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HONG-OU-MANDEL QUANTUM EFFECT ON “EXPANDED GRAPHITE - CNTs” COMPOSITES

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We investigated influence of multiwalled carbon nanotubes (CNTs) on spectral characteristics of composites “thermo-expanded graphite – carbon nanotubes (TEG–CNTs)”. The introduction of CNTs in an amount of 0-3% by weight of TEG composites results in a significant increase in the strength characteristics and thermal stability of the composites. This result indicates that CNTs is ideal filler for composites based on TEG compositions and structures. Measurements the giant two-polar oscillations with very small half-width 0.5 cm^{-1} testify the strong interaction of surface polaritons with photons. When frequencies of local oscillations of surface bonds of carbon nanotubes and modes along “nanotube-TEG” boundaries matches, then the light absorption increases 10^2–10^5 times.

Thus, IR absorption with two-polar oscillations was measured at 0% of nanotubes in TEG at frequency of 2750 cm^{-1}. It is own optical mode in the thermally expanded graphite. 5 peaks with two-polar oscillations were measured in the IR absorption spectra at 1% of carbon nanotubes. And 8 peaks with two-polar oscillations were measured at 3 % of carbon nanotubes at optical mode frequencies along the boundaries of thermally expanded graphite - carbon nanotubes. When frequencies of local oscillations of carbon nanotubes and composite’s modes matches, then the light absorption extremely increases (in 10^2–10^5 times), and two-polar IR absorption oscillations with negative components are formed.

In general, two-photon interference is a result of quantum entanglement of dipole-active oscillations and splitting of photons according to the Hong-Ou-Mendel (HOM) quantum effect. Two-photon entanglement is built on the basis of the most entanglement states, also known as Bell's states. The HOM–quantum effect on composites “expanded graphite-carbon nanotubes” is promising for the development of highly coherent optical quantum computers.

***Keywords**: thermo-expanded graphite (TEG), multiwalled carbon nanotubes (CNTs), disclination defects, IR absorption spectra, Hong-Ou-Mandel (HOM) quantum effect*

INTRODUCTION

In this paper, we investigated influence of multiwalled carbon nanotubes (CNTs) on the characteristics of composites “expanded graphite-CNTs” at 0–3 wt. % of CNTs. The methodologies of obtaining composite materials filled with CNTs with their homogeneous distribution in matrices of different nature were presented in [1]. The grid of the nanoscale filler creates a layer of the matrix itself in the nanoscale state, which has significantly improved properties due to the surface energy than in the usual one. It is shown that the thermally expanded graphite (TEG) is a clustered nanoscale system (Fig. 1), which is characterized by long, cylindrical, conical, and slot defects with average diameters of cross sections (0.7–20 nm). It is due to convolution and break several layers of graphene, which related to disclination defects (Fig. 1 *a*, *b*). CNTs were obtained by the method of catalytic chemical vapor deposition (CCVD) by pyrolysis of hydrocarbons on complex metal oxide catalysts [2]. TEG structure formation by rapid heating of residual compounds of graphite and the physical model of TEG as a nanoscale system were clarified in monography [1]. Study of the reaction of intercalation of dispersed natural graphite by anode oxidation with sulfuric acid were provided in [2] too. That include determination of regularities and mechanism of formation of TEG powder into a continuous material and its deformation behavior depending on the conditions of synthesis and density, as well as the possibility and limits of regulation of physico-chemical properties of TEG and materials based on it, for modification by organic compounds and highly modular components (carbon fibers).

The features of physical-chemical processes of formation of intercalated compounds in the dispersed system of natural graphite – sulfuric acid by anodic oxidation were investigated in [2]. It is proved that the TEG is a nanoscale cluster-assembled system, which is characterized by the presence of elongated cylindrical, conical and slit-like defects with average diameters of sections from 0.7 nm to more than 20 nm (Fig. 1 *a–c*), due to the folding and bending of several layers of graphene with a possible mechanism for the formation of orientation defects (disclination). Graphene is a monatomic layer of sp^2-hybridized carbon atoms that form a hexagonal lattice.

It was found that secondary electrochemical “intercalation” in weak electrolytes delaminates dense material from TEG into its nano-sized structural elements and forms graphene nanoparticles, which self-organize into 3D structures on electrically conductive substrates [1, 3]. From the data [3] obtained using the adsorption of nitrogen, *n*-hexane and water vapors, and the electrochemical measurements it is shown that the thermally expanded graphite possesses non-homogeneously - porous structure and expressly hydrophobic surface with ~ 10^{18} active centers per one square meter.

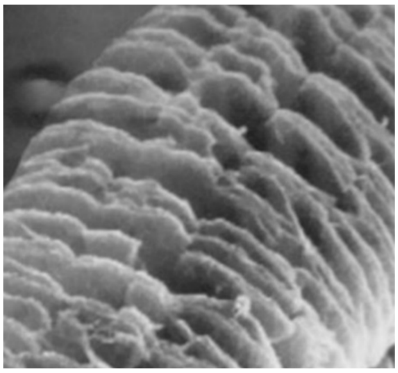

Fig. 1 *a*. Disclination defects in the thermally expanded graphite

60 mcm

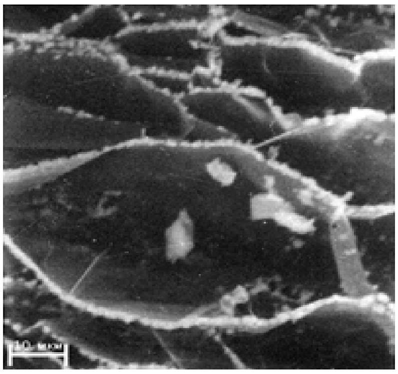

Fig. 1 *b*. SEM image of TEG structure

10 mcm

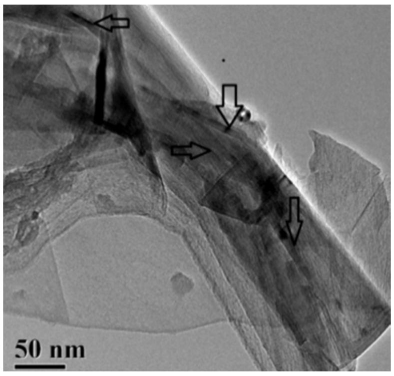

Fig. 1 *c*. SEM image of composite “TEG-CNTs”

50 nm

RESULTS AND DISCUSSION

IR absorption. Earlier we evaluated that IR peaks dependences on the carbon nanotubes content in polymer composites correspond to 1D Gaussian curve for the diffusion equation in the electric field between electrons of nanotubes and protons in polymer according to “semiconductor” model of the composite structuring. And we measured IR reflectance maxima of composites “rubber-carbon nanotubes” at 0–10 % of CNTs in the spectral area of the rubber CH deformation vibrations at frequencies 1297, 1465 and 1728 cm^{-1} and valence vibrations at frequencies 2844 and 2916 cm^{-1} [4–15].

Fig. 2 of this paper shows IR absorption spectra of TEG without CNTs in the spectral area 0–8000 cm^{-1} with intensive negative absorption peak at 2750 cm^{-1} [2]. It is spectral position of the second order mode of D vibration (2D band) at 2713 cm^{-1} [12] with higher intensity than is usually observed for the second order vibration. The latter fact could be an evidence of similarity of carbon nanostructures manifesting a strong electron-phonon interaction and strong dispersion dependence of D-mode. More conductive materials exhibit stronger electron-phonon interaction than semi-conductive ones. A low intensive band at 2451 cm^{-1} called by D'' by Vidano and Fischbach [17, 18], consists of the sum of D and D$_1$ modes (D$_1$ – sp^3 at 1060–1080 cm^{-1}). In our case of composites “TEG – CNTs”, the position of G-mode at 1581cm^{-1} does testify the formation of good crystalline structure of composites “TEG – CNTs” (its theoretical value for graphite and graphene is 1580 cm^{-1}), relative intensity $I_{2D}/I_G = 1.12$.

The analysis of fitted data for 2D mode suggests that the obtained composites “expanded graphite-CNTs” are of good crystalline structure and exhibit the metal-like conductive properties (inharmonic of 2D mode is about 10 cm^{-1}) [18].

Fig. 3 shows IR absorption spectra of composites “TEG - CNTs” at 1% of CNTs in the spectral area 0–8000 cm^{-1} with 5 intensive negative absorption peaks at 2000 cm^{-1} (two peaks), 2500 and 4000 cm^{-1} (two peaks).

Fig. 4 shows IR absorption spectra of composites “TEG – CNTs” at 3 % of CNTs in the spectral area 0–8000 cm^{-1} with 8 intensive negative absorption peaks at 1100, 1300, 1500 *(2),* 2000 *(2),* 3700, 4200 and 5230 cm^{-1}.

Carbon nanotubes are characterized by extremely high specific strength characteristics (breaking strength at ~1.8 TPA), electro- and thermal conductivity, *etc*. In this regard, they have huge perspectives for use in modern technologies. The feature that distinguishes CNT from other nanoparticles is the uniquely high aspect ratio (η) (ratio of length to diameter) that exceeds 10^3. In this case, the percolation threshold Fη (Fη ≈1/η), that is, the concentration at which a continuous grid of CNTs is formed, provided they are uniformly distributed in the polymer matrix, can be ≈ 0.1 wt.%. Therefore, the use of CNTs as a modifier of filled TEG, even with a minimum content of 0.1 wt.%, can provide an increased level of strength of composites “TEG – CNTs”.

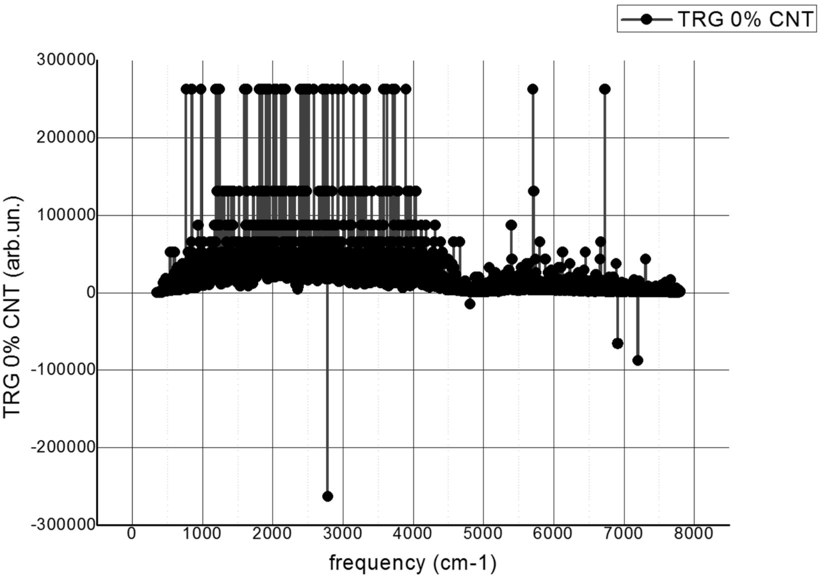

Fig. 2. IR absorption spectra of TRG without CNTs in the spectral area 0–8000 cm^{-1} with intensive negative absorption peak at 2750 cm^{-1} [2]

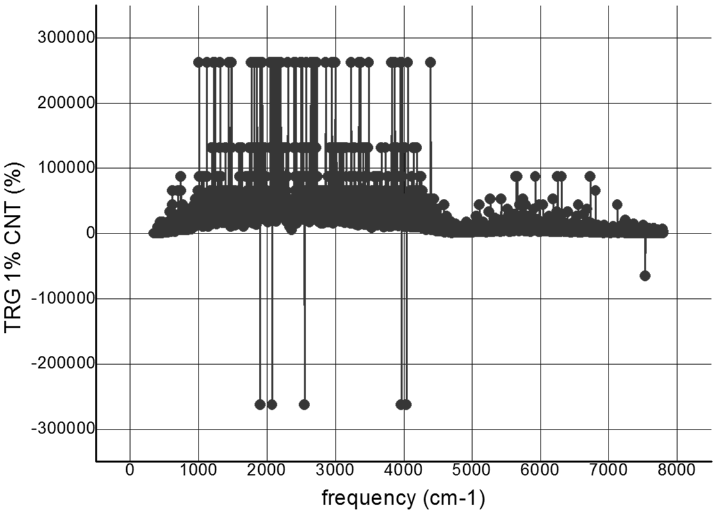

Fig. 3. IR absorption spectra of composites “TRG – CNTs” at 1% of CNTs in the spectral area 0–8000 cm^{-1} with 5 intensive negative absorption two-polar peaks near 2000 (2), 2500 and 4000 cm^{-1}(2)

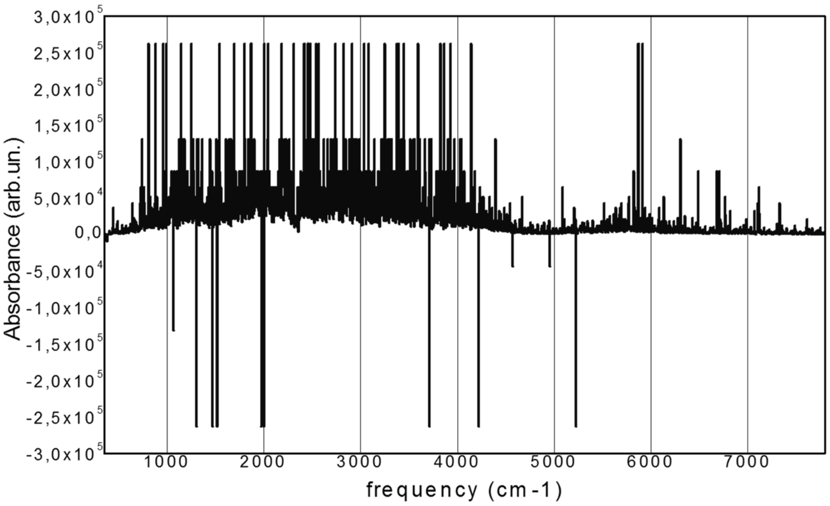

Fig. 4. IR absorption spectra of composites "TRG – CNTs" at 3 % of CNTs in the spectral area 0–8000 cm^{-1} with 8 intensive negative absorption two-polar peaks.

IR absorption spectra. IR absorption spectra of composites "TEG - carbon nanotube" included the giant two-polar oscillations (Figs. 2–4). IR absorption spectra of TEG without CNTs include intensive negative absorption peak at 2750 cm^{-1} as spectral position of the second order mode of D vibration (2D band) at 2713 cm^{-1} (Fig. 2). IR absorption spectra of composites "TEG – CNTs" at 1% of CNTs have 6 intensive negative absorption peaks at 1500 (two peaks), 2000 (two peaks), 2500 and 4000 cm^{-1} in the spectral area 0–8000 cm^{-1}. And IR absorption spectra of composites "TEG – CNTs" at 3% of CNTs include 8 intensive negative absorption peaks at 1300, 1500 (two peaks), 2000 (two peaks), 3700, 4200 and 5230 cm^{-1}. Measurements the giant two-polar oscillations with very small half-width 0.5 cm^{-1} testify the strong interaction of surface polaritons with photons. When frequencies of local oscillations of surface bonds of carbon nanotubes and modes along TEG/nanotube boundaries matched (Fig. 1 *c*), then the light absorption increases 10^2–10^5 times (Figs. 4–5). Furthermore, two-photon oscillations are perspective for high-coherent optical quantum computers on composites. Thus, vertically polarized light along carbon nanotubes and horizontally polarized light for vibrations resulted in beams splitting, two-photon interference and quantum Hong-Ou-Mandel effect [19, 20].

Earlier we investigated high-resolution IR absorption spectra and 1D, 2D polaritons in periodical 2D macroporous silicon structures with nano-coatings of SiO_2 and CdS, ZnO nanoparticles that resulted in detection of dipole-active TO vibrations, photon splitting and giant two-polar absorption oscillations with amplitudes of $\pm 10^7$ arb. un. [21].

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 oscillations with negative components were at spectral positions of "D-band" and "2D-band" of sp^3 hybridization in composites derived from polypropylene, polyamide-6, polyamide-12 and polyvinyl chloride after adding CNTs to polymers [22].

After vulcanization IR absorption spectra of composite "rubber-carbon nanotubes" includes some giant two-polar oscillations in spectral area of the rubber C–H bond deformation and valence vibrations [13].

CONCLUSIONS

The introduction of CNTs in an amount of 0–3 % by weight of TEG compositions results in a significant increase the strength characteristics and thermal stability of the compositions. This result indicates that CNT is ideal filler for composites based on TEG composition and structure.

Measurements of the giant two-polar oscillations with very small half-width 0.5 cm^{-1} testify the strong interaction of surface polaritons with photons. When frequencies of local oscillations of surface bonds of carbon nanotubes and modes along TEG/nanotube boundaries

matched, then the light absorption increases 10^2–10^5 times.

IR absorption with two-polar oscillations was measured at 0 % of nanotubes in TEG at frequency of 2750 cm^{-1}. It is own optical mode in the thermally expanded graphite. 5 peaks with two-polar oscillations were measured in the IR absorption spectra at 1% of carbon nanotubes. 8 peaks with two-polar oscillations were measured at 3% of carbon nanotubes at optical mode frequencies along the boundaries of thermally expanded graphite - carbon nanotubes. In general, when frequencies of local oscillations of carbon nanotubes and composite’s modes matches, then the light absorption increases 10^2–10^5 times, and two-polar IR absorption oscillations with negative components are formed.

In general, two-photon interference is the result of quantum entanglement of dipole-active oscillations and splitting of photons according to the quantum Hong-Ou-Mendel effect (HOM). Two-photon entanglement is built on the basis of the most entanglement states, also known as Bell's states. The HOM effect is promising for the development of highly coherent optical quantum computers.

Ефект Хонг-Оу-Менделя в композитах «терморозширений графіт – вуглецеві нанотрубки»

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Досліджено вплив багатошарових вуглецевих нанотрубок (ВНТ) на спектральні характеристики композитів «терморозширений графіт – вуглецеві нанотрубки (ТРГ–ВНТ)». Введення ВНТ у кількості 0–3 % від маси ТРГ призводить до значного підвищення міцнісних характеристик і термостабільності отримуваних композитів. Цей результат вказує на те, що ВНТ є ідеальним наповнювачем для композитів на основі ТРГ. Вимірювання гігантських двополярних осциляцій з дуже малою напівшириною 0,5 см$^{-1}$ свідчать про сильну взаємодію поверхневих поляритонів з фотонами. При збігу частот локальних коливань поверхневих зв’язків вуглецевих нанотрубок і мод уздовж кордонів «ТРГ–ВНТ» поглинання світла збільшується в 10^2–10^5 разів.

Так, ІЧ-поглинання з двополярними коливаннями було виміряно у ТРГ при відсутності нанотрубок (0 % ВНТ) на частоті 2750 см$^{-1}$, що є власною оптичною модою в термо-розширеному графіті. У спектрах ІЧ-поглинання композиту ТРГ–ВНТ при введенні 1% вуглецевих нанотрубок виміряно 5 піків із двополярними коливаннями. При вмісті 3% вуглецевих нанотрубок в композиті виміряно 8 піків з двополярними коливаннями на частотах оптичної моди вздовж меж ТРГ–ВНТ. Загалом, коли частоти локальних коливань вуглецевих нанотрубок і мод композиту збігаються, поглинання світла надзвичайно зростає (в 10^2–10^5 разів), і утворюються двополярні коливання ІЧ-поглинання з негативними складовими.

Загалом, двофотонна інтерференція є результатом квантового заплутування дипольно-активних коливань і розщеплення фотонів відповідно до квантового ефекту Хонг-Оу-Менделя (HOM). Двофотонна заплутаність побудована на основі станів заплутаності, також відомих як стани Белла. Використання HOM-квантового ефекту на композитах «ТРГ–ВНТ» є перспективним при розробці висококогерентних оптичних квантових комп’ютерів.

Ключові слова: *терморозширений графіт, багатостінні вуглецеві нанотрубки, ефект Хонг-Оу-Менделя, спектри ІЧ поглинання*

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