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## **P-N junctions dynamics in graphene channel induced by ferroelectric domains motion**

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### **Abstract**

The p-n junctions dynamics in graphene channel induced by stripe domains nucleation, motion and reversal in a ferroelectric substrate is explored using self-consistent approach based on Landau-Ginzburg-Devonshire phenomenology combined with classical electrostatics. Relatively low gate voltages are required to induce the hysteresis of ferroelectric polarization and graphene charge in response to the periodic gate voltage. Pronounced nonlinear hysteresis of graphene conductance with a wide memory window corresponds to high amplitudes of gate voltage. Also we revealed the extrinsic size effect in the dependence of the graphene channel conductivity on its length. We predict that the considered nanostructure "top gate/dielectric layer/graphene

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channel/ferroelectric substrate" can be a promising candidate for the fabrication of new generation of modulators and rectifiers based on the graphene p-n junctions.

## I. INTRODUCTION

Since the discovery of graphene by Novoselov and Geim [1, 2] its unique electronic, elastic and optical properties [3, 4] are very attracting for fundamental research of 2D semiconductors and promising for advanced application possibilities in modern nanoelectronics, optoelectronics nanoplasmonics, sensors and memory technologies. Following the development of graphene as a paradigm for 2D semiconductors, the remarkable properties of the p-n-junction (**pnJ**) in graphene have been realized experimentally [5, 6, 7] and studied theoretically [8, 9]. The first pnJ in graphene were realized by means of two gates doping of the one region with electrons and the other with holes [5]. The next pnJ in a back gated graphene channel was realized by a top gate superimposed on the dielectric layer above the channel [6]. For all these cases [5-8, 9] the resistance of pnJ is quite low, since the electric field in the pnJ is typically high due to the low screening ability of two-dimensional Dirac quasiparticles (electrons or holes). Several seminal studies of the pnJs operation in graphene were devoted on the physical manifestations of Andreev reflection, Klein tunneling, quantum Hall effect and Veselago lensing [10, 11, 12].

Remarkably the usage of multiple gates doping of graphene channel by the opposite types of carriers is not the only possibility to design pnJs in graphene. The alternative promising and much less explored way was not revealed till Hinnefeld et al [13] and Baeumer et al [14], who created a pnJ in graphene using the ferroelectric substrates  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  and  $\text{LiNiO}_3$ , respectively. Further, Baeumer et al [14] considered the pnJ in graphene induced by a ferroelectric domain wall. The principal idea of the latter work is that if graphene is imposed on a  $180^\circ$ -ferroelectric domain wall, a pnJ can arise without applying any additional gates, doping or screening, due to the charge separation by an electric field of a ferroelectric domain wall – surface junction. The physical origin of pnJ appearance can be understood from Zheng et al [15] and Yusuf et al [16] results, who decided to combine ferroelectrics with graphene, since a pronounced free charge accumulation can take place at the graphene - ferroelectric interface [16]. Using the possibility of hysteretic ferroelectric gating, Zheng et al [15] demonstrated a symmetrical bit writing in graphene-on-ferroelectric FETs with an electro-resistance change of over 500%, as well as a reproducible, nonvolatile switching. Recently graphene-ferroelectric metadevices for nonvolatile memory and reconfigurable logic-gate operations have been proposed [17].

Kurchak and Strikha pointed out that adsorbed charges dynamics leads to the unusual conductivity effects in the graphene channel on an organic ferroelectric substrate [18]. Morozovska et al [19, 20] have shown that the effective screening of the spontaneous polarization discontinuity at the ferroelectric surface by graphene free charge strongly decreases the electrostatic energy of the

graphene-on-ferroelectric and can cause versatile phenomena, such as the charge density modulation induced by the pyroelectric effect at the graphene/ferroelectric interface [19] and the triggering of the space charge modulation in multilayer graphene by ferroelectric domains [20]. Finite-size effects can strongly influence the nonlinear hysteretic dynamics of the stored charge and electro-resistance in the multilayer graphene-on-ferroelectric structures; in this case, the domain stripes of different polarities can induce domains with  $p$ - and  $n$ -type conductivity, and with pnJ potentials [21]. A theoretical model for the electric field and ballistic transport in a single-layer graphene channel at a  $180^\circ$ - ferroelectric domain wall have been developed recently [22]. The results of [22] have been expanded for the case of different types of current regimes in Ref.[23]. In [23] we presented the theory of the conductivity of pnJ in graphene channel, placed on ferroelectric substrate, caused by ferroelectric domain wall for the case of arbitrary current regime: from purely ballistic to diffusive one.

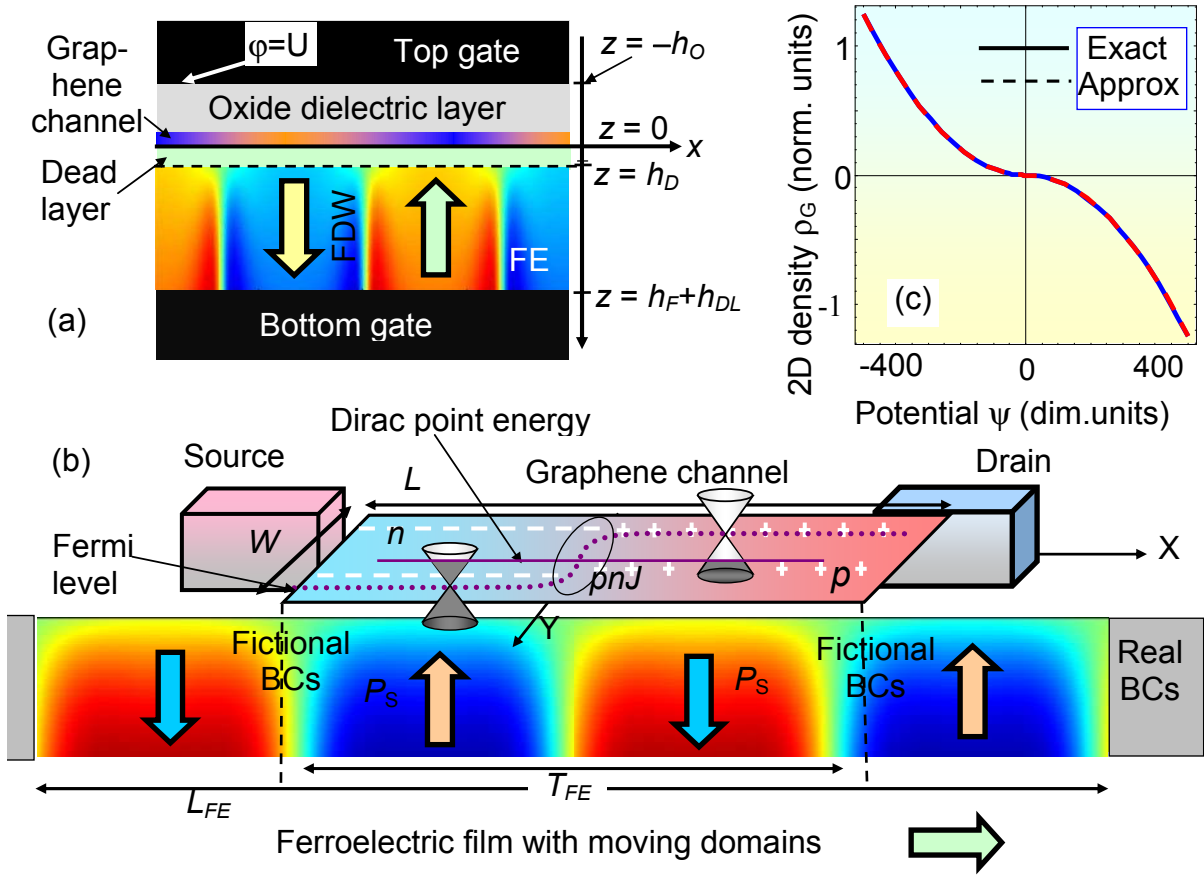
Here, complementary to Refs.[21-23], we consider the multi-domain states dynamics in a ferroelectric substrate. Using a self-consistent approach based on Landau-Ginzburg-Devonshire (**LGD**) phenomenology combined with classical electrostatics, here we study p-n junctions dynamics in graphene channel induced by stripe domains nucleation, motion and reversal in a ferroelectric substrate. We regard that 2D-electrons in a single-layer graphene sheet have a linear Dirac density of states.

## II ANALYTICAL TREATMENT

### A. Problem statement

The geometry of the considered problem is shown in **Fig. 1(a)**. Top gate is deposited on oxide layer; then 2D-graphene layer (channel) is separated from a ferroelectric substrate by ultra-thin paraelectric dead layer, originated due the imperfect deposition process of graphene on the ferroelectric. The ferroelectric substrate is in ideal electric contact with the bottom gate electrode. Periodic voltage is applied to the top gate. The voltage can induce  $180$ -degree ferroelectric domain walls (**FDW**) motion in the ferroelectric substrate.

Schematic of the pnJ created in the graphene channel by domain walls moving in the ferroelectric substrate is shown in **Fig. 1(b)**. Since the lateral dimension of a ferroelectric film  $L_{FE}$  is typically much higher than the graphene channel length  $L$ , odd, even or fractional number of domain walls can pass along the channel during the period of the gate voltage depending on the interrelation between the graphene channel length  $L$  and the period  $T_{FE}$  of the domain structure in a ferroelectric film.



**FIG. 1.** (a) Schematics of the 180°-domain wall structure near the ferroelectric surface in the heterostructure "top gate – oxide dielectric layer – graphene channel – paraelectric dead layer – ferroelectric film – bottom gate". (b) Schematics of the pnJ induced in the graphene channel by domain walls moving in the ferroelectric substrate. (c) Dependence of the 2D charge density of graphene on the dimensionless variable  $\psi = \frac{e\phi + E_F}{k_B T}$ .

Solid curve is exact expression (1), dashed curve is Pade-exponential approximation (2).

Below we describe how we model each of the layers in the hetero-structure.

**Single-layer graphene channel.** We treat a single-layer graphene as an infinitely thin sheet for which that the two-dimensional (2D) electron density of states (DOS) is  $g_n(\epsilon) = g_p(\epsilon) = 2\epsilon/(\pi\hbar^2 v_F^2)$  (see e.g. [24]). Hence the 2D concentration of electrons in the conduction band and holes in the valence band of graphene are  $n_{2D}(\phi) = \int_0^\infty d\epsilon g_n(\epsilon) f(\epsilon - E_F - e\phi)$  and  $p_{2D}(\phi) = \int_0^\infty d\epsilon g_p(\epsilon) f(\epsilon + E_F + e\phi)$ , respectively

( $E_F$  is a Fermi energy level, these expressions correspond the gapless graphene spectrum). For pure planar graphene sheet we introduce a new variable,  $\psi = \frac{e\phi + E_F}{k_B T}$ . Using the variable the graphene charge density  $\sigma_G(\psi) = e(p_{2D}(\psi) - n_{2D}(\psi))$  is equal to

$$\sigma_G(\Psi) = \frac{2(k_B T)^2 e}{\pi \hbar^2 v_F^2} (\text{Li}_2(-\exp(\Psi)) - \text{Li}_2(-\exp(-\Psi))) \quad (1)$$

Here  $\text{Li}_n(z) = \sum_{m=1}^{\infty} \frac{z^m}{m^n}$  is the polylogarithm function (see **Appendix A** of **Suppl. Mat.** [25]).

Corresponding Pade-exponential approximation of Eq.(2) valid for arbitrary  $\Psi$  values is

$$\sigma_G(\Psi) \approx \frac{2(k_B T)^2 e}{\pi \hbar^2 v_F^2} \left( \frac{1}{\eta(\Psi)} - \frac{1}{\eta(-\Psi)} \right), \quad (2)$$

where the function:  $\eta(\Psi) = \exp(\Psi) + 2 \left( \Psi^2 + \frac{\Psi}{2} + \frac{2\pi^2}{12 - \pi^2} \right)^{-1}$ . Since the problem of dielectric

permittivity of 2D-graphene layer is still under debate (see e.g. [26]), our results obtained for the DOS from Eqs.(1) are free from the problems inherent for 3D Boltzmann or Debye-Thomas-Fermi approximations [21, 22].

**Dielectric and dead layers.** Equations of state  $\mathbf{D} = \epsilon_0 \epsilon_O \mathbf{E}$  and  $\mathbf{D} = \epsilon_0 \epsilon_{DL} \mathbf{E}$  relate the electrical displacement  $\mathbf{D}$  and electric field  $\mathbf{E}$  in the oxide dielectric and ultrathin dead layers of thicknesses  $h_O$  and  $h_{DL}$ , respectively,  $\epsilon_0$  is a universal dielectric constant. Despite there are different types of "dead" (or "passive") layers [27, 28], most of them are treated as ultrathin sub-surface layers located under the surface of ferroelectric substrate, where the spontaneous polarization is absent (or negligibly small) due to the surface contamination, zero extrapolation length and/or strong depolarization field [27, 28]. The matter in the dead layer is of almost the same chemical composition than in the bulk of ferroelectric substrate, but it is in the paraelectric phase induced by the surface confinement effect, and so the relative dielectric permittivity of the dead layer  $\epsilon_{DL}$  is rather high  $\sim 10^2$  (as it should be for paraelectrics at room temperature [29]), in comparison with unity for a physical gap. The potential  $\varphi_{DL}$  satisfies Laplace's equation inside the dead layer.

**Ferroelectric substrate.** As a substrate we consider a ferroelectric film of thickness  $l$  with ferroelectric polarization  $P_3^f$  directed along its polar axis  $z$ , with 180-degree domain wall – surface junctions [see **Fig. 1(a)**]. Also we assume that the dependence of polarization components on the inner field  $\mathbf{E}$  can be linearized for transverse components as  $P_1 = \epsilon_0 (\epsilon_{11}^f - 1) E_1$  and  $P_2 = \epsilon_0 (\epsilon_{22}^f - 1) E_2$ . Ferroelectric is dielectrically isotropic in transverse directions, i.e. relative dielectric permittivity are equal,  $\epsilon_{11}^f = \epsilon_{22}^f$ . Polarization  $z$ -component is  $P_3(\mathbf{r}, E_3) = P_3^f(\mathbf{r}, E_3) + \epsilon_0 (\epsilon_{33}^b - 1) E_3$ , where a so-called relative "background" permittivity  $\epsilon_{ij}^b$  is introduced [27]. The values of  $\epsilon_{ij}^b$  are not related with a soft ferroelectric mode and limited by the linear dielectric response of the lattice, so for the most of ferroelectrics perovskites they are within the range (4 – 7) (see Ref.[30] for its determination and refs therein). Hence the ferroelectric permittivity  $\epsilon_{33}^f$  related with the soft mode is much higher than the background,  $\epsilon_{33}^f \gg \epsilon_{33}^b$ . Inhomogeneous spatial distribution of the ferroelectric polarization  $P_3(x, y, z)$  is determined from the time-dependent LGD type Euler-Lagrange equation,

$$\Gamma \frac{\partial P_3}{\partial t} + aP_3 + bP_3^3 + cP_3^5 - g\Delta P_3 = E_3. \quad (3a)$$

$\Gamma$  is a Landau-Khalatnikov relaxation coefficient [31], and  $g$  is a gradient coefficient,  $\Delta$  stands for a 3D-Laplace operator. Corresponding boundary conditions are of the third kind [32],

$$\left( P_3 - \Lambda_+ \frac{\partial P_3}{\partial z} \right) \Big|_{z=h_D} = 0, \quad \left( P_3 + \Lambda_- \frac{\partial P_3}{\partial z} \right) \Big|_{z=h_{DL}+h_F} = 0 \quad (3b)$$

The physical range of extrapolation lengths  $\Lambda_{\pm}$  is (0.5 – 2) nm [33]. Constants  $a = \alpha_T(T - T_C)$ ,  $b$  and  $c$  are the coefficients of LGD potential expansion on the polarization powers (also called as linear and nonlinear dielectric stiffness coefficients). Quasi-static electric field is defined via electric potential as  $E_3 = -\partial\phi/\partial z$ . The potential  $\phi_f$  satisfies Poisson equation inside a ferroelectric film.

Also we suppose that the polarization relaxation time  $\tau_{LK} = \Gamma/|a_3| \sim 10^{-11}$  s is much higher than the graphene charge relaxation time  $\tau_M = \varepsilon\varepsilon_0/(e\eta n_s) \sim 10^{-12}$  s ( $\varepsilon_0$  is a universal dielectric constant,  $\varepsilon$  is a relative dielectric permittivity,  $\eta$  is a mobility of carriers in graphene), and so adiabatic approximation can be used for the charge description [see **Fig.S1** in **Appendix B** of **Suppl. Mat.** [25]]. The inequality  $\tau_{LK} \gg \tau_M$  works well in the vicinity of ferroelectric phase transition, where  $\tau_{LK}$  diverges due to the critical slowing down effect.

Hence, for the problem geometry shown in the **Fig. 1(a)** the system of electrostatic equations acquires the form:

$$\Delta\phi_O = 0, \quad \text{for } -h_O < z < 0, \quad (\text{oxide dielectric layer "O"}) \quad (4a)$$

$$\Delta\phi_{DL} = 0, \quad \text{for } 0 < z < h_{DL}, \quad (\text{dead layer "DL"}) \quad (4b)$$

$$\left( \varepsilon_{33}^b \frac{\partial^2}{\partial z^2} + \varepsilon_{11}^f \Delta_{\perp} \right) \phi_f = \frac{1}{\varepsilon_0} \frac{\partial P_3^f}{\partial z}, \quad \text{for } h_{DL} < z < h_{DL} + h_F. \quad (\text{ferroelectric "f"}) \quad (4c)$$

3D-Laplace operator is  $\Delta$ , 2D-Laplace operator is  $\Delta_{\perp}$ . Boundary conditions to the system (4) are fixed potential at the top ( $z = -h_O$ ) and bottom ( $z = h_{DL} + h_F \approx h_F$ ) gate electrodes; the continuity of the electric potential at the graphene layer ( $z = 0$ ) and the equivalence of difference of the electric displacement normal components,  $D_3^O = \varepsilon_0\varepsilon_O E_3$  and  $D_3^{DL} = \varepsilon_0\varepsilon_{DL} E_3$ , to the surface charges in graphene  $\sigma_G(x, y)$ ; and the continuity of the displacement normal components,  $D_3^f = \varepsilon_0\varepsilon_{33}^b E_3 + P_3^f$  and  $D_3^{DL} = \varepsilon_0\varepsilon_{DL} E_3$ , at dead layer/ferroelectric interface.

Explicit form of the boundary conditions (**BCs**) in  $z$ -direction is:

$$\phi_O(x, y, -h_O) = U(t), \quad (5a)$$

$$\phi_O(x, y, 0) = \phi_{DL}(x, y, 0), \quad D_3^O(x, y, 0) - D_3^{DL}(x, y, 0) = \sigma_G(x, y), \quad (5b)$$

$$\phi_d(x, y, h_{DL}) = \phi_f(x, y, h_{DL}), \quad D_3^{DL}(x, y, h_{DL}) - D_3^f(x, y, h_{DL}) = 0, \quad (5c)$$

$$\varphi_f(x, y, h_{DL} + h_F) = 0. \quad (5d)$$

The gate voltage is periodic with a period  $T_g$ ,  $U(t) = U_{\max} \sin(2\pi t/T_g)$ .

To generate moving domains ferroelectric film thickness should be above the critical thickness  $l_{cr}$  of the size-induced phase transition into a paraelectric phase. The critical thickness of the single-domain ferroelectric state instability strongly depends on the dielectric and dead layer thicknesses, namely [34],  $l_{cr}(T) \approx \frac{1}{\alpha_T(T_C - T)} \left( \frac{2g}{\lambda + L_C} + \frac{h_{DL}}{\epsilon_0 \epsilon_{DL}} + \frac{h_O}{\epsilon_0 \epsilon_O} \right)$ , where  $L_C = \sqrt{g/\epsilon_0 \epsilon_{33}^b} \sim 0.1$  nm is a correlation length in z-direction that is very small due to the depolarization field effect [35, 36].

## B. The "fictional" boundary conditions at the source and drain planes

The domains origin can be energetically favorable above the critical thickness, since they minimize the depolarization field energy in the gap and dielectric layer [37]. The period of domain stripes depends on the thicknesses of oxide dielectric layer  $h_O$ , dead layer  $h_{DL}$  and film  $h_F$  in a self-consistent way.

We note that exact domain dynamics will be determined by the interplay of the driving forces, memory effects in materials due to defects, and parity effects. Here, we consider only the latter, assuming no pinning on the top surface.

Contrary to the popular belief that the controlled motion of a solitary domain wall (or a small number of them) in a ferroelectric film under a channel is relatively easy to carry out, this task is hardly feasible for thin films, which lateral surfaces  $x = \pm L_{FE}/2$  (designated "real BCs" in **Fig.1 (b)**) border with air or solid. First of all, because these surfaces are themselves topological defects, on which a partial or complete pinning of the domain walls occurs, these walls can be "flipped" only hypothetically by applying a very large electrical voltage to the gate that is rather difficult in reality because of the electric breakdown of a thin oxide dielectric layer, as well as of a thin ferroelectric film. In addition, large fields in thin films do not agree with the requirements for energy saving and miniaturization of heterostructures. In reality there are many domain stripes in a thin ferroelectric film, which expand or contract with the film polarization reversal, rather than a "domain" running along the film in a transverse direction. Thus the motion, birth or annihilation of domain walls inside the film are likely, at that the polarization values on the surfaces  $x = \pm L_{FE}/2$  are determined by the screening conditions and pinning centers. In this case, the domain walls naturally move along the channel, but sometimes towards each other.

In this case, it is reasonable to assume general boundary conditions of the third kind for polarization,  $P_3 \pm \lambda_{\pm}^F \frac{\partial P_3}{\partial x} \Big|_{x=\pm \frac{L_{FE}}{2}} = 0$ , with different extrapolation lengths  $\lambda_{\pm}^F$  at different lateral surfaces



of the film  $x = +L_{FE}/2$  and  $x = -L_{FE}/2$ . In general, the values of  $\lambda_{\pm}^F$  are unknown, but, as a rule, they are regarded not negative (including special cases of zero and infinitely large value) [32, 33, 35, 37, 38]. It turns out that for any positive  $\lambda_{\pm}^F$  the domains expand and contract more often when a periodic voltage is applied to the film, rather than a solitary domain wall moves in the transverse direction. The equilibrium period of the domain structure is practically independent of the boundary conditions and is determined by the film thickness  $h_F$ , temperature, and screening conditions on its polar surface  $z = h_{DL}$  [37]. Since for a realistic situation  $L_{FE} \gg L_c$  (where  $L_c \equiv \sqrt{-a/g}$  is a correlation length that is the same order of lattice constant far from Curie temperature) and  $L_{FE} \gg L$ , the solution for the polarization of the film "under" the channel (i.e., at  $-L/2 \leq x \leq L/2$ ) practically does not depend on the specific type of BCs at the surfaces  $x = \pm L_{FE}/2$ . Taking into account that  $L_{FE} \gg L$ , it is possible to perform calculations on a computational cell with a transverse dimension  $-L/2 \leq x \leq L/2$ .

Since the lateral dimension of a ferroelectric film  $L_{FE}$  is typically much higher than the graphene channel length  $L$ , *odd*, *even* or *not fractional* number of domain walls can pass along the channel during the period of the gate voltage  $T_g$  depending on the interrelation between the graphene channel length  $L$  and the period  $T_{FE}$  of the domain structure in a ferroelectric film [**Fig.1(b)**]. The realistic situations can be modelled by a "fictional" boundary conditions (**BCs**) on polarization component  $P_3$ , its derivative  $\frac{\partial P_3}{\partial x}$ , electric potential  $\varphi_f$  and its derivative  $\frac{\partial \varphi_f}{\partial x}$  at the lateral boundaries  $x = \pm L/2$  of the computation box,  $x = \pm L/2$ , which will be listed below.

The *periodic*, *antiperiodic* or *mixed parity BCs* imposed on polarization and electric potential in transverse x-direction model the appearance and motion of different parity (*even* or *odd*, or *fractional*) of pnJs along the channel of length  $L$  over a voltage period, respectively. Appeared that *periodic BCs* for polarization and potential, and their derivatives,

$$P_3\left(-\frac{L}{2}, y, z\right) = P_3\left(+\frac{L}{2}, y, z\right), \quad \left. \frac{\partial P_3}{\partial x} \right|_{x=-\frac{L}{2}} = \left. \frac{\partial P_3}{\partial x} \right|_{x=+\frac{L}{2}}, \quad (\text{periodic}), \quad (6a)$$

$$\varphi_f\left(-\frac{L}{2}, y, z\right) = \varphi_f\left(+\frac{L}{2}, y, z\right), \quad \left. \frac{\partial \varphi_f}{\partial x} \right|_{x=-\frac{L}{2}} = \left. \frac{\partial \varphi_f}{\partial x} \right|_{x=+\frac{L}{2}}, \quad (\text{periodic}), \quad (6b)$$

allow the motion of the even number of domain walls in the ferroelectric and so the even number of pnJs are moving in the channel. Thus the asymmetry of the graphene channel conductance and rectification effect is absent in this case for the reasons discussed below.

In contrast the *mixed parity BCs*,

$$P_3\left(-\frac{L}{2}, y, z\right) = P_3\left(+\frac{L}{2}, y, z\right), \quad \frac{\partial P_3}{\partial x}\Big|_{x=-\frac{L}{2}} = \frac{\partial P_3}{\partial x}\Big|_{x=+\frac{L}{2}} \quad (\text{periodic}), \quad (7a)$$

$$\varphi_f\left(-\frac{L}{2}, y, z\right) = -\varphi_f\left(+\frac{L}{2}, y, z\right), \quad \frac{\partial \varphi_f}{\partial x}\Big|_{x=-\frac{L}{2}} = -\frac{\partial \varphi_f}{\partial x}\Big|_{x=+\frac{L}{2}} \quad (\text{antiperiodic}), \quad (7b)$$

can lead to the asymmetry of the graphene channel conductance and rectification effect, which will be demonstrated in the next section.

The completely *antiperiodic BCs*:

$$P_3\left(-\frac{L}{2}, y, z\right) = -P_3\left(+\frac{L}{2}, y, z\right), \quad \frac{\partial P_3}{\partial x}\Big|_{x=-\frac{L}{2}} = -\frac{\partial P_3}{\partial x}\Big|_{x=+\frac{L}{2}} \quad (\text{antiperiodic}), \quad (8a)$$

$$\varphi_f\left(-\frac{L}{2}, y, z\right) = -\varphi_f\left(+\frac{L}{2}, y, z\right), \quad \frac{\partial \varphi_f}{\partial x}\Big|_{x=-\frac{L}{2}} = -\frac{\partial \varphi_f}{\partial x}\Big|_{x=+\frac{L}{2}} \quad (\text{antiperiodic}), \quad (8b)$$

can lead to the motion of the odd number of domain walls in the ferroelectric and hence the odd number pnJs can move in the channel. The asymmetries of the graphene channel conductance and rectification effect are possible in the case. The physical consequences of the BCs are described below.

One can try to understand the effect of the BCs on the graphene conduction kinetics induced by the moving domain walls using the analytical results [22, 23] for a thermodynamic state as an instant "snapshot" at some moment of time. Because of the results [22, 23], it is natural to expect that for the *even* number ( $2k$ ) of walls between source and drain electrodes of graphene channel its conduction  $G_+^{total}$  and  $G_-^{total}$  are the same for both polarities of the gate voltage

$$\frac{G_+^{total}}{G_-^{total}} = 1, \quad (9a)$$

because for each polarity there are  $k$  pnJs with conduction, given by eq.(8) of [23] (electrons are tunneling through the barrier), and  $k$  pnJs with conduction, given by eq.(10) of [23] (electrons do not feel the barrier). For the *odd* number of the walls  $2k+1$ , Eq.(14) of Ref.[23] can be modified as:

$$\frac{G_+^{total}}{G_-^{total}} = \frac{\beta(L + \lambda) + \lambda k(1 + \beta)}{\beta(L + \lambda) + \lambda k(1 + \beta) + \lambda}, \quad (9b)$$

where the factor  $\beta$  is equal to  $\sqrt{\frac{\pi\alpha c}{4\epsilon_{33}^f v_F}}$ ,  $\lambda$  is the electron mean free path,  $\epsilon_{33}^f$  is the ferroelectric permittivity,  $v_F$  is the Fermi velocity of electrons in graphene,  $\alpha = 1/137$  is fine structure constant,  $c$  is light velocity in vacuum.

For a pronounced diffusion regime of current,  $\beta L \gg \lambda$ , and/or for the great number of the walls,  $k \gg k_{cr}$ , where  $k_{cr} = 1 + \frac{\beta L}{(1 + \beta)\lambda}$ , the right-hand-side in Eq.(9b) tends to unity (as in Eq. (9a)), and a conduction of graphene channel is described by a well known expression [9]:

$$G = \frac{\lambda(n_{2D})}{L} \frac{2e^2}{\hbar\pi^{3/2}} w\sqrt{n_{2D}} \quad (10)$$

Here  $w$  is graphene channel width,  $n_{2D}$  is 2D charge concentration in the channel, determined by Eq.(1). The mean free path  $\lambda(n_{2D})$  depends on the concentration  $n_{2D}$ . In particular,  $\lambda(n_{2D}) \sim \sqrt{n_{2D}}$  for the scattering at ionized centers in the substrate and in the temperature range far from Curie temperature [9]. Therefore the conduction is a linear function of concentration in this case.

### III. NUMERICAL RESULTS AND DISCUSSION

Below we present results of numerical modeling of the problem (1)-(5). We study numerically the modulation of the graphene channel conductance caused by a domain structure moving in a ferroelectric substrate. Parameters used in the calculations are listed in **Table I**.

**Table I.** Parameters designations and numerical values

| Parameter, constant or value                         | Numerical value and dimensionality   |
|--|--|
| oxide dielectric thickness                           | $h_O = (4 - 10)$ nm  |
| dielectric (dead) layer thickness                    | $h_{DL} = (0 - 4) \times 10^{-10}$ m   |
| ferroelectric film thickness                         | $h_F = (50 - 500)$ nm  |
| graphene channel length                              | $L = (20 - 200)$ nm  |
| universal dielectric constant                        | $\epsilon_0 = 8.85 \times 10^{-12}$ F/m (e/Vm)   |
| permittivity of the dielectric layer                 | $\epsilon_{DL} = 100$ (typical range 10 – 300)   |
| Ferroelectric permittivity of the ferroelectric film | $\epsilon_{33}^f = 500$ , $\epsilon_{11}^f = \epsilon_{22}^f = 780$ (Pb(ZrTi)O <sub>3</sub> -like)   |
| Background permittivity of the ferroelectric film    | $\epsilon_{11}^b = \epsilon_{22}^b = \epsilon_{33}^b = 4$ (Pb(ZrTi)O <sub>3</sub> , BaTiO <sub>3</sub> , or other ferroelectric perovskite)  |
| Landau-Ginzburg-Devonshire potential coefficients    | $\alpha_T = 2.66 \times 10^5$ C <sup>-2</sup> ·mJ/K, $T_C = 666$ K (PbZr <sub>x</sub> Ti <sub>1-x</sub> O <sub>3</sub> , $x \approx 0.5$ ), $P_S^{bulk} = (0.5 - 0.7)$ C/m <sup>2</sup><br>$b = 1.91 \times 10^8$ J C <sup>-4</sup> ·m <sup>5</sup> , $c = 8.02 \times 10^8$ J C <sup>-6</sup> ·m <sup>9</sup> |
| Temperature  | $T = 298$ K (room temperature)   |
| dielectric anisotropy of ferroelectric film          | $\gamma = \sqrt{\epsilon_{33}^f / \epsilon_{11}^f} = 0.8$  |
| extrapolation length                                 | $\Lambda_+ = \Lambda_- = \infty$   |
| Plank constant                                       | $\hbar = 1.056 \times 10^{-34}$ J·s = $6.583 \times 10^{-16}$ eV·s   |
| Fermi velocity of electrons in graphene              | $v_F \approx 10^6$ m/s   |

For the enlisted parameters we get factor  $\beta = 0.185$ , the product  $\beta L \approx (4 - 400)$  nm for the channel length  $L = (20 - 2000)$  nm, and  $k_{cr} = 1 + 0.16L/\lambda$ . So that the ratio  $G_+^{total} / G_-^{total}$  can be noticeably different from unity for  $\lambda \geq 0.185L$  and domain wall number  $k \leq 1 + 0.16L/\lambda$ .

The polarization component in the ferroelectric film  $P_3$ , variation of 2D-concentration of free carriers in the graphene channel  $\Delta n_G = (p_{2D} - n_{2D})$ , and the effective conductance ratio

$\Delta\eta(U_{\max}) = \frac{\Delta n_G(+U_{\max})}{\Delta n_G(-U_{\max})}$  were calculated in dependence on the gate voltage  $U(t) = U_{\max} \sin(2\pi t/T_g)$ .

Results of the calculations are presented and analyzed in the next subsections.

### A. Hysteresis loops of ferroelectric polarization and carrier concentration at low voltages

The hysteresis loops of the average polarization  $P_3(U)$  and concentration variation  $\Delta n_G(U)$  are shown in **Figs.2(a)-(f)**. Only the steady regime is shown, for all the cases the transient process was cut off. Black, red and magenta loops correspond to the different amplitudes of gate voltage  $U_{\max} = (2, 5, 10)$  V. At relatively low voltages ( $\leq 2$  V for chosen material parameters) polarization and concentration loops of quasi-elliptic shape do not reveal any ferroelectric peculiarities [see black curves in **Fig. 2(a)-(f)**]. This happens because the film state is poly-domain, and the domain walls do not disappear with the gate voltage changing from  $-U_{\max}$  to  $+U_{\max}$ , but they are moved by the electric field. Actually, the domain walls are moving to minimize the system energy when the voltage higher than the coercive one is applied to the top gate. Note that the domain wall pinning effects, which can affect on the coercive field, are not considered in our modeling.

With the increase of the gate voltage amplitude  $U_{\max}$  to (5 – 10) V the domain walls start to collide and the domains with opposite polarization orientation almost "annihilate" for definite periodic moments of time  $t$ , and then the polar state of the film with some degree of unipolarity partially restores [see red and magenta curves in **Figs.2(a)-(c)** and **Videos 1 - 3**]. The loops asymmetry and rectification ratio is defined by the symmetry of the BCs.

#### A1. Impact of periodic boundary conditions

Completely symmetric loops of polarization  $P_3(U)$  and concentration  $\Delta n_G(U)$  variation correspond to the periodic BCs [**Figs.2(a)** and **2(d)**]. The amount of positive (holes) and negative (electrons) charges is equal entire the period of the gate voltage for the periodic BCs. The rectification effect is absent in the case of periodic BCs, since the effective ratio  $\Delta\eta(U_{\max}) \equiv -1$  for all  $U_{\max}$  [see **Figs.2(g)**], because the even number of domain walls is moving in the ferroelectric substrate at any time [see **Video 1**].

A typical distributions of electric potential and polarization for periodic BCs proved that two domain walls separating three domains with "up" and "down" polarizations are present in the ferroelectric film [see **Fig.3(a)**]. An example of the spatial-temporal evolution of the polarization component  $P_3$  and corresponding free charge concentration calculated along the graphene channel at certain times over a period are shown in **Figs. 4(a)** and **4(d)**, respectively. Two pnJs produced by the two moving domain walls are seen at certain times over one period. The relative depth (i.e. maximum to minimum ratio) and position ( $\Delta n_G = 0$ ) of the pnJs are time-dependent.

Spatial-temporal distributions of 2D-concentration of the free charge  $\Delta n_G(x, t)$  calculated for the periodic BCs at small amplitude of the gate voltage ( $U_{\max} = 2$  V) is shown in **Fig.5(a)**. Maximal

positive (holes) and negative (electrons) charge densities are equal entire the period of the gate voltage for the periodic BCs [corresponding color scale of the carriers density ranges from  $+8.7 \times 10^{17} \text{ m}^{-2}$  to  $-8.7 \times 10^{17} \text{ m}^{-2}$  in **Fig.5(a)**], reflecting the fact that negative and positive polarizations are equivalent entire the period for periodic BCs [see symmetric polarization loops in **Figs.2(a)**].

### A2. Impact of mixed boundary conditions

Asymmetric loops of  $P_3(U)$  and  $\Delta n_G(U)$  correspond to the mixed BCs [**Figs.2(b)** and **2(e)**]. The vertical asymmetry and horizontal shift of polarization loop are much stronger than the asymmetry of concentration variation because the polarization acts on the charge indirectly via the depolarization field. The asymmetry of  $P_3(U)$  and  $\Delta n_G(U)$  becomes weaker with maximal voltage increase [compare different loops in **Figs.2(b)** and **2(e)**]. The rectification effect is present in the case of low voltages, since the effective ratio  $\Delta\eta(U_{\max}) \gg 1$  for  $1 < U_{\max} < 3$ , the ratio changes sign at  $U_{\max} = 4$ , and then  $\Delta\eta(U_{\max}) \rightarrow -1$  for higher voltages [see **Figs.2(h)**], because asymmetric domain walls are moving in the ferroelectric film [see **Video 2**]. An instant distributions of electric potential and polarization show that two domain walls with complex shape near the surface, which separate three domains (two with "up" and one with "down" polarizations), are located in the ferroelectric film [see **Fig.3(b)**]. The spatial-temporal evolution of the polarization and corresponding free charge concentration calculated along the graphene channel are shown in **Figs. 4(b)** and **4(e)**, respectively. Two pnJs produced by the two moving domain walls exist inside the channel at all times over one period. Also there are two halves of "positive" pnJs at the channel boundaries and, most probably, these peculiarities create the asymmetry of the total conductance.

Spatial-temporal distributions of 2D-concentration of the free charge  $\Delta n_G(x, t)$  calculated for the mixed BCs is shown in **Fig.5(b)**. The charge density symmetry is broken for the mixed BCs [corresponding color scale ranges from  $+3.6 \times 10^{17} \text{ m}^{-2}$  to  $-4.4 \times 10^{17} \text{ m}^{-2}$  in **Fig.5(b)**], reflecting the fact that polarization has noticeable degree of unipolarity for the mixed BCs [see asymmetric polarization loops in **Figs.2(b)**].

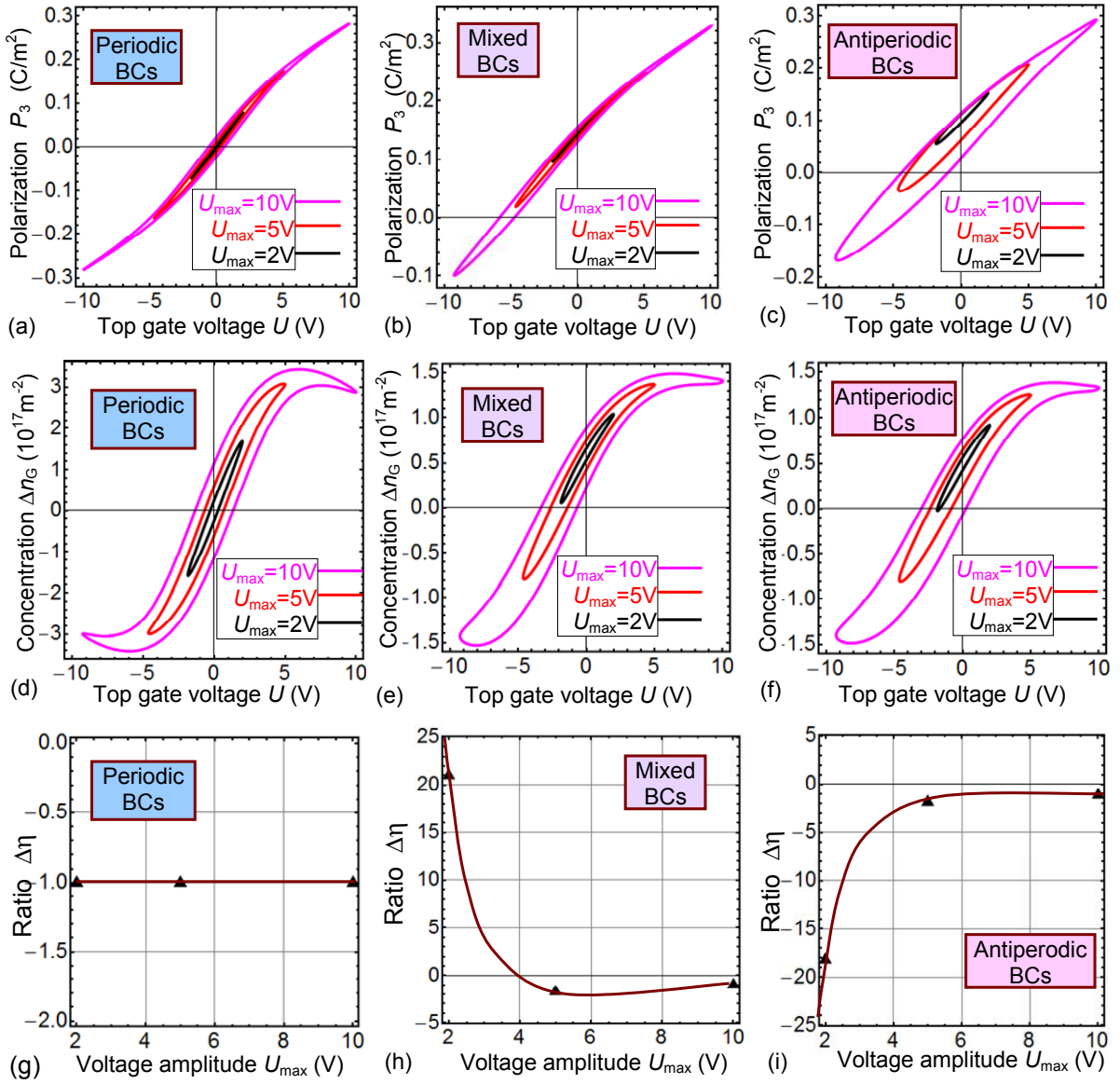
### A3. Impact of antiperiodic boundary conditions

The antiperiodic BCs also lead to asymmetric loops of polarization  $P_3(U)$  and concentration  $\Delta n_G(U)$  [**Figs.2(c)** and **2(f)**]. As well as in the previous case, the vertical asymmetry and horizontal shift of  $P_3(U)$  loop are much stronger than the asymmetry of  $\Delta n_G(U)$  loop because the polarization acts on the charge indirectly via the depolarization field. The asymmetry of  $P_3(U)$  and  $\Delta n_G(U)$  becomes weaker with maximal voltage increase [compare different loops in **Figs.2(c)** and **2(f)**]. The rectification effect is evident in the case of low and moderate voltages, since the effective ratio

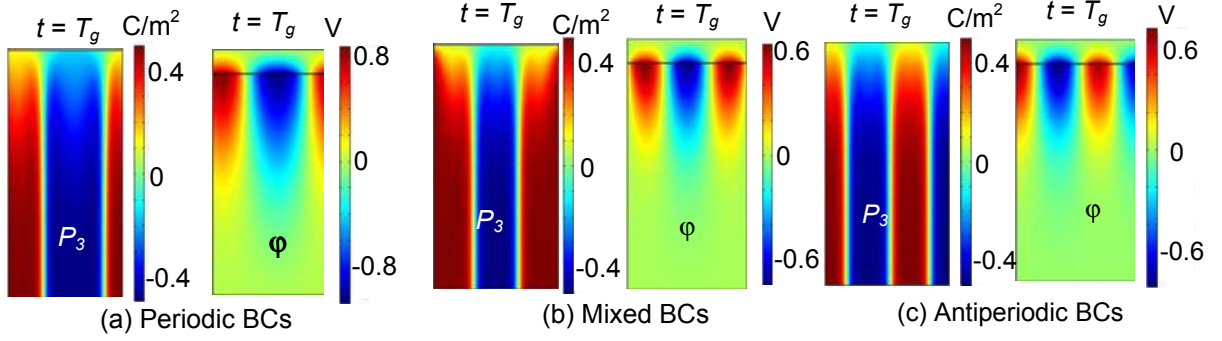
$\Delta\eta(U_{\max}) \ll 1$  for  $1 < U_{\max} < 4$ , and for higher voltages then the ratio saturates,  $\Delta\eta(U_{\max}) \rightarrow -1$  [see **Figs.2(i)**], because the odd number of domain walls is moving in the ferroelectric film most of the time [see **Video 3**]. A typical distributions of electric potential and polarization show that three domain walls, which separate four domains (two with "up" and two with "down" polarizations), are present in the ferroelectric film [see **Fig.3(c)**]. A typical spatial-temporal evolution of the polarization and corresponding graphene charge is shown in **Figs. 4(c)** and **4(f)**, respectively. Three pnJs produced by the three moving domain walls exist at almost all times over one period [see **Video 3**], at that their relative depth and position are time-dependent.

Spatial-temporal distributions of 2D-concentration of the free charge  $\Delta n_G(x,t)$  calculated for the antiperiodic BCs is shown in **Fig.5(c)**. The charge density symmetry breaking for the antiperiodic BCs is evident from the figure, where the carrier density scale ranges from  $+3.7 \times 10^{17} \text{ m}^{-2}$  to  $-4.2 \times 10^{17} \text{ m}^{-2}$ , caused by the polarization asymmetry shown in **Figs.2(c)**.

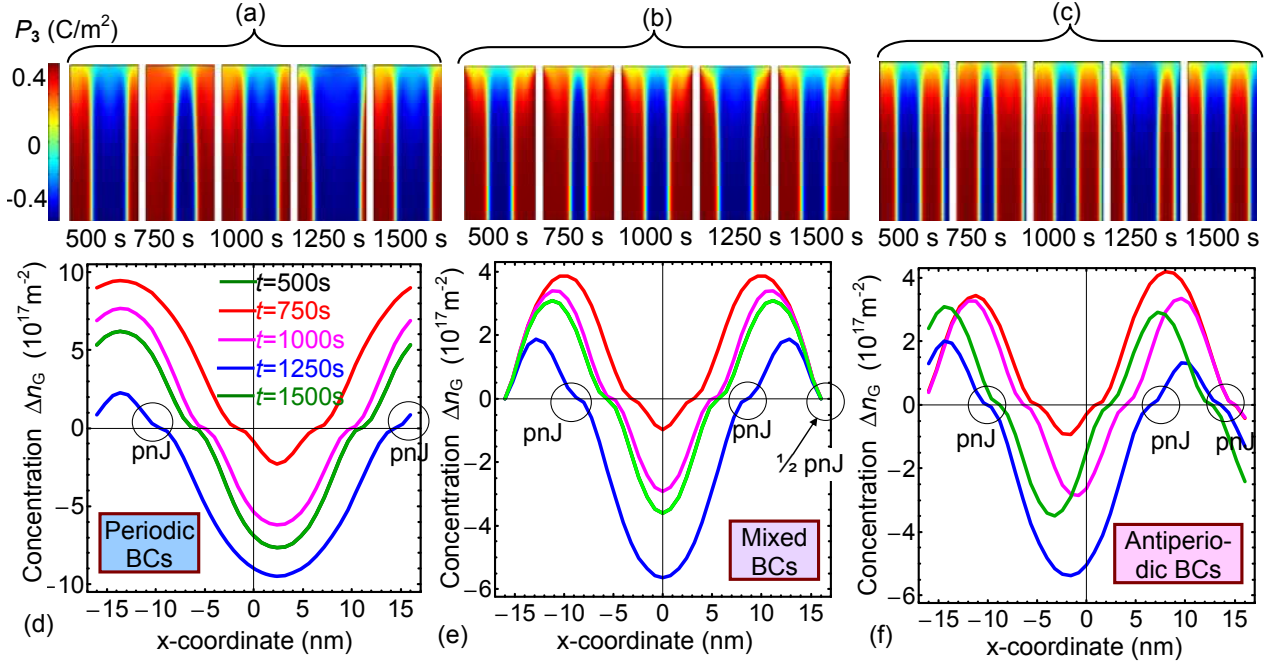
Note that for low voltage amplitudes  $U_{\max} < 10 \text{ V}$  the maximal charge density  $\sigma_G = e\Delta n_G \sim (0.024 - 0.048) \text{ C/m}^2$  recalculated from the  $\Delta n_G \sim (1.5 - 3) \times 10^{17} \text{ m}^{-2}$  appeared noticeably smaller than the spontaneous polarization of the ferroelectric film  $P_S \sim 0.5 \text{ C/m}^2$ . This happens because of the electric potential drop in the oxide dielectric layer and corresponding appearance of depolarization field in it.



**FIG. 2.** Hysteresis loops of ferroelectric polarization  $P_3(U)$  (a, b, c), carriers concentration variation in graphene channel  $\Delta n_G(U)$  (d, e, f) and the conductance ratio  $\Delta\eta(U_{\max})$  (g, h, i) calculated for periodic (a, d, g), mixed (b, e, h) and antiperiodic (c, f, i) boundary conditions (BCs). Black, red and magenta loops in plots (a)-(f) correspond to the different amplitudes of gate voltage  $U_{\max} = (2, 5, 10)$  V. The thicknesses of ferroelectric film is  $h_f = 75$  nm, oxide dielectric thickness  $h_o = 8$  nm, dead layer thickness  $h_{DL} = 0.4$  nm, gate voltage period  $T_g = 10^3$  s. Other parameters are listed in **Table I**. For animations of the ferroelectric domain structure evolution along one period of applied voltage, see **Videos 1, 2** and **3**, corresponding to periodic, mixed and antiperiodic BCs respectively.

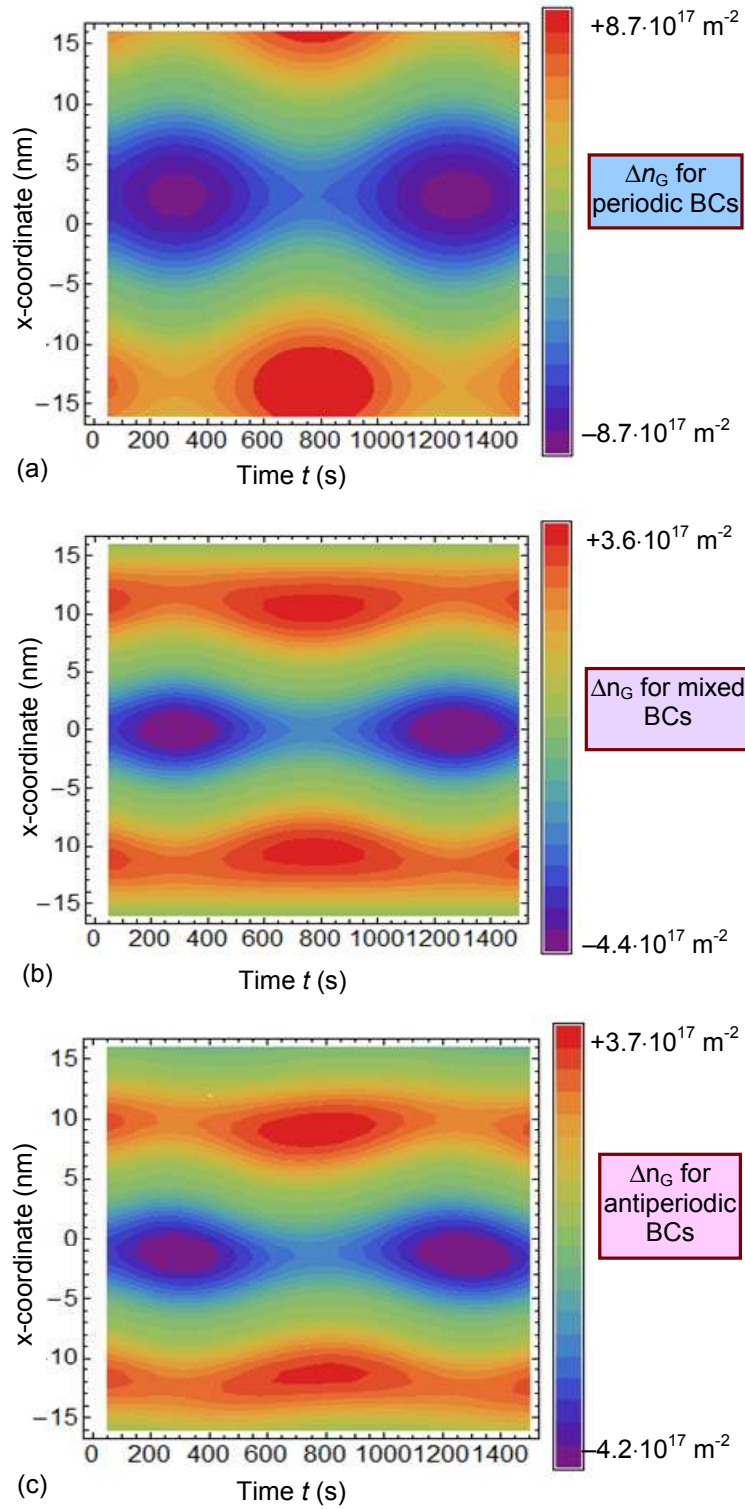


**FIG. 3.** Spatial distribution of a ferroelectric polarization component  $P_3$  and electric potential  $\phi$  calculated for the periodic (a), mixed (b) and antiperiodic (c) boundary conditions (BCs) at time  $t = T_g$ ,  $U_{\max} = 10$  V and  $T_g = 1000$  s. Other parameters are the same as in Fig.2.



**FIG. 4.** Spatial distribution of polarization component  $P_3$  in a ferroelectric film calculated at certain times over a period, 500, 750, 1000, 1250 and 1500 s (specified in the plots) for the periodic (a), mixed (b) and antiperiodic (c) BCs. 2D-concentration of the free charge  $\Delta n_G(x, t)$  calculated along the graphene channel at certain times over a period, 500, 750, 1000, 1250 and 1500 s (specified in the plots) for the periodic (d), mixed (e) and antiperiodic (f) BCs. Gate voltage amplitude  $U_{\max} = 5$  V and period  $T_g = 10^3$  s. Other parameters are the same as in Fig.2. The transient process that almost vanish enough rapidly is not shown (so that we started to show the plots from times  $t \geq 500$  s).





**FIG. 5.** Spatial-temporal distributions of 2D-concentration of the free charge  $\Delta n_G(x, t)$  calculated along the graphene channel for the periodic (a), mixed (b) and antiperiodic (c) BCs. Gate voltage amplitude  $U_{\max} = 2 \text{ V}$  and period  $T_g = 1000 \text{ s}$ . Other parameters are the same as in Fig.2.

To resume, the aperiodicity of electric boundary conditions for polarization at the lateral surfaces of ferroelectric substrate can induce the voltage asymmetry of the graphene channel conductance hysteresis loops.

## **B. Hysteresis loops of ferroelectric polarization and carrier concentration: transition from low to high voltages**

For comparative analysis of the system behavior at low, intermediate and high gate voltages we calculated the dependences of domain structure evolution and ferroelectric polarization in the substrate and the surface charge concentration in graphene channel on the gate voltage. Results for hysteresis loops of ferroelectric polarization  $P_3(U)$  and surface charge concentration variation  $\Delta n_G(U)$  are shown in **Fig.6** for periodic, mixed and antiperiodic BCs, respectively. Black, brown, red, blue and magenta loops correspond to the different amplitudes of gate voltage  $U_{\max} = (2, 5, 10, 15, 20)$  V.

At relatively low voltages (less than 5 V for chosen material parameters) polarization and concentration loops of quasi-elliptic shape are looking very slim for the chosen voltage window (20 V) and do not reveal any ferroelectric peculiarities [see red and brown curves in **Fig. 6**]. This happens because the film state is poly-domain, and the domain walls do not disappear with the gate voltage changing from  $-U_{\max}$  to  $+U_{\max}$ ; they are moved in the electric field. With the increase of the gate voltage amplitude  $U_{\max}$  to (5 – 10) V the loops shape start to deviate from elliptic one [compare red and brown curves in **Fig.6**]. This happens because domain walls start to collide and the domains with opposite polarization orientation almost "annihilate" for definite periodic moments of time  $t$ , and then the film polar state with significant degree of unipolarity partially restores. The degree of unipolarity strongly increases with  $U_{\max}$  increase to values higher than 10 V. We observed that the stationary switching regime is single-domain independently on the poly-domain seeding for  $U_{\max} \geq 15$  V. Corresponding hysteresis loops undergo the voltage-induced phase transition with  $U_{\max}$  increase above 10 V acquiring a pronounced loop opening with high remnant polarization, charge and coercive voltage (i.e. memory window) and square-like, irregular or rhomboidal shape depending on the BCs [compare blue and magenta loops with e.g. red ones in **Fig. 6**].

Notice that for the voltages higher than 10 V one needs especially good quality of dielectric layers, because the field in the oxide layer can be greater than the breakdown one (about 1 V/nm for SiO<sub>2</sub> [39]). Nevertheless electric potential drops in the dielectric layer and gap, the graphene charge recalculated from the carrier concentration appeared of the same order than the spontaneous polarization of the ferroelectric film at  $U_{\max} \geq 15$  V. This happens because the gate voltages are high enough to increase the polarization of the ferroelectric film entire the period.

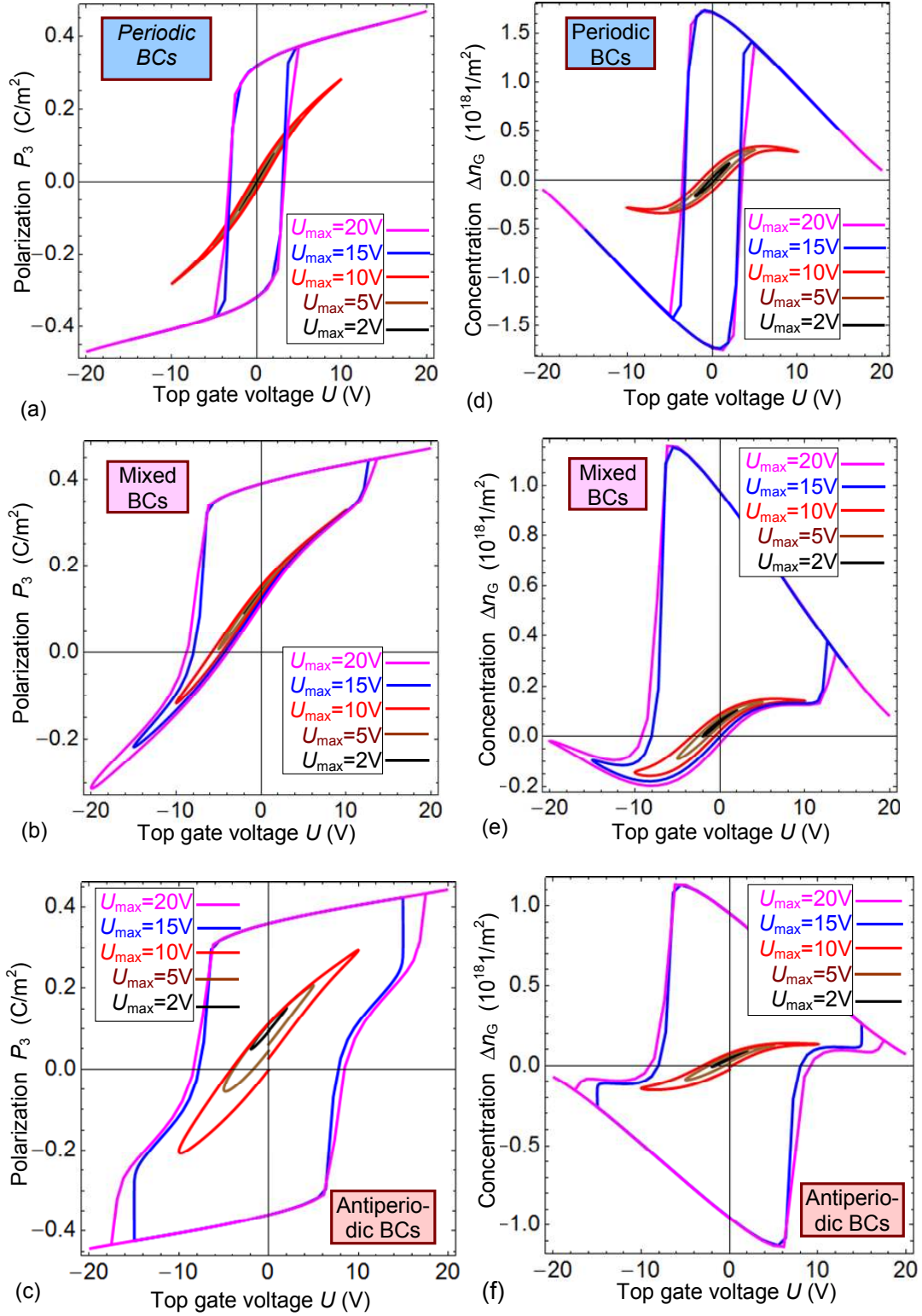
Symmetric square-shaped polarization loops with high coercive voltage correspond to the periodic BCs and voltage amplitude  $U_{\max} \geq 20$  V [see blue and magenta curves in **Fig.6(a)**]. The modulation effect of the graphene carrier concentration loop is evident in this case and the rectification

effect is absent [see blue and magenta curves in **Fig.6(d)**], since even number of domain walls are moving in the ferroelectric substrate. Spatial distributions of domain structure in ferroelectric film below the channel along one period is demonstrated by **Video 1**.

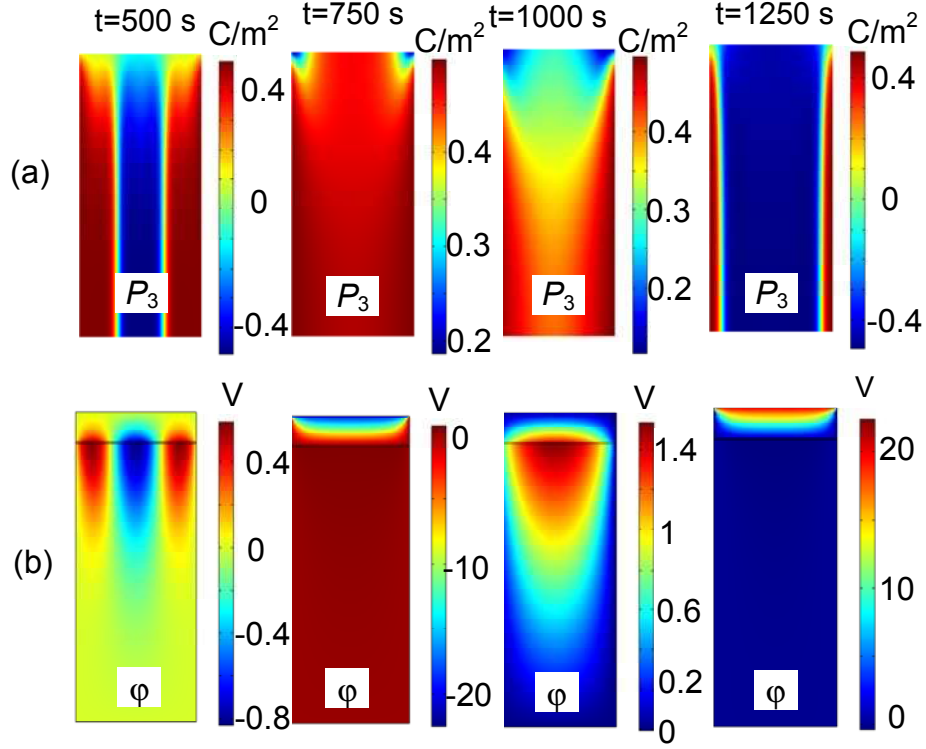
Strongly asymmetric polarization loops with high coercive voltage correspond to the mixed BCs and  $U_{\max} \geq 15$  V [see blue and magenta curves in **Fig. 6(b)**]. The rectification effect of the carrier concentration loops with pronounced memory window is evident in the case [see blue and magenta curves in **Fig. 6(e)**]. Corresponding spatial distributions of domain structure in ferroelectric film and electric potential in the system calculated for different moments of time (500, 750, 1000 and 1250 s) are shown in **Figs. 7(a) and 7(b), respectively**. From **Fig. 7(a)** odd number of domain walls are moving in the ferroelectric substrate.

Almost symmetric square-shaped polarization loops with a constriction, which have the highest coercive voltage, correspond to the antiperiodic BSc and  $U_{\max} \geq 15$  V [see blue and magenta curves in **Fig. 6(c)**]. The of the carrier concentration loops have rhombus shape with rounded corners and the highest coercive voltage in comparison with periodic and mixed BCs [see blue and magenta curves in **Fig.6(f)**]. In the case modulation effect of concentration is evident and the rectification effect is absent, since the switching process acquires almost single-domain scenario at high voltage amplitude and the fraction of domain walls moving in the ferroelectric substrate becomes rather small. Hence the loops asymmetry inherent to the lower voltage amplitudes [see red curves in **Figs.6(c) and 6(f)**] vanishes with  $U_{\max}$  increase to (15 – 20) V.

In contrast to the estimates made in the end of subsection III.A3 for low voltages  $U_{\max} < 5$  V, at high voltages  $U_{\max} > 15$  V the maximal modulation of the graphene charge density  $e\Delta n_G \sim (0.16 - 0.24)C/m^2$  recalculated from the 2D-concentration of carriers  $\Delta n_G \sim (1 - 1.5) 10^{18} m^{-2}$  appeared of the same order than the spontaneous polarization of the ferroelectric film  $P_S \sim 0.5C/m^2$ .



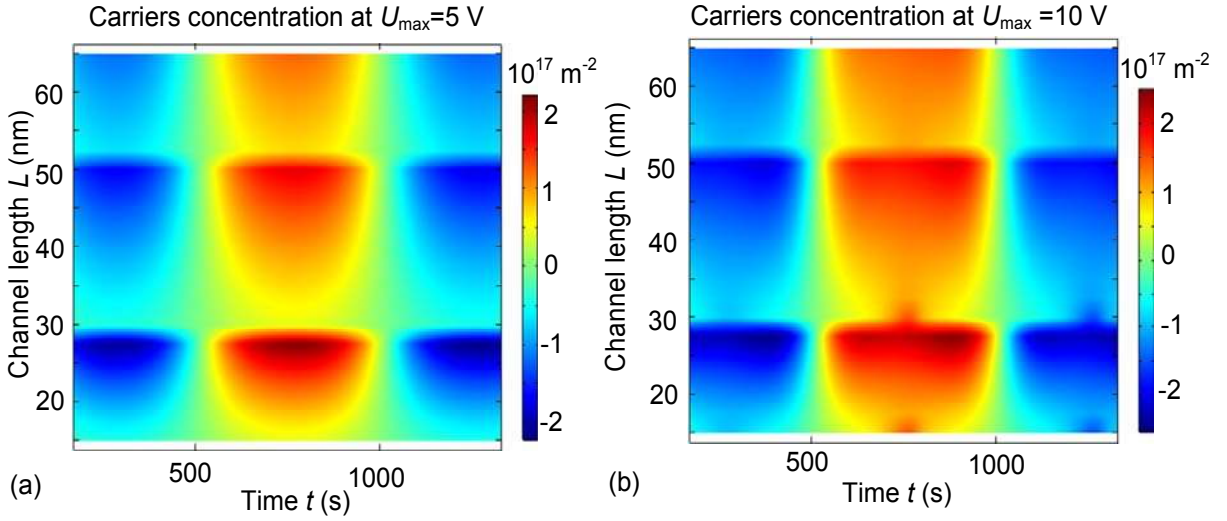
**FIG. 6.** Hysteresis loops of ferroelectric polarization (a, b, c) and surface charge concentration in graphene channel (d, e, f) calculated in dependence on the gate voltage for periodic (a, d), mixed (b, e) and antiperiodic (c, f) BCs. Black, brown, red, blue and magenta loops correspond to the different amplitudes of gate voltage  $U_{\max} = (2, 5, 10, 15, 20)$  V. The thicknesses of ferroelectric film  $h_f=75$  nm, oxide dielectric thickness  $h_o=8$  nm, dead layer thickness  $h_{DL}=0.4$  nm, gate voltage period  $T_g=10^3$  s. Other parameters are listed in **Table I**.



**FIG. 7.** Spatial distribution of ferroelectric polarization in  $P_3$  in a ferroelectric film **(a, the top row)** and electric potential  $\varphi$  **(b, the bottom row)** calculated for the mixed BCs at  $U_{\max} = 20$  V for different moments of time (500, 750, 1000 and 1250 s). Other parameters are the same as in **Fig.6**.

### C. Dependence of the graphene channel conductivity on its length

Also we revealed that the conductivity of graphene channel (that is proportional to the total carrier concentration variation  $\Delta n_G(L, t)$ ) depends on its length  $L$  in a rather non-trivial way. Corresponding contour maps of the average concentration variation  $\Delta n_G(L, t)$  in coordinates "channel length  $L$  – time  $t$ " are shown in **Figs. 8**. The phenomenon we called "extrinsic size effect" consists in a quasi-periodic modulation of the conductivity amplitude with the channel length for periodic BCs, at that the modulation extremes are the most pronounced for  $L$  around 27.5 nm (the first maxima and minima depending on the time moment), 50 nm (the second maxima and minima) and slightly more that 70 nm (the third maxima and minima are not shown in the figure) at the parameters listed in **Table I** and room temperature. The modulation degree strongly increases with the gate voltage amplitude increase above 5 V [compare **Fig.8(a)** for  $U_{\max} = 5$  V with **Fig.8(b)** for  $U_{\max} = 10$  V], but becomes less pronounced with  $L$  increase [compare the contrast for the modulation maxima and minima at  $L=27.5$  nm and 50 nm]. Also the distance  $\Delta L$  between the maxima is voltage-independent and slightly increases with  $L$  increase [compare **Fig.8(a)** with **Fig.8(b)**].



**FIG. 8.** Dependence of the graphene channel conductivity on its length and time calculated for the periodic BCs at  $U_{\max} = 5$  V **(a)** and  $U_{\max} = 10$  V **(b)**. Other parameters are the same as in **Fig.5**.

Despite the peculiarities shown in **Figs.8** complicate the analytical treatment of the problem, we expect that the extrinsic size effect should vanish for long channels, which length is much longer than the intrinsic period of domain structure in a ferroelectric substrate. Rigorously speaking, we relate the origin of the size effect with the interference of the domain structure period and the channel length. Actually the period of the 180-degree domain structure  $W$  is the intrinsic characteristic of the ferroelectric film that is defined by the film thickness, polarization screening conditions at the film - graphene and film - bottom gate interfaces ( $z = 0$  and  $z = h_F$ ), lateral surfaces ( $x = +L_{FE}/2$  and  $x = -L_{FE}/2$ ), surface energy (via extrapolation lengths  $\Lambda_{\pm}$  and  $\lambda_{\pm}^F$ ) and temperature [37]. Since the strong inequality  $L_{FE} \gg L$  is regarded valid for the considered geometry, the period  $W$  becomes virtually independent on  $L_{FE}$  and extrapolation lengths  $\lambda_{\pm}^F$ , but the relation between  $W$  and  $L$  can be arbitrary (because  $L$  changes independently in **Fig.8**), at that the number of domains in the channel of length  $L$  is defined by two factors, the ratio  $L/W$  and the symmetry type of the virtual BCs at  $x = \pm L/2$ . As it was established in subsections II.B and III.A [see Eqs.(9) and **Figs.2(d)-(f)**], since each domain walls creates a pnJ in the graphene channel, the sign and values of the average concentration modulation  $\Delta n_G(U, t)$  is defined by the total number  $k$  of the domain walls in the channel at low gate voltage amplitudes  $U_{\max} \leq 10$  V. For a particular case of periodic BCs the number  $k$  should be even [see comments to Eq.(9a)]. Thus the case when the integral part of the ratio  $L/W$  approaches an even number corresponds to the  $\Delta n_G(U, t)$  extremes in **Figs.8** So that at low gate voltages the channel conductivity modulation naturally depends on the commensurability of the channel length  $L$  and the intrinsic period of the domain structure  $W$ . Allowing for the single-domain

switching domination with the voltage increase the value of  $\Delta n_G(U, t)$  is defined by the total degree of unipolarity of ferroelectric substrate for the case of high voltages  $U_{\max} \geq 15$  V.

Note that extrinsic size effect can be expected for the mixed BCs. However, according to our additional calculations, its manifestation appeared less pronounced and irregular than the one for the periodic (or antiperiodic) BCs. The result supports the interference origin of the size effect since the mixed BCs allow fractional number of the domain walls moving along the graphene channel and the commensurability of  $L$  and  $W$  are not required for the case.

To resume results of the section III we predict that the considered nanostructure "top gate/dielectric layer/graphene channel/ferroelectric substrate" can be a promising candidate for the fabrication of new generation of modulators and rectifiers based on the graphene p-n junctions.

#### IV. CONCLUSION

Using a self-consistent approach based on Landau-Ginzburg-Devonshire phenomenology combined with classical electrostatics we studied p-n junctions dynamics in graphene channel induced by stripe domains nucleation, motion and reversal in a ferroelectric substrate.

Relatively low amplitudes of gate voltages ( $U_{\max} \leq 5$  V) are required to induce the hysteresis of ferroelectric polarization and graphene charge in dependence on the periodic gate voltage. The harmonic-like charge concentration modulation caused by a domain structure alternates with its homogeneous distribution caused by a single-domain states under the periodic change of applied voltage.

Pronounced nonlinear hysteresis of graphene conductance with a wide memory window corresponds to high amplitudes of gate voltage  $U_{\max} \geq 15$  V, because the degree of unipolarity of the ferroelectric film strongly increases with  $U_{\max}$  increase and the stationary switching regime becomes single-domain independently on the poly-domain seeding.

It was demonstrated that the asymmetry type of electric boundary conditions for polarization at the lateral surfaces of ferroelectric substrate is responsible for the asymmetry of the hysteresis loop of graphene channel conductance between the source and drain electrodes. In particular, asymmetric loops of polarization and charge correspond to the mixed and antiperiodic boundary conditions. Completely symmetric loops of ferroelectric polarization and graphene charge concentration variation correspond to the periodic boundary conditions.

For the periodic BCs we revealed the extrinsic size effect in the dependence of the graphene channel  $L$  conductivity on its length that consists in a quasi-periodic modulation of the conductivity amplitude with  $L$ . The origin of the size effect can be related with the interference of the intrinsic period of the domain structure  $W$  and the channel length  $L$ . Namely, the conductivity extremes correspond to the situation when the integer part of the ratio  $L/W$  approaches an even number. The



size effect can be expected for the mixed BCs, but its manifestation would be less pronounced than the one for the periodic (or antiperiodic) BCs, since the mixed BCs allow fractional number of the domain walls moving along the graphene channel and the commensurability of  $L$  and  $W$  are not required for the case. The result supports the interference origin of the size effect.

We expect that predicted effects become enough pronounced to be observed experimentally for the case of thin oxide dielectrics ( $<10$  nm), intimate electric contact or ultra-thin gap ( $<0.5$  nm) between the ferroelectric and graphene sheet, short lengths of graphene channels (20 – 100) nm and thin ferroelectric substrates ( $<100$  nm). The first inequality mentioned above imposes demands for the dielectric with a high breakdown field. At that it is extremely important to control the number of domain walls moving in the channel.

Taking into consideration the effects obtained in our work we predict that the considered nanostructure "top gate/dielectric layer/graphene channel/ferroelectric substrate" can be promising candidates for the fabrication of new generation of modulators and rectifiers based on the graphene p-n junctions.

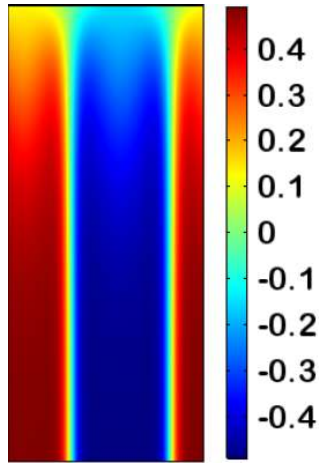
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**Authors' contribution.** A.I.K. and E.A.E. performed numerical calculations and make figures. M.V.S. generated the research idea and motivation, derived analytical expression for graphene conductance, strongly contributed to the results analyses and manuscript improvement. S.V.K. contributed to the interpretation of the results and manuscript writing. A.N.M. formulated the problem mathematically, performed most of analytical calculations, select parameters and wrote the manuscript.

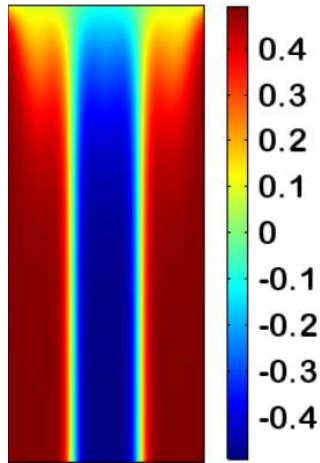
#### APPENDIX:

Animations of the ferroelectric domain structure evolution along one period of applied voltage are demonstrated by **Videos 1, 2 and 3**, corresponding to periodic, mixed and antiperiodic BCs, respectively. Each image is obtained by calculating at specific time intervals within a period.

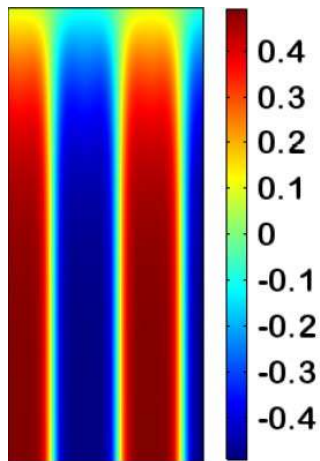




**VIDEO 1.** [URL will be provided by Publisher to Video\_1.gif] Animations of the ferroelectric domain structure evolution along one period of applied voltage for periodic BCs. Gate voltage amplitude  $U_{\max} = 10$  V and period  $T_g = 10^3$  s. Other parameters are the same as in Fig.2.



**VIDEO 2.** [URL will be provided by Publisher to Video\_2.gif] Animations of the ferroelectric domain structure evolution along one period of applied voltage for mixed BCs. Gate voltage amplitude  $U_{\max} = 10$  V and period  $T_g = 10^3$  s. Other parameters are the same as in Fig.2.



**VIDEO 3.** [URL will be provided by Publisher to Video\_3.gif] Animations of the ferroelectric domain structure evolution along one period of applied voltage for antiperiodic BCs. Gate voltage amplitude  $U_{\max} = 10$  V and period  $T_g = 10^3$  s. Other parameters are the same as in **Fig.2**.

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