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جامعة المرقب

العدد العشرون
يناير 2022م

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Influence of Hydrogen content on structural and optical properties of doped nano-a-Si:H/a-Ge: H multilayers used in solar cells

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Abstract

Doped-nanomultilayers of a-Si:H/a-Ge:H thin films are used as a new type of narrow band gap materials for amorphous silicon-based solar cells. High efficiency solar cells are necessary to convert solar energy to electrical energy at low cost until. Doped-nanomultilayers of a-Si:H/a-Ge: H were prepared at 200°C by alternating deposition from SiH₄ and GeH₄ plasmas in a computer-controlled four chamber glow-discharge deposition system with capacitively coupled diode reactors. IR, XRD and SEM were used to study the structural changes after annealing at 300 for 8 h. The annealed doped-nanomultilayers exhibit surface and bulk degradation with formation of bumps and craters. The measurements results reveal that the optical energy gap is decreased with increasing the annealing temperature and/or time is partially due to formation of H bubbles in the Ge layers and partially due to crystallization effects.

Keywords Doped, nano-multilayers (NMLs), a-Si:H/a-Ge: H, hydrogen content (N_H), structure, crystallization, optical properties

I. Introduction

Amorphous silicon based semiconductors doped-nanomultilayers of great interest for device applications. doped-nanomultilayers of a-Si:H/a-Ge:H are used as a new type of narrow band gap materials for amorphous silicon-based solar cells. Since the quality of a-Si_{1-x}Ge_x:H is not as good as that of a-Si:H, some attempts were carried out to improve the optical and electrical properties of doped-nanomultilayers of a-Si_{1-x}Ge_x:H by producing it from ultrathin layers of doped-nanomultilayers of a-Si:H/a-Ge:H [1-7].



II. Experimental

The boron (B) and phosphorous (P) doped a-Si:H/a-Ge:H multilayers was deposited by alternating deposition from SiH₄ and GeH₄ mixed with B₂H₄ and PH₃ gases, respectively plasmas in a computer-controlled four chamber glow-discharge deposition system with capacitively coupled diode reactors. At a substrate temperature of 200 °C, a RF of 13.6 MHz, a RF power of 10 W, a pressure of about 0.18 mbar and a gas flow of 5 sccm in the SiH₄ chamber and 0.32 mbar and a gas flow of 0.25-2 sccm of GeH₄ mixed with 1% B₂H₄ to prepare p-type and a gas flow of 0.25-2 sccm of GeH₄ doped with 1% PH₃ for preparing n- type a-Si:H/a-Ge:H multilayers, P- and B-doped multilayers, with barrier layer thickness of 3 nm and well layer thickness of 2.8 and 3 nm were prepared for the measurements.

The individual thickness of a-Si:H barrier layer dSi was varied by changing the deposition time, while the well layer thickness dGe of a-Ge:H was controlled by changing the hydrogen dilution ratio [H₂]/[GeH₄] as well as by changing the deposition time. The growth rate was kept near 0.1 nms⁻¹ for a-Si:H and ranged from 0.1 to 0.4 nms⁻¹ for a-Ge:H layers. The total film thickness measured by the Dectak surface profiler was in the range of 300 to 550 nm and the total number of periods was controlled between 60 and 100.

The X-ray diffraction (XRD) of the prepared samples has shown that the interface between a-Ge:H well layer and a a-Si:H barrier layer is atomically abrupt.

The period thickness measured by XRD was in good agreement with the period calculated from the total film thickness measured with a Dectak surface profiler and the growth rates of the individual layers in the bulk.

Infrared absorption spectra were measured in the range between 400 and 2200 cm⁻¹ using a Nicolet Fourier transform infrared spectrometer (model 740). After base line correction, the IR absorption peaks were fitted by Gaussian to obtain the integrated absorption intensity I*. As the film thickness was usually below 1 μm, the correction proposed by Langford et al. was employed to obtain the integrated absorption I.

The optical band gap E_g was deduced from transmission and reflection measurements using JASCO V-570 UV - vis Spectrophotometer-Instructions.

The samples were characterized by the scanning electron microscopy (SEM) after annealing at 300 °C for 8 h. For electrical measurements, a co-planer method was used for the dark and photo-conductivity in



vacuum. The light source of a tungsten lamp of intensity 100mW/cm^2 was employed.

III. Results and discussion

3.1. The hydrogen content calculating by fitting Gaussian distribution

Typical infrared (IR) absorption spectra for a-Si:H(3 nm)/a-Ge:H multilayers of $d_{\text{Ge}}=2.8$ nm doped with P and of $d_{\text{Ge}}= 3$ nm doped with B are shown in figure (1). the absorption peak near 1880 cm^{-1} is attributed to the stretching vibration of Ge-H groups incorporated into bulk material, while the absorption near 2100 cm^{-1} is associated with the vibration of Si-H and/or Si-H₂ groups located at internal surfaces of voids. The absorption peak near 2000 cm^{-1} was attributed to Ge-H or Ge-H₂ groups at void surface and Si-H groups in compact material [1,8-10]. It is seen that after annealing for 8 h at 300°C , the absorbance integrated intensity of the wagging, bending and stretching bond decreases for the two samples. In the stretching mode range of the wave number, the spectra show that the integrated absorption intensity of Ge-H and Si-H stretching bonds is decreasing after annealing at 300°C , indicating that hydrogen moves around in Ge layers and is partially evolved, thus causing a change in the atomic density of the Ge network and also a change in the ratio of $d_{\text{Ge}}/(d_{\text{Si}}+ d_{\text{Ge}})$ which is correlated with the structure factor [11]. The hydrogen evolved from Ge leads to structural relaxation caused by the annealing preferentially occurs [5,6,9,12 – 14,16].

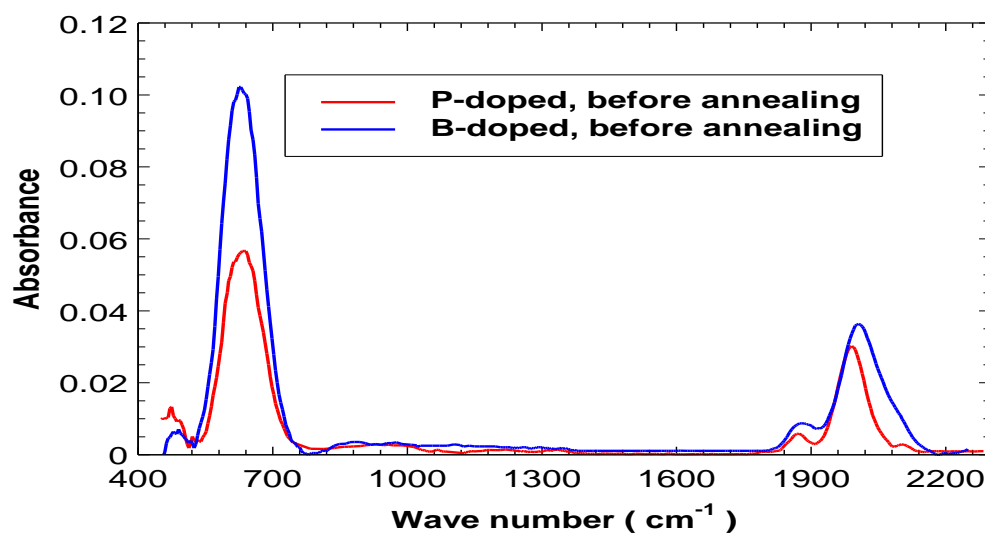


Figure (1): IR- spectra of untreated P- and B- doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{\text{Ge}}= 2.8$ and 3 nm.

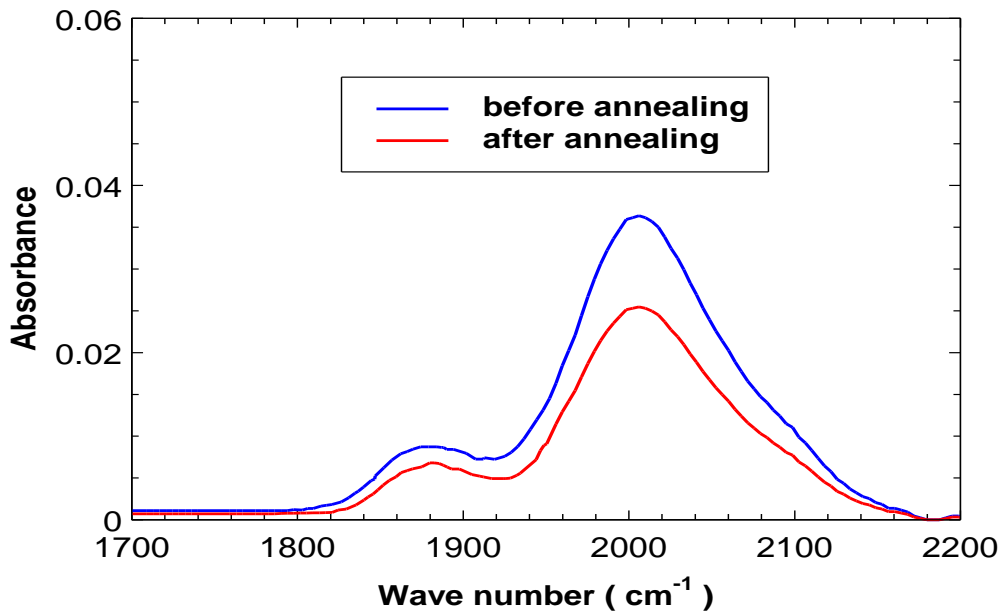


Figure (2) :IR- spectra in the stretching mode of P- doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}=2.8$ nm before and after annealing at 300°C for 8 h.

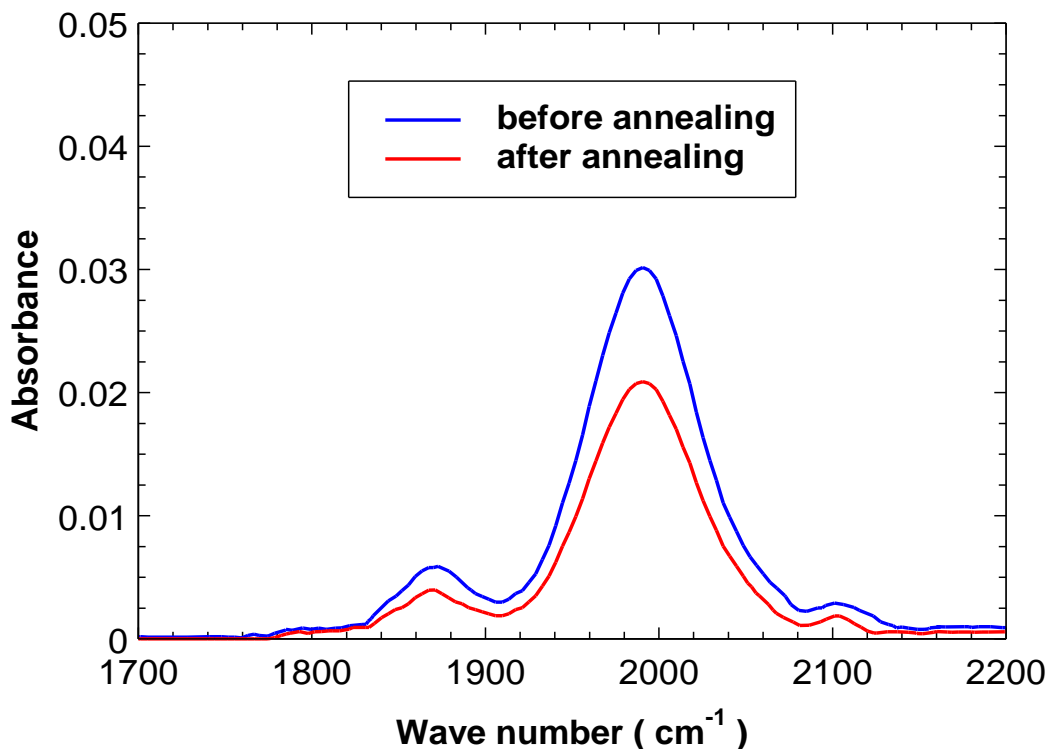


Figure (3): IR- spectra in the stretching mode for B-doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}= 3$ nm before and after annealing at 300°C for 8 h.

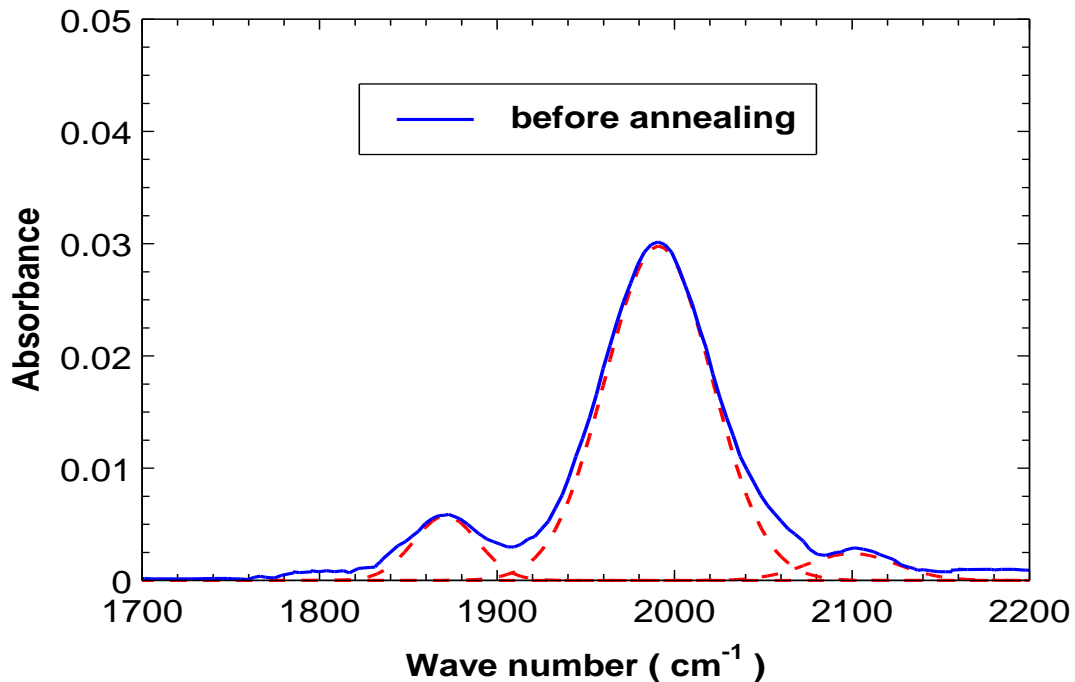


Figure (4): Fitting of IR- spectra in the stretching mode range for B-doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}= 3$ nm before annealing at 300°C for 8 h.

The hydrogen content (N_H) of P- and B-doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge} = 2.8$ and 3 nm calculated by fitting the stretching mode (See figures 2 and 3) before and after annealing at 300°C for 8 h is given in table (1). The fitting was done by using the Gaussian distribution. For brevity see figure (4) for B- doped a-Si:H/a-Ge:H multilayers before annealing.

Table (1): The hydrogen content for the two samples before and after annealing.

The samples	Hydrogen content (N_H) cm^{-3} before annealing	Hydrogen content (N_H) cm^{-3} after annealing
P-doped a-Si:H(3 nm)/a-Ge:H	2.97×10^{21}	2.03×10^{21}
B-doped a-Si:H(3 nm)/a-Ge:H	3.98×10^{21}	2.70×10^{21}

It is seen from table (1) that the total hydrogen content (N_H) is decreased after annealing due to the evolution of hydrogen from the network and producing bubbles on the surface of the films as are shown by SEM.



3. 2. The X-ray diffraction (XRD)

The structural change of P-doped a-Si:H(3 nm)/a-Ge:H multilayers (n-type) of $d_{Ge}=2.8$ nm after annealing at 300°C for 8 h have been investigated by XRD . The phases appeared in the x-ray spectra are Ge-Ge(211), Ge-Ge(220), Ge-Ge(400) and Ge-Ge(332).The crystallization occurs in a-Ge:H layers only because the crystallizing temperature of germanium at 300°C is lower than that of silicon near 550°C .

Similarly the structural changes for B-doped a-Si:H(3 nm)/a-Ge:H multilayers (p-type) of $d_{Ge}= 3$ nm annealed at 300°C for 8 h have been investigated by XRD . The phases appeared in the x-ray spectra are Ge-Ge(411) phases only. As reported the crystallization occurs in the a-Ge:H layers only because the crystallizing temperature of the germanium at 300°C is lower than that of silicon.

3.3. Scanning electron microscopy (SEM)

The P- and B-doped a-Si:H(3 nm)/a-Ge:H multilayers annealed at 300°C for 8 h were characterized by the scanning electron microscopy (SEM) as shown in figures (7) and (8), respectively. It is seen from the images that bumps appear on the surface of the films annealed at 300°C for 8 h. the hydrogen forming the bubbles arises from the rupture of the Si-H and Ge-H bonds activated by the thermal energy of the annealing temperature and by the energy released from the recombination of thermally generated electron hole pairs [13,15,16].

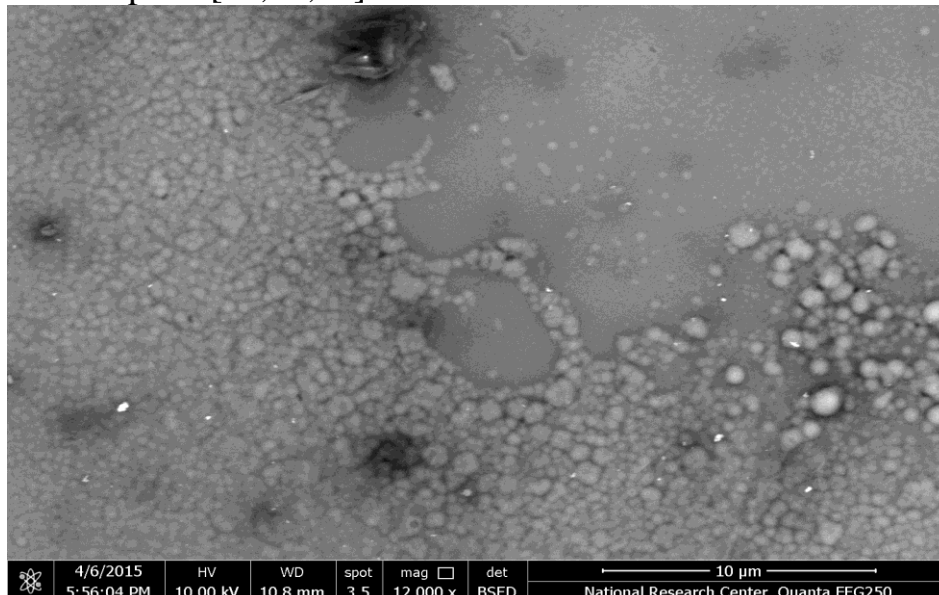


Figure (7): SEM images for P-doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}=2.8$ nm after annealing at 300°C for 8 h.

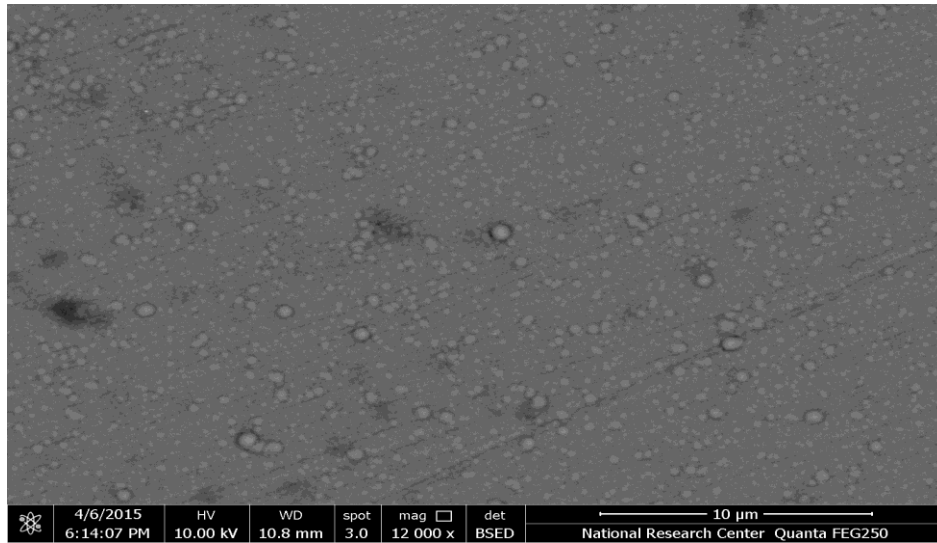


Figure (8): SEM images for B-doped a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}=3$ nm after annealing at 300°C for 8 h.

3. 4. Optical properties of phosphorus and boron doped Nano- a-Si:H(3 nm)/a-Ge:H multilayers

The optical energy gap E_g and Urbach energy E_u for P- and B-doped a-Si:H(3 nm)/a-Ge:H multilayers were determined using plots represented in figures (9) and (10) for untreated films and figures (11) and (12) for films annealed at 300°C for 8 h, respectively. The values of E_g and E_u are given in tables (2) and (3).

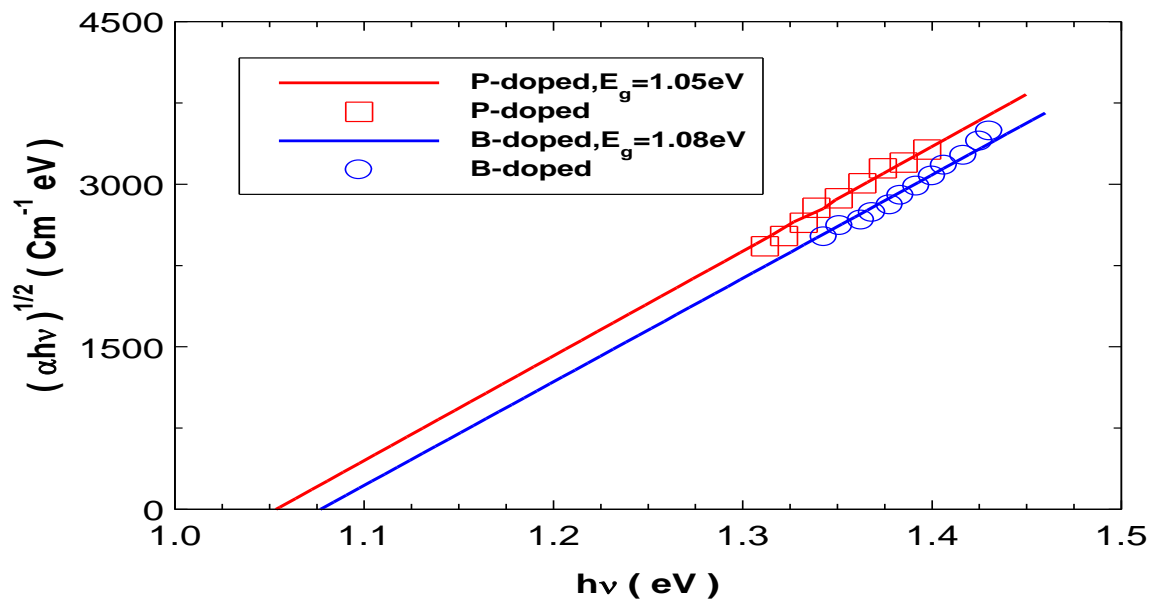


Figure (9): $(\alpha hv)^{1/2}$ vs hv for untreated a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}=2.8$ nm (P- doped) and $d_{Ge}=3$ nm (B-doped).

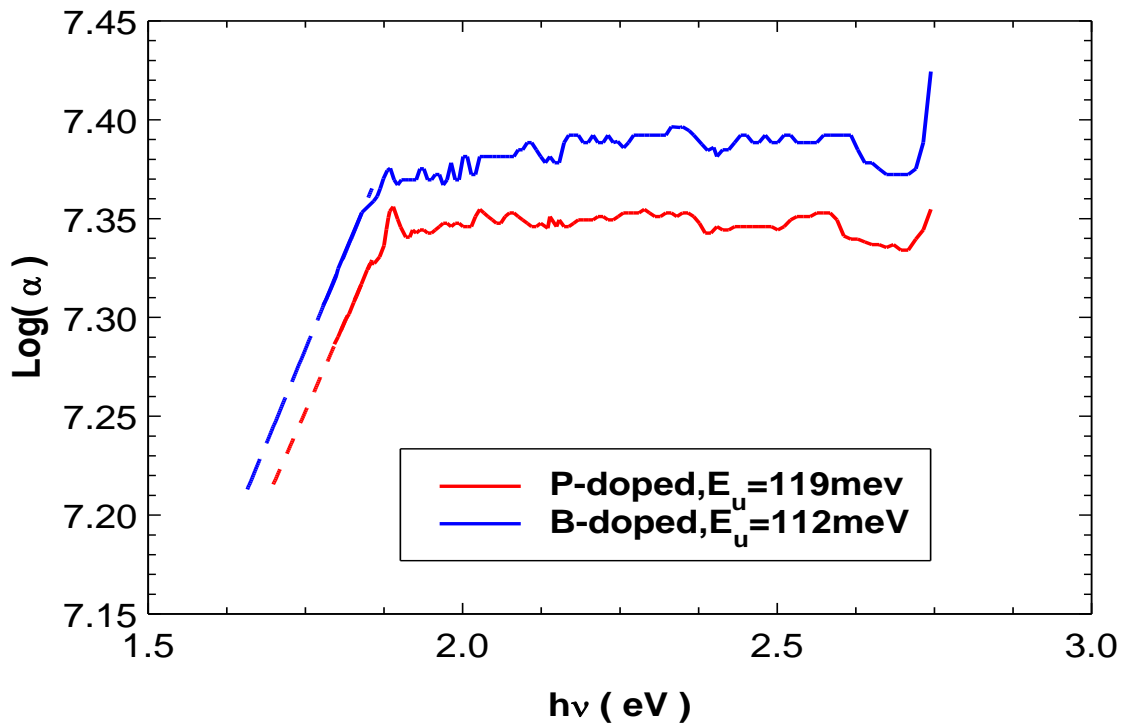


Figure (10): $\text{Log}(\alpha)$ Vs. $h\nu$ for untreated a-Si:H(3 nm)/a-Ge:H multilayers of thickness $d_{\text{Ge}}=2.8$ nm (P-doped) and $d_{\text{Ge}}=3$ nm (B-doped).

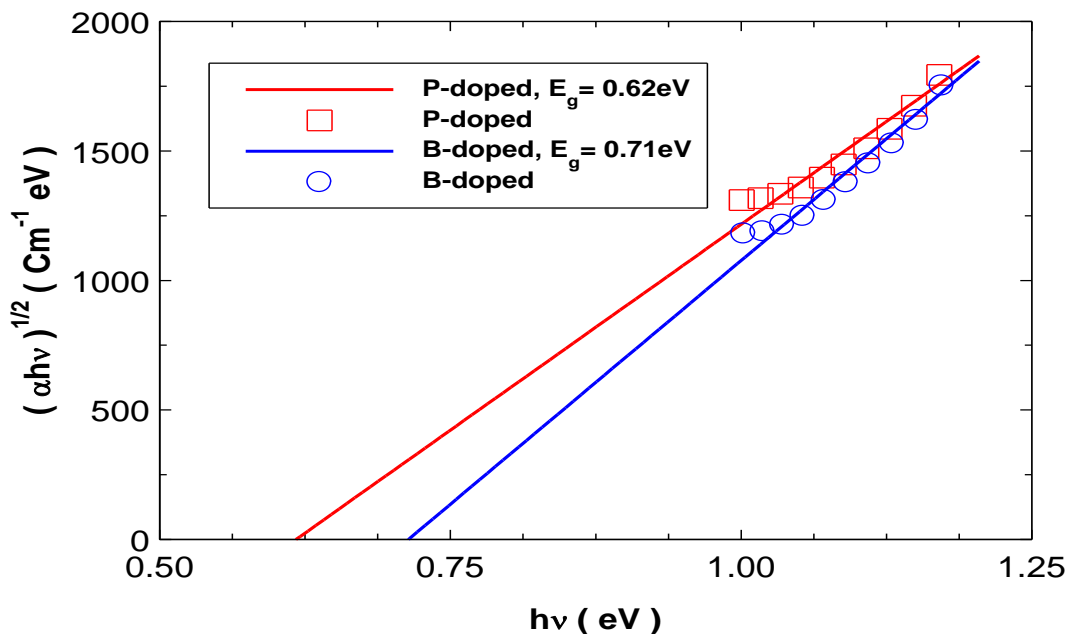


Figure (11): $(\alpha h\nu)^{1/2}$ vs $h\nu$ for a-Si:H(3 nm)/a-Ge:H multilayers of thickness $d_{\text{Ge}}=2.8$ nm (P-doped) and $d_{\text{Ge}}=3$ nm (B-doped), after annealing at 300°C for 8 h.

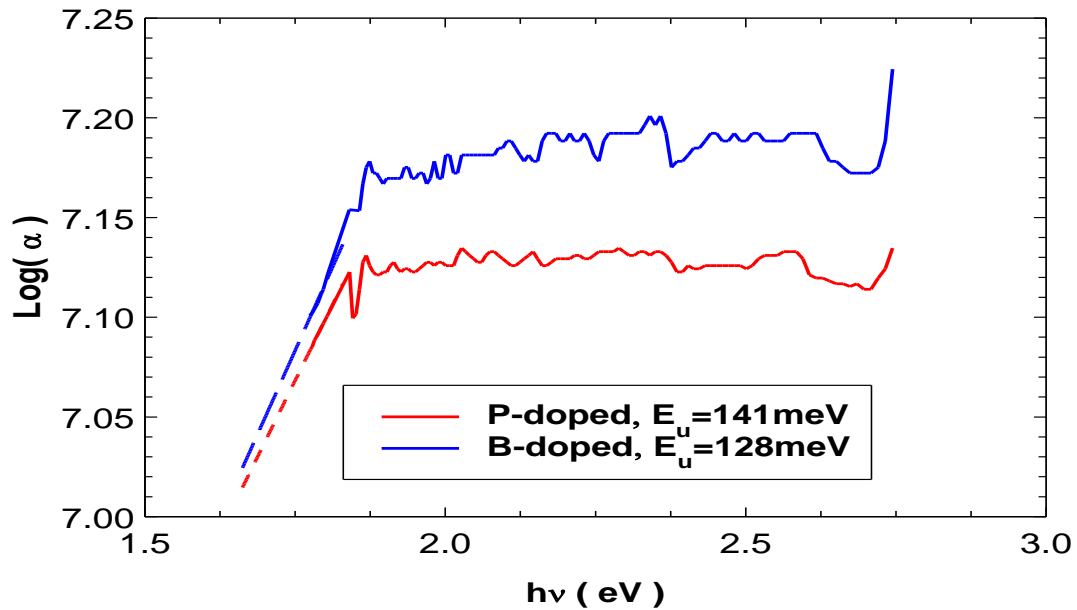


Figure (12): Log (α) Vs. $h\nu$ for a-Si:H(3 nm)/a-Ge:H multilayers of thickness $d_{Ge}=2.8$ nm (P- doped) and $d_{Ge}= 3$ nm (B-doped) after annealing at 300°C for 8 h.

Table (2): The optical band gap E_g and Urbach energy, E_u for untreated a-Si:H(3 nm)/a-Ge:H multilayers of thickness $d_{Ge}=2.8$ nm (P-doped) and $d_{Ge}= 3$ nm (B-doped).

Samples	N_H (cm^{-3})	E_g (eV)	E_u (meV)
(P-doped)	3.98×10^{21}	1.05	119
(B-doped)	2.97×10^{21}	1.08	112

For untreated films the incorporation of boron (B) or phosphorus (P) induce a decrease of the hydrogen content [17] which leads to a decrease of the optical energy gap and an increase of Urbach energy as shown in table (2). After annealing at 300°C for 8 h, the total hydrogen content is more reduced which leads to a more decrease in the optical energy gap and more increase in Urbach energy as shown in table (3).

Table (3): The optical band gap E_g and Urbach energy, E_u for a-Si:H(3 nm)/a-Ge:H multilayers of $d_{Ge}= 2.8$ nm (P-doped) and $d_{Ge}= 3$ nm (B-doped) after annealing at 300°C for 8 h.

Samples	N_H (cm^{-3})	E_g (eV)	E_u (meV)
(P-doped)	2.70×10^{21}	0.62	141
(B-doped)	2.03×10^{21}	0.71	128



IV. Conclusion

The total hydrogen content and annealing play an important role for determining the optical absorption edge. The results showed considerable improvements for the optical band gap and structural by controlling the hydrogen content and hydrogen configuration upon annealing. The data shows that the optical energy gap is decreased with rising annealing temperature while the Urbach energy is increased mainly due to the decreased total hydrogen content in agreement with previous works.

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