dangling bonds at the surface,[10] the substrates were etched in 5% hydrofluoric acid (HF) for 3 min, cleaned with DI water, and blow dried in nitrogen (N₂) flow. Afterward, the samples were transferred into a high-vacuum chamber (PLS 570, Pfeiffer Vakuum, p ≈ 5 × 10⁻⁷ mbar), and 32 nm-thick nickel–chromium (NiCr, 80:20) back contacts were sputtered through a mask onto the unpolished side of the silicon wafer. Sputtering deposition was conducted using an RF pulsed magnetron sputtering source, working at a frequency of f = 60 kHz and a power of P = 50 W. The sputtering gas used was argon.

Once the NiCr contacts were deposited, the substrates were again cleaned, wet etched, and transferred back into the vacuum chamber. After etching the polished side of the silicon substrates (here referred to as the “front side”) in an argon plasma for 5 min, 5 nm of amorphous silicon (target: p-type, boron-doped, 17 mΩ cm) was sputtered using RF-pulsed magnetron sputtering deposition (f = 60 kHz, P = 90 W). During deposition, the substrates were kept at 430 °C. A defined flow rate of hydrogen in the range from 0 to 50 sccm was introduced during sputtering to saturate the dangling bonds of amorphous silicon.

With the amorphous silicon on top, the samples were cleaned in acetone, ethanol, and DI water and etched in 5% HF for 3 min. HF etching is known to remove surface oxide layers and therefore supports the formation of an ohmic contact between a-Si and metal contacts.[14] Here, NiCr front contacts (32 nm thick) were sputtered (with the same parameters as mentioned earlier) on the amorphous silicon layer. The final sample structure is schematically shown in Figure 1.

The film thickness of the sputtered amorphous silicon was verified with a J. A. Woollam Inc. M-2000/T-solar variable angle spectroscopic ellipsometer. The samples were placed in the electrical characterization setup, where their back and front contacts were connected to a Keithley 2601A SourceMeter. Liquid indium–gallium (InGa) was used to improve the connection of the measurement device with the sample back contact, whereas the front sample contacts were connected through metal probes. The electrical characterization was conducted as a function of temperature in darkness in the temperature range from 150 to 350 K with a temperature step width of 10 K in vacuum conditions (p ≈ 10⁻¹ mbar).

3. Results

3.1. Dark Current–Voltage Measurements

To verify if NiCr provides ohmic contacts with the amorphous as well as crystalline silicon, current–voltage measurements were carried out on NiCr/a-Si and NiCr/c-Si test samples. Our measurements (not shown here) confirmed a linear ohmic behavior for NiCr/a-Si as well as the NiCr/c-Si junction.

As NiCr provides ohmic contacts, these contacts contribute to series resistance whereas the rectifying behavior observed stems from the a-Si/c-Si junction. We investigated the current density versus voltage dependence of the prepared samples. The measurements showed increased current densities with increasing temperature (see for example the sample prepared with a hydrogen flow rate of 0 sccm in Figure 2) as expected.

![Figure 1. Schematic side view of the sample structure with the connected measurement device.](image1)

![Figure 2. Typical temperature-dependent current density–voltage measurements of the a-Si/c-Si samples shown for a sample prepared without hydrogen flow.](image2)
for diodes. The current density–voltage measurement in the logarithmic plot also shows a clearly distinguishable linear region for low positive voltages, which disappears for higher temperatures.

### 3.2. Hydrogen Content

The hydrogen content of the prepared samples (Figure 3) was estimated from previous results reported by Schäfer. To estimate the hydrogen content at a deposition temperature of 430 °C, a function describing the hydrogen content as a function of the deposition temperature was extracted by linear fitting of the hydrogen content obtained from Fourier-transform infrared spectroscopy (FTIR) measurements for different hydrogen flow rates. This procedure then allowed the hydrogen content of the samples to be extracted that were prepared using a deposition at 430 °C by extrapolation.

It should be mentioned that the line shape of the silicon–hydrate stretching modes in the measured FTIR spectra did not change significantly with the hydrogen content.

### 3.3. Barrier Heights

The barrier height at the interface of the two semiconductors plays a key role for charge separation in solar cells. In the case of amorphous and crystalline silicon, the band offset is a critical parameter contributing to the barrier height. The bandgap of amorphous silicon can be changed by introducing hydrogen, as shown in earlier work, and herewith the band offsets. Furthermore, hydrogen saturates dangling silicon bonds and reduces therefore the density of trap states within the bandgap. The study of samples with varying hydrogen content is therefore a promising approach toward a better understanding of the role of hydrogen in amorphous silicon.

All measurements in this work were carried out on 5 nm p-type a-Si, including the samples used to determine the activation energy. A band bending will occur at the heterojunction and will certainly influence the barrier height at the interface. However, Dao et al. suggested that for interfaces with a high defect density, defect states within the bandgap play a significant role with respect to the barrier height. Therefore, our measured barrier height represents an “effective” barrier height considering band bending, band offset, bandgap widening, and defect state density.

To evaluate the influence of hydrogen on the barrier height between a-Si and c-Si, various flow rates of hydrogen were introduced during the sputtering deposition process. In the thermionic emission model, where only the height of the barrier but not its shape is considered, the current density follows

\[
J = J_0 \exp \left( \frac{-eV}{\eta kT} \right) - 1
\]

(1)

where \( V \) is the applied voltage, \( \eta \) is the ideality factor, \( k \) is Boltzmann’s constant, and \( T \) is the temperature.

The saturation current density \( J_0 \) is defined by

\[
J_0 = A^* T^2 \exp \left( \frac{\Phi}{kT} \right)
\]

(2)

where \( A^* \) is the effective Richardson constant for thermionic emission and \( \Phi \) is the barrier height.

By applying the natural logarithm, \( \ln(J_0 T^{-2}) \) over \( 1000/T \) is shown. The slope of the linear fit provides \( \Phi/k \) (Richardson plot). The corresponding graph is shown in Figure 4, where instead of \( J_0 \), rather, the directly measured saturation current \( I_s \) is shown.

The barrier heights extracted from the Richardson plot are shown in Table 1. It can be observed that for very low and very high hydrogen flow rates, the barrier heights are in the range of about 400 meV. In contrast, an intermediate flow rate of 5–10 sccm hydrogen induced low barrier heights ranging from about 100 to 200 meV.

### 3.4. Thermal Activation

The thermal activation energies give valuable insight into conduction mechanisms that govern the electrical conduction.

![Figure 3](image-url)  
**Figure 3.** Hydrogen content of thin films obtained by extrapolation from FTIR results presented in a study by Schäfer et al. As no data were available for samples sputtered with a hydrogen flow rate of 50 sccm, results for a hydrogen flow rate of 60 sccm are shown instead.

![Figure 4](image-url)  
**Figure 4.** Richardson plot of the a-Si/c-Si samples with different hydrogen flow rates.
A useful approach to calculate and analyses thermal activation energies is based on the analysis of the temperature dependence of electrical conductivity\cite{50}

\[ \sigma = \sigma_0 \exp \left( \frac{-E_A}{kT} \right) \]

with \( \sigma = \frac{1}{R} \) being the electrical conductivity and \( R \) the electrical resistance of the device.

The analysis of \( \ln(R/J) \) over \( 1000/T \) (Arrhenius plot) is a convenient way to extract the thermal activation energies \( E_A \) of the charge carriers involved in the transport (Figure 5).

The slope of the linear fit corresponds to the activation energies which are shown in Table 2. Films prepared with intermediate hydrogen flow rates lead to higher activation energies \( E_{A,2} \). Regarding \( E_{A,1} \), no relation to the hydrogen flow rate could be observed.

### 4. Discussion

The vanishing linear region in the current–voltage measurement indicates that the commonly used exponential dependence of the current density \( J \) on voltage \( V \) for diodes as in the thermionic emission model (1) is valid just in the low forward-bias region (\( V < 0.25 \text{ V} \)) at sufficiently low temperatures. This result is similar to the findings by Marsal et al.\cite{20} who observed the low forward-bias linear region for n-amorphous/p-crystalline silicon heterojunctions below 0.4 V and suggested that this can be attributed to the recombination at the amorphous side of the space-charge region. Song et al.\cite{45} located this conduction mechanism for voltages below 0.3 V. In contrast to other reported works,\cite{51,52} no indications for a second conduction mechanism within the low bias region such as multitunneling by capture emission (MTCE) were found. For higher forward bias, the current increases according to the power law \( I \approx V^n \), as described by Sze and Ng.\cite{45} Marsal et al.\cite{20} as well as Song et al.\cite{48} determined this region for voltages above 0.6 V and attributed this to space-charge-limited currents (SCLC) as the dominant transport mechanism for high applied voltages.\cite{51,54}

The estimated hydrogen content of the samples shows a subtle increase up to 10 sccm followed by a slight decrease; this variation is, however, within the error bars of our estimation (Figure 3) and should be treated with care. It is, nevertheless, worth mentioning that Schäfer\cite{43} and Habler et al.\cite{55} observed a decreasing hydrogen content for samples prepared with higher hydrogen flow rates. They attributed this observation to changes in the growth mode due to hydrogen excess.

For intermediate hydrogen flow rates in the range of 5–10 sccm, corresponding to a hydrogen content of about 3.5% present in the films, very low barriers were observed. The literature suggests that barrier heights are influenced by the density of defect states at the interface.\cite{19} Furthermore, the bandgap and consequently, the band offset of silicon layers can be altered by changing the hydrogen flow rate during thin-film deposition. From the data on hand, it cannot be determined as to which effects led to the observed barrier heights.

However, by analyzing the thermal activation energies, it can be observed that two different activation energies are involved in electrical conduction (Figure 5) and hence, two different conduction mechanisms. After Batabyal et al.\cite{15} and Nobis,\cite{56} activation energies \( E_{A,1} \) of about 100 meV correspond to the activation of the nearest neighbor hopping transport in the low-temperature range. Here, electrons perform hopping processes which take place within the mobility gap where charges can move via hopping processes between the localized states near the Fermi level.\cite{18} At higher temperatures, charge carriers conduct the electric current through delocalized states in the conduction band with typical activation energies \( E_{A,2} \) of about 500 meV.\cite{15,56,57} Assuming that the activation energy for band conduction can be interpreted as the difference between Fermi energy

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**Table 1.** Barrier heights for samples with different hydrogen flow rates obtained from the Richardson plot (Figure 2).

| Hydrogen flow rate [sccm] | Barrier height [meV] |
|---------------------------|---------------------|
| 0                         | 410 ± 12            |
| 5                         | 112 ± 6             |
| 10                        | 180 ± 9             |
| 20                        | 370 ± 6             |
| 50                        | 370 ± 10            |

**Table 2.** Activation energies for two different conduction mechanisms at low \( (E_{A,1}) \) and high temperatures \( (E_{A,2}) \).

| Hydrogen flow rate [sccm] | \( E_{A,1} \) [meV] | \( E_{A,2} \) [meV] |
|---------------------------|---------------------|---------------------|
| 0                         | 45 ± 4              | 243 ± 9             |
| 5                         | 19 ± 17             | 380 ± 15            |
| 10                        | 63 ± 5              | 493 ± 13            |
| 20                        | 129 ± 5             | 342 ± 13            |
| 50                        | 40 ± 5              | 200 ± 24            |

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**Figure 5.** Arrhenius plot of the a-Si/c-Si samples prepared using different hydrogen flow rates. The arrows indicate that the left linear fits correspond to low-temperature activation energies \( E_{A,1} \), whereas the ones on the right are used to determine high-temperature activation energies \( E_{A,2} \). The absolute transition temperature from \( E_{A,1} \) to \( E_{A,2} \) depends on the hydrogen flow rate.
$E_F$ and valence band $E_V$.\textsuperscript{[2]} The presented results agree with Dao et al.\textsuperscript{[3]} which determined a difference $E_F - E_V$ of 405 meV for a-Si:H/c-Si solar cells.

5. Conclusion

Amorphous p-type silicon films with a thickness of 5 nm were deposited via RF-pulsed magnetron sputtering on crystalline silicon. From deposition to deposition, the process parameter altered was the hydrogen flow rate. The hydrogen content present in the films in relation to the hydrogen flow rate was calculated using data published previously on films deposited using the same setup but different temperatures.\textsuperscript{[4]} The estimated hydrogen content of the samples shows only a slight increase up to 10 sccm followed by a slight decrease at higher hydrogen flow rates, which is within the error bars of determination.

A current–voltage dependence predicted by the thermionic emission model was found for the low forward-bias region below 0.25 V. This model was used to evaluate the influence of hydrogen flow rate on the barrier heights. An intermediate hydrogen flow rate of 5 sccm led to a minimal barrier height of (112 ± 6) meV, whereas for lower and higher flow rates the barrier was significantly increased to about 400 meV. It remains unclear whether the saturation of interface defects states or a bandgap widening are responsible for the observed barrier height characteristics.

Temperature-dependent current–voltage measurements revealed that the electrical conduction mechanism changes from nearest neighbor hopping (for low temperatures) to band conduction at around 200–250 K, depending on the amount of hydrogen atoms present.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

a-Si/c-Si heterojunctions, electrical measurements, interface properties, semiconductor heterojunctions, solar cells

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