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PACS 77.22, 77.70

RIVAL MECHANISMS OF HYSTERESIS IN THE RESISTIVITY OF GRAPHENE CHANNEL

A model for rival mechanisms of hysteresis that appears in the dependence of the resistivity of graphene channels created on substrates of various nature on the gate voltage has been developed. Two types of hysteresis were distinguished: direct (associated with the presence of adsorbates with dipole moments on the surface and the interface) and inverse (associated with the capture of charge carriers from the graphene layer by the localized states at the interface graphene–substrate). A capability of discerning between those channels by varying the rate of gate voltage sween was discussed. A good agreement is obtained between our theoretical predictions and the experimental data available in the literature.

Keywords: graphene, mechanisms of hysteresis, adsorbates, surface dipoles.

1. Introduction

One of the remarkable properties of graphene, which was obtained for the first time in 2004 [1], consists in a symmetry of its band spectrum with respect to the Dirac point and a resulting symmetry in the dependence of the graphene conductivity (resistivity) on the gate voltage [2] with respect to the zero bias (when the Fermi level crosses the Dirac point). However, such a perfect symmetry of current-voltage characteristics with respect to the zero bias voltage at the gate is observed rather rarely for graphene channels in real graphene structures. As a rule, a shift of the voltage that corresponds to the electroneutrality point from zero is observed, which is induced by a certain "chemical" doping of graphene specimens in the course of their fabrication. At the same time, a hysteresis in the dependence of the specimen resistance (conductance) on the gate voltage was repeatedly observed in experiments for graphene channels on substrates of various nature.

Attention to hysteresis phenomena in the resistance of one- and multilayered graphene channels substantially grew after the intense study of graphene on ferroelectric substrates has been started (see review [3] and references therein). The presence of a hysteresis loop in the dependence of the ferroelectric polarization on the applied field strength allows neatly distinguishable states with different resistances of graphene "channel" (states "0" and "1" for logic elements) to be created. For the first time, such a logic element was created on the basis of graphene deposited onto a substrate made of a liquid-crystal ferroelectric polyvinylidene fluoride–trifluoroethylene (PVDF–TrFE) [4]. Later, the fabrication technology of such elements was improved [5, 6]. However, from the viewpoint of practical applications, their shortcoming consists in the necessity to apply considerable switching voltages to the gate (some tens of volts), which is a consequence of a large coercive field that is necessary to vary the polarization direction in the ferroelectric PVDF–TrFE.

From this viewpoint, the application of a substrate made of ferroelectric ceramics $\mathrm{Pb(Zr}_x\mathrm{Ti}_{1-x})\mathrm{O}_3$ (PZT) turned out a fortunate alternative. It is a high-technology substrate characterized by a very high dielectric permittivity (up to 3850 near the morphotropic phase border at $x = 0.52$ [7]). Rather reliable systems with two stable states, which may be taken as a principle of operation for elements of the ferroelectric-based nonvolatile memory of a new generation, have already been created on the basis of one- and multilayered graphene on the PZT substrate. An opportunity of using one- and multilayered graphene on the ferroelectric substrate for the creation of an efficient modulator of radiation in the near and mid-infrared range for on-chip optical interconnections was also demonstrated [11].

In work [12], a model was proposed which considers the capture of charge carriers by states at the interface graphene–ferroelectric and the screening of the electric field in the substrate by their charges (with a corresponding reduction of the free carrier concentration in graphene). This simple model allowed the behavior of the resistance in a graphene channel with gate voltage variation, which was observed in the experiment [8], to be described qualitatively. In work [13], the finite energy width of the impurity state band was additionally taken into account, which allowed us to explain the increase of "memory window" followed by its saturation as the switching voltage at the gate grows.

However, the hysteresis phenomena in the graphene channel were observed not only for systems on a ferroelectric substrate, but for usual and SiO_2 substrates most widespread for today [14–18]. It should also be emphasized that the p-doping at the zero gate voltage is often observed in graphene on a quartz substrate. The interpretations of those results in different works are substantially different.

Really, in the ideal "doped-by-gate" graphene, the electroneutrality point (it takes place when the Fermi level crosses the Dirac point in the band spectrum) corresponds to the zero gate voltage V_g (see, e.g., work [19]). However, the majority of graphene-based field-effect transistors usually reveal the p-doping (i.e. the electroneutrality point lies at a positive V_g), and, at the same time, the dependences of their channel resistance on V_g have a hysteresis loop. Moreover, the physical origins of those two phenomena can be different. In particular, the p-doping can be induced both by adsorbates located on the free surface of graphene and/or at the interface graphene–quartz [14, 15] and by electrochemical processes with the participation of graphene [16, 17]. At the same time, two different types of hysteresis can be observed: direct or inverse [15] (or, in the terminology of work [18], negative or positive). The direct hysteresis corresponds to a displacement of the electroneutrality point toward negative V_g's, when the gate voltage changes consecutively firstly to positive and then to negative V_g. The inverse hysteresis corresponds to a displacement of the electroneutrality point toward positive V_g. In work [15], the direct hysteresis was associated with polarization effects with the participation of adsorbates on the graphene surface, and the inverse one with the capture of charge carriers from graphene by traps at the interface and a subsequent screening of the field in the substrate by that bound charge. Note that the latter mechanism is completely similar to that, the quantitative model of which was developed in works [12, 13].

In particular, in a damp environment, there is a layer of adsorbed water at room temperature [14, 15] or ice at low temperatures [15] on the graphene surface, with the adsorbed water molecules possessing the dipole moment. As to traps, their role can be played by the interface states [15]. Till now, the dependences of the hysteresis on such factors as the regimes of graphene fabrication, heating or annealing of a system, the rate of gate voltage switching, the usage of heavy water instead of ordinary one, and so forth have been studied experimentally [18]. However, no quantitative model, which would allow the transition from the hysteresis of one type to another one to be described, has been developed till now.

In this work, we have developed such a model considering both the adsorbed layer of dipoles on the free graphene surface and the capture of charge carriers from graphene. The comparison of predictions given by the theory with available experimental data has also be done.

2. Theoretical Model

In what follows, we assume that there is a layer of adsorbed molecules – e.g., of electrolyte, water, or ice – on the free graphene surface. The molecules are dipoles, and, owing to some physical and chemical reasons [15], they are oriented by their negative charges toward the graphene layer (Fig. 1, *a*). This means that, provided the zero bias voltage at the gate, there is a 2D concentration of holes in graphene, which is related to the polarization of the adsorbed layer of molecules by the simple relation

$$n_{po} = P_o/e. \tag{1}$$

We also assume that, up to a certain value of positive voltage at the gate, this polarization remains constant. (This voltage value is connected with a critical electric field near the graphene surface (Figs. 1, *a* and *b*). The field itself is determined by the geometry of the problem, because, in effect, the graphene channel does not cover the whole substrate surface, and the electric field lines pass around its sides to terminate at charges on the free graphene surface.) Note that

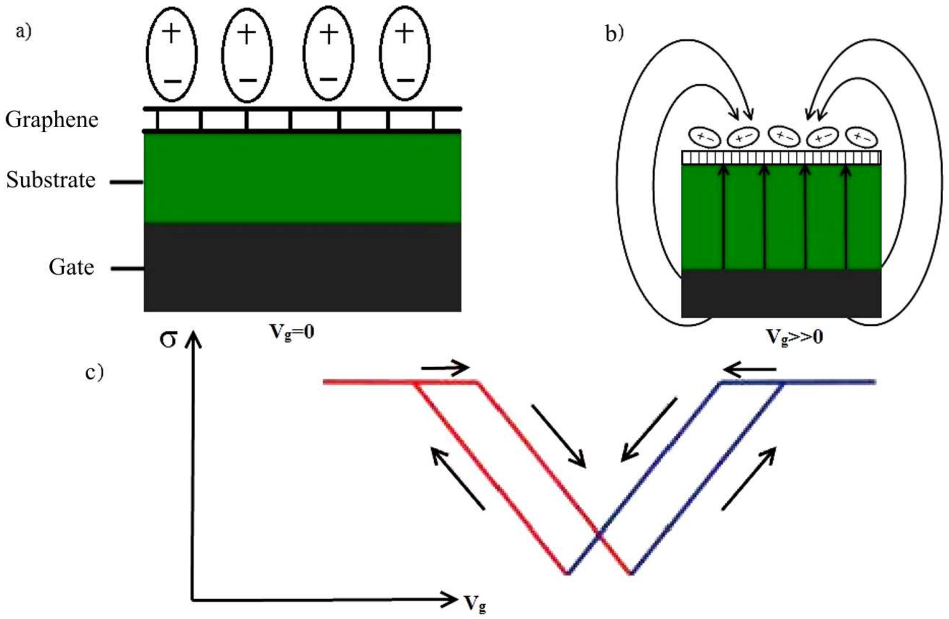

Fig. 1. "Direct" hysteresis in graphene. Dipoles in the adsorbate layer are so oriented that they "dope" the graphene channel with holes at zero V_g (*a*). After the critical value of electric field has been achieved, the polarization in the adsorbate layer becomes destroyed (*b*). Dependences of the concentration and the conductivity on the gate voltage create a closed hysteresis loop (*c*)

this assumption corresponds to the nonlinear dynamics of dipoles bound with the graphene surface (for free dipoles, the orientation of which is described by the Langevin laws, this effect does not take place). Then, the concentration of free carriers in graphene is described as follows:

$$n = \kappa V_g/4\pi ed - n_{po}, \tag{2}$$

where d is the substrate thickness, and κ the substrate dielectric permittivity. The electroneutrality point ($n = 0$) corresponds to a positive voltage equal to

$$V_{NN} = 4\pi edn_{po}/\kappa = 4\pi dP_o/\kappa. \tag{3}$$

At voltages higher than the critical one, the polarization becomes destroyed (see Fig. 1, *b*); for simplicity, we consider it to equal zero. Then, the concentration is described by the usual formula for the plane capacitor:

$$n = \kappa V_g/4\pi ed. \tag{4}$$

If the gate voltage changes rapidly and the polarization has no time to restore spontaneously, the concentration reaches the electroneutrality point at $V_g = 0$. A further increase of the negative gate voltage gives rise to that the polarization P_o ultimately becomes restored, and the loop of direct hysteresis gets closed (Fig. 1).

There are plenty of models for the critical field in the dipole layer. We use the simplest static model, in which the relative concentration of dipoles in this layer, δ, depends nonlinearly on the applied voltage V [20, 21]; namely,

$$V(\delta) = V_S - (RT/F)(\ln(\delta/(1-\delta)) - J(\delta - 0.5)). \tag{5}$$

The quantity J in Eq. (5) has the sense of a dimensionless exchange constant, which equals identically zero in the "ideal" linear case; the relative concentration falls within the interval $0 < \delta < 1$, and the actual concentration is determined by the relation $N = N_0\delta$; R is the universal gas constant; F the Faraday constant; and V_S is a certain equilibrium value corresponding to the steady state of dipoles (for the sake of simplicity, all changes can be reckoned from this state). In the dynamic case, the dependence $V(\delta)$ transforms into the relaxation equation [21]

$$-\frac{1}{\tau}\frac{\partial\delta}{\partial t} + \ln\left(\frac{\delta}{1-\delta}\right) - J\left(\delta - \frac{1}{2}\right) = \theta, \tag{6a}$$

where τ is the characteristic time of dipole relaxation. We also introduced the dimensionless voltage $\theta = (V - V_S)\, F/(RT)$. From Fig. 2, one can see that a well-pronounced bistable behavior is observed at positive $J > J_{\mathrm{cr}}$, where $J_{\mathrm{cr}} = 4$. This fact has an elementary explanation, because

$$\ln\left(\frac{\delta}{1-\delta}\right) \approx 4\left(\delta - \frac{1}{2}\right) + \frac{16}{3}\left(\delta - \frac{1}{2}\right)^3 + \frac{64}{5}\left(\delta - \frac{1}{2}\right)^5$$

and Eq. (6a) has the form

$$-\frac{1}{\tau}\frac{\partial \delta^*}{\partial t} - (J-4)\,\delta^* + \frac{16}{3}\delta^{*3} = \theta \tag{6b}$$

in a vicinity of the transition point $\delta^* = \delta - \frac{1}{2}$.

Hence, our assumption can correspond to a situation described by formula (5) with a large enough value of exchange constant J, when the hysteresis loop in Fig. 2 is characterized by rather a high coercive field $E_C = (J-4)^{3/2}/6$. However, since the steady state energy of dipoles oriented as shown in Fig. 1,*a* is lower by ΔE than the same energy for the orientation shown in Fig. 1,*b*, a spontaneous restoration of the dipole polarization $P(t) \propto eaN_0\delta^*(t)$, where a is the arm of an elementary dipole, will take place starting from the time moment when the electric field strength becomes lower than the critical value E_C. The restoration formula reads

$$P(t) = P_o[1 - \exp(-t/\tau)]. \tag{7}$$

In Eq. (5), we adopted for simplicity that the relaxation is characterized by a unique time τ (this is a natural approximation for a simple dipole layer; but, generally speaking, it is rarely realized in real systems [22]). However, in the framework of this simple approximation, the voltage distance between the electroneutrality points in the hysteresis loop (Fig. 1) is determined by the relation

$$|V_{NN}^1 - V_{NN}^2| = 4\pi d[P_o - P(t)]/\kappa =$$

$$= 4\pi d P_o \exp(-t/\tau)/\kappa. \tag{8}$$

Therefore, if the switching time in the system becomes of the same order of magnitude as the relaxation time τ, the hysteresis loop diminishes and ultimately vanishes. For the geometry exhibited in Fig. 1, $\kappa = (\varepsilon_S + \varepsilon_l)/2$ in formula (8) in the quasistatic case where ε_S and ε_l are the dielectric permittivities of the substrate and the medium over graphene, respectively.

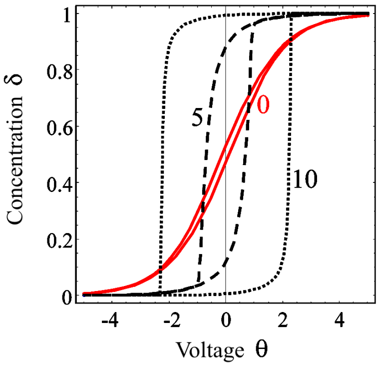

Fig. 2. Dependences of the relative concentration of dipoles (it is proportional to their polarization) on the applied voltage. The numbers near the curves correspond to the values $J = 0$, 5, and 10. The frequency of applied voltage variation $\omega = 0.1/\tau$

To describe the phenomenon of the inverse hysteresis, let us use the approach developed in works [12, 13]. Here, we consider a monoatomic graphene layer doped by means of the gate voltage. The dependence of the Fermi energy in graphene on the concentration of free charge carriers n is given by the known relation (see, e.g., work [19])

$$E_{\mathrm{F}} = \hbar v_{\mathrm{F}} (\pi n)^{1/2}, \tag{9}$$

where $v_{\mathrm{F}} = 10^8$ cm/s. Suppose that there are localized states with the energy E_T and the 2D concentration n_T at the interface graphene–substrate. While V_g increases in the interval for which $E_{\mathrm{F}} < E_T$, the concentration of charge carriers in graphene is determined by expression (4). However, at $E_{\mathrm{F}} = E_T$, the electrons from graphene start to occupy the interface states, and, as was shown in work [13], this process of state filling takes place in the voltage interval

$$\frac{4\pi ed}{\kappa}\frac{E_T^2}{\pi\hbar^2 v_{\mathrm{F}}^2} \le V_g < \frac{4\pi ed}{\kappa}\frac{E_T^2}{\pi\hbar^2 v_{\mathrm{F}}^2} + \frac{4\pi ed n_T}{\kappa}. \tag{10}$$

The negative charge bound at filled interface states screens the field in the substrate. Therefore, the concentration of free charge carriers in graphene remains constant,

$$n = \frac{E_T}{\sqrt{\pi}\hbar v_{\mathrm{F}}}, \tag{11}$$

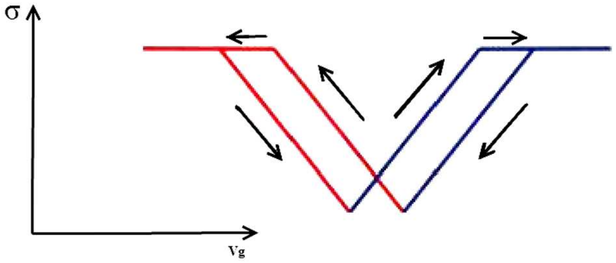

Fig. 3. "Inverse" hysteresis of the conductivity in graphene. The direction of the hysteresis loop path-tracing is opposite to that exhibited in Fig. 1, *c*

in the voltage interval (10). At higher voltages, when all interface states are already filled by electrons, the evident relation takes place,

$$n(V_g) = \kappa V_g/4\pi ed - n_T. \tag{12}$$

The next assumption of works [12, 13] consists in that the lifetime of electrons at the interface states considerably exceeds the switching time for the system. Therefore, relation (12) remains valid even if the gate voltage diminishes, because the charge carriers captured by the states remain at them, although the Fermi level in graphene becomes lower than those states on the energy scale.

Hence, the dependence of the concentration n on the voltage V_g has the form shown in Fig. 3. Here, the arrows mark the direction of voltage sweep (increase or decrease). If the voltage diminishes after the switching, the value of n reaches the electroneutrality point (the Dirac point) at the positive voltage

$$V_{NP} = 4\pi ed n_T/\kappa \tag{13}$$

determined by the concentration of interface states n_T. Note that the n-values to the left from the Dirac points in Fig. 3 correspond to the hole concentrations. At a certain high negative V_g (the specific value depends on the model of localized states), the captured electrons should ultimately recombine with holes in the graphene layer. Unfortunately, the data on the microscopic nature of level potential, which would allow a more exact model of this process to be developed, are absent. Therefore, the assumption was made in work [13] that the dependence of the free charge carrier concentration n on the gate voltage is symmetric with respect to the voltage $V_{NP}/2$; i.e. the depopulation of the level starts at a certain negative voltage and terminates at a voltage higher by V_{NP}. Then, the dependence of the concentration on the voltage has form (4) again. Hence, the hysteresis loop in the dependence of the concentration on the gate voltage becomes closed.

It is clear that if the interval of a gate voltage variation is narrow, i.e. if the condition $E_F < E_T$ is satisfied, no hysteresis is observed. The case where the the switching voltage V_{sweep} falls in interval (10), i.e. if the level filling is not complete, is of special interest. In this case, the antihysteresis curve broadens, and the electroneutrality points move away from each other in the voltage scale as V_{sweep} increases. The distance between them is now determined by the relation

$$V_{DP}(V_{\text{sweep}}) = 4\pi ed n_T(V_{\text{sweep}})/\kappa, \tag{14}$$

where $n_T(V_{\text{sweep}})$ is the concentration of interface states filled by electrons before the switching time moment.

Hysteresis in the dependence of the concentration on the gate voltage should have a counterpart in a similar dependence for the total resistivity (recall that it has the dimension of ohms for 2D structures). Really, the total resistivity of a graphene layer is given by the relation

$$\rho(V_g) \approx 1/\sigma(V_g) + 1/\sigma_{\text{intr}}(T) + 1/\sigma_{\min}. \tag{15}$$

The first term in (15) corresponds to the conductivity of doped graphene,

$$\sigma(V_g) = en\mu. \tag{16}$$

In Eq. (16), the concentration n is determined from one of the V_g-dependences obtained above, and μ is the charge carrier mobility. The second term in (15) describes the intrinsic conductivity of graphene. It has to be taken into account, when the Fermi level is located near the electroneutrality point. It should be recalled that the concentration of charge carriers in intrinsic graphene depends on the temperature T as follows:

$$n_{\text{intr}}(T) = \frac{2(kT)^2}{\pi(\hbar v_F)^2}, \tag{17}$$

where k is the Boltzmann constant. According to Eq. (17), the concentration is $n_{\text{intr}} \approx 1.1 \times 10^{11}$ cm^{-2} at room temperature. That is why this term should be taken into account in an interval of about ±0.1 V around the electroneutrality point if the substrate thickness is of the order of 300 nm, and the substrate dielectric permittivity is of the order of 400. If the gate voltages fall within this interval, the conductivity has the same order as that described by expression (16). At last, the value $\sigma_{\min} \approx 4e^2/\hbar$ corresponds to the minimum conductivity near the Dirac point, which has the quantum-mechanical nature (see, e.g., work [19]).

We should emphasize that, if the both mechanisms (of direct and inverse hystereses) act simultaneously, then, according to Eq. (8), the mechanism of direct hysteresis should expectedly dominate at high switching rates (the polarization has no time to restore, and the electroneutrality points in Fig. 1,c are located far away from each other). At the same time, at low switching rates, the polarization restores, and the electroneutrality points approach each other and ultimately merge together. The phenomenon of inverse hysteresis associated with the capture of charge carriers by the interface states can dominate against this background.

3. Comparison with Experiment

In work [15], the hysteresis phenomenon in one- and multilayered graphene obtained using the method of chemical deposition from the vapor phase onto a SiO_2 substrate was studied. The gate voltage was varied in the interval from −80 to +80 V, and the rate of its variation was changed considerably. Figure 4 schematically illustrates the dependences of the graphene channel conductivity on the voltage at the electrolytic gate (the aqueous solution of KCl was used as an electrolyte) for different voltage sweep rates. If the rate of voltage sweep decreases, the distance between the electroneutrality points also decreases until they practically merge together at a rate of 0.0625 V/s. One can see that, at even lower rates, the "inverse" mechanism of hysteresis dominates. Such a behavior is in perfect agreement with the predictions made in the previous section of this work.

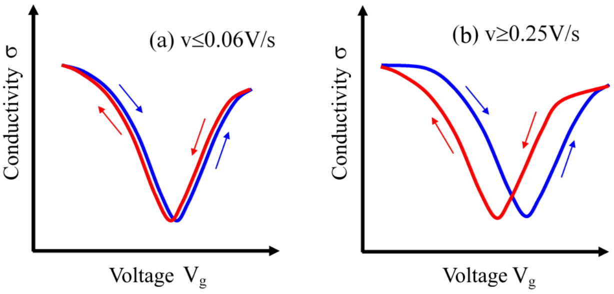

Fig. 4. Schematic plots of the dependences of the graphene channel conductivity on the voltage at the upper electrolytic gate V_g for various rates v of this voltage sweep (according to the experimental data of work [15])

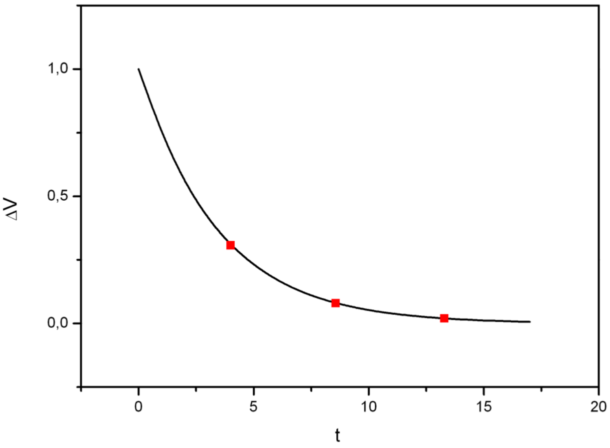

Fig. 5. Dependence of the "memory window" (in volt units) on the time needed for the system to return back from the switching point to the electroneutrality one (in second units): experimental data of work [15] (symbols) and their approximation by formula (8) (solid curve)

In Fig. 5, the dependence of the "memory window" (the distance between the electroneutrality points, $\Delta V = |V_{NN}^1 - V_{NN}^2|$) on the time interval needed for the the system to return back from the switching point to the electroneutrality one is depicted by symbols. Here, we used the experimental data of work [15]. The curve in the same figure shows the approximation of experimental data by formula (8) with $\tau = 3.4$ s and the preexponential coefficient to be approximately 1. (Generally speaking, the preexponential coefficient is more cumbersome in this case than the formula for the plane capacitor (4) predicts. However, we cannot derive its exact form due to the lack of the exhaustive information concerning the geometry of the problem.) It is evident from Fig. 5 that a reduction of the "memory window" as the time interval of surface dipole relaxation increases is described well by the exponential dependence (8). It is worth to note that, owing to a strong coupling between the negative poles of dipoles with the graphene surface (see, e.g., work [23]), the time τ that is in-

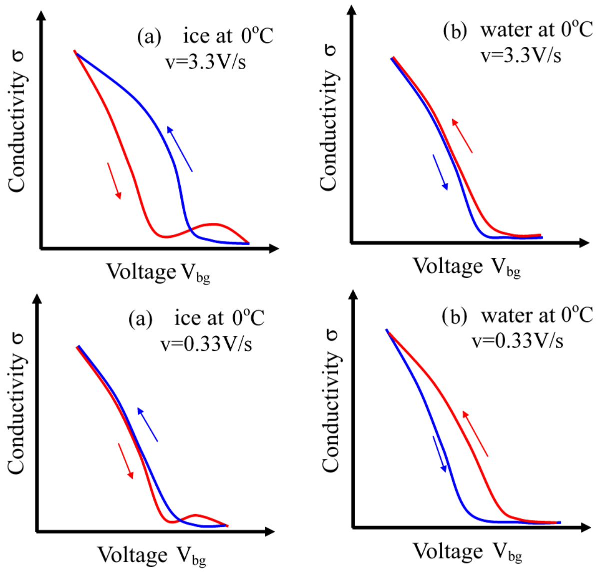

Fig. 6. Schematic plots of the dependences of the graphene channel conductivity on the gate voltage V_{bg} at various rates v of this voltage sweep and at temperatures above (the adsorbate is water) and below (the adsorbate is ice) the freezing point (according to the experimental data of work [15])

cluded into this dependence exceeds, by many orders of magnitude, the relaxation times that are characteristic of the polarization induced by free dipoles and described by a formula of the Langevin type.

At the same time, the relaxation time for the concentration of charge carriers captured by the interface states is much longer. It equals hours at room temperature and tens of days at the nitrogen one [8]. Therefore, on the exponential time scale of work [15], this concentration can be regarded as dependent only on the gate voltage change.

We also examined the dependence of the graphene channel conductivity on the voltage at the standard Si gate at various sweep rates for this voltage both at high temperatures, when the adsorbed water molecules form a water layer, and at temperatures below the water freezing point, when the covering layer is ice. The chemical nature of those molecules is identical, but the mobility is different, namely, it is substantially higher in the liquid and lower in the crystal. This difference explains a discrepancy between the corresponding experimental dependences. In particular, in the case of ice, the direct hysteresis took place at high rates of voltage variation, and the inverse one at low rates. At the same time, in the case of water where the time of spontaneous restoration of a polarization is very short owing to a high mobility of dipoles, the inverse hysteresis was observed at any rate of voltage variation among those that were used in the experiment (Fig. 6).

4. Conclusions

The results of comparison between the experimental data and the theoretical dependences, which was done in the previous section, testify that the model proposed in this work can describe the hysteresis phenomenon in the dependence of the conductivity in graphene channels created on the substrates of various origins on the gate voltage. The model provides a comprehension, at the quantitative level, of parameters inherent to the interface states and the polarization of an adsorbate bound with the free graphene surface and predicts an opportunity to distinguish between two hysteresis types, direct and inverse ones. The former is associated with the repolarization of dipoles bound at the graphene surface. Under experimental conditions, it is water molecules that most often play this role. The latter is connected with the capture of free charge carriers by the interface states. The resolution between the hysteresis types can be made by varying the rate of gate voltage sweep. This becomes possible owing to the hierarchy of spontaneous relaxation times that describe two indicated processes. Namely, these are seconds for the relaxation of the polarization induced by adsorbed dipoles, and hours and days for the relaxation of the concentration of charge carriers captured at the interface states.

The approximations used in this work are standard in the graphene physics and the physics of polarizable media. At the same time, the introduction of the parameter τ for the spontaneous relaxation of a polarization requires a further substantiation, because the approximation applied by us is simplified. The model proposed can be specified on the basis of a deeper understanding of the physicochemical nature of the coupling between adsorbed dipoles and the graphene surface, on the one hand, and the model of creation of localized states at the interface graphene–dielectric substrate, on the other hand. A further experimental research of the hysteresis phenomena in the dependence of the graphene channel conductivity on the gate voltage can provide us with a valuable material for a better understanding of the parameters of this physico-chemical structure.

The work was supported by the State Fund for Fundamental Researches of Ukraine, the National Academy of Sciences of Ukraine, and the Science and Technology Center in Ukraine. The authors are grateful to V.G. Litovchenko for his permanent attention to this work.

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Received 19.12.12.
Translated from Ukrainian by O.I. Voitenko

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КОНКУРЕНТНІ МЕХАНІЗМИ ГІСТЕРЕЗИСУ ОПОРУ В ГРАФЕНОВОМУ КАНАЛІ

Резюме

Побудовано модель для конкурентних механізмів гістерезису залежності опору графенового каналу на підкладці різної природи від напруги на затворі: прямого (зумовленого наявністю на поверхні й інтерфейсі адсорбатів з дипольним моментом) і оберненого (зумовленого захопленням носіїв з шару графену на локалізовані стани на інтерфейсі графен–підкладка). Обговорено можливість дискримінації цих каналів шляхом варіації швидкості зміни напруги на затворі. Відзначено добру відповідність передбачень теорії з наявними в літературі експериментальними даними.