Physics of the Solid State, 2025, Vol. 67, No. 9

05,11

Negative magnetoresistance in high magnetic fields in a carbon nanostructure obtained by PECVD

© *A.A. Kharchanka, A.K. Fedotov, J.A. Fedotova*

Institute for Nuclear Problems of Belarusian State University,
Minsk, Republic of Belarus

E-mail: XaaTM@mail.ru

Received December 31, 2024
Revised July 19, 2025
Accepted September 23, 2025

The negative magnetoresistance effect (NME) is investigated in quasi-two-dimensional graphite layers on the surface of which the island-like nuclei of vertical graphene (VGN), containing a high density of paramagnetic defects (PMC) with uncompensated spins, were formed. A hypothesis is proposed for a possible reason for the NME observation, which is formally described by the theory of quantum corrections to the Drude conductivity with the magnetic field oriented perpendicular to the layer plane. The model is based on the assumption that the magnetic flux Φ is distributed non-uniformly along the plane of the C-layer due to the fact that the islands of the VGN phase contain a high density of PMC with uncompensated spins. Ferromagnetic regions are induced in the vicinity of the PMC clusters due to the parallel alignment of spins on the PMC due to the exchange interaction. It is assumed that the described redistribution of the magnetic flux Φ should lead to its concentration in the regions of the location of the islands-nuclei of the VGN phase and its significant weakening (rarefaction) between the islands. This explains the apparent observation of the OMR effect described by the theory of quantum corrections at high values of the external magnetic field B given by the superconducting solenoid (up to 8 T), although in reality most of the conducting C-layer is in a significantly weakened field..

Keywords: quantum corrections to conductivity, weak localization, negative magnetoresistive effect, vertical graphene, carbon nanostructure, magnetism in carbon structures.

DOI: 10.61011/PSS.2025.09.62353.358-25

1. Introduction

The literature has abundance of scientific studies that are dedicated to producing graphite- and graphene-like nanostructures and studying a relation between a structure and many application properties (see [1–9] and others and references therein). However, this relation is still incompletely understood in relation to electrical properties of the graphite-like nanostructured materials. One of these problematic issues is the nature of the origin of the negative magnetoresistive (NMR) effect in some carbon-based materials. In many studies (see, for example, studies on graphite and a polycrystalline single- and multi-layer CVD graphene [10,11], etc.) this effect is interpreted based on an Altshuler-Aronov theory of quantum corrections (QC) to Drude conductivity ([10], etc.), which are induced by dephasing of electron wave functions when they move under the conditions of the so-called weak localization (WL). However, this interpretation does not take into account that the quantum corrections shall be suppressed already in the weak magnetic fields according to the theory (at most 50−100 mT). Therefore, it is entirely correct to use this model to explain existence of the NMR effect in the much stronger magnetic fields (including from 1 to 8 T). The second problem is an influence of the magnetic properties on charge carrier transport in some carbon-based structures. It is theoretically predicted in [11,12] that magnetic centers are formed even without the magnetic ions in the samples, which was experimentally found later in the studies [13–15].

The article proposes a hypothesis that explains possible causes of observation of the NMR effect in the external magnetic fields above 1 T, which is formally described within the framework of the theory of WL quantum corrections. Within the framework of this hypothesis, it is assumed that a density of the magnetic flux Φ is highly inhomogeneously distributed over the sample area, if the latter includes areas (including nanoscale ones), which have a large concentration of the paramagnetic centers (PMC) with uncompensated spins. As a result, these areas can have a high ferromagnetic moment even without ferromagnetic ions due to the parallel alignment of spins on the PMCs by the exchange interaction. This situation can result, for example, in a high concentration of the magnetic flux within the said hypothetical clusters and its sharp attenuation between these clusters.

Specific features of magnetic transport in the described conditions are interesting both in terms of a correct understanding of the WL effect as an well as indirect confirmation of the formation of magnetic inclusions without using magnetic alloying elements in thin carbon layers. A research object included graphite layers of the thickness of 35 nm that were produced at the initial stages of synthesis of the so-called vertical graphene nanosheets (VGN) [15–17]. According to the studies [18–21], at a certain stage of synthesis breaks of this layer exhibit nucleus islands of the future VGN phase with the increased defect concentration. These defects can be the PMCs with uncompensated spins and act as a source of induced ferromagnetism.

2. Methodology

The studied structure was grown by microwave-plasma-enhanced chemical vapor deposition (PECVD) in an Innovative Plasma Systems GmbH IPLAS [18–20] installation. The time of carbon deposition onto the substrate for these samples was selected based on the current understanding of the formation process of the VG-structures, which are formulated in the studies [16,17,22], when the first stage of deposition produces the „nuclei" of the nanographite phase. At the second stage, these „nuclei" grow and merge into a thin locally-damaged graphite sublayer that fully covers the substrate. At the third stage, the islands of the VGN phase are nucleated in the most damaged areas of this sublayer. At the fourth stage of growth, a vertical graphene-like VGN structure that is perpendicular to the substrate is formed. According to the formulated task, the PECVD growth was stopped at the third stage in the presented study for the charge carrier transport investigation of the graphite sublayer at the initial (island) stage of VGN nucleation.

The sample surface was studied using a scanning electron microscope (SEM) FEI Quanta 200 FEG with resolution of 1.2 nm, which included a field electron emission source in the Schottky-type electron gun. SEM images in a cross section were obtained using a two-beam system Helios Nanolab 650 [18–20].

Temperature and magnetic-field dependences of layer electrical resistance $R_\square$ and conductivity $\sigma_\square$ were measured within the temperature range $2 < T < 300$ K in a noncryogenic measurement system (Cryogenics Ltd, Great Britain) based on a closed-cycle refrigerator [20,23,24]. The relative magnetoresistive effect was determined by the following relationship $MR(B) = [R_\square(B,T) - R_\square(0,T)]/R_\square(0,T)$.

3. Results

The SEM images of the surface and the cross section are shown in Figure 1 (see also [18–20]). As it is clear in Figure 1, *b*, the surface is a quite homogeneous structure with a lot of non-overlapping bright areas of the sizes of up to 20−30 nm. As it is clear from transverse cleavages in Figure 1, *a*, according to the above-described growth diagram, the sample includes a nanographite buffer C-sublayer of the average thickness of about 35 ± 5 nm, on which the islands of the future VGN-phase are randomly distributed without overlapping.

Prior to starting measurements of the temperature dependences of conductivity (resistance), current-voltage characteristics of the sample were measured at the temperatures of 2 and 300 K (the curves *1* and *2*, respectively, in the inset of Figure 2). The latter were linear [20], thereby indicating ohmic properties of used electrodes.

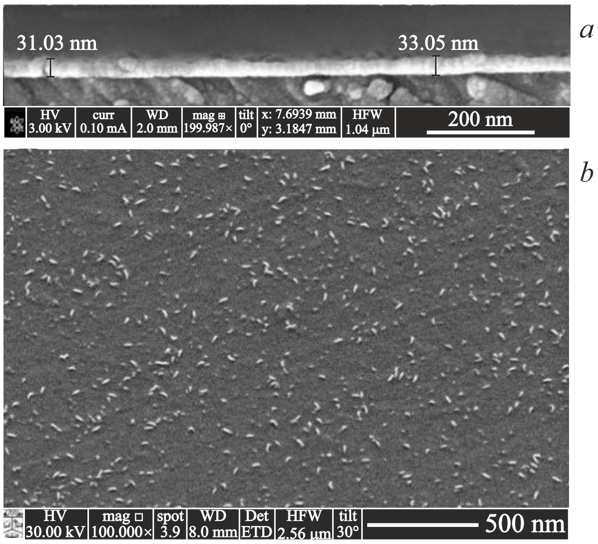

Figure 1. SEM images of the cross section (*a*) and the surface (*b*) of the sample in the backscattered electron mode.

The temperature dependence of layer conductivity $(\sigma_\square(T))$ is shown in Figure 2. The experimental curve (dots) can be described within the framework of QCs to Drude conductivity in the conditions of superposition of the two-dimensional (2D) and three-dimensional (3D) WL corrections [25]:

$$\sigma_\square(T) = \sigma_D(T) - \left(G_0\,\mathrm{Ln}\left(\frac{\tau_{\varphi 2\mathrm{D}}(T)}{\tau(T)}\right)\right)_{2\mathrm{D}} - \left(\mathrm{const}_{3\mathrm{D}} - G_0\left(D\tau_{\varphi 3\mathrm{D}}(T)\right)^{-1/2}\right)_{3\mathrm{D}}, \tag{1}$$

where σ_D is Drude conductivity, T is the temperature, τ is a time of relaxation of electron momentum under elastic scattering, τ_φ is a time of relaxation of the electron wave function (the dephasing time), D is an electron diffusion constant, $G_0 = e^2/(\pi h) \approx 1.23 \cdot 10^{-5}\,\Omega^{-1}$, e is the electron charge, h is the Planck constant.

It is known [26] that τ_φ follows a power-law temperature dependence, $\tau_\varphi = AT^{-p}$ type, where A is a proportionality constant and the value of the exponent p is determined by a mechanism of charge carrier scattering in the WL conditions. In this case, the equation (1) is written as:

$$\sigma_\square(T) = \sigma_D(T) - G_0\,\mathrm{Ln}\left(\frac{A_{2\mathrm{D}}T^{-p_{2\mathrm{D}}}}{\tau(T)}\right) - \mathrm{const}_{3\mathrm{D}} + G_0(DA_{3\mathrm{D}}T^{-p_{3\mathrm{D}}})^{-1/2}, \tag{2}$$

where $p_{2\mathrm{D}}$ and $p_{3\mathrm{D}}$ are parameters p for the 2D and 3D WL corrections, respectively.

Assuming that $\sigma_\square$ weakly depends on the temperature and taking into account that $\tau_\varphi \gg \tau$, it follows that a

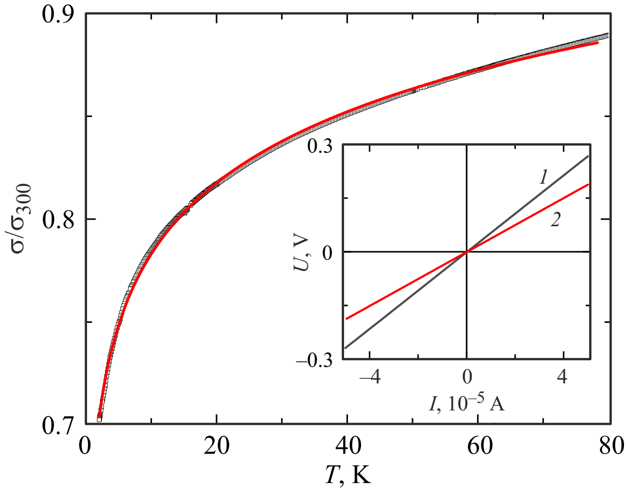

Figure 2. Temperature dependence of layer conductivity of the sample (the black dots) and its approximation by the formula (3) (the red curve). The inset includes current-voltage characteristics at the temperature of 2 (*1*) and 300 K (*2*).

change of the temperature dependence of conductivity will be mainly contributed by the dependence $\tau_\varphi(T)$. If conductivity components that weakly depend on the temperature are designated as

$$\sigma_0 = \sigma_D(T) + G_0\,\mathrm{Ln}(\tau) - G_0\,\mathrm{Ln}(A) - \mathrm{const}_{3\mathrm{D}},$$

then the temperature dependence of conductivity in the WL conditions will be written as:

$$\sigma(T) = \sigma_0 + p_{2\mathrm{D}} G_0\,\mathrm{Ln}(T) + \sqrt{DA_{3\mathrm{D}} G_0^2}\,T^{p_{3\mathrm{D}}/2}. \qquad (3)$$

It is clear from Figure 2 that the experimental data are well described by the equation (3) (the red curve in Figure 2) with an exponent $p_{2\mathrm{D}} = 1$ and $p_{3\mathrm{D}} = 1$, which correspond to a mechanism of elastic scattering by impurities [26]. At the same time, the prevailing mechanism that is responsible for the temperature change of conductivity is a contribution by the two-dimensional quantum correction (the second summand in the relationship (3)).

The mechanisms of conductivity were refined by additionally studying the relative magnetoresistive effect in the longitudinal and the transverse magnetic field. Within the temperature range up to 10 K and with longitudinal orientation of B in relation to a plane of the studied carbon sublayer (Figure 3, the black squares (1)), the curves $MR(B)$ exhibit positive magnetoresistance (PMR) due to the impact by the Lorentz force on the drift of the charge carriers. It means that the thickness of the carbon sublayer allows displacement/deviation of the charge carriers in a direction perpendicular to the sample plane under the effect of the magnetic field oriented within the sample plane.

As known, in the WL conditions, application of the magnetic field at a fixed temperature shall attenuate the QC contribution to Drude conductance, which is manifested as NMR. Indeed, as it is clear from Figure 3 (the blue circles (2)), in case of perpendicular orientation of the magnetic curve a progress of the curve $MR(B)$ can be described by the Altshuler–Aronov theory for interference-induced QCs to Drude conductivity [4] (the green curve (3)):

$$\frac{\Delta\rho_\square(B,T)}{\rho_\square(0,T)} = -\frac{e^2}{\pi h \rho_\square(0,T)} F(x), \qquad (4)$$

where

$$F(x) = \ln(x) + \psi(0.5 + x^{-1}), \qquad (5)$$

ψ is a digamma function. Here, the parameter $x = B/B_\varphi$ is determined by a ratio of induction of the external magnetic field B to a certain characteristic field B_φ that depends on a nature of the process of charge carrier scattering.

As it is clear from Figure 3, the field dependence of relative magnetoresistance is well described by the equation (4). However, the NMR increase is observed up to the magnetic field values of about 8 T, whereas according to the theory [4] the NMR effect in the WL theory shall be suppressed at very weak values $B \sim 100{-}200$ mT. In addition to shifting of the NMR effect into the strong magnetic fields, there is an overestimated value of the parameter $B_\varphi \approx 1$ T, which in fact shall be close to $B_\varphi \approx 0.05$ T [23]. To explain the above, we can use the above-described hypothesis, which is shown in Figure 4 as schematic diagrams that show the conditional paths of the charge carrier motion along the graphite sublayer, including between the VGN-phase island clusters, both in the zero magnetic field (Figure 4, *a, b*) and in the magnetic field directed either parallel (Figure 4, *c* and *d*) or perpendicular (Figure 4, *e* and *f*) to the sample plane.

According to the diagram in Figure 4, *a, b*, the following path portions of the two types are selected in the zero magnetic field on the current lines (the red current line *j*)

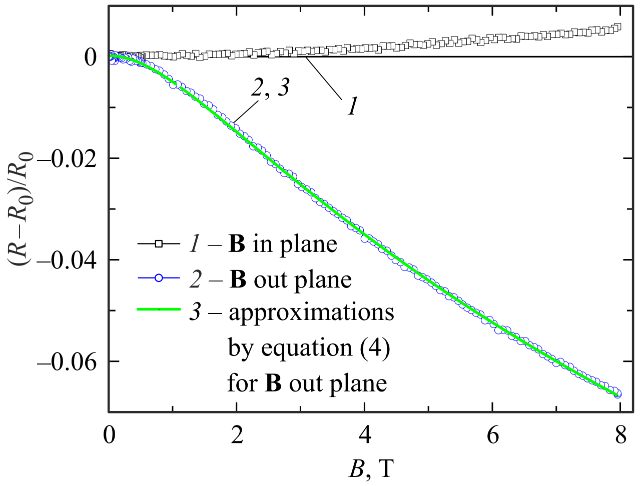

Figure 3. Field dependence of relative magnetoresistance $MR(B)$ for longitudinal (*1*) and perpendicular (*2*) orientation of the magnetic field in relation to the plane of the carbon sublayer at temperature of $T = 7$ K. The green curve (*3*) is approximations by the equation (4) at perpendicular orientation of the magnetic field.

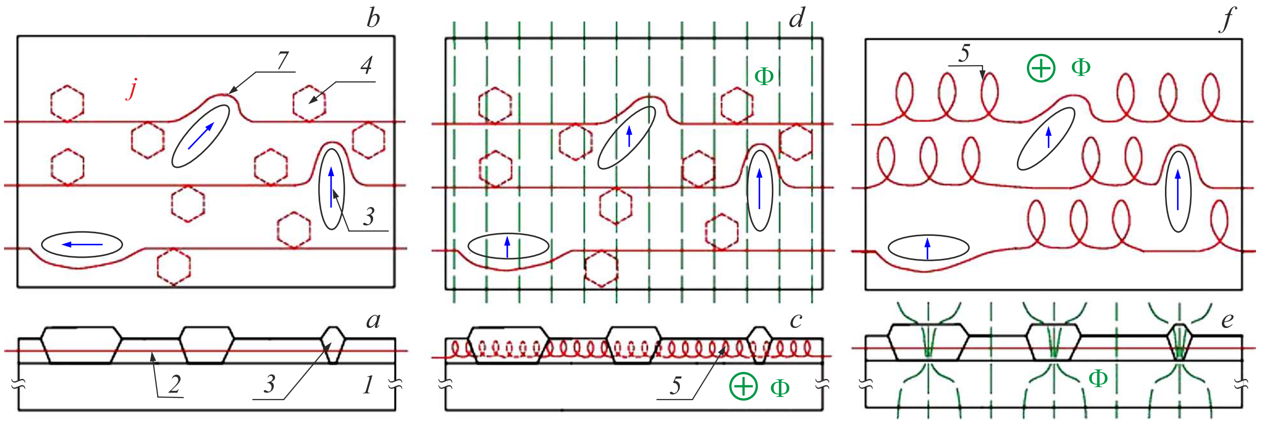

Figure 4. Schematic image of charge carrier transport in the sample without the magnetic field (a, b) or with orientation of the vector B along (c, d) or perpendicular (e, f) to the plane of the carbon sublayer. *1* — the substrate, *2* — the conducting nanographite sublayer, *3* — the VGN-phase islands with clusters of paramagnetic centers (defects), *4* — self-intersecting carrier paths that provide a contribution to conductivity by the WL mechanism, *5* — carrier motion under the Lorentz force, *7* — paths that envelop the VGN-phase islands. The magnetic flux Φ is designated by the green lines, the current lines j are designated by the red lines, the blue arrows (*6*) inside the VGN-phase islands (*3*) indicate magnetic moments M from the clusters enriched by the paramagnetic centers in the islands.

during the motion along the nanographite sublayer in the WL conditions under the effect of the electric field: (1) the path portions of the 4 type, which correspond to charge carrier diffusion along circular or self-intersecting paths, which is described by the Altshuler–Aronov theory of quantum corrections [4] and (2) the current line portions of the 7 type, which symbolize electron scattering on the most damaged (defective) portions of the graphite sublayer near the VGN islands and result in their envelopment by electrons (Figure 4, *b*).

When the sample is placed in the longitudinal magnetic field (Figure 4, *d*), with the magnetic flux Φ being parallel to the graphite-sublayer plane the areas of the 4 type with the self-intersecting paths, which are caused by the WL effects, and the paths with envelopment of the VGN-phase islands are preserved. At the same time, cycloids of the 5 type, which are oriented perpendicular to the graphite-sublayer plane (Figure 4, *c*), appear due to the influence of the Lorentz force on carrier motion (because of mutual perpendicularity of the vectors of current and the magnetic field), which is manifested as PMR on the dependences $MR(B)$ at the low temperatures.

When the magnetic flux Φ is directed normal to the graphite-sublayer plane and corresponds to wave-function-dephasing values of the magnetic fields B (Figure 4, *e* and *f*), then, quantum corrections according to the theory of WL interference effects are suppressed (a role of electron interference vanishes from the self-intersecting path portions of the 4 type) and the latter is manifested as NMR on the curves $MR(B)$. At the same time, the effect of the Lorentz force enables cycloidal current line portions (of the 5 type) that lie in the graphite-sublayer plane. The current line portions of the 7 type, which symbolize a bend of the current lines around the most damaged graphite-sublayer portions near the VG islands are preserved as well.

A phenomenological hypothesis that is schematically shown in Figure 4 and above described charge carrier motion along the graphite sublayer with the islands makes it possible to explain divergence between the experiment and the theory, interpreting it based on heterogeneous distribution of the density of the magnetic flux Φ over an area of the graphite sublayer shown in Figure 4, *e*.

We remind that this hypothesis is based on a representation of existence of heterogeneous distribution Φ due to the high concentration of paramagnetic defects (see Figure 2, *c* [21]), thereby resulting in formation of the ferromagnetic moment around the nucleus islands around the VGN phase. As a result, real strength of the magnetic field B between the islands becomes extremely small (as compared to the average field externally created by a superconducting solenoid), corresponding to a prediction of the theory of quantum corrections to Drude conductivity in the carbon sublayer. As a result, the overestimated value of the parameter B_φ, which is calculated by us based on the WL theory from the equation (6), as well as shifting of the NMR effect from WL into the much higher fields are seeming, so is no observance of the PMR effect in our measurements.

4. Conclusion

It was found in the graphite layers of the thickness of 35 nm with inclusions of the VGN-phase islands, which are produced by microwave-plasma-enhanced chemical vapor deposition (PECVD), at the initial stages of synthesis of the vertical graphene nanosheets, that the temperature curves of conductivity were described by Drude conductivity in the conditions of weak localization (as a combination of the 2D and 3D quantum corrections). It is shown that the field dependence of relative negative magnetoresistive (NMR) effect is described by the equation for weak localization with the highly overestimated value of the parameter $B_\varphi \approx 1\,\mathrm{T}$ at the temperature of $T = 7\,\mathrm{K}$.

It is hypothesized to explain causes of observation of the NMR effect in the quite strong magnetic fields ($> 1\,\mathrm{T}$), which is formally described within the theory of quantum corrections (in the conditions of weak localization). A key provision of the hypothesis is origination of high heterogeneity in distribution of the density of the magnetic flux Φ over the sample. This heterogeneity is caused by presence of the nanoscale areas with the high concentration of the paramagnetic centers (PMC) with uncompensated spins. Due to exchange interaction, the PMC spins in such areas can be aligned in parallel, forming locally-ferromagnetic ordering even without involvement of the ions of the ferromagnetic materials. It induces sharp space heterogeneity of Φ: a strong concentration of the magnetic flux inside the areas with the high PMC concentration and its significant attenuation in spaces between them.

Funding

The study was financially supported by the State Program of Scientific Research of the Republic of Belarus „Photonics and electronics for innovations“ for 2021–2025 subprogram „Micro- and nano-electronics“, Agreement No. 20212560.

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] X. Zhang, Q. Xue, D. Zhu. Physics Letters A **320**, 471 (2004).

[2] Y. Hishiyama, H. Irumano, Y. Kaburagi, Y. Soneda. Phys. Rev. B **63**, 245406 (2001).

[3] Y. Kaburagi, Y. Hishiyama. Carbon **36**, 1671 (1998).

[4] E. McCann, K. Kechedzhi, V.I. Fal'ko, H. Suzuura, T. Ando, B. Altshuler. Phys. Rev. Lett. **97**, 146805 (2006).

[5] O. Mermer, G. Veeraraghavan, T. Francis, Y. Sheng, D. Nguyen, M. Wohlgenannt, A. Köhler, M.K. Al-Suti, M.S. Khan. Phys. Rev. B **72**, 205202 (2005).

[6] A.S. Alexandrov, V.A. Dediu, V.V. Kabanov. Phys. Rev. Lett. **108**, 186601 (2012).

[7] E. Andrade, F. López-Urías, G.G. Naumis. Phys. Rev. B **107**, 235143 (2023).

[8] C. Zhao, Q. Huang, L. Valenta, K. Eimre, L. Yang, A.V. Yakutovich, W. Xu, J. Ma, X. Feng, M. Juríček, R. Fasel, P. Ruffieux, C.A. Pignedoli. Phys. Rev. Lett. **132**, 046201 (2024).

[9] R. Sakano, T. Hata, K. Motoyama, Y. Teratani, K. Tsutsumi, A. Oguri, T. Arakawa, M. Ferrier, R. Deblock, M. Eto, Kensuke Kobayashi. Phys. Rev. B **108**, 205147 (2023).

[10] A.I. Romanenko, O.B. Anikeeva, A.V. Okotrub, L.G. Bulusheva, V.L. Kuznetsov, Yu.V. Butenko, A.L. Chuvilin, C. Dong, Y. Ni. FTT **44**, *3*, 468 (2002). (in Russian).

[11] R. Ur Rehman Sagar, X. Zhang, J. Wang, C. Xiong. J. Appl. Phys. **115**, 123708 (2014).

[12] K. Kusakabe, M. Maruyama. Phys. Rev. B **67**, 092406 (2003).

[13] P. Esquinazi, Handbook of magnetism and advanced magnetic materials, vol. 4 (2007).

[14] J. Červenka, C. Flipse. Phys. Rev. B **79**, 195429 (2009).

[15] S. Ghosh, K. Ganesan, S. R. Polaki, T. Ravindran, N.G. Krishna, M. Kamruddin, A. Tyagi. J. Raman Spectrosc. **45**, 642 (2014).

[16] S.K. Behura, I. Mukhopadhyay, A. Hirose, Q. Yang, O. Jani. Phys. Status Solidi A **210**, 1817 (2013).

[17] J. Zhao, M. Shaygan, J. Eckert, M. Meyyappan, M.H. Rummeli. Nano Lett. **14**, 3064 (2014).

[18] E. Rajackaite, D. Peckus, R. Gudaitis, M. Andrulevičius, T. Tamulevičius, D. Volyniuk, Š. Meškinis, S. Tamulevičius. Surface and Coatings Technology **395**, 125887 (2020).

[19] E. Rajackaite, D. Peckus, R. Gudaitis, T. Tamulevičius, S. Meškinis, S. Tamulevičius. Surfaces and Interfaces **27**, 101529 (2021).

[20] A.A. Maksimenko, E. Rayatskaite, Sh. Meshkinis, T. Tamulevichyus, S. Tamulevichyus, A.A. Kharchenko, A.K. Fedotov, Yu.A. Fedotova. Zhurnal BGU. Fizika **3**, 89 (2020). (in Russian).

[21] L. Zhang, Z. Sun, J.L. Qi, J. Shi, T. Hao, J. Feng. Carbon **103**, 339 (2016).

[22] S. Ghosh, K. Ganesan, S. Polaki, T. Mathews, S. Dhara, M. Kamruddin, A. Tyagi. Appl. Surf. Sci. **349**, 576 (2015).

[23] Yu.A. Fedotova, A.A. Kharchenko, A.K. Fedotov, M.V. Chichkov, V.D. Malinkovich, A.O. Konakov, S.A. Vorob'eva, Yu.V. Kasyuk, V.E. Gumennik, M. Kula, M. Mitura-Nowak, A.A. Maksimenko, J. Przewoźnik, Cz. Kapusta. FTT **62**, *2*, 368 (2020). (in Russian).

[24] A. Fedotov, V. Shepelevich, I. Svito, V. Sivakov. Phys. Rev. B **97**, 075204 (2018).

[25] T.A. Polyanskaya, Yu.V. Shmartsev. FTP **23**, *1*, 3 (1989). (in Russian).

[26] V. Pudalov. Proceedings of the International School of Physics „Enrico Fermi“ **157**, 335 (2004).

Translated by M.Shevelev