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1D AND 2D POLARITONS IN MACROPOROUS SILICON STRUCTURES WITH NANO-COATINGS

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In this paper, we used high-resolution IR absorption spectra to investigate 1D and 2D polaritons in periodical 2D macroporous silicon structures with nano-coatings of SiO_2 and CdS, ZnO nanoparticles. The application of high-resolution IR absorption spectroscopy resulted in detection of dipole-active TO vibrations, photon splitting and giant two-polar absorption oscillations with amplitudes of $\pm 10^7$arb.un. As a result, the dispersion law in yz surfaces of macropores change to z direction along macropores. It means additional degree of freedom as vertically polarized light in z direction and horizontally polarized light in x direction resulted in beams splitting and two-photon interference - Hong-Ou-Mandel effect. In our case, 2D resonances of Wannier-Stark electro-optical effect in yz plane correspond to constructive interference of the two-photon states (bosonic behavior), and two-polar resonances in ±z direction are determined by destructive interference of the two-photon states (fermionic behavior). Two-polar oscillations of 1D -polaritons have the ultra-small half-width 0.4–0.6 cm^{-1} and minimal Rabi frequency of samples 1.0 cm^{-1} equaled to the resolution of spectral measurements. Furthermore, two-photon interference and 1D polaritons are perspective for high-coherent optical quantum computers on macroporous silicon with nano-coatings and, in addition, for lasers and new metamaterials.

***Keywords**: macroporous silicon, nano-coatings, 1D and 2D polaritons, beams splitting, two-photon interference*

INTRODUCTION

Macroporous silicon structures prepared by photoanodic etching are one of the promising materials for the development of 2D photonic crystals [1, 2] and for the measuring of photonic band gaps [3–6]. Presence of periodically located cylindrical macropores provides large effective macropore surface and enhanced photo-physical characteristics of macroporous silicon [7–9]. Franz-Keldysh oscillations in the electro-reflectance spectra of macroporous silicon structures without nanocoatings in the area of direct band-to-band transition were measured [10]. At the wavelengths, compared with the optical period of the macroporous silicon structures, the measured IR absorption is two orders of magnitude higher of the silicon single crystal. The formation of steps and the change in optical absorption by the 3/2 law confirmed realization of the impurity Franz-Keldysh effect [11]. In addition, for 2D macroporous silicon structures with coatings of surface nanocrystals, oscillations of IR absorption increase to maximal amplitudes in the spectral region of surface states [1]. The dependence of the oscillation maxima on its number is linear with a constant period. The analysis of IR absorption oscillations was performed within the model of resonant scattering of electrons on surface states in a strong electric field with a difference between two resonant energies equal to the Wannier-Stark step $\Delta E = Fa$ (F – the electric field strength, and a– the silicon lattice constant) [1]. The shift and deviations of oscillation peaks, the broadening parameter Γ of the Wannier–Stark ladders and the influence of "quantum superiority" on coherence of Wannier levels were analyzed in [2]. In addition, we proposed a high coherent optical quantum computer based on a silicon matrix with macropores and a layer of nanocrystals on the macropore surface for the implementation of Wannier-Stark quantum electro-optical effect [12].

In this work, we investigated high-resolution optical absorption spectra of 2D macroporous silicon structures with nano-coatings. We performed a comparative analysis of the results on electro-optical effects in macroporous silicon of 1D and 2D exciton-polaritons on macropores**.** Here we achieved excellent parameters of 1D

exciton-polaritons at room temperature for high-coherent optical phenomena implementations in quantum computers, lasers and metamaterials.

PROCEDURE

The samples to be studied were made of silicon wafers with thickness H = 520 μm, resistivity of 4.5 Ω·cm, 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 the backside illumination of a silicon substrate (thickness H = 520 μm) [13, 14]. Macropores were etched in the form of a square lattice of parallel air cylinders with diameter Dp = 2±0.2 μm, period 4 μm, depth hp = 50÷100 μm, and concentration $Np = 6.25 \cdot 10^6$ cm^{-2} (Fig. 1). Addition anisotropy etching in 10 % solution of KOH permits to remove microporous layers from the macropores surface.

SiO_2 nano-coatings were formed in the diffusions to ve after treatment of macroporous silicon substrates in the nitrogen atmosphere [15]. The oxide layers (thickness of 10 and 20 nm) were obtained on macroporous silicon samples in dry oxygen during 40÷60 min at the temperature of 1050 °C. The oxide thickness was measured using ellipsometry.

CdS nanoparticles of 1.8–2 nm in size [16] were deposited on the oxidized surface of macropores from the colloidal solutions in polyethyleneimine (PEI). Method of synthesis of ZnO nanoparticles in isopropanol and from solution of zinc acetate of $Zn(CH_3COO)_2$ in ethanol have been developed in [17].

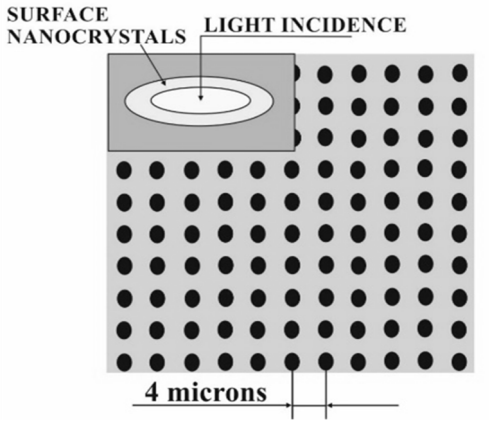


Fig. 1. Scheme of two-dimensional macroporous silicon structures with layer ncCdS+SiO_2; macropore diameter D_p = 2 μm and period 4 μm; *insertion*: normal incidence of IR radiation on a sample (along the pores)

Table shows the parameters of nano-coatings on 2D macroporous silicon structures. We performed optical investigations within the spectral range 300…7800 cm^{-1} by using an IR Fourier spectrometer "PerkinElmer" (Spectrum BXII). High-resolution optical absorption spectra were measured in the 200÷8500 cm^{-1} spectral range on a spectrometer BrukerVertex70Vwith a resolution of 1 cm^{-1}.

The optical absorption spectra were measured at normal incidence of IR radiation on a sample (along the cylindrical macropores, Fig. 1, insertion) in the air at room temperature.

Table. The parameters of nano-coatings on 2D macroporous silicon structures and splitting of surface modes

No of sample	Boundary "Si – nanocoating"	Thickness d_1 of SiO_2 layer, nm	Thickness d_2 of "PEI-Nclayer", nm	d/λ	$(d/\lambda)^{1/2}$	Splitting of surface modes, cm^{-1}	Rabi frequency (cm^{-1})
1	Si-SiO_2+"PEI-ncCdS"	10	16	$6.25 \cdot 10^{-4}$	0.0025	–	1.0
2	Si-SiO_2+"PEI-ncCdS"	20	16	$1.25 \cdot 10^{-3}$	0.0353	6	1.0
3	Si-SiO_2+"PEI-ncCdS"	20	28	$3.0 \cdot 10^{-3}$	0.055	14	1.0
4	Si-SiO_2+"PEI-ncZnO"	20	28	–	–	–	1.0

EXPERIMENT AND DISCUSSION OF RESULTS

Figures 2 *a*, *b* show comparison of the absorption spectra of the structure #1: after changing resolution of measurement from 2 cm^{-1} (curves 1) to 1 cm^{-1} (curves 2) the period of oscillations in optical absorption spectra decreases by 3 times and the optical absorption increases by 60–100 times.

Figures 3 *a*, *b* shows fragments of IR optical absorption spectra of structure #1 (Table) with a resolution of 1 cm^{-1}. Giant two-polar oscillations

of the absorption appear in the spectral region of Si-Si bonds and in the 5500–7500 cm^{-1} spectral region of P_b centers [15, 19].

Si-Si-bonds represent the transverse fluctuations of silicon atoms on the surface of the macropores, *i.e.*, transverse phonons [19]. Fig. 3 *a* and Fig. 4 *a*, *b* show fragments of the absorption spectra of 2D macroporous silicon structures with nano-coatings from Table in the spectral region of Si-Si-bonds: Fig. 3 *a* includes one minimum for sample #1, Fig. 4 *a* includes two minima for sample #2, and Fig. 4 *b* includes three minima for sample #3. Samples #1 and #2 differ in the thickness of the SiO_2 layer on the macropore surface (Table).

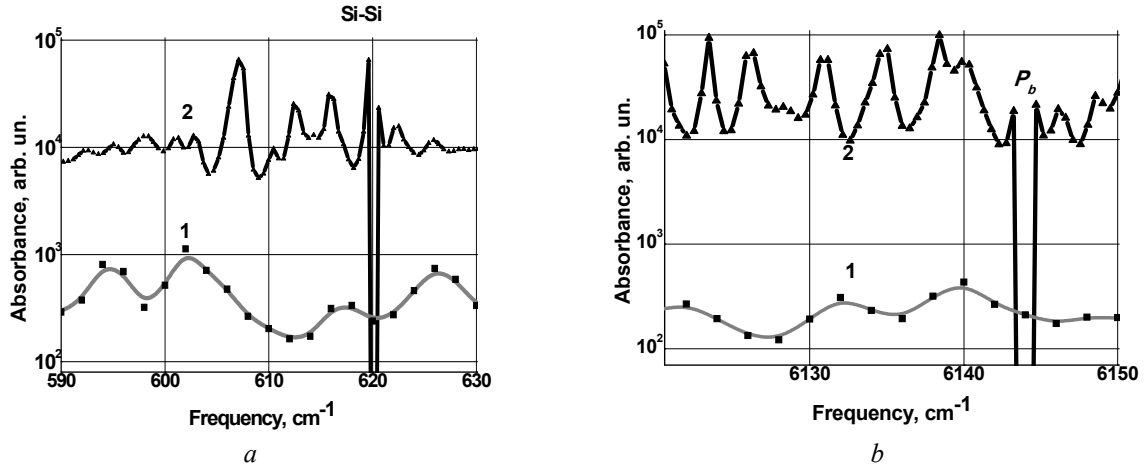


Fig. 2. Fragments of the absorption spectra of the structure #1 for the resolution of measurements 2 cm^{-1} (curves 1) and resolution 1 cm^{-1} (curves 2) in the spectral ranges: *a* – 590–625 cm^{-1}; *b* – 6130–6150 cm^{-1}

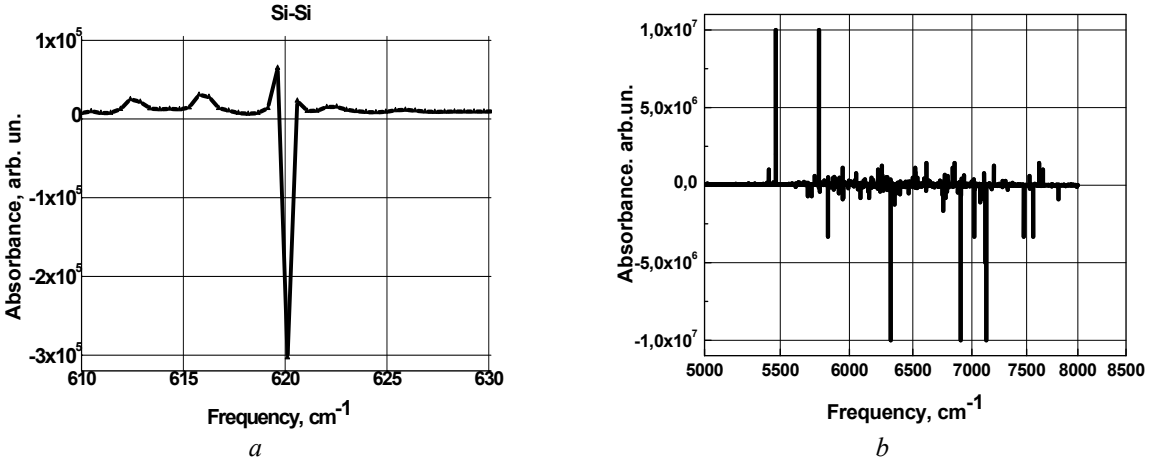


Fig. 3. The optical absorption by 2D structures of macroporous silicon with nano-coatings for sample #1 from Table measured at resolution of 1 cm^{-1}: *a* – at spectral region of Si-Si-bonds; *b* – at spectral region of P_b centers

At thickness of the SiO_2 layer d_1 = 10 nm the surface polaritons are formed at the frequency ω_S (Fig. 3 *a*) equal to the frequency of transverse phonon ω_{TO} = 620 cm^{-1} [19]. When the SiO_2 thickness is d_1 = 20 nm on the boundary "SiO_2–macropore surface", the splitting of mode is formed (Fig. 4 *a*). The thickness of "PEI-ncCdS" layer increase to d_2 = 28 nm for sample #3 (Table). Thus additional modes and the additional surface polaritons are formed for sample #3 (Fig. 4 *b*) due to resonance interaction of dipole-active oscillations with surface polaritons.

In general, the interaction of an external electromagnetic field with dipole-active states leads to polariton formation [20, 21]. These resonances lead to the splitting of surface polaritons and to the appearance of the slits in its spectrum with width proportional to $(d/\lambda)^{1/2}$ [20].

Table shows minimal value of $d/\lambda = (0.625–3)\cdot 10^{-3} << 1$ and small splitting of surface polariton modes, equaled to 6–14 cm^{-1} for structures #2 and #3 (Fig. 4 *a*, *b*). It is proportional to $(d_2/d_3)^{1/2}$, where d_2 = 20 nm and d_3 = 20+28 = 48 nm. So, for 2D macroporous silicon structures with nano-coatings surface polaritons are formed at boundary Si-SiO_2 for samples #1 (Fig. 3 *a*), and at boundaries Si-SiO_2 and SiO_2+"PEI-ncCdS" for samples #2 and #3 (Table), The results obtained are fully consistent with data for phonon polaritons in microresonators [20].

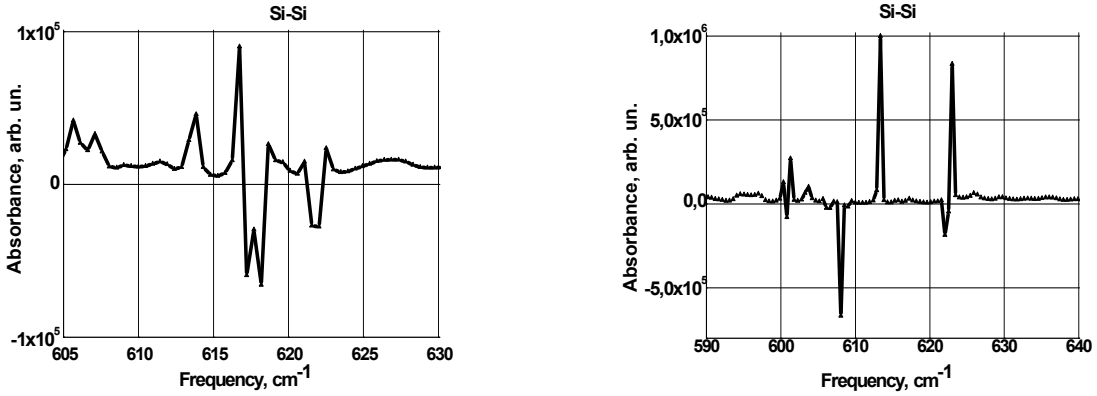


Fig. 4. The optical absorption by 2D structures of macroporous silicon with nano-coatings: *a* – sample #2, *b* – sample #3 from Table measured at resolution of 1 cm^{-1} near Si-Si-bonds

Measurements the giant two-polar oscillations with very small half-width 0.5 $cm^{-1}$and Rabi frequency 1.0 cm^{-1} (Table) testify the strong interaction of surface polaritons with photons. Moreover, by changing the thickness of the nano-coatings, it is possible to achieve a match in the frequency of interference modes with frequencies of slit oscillations of surface bonds on boundaries Si-SiO_2 and SiO_2+"PEI-ncCdS". When the frequencies of local oscillations of surface bonds and slit modes matched, then the light absorption increases up to 10^5 times on the frequencies of slit oscillations of surface bonds [21].

Earlier we observed the oscillating structure in the absorption spectra of macroporous silicon structures with surface nanocrystals [1, 2]. The form of oscillations and constant oscillation period indicate the resonant character of scattering (the Wannier–Stark electro-optical effect). According to method of experimental observation of Wannier–Stark ladders [22], the scattering amplitude has resonant behavior at electron scattering on impurities.

The wave function in the Wannier representation is:

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

Here the first (second) term describes the incident wave (scattered waves); *j*– numbers the lattice site, $\hat{G}_0(E)$ is the Green operator, V_0 is the impurity potential. The complex energies of resonances in electron scattering are

$$1/V_0 = \langle 0|\hat{G}_0(E)|0\rangle \qquad (2)$$

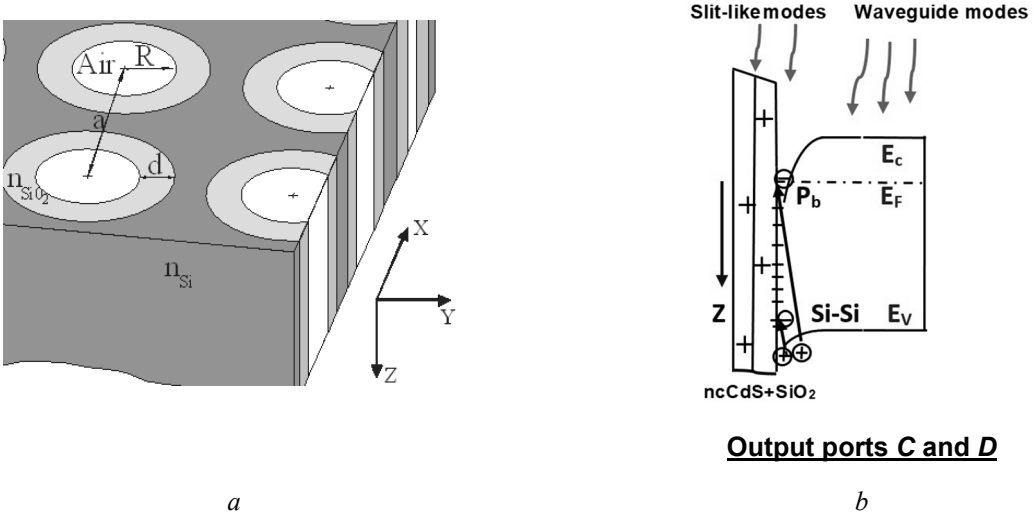
at $E = \varepsilon - i\Gamma$ (Γ>0). The difference of two neighboring resonance energies is equaled approximately to the value of the step in the Wannier–Stark ladder. The dispersion law is considered in form:

$$E(\kappa) = E_0 - \Delta\,(cos\ k_y a + cos\ k_z a), \qquad (3)$$

where k is a quasi-momentum with components k_y, k_z, E_0 is the energy corresponding to the midgap, Δ – the energy equal to 1/6 of the band gap, a – Si lattice parameter.

In our case, an electric field of "silicon-nanocoating" heterojunctions on the macropore surface is directed perpendicularly to the macropore surface (Fig. 5 *a*), 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. After illumination, electrons are accelerated in the

electric field of the enriched electric potential (Fig. 5 *b*), oscillate and scatter by surface states in the radial direction *x* relative to the macropore, in the plane (*y, z*) (Fig. 5 *a*), which is the plane of resonance scattering with infinity amplitude [22]. It results in additional vertically polarized light in *z* direction and horizontally polarized light in *x* direction and the infinity growth of dielectric constants and corresponding changes in IR absorption at room temperature.



a *b*

Fig. 5. *a* – a fragment of the macroporous silicon structure with the nanocoating with the radius of macropore *R* and the surface nanocoating thickness *d*; *b* – beam splitter with input and output ports. Scheme of band bending on the surface of macropores between macroporous silicon matrix with Fermi level E_F and layers ncCdS and SiO_2 (circles – electrons and holes, lines – impurity states)

For 2D macroporous silicon structures with nano-coatings, band bendings on the surface of the macropores are significant (Fig. 5 *b*). Under these conditions, surface polaritons interact with photons strongly [20, 21] due to resonances of dipole-active vibrations and surface modes at boundaries Si-SiO_2 and SiO_2+"PEI-ncCdS" on macropores. Shape of oscillations [12] corresponds to the interference of polaritons as the eigenstates of the system "nanocoating – silicon matrix – waveguide modes" (Fig. 6). In our case, an additional degree of freedom due to creation of vertically polarized light in *z* direction and horizontally polarized light in *x* direction (Fig. 5 *a*) permit to interpret the obtained result as two-photon interference – Hong-Ou-Mandel effect [23]. In this case, macropore is a beam splitter (BS) with maximum and minimum coincidences for measurements with parallel and perpendicularly polarizations, respectively. Beam splitter includes input ports A and B, and output ports labelled C and D. The four ways of the two photons can exit from the beam splitter through the same port or different ports. Two-photon states corresponded to photons exiting through the same output ports demonstrate constructive interference (bosonic behavior). The two-photon states corresponding to photons exit through the different output ports demonstrate destructive interference (fermionic behavior).

In case [12], presented in Fig. 6, we observed the "constructive" interference of the two-photon states corresponded to photons exited through the same output ports C and D (Fig. 5 *b*, bosonic behavior). Maximum coincidences for measurements of the IR absorption spectrum of macroporous silicon structures with surface ZnO nanocrystals (Fig. 6, grey curve 1) and the same spectrum of IR absorption, moved for 2 period (Fig. 6, black curve 2) confirmed the high coherence of "constructive" interference of the two-photon states.

In addition, we measured giant two-polar oscillations of the absorption in spectra of Fig. 4 and Fig. 5 at TO-vibrations of Si-Si-bonds [24] and P_b centers [25]. Interaction of TO phonons with the scattered electrons in *z* direction (Fig. 5 *b*) enhances k_z – vibrations, and the

dispersion law (3) transforms to $E(\kappa) = E_0 - \Delta\ (cos\ k_z a)$. Amplitude of the resonance scattering became dependent on the secant function $(cos\ k_z a)^{-1} = sec\ k_z a$), and the slit-like modes [28, 29] are formed on boundaries "silicon matrix – nanocoatings". Fig. 7 *a–d* show fragments of optical absorption from Fig. 3 *b* for sample #1 and a resolution of 1 cm^{-1}: *a* – in spectral area of P_b centers; *b* – theoretical calculations the secant function, and *c*, *d* – fragments of optical absorption spectra *a* and *b* enlarged on the *y* axis. Fig. 7 *a–d* confirms qualitatively the model of the two-polar giant oscillations in optical absorption spectra depended on the function *sec* $k_z a$. In this case, the two-photon waves exit from the beam splitter through the different ports, and we observe destructive interference of the two-photon states corresponding to fermionic behavior of two-polar oscillations.

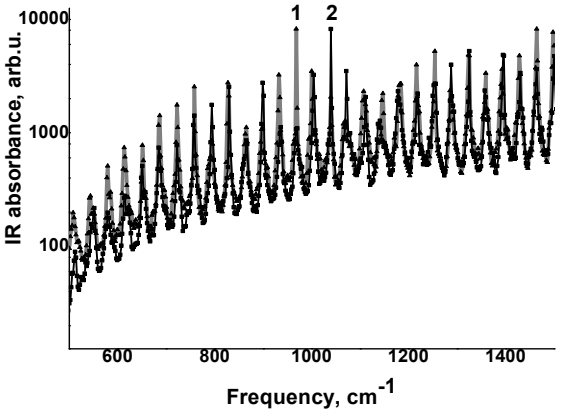


Fig. 6. Dependences of the IR absorption of macroporous silicon structures with surface ZnO nanocrystals (structure #4) in the spectral range 500–1500 cm^{-1} (*1*), the same spectrum of IR absorption, moved for 2 period (*2*)

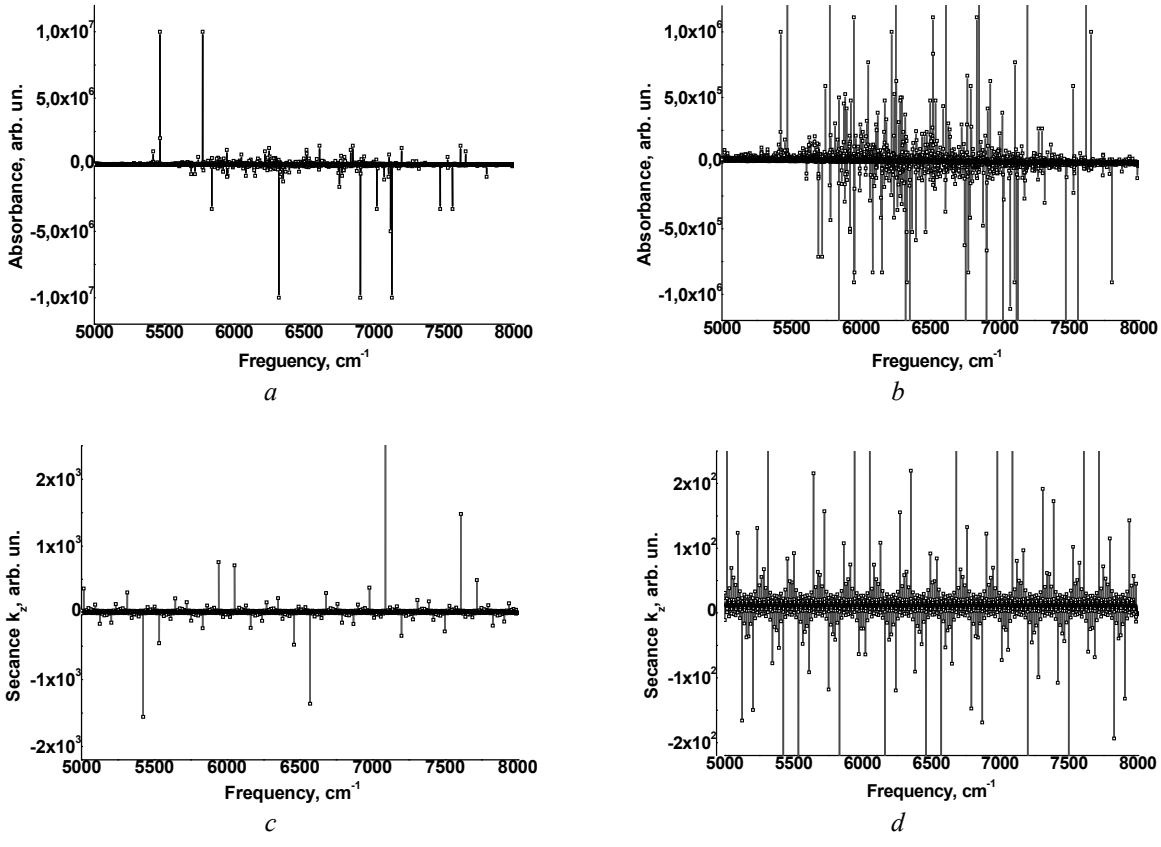


Fig. 7. Fragments of optical absorption of sample #1 measured with a resolution of 1 cm^{-1} in spectral area 5000–8000 cm^{-1} of P_b centers (*a*); *b* – theoretical calculations by the secant function; (*c*), (*d*) – fragments of optical absorption spectra (*a*) and (*b*) with reduced *y* axi

It means that 2D resonances of Wannier-Stark electro-optical effect in *yz* plane (Fig. 6) transforms into 1D resonances in one direction *z* of surface waves (Fig. 7) and into the one-dimensional (1D) polaritons. Such transformation results in the giant amplitudes of absorption oscillations ($\pm 10^7$ arb. un.) and measurements the ultra-small half-width of oscillations (0.5 ± 0.1 cm^{-1}) and minimal Rabi frequency of samples (1.0 cm^{-1}) equaled to resolution of spectral measurement (Table).

The quantum statistics of atoms is observed typically in the behavior of an ensemble via macroscopic observables, in addition, quantum statistics modifies the behavior of even two particles. The basis of the two-photon interaction provides the mixing between the two input modes via the action of the beam splitter (BS) [23] with the diagonal coherent state representation; in particular, they give the explicit example of what happens when two photons, one horizontally and one vertically polarized, are incident from different input ports. In this case, one observe constructive interference of the two-photon states corresponding to photons exiting through the same output ports (bosonic behavior), and destructive interference of the two-photon states corresponding to photons exiting opposite output ports (fermionic behavior). So far, two-particle interference has been investigated on massive particles, such as electrons, atoms, plasmons, and more complex quantum systems [24].

In our case, 2D resonances of Wannier-Stark electro-optical effect in *yz* plane correspond to constructive interference of the two-photon states (bosonic behavior, 2D polaritons), and two-polar resonances in $\pm z$ direction are determined by 1D polaritons (destructive interference of the two-photon states, fermionic behavior).

CONCLUSIONS

1D polaritons with the giant two-polar oscillations in IR absorption spectra of macroporous silicon with nano-coatings were measured. The generated photoelectrons link heavily with holes in the macropore potential pits forming 2D interference polaritons at room temperature. Increase the resolution of IR spectra measurements to 1 cm^{-1} resulted in generation of dipole-active vibrations and change of dispersion law in *yz* plane of macropore surface to *z* direction along macropores. Thus, 2D polariton transforms into 1D polariton according to secant law in spectral area of TO-vibrations of Si-Si-bonds and P_b centers.

Surface polaritons interact with photons strongly due to resonances of dipole-active vibrations and surface modes at boundaries Si-SiO_2 and SiO_2+"PEI-ncCdS" on macropores. Shape of oscillations corresponds to the interference of polaritons as the eigenstates of the system"nanocoating – silicon matrix – waveguide modes". In our case, the vertically polarized light along macropores (*z* direction) and horizontally polarized light (*x* direction) permit the explanation of results as two-photon interference (the Hong-Ou-Mandel effect). In this case, macropore is a beam splitter (BS) with maximum and minimum coincidences for measurements with parallel and perpendicularly polarizations, respectively. Beam splitter includes input ports A and B, and output ports labelled C and D. The four ways of the two photons can exit from the beam splitter through the same port or different ports. We observedconstructive interference of the two-photon states corresponding to photons exiting through the same output ports (bosonic behavior).Furthermore, 1D polaritons are perspective for high-coherent optical quantum computers on macroporous silicon with nano-coatings, for lasers and new metamaterials.

1D та 2D поляритони в структурах макропористого кремнію з нанопокриттями

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У даній роботі ми використали високо-роздільне ІЧ поглинання для дослідження 1D та 2D поляритонів в періодичних структурах макропористого кремнію з нанопокриттями SiO_2 та наночастинок CdS, ZnO. ІЧ поглинання з високою роздільною здатністю призводить до генерації дипольно-активних поперечних коливань, розщеплення фотонів і вимірювання гігантських двополярних осциляцій поглинання з амплітудами $\pm 10^7$відн.од. Це означає додатковий ступінь свободи, оскільки вертикально поляризоване світло в напрямку z і горизонтально поляризоване світло в напрямку x призводить до розщеплення променів і до двофотонної інтерференції – ефекту Хонг-Оу-Менделя. В нашому випадку 2D резонанси електро-оптичного ефекту Ваньє-Штарка у площині yz відповідають «конструктивній» інтерференції двофотонних станів (бозони, 2D поляритони), а двополярні резонанси у ±z напрямках визначаються «деструктивною» інтерференцією двофотонних станів (ферміони, 1D поляритони). 1D-поляритони є перспективними для високо когерентних оптичних квантових комп'ютерів на основі макропористого кремнію з нанопокриттями, для лазерів і нових метаматеріалів.

***Ключові слова**: макропористий кремній, нанопокриття, 1D та 2D поляритони, двофотонна інтерференція*

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