

*Full Length Research Paper*

# Optical absorption of the hydrogenated evaporated amorphous silicon

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**This study focuses on the study characteristic of the optical absorption of thin films of evaporated hydrogenated amorphous silicon (a-Si: H) prepared in a built Ultra-High Vacuum (UHV). Atomic hydrogen is produced using a plasma discharge tube directed towards the substrate holder. The optical absorption coefficient was obtained in the range of 0.5 to 2.5 eV using optical transmission measurements and Photothermal Deflection Spectroscopy (PDS). The results obtained are comparable to those of films prepared by decomposition of silane. The refractive index of the films decreases when the concentration of hydrogen increases.  $E_G$  bandwidth increases (1.45 to 1.60 eV), the parameter threshold Urbach  $E_0$  decreases (from 149 to 62 meV) and finally the number of spins ( $4 \times 10^{19}$  to  $6.5 \times 10^{16} \text{ cm}^{-3}$ ) depending on the concentration of hydrogen increases.**

**Key words:** Hydrogenated amorphous silicon, optical absorption, density of electronic states, urbach absorption.

## INTRODUCTION

The pure evaporated or sputtered amorphous silicon (a-Si, amorphous silicon without hydrogen) is a material that contains a high concentration of dangling bonds on the order of  $10^{19} \text{ cm}^{-3}$  which gives rise to an electronic paramagnetic resonance signal (Brodsky and Tittle, 1969; Thomas et al., 1974) and it also exists a weak covalent bonds concentration of the order of  $3 \times 10^{21} \text{ cm}^{-3}$  (Thomas et al., 1978; Kaplan et al., 1978; Street, 1991; Nickel, 1999). The electron energy levels of the dangling bond lie in between the bonding (valence band) and antibonding (conduction band) and give high optical absorption at low photon energy (Kré et al., 2007). Furthermore, the dangling bonds have the effect of pinning the Fermi level thereby reducing the doping efficiency in pure amorphous silicon. These defects produce a large electronic density of states in the normally forbidden band gap which makes evaporated or sputtered a-Si worthless for electronic devices.

Over the years, it has been demonstrated that the midgap electronic density of states in amorphous silicon can be reduced by several orders of magnitude when

hydrogen is introduced during preparation (deposition) of the material. Hydrogen passivates the dangling bonds making it possible for the resulting alloy, hydrogenated amorphous silicon (a-Si: H) to be doped by means of boron and phosphorus, has properties which are similar to that of crystalline semiconductors as photoconductive properties (Street, 1991). In other words the hydrogen in a-Si: H is responsible for the fact that low defect densities of  $10^{16} \text{ cm}^{-3}$  (Stutzmann, 1989) can be obtained compared to pure a-Si. However, hydrogen not only saturates the dangling bonds and decreases the density of defect states but may also play active roles in reducing the structural disorder, enlarging the band gap and participating in the carrier induced metastable degradation of a-Si: H electrical properties. Therefore, to make this material adaptable for electronic and optoelectronic applications, the presence of hydrogen in the network of a-Si is necessary (Dong and Drabold, 1998; Fedders and Drabold, 1993; Biswas et al., 1989; Fedders and Carlsson, 1989). However, the action of hydrogen on the electronic properties of a-Si is still relevant (Kolodziej, 2004; Biswas and Pan, 2003; Fritzsche, 1995; Staebler and Wronski, 1980) because the properties of films of a-Si and a-Si: H depends not only on the technique but also the deposition parameters (Acco et al., 1996; Leadbetter

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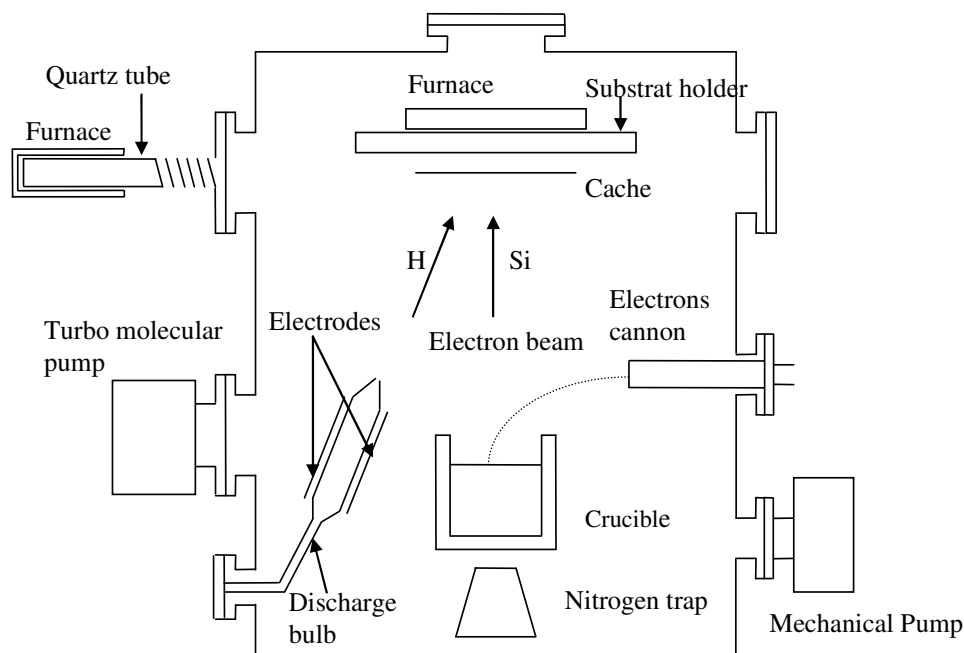


Figure 1. Diagram of the evaporation and hydrogenation of the films of a-Si.

et al., 1981).

The hydrogenated amorphous silicon a-Si: H and microcrystalline thin film are generally obtained by chemical deposition vapor plasma (PECVD for "Plasma Enhanced Chemical Vapor Deposition) (Shinar et al., 1994). Deposition techniques used for a-Si: H are radio frequency (13.56 MHz) capacitively coupled plasma enhanced chemical vapor deposition RFPECVD (Vanecek et al., 1991) with growth rates are around 1 - 3 Å. However, efforts of increasing the RFPECVD growth rate by means of extreme deposition conditions (higher power and pressure) are used as, very high frequency (VHF) range (30 - 300 MHz) PECVD (VHFPECVD) (Mai et al., 2005), glow discharge PECVD (GWPECVD) (Gallagher, 1988; Finger et al., 1992), microwave down stream CVD (MWCVD) (Johnson et al., 1991; Jasinski, 1995), electron cyclotron resonance CVD (ECRCVD) (Zhang et al., 1995; Dalal et al., 1993), reactive magnetron sputtering (RMS) (Liang et al., 1994), hot wire CVD (HWCVD) (Vanecek et al., 1991; Mahan et al., 1991; Deng and Povolny, 2003), ion implantation at different H concentrations (Acco et al., 1996; Will et al., 1998) and the remote expanding thermal plasma (ETP) deposition technique (Severens et al., 1995). Growth rates of up to 30 Å/s have been reported with good film properties (Heintze and Zedlitz, 1993; van Sark et al., 1995; Kroll et al., 1998). Although there are many differences between these techniques, the material properties of the a-Si: H deposited are very similar.

Concerning the hydrogenation of pure evaporated thin films of a-Si, generally two techniques are used: the post-hydrogenation (Kaplan et al., 1978; Thomas, 1980) and the hydrogenation during the growth of the layers. The

first permits to get a material whose properties are similar to those of a-Si: H prepared by silane decomposition was obtained after atomic hydrogen diffusion into a-Si evaporated in ultra high vacuum (UHV) however; this technique requires very thin films.

The hydrogenation during the growth of the layers allows to obtain films of pure a-Si: H with a large range of hydrogen concentration. This technique has the advantage to get some thin films with evaporation of a-Si of the order of 10 Å/s (Kr e et al., 2007). Therefore, this technique could allow to obtain films of a-Si: H with optical properties comparable to those of the best samples of a-Si: H obtained by silane decomposition. So, thin films of a-Si: H which are the subject of our study were obtained by incorporate hydrogen during deposition of films of a-Si.

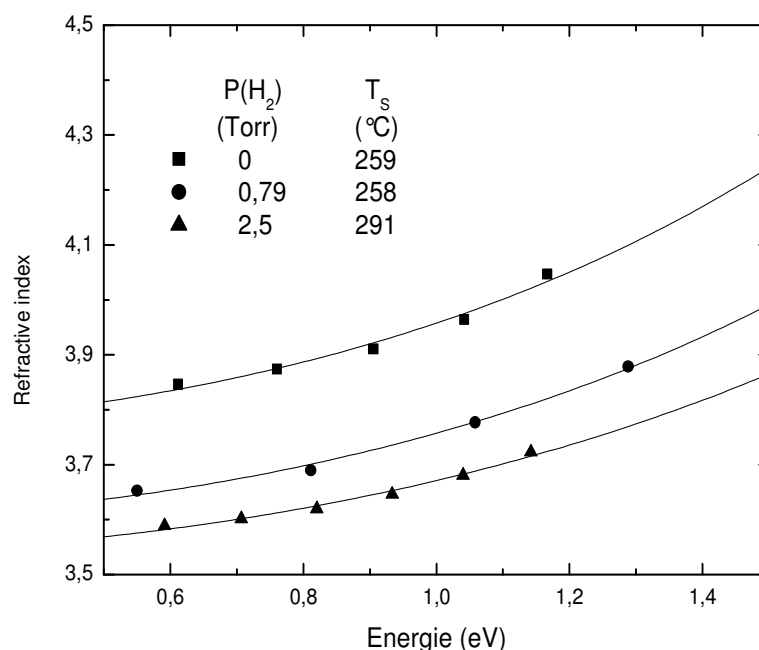
In this work, we try to characterize these thin films of a-Si: H using transmission measurements and optical reflection of the photothermal deflection spectroscopy (PDS) (Jackson et al., 1981; Jackson and Amer, 1982) and electron paramagnetic resonance spectroscopy (EPR) (Thomas et al., 1978).

#### PREPARATION OF a-Si: H FILMS

The films of a-Si: H were prepared in ultrahigh vacuum UHV chamber (Figure 1). A discharge tube containing hydrogen gas is directed towards the substrate holder. The discharge tube was performed using a generator Radio Frequency (RF) with a frequency of 13.56 MHz using a capacitive coupling. An impedance matching is used to connect the generator to the electrodes. The power generator at the deposit was 30 W. Before each series of repository, a degassing process is carried out of the frame (Kr e et al., 2007). After all these operations of degassing and steaming, we get a

**Table 1.** Deposition parameters of films of a-Si: H.

Film	P(H <sub>2</sub> ) (Torr)	P(frame) (10 <sup>-5</sup> Torr)	T <sub>S</sub> (°C)	Thickness (nm)	V <sub>S</sub> (Å/s)
A	0	5.1x10 <sup>-3</sup>	259	1.05	2.5
B	0.6	0.35	257	0.62	3.8
C	0.49	0.95	256	0.88	3.2
D	0.79	1.45	258	1.12	3.9
E	1.2	2.13	259	0.77	2.4
F	2.4	4.73	262	1.73	1.9
G	2.5	4.78	291	1.46	1.6


**Figure 2.** Change in refractive index as a function of energy for two films of a-Si:H and that of a-Si.

pressure that is located in the range of  $10^{-10}$  to  $4 \times 10^{-10}$  Torr before deposition. Remember that many factors are crucial in preparing film a-Si:H: the hydrogen pressure in the discharge tube  $P(H_2)$ , the flow of hydrogen characterized by the opening of the bulb. The pressure in the P (frame) during deposition, the substrate temperature  $T_S$ , the evaporation rate of silicon  $V_S$  and finally the electric power generator  $P_W$ . The rates of evaporation (2 - 4 Å/s) are superior to those of a-Si: H commercial films with frequency of 13.56 MHz.

For this study, we have chosen primarily to vary the pressure of hydrogen  $P(H_2)$  for a given hydrogen flow and keep the possible other parameters constant. We chose a substrate temperature of about 260°C close to that to optimize the properties of samples obtained by decomposition of silane. The deposition parameters of films of a-Si: H deposited on quartz substrates are shown in Table 1.

It should be noted, first, that the film B was obtained with a discharge tube differs from that used for other samples on the other hand, the film G was obtained at a temperature of 291°C higher than that of other samples and finally the film A is a film of pure a-Si.

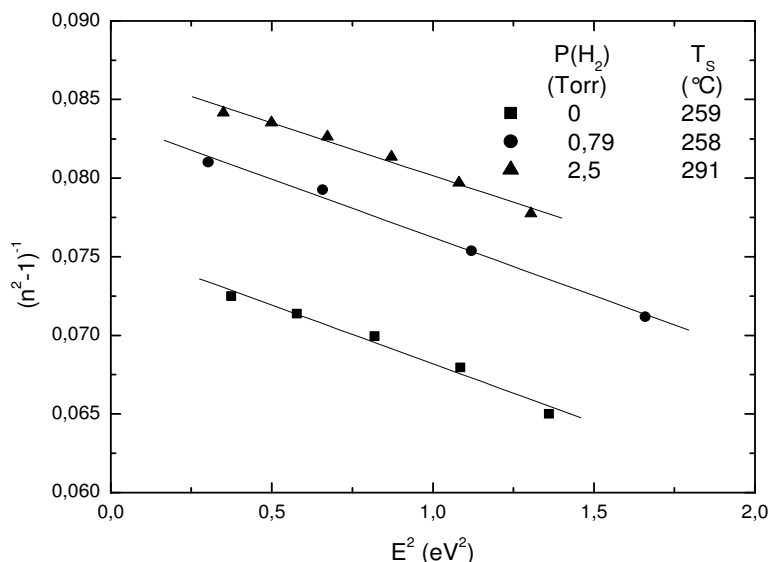
## RESULTS AND DISCUSSION

### Refractive index as a function of energy

The refractive index of films in the region of low energies has been obtained from transmission spectra and reflection optics (Swanepoel, 1983). Figure 2 shows the variation of the refractive index ( $n$ ) versus photon energy for two samples of a-Si: H and that of a-Si. The variation of the refractive index as a function of energy was adjusted with the simple oscillator model of Wemple-DiDomenico (Wemple and DiDomenico, 1971).

$$\varepsilon_1(\hbar\omega) = n^2(\hbar\omega) = 1 + \frac{E_0 E_d}{E_0^2 - (\hbar\omega)^2} \quad (1)$$

where  $E_0$  is the oscillator energy (almost energy peak of



**Figure 3.** Curves of variation of  $(n^2 - 1)^{-1}$  depending on  $(\hbar\omega)^2$ .

the spectrum from  $\varepsilon_2(\hbar\omega)$  and  $E_d$  the dispersion energy or the oscillator strength. Representing  $(n^2 - 1)^{-1}$  as a function of  $E = \hbar\omega$ .

We adjust the points with a linear function (Figure 3). The parameters  $E_0$  and  $E_d$  are directly determined from the slope  $(E_0 E_d)^{-1}$  and intercept  $E_0/E_d$ . The value of the refractive index for  $\hbar\omega = 0$  ( $\lambda \rightarrow \infty$ ) is determined using the following expression:

$$n(0) = \left(1 + \frac{E_d}{E_0}\right)^{1/2} \quad (2)$$

The values of dispersion parameters are listed in Table 2. The results of Table 2 show that  $E_d$  decreases while  $E_0$  increases with increasing hydrogen in the films. According to Wemple and DiDomenico (1971), the parameter  $E_d$  follows the following empirical relationship:

$$E_d = \beta N_c Z_a N_e \quad (3)$$

where  $N_c$  is the coordination number,  $Z_a$ , chemical valence,  $N_e$  the effective number of valence electrons per atom and  $\beta$  a multiplier ranging from 0.32 for germanium to 0.39 for diamond. Using the values of  $N_c = 4$ ,  $Z_a = 4$  and  $\beta = 0.36$ , the values of  $N_e$  calculated are almost equal to 7 for all three samples.

This constant value could suggest that hydrogen does not induce a rearrangement of the effective number of valence electrons. The increase of  $E_0$  may suggest a broadening of the optical gap with the presence of hydrogen in the films. The values of refractive indices are

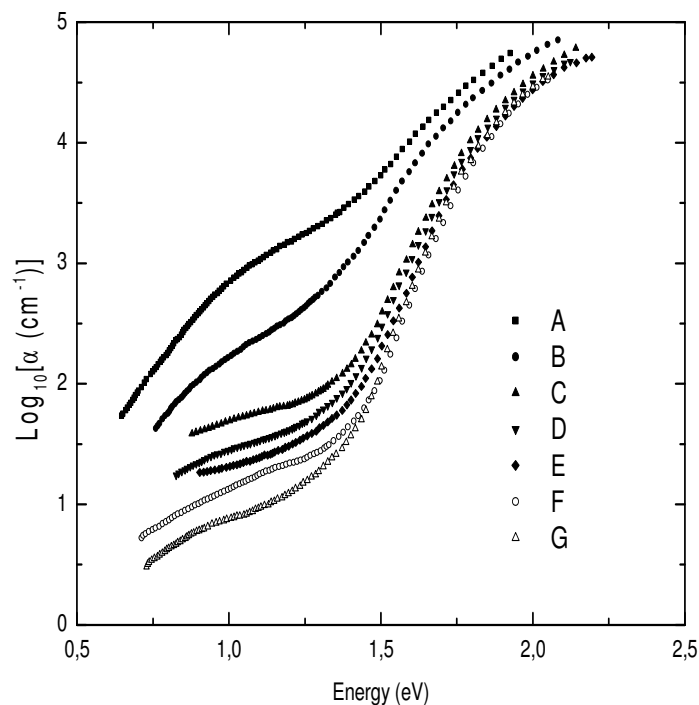
**Table 2.**  $E_d$ ,  $E_0$  and  $n(0)$  parameters of model Wemple-Di Domenico.

Film	$E_d$ (eV)	$E_0$ (eV)	$n(0)$
A	42.07	3.18	3.77
D	40.17	3.36	3.60
G	41.34	3.59	3.54

similar to those of samples of a-Si: H obtained by PECVD (Wronski et al., 1982; Jackson and Amer, 1982). Moreover, we observe a decrease in refractive index with increasing concentration of hydrogen. This variation is related to the decrease in atomic density of films of a-Si: H when the concentration of hydrogen increases (Freeman and Paul, 1979).

### Optical absorption spectra

The optical absorption spectra of films of a-Si: H are presented in Figure 4. The change of spectra as a function of hydrogen pressure in the discharge tube shows the change brought by the hydrogen in the formation of films. The shift of absorption edge towards higher energies indicates a broadening of the optical gap. For energies below the optical gap, we observe the exponential Urbach absorption of almost three orders of magnitude on the spectrum of the film G. The decrease of absorption at low energies suggests the reduction of deep states. To analyze this part of the spectra, we use the absorption coefficient corresponding to the energy 1.2 eV in comparison with the spin density. The optical gap and the width of Urbach threshold will be determined using methods commonly used.



**Figure 4.** Absorption spectra of films of a-Si: H.

**Table 3.** Values of optical gap  $E_G$ , the width of the absorption threshold of Urbach  $E_0$  and the spin density  $N_s$ .

Sample n°	P(H <sub>2</sub> ) (Torr)	$E_G$ (eV)	$E_0$ (meV)	$N_s$ (cm <sup>-3</sup> )
A	0	1.45	149	$4.03 \times 10^{19}$
B	0.61	1.50	113	$2.30 \times 10^{18}$
C	0.49	1.58	79	$3.22 \times 10^{17}$
D	0.79	1.59	76	$1.76 \times 10^{17}$
E	1.20	1.60	71	$1.54 \times 10^{17}$
F	2.40	1.63	65	$6.41 \times 10^{16}$
G	2.50	1.60	62	$6.49 \times 10^{16}$

### Optical gap

The optical gap values are determined using the model Tauc (Tauc et al., 1966) by extrapolating the linear part of the curve  $(\alpha\hbar\omega)^{1/2}$  according to  $\hbar\omega$  using the following equation:

$$(\alpha\hbar\omega)^{1/2} = B(\hbar\omega - E_G) \quad (4)$$

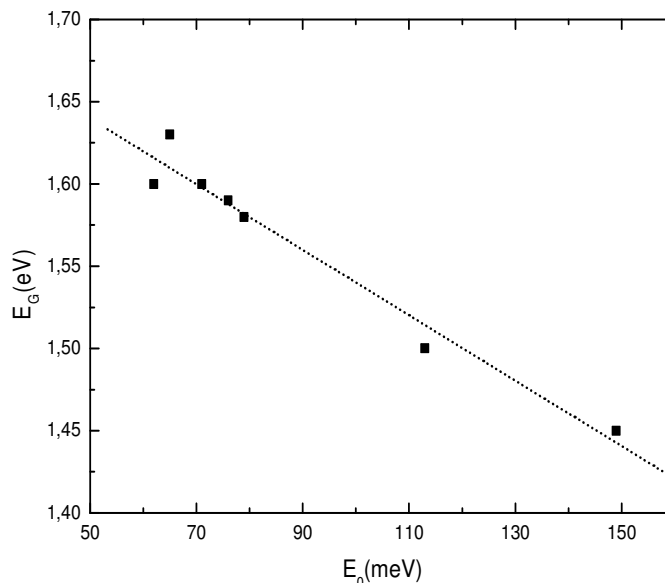
Assume in this case that the valence band and conduction are parabolic and the matrix elements of the momentum are constant. The value of  $E_G$  is the intersection of the linear part of the curve  $(\alpha\hbar\omega)^{1/2}$  with the energy axis. For all samples, the value of B is about  $3.6 \times 10^5 \text{ cm}^{-1/2} \text{ eV}^{1/2}$  which is consistent with that

obtained by other authors (Freeman and Paul, 1979). The values of  $E_G$  for all samples are presented in column 3 of Table 3.

### Width of the absorption Urbach edge

The energy width of the tails of localized states of absorption Urbach  $E_0$  quantifies the disorder in the films of a-Si: H (Wronski et al., 1982). It is determined by considering the exponential part of the spectrum defined by the following expression (Jackson and Amer, 1982; Cody et al., 1981; Abeles et al., 1980).

$$\alpha(\hbar\omega) = \alpha_0 \exp\left(\frac{\hbar\omega}{E_0}\right) \quad (5)$$



**Figure 5.** Variation of optical gap  $E_G$  according to the Urbach parameter  $E_0$ .

where  $\alpha_0$  is a constant.

An evaluation of  $E_0$  can be deduced from the plot of  $\ln \alpha$  versus  $\hbar\omega$ . The values of  $E_0$  including those of the spin density  $N_S$  samples studied were also recorded respectively in columns 4 and 5 of Table 3.

Compared with non-hydrogenated sample, we see a sharp decrease of the parameter  $E_0$  and over time increase the optical gap  $E_G$  when the pressure of hydrogen increases. The value of 62 meV of the sample 1083 is slightly higher than the value of 47 meV reported for samples of a-Si: H obtained by PECVD (Wronski et al., 1982; Jackson and Amer, 1982). However it is comparable to those of samples prepared by sputtering (Jousse et al., 1985).

Figure 5 shows a linear variation of optical gap as a function of the parameter of the Urbach following relationship:

$$E_G \text{ (eV)} = 1.74 - 2E_0 \text{ (eV)} \quad (6)$$

These results are in good agreement with the model Cody expressing that the main influence of hydrogen results from its effect on the disorder through relaxation of the network (Cody et al., 1981; Aljishi et al., 1990; Ewald et al., 1979).

### Deep defect density

To determine the nature of deep states, we have shown in Figure 6, the variation of spin density  $N_S$  depending on  $\alpha$  (1.2 eV), the absorption coefficient at 1.2 eV (Jackson and Amer, 1982). A linear correlation can be established between  $N_S$  and  $\alpha$  (1.2 eV) in the range of the measures:

$$N_S \text{ (cm}^{-3}\text{)} = 1 \times 10^{15} \alpha^{1.3501} \quad (7)$$

where  $\alpha$  is the absorption coefficient at 1.2 eV and is expressed in  $\text{cm}^{-1}$ .

The correlation between these two quantities shows that the absorption at low energy is mainly due to paramagnetic centers (spins) responsible for the EPR signal. These centers would be singly occupied dangling bonds. This conclusion is in good agreement with that Jackson and Amer (1982). It is important to note that the samples F and G have a spin density of about  $10^{17} \text{ cm}^{-3}$ . This value is close to that of the best films of a-Si: H obtained by decomposition of silane ( $10^{15}$  to  $10^{16} \text{ cm}^{-3}$ ).

### Conclusion

In this work, we characterized some optical properties and confirm the actual presence of hydrogen in samples of a-Si: H preparations other than by decomposition of silane. Indeed, the device is to evaporate the silicon in the presence of atomic hydrogen allows obtaining samples whose optical properties are comparable to the best samples obtained by decomposition of silane. We obtained spectra on a considerable decrease of absorption at low energy due to a decrease of deep states associated with dangling bonds. Changes in the absorption edge and a shift of spectra to high energies have been obtained. The best films of a-Si: H that we obtained have a lower concentration of defects at  $10^{17} \text{ cm}^{-3}$ . This value is comparable to film a-Si: H obtained by decomposition of silane ( $\sim 10^{16} \text{ cm}^{-3}$ ) while the spin density corresponding to non-hydrogenated layer is about  $10^{19} \text{ cm}^{-3}$ . A study on the nature of Si-H bonds

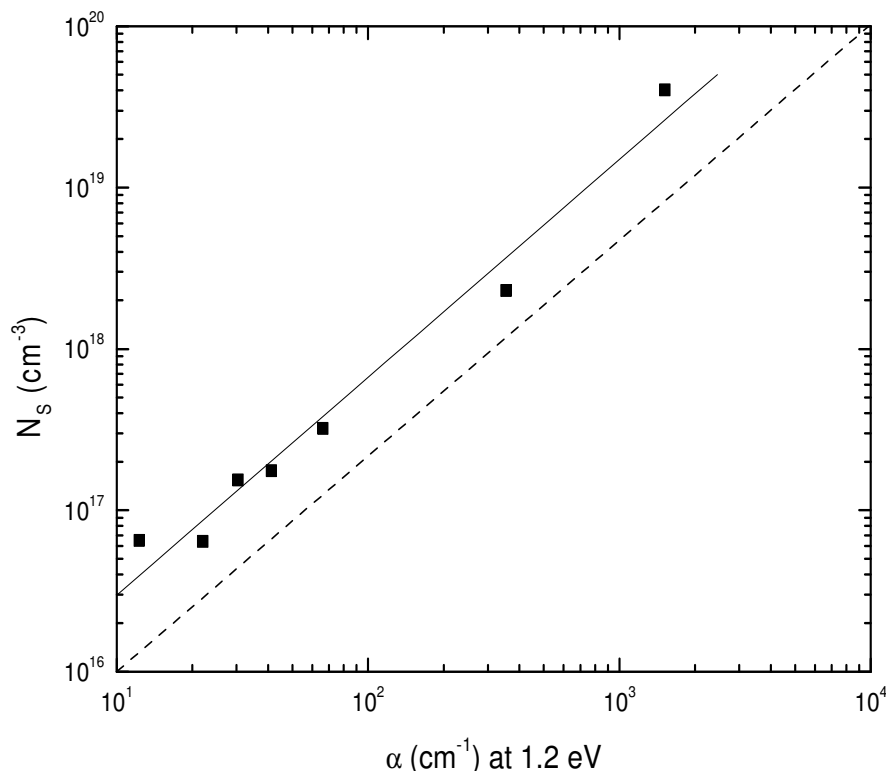


Figure 6. Spin density as a function of absorption coefficient at 1.2 eV.

should be considered to determine bonded hydrogen concentration in the films.

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