Transient thermal gratings and carrier-induced gratings in diffusion experiments

M. Niehus, S. Koynov, T. Múrias, and R. Schwarz
Instituto Superior Técnico, Departamento de Física, P-1096 Lisboa, Portugal
Contact: manfredo@fisica.ist.utl.pt

Key Words: Transient Grating, Thermal Diffusion, Ambipolar Diffusion, Photoreflection

Abstract

Complications can arise during measurements of the ambipolar diffusion coefficient, D_{amb}, of charged carriers by the transient grating method (TG) due to the concomitant decay of a transient thermal grating. The thermal diffusion coefficient, D_{th}, of hydrogenated amorphous silicon (a-Si:H) is of the same order of magnitude as D_{amb}, about 10^{-2} to 1 cm^2/s. For calibration purposes we measured the thermal change of refractive index, dn/dT, by optical reflection. We monitored the temperature decay after pulsed laser excitation using transient photoreflectance (TPR). A thermal diffusion process is confirmed by a near square-root time dependence. In grating experiments, the initial temperature and its time dependence can be monitored by TPR. In order to determine D_{amb} a good thermal connection to the substrate and a high quality material is needed.

1. Introduction

Laser pulses of ps and ns duration have been used in a number of measurements of the ambipolar diffusion coefficient, D_{amb}, of charged carriers by the transient grating method (TG) [1-4]. For example, in hydrogenated amorphous silicon (a-Si:H) the diffusion coefficient in the µs-time regime was found to range from 0.55 to 1.1×10^{-2} cm^2/s [1]. Laser pulse energies up to several mJ/cm^2 and concomitant local heating, however, may distort the expected result due to the superposition of a transient thermal grating (TTG) in addition to the transient carrier grating (TCG) when the thermal diffusion coefficients, D_{th} and D_{amb}, are of comparable magnitude. This problem was discussed in 1992 by Devlen and Schiff [5] who suggested that in the case of a-Si:H the typical film thickness was usually less than the grating periods employed which would reduce the thermal effects considerably.

The problem can be resolved for certain materials in the ps-time regime. As in the case of crystalline silicon, also in a-Si:H the contributions to the change of refractive index from sample heating and from carrier generation are of opposite sign:

$$\Delta n = \frac{\partial n}{\partial T}\Delta T - \sigma_{eh} N \qquad (1)$$

where σ_{eh} can be calculated within the Drude model [6]. Galeckas et al. [7] showed by transient photoinduced reflection (TPR) that initially the carrier density gave a fast negative signal (several 100 ps) followed by a slower positive signal due to sample heating to the ns- and µs-

Key Words: A200, D210, O110, R150, R160, T150, T280

range. It should then be expected that transient grating experiments performed on the same samples are indeed governed by carrier diffusion during the short time window of the experiment [3].

To separate thermal and carrier-induced contributions to the transient grating decay measurements in the ns-time regime is more difficult. In the experiments described below, we have therefore first measured the transient temperature increase by TPR. This measurement would correspond to measuring the zero-order-diffracted beam in the transient grating method. We have chosen a sample structure where heat conduction to the substrate is reduced for obtaining a larger temperature increase. The decay should then follow a square-root time dependence as predicted by diffusion theory. From a calibration of the TPR results we then quantify the thermal part in eq. (1). Finally, we will discuss the expected time dependence of the grating decay taking into account the absolute thermal and carrier diffusion coefficients.

2. Transient photoreflection measurements

A schematic used for the reflection measurements is shown in Fig. 1. The laser source is the frequency-doubled 532 nm line of a Q-switched Nd:YAG laser system. The power densities used are up to 20 mJ/cm^2 and the pulse duration is about 5 ns. As probe beam we used a HeNe laser (633 nm) with a cw power of 2 mW. The reflected HeNe beam was guided through an optical fiber to a photomultiplier tube with a 200 MHz bandwidth transient digitizer for data acquisition.

Figure 2 shows a typical TPR result from an amorphous multilayer sample. It consisted of 70 periods of a 2.5 nm thick a-Si:H and a 25 nm thick a-SiC:H bilayer, deposited on a Corning 7059 glass substrate. The TPR signal is the (normalized) change of reflectivity $\Delta R/R$ which is proportional to the (normalized) change of refractive index $\Delta n/n$ for the wavelength employed here. The TPR signal should therefore be a direct measure of the temperature near the sample surface.

But to know exactly what temperature we measure with the TPR signal of Fig. 2 we need to translate the signal height into a temperature increase via the thermal coefficient, dn/dT. This coefficient is obtained, for example, from the shift of the interference fringes in optical transmission spectroscopy, as shown for an a-Si:H sample in Fig. 3. Since fringe spacing depends on the product $n*d$, the shift will depend on both the change of refractive index n and on thermal expansion of the film of thickness d. However, as the thermal expansion is two orders of magnitude lower than the change of the refractive index, it is negligible. So we obtain $dn/dT = 5.6\text{x}10^{-4}$ K^{-1}, consistent with data from the literature [7].

In addition, we learn from Fig. 3 that depending on the position of the probe wavelength relative to a fringe maximum we might observe a different sign in the TPR signal. The change of absorption coefficient, in particular of the band gap E_g, is not important in our case, since the probe laser energy is higher than E_g.

3. Discussion

An estimate of the parameters involved in transient photoreflectance and in transient grating experiments will show the effect of sample heating. The efficiency of diffraction, η, into the first order direction in a grating experiment is given by [8]:

$$\eta = \frac{I_1}{I_0} = \left(\frac{\pi d}{\lambda}\Delta n\right)^2 + \left(\frac{d}{4}\Delta k\right)^2 \qquad (2)$$

Neglecting the change of optical absorption and considering the two terms of eq. (1) we obtain:

$$\eta = \left(\frac{\pi d}{\lambda}\right)^2 \left(\left(\frac{\partial n}{\partial T}\Delta T\right)^2 + \left(\sigma_{eh} N\right)^2 - 2\frac{\partial n}{\partial T}\Delta T \sigma_{eh} N\right) \qquad (3)$$

This means that there is a possibility that both contributions will cancel at a certain time. However, for t = 0 just after the pump laser pulse, and with sample thickness d 1 m and pump wavelength = 532 nm, we can estimate the following relative contributions:

$$\frac{\partial n}{\partial T} = 5.6\times10^{-4} K^{-1} \qquad \sigma_{eh} \approx 10^{-21} cm^3$$
$$\Delta T \approx 100\,^{\circ}C \qquad N \approx 10^{19} cm^{-3} \qquad \Rightarrow \frac{\eta_{th}}{\eta_c}(t=0) \approx 30$$
$$\eta_{th} \approx 0.078 \qquad \eta_c \approx 0.0025$$

The first and second column shows the thermal and the carrier-induced contributions, respectively. For the modest temperature we have chosen the initial thermal term is relatively large indeed.

3.1 Photoreflectance

If we consider a radially symmetric configuration to represent the TPR experiment then we can state the heat diffusion equation in the following form [9]:

$$k\nabla^2 T(r,t) = \frac{k}{D_{th}}\frac{\partial T(r,t)}{\partial t} - \frac{E_g}{\tau} N(r,t) - Q(r) \qquad (4)$$

where κ is the thermal conductivity and D_{th} the thermal diffusion coefficient. The source term $Q(r)$ depends on the absorption coefficient for the pump laser, its excess energy with respect to the band gap E_g, and on the laser beam profile. It vanishes for times $t > 0$. The second term on the right-hand side takes those carriers into account which recombine non-radiatively with a characteristic lifetime τ. Obviously, this term will lead to substantial complications since the

coupled second order partial differential equation governing carrier diffusion (and recombination) has to be solved simultaneously. However, assuming this term to be negligible the solution *T(r,t)* in the case of radially symmetric heat diffusion will be of the form:

$$T(r,t) \propto \frac{1}{\sqrt{t}} * \exp\left(-\frac{r^2}{4D_{th}t}\right) \qquad (5)$$

This square-root time dependence is tested for the TPR data in the double-logarithmic representation of Fig. 4. Since the probe beam is directed into the center of the pump beam spot, we have $r = 0$. The TPR data has a slightly faster decay that could be due to loss of heat into the substrate. The generally slow temperature decay may be attributed to the layered sample structure.

By definition, the TPR signal amplitude should be linear with the laser pulse energy, as is verified in Fig. 5. The decay at smaller laser intensity is slower which could be explained by the small and retarded heat release from trapped carriers.

3.2 Transient gratings

Let us now compare the expected grating decay curves under the two cases of purely thermal or purely carrier-induced transient gratings. The solution of the heat diffusion equation under illumination with a sinusoidal grating will lead to the following decay rate $1/\tau_{th}$ [8]:

$$\frac{1}{\tau_{th}} = \left(\frac{2\pi}{\Lambda}\right)^2 D_{th} \qquad (6)$$

In contrast, the carrier-induced grating decay is governed by two terms, the first one is governed by the recombination rate, and the second one is quite similar to the previous case:

$$\frac{1}{\tau_{carr}} = \frac{1}{\tau_{rec}} + \left(\frac{2\pi}{\Lambda}\right)^2 D_{amb} \qquad (7)$$

Figure 6 shows the experimental results from ref. (4) for the case of a n-type a-Si:H sample. The transient grating was induced in those experiments by two interfering laser beams of similar intensity as employed for the case of the TPR measurement described above. The resulting ambipolar diffusion coefficients calculated using eqs. (2) and (7) usually lie in the range of 0.5 to 2×10^{-2} cm^2/s in intrinsic, p- or n-type doped a-Si:H, and in microcrystalline Si films [4].

To test whether the results in Fig. 6 could come from the decay of a thermal grating we have plotted also the two types of grating decays using typical D_{th} and D_{amb} from the literature [4,7,9]. It is obvious that the thermal contribution, even though it might be important in magnitude at short times, will decay much faster than the carrier-induced grating. The thermal contribution in the experimental data can safely be assumed to be negligible, as we would have

observed an initial fast peak in addition to the exponential decay curves. Additionally, in the case of heat flow to the substrate, the temperature decrease would be much faster [5].

We can conclude that the film temperature is a very crucial parameter in TG experiments. It could conveniently be measured using the zero-order reflected probe beam intensity.

4. Conclusion

A crucial parameter to discriminate between transient thermal gratings and carrier-induced transient gratings in diffusion experiments is the magnitude of the film temperature. The initial temperature and its time dependence can be conveniently measured through transient photoreflection measurements. In the case of thin films of amorphous or microcrystalline silicon, a good thermal connection to the substrate of sufficiently large thermal conductivity, the use of high quality samples with large carrier lifetime, and the fact that thermal diffusion is much faster than ambipolar diffusion of charged carriers will ensure that carrier diffusion after excitation with ns-pulses can be well separated from thermal effects.

Acknowledgments: The authors thank E. Schiff for helpful discussions. M.N. acknowledges support through a stipend from the Foundation of Science and Technology FCT. R.S. acknowledges financial support from the German Academic Exchange Service DAAD. S.K. acknowledges financial support from NATO grant Nº426/10//10/12/97. This work is supported by the FCT project PRAXIS/PCEX/P/FIS/7/98.

References

[1] S. Komuro, Y. Aoyagi, Y. Segawa, S. Namba, A. Masuyama, H. Okamoto, and Y. Hamakawa, Appl. Phys. Lett. **42** (1983) 79.
[2] V.J. Newell, T.S. Rose, and M.D. Fayer, Phys. Rev. **32** (1985) 8035.
[3] M. Petrauskas, J. Kolenda, A. Galeckas, R. Schwarz, F. Wang, T. Muschik, T. Fischer, and H. Weinert, Mat. Res. Soc. Symp. Proc. **258** (1992) 553.
[4] R. Schwarz, S. Grebner, C.E. Nebel, M. Lanz, and M. Stutzmann, Mat. Res. Soc. Symp. Proc. **420** (1996) 723.
[5] R.I. Devlen and E.A. Schiff, J. of Non-Cryst. Sol. **141** (1992) 106.
[6] J.P. Woerdman, Philips Res. Reports Suppl. **7** (1977).
[7] A. Galeckas, M. Petrauskas, F. Wang, and R. Schwarz, phys. stat. sol. **(b) 190** (1995) 587.
[8] H.J. Eichler, P. Günther, and D.W. Pohl, *Laser-Induced Dynamic Gratings*, (Springer, Berlin, 1986).
[9] M. Liu, M.B. Suddendorf, M.B. Somekh, and S.J. Sheard, Semicond. Sci. Technol. **8** (1993) 1639.

Figure Captions

Fig.1: Experimental schematic of the transient photoreflectance measurements
(M: Mirror; L: Lens; PMT: Photomultiplier tube; t: Pulse width)

Fig. 2: TPR signal from an amorphous a-Si:H/a-SiC:H multilayer film

Fig. 3: Optical reflection spectra in a-Si:H for two temperatures. The resulting thermal coefficient of the refractive index is $dn/dT = 5.6 \times 10^{-4}\ K^{-1}$

Fig.4: Double-logarithmic representation of the TPR signal shown in Fig. 2. Lateral thermal diffusion in a thin film is indicated by the dotted line.

Fig.5: (a) Transient photoreflection for different laser beam intensities
(b) Linear beam intensity dependence of the peak TPR signal
(line is drawn as guide for the eye)

Fig.6: Experimental transient grating experiment in a n-type a-Si:H sample as described in ref. 4. The solid lines are calculated results of a pure carrier grating. The dotted lines inside the very fast time domain are calculated for a pure thermal grating.

Fig.1 (of 6) Experimental schematic of the transient photoreflectance measurements (M: Mirror; L: Lens; PMT: Photomultiplier tube; t: Pulse width)

File: Fig1.wmf

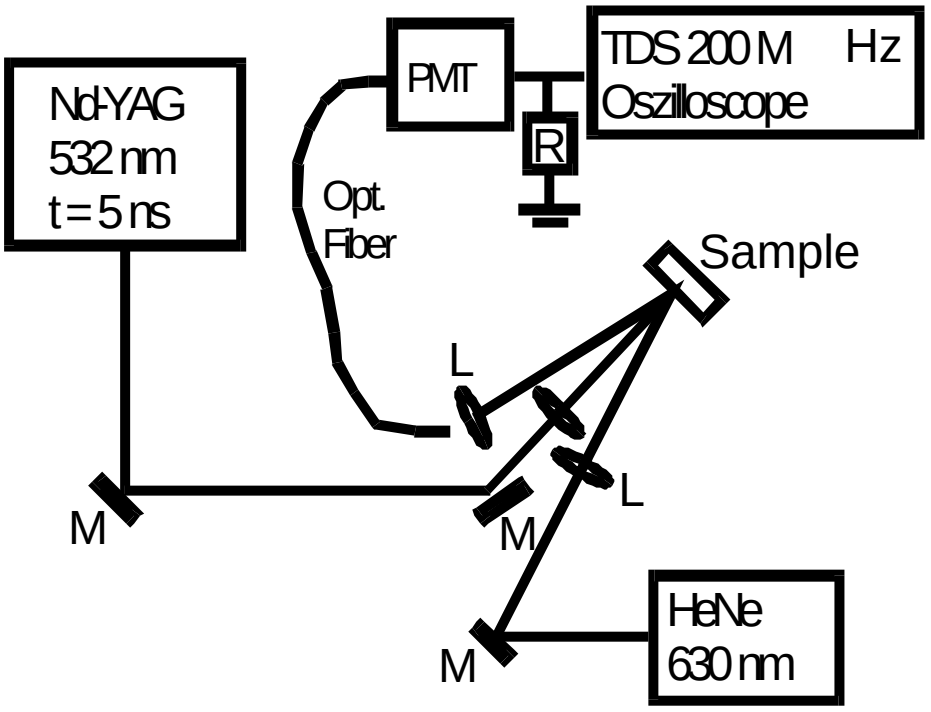


Fig.2 (of 6) TPR signal from an amorphous a-Si:H/a-SiC:H multilayer film

File: Fig2.wmf

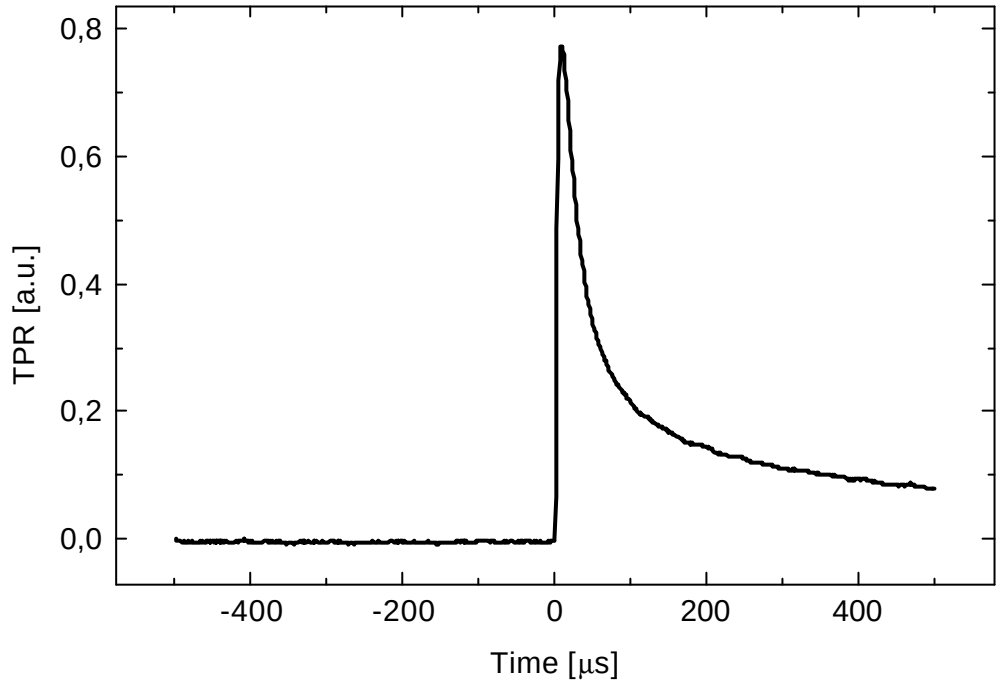


Fig.3 (of 6) Optical reflection spectra in a-Si:H for two temperatures. The resulting thermal coefficient of the refractive index is $dn/dT = 5.6 \times 10^{-4}\ K^{-1}$

File: Fig3.wmf

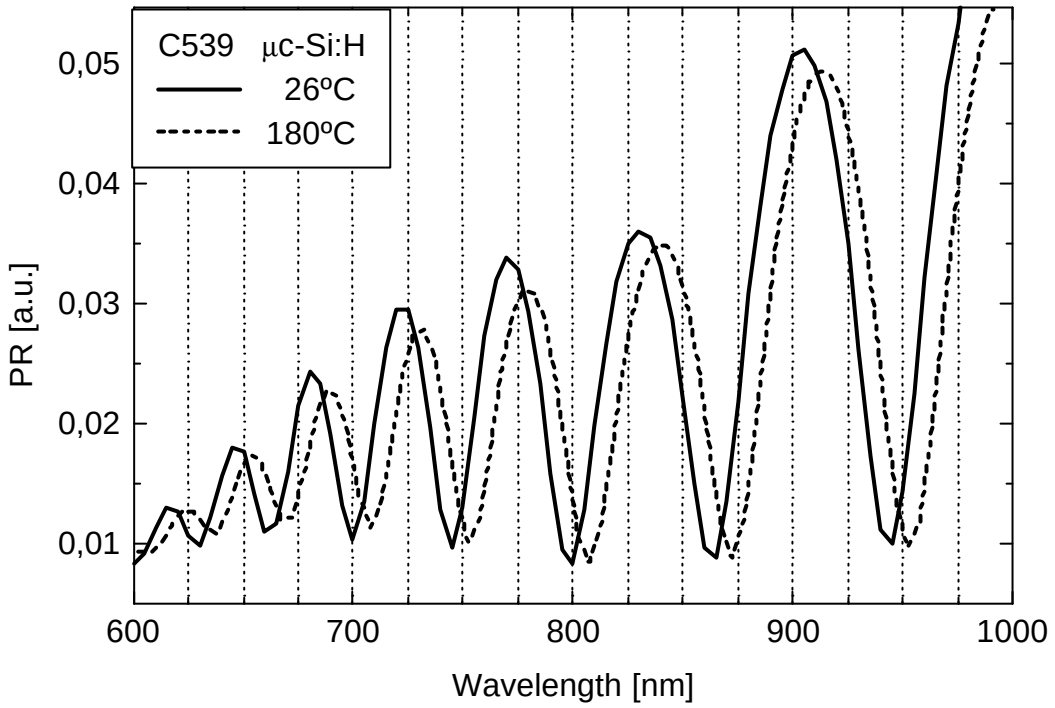


Fig 4 (of 6) Double-logarithmic representation of the TPR signal shown in Fig. 2. Lateral thermal diffusion in a thin film is indicated by the dotted line.

File: Fig4.wmf

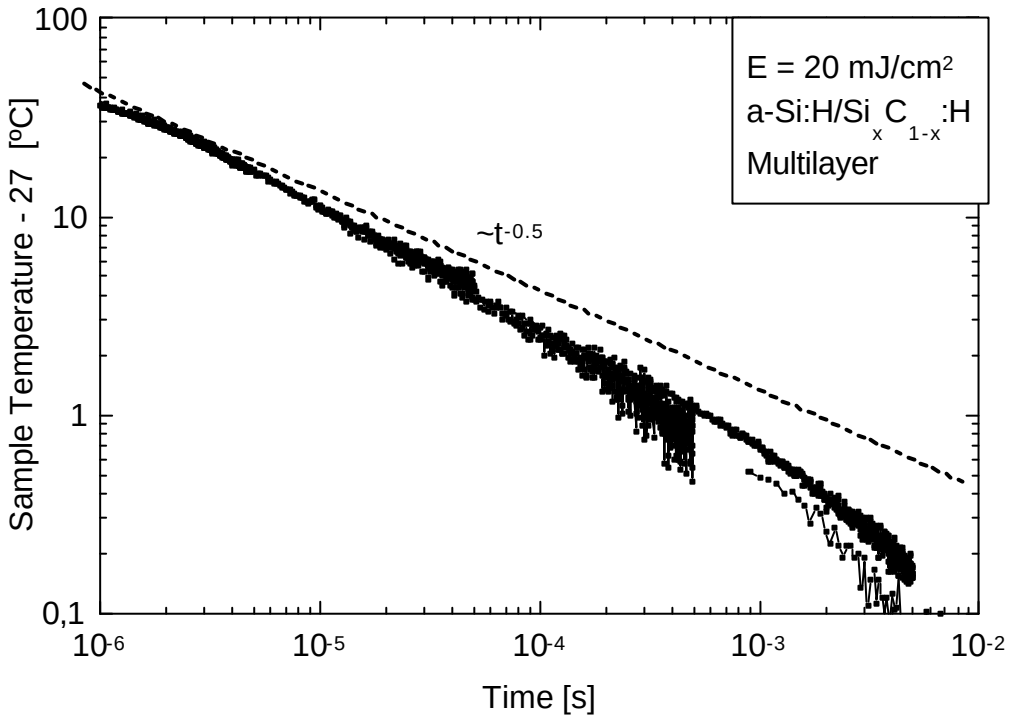
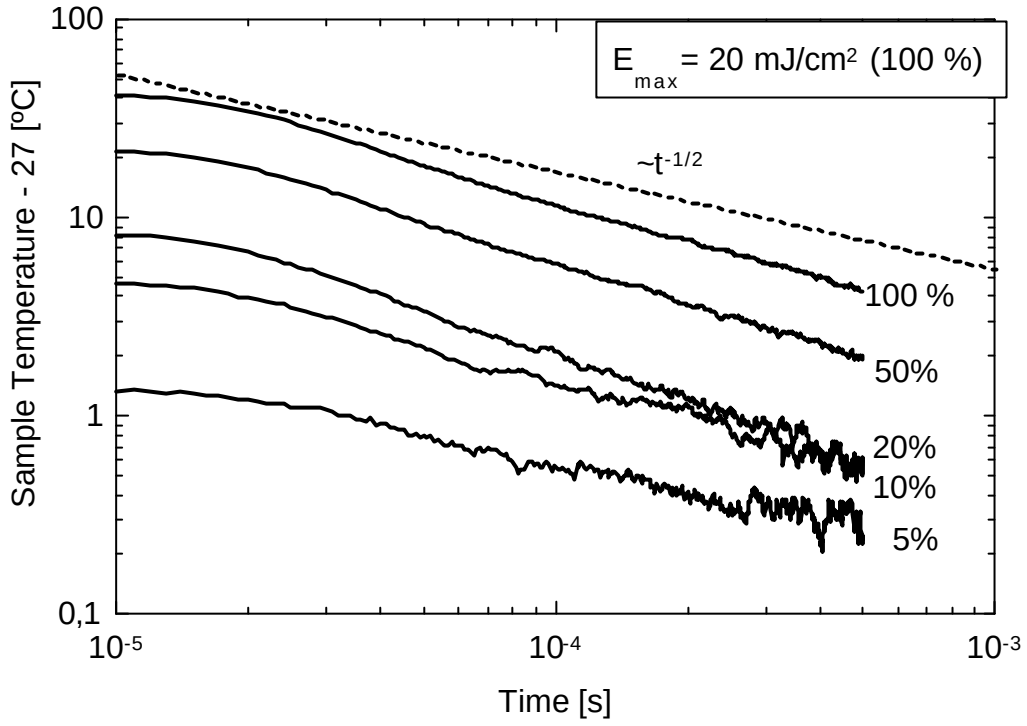


Fig.5 (of 6) (a) Transient photoreflection for different laser beam intensities
(b) Linear beam intensity dependence of the peak TPR signal
(line is drawn as guide for the eye)
Files: Fig5a.wmf (top), Fig5b.wmf (bottom)



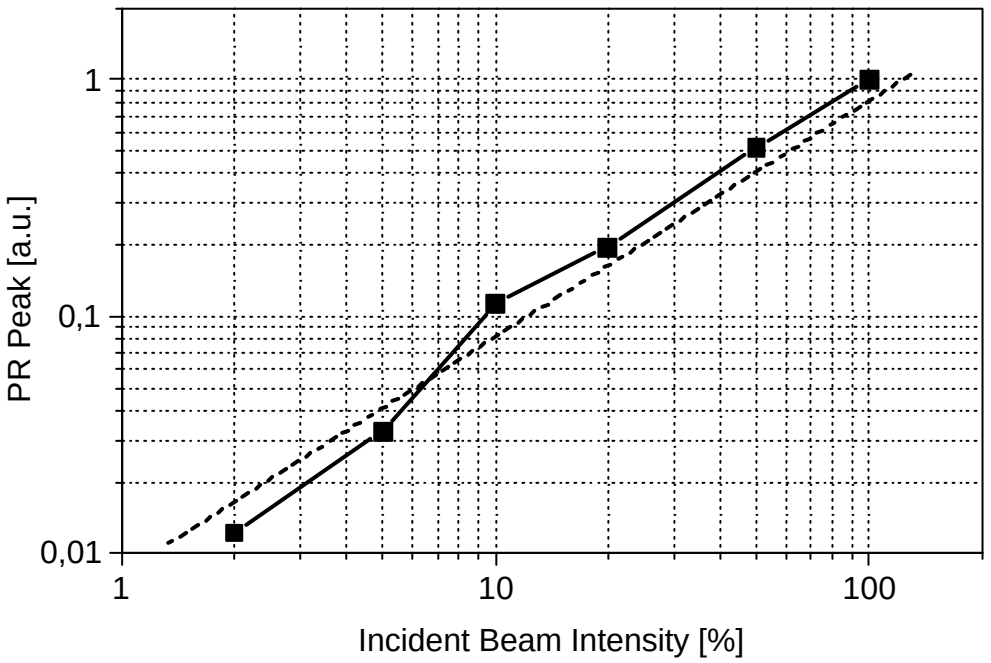


Fig.6 (of 6) Experimental transient grating experiment in a n-type a-Si:H sample as described in ref. 4. The solid lines are calculated results of a pure carrier grating. The dotted lines inside the very fast time domain are calculated for a pure thermal grating.

File: Fig6.wmf