

ANOMALOUS-SIGN PHOTO-EMF IN MACROPOROUS SILICON AT PHOTON ENERGIES COMPARABLE TO THAT OF INDIRECT BAND-TO-BAND TRANSITION

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Experimental and theoretical temperature dependences of photo-emf generated in macroporous silicon at photon energies comparable to that of the indirect interband transition in silicon have been studied. The photo-emf was found to saturate or change its sign to negative at temperatures lower than 130 K owing to the light absorption due to phototransitions via surface states located closely to the conduction band in silicon. In this case, the surface band bending increases due to the growth of a negative charge of the semiconductor surface. Equilibrium electrons in the bulk and light-excited holes on the macropore surface recombine through the channel of multistage tunnel recombination between the conduction and valence bands.

silicon specimens owing to a larger surface area in the former.

In this work, we continued works [4, 5] by studying the temperature dependences of the photo-emf emerging in macroporous silicon at photon energies that are comparable with the energy of the indirect band-to-band transition in silicon ($\lambda = 0.7 \div 0.95 \mu\text{m}$). The calculations of the temperature dependence of the surface photo-emf were carried out by using the balance equation for the generation-recombination process involving nonequilibrium charge carriers and taking into account the thermoactivated and tunnel mechanisms of recombination, as well as the recombination in a space charge region (SCR) under the open circuit condition. To explain the anomalous photo-emf sign, we consider the light absorption that occurs with the participation of surface states located near the conduction band.

1. Introduction

Macroporous silicon fabricated by photoanodic etching occupies a special place among two-dimensional photonic crystals. This fact is associated with the possibilities to use this material to manufacture structures with necessary geometries [1] and to form additional optical absorption bands in it. An additional advantage of this substance is its photo-electric properties. It should be noted that silicon is an extremely attractive material for integrated optics at the 1.55- μm telecommunication wavelength owing to the compatibility with circuits including MOS structures. In last years, there appeared the reports concerning a photocurrent running in a ridge waveguide fabricated on the basis of a silicon-dielectric structure as a result of the absorption by surface states [2, 3]. Further works should reveal the source and the mechanism of this photocurrent. According to the results of our researches of structures made of macroporous silicon [4, 5], their photoconductivity at low temperatures is governed by the tunnel mechanism of a current flow in the near-surface region of the silicon matrix. In this case, the contribution of the tunneling to the photoresponse and the dark conductivity of macroporous silicon is substantially higher than that in single-crystalline

2. Experimental Technique

We studied macroporous silicon structures fabricated on silicon wafers characterized by the [100] orientation, the thickness $H = 400 \div 450 \mu\text{m}$, the n -type of conductivity, the equilibrium concentration of electrons $n_0 = 10^{15} \text{ cm}^{-3}$, and the specific resistance of $4.5 \Omega \times \text{m}$. Electrochemical etching at the illumination of the back side of silicon substrates [6, 7] was used to form cylindrical macropores of the depth $h_p = 100 \div 150 \mu\text{m}$. Macropores were arranged irregularly. Their average diameter $D_p = 2 \div 9 \mu\text{m}$, and the average concentration $N_P = (0.5 \div 2) \times 10^6 \text{ cm}^{-2}$. The specific area of the surface of macropores (per unit volume of macroporous silicon) was $S_p = 1200 \div 2500 \text{ cm}^2/\text{cm}^3$. The table presents the parameters of those specimens, for which the experimental photo-emfs *versus* the temperature are depicted in Figs. 1, 2, and 5.

Non-Ohmic contacts “In/single-crystalline n -Si” and “In/macroporous n -Si” were prepared by the thermal deposition of indium in a planar four-probe configuration

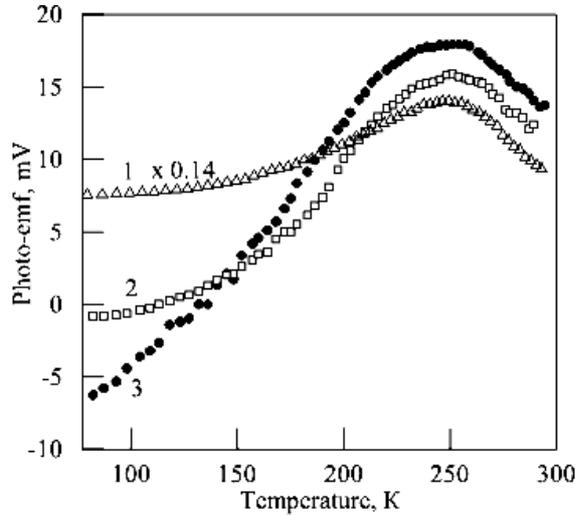


Fig. 1. Experimental temperature dependences of the photo-emf in macroporous silicon (specimen 1) at the illumination with an intensity of $(2 \pm 0.3) \times 10^{14}$ photon/($\text{cm}^2 \cdot \text{s}$) and various light wavelengths $\lambda = 0.7$ (1), 0.94 (2), and $0.95 \mu\text{m}$ (3)

with a distance of 4 mm between contacts and a transient resistance of $4 \div 10 \Omega \times \text{cm}^2$ [8]. The four-probe configuration was used to monitor the dark concentration of free charge carriers. The temperature dependences of the photo-emf in macroporous silicon structures were measured in the range $77 \div 300$ K in the open circuit regime, and making use of two contacts, one of them being illuminated. For illumination, a GaP-based light-emitting diode with a wavelength of $0.7 \mu\text{m}$ (the red spectral range) and IR light-emitting diodes – GaAs (the wavelength of $0.94 \mu\text{m}$) and GaAlAs/GaAs (the wavelength of $0.95 \mu\text{m}$) ones – were used.

3. Temperature Dependence of Surface Photo-emf in Macroporous Silicon Structures

Figure 1 illustrates the experimental temperature dependences of the photo-emf generated in macroporous silicon at its excitation with red and infra-red light with wavelengths of 0.7 , 0.94 , and $0.95 \mu\text{m}$ and an intensity of $(2 \pm 0.3) \times 10^{14}$ photon/($\text{cm}^2 \times \text{s}$). The figure corresponds to the case where the photo-emf values V_{OC} were less than kT/q . It is evident from the figure that, being

Structure parameters of macroporous silicon

Specimen No.	H , μm	h_p , μm	D_p , μm	N_p , cm^{-2}	S_p , cm^2/cm^3
1	400	130	6	10^6	1884
2	400	100	6 ± 3	$1.3 \cdot 10^6$	2448

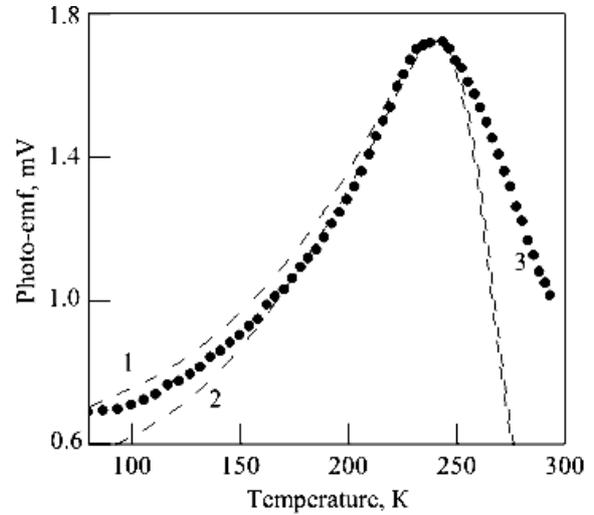


Fig. 2. Temperature dependences of the photo-emf in macroporous silicon at the illumination with the light wavelength $\lambda = 0.7 \mu\text{m}$ and the intensity $I = (2 \pm 0.3) \times 10^{14}$ photon/($\text{cm}^2 \cdot \text{s}$): the theoretical dependences for the parameter values $\varepsilon_{T0} = 0.02$ and $\varepsilon_1 = 0.08$ (curve 1), and $\varepsilon_{T0} = 0.01$ and $\varepsilon_1 = 0.06$ (curve 2), and the experimental data (curve 3) for specimen 2

excited with red light with a wavelength of $0.7 \mu\text{m}$, the photo-emf has a maximum in the range $200 - 260$ K and saturates at $T < 120$ K. If the specimen is illuminated with infra-red light ($\lambda = 0.94$ and $0.95 \mu\text{m}$), the photo-emf has a maximum in the same temperature range. At the same time, the photo-emf continues to decrease at lowering the temperature and, at temperatures of about 130 K, changes its sign. At the insignificant reduction of the wavelength from $\lambda = 0.95 \mu\text{m}$ to $\lambda = 0.94 \mu\text{m}$, the negative photo-emf decreases.

It should be noted that the temperature dependences of the photo-emf in microporous silicon were studied in work [9]. In that work, the intensity of illumination was rather high, so that an almost complete straightening of bands was achieved. In this case, the photo-emf magnitude can be used to determine the initial surface band bending (before illumination). We note that, in the case of interband light absorption, when the illumination generates electron-hole pairs, the surface band bending can only diminishes at the illumination in comparison with its value in the illumination absence. In this case, the photo-emf sign is always positive for a semiconductor of the n -type and negative for a semiconductor of the p -type, which is a usual practice. In works [10, 11], the temperature dependence of the photo-emf in semiconductors of group A_2B_6 was studied in another limiting case where the photo-emf is comparable with kT/q . It was found that, when the photo-emf was excited with

light with the energy of photons lower than the energy gap in CdTe, the photo-emf changed its sign at temperatures below 150 K. This means that the surface band bending does not decrease at the illumination, as usual, but increases. Such a photo-emf will be referred to as anomalous in what follows. Work [12] presents a theoretical model which allows one to explain the variation of the photo-emf sign by considering the tunnel recombination of equilibrium electrons in the bulk and nonequilibrium holes at the surface of semiconductor.

The temperature dependence of the normal-sign surface photo-emf V_{OC} can be calculated taking advantage of the generation-recombination balance equation for nonequilibrium charge carriers [13] and making allowance for three mechanisms of recombination under the open circuit condition. The corresponding equation looks like

$$I_G = I_{01} \frac{n_i^2(T)}{n_i^2(300)} \left(\exp\left(\frac{qV_{OC}}{kT}\right) - 1 \right) + I_{02} \frac{n_i(T)}{n_i(300)} \times \left(\exp\left(\frac{qV_{OC}}{2kT}\right) - 1 \right) + I_{03} \left(\exp\left(\frac{qV_{OC}}{\varepsilon_T}\right) - 1 \right). \quad (1)$$

Here, I_G is the surface density of the photogeneration current, the first term on the right-hand side describes the recombination current density, when the superbarrier mechanism of recombination at the surface and in the bulk dominates, the second term is the recombination current density in the SCR, and the third one is the density of the tunnel recombination current. The quantity I_{01} is the saturated current density, when the superbarrier mechanism of recombination dominates and at room temperature; I_{02} is the saturated current density at recombination in the SCR and at room temperature; I_{03} is the saturated tunnel current density; and ε_T is a characteristic energy of tunneling. According to the results of work [14], in the temperature range 77 – 300 K, the temperature dependence of the concentration of intrinsic charge carriers in silicon $n_i(T)$ can be described by the expression

$$n_i(T) = 2.91 \times 10^{15} T^{1.6} \exp\left(\frac{6561}{T}\right). \quad (2)$$

In Fig. 2, the theoretical (curves 1 and 2, calculated making use of Eqs. (1) and (2)) and experimental (curve 3) temperature dependences of the photo-emf V_{OC} in macroporous silicon subjected to the light illumination with $\lambda = 0.7 \mu\text{m}$ are depicted in the range, for which $V_{OC} < kT/q$. The figure evidences for rather a good agreement between the theory and the experiment.

When calculating the photo-emf, we used the following temperature dependence of the characteristic tunneling energy:

$$\varepsilon_T = \varepsilon_{T0} + \varepsilon_1(T/300)^3, \quad (3)$$

which, in particular, allowed the temperature dependences of current-voltage characteristics obtained in work [13] to be explained.

4. Photo-emf in Macroporous Silicon Structures at the Participation of Surface States in the Trapping of Majority Charge Carriers

It should be noted that expression (1) quantitatively describes the emerging photo-emf in the case of inverse band bending at the surface. In the case of depleting band bending in silicon, the effect of surface sticking is important. The latter consists in that the overwhelming number of minority charge carriers are trapped by surface states. In this case, the photo-emf grows, and its sign remains normal [15, 16]. By analogy with those data, let us make allowance for the light absorption with the participation of surface states in the trapping of majority charge carriers in order to explain the anomalous photo-emf sign. In particular, let us consider photo-induced transitions between the valence band and the surface electron states (SESs) located close enough to the conduction band in macroporous silicon. We suppose that the depleting band bending is realized at the macropore surface in the absence of illumination. For the band bending to be larger at the illumination, which corresponds to the anomalous photo-emf sign, it is necessary that the occupation of surface states, which govern band bending, increase. In this case, the positive charge in the SCR of the semiconductor grows, the nonequilibrium band bending at the semiconductor surface increases, and the photo-emf sign becomes anomalous. From the continuity equation that describes the nonequilibrium SES occupation with electrons owing to photo-induced transitions between the valence band and the SESs [12], we obtain, in the stationary case, the following expression for the nonequilibrium electron concentration n_t :

$$n_t = \frac{(\gamma_t I + C_{nt} n_s - C_{pt} p_{1t}) N_t}{C_{nt}(n_s + n_{1t}) + C_{pt}(p_s + p_{1t}) + \gamma_t I}, \quad (4)$$

where N_t is the SES concentration; γ_t is the coefficient that describes the probability of light absorption with the participation of SESs; I is the intensity of illumination, n_s and p_s are, respectively, the electron and hole

concentrations at the surface; C_{nt} and C_{pt} are, respectively, the coefficients of electron and hole trapping by SESs; $n_{1t} = n_i \exp(E_t/kT)$ and $p_{1t} = n_i \exp(-E_t/kT)$ are the Shockley–Reed factors for electrons and holes, respectively; and E_t is the surface level depth with respect to the energy gap middle point. In this case, interesting for analysis are surface levels located near the conduction band. In the absence of illumination, such states are poorly occupied with electrons, i.e. $n_{t0} \ll N_t$. At illumination, the concentration of electrons on them grows. Since the value of n_{1t} is much higher than the concentration of electrons or holes at the surface in the range of room temperatures, a larger fraction of electrons excited—due to thermal activation—on SESs transits into the conduction band (the transition scheme is illustrated in Fig. 3,*a*); afterwards, they are driven by an electric field from the semiconductor surface to the SCR boundary. The ultimate result of this process is the appearance of photoelectrons in the conduction band. Photoelectrons are excited due to phototransitions with the participation of SESs, and their integral concentration is practically equal to the Gibbs excess for photoholes. This case corresponds to the normal sign of the photo-emf.

When the temperature drops down, another situation comes into being (see the corresponding scheme in Fig. 3,*b*). In this case, the probability of thermoactivated electron transition from the surface level into the conduction band becomes small, i.e. the inequality $C_{pt}p_s > C_{nt}n_{1t}$ is fulfilled. In the intermediate temperature range, where $C_{pt}p_s \approx C_{nt}n_{1t}$, the electron concentration on the surface level increases, as the temperature decreases. Accordingly, the surface band bending grows, and the photo-emf changes its sign.

In the case where the depleting band bending is realized at the macropore surface, the temperature dependence of the anomalous photo-emf associated with photo-induced transitions between the valence band and the surface states can be derived by solving the neutrality equation for the surface state and SCR charges in the semiconductor:

$$\begin{aligned}
 & ((\gamma_t I + C_{nt} n_0 \exp y_s) N_t) / (C_{nt} (n_{1t} + n_0 \exp y_s) + \\
 & + C_{pt} (n_i^2 / n_0 + \Delta p) \exp(-y_s) + \gamma_t I) + n_{t0}^* = \\
 & = 2L_D n_0 \sqrt{-y_s}, \quad (5)
 \end{aligned}$$

where n_{t0}^* is the electron occupation of SESs in the absence of illumination, y_s the surface band bending, Δp

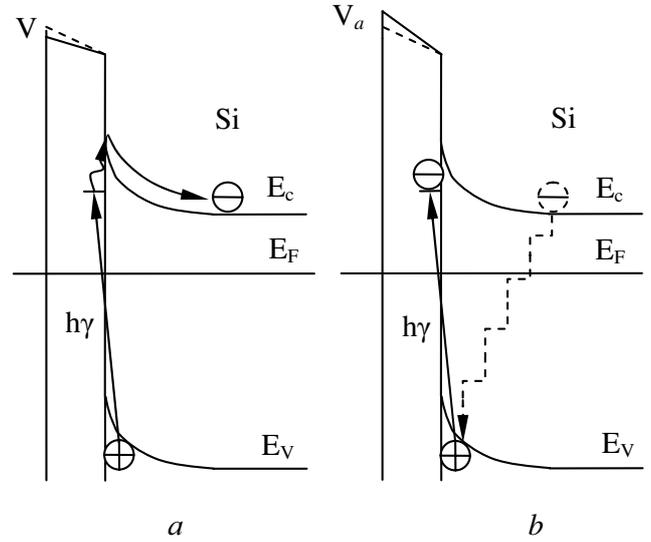


Fig. 3. Scheme of surface level occupation with light-excited electrons at (a) high ($C_{pt}p_s < C_{nt}n_{1t}$) and (b) low ($C_{pt}p_s > C_{nt}n_{1t}$) temperatures

the excess hole concentration in the semiconductor bulk arising owing to an incomplete recombination of equilibrium electrons in the bulk and nonequilibrium holes at the surface, and L_D the Debye screening length.

In order that Eq. (5) be valid in the case $\Delta p \rightarrow 0$, it is necessary to provide that the equilibrium electrons that are located in the vicinity of the SCR boundary could approach the surface and recombine with light-excited holes. This can be done either through the channel of multistage tunnel recombination between the conduction band in the bulk and the valence band at the surface [12], or by means of an exchange between electrons and holes through the external circuit. In the latter case, the time constant $\tau = RC$, where R is the load resistance and C is the SCR capacity, should not be very large. Moreover, when the exchange occurs through the external circuit, it is not, actually, the open circuit situation, but a case where the photo-emf and photo-emf values are rather close. Estimations show that, at large enough R -values, at which the photo-emf and the photo-emf are practically coincide, the value of τ is much less than a second, which allows the signal to be measured under the condition of recharging through the external circuit.

The amplitude of anomalous photo-emf is determined as follows:

$$V_a = \frac{kT}{q} (y_s - y_{s0}), \quad (6)$$

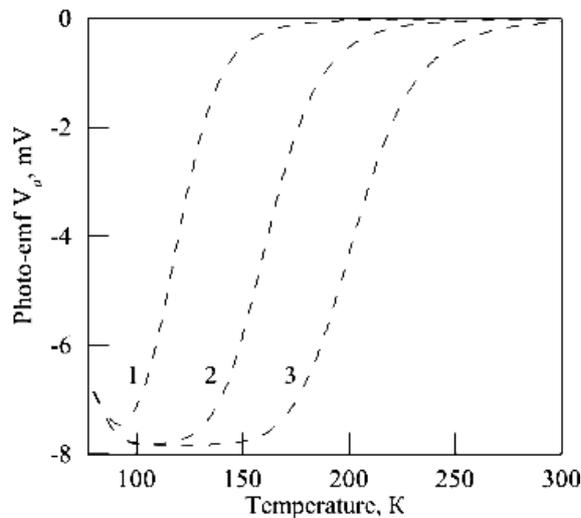


Fig. 4. Theoretical temperature dependences of the anomalous photo-emf V_a associated with phototransitions between the valence band and SESs for various surface level energies $E_t = 0.45$ (1), 0.4 (2), and 0.35 eV (3)

where the quantity y_s is determined from the solution of Eq. (5), and y_{s0} from the solution of the same equation, but at $I = 0$.

In Fig. 4, the theoretical temperature dependences of V_a are exhibited. Here, the control parameter is the surface level depth E_t reckoned from the energy gap middle point. The figure demonstrates that the larger the E_t -value, the lower is the temperature of anomalous photo-emf appearance, which correlates with the degree of SES occupation by electrons. In addition, at temperatures close to 77 K, the amplitude of anomalous photo-emf starts to decrease, which is connected with the inequality $\Delta p \neq 0$. It should be noted that the experimental dependences of the photo-emf on the temperature (Fig. 1) do not demonstrate this feature, i.e. $\Delta p = 0$ for them.

In the general case where the energy of photons is higher than that of the indirect band-to-band transition in silicon, both interband phototransitions and phototransitions with the participation of surface states are observed. The higher the energy of photons, the higher is the probability of interband transitions, whereas the contribution of phototransitions with the participation of surface states to the photo-emf becomes rather small in this case. This situation is realized in macroporous silicon subjected to the light illumination with a wavelength of $0.7 \mu\text{m}$, when the photo-emf does not change its sign. Concerning the contribution of phototransitions with the participation of surface states in macroporous silicon, its growth is favored by two circumstances. The

first is the density of surface states in macroporous silicon, the second is a large area of the macropore surface, which is two orders of magnitude larger than the area of silicon surface.

Let us consider the effective density of surface states in macroporous silicon in more details. Surface states in macroporous silicon that absorb light are mainly located at the walls of macropores. Their number is much larger than the number of surface states at a planar geometrical surface, because the total area of macropores in macroporous silicon is almost two orders of magnitude larger than the geometrical area of the surface. Moreover, the density of surface states in macroporous silicon (i.e. their number per unit surface area) is rather high, being estimated as more than or equal to 10^{12} cm^{-2} [4]. Then, the effective density of surface states calculated with regard for the ratio between the total area of macropores to the geometrical area of the surface can reach 10^{14} cm^{-2} . It is a very large value which substantially exceeds the concentration of surface states determined in work [3], where the anomalous photo-emf at the impurity light absorption was observed experimentally. Therefore, the probability of light absorption by surface states in macroporous silicon has to be considerably larger than the probability of light absorption in a semiconductor with a planar interface.

Several mechanisms describing the emergence of the photo-emf with the anomalous sign in semiconductors and semiconducting structures are known for today. The first one is realized in silicon structures with $p-n$ transitions owing to a non-Ohmic character of rear contacts, the second one in bulk inhomogeneous semiconductors, and the third one consists in the change of the photo-emf sign due to the light absorption by surface levels. While the first two mechanisms are usually realized at room temperature, the third one is active only at temperatures low enough, close to the temperature of liquid nitrogen [3]. In our case, the photo-emf sign change occurred only at temperatures below 130 K, which is typical of the third mechanism.

A qualitative difference between the $V_{OC} > kT/q$ and $V_{OC} < kT/q$ cases consists in that, when the illumination intensity is high and provided that there are photo-induced transitions with the participation of surface states, the occupation of SES by electrons can saturate at a temperature reduction in the former case, because the electron concentration at the surface level cannot exceed the concentration of surface levels themselves. In this case, the anomalous-sign photo-emf connected with phototransitions with SES participation saturates as well. The confirmation of this fact is the ex-

perimental temperature dependences of the photo-emf at the illumination of silicon with light with the wavelength $\lambda = 0.95 \mu\text{m}$ (see Fig. 5). For one macroporous silicon specimen, the photo-emf sign does not change at low temperatures; however, the corresponding photo-emf magnitude becomes extremely low. For another specimen, the photo-emf changes its sign at a temperature of about 130 K. Those dependences can be qualitatively explained, if we consider that the photo-emf is governed by both the interband transitions (they give a normal sign) and the phototransitions between the valence band and SESs (they give an anomalous sign) and admit that the SES concentration in the second specimen is higher. It should be noted that the specific macropore surface in the second specimen is 1.3 times larger in comparison with the corresponding parameter in the first specimen (see the Table). Therefore, the total SES concentration per unit volume of the macroporous structure is higher for the second specimen.

At a lower SES concentration (the first specimen), the saturation of the anomalous-sign photo-emf occurs earlier than the photo-emf saturation in the second specimen with a higher SES concentration. Then, if we suppose that the anomalous-sign photo-emf saturation already takes place in the first specimen, owing to the saturation of the surface level occupation with electrons at the used illumination intensity, and does not occur in the second specimen, the obtained difference between the photo-emf behaviors in those two specimens becomes clear. Since the anomalous-sign photo-emf in the temperature range, where the condition $C_{pt}p_s > C_{nt}n_{1t}$ is satisfied, is higher in the second specimen, the total photo-emf in it must be much lower than that in the first specimen, as is really observed experimentally.

5. Conclusion

The experimental and theoretical temperature dependences of the photo-emf in macroporous silicon in the range $T = 77 - 300$ K have been studied for photon energies comparable with the energy of the indirect band-to-band transition in silicon ($\lambda = 0.7 - 0.95 \mu\text{m}$). In the range lower than 130 K, the experimental temperature dependences of the photo-emf either saturate or change their sign into negative (the anomalous photo-emf).

To explain the photo-emf sign change in macroporous silicon at low temperatures, we considered, besides the interband light absorption, phototransitions with the participation of surface electron states located near the conduction band. In this model, the surface band bending grows due to an increase of the negative charge at the

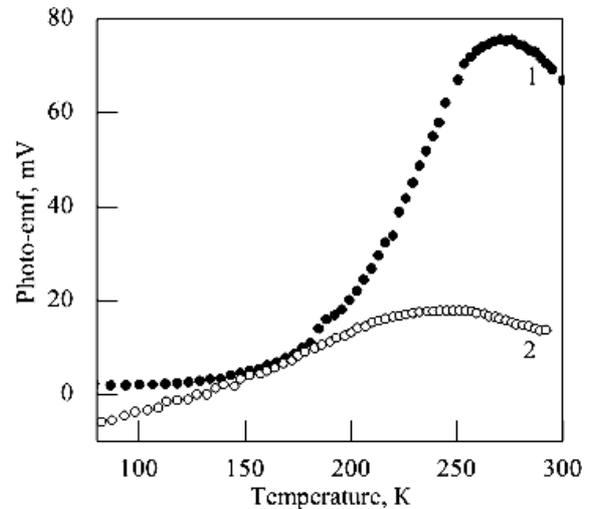


Fig. 5. Experimental temperature dependences of the nonlinear photo-emf in macroporous silicon illuminated with light with the wavelength $\lambda = 0.95 \mu\text{m}$ at the intensity $I = (1 \pm 0.3) \times 10^{16}$ photon/($\text{cm}^2 \cdot \text{s}$) for specimens 2 (curve 1) and 1 (curve 2)

semiconductor surface. Since, the band-to-band light absorption in the semiconductor increases with the growth of the photon energy, the anomalous-sign photo-emf is not observed in macroporous silicon illuminated in the visible spectral range.

In general, the contribution of phototransitions with the participation of surface states in macroporous silicon increases owing to a high concentration of surface states. It is related to a large specific area of macropore surfaces, which is two orders of magnitude larger than the area of the silicon specimen surface. If the temperature decreases, the probability of the thermoactivated electron transition from a surface level into the conduction band diminishes, and the tunnel process dominates. The recombination of equilibrium electrons with holes excited by illumination at the surface occurs in the vicinity of the surface charge region boundary and through the channel of multistage tunnel recombination between the conduction and valence bands, or as a result of the electron-hole exchange through the external circuit.

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ФОТОЕРС АНОМАЛЬНОГО
ЗНАКА В МАКРОПОРИСТОМУ
КРЕМНІЙ ПРИ ЕНЕРГІЯХ ФОТОНІВ, СУМІРНИХ
З ЕНЕРГІЄЮ НЕПРЯМОГО ЗОНА-ЗОННОГО ПЕРЕХОДУ

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

Резюме

Досліджено експериментальні та теоретичні температурні залежності фотоерс у макропористому кремнії для енергій фотонів, сумірних з енергією непрямого зона-зонного переходу в кремнії. Встановлено, що фотоерс у діапазоні температур, менших за 130 К, насичується або змінює знак на від'ємний, що пов'язано з поглинанням світла за участі фотопереходів через поверхневі стани, близькі до зони провідності кремнію. При цьому зростає поверхневий вигин зон за рахунок збільшення від'ємного заряду поверхні напівпровідника. Рекомбінація рівноважних електронів в об'ємі зі збудженими освітленням дірками на поверхні макропор відбувається по каналу багатовипуклої тунельної рекомбінації між зоною провідності та валентною зоною.