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PHOTOEFFECT PECULIARITIES IN MACROPOROUS SILICON STRUCTURES

L.A. Karachevtseva*, V.F. Onyshchenko, A.V. Sachenko

Lashkaryov Institute of Semiconductor Physics of the National Academy of Sciences of Ukraine
41 Nauki Prsp., Kyiv 03028, Ukraine

The effects of increase in photoconductivity in the macroporous silicon structures have been examined as a function of the distance between cylinder macropores. The ratio of macroporous silicon photoconductivity to bulk silicon one has been found to achieve a maximum at the distance between macropores equal to the double thickness of the Shottky layer what corresponds to the experimental results. The relaxation time of photoconductivity for macroporous silicon structures was found to be defined by the light modulation of the barrier on macropore surfaces whereas its relaxation to occur according to the logarithmic law. If T>180 K, the temperature dependence of the relaxation time of photoconductivity is defined by a thermo-emission mechanism of the current transport in the space charge region and below 100 K the relaxation time is controlled by the processes of tunnel current flow.

INTRODUCTION

Macroporous silicon structures, unlike those of micro- and mesoporous one, are a material having a system of cylindrical air channels arranged in parallel one to another and normally to the substrate surface. Such structures are characterized by a large effective surface area what has a considerable effect on optical, electro-physical, and photo-electrical properties of the material. Macroporous silicon is promising for use in the infra-red spectral range owing to the optical absorption enhance [1], the photoconductivity enhance in the spectral range of the band-to-band light excitation [2], and to the possibility to measure additional photoconductivity bands [3]. The electron conductivity, concentration, and mobility as dependent on the macropore size and period have been studied for two-layer structures of "macroporous silicon–silicon" [4]. Both the microstructure of the macropore surfaces and the built-in electric field were found to depend on the parameters of the electrochemical process: the initial voltage and the current density [5]. At the same time, the sign of the main maximum in electroreflection spectra and the dependence of its amplitude on the static bias correspond to the formation of inversion (Schottky) layers at the macropore surfaces. That is why the conductivity and photoconductivity of the macroporous silicon structures are defined in this article as dependent on the distance between cylindrical macropores, the Shottky layers at the macropore surfaces being taking into account. The photoconductivity relaxation time and its temperature dependence for macroporous silicon structures have been studied experimentally and analyzed theoretically. The analysis was carried out taking into account the mechanism of current transport through a barrier in the space charge region at the macropore surfaces. The models of "frozen" or barrier photoconductivity considered previously in [6–8] were mathematically based, as a rule, on a linear model; i. e. the variation of the surface band bending under illumination was believed to be less than *kT/e*, the photoconductivity being depending linearly on the illumination intensity. In this work, we examine the case when the variation of the surface band bending under illumination is larger than *kT/e*.

METHODOLOGY

Specimens were made up of silicon plates characterized by the [100] orientation and the *n*-type of conductivity ($n_0 = 10^{15}$ cm^{-3}). The method of electrochemical etching at illuminating the back side of a silicon substrate of the thickness d = 400÷450 μm [9, 10] was used to form cylindrical macropores of the depth h = 140÷150 μm with diameter D_p = 2÷9 μm and concentration $N_P = (0.5÷2)\cdot 10^6$ cm^{-2}. The ohmic contacts In/single-crystalline *n*-Si and In/macroporous *n*-Si with a transient resistance

* Corresponding author *lakar@isp.kiev.ua*

of 4÷10 Ω·cm^2 were fabricated by thermal deposition of indium in a planar 4-probe configuration; the distance between the contacts was of 4 mm [4].

The photoconductivity spectra were measured at wavelengths λ = 0.8÷1.5 μm that included the band-to-band light excitation. The relaxation of photoconductivity in the samples of macroporous and single-crystalline silicon (see Table 1) was measured provided light fell normally to the surface, the contacts were screened, and the nonequilibrium charge carriers were pulse-excited with a GaAs laser diode with a 40 ns pulse duration and the wavelength of 0.88 μm. The laser pulse intensity at measurements was of 10^{16} photon/(cm^2·s).

Table 1. Macroporous silicon parameters

N	H, μm	h_P, μm	D_p, μm	a-D_p, μm	$\rho_{эф}$, Ω·cm	n, 10^{16} cm^{-3}
1	450	140	2	2.5	4.6	0.23
2	400	150	3	1.5	1.17	2.5
3	400	150	9	1	0.44	1.8
4	450	Si single crystal			3.8	0.22

DEPENDENCE OF CONDUCTIVITY AND PHOTOCONDUCTIVITY ON PORE GEOMETRY IN MACROPOROUS SILICON STRUCTURES

To find the dark conductivity and photoconductivity, we consider macroporous silicon with a regular structure of cylinder macropores with a period a (Fig. 1).The elementary cell consists of macropores with diameter D_p and the distance between pores $a - D_p$ (Fig. 1, inset). The ratio of the cylinder macropore surface area to that of the crystal surface is equal to:

$$\frac{S_p}{S_0} = \frac{\pi(D_p + 2w)^2}{2a^2}. \qquad (1)$$

That takes into account the Schottky layers of thickness w around a macropore. Here, S_0 is the area of the elementary surface cell limited to lines between the centers of macropores, and S_p is the part of S_0 occupied by macropores including the Shottky layer. The thickness of the Shottky layer on the macropore surface is determined by the formula from [11]:

$$w = 0.182\left(\frac{T}{300}\frac{10^{15}}{n_0}\right)^{0.5}\sqrt{2\ln\left(\frac{n_0}{n_i(T)}\right)}. \qquad (2)$$

Here, T is the absolute temperature, n_0 and $n_i(T)$ are the equilibrium electron concentrations in silicon matrix and the intrinsic charge carrier concentration in silicon, respectively. The Shottky layer thicknesses are equal to w = 0.9÷1.15 μm at T = 77÷300 K ($n_0 = 10^{15}$ cm^{-3}) for the examined macroporous silicon structures and determine the dark resistance of silicon matrix for the structures with the distance between macropores $a - D_p \approx 0.6÷3.6$ μm.

The dark resistance of a structure with regular cylinder macropores was determined as two series resistances of the silicon matrix (1) with area $S_0 - Sp$ and thickness h, and of the silicon substrate (2) with area S_0 and thickness $d - h$. This gives:

$$R_{Tp} = R_0\frac{d-h}{d}\left(1 + \frac{h}{d-h}\frac{1}{1-\dfrac{\pi(D_p + 2\cdot w)^2}{2\cdot a^2}}\right), \qquad (3)$$

where R_0 is the dark resistance of a crystal without pores. According to (3), the resistance of the crystal increases in the presence of macropores more rapidly when there are Shottky layers around pores with $w \to D_p/2$.

Under the conditions estimated earlier, the ratio of the photoconductivity of the macroporous silicon structure with highly depleted or inversed surface potential to that of the bulk silicon is proportional to the ratio of the total pore surface area S_{sp} to the structure surface area S_0:

$$\frac{S_{sp}}{S_0} = \frac{\pi h D_p}{a^2}, \quad a - D_p \geq 2w. \qquad (4)$$

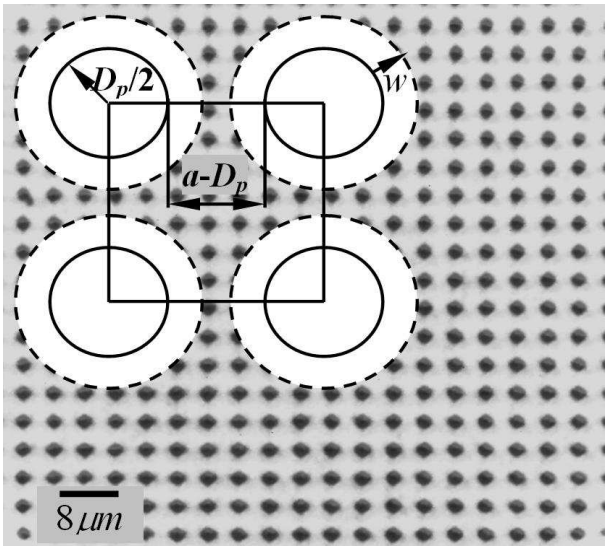

Fig.1. Two-dimensional macroporous silicon photonic structure with cylindrical macropores and its elementary cell consisted of four neighboring pores.

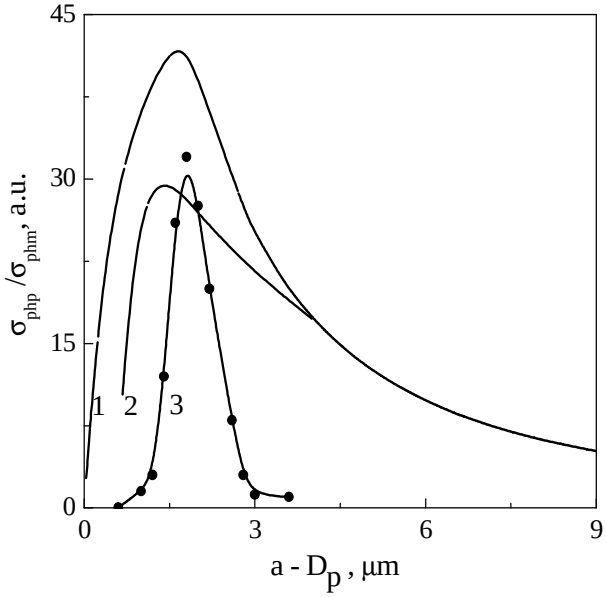

Fig.2. Ratio of macroporous silicon photoconductivity to bulk silicon photoconductivity versus distance between pores: (1) theoretical calculations for D_p = 2 μm, n_0 = 10^{15} cm^{-3}; (2) experimental data and (3) theoretical calculations for macroporous silicon structure with D_p = 0.5÷6 μm, a – D_p = 0.65÷ 4 μm, n_0 = 10^{15} cm^{-3}.

This ratio amounts to the value of 50÷100 for the macroporous silicon structures under study. When the distance between macropores is less than double thickness of a Shottky layer ($a - D_p < 2w$), the resulted depleted layer becomes thinner, and the ratio of the macroporous silicon photoconductivity σ_{php} to that of the bulk silicon σ_{phm}, diminishes:

$$\frac{\sigma_{php}}{\sigma_{phb}} \approx \begin{cases} \dfrac{\pi h D_p}{a^2}, & \text{for } a - D_p \geq 2w \\ \dfrac{\pi h D_p}{a^2}\dfrac{a - D_p}{2w}, & \text{for } a - D_p < 2w \end{cases} \qquad (5)$$

The sharp decrease in this ratio is due to the overlapping of Shottky layers and a decrease of the conductivity modulation under illumination. The dependences of the ratio (5) on the distance between pores $a - D_p$ are shown in Fig. 2 with a maximum value of 44 when $a - D_p = 2w$. It should be noted that the equation (5) is valid when macroporous silicon becomes excited uniformly along the full depth of macropores. The maximum value of the photoconductivity ratio experimentally measured is of 32 (Fig. 2) for silicon optical absorption depth of 50 μm (the absorption coefficient is of $2 \cdot 10^2$ cm^{-1} for λ = 0.95 μm) at $a - D_p \approx$ 2 μm what corresponds to the double thickness of a Shottky layer. The experimental data are in a good agreement with the theoretical calculations.

According to the model examined, the main contribution to the macroporous silicon photoconductivity is made by illumination-induced modulations of the Shottky layer conductivity. Here, the Shottky layer thickness is less than that in darkness and corresponds to the additional electron conductivity. In the case of conductivity inversion, the Gibbs excess of electrons under illumination is given by:

$$\Delta N = 0.182 \cdot 10^{-4}\left(\frac{T}{300}\frac{10^{15}}{n_0}\right)^{0.5}\left[\sqrt{2\ln\left(\frac{n_0}{n_i(T)}\right)} - \sqrt{2\ln\left(\frac{n_0}{n_i(T)}\right) - \Delta y_s}\right], \quad (6)$$

where $\Delta y_S = \Delta e\varphi_{s0}/kT$ is a change in the dimensionless surface potential under the illumination. The value Δy_S is found from the electroneutrality equation that includes a surface level charge, a non-equilibrium holes charge and a charge of completely ionized donors in the Shottky layer under illumination. The surface states were approximated by two discrete surface levels [12]. Thus, the ratio of macroporous silicon photoconductivity to bulk silicon dark conductivity is determined by a formula:

$$\sigma_{ph}{}^{p} / \sigma_{d}{}^{m} = e\mu_n \kappa \frac{\Delta N}{w}\frac{\pi h D_p}{a^2}\frac{a - D_p}{2w}R_0, \quad (7)$$

where, μ_n is the electron mobility and κ is a coefficient considerably less than unity which takes into account the fact that Shottky layers do not fill completely the space between pores. The ratio $\sigma_{ph}{}^{p} / \sigma_{d}{}^{m}$ is logarithmically dependent on Δy_S, and the photosensitivity of the crystal with pores is larger than that of bulk silicon.

PHOTOCONDUCTIVITY RELAXATION TIME IN MACROPOROUS SILICON STRUCTURES

The experimental plots of the photoconductivity relaxation (Fig. 3) in macroporous silicon cannot be approximated by an exponential law. The characteristic relaxation times of photoconductivity in macroporous silicon are of hundreds of microseconds at room temperature and tens of milliseconds at T < 200 K what are almost an order of magnitude larger than that in single-crystalline silicon.

Fig. 4 (curves *1*-*3*) shows the temperature dependence of the photoconductivity relaxation time τ_{ef} in macroporous silicon. Within the temperature range of 180÷300 K, the τ_{ef} temperature dependence has an activation character with the activation energy value of (0.30±0.1 eV). At T<100 K, the τ_{ef} value becomes practically independent on the temperature. The temperature dependence of the relaxation time of photoconductivity in single-crystalline silicon (Fig. 4, curve *4*) is characterized by a lower activation energy (less than 0.08 eV), and it correlates to the temperature dependence of the capture cross-section for a repulsive centre [13].

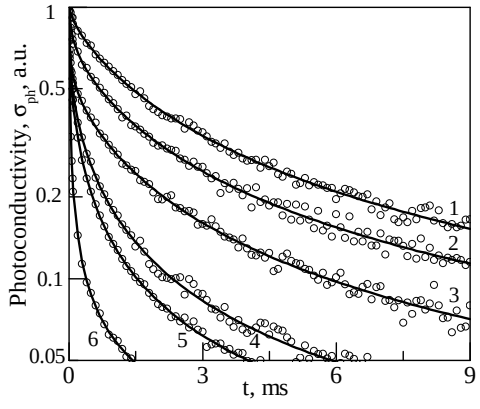

Fig. 3. Experimental (circles) and theoretical (curves) dependences of photoconductivity relaxation in macroporous silicon specimen 1 at various temperatures: 80 (*1*), 100 (*2*), 160 (*3*), 220 (*4*), 240 (*5*), and 300 K (*6*).

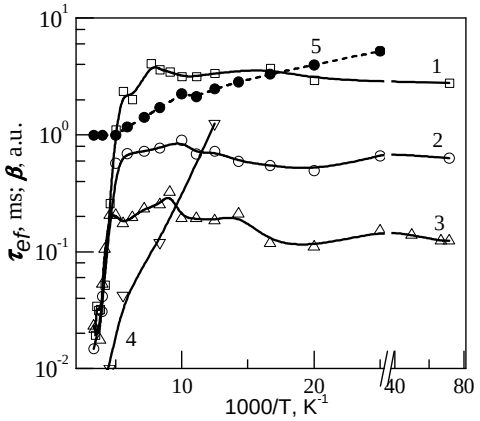

Fig. 4. Temperature dependences of the photoconductivity relaxation time τ_{ef} for macroporous silicon specimens 1 (*1*), 2 (*2*), 3 (*3*) from Table, the single-crystalline one *(4)* and the calculated non-ideality factor β for specimen 2 *(5)*.

The previous results [14] on the current-voltage characteristics of the barrier contact "indium–macroporous silicon" not subjected to illumination are evidence of the realization of both thermoemission and tunnel mechanisms of the current transport by charge carriers. The thermoemission one has the activation energy of about 0.25 eV and dominates within T = 180÷300 K what correlates with the data on the temperature dependence of the photocarrier lifetime (Fig. 4, curves *1-3*). Hence, the activation temperature dependence of the photocarrier lifetime in macroporous silicon structures is controlled by the potential barrier in the subsurface region of macropores. It is connected with the fact that the barrier decreases the recombination between photoelectrons in the silicon matrix and holes at the silicon surface. At the temperature T<100 K, the lifetime of photocarriers becomes temperature-independent (Fig. 4). According to [14], at T = 77÷100 K, the non-ideality factor β for nonlinear current-voltage characteristics grows from 1.5 to 5 with lowering temperature what is typical for the tunnel mechanism of current transport.

THEORETICAL ANALYSIS OF THE PHOTOCONDUCTIVITY RELAXATION IN MACROPOROUS SILICON STRUCTURES

According to the above results, the photoconductivity relaxation in macroporous silicon structures has some specific features as compared with that in single-crystalline silicon. The origin of those features is that, owing to the developed surface of macropores, light-generated electron-hole pairs are brought mainly into the space charge region at the macropore surface, which is for n-Si a potential well for holes and a potential barrier for electrons [11]. While analyzing the photoconductivity relaxation in macroporous silicon, the fact is to be taken into account that the barrier height $e\varphi_s$ decreases under illumination and increases during the process of photoconductivity relaxation. This phenomenon should result in a non-exponential character of relaxation (Fig. 3) and in the growth of the effective lifetime value as compared with that in single-crystalline silicon according to experimental results (Fig. 4).

To simulate the photoconductivity kinetics, let us consider the macroporous silicon structure with cylindrical macropores presented in the inset in Fig. 1. When falling perpendicularly to the macroporous silicon surface (parallel to cylindrical macropores), the light is absorbed by silicon located between macropores. In accordance with experimental data [4, 6, 11], let us assume that, in the space charge region adjacent to the surface of cylindrical pores, there exist equilibrium band bendings which correspond to the inversion of conductivity. The photoconductivity relaxation in macroporous silicon was estimated, taking into account the assumption that the photoconductivity is controlled by nonequilibrium electrons ΔN which partially fill the equilibrium Schottky layer, i. e.

$$\sigma_{ph} = q\mu_n \Delta N \,. \qquad (8)$$

Let the kinetic equation for finding the law of photoconductivity relaxation be written down in the form

$$\frac{d\Delta N}{dt} = -\, j_r(t) \,. \qquad (9)$$

The quantity $j_r(t)$ is a recombination flux through the macroporous surface determined from the current-voltage characteristics of a surface barrier (like those for Schottky contact or *p–n*-junction) provided that the circuit is broken:

$$j_r(t) = \frac{J_s}{q}\left(\exp\left(\frac{\Delta y_s(t)}{\beta}\right) - 1\right). \qquad (10)$$

Here J_s is the saturation current surface density, β is the coefficient of non-ideality of current-voltage characteristics, and $\Delta y_s(t)$ is the change in the surface potential after illumination. When substituting Eq. (10) into Eq. (9), we obtain for $\Delta y_s(t) > 1$ a logarithmic relaxation law:

$$\Delta N(t) \cong 2L_D n_0 \times \\ \times\left(\sqrt{y_0} - \sqrt{y_0 - \Delta y_s(t=0) + \beta \ln(1 + t/\tau_r)}\right) \qquad (11)$$

$$\tau_r = \frac{e\beta n_0 L_D}{J_s \sqrt{y_0}} \exp\left(-\frac{\Delta y_s(t=0)}{\beta}\right). \qquad (12)$$

Here L_D is the Debye screening length, $y_0 = e\varphi_{s0}/kT$ is the dimensionless equilibrium band bending on the macropore surface. The magnitude of the effective photoconductivity relaxation time (τ_{ef}) necessary to reduce the photoconductivity by half is determined from the equation

$$\frac{\sqrt{y_0} - \sqrt{y_0 - \Delta y_s(t=0) + \beta \ln(1 + \tau_{ef}/\tau_r)}}{\left(\sqrt{y_0} - \sqrt{y_0 - \Delta y_s(t=0)}\right)} = \frac{1}{2}. \qquad (13)$$

Let us analyze the temperature dependence of the effective time τ_{ef} in macroporous silicon. Within the range of high enough temperatures,

where the thermoemission mechanism of current flow dominates (β = 1), the quantity $\tau_r \sim \exp((E_g - \Delta y_s(t=0)/kT)$, and τ_{ef} value grows exponentially with the temperature decrease. The situation is not changed essentially in the intermediate temperature region, where the recombination mechanism of the current flow dominates, i. e. when β=2 and $\tau_r \sim \exp(E_g/2kT)$. The rate of the τ_{ef} growth becomes slower as compared with those in the case considered above. Fundamental modifications occur at the low enough temperatures, when the tunnel mechanism of the current flow becomes dominant. In this case, $\beta = \varepsilon_T / kT$, where ε_T is the characteristic tunnel energy. Therefore, the ratio $\Delta y_s(t=0)/\beta$ is equal to $ln(J_{ph}/J_s)$ where J_{ph} is the surface density of photogeneration current and should not depend on the temperature, if the quantity J_s is temperature independent. That is why the lifetime of photocarriers τ_{ef} would be either independent or weakly dependent on the temperature in the low-temperature range when the tunnel mechanism of current dominates.

By fitting the theoretical dependences of the photoconductivity relaxation (11) to experimental ones (Fig. 3), the magnitudes of the surface potential in darkness, y_0, and under illumination, $y_0 - \Delta y_s(t=0)$, were determined. In particular, the dimensionless y_0-value at T = 300 K is approximately equal to $|y_0|$ = 12, which corresponds to the equilibrium surface band bending of about 0.31 eV and to the experimental temperature dependence of the photoconductivity relaxation time τ_{ef} with the activation energy E_a = 0.31 ± 0.01 eV (Fig. 4, curves 1–3). In Fig. 4 (curve 5) the temperature dependence of the β factor is depicted, which is obtained after (13) and the experimental data from (Fig. 3). The figure demonstrates that the dependence corresponds to the thermoemission mechanism of current flow (β = 1) at T = 180÷300 K and to the tunnel one ($\beta > 2$) at T <100 K in analogy to the experimental τ_{ef} temperature dependences.

CONCLUSIONS

The effects of the photoconductivity increase in the periodical macroporous silicon structures have been examined as dependent on the distance between macropores, an inversion of conductivity at the macropore – silicon interface (Shottky layer) being taken into account. The ratio of macroporous silicon photoconductivity to bulk silicon photoconductivity has been shown to achieve a maximum at the distance between macropores equal to two thicknesses of Shottky layer what corresponds to the experimental data. The increase in photosensitivity is due to the large macropore total surface area and the existence of Shottky layers in macropore subsurface region.

The photoconductivity kinetics in macroporous silicon has been found experimentally to have a nonexponential law of relaxation. Within the range T = 180÷300 K, the temperature dependence of the photoconductivity relaxation time has an activation character with the activation energy of 0.3±0.01eV and does not depend on the temperature at T < 100 K. The character of the photoconductivity relaxation in macroporous silicon has been analyzed theoretically as dependent on the mechanism of current flow through the subsurface region of space charge. The effective relaxation time of photoconductivity has been found to be controlled by the light modulation of the barrier on the macropore surfaces whereas the relaxation itself follows the logarithmic law. The temperature dependence of the photoconductivity relaxation time is determined by the thermoemission mechanism of the current flow through the subsurface space charge region at T>180 K, and, below 100 K, by the tunnel processes of current flow. In the last case, the tunnel probability, the photocarrier lifetime, and cross-section of photocarrier capture do not depend on the temperature. The dimensionless y_0-value is about 12 at room temperature what corresponds to the equilibrium surface band bending of about 0.31 eV and correalte with the data on activation energy E_a of the experimental temperature dependence of the photocarrier lifetime.

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Особливості фотоефекту в структурах макропористого кремнію

Л.А. Карачевцева, В.Ф. Онищенко А.В. Саченко

Інститут фізики напівпровідників ім. В.Є. Лашкарьова Національної академії наук України, просп. Науки, 41, Київ 03028, Україна, lakar@isp.kiev.ua

Досліджені ефекти підвищення фотопровідності в структурах макропористого кремнію в залежності від відстані між циліндричними макропорами. Встановлено, що відношення фотопровідності макропористого кремнію до фотопровідності монокристалічного кремнію досягає максимуму при відстані між порами, яка дорівнює двом товщинам шару Шотткі, що відповідає результатам експерименту. Час релаксації фотопровідності структур макропористого кремнію визначається модуляцією світлом бар'єру на поверхні макропор, а її релаксація відбувається за логарифмічним законом. При T>180.K температурна залежність часу релаксації фотопровідності визначається термоемісійним механізмом проходження струму в області просторового заряду, а при T<100.K - тунельними процесами струмопереносу.

Особенности фотоэффекта в структурах макропористого кремния

Л.А. Карачевцева, В.Ф. Онищенко, А.В. Саченко

Институт физики полупроводников им. В.Е. Лашкарева Национальной академии наук Украины, просп. Науки, 41, Киев 03028, Украина, lakar@isp.kiev.ua

Исследованы эффекты повышения фотопроводимости в структурах макропористого кремния в зависимости от расстояния между цилиндрическими макропорами. Установлено, что отношение фотопроводимости макропористого кремния к фотопроводимости монокристаллического кремния достигает максимума при расстоянии между порами, равном двум толщинам слоя Шоттки в соответствии с результатами эксперимента. Время релаксации фотопроводимости структур макропористого кремния определяется модуляцией светом барьера на поверхности макропор, а ее релаксация происходит по логарифмическому закону. При T>180.K температурная зависимость времени релаксации фотопроводимости определяется термоэмиссионным механизмом прохождения тока в области пространственного заряда, а при T<100K - туннельными процессами токопереноса.