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Oscillation of photoconductivity in hydrogenated amorphous silicon

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Abstract. The photoconductivity oscillations in glow discharge a-Si:H films at elevated temperatures (above 150 °C) have been investigated. The correlation between this phenomenon and structural inhomogeneity of the material has been determined. The parameters of oscillations have been shown to be controlled by hydrogen diffusive motion.

1. Introduction

The investigation of optoelectrical properties at high temperatures is known to give valuable information about thermal equilibrium processes in a-Si:H and related alloys [1, 2]. These processes were shown to be controlled by structural rearrangements in the silicon network activated by the diffusive motion of hydrogen [3–5]. Hence the influence of structural inhomogeneities on relaxation processes may be easily deduced from optoelectrical measurements at elevated temperatures.

By detailed investigation of the photoconductivity of a-Si:H inhomogeneous films we have found the phenomenon of photoconductivity oscillations at temperatures above 150 °C. In this work we present the experimental data and carry out an analysis of the effect found.

2. Experiment

The a-Si:H films were prepared by an RF (13.56 MHz) glow-discharge decomposition of the silane mixture (10% SiH_4 + 90% H_2). The films were deposited on Corning 7059 glass at an RF power density of 0.3 W cm^{-2} at a substrate temperature of 220 °C and at a pressure in the reactor of 0.37 Torr. To promote the formation of an inhomogeneous structure, the RF power density was higher than the optimal value (0.1 W cm^{-2}). The thickness of the films was 0.5 μm. To exclude the influence of plasma-induced defects on the results of measurements, the films after deposition were annealed at 220 °C for 0.5 h in a vacuum of 10^{-5} Torr. The quality of films was determined by measurements of the dark conductivity and photoconductivity at room temperature. The dark conductivity was $2 \times 10^{-9}\ \Omega^{-1}\ cm^{-1}$ and the photoconductivity $8 \times 10^{-6}\ \Omega^{-1}\ cm^{-1}$ (at a light intensity of 10^{15} cm^{-2} s^{-1} and $\lambda = 0.633\ \mu m$).

The microstructure of the films was analysed by infrared (IR) spectroscopy data. The absorption line at 2075 cm^{-1} in undoped a-Si:H indicated structural inhomogeneity of the material which is connected with the presence of clustered Si–H and SiH_2 groups [6, 7]. The presence of residual impurities (O, N and C) in the films was controlled by depth profile

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Auger analysis. The impurities were found to be concentrated in a surface layer less than 30 Å thick. This layer was withdrawn by Ar ion bombardment (2 keV) before formation of aluminium contacts for photoconductivity measurements.

The aluminium coplanar electrodes were deposited on the top of a-Si:H films, and the distance between the contacts was 0.6 mm. The measured current exhibited a linear dependence on applied voltage in the range 0–15 V. This indicated the absence of the influence of surface band bending and metal–semiconductor interface on the results of measurements.

The stabilized power source was used to maintain the electric field strengths at 2×10^2 and 6×10^2 V cm^{-1}. The measuring cell was housed in a screened framework to eliminate the influence of external electric and magnetic fields.

To observe the oscillations the dependences of photoconductivity were measured during illumination with an He–Ne laser ($\lambda = 0.633$ μm) at constant temperatures above 150 °C. The intensities of light were 10^{15} and 10^{17} cm^{-2} s^{-1}. To exclude the influence of possible fluctations of light intensity on the results of measurements the stability of laser illumination was controlled during photoconductivity measurements.

Random errors in all experimental data were determined as the highest deviation from the average value by measurements of ten samples prepared under the same conditions. These values did not exceed the size of experimental points presented on all plots.

3. Results and discussion

Figure 1 shows the conductivity of a-Si:H films during light illumination and after the laser has been switched off at different temperatures, light intensities and applied electric fields. Oscillations of the photoconductivity are seen on all curves. The dark conductivity after the laser is switched off is lower than it was before illumination. Such a change in the dark conductivity under light illumination is known as the Staebler–Wronski effect and is usually observed for doped and undoped a-Si:H films [8, 9].

As one can see from figure 1 the oscillations of the photoconductivity have a sinusoidal character and its amplitudes are damped. The increase in light itensity results in both an increase in amplitude and a decrease in the period of oscillations. The magnitude of the electric field does not affect the period of oscillations and slightly changes the value of the amplitude.

The analysis of the photoconductivity behaviour has been performed by decomposition of experimental curves into a base line about which oscillations occur and the oscillations themselves. The results of this procedure for the a-Si:H film at 150 °C (light intensity, 10^{17} cm^{-2} s^{-1}; field strength, 6×10^2 V cm^{-1}) are shown in figure 2. It was found that the base line can be satisfactorily described by a stretched-exponential dependence [10]:

$$\sigma(t) = \{\sigma_{\infty 1} - (\sigma_{\infty 1} - \sigma_{01}) \exp[-(t/\tau_1)^{\beta 1}]\} \tag{1}$$

where $\sigma(t)$ is the photoconductivity at time t, σ_{01} the initial value of photoconductivity at time $t = 0$; $\sigma_{\infty 1}$ the steady state photoconductivity, τ_1 the characteristic time and β_1 the dispersive parameter. The results of mathematical simulation of light-induced changes in a-Si:H according to dependence (1) are shown in figure 2 as a full curve. The parameters of the stretched exponents determined from the best correlation between simulating curves and experimental data are presented in table 1. As one can see, the characteristic time τ_1 decreases with increasing light intensity, electric field and temperature.

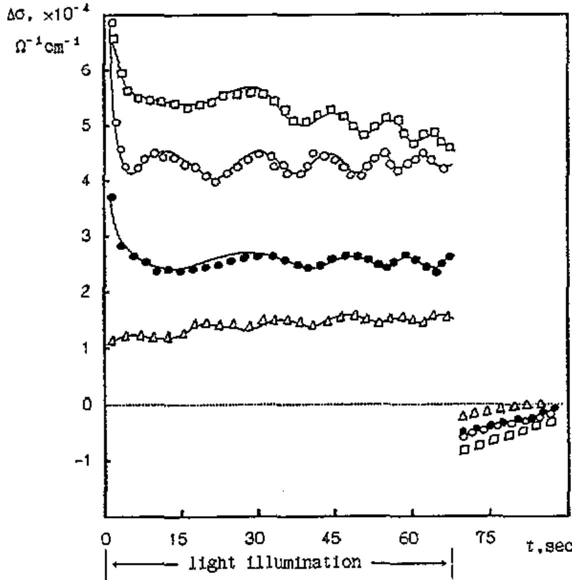

Figure 1. Light-induced conductivity changes $\Delta\sigma$ with time t for a-Si:H films at various temperatures T, light intensities I and applied electric field strengths U:——, simulated curves; ●, $T = 150\,°C$, $I = 10^{15}\ cm^{-2}\ s^{-1}$, $U = 2\times10^{2}\ V\ cm^{-1}$; ○, $T = 150\,°C$, $I = 10^{17}\ cm^{-2}\ s^{-1}$, $U = 2\times10^{2}\ V\ cm^{-1}$; □, $T = 150\,°C$, $I = 10^{17}\ cm^{-2}\ s^{-1}$, $U = 6\times10^{2}\ V\ cm^{-1}$; Δ, $T = 200\,°C$, $I = 10^{17}\ cm^{-2}\ s^{-1}$, $U = 2\times10^{2}\ V\ cm^{-1}$.

Table 1. The fitting parameters of the simulation of photoconductivity oscillations at various temperatures, light intensities and applied electric fields.

Temperature (°C)	Light intensity ($cm^{-2}\ s^{-1}$)	Electric field strength ($V\ cm^{-1}$)	τ_1	β_1	τ_{osc}	β_{osc}	T_{osc}	τ_2	φ
150	10^{15}	2×10^{2}	120	0.7	120	0.8	35	110	2.5
150	10^{17}	2×10^{2}	115	0.9	100	1.0	26	95	4.5
150	10^{17}	6×10^{2}	90	0.7	100	1.0	28	75	4.0
200	10^{17}	2×10^{2}	28	0.9	40	0.9	45	50	2.3

The oscillations (inset in figure 2) could be mathematically simulated by the following dependence:

$$\sigma(t) = \{\sigma_{osc}\exp[-(t/\tau_{osc})^{\beta_{osc}}]\}\sin\{2\pi t/[T_{osc}\exp(-t/\tau_2)] + \varphi\}. \qquad (2)$$

The stretched-exponential dependence in the first set of curly brackets describes the damping of the oscillation amplitude σ_{osc} with the parameters τ_{osc} and β_{osc}. The sinusoidal function describes the oscillations of photoconductivity with the exponentially damped period of $T_{osc}\exp(-t/\tau_2)$: τ_2 is the characteristic time. The parameter φ is used only for the initial shift in the sinusoidal function for best coincidence between the experimental and simulated curves.

Equation (2) contains six parameters, but the parameters (T_{osc} and τ_2) controlling the decrease in the oscillation period may be fitted independently from the parameters σ_{osc}, τ_{osc} and β_{osc} which determine the damping of the oscillation amplitude. Moreover the parameter φ is independent of the other parameters. For these reasons the problem of fitting was greatly

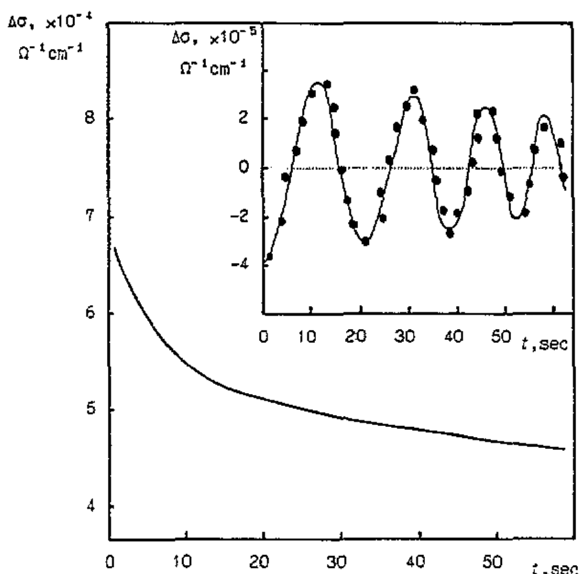

Figure 2. The base line and own oscillations (inset) of light-induced conductivity changes $\Delta\sigma$ on time t for a-Si:H film at a temperature of 150 °C, light intensity of 10^{17} cm^{-2} s^{-1} and an applied electric field strength 6×10^2 V cm^{-1}: ——, simulated curves.

simplified. Firstly the parameters T_{osc} and τ_2 describing the period of oscillations and φ were fitted; then we fitted the parameters σ_{osc}, τ_{osc} and β_{osc} which relate to the amplitude of the oscillations. The fitting of simulated and experimental curves was carried out by using the least-squares method with an error of less than 0.5%.

The results of simulation of oscillation by equation (2) are shown as a full curve in the inset in figure 2. The complete description of the experimental curve of light-induced photoconductivity changes was obtained by superposition of dependences (1) and (2). The results of such simulation are presented in figure 1 as full curves. Good coincidence between simulated curves and experimental data is seen.

The parameters of modelling for a-Si:H films at various temperatures, exciting light intensities and applied electric fields are presented in table 1. It can be seen that the period of oscillations increases with increase in temperature and decrease in light intensity. Moreover the characteristic times which describe both the change in base line and the damping of oscillations have similar values. It leads to information about the common nature of processes which control the damping of oscillations and decay of photoconductivity under illumination.

The low-frequency oscillations of the photocurrent are known to be found in II–VI compounds such as ZnSe [11] and CdS [12], but that phenomenon has been observed only at low temperatures of about 77 K and under simultaneous irradiation by visible and infrared light. Some attempts have been made to describe this effect but no satisfactory explanation has been obtained.

The oscillations of photoconductivity in a-Si:H were shown to be observed without additional IR irradiation. As is seen from the results presented in table 1, the periods of oscillations at high temperatures are too long for processes of trapping and detrapping of excess charge carriers to occur. On the other hand, at temperatures above 150 °C, thermal equilibration processes in a-Si:H films are observed [2]. According to the usual models

[13–17] these processes are assisted by hydrogen diffusive motion.

The hydrogen diffusion coefficient D_H in a-Si:H is reported to be 10^{-20} cm^2 s^{-1} at 160 °C [13]. A higher diffusion coefficient was also observed in undoped a-Si:H deposited under non-optimal growth conditions [14, 15]. The increase in D_H by about four orders of magnitude was shown to be connected with the presence of large voids or columnar growth morphology in the macroscopic film structure [16, 17].

According to IR spectroscopy analysis the a-Si:H films investigated contain clustered hydrogen and SiH_2 groups which are usually connected with the presence of microvoids, micropores, etc, in the material [6, 7]. Assuming that oscillations are caused by hydrogen interatomic hopping (0.5 nm) and considering the period of oscillations to be 35 s (see table 1) one can obtain a value for the diffusion coefficient of 1.8×10^{-17} cm^{-2} s^{-1}. This value of D_H is reasonable for inhomogeneous a-Si:H films [16, 17]. Moreover it means that hydrogen hopping occurs in a spatial region which contains clustered hydrogen. On the other hand this value of D_H and the characteristic time of damping of 120 s give the distance of hydrogen motion as about 1 nm. Thus the damping of oscillations could be connected with the removal of hydrogen from clustered Si–H bonds. The stretched-exponential character of damping in this case follows from the distribution of weak Si–Si bonds near clustered Si–H bonds.

In previous work [18] we have considered the model of the microstructure for inhomogeneous a-Si:H films. The microstructure of this material consists of homogeneous region of the silicon network (material bulk) separated by internal boundaries containing mostly distorted Si–Si bonds and clustered Si–H and SiH_2 groups (figure 3).

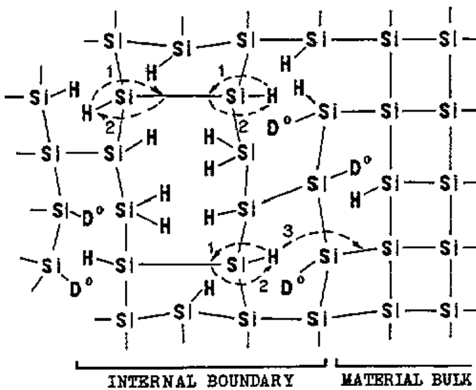

Figure 3. The scheme of microstructure for inhomogeneous a-Si:H films (D^0 are dangling bonds).

The recombination of excess charge carriers induced by light illumination firstly occurs at the states formed by weak Si–Si bonds at internal boundaries and lowers its defect formation energy [4]. The creation of metastable defects in this case occurs owing to hydrogen interatomic hopping from clusters to those states (process 1 in figure 3). On the other hand the sites which hydrogen leaves become recombination centres and thus are the potential sites for hydrogen back hopping (process 2 in figure 3). Hence reversible hydrogen motion in the single-hop limit may take place [3]. The corresponding periodic change in recombination centre density leads to oscillations in the photoconductivity. At high temperatures the hydrogen atoms are homogeneously redistributed in the material and create metastable defects in the bulk (process 3 in figure 3). Thus the damping of photoconductivity oscillations is connected with the diffusion of hydrogen from internal boundaries into the bulk. Therefore these processes will be observed in structurally inhomogeneous material

with nearest positions of clustered hydrogen and defect creation sites. However, the question of whether the oscillations are connected with synchronized hopping of hydrogen still remains unclear. If it is, what is the nature of such self-organization?

4. Conclusions

In conclusion, photoconductivity oscillations in a-Si:H at elevated temperatures (above 150 °C) were found. The oscillations are damped and their amplitude and period depend on the temperature, light intensity and applied electric field. This effect was shown to be connected with the structural inhomogeneity of the material, and the time scale of oscillation coincides with that of hydrogen interatomic hopping at elevated temperatures.

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