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HONG-OU-MANDEL QUANTUM EFFECT ON “POLYMER - MULTIWALL CARBON NANOTUBES” COMPOSITES

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The possibilities to enhance the properties of nanostructured surfaces are evaluated on “polymer-multiwall carbon nanotube” composites. Influence of sp^3 hybridization bonds is investigated in composites derived from polypropylene, polyamide-6, polyamide-12 and polyvinyl chloride after adding CNTs to polymers. IR absorption of “polymer-CNTs” films exceeds that of polymer by $10-10^3$ times in the entire measured spectral range. In addition, two-polar IR absorption are measured on composites with negative components at spectral positions of “D-band” and “2D-band” of sp^3 hybridization. In this case, the greater oscillation amplitudes of C-C, CH_2 and CH_3 bonds correspond to a higher absorption at the vibration frequencies $\gamma_{\omega}(CH)$ and $\gamma_{\omega}(CH_2)$. Two-polar oscillations of absorption with a negative component in the spectral band ranges “D” and “2D” of sp^3 hybridization in nanotubes have been measured for the composites. Frequencies of 2D-band correspond to the second order frequencies of D-band. The intensity of 2D band increases with an increase in the concentration of defects. The absorption of light increases when the frequencies of local oscillations of surface bonds in carbon nanotubes correspond to the frequencies of slotted modes along the boundary of the “nanotube polymer” (surface polaritons). Two-polar oscillations have an ultra-small half width $0.4-0.6\text{ cm}^{-1}$, which indicates a strong interaction of surface polaritons with photons. Vertically polarized light along carbon nanotubes and horizontally polarized light of D and 2D bands resulted in light beams splitting, two-photon interference and realization of the quantum Hong-Ou-Mandel effect.

Keywords: polymer-multiwall carbon nanotube composites, beams splitting, two-photon interference, quantum Hong-Ou-Mandel effect

INTRODUCTION

In previous papers [1, 2] we analyzed high-resolution IR absorption spectra of macroporous silicon to investigate phonon-polaritons resulted in detection of dipole-active transverse optical (TO) vibrations, photon beam splitting and giant two-polar absorption oscillations with high amplitudes of $\pm 10^7$ arb. un. 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 in macroporous silicon. 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 eigen states 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). As a result, macropore is a beam splitter (BS) with maximum and minimum coincidences for measurements with parallel and perpendicular 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 observed constructive interference of the two-photon states corresponding to photons exiting through the same output ports (bosonic behavior). Two-polar resonances in $\pm z$ direction are determined by 1D polaritons (destructive interference of the two-photon states, fermionic behavior). Furthermore,

1D polaritons are perspective for high-coherent optical quantum computers on macroporous silicon with nanocoatings, for lasers and new metamaterials.

In this paper, we analyzed high-resolution IR absorption spectra of “polymer–multiwall carbon nanotubes (CNTs)” composites to investigate phonon-polaritons resulted in detection of dipole-active TO vibrations, photon splitting and two-polar absorption oscillations with amplitudes up to $\pm 10^5$ arb. un. We compared obtained results with *macroporous silicon structures*.

Multiwall CNTs are among the most anisotropic materials known and have extremely high values of Young’s modulus [3]. Carbon nanotube aspect ratio of length to diameter is more than 10^3 ; this distinguishes it from other nanoparticles. New composites with CNTs as additives are investigated intensively during the last decade. Composites characterized by extremely high specific strength properties, electrical and thermal conductivity [4]. The presence of CNTs in the matrix improves the composite biocompatibility [5]. CNTs exhibit both semiconducting and metallic behavior dependent on their chirality [6]. The researchers have successfully demonstrated field-effect transistors based on semiconducting CNTs [7]. Metallic CNTs are considered as a potential

solution for on-chip interconnects with a current density well above 10^6 A/cm² [8, 9].

Earlier the opportunity to enhance the properties of nanostructured surfaces was investigated on films of “polymer–multiwall CNTs” composites and the composites based on 2D macroporous silicon structures with nanocoatings [1]. Influence of sp^3 hybridization bonds on polymer crystallization and strengthening we evaluated for composites of polypropylene and polyamide with multiwall CNTs [10]. The IR absorption peak dependences on CNT content at frequencies of sp^3 hybridization bonds we described by 1D Gaussian curves for the diffusion equation in the electric field (Fig. 1 *a*). Distance between nanotubes in composite depends on the concentration of CNTs (N_{CNT}), its content (% CNT) and the nanotube volume (V_{CNT}) are equaled to: $a = (N_{\text{CNT}})^{1/2} = (\% \text{ CNT}/100V_{\text{CNT}})^{1/2}$ [10]. IR absorption maximum for sp^3 bond hybridization (D) of composites corresponds to the average distance $a = 0.35$ micron between the cylindrical CNT with diameter of 20 nm and a length of 2 microns. The electric field between nanotubes and polymer matrix (Fig. 1 *b*) has a thickness w of space charge and is equaled to $w = (a_m - d)/2 = 0.17$ μm for maximum of IR absorption. The electric field intensity between nanotubes and polymer matrix is equal to $6.3 \cdot 10^3$ V/cm at 0.25 % CNT [11].

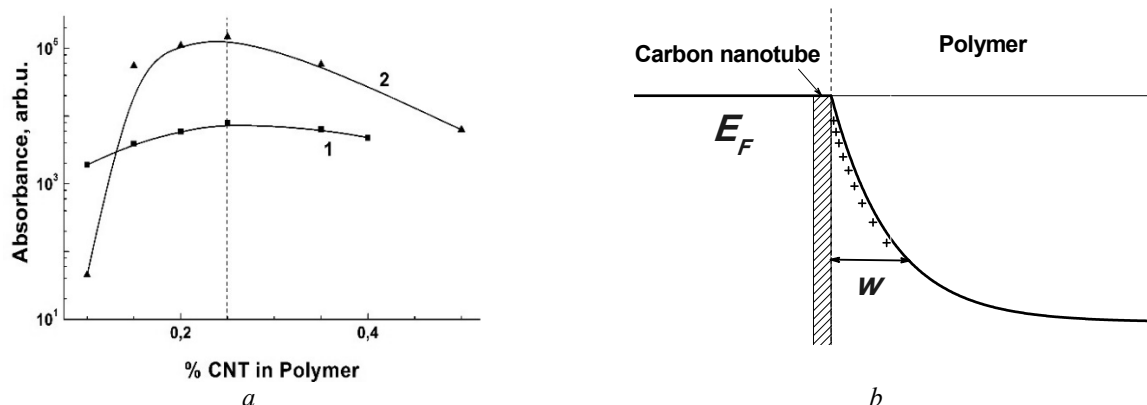


Fig. 1. *a* – IR absorption by sp^3 hybridization bonds (D) in composites based on polypropylene (1) and polyamide-6 (2) vs. CNT content in polymer; *b* – the electric field intensity between nanotubes and polymer matrix has a thickness w

In this paper, the opportunities to enhance the properties of nanostructured surfaces are investigated on “polymer-multiwall CNTs” composites. Influence of sp^3 hybridization bonds on polymer strengthening is studied in

composites polypropylene (PP) – *a*, polyamide-6 (PA6) – *b*, polyamide-12 (PA12) – *c*, and polyvinyl chloride (PVC) – *d* after adding CNTs to polymers with concentration of 0.25 %.

PROCEDURE AND RESULTS

Carbon high purity multiwall CNTs of 2 μm length and 20 nm diameter (Fig. 2) were obtained by catalytic pyrolysis of unsaturated hydrocarbons [12, 13]. Nanoparticle morphology we investigated by the atomic force microscopy (AFM, NanoScopeIIIa Dimension 3000TM).

Carbon nanotubes in “polymer–CNTs” composites we made using a solvent dispersion

method to more uniformly disperse the filler. A screw extruder we used to manufacture specimens. The effect of low CNT concentrations on the mechanical and interfacial properties of polymers were investigated using tensile and impact tests. Low concentrations of CNTs resulted in significant increases in Young’s modulus, impact strength and interfacial adhesion.

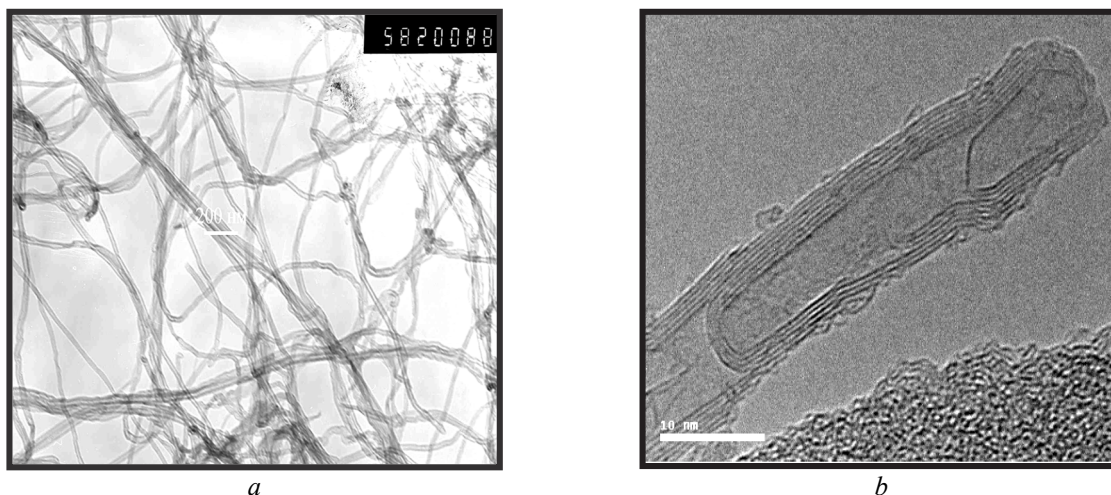


Fig. 2. *a* – group of multiwall carbon nanotubes, *b* – alone multiwall carbon nanotube

The composites were made of polypropylene, polyamide-6, polyamide-12 and polyvinyl chloride filled by a mixture of CNTs with the polymer powder and dried. Thin polymeric films (100–150 μm thick) without and with CNTs were prepared using a Thermo HYDROPRESS. We obtained compression and tension tests of the polymeric materials and their composites using tensile machine 2167-R50 with the automatic recording of the deformation diagrams.

High-resolution optical absorption spectra we measured in the 200–8500 cm^{-1} spectral range on a spectrometer BrukerVertex70V with a resolution of 1 cm^{-1} . We obtained optical absorption spectra at normal incidence of IR radiation on a sample in the air at room temperature. IR absorption of polymer composite films exceeds that of polymer films essentially. According to Fig. 3, IR absorption exceeds the absorption of polypropylene by 10^3 – 10^5 times in the entire spectral range measured after adding CNTs to polypropylene (concentration of 0.25 %).

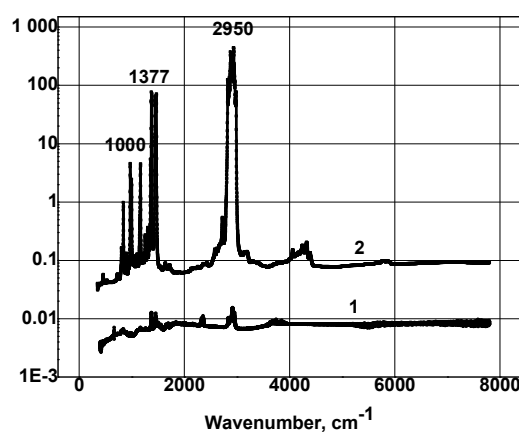


Fig. 3. IR absorption spectra of polypropylene (1) and “polypropylene-CNTs” composite (2)

After adding carbon nanotubes to polyamide-12 (PA-12), the IR absorption of composites exceeds the absorption of polymers by 10^3 – 10^5 times in the entire measured spectral range (Fig. 4 *c*). IR absorption increases between 1400–1500 cm^{-1} (Fig. 4 *d*) [16] for polyvinyl chloride (PVC) after adding CNTs to polymers.

The intensity of vibrations increases for C–C bonds (835 and 1000 cm^{-1}), $\gamma_r(\text{CH}_3) - 970\text{ cm}^{-1}$, $\gamma_\omega(\text{CH}_3) - 1170\text{ cm}^{-1}$, at the frequency of sp^3 hybridization (D) – 1360 cm^{-1} , as well as oscillations: $\delta(\text{CH}_3) - 1380\text{ cm}^{-1}$, $\delta(\text{CH}_2) - 1440\text{ cm}^{-1}$, $\delta(\text{CH}_3) - 1470\text{ cm}^{-1}$. At the same time, IR absorption increases significantly at frequencies 690 cm^{-1} – Amide V and at a

frequency of 1358 cm^{-1} , which corresponds to fluctuations of $\gamma_\omega(\text{CH}_2)$ and frequency of sp^3 hybridization (D) [15, 16]. Higher C–C fluctuations, CH, CH_2 and CH_3 bond absorption correspond to higher absorption at the frequencies of $\gamma_\omega(\text{CH})$ and $\gamma_\omega(\text{CH}_2)$ vibrations [16]. In addition, two-polar IR absorption was measured (Fig. 4).

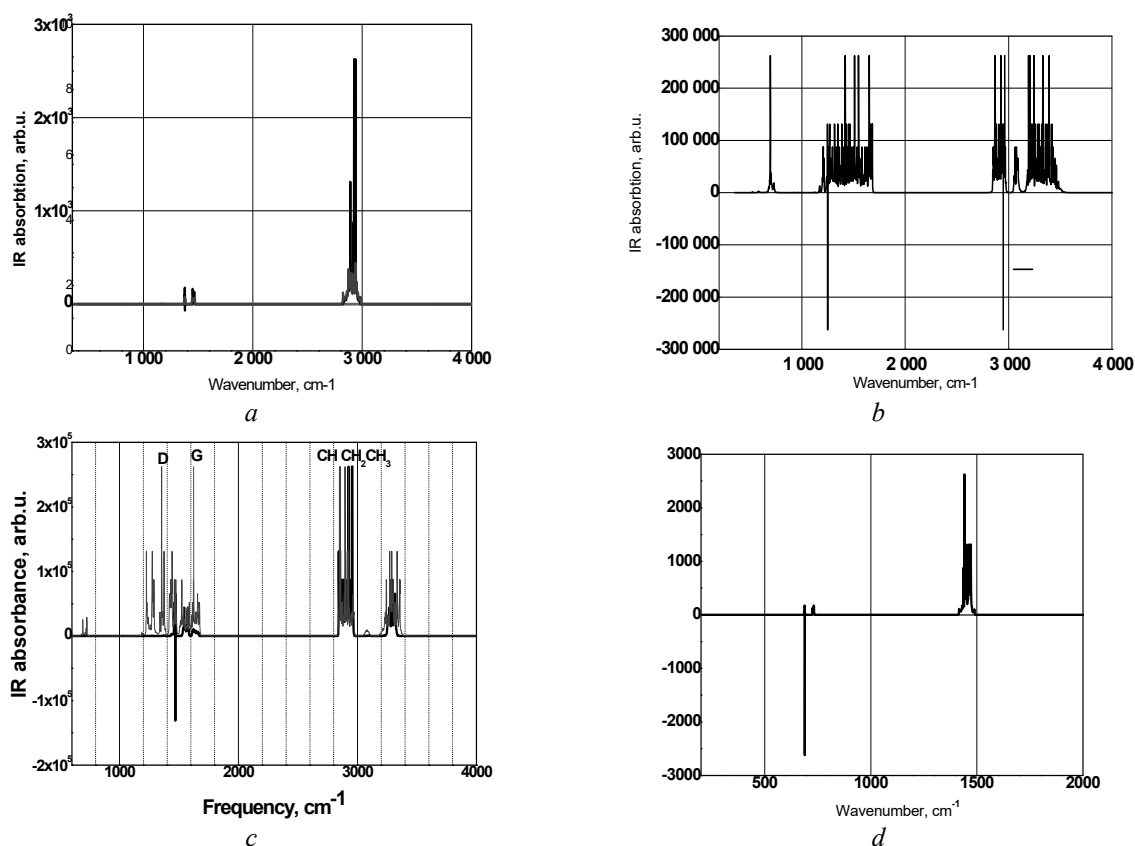


Fig. 4. IR absorption spectra of “polymer-CNTs” composites for films of polypropylene – *a*, polyamide-6 – *b*, polyamide-12 – *c* and polyvinyl chloride – *d*

Table 1 shows types of bonds for two-polar IR absorption from Fig. 4 after adding carbon nanotubes to polymers. Two-polar IR absorptions were measured on polypropylene (PP) composite at 1379 cm^{-1} , on polyamide-6 (PA6) in spectral area $\gamma_\omega(\text{CH}_2)$ vibrations, $\delta(\text{CH}_3)$ (E) for frequency of sp^3 hybridization bonds -1260 (D) and -2950 (2D); polyamide-12 (PA12) -1358 (D), 1529 (G), 1632 , -2904 (2D), and polyvinyl chloride (PVC) -690 cm^{-1} .

According to Fig. 4 and Table 1, two-polar IR absorptions we measured on:

– PP composite at 1377 cm^{-1} , -1379 cm^{-1} in spectral area of $\gamma_\omega(\text{CH})$ vibration for frequency of

sp^3 hybridization bonds (D) [14] and intensive positive peaks 2850 cm^{-1} and 2950 cm^{-1} ;

– PA6 at 1260 cm^{-1} of sp^3 hybridization bonds (D) and -2950 cm^{-1} of double sp^3 hybridization bonds (2D) [12].

IR absorption spectra of PA12 include negative peaks -1358 cm^{-1} (D), and -2904 cm^{-1} (2D) [15, 16] and intensive positive peaks at 1249 cm^{-1} and in area of $2800\text{--}3000\text{ cm}^{-1}$, $3200\text{--}3500\text{ cm}^{-1}$.

IR absorption spectra of PVC negative IR absorption band at -690 cm^{-1} (Amide V) [15] and intensive positive peaks at 1430 between $1400\text{--}1500\text{ cm}^{-1}$.

In general, two-polar IR absorption bands we measured on all investigated composites with negative IR absorption at spectral positions of sp^3 hybridization bonds and/or double sp^3 hybridization bonds of carbon multiwall nanotubes in polymer composites. A group of peaks (1260–1379) cm^{-1} (Table 1), called “D-band”, is assigned to the presence of disorder in graphitic materials [17]. Thus, the origin of D-band in nanotubes is a level of “disorder” in

graphite. D-band is active for nanotubes satisfying a certain chirality due to double resonance conditions [18]. In addition, the D-band of isolated CNTs can be decomposed into two bands; their separation depends upon the incident laser energy. A group of peaks in the 1550–1600 cm^{-1} range constitutes the G-band. In graphite, a single peak is present at 1582 cm^{-1} , corresponded to the tangential vibrations of the carbon atoms [18].

Table 1. Types of bonds for IR absorption growth after adding to polymers carbon nanotubes

PP		PA6		PA12		PVC	
Type of bonds	Frequency, cm^{-1}	Type of bonds	Frequency, cm^{-1}	Type of bonds	Frequency, cm^{-1}	Type of bonds	Frequency, cm^{-1}
	1377		1249,	$\gamma_{\omega}(\text{CH}_2)$	763, -1358 (D),		689
$\gamma_{\omega}(\text{CH})$	-1379 (D)	$\gamma_{\omega}(\text{CH}_2)$	-1260 (D),		1529 (G), 1632,	$\gamma_{\omega}(\text{CH}_2)$	-690
	-2850,		-2950 (2D)	$\nu_{\alpha}(\text{CH}_2)$	-2904 (2D), 3406		1329
	2950 (2D)						

Double (2D) sp^3 hybridization bonds 2950, 2904, 3240 cm^{-1} (Table 1), are second-order mode of D-band [12]. 2D-band increases in intensity with increase in defects as part of the density of states. 2D-band indicates the long range order in a sample, and arises from a two-phonon second-order scattering process. It results in creation of inelastic phonons [12].

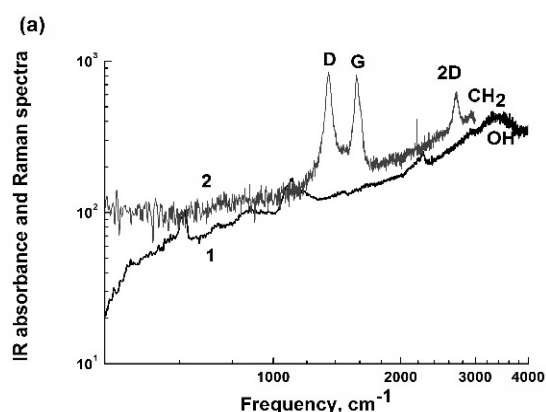


Fig. 5. IR absorption (1) and Raman (2) spectra of multiwall CNTs

Intensive peaks of sp^3 -hybrid orbitals (D), sp^2 -hybrid orbitals (G), 2D and CH_2 bonds were measured in the IR spectra (Fig. 4, curve 1) and Raman spectra (Fig. 4, curve 2) of multiwall CNTs [10].

In paper [1], we investigated two-polar oscillations in absorption spectra of periodical 2D macroporous silicon structures with nano-coatings of SiO_2 and CdS nanoparticles.

The optical absorption spectra we measured at normal incidence of IR radiation along the cylindrical macropores in the air at room temperature. Fig. 6 a, c show fragments of two-polar IR absorption in macroporous silicon structures [1] and Fig. 6 b, d show fragments of two-polar IR absorption “polymer (PVC)–CNTs” composite. 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].

The application of high-resolution IR absorption spectroscopy [1] is determined by detection of bipole-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. 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) [20]. 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 observed constructive interference of the two-

photon states corresponding to photons exiting through the same output ports (bosonic behavior). Two-polar resonances in $\pm z$ direction are connected with 1D polaritons (destructive interference of the two-photon states, fermionic behavior).

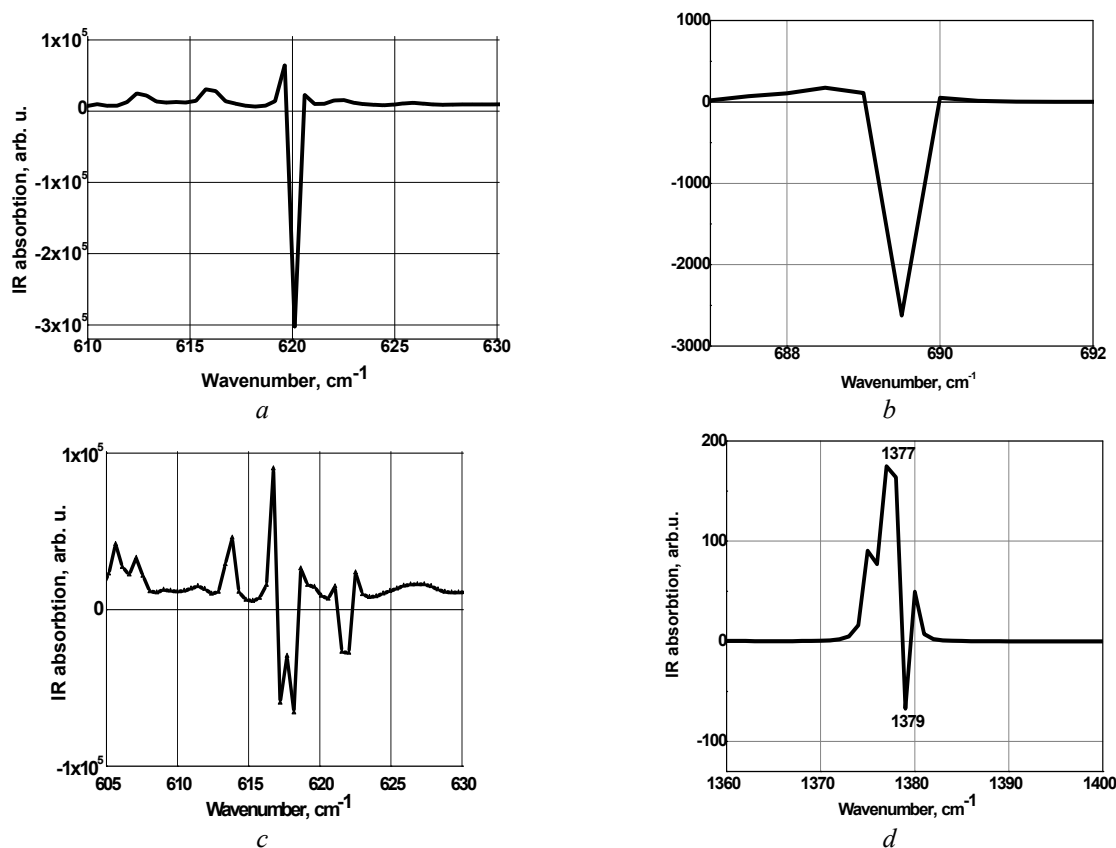


Fig. 6. Two-polar IR absorption in macroporous silicon structures (*a, c*) and “polymer (PVC)–CNTs” composites (*b, d*)

Fig. 6 *a* shows optical absorption by 2D structures of macroporous silicon with nano-coatings measured at resolution of 1 cm^{-1} : *a* – at spectral region of Si-Si-bonds; *b* – at spectral region of P_b centers [1].

In [1], we used high-resolution IR absorption spectra to investigate periodical 2D macroporous silicon structures with nano-coatings of SiO_2 and CdS, ZnO nanoparticles. The application of high-resolution IR absorption spectroscopy permitted to detect 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, beams splitting and two-photon interference – Hong-Ou-Mandel effect. In this case, 2D resonances of Wannier-Stark electro-optical effect in yz plane correspond to constructive interference of the two-photon states (bosonic behavior), and dipolar resonance in $\pm z$ direction is determined by destructive interference of the two-photon states (fermionic behavior). Bipolar oscillations of 1D-polaritons have the ultra-small half-width $0.4\text{--}0.6 \text{ cm}^{-1}$ 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 nanocoatings and, in addition, for lasers and new metamaterials.

Optical properties of crystals are determined by interaction of an external electromagnetic field with dipole-active states of the crystal bulk and with defects of crystal structure. This interaction leads to the formation of a linked state of the oscillate movement of charged particles (dipole) with the electromagnetic field, named polaritons [18, 19]. For 2D macroporous silicon structures with nanocoatings band bending on the surface of the macropores are significant. Under these conditions, surface polaritons interact with photons strongly [22, 23] due to resonances of dipole-active Si-Si-bond vibrations and surface modes on boundaries Si-SiO₂ and SiO₂+PEI-ncCdS on macropores. Si-Si-bonds represent the transverse fluctuations of silicon atoms on the surface of the macropores, *i.e.*, transverse phonons. Measurements the giant two-polar oscillations with very small half-width 0.5 cm⁻¹ (Fig. 3) testify the strong interaction of surface polaritons with photons. Moreover, by changing the thickness of the nanocoatings, it is possible to achieve a match in the frequency of interference modes with frequencies of surface bond oscillations on boundaries Si-SiO₂ and SiO₂+PEI-ncCdS (slotted modes [19]). When the frequencies of local oscillations of surface bonds and slotted modes matched, then the light absorption increases up to 10⁵ times on the frequencies of slit oscillations of surface bonds [20]. For macroporous silicon, 2D resonances of Wannier-Stark electro-optical effect in γz 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) [2]. For composites polymer-CNT two-polar resonance in $\pm z$ direction are determined by 1D polaritons (destructive interference of the two-photon states, fermionic behavior) in spectral area of sp^3 hybridization bonds. When frequencies of local oscillations of surface bonds of carbon nanotubes and slotted modes along nanotube-polymer boundaries matched, then the light absorption increases in 10²–10⁵ times (Figs. 3 and 5) on the frequencies of slit oscillations of surface bonds.

Measurements the giant two-polar oscillations with very small half-width 0.5 cm⁻¹ in IR

absorption spectra of polyvinyl chloride include negative IR absorption band at -690 cm⁻¹ (Amide V) [14] and intensive positive peaks at 1430 between 1400–1500 cm⁻¹. Thus, vertically polarized light along carbon nanotubes and horizontally polarized light for D and 2D bands resulted in beams splitting and two-photon interference and quantum Hong-Ou-Mandel effect [20].

In general, two-polar IR absorption bands are measured on all investigated composites from Table 1 with negative IR absorption at spectral positions of sp^3 hybridization bonds and/or double sp^3 hybridization bonds of carbon multiwall nanotubes in polymer composites. In this case, two-polar oscillations 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 [1, 2].

CONCLUSIONS

In this paper, the opportunity to enhance the properties of nanostructured surfaces is demonstrated on “polymer–multiwall CNTs” composites. Influence of sp^3 hybridization bonds on polymer strengthening is investigated in composites polypropylene (PP), polyamide-6 (PA6), polyamide-12 (PA12) and polyvinyl chloride (PVC) after adding CNTs to polymers (concentration of 0.25 %).

“Polymer–CNTs” composites we made using a solvent dispersion method to more uniformly disperse the filler. The composites were made of PP, PA6, PA12 and PVC filled by a mixture of CNTs with the polymer powder and dried. Thin polymeric films (100–150 μm thick) with out and with CNTs were prepared using Thermo HYDROPRESS.

IR absorption of “polymer–CNTs” films exceeds that of polymer by 10–10³ times in the entire spectral range measured after adding nanotubes with concentration of 0.25 %. Higher C–C fluctuations, CH, CH₂ and CH₃ bond absorption correspond to higher absorption at the frequencies of $\gamma_{\omega}(\text{CH})$ and $\gamma_{\omega}(\text{CH}_2)$ vibrations. In addition, two-polar IR absorption measured after adding carbon nanotubes to polymers with negative IR absorption at spectral positions of sp^3 hybridization bonds of “D-band” and of double “2D-band”. 2D-band is the second-order mode of D-band and increase in intensity with the defect density growing. Two-polar oscillations have the ultra-small half-width 0.4–0.6 cm⁻¹ that testify the

strong interaction of surface polaritons with photons. The light absorption increases 10^2 – 10^5 times when the frequencies of local oscillations of surface bonds in carbon nanotubes matched slotted

modes along “nanotube/polymer” boundaries resulted in the photon beams splitting and two-photon interference – Hong-Ou-Mandel effect.

Квантовий ефект Хо-Оу-Менделя в композитах «полімер-багатостінні вуглецеві нанотрубки»

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В даній роботі можливості підсилення властивостей наноструктурованих поверхонь були виявлені на композитах «полімер-багатостінні вуглецеві нанотрубки». Вплив зв'язків sp^3 гібридизації досліджено в композитах на основі поліпропілену, поліаміду-6, поліаміду-12 і полівінілхлориду після додавання вуглецевих нанотрубок до полімера. ІЧ-поглинання плівок композитів перевищує поглинання полімера у 10^2 – 10^3 разів. При цьому більша амплітуда коливань C–C, CH, CH₂ та CH₃ зв'язків відповідає більш високому поглинанню на частотах коливань $\gamma_{\omega}(CH)$ і $\gamma_{\omega}(CH_2)$. Крім того, на композитах виміряно двополярні осциляції поглинання з негативною компонентою в спектральних діапазонах смуг «D» і «2D» sp^3 гібридизації в нанотрубках. 2D-смуга відповідає моді другого порядку по відношенню до D-смуги. Інтенсивність 2D-смуги збільшується при зростанні концентрації дефектів. Поглинання світла збільшується, коли частоти локальних коливань поверхневих зв'язків в вуглецевих нанотрубках відповідають частоті цилінричних мод уздовж межі «нанотрубка/полімер» (поверхневі поляритони). Двополярні коливання мають надмалу напівширину 0.4 – 0.6 cm^{-1} , що свідчить про сильну взаємодію поверхневих поляритонів з фотонами. Вертикально поляризоване світло уздовж вуглецевих нанотрубок і горизонтально поляризоване світло в D і 2D смугах призводять до розгалуження фотонів, до двофотонної інтерференції та реалізації квантового ефекту Хо-Оу-Менделя.

Ключові слова: композити «полімер-багатостінні вуглецеві нанотрубки», розгалуження фотонів, двофотонна інтерференція, квантовий ефект Хо-Оу-Менделя

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