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Optical nonlinearities and thermal lens effect of a-Si:H films investigated by Z-scan technique

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Abstract

Nonlinear optical effects have been studied in hydrogenated amorphous silicon films through the single beam Z-scan technique using a modulated CW laser in the millisecond time-scale regime. In these experiments, the samples were moved through the focal region of a focused gaussian laser beam (λ = 532 nm) while the light transmittance in the far field was measured. The thermal lens effect was observed in the transmittance signal and the time-resolved Z-scan mode could therefore be used to achieve the samples thermal diffusivity ($D \sim 3 \times 10^{-3}$ cm^2/s) and thermal conductivity ($K \sim 5 \times 10^{-3}$ W/Kcm). The Thermal Lens Model was used to determine the samples temperature coefficient of the optical path length change (ds/dT), and the effective thermal nonlinear refractive index (n_2) was estimated by the on-axis non-linear phase shift at focus measurements.

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Keywords: hydrogenated amorphous silicon; thermal lens effect; Z-scan technique; thermal diffusivity; PECVD.

1. Introduction

Hydrogenated amorphous silicon (a-Si:H) is an important material to electronic industry. It has been largely used in electronics devices such as solar cells, thin-film transistor (TFT) in active matrix liquid crystal display and sensor devices. Nowadays, silicon photonics is a subject of great interest for the future of computing, information technology and telecommunication because silicon based photonic devices, such as integrated lasers, waveguides and optical switching on a chip, can generate, process and control information through light signals beyond today limits imposed by electronics [1, 2]. Amorphous silicon has its place in that technologic area [3]. Thus, the study of nonlinear optical properties of silicon is fundamental. In particular, this work contributes to the study of nonlinear optical effects caused by thermo-optical mechanisms.

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2. Theoretical Background

When light is absorbed by a material and non-radioactive decay occurs, the absorbed photon energy is converted into heat, what causes a local increase of the material temperature. The temperature change in that material causes changes in its optical path length, affecting the light propagation through it. This nonlinear optical effect is known as the thermal lens effect [4, 5]. The Thermal Lens Model (TLM) predicts how the transmission of the light through the sample will vary as a function of the sample position *z* and the time *t,* when the sample is subjected to the Z-scan experiment (see experimental section). The TLM normalized transmittance T_N is [5, 6]:

$$T_N(z,t) = \frac{I(z,t)}{I(z,0)} = \left\{ 1 + \left[\frac{\theta}{1 + t_{c0}(1+x^2)/2t} \right] \frac{2x}{1+x^2} + \left[\frac{\theta}{1 + t_{c0}(1+x^2)/2t} \right]^2 \frac{1}{1+x^2} \right\}^{-1} \quad (1)$$

where I is the transmitted on-axis light intensity, $x = z/z_0$, z is the sample position, z_0 is the light diffraction length given by $z_0 = \pi w_0^2/\lambda$, w_0 is the beam waist, λ is the light wavelength, t_{c0} is the thermal lens characteristic time at focus and θ is the phase shift due the thermal lens effect given by the set of equations (2), (3) and (4) [4-6]:

$$\theta = -\frac{\alpha P L_{eff}}{\lambda \mathrm{K}} \frac{ds}{dT} \quad (2)$$

$$\mathrm{K} = D\rho c \quad (3)$$

$$D = \frac{w_0^2}{4t_{c0}} \quad (4)$$

where α is the sample linear absorption coefficient, P is the incident power, L is the sample length, $L_{\mathrm{eff}} = (1 - e^{-\alpha L})/\alpha$ is the effective length, ds/dT is the temperature coefficient of the optical path length and K is the sample thermal conductivity, D is the sample thermal diffusivity, ρ is the density and c is the specific heat. The effective nonlinear refractive index (n_2), caused by a phase shift due the thermal lens effect, can be estimated by eq. (5) [6], considering dn/dT ~ ds/dT, where dn/dT is the temperature coefficient of the refractive index:

$$n_2 = \frac{\alpha w_0^2}{4\mathrm{K}} \frac{dn}{dT} \quad (5)$$

3. Experimental

3.1 Samples

The samples were deposited by PECVD on glass substrate (plasma source setup: P_s = 25W; f_s = 13.56 MHz) from silane (SiH_4), hydrogen (H_2) and hydrogen diluted phosphine (4.94 % PH_3 in H_2). The gases input flows were 10 sccm (SiH_4) and 250 sccm (H_2). The flow of PH_3 was varied from 0 to 40 sccm. Depositions were made with substrate temperature from 50 to 200 °C. The a-Si:H films were about 200 nm thick.

3.2 Z-scan setup

The Z-scan experiment performed in this work is schematically shown in fig. 1. The sample was translated through a focused gaussian laser beam while the transmitted intensity was measured in the far field [7, 8]. The radius (and the area) of a focused gaussian beam changes with *z,* and has its minimum (w_0 ~ 32 μm) at focus (z = 0) where the light intensity is maximum. Around the focus position the intensity is big enough to produce optical nonlinearities. A Nd:YVO_4 CW laser (λ = 532 nm) was modulated by a mechanical chopper to produce milliseconds pulses (pulse width ~ 40 ms). The on-axis intensity measurements (sensible to the phase changes) are achieved using an aperture before the photodetector D1, while the photodetector D2 measures the total intensity change.

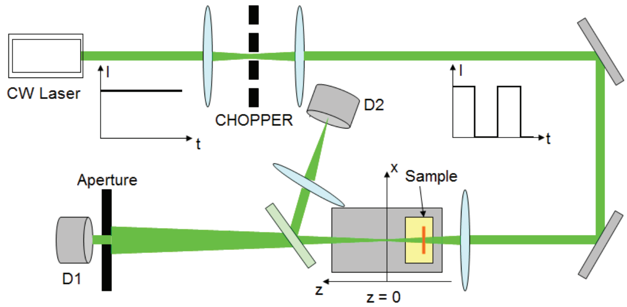

Fig. 1 – Z-scan setup used in the experiments. D1 is a photodetector used for measuring on-axis intensity changes and D2 is a photodetector used for measuring total intensity changes.

4. Results and discussion

Fig. 2 shows the experimental time-resolved Z-scan data for the sample with substrate temperature of 50 ^oC and 2.5 sccm of PH_3 flow. The continuous line is the theoretical fitting of eq. 1. Fig. 2a shows the normalized transmittance as a function of z (at t = 30 ms). The curve valley-peak behavior is typical of a converging thermal lens. Fig. 2b and 2c show the temporal evolution of the normalized transmittance at a fixed position before and after the focus and the fitting of eq. 1. The transmitted intensity decreases (2b) or increases (2c) some milliseconds after the pulse beginning, showing that the phenomenon origin is thermal. The t_{c0} and θ values achieved through the fittings for this sample and for all others are shown in table 1. The samples were identified by substrate temperature and PH_3 flow values used during deposition.

Table 1 - All samples´ t_{c0} and θ.

T_{sub} (°C)	PH_3 flow (sccm)	tc_0 (ms)	θ (rad)
200	10	0.4 ± 0.2	-0.099 ± 0.006
150	10	1.0 ± 0.2	-0.165 ± 0.008
200	10	2.3 ± 0.3	-0.158 ± 0.004
90	10	0.7 ± 0.3	-0.072 ± 0.004
70	10	2.6 ± 0.6	-0.096 ± 0.004
50	40	3.4 ± 1.8	-0.052 ± 0.006
50	20	0.4 ± 0.3	-0.035 ± 0.006
50	10	4.0 ± 1.8	-0.031 ± 0.005
50	2.5	1.0 ± 0.2	-0.094 ± 0.004

Table 2 summarizes the achieved values for the thermal diffusivity D from eq. 4 with experimental t_{c0} shown in table 1, thermal conductivity K from eq. 3 (the values ρ = 2.21 g/cm^3 and c = 0.62 J/gK for the a-Si:H were from [9]), the temperature coefficient of the optical path length ds/dT from eq. 2 and effective nonlinear refractive index n_2 from eq. 5.

The values obtained for the thermal diffusivity have the same order of magnitude of the values previously reported for thin films of similar compositions. For the sample deposited with T_{sub} = 150 ^oC, for example, D = (2.0 ± 0.4) x 10^{-3} cm^2/s. Messias et al. [10] measured D = 1.9 x 10^{-3} cm^2/s for a-Si:H thin films by thermal lens (TL) technique while Anjos et al. [11] measured D = 1.6 x 10^{-3} cm^2/s for amorphous silicon nitride thin films also by TL technique.

The values obtained for d*s*/d*T* have the same order of magnitude of values measured by TL for some laser glasses [4], but is at least 2 orders of magnitude smaller than the value of a 5-μm-thick waveguide layer of a-Si:H measured by interference in Fabry-Perot cavities [12]. Further investigations have to be carried out to determine the influence of the film´s thickness in the d*s*/d*T* results for the a-si:H when the Z-scan technique is used.

The values of n_2 are only estimatives of what order of magnitude is the thermal nonlinear change in the refractive index to be compared with ultrafast electronic nonlinear change in the refractive index. In ultrafast nonlinearities, n_2 has another meaning: it is related to the real part of the third order optical susceptibility. However, this estimative confirms the theoretical and general estimative made by Boyd [13], that $n_{2(\text{effective thermal})} \sim 10^{-5}$ $cm^2/W >> n_{2(\text{electronic})}$.

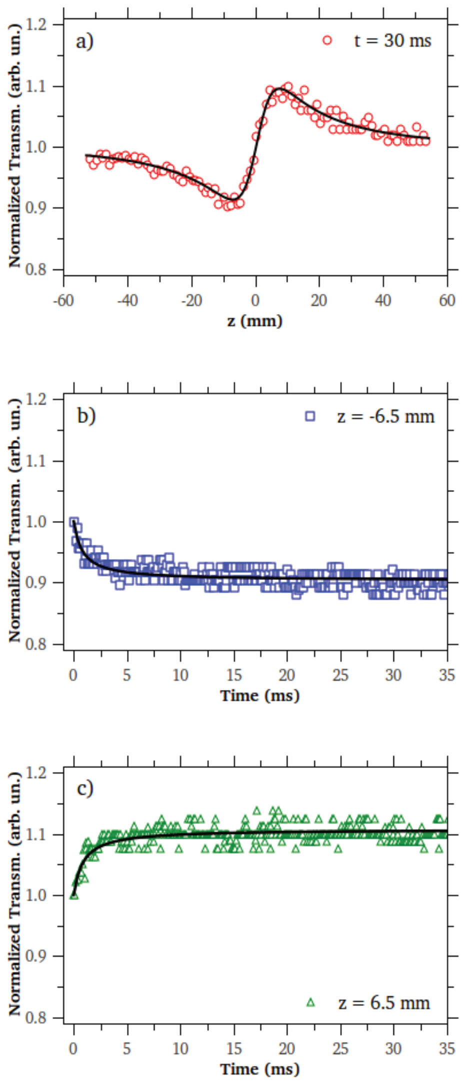

Fig. 2 - Experimental time-resolved Z-scan data. The continuous line is the theoretical fitting of eq. 1. (a) shows the normalized transmittance as a function of *z* (at *t* = 30 ms). (b) and (c) show the temporal evolution of the normalized transmittance at a fixed position before and after the focus and the fitting of eq. 1.

Table 2 – Optical and thermal properties obtained for the samples.

T_{sub} (°C)	PH_3 flow (sccm)	D (10^{-3} cm²/s)	K (10^{-3} W/Kcm)	ds/dT (10^{-7} K^{-1})	$n_2^{(ef)}$ (10^{-5} cm²/W)
200	10	5.3 ± 2.6	7.2 ± 3.6	20 ± 10	3.8
150	10	2.0 ± 0.4	2.7 ± 0.5	10.5 ± 2.2	4.0
200	10	0.9 ± 0.1	1.3 ± 0.2	4.59 ± 0.79	2.2
90	10	3.7 ± 1.4	5.0 ± 1.9	6.7 ± 2.7	0.37
70	10	1.0 ± 0.2	1.4 ± 0.3	2.87 ± 0.71	0.64
50	40	0.8 ± 0.5	1.0 ± 0.5	1.68 ± 0.90	0.26
50	20	6.2 ± 4.3	8.6 ± 5.8	8.8 ± 6.3	0.25
50	10	0.6 ± 0.3	0.9 ± 0.4	0.68 ± 0.33	0.21
50	2.5	3.3 ± 0.7	4.5 ± 0.9	8.7 ± 2.1	0.88

Fig. 3 shows the achieved values of t_{c0} (fig. 3a) and D (fig 3b) to the samples with PH_3 flow of 10 sccm and different substrate temperatures. Apparently, t_{c0} decreases with substrate temperature increasing, and D increases with substrate temperature increasing, but more investigations is necessary to identify why this occurs. One possibility is the increasing of the silicon crystalline fraction with the substrate temperature, changing, in that way, the t_{c0} and D values.

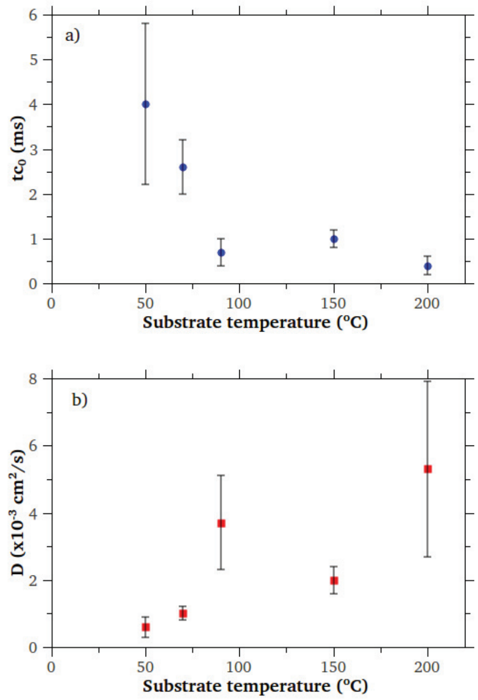

Fig. 3 - Values of t_{c0} (a) and D (b) for samples with different substrate temperature and 10 sccm of PH_3 flow.

5. Conclusion

The Z-scan technique was useful to measure the a-Si:H films characteristic time and thermal diffusivity. The values are consistent with the literature [4,10-11]. Apparently, t_{c0} and D depend on the substrate temperature, when the sample is deposited with 10 sccm of PH_3. The characteristic times of tenths and units of milliseconds indicates that the nonlinear effect is caused by thermal mechanism, as does the estimative of the effective nonlinear refractive index. Moreover, it was possible to estimate the a-Si:H thermal conductivity and the temperature coefficient of the optical path length with this experiments.

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