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Quantum confinement modeling of electrical and optical processes in nanocrystalline silicon

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The paper presents a quantum confinement model for the electrical transport, the phototransport and the photoluminescence phenomena in nanocrystalline silicon. The infinite rectangular quantum well was proved to be the best choice for the investigated systems – nanocrystalline porous silicon and silicon nanodots embedded in an amorphous silicon dioxide matrix. Previous microstructure investigations have shown that the nanocrystalline porous silicon is formed by a nanowire network, so that the electron Hamiltonian is the sum of a one-dimensional Bloch-like Hamiltonian and a two-dimensional infinite rectangular quantum well. In the case of the silicon nanodots, the quantum well is three-dimensional. In both cases, the quantum well introduces quantum confinement levels in the band gap, the investigated phenomena being related with transitions between these levels.

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1. Introduction

Nanocrystalline silicon (nc-Si) is extremely interesting, for both fundamental and practical applications. The fundamental research is centered on its new specific properties and on the new phenomena that appear at nanometric scale. These are due in the first place to the quantum confinement (QC) and in the second one to the surface/interface effects. Indeed, the linear size of a nanocrystal is less than 20 interatomic distances and the surface/volume ratio is more than 10^8 m^{-1}. The practical applications are oriented towards the fabrication of new structures and devices. The compatibility of the nanocrystalline silicon-based materials with the classic mono- and/or polycrystalline silicon (bulk or thin films) permits the use of these new materials for the integrated micro- and optoelectronics, photonic crystals, biomedical applications or efficient sensors [1 – 10].

Various mechanisms for the electrical transport in nc-Si and Si-based nanostructures were investigated. We will consider only the nanocrystalline porous silicon (nc-PS) and the Si nanodots embedded in an amorphous SiO_2 matrix (Si – SiO_2). For nc-PS, the hopping processes at low temperatures with thermal activation at high temperatures [11], Poole-Frenkel tunneling [12], fractal percolation [13], generation-recombination phenomena in the depletion region [14], and dangling bonds governing the Al / nc-PS junction [15], were considered. For Si – SiO_2, the transport inside the nanodots is ballistic [16], and tunneling under Coulomb blockade occurs between nanodots [17]. A special role is played by the QC. It produces the enlargement of the band gap [18] and the breakdown of the momentum conservation rule [19], as well as the appearance of new energy levels, that introduce (additional) activation energies in the temperature dependence of the dark current (TDDC) [20, 21] and supplementary optical transitions [19, 22].

The phototransport (PT) was much less investigated [6, 23 – 25]. On the contrary, the photoluminescence (PL) phenomena were intensely investigated [1 – 4, 19, 22, 26 – 28]. It is now admitted the dominant role played by the QC, while the surface/interface contributions are related to the passivation of some non-radiative states and the formation of some radiative ones.

Our team investigated the electrical transport, the PT and the PL phenomena in nc-PS and Si – SiO_2 systems [29 – 38], in relation with their microstructure [31, 34, 39]. The microstructure of our nc-PS samples proved a double scale of porosity: an alveolar columnar structure of macropores (1 – 3 μm diameter, separated by walls of 100 – 200 nm thickness), and a nanoporosity of the alveolar walls, formed by a nanowire network (1 – 5 nm diameter and 5 – 35 μm length of the nanowires, leading to a length/diameter ratio of the order of 10^3 – 10^4) [31]. The Si – SiO_2 samples have variable nc-Si volume concentration (from $x \approx 0\%$ to $x \approx 100\%$), while the mean nanodot diameters vary slowly with x [34, 39].

In the following, we will present an unitary QC model for the TDDC, the PT and the PL phenomena in nc-Si. Section 2 discusses the model, while Section 3 analyzes the experimental results. The last Section summarizes the conclusions.

2. Modeling

One can introduce QC by supposing that the nanocrystallite surface/interface behaves as the wall of a quantum well, generating QC energy levels situated in the band gap. This is due to the fact that, at absolute zero temperature, the fundamental QC level represents the maximum occupied level, i.e. the top of the valence band. Different kinds of quantum wells were investigated: rectangular (either finite or infinite) [29 – 34], parabolic [40], Woods-Saxon [41] etc. The diferent shapes lead to different energy differences between consecutive levels, allowing the choice of the best shape for the modeling. For instance, a 1D infinite rectangular quantum well (IRQW) gives differences increasing like odd natural numbers, while a parabolic one gives equidistant levels. On the other hand, the differences between the first 3 – 4 levels in an infinite and a finite (2 – 5 eV depth) 1D rectangular quantum well for 1 – 10 nm width is less than 5% [30]. Thus, the experimental results can be described with good accuracy (with respect to the size and shape dispersion of the nanocrystallites) by the IRQW model. One has also to remember that the IRQW is independent of the kind of investigated nanomaterial.

As a first approximation, one can admit that the nanowires have cylindrical symmetry. Then, the electron Hamiltonian is given by the sum of a 1D Bloch-like longitudinal part and a 2D cylindrical IRQW transversal part, so that the electron energy is

$$\mathrm{E} = \varepsilon_{\mathrm{n,k_z}} + \frac{2\hbar^2}{\mathrm{m^*d^2}}\mathrm{x}^2_{\mathrm{l,p}} = \left(\varepsilon_{\mathrm{n,k_z}} + \frac{2\hbar^2}{\mathrm{m^*d^2}}\mathrm{x}^2_{0,1}\right) + \frac{2\hbar^2}{\mathrm{m^*d^2}}\left(\mathrm{x}^2_{\mathrm{l,p}} - \mathrm{x}^2_{0,1}\right) \equiv \varepsilon^{\mathrm{s}}_{\mathrm{n,k_z}} + \mathrm{E}_{\mathrm{l,p-1}}, \tag{1}$$

where ε^s_{n,k_z} is the longitudinal Bloch energy, shifted so that the energy of the QC transversal levels

$$E_{l,p} = \frac{2\hbar^2}{m^* d^2}\left(x^2_{l,p+1} - x^2_{0,1}\right) \tag{2}$$

are measured from the top of the valence band (by convention, $E_{0,0} \equiv E_v \equiv 0$), m^* = 0,66·m_e is the transversal effective electron mass, d is the mean nanowire diameter and $x_{l,p}$ is the p-th zero of the Bessel function $J_l(x)$, l being the orbital quantum number. In the following, the QC levels will be quoted (l, p).

The silicon nanodots in the Si – SiO$_2$ can be approximated as spherical [34]. For diameters less than about 5 nm, there are no more energy bands, but permitted quasibands (formed by neighboring levels), separated by (relatively) large intervals – forbidden bands. Because of the small number of atoms in a nanodot, the forbidden bands are increased with respect to the bulk crystals (with more than 100% for sizes of the order of 3 nm) and tend to become "direct". This is related with the fact that the momentum conservation law is no longer valid, due to the reduced number of states in a quasiband [20]. Then, the QC energy levels are

$$E \equiv E_{l,p} = \frac{2\hbar^2}{m^* d^2}\left(x^2_{l,p+1} - x^2_{0,1}\right), \tag{3}$$

where $m^* = m_e$ and $x_{l,p}$ is the p-th zero of the spherical Bessel function $j_l(x)$, l being the orbital quantum number.

Eqs. (1 – 3) are valid in the effective mass approximation, when $E \propto d^{-2}$. Rigorous calculations made by the linear combination of atomic orbitals method [19] proved that a better expression for the diameter dependence of the band gap is $E \propto d^{-\alpha}$, with α = 1.02 for cylindrical nanowires and α = 1.39 for spherical nanodots. More recently, exciton measurements [19] suggested even lower values for the exponent (α = 0.6 – 0.8). However the effective mass approximation could be used if one introduces a size-dependent effective mass [42]. Therefore the estimation of the nanocrystal diameters from Eqs. (2, 3) is not rigorous, but represents a good approximation.

Considering the energy differences between the QC levels given by Eqs. (2, 3), it results that, for diameters under 10 nm, these differences are at least one order of magnitude smaller than the thermal agitation energy $k_B T$, even at room temperature (RT). Then the temperature dependence of the carrier concentration is given by the Boltzmann law $n \propto \exp\left(-E_a / k_B T\right)$. The activation energy E_a is the absolute value of the difference between the energy of the last occupied level and the following one. As the drift velocity is constant (Ohmic behavior in nanowires and ballistic transport in nanodots [16]), the TDDC is Arrhenius-like. When a level is practically filled (the number of independent quantum states on a level is proportional with d^2 in nc-PS and with d^3 in Si – SiO$_2$), a following one starts to be excited, so that the activation energy is modified rather abruptly.

The ratio of two consecutive activation energies depends on the kind of nanocrystals. For nanodots, the ratio is

$$R_d = \frac{E''_a}{E'_a} = \frac{\varepsilon_{l'',p''} - \varepsilon_{l',p'}}{\varepsilon_{l',p'} - \varepsilon_{l,p}} = \frac{x^2_{l'',p''+1} - x^2_{l',p'+1}}{x^2_{l',p'+1} - x^2_{l,p+1}}. \tag{4}$$

For nanowires, the carriers are always excited from the valence band, so that

$$R_w = \frac{x^2_{l'',p''+1} - x^2_{0,1}}{x^2_{l',p'+1} - x^2_{0,1}}. \tag{5}$$

The value of the ratio depends on the kind of exitation. For thermal excitation (weak applied field, $eU << k_B T$), the first three levels are given by the quantum numbers p = 0, l = 0, 1, 2, so that R_w = 2.29 si R_d = 1.26. For high field excitation ($eU >> k_B T$), one has angular momentum conservation, therefore the first three levels correspond to l = 0, p = 0, 1, 2, and R_w = 2.80, R_d = 1.67. If the high field excitation starts at a temperature where the (1, 0) level is already thermally excited, the ratios are R_w = 2.25 and R_d = 1.50. In the case of the PT and PL phenomena, the optical selection rule implies $\Delta l = \pm 1$, leading to different values for the ratios R_w, R_d.

3. Results and discussion

The experimental investigations of the TDDC and PL on both nc-PS and Si – SiO$_2$ were presented in previous papers [29 – 36]. PT phenomena were investigated only on nc-PS. Indeed, in spite of the fact that the Si – SiO_2 samples are photosensitive, the currents tunneling between nanodots are too small to allow the measurement of the photocurrents with enough precision for a spectral analysis.

The TDDC in nc-PS was measured on both fresh and stabilized samples (through native oxidation), between liquid nitrogen temperature (LNT) and RT [29 – 33]. For the fresh samples, one activation energy, $E_f = 0.52 \pm 0.03$ eV, was observed, while for the stabilized samples the first energy is slightly shifted to $E_1 = 0.55 \pm 0.05$ eV and a second one, $E_2 = 1.50 \pm 0.30$ eV, appears at about 280 K. The shift of the first activation energy was interpreted as due to the thinning of the nanowires by oxidation (with one interatomic distance).

The PL measurements on stabilized nc-PS at RT proved one broad maximum, at $\lambda = 656 \pm 125$ nm ($E = 1.89 \pm 0.36$ eV) [35]. The maximum presents a gaussian shape, in agreement with the microstructure investigations [31].

The spectral dependence of the PT in nc-PS was measured on stabilized samples, in both sandwich structure (through a 325 nm thick semitransparent top Al electrode, on natively oxidized samples) and coplanar structure (paralel Al electrodes at 2 mm distance for natively oxidized samples and 3 mm distance for anodically oxidized ones) [36, 37]. The observed maxima and/or shoulders of the photocurrent are presented in Table 1. The maximum quoted F is but a small shoulder that appears only in two curves, in slightly different positions. This is why it was considered a false maximum and will be ignored in the following.

Table 1. Photocurrent maxima observed in nc-PS at RT.

No.	Para-meters	Sandwich (native oxidation)			Coplanar				Mean values
					Native oxidation			Anodical oxidation	
		$U = 1$ V	$U = 4$ V	$U = 20$ V	$U = 1$ V	$U = 4$ V	$U = 20$ V	$U = 1,5$ V	
1.	λ (nm)	525	500	500	–	–	500	500	505
	E (eV)	2.36	2.48	2.48	–	–	2.48	2.48	2.46
2.	λ (nm)	575	–	575	575	575	575	575	575
	E (eV)	2.16	–	2.16	2.16	2.16	2.16	2.16	2.16
3.	λ (nm)	–	625	625	650	–	625	–	631
	E (eV)	–	1.98	1.98	1.91	–	1.98	–	1.96
4.	λ (nm)	700	700	725	725	–	725	700	713
	E (eV)	1.77	1.77	1.71	1.71	–	1.71	1.77	1.74
F.	λ (nm)	775	–	–	–	750	–	–	763
	E (eV)	1.60	–	–	–	1.65	–	–	1.63
5.	λ (nm)	825	825	825	825	825	825	–	825
	E (eV)	1.50	1.50	1.50	1.50	1.50	1.50	–	1.50
6.	λ (nm)	–	–	875	875	875	875	–	875
	E (eV)	–	–	1.42	1.42	1.42	1.42	–	1.42
7.	λ (nm)	925	925	950	925	925	950	925	932
	E (eV)	1.34	1.34	1.31	1.34	1.34	1.31	1.34	1.33
8.	λ (nm)	–	1,025	1,025	1,025	1,025	1,025	–	1,025
	E (eV)	–	1.21	1.21	1.21	1.21	1.21	–	1.21

The temperature dependence of the photocurrent (TDPC) was investigated on anodically oxidized samples between LNT and RT [37]. Two activation energies were observed on the whole temperature interval, function of the illumination wavelength: $E_1 = 0.21$ eV for $\lambda = 500$, 575 and 600 nm, and $E_2 = 0.13$ eV for $\lambda = 650$ and 1.100 nm.

If one attempts to identify the different observed energies on the basis of the relations (1, 2, 5), one discovers that all the PT maxima but No. 6 are consistent with a mean nanowire diameter $d = 3.25$ nm. This value is also in good agreement with both the microstructure investigations and the TDPC, TDDC and PL measurements. If one defines the relative energy error as

$$\sigma_r = \frac{E_{\exp}}{\Delta E_{teor}} - 1 , \qquad (6)$$

the choice of the mean diameter leads to the condition $\sum \sigma_r = 0$ for the PT maxima. The maximum No. 6 cannot be identified with a transition between QC levels. This maximum corresponds to a transition between Si/SiO_2 interface states.

The first TDPC maximum is also identifiable with a transition between QC levels. The second one (observed at higher illumination wavelengths) is too small compared with the QC levels, but close to the surface trapping levels, located in the forbidden band [29, 43].

The PL maximum can be identified with high accuracy with the (2, 2) → (1, 1) transition. However, it can also be identified with the (0, 3) → (1, 1) transition (corresponding to No. 3 PT maximum), the error being still much smaller than the experimental broadening of the maximum.

Finally, both TDDC maxima were also identified with transitions between QC levels. In fact, it was this identification that originated the model previously presented and allowed to choose the IRQW as the best description of the investigated phenomena. All the identified transitions and the corresponding errors are presented in Table 2.

Table 2. QC transitions identified in nc-PS.

nc-PS	No.	E_{exp} (eV)	Transition	σ_r (%)
PT	1	2.46	(2, 3) → (1, 2)	− 2.63
	2	2.16	(1, 2) → (0, 0)	+ 0.99
	3	1.96	(0, 3) → (1, 1)	− 0.19
	4	1.74	(1, 2) → (2, 0)	+ 3.01
	5	1.50	(3, 1) → (2, 0)	− 0.54
	6	1.42	−	−
	7	1.33	(0, 2) → (1, 0)	+ 0.93
	8	1.21	(2, 1) → (1, 0)	− 1.57
	TDPC	0.21	(1, 0) → (0, 0)	+ 5.00
		0.13	−	−
PL	1	1.89	(2, 2) → (1, 1) (0, 3) → (1, 1)	+ 0.43 − 3.86
TDDC	1	0.55	(0, 1) → (0, 0)	+ 1.85
	2	1.50	(0, 2) → (0, 0)	− 0.66

The PL measurements on Si – SiO_2 samples were performed for different nc-Si volume concentrations (from $x = 50\%$ - zone 1 to $x = 70\%$ - zone 8) [38]. The results are presented in Table 3. When the nc-Si concentration increases, the envelopes of the experimental curves evolve from the envelope of a Poisson histogram to a gaussian, in agreement with the microstructure investigations [34, 39]. The TDDC measurements performed at $x = 66\%$ [34] gave three activation energy values at small bias (4 and 5 V), namely $E_1 = 0.22 \pm 0.02$ eV, $E_2 = 0.32 \pm 0.02$ eV and $E_3 = 0.44 \pm 0.02$ eV. At high bias (25 V), only E_2 and E_3 values can be observed. The analysis of the $I - V$ characteristic at the same concentration, based on the Si/SiO_2 potential barrier measured on nc-PS (2.2 eV, see [29, 33]), prove that the mean number of tunneled barriers between nanodots is $N = 87$, so that a variation of 20 V for the applied bias gives a mean voltage of 0.23 V per barrier (practically E_1/e).

Table 3. PL maxima observed in Si – SiO_2 at RT.

No.	Parameters	Zone 1	Zone 2	Zone 4	Zone 6	Zone 8	Mean values
1.	λ (nm)	425	425	425	400	400	415
	E (eV)	2.92	2.92	2.92	3.10	3.10	2.99
2.	λ (nm)	435	435	435	435	435	435
	E (eV)	2.85	2.85	2.85	2.85	2.85	2.85
3.	λ (nm)	455	460	460	455	455	457
	E (eV)	2.73	2.70	2.70	2.73	2.73	2.71
4.	λ (nm)	480	480	480	475	475	478
	E (eV)	2.58	2.58	2.58	2.61	2.61	2.59

Table 4. QC transitions identified in Si – SiO_2.

Si – SiO_2	No.	E_{exp} (eV)	Transition	σ_r (%)
PL ($d = 4.92$ nm)	1	2.99	(1, 6) → (2, 1)	+ 0.95
	2	2.85	(2, 5) → (1, 0)	− 1.10
	3	2.71	(0, 6) → (1, 1)	+ 1.47
	4	2.59	(2, 5) → (1, 1)	− 2.17
TDDC ($d = 5.28$ nm)	1	0.22	(1, 1) → (1, 0)	+ 0.61
	2	0.32	(1, 2) → (1, 1)	− 2.66
	3	0.44	(1, 3) → (1, 2)	+ 1.77

The transitions between QC levels corresponding to each observed energy were identified from the relations (3, 4) and are presented in Table 4. The mean nanodot diameters differ slightly for PL and TDDC measurements due to the diferent mean nc-Si concentrations.

4. Conclusions

The study of the electrical and optical processes in nc-Si proved the dominant role played by the QC. A simple unitary QC model, based on the IRQW potential, with estimated theoretical errors smaller than 5%, was proposed. This model allows for the identification of seven of the eight PT maxima and one of the two TDPC activation energies observed in nc-PS, as well as of all the PL maxima and all the TDDC activation energies observed in nc-PS and Si – SiO_2, as transitions between QC levels. The differences between the computed and the experimental energy values are in all these cases smaller than 4%.

The only elements that cannot be correlated with the QC model are: (i) one PT maximum in nc-PS (λ = 875 nm, $E_6 = 1.42$ eV), probably due to a radiative state at the Si/SiO_2 interface, and (ii) one TDPC activation energy in nc-PS ($E_2 = 0.13$ eV), probably due to some trapping centers situated either at the the Si/SiO_2 interface or at the crystallites interface.

The mean diameters of the nanocrystals estimated from the model (3.25 nm for the nc-PS nanowires and

4.92 – 5.28 nm for Si – SiO_2 nanodots) are in good agreement with the microstructure measurements.

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