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Published in:
Philosophical Magazine

DOI:
[10.1080/14786435.2018.1532120](https://doi.org/10.1080/14786435.2018.1532120)

Publication date:
2019

Document Version
Peer reviewed version

[Link to publication in Discovery Research Portal](#)

Citation for published version (APA):

Belgacem, H., & Reynolds, S. (2019). A new approach for determination of free carriers lifetime and density of localised states in disordered semiconductors. *Philosophical Magazine*, 99(2), 131-147.
<https://doi.org/10.1080/14786435.2018.1532120>

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Journal:	<i>Philosophical Magazine & Philosophical Magazine Letters</i>
Manuscript ID	TPHL-18-Jun-0063.R1
Journal Selection:	Philosophical Magazine
Date Submitted by the Author:	08-Sep-2018
Complete List of Authors:	Belgacem, Hocine; Faculty of Matter Science, University Hadj Lakhdar Batna 1, Department of Physics Reynolds, Stephen; University of Dundee, Electronic Engineering and Physics;
Keywords:	a-Si:H, amorphous semiconductors, electronic density of states, electronic properties, localized states, materials characterisation, thin films, transport properties
Keywords (user supplied):	Transient Photocurrent, Recombination lifetime, Laplace transform

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A new approach for determination of free carriers lifetime and density of localized states in
disordered semiconductors

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Abstract

A new method for measuring the free carriers lifetime (τ_R) and the density of localized states (DOS) in amorphous semiconductors is described. The method is based on the analysis of transient photoconductivity (TPC) data using Laplace transform technique. This technique involves Laplace transform of TPC data, it is a simple analysis of the solution of the basic linearized multiple trapping equations for free and trapped electrons. It is demonstrated by application to simulated and experimental TPC data measured on a typical disordered semiconductor, the hydrogenated amorphous silicon (a-Si:H). An introduced τ_R in the computing of the TPC using an arbitrarily proposed DOS model is recovered with high accuracy. For the experimental case, the determination of the exact DOS and the estimated τ_R from the experimental TPC data allow to reconstruct accurately this last. The performance and limitations of the technique are studied by means of computer simulations. Limitations are essentially the low temperatures of measurement of TPC when the recombination phenomenon is not observed at short times.

Keywords: Transient Photocurrent; Recombination lifetime; DOS; Laplace transform

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1. Introduction

Photodecay decay experiments have been used to measure the recombination lifetime of photogenerated carriers in semiconductors [1]. In amorphous semiconductors, the TPC decay experiments also enable us to determine the characteristic recombination time τ_c from the transition point from the pre-transit photocurrent I_{ph} vary as $t^{-(1-\alpha)}$ to the post-transit one vary as $t^{-(1+\alpha)}$ in case of the monomolecular recombination process [2] and by assuming an exponential decreasing density of localized states $g(E) = g(E_c) \exp\left(\frac{-E}{kT_c}\right)$, where $\alpha = \frac{T}{T_c}$ for $T < T_c$ is the dispersion parameter. Transitions to the post-transit region occur at various times, increasing with increasing τ_R . The τ_c can be related to τ_R by assuming the thermalization of photogenerated carriers into an exponential band tail of localized defects [2]. However, the precise measurement of τ_c suffers from the gradual transitions in the photocurrent from $I_{ph} \propto t^{-(1-\alpha)}$ to $I_{ph} \propto t^{-(1+\alpha)}$. In addition, it has been shown that the thermalization approximation does not work for structured distributions of localized tail states [3] and for exponential band tail states whose width is smaller than kT , where kT is the thermal energy. Information on τ_R is important for the understanding of nature of recombination centers in amorphous semiconductors. Since the transient photocurrent is intimately related to the energetic distribution of localized states and τ_R , it may be used to study them, and various techniques have been advanced for doing so [4-6]. The DOS and τ_R are key material quality indicators, of importance in the design of improved solar cells, photodetectors and thin-film transistors. A simple and reliable method is introduced to evaluate τ_R .

We develop in this paper a new method for measuring τ_R and DOS $g(E)$ in disordered semiconductors from an experimental transient photocurrent data. The present method consists of three steps: first, τ_R is estimated from the experimental TPC data using Laplace

transform technique. Secondly, exact DOS $g(E)$ is determined from the same experimental TPC data using the same Laplace transform technique [7-10]. Thirdly, computer-generated transient photocurrent using the calculated τ_R and DOS $g(E)$ must reproduce the experimental one, and thus to valid the method. The method is applied to the study of the effects of random noise and truncated data on τ_R for simulated TPC on the one hand and the effect of temperature on τ_R in undoped a-Si: H sample on the other hand.

2. Theory

2.1. Recombination lifetime determination

In the TPC technique, the transient photocurrent is determined by the time dependence of the photoconductivity following carrier excitation by means of a short pulse of illumination in samples having a coplanar electrode configuration [2]. For the theoretical analysis of this measurement, it suits to assume unipolar conduction, equal capture cross section for all localized states and small signal condition (the injected charge density is low enough that the localized states are not saturated and that the internal electric field is not perturbed) [7]. The small signal condition ensures the monomolecular recombination kinetics of photogenerated carriers and avoids space charge effects. We assume the transient photocurrent is carried solely by carriers in extend states, which interact through trapping and release with a distribution of localized states. Then, the basic linearized multiple trapping equations for free and trapped electrons respectively are:

$$\frac{dn(t)}{dt} = -\sum_i \frac{dn_i(t)}{dt} - \frac{n(t)}{\tau_R} + N_0 \delta(t), \quad (1)$$

$$\frac{dn_i(t)}{dt} = \omega_i n(t) - \gamma_i n_i(t), \quad (2)$$

where $n(t)$ is the free carrier density at time t , $n_i(t)$ the trapped carrier density at the i^{th} localized state at time t , N_0 the pulsed electron density (the excess free electron density at

the initial time $t = 0$), τ_R the recombination lifetime, $\omega_i = \sigma v g(E_i) \Delta E$ the capture rate constant, $\gamma_i = \nu \exp\left(\frac{-E_i}{kT}\right)$ the release rate constant, $E_i = i\Delta E$ the i^{th} energy level below (or above) a mobility edge, σ the capture cross section, v the thermal velocity and ν the attempt-to-escape frequency. The delta function in Eq. (1) defines the initial condition for the transient photoconductivity experiment. We set the conduction band mobility edge at $E_c = 0$, so that E is negative.

Eqs. (1) and (2) may be solved to yield the Laplace transform of the free carrier density $n(t)$. The Laplace transform of $n(t)$ is defined as:

$$\hat{n}(s) = \int_0^\infty n(t) e^{-st} dt$$

The solution of Eqs. (1) and (2) for $\hat{n}(s)$ is:

$$\hat{n}(s) = \frac{N_0}{s + \frac{1}{\tau_R} + \sum_i \frac{s\omega_i}{s + \gamma_i}}, \quad (3)$$

that is to say

$$\frac{1}{\tau_R} = \frac{N_0}{\hat{n}(s)} - s \left(1 + \sum_i \frac{\omega_i}{s + \gamma_i} \right), \quad (4)$$

in which s is the Laplace variable.

For the TPC experiment, the photocurrent is given by $I(t) = q\mu_0 F S n(t)$, where q is the electron charge, μ_0 the microscopic mobility, F the applied electric field and S the conduction cross section, which is transformed into

$$\hat{I}(s) = q\mu_0 F S \hat{n}(s). \quad (5)$$

Inserting Eq. (5) into Eq. (4), one obtains

$$\frac{1}{\tau_R} = \frac{q\mu_0 F S N_0}{\hat{I}(s)} - s \left(1 + \sum_i \frac{\omega_i}{s + \gamma_i} \right), \quad (6)$$

for $s = 0$ one obtains

$$\tau_R = \frac{\hat{I}(0)}{I(0)} = \frac{\int_0^\infty I(t)dt}{I(0)}, \quad (7)$$

where $I(0) = q\mu_0 FSN_0$, which is estimated from the extrapolated value of $I(t)$ to $t = 0$.

$\hat{I}(0)$ can be carried out by a simple numerical integration over N sampling points t_j :

$$\hat{I}(0) = \sum_{j=2}^{N-1} I(t_j) \Delta t_j, \quad (8)$$

where $\Delta t_j = \frac{t_{j+1} - t_{j-1}}{2}$.

Eq. (7) is a simple and adequate relationship that leads to the estimate of τ_R . Like one sees τ_R depends only on experimental $I(t)$ data and initial photocurrent $I(0)$, i.e. generated excess density N_0 .

To investigate the potential accuracy of the τ_R determination procedure, it is necessary to simulate accurately the transient photocurrent for the following two representative localized state distributions in amorphous and polymeric semiconductors: (i) an exponential distribution

$g(E) = g(E_c) \exp\left(\frac{-E}{kT_c}\right)$ and (ii) an exponential distribution on which a Gaussian distribution

has been superimposed $g(E) = g(E_c) \exp\left(\frac{E}{kT_c}\right) + g_g \exp\left[-\left(\frac{E - E_g}{E_w}\right)^2\right]$, where $g(E_c)$ is the

density of localized states at the mobility edge, T_c the characteristic temperature and g_g , E_g and E_w the peak value, the energy position from the mobility edge and the energy width of the Gaussian distribution, respectively.

In the present work $n(t)$ was carried out by means of an analytical expression for the inverse Laplace transform of $\hat{n}(s)$, obtained from Eq. (3), using Heaviside's expansion theorem [11].

$$n(t) = L^{-1} \{ \hat{n}(s) \} = \sum_{i=1}^{n+1} \frac{P(\alpha_i)}{Q'(\alpha_i)} e^{\alpha_i t}, \quad (9)$$

In Eq. (9) the α_i are the $n+1$ zeroes of the denominator $Q(s)$ and $Q'(\alpha_i)$ is the first derivative of $Q(s)$ taken at points α_i . $\hat{n}(s)$ is of the form of $\frac{P(s)}{Q(s)}$.

Fig. 1b shows the TPC, $n(t)$, computed at 300 K for the proposed exponential DOS model of Fig. 1a with various values of τ_R . The inflection points in the transients are due to the monomolecular recombination of the photogenerated free carriers. The characteristic recombination time determined from the inflection point, τ_c , is much larger than τ_R , this results from frequent trapping and detrapping of free carriers into the exponential localized-state distributions. The value of τ_c increases with τ_R . The τ_R used in the simulation of $n(t)$, are, respectively, 10^{-8} , 10^{-7} and 10^{-6} s. The corresponding calculated τ_R using Eq. (7) are, respectively, 9.0165×10^{-9} , 8.8164×10^{-8} and 8.7647×10^{-7} s. It can be seen that the introduced τ_R were recovered with high accuracy.

Fig. 2b shows the TPC, $n(t)$, computed at 350 K for the proposed exponential plus Gaussian DOS model of Fig. 2a with $\tau_R = 10^{-8}$ s. As in the case of computed TPC, $n(t)$, for an exponential DOS model, One can see well the inflection point in the transient due to recombination. The corresponding calculated τ_R using Eq. (7) is 8.7256×10^{-9} s. It can be seen here also that the proposed method recovers very precisely the introduced τ_R .

Figs. 3a, 3b, 3c and 4 show the TPC, $n(t)$ (symbol), of Figs. 1b ($\tau_R = 10^{-6}, 10^{-7}, 10^{-8}$ s) and 2b respectively, and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$ (Eq. (4)). One can see that $\tau_R(s^{-1})$ remains constant till the characteristic recombination time τ_c and equal to introduced value of τ_R in the simulation of $n(t)$. Normally τ_R is independent of s , but most important is that $\tau_R(s^{-1})$ is constant and equal to introduced τ_R for small values of s and for

$s = 0$ also of course (Eq. (7)). An important observation, the curve $\tau_R(s^{-1})$ deviates from introduced τ_R exactly at the moment when recombination begins, the curves $n(t)$ and $\tau_R(s^{-1})$ intersect at this specific time. Which means that the proposed method is not valid when the recombination phenomenon is not observed, this happens for low temperatures of measurement of $n(t)$, at short times.

2.2. Density of localized states

Eq. (6) can also be written as follows:

$$\sum_i \frac{g(E_i)}{s + \gamma_i} = \frac{1}{s \sigma v \Delta E} \left(\frac{q \mu_0 F S N_0}{\hat{I}(s)} - s - \frac{1}{\tau_R} \right), \quad (10)$$

We note here τ_R is experimentally known (Eq. (7)).

Eq. (10) is an integro-differential equation for the DOS $g(E)$, termed a Fredholm integral equation of the first kind [12] which may arise from an ‘ill-conditioned problem’. The inversion of this equation to obtain the DOS requires care, it needs a special resolution method. The resolution method used here is an exact matrix solution for $g(E)$ based on Tikhonov regularization [13,14]. Written in a discrete form, Eq. (10) yields

$$\sum_{i=1}^M g(i) A(j, i) = b(j) \quad \text{for } j = 2, \dots, N-1, \quad (11)$$

where i and j are the energy and time indexes respectively, and $g(i) = g(E_i)$. The matrix elements $A(j, i)$ and the vector elements $b(j)$ are respectively,

$$A(j, i) = \frac{1}{s(j) + \gamma(i)},$$

$$b(j) = \frac{1}{s(j) \sigma v \Delta E} \left(\frac{q \mu_0 F S N_0}{\hat{I}(j)} - s(j) - \frac{1}{\tau_R} \right),$$

where $\gamma(i) = \nu \exp\left(\frac{-E_i}{kT}\right)$, $s(j) = \frac{1}{t_j}$ and $\hat{I}(j) = \hat{I}(s_j)$.

The DOS vector is then given by

$$g = A^{-1}b. \quad (12)$$

This expression constitutes the basis of the transient method that returns for $\hat{I}(j)$ (i.e. the TPC data) a DOS distribution $g(E)$ of localized states.

3. Experiment

3.1. Preparation of sample

[15] A P-doped sample of a-Si:H was prepared by RF glow discharge decomposition of SiH_4 with 3 vppm PH_3 . Coplanar Al electrodes of 0.5 mm gap and 5 mm width were deposited on top of the film of 1 μm thickness, and the voltage applied across the gap was 400 V (i.e. an electric field of 8000 Vcm^{-1}). The light source employed was a Laser Science VSL337 N_2 -pumped 5 ns pulse dye laser, 640 nm, producing pulsed carrier densities of up to 10^{20} cm^{-3} , varied by means of neutral density filters. Single shots or low frequency ($< 1\text{Hz}$) laser pulses were used to allow complete carrier relaxation. The TPC signal was amplified where necessary, and measured using a PC-controlled Tektronix TDS380 oscilloscope as a transient recorder. A temperature range from 120–400 K was covered using a cryostat operating at a chamber pressure of typically 10^{-4} Torr. The dark Fermi level was estimated from the dark conductivity measurement to be 0.5 eV below the conduction band mobility edge.

3.2. Results

In the following, the free carriers lifetime τ_R and the energy profile of the DOS $g(E)$ are determined from an experimental TPC data. TPC are then generated by numerical simulation, using these τ_R and DOS, and compared to the experimental ones.

Fig. 5 shows a set of five TPC decays for the P-doped a-Si:H sample at 150, 200, 250, 290 and 310 K measured with an excitation density of $N_0 = 10^{16} \text{ cm}^{-3}$.

To valid the proposed method for the experimental case, we determinate τ_R and exact DOS $g(E)$ from the experimental TPC data of Fig. 5. We then simulate TPC using these τ_R and DOS $g(E)$ in order to reproduce the experimental ones. Using Eq. (7) for measured TPC decays at 200, 250 and 310K of Fig. 5, we find respectively the following values of τ_R : 1.2933×10^{-6} s, 9.7965×10^{-7} s and 8.552×10^{-7} s. And by inserting these values of τ_R into Eq. (10) for each case (200, 250 and 310K), we determine the energy profile of the DOS $g(E)$ after calculating the corresponding $\hat{I}(s)$ (Eq. (5)).

Fig. 6 shows the portion DOS $g(E)$, calculated by the inversion of Eq. (10), in the P-doped sample of Fig. 5 for the temperatures 200, 250 and 310K. The solid line is the DOS model obtained by fitting of the experimental DOS to the DOS expression:

$$g(E) = g(E_c) \exp\left(\frac{E}{kT_{c1}}\right) \left[1 - \frac{1}{1 + \exp\left(\frac{E - E_d}{kT_{c2}}\right)} \right] + g_g \exp\left[-\left(\frac{E - E_g}{E_w}\right)^2\right]. \quad (13)$$

Equation (13) indicates that the DOS model presents two exponential distributions parts on which a Gaussian distribution has been superimposed. One exponential distribution above E_d with high characteristic temperature T_{c1} and the other below E_d with low characteristic temperature $T_c = \frac{T_{c1}T_{c2}}{T_{c1} + T_{c2}}$. Least squares fitting gives $g(E_c) = 3.2279 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_d = 0.12417 \text{ eV}$ below E_c , $T_{c1} = 1321.3 \text{ K}$ and $T_{c2} = 211.49 \text{ K}$, resulting in $T_c \approx 182.3 \text{ K}$. For the Gaussian distribution, $g_g = 3.1292 \times 10^{15} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_g = 0.36888 \text{ eV}$ below E_c and $E_w = 29.685 \text{ meV}$. The result is a well-defined steep exponential conduction band tail of characteristic temperature T_c with a slight deviation towards E_c at the top to which is added a Gaussian distribution of energy width E_w and whose the peak, of value g_g , is positioned at E_g below the mobility edge E_c . The doping effect on the tail shape is remarkable, the DOS

with energies around the donor (P) band peak at a certain level E_d between 0.1eV and 0.2eV below E_c [16] is found to increase with doping. The DOS is then, a broad distribution above E_d , a narrow tail below it and a deeper Gaussian distribution, so that the DOS profile can be fitted with the DOS model of Eq. (13).

The parameters used to perform τ_R and the DOS $g(E)$ are the following:

$$\mu_0 = 10 \text{ cm}^2 \text{ s}^{-1} \text{ V}^{-1}, C_n = \sigma v = 2 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1} \text{ and } \nu = 10^{12} \text{ s}^{-1}.$$

Fig. 7 shows the computed TPC decays (solid lines) for the P-doped sample at temperatures 200, 250 and 310K using the DOS model of Fig. 6. The corresponding experimental TPC decays of Fig. 5 are also presented (symbols). It turns out that, the modelled TPC curves line up quite rigorously with the experimental ones.

4. Discussion

4.1. Temperature effect

Limitations of the method are essentially the effect of low temperatures of TPC measurement on τ_R when the recombination phenomenon is not observed at short times. Fig.

9 shows the TPC, $n(t)$, computed at 150K between initial time $t_i = \frac{1}{\nu}$ and various values of

final time $t_f = \frac{1}{\nu} \exp\left(\frac{E_c - E}{kT}\right)$ for the proposed exponential plus Gaussian DOS model of Fig.

8 with $\tau_R = 10^{-8} \text{ s}$. The chosen t_f are, respectively, 1, 10^2 and 10^4 s . The corresponding calculated τ_R using Eq. (7) are, respectively, 2.9693×10^{-9} , 9.8854×10^{-8} and $7.3038 \times 10^{-7} \text{ s}$.

It can be seen here that the proposed method does not recovers the introduced τ_R . We can retrieve the 'exact' value of the pre-proposed τ_R for low temperatures, provided to take extremely long times, which is not feasible experimentally. In conclusion we can say that the method is influenced by the temperature, it is valid only when the phenomenon of

recombination is observable, that is to say in the case of high temperatures. Higher temperature is better, as it shifts the recombination feature to shorter times, into the accessible time-region. Therefore, at low temperatures there is a little evidence of the effect of recombination on TPC over the experimental timescale.

4.2. Truncated data

Fig. 10 shows the TPC, $n(t)$ (solid line), computed at 350 K between initial time $t_i = 1$ ps and final time $t_f = 1$ s for the proposed exponential plus Gaussian DOS model of Fig. 2a with $\tau_R = 10^{-8}$ s and the truncated $n(t)$ (symbol o) at time 10 ns after t_i , i.e. 10 ns is the new starting time t_i . For the full data $n(t)$, τ_R was already been calculated (section 2.1), it is worth 8.7256×10^{-9} s and for the truncated one, τ_R is equal to 8.5912×10^{-9} s, it was calculated in the same way as in the case of complete data (Eq. 7). Fig. 11 shows the same full TPC, $n(t)$ (solid line) as of Fig. 10 and the truncated $n(t)$ (symbol o) at time $100 \mu\text{s}$ before t_f , i.e. $100 \mu\text{s}$ is the new final time t_f . For the truncated one, τ_R is equal to 8.7217×10^{-9} s. As we see the estimation of τ_R is not influenced by the missing of short or long time data. This implies that there must be some influence of recombination at short times, long before the 'final' recombination decay which we normally expect (and see) at long times.

4.3. Noisy data

Accuracy of measuring τ_R has been already investigated by application to simulated (perfect) data, but how good is it when used on real (imperfect) data, subject to noise? To do this, we simulate TPC, $n(t)$, for given DOS model and τ_R , and add Gaussian noise whose amplitude is a constant fraction of $n(t)$. Random noise is approximately Gaussian of similar amplitude over whole time range. Noise was introduced by multiplying each point of $n(t)$ by a random number from a Gaussian distribution whose mean value is 1. The standard deviation of the distribution was varied between 10% and 40%.

Fig. 12 shows the simulated (smoothed) TPC, $n(t)$ (solid line), of Fig. 2b and the noisy $n(t)$ (symbol o) when noise level is equal to 10%. Eq. (7) gives $\tau_R = 8.7256 \times 10^{-9}$ s for simulated $n(t)$ and 9.0484×10^{-9} s for noisy $n(t)$ when the used τ_R in the simulation of $n(t)$ is 10^{-8} s. It can be seen that the estimation of τ_R is not influenced by noise.

Fig. 13 shows the deviation $\frac{\text{smoothed data} - \text{noisy data}}{\text{smoothed data}}$ versus time in the case of 10% noise. As we see Gaussian noise is approximately uniform over entire time range.

Fig. 14 shows estimated τ_R versus noise level, this last varying between 10% and 40%. τ_R remains practically constant, and equal to 10^{-8} s, over whole noise level range. The method is 'noise tolerant', it is capable of returning optimal resolution for a given set of noisy data even at quite high input noise levels.

1
2 **5. Conclusion**
3

4 We have proposed a new method for determining the free carriers lifetime τ_R and the
5 density of localized states DOS in amorphous semiconductors from the transient photocurrent
6 TPC data. This technique involves Laplace transform of TPC data, it is a simple analysis of
7 the solution of the basic linearized multiple trapping equations for free and trapped electrons.
8 It has been tested by applying it to simulated and experimental TPC data measured on a
9 typical disordered semiconductor, the hydrogenated amorphous silicon (a-Si:H). An
10 introduced τ_R in the computing of the TPC using an arbitrarily proposed DOS model is
11 recovered with high accuracy. For the experimental case, the estimated τ_R and the
12 determination of the exact DOS from the experimental TPC data allow to reconstruct
13 accurately this last. Limitations of the technique are essentially the effect of low temperatures
14 of TPC measurement on τ_R when the recombination phenomenon is not observed at short
15 times. On the other hand, the determination of τ_R is not at all influenced by the missing of
16 short or long time data and the noisy data.
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Acknowledgements

The authors thank Amorphous Semiconductors group at Dundee University for the measurement facilities. The Algerian Ministry of Higher Education and Research is acknowledged for his financial support.

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Figure captions

Fig. 1. (a) - Exponential DOS model with $g(E_c) = 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$ and characteristic temperature $T_c = 400 \text{ K}$.

(b) - Simulated TPC, $n(t)$, at 300 K using DOS of (a) for different τ_R values.

Fig. 2. (a) - Exponential plus Gaussian distribution of states with $g(E_c) = 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$, $T_c = 400 \text{ K}$, $g_g = 10^{19} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_g = -0.35 \text{ eV}$ and $E_w = 40 \text{ meV}$.

(b) - Simulated TPC, $n(t)$, at 350 K using DOS of (a) for $\tau_R = 10^{-8} \text{ s}$.

Fig. 3. (a) - $n(t)$ (symbol o) of fig 1 (b) for $\tau_R = 10^{-6} \text{ s}$ and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$. For clarity, the curve $n(t)$ is multiplied by the factor 10^{-20} .

(b) - $n(t)$ (symbol o) of fig 1 (b) for $\tau_R = 10^{-7} \text{ s}$ and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$. For clarity, the curve $n(t)$ is multiplied by the factor 10^{-21} .

(c) - $n(t)$ (symbol o) of fig 1 (b) for $\tau_R = 10^{-8} \text{ s}$ and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$. For clarity, the curve $n(t)$ is multiplied by the factor 10^{-22} .

Fig. 4 - $n(t)$ (symbol o) of fig 2 (b) and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$. For clarity, the curve $n(t)$ is multiplied by the factor 10^{-21} .

Fig. 5 - Measured TPC decays for the P-doped a-Si:H sample at five different temperatures (150, 200, 250, 290 and 310 K) with an excitation density of $N_0 = 10^{16} \text{ cm}^{-3}$.

Fig. 6 - Experimental DOS $g(E)$ in the P-doped sample of Fig. 5 for the temperatures 200, 250 and 310 K (symbols). The solid line is the DOS model obtained by fitting of the experimental DOS with Eq. (13).

Fig. 7 - Simulated TPC decays (full curves) for the P-doped sample at temperatures 200, 250 and 310 K using the DOS model of Fig.7, showing good reconstruction of the corresponding experimental ones (symbols). The curves are offset vertically for clarity.

Fig. 8 - Exponential plus Gaussian distribution of states for various values of final time t_f ,
with $g(E_c) = 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$, $T_c = 400 \text{ K}$, $g_g = 10^{19} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_g = -0.3 \text{ eV}$ and
 $E_w = 40 \text{ meV}$. The curves are offset vertically for clarity.

Fig. 9 - Simulated TPC, $n(t)$, at 150 K using corresponding DOS of Fig. 8 for $\tau_R = 10^{-8} \text{ s}$.
The curves are offset vertically for clarity.

Fig. 10 - Full TPC, $n(t)$ (solid line), of Fig. 2b and truncated $n(t)$ (symbol o) at time 10 ns.

Fig. 11 - Full TPC, $n(t)$ (solid line), of Fig. 11 and truncated $n(t)$ (symbol o) at time 100 μs .

Fig. 12 - Smoothed TPC, $n(t)$ (solid line), of Fig. 2b and noisy $n(t)$ (symbol o) (10% noise).

Fig. 13 - Deviation $\frac{\text{smoothed data} - \text{noisy data}}{\text{smoothed data}}$ versus time (10% noise).

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Figure 2

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A new approach for determination of free carriers lifetime and density of localized states in
disordered semiconductors

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Abstract

A new method for measuring the free carriers lifetime (τ_R) and the density of localized states (DOS) in amorphous semiconductors is described. The method is based on the analysis of transient photoconductivity (TPC) data using Laplace transform technique. This technique involves Laplace transform of TPC data, it is a simple analysis of the solution of the basic linearized multiple trapping equations for free and trapped electrons. It is demonstrated by application to simulated and experimental TPC data measured on a typical disordered semiconductor, the hydrogenated amorphous silicon (a-Si:H). An introduced τ_R in the computing of the TPC using an arbitrarily proposed DOS model is recovered with high accuracy. For the experimental case, the determination of the exact DOS and the estimated τ_R from the experimental TPC data allow to reconstruct accurately this last. The performance and limitations of the technique are studied by means of computer simulations. Limitations are essentially the low temperatures of measurement of TPC when the recombination phenomenon is not observed at short times.

Keywords: Transient Photocurrent; Recombination lifetime; DOS; Laplace transform

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1. Introduction

Photodecay experiments have been used to measure the recombination lifetime of photogenerated carriers in semiconductors [1]. In amorphous semiconductors, the TPC decay experiments also enable us to determine the characteristic recombination time τ_c from the transition point from the pre-transit photocurrent I_{ph} vary as $t^{-(1-\alpha)}$ to the post-transit one vary as $t^{-(1+\alpha)}$ in case of the monomolecular recombination process [2] and by assuming an exponential decreasing density of localized states $g(E) = g(E_c) \exp\left(\frac{-E}{kT_c}\right)$, where $\alpha = \frac{T}{T_c}$ for $T < T_c$ is the dispersion parameter. Transitions to the post-transit region occur at various times, increasing with increasing τ_R . The τ_c can be related to τ_R by assuming the thermalization of photogenerated carriers into an exponential band tail of localized defects [2]. However, the precise measurement of τ_c suffers from the gradual transitions in the photocurrent from $I_{ph} \propto t^{-(1-\alpha)}$ to $I_{ph} \propto t^{-(1+\alpha)}$. In addition, it has been shown that the thermalization approximation does not work for structured distributions of localized tail states [3] and for exponential band tail states whose width is smaller than kT , where kT is the thermal energy. Information on τ_R is important for the understanding of nature of recombination centers in amorphous semiconductors. Since the transient photocurrent is intimately related to the energetic distribution of localized states and τ_R , it may be used to study them, and various techniques have been advanced for doing so [4-6]. The DOS and τ_R are key material quality indicators, of importance in the design of improved solar cells, photodetectors and thin-film transistors. A simple and reliable method is introduced to evaluate τ_R .

We develop in this paper a new method for measuring τ_R and DOS $g(E)$ in disordered semiconductors from an experimental transient photocurrent data. The present method consists of three steps: first, τ_R is estimated from the experimental TPC data using Laplace

transform technique. Secondly, exact DOS $g(E)$ is determined from the same experimental TPC data using the same Laplace transform technique [7-10]. Thirdly, computer-generated transient photocurrent using the calculated τ_R and DOS $g(E)$ must reproduce the experimental one, and thus to valid the method. The method is applied to the study of the effects of random noise and truncated data on τ_R for simulated TPC on the one hand and the effect of temperature on τ_R in undoped a-Si: H sample on the other hand.

2. Theory

2.1. Recombination lifetime determination

In the TPC technique, the transient photocurrent is determined by the time dependence of the photoconductivity following carrier excitation by means of a short pulse of illumination in samples having a coplanar electrode configuration [2]. For the theoretical analysis of this measurement, it suits to assume unipolar conduction, equal capture cross section for all localized states and small signal condition (the injected charge density is low enough that the localized states are not saturated and that the internal electric field is not perturbed) [7]. The small signal condition ensures the monomolecular recombination kinetics of photogenerated carriers and avoids space charge effects. We assume the transient photocurrent is carried solely by carriers in extend states, which interact through trapping and release with a distribution of localized states. Then, the basic linearized multiple trapping equations for free and trapped electrons respectively are:

$$\frac{dn(t)}{dt} = -\sum_i \frac{dn_i(t)}{dt} - \frac{n(t)}{\tau_R} + N_0 \delta(t), \quad (1)$$

$$\frac{dn_i(t)}{dt} = \omega_i n(t) - \gamma_i n_i(t), \quad (2)$$

where $n(t)$ is the free carrier density at time t , $n_i(t)$ the trapped carrier density at the i^{th} localized state at time t , N_0 the pulsed electron density (the excess free electron density at

the initial time $t = 0$), τ_R the recombination lifetime, $\omega_i = \sigma \nu g(E_i) \Delta E$ the capture rate constant, $\gamma_i = \nu \exp\left(\frac{-E_i}{kT}\right)$ the release rate constant, $E_i = i\Delta E$ the i^{th} energy level below (or above) a mobility edge, σ the capture cross section, ν the thermal velocity and ν the attempt-to-escape frequency. The delta function in Eq. (1) defines the initial condition for the transient photoconductivity experiment. We set the conduction band mobility edge at $E_c = 0$, so that E is negative.

Eqs. (1) and (2) may be solved to yield the Laplace transform of the free carrier density $n(t)$. The Laplace transform of $n(t)$ is defined as:

$$\hat{n}(s) = \int_0^\infty n(t) e^{-st} dt$$

The solution of Eqs. (1) and (2) for $\hat{n}(s)$ is:

$$\hat{n}(s) = \frac{N_0}{s + \frac{1}{\tau_R} + \sum_i \frac{s\omega_i}{s + \gamma_i}}, \quad (3)$$

that is to say

$$\frac{1}{\tau_R} = \frac{N_0}{\hat{n}(s)} - s \left(1 + \sum_i \frac{\omega_i}{s + \gamma_i} \right), \quad (4)$$

in which s is the Laplace variable.

For the TPC experiment, the photocurrent is given by $I(t) = q\mu_0 F S n(t)$, where q is the electron charge, μ_0 the microscopic mobility, F the applied electric field and S the conduction cross section, which is transformed into

$$\hat{I}(s) = q\mu_0 F S \hat{n}(s). \quad (5)$$

Inserting Eq. (5) into Eq. (4), one obtains

$$\frac{1}{\tau_R} = \frac{q\mu_0 F S N_0}{\hat{I}(s)} - s \left(1 + \sum_i \frac{\omega_i}{s + \gamma_i} \right), \quad (6)$$

for $s = 0$ one obtains

$$\tau_R = \frac{\hat{I}(0)}{I(0)} = \frac{\int_0^\infty I(t)dt}{I(0)}, \quad (7)$$

where $I(0) = q\mu_0 FSN_0$, which is estimated from the extrapolated value of $I(t)$ to $t = 0$.

$\hat{I}(0)$ can be carried out by a simple numerical integration over N sampling points t_j :

$$\hat{I}(0) = \sum_{j=2}^{N-1} I(t_j) \Delta t_j, \quad (8)$$

where $\Delta t_j = \frac{t_{j+1} - t_{j-1}}{2}$.

Eq. (7) is a simple and adequate relationship that leads to the estimate of τ_R . Like one see τ_R depends only on experimental $I(t)$ data and initial photocurrent $I(0)$, i.e. generated excess density N_0 .

To investigate the potential accuracy of the τ_R determination procedure, it is necessary to simulate accurately the transient photocurrent for the following two representative localized state distributions in amorphous and polymeric semiconductors: (i) an exponential distribution

$g(E) = g(E_c) \exp\left(\frac{-E}{kT_c}\right)$ and (ii) an exponential distribution on which a Gaussian distribution

has been superimposed $g(E) = g(E_c) \exp\left(\frac{E}{kT_c}\right) + g_g \exp\left[-\left(\frac{E - E_g}{E_w}\right)^2\right]$, where $g(E_c)$ is the

density of localized states at the mobility edge, T_c the characteristic temperature and g_g , E_g and E_w the peak value, the energy position from the mobility edge and the energy width of the Gaussian distribution, respectively.

In the present work $n(t)$ was carried out by means of an analytical expression for the inverse Laplace transform of $\hat{n}(s)$, obtained from Eq. (3), using Heaviside's expansion theorem [11].

$$n(t) = L^{-1} \{ \hat{n}(s) \} = \sum_{i=1}^{n+1} \frac{P(\alpha_i)}{Q'(\alpha_i)} e^{\alpha_i t}, \quad (9)$$

In Eq. (9) the α_i are the $n+1$ zeroes of the denominator $Q(s)$ and $Q'(\alpha_i)$ is the first derivative of $Q(s)$ taken at points α_i . $\hat{n}(s)$ is of the form of $\frac{P(s)}{Q(s)}$.

Fig. 1b shows the TPC, $n(t)$, computed at 300K for the proposed exponential DOS model of Fig. 1a with various values of τ_R . The inflection points in the transients are due to the monomolecular recombination of the photogenerated free carriers. The characteristic recombination time determined from the inflection point, τ_c , is much larger than τ_R , this results from frequent trapping and detrapping of free carriers into the exponential localized-state distributions. The value of τ_c increases with τ_R . The τ_R used in the simulation of $n(t)$, are, respectively, 10^{-8} , 10^{-7} and 10^{-6} s. The corresponding calculated τ_R using Eq. (7) are, respectively, 9.0165×10^{-9} , 8.8164×10^{-8} and 8.7647×10^{-7} s. It can be seen that the introduced τ_R were recovered with high accuracy.

Fig. 2b shows the TPC, $n(t)$, computed at 350K for the proposed exponential plus Gaussian DOS model of Fig. 2a with $\tau_R = 10^{-8}$ s. As in the case of computed TPC, $n(t)$, for an exponential DOS model, one can see well the inflection point in the transient due to recombination. The corresponding calculated τ_R using Eq. (7) is 8.7256×10^{-9} s. It can be seen here also that the proposed method recovers very precisely the introduced τ_R .

Figs. 3a, 3b, 3c and 4 show the TPC, $n(t)$ (symbol), of Figs. 1b ($\tau_R = 10^{-6}, 10^{-7}, 10^{-8}$ s) and 2b respectively, and $\tau_R(s^{-1})$ (solid line) for corresponding $\hat{n}(s)$ (Eq. (4)). One can see that $\tau_R(s^{-1})$ remains constant till the characteristic recombination time τ_c and equal to introduced value of τ_R in the simulation of $n(t)$. Normally τ_R is independent of s , but most important is that $\tau_R(s^{-1})$ is constant and equal to introduced τ_R for small values of s and for

$s = 0$ also of course (Eq. (7)). An important observation, the curve $\tau_R(s^{-1})$ deviates from introduced τ_R exactly at the moment when recombination begins, the curves $n(t)$ and $\tau_R(s^{-1})$ intersect at this specific time. Which means that the proposed method is not valid when the recombination phenomenon is not observed, this happens for low temperatures of measurement of $n(t)$, at short times.

2.2. Density of localized states

Eq. (6) can also be written as follows:

$$\sum_i \frac{g(E_i)}{s + \gamma_i} = \frac{1}{s \sigma v \Delta E} \left(\frac{q \mu_0 F S N_0}{\hat{I}(s)} - s - \frac{1}{\tau_R} \right), \quad (10)$$

We note here τ_R is experimentally known (Eq. (7)).

Eq. (10) is an integro-differential equation for the DOS $g(E)$, termed a Fredholm integral equation of the first kind [12] which may arise from an ‘ill-conditioned problem’. The inversion of this equation to obtain the DOS requires care, it needs a special resolution method. The resolution method used here is an exact matrix solution for $g(E)$ based on Tikhonov regularization [13,14]. Written in a discrete form, Eq. (10) yields

$$\sum_{i=1}^M g(i) A(j, i) = b(j) \quad \text{for } j = 2, \dots, N-1, \quad (11)$$

where i and j are the energy and time indexes respectively, and $g(i) = g(E_i)$. The matrix elements $A(j, i)$ and the vector elements $b(j)$ are respectively,

$$A(j, i) = \frac{1}{s(j) + \gamma(i)},$$

$$b(j) = \frac{1}{s(j) \sigma v \Delta E} \left(\frac{q \mu_0 F S N_0}{\hat{I}(j)} - s(j) - \frac{1}{\tau_R} \right),$$

where $\gamma(i) = \nu \exp\left(\frac{-E_i}{kT}\right)$, $s(j) = \frac{1}{t_j}$ and $\hat{I}(j) = \hat{I}(s_j)$.

The DOS vector is then given by

$$g = A^{-1}b. \quad (12)$$

This expression constitutes the basis of the transient method that returns for $\hat{I}(j)$ (i.e. the TPC data) a DOS distribution $g(E)$ of localized states.

3. Experiment

3.1. Preparation of sample

[15] A P-doped sample of a-Si:H was prepared by RF glow discharge decomposition of SiH_4 with 3 vppm PH_3 . Coplanar Al electrodes of 0.5 mm gap and 5 mm width were deposited on top of the film of 1 μm thickness, and the voltage applied across the gap was 400 V (i.e. an electric field of 8000 Vcm^{-1}). The light source employed was a Laser Science VSL337 N_2 -pumped 5 ns pulse dye laser, 640 nm, producing pulsed carrier densities of up to 10^{20} cm^{-3} , varied by means of neutral density filters. Single shots or low frequency ($< 1 \text{ Hz}$) laser pulses were used to allow complete carrier relaxation. The TPC signal was amplified where necessary, and measured using a PC-controlled Tektronix TDS380 oscilloscope as a transient recorder. A temperature range from 120–400 K was covered using a cryostat operating at a chamber pressure of typically 10^{-4} Torr. The dark Fermi level was estimated from the dark conductivity measurement to be 0.5 eV below the conduction band mobility edge.

3.2. Results

In the following, the free carriers lifetime τ_R and the energy profile of the DOS $g(E)$ are determined from an experimental TPC data. TPC are then generated by numerical simulation, using these τ_R and DOS, and compared to the experimental ones.

Fig. 5 shows a set of five TPC decays for the P-doped a-Si:H sample at 150, 200, 250, 290 and 310 K measured with an excitation density of $N_0 = 10^{16} \text{ cm}^{-3}$.

To valid the proposed method for the experimental case, we determinate τ_R and exact DOS $g(E)$ from the experimental TPC data of Fig. 5. We then simulate TPC using these τ_R and DOS $g(E)$ in order to reproduce the experimental ones. Using Eq. (7) for measured TPC decays at 200, 250 and 310K of Fig. 5, we find respectively the following values of τ_R : 1.2933×10^{-6} s, 9.7965×10^{-7} s and 8.552×10^{-7} s. And by inserting these values of τ_R into Eq. (10) for each case (200, 250 and 310K), we determine the energy profile of the DOS $g(E)$ after calculating the corresponding $\hat{I}(s)$ (Eq. (5)).

Fig. 6 shows the portion DOS $g(E)$, calculated by the inversion of Eq. (10), in the P-doped sample of Fig. 5 for the temperatures 200, 250 and 310K. The solid line is the DOS model obtained by fitting of the experimental DOS to the DOS expression:

$$g(E) = g(E_c) \exp\left(\frac{E}{kT_{c1}}\right) \left[1 - \frac{1}{1 + \exp\left(\frac{E - E_d}{kT_{c2}}\right)} \right] + g_g \exp\left[-\left(\frac{E - E_g}{E_w}\right)^2\right]. \quad (13)$$

Equation (13) indicates that the DOS model presents two exponential distributions parts on which a Gaussian distribution has been superimposed. One exponential distribution above E_d with high characteristic temperature T_{c1} and the other below E_d with low characteristic temperature $T_c = \frac{T_{c1}T_{c2}}{T_{c1} + T_{c2}}$. Least squares fitting gives $g(E_c) = 3.2279 \times 10^{21} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_d = 0.12417 \text{ eV}$ below E_c , $T_{c1} = 1321.3 \text{ K}$ and $T_{c2} = 211.49 \text{ K}$, resulting in $T_c \approx 182.3 \text{ K}$. For the Gaussian distribution, $g_g = 3.1292 \times 10^{15} \text{ cm}^{-3} \text{ eV}^{-1}$, $E_g = 0.36888 \text{ eV}$ below E_c and $E_w = 29.685 \text{ meV}$. The result is a well-defined steep exponential conduction band tail of characteristic temperature T_c with a slight deviation towards E_c at the top to which is added a Gaussian distribution of energy width E_w and whose the peak, of value g_g , is positioned at E_g below the mobility edge E_c . The doping effect on the tail shape is remarkable, the DOS

with energies around the donor (P) band peak at a certain level E_d between 0.1eV and 0.2eV below E_c [16] is found to increase with doping. The DOS is then, a broad distribution above E_d , a narrow tail below it and a deeper Gaussian distribution, so that the DOS profile can be fitted with the DOS model of Eq. (13).

The parameters used to perform τ_R and the DOS $g(E)$ are the following:

$$\mu_0 = 10 \text{ cm}^2 \text{ s}^{-1} \text{ V}^{-1}, C_n = \sigma v = 2 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1} \text{ and } \nu = 10^{12} \text{ s}^{-1}.$$

Fig. 7 shows the computed TPC decays (solid lines) for the P-doped sample at temperatures 200, 250 and 310K using the DOS model of Fig. 6. The corresponding experimental TPC decays of Fig. 5 are also presented (symbols). It turns out that, the modelled TPC curves line up quite rigorously with the experimental ones.

4. Discussion

4.1. Temperature effect

Limitations of the method are essentially the effect of low temperatures of TPC measurement on τ_R when the recombination phenomenon is not observed at short times. Fig. 9 shows the TPC, $n(t)$, computed at 150K between initial time $t_i = \frac{1}{\nu}$ and various values of final time $t_f = \frac{1}{\nu} \exp\left(\frac{E_c - E}{kT}\right)$ for the proposed exponential plus Gaussian DOS model of Fig. 8 with $\tau_R = 10^{-8} \text{ s}$. The chosen t_f are, respectively, 1, 10^2 and 10^4 s . The corresponding calculated τ_R using Eq. (7) are, respectively, 2.9693×10^{-9} , 9.8854×10^{-8} and $7.3038 \times 10^{-7} \text{ s}$. It can be seen here that the proposed method does not recover the introduced τ_R . We can retrieve the 'exact' value of the pre-proposed τ_R for low temperatures, provided to take extremely long times, which is not feasible experimentally. In conclusion we can say that the method is influenced by the temperature, it is valid only when the phenomenon of

recombination is observable, that is to say in the case of high temperatures. Higher temperature is better, as it shifts the recombination feature to shorter times, into the accessible time-region. Therefore, at low temperatures there is a little evidence of the effect of recombination on TPC over the experimental timescale.

4.2. Truncated data

Fig. 10 shows the TPC, $n(t)$ (solid line), computed at 350 K between initial time $t_i = 1$ ps and final time $t_f = 1$ s for the proposed exponential plus Gaussian DOS model of Fig. 2a with $\tau_R = 10^{-8}$ s and the truncated $n(t)$ (symbol o) at time 10 ns after t_i , i.e. 10 ns is the new starting time t_i . For the full data $n(t)$, τ_R was already been calculated (section 2.1), it is worth 8.7256×10^{-9} s and for the truncated one, τ_R is equal to 8.5912×10^{-9} s, it was calculated in the same way as in the case of complete data (Eq. 7). Fig. 11 shows the same full TPC, $n(t)$ (solid line) as of Fig. 10 and the truncated $n(t)$ (symbol o) at time $100 \mu\text{s}$ before t_f , i.e. $100 \mu\text{s}$ is the new final time t_f . For the truncated one, τ_R is equal to 8.7217×10^{-9} s. As we see the estimation of τ_R is not influenced by the missing of short or long-time data. This implies that there must be some influence of recombination at short times, long before the 'final' recombination decay which we normally expect (and see) at long times.

4.3. Noisy data

Accuracy of measuring τ_R has been already investigated by application to simulated (perfect) data, but how good is it when used on real (imperfect) data, subject to noise? To do this, we simulate TPC, $n(t)$, for given DOS model and τ_R , and add Gaussian noise whose amplitude is a constant fraction of $n(t)$. Random noise is approximately Gaussian of similar amplitude over whole time range. Noise was introduced by multiplying each point of $n(t)$ by a random number from a Gaussian distribution whose mean value is 1. The standard deviation of the distribution was varied between 10% and 40%.

Fig. 12 shows the simulated (smoothed) TPC, $n(t)$ (solid line), of Fig. 2b and the noisy $n(t)$ (symbol o) when noise level is equal to 10%. Eq. (7) gives $\tau_R = 8.7256 \times 10^{-9}$ s for simulated $n(t)$ and 9.0484×10^{-9} s for noisy $n(t)$ when the used τ_R in the simulation of $n(t)$ is 10^{-8} s. It can be seen that the estimation of τ_R is not influenced by noise.

Fig. 13 shows the deviation $\frac{\text{smoothed data} - \text{noisy data}}{\text{smoothed data}}$ versus time in the case of 10% noise. As we see Gaussian noise is approximately uniform over entire time range.

Fig. 14 shows estimated τ_R versus noise level, this last varying between 10% and 40%. τ_R remains practically constant, and equal to 10^{-8} s, over whole noise level range. The method is 'noise tolerant', it is capable of returning optimal resolution for a given set of noisy data even at quite high input noise levels.

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5. Conclusion

We have proposed a new method for determining the free carriers lifetime τ_R and the density of localized states DOS in amorphous semiconductors from the transient photocurrent TPC data. This technique involves Laplace transform of TPC data, it is a simple analysis of the solution of the basic linearized multiple trapping equations for free and trapped electrons. It has been tested by applying it to simulated and experimental TPC data measured on a typical disordered semiconductor, the hydrogenated amorphous silicon (a-Si:H). An introduced τ_R in the computing of the TPC using an arbitrarily proposed DOS model is recovered with high accuracy. For the experimental case, the estimated τ_R and the determination of the exact DOS from the experimental TPC data allow to reconstruct accurately this last. Limitations of the technique are essentially the effect of low temperatures of TPC measurement on τ_R when the recombination phenomenon is not observed at short times. On the other hand, the determination of τ_R is not at all influenced by the missing of short or long-time data and the noisy data.

Acknowledgements

The authors thank Amorphous Semiconductors group at Dundee University for the measurement facilities. The Algerian Ministry of Higher Education and Research is acknowledged for his financial support.

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