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Physica E 22 (2004) 852–859

PHYSICA E

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A new approach to determine radiative recombination lifetime in quantum well solar cells

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Received 3 September 2003; accepted 22 September 2003

Abstract

In this paper, the authors present and extend on an existing model, which has been developed to determine the radiative recombination lifetimes in quantum well solar cells. Given the fact that recombination reduces cell performance, the main future use of this new model is to aid in optimisation of cell designs and increase cell efficiency. In this work the authors introduce a coefficient defined as the delta factor which is based on material parameters into the existing model. The introduction of this factor into the existing model has shown an improvement in radiative recombination lifetime determination of approximately 11% when comparison is made with the previous model. This has lead to an overall average improvement in lifetime determination of approximately 9% when comparisons are made between the new model and experimental data. This is a significant improvement in lifetime determination, which will benefit cell designers.

PACS: 85.35.B; 73.63.H; 84.60.J

Keywords: Computational modelling; Radiative recombination lifetime; Quantum well solar cell

1. Introduction

In the past few years, quantum well solar cells (QWSC) have attracted great research attention. This revolutionary device invented by Professor Keith Barnham of Imperial College shows great potential in surpassing the 40% efficiency mark and perhaps the 50% mark under solar concentration [1]. Although progress has been fruitful these efficiency figures have yet to be achieved [2,3]. One major factor, which has contributed to lower cell performances, has been carrier recombination. Recombination reduces the number of photo-generated carriers, which inevitably reduces cell performance. As mentioned in a previous paper, recombination occurs in three main process [4], Auger, SRH and radiative. In this paper, the author presents the latest results regarding a new approach to mathematically determine radiative recombination lifetimes for QWSC. It is intended that the model developed here will improve radiative recombination lifetime determination and be used to optimise future cell designs and improve cell efficiency.

A detailed review of the fundamentals of recombination lifetime modelling has been performed by Pratt et al. [4]. Since then many researchers have conducted calculations and experimentation for various electronic devices [5–13,15,17]. Our choice to study

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doi:10.1016/j.physe.2003.09.039

radiative recombination is seen through Barnham's original work [1]. It is assumed to be the dominant form of recombination within the QWSC. The new approach presented here is based on the radiative recombination equation of Pratt et al. [4] combined with a new approach to determine the thermal generation rate using a Burr distribution and the optical parameters of the QWSC [12]. The simple concept was to use the Burr distribution combined with the optical parameters of the QWSC to determine the probability of an electron–hole pair forming through the process of impact ionisation [13]. From this point the radiative recombination lifetime can be easily determined. The authors in Ref. [12] have presented and established a detailed approach used to form these links and draw the conclusions that the Burr distributions are simply the approximations of the more complex Giest–Wang distribution [14].

The work presented here is an extension and improvement of the above Burr distribution-based model [12]. The authors show how considering the semiconductor parameters such as dielectric parameters, photo-carrier masses and operating temperature can improve the determination of the lifetime results generated. Furthermore, comparisons between the new, previous (Burr-based) and experimental data are drawn to show the improvement and any discrepancies in the models are discussed.

The equation for radiative recombination lifetime is given by [4]

$$\tau_R = \frac{n_i^2}{G_R(n_0 + p_0)}, \tag{1}$$

where n_i is the intrinsic carrier concentration, n_0 the background concentration, $p_0 = n_i^2/n_0$ the hole concentration and G_R the thermal generation rate.

The authors have previously given a detailed definition of the Burr distribution can be combined with QWSC's reflectivity to obtain radiative recombination lifetime results [12] (Fig. 1).

Following the approach documented in Ref. [12] and introducing the delta function (δ), which is dependent upon semiconductor parameters, we obtain

$$G_R = 2 \times P \times N \times \delta, \tag{2}$$

where according to Ref. [12] P is the Burr distribution probability density function of an electron–hole pair forming and is defined as

$$P = \frac{CD}{B}\left(\frac{x-A}{B}\right)^{-C-1} \times \left(1 + \left(\frac{x-A}{B}\right)^{-C}\right)^{-D-1}, \tag{3}$$

where x is the variable being distributed, A the location of the peak distribution, B the scale of the distribution, C, D the shape of the distribution.

The above four parameters can be treated as fitting parameters. Typically the parameters are $A = 0$ and $B = 1$. As for C and D they can range between 3 and 6. Exceeding 6 often leads to lifetimes which are too short, whilst values of 1–2 give lifetimes typically too long. A successful attempt would be to ensure that the final distribution plot has a peak value nearest to PDF = 1 but does not exceed the PDF maximum value of 1. If this occurs then the parameters chosen would most likely give incorrect lifetimes. A PDF value of less than 0.85 can also lead to the same erroneous results. Given these rules the determination of the parameters can be quickly isolated. Typically, parameters of integer value are sufficient to generate reasonable results. If one desires accuracy a more sophisticated calculation can be easily performed with the use of a computer program and the use of iterative loops. This was not performed given the accuracy of the experimental data was taken from a graph from which the authors could only determine lifetimes of accuracy to only two decimal places. For the calculations the values of $A = 0$, $B = 1$, $C = 4$, $D = 4$ were used.

The use of fitting parameters may deem the theory to be a function of trial and error but it is not an uncommon practice. Nelson et al. [15] use fitting parameters to determine steady state carrier escape of particles from a single quantum well. But nonetheless a link to existing theory does exist and is discussed in detail in Ref. [12].

The number of photons absorbed by the cell is given as [12]

$$N = A(\lambda) \times \mathrm{NI}, \tag{4}$$

where NI is the number of incident photons on the cell.

$$A(\lambda) = 1 - R(\lambda),$$

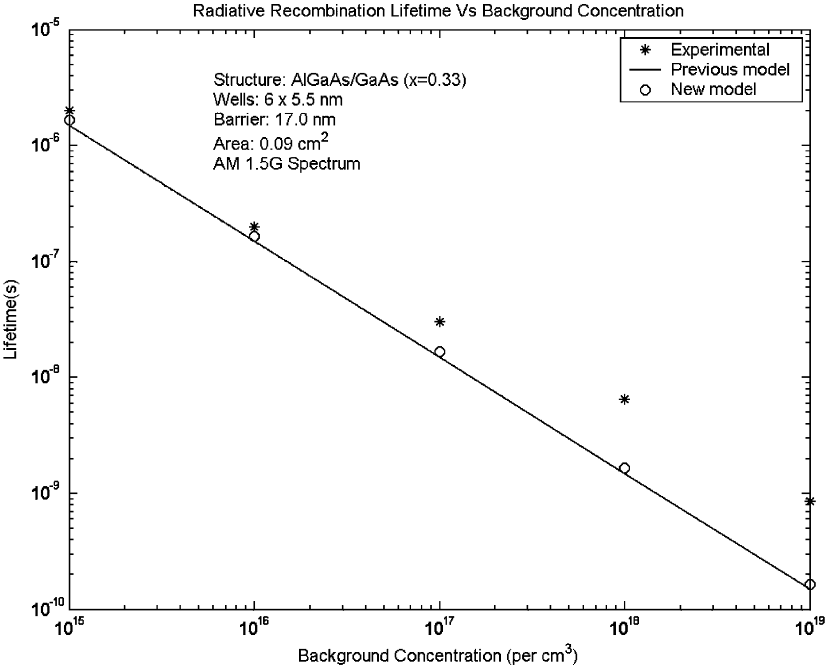

Fig. 1. Radiative recombination lifetime versus background concentration. Clearly a linear relationship is observed and as concentration increases the radiative recombination model breaks down and the dominant effects of Auger recombination take over.

$R(\lambda)$ (as a percentage) as a function of wavelength is determined using Ref. [16].

Assuming the use of standard solar spectrums AM0 or AM1.5G. The number of incident photons will be related to the spectrum radiance, cell area, time of exposure and spectrum.

$$\mathrm{NI} = \frac{W \times \mathrm{CA} \times T}{e} \times \frac{1}{\mathrm{SE}}, \tag{5}$$

where $W = 0.135\ \mathrm{mW/cm^2}$ (AM0) or $0.100\ \mathrm{mW/cm^2}$ (AM1.5G), $\mathrm{CA} = 0.09\ \mathrm{cm^2}$ is the area of the cell, and T the time of cell radiation. This was assumed to be 100 sec in order for steady-state cell operation to be achieved, and $e = 1.6 \times 10^{-19}$ C.

$$\mathrm{SE} = \frac{hc}{\lambda} \text{ is the solar energy spectrum (units eV),} \tag{6}$$

where λ is the appropriate wavelength spectrum. In this case, 300–1100 nm for AM1.5G.

Clearly the approach documented here is based on simple statistics and the determination of the expectation value. In simple terms if one knows the number of photons absorbed by the cell and the probability of an electron–hole pair forming then the generation rate is determinable. Using this and combining with Eq. (1) above, the radiative recombination lifetime is determined as a function of doping concentration.

The number of photons absorbed by the cell is dependent upon the number of incident photons on the cell. This is dependent upon the spectral irradiance and the cell's absorption coefficient. This is in turn dependent upon the cell's reflectivity. Using Eqs. (4)–(6) above allows one to determine this.

However, not all absorbed photons will produce an electron–hole pair. This follows given that photons of energy less than the semiconductor energy gap will not produce photo-carriers whilst photons of higher energy tend to produce phonons, which produce heat in the material and hence reduce semiconductor performance [13]. In order to consider these factors the probability function (Eq. (3) above) was introduced and modelled upon these above assumptions.

The theory behind the probability function used in this work is based on the work of Wolf et al. [13] and Giest and Wang [14]. The present author has described this link in detail in Ref. [12]. Concisely presented here, the conclusion is; the complex Giest–Wang distributions can be simply approximated using the Burr distribution. This is clearly seen in Figs. 2 and 3. Furthermore, support for the theory is seen through the probability density functions; they model the assumptions documented above. For energies below the energy gap the distribution show a zero likelihood of an electron–hole pair forming. As we move to the energy corresponding to the energy gap, the likelihood of an electron–hole pair forming is approximately 1 and as we go up in energies beyond the energy gap and exponential decay in the likelihood of an electron–hole pair forming is observed. This corresponds with the assumption that energies in this region are likely to form phonons, which reduce cell performance.

Having introduced the probability approach one must show the similarities and differences compared to existing theories. This is performed next.

According to Pratt et al. [18] the generation rate is defined as

$$G_{\mathrm{R}} = \frac{8\pi n^2}{h^3 c^2} \int_{E_{\mathrm{g}}}^{\infty} \frac{a(E)E^2}{\exp(E/kT) - 1}\,\mathrm{d}E, \tag{7}$$

where $a(E)$ is the absorption coefficient, n the refractive index, h the Planck constant, c the velocity of light, and E the energy spectrum, k the Boltzmann's constant and T the operating temperature in kelvin.

Following the approach, the above equation can be simplified to

$$G_{\mathrm{R}} = 1.29 \times 10^{30} (kT)^{9/2} \exp\left(\frac{-E_{\mathrm{g}}}{kT}\right) \times \left[\left(\frac{E_{\mathrm{g}}}{kT}\right)^2 + 5\left(\frac{E_{\mathrm{g}}}{kT}\right) + 8.75\right]. \tag{8}$$

From other work of Pratt et al. [4]

$$G_{\mathrm{R}} = 5.8 \times 10^{13} \varepsilon_{\infty}^{1/2} \left[\frac{m_0}{m_{\mathrm{e}} + m_{\mathrm{h}}}\right]^{\frac{3}{2}} \left[\frac{300}{T}\right]^{\frac{3}{2}} \times [E_{\mathrm{g}}^2 + 3kTE_{\mathrm{g}} + 3.75k^2T^2], \tag{9}$$

where ε_{∞} is the free space permittivity, ε the relative dielectric for GaAs=12.91, m_0 the mass of an electron, m_{e} the effective mass of an electron and m_{h} the mass of a hole.

If we closely look at the above Eqs. (7) and (8) it is obvious that our approach is similar. On a superficial overview one can observe the similarities to the previous work. Our approach to determine the number of photons absorbed by the cell (N above/Eq. (4)) through the use of the absorption coefficient and the energy spectrum is similar to that of the term $a(E)E^2$ above. Introducing the probability function and solving the function through the determination of the root mean square (RMS) [12] takes the form of the integral.

The reasoning behind the delta factor is seen through the work of Wolf et al. [13]. According to Wolf et al., the generation rate is proportional to the absorption coefficient and the transitional matrix. In our approach the absorption coefficient and the transitional matrix are determined using the approach documented in Ref. [16]. However, in our case the introduction of the probability function has changed the proportionality relationship. Therefore, to compensate for this the authors have defined a new constant associated with the proportionality. This new constant (delta factor) is shown related to the photo-carrier masses, dielectric constants and operating temperature.

The dependency of the delta factor on the above material parameters is shown in the previous work of Pratt et al. [4] in Eq. (9) above. The work shows the generation rate above can be represented as a function of semiconductor energy gap, particle masses, dielectric constants and temperature. Given this it would make sense to relate our constant in a similar nature.

To determine the delta factor one relies on the fitting parameter approach [15] justified above. After numerous simulations the delta factor is defined as

$$\delta = \varepsilon_{\infty} \varepsilon^{0.5} \left(\left(\frac{m_0}{m_{\mathrm{e}}} + \frac{m_0}{m_{\mathrm{hh}}} \right)^{-3} \left(\frac{T}{300} \right)^{\frac{3}{2}} \right), \tag{10}$$

where the parameters are based on those of Eq. (9) and m_{hh} is the mass of a heavy hole.

Following the approach documented above and in Ref. [12] the radiative recombination lifetime as a function of background concentration is easily determined.

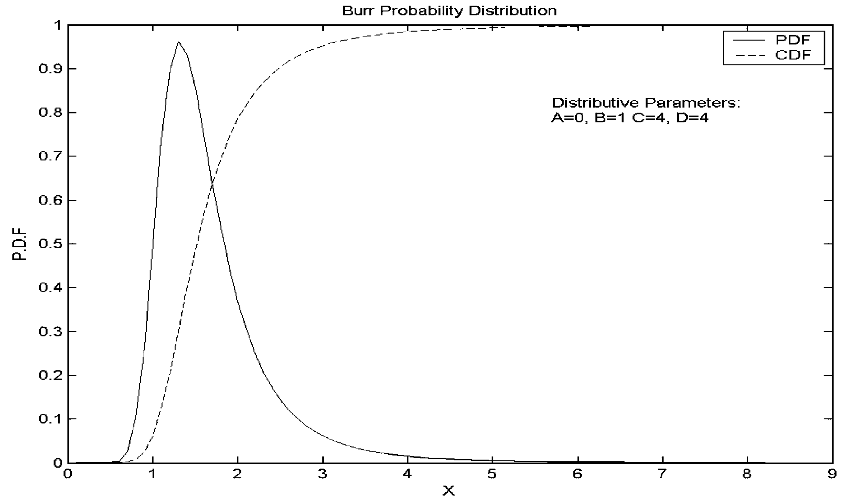

Fig. 2. Burr probability distribution used for lifetime analysis Ref. [12].

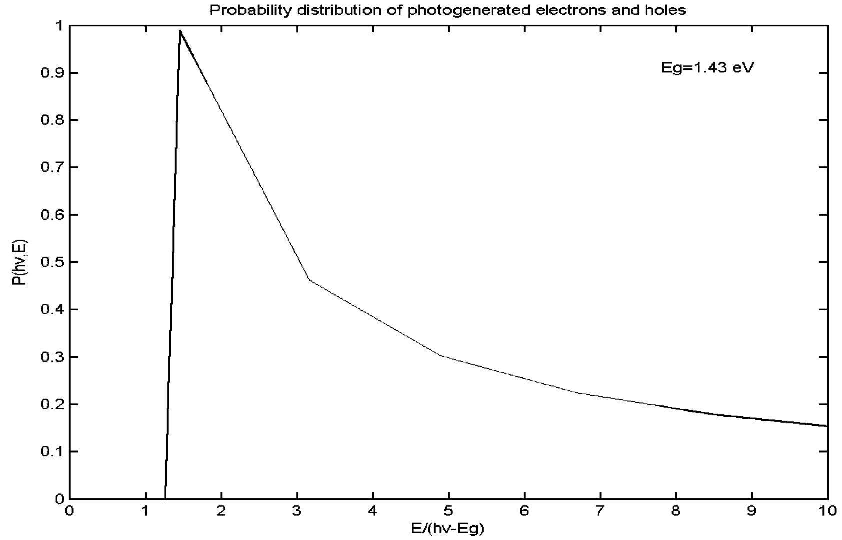

Fig. 3. Probability distribution proposed by Geist and Wang Ref. [13].

It must be made clear that the present authors are not claiming the present approach is equivalent to those above. However, from a superficial level the present work shows similarity in definition and clear differences, which justify the approach. Furthermore, the results obtained in comparison (discussed later) to previous (Burr) model and experimental data match accurately. Clearly the approach presented and that existing of Pratt are different. The work of Pratt [18] is based on a simple semiconductor material whilst that presented here is of a QWSC. Clearly quantum confinement will change the parameters.

Table 1
Comparison table of radiative recombination lifetime versus background concentration

Model	Background concentration (per cm^3)		
	10^{15}	10^{17}	10^{19}
New model	1.65 µs	0.0165 µs	0.00165 µs
Previous (Burr) model Ref. [12]	1.48 µs	0.0148 µs	0.00148 µs
Experimental Refs. [9,12]	2.00 µs	0.0250 µs	0.00800 µs
Discrepancy (new–previous)	11%	11%	11%
Discrepancy (previous–experimental)	35%	41%	81%
Discrepancy (new–experimental)	17%	34%	79%
Model improvement	18%	7%	2%

Comparison between new, previous and experimental data shows an overall improvement in determining radiative recombination lifetime. An average improvement of 11% is seen between new and previous models. An average improvement of 9% between model and experimental data is determined.

2. Computer-generated results

In accordance with the previous model (Burr-based) and experimental data documented in Ref. [12] the same QWSC device was considered and new simulations were performed with the delta factor introduced. Fig. 1 shows the linear relationship between radiative recombination lifetime and background concentration for the documented device under AM 1.5G spectrum. Furthermore, Table 1 shows the comparison and the accuracy of the new model compared to the previous (Burr-based model) as well as a comparison with experimental data.

3. Discussion

Investigation into radiative recombination lifetime modelling has been performed extensively. As mentioned before, there have been numerous papers which have performed theoretical and experimental investigations. These have included various structures including low-dimensional structure (quantum wells). In more recent time focus has been on solar cell performance. Work performed by Wolfe et al. on SiGe alloy comprised of a detailed calculation of cell performance through band structure, impact ionisation and probability density functions of photo-carrier generation [13]. A detailed balance theory was placed forth by Araujo and Marti [17]. This was done through a detailed evaluation of the absorption properties and optimisation of the cell structure through detailed continuity equations relating to the spectrum and photo-carrier generation.

The work presented here focuses on a new approach used to determine the radiative recombination lifetime of photo-carriers in QWSCs. It is intended that the work will aid in understanding the fundamentals of photo-carrier transport within the device and aid in optimisation of the device to increase cell performance. Our approach is based on a new probabilistic approach to determine the thermal generation rate through the process of impact ionisation [12,13]. This is combined with the original equations defined by Pratt et al. [4]. Recent results presented here have shown the introduction of a constant defined above as the delta factor which is based on semiconductor parameters improved determination of the radiative recombiantion lifetimes.

Although similar avenues as seen in the existing work are being explored, the main differences and advantages of the approach documented here is it is intended to be a simplified approach in computation whilst still giving reasonably accurate results. This is obvious given there is no need to calculate complex quantum mechanical band structures and the use of complex integrals which often have to be solved numerically. Further insight into the relationship between the radiative recombination lifetime and the optical parameters (cell reflectivity) are explored.

From Fig. 1 we observe the radiative recombination lifetimes versus background concentration for an aluminium gallium arsenide–gallium arsenide (AlGaAs/GaAs) QWSC. According to Ref. [12] the background concentration simulated was the range 10^{15}–10^{19}/cm^3. It is clear a linear relationship is established and overall the simulated values match well with the experimental data. From the graph we observe values of lifetime ranging from 1.5 to 2.0 µs at low concentration (10^{15}/cm^3) through to 1.5–8 ns at high concentration (10^{19}/cm^3). Furthermore, we observe the radiative recombination model breaking down as background concentration increased. This is expected and has been well documented by Andreev et al. [5] and discussed in detail in Ref. [12]. As concentration increases the effects of Auger recombination become dominant above the 10^{17}/cm^3 concentration level. This is clearly observed. Although this is a break down of the model it is not a concerning matter given that according to Barnham's original design methodology [1] a background concentration of $n_\mathrm{d} \leqslant 10^{15}\ \mathrm{cm}^{-3}$ is a sufficient upper limit.

From Table 1 we observe and compare results of the new and previous (Burr-based) models as well as their performance with experimental data. Three main points across the graphs in Fig. 1 have been taken to make the comparison. These correspond to the respective radiative recombination lifetimes associated with background concentration levels 10^{15}, 10^{17} and 10^{19}/cm^3. In the case of the 10^{15}/cm^3 concentration level: we observe a new model lifetime of 1.65 µs, a previous model lifetime of 1.48 µs and an experimental lifetime of 2.0 µs. From these figures we determine the discrepancy between new and previous model to be approximately 11%. A discrepancy between previous model and experimental to be approximately 35% and a discrepancy between new model and experimental to be approximately 17%. Clearly the last two values show an improvement in overall lifetime determination of the new model with the introduction of the delta factor to have improved by 18%.

As we increase background concentration to 10^{17} and 10^{19}/cm^3 we observe and overall improvement of approximately 7% and 2%, respectively. This leads to an average model improvement of approximately 9% over the background concentration range. The reduction in improvement as concentration increases can be attributed to the increased Auger recombination dominance discussed above and in detail in Refs. [5,12]. The discrepancies between new model and experimental results are slightly large (average discrepancies of 43% over the concentration range). This is due to the model breaking down as the background concentration increases. The effects of Auger recombination become more dominant as described by Andreev et al. [5]. Nonetheless a significant improvement in lifetime determination is observed and will lead to greater accuracy in lifetime modelling.

4. Conclusion

A new approach to determine radiative lifetime for an AlGaAs/GaAs QWSC has been presented. The new approach is based on a Burr distribution model combined with a previous model to determine the photo-generation of electron–hole pairs within the semiconductor layers and semiconductor parameters such as dielectric constants and particle mass to. The new model is compared to the previous model and shows an improvement of 11%. The new model results are then compared to existing experimental data and an improvement of 9% is observed. The discrepancies with the results are discussed and conclusions are determined. The approach is valid and would be an aid to semiconductor specialists.

References

[1] K.W.J. Barham, G. Duggan, J. Appl. Phys. 67 (1990) 3490.

[2] D.B. Bushnell, N.J. Ekins-Daukes, K.W.J. Barnham, J.P. Connolly, J.S. Roberts, J. Clark, G. Hill, M. Mazzer, Appl. Phys. Lett. 75 (1999) 4195.

[3] N.J. Ekins-Daukes, J. Zhang, D.B. Bushnell, K.W.J. Barnham, Strain-balanced materials for high-efficiency solar cells, 28th IEEE PV Specialist Conference, Anchorage, AL, USA, September 2000, pp. 1273–1276.

[4] R.G. Pratt, J. Hewett, P. Capper, C.L. Jones, N. Judd, J. Appl. Phys. 60 (1986) 2377.

[5] A.D. Andreev, G.G. Zegrya, IEE Proceedings Optoelectron 144 (1997) 336.

[6] B.K. Ridley, Phys. Rev. B 41 (1990) 12190.

[7] G.G. Zegyra, E.B. Dogonkine, A.S. Polkovnikov, Theoretical investigation of carrier relaxation process in quantum wells and their effect on radiative and Auger recombination, Semiconductor Conference Proceedings, CAS 99, 1999, pp. 351–354.

[8] A. Cuevas, M. Stocks, D. McDonald, M. Kerr, C. Samundsett, IEEE Trans. Elec. Dev. 46 (1999) 2026.

[9] P.J. Bishop, M.E. Daniels, B.K. Ridley, K. Woodbridge, Phys. Rev. B 45 (1992) 6686.

[10] J.W. Orton, P. Dawson, D.E. Lacklison, T.S. Cheng, C.T. Foxon, Semicond. Sci. Technol. 9 (1994) 1616.

[11] H.J. Polland, K. Leo, K. Rother, K. Ploog, Phys. Rev. B 38 (1988) 7635.

[12] F.K. Rault, A. Zahedi, Microelectron. J. 34 (2003) 265.
[13] M. Wolf, R. Brendel, J. Appl. Phys. 83 (1998) 4213.
[14] J. Geist, C.S. Wang, Phys. Rev. B 27 (1983) 4841.
[15] J. Nelson, M. Paxman, K.W.J. Barnham, J.S. Roberts, C. Button, IEEE JQE 29 (1993) 1460.
[16] F.K. Rault, A. Zahedi, Sol. Energy Mater. Sol. Cells 79 (2003) 470.
[17] G. Araujo, A. Marti, Sol. Energy Mater. Sol. Cells 33 (1994) 213.
[18] R. Pratt, J. Hewett, P. Capper, C.L. Jones, M.J. Quelch, J. Appl. Phys. 54 (1983) 5152.