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# Photocurrent and photoabsorption transients in amorphous solids in the presence of optical bias

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**Abstract.** An approximate treatment of the transient response of an illuminated ('optically biased') amorphous semiconductor to a short light pulse is presented. The problem is formulated in terms of Rose's multiple-trapping model and is studied in two specific cases, corresponding to strongly non-equilibrium and quasi-equilibrium trapped-carrier distributions. In both approximations, the formulae describing the photocurrent and photoabsorption transients in some characteristic time intervals are derived. The accuracy of the formulae is verified by numerical calculations, performed for the exponential distribution of traps. The above results are consistent with those obtained previously by other authors, particularly by Pandya and Schiff. The given formulae make it possible to determine the trap distribution in the energy gap as well as some trap parameters from experimental data without making any model assumptions.

#### 1. Introduction

Investigations of photoconductivity and related phenomena provide valuable information about the carrier transport and recombination mechanisms in amorphous semiconductors. In the last few years, many experimental techniques have been developed for this purpose. Among other things, the influence of auxiliary illumination ('optical bias') on the photocurrent (PC) and photoabsorption (PA) transients has been extensively studied, mainly for a-Si:H [1–6]. It has been established that the decay rate of the PC and PA increases with increasing bias intensity.

The photoconductive properties of disordered semiconductors are commonly interpreted in terms of a multiple-trapping (MT) model. Usually, the optical bias effects are associated with filling of deeper traps, which reduces the trapping rate of free carriers and (for bimolecular recombination) enhances their recombination rate [7-10]. An alternative interpretation of these effects is given in [6, 11]. Despite these controversies, one can state that the measurements of the transient PC and PA in the presence of additional illumination yield valuable information on the energy distribution of traps and enable us to test the validity of the MT model.

The most comprehensive theoretical analysis of optical bias effects was carried out by Pandya and Schiff (PS) [9]. They obtained general solutions of the linearized MT equations, which correspond to a small photoexcitation pulse, by means of Fourier transformation. Next, they calculated the PC and PA transients for an exponential distribution of traps both analytically and numerically. It was concluded that the form of the transient responses depends mainly on the degree of carrier equilibration at the onset of recombination, as well as

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10381

# 10382 P Grygiel and W Tomaszewicz

on the recombination mechanism (monomolecular or bimolecular). A simple interpretation of the results obtained in terms of progressive carrier thermalization was also given.

In this paper a quantitative treatment of PC and PA transients with optical bias, using the carrier thermalization concept, is presented. The MT equations are solved approximately in two specific cases, corresponding either to strongly non-equilibrium [12, 13] or to quasi-equilibrium trapped-carrier distributions. The latter approach is similar to that introduced in [14, 15] and will be presented in detail elsewhere [16]. The resulting formulae for the PC and PA are relatively simple, and make it possible to determine the trap distribution as well as some trap parameters from experimental data. For the case of exponential distribution of traps, the formulae reproduce all of the results obtained by PS with satisfactory accuracy, as established by numerical calculations. The paper is mainly concerned with the case of bimolecular carrier recombination. The MT equations are then identical to those formulated within photoconductivity theory by Rose [17]. The case of monomolecular recombination is considered briefly in appendix C.

#### 2. Formulation of the problem

In the photoconductivity model considered, a wide distribution of trapping levels for majority carriers (e.g. electrons) and a single type of recombination centre are assumed to exist in the energy gap. The photogenerated free electrons may be temporarily captured by the traps or may recombine with the holes captured by recombination centres. The lifetime of the free holes is neglected. The resulting PC and PA are therefore proportional to the free-, and trapped-electron concentrations, respectively. According to the charge neutrality condition, the total concentration of electrons is equal to the concentration of holes in recombination centres.

The above model of photoconductivity corresponds to the following set of equations:

$$\frac{d}{dt}[n(t) + n_t(t)] = f(t) - b_r n(t)[n(t) + n_t(t)]$$
(1)

$$\frac{\mathrm{d}n'_t(t,E)}{\mathrm{d}t} = b_t [N_t(E) - n'_t(t,E)]n(t) - \frac{n'_t(t,E)}{\tau_r(E)}$$
(2)

$$n_t(t) = \int_{E_t^0}^{E_t} n'_t(t, E) \, \mathrm{d}E \tag{3}$$

where the time and energy variables are denoted by t and E, respectively, n(t) and  $n_t(t)$  are the free- and trapped-carrier densities,  $n'_t(t, E)$  is the density of trapped carriers per unit of energy and f(t) is the carrier generation rate. The meaning of the remaining notation is as follows:  $b_t$ ,  $b_r$ : carrier capture and recombination coefficients, respectively;  $N_t(E)$ : the trap density per energy unit;  $\tau_r(E) = v^{-1} \exp(E/kT)$ : the mean lifetime of trapped carriers (v is the frequency factor, k is the Boltzmann constant, and T the absolute temperature); and  $E_t^0$  and  $E_t$ : lower and upper limits of the trap distribution (the energy is measured from the edge of the conduction band).

It is convenient here to split each of the carrier densities and the carrier generation rate into two terms, relating to the steady-state bias illumination and to the additional light pulse. In the following, these terms will be indicated by the superscript '0' and the symbol ' $\Delta$ ', respectively. Therefore, we will write  $n(t) = n^0 + \Delta n(t)$ ,  $n_t(t) = n_t^0 + \Delta n_t(t)$ , etc. Throughout the paper, we shall consider the case of relatively weak additional excitation, subject to the conditions  $|\Delta n(t)| \ll n^0$  and  $\Delta n_t(t) \ll n_t^0$ . We shall assume also that

 $n^0 \ll n_t^0$ . Then, from equations (1)–(3) one gets

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[ \Delta n(t) + \Delta n_t(t) \right] = -b_r \left[ n_t^0 \,\Delta n(t) + n^0 \,\Delta n_t(t) \right] \tag{4}$$

$$\frac{\mathrm{d}\,\Delta n_t'(t,E)}{\mathrm{d}t} = b_t [N_t(E) - n_t'^0(E)] \Delta n(t) - \left[b_t n^0 + \tau_r^{-1}(E)\right] \Delta n_t'(t,E)$$
(5)

$$\Delta n_t(t) = \int_{E_t^0}^{E_t} \Delta n'_t(t, E) \, \mathrm{d}E.$$
(6)

Since the additional light pulse, generating carriers from the time moment t = 0, is assumed to be very short, the generation term  $\Delta f(t)$  on the RHS of equation (4) was omitted. The initial conditions for the above equations may then be formulated as follows:  $\Delta n(0) > 0$  and  $\Delta n'_t(0, E) = 0$ .

One should notice that within the approximation considered, the decay of the carrier density  $\Delta n(t) + \Delta n_t(t)$  is related to two different effects, described by the terms on the RHS of (4). The first process is the 'monomolecular' recombination of free carriers, having density  $\Delta n(t)$ , at a rate proportional to  $b_r n_t^0$ . The second process consists in the recombination of trapped carriers of density  $\Delta n_t(t)$ , the rate of recombination being proportional to  $b_r n^0$ . As will be shown later, the second effect is significant solely in the long-time limit.

From equations (1)–(3) one also obtains the equations relating the steady-state carrier densities to the generation rate  $f^0$ . It is sufficient here to characterize the optical bias intensity using the value of the free-carrier density  $n^0$ . The trapped-carrier density may be calculated from

$$n_t^{\prime 0}(E) = \frac{b_t n^0 N_t(E) \tau_r(E)}{1 + b_t n^0 \tau_r(E)}.$$
(7)

Equation (5) may be integrated with respect to the time and energy variables, which, together with equation (6) and the initial conditions, leads to

$$\Delta n_t(t) = \int_0^t \Phi(t - t') \,\Delta n(t') \,\mathrm{d}t' \tag{8}$$

where

$$\Phi(t) = b_t \int_{E_t^0}^{E_t} \frac{N_t(E)}{1 + b_t n^0 \tau_r(E)} \exp\left\{-\left[b_t n^0 + \tau_r^{-1}(E)\right]t\right\} dE.$$
(9)

The function  $\Phi(t)$  determines the probability that a carrier, being free at the initial moment (t = 0), is trapped within the time unit and stays in a trap up to the time t.

In the next section the approximate solutions of equations (4) and (8) are derived. The accuracy of the approximate solutions is estimated by comparison with the exact numerical results, obtained for a special case of exponential trap distribution,

$$N_t(E) = \frac{N_{tot}}{kT_c} \exp\left(-\frac{E}{kT_c}\right) \qquad E_t^0 = 0 \qquad E_t = \infty$$
(10)

where  $N_{tot}$  is the total trap density and  $T_c$  the characteristic temperature of the trap distribution.

# 3. Analytical solutions

#### 3.1. Carrier thermalization

The progressive carrier thermalization in the presence of optical bias is characterized by the positions of the demarcation level  $E_0(t)$  and the quasi-Fermi level  $E_{f0}$  in the energy gap. These energies are defined implicitly by  $\tau_r(E_0) = 1.8t$  (the correction factor 1.8 was introduced in [13]) and  $\tau_r(E_{f0}) = 1/b_t n^0$ , which yields

$$E_0(t) = kT\ln(1.8\nu t) \tag{11}$$

$$E_{f0} = kT \ln(\nu/b_t n^0).$$
(12)

The energy  $E_0(t)$  is the maximum trap depth for which the trapped carriers are in thermal equilibrium with the carriers in the conduction band.

The onset of the carrier thermalization is given by  $t_D = \tau_r(E_t^0)$ , while the time required for establishing the complete carrier equilibrium equals  $t_T = 1/b_t n^0$ . In what follows, we shall assume that  $t_T \gg t_D$ , which corresponds to the case of a wide trap distribution and relatively low bias intensity. As was shown by PS, another important parameter is the time of the onset of carrier recombination  $t_R$ , which will be defined later. If  $t_T \ll t_R$ , the carrier thermalization is complete before the recombination starts. Otherwise, the thermal equilibrium of carriers cannot be established. These cases are termed by PS as 'A-trapping' and 'B-trapping', respectively.

The MT equations can be simplified in two limiting cases. The non-equilibrium approach is based on the assumption that the majority of carriers occupy traps of depth  $E > E_0(t)$ , whereas the quasi-equilibrium approach corresponds to the opposite situation. Their adequacy depends on the stage of carrier thermalization and, for the dispersive transport regime ( $t_D \ll t \ll t_T$ ), on the form of the trap distribution. A rough criterion may be formulated in terms of an energy-dependent dispersion parameter [16]:

$$\alpha(E) = -kT \frac{\mathrm{d}\ln N_t(E)}{\mathrm{d}E} \tag{13}$$

characterizing the decay rate of the trap density. The non-equilibrium approximation gives good results when  $\alpha(E) < 0.5$ , and the quasi-equilibrium approximation gives good results when  $\alpha(E) > 0.5$  ( $E_t^0 \leq E \leq E_t$ ). In the following subsections, we shall present the formulae for the PC and PA obtained in the frameworks of the two approaches. Some additional approximations, under which the above-mentioned formulae are derived, are discussed in appendices A and B.

#### 3.2. The non-equilibrium case

In the case considered, equation (8), which describes the kinetics of the carrier trapping/detrapping, may be approximated [12, 13] by

$$\Delta n_t(t) \approx \Phi(t) \int_0^t \Delta n(t') \, \mathrm{d}t' \tag{14}$$

which leads to the equation

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[ \frac{\Delta n_t(t)}{\Phi(t)} \right] \approx \Delta n(t). \tag{15}$$

A plot of the function  $\Phi(t)$  for the exponential trap distribution (10) is shown in figure 1. Below, we shall give the solutions of equations (4) and (15) for various characteristic time intervals.



**Figure 1.** The function  $\Phi(t)$  calculated from the exact formula (9) (points) and the approximate formulae (16), (24) and (34) (dashed lines) for the exponential trap distribution.  $n_0 = 10^{-10}$ ,  $b_r = 20$ . The quantities on the plot and in the caption are dimensionless (cf. section 4.1).

3.2.1. Interval I.  $t \ll t_D$ . In this time region, carrier emission from the traps is negligible, and thus only the processes of carrier trapping and recombination are significant. The function  $\Phi(t)$ , defined by equation (9), does not depend on time (cf. figure 1), and can be approximated by

$$\Phi(t) \approx b_t \int_{E_t^0}^{E_{f0}} N_t(E) \, \mathrm{d}E = 1/\tau_t^0.$$
(16)

Here,  $\tau_t^0$  is the mean carrier trapping time in the presence of the bias illumination.  $\tau_t^0$  is an increasing function of the bias intensity, due to the corresponding shift of the quasi-Fermi level  $E_{f0}$ .

Inserting the above expression into equation (15), one obtains

$$\frac{\mathrm{d}\,\Delta n_t(t)}{\mathrm{d}t} \approx \frac{\Delta n(t)}{\tau_t^0}.\tag{17}$$

Equations (4) and (17) can be integrated exactly. However, for  $t \ll t_T$ ,  $n^0 \Delta n_t(t) \ll n_t^0 |\Delta n(t)|$  (see appendix A). Then, the second term on the RHS of (4) may be omitted:

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[ \Delta n(t) + \Delta n_t(t) \right] \approx -b_r n_t^0 \,\Delta n(t). \tag{18}$$

The solution of equations (17) and (18) is

$$\Delta n(t) = \Delta n(0) \exp(-t/\tau_e)$$
<sup>(19)</sup>

$$\Delta n_t(t) = \Delta n(0) \frac{\tau_e}{\tau_t^0} \left[ 1 - \exp(-t/\tau_e) \right].$$
<sup>(20)</sup>

The time  $\tau_e$  determined from

$$1/\tau_e = 1/\tau_t^0 + b_r n_t^0 \tag{21}$$

is the mean lifetime of a free carrier which depends on the trapping and recombination rates of the carriers. According to the above equations, the PC decays exponentially with time, whereas the PA increases monotonically. 3.2.2. Interval II.  $t_D \ll t \ll t_T$ . The time interval considered corresponds to the dispersive transport regime. Equation (9) thus simplifies to the form

$$\Phi(t) \approx b_t \int_{E_0(t)}^{E_{f0}} N_t(E) \, \mathrm{d}E \tag{22}$$

which is similar to the form without optical bias [12, 13]. The only difference is that the upper integration limit now equals  $E_{f0}$ , because of the saturation of deeper traps.

For the exponential trap distribution and relatively low bias intensity, when  $E_{f0}$  may be replaced by infinity, this formula yields (cf. figure 1)

$$\Phi(t) \approx \frac{\alpha}{\tau_t} (1.8\nu t)^{-\alpha} \qquad \alpha < 1.$$
(23)

Here,  $\alpha = T/T_c$  is the dispersion parameter, and  $\tau_t = 1/b_t N_{tot}$  is the mean carrier trapping time with no optical bias.

The set of equations (4) and (15) can easily be solved subject to the conditions  $|d \Delta n(t)/dt| \ll |d \Delta n_t(t)/dt|$  and  $n^0 \Delta n_t(t) \ll n_t^0 |\Delta n(t)|$ . As estimated in appendix A, these inequalities hold for  $t \gg t_D$  and  $t \ll t_T$ , respectively. In this case, equation (4) reduces to

$$\frac{\mathrm{d}\,\Delta n_t(t)}{\mathrm{d}t} \approx -b_r n_t^0 \,\Delta n(t). \tag{24}$$

The solution of equations (15) and (24) is as follows:

$$\Delta n(t) = \Delta n(0) \frac{\mathrm{d}}{\mathrm{d}t} \left[ \frac{1}{\Phi(t) + b_r n_t^0} \right]$$
(25)

$$\Delta n_t(t) = \Delta n(0) \frac{\Phi(t)}{\Phi(t) + b_r n_t^0}.$$
(26)

The above formulae simplify in two time intervals. One can define the onset  $t_R$  of carrier recombination by the implicit equation

$$\Phi(t_R) = b_r n_t^0. \tag{27}$$

Since the function  $\Phi(t)$  decreases monotonically with time, for  $t \ll t_R$  one gets

$$\Delta n(t) \approx \Delta n(0) \frac{\mathrm{d}}{\mathrm{d}t} \left[ \frac{1}{\Phi(t)} \right]$$
 (28)

$$\Delta n_t(t) \approx \Delta n(0). \tag{29}$$

On the other hand, if  $t \gg t_R$ , one obtains

$$\Delta n(t) \approx \frac{\Delta n(0)}{(b_r n_t^0)^2} \left[ -\frac{\mathrm{d}\Phi(t)}{\mathrm{d}t} \right]$$
(30)

$$\Delta n_t(t) \approx \frac{\Delta n(0)}{b_r n_t^0} \,\Phi(t). \tag{31}$$

The formulae given are similar to those determining the current intensity in a time-of-flight experiment [12, 13]. Equations (28) and (29) describe progressive carrier thermalization with negligible recombination. In this case the optical bias influences the PC transient only via the position of the quasi-Fermi level in the forbidden gap (cf. equation (22)). Equations (30) and (31) correspond to carrier emission from the traps and the subsequent carrier recombination, without significant retrapping.

In the particular case of exponential trap distribution, when  $\Phi(t)$  is given by equation (23), from equation (27) one obtains

$$t_R \approx \frac{1}{\nu (\tau_t b_r n_t^0)^{1/\alpha}}.$$
(32)

Here, some numerical coefficients of the order of unity were omitted. According to equations (28), (30) and (31), the corresponding PC and PA transients are power functions of time:  $\Delta n(t) \propto t^{-(1-\alpha)}$  for  $t \ll t_R$ ;  $\Delta n(t) \propto t^{-(1+\alpha)}$  and  $\Delta n_t(t) \propto t^{-\alpha}$  for  $t \gg t_R$ .

3.2.3. Interval III.  $t \gg t_T$ . In this time region the assumption of non-equilibrium carrier distribution is justified only if  $t_R \ll t_T$  (the B-trapping case of PS). As results from equation (9), the ultimate decay of  $\Phi(t)$  is given by

$$\Phi(t) \approx \frac{\exp(-b_t n^0 t)}{n^0} \int_{E_0(t)}^{E_t} \frac{N_t(E)}{\tau_r(E)} \, \mathrm{d}E.$$
(33)

Since the above integral varies slowly with time, the final decay of  $\Phi(t)$  is approximately exponential with a time constant  $1/b_t n^0 = \tau_r(E_{f0})$ . Thus, for  $t > t_T$  the carriers are emitted mainly from the traps in the vicinity of the quasi-Fermi level. For the exponential distribution of traps (10), from equation (33) one gets

$$\Phi(t) \approx \frac{\alpha \nu N_{tot}}{(1+\alpha)n^0} (1.8\nu t)^{-(1+\alpha)} \exp(-b_t n^0 t) \qquad \alpha < 1$$
(34)

(cf. figure 1).

When solving equations (4) and (15), the assumption that  $|d \Delta n(t)/dt| \ll |d \Delta n_t(t)/dt|$  may be used again, but both terms on the RHS of (4) must be retained. The resulting solution has then an involved form (see appendix A). According to this, the final decay of the PC and PA is given by

$$\Delta n(t) \approx \frac{c \,\Delta n(0)}{(b_r n_t^0)^2} \left[ -\frac{\mathrm{d}\Phi(t)}{\mathrm{d}t} - b_r n^0 \Phi(t) \right]$$
(35)

$$\Delta n_t(t) \approx \frac{c \,\Delta n(0)}{b_r n_t^0} \Phi(t) \tag{36}$$

where  $e^{-1} < c < 1$ . For exponential trap distribution, the long-time PC asymptotics is described by  $\Delta n(t)$ ,  $\Delta n_t(t) \propto t^{-(1+\alpha)} \exp(-b_t n^0 t)$ .

It is seen from equations (30) and (35) that the sign of the PC transient might change from '+' to '-' before the ultimate PC behaviour is established. One can give an approximate criterion for the existence of this effect. Inserting the asymptotic formula  $\Phi(t) \propto \exp(-b_t n^0 t)$  into (35), one obtains the equation

$$\Delta n(t) \propto (b_t - b_r) \exp(-b_t n^0 t) \tag{37}$$

with a positive proportionality constant. This implies that the PC sign reversal occurs if  $b_r > b_t$ .

#### 3.3. The quasi-equilibrium case

In the case of a quasi-equilibrium distribution of trapped carriers, equation (8) may be approximated by [16]

$$\Delta n_t(t) \approx \left[ \int_0^t \Phi(t') \, \mathrm{d}t' \right] \Delta n(t). \tag{38}$$



**Figure 2.** The function  $\Theta(t)$  calculated from the exact formula (41) (points) and the approximate formulae (47) and (48) (solid and dashed line, respectively).  $n_0 = 10^{-10}$ ,  $b_r = 20$ . The quantities on the plot and in the caption are dimensionless (cf. section 4.1).

Introducing the function  $\Theta(t)$  which obeys the relation

$$\Theta^{-1}(t) = 1 + \int_0^t \Phi(t') dt$$
(39)

equation (38) may be rewritten as

$$\Delta n_t(t) \approx \left[\Theta^{-1}(t) - 1\right] \Delta n(t). \tag{40}$$

It is seen that the function  $\Theta(t)$  is equal to the ratio of the free-carrier density to the total carrier density,  $\Theta(t) = \Delta n(t) / [\Delta n(t) + \Delta n_t(t)]$ . Inserting the function  $\Phi(t)$  defined by (9) into equation (39), one obtains

$$\Theta^{-1}(t) = 1 + b_t \int_{E_t^0}^{E_t} \frac{N_t(E)\tau_r(E)}{\left[1 + b_t n^0 \tau_r(E)\right]^2} \left\{ 1 - \exp\left[-(b_t n^0 + \tau_r^{-1}(E))t\right] \right\} dE.$$
(41)

A plot of the function  $\Theta(t)$  for the exponential distribution of traps given by (10) is presented in figure 2.

Below, the solutions of equations (4) and (40) are given for the time intervals II and III, defined in the preceding section (in interval I the quasi-equilibrium approach cannot be valid). It is assumed that  $\Theta(t) \ll 1$ , which is usually the case for  $t \gg t_D$ . Only the expressions determining the free-carrier density are presented. Those for the trapped-carrier density may be obtained from the relationship

$$\Delta n_t(t) \approx \Delta n(t) / \Theta(t) \tag{42}$$

which results from equation (40).

3.3.1. Interval II.  $t_D \ll t \ll t_T$ . In the time interval considered, equation (41) for  $\Theta(t)$  may be approximated by

$$\Theta^{-1}(t) \approx b_t \int_{E_t^0}^{E_0(t)} N_t(E) \tau_r(E) \,\mathrm{d}E.$$
 (43)

The set of equations (4) and (40) can be integrated without further approximations. However, for  $t \ll t_T$  the second term on the RHS of equation (4) may be dropped again (cf. appendix B). The solution of the resulting set of equations (18) and (40) is

$$\Delta n(t) = \Delta n(0) \Theta(t) \exp\left[-b_r n_t^0 \int_0^t \Theta(t') dt'\right].$$
(44)

The exponential factor in this formula describes the influence of carrier recombination. Thus, the carrier recombination onset  $t_R$  is now determined by

$$\int_{0}^{t_{R}} \Theta(t') \, \mathrm{d}t' = 1/b_{r} n_{t}^{0}. \tag{45}$$

Equation (44) is valid only for  $t < t_R$ , since for longer times the quasi-equilibrium approach is inadequate. Instead, equation (30) from the previous subsection must be used. For the time  $t \ll t_R$ , when the carrier recombination is negligible, from equation (44) one gets

$$\Delta n(t) \approx \Delta n(0) \,\Theta(t). \tag{46}$$

The PC decay is then due solely to carrier thermalization. One can notice that in the approximation considered the function  $\Theta(t)$  and the PC for  $t \ll t_R$  do not depend on the bias intensity. An analogous formula was obtained for the initial current intensity in the time-of-flight regime [16].

For the exponential trap distribution (10), the function  $\Theta(t)$  is approximately given, according to equation (43), by

$$\Theta(t) \approx \frac{(1-\alpha)\tau_t \nu}{\alpha} (1.8\nu t)^{-(1-\alpha)} \qquad \alpha < 1$$
(47)

(cf. figure 2). From the above equations one then obtains essentially the same results as in the previous subsection. Equation (45) again yields formula (32) for the recombination onset  $t_R$  (except for some multiplicative coefficients). Also, equation (46) gives the same time dependence of the PC,  $\Delta n(t) \propto t^{-(1-\alpha)}$  for  $t \ll t_R$ , as equation (28).

3.3.2. Interval III.  $t \gg t_T$ . In the time region considered, the approximate thermal equilibrium between the free and trapped carriers may exist only if  $t_R \gg t_T$  (the A-trapping case of PS). According to equation (41), the function  $\Theta(t)$  does not depend on time (cf. figure 2). Denoting the limiting value of this function by  $\Theta^0$ , one gets

$$\Theta^{0^{-1}} \approx b_t \int_{E_t^0}^{E_{f0}} N_t(E) \tau_r(E) \, \mathrm{d}E.$$
(48)

From the solution of equations (4) and (40) given in appendix B it follows that the PC decays exponentially for large times:

$$\Delta n(t) = \Delta n(0) \Theta^0 \exp(-2b_r n^0 t). \tag{49}$$

The carrier recombination onset is therefore given by  $t_R = 1/2b_r n^0$ . For  $t_T \ll t \ll t_R$  the PC has a constant value,  $\Delta n(t) \approx \Delta n(0) \Theta^0$ .

All of the analytical results obtained for the interval II for the exponential trap distribution as well as for the interval III may be compared with the corresponding results of PS (region I was not considered earlier). In general, there is complete qualitative agreement between the given formulae. The small discrepancies concern only the numerical values of some coefficients and the final asymptotics of the PC and PA decay in the B-trapping case, which according to PS should be purely exponential.

# 4. Numerical results

# 4.1. The numerical method

In order to verify the accuracy of our approximate formulae derived in the preceding section, we carried out numerical calculations of the PC and PA for the exponential trap distribution given by (10).

# 10390 P Grygiel and W Tomaszewicz

To reduce the number of parameters, the MT equations (1)–(3) were rewritten in dimensionless form. The following quantities were selected as the basic units:  $N_{tot}$ : the unit of the trap and carrier densities;  $1/C_t N_{tot}$ : the time unit; kT: the energy unit. The continuous trap distribution (10) was approximated by the set of L = 150 equidistant discrete levels, separated by the energy  $\Delta E = kT$ . The resulting set of stiff differential equations has been solved using Gear's algorithm [18]. A sparse form of the Jacobian of MT equations allowed us to optimize the original Gear procedure. Special subroutines were developed for the storage of non-zero Jacobian elements and for the solution of the corresponding linear equations. These modifications made the computing process less memory consuming and much more efficient.

The intensity of the bias illumination was characterized by the value of the stationary (dimensionless) density  $n^0/N_{tot}$  of free carriers. Although the numerical results given below refer to rather low values of  $n^0/N_{tot}$ , it was established that the PC and PA transients do not change appreciably up to  $n^0/N_{tot} \approx 10^{-3}$ . The steady-state densities of the carriers, captured at each energy level, were computed using the discretized formula (7).

#### 4.2. Comparison of the analytical and numerical results

In the figures below, the results of the analytical and numerical calculations of the PC and PA transients are compared. The analytical and numerical curves are indicated by points and solid lines, respectively. All of the quantities in the figures and in their captions are dimensionless. For simplicity, we have not introduced new symbols for these quantities.



**Figure 3.** PC transients evaluated analytically (solid lines) and numerically (points) for exponential trap distribution at different bias illuminations. The limits  $t_D$ ,  $t_R$  and  $t_T$  of the corresponding time intervals are marked by the arrows. The calculations were carried out for a high value of the recombination coefficient,  $b_r = 20$ , and for a relatively high value of the dispersion parameter  $\alpha = T/T_c$ .

Each figure presents the results corresponding to three different intensities of bias illumination, i.e. to different positions of the quasi-Fermi level below the mobility edge. The individual figures differ in, among other factors, the value of the dispersion parameter



**Figure 4.** PC transients for a case similar to that represented in figure 3. The dispersion parameter  $\alpha$  has an intermediate value; the remaining parameters are identical to those of figure 3.



**Figure 5.** PC transients for a case similar to that represented in figure 3. The dispersion parameter  $\alpha$  has a relatively small value; the remaining parameters are identical to those of figure 3.

 $\alpha = T/T_c$ , which characterizes the decay rate of the exponential trap distribution. In all of the figures the PC or PA in the initial time interval,  $t < t_D$ , were computed using equations (19) or (20), respectively.

Figures 3–5 show the PC transients obtained for a relatively high value of the recombination coefficient (case B of PS,  $t_R \ll t_T$ ). For  $t > t_D$  the PC were calculated



Figure 6. PA transients corresponding to the same case as for the PC in figure 5.



Figure 7. PC transients evaluated analytically and numerically for exponential trap distribution at different bias illuminations. The calculations were carried out for a low value of the recombination coefficient,  $b_r = 10^{-8}$ , and for a relatively small value of the dispersion parameter  $\alpha$ .

from equations (A10) and (A1). The exception are the PC shown in figure 3, where for the time interval  $t_D < t < t_R$  equation (B1) was used. This was done because for  $\alpha > 0.5$  the quasi-equilibrium formulae should give better accuracy compared to the non-equilibrium ones (cf. section 3.1). All of the features of the PC discussed in the preceding section can be recognized on the plots. In the initial time interval,  $t < t_D$ , the PC decays exponentially. For larger times, the PC transient changes gradually from  $t^{-(1-\alpha)}$  to  $t^{-(1+\alpha)}$  in the vicinity of the time  $t_R$ , which marks the onset of recombination. Close to the thermalization time  $t_T$  the PC becomes negative and its final decay is nearly exponential. Figure 6 presents the PA transients for identical values of the parameters as for the PC represented in figure 5. For  $t > t_D$  the transients were plotted using equations (A10) and (A2). After the initial increase, the PA behaves as  $t^{-\alpha}$ . The final decay of the PA is exponential, and is characterized by the same rate constant as that of the PC.

Figures 7-9 present the PC transients calculated for a low value of the recombination



**Figure 8.** PC transients for a case similar to that represented in figure 7. The dispersion parameter  $\alpha$  has an intermediate value; the other parameters are identical to those of figure 7.



**Figure 9.** PC transients for a case similar to that represented in figure 7. The dispersion parameter  $\alpha$  has a relatively high value; the other parameters are identical to those of figure 7.

coefficient (case A of PS,  $t_R \gg t_T$ ), and the same bias intensities as for figures 3–6. In the time region  $t > t_D$  the PC were computed using equation (B1), except the PC in figure 7, which was calculated for  $t_D < t < t_T$  from equations (A10) and (A1). The character of the PC differs significantly from those of the previous ones. For  $t > t_D$  the PC decays according to the expression  $t^{-(1-\alpha)}$  up to the time  $t_T$ . Then the PC transient levels off to a constant value, and this is followed by a final exponential decay near to  $t_R$ . Figure 10 illustrates the PA transients, computed from equations (B1) and (40) with parameters identical to those of figure 9. Over the time interval  $t_D \ll t \ll t_R$  the PA is essentially constant and for longer times it exhibits the same exponential decay as the PC.

All of the above figures illustrate the influence of the optical bias intensity on the PC and PA. In particular, the decrease of the values of  $t_R$  and  $t_T$  with increasing illumination level is clearly seen.

In general, the PC and PA time dependencies, calculated analytically and numerically,



Figure 10. PA transients corresponding to the same case as for the PC in figure 9.

remain in good agreement. The more significant discrepancies concern only the value of the recombination onset time  $t_R$  in the A-trapping case (cf. figures 9 and 10). The reason for these divergences is not clear to us. One can notice that the accuracy of the nonequilibrium formulae somewhat ameliorates with decreasing  $\alpha$ -parameter. The accuracy of the quasi-equilibrium formulae exhibits the opposite trend. Summarizing, the approximate expressions derived are found to describe all of the features of the PC and PA in the presence of bias illumination with a rather satisfactory accuracy.

#### 5. Conclusions

The analytical and numerical treatment of the PC and PA in the presence of optical bias, given in sections 3 and 4, enables us to formulate the following conclusions. The forms of the PC and PA transients for the initial time region (I) and, in a first approximation, for the final time interval (III), are independent of the trap distribution in the energy gap. In contrast, for the dispersive transport regime (II), the shape of the PC and PA curves depends on the energetic trap distribution as well as on the bias intensity. In this time region, the non-equilibrium and quasi-equilibrium approaches give consistent results, at least for exponential trap distribution. Combining these approaches, if necessary, one can describe all of the features of PC and PA transients with good accuracy. The results obtained are consistent with those obtained by other authors, particularly PS.

The methods of determination of the MT model parameters from the measured PC and PA were considered in earlier papers. Therefore, we shall confine ourselves here to emphasizing certain points. According to equations (19)–(21), the investigation of PC or PA transients in the initial interval makes it possible to calculate the mean lifetime  $\tau_e$  of free carriers. On the other hand, the analysis of the PC and/or PA in the dispersive transport region enables us in principle to determine the shape of the trap distribution in the energy gap. The corresponding formulae, given in section 3, seem to be suitable for this purpose because of their simplicity. Using these formulae, one can determine the form of the function  $\Phi(t)$  or  $\Theta(t)$ , which is directly related to the energetic trap profile, according to (22) or (43). A particularly simple formula results from equations (30) and (22), corresponding to the B-trapping case:

$$\Delta n(t) \propto N_t [E_0(t)]/t \qquad t_R \ll t \ll t_T.$$
(50)

The PC is here proportional to the trap density at the energy level  $E_0(t)$ , divided by the time *t*. An analogous formula was obtained in the case of PC decay from the steady state under strong recombination [19]. The above-mentioned analysis can be carried out for experimental data corresponding to different intensities of bias illumination. Therefore, the consistency of the PC and PA interpretation in terms of the MT model considered may be verified.

This paper is concerned mainly with the case of bimolecular carrier recombination. However, essentially the same results are valid for the monomolecular recombination case up to the thermalization time  $t_T$  (cf. appendix C). This implies that the above conclusions apply also to monomolecular carrier recombination.

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#### Appendix A. The non-equilibrium approach

The set of equations (4) and (15) does not seem to have an exact solution. Therefore, the approximations made in section 3.2 may be justified only in semi-consistent way. We shall restrict ourselves to discussing these approximations for the time interval II. Some of the solutions obtained apply also to the final time interval III.

#### A1. Interval II. $t_D \ll t \ll t_T$

From equation (15) it is apparent that the carrier densities can be expressed as

$$\Delta n(t) = \Delta n(0) \frac{\mathrm{d}u(t)}{\mathrm{d}t} \tag{A1}$$

$$\Delta n_t(t) = \Delta n(0) \,\Phi(t) u(t). \tag{A2}$$

The solution of the approximate equations (24) and (15) is then given by

$$u(t) = \frac{1}{\Phi(t) + b_r n_t^0}.$$
 (A3)

Let us investigate first the simplifying assumption  $|d \Delta n(t)/dt| \ll |d \Delta n_t(t)/dt|$ . For this purpose, equation (24) may be replaced by

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[ \Delta n(t) + \Delta n_t(t) \right] \approx -b_r n_t^0 \,\Delta n(t). \tag{A4}$$

The solution of equations (A4) and (15) is

$$u(t) = \int_0^t \exp\left\{-\int_{t'}^t \left[\Phi(t'') + b_r n_t^0\right] dt''\right\} dt'.$$
 (A5)

The above expression reduces to (A3) subject to the condition

$$\int_0^t \left[ \Phi(t') + b_r n_t^0 \right] \mathrm{d}t' \gg 1.$$
(A6)

10396 P Grygiel and W Tomaszewicz

This may be proved as follows:

$$u(t) = \int_{0}^{t} \frac{1}{\Phi(t') + b_{r}n_{t}^{0}} \frac{d}{dt'} \exp\left\{-\int_{t'}^{t} \left[\Phi(t'') + b_{r}n_{t}^{0}\right] dt''\right\} dt'$$

$$\approx \frac{1}{\Phi(t) + b_{r}n_{t}^{0}} \int_{0}^{t} \frac{d}{dt'} \exp\left\{-\int_{t'}^{t} \left[\Phi(t'') + b_{r}n_{t}^{0}\right] dt''\right\} dt'$$

$$\approx \frac{1}{\Phi(t) + b_{r}n_{t}^{0}}.$$
(A7)

Taking into account relationship (39), inequality (A6) may be rewritten as

$$\frac{1}{\Theta(t)} - 1 + b_r n_t^0 t \gg 1. \tag{A8}$$

Since usually  $\Theta(t) \ll 1$  for  $t \gg t_D$ , one can state that the inequality  $|d \Delta n(t)/dt| \ll |d \Delta n_t(t)/dt|$  holds for  $t \gg \min(t_D, 1/b_r n_t^0)$ .

Let us consider now the assumption  $n^0 \Delta n_t(t) \ll n_t^0 |\Delta n(t)|$ . To obtain the corresponding criterion, equation (24) may be replaced by

$$\frac{\mathrm{d}\,\Delta n_t(t)}{\mathrm{d}t} \approx -b_r \left[ n_t^0 \,\Delta n(t) + n^0 \,\Delta n_t(t) \right]. \tag{A9}$$

The solution of equations (A9) and (15) has the form

$$u(t) = \frac{1}{\Phi(t) + b_r n_t^0} \exp\left[-\int_0^t \frac{b_r n^0 \Phi(t')}{\Phi(t') + b_r n_t^0} dt'\right].$$
 (A10)

This expression yields (A3) provided that

$$\int_{0}^{t} \frac{b_{r} n^{0} \Phi(t')}{\Phi(t') + b_{r} n_{t}^{0}} \, \mathrm{d}t' \ll 1.$$
(A11)

Taking into account equation (39), the value of the above integral may be estimated as follows:

$$\int_{0}^{t} \frac{b_{r} n^{0} \Phi(t')}{\Phi(t') + b_{r} n_{t}^{0}} \, \mathrm{d}t' < \min\left\{\frac{n^{0}}{n_{t}^{0}} \left[\frac{1}{\Theta(t)} - 1\right], b_{r} n^{0} t\right\}.$$
(A12)

Making use of equation (7), one obtains the relationship

$$n_t^{\ 0} = \left(\frac{1}{\Theta^0} - 1\right) n^0 \tag{A13}$$

which is analogous to (40), with  $\Theta^0$  given by equation (48). In the following, it will be assumed that  $\Theta(t), \Theta^0 \ll 1$ . Then

$$n^0 \approx \Theta^0 n_t^0 \tag{A14}$$

$$\int_0^t \frac{b_r n^0 \Phi(t')}{\Phi(t') + b_r n_t^0} dt' < \min\left[\frac{\Theta^0}{\Theta(t)}, b_r n^0 t\right].$$
(A15)

In principle,  $\Theta(t) \gg \Theta^0$  for  $t \ll t_T$ . One can therefore conclude that the inequality  $n^0 \Delta n_t(t) \ll n_t^0 |\Delta n(t)|$  is valid for  $t \ll \max(t_T, 1/b_r n^0)$ .

# A2. Interval III. $t \gg t_T$

In this time interval, solution (A10) is also valid, subject to  $|d \Delta n(t)/dt| \ll |d \Delta n_t(t)/dt|$ . For sufficiently large times the integral in equation (A10) tends to a constant value:

$$\int_{0}^{\infty} \frac{b_r n^0 \Phi(t')}{\Phi(t') + b_r n_t^0} \, \mathrm{d}t' = c_0 < 1 \tag{A16}$$

(cf. equation (A15)). Then, from equations (A10) and (A16), together with (A1) and (A2), one obtains formulae (35) and (36) with  $c = \exp(-c_0)$ .

## Appendix B. The quasi-equilibrium approach

The exact solution of equations (4) and (40) has the following form:

$$\Delta n(t) = \Delta n(0) \Theta(t) \exp\left\{-b_r \left[n_t^0 \int_0^t \Theta(t') dt' + n^0 t\right]\right\}.$$
(B1)

We shall assume here, as before, that  $\Theta(t)$ ,  $\Theta^0 \ll 1$ .

# B1. Interval II. $t_D \ll t \ll t_T$

Making use of (A14), one can notice that the second term in the exponent is negligible if  $\Theta(t) \gg \Theta^0$ . From (B1) one then gets formula (44). This shows again that  $n^0 \Delta n_t(t) \ll n_t^0 |\Delta n(t)|$  for  $t \ll t_T$ .

## B2. Interval III. $t \gg t_T$

For  $t \gg t_T$  the integral in (B1) may be approximated by

$$\int_0^t \Theta(t') \, \mathrm{d}t' \approx \Theta^0 t. \tag{B2}$$

Then, from (B1) and (A14) one obtains formula (49).

#### Appendix C. Monomolecular recombination

The kinetics of monomolecular recombination (MR) of the carriers is described by

$$\frac{\mathrm{d}}{\mathrm{d}t} \left[\Delta n(t) + \Delta n_t(t)\right] = -\frac{\Delta n(t)}{\tau_R} \tag{C1}$$

where  $\tau_R$  is the mean recombination time. This equation has the same form as equation (4) for bimolecular recombination (BR), in which the product  $b_r n_t^0$  is replaced by  $1/\tau_R$  and the second term on the RHS, describing the recombination of trapped carriers, is omitted.

As indicated before, in the case of BR the latter term in equation (4) is negligible for time  $t \ll t_T$ . This implies that the formulae relating to MR may be obtained from those corresponding to BR for  $t \ll t_T$ , given in section 3, by replacing  $b_r n_t^0$  by  $1/\tau_R$ . The formulae obtained for the dispersive transport region apply also in the final time interval,  $t \gg t_T$ . In particular, for  $t_R \ll t_T$  (the B-trapping case of PS) the ultimate PC and PA decay are described by the equations

$$\Delta n(t) \approx \Delta n(0) \tau_R^2 \left[ -\frac{\mathrm{d}\Phi(t)}{\mathrm{d}t} \right]$$
(C2)

$$\Delta n_t(t) \approx \Delta n(0) \,\tau_R \Phi(t) \tag{C3}$$

which follow from (30) and (31). In this case the PC sign reversal does not occur. For the exponential trap distribution from (34), (C2) and (C3), one gets essentially the same expressions as for the BR:  $\Delta n(t)$ ,  $\Delta n_t(t) \propto t^{-(1+\alpha)} \exp(-b_t n^0 t)$ . For  $t_T \ll t_R$  (the Atrapping case of PS) the final decay of the PC is given by the equation

$$\Delta n(t) = \Delta n(0) \Theta^0 \exp(-b_r n^0 t) \tag{C4}$$

which results from (44), (A14) and (B2). This equation has almost the same form as equation (49), which is for BR.

In conclusion, the PC and PA transients for MR and BR should have identical forms in the time interval  $t \ll t_T$  and almost the same ultimate behaviours. The dependence of the PC and PA on the bias intensity would be, however, different. This is because the rate of MR of the free carriers, proportional to  $1/\tau_R$ , is independent on the optical bias, unlike the BR rate. The above-mentioned results concerning the MR are consistent with those from the paper by PS. The formulae obtained were also verified numerically in a way similar to that described in section 4.

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