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Nonlinear dependence of optical gap of a-$Si_{1-x}Ge_x$:H films on Ge content (x < 0.4)

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The dependence of optical band gap of a-$Si_{1-x}Ge_x$:H films on Ge content is discussed. The films are deposited by magnetron co-sputtering of c-Si target with c-Ge chips on it in Ar + H_2 atmosphere. It has been observed that concentration of the bonded hydrogen decreases with Ge content in the films. The results of study show that variation of the optical band gap of a-$Si_{1-x}Ge_x$:H films on Ge concentration follows the nonlinear law. This is related to the nonlinear changes of H concentration in the films.

Keywords: amorphous semiconductors, a-SiGe:H films, optical properties, magnetron sputtering.

1. Introduction

Hydrogenated amorphous silicon and its alloys have attracted significant interest as they could be used in low cost high efficiency solar cells [1]. By alloying a-Si:H with Ge it is possible to narrow the band gap and thus improve the long-wavelength response. However, it has been generally observed that all the properties, optical, structural, electrical, which are important for optoelectronic application, change with alloying [2–4]. That is why each investigation in this area is of great importance for both the basic and applied research.

In this work we focus on nonlinear dependence of the optical band gap of a-$Si_{1-x}Ge_x$:H on increasing Ge content ($0 < x < 0.4$). The change of H concentration is regarded as well.

2. Experimental

Thin films of a-$Si_{1-x}Ge_x$:H have been deposited by reactive magnetron co-sputtering of c-Si and Ge in Ar + H_2 gas mixture as described in Ref. 4. The film thickness was about 1 μm. The composition of the alloys was obtained using Rutherford backscattering method. The Ge content, x, changes from 0.06 to 0.38. The Tauc optical band gap, E_{opt}, was determined from optical transmission in the region of 400–2500 nm on Perkin-Elmer photospectrometer 330 by a technique reported earlier [4,5]. Concentration of the bonded H, C_H, was calculated from wagging vibration modes around 640 cm^{-1} in IR absorption spectra [4].

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3. Results and discussion

The change of the optical gap on Ge content in a-$Si_{1-x}Ge_x$:H film, deposited at two different substrate temperatures, is presented in Fig. 1. It could be expressed by linear dependence, as in Refs. 3,6, and 7, assuming a high solubility of Ge in Si.

$$E_{opt} = 1.81 - 0.96\,x \qquad \text{at } T_s = 200\,^{\circ}C,$$
$$E_{opt} = 1.63 - 1.25\,x \qquad \text{at } T_s = 275\,^{\circ}C.$$

These two relationships show that not only the value of the optical gap but also its dependence on Ge content are influenced by the substrate temperature. We noted that the parameters in these equations are different for the films deposited at different substrate temperatures and are different from those reported by other authors [3,6,7]. This difference could be explained with different concentration of hydrogen in the films, studied and presented by different groups. Data on H content in the studied films are not reported in the above mentioned references. More detailed observation of the curves presented in Fig. 1 points out that linear law cannot fit well the experimental points.

The change of the optical gap of the amorphous silicon-based alloys on the hydrogen concentration is a well-known fact, described in literature. Now, let us see the changes in concentration of the bonded hydrogen in a-$Si_{1-x}Ge_x$:H films. Our results on H concentration in the films show that C_H decreases with Ge content, as it is demonstrated in Fig. 2. Decrease in H concentration with Ge in a-$Si_{1-x}Ge_x$:H films is a well-known and has been observed by different authors [7,8]. Moreover, it has to be noticed that the dependence of C_H on x, presented in Fig. 2, follows nonlinear law and can be described by the equations

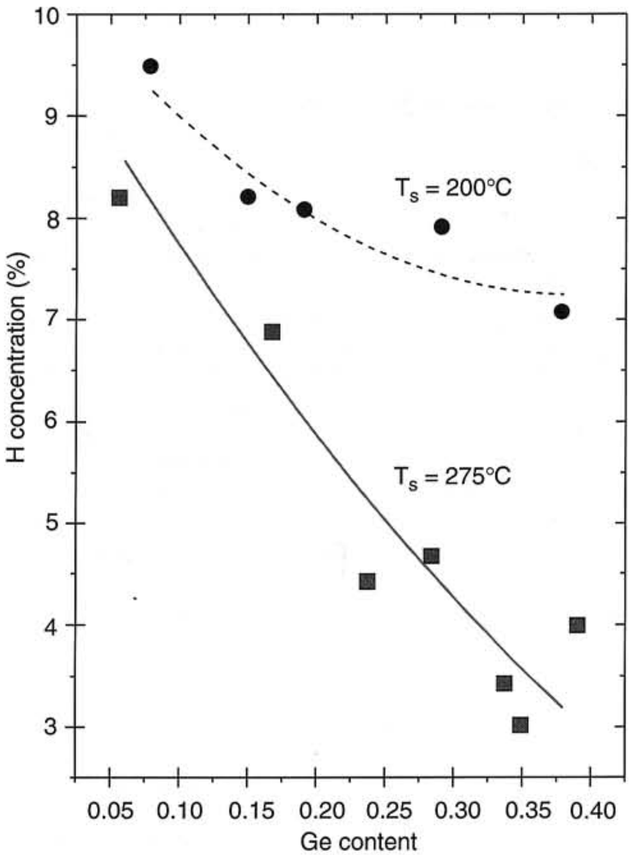

Fig. 1. Hydrogen concentration as a function of Ge content for two series of a-$Si_{1-x}Ge_x$:H films deposited at different substrate temperatures – 200 and 275°C.

$$C_H\,(at\%) = 10.4 - 16.0x + 21.0\,x^2 \quad \text{at } T_s = 200°C,$$

$$C_H\,(at\%) = 9.8 - 22.0x + 13.0\,x^2 \quad \text{at } T_s = 275°C.$$

It is possible to find the law which is followed by the values of the optical gap with Ge content, using the experimental data presented in Fig. 1. The dependence of optical gap on x could be expressed as follows, having in mind high solubility of Ge in Si

$$E_{opt} = A - B\,x, \text{ where } A = E_{opr} \text{ at } x = 0,$$

and

$$B = (dE_{opt}/dx)_{CH=const} + (\delta E_{opt}/\delta C_H)\,(\delta C_H/\delta x).$$

As showed our previously published results [10], the change of the optical gap only due to the alloying with Ge, for $x < 0.28$, is $dE_{opt}/dx = 1.8\times10^{-2}$ eV/at%Ge and for $x > 0.28$ it is 1.1×10^{-2} eV/at%Ge. Assuming that the gradient of change of the gap of a-Si_{1-x} Ge_x:H films with H concentration is the same as in a-Si:H one – $\delta E_{opt}/\delta C_H$ = 0.02 eV/at%H [11] we obtain the following expressions for the dependence of the optical gap on the Ge content in different regions of x

$$\left.\begin{array}{ll}\text{for } x < 0.28 & E_{opt} = 1.82 - 1.22x + 0.84\,x^2\ [eV] \\ \text{for } x > 0.28 & E_{opt} = 1.82 - 1.42x + 0.84\,x^2\ [eV]\end{array}\right\} \text{ at } T_s = 200°C,$$

and

$$\left.\begin{array}{ll}\text{for } x < 0.28 & E_{opt} = 1.63 - 2.45x + 0.52\,x^2\ [eV] \\ \text{for } x > 0.28 & E_{opt} = 1.63 - 1.65x + 0.52\,x^2\ [eV].\end{array}\right\} \text{ at } T_s = 275°C$$

These dependencies are plotted by dashed lines in Figs. 2(a) and (b).

For the whole region of x, under consideration, the following expressions are obtained by fitting the two curves in two regions of x with one equation

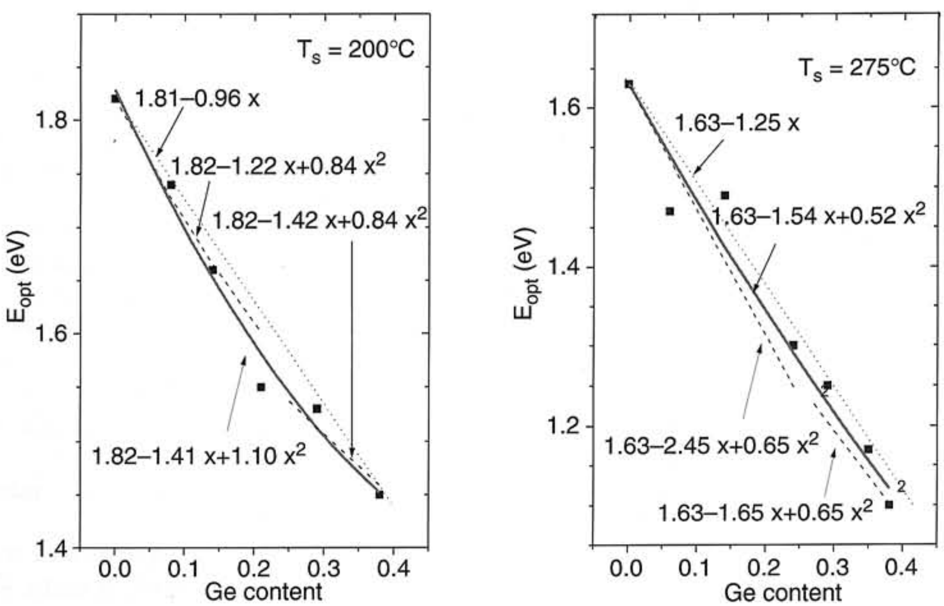

Fig. 2. Optical band gap versus Ge content in a-$Si_{1-x}Ge_x$:H films, deposited at different substrate temperatures: T_s = 200 (a) and 275°C (b), respectively. The experimental points are fitted with different laws.

$$E_{opt} = 1.82 - 1.41x + 1.10\,x^2\ [eV] \qquad \text{at } T_s = 200°C,$$
$$E_{opt} = 1.63 - 1.54x + 1.10\,x^2\ [eV] \qquad \text{at } T_s = 275°C.$$

The calculated values of E_{opt} by using the last expressions are presented by solid lines in Figs 1(a) and 1(b) which show good coincidence with the experimental data. A better fitting is possible to obtain if a change of $\delta E_{opt}/\delta C_H$ with Ge content is supposed, too, but similar data are missing in literature by now.

Conclusions

The results of this study show, for the first time by our knowledge, that the dependence of the optical gap on Ge concentration in a-$Si_{1-x}Ge_x$:H films can be described by polinom of the second order better than by a linear law as it was presented till now. This behaviour could be related to the decrease in hydrogen concentration in the films with increasing Ge concentration.

References

1. H. Haku, K. Sayama, E. Maruyama, H. Dohjoh, N. Nakamura, S. Tsuda, S. Nakano, Y. Kishi, and Y. Kuwano, "High-performance a-SiGe solar cells using a super chamber method," *Jap. J. Appl. Phys.* **30**, 2700–2704 (1991).
2. D. Dimova-Malinovska and L. Nedialkova, "Influence of Ge content on vibrational and optical properties of a-$Si_{1-x}Ge_x$:H thin films," *Solar Energy Mat. & Solar Cells* **31**, 283–289 (1993).
3. K.D. Mackenzie, J.R. Eggert, D.J. Leopold, Y.M. Li, S. Lin, and W. Paul, "Structural, electrical and optical properties of a-$Si_{1-x}Ge_x$:H and an inferred electronic band structure," *Phys. Rev.* B **31**, 2198–2212 (1985).
4. D. Dimova-Malinovska, L. Nedialkova, and V. Kudoyarova, "Compositional dependence of the optical properties and the H local bonding in a-$Si_{1-x}Ge_x$:H (x 0.4) thin films," *Solar Energy Mat. & Solar Cells* **30**, 27–35 (1993).
5. D. Dimova-Malinovska and A. Krapchev, "Determination of the refractive index and the thickness of the amorphous silicon films," *Bulg. J. Phys.* **17**, 109–113 (1990).
6. S. Wagner, V. Chu, J.F. Conde, and J.Z. Liu, "The optoelectronic properties of a-Si,Ge:H(F) alloys," *J. Non-Cryst. Solids* **114**, 453–458 (1989).
7. C. Godet, A. Lloret, and J.P. Stoquert, "Influence of the dilution gas nature and deposition temperature on the photoconductivity of glow-discharge deposited amorphous silicon-germanium-hydrogen alloys," *Proc. EC PV Solar Energy Con.*, Kluwer Acad. Publ., Dordrecht/Boston/London, 70–73 (1989).
8. M. Kumeda, Y. Tsujimura, Y. Yonezawa, A. Morimoto, and T. Shimizu, "NMR and ESR studies on a-$Si_{1-x}Ge_x$:H prepared by magnetron sputtering," *Solid State Com.* **55**, 409–412 (1985).
9. D. Dimova-Malinovska, L. Nedialkova, M. Tzolov, and N Tzenov, "Modification of the optical and structural properties of hydrogenated amorphous silicon by alloying with germanium and carbon," *Solar Energy Mat. & Solar Cells* **53**, 333–339 (1998).
10. W. Paul and D.A. Anderson, "Properties of amorphous hydrogenated silicon, with special emphasis on preparation by sputtering," *Solar Energy Mat*erials **5**, 229–316 (1980).