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# Crystalline Volume Fraction Effect on the Electronic Properties of Hydrogenated Microcrystalline Silicon μc-Si:H Investigated by Ellipsometry and AMPS-1D Simulation

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The main objective of the present work is to study experimentally and by simulation, using the one dimensional analysis of microelectronic and photonic structures program (AMPS-1D), the correlation between the crystalline volume fraction ($F_c$) and the transport properties of hydrogenated microcrystalline silicon thin films (μc-Si:H). The $F_c$ was determined by spectroscopic ellipsometry (SE) and the electrical conductivity measurements. The μc-Si:H samples were deposited by radio-frequency magnetron sputtering technique of a crystalline silicon target, under an argon (Ar) gas mixture of 70 % of hydrogen ($H_2$) and 30 % of Ar, at three different total pressures (2, 3 and 4 Pa) and changing substrate temperatures (25, 100, 150 and 200 °C). The dark conductivity was measured in a coplanar configuration in an optical cryostat under applied electrical field and controlling current with an electrometer. In the simulation studies of the dark conductivity using the AMPS-1D, we modelled the films as an alternation of amorphous and crystalline regions with different crystalline volume fractions Fc (from 0 to 80 %). The results evidently demonstrated that the conductivity depends on the width of the area separating amorphous and crystalline regions. We found a strong correlation between the μc-Si:H films activation energy and the crystalline volume fraction where the grain size-to-thickness ratio plays a crucial role.

**Keywords:** Microcrystalline structure, Amorphous silicon, Dark conductivity, AMPS-1D, Activation energy, Electrical properties.

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## 1. INTRODUCTION

Actually, the intrinsic microcrystalline silicon was mainly studied in devices with thin film solar cells [1, 2], transistors [3] and diodes [4]. Several applications of hydrogenated amorphous silicon are replaced by microcrystalline silicon in our days because of its stability under light shining and its great conductivity. A proper description of the structural properties of such material can be quite a complicated task, as shape and size of the crystalline grains can be highly variable and their distribution within the amorphous matrix is probably different for each sample. Microcrystalline silicon is a two-phase material. Its composition can be interpreted as a series of grains of crystalline silicon imbedded in an amorphous silicon matrix, with a high concentration of dangling bonds in the transition regions [5, 6]. Hence, each phase of μc-Si:H plays an important role in electronic transport.

This work presents a study of the effect of the microstructure on the transport properties by the conductivity measurements and AMPS-1D simulation program used to determine the activation energy. The latter was determined recently by C. Qingdong et al. [7].

## 2. EXPERIMENTAL DETAILS

A series of undoped μc-Si:H thin films with different thicknesses were deposited on corning substrates at temperatures ($T_s$) of (25-150-100-200 °C) in a parallel-plate glow discharge plasma deposition system operating at a standard rf frequency of 13.56 MHz using Ar and $H_2$ as feed gases. Film thickness was calculated from the interference pattern of the optical transmission spectrum in the visible and near infrared regions. The microstructural studies were carried out using spectroscopic ellipsometry (SE) and X-ray diffraction (XRD). The dark conductivity $\sigma_d(T)$ measurements were carried out on a large number of annealed samples with different thicknesses, microstructures and morphological properties, using coplanar geometry in different experimental set-ups (above room temperature, 300-450 K). In the temperature range above room temperature, $\sigma_d(T)$ follows Arrhenius type thermally activated behavior: $\sigma_d(T) = \sigma_0 \cdot \exp(-E_a/kT)$, where $\sigma_0$ is known as the conductivity prefactor and $E_a$ as the activation energy. In the simulation study of the dark conductivity by the AMPS-1D program, samples were modelled as an alter-nation of amorphous and crystalline phases with different crystalline volume fractions ($F_c$) from 0 to 80 %.

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## 3. RESULTS AN DISCUSSION

The average of µc-Si:H grain size ($\delta$) was evaluated from XRD measurements. For the series of intrinsic samples deposited at 3 and 4 Pa, $\delta$ varies from ~ 6 nm to ~ 9.5 nm, respectively. This result allows to classify our samples into three categories, types of microstructure that can be visualized as Type 1: films having small grains and amorphous tissues and Type 2: films having closely packed columnar crystals consisting of big grains with very less hydrogenated amorphous silicon a-Si:H tissues in the columnar boundary region ($\delta$ > 6nm) and amorphous tissues Type (see Fig. 1).

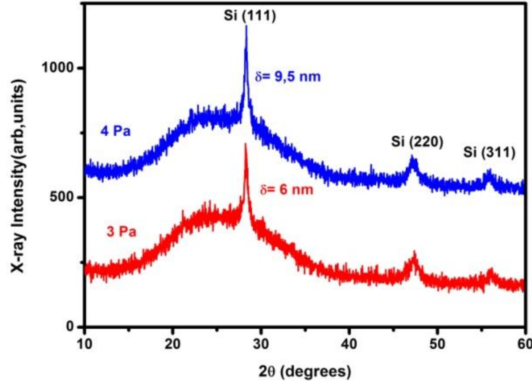



**Fig. 1** – XRD spectra in µc-Si:H samples deposited at 200 °C, varying pressure between 3 and 4 Pa

XRD was employed to characterize the samples. Three preferred orientations in the (111), (200) and (311) directions are observed. The crystallite size in the as deposited state is estimated from the full-width-half-maximum (FWHM) of the (111)-peak. In the material, microstructural studies have been demonstrated the presence of small and large crystallite grains and a moderate amount of disorder phase in the columnar boundary regions. Using the Bruggeman effective medium approximation and the Tauc-Lorentz model, we analyzed the data of spectroscopic ellipsometry (Fig. 2). The results obtained strongly indicate that the samples deposited at 2 Pa, are completely amorphous.

When the pressure is raised up to 3 and 4 Pa under different $T_s$ the presence, in these films, of a mixture of amorphous as well as crystalline silicon structure with different grain size. They clearly indicate that crystallization occurs in all samples elaborated at 3 and 4 Pa at $T_s$ less than room temperature, and there is a combined effect of both pressure and temperature on the crystalline fraction. The samples are well crystallized with a crystalline volume fraction varying from about 60 to 90 %. The substrate deposition temperature has nearly no clear effect either on the size of the crystallites or on the average crystalline volume fractions. The results obtained from spectroscopic measurements are summarized in Table 1.

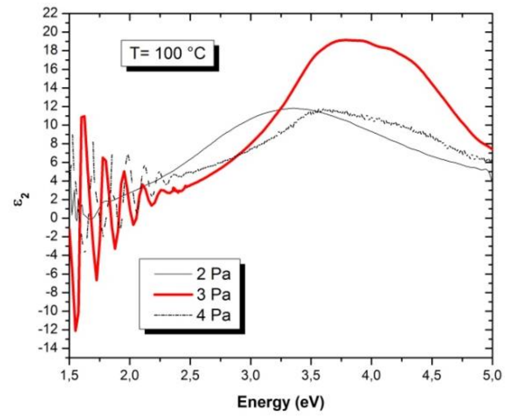



**Fig. 2** – Spectroscopic ellipsometry in µc-Si:H samples deposited at 100 °C, varying pressure between 2, 3 and 4 Pa

**Table 1** – Summary of the growth conditions, thickness, $F_c$, values of the grain size and activation energy

| **Pressure, Pa** | 2 | | 3 | | | | 4 | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| $T_s$, °C | 100 | 200 | 25 | 100 | 150 | 200 | 25 | 100 | 150 | 200 |
| $d_f$, µm) | 5.23 | 4.95 | 0.85 | 0.98 | 0.98 | 0.85 | 1.04 | 0.85 | 0.91 | 0.91 |
| $F_c$, % | 0 | 0 | 71 | 85 | 87 | 86 | 76 | 60 | 59 | 62 |
| $\delta$, nm | 0 | 0 | 7.3 | 3.7 | 6.1 | 6.0 | 9.01 | 12.5 | 10 | 9.7 |
| $(\delta/d_f)\cdot 10^{-3}$ | 0 | 0 | 8.6 | 3.8 | 6.22 | 5.88 | 8.66 | 14.7 | 10.98 | 10.44 |
| $E_a$, eV | 0.99 | 0.83 | 0.380 | 0.086 | 0.390 | 0.193 | 0.400 | 0.130 | 0.199 | 0.216 |

In order to understand the effect of the $F_c$ on the electronic properties of the µc-Si:H films, a detailed study has been conducted. Structural analysis spectroscopic ellipsometry combined with electrical measurements of the dark conductivity were used to characterize the films. Figs. 3a, 3b, 3c show the dark conductivity as a function of temperature measured in a coplanar configuration in an optical cryostat under applied electrical field of $10^3$ V/cm and controlling current with an electrometer. All the samples show one regime of conduction in the temperature range varying between 290 and 420 K. The conductivity follows an Arrhenius-type thermally activated behaviour $\sigma_d(T) = \sigma_0 \cdot \exp(-E_a/kT)$, where $\sigma_0$ is known as conductivity pre-factor and $E_a$ as activation energy. $E_a$ and $\sigma_0$ are valuables for understanding the mechanism and physics of electrical transport in the material. For the samples deposited at 2 Pa, the dark conductivity curves show an activated energy around 0.9 eV. The latter clearly indicates that these samples are amorphous. The results are in good agreement with spectroscopic ellipsometry spectra.

In general, samples deposited at 4 Pa ($T_s$ = 150 and 200 °C) have nearly the same $F_c$ and the same $E_a$. However, in the case of 3 Pa samples have a high $F_c$ ($T_s$ = 150 and 200 °C), it is clear that they have the same $F_c$ and different $E_a$. Thus we introduced the crystallite size-to-thickness ratio ($\delta/d_f$) to explain precisely the dependence of activation energy because the $F_c$ is not the only parameter that can affect somehow the $E_a$.

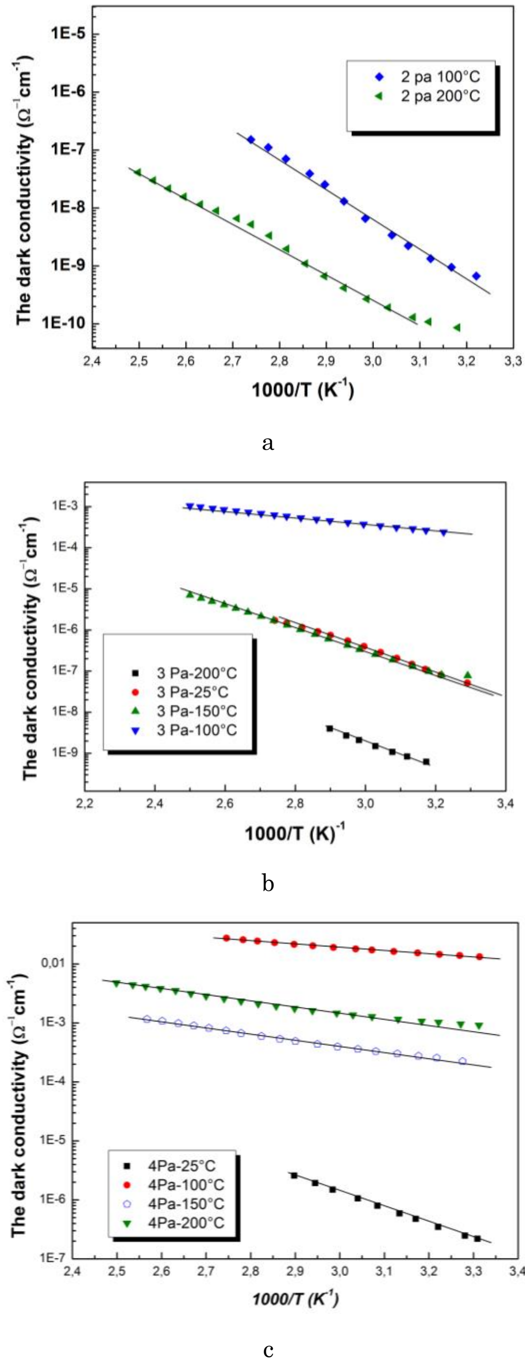



**Fig. 3** – The dark conductivity of the samples deposited at 2 (a), 3 (b) and 4 Pa (c) under different $T_s$

According to the curves representing samples deposited at 3 Pa (25 and 150 °C) and 4 Pa (25 °C), it is shown that the samples have an activation energy corresponding to around 0.4 eV. The samples deposited at 3 Pa (200 °C) and 4 Pa (150, 200 °C) have activation energy around 0.2 eV. The difference in activation energy is due to two different conduction mechanisms operating in the temperature measurement range. It appears that the conduction does not depend on deposition temperature as indicated in Table 1. The connection between the conductivity and the deposition pressure is not that obvious, however it becomes clear with the crystallite rate.

The activation energy value of 0.9 eV found for samples deposited at 2 Pa is a feature of intrinsic amorphous. When the crystallization takes place, the $E_a$ value decreases gradually around 0.4 eV for the $F_c$ situated between 70 % and 90 % with some exceptions. In this case, the dark conductivity is limited by the potential barrier between the amorphous and crystalline regions. The band gap of crystalline silicon (1.12 eV) is smaller than the band gap of a-Si:H (1.8 eV). Therefore, the barriers at grain boundaries are due to the offset value of conduction band (Fig. 4b).

There are some samples which contain around 60 % of $F_c$, the activation energy close to 0.2 eV excepting sample deposited at 3 Pa and 200 °C. The quite small value of $E_a$ might be due to the fact that the Fermi level $E_f$ is pinned near the conduction band by defects in the distorted network at the transition between crystallites and the surrounding amorphous matrix as it has been previously suggested [8, 9]. On the other hand, this small value of $E_a$ has been discussed by several authors who claim that conduction transport of intrinsic μc-Si:H is attributed by Variable Range Hopping VRH transport in the temperature range of 270 to 450 K [10]. From data analysis, it has been obtained that the $F_c$ affects the hopping conduction among the clusters of grains, where the significant $F_c$ exceeds 60 %, leads to separating distance between the grain boundaries of a few atomic spacings which can favor either the VRH or the tunnel conduction.

To explore the correspondence between the $F_c$ and electrical transport behaviour, we have based on the simulation studies of the dark conductivity by the AMPS-1D program. We considered all the modelled samples as an alternation of amorphous and crystalline regions with different crystalline volume fractions varying between 0 % and 80 % [11]. The one dimensional device simulation program AMPS-1D solves simultaneously the Poisson equation and the electron and the hole continuity equations by using the method of finite differences and the Newton-Raphson numerical method. The recombination and the trapping mechanism are described by the Shockley-Read-Hall model [12, 13]. We note that, neither tunnelling nor the VRH conduction is contained by AMPS-1D program. The parameters used to simulate the device layers are summarized in Table 2 and all the simulation findings are reported in Table 3.

The simulation results obtained show that the activation energy is nearly in agreement with the experiment results. It continuously decreases from 0.9 eV at $F_c$ less than 10 % till the value of 0.3 eV for the $F_c$ extremely more than 80 % as reported in Fig. 4a. As soon as the volume crystalline fraction takes place, the offsets between the amorphous and crystallites region appear.

We modeled, using AMPS-1D simulation, the films of μc-Si:H as an alternation of amorphous and crystalline silicon a-Si:H/c-Si with three values of $F_c$ ($F_c$ = 10, 60 and 80 %). The discontinuity of bands is numerically calculated. The offsets are considered as a high barrier which reaches 0.5 eV when the $F_c$ ranges from 10 % to 80 %. However, the edge of the barrier is not straight, but it has a curve shape as indicated in Fig. 4b. That is

**Table 2** – Set of parameters used to simulate μc-Si:H

| Parameters | Materials | |
|---|---|---|
| | a-Si:H (i) | c-Si (i) |
| Electronic affinity, eV | 3.93 | 4.01 |
| Mobility gap, eV | 1.80 | 1.12 |
| Effective density of state $N_c$, $N_v$, cm$^{-3}$ | $10^{21}/10^{21}$ | $(4.3/1.6)10^{19}$ |
| Electron mobility, cm$^3$/V/S | 20 | 1250 |
| Hole mobility, cm$^3$/V/S | 02 | 450 |
| The DOS in donor-like Gaussian, cm$^{-3}$ | $8.10^{15}$ | $10^{12}$ |
| The DOS in acceptor-like Gaussian, cm$^{-3}$ | $8.10^{15}$ | $10^{12}$ |
| Standard deviation in donor-like Gaussian, eV | 0.10 | 0.015 |
| Standard deviation in acceptor-like Gaussian, eV | 0.10 | 0.015 |
| Position donor-like Gaussian from $E_c$, eV | 0.87 | 0.46 |
| Position donor-like Gaussian from $Ev$, eV | 0.89 | 0.66 |
| Energy slope donor-like bandtail, eV | 0.048 | // |
| Energy slope donor-like bandtail, eV | 0.036 | // |

**Table 3** – The simulation results

| Crystalline volume fraction $F_c$, % | Activation energy $E_a$, eV |
|---|---|
| a-Si:H | 0.89 |
| 10 | 0.89 |
| 20 | 0.87 |
| 30 | 0.85 |
| 40 | 0.81 |
| 50 | 0.75 |
| 60 | 0.65 |
| 70 | 0.48 |
| 80 | 0.31 |

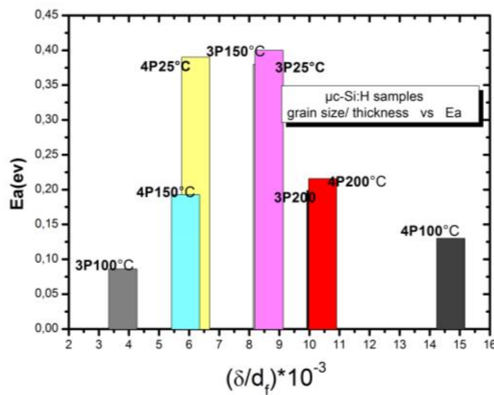
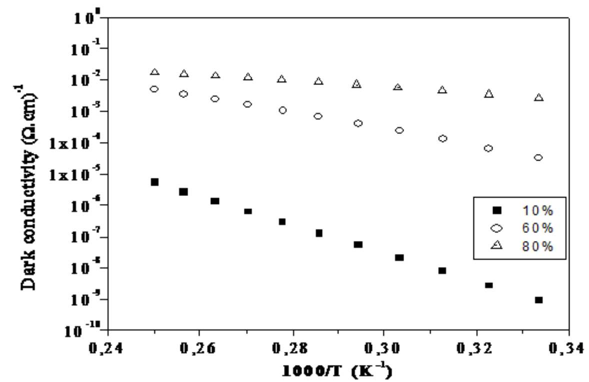


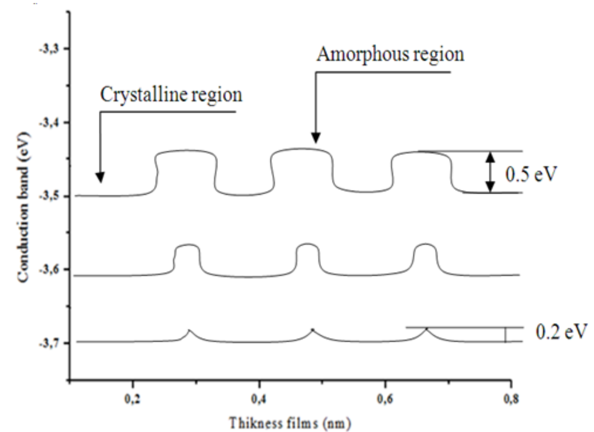

**Fig. 4** –The variation of the activation energy of μc-Si:H samples versus the report (grain size/ thickness)

Similarly, no systematic variation in $F_c$ and $E_a$ is observed for the μc-Si:H samples (Table 1). The samples deposited at 4 Pa (150 and 200 °C) have approximately the same $F_c$ ~ 60 %, $d_f$ thickness and $E_a$. However in the case of 3 Pa (150 and 200 °C), the samples have nearly the same $F_c$ ~ 86 %, δ grain size and a big difference in $E_a$. Fig. 4 indicated the variation of $E_a$ as a function of ($\delta/d_f$). According to this graph one can summarize the following important results:

The variation of $E_a$ as a function of ($\delta/d_f$) has a




a




b

**Fig. 4** – The dark conductivity simulation for $F_c$ (10 %, 60 % and 80 %) (a) and a band diagram for a layer of a device simulated (b)

Gaussian form.

1. The samples deposited at room temperature are situated at the top of the Gaussian.
2. The samples elaborated at 100 °C are situated in the bottom of the Gaussian.
3. The samples obtained at 200 °C are situated in the half-height of the Gaussian.

On the other hand, the samples deposited at 150 °C are in the middle, between the top and the half-height of the Gaussian, and this means that the activation energy $E_a$ is related to the deposition conditions such as pressure and temperature.

## 4. CONCLUSIONS

In the present work, the hydrogenated microcrystalline thin films, prepared using rf magnetron sputtering technique, were investigated by means of spectroscopic methods and using AMPS-1D simulation program to determine the grain size, crystalline volume fraction and the activation energy. The findings clearly indicated that the conduction process strongly depends on the offsets, between amorphous and crystalline phases, considered as a potential barrier. The μc-Si:H films activation energy does depend on the crystalline volume fraction and strongly related to the ratio of grain size-to-thickness.

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## Вплив кристалічної об'ємної фракції на електронні властивості гідрогенізованого мікрокристалічного кремнію, дослідженого методом еліпсометрії та моделюванням мікроелектронних і фотонних структур

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Метою даної роботи є експериментальне і модельне дослідження кореляції між об'ємною фракцією кристалів і транспортними властивостями тонких плівок гідрогенізованого мікрокристалічного кремнію, використовуючи одновимірний аналіз програми мікроелектронних і фотонних структур (AMPS-1D). Об'ємну фракцію кристалів визначали спектроскопічною еліпсометрією і вимірюваннями електропровідності. Зразки гідрогенізованого мікрокристалічного кремнію осаджували методом радіочастотного магнетронного розпилення кристалічної мішені кремнію у газовій суміші аргону і водню при трьох різних сумарних тисках (2, 3 і 4 Па) та змінюючи температуру підкладки (25, 100, 150 і 200 °C). Темна провідність вимірювалася в копланарній конфігурації в оптичному кріостаті при прикладанні електричного поля і керуючого струму. У дослідженнях темної провідності з використанням AMPS-1D ми моделювали плівки як чергування аморфних і кристалічних областей з різними кристалічними об'ємними фракціями (від 0 до 80 %). Результати показали, що провідність залежить від ширини ділянки, що розділяє аморфні та кристалічні області. Була виявлена сильна кореляція між енергією активації плівок гідрогенізованого мікрокристалічного кремнію і об'ємною часткою кристалічних речовин, де співвідношення розміру зерна до його товщини грає вирішальну роль.

**Ключові слова:** Мікрокристалічна структура, Аморфний кремній, Темна провідність, AMPS-1D, Енергія активації, Електричні властивості.