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QUASI-GUIDED AND PHOTONIC MODES IN 2D MACROPOROUS SILICON STRUCTURES WITH SiO₂ NANOCOATINGS

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We investigated the IR light absorption oscillations in 2D macroporous silicon structures with SiO_2 nanocoatings 70–800 nm thick. The Wannier–Stark electro-optical effect due to the strong electric field on Si–SiO_2 interface and an additional electric field of quasi-guided optical modes were analyzed. The photonic modes and band gaps were also considered as peculiarities in absorbance spectra of macroporous silicon structures with a thick SiO_2 nanocoating.

INTRODUCTION

Macroporous silicon is a promising material for development of 2D photonic structures with required geometry and large effective specific surface [1, 2]. This determines optical and electro-optical characteristics of macroporous silicon structures [3–7]. With periodical arrangement of cylindrical macropores, the material is a photonic crystal and has photonic bandgap that reacts to variation of permittivity in macropores. In view of the potential barrier on a macropore surface, one should take into account recharging of the local surface centres at the energies below that of the indirect interband transition. The near-IR optical absorption in 2D photonic macroporous silicon structures was investigated in [6], with allowance made for the linear electro-optical effect. The experimental absorption spectra of macroporous silicon agree well with the corresponding spectral dependences of the electro-optical energy and the imaginary part of permittivity in the weak electric field approximation, thus confirming realization of the impurity Franz–Keldysh effect. In [7] we investigated the near-IR light absorption oscillations in 2D macroporous silicon structures with microporous silicon layers and CdTe, ZnO, CdS surface nanocrystals taking into account the electro-optical effect within the strong electric field approximation. The model [8, 9] of the resonance electron scattering on impurity states in an electric field of «silicon–nanocoating» heterojunction on macropore surface and realization of the

Wannier–Stark effect on randomly distributed surface bonds were confirmed. In this case, the Wannier–Stark effect is realized as a result of large-time electron scattering as compared with the period of its oscillations in the strong electric field of «silicon-nanocoating» interface.

In this paper the near-IR light absorption oscillations of 2D macroporous silicon structures with SiO₂ nanocoatings of 5–800 nm thick were investigated taking into account the Wannier–Stark electro-optical effect due to the strong electric field on Si–SiO₂ interface and an additional electric field of quasi-guided modes. The photonic modes and band gaps were considered as peculiarities in absorbance spectra of macroporous silicon structures with a thick SiO₂ nanocoating. A comparison of the results obtained for SiO₂ nanocoatings and II–VI nanocrystal surface was made.

PROCEDURE

The samples to be studied were made of silicon wafers characterized by the (100) orientation and n-type of conductivity (the electron concentration $n_0 = 10^{15}$ cm⁻³). We used the technique of electrochemical etching at illumination of the back side of a silicon substrate (thickness $H = 520\,\mu m$) [3]. The square-lattice periodic structures with macropore depth $h_p = 90$–$100\,\mu m$, diameter $D_p = 2\,\mu m$ and concentration $N_p = 5 \times 10^6$ cm⁻² were formed. The initial specimens are complex micropore-macropore silicon structures consisting of 100 nm micropore

layers on macropore walls. An additional anisotropic etching in 10 % aqueous solution of KOH was used to remove the microporous layers from macropore walls.

SiO$_2$ nanocoatings were formed in the diffusion stove in the nitrogen atmosphere. The layers of oxide (thickness of 5–200 nm) have been formed on macroporous silicon samples in dry oxygen during 40–60 minutes at a temperature of 1050–1200 °C. The 800 nm oxide was formed for 50 minutes at the temperature of 1100 °C in wet oxygen using a steam generator with deionized water. The oxide thickness was measured using ellipsometry.

The chemical states on the surface of macroporous silicon structures with nanocoatings and the electric field at the Si–SiO$_2$ interface were identified by IR absorption using a Perkin-Elmer Spectrum BXII IR Fourier spectrometer. The optical absorption spectra were recorded at normal incidence of IR radiation on the sample (along the main axes of cylindrical macropores – see Fig. 1). The experiments were carried out at room temperature in air. The error of spectral measurements was about 2 cm^{-1}.
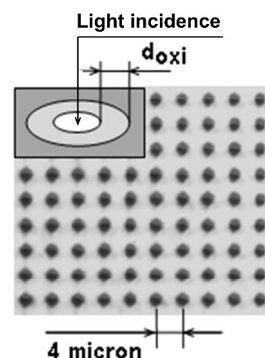

Fig. 1. 2D macroporous silicon square-lattice structure with period $a = 4$ µm. Insertion: fragment of the cylindrical macropore with SiO$_2$ nanocoating and direction of light incidence on the sample (along the main axis of cylindrical macropore)

EXPERIMENTAL

For macroporous silicon structures with SiO$_2$ nanocoatings more 10 nm thick, the light absorption increases and an oscillating structure occurs (Fig. 2 *a*, curves 1 and 2) in analogy to macroporous silicon structures with surface nanocrystals [7]. We observed an essential absorption growth in the spectral region of Si–O, Si–H, O–H bonds and organic compounds. The amplitude of oscillations is maximal in the spectral ranges of surface level absorption (Fig. 2 *b*).
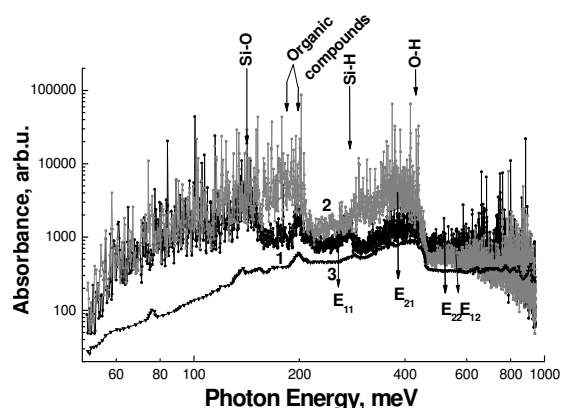

Fig. 2 *a*. Absorption spectra of macroporous silicon structures with SiO$_2$ nanocoating: 70 (1) and 800 nm (2) thick and without coating (3)

Fig. 2 *b*. Fragment of absorption spectra of macroporous silicon structures with: (1) SiO$_2$ nanocoating, (2) ZnO nanocrystals from [7] and (3) without coating

The plots of oscillation maxima of macroporous silicon structures with SiO$_2$ nanocoating on oscillation number have bends (Fig. 3 *a*) at the energies of 250 (curve 1), 400 (curve 2), and 700 meV (curve 3). And the oscillation period fluctuates about a constant value at low spectral energies and becomes quadratic at
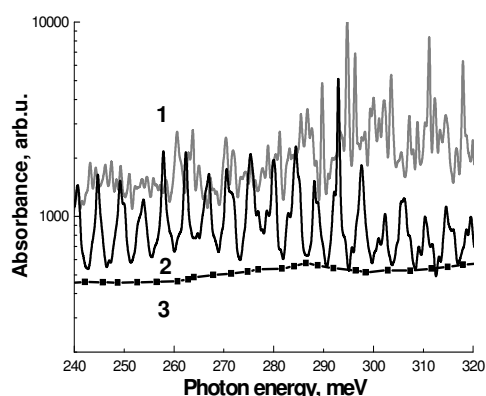
photon energies depending on SiO$_2$ nanocoating thickness (Fig. 3 *b*).

In addition, we observed (Fig. 4) the peculiarities at photon energies of 100–200 meV and 220–480 meV in the absorption spectra of macroporous silicon structures with SiO$_2$ nanocoating with thickness of 800 nm. The

peculiarities are similar to those observed in reflectance spectra of macroporous silicon photonic crystals at near-normal (5°) light incidence [10]. The critical points $E_0 = 110$ meV and $E_{02} = 220$ meV in absorbance may be related to singularities in

diffracted intensity $D_i(\omega) = C_i(h\omega - E_{0i})^{-1/2}$ due to a photonic modes excitation as an «absorption» process which includes intensity of diffracted beam: $A = A_{0i} + (D_i)^{-1}$.
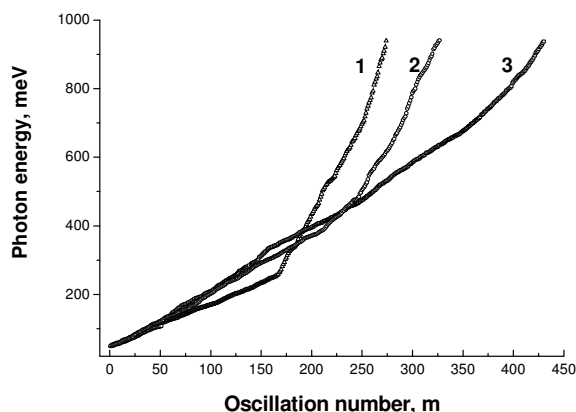

Fig. 3 *a*. The spectral position of oscillation maxima in the macroporous silicon structures with SiO_2 nanocoatings of 70 (1), 200 (2) and 800 nm (3) thickness
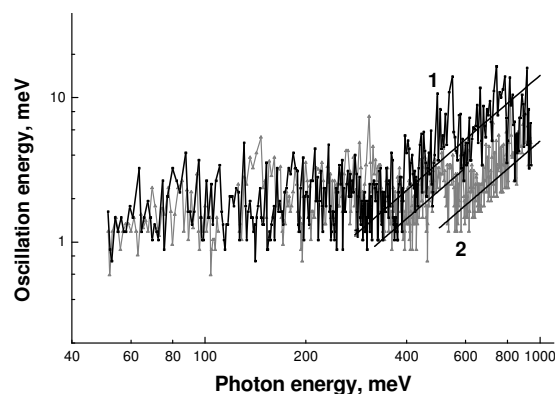
Fig. 3 *b*. Spectral dependencies of the oscillation energy in the macroporous silicon structures with SiO_2 nanocoatings of 200 (1) and 800 nm (2) thickness
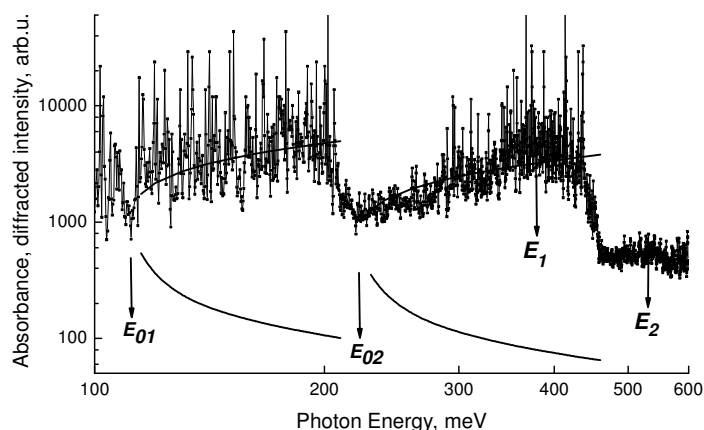

Fig. 4. Absorption spectra of macroporous silicon structures with SiO_2 nanocoating of 800 nm thickness: E_1 and E_2 – the minimal energies of the quasi-guided mode formation; E_{01} and E_{02} – critical points of peculiarities in absorption spectra of macroporous silicon structure. Below: spectral dependence of the diffracted beam intensity $D_i(\omega) \sim (h\omega - E_{0i})^{-1/2}$

DISCUSSION

Wannier–Stark ladders. We observed the giant oscillations in absorption spectra of macroporous silicon structures with SiO_2 nanocoatings of 10–800 nm thickness. The amplitude of oscillations is maximal in the spectral ranges of surface level absorption. The data obtained indicate a strong effect of impurity states on the surface of macroporous silicon structures with SiO_2 nanocoatings. This may result from

scattering of both electromagnetic radiation and electrons on the impurity states. The form of oscillations (Fig. 2 *b*) indicates resonant character of scattering. We observed well-separated oscillations in the spectral ranges of the surface bond absorption in the absorption spectra of macroporous silicon structures with surface nanocrystals [7] and explained by the realization of the Wannier–Stark effect on randomly distributed surface bonds. A method of experimental

observation of Wannier–Stark ladders was proposed in [8]. It was shown that the scattering amplitude had resonant behavior in the case of electron scattering on impurities. If the electric field is directed along the *x*-axis of the crystal, then electron scattering occurs in the plane (y, z), and the difference between two resonant energies is approximately equal to the Wannier–Stark ladder.
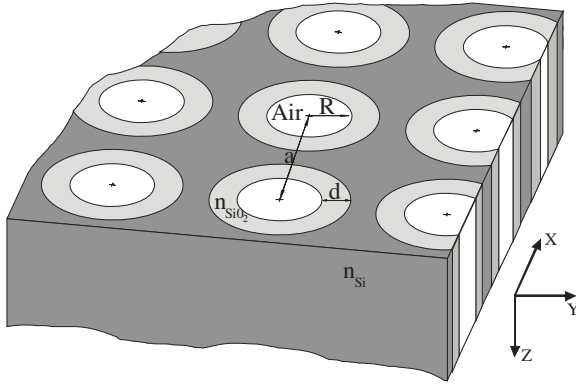
Fig. 5. The fragment of the system considered

In our case, an electric field of «silicon-nanocoating» heterojunctions on the macropore surfaces is directed perpendicularly to the surface too (Fig. 5), and surface states that scatter electrons are concentrated perpendicularly to the *x*-direction in the plane (y, z) that is the plane of resonant scattering.

Let us consider a semiconductor with the dispersion law $E(k) = E_0 - \Delta (cosk_y a + cosk_z a)$, where k is a quasi-momentum with components k_y, k_z, E_0 is the energy corresponding to the midgap, Δ is the energy equal to 1/6 the band gap, *a* is the lattice parameter. The wave function in the Wannier representation was written as [8]:

$$\langle j|\psi_E\rangle = \langle j|\Phi_E\rangle + \frac{\langle j|\hat{G}_0(E)|0\rangle V_0\langle 0|\Phi_E\rangle}{1-V_0\langle 0|\hat{G}_0(E)|0\rangle}. \quad (1)$$

Here the first (second) term describes the incident wave (scattered waves); j – the number of the lattice site, $\hat{G}_0(E)$ is the Green operator, V_0 is the impurity potential. The complex energies for which the denominator of the second term becomes zero correspond to the resonances in electron scattering

$$1/V_0 = \langle 0|\hat{G}_0(E)|0\rangle$$

at $E = \varepsilon - i\Gamma$ (Γ>0). The difference of two neighboring resonance energies ΔE is approximately equal to the value of the step Fa in

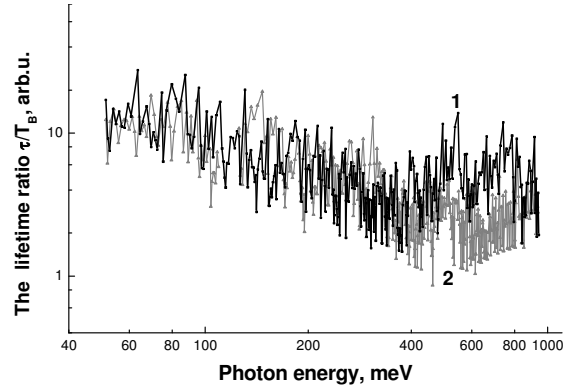the Wannier–Stark ladder for electrical field strength *F*.

Fig. 6. Spectral dependence of the lifetime ratio τ/T_B for macroporous silicon structures with SiO₂ nanocoatings of 200 (1) and 800 nm (2) thickness

The fact is that the levels of the Wannier–Stark ladder have a certain width Γ, while its detection requires that this width be less than the difference of energies of adjacent levels, $\Gamma < Fa$. [9]. The Wannier–Stark ladder is not broken by impurities if the intervals between the transitions due to scattering from impurity atoms with lifetime τ are greater than the period of electron oscillations in external field, T_B ($\tau/T_B > 1$), where $T_B = 2\pi h/Fa$, τ is equal to $1/W$ (W is the probability for an electron to leave the state per unit time due to scattering from an impurity atom at lattice site). In [9] the following estimate of the probability W for an electron to leave the state per unit time due to scattering from an impurity atom at lattice site was obtained: $W < 2V_0 Ni/(Nh)$, where V_0 is the impurity potential, N_i the impurity concentration and $N \approx (a^2)^{-1}$ the density of states. As a result, the inequality $\tau/T_B > 1$ passes to $N_i < \Delta E/(4\pi a^2 V_0)$. Using the last inequality, we find a numerical estimate of impurity concentration.

The surface state concentration in macroporous silicon structures changes from 10^{10}cm^{-2} to 10^{11}cm^{-2} [11] and $N_i^{max} > 10^{11}$ cm^{-2} for the investigated spectral range. Spectral dependence of the lifetime ratio τ/T_B for macroporous silicon structures with SiO₂ nanocoatings of 200 (1) and 800 nm (2) thickness are presented on (Fig. 6). The inequality $\tau/T_B > 1$ for the lifetime ratio is satisfied in the whole spectral region investigated for macroporous silicon structures with SiO₂ nanocoatings taking into account that the surface impurity concentration for macroporous silicon structures, N_i, is less than 10^{11} cm^{-2}.

Quasi-guided modes. Usually, the basic sources of external electric field at a semiconductor surface are the charge of the electron levels and the built-in charge in the semiconductor surface oxide [12]. Spectral dependences of the electric field strength F on macroporous silicon surface for structures with SiO_2 nanocoatings of 200 and 800 nm thickness are presented in Fig. 7 (curves 1–2). The oscillation period and electric field strength of macroporous silicon structures with SiO_2 nanocoatings fluctuate about a constant value at low spectral light energies and have quadratic low at high spectral light energies (Fig. 7, curves 3–4).
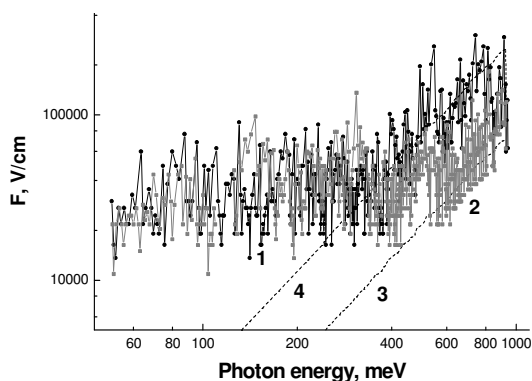

Fig. 7. Spectral dependences of the electric field strength F on macroporous silicon surface for structures with SiO_2 nanocoatings of 200 (1) and 800 nm (2) thickness; spectral dependencies of the electric field strength as a result of quasi-guided mode formation in the silicon matrix (3–4)

The electric field strength F becomes quadratic at light wavelengths equal to the geometrical sizes of silicon matrix and SiO_2 nanocoatings (Fig. 7, curves 3–4 and Table). Such dependence corresponds to the quasi-guided mode formation [13] in the silicon matrix (minimal distance between macropores) with $2\rho_{Si} = a - (D_p + d_{SiO_2})$ and in silicon column with $2\rho_{Si} = 1.4[a - (D_p + d_{SiO_2})]$. Comparison of the sample and quasi-guided mode characteristics in Table has confirmed that the mode parameter $Q_{Si} \sim k\rho_{Si}$ [13] is determined by the beginning of the electric field strength quadratic growth in Fig. 7.

In general, at grazing angle of light incidence, the electric field of the reflected electromagnetic wave changes the local electric field in the near-surface region of the macrospores walls with thickness $d \approx 0.1\lambda$ for wavelength λ [14]. Let us consider that d is determined by the electric component of electromagnetic wave with $\hbar\omega$ and by the change of built-in electric field ΔF_s ($d = \hbar\omega/(e \cdot \Delta F_s)$). Indeed, under our experimental condition of the grazing angle of light incidence onto the macropore surface, the electric field strength on macroporous silicon surface for structures with SiO_2 nanocoatings is about $F_s + \Delta F_s$, with $\Delta F_s \approx \hbar\omega/(0.1\lambda_l) \sim \hbar\omega^2$ according to the experiment (Fig. 7). Light wavelength is equal to $\lambda_l = \lambda/n_i$ (n_i is effective refractive index of pores with SiO_2 nanocoatings or refractive index of SiO_2 nanocoatings).

Table. Structure characteristics and quasi-guided mode parameters

d_{SiO_2}, nm	$D_p + d_{SiO_2}$, μm	$a - (D_p + d_{SiO_2})$, μm	Mode's type	$2\rho_{Si}$, μm
70	2.07	2.4	Quasi-guided modes in the silicon matrix	$2[a - (D_p + d_{SiO_2})] = 4.8$ (0.26 eV)
			Quasi-guided modes in the silicon matrix	$[a - (D_p + d_{SiO_2})] = 2.2$ (0.56 eV)
200	2.2	2.27	Quasi-guided modes In the silicon matrix	$2[a - (D_p + d_{SiO_2})] = 4.1$ (0.30 eV)
			Quasi-guided modes in the silicon column	$1.4[a - (D_p + d_{SiO_2})] = 3.65$ (0.34 eV)
800	2.8	1.67	Quasi-guided modes in the silicon matrix	$2[a - (D_p + d_{SiO_2})] = 3.25$ (0.38 eV)
			Quasi-guided modes in the silicon column	$1.4[a - (D_p + d_{SiO_2})] = 2.34$ (0.53 eV)

Photonic modes. The peculiarities at photon energies 100-200 and 220–480 meV in absorption spectra of macroporous silicon structures with SiO_2 nanocoating of 800 nm thickness (Fig. 4) are similar to those observed in reflectance spectra of macroporous silicon photonic crystals at the near-

normal (5°) light incidence [10]. Such anomalies in absorbance of macroporous silicon structures are due to matching of the external field with a Bloch state, which behaves like a one-dimensional (1D) critical point, with density of states given by photon dispersion in the vertical direction along macropores only, whereas the in-plane momentum and the parallel vector k are conserved. Onset of a diffracted beam corresponds to a complex wave-vector component q that passes through zero and becomes real [15]. The out-of-plane dispersion of all bands is quadratic in q around q = 0 [16], with a threshold at $E(\omega,q) = E_0$ and a diffracted intensity $D(\omega) \sim (h\omega - E_0)^{-1/2}$, like for a 1D density of states. A structure in absorbance (Fig. 4) marks the onset of a photonic mode that is excited and remains propagating for higher frequencies [10].

The diffracted intensity $D_i(\omega)$ is removed from the absorbance (Fig. 4), that has the form of the square root at $E(\omega,q) > E_0$, proving that the measured features in absorbance correspond to the 1D critical point [17], and the calculated diffracted intensities of allowed modes at normal incidence have the form of the inverse square root close to thresholds $E_{01} = 110$ meV and $E_{02} = 220$ meV. Such bands do not appear in absorption curves of macroporous silicon structures with SiO₂ layers of the less thickness (Fig. 2 *b*). They may have allowed bands, but very weak spectral strength.

The threshold energies E_{01} and E_{02} (Fig. 4) correspond to normalized frequencies $\omega a/2\pi c = 0.36$ and 0.71. The results obtained are similar to calculations for 2D square lattice composed of circular air-rods in dielectric material with dielectric constant 2.1 and filling factor 0.25 [18]. There are two band gaps, one of then being between the first and the second 1D eigenmodes (A modes), and another between the second and third A modes. The normalized frequency ranges of this band gaps are 0.36–0.39, 0.71–0.73 for E-polarization and 0.36–0.40, 0.73–0.74 for H-polarization. Our structure is three-component one, with averaged refractive index 1.4, and the structure from [18] is two-component, with averaged refractive index 1.3. And pronounced structure observed in experimental reflectance spectra at near-normal (5°) light incidence on macroporous silicon structure with averaged refractive index 3 around higher normalized frequency 0.47 [10]

corresponded to the allowed band with symmetry Γ_5^-.

The quasi-guided mode formation in the silicon matrix at energies $E_1 = 0.38$ eV and $E_2 = 0.53$ eV (Table) does not appear in absorption spectra (Fig. 2 *a* and Fig. 4) and does not coincide with the photonic modes as peculiarities in absorbance (and reflectance) spectra of macroporous silicon structures. The quasi-guided and photonic modes do not appear in absorption spectra of 2D macroporous silicon structures with microporous silicon layers and CdTe, ZnO surface nanocrystals [7].

CONCLUSIONS

The near-IR light absorption oscillations in the 2D macroporous silicon structures with SiO₂ nanocoatings of 5–800 nm thickness have been investigated taking into account the electro-optical effect within the strong electric field approximation. We observed the oscillating structure in the absorption spectra of macroporous silicon structures with SiO₂ nanocoatings. The amplitude of oscillations is maximal in the spectral ranges of surface level absorption as a result of the resonance electron scattering on impurity states with the difference between two resonance energies equal to the Wannier–Stark ladder.

The oscillation period and electric field strength in the macroporous silicon structures with thick SiO₂ nanocoatings fluctuate about a constant value at low photon energies and become quadratic in photon energy, depending on the geometrical sizes of silicon matrix and SiO₂ nanocoatings. The relevant electric field strength growth corresponds to the quasi-guided mode formation in the silicon matrix (minimal distance between the macropores) and in the silicon column.

The peculiarities in absorption spectra of macroporous silicon structures with thick SiO₂ nanocoating are similar to those observed in reflectance spectra of macroporous silicon photonic crystals at near-normal light incidence. The critical points in absorbance are related to photonic modes excitation as an «absorption» process that includes intensity of diffracted beam. Photonic modes do not coincide with quasi-guided modes in the silicon matrix and do not appear in absorption spectra of 2D macroporous silicon structures with surface nanocrystals.

Квазі-направлені та фотонні моди в двовимірних структурах макропористого кремнію з нанопокриттями SiO₂

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Досліджені осциляції ІЧ поглинання світла у двовимірних структурах макропористого кремнію з нанопокриттями SiO₂ завтовшки 70–800 нм. Проаналізовано електрооптичний ефект Ваньє-Штарка в сильному електричному полі на межі Si–SiO₂ і додаткове електричне поле квазі-направлених оптичних мод. Розглянуто також фотонні моди та заборонені зони як особливості в спектрах поглинання двовимірних структур макропористого кремнію з товстим нанопокриттям SiO₂.

Ключові слова: *макропористий кремній, нанопокриття SiO₂, квазі-направлені та фотонні моди*

Квази-направленные и фотонные моды в двумерных структурах макропористого кремния с нанопокрытиями SiO₂

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Исследованы осцилляции ИК поглощения света в двумерных структурах макропористого кремния с нанопокрытиями SiO₂ толщиной 70–800 нм. Проанализированы электрооптический эффект Ваньє-Штарка в сильном электрическом поле на границе Si–SiO₂ и дополнительное электрическое поле квази-направленных оптических мод. Рассмотрены также фотонные моды и запрещенные зоны как особенности в спектрах поглощения двумерных структур макропористого кремния с толстым нанопокрытием SiO₂.

Ключевые слова: *макропористый кремний, нанопокрытие SiO₂, квази-направленные и фотонные моды*

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