Frequency-resolved photoconductivity in a-SiGe:H

R. Kaplan

Department of Physics, University of Erciyes, 38039 Kayseri, Turkey

Abstract

Frequency-resolved photocurrent measurements were carried out on hydrogenated amorphous silicon–germanium (a-SiGe:H) thin film alloys between 20 and 290 K, by using the quadrature frequency-resolved spectroscopy method which yields lifetime distributions directly. The results show that the frequency-resolved photocurrent depends on excitation light intensity and temperature and thus they give information about recombination kinetics. © 1998 Elsevier Science S.A. All rights reserved.

Keywords: Amorphous SiGe:H; Photoconductivity; Carrier lifetime; Recombination

1. Introduction

The interest in a-SiGe:H alloys has come largely from their potential use in photo-voltaic devices to achieve efficient solar energy conversion [1–7]. Due to the lower optical band gap, these alloy films show better photoresponse at longer wavelengths than a-Si:H film. This property improves the collection efficiency in that part of the solar radiation spectrum with a corresponding increase in the device efficiency when incorporated into a multijunction solar cell as an intrinsic layer. The photoconductive properties of a-SiGe:H alloys are therefore of interest. The applications require optimization of the carrier recombination and transport mechanisms that are dominant in these materials.

The frequency-resolved photocurrent measurement as a function of temperature and light intensity in an amorphous semiconductor is a valuable tool in achieving a good understanding of the recombination processes and distribution of localized states which control the photocurrent. However, in spite of the extensive studies of localized states of a-SiGe:H alloys, the interpretation of experimental data remains difficult.

Lifetime analysis can be studied by means of the frequency-resolved spectroscopy (FRS) [8] method. The method has been widely applied to the study of lifetime and recombination processes [9–16]. In this method, the in-phase and quadrature photoluminescence response in amorphous materials have been treated in detail and showed that the quadrature response function gives the lifetime distribution of the system whereas the in-phase response is the integral of the lifetime distribution between the limits $\tau \propto 2\pi f^{-1}$ and ∞. This treatment has been applied to frequency-resolved photocurrent response in amorphous materials [17–26].

In this work, the results of frequency-resolved photocurrent spectroscopy measurements performed on a-SiGe:H thin film alloys will be presented. The photocurrent response was determined over the frequency range 10 Hz to 100 kHz and as a function of excitation intensity (between 10 μW and 2.3 mW) and temperature (between 20 and 290 K). The results are discussed in terms of models that try to explain the photocurrent response in relation to the localized states.

2. Experimental procedures

The samples used in this work are ion-bombarded thin films of a-SiGe:H alloys. They were obtained from Dundee University. The details of their preparation conditions and properties are listed in Tables 1 and 2. Al electrodes separated by a 0.1 mm gap were deposited onto the films by using our own vacuum coating unit. Cu wires were then placed on the Al layers with electrically conductive silver paint. Current–voltage (*I–V*) measurements taken in the dark proved that the contacts were perfectly ohmic.

The quadrature frequency resolved photocurrent (FRPC) response of the samples in the frequency interval between 10 Hz and 100 kHz was measured as a function of the intensity of the excitation light and temperature. FRPC measurements were made with a lock-in amplifier (SR 530 Stanford Research System). The advantage of this

Table 1
Amorphous Si–Ge alloys. Preparation: T_d is the deposition temperature; the next two columns of the table show gas phase ratios of Ge and Si; P is the plasma pressure and d is the resulting sample thickness

Sample	T_d (°C)	Ge/Ge+Si (%)	Ge/Ge+Si+H$_2$ (%)	RF frequency (MHz)	RF power (W)	P (Torr)	d (μm)
10	229	10	100	5	◡	0.6	0.99
16	230	30	100	5	◡	0.6	0.90
51	210	10	10	13	50	0.5	0.35
61	192	30	50	13	50	0.5	0.19

frequency-locked AC measurement is that it rejects any stray light, dark current and other unmodulated signals.

The photocurrent was excited by a HeNe laser beam (1.96 eV) which was modulated by an acousto–optic modulator (IntraAction Corp., Model AOM-125). The modulation amplitude amounted to 46% of the bias light intensity. The samples were placed in a helium exchange-gas cryostat in which the temperature could be varied between room temperature (295 K) and 20 K. The vacuum pressure of the cryostat was about 10^{-6} Torr. Using neutral density filters, the intensity of the modulated excitation beam was varied between 2.31 mW and 10 μW (1 mW corresponds to $\sim 1 \times 10^{19}$ photons/s per cm^2).

3. Results and discussion

We performed measurements on four different a-SiGe:H samples whose properties are given in Tables 1 and 2. However, the photocurrent behavior of each of these samples is found to be very similar. So, the results presented here are common for all of them, except for Fig. 5.

Since the energy of the excitation light is bigger than the optical band gap of these samples, we assume that the carriers are photoexcited between extended states and then a trap limited recombination occurs.

Fig. 1 shows the FRPC response of one of the samples, sample 61 in Tables 1 and 2, for different excitation intensities at room temperature (295 K). The measurements were taken under an electric field of 1×10^4 V/cm. Each curve is normalized with respect to its maximum value. As can be seen from the figure there is a single peak in each curve which shifts to higher frequencies as the intensity increases. Further, the spectra broadens with decreasing intensities. Wagner et al. [17] suggest that for a system with a single characteristic time τ the quadrature frequency-resolved

spectrum is a symmetric line of half-width 0.7 decades peaked at the frequency:

$$\tau = \frac{1}{2\pi f_{peak}} \tag{1}$$

Using this relation, from Fig. 1 the lifetimes of about 126.6, 44.9, 14.2 and 7.1 μs were calculated for the intensities of 10, 20, 80 μW and 2.31 mW, respectively. The distribution of lifetimes ($\tau = (2\pi f_{peak})^{-1}$) with respect to generation rate (proportional to the excitation intensity) have been used to determine what kind of recombination occurs between the photoexcited carriers [27,28,15]. It has been suggested that in the geminate type of recombination the distribution of lifetimes will be insensitive to generation rates, whereas in the distant pairs (DP) model, where it is assumed that recombination takes place between nearest available neighbors non-geminately, the lifetimes should decrease with increasing generation rate. The result of Fig. 1 shows that the lifetime is dependent on the generation rate and therefore can be interpreted as supporting the DP model.

The frequency dependence of FRPC response at 2.3 mW for different temperatures under an electric field of 1×10^4 V/cm is given in Fig. 2 where the logarithmic scale shows the correct relation between the absolute intensity values. The peak frequency and thus the lifetime seems to be almost independent of temperature. In a-Si:H, the peak frequency is excitation intensity-dependent and becomes an activated process at high temperatures (60–295 K), while it tends to be constant below about 60 K [29]. The recombination takes place by tunneling transition from localized conduction band tail states to neutral dangling bonds at low temperatures. Since the tunneling processes are only slightly temperature-dependent, the peak frequency will be therefore nearly constant.

In both the temperature- and the intensity-dependent

Table 2
Amorphous Si–Ge alloys. Properties: E_σ is the dark current activation energy; σ_o is the pre-exponential; σ_{RT} is the room temperature dark conductivity; E_g is the optical gap and σ_{ph} is the photoconductivity excited by 7×10^{14} photons/s per cm^2

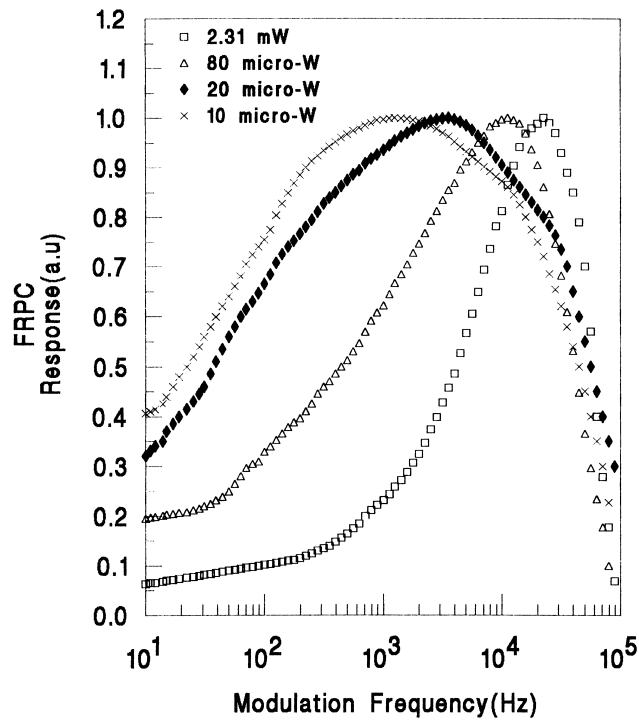| Sample	E_σ (eV)	σ_{RT} (Ω^{-1} cm^{-1})	σ_o (Ω^{-1} cm^{-1})	E_g (eV)	σ_{ph} (Ω^{-1} cm^{-1})
10	0.78	5.3×10^{-10}	1.2×10^4	1.42	5×10^{-9}
16	0.53	2×10^{-6}	2.8×10^3		
51	0.83	1.55×10^{-10}	2.3×10^4	1.78	1.7×10^{-6}
61	0.73	3.2×10^{-9}	9.3×10^3		

Fig. 1. FRPC response of a-SiGe:H at 295 K, for the indicated excitation intensities. Each curve is normalized with respect to its maximum value (a.u, arbitrary units).

FRPC measurements mentioned above the peak position, i.e. f_{peak} was independent of the applied electric field.
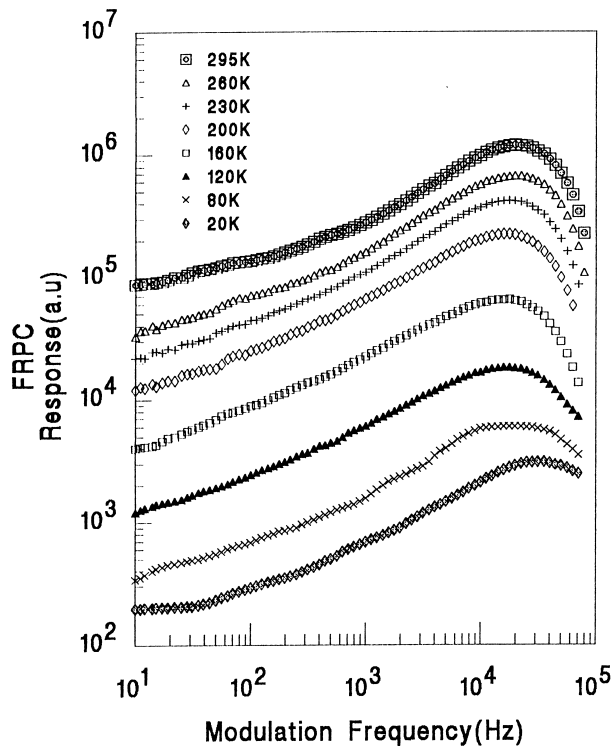

Fig. 2. FRPC response of a-SiGe:H at the indicated temperatures for an excitation intensity of 2.3 mW (a.u, arbitrary units).

Fig. 3 shows the temperature dependence of photocurrent for different excitation intensities determined at an electric field of 1×10^4 V/cm and 100 Hz. This behavior did not change for frequencies up to 10 kHz. All curves almost yield straight lines for high temperatures (above about 130 K). From the slopes of lines, an average activation energy value of 70.9 ± 2.3 meV was calculated. A smaller activation energy value of about 40 meV was obtained for a-Si:H [31] at high temperatures (100–300 K). The higher value of the photocurrent activation energy seen in a-SiGe:H could be related with increasing of the conduction band tail due to extra chemical disorder and the possibility of both Si and Ge dangling bonds. At high temperatures, the transport is determined by delocalized electrons in extended states in a conduction band and this transport is controlled by the trapping of carriers in tail band states [30]. At low temperatures (below about 100 K) the intensity of photocurrent does not show an activated behavior.

An important characteristic of photoconductivity is the dependence of photocurrent on the incident light intensity. It is now known that the nature of this dependence differs in various semiconductors. It may be described that the intensity of photocurrent is proportional to the generation rate [32]

$$I_{ph} \propto G^{\nu} \tag{2}$$

where the exponent ν is to be determined. Fig. 4 shows this dependence for different samples of a-SiGe:H listed in Tables 1 and 2 at room temperature (295 K). The exponent $v \, (\equiv \nu)$ was calculated from $(d(\ln I_{ph})/d(\ln G))$. These values are typical for the alloys a-SiGe:H [30]. The temperature
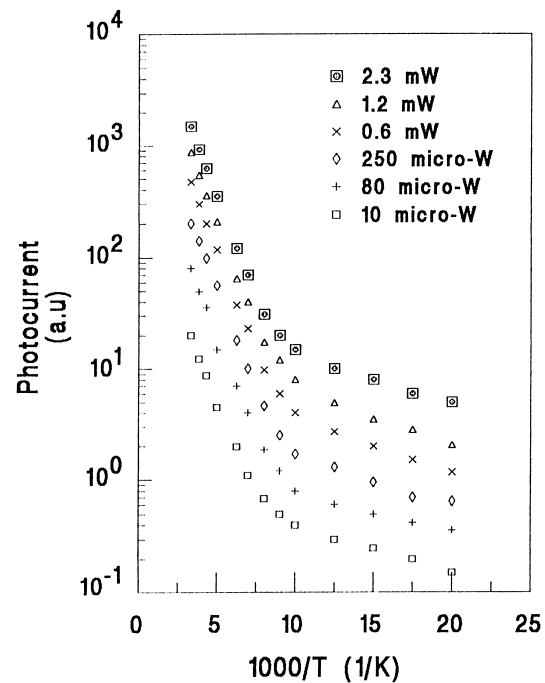

Fig. 3. Temperature dependence of photocurrent in a-SiGe:H for different excitation intensities measured at 100 Hz (a.u, arbitrary units).
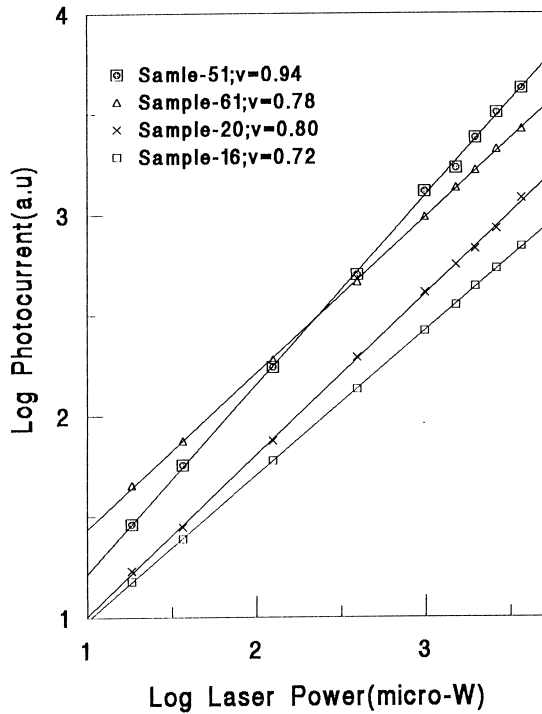
Fig. 4. Excitation intensity dependence of the photocurrent for different samples of a-SiGe:H measured at 100 Hz (a.u, arbitrary units). $T = 295$ K; ν ($\equiv v$) denotes the exponent in the power-law relationship $I_{ph} \propto G^\nu$.
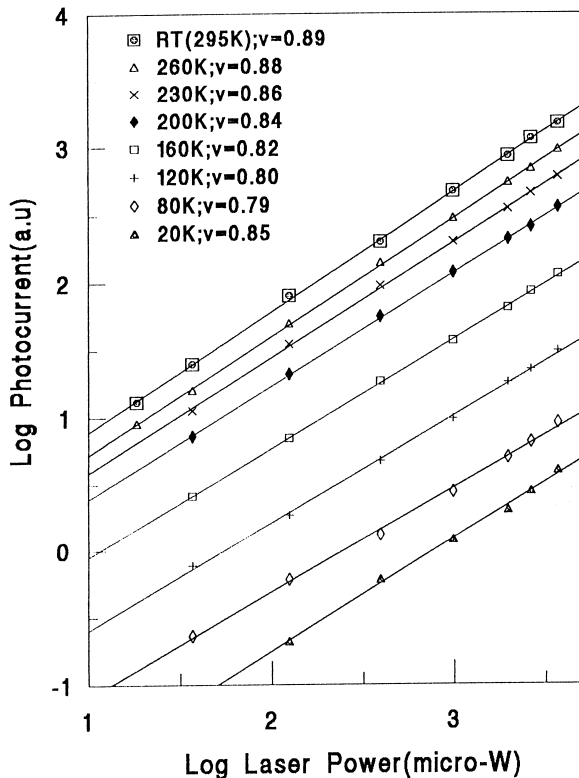

Fig. 5. Excitation intensity dependence of the photocurrent at the indicated temperatures measured at 100 Hz (a.u, arbitrary units); ν denotes the exponent in the power–law relationship $I_{ph} \propto G^\nu$.

dependence of ν is given in Fig. 5 for sample 61. Other samples also give similar results. The ν values calculated from the slopes of the lines are quite close to each other at all temperatures chosen. The average value of this virtually temperature independent exponent is 0.84 ± 0.02. Any small change in ν with temperature might be related to a higher density of defects in the thin films which play the role of recombination centers [30]. For a-Si:H, ν was determined as 0.92 ± 0.05 (average) at the temperatures between 20 K and room temperature [31]. Rose [32] suggests that $\nu = 1$ corresponds to monomolecular recombination and $\nu = 0.5$ to bimolecular recombination. However in the case of continuous distribution of traps the value of ν may be anywhere between 0.5 and 1.0 depending upon the intensity and the temperature range. Unfortunately, the model of Rose cannot explain the temperature independence of ν. Shklovskii et al. [33] and Searle [28] have developed geminate and DP models, respectively, for the photoconductivity processes in amorphous semiconductors. These models are essentially the same in the absence of significant geminate pair effects and allow, in particular, analytical relations to be obtained for the correlation between the excitation light intensity, the carrier density and the corresponding lifetime. According to these models, at low temperatures, photoexcited carriers can lose energy only by tunneling to lower energy states, such as localized tail states or by recombination. Since the tunneling processes are only slightly temperature-dependent, the exponent ν will be therefore nearly constant. The predicted values of the models for ν are slightly less than unity, which are in agreement with our result.

The ν values were found to be independent of the applied electric field in the whole temperature range indicated in Fig. 5.

4. Conclusion

The quadrature FRPC response of the ion-bombarded a-SiGe:H alloys between 10 Hz and 100 kHz was measured as a function of the intensity of the excitation light and temperature. The temperature range covered was 20–295 K. It was determined that the carrier lifetime depends on the intensity of the excitation light. This may be interpreted as supportive as the distant-pair model where the lifetime distribution is determined by the spatial distribution of the photocarriers. The lifetime was also found to be temperature-independent at a constant excitation intensity of 2.31 mW. At low temperatures, the recombination takes place by tunneling transitions from localized conduction band tail states to neutral dangling bonds. Since the tunneling processes are only slightly temperature-dependent, the lifetime will be therefore nearly constant.

The intensity of the photocurrent at a fixed frequency is also found to be temperature-dependent. The high temperature regions of photocurrent versus inverse temperature

curves show an activated behavior with an average activation energy of 70.9 ± 2.3 meV for all excitation intensities (2.3 mW to 10 μW). This activation energy might be related to increasing of the density of the localized states which play role of the traps together with the tail band states.

The exponent ν in the power law relationship of the intensity of the photocurrent on the generation rate was determined as between 0.89 and 0.79. The small change in ν with temperature might be related to a higher density of defects in the thin films which play the role of recombination centers. The average value of this virtually temperature independent exponent is 0.84 ± 0.02, which is in contradiction with the predictions of the multiple-trapping model. An explanation for this result may be given by the models proposed by Shklovskii et al. [33] and Searle [28] based on the theory of energy-loss hopping of photocarriers through a distribution of localized band-tail states.

Acknowledgements

References

[1] A. Rose, Concepts in Photoconductivity and Allied Problems, , Robert E. Krieger, New York, 1978.
[2] A. Catalano, R.R. Arya, B. Fieselmann, et al., J. Non-Cryst. Solids 115 (1989) 14.
[3] D.E. Carlson, Phil. Mag. B63 (1991) 305.
[4] W. Paul, J.H. Chen, E.Z. Liu, A.E. Wetsel, P. Wickboldt, J. Non-Cryst. Solids 164–166 (1993) 1.
[5] J.K. Rath, A.R. Midya, S. Ray, Phil. Mag. B71 (1995) 851.
[6] F. Zhong, C.C. Chen, J.D. Cohen, J. Non-Cryst. Solids 198–200 (1996) 572.
[7] S. Hazra, A.R. Middya, S. Ray, Phil. Mag. B 75 (1997) 859.
[8] S.D. Depinna, D.J. Dunstan, Phil. Mag. B50 (1984) 579.
[9] R. Stachowitz, M. Schubert, W. Fuhs, J. Non-Cryst. Solids 164–166 (1993) 583.
[10] M. Schubert, R. Stachowitz, R. Saleh, W. Fuhs, J. Non-Crys. Solids 164-166 (1993) 555.
[11] H. Oheda, J. Non-Cryst. Solids 164–166 (1993) 559.
[12] J. Ristein, Phil. Mag. B70 (1994) 963.
[13] R. Stachowitz, M. Schubert, W. Fuhs, Phil. Mag. B70 (1994) 1219.
[14] H. Oheda, Phys. Rev. B52 (1995) 16530.
[15] M. Schubert, R. Stachowitz, W. Fuhs, J. Non-Cryst. Solids 198–200 (1996) 251.
[16] C. Ogihara, H. Ishimura, T. Kinoshita, K. Ikeda, K. Morigaki, J. Non-Cryst. Solids 198–200 (1996) 255.
[17] D. Wagner, P. Irsigler, D.J. Dunstan, J. Phys. C: Solid State Phys. 17 (1984) 6793.
[18] Y. Nitta, K. Abe, K. Hattori, H. Okamoto, Y. Hamakawa, J. Non-Cryst. Solids 97/98 (1987) 695.
[19] T.M. Searle, M. Hopkinson, M. Edmeades, S. Kalem, I.G. Austin, R.A. Gibson, in: M.A. Kastner, G.A. Thomas, S.R. Ovshinsky (Eds.), Disordered Semiconductors, Plenum Press, New York, 1987.
[20] K. Abe, H. Okamoto, Y. Nitta, Y. Tsutsumi, K. Hattori, Y. Hamakawa, Phil. Mag. B58 (1988) 171.
[21] K.P. Homewood, P.G. Wade, D.J. Dunstan, J. Phys. E: Sci. Instrum. 21 (1988) 84.
[22] K. Hattori, Y. Niwano, H. Okamoto, Y. Hamakawa, J. Non-Cryst. Solids 137/138 (1991) 363.
[23] C. Main, D.P. Webb, R. Bruggemann, R. Reynolds, J. Non-Cryst. Solids 137/138 (1991) 951.
[24] H. Oheda, J. Non-Crys. Solids 137/138 (1991) 1147.
[25] C. Main, R. Bruggemann, D.P. Webb, S. Reynolds, J. Non-Cryst. Solids 164–166 (1993) 481.
[26] R. Kaplan, Appl. Phys. A64 (1997) 171.
[27] M. Bort, W. Fuhs, S. Liedtke, R. Stachowitz, Phil. Mag. Lett. 64 (1991) 227.
[28] T.M. Searle, Phil. Mag. Lett. 61 (1990) 251.
[29] R. Kaplan, Turkish J. Phys. 21 (1997) 605.
[30] D.D. Malinovska, L. Nedialkova, D. Nesheva, in: N. Kirov (Ed.), New Physical Problems in Electronic Materials, World Scientific, Singapore, 1991.
[31] R. Kaplan, Turkish J. Phys. 21 (1997) 868.
[32] A. Rose, Concepts in Photoconductivity and Allied Problems, , Robert E. Krieger, New York, 1978.
[33] B.I. Shklovskii, H. Fritzsche, S.D. Baranovskii, J. Non-Cryst. Solids 114 (1989) 325.