

**QUANTUM SIZE EFFECTS ARISING FROM NANOCOMPOSITES  
PHYSICAL DOPING WITH NANOSTRUCTURES HAVING HIGH  
ELECTRON AFFINITY**

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**Abstract**

This article considers main problems in application of nanostructured materials in high technologies. Theoretical development and experimental verification of methods for creating and studying the properties of physically doped materials with spatially inhomogeneous structure on micro- and nanometer scale are proposed. Results of studying 11 quantum size effects exposed to nanocomposites physical doping with nanostructures with high electron affinity are presented. Theoretical and available experimental data were compared in regard to creation of nanostructured materials, including those with increased strength and wear resistance, inhomogeneous at the nanoscale and physically doped with nanostructures, i.e., quantum traps for free electrons. Solving these problems makes it possible to create new nanostructured materials, investigate their varying physical properties, design, manufacture and operate devices and instruments with new technical and functional capabilities, including those used in the nuclear industry. Nanocrystalline structures, as well as composite multiphase materials and coatings properties could be controlled by changing concentrations of the free carbon nanostructures there. It was found out that carbon nanostructures in the composite material significantly improve impact strength, microhardness, luminescence characteristics, temperature resistance and conductivity up to 10 orders of magnitude, and expand the range of such components' possible applications in comparison with pure materials, for example, copper, aluminum, transition metal carbides, lumino-phores, semiconductors. (thermoelectric) and silicone (siloxane, polysiloxane, organosilicon) compounds

**Keywords**

*Physical doping, high-power microwave devices, allotropic carbon nanostructures, plasma metal coating, nanocomposite based on transition metal carbides, nanocomposite properties control, dry friction coefficient, nanocomposite wear and heat resistance, charged layer*

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**Introduction.** Operation of modern devices used in cellular networks is based on inventions made by Popov (1895), Tesla and Marconi, who were pioneers in radio communication. However, American and European companies appeared to be the pathfinders in creating the real cellular networks. Now, the United States and China are struggling for leadership in this breakthrough technologies area. Potential benefits of the 5G networks could be tremendous, as their advocates argue. Companies owning patents would earn billions. A zone was deployed in China in 2019, where the 5G mobile network operates; the average data transmission rate there is 3.2–3.5 Gbps. While the maximum rate could reach 35 Gbps. *Huawei* (China) is planning to release not only the 5G smartphones, but other devices as well. For example, the company introduced a Wi-Fi router, where a SIM card could be installed, and the 5G Internet could be provided. 5G technology is not limited to simple web surfing. High data transmission rates, low signal processing delays and increased volume of transmitted information make it possible to expanding application of this technology in other areas. For example, high-quality images from drone cameras could be transmitted to a monitor or to the virtual reality glasses, 4D video could be broadcasted or watched without any delay, etc.

Confrontation between the United States and China in the high technologies area creates problems for Russia as well. Any technology is based not only on discoveries and patents, but also on certain minerals used to manufacture chips, integrated circuits, etc. At present, China is the largest producer and exporter of rare earth metals (REM) providing, in various data, 90–97 % of the global demand (taking into consideration rare earth metals extraction in Congo). According to the experts' forecasts, demand for rare earth metals in Russia by 2020 would reach 5–7 thousand tons/year, and according to a more optimistic forecast it would account for  $\approx$  13 thousand tons/year.

To ensure a breakthrough in the high technologies area, the Russian government decided to recover its REM producer position in the global arena. The group of metals playing a key role in modern science and technology would be extracted and processed in Russia. At least, plans were already announced to start developing one of the world's largest deposits located in Yakutia. Tomtorskoye REM deposit is one of the largest in the world. Its predicted resources are estimated at 154 million tons of ore. Tomtorskoye deposit in Yakutia would be launched in 2021–2022. Tomtorskoye deposit is the only one in the country with niobium and REM reserves. Uranium (0.005 %) and thorium (0.15 %) are the associated elements in the extracted ore. If the first problem in high-tech is the rare earth metals production, then the second is their economical use and efficient operation in high-tech devices. It is possible

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to save such metals with their careful use, both in the rare-earth metals production and their products manufacture, and with an increase in working life and operation efficiency of devices based on REM. These instruments, devices or gadgets parameters are determined by discovering and efficient practical application of various quantum size effects.

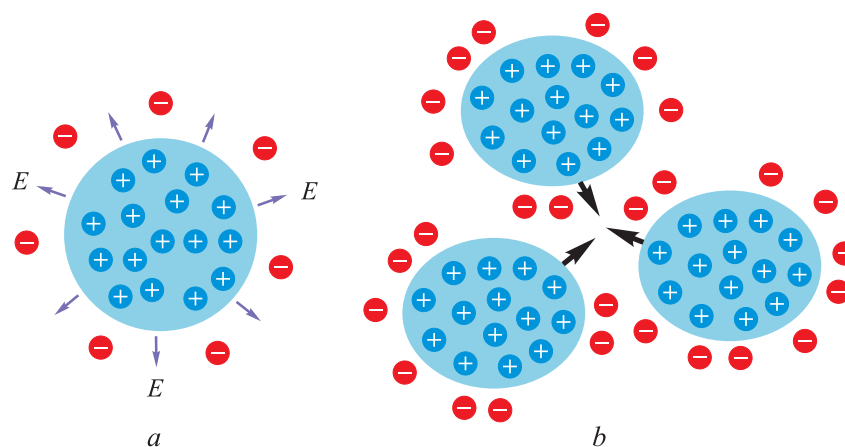
**Problem statement.** Any breakthrough technologies and recent achievements are based lately on certain discoveries in creating and introducing new materials, as well as on finding the quantum size effects in micro- and nanoworld, which make it possible to significantly increase efficiency of the nanocomposites' operation in devices for various purposes. Here, the influence of polarization forces arising in composite materials physically doped with nanostructures with high affinity to free electrons will be analyzed on significant alteration such composites physical properties and them long-term operation.

Interest in nanostructured carbon materials was manifested in several modern experimental works carried out in Russia and abroad (see [1]). This is due to unique properties arising from the composite material nanostructuring with carbon structures, since these are 10 times more durable and plastic, luminescent characteristics are increasing, conductivity could be changed in the range of up to ten orders of magnitude, etc. Currently, scientific literature misses any generally accepted clear understanding of the mechanisms leading to the unique properties' emergence in such nanostructured composites, there is no description of the complex of properties responsible for their technical characteristics.

Physical doping mechanisms and processes responsible for significant alteration in the properties of nanocomposite materials during their physical doping with carbon nanostructures was revealed for the first time in [2, 3] (Fig. 1).

This paper analyzes the main achievements obtained in Russia in various experimental studies on the effect of physical doping processes upon the nanocomposites' properties based on various materials, including copper, aluminum, transition metal carbides, luminophore, semiconductors (for example, thermoelectrics) and silicone (siloxane, polysiloxane, organosilicon) compounds.

Method of physical doping was investigated in order to compare and propose general principles of significant modification in the nanocomposite properties by physical doping with nanostructures that have high affinity to electrons; therefore, they are efficient quantum traps for free electrons in the nanocomposites. It turns out that any carbon nanostructure possesses such properties [3].



**Fig. 1.** Charge separation at physical doping:

*a)* composite material properties modification by layers of a volumetric charge created by the electron traps (red balls with electrons); *b)* diagram illustrating Coulomb compression of the polarized complex structures (arrows show direction of the Coulomb forces compressing the nanostructured polarizable composite)

Free carbon is found both in nature and in many products of micro- and nanoelectronics in various allotropic forms. Carbon is the only element in the periodic table having a variety of allotropic forms. There are stable isomers with one (carbines — linear structures), two (graphenes — two-dimensional structures) or three (diamonds — three-dimensional structures) dimensions. Possibility of different bonds of the carbon atom electronic states in molecules (with different topology in atomic bonds) forming topologically different but mutually compatible bonds could lead to a wide range of electrical properties connected to a specific geometry of nanostructures. Schematically, carbon nanostructures could function as a conductor in the  $sp^1$  — carbine — and  $sp^2$ -chain of the graphene and graphite planar structure, or as a wide-band gap dielectric with  $sp^3$ -bonds, for example, a diamond, etc.

Origin of these properties could be directly attributed to the type of hybridization:  $sp^1$ ,  $sp^2$  or  $sp^3$ . The first two types possess potential to create bonds that are electrically conductive, and the third type has the thermal insulating properties. These properties suggest that nanostructured composite materials have a powerful potential for their use in transistors and other electronic components in the next modifications of data transmission systems and other applications of ‘smart’ nanocomposites, including devices to be used in the next-generation cellular networks [1, 4].

Namely, it is the volumetric charge structuring that determines new properties of the nanocomposites based on copper, aluminum, transition metal

carbides, luminophores, semiconductors and other nanocomposites physically doped with carbon structures. Physically doped nanocomposite diagram (see Fig. 1) generally corresponds to charge separation in the copper–carbon [3] or carbide–carbon [1] nanocomposite. Structures with plus signs are metal [3], metal carbide [1] or semiconductor (thermoelectric, etc. [3]) nanocrystals, and with minus signs — doping nanostructures with high affinity to electrons.

Nanocomposites could be created using various methods:

- anopowder mixing in mills with subsequent compaction and annealing;
- high-speed plasma magnetron sputtering with formation of a nanocomposite film on the substrate surface;
- impurity spraying or deposition on the surface of a physically doping material (for example, luminophore);
- etching, mechanical action on the material with changing its surface or internal structure at bombardment with high-energy particles, creation of cavities, caverns, guide lines and even at using the crystal lattice planes, etc.

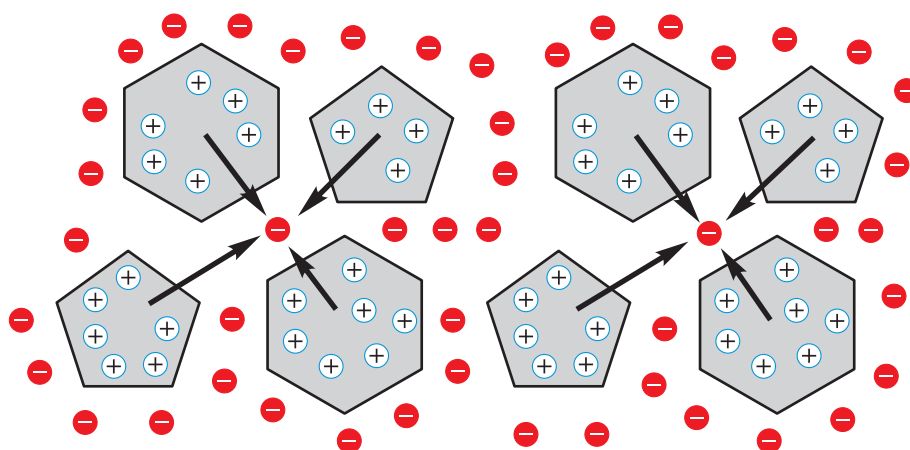
Free electron de Broglie wavelengths usually exceed the interatomic distances, and this contributes to extrusion of both electrons and atomic excitations into the cavities and caverns area. This leads to electrons' localization and charge separation in the so structured nanocomposites. We will not dwell on porous silicon structures. Here, material properties could also widely vary depending on the porosity. (Our approach can be used to explain similar effects in porous materials as well.) Let us concentrate on discussing application of hollow fullerenes and nanotubes in technology.

**Results obtained.** Based on many years of research on nanostructured materials, we came to practically interesting results in the use of nanocomposites and classifications of the types of material doping. Let us consider classification of types of the differently doped materials.

*Classification of doped materials.* For convenience, there are two general types of doping, i.e., chemical and physical doping of composites [6–11]. When crystals are chemically doped with foreign atoms, a foreign atom is introduced into the crystal lattice. This disturbs the lattice and leads to wave oscillations at the nanoscale of the relative dielectric permittivity of the doped material [6–11]. Such doping is introduced in manufacturing transistors used in cellular network devices. Physical doping methods are not yet widely used there. (However, any structuring at the micro- and nanoscale level could be attributed to the material physical doping, for example, to formation of nano- and microstructures in the integrated circuits production, etc.).

During physical doping with nanostructures having high affinity to electrons, portion of the electrons is extracted from the crystal into the doped

micro- or nanostructure. This leads to Coulomb interaction between a positively charged micro- or nanocrystal and the negatively charged doped nanostructure (see Fig. 1 and Fig. 2). This is manifested, for example, in the nanocrystal luminescence characteristics alteration, Coulomb hardening material, elimination of recrystallization in the doped nanostructured material and in other quantum size effects. Methods of obtaining nanostructured composite multiphase nanocomposites of various nature physically doped with carbon nanostructures and studying their properties were tested using the example of physical doping of copper, aluminum, luminophores, semiconductors (thermoelectric and silicone compounds) and transition metal carbide nanocrystals. Not only the results of studies of the nanocomposite properties could be found in the references to experimental works, but also the methods for their creation. Let us list the main 11 quantum size effects experimentally and analytically studied in Russia.



**Fig. 2.** Nanocomposite physical doping diagram: arrows show direction of the Coulomb forces that compress the nanostructured polarized composite; circles with the “-” sign inside are nanostructures with trapped electrons; circles with the “+” sign inside are the positive ion in a positive nanocrystal with free electrons gone; hexagons are the MeC (carbide nanostructures of metal — Me); pentagons are the Me (metal nanostructures)

Basic material nanocrystals’ encirclement with nanostructures of the carbon allotropic forms is used in retraction of free electrons of the physically doped material by polarization forces into the electron quantum traps being thus negatively charged. According to Fig. 2, it is possible to physically dope not only materials with free electrons (metals), but also any other materials. To do this, it is necessary to add small amounts of doped impurity to the composite, as well as a small amount of metal or impurities with free electrons.

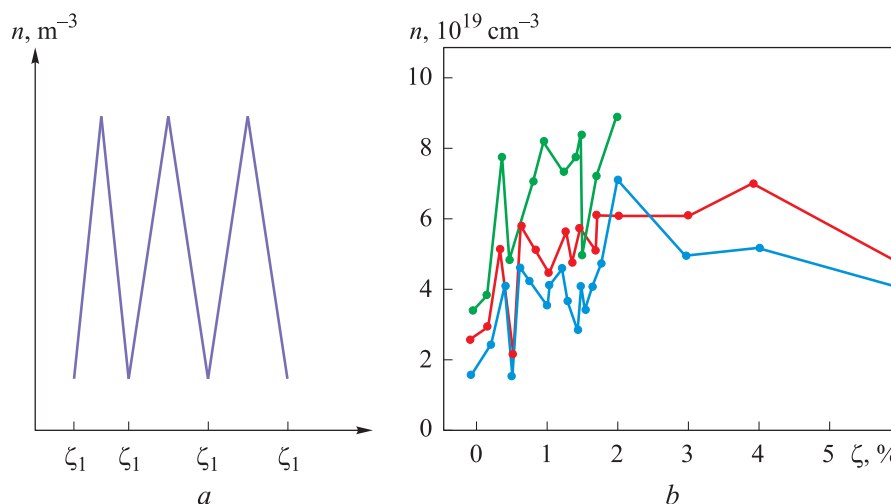
Positively charged crystallites of the basic material would not recrystallize, which ensures continuous functioning of the nanostructured materials during physical doping. This could be applied not only to materials obtained as a result of magnetron sputtering, but also as a result of compacting mixtures and of other methods of creating nanocomposites with physically doped impurities acting as the electron traps previously located in nanocrystals of the basic material.

**11 quantum size effects in physical doping of nanostructured composites.** Violation of electroneutrality (polarization) at the nanoscale (see Figs. 1, 2) leads to 11 polarization quantum size effects described in [1–3, 5–10] and experimentally studied in [1, 3, 5, 10, 12–16].

*Effect 1* — Coulomb hardening (contraction, focusing, reduction, compression, cumulation) of positively charged nanocrystals by nanolayers of the negatively charged electron traps (Fig. 1 *b*, see Fig. 2) [1–3, 6–10]. This quantum size effect was studied experimentally using the example of carbon nanostructure physical doping in materials, such as semiconductors (thermoelectrics) [5], copper [3, 10, 12, 14, 15], aluminum [13] and transition metal carbides [1]. According to experimental observations, copper and aluminum were strengthened with physically doped fullerenes up to 10 GPa, and copper films — with graphite nanostructures up to 4 GPa [14]. Transition metal carbides' strengthening by physical doping of films with carbon nanostructures (see Fig. 2) by magnetron sputtering was registered up to 31 GPa [1]. In this case, strengthening by physical doping of nanostructures with sizes ranging from 20 nm to 100 GPa is considered theoretically possible [2]. According to [17], adhesion forces and compression counteracting forces in crystalline materials are equal to each other. Therefore, material strengthening corresponds to the same increase in the nanocomposite microhardness [10, 17]. Note that nanocomposite materials could be strengthened by creating nanosized graphene shells covering the composites.

*Effect 2* — generation of electric fields (up to  $10^{11}$  V/m) and potentials that 'heat' electrons in the nanocrystal surface region (Fig. 1 *a*) [2, 8, 9]. With such physical doping, surface properties of the volumetrically charged nanocrystals are also changing, for example, due to an increase in the free electron average energy at the nanocrystal surface. This leads not only to dynamic strengthening of the physically doped nanocrystals and weak modification of their crystal lattice, but also to alteration in the physical and chemical properties due to the electric fields' catalytic properties. Thus, amplification of the luminophore luminescence characteristics physically doped with carbon nanostructures occurs up to 50 %, according to [15].

*Effect 3* — dependence of the electric field strength on the nanocrystal surface upon the nano-trap concentration on the surface of a physically doped material [2]. This leads to alteration in the electron energy at its surface, and, consequently, to alteration in the number of electrons captured in the electron quantum resonator (nano-trap). If only the resonance spectrum of localized electrons is possible in a quantum resonator, as in the  $C_{60}$  fullerene [5–10], then the Vysikaylo concentration-polarization-quantum-size effect of the second type would be observed in the corresponding range of parameters (Fig. 3). Applying the quantum size effect, properties of the thermoelectrics could be modified by changing nanocarbon particles' concentration in the nanocomposite [5–10]. In experiments [7], it became possible to increase its efficiency by 30 % by using physical doping of thermoelectric by fullerenes.



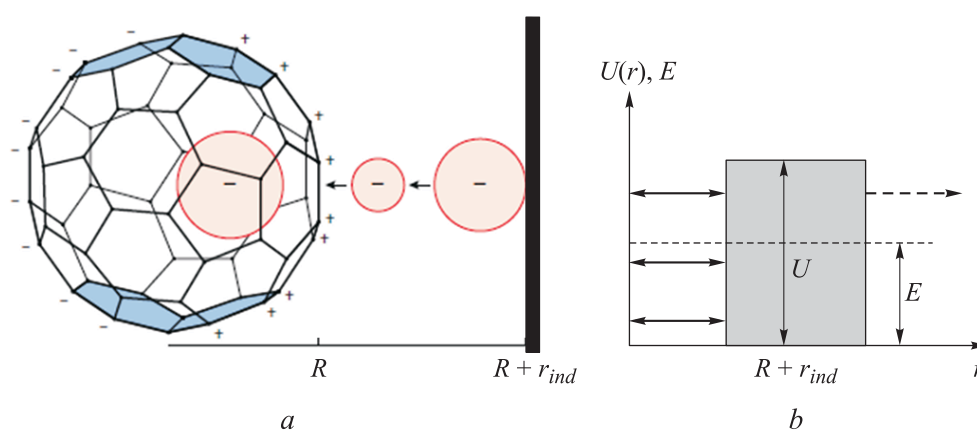
**Fig. 3.** Concentration quantum size effect:

*a)* characteristic dependence of the nanocomposite parameter, for example, of the  $n$  ions or electrons concentration on the  $C_{60}$  quantum resonator volumetric content in the composite (profiles shift with alteration in the nanocrystallite characteristic radius forming a quantum size pair with a relative concentration of traps, which changes parameters of nanostructured composite in the mesoworld [6–10]); *b)* results of experimental measurements in the electron concentration resonance profiles with alteration in the trap relative concentration at doping with  $C_{60}$  fullerenes of semiconductor nanocrystals (thermoelectrics) at different temperatures (77–295 K); nanocrystallite characteristic radius after annealing is  $R \approx 17$  nm [5]

*Effect 4* — cumulation (localization) of free electrons in a trap, i.e., quantum resonator. For example, fullerene according to experiments [18] could accumulate up to six electrons per a single  $C_{60}$  with resonant kinetic energy ranging from 0.24 to 12 eV [19–21]. This could lead to the Fermi gas energy cumulation of up to 30 eV in a single fullerene. The  $C_{60}$  ideal shape allowed the



author of [6–10] to introduce a mathematical model developed by Gamow to describe the  $\alpha$ -particle penetration through a potential barrier. The Gamow model modified in [6–10] made it possible to analytically calculate the resonance energy levels of trapped electrons for all spherical quantum resonators ( $C_{60}$ ,  $C_{70}$  and senior fullerenes). From comparison with experiments, it became possible to determine the effective polarization sizes for various fullerenes ( $r_{ind}$  for  $C_{60}$  0.26 nm and for  $C_{70}$  0.28 nm), where polarization potential barrier about 18 eV high appears for electrons escaping the fullerene and bringing the electron back into the fullerene cavity (Fig. 4).



**Fig. 4.** The scheme of the cumulative polarization electron capture with the resonant energy from 0.2 to 20 eV into a hollow  $C_{60,70}$  molecules [6–10]. The endoion radius (the location of reflecting ‘mirror’  $R_i = (R_{C_{60,70}} + r_{ind})$ ); *a*) the 4D space-time process of formation of a standing de Broglie wave in a polarizing quantum resonator; *b*) the diagram of a metastable (quasi-open) quantum particle with the polarization mirror (of finite size), which captures the electron with energy  $E > 0$  into the polarization trap of the characteristic size  $R + r_{ind}$ . The polarization mirror is darkend. This problem is the first Helmholtz boundary value problem [6–10]

Satisfactory agreement between theoretical model [6-12] and experimental studies in [19–21] made it possible to calculate the effective polarization sizes ( $2(R+r_{ind})$ ) for all types of senior fullerenes [6–10]. Here, based on the comparison of analytical calculations with experiments [19–21], splitting of levels with the  $n$  principal quantum number was discovered into two levels: ( $n$ ) sin-solutions and ( $n-1/2$ ) cos-solutions of the Schrödinger equation (reduced to the Helmholtz first boundary value problem) for hollow spherically symmetric quantum resonators. These studies led to discovery of the cumulative quantum mechanics in [6–10], where the  $\psi$ -functions could cumulate indefinitely to the

center of hollow spherically or cylindrically symmetric quantum resonators for de Broglie waves of quantum particles [6–10]. In this case, regularization of unbounded solutions for the  $\psi$ -eigenfunctions in hollow quantum resonators occurs as a result of considering the  $4\pi r^2$  geometric regularization coefficient, when calculating the probability of finding a particle in the  $dr$  —  $W = 4\pi r^2 |\psi|^2 dr$  layer for spherically symmetric resonators and, accordingly,  $2\pi r$  with the resonator cylindrical symmetry.

Total resonant energy spectrum ( $E_{n-1/2}, E_n$ ) of quantum particles is not depending on the hollow quantum nanoresonator symmetry, but only on the size of the  $R$  hollow quantum resonator. Analytical studies carried out in [6–10] and their results comparison with experiments [17–19] demonstrated that de Broglie hypothesis that particles behave like waves for hollow quantum resonators also works in the case of cumulative quantum phenomena in hollow polarizable spherically and cylindrically symmetric resonators, as in the case of ordinary acoustic resonators [6–10] used to study the sonoluminescence (SL) [22]. A model is proposed here explaining the appearance of a single-bubble SL based on considering the energy additional resonant absorption by a single cavitation bubble in the symmetric acoustic field [22]. A more convincing example of the classical phenomenon of the cos-wave transition into a particle and back is a drop falling into the center of a cup with a liquid (Fig. 5).

**Fig. 5.** Illustration of the particle — wave (cos-wave) — particle transition or of the cumulative drop rebound from the surface of liquid in the acoustic resonator with a boundary (response to a drop of liquid to a resonator with liquid in the Earth gravitational field)



Quantum dimensional polarization effects should also be considered in the interaction of atoms and molecules in the dense media [6–10, 23]. This leads to an increase in the three-body interaction effective calculated cross sections of atoms, molecules and more complex atomic structures, and to solving a number of paradoxes (discrepancies between experimental results and theoretical concepts) [5–11, 23, 24].

*Effect 5* — consequence of the quantum size Effect 4. Rather, it is a consequence of the possibility of trapping electrons in a cavern, cavity or crack. When materials are exposed to energy loading, for example, when they are exposed to neutron irradiation, any form of electromagnetic radiation, friction, bending and other functional energy loads, there appears accumulation of energy arriving from the outside by the Fermi gas localized in the material. Electrons in the material are staying in the atomic nuclei Coulomb potential wells. And as the electron total energy in the Coulomb well increases, its kinetic energy decreases. According to the quantum mechanics basics, de Broglie wavelength of such an electron increases; therefore, the atom swells. According to the Newton's third law, medium is opposing the excited atom. Therefore, excitations tend to be displaced to the surface and to be self-focused to internal inhomogeneities of the functioning material. As the number of excited atoms increases, noticeable local swelling is observed in loaded materials, as well as crack growth and alterations in other physical and chemical properties leading to deterioration in the material functional properties and even to destruction thereof. Inflation and self-focusing processes (cumulation and localization) in swelling promote formation and growth of cracks and localization of free electrons in cracks or cavities. Further filling of cracks with free electrons, as the most mobile and volumetric gas, is determined by displacement of free electrons from the material into the crack and reflection of electrons by the crack boundaries back into the crack. This occurs in the same way, as in the case of an electron polarization capture by fullerene (see Fig. 4). Electron energy distribution function (EDF) is formed in a crack with a large number of electrons, as a result of electron-electron collisions, since the crack geometry, as a rule, has a conical shape, it is accompanied by displacement of high-energy electrons into the crack tip region. Increase in the Fermi gas pressure localized in a crack and geometry of energy cumulation to the crack tip leads to a catastrophic crack growth and material destruction, which is in no time to dissipate (disseminate) external energy cumulating into the crack by the functioning material. According to the above, the Fermi gas is responsible for all this, more precisely, increase and cumulation of total energy in the crack in regard to a small fraction of the functioning material electrons. Increase in the excited electrons number in the cavity or crack region leads first to its slow and then catastrophic explosive growth, as the material fatigues, and the electron excessive kinetic energy accumulates in it transforming into potential energy concentrated in the cracks. In this case, crack strength due to its geometry is decreasing as it grows; and surface and volume, where the energy is accumulated by free electrons, are growing. Therefore, introduction into physically doped nanocomposites of the strongest nanometer traps for the  $C_{60}$  free

electrons (with resonance energy spectrum of 0.24–12 eV) in small amounts, which do not significantly change useful properties of nanocomposite materials, makes it possible, under a number of conditions, to reduce the number of free electrons and thereby weaken catastrophic swelling and crack growth in functional materials exposed to energy loading. These  $C_{60}$  spherical cavities are able to concentrate up to six electrons with the total energy of up to 30 eV [10]. In this case, the fullerene skeleton strength corresponds to the graphene strength and exceeds the diamond strength. In this regard, such a spherically symmetric trap, unlike cavity or crack, is not increasing in volume and is not swelling, which significantly increases the threshold of a nanocomposite catastrophic destruction. Such physical doping increases the nanocomposite useful life, its reliability and duration of functioning under the operation conditions.

Significant increase in reliability at the nanocomposite physical doping would undoubtedly stay in demand in the nuclear industry for physical TFE and their protective containment doping [25]. (In certain sense, this phenomenon, i.e., physical doping with fullerenes, is similar to drilling a hole in glass with a crack in the region of its end and beginning. This method was used by auto enthusiasts. The difference is significant only in characteristic dimensions of the structures.)

*Effect 6* — levitation (hovering, repulsion) of positively charged nanostructures of the physically doped material one above the other (see Figs. 1, b, 2). This effect counteracts recrystallization processes in the nanocomposites, which significantly increases their functioning period [1, 8–10].

*Effect 7* — nanofullerite (fullerene nanocrystals) Coulomb melting, which occurs when the  $k$  electrons are captured by fullerene molecules in the fullerite. In fullerite crystal, the  $C_{60}$  molecules are interconnected by weak van der Waals forces. The binding energy of a single fullerene molecule in the fullerite nanocrystal is not exceeding 1.6 eV [26]. In this case, potential energy of the Coulomb interaction of two negative ions in fullerite with the  $k$  electrons is of the  $1.4k^2$  eV order (average distance between fullerenes in fullerite is  $D \approx 1$  nm). Therefore, capture of several electrons from a nanocrystal physically doped with fullerenes leads to a Coulomb sputtering of fullerite nanostructures and monolayer covering with negatively charged fullerenes of the doped positively charged nanocrystal (see Fig. 1 b) [1, 9, 10]. One  $C_{60}$  fullerene molecule is able to capture up to six electrons [18]. Note that, in principle, neither nanotubes, due to their entanglement, nor nanographenes, due to their strength, are possessing such property to decompose into strictly nanoscale structures. In experiments, it is namely this property of fullerites that made it possible to create nanocomposite materials by physical thermoelectrics [5], copper [12] and alu-

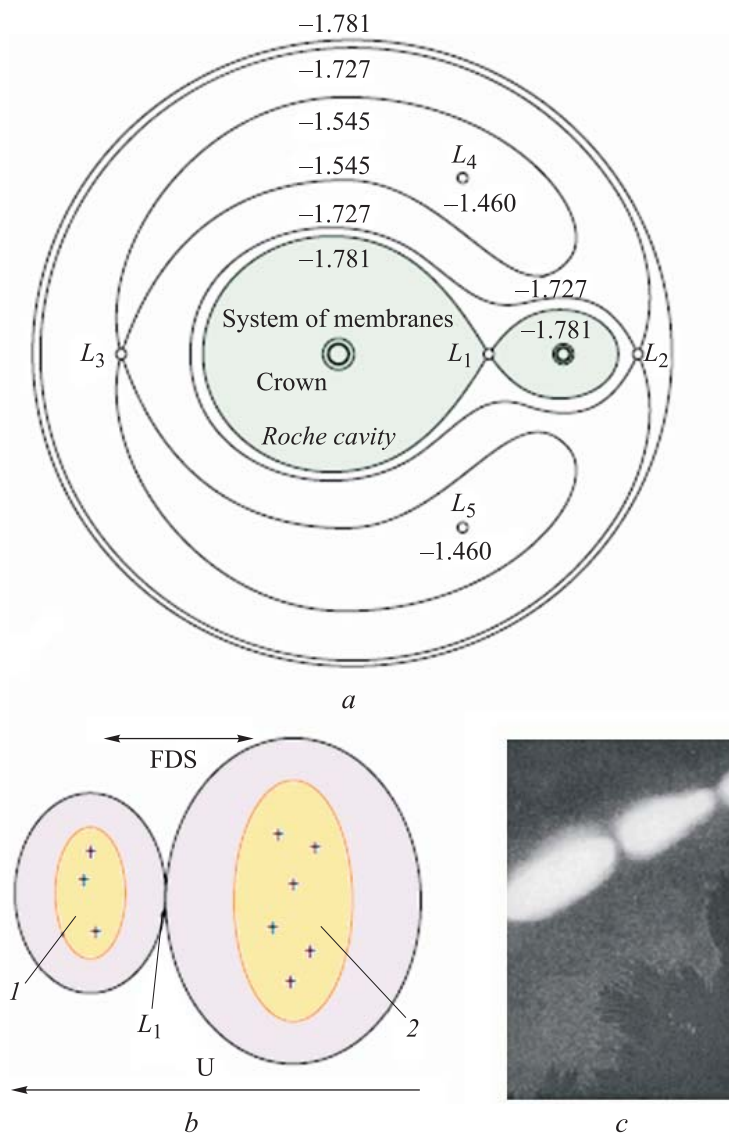
minum [13] doping with  $C_{60}$  fullerenes having unique functional and strength properties. Its most significant progress in increasing strength properties was observed during physical doping of transition metal carbides [1]. The theory in [2, 10] predicts possibility to increase microhardness of physically doped nanocomposite up to 100 GPa.

*Effect 8* — electron fluxes cumulation at libration points between positively charged nanostructures (Fig. 6 *a, b*). These effects were so far studied in the gas-discharge plasma [27] and in insufficient detail in the nanocomposite materials. This cumulative-quantum size effect caused by capturing free electrons with resonant energy in a quantum resonator and formation of positively charged nanocrystals of the doped material could be used to control conductivity, thermal conductivity and other properties of the nanocomposites.

*Effect 9* — counteraction of electrons in the trap to external action on the trap and on the entire nanocomposite material (see Fig. 4). In this case, electrons already with the  $D = 2(R + r_{ind})$  sizes from a nanocrystal physically doped with traps counteract the external influence keeping the physically doped material from destruction;  $R$  is the trap radius;  $r_{ind}$  is the characteristic induction size to which the electron leaves the trap (see Fig. 4 *b*). In experiments, this effect was observed when metal products were coated with graphene sheets, and it was used to explain the white dwarf properties in astrophysics by Frenkel and earlier in the work by Fowler. It turns out that any electrons localized in the Coulomb well in any quantum resonators with sizes ranging from  $10^{-15}$  (in this case, negative mesons play the role of electrons [17]) to  $10^{26}$  m have dual properties to focus and counteract compression.

General diagrams of complex gravitating (Fig. 6 *a*) and electrically (Fig. 6 *b*) interacting cumulative dissipation structures are shown in Fig. 6. In the case of gravitating systems (see Fig. 6 *a*), the Roche surface diagram consists of two closed cavities surrounding both material points (two stars or stars and planets, for example, the Sun is the massive point on the left and Jupiter is the massive point on the right) and having the  $L_1$  common point. The  $L_{2-5}$  Lagrange — Euler libration and cumulation points are determined by consideration of interference of long-range nonlocal gravitational and centrifugal potentials or of the gravitating bodies inertia ( $m(\mathbf{V} \cdot \nabla)\mathbf{V}$  term in the momentum transfer equation). In the case of electrically charged systems interaction (see Fig. 6 *b*), equipotential surfaces consist of two closed cavities 1 and 2, for example, two positively charged 3D finite plasma structures, i.e., cathode spot 1 on the left and positive column 2 on the right (or two positively charged striations) and having the  $L_1$  common point, i.e., the electron fluxes cumulation point ( $L_1$  is the electron

fluxes cumulation point or the focus for electrons);  $U$  is the external potential drop, electrons there are moving from point 1 to point 2. This example illustrates the method of generalized mathematical transposition (transfer) of mathematical models from astrophysics to the electrical phenomena area [27].



**Fig. 6.** Cumulative dissipation structure diagrams:

*a*) cross-section of surfaces with equal potential (taking into account the centrifugal potential) in the Roche model in the orbital plane of a binary system; *b*) cross section with equal potentials according to the Vysikaylo — Roche model [27] in the cross section plane of two positively charged plasma structures; *c*) illustration of the long-range convective non-locality — beaded lightning with points of electron fluxes cumulation and Faraday dark spaces (FDS) between luminous positively charged plasmoids [27]

*Effect 10* — significant alteration in conductivity of materials physically doped with nanostructures of the carbon allotropic forms. For example, nanotubes use could lead to effects of quantum ballistic conductivity in negative ions from nanotubes that capturing electrons. The method of physical doping with nanotubes makes it possible to change conductivity of the initial siloxane by 10 orders of magnitude [28]. As shown in [5–10], if for the  $C_{60}$  fullerenes polarization length, to which, according to the quantum mechanics, an electron localized by the polarization potential could move is from the order of  $r_{ind} = 0.26$  nm, and for  $C_{70}$   $r_{ind} = 0.28$  nm (see Fig. 4), then, for a carbon nanotube the  $r_{ind}$  polarization length is determined by the  $L$  nanotube length. This leads to a significant decrease in the lower level of the quantum resonator intrinsic (resonant) energy spectrum for electrons and its tendency to a continuous spectrum with the increasing  $L$  length. Consequently, much more free electrons with resonance energy can be localized around and in a nanotube, which can significantly increase the efficiency of nanotubes in emission phenomena as compared to fullerenes. Decrease in the nanotube lower energy level leads to an increase in the ejection frequency of electrons previously captured by the nanotube. While the analytical calculations provided in [6–10] for fullerenes were confirmed in a number of experiments [1, 3, 5, 10, 12–16], such calculations in regard to nanotubes present ambiguous results.

These ambivalent (dual) conclusions for polarized carbon nanotubes should be verified in experiments with nanotubes having the  $L = \text{const}$  given length.

*Effect 11* — quantum-size polarization effects should be taken into account in the multiparticle interaction of atoms and molecules in dense media [10]. This accounting leads to an increase in the capture time of several particles into the polarization trap (in the polarization barrier region common for several particles) and to an increase in the effective polarization cross section by  $2Rr_{ind} + r_{ind}^2$ . Here  $R$  is the cross-section radius without taking polarization into account and  $r_{ind}$  — takes into account the polarizing length (see Fig. 4). In particular, this leads to an increase in the effective calculated cross sections for three-body interactions of atoms, molecules and more complex atomic structures, as well as to solution of several paradoxes (discrepancy between experimental results and theoretical concepts) [23, 24].

**Results and discussion.** Currently, fundamental changes are taking place in the new technologies. This is due to the fact that the era of creating unique devices and of using materials in them came, and their properties are determined by structural elements of the nanoscale range ( $\approx 100$  nm). Here, the most important characteristic features of nanosystems are determined not by a specific

decrease in particles, elements or structures in devices, but by fundamentally new qualitative phenomena inherent in the nanoscale, when the influence of quantum mechanics laws, electric potentials and dimensional surface effects on macroscopic parameters of the products obtained is manifested. It is believed that nanotechnology includes technologies, where design of materials in new devices is determined by characteristic dimensions of the composites, i.e., 1–100 nm. Indeed, characteristic quantum size of a free electron at room temperature or its de Broglie wavelength is  $l_e = 12.25 [\text{Å}]/\varepsilon \approx 7.6 \text{ nm}$ . Here  $\varepsilon$  is the electron kinetic energy. In media, disturbance influence with the  $l$  characteristic size (or  $l_e = 7.6 \text{ nm}$ ) has a significant effect on sizes several times larger, on the order of  $10l$  (or 76 nm).

Consequently, nanotechnologies are technologies used in conditions comfortable for humans (at the room temperature), where polarization capture of one or several free electrons could be the main element in designing composite materials [1, 10, 16]. Electron has a negative charge. Coulomb potentials (wells and barriers) that modify nanoobject properties arise as a result of forming and modifying double layers of a volumetric charge in neutral nanostructures [1, 6–10] (see Figs. 1, 2, 4). Coulomb forces appear to be all decisive at the nanoscale level and are of long-range character. Therefore, addition of nanoclusters or nanostructures capable of capturing electrons with resonant energy (quantum traps for free electrons) could significantly modify the nanocomposite macro properties. This phenomenon is caused by violation of electroneutrality at the nanosizes (see Figs. 1, 2, 4). Even in these phenomena, quantum nature of the nanotechnological processes is fundamentally changing the way materials, instruments and complex devices are created. Unlimited possibilities open up for synthesizing nanoscale structures with various potential polarization barriers and, what is most importantly, with self-assembly into micro- and macrostructures having unique properties and functions (see Figs. 1–4, 6). This understanding is truly a revolutionary step leading to radical changes in the technology of new devices, instruments and materials manufacture for various industries. Carbon structures have high electron affinity, while hollow structures such as fullerenes, nanotubes and other closed structures bounded by the graphene planes are able to capture resonant electrons and modify the properties of nanocomposites containing the nanostructured carbon (see Figs. 1–4, 6).

In order to create modern 4G and 5G technologies, it is very important to use high-temperature materials based on chemical compounds of the transition metals pertaining to IV–VI groups of the periodic table of elements with nitrogen, boron, silicon and carbon. The work shows that there are methods of phys-



ical doping, which include such materials that make it possible to effectively improve performance characteristics thereof. According to elaborated and experimentally verified methods of physical doping of a number of nanocomposites, varying composition of the resulting multiphase composite could significantly change not only its structure, but also useful properties of instruments and devices. These materials and nanocomposites made from them are of interest for instruments and devices used in the 5G networks.

At present, interest was already shown in nanocomposites physically doped with the carbon allotropic forms. Analysis of properties of a certain number of nanocomposites was carried out. This work summarizes experience obtained by comparing analytical calculations and experimental studies conducted by various authors.

Research in quantum size effects caused by nanocomposites physical doping with carbon nanostructures was first performed in theoretical works [6–9] using the example of nanocomposite doping with fullerene molecules characterized by high affinity to free electrons. It was found in experiments (see [18]) that up to six electrons could be captured by a single fullerene molecule. According to theoretical concepts of the fullerene endoion intrinsic energy spectrum [6–9], energy in this case up to 30 eV could be focused on a single fullerene. According to indirect data in [5], capture of up to six electrons per one fullerene was observed in semiconductors.

Polarization quantum size effects were described in [6–9]. And according to analytical estimates, upon physical nanocomposite doping with fullerenes with characteristic core size of about 20 nm, the composite could be strengthened up to 100 GPa. Effect of nanocomposite strengthening by physical doping of metals (copper and aluminum) with fullerenes was confirmed in [12, 13]. In these cases, microhardness increased up to 10 times (up to 10 GPa). Copper nanocomposite physical doping with graphite nano-structures ensured obtaining nanocomposites with a strength four times higher than the initial samples [3, 14, 16]. Thus, physical doping with fullerenes to increase microhardness or adhesion of the nanocomposite parts is several times more effective than with other allotropic forms of nanocarbon. This is due to Coulomb melting of fullerene (Effect 7) in the nanocomposite when stirring it in mills [5, 10, 12, 13].

Discovery of the concentration-polarization effect of the second kind (Effect 3) described in [6–10], was confirmed in experiments [5] (see Fig. 2), where it became possible to increase efficiency of the thermoelectric physically doped with fullerene molecules by 30 % using this effect.

When studying composite coatings, it was found out that presence of the free carbon nanostructures in a composite regardless of the composition

makes this material resistant to wear [1, 3, 10, 14, 16]. As established in [28], method of physical doping with nanotubes (Effect 10) makes it possible to change conductivity of the initial siloxane by 10 orders of magnitude.

A special position among nanocomposites is occupied by materials based on oxides and carbides having high melting point ranging from 2,580 °C (molybdenum semi-carbide) to 3,880 °C (tantalum carbide with hardness of up to 31 GPa), wear resistance, corrosion resistance, resistance to molten metals, low vapor pressure and low evaporation rate. Studies are of particular interest that include thermal properties of nanocomposite coatings based on carbides and methods of applying them to working surface of the machine tools. Carbides in their pure form, like oxides, have increased brittleness. To increase their plastic properties, a metal phase is usually introduced into the carbide composition, and the grains are ground and then sintered. In principle, this is also one of the methods for material physical doping. However, nanocrystals start to recrystallize during long-term functioning of a nanocomposite in the device.

To preserve fine-grained (fine-dispersed, fineness) of metals, carbides, semiconductors and luminophores during long-term operation of devices based on them, an original method of composite nanocrystal physical doping with carbon structures being the traps for free electrons was proposed. Carbon structures are polarizing and pull on themselves a portion of the composite free electrons. They are charging negatively, and the carbide nanocrystals that donated free electrons are charging positively (see Figs. 1 *b*, 2). In this regard, positively charged carbide nanocrystals do not recrystallize with time (Effect 6) [1, 9, 10].

Coatings of carbide nanocomposites physically doped with graphite nanostructures have the prospect of being used in cutting metalworking and shaping tools operating in modern high-speed cutting, rolling and broaching modes that increase the processing productivity and significantly reduce its cost. In addition, these coatings are characterized by compatibility with such viscous and ductile metals as copper, aluminum and their alloys, and could be used to protect spinnerets and dies when drawing wires, as well as in rolling and sizing rolls of rolling mills when producing sheets and foil from these materials.

For the most part, practically all carbide composites obtained in [1] as a result of physical doping with graphite nanostructures had a relatively low friction coefficient, less than 0.3. Among this group, samples of the Nb:C series (50:50) had an abnormally low friction coefficient of 0.13–0.16 at loads on a

ball with diameter of 5 mm made of VK-8 up to 100 N. Along with this, they also have specific electrical and thermal properties, and could be used as materials for heating elements in high-temperature furnaces, high-temperature thermocouples and cathodes of the thermoelectric devices [3], as well as in fuel elements with the uranium addition [25], etc.

Using the example of measuring the carbide microhardness, it was established according to the experiments performed in [1] that with an increase in the graphite nanostructures' content it becomes possible to obtain the maximum microhardness, which is 20–60 % higher than for the compact materials. At the same time, these solid nanocomposites have a fairly low friction coefficient. Increase in the material hardness could possibly be associated with the following factors [1]:

- increase in the grain boundaries length with ultradispersed and nanocrystalline structures;
- alteration in carbon composition and carbide phase growth;
- increase in internal stresses (compression);
- volumetric charge separation and Coulomb compression of the nanostructured material (see Figs. 1 *b*, 2).

Nanocomposite physical doping based on transition metal carbides made it possible to obtain durable (Coulomb compressed, see Figs. 1, 2) materials with high microhardness of up to 30 GPa, low dry friction coefficient of up to 0.13–0.16, high thermal stability of up to 3,000 °C, thermal stability in nanocrystalline state over 1,200 °C, high wear resistance and wear rate less than  $10^{-17}$  m<sup>3</sup>/Nm [3].

High-speed ion-plasma magnetron sputtering (HIPMS) appears to be a modern method of carbide coating without the use of reaction gases, such as acetylene, methane, propane, etc. (explosive process) [1, 3, 10, 14, 16]. This method makes it possible to carry out under certain conditions (changing power density of the plasma discharge in the range of 40–125 W/cm<sup>2</sup>) combined sputtering from mosaic targets of various compositions of materials, for example, metal — graphite, metal — carbide — graphite, carbide — metal, carbide–graphite, and to vary in a wide range both composition (ratio of the areas occupied the metal, carbide and graphite), and dispersion of a crystallite structure by changing the discharge power. Rates of coating with carbides and metals using HIPMS at such power densities of material sputtering (metal and graphite) range from 0.1 to 0.7 μm/min, which is almost an order of magnitude higher than with conventional magnetron sputtering (MRS-PVD) and vacuum arc sputtering (ARC-PVD). This method makes it possible to practically pass to a

different mechanism of interaction between carbon and metals during their sputtering by ion bombardment.

When using gases, carbides are formed directly on the target, and then carbides are molecularly transferred by sputtering onto the substrate. But in the case of HIPMS, carbon and metal atoms are jointly sputtered from mosaic targets and interact between each other both in the vapor flow and on the substrate. In this case, we are able to obtain not only carbides, but also other phases, including nanocarbon and metal nanostructures. This is confirmed by the process of applying a composite metal-carbon coating, where the metal is not interacting with carbon, for example, copper-carbon. In this case, the coating has a two-phase composition consisting of metal nanostructures and nanographite phase [3, 10, 14, 16].

Results of analyzing the comprehensive study of phase composition, structure, mechanical and electrical properties of the multiphase nanostructured composites based on metal carbides and obtained by magnetron sputtering from a composite (mosaic) target in a high-speed mode and with a discharge power density of more than  $40 \text{ W/cm}^2$  demonstrate the following [1].

1. Composite coatings based on the transition metal carbides with nanostructure could be obtained by combined sputtering of metal and graphite from a mosaic target.

2. High-speed materials sputtering mode in the discharge power density range of  $37\text{--}125 \text{ W/cm}^2$  makes it possible to vary the composite phase composition, nanostructure and its properties, including those mechanical: hardness, wear resistance and dry friction coefficient.

3. High-speed sputtering mode significantly affects dynamics of generating the vapor flow and formation of condensate in it with several nanophases: nanophase based on carbide, nanophase based on metal and graphite nanophase. The latter is present in all compositions.

4. Free carbon influence in the nanographite form on the composite properties significantly reduces the friction coefficient, increases wear resistance, thermal stability, strength at high temperatures and contributes to production of materials with carbide and metal ultradispersed nanocrystalline structure.

5. Process of metal and graphite combined sputtering is much simpler, technologically advanced and environmentally friendly in comparison with the process of spraying metals in the atmosphere of reaction gases: acetylene, methane, propane, etc.

Technological process of forming nanocomposite carbon with physically doped nanostructures was investigated in [2, 3, 10, 14, 16], and appears to be a breakthrough approach.

Special attention among the new nanocomposites is paid to film materials, which recently became widely used to modify the working surfaces of machines and mechanisms significantly increasing service characteristics of most components and metalworking tools. Improving properties of the material surface layers in components and contact conditions of work leads to an increase in their service life and, accordingly, to significant savings in materials and energy.

Let us list the main problems that we solved from 2010 to 2020:

1. Theoretical models and methods for creating nanocomposite materials with unique properties were developed.

2. Implementation included:

a) selection of candidate materials and their concentration for coating layers of transition metal carbides physically doped with carbon nanostructures;

b) development and manufacture of targets;

c) synthesis by the coating magnetron sputtering method using transition metal carbides with different relative carbon content physically doping the carbide nanocrystals;

d) experiments and calculations to establish carbide nanograin structure and composition of the intergranular interface layers, i.e., carbon nanostructures in sputtered composite targets and synthesized coating layers of transition metal carbides with different relative carbon content;

e) target composition optimization for forming coatings of transition metal carbides with different relative carbon contents;

f) development of composite multiphase targets providing stable magnetron sputtering for the coating magnetron synthesis of transition metal carbides with different relative carbon nanostructure content.

3. Nanocomposites made of nanocrystals of transition metal carbides and physically doped with carbon nanostructures were created, and their properties were investigated.

These high-temperature nanostructured materials based on chemical compounds of transition metals of the IV–VI groups in the periodic table of elements with carbon are of interest both from the standpoint of fundamental science and for creation of 4G and 5G modern technologies. Our studies showed that a special place among these compounds is occupied by carbides with the high melting point ranging from 2,580 °C, i.e., molybdenum semi-carbide, up to 3,880 °C, i.e., tantalum carbide, with hardness of up to 30 GPa, wear resistance, corrosion resistance, resistance to molten metals, low vapor pressure and low evaporation rate. Nanostructured composites possess a set of properties that are important for solving many problems in micro- and nanoelectronics: high melting point, high hardness, wear resistance, corrosion resistance, resistance

to molten metals, low vapor pressure and low evaporation rate, weak recrystallization of nanostructures. These properties determine high efficiency of using composites nanostructured with nanostructures of the carbon allotropic forms, and hence scientific and applied significance of the nanocomposite physical doping method.

4. A method was elaborated to obtain and control properties of the nanostructured composite multiphase coatings (based on transition metal carbides) with high microhardness of up to 30 GPa, low dry friction coefficient of up to 0.13–0.16, with high thermal stability of up to 3,000 °C and thermal stability in the nanocrystalline state over 1,200 °C. Nanocrystalline structure and properties of the carbide composite multiphase coatings were controlled by changing free carbon concentrations in them with formation of the amorphous nanographite phase in the material content. It was found out that presence of nanographite in the composite significantly increases the impact toughness and broadens the range of possible applications in comparison with pure carbides.

5. New technological process of the nanostructured composite material physical doping was elaborated, which appears to be a breakthrough and is 'green' (environmentally friendly).

First, using the PMRM technique, it is possible to sputter any materials from one target, setting their concentration by a simple ratio of the areas in the zone of maximum target erosion. Second, any carbides and metals physically doped with carbon nanostructures could be obtained by simple combined metal and graphite sputtering without supplying explosive gases to the chamber working volume. They are simply not required in this process. Third, controlling the sputtering mode (plasma discharge power density) makes it possible to change structure and phase composition (free amorphized graphite content) in a wide range and control accordingly the properties, for example, hardness, wear resistance and friction coefficient, as well as the most important performance characteristics of composite coatings based on carbides and metals, such as copper, aluminum, etc. When using HIPMS for applying carbide and other carbon-containing composites, explosive gases and their mixtures with hydrogen will not be introduced, unlike other technologies used in research and production of the coatings.

6. Theory of quantum size effects caused by the nanocrystal free electrons capture by doped nanostructures that act as traps for the composite free electrons (on the example of carbon nanostructures) was elaborated in [2, 6–10] and verified by experiments [1, 3, 12–16, 19–21].

It is rather difficult now to discuss possible implementation of commercial 5G networks and relevant devices, provided that a common standard has not yet been introduced, and there is strong opposition from the United States, which is constraining development of China. However, manufacturers in China tackled the next-generation networks very seriously. Their developments are even ahead of the standard release. If companies participating in this project manage to achieve their goals and agree on cooperation, then the entire world would be able to obtain a unified, stable, convergent and highly available next-generation network. Representatives of the International Telecommunication Union declare their hopes that the 5G network would become a point in the development of wireless networks. It is believed that there will be no significant alteration in the architecture, and we will only face minor revision of the radio part that includes research, development and application of technology not only of chemically alloyed composites, but also introduction of physical doping into production of technologically new nanocomposites and devices based on them and in practice. Such implementation is necessary starting with electronic (various computers, mobile phones and other gadgets) and processing industries, where ultra-strong temperature-resistant materials are required, and ending with nuclear industry, where the role of reliability and safety of the working devices operation is essential, and which could be significantly increased by using physical doping of fuel elements and of their coatings with carbon nanostructures [25] or silicon. Results obtained and discovered quantum size effects are promising in terms of their application in new devices and development of methods for controlling the composite material properties and for analytical prediction of their characteristics based on the principles of physical and chemical doping and creation of nano-traps for free electrons or 4D quantum dots of various nature.

Foundations of the mechanism for generating a strong bond between physically doped crystals and carbon nanostructures were investigated, and selection of optimal compositions and coating content for nanoelectronics and optoelectronics components was made. The physical doping method developed in [1, 6–10, 14–16] has no global analogues and would be undoubtedly used to solve problems in nanoelectronics and optoelectronics.

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