Suppression of the superconducting transition temperature of doped graphene due to thermal fluctuations of the order parameter

V. M. Loktev*
Bogolyubov Institute for Theoretical Physics, Metrologichna Str. 14-b, Kiev 143 03680, Ukraine

V. Turkowski†
Department of Physics and NanoScience and Technology Center, University of Central Florida, Orlando, Florida 32816, USA
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In this Brief Report, we analyze the superconducting properties of doped single- and double-layer graphene systems by taking into account the fluctuations of the superconducting order parameter. Our analysis is rather general, and corresponds to a phenomenological electron-electron (hole-hole) attraction defined by its strength and range, and is independent of the origin of attraction. We show that in this model, similar to the case of two-dimensional doped metal, the thermal fluctuations of the order-parameter result in a significant reduction in the Berezinskii-Kosterlitz-Thouless critical temperature T_c comparing to the mean-field temperature T_c^{MF}, and there is a pseudogap phase with a suppressed density of states at temperature range $T_c<T<T_c^{\mathrm{MF}}$. At low doping n_f, the critical temperature is proportional to n_f in the double-layer case, and it is exponentially suppressed in the case of a single layer.

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I. INTRODUCTION

The carbon-based structures show a great promise for different applications, especially in nanotechnologies. In order to understand and predict the physical properties of different carbon-based systems, one needs to understand general properties of the basic units of such structures. One of the most important such units is the single layer of graphene, which can be considered as the basis for two-dimensional (2D) (one- to several-layer graphene), one-dimensional (1D) (carbon nanotubes and nanoribbons), and zero-dimensional (0D) (fullerene molecules, graphene flakes, etc.) structures. Over the last years, tremendous progress has been made in understanding some of the physical properties of graphene, including the electronic properties (see, e.g., Ref. 1 and references therein). One of the most interesting questions is the possibility of superconductivity (SC) in single- and several-layer graphene structures. Recently, some evidences of SC in graphite and graphite-sulfur composites were found experimentally.[2–5] The possibility of SC in graphene was studied theoretically in Refs. 6–13. In particular, the quantum critical-point scenario of SC in undoped graphene, when the Fermi surface consists of Dirac points, was analyzed in Refs. 6 and 8. It was shown that in this case SC takes place only when the coupling is bigger than some critical value. Different mechanisms of SC were discussed, like the phonon, plasmon,[9] resonant-valence bond, and the density-wave scenarios.[6,10,11] In the case of electron- or hole-doped graphene, one does not require a large value of the coupling to get a SC state, since in this case the Fermi surface is finite. SC in doped graphene within the framework of different models was analyzed in Refs. 6, 7, 9, 10, 12, and 13. In particular, the Bardeen-Cooper-Schrieffer (BCS) scenario with the electron-electron attraction, which depends on two parameters—the strength and the range—was analyzed in Ref. 12. In Ref. 13, the possibility of strong enhancement of SC ($T_c \sim 10$ K) due to a van Hove singularity in the density of states (DOS) at ~3 eV from the Dirac points was discussed. However, the question whether one can reach such large values of the doping on practice remains open. Generally speaking, the results obtained in the papers mentioned above are based on the mean-field analysis. However, it is well known that in 2D systems the fluctuations can significantly affect the SC properties. In this case, the Berezinskii-Kosterlitz-Thouless (BKT) scenario of SC with the algebraically ordered order parameter below the temperature $T_c \equiv T_{\mathrm{BKT}}$, which is lower than the mean-field critical temperature T_c^{MF}, must be considered. Below, we show on the example of the model considered in Ref. 12 that indeed the thermal fluctuation leads to a drastic suppression of SC in the single- and double-layer doped graphene. In the case of one-layer, another factor for the suppression comes from the reduction in the DOS ($\sim\sqrt{n_f}$) with carrier density decreasing. We also show that at temperatures $T_c<T<T_c^{\mathrm{MF}}$ both systems demonstrate a so-called pseudogap phase with a significant reduction in the one-electron (hole) DOS in the gap region due to the thermal fluctuations of the SC order parameter.

II. MODEL AND THE MEAN-FIELD RESULTS

The effective Hamiltonian of the SC system of doped graphene can be written in the following form:

$$H(\tau) = \sum_{\sigma} \psi_{\sigma}^{\dagger}(\tau,\mathbf{r})[\varepsilon(\nabla)-\mu]\psi_{\sigma}(\tau,\mathbf{r}) - \frac{1}{2}\int d\mathbf{r}_1 \int d\mathbf{r}_2 \times \psi_{\uparrow}^{\dagger}(\tau,\mathbf{r}_2)\psi_{\downarrow}^{\dagger}(\tau,\mathbf{r}_1)V(\mathbf{r}_1-\mathbf{r}_2)\psi_{\downarrow}(\tau,\mathbf{r}_1)\psi_{\uparrow}(\tau,\mathbf{r}_2), \quad (1)$$

where $\psi_{\sigma}^{\dagger}(\tau,\mathbf{r})[\psi_{\sigma}(\tau,\mathbf{r})]$ is the creation (annihilation) operator of fermion on site $\mathbf{r}$ at imaginary time τ with spin $\sigma=\uparrow,\downarrow$; $\varepsilon(\nabla)$ and μ are the free quasiparticle dispersion relation and the chemical potential, and $V(\mathbf{r}_1-\mathbf{r}_2)$ is the fermion-fermion attraction potential. In the case of single graphene

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layer, the quasiparticle spectrum around the Dirac points can be approximated by $\varepsilon^{\pm}(\mathbf{k})=\pm v_F|\mathbf{k}|$, where the Fermi velocity $v_F=3ta/2\sim 10^6$ m/s ($t=2.8$ eV is the nearest-neighbor hopping and $a=1.42$ Å is the carbon-carbon distance). In the two-layer case, the low-energy spectrum can have several forms depending on the kinetic processes taken into account. In this Brief Report, we consider the case with a nonzero nearest-neighbor intralayer hopping t between the different sublattice A and B sites and the nearest-neighbor interlayer hopping $t_\perp$ between the A sites ($t_\perp \simeq 0.4$ eV in graphite). In this case, there are four parabolic bands with the dispersions $\varepsilon_1^{\pm}=\pm(t_\perp+v_F^2|\mathbf{k}|^2/t_\perp)$, and $\varepsilon_2^{\pm}=\pm v_F 2|\mathbf{k}|^2/t_\perp$ (for details, see, e.g., Ref. 1). At zero doping, the band $\varepsilon^{-}(\mathbf{k})$ is filled in the single-layer case, and the bands $\varepsilon_1^{-}(\mathbf{k}),\varepsilon_2^{-}(\mathbf{k})$ are filled in the case of two layers. For definiteness, we consider the electron-doped case, where the conduction band is $\varepsilon^{+}(\mathbf{k})\equiv\varepsilon(\mathbf{k})$ in the single-layer case, and $\varepsilon_2^{+}(\mathbf{k}\equiv\varepsilon^{b}(\mathbf{k})$ in the double-layer case. In the last case, one can also take into account the interlayer hopping processes between the different sublattice sites A_1 and (A_2), and B_2 and (B_1) (with the hopping parameter ~ 0.3 eV$<t_\perp$ in graphite). In this case, the dispersion will consist of a set of linear Dirac dispersions, and the situation will be qualitatively similar to the single-layer case. The interlayer hopping between the nearest-neighbor B sites can be neglected, since it is much smaller than the hopping parameters discussed above (-0.04 eV in graphite). It is important to notice, that in the undoped case the effective Hamiltonian must be written in terms of the Dirac fermions.[1] However, in the finite doping case, the electron-hole symmetry is broken, and one can use Hamiltonian (1) with standard "nonrelativistic" Fermi quasiparticles.

We assume that the SC attraction is caused by the following model interparticle potential:

$$V_{\mathbf{p}}=V_0\theta(\xi_0-|\varepsilon(\mathbf{p})-\mu|), \tag{2}$$

where V_0 is the amplitude of the attraction and ξ_0 is its BCS range. Since the dependence of the SC properties on ξ_0 is rather simple (SC increases with ξ_0 growth), we shall study the V_0 dependencies and put $\xi_0=\infty$, which assumes that the interaction range is bigger than the bandwidth and puts the upper limit on SC for the given value of V_0. Large values of ξ_0 are consistent with the low-doping case mainly considered in this Brief Report, since in this case $\xi_0>\mu$. The source of electron-electron attraction in doped graphene, which can lead to SC, is not established yet. The most often discussed possible mechanisms include phonon or screened acoustic phonon exchange, and the electron-electron correlation scenario (see, e.g., Refs. 9 and 10, correspondingly). The exact values of V_0 and ξ_0 can be obtained from *ab initio* calculations for different interactions, though they are not available so far. The results obtained in this Brief Report allow one to estimate the values of V_0 and ξ_0 from the experimental values of n_f and T_c, and consequently might help establish the mechanism of SC in graphene. For simplicity, we shall also consider the pairing in the s-wave channel when the SC gap $\Delta(\mathbf{k})$ is momentum independent. However, the results obtained below are rather general and can be straightforwardly generalized on the cases of more complicated interactions and different symmetries of the order parameter.

In order to study the SC properties of the systems, it is convenient to consider the partition function

$$Z=\int D\psi^{\dagger}D\psi\exp\left[-\int_0^{1/T}d\tau\times\left(\sum_{\sigma}\int d\mathbf{r}\psi_{\sigma}^{\dagger}(\tau,\mathbf{r})\partial_{\tau}\psi_{\sigma}(\tau,\mathbf{r})+H(\tau)\right)\right], \tag{3}$$

which can be obtained by performing the path integration over the wave functions. In order to find Z, one can introduce the Nambu spinor operators

$$\Psi(\tau,\mathbf{r})=\begin{bmatrix}\psi_{\uparrow}(\tau,\mathbf{r})\\ \psi_{\downarrow}^{\dagger}(\tau,\mathbf{r})\end{bmatrix},\quad \Psi^{\dagger}(\tau,\mathbf{r})=[\psi_{\uparrow}^{\dagger}(\tau,\mathbf{r}),\psi_{\downarrow}(\tau,\mathbf{r})]. \tag{4}$$

Then, to split the four-fermion product [see Eq. (1)], one can use the Hubbard-Stratonovich transformation and show that the partition function in the SC state is equivalent to

$$Z=\int D\Psi^{\dagger}D\Psi D\Phi^{\dagger}D\Phi e^{-S(\Psi^{\dagger},\Psi,\Phi^{\dagger},\Phi)}, \tag{5}$$

where

$$\begin{aligned}S(\Psi^{\dagger},\Psi,\Phi^{\dagger},\Phi)=\int_0^{1/T}d\tau\int d\mathbf{r}_1\int d\mathbf{r}_2\Bigg\{&\delta(\mathbf{r}_1-\mathbf{r}_2)\sum_{\sigma}\psi_{\sigma}^{\dagger}(\tau,\mathbf{r}_1)\\ &\times[\partial_{\tau}+\tau_z(\varepsilon(\nabla_{\mathbf{r}_2})-\mu)]\psi_{\sigma}(\tau,\mathbf{r}_2)\\ &+\frac{1}{2}\frac{|\Phi(\mathbf{r}_1,\mathbf{r}_2)|^2}{V(\mathbf{r}_1-\mathbf{r}_2)}\\ &-\frac{1}{2}\Psi^{\dagger}(\tau,\mathbf{r}_1)\tau_{+}\Psi(\tau,\mathbf{r}_2)\Phi(\tau,\mathbf{r}_1,\mathbf{r}_2)\\ &-\frac{1}{2}\Phi^{\dagger}(\tau,\mathbf{r}_1,\mathbf{r}_2)\Psi^{\dagger}(\tau,\mathbf{r}_1)\tau_{-}\Psi(\tau,\mathbf{r}_2)\Bigg\}\end{aligned} \tag{6}$$

is the effective action, $\tau_{\pm}=1/2(\tau_x\pm i\tau_y)$ and τ_z are the Pauli matrices, and $\Phi(\tau,\mathbf{r}_1,\mathbf{r}_2)\sim V(\mathbf{r}_1-\mathbf{r}_2)\psi_{\downarrow}(\tau,\mathbf{r}_1)\psi_{\uparrow}(\tau,\mathbf{r}_2)$ is the complex SC order-parameter function. Integration over the Nambu spinors in Eq. (5) leads to the thermodynamical potential $\Omega(\Phi^{\dagger},\Phi)$ defined as follows:

$$e^{-\Omega(\Phi^{\dagger},\Phi)/T}=\int D\Psi^{\dagger}D\Psi e^{-S(\Psi^{\dagger},\Psi,\Phi^{\dagger},\Phi)}. \tag{7}$$

In the mean-field approximation, we assume that in the momentum representation $\Phi(\tau,\mathbf{r}_1,\mathbf{r}_2)$ is momentum and time independent and equal to $\Delta(\tau,\mathbf{k})\equiv\Delta=$const. Then, minimization of $\Omega(\Delta)$ with respect to Δ together with the particle-number equation $(1/V)\partial\Omega/\partial\mu=-n_f$ (V is the volume of the system) result in the following system of equations for the SC gap and the chemical potential:

$$1=\frac{1}{2}\sum_{\mathbf{k}}V(\mathbf{k})\tanh\left(\frac{E(\mathbf{k})}{2T}\right)\frac{1}{E(\mathbf{k})}, \tag{8}$$

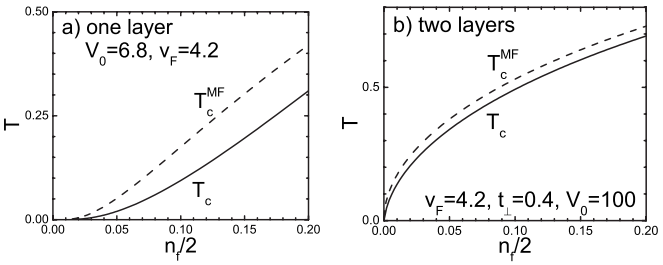

FIG. 1. The doping dependence of the mean-field and the BKT critical temperatures in the case of one-layer (top) and two-layer (bottom) graphene. All values of the parameters are given in eV.

$$n_f = 2\sum_{\mathbf{k}} \left[1 - \tanh\left(\frac{E(\mathbf{k})}{2T} \right) \frac{\varepsilon(\mathbf{k}) - \mu}{E(\mathbf{k})} \right], \tag{9}$$

where $E(\mathbf{k}) = \sqrt{(\varepsilon(\mathbf{k}) - \mu)^2 + \Delta^2}$ and $n_f = N_e/N - 1$ is the density of doped electrons (N_e is the number of electrons and N is the number of sites, such that $n_f = 0$ in the undoped case). The solution of this system of equations gives the doping and the temperature dependence of the SC gap Δ and also the doping dependence of the mean-field SC critical-temperature T_c^{MF} (at $\Delta = 0$). Our analysis shows that at low doping, the mean-field gap and T_c^{MF} are proportional to $\sqrt{n_f}$ in the two-layer case and it is exponentially suppressed when approximately $\sqrt{n_f} < \mathrm{const}/V_0$, where the constant is on order of 1 eV for the model parameters mentioned above, in the case of one-layer (see Fig. 1). The reason for this difference is caused by different doping dependencies of the single-electron DOS on the Fermi-level $N(\varepsilon_F)$, which is proportional to $\sqrt{n_f}$ in the single-layer case and is constant in the case of two layers. This can be shown in the easiest way in the weak-coupling case. In fact, the effective BCS-like attraction in both cases is $\lambda \sim N(\varepsilon_F) V_0$, and the BCS gap and the mean-field critical temperature have the following dependencies at low doping: $\sim \sqrt{W \varepsilon_F} \exp(-A/\lambda)$, where A is a numerical constant and W is the bandwidth, which leads to the exponential suppression of the gap and T_c^{MF} in the single-layer case. Below, we demonstrate that the real T_c is much lower than these values due to the thermal fluctuations in the (quasi-)two-dimensional systems.

III. FLUCTUATIONS

In order to study the fluctuation effects in the system, we represent the fermionic operators as the product of the neutral fermions and the phases $\psi_\sigma(\tau, \mathbf{r}) = \chi_\sigma(\tau, \mathbf{r}) \exp[i\theta(\tau, \mathbf{r})/2]$ and $\psi_\sigma^\dagger(\tau, \mathbf{r}) = \exp[-i\theta(\tau, \mathbf{r})/2] \chi_\sigma^\dagger(\tau, \mathbf{r})$ such that

$$\Psi(\tau, \mathbf{r}) = e^{i\tau_z \theta(\tau, \mathbf{r})/2} \Upsilon(\tau, \mathbf{r}) \equiv e^{i\tau_z \theta(\tau, \mathbf{r})/2} \begin{pmatrix} \chi_\uparrow(\tau, \mathbf{r}) \\ \chi_\downarrow^\dagger(\tau, \mathbf{r}) \end{pmatrix},$$

$$\Psi^\dagger(\tau, \mathbf{r}) = \Upsilon^\dagger(\tau, \mathbf{r}) e^{-i\tau_z \theta(\tau, \mathbf{r})/2} \equiv [\chi_\uparrow^\dagger(\tau, \mathbf{r}), \chi_\downarrow(\tau, \mathbf{r})] e^{-i\tau_z \theta(\tau, \mathbf{r})/2}. \tag{10}$$

In this case, the gap functions become $\Phi(\tau, \mathbf{r}_1, \mathbf{r}_2) = \Delta(\tau, \mathbf{r}_1, \mathbf{r}_2) \exp[i\theta(\mathbf{R})]$, $\Phi^\dagger(\tau, \mathbf{r}_1, \mathbf{r}_2) = \Phi^*(\tau, \mathbf{r}_1, \mathbf{r}_2)$. In the last expressions, $\Delta(\tau, \mathbf{r}_1, \mathbf{r}_2) = |\Phi(\tau, \mathbf{r}_1, \mathbf{r}_2)|$ is the modulus of the order parameter, and $\theta(\mathbf{R}) \simeq [\theta(\mathbf{r}_1) + \theta(\mathbf{r}_2)]/2$, where $\mathbf{R} = (\mathbf{r}_1 + \mathbf{r}_2)/2$ is the center-of-mass coordinate of two electrons, is its phase. This approximation is valid when the space dependence of the phase is weak, i.e., when the thermal fluctuations are not strong. Below, we neglect the time-(quantum-) fluctuations, which are important $T \to 0$, and assume that the order-parameter modulus (the gap) is a function of the relative electron coordinate $\mathbf{r}_1 - \mathbf{r}_2$

$$\Phi(\tau, \mathbf{r}_1, \mathbf{r}_2) \simeq \Delta(\mathbf{r}_1 - \mathbf{r}_2) e^{i\theta(\mathbf{R})}. \tag{11}$$

The last approximation means that the dynamics of the Cooper pairs is described by the order-parameter modulus, and its symmetry depends on the relative pair coordinate, but the motion of the condensate is defined by the slowly varying phase of the order parameter, and its space dependence can be described by the center-of-mass coordinate.

Substitution of Eqs. (10) and (11) into Eq. (7) and functional integration over the neutral fermionic fields $\Upsilon^\dagger$ and Υ yield the following equation for the thermodynamic potential as a function of the phase of the order parameter:

$$\Omega(\Delta, \partial_\tau \theta, \nabla \theta) = T \int_0^{1/T} d\tau \int d\mathbf{r}_1 \int d\mathbf{r}_2 \frac{1}{2} \frac{|\Delta(\mathbf{r}_1 - \mathbf{r}_2)|^2}{V[(\mathbf{r}_1 - \mathbf{r}_2)} - \mathrm{Tr} \ln(G^{-1} - \Sigma) + \mathrm{Tr} \ln G^{-1}, \tag{12}$$

where $G_{\mathbf{k}}(t, t') = -i\langle T(\Upsilon_{\mathbf{k}}(t) \Upsilon_{\mathbf{k}}^\dagger(t')) \rangle$ is the time-ordered mean-field matrix Green's function, which in the Matsubara frequency-momentum representation reads $G_{\mathbf{k}}(i\omega_n) = \{i\omega_n - \tau_z[\varepsilon(\mathbf{k}) - \mu] - \tau_x \Delta(\mathbf{k})\}^{-1}$, and Σ is the nonhomogeneous Green's function self-energy, which depends on the phase gradients

$$\Sigma(\mathbf{r}_1, \mathbf{r}_2) = \delta(\mathbf{r}_1 - \mathbf{r}_2)[e^{-i\tau_z \theta(\mathbf{r}_1)/2} \tau_z \varepsilon(-i\nabla_{\mathbf{r}_2}) e^{i\tau_z \theta(\mathbf{r}_2)/2} - \tau_z \varepsilon(-i\nabla_{\mathbf{r}_2})]. \tag{13}$$

In Eq. (12), Tr means the space-time integration and the matrix trace (for details, see, e.g., Ref. 14). Expanding the logarithm in terms of the powers of Σ

$$\mathrm{Tr} \ln[G^{-1} - \Sigma] = \mathrm{Tr} \ln G^{-1} + \mathrm{Tr} \sum_n \frac{1}{n} (G\Sigma)^n, \tag{14}$$

one can get the following expression for the second-order expansion of the thermodynamical potential in the limit of small fluctuations of the phase of the order parameter:

$$\Omega(\Delta, \theta) = \frac{\mathcal{J}}{2} \int d^2 r (\nabla \theta)^2, \tag{15}$$

where the stiffness $\mathcal{J}$ is

$$\mathcal{J} = \frac{v_F^2}{8T} \int \frac{d^2 k}{(2\pi)^2} \frac{1}{\cosh^2[E(\mathbf{k})/2T]}, \tag{16}$$

in the case of one layer, and

$$\mathcal{J}^b = \frac{v_F^2 n_f}{2t_\perp} - \frac{v_F^4}{4t_\perp^2} \frac{1}{T} \int \frac{d^2 k}{(2\pi)^2} \frac{\mathbf{k}^2}{\cosh^2[E(\mathbf{k})/2T]} \tag{17}$$

in the case of two layers.

In analogy with the 2D spin-XY model, the equation for the critical temperature of the BKT transition T_c, below which the phases of order parameter (the spin $\nabla\theta$ orientation in the XY-model case) become algebraically ordered, has the following form:

$$T_c = \frac{\pi}{2}\mathcal{J}[\Delta(\mu,T_c),\mu,T_c], \tag{18}$$

where function $\mathcal{J}$ is defined in Eqs. (16) and (17). The doping and the coupling dependence of T_c can be found by solving the system of Eqs. (8), (9), and (18).

The numerical analysis shows that T_c is significantly lower than T_c^{MF} in both cases especially in the low-doping limit (Fig. 1). When the coupling is not too strong in the two-layer case, there is a universal dependence $T_c=\epsilon_F/8 \simeq \sqrt{3}\pi t^2 N_{\mathrm{cell}}/t_\perp \sim 10^5 K N_{\mathrm{cell}}$, where ϵ_F is the Fermi energy and $N_{\mathrm{cell}}=n_f V_{\mathrm{cell}}$ is the number of doped carriers per unit cell (V_{cell} is the volume of the unit cell), while in the single-layer case it is exponentially suppressed, similar to T_c^{MF}. Our analysis shows that T_c starts to grow rapidly at densities n_f larger than the critical-value $n_f^{\mathrm{cr}} \sim 1/V_0^2$. In particular, in order to get the critical temperature bigger than 1K, one needs to have the effective-coupling $\sqrt{n_f}V_0 \sim 1$ eV. One can in principle argue that in the single-layer case, T_c can be higher due to a van Hove singularity, but it is still not clear whether such high values of doping ($\epsilon_F \sim 5$ eV) can be achieved. Besides the reduction in the value of the critical temperature, another important consequence of the thermodynamic fluctuations is the presence of a finite DOS at temperatures $T_c < T < T_c^{\mathrm{MF}}$ in the gap region, or pseudogap phase (Fig. 2) which can be observed experimentally in graphene systems. Similar phase was observed in high-temperature superconductors, and the role of thermal fluctuations as a possible scenario of the formation of this phase is a topic of hot debates (see, e.g., Ref. 15 and references therein).

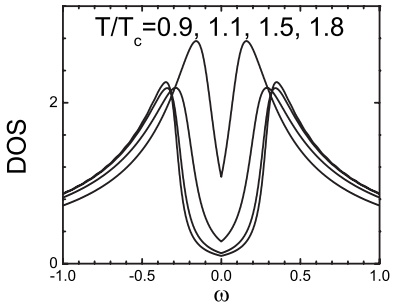

FIG. 2. An approximate curve of the DOS at $n_f=0.2$, when $T_c^{\mathrm{MF}} \simeq 1.86 T_c$, and different values of temperature in the case of one-layer graphene. The model parameters are the same as in Fig. 1 (for details of calculations, see Ref. 16).

IV. CONCLUSIONS

We have studied the doping and the coupling dependencies of the SC properties of single- and double-layer graphene systems by taking into account the thermodynamical fluctuations of the SC order parameter. We have shown that the fluctuations lead to a significant reduction in the critical temperature. Namely, for the realistic values of the model parameters, the critical-temperature T_c is exponentially suppressed at doping $n_f < 1$ eV$/V_0$ in the case of single-layer graphene, and $T_c=\epsilon_F/8$ at low doping in the case of two layers. In addition, in the case of both models the thermodynamic fluctuations lead to a pseudogap phase, with a finite DOS in the SC gap region at temperatures over T_c ($T_c < T < T_c^{\mathrm{MF}}$), which can be observed experimentally.

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*vloktev@bitp.kiev.ua
†vturkows@mail.ucf.edu

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