Electric bias control on impurity effects in bigraphene

Y.G. Pogorelov,1 M.C. Santos,2 V.M. Loktev3,4
1 IFIMUP-IN, Departamento de Física, Universidade do Porto,
Rua do Campo Alegre 687, Porto 4169-007, Portugal,
2 Departamento de Física, Universidade de Coimbra, R. Larga, Coimbra 3004-535, Portugal,
3 Bogolyubov Institute for Theoretical Physics, NAN of Ukraine, 14b Metrologichna str., Kiev 03680, Ukraine
4 National Technical University of Ukraine KPI, Pr. Peremogy 37, Kiev 03056, Ukraine

Formation of localized impurity levels within the band gap in bigraphene under applied electric field is considered and the conditions for their collectivization at finite impurity concentration are established. It is shown that a qualitative restructuring of quasiparticle spectrum within the initial band gap and then specific metal-insulator phase transitions are possible for such disordered system and can be effectively controlled by variation of the electric field bias at given impurity perturbation potential and concentration. Since these effects can be expected at low impurity concentrations but at not too low temperatures, they can be promising for practical applications in nanoelectronics devices.

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1. INTRODUCTION

Between various derivitives from the basic graphene system1–3, a special interest is attributed to its bilayer combination, the bigraphene4. This interest is mainly due to the important possibility to realize here a case of semiconductor with controllable band gap due to an applied electric field5–7. It should be noted that similar crystalline structure of two planes with hexagonal lattices is now recognized for a whole family of materials, either really fabricated or theoretically predicted. Besides the two known modifications of bigraphene itself, called the Bernal structure8 and its alternative A-A structure9, it includes also the bilayers of silicene, silicon analog to graphene10, the bilayers of boron nitride11 or its bilayered combination with graphene12, the bilayered chalcogenides of transition metals (pure or alloyed)13, etc. However, the most reliable structure for external tuning and the easiest for theoretical study is seen in the Bernal-stacked bigraphene, hence chosen here as the basic host system for studying impurity effects. By the analogy with the known effects of dopants in common semiconductor systems, this rises first the question about possibility for specific localized impurity levels within this gap14,15, like those known for common donor and acceptor levels by impurities in conventional semiconductors with fixed band structure16,17. Next, it is known that, at high enough impurity concentration, an intensive interaction can take place between these levels that can essentially modify the band spectrum near the gap edge18,19 and even produce a phase transition from insulating to metallic state20 with important practical applications. Yet much more flexible field of electronic properties of a doped semiconducting system is expected under the conditions of continuously variable band gap and hence controllable phase transitions, like the situation in magnetic excitation spectra of some magnetic crystals with impurities controllable by applied magnetic field21,22. Up to the authors’ knowledge, such a possibility have not been known before for fermionic systems and it can open interesting perspectives for future nanoelectronics.

The paper is organized as follows. In Sec. 2, the second quantization Hamiltonian is defined for a Bernal-stacked biased bigraphene (free from impurities) and the related matrix representation for Green functions (GF) is built, giving rise to its 4-subband electronic spectrum. The next Sec. 3 introduces the model impurity perturbation and treats formation of impurity levels and the conditions for subsequent formation of impurity bands from these levels, based on specific forms of self-energy matrices entering the Green function matrices. A more detailed study of such impurity bands, including the estimates for characteristic mobility edges between the band-like and localized states, is presented in Sec. 4. Eventually, the possibility for specific metal-insulator phase transition in doped bigraphene under the external electric field bias (at fixed impurity concentration) and the resulting transport effects are considered in Sec. 5. The final Sec. 6 discusses the main conclusions and indications for practical applications of the described impurity effects.

2. BIGRAPHENE UNDER APPLIED FIELD

As well known, the relevant electronic dynamics of a graphene sheet is generated by the carbon sp3 orbitals (whose energy level can be chosen as the energy reference) in the simplest approximation of the hopping parameter t between nearest neighbor carbons from different sublattices at distance a in the honeycomb lattice2. The bigraphene structure yet involves the interlayer hopping $t_z$ by vertical links between nearest neighbors from two sublattices, conventionally denoted A and B, in different layers, as shown in Fig. 1. With an account taken...
of the electric bias \( V = eE d \) between the layers (with the electron charge \( e \), applied electric field \( E \), and the interlayer spacing \( d \)), this defines the tight-binding (Fourier transformed) Hamiltonian \( 4 \times 4 \) matrix:

\[
\hat{H}_k = \begin{pmatrix}
V/2 & \gamma_k & 0 & t_z \\
\gamma_k^* & V/2 & 0 & 0 \\
0 & 0 & -V/2 & \gamma_k \\
0 & 0 & \gamma_k^* & -V/2
\end{pmatrix},
\]

(1)

where the wave vector \( k \) lies in the first Brillouin zone (see inset in Fig. 1) and the in-plane dispersion follows from the sums \( \gamma_k = t \sum_{\delta} e^{i k \delta} \) over near neighbor vectors \( \delta \) of the honeycomb lattice, suitably approximated as \( \gamma_k \approx \xi_k e^{i \varphi_k} \) with \( \xi_k = \hbar v_F |k - K| \), the Fermi velocity \( v_F = 3ta/2h \), and \( \varphi_k = \arctan(k_y/(k_x - K_x)) \) near the Dirac points \( K = \pm (4\pi/3\sqrt{3}a, 0) \) (the relevant range of \( |k - K| \approx Kt_z/W \) is really narrow, since \( t_z \) is weak besides the total bandwidth \( W \), see below). This matrix enters the second-quantization Hamiltonian (in absence of impurity perturbation):

\[
H_0 = \sum_k \psi_k^\dagger \hat{H}_k \psi_k,
\]

(2)

where the spinors \( \psi_k = (a_{1k}^\dagger, b_{1k}^\dagger, a_{2k}^\dagger, b_{2k}^\dagger) \) are made of Fourier transformed second quantization operators \( a_{jk} = N^{-1} \sum_n a_{jn} e^{ikn} \) and \( b_{jk} = N^{-1} \sum_n b_{jn} e^{ikn} \) with the on-site operators \( a_{jn} \) and \( b_{jn} \) for \( A \) and \( B \)-type sites respectively from \( n \)th unit cell in \( (j = 1, 2) \)-th layer and \( N \) is the number of cells in a layer. Generally, the energy spectrum is defined through the matrix of Fourier transformed two-time GFs\(^{22,23} \) \( \hat{G}_k(\xi) = \langle \langle \psi_k^\dagger(\xi) \psi_k^\dagger(\xi) \rangle \rangle \), as solutions of the dispersion equation:

\[
\Re \det \hat{G}_k^{-1} = 0.
\]

(3)

Thus, for the non-perturbed system by Eq. 2 the GF matrix reads \( \hat{G}_k(\xi) = (\xi - \hat{H}_k)^{-1} \) and, after diagonalization of \( \hat{H}_k \) in spinor indices, its dispersion near the Dirac points is suitably expressed through the radial variable \( \xi_k \equiv \xi \). It includes two positive energy subbands:

\[
\varepsilon_\nu(\xi) = \sqrt{\frac{t_z^2}{2} + \frac{V^2}{4} + \xi^2 - (-1)^\nu \sqrt{\frac{t_z^4}{4} + \xi^2 (t_z^2 + V^2)}},
\]

(4)

the "external" \( (\nu = 1) \) and "internal" \( (\nu = 2) \) ones, and their negative energy counterparts, as shown in Fig. 2a. The most relevant feature of this spectrum is the bias-controlled energy gap between the extrema \( \varepsilon_g = \pm V/(2 \sqrt{1 + (V/t_z)^2}) \) of two internal subbands, attained along a circle around each Dirac point (the so-called "mexican hat") whose radius in the \( \xi \)-variable is \( \varepsilon_0 = \sqrt{\varepsilon_g^2 + V^2/4} \). The physical characteristics of this system follow from the GF matrix as, for instance, the density of states (DOS) by electronic quasiparticles:

\[
\rho(\varepsilon) = \frac{1}{\pi} \Im \text{Tr} \hat{G}(\varepsilon),
\]

(5)

where \( \hat{G}(\varepsilon) = (2N)^{-1} \sum_k \hat{G}_k(\varepsilon) \) is the local GF matrix. In what follows, the sum in \( k \) over triangular halves of the Brillouin zone is approximated by the \( \xi \)-integration:

\[
\frac{1}{2N} \sum_k f_k(\varepsilon) \approx \frac{2}{W^2} \int_0^W f(\xi, \varepsilon) \xi d\xi,
\]
over two equivalent circles around Dirac points (inset in Fig. 1) of the $\varepsilon$-radius $W = \hbar v_F \sqrt{K/a}$, well justified at low energies, $|\varepsilon| \ll W$. For the pure bigraphene system by Eq. 1, the result for Eq. 5 is generated by the explicit diagonal elements of non-perturbed local GF matrix:

$$G_{11}^{(0)}(\varepsilon) = \frac{\varepsilon - \varepsilon_2}{W^2} \sqrt{\frac{2\varepsilon \varepsilon_2}{(t_z^2 + V^2)(\varepsilon^2 - \varepsilon_2^2)}} \left[ \pi - \arctan \left( \frac{(t_z^2 + V^2)(\varepsilon^2 - \varepsilon_2^2)}{\varepsilon^2 + \varepsilon_2^2} \right) \right] + \ln \sqrt{\frac{(\varepsilon^2 - \varepsilon_2^2)(\varepsilon^2 - \varepsilon_2^2)}{W^2}},$$

$$G_{22}^{(0)}(\varepsilon) = G_{11}^{(0)}(\varepsilon) - t_z^2 \frac{(\varepsilon - \varepsilon_2)}{W^2} \sqrt{\frac{t_z^2 + V^2}{(t_z^2 + V^2)(\varepsilon^2 - \varepsilon_2^2)}} \left[ \pi - \arctan \left( \frac{(t_z^2 + V^2)(\varepsilon^2 - \varepsilon_2^2)}{\varepsilon^2 + \varepsilon_2^2} \right) \right], \quad (6)$$

These reveal the inverse square root divergences at the gap edges $\pm \varepsilon_2$ (of Im $G$ beyond the gap and of Re $G$ within the gap), also note the finite steps of Im $G$ at the limiting energies $\varepsilon_{1,2} \equiv \varepsilon_{1,2}(0)$ of the two subbands. The resting diagonal elements are simply $G_{33}^{(0)}(\varepsilon) = -G_{22}^{(0)}(\varepsilon)$ and $G_{44}^{(0)}(\varepsilon) = -G_{11}^{(0)}(\varepsilon)$, so that finally DOS is a function of $\varepsilon^2$, as shown in Fig 2b in agreement with the known previous calculations. It presents BCS-like divergences near $\varepsilon_2^r$, finite steps at $\varepsilon_{1,2}^r$, and gets to coincidence with the known linear DOS for monolayer graphene beyond $\varepsilon_1^r$, due to joint (non-linear) contributions from both subbands.

Within the gap, only real parts of $G_{jj}^{(0)}(\varepsilon)$ are non-zero, and their divergences near $\varepsilon_2^r$ are important for appearance, under the effect of localized impurity perturbations, of in-gap localized levels and related collective states which is the main focus for the analysis below.

3. IMPURITY LEVELS AND IMPURITY SUBBANDS

In order to study the effects of localized perturbations on electronic states in bigraphene, we adopt the Lifshitz model of impurity perturbation, where the perturbation potential is identical for all impurity sites randomly distributed among the lattice sites. This model looks more adequate to the case of substitutional impurities in graphene, than its unitary limit in Ref. or the alternative choice of Anderson model with random perturbations at each lattice site in Ref. Also we do not consider the long-range impurity potentials as for Coulomb or screened-Coulomb centers.

Let us begin from the simplest point-like perturbation by a Lifshitz impurity, producing a certain shift $U$ of the on-site energy at the impurity site. In accordance with the composition of spinors, the A- and B-sites from first plane can be referred to the types $j = 1, 2$ respectively and those from second plane to $j = 3, 4$, then $p_j$ denote the defect sites of $j$th type with relative concentrations $c_j = \sum_{p_j} N^{-1}$ such that the total impurity concentration $\sum_j c_j = c \ll 1$. Then the perturbation operator written in terms of local Fermi operators is

$$H_1 = U \left( \sum_{p_1} a_{p_1}^\dagger a_{p_1} + \sum_{p_2} b_{p_2}^\dagger b_{p_2} + \sum_{p_3} a_{p_3}^\dagger a_{p_3} + \sum_{p_4} b_{p_4}^\dagger b_{p_4} \right), \quad (7)$$

or, in terms of $\psi$-spinors by Eq. 2, it takes the form of scattering operator:

$$H_1 = \frac{1}{N} \sum_{j, p_j, k, k'} e^{i(k' - k) \cdot p_j} \psi_{kj}^\dagger \hat{U}_j \psi_{k'j}. \quad (8)$$

where the diagonal matrix $\hat{U}_j$ has a single non-zero element $U$ at the $jj$ site. Considering now the Hamiltonian in presence of impurities $H_0 + H_1$ and following a similar routine to Refs. and following a similar routine to Refs., we arrive at solutions for the GF matrix in two specific forms adequate for two alternative types of excitation states in a disordered system: the band-like (extended) states and localized states. Thus, the first of these types is better described by the so-called renormalized form (RF):

$$\hat{G}_k = \left[ \left( \hat{G}_k^{(0)} \right)^{-1} - \hat{\Sigma}_k \right]^{-1} \quad (9)$$

where the self-energy matrix is additive in different types of impurity centers: $\hat{\Sigma}_k = \sum_j \hat{\Sigma}_{j,k}$, with the partial matrices given by the related RF group expansions in complexes of impurity centers (of the same $j$-type, involved in multiple scattering processes):

$$\hat{\Sigma}_{j,k} = c_j \hat{T}_j \left[ 1 + c_j \sum_{n \neq 0} \left( e^{-i k \cdot n} \hat{A}_{j,n} + \hat{A}_{j,n}^\dagger \hat{A}_{j,-n} \right) \times \left( 1 - \hat{A}_{j,n}^\dagger \hat{A}_{j,-n} \right)^{-1} + \ldots \right]. \quad (10)$$

Each T-matrix $\hat{T}_j = \hat{U}_j \left( 1 - \hat{G}_j \right)^{-1}$ describes all the scatterings on a single impurity center of $j$th type, and
the next to unity term in r.h.s. of Eq. 10 accounts for scatterings on pairs of \( j \)-impurities through the matrices \( A_{j,n} = T_j(2N)^{-1} \sum_{k \neq j} \hat{G}_k e^{i kn} \) of indirect interaction (via band-like excitations) in such pairs at separation \( n \).

Notice the excluded quasimomentum \( k \) (for given \( \Sigma_k \)) in this sum, also the RF GE excludes coinciding quasimomenta in all the multiple sums for products of interaction matrices\(^{21} \). The omitted terms in Eq. 10 relate to all scattering processes in groups of three and more impurities, and their general structure can be found in similarity with the known group integrals from the Ursell-Mayer classical theory of non-ideal gases.

Otherwise, for the range of localized states the non-renormalized form (NRF):

\[
\hat{G}_k = \hat{G}_k^{(0)} - \hat{G}_k^{(0)} \hat{\Sigma}_k^{(0)},
\]

is more adequate. Here the respective NRF self-energy matrix \( \hat{\Sigma} = \sum_j \hat{\Sigma}_j \) has a similar structure to the RF one by Eq. 10 but with the NRF matrices \( \hat{T}_j^{(0)} = U_j(1 - \hat{G}_k^{(0)} U_j)^{-1}, \hat{G}_k^{(0)} = (2N)^{-1} \sum_k \hat{G}_k^{(0)} \), and with no restrictions in all the quasimomentum sums for the products of NRF interaction matrices \( A_{j,n}^{(0)} = \hat{T}_j^{(0)} (2N)^{-1} \sum_k \hat{G}_k^{(0)} e^{i kn} \) (that are only present in their even combinations \( A_{j,n}^{(0)} A_{j,-n}^{(0)} \)).

The best known effect of local perturbations consists in emergence of localized energy levels in the band gap and those were already indicated for impurities in bigraphene\(^{14,15} \). As known from general theory\(^{21,24} \), such levels are most pronounced at sufficiently low concentrations of impurities (so that their indirect interactions can be neglected) and given by the poles of T-matrices. In the present case, the matrices \( \hat{T}_j \) give rise to four different local levels \( \epsilon^{(j)} \) within the band gap, and their locations depend on the magnitude and sign of perturbation parameter \( U \) (like the known situations in common doped semiconductors\(^{16,17} \)) but yet on the applied field \( V \) (as a specifics of doped bigraphene). For definiteness, choose \( U \) negative and comparable in magnitude to the bandwidth \( W \) of non-perturbed spectrum. The positions of four impurity levels \( \epsilon^{(j)} \) by each type of impurity centers are the roots of related Lifshitz equations:

\[
U G_{ij}^{(0)}(\epsilon^{(j)}) = 1,
\]

where using of Eq. 6 provides their dependence on the applied bias \( V \) as shown Fig. 3. It is seen that generally they stay rather shallow at growing \( V \) though can interchange their order in this course (as, e.g., the deepest one passes from \( \epsilon^{(2)} \) to \( \epsilon^{(4)} \) at \( V \approx 2.2 t_z \) for the choice of \( U = -W/2 \)). It can be noted that for the commonly used value of \( t_z \approx 0.35 \) eV, this interchange bias would amount to \( \approx 0.77 \) eV, to compare with the experimentally realized (to the moment) \( V \) values up to \( \approx 0.36 \) eV\(^5 \).

The well known property of localized states by shallow energy levels is their long effective radius\(^{21} \), also indicated for impurities in biased bigraphene\(^{14} \), defining intensive interactions between them already at their very low concentrations. Such interactions were shown to allow, at certain conditions, collectivization of impurity states to form specific band-like states within narrow energy bands (called impurity bands) around the initial localized levels\(^{18} \). As will be seen below, this effect is possible either in the present case of multiple localized levels, with the most essential specifics of their joint participation to form the lowest impurity subband of much stronger dispersion than higher lying subbands (if those permitted).

Formally, in similarity to the non-perturbed case, the band spectrum for the disordered system can be evaluated from the dispersion equation, Eq. 3, with the GF matrix by RF Eqs. 9,10. Of course, if treated rigorously, it presents a tremendous problem of developing infinite sequence of renormalization procedures in all possible terms of the corresponding GE, and there is no reasonable hope for its exact solution. The popular way to avoid this problem by restriction of the full self-energy to its self-consistent T-matrix form (that is, neglecting all the interactions between impurities)\(^{25} \), though provides some explicit solutions with impurity bands but is known to fail just in reproducing the observed (when available) narrow dispersion of these bands. On the other hand, validity of the simplest NRF, Eq. 11, is only limited to the energy ranges of fully localized states.

A more consistent approach can be suggested using partial renormalizations of the full self-energy in Eq. 10, first substituting there the NRF T-matrix and interaction matrices and then subsequently introducing such approximate self-energies into the next generations of GF and interaction matrices, checking convergence of the ob-
tained GE’s in order not to extend the renormalizations to irrelevant GE terms. Namely, it is reasonable to define the $l$th generation GF matrix $G_k^{(l)}$ by an analog to Eq. 9 with the respective self-energy $\tilde{\Sigma}_k^{(l)}$ by an analog to Eq. 10 but containing T-matrices $T^{(l-1)}$ and interaction matrices $A^{(l-1)}_m$ built with use of the preceding generation $\tilde{G}_k^{(l-1)}$ matrices. This algorithm leads to the true RF at $l \to \infty$ and, for the band-like energy ranges where the true RF GE converges, even its first non-trivial $l = 1$ approximation can be reasonable.

Then, in the first step of this routine, the formal solutions of Eq. 10 with the self-energies in the NRF T-matrix approximation, $\tilde{\Sigma}_{jk} \approx c_j T_{jk}^{(0)}$, display four narrow subbands near four impurity levels $\varepsilon_j$, besides the four broad principal bands $\pm \varepsilon_g(\xi)$ (here only slightly modified compared to Eq. 4). An example of such modified spectrum (at a natural choice of equal partial concentrations $c_j$) for the cases of Fig. 3 is shown in Fig. 4. The lowest impurity subband, conventionally denoted here as $\varepsilon^{(2)}(\xi)$ by its proximity to the lowest $\varepsilon^{(2)}(\xi)$ level, is seen to strongly dominate in its dispersion over all the resting ones, and the direct analysis of Eq. 10 shows that this domination is just due to a constructive interplay between all $\varepsilon^{(j)}$.

Note that all impurity subbands in this approximation produce BCS-like divergences in DOS, as well near their merging to the levels $\varepsilon^{(j)}$ as near their terminations. However, since quasimomentum is not true quantum number in a disordered system, the analysis of its real energy spectrum, especially for the in-gap states, should also take account of the damping $\Gamma_j(\xi)$ of each $\varepsilon^{(j)}(\xi)$ state, resulting from $\text{Im} \tilde{\Sigma}_j$. Hence one can consider these states Bloch-like (or conducting) only if the Ioffe-Regel-Mott (IRM) criterion is fulfilled or the GE, Eq. 10, is convergent at related energies, otherwise they should pertain to the localized type. As will be seen, all the formal DOS singularities fall within the localized energy ranges and so are effectively broadened.

The mentioned criteria permit to estimate also the division points between band-like and localized ranges known as Mott’s mobility edges20. Of course, such mobility edges can be found both near the limits of principal and impurity bands but our main focus here will be put on the most dispersive impurity band, like $\varepsilon^{(3)}(\xi)$ in the above example, as the most interesting object for practical purposes. Finally, a certain special value $V_\Lambda$ of bias control (at given impurity concentrations $c_j$ and perturbation parameter $U$) can be indicated, such that mobility edges from both sides of a conducting impurity band will go to merger. This collapse of impurity band will manifest a kind of Anderson transition in a disordered system, realized in a controllable way at $V \to V_\Lambda$.

4. CONDITIONS FOR EXISTENCE OF IMPURITY SUBBANDS

As known from studies on many disordered systems where a localized impurity level $\varepsilon_{imp}$ near an edge $\varepsilon_g$ of pure crystal energy band can give rise, at high enough impurity concentration, to a specific impurity band $\varepsilon_{imp}(\mathbf{k})$21, the latter is restricted by the general IRM criterion:

$$\mathbf{k} \cdot \nabla_k \varepsilon_{imp}(\mathbf{k}) \gg \Gamma_{imp}(\varepsilon_{imp}(\mathbf{k})), \quad (13)$$

where the linewidth $\Gamma_{imp}(\varepsilon)$ of a Bloch-like state with quasi-momentum $\mathbf{k}$ and energy $\varepsilon$ is defined as the imaginary part of the corresponding self-energy. For the present multiband system, this criterion should be formulated for each of $\varepsilon^{(j)}(\xi)$ subbands by expanding the general determinant from Eq. 3 near a given energy $\varepsilon$ in a complex form: $\det G^{-1}_k \approx [\varepsilon - \varepsilon^{(j)}(\xi) + i\Gamma_j(\varepsilon)]/D_j(\varepsilon)$, providing the corresponding linewidth $\Gamma_j(\varepsilon)$ (besides a certain factor $D_j(\varepsilon)$ of energy to cube dimension).

In the adopted Lifshitz perturbation model, each partial T-matrix $T_j$ (regardless of its renormalization) has a single non-zero element at the $jj$ site (alike $U_j$ itself): $T_j = U/(1 - UG_{jj})$, and, in the above suggested first step
renormalization, the imaginary part of self-energy $\Sigma_j(\epsilon)$ for $\epsilon$ within the bandgap is only due to the GE terms next to unity in Eq. 10 where the pair term dominates once GE is convergent. It can be also shown that the most relevant contribution to $\text{Im}\Sigma_j(\epsilon)$ comes from the $j$th matrix element of the GE pair term while those from other its elements (though generally non-zero) are strongly reduced by the quantum interference effects. This relevant contribution:

$$B_j(\epsilon) = \text{Im} \sum_{n>a} A_{jn}^{(0)*} A_{jn}^{(0)} n, \quad B_j(\epsilon) = \sum_{n>a} \frac{A_{jn}^{(0)*} A_{jn}^{(0)} n}{1 - A_{jn}^{(0)*} A_{jn}^{(0)} n},$$  \tag{14}

\[ \text{can be obtained from the residues at zeros of the denominator, using the explicit spatial behavior of scalar interaction functions (see Appendix A for details):} \]

$$A_{jn}^{(0)}(\epsilon) = \frac{T_j^{(0)}(\epsilon)}{2N} \sum_k e^{i\mathbf{k} \cdot \mathbf{n}} \left( G_k^{(1)} \right)_{jj} \approx \sqrt{\frac{r_{j,\epsilon}}{n}} \sin \frac{n}{r_0} \cos(\mathbf{K} \cdot \mathbf{n}) e^{-n/r_0}, \quad \tag{15}$$

where the characteristic scales:

$$r_0 = \frac{\hbar v_F}{\xi_0}, \quad r_\epsilon = r_0 \sqrt{\frac{\epsilon^2 + \epsilon_0^2}{(t_f^2 + V^2)(\epsilon_g^2 - \epsilon^2)}},$$

$$r_{j,\epsilon} = r_0 \left( \frac{\pi/2}{1 - U G_{jj}(\epsilon)} \right)^2.$$  

A similar behavior with two oscillating factors in effective inter-impurity interactions was previously indicated for the impurity states within superconducting gap in ferropnictides$^{31}$ where a faster cosine factor had Fermi wavelength. But the present case is simplified by the specific symmetry of $\mathbf{K}$ in the Brillouin zone, so that the square of $\cos(\mathbf{K} \cdot \mathbf{n})$ for all separations $\mathbf{n}$ between lattice sites of the same $j$th type only takes the values 1 and 1/4 (with respective densities 1/3 and 2/3) whose contributions can be then simply summed in Eq. 14. Then the most relevant feature is the sensible decrease of $r_{j,\epsilon}$ away from the impurity level. Therefore $r_\epsilon \gg r_{j,\epsilon}$ and the last exponential factor in Eq. 15 can be approximately taken unity at all distances $n < r_{j,\epsilon}$ contributing to Eq. 14, except for a very narrow vicinity of the impurity level:

$$\epsilon^{(j)} - \epsilon \approx \left( \frac{\epsilon_g - \epsilon^{(j)}}{\epsilon_g^{5/2} / \epsilon_g^{5/2}} \right), \quad \tag{16}$$

considered separately below. Otherwise, the contribution by Eqs. 14, 15 will vanish at all if $r_{j,\epsilon}$ gets shorter of $r_0$ (also alike the case of Ref.$^{31}$) that can occur for energies far enough from the impurity level: $\epsilon^{(j)} - \epsilon \approx \epsilon_g - \epsilon^{(j)}$, and as seen from Fig. 4 this is possible for the most dispersive $\cos^2(\mathbf{K} \cdot \mathbf{n})$ subband. Then, following the algorithm of subsequent integrations in Eq. 14, first over the poles of fast oscillating sine and then over its residues with a slow envelope function$^{31}$, we obtain explicitly for this subband (within the above indicated limits):

$$B_2(\epsilon) = \frac{3\pi}{8} \left( \frac{r_{2,\epsilon}}{a} \right)^2, \quad \tag{17}$$

with the prefactor resulted just from the weighting of $\cos^2(\mathbf{K} \cdot \mathbf{n})$ values. Then the above suggested expansion of $\det G^{-1}_k$ for $\epsilon$ near enough to $\epsilon^{(j)}$ (so that all $\Sigma_j$ but $\Sigma_1$ can be neglected) provides the linewidth:

$$\Gamma_2(\epsilon) \approx c_2 \left( \epsilon^{(2)} - \epsilon \right) B_2(\epsilon), \quad \epsilon^{(2)} - \epsilon \ll \epsilon_g - \epsilon^{(2)}, \quad \tag{18}$$

while its finite values for a more distant energy range could result from the higher order GE terms (of course, if exclude all other possible relaxation processes, as thermal phonons, electron-electron collisions, etc.) From Eq. 18, the IRM criterion is reduced to the inequality

$$c_2 B_2(\epsilon) \ll 1 \quad \text{(agreeing with the GE convergence) and, assuming Eq. 17 valid, leads to an estimate for the mobility edge:}$$

$$\epsilon^{(2)} - \epsilon_c \sim c^{1/4} \sqrt{\frac{W}{2\epsilon_0}} \left( \epsilon_g - \epsilon^{(2)} \right), \quad \tag{19}$$

and all the states closer to the impurity level $\epsilon^{(2)}$ are localized (on a certain 2nd type impurity center and on some its neighbor 2nd type impurity centers). When going to $\epsilon^{(2)}$ as close as by Eq. 16, the diverging $B_2(\epsilon)$ by Eq. 17 should saturate at $B_2(\epsilon^{(2)}) = (\pi/2)(3\epsilon_2/2)/8a^2$, however this limit could only apply if such closeness is reached before the above mobility range by Eq. 19.

The first conclusion from this estimate is that existence of the impurity subband itself is only assured if its bandwidth $\approx \epsilon^{(2)} - \epsilon_{g,2}$ surpasses the width of localized range around $\epsilon^{(2)}$ by Eq. 19. This is fulfilled when the total impurity concentration exceeds the critical value:

$$c_{cr} \sim \left( \frac{t_z}{W} \right)^{8/3} \left( \frac{|U|}{W} \right)^{4/3} \left( \frac{V}{W} \right)^{2/3} \times \frac{(1 + \cos \varphi)(1 + \cos^2 \varphi)}{\cos^{4/3} \varphi}, \quad \tag{20}$$

(it is obtained approximating Eq. 6 only to its diverging terms). Smallness of this value is mainly due to the essential factors of interlayer coupling, impurity perturbation, and applied bias, while a rather slow function of the "bias angle" $\varphi = \text{arctan} V/t_z$ stays of the order of unity for not too high $V$. Thus, for the sample choice of parameters, $W = 20t_z$, $|U| = 10t_z$, and $V = 2t_z$, we obtain $c_{cr} \sim 1.5 \cdot 10^{-5}$. Then for the example of $c = 0.01$ chosen in Fig. 3, the mobility edge $\epsilon_{c,2}$ extends from $\epsilon^{(2)}$ to about 0.38 of the distance $\epsilon_g - \epsilon^{(2)}$ while the dispersion of $\epsilon^{(2)}(\xi)$ subband is about four times bigger (see Fig. 4). At least, from comparison of Eqs. 19 and 16 it follows that for $c > c_{cr}$ the latter vicinity always occurs.
within the localized range and so cannot influence the above obtained estimates.

In summary, only the most dispersive impurity sub-band can be considered to really emerge beyond its mobility edge, with its main specifics in anomalously strong variation of the lifetimes $\tau(\varepsilon)$ along very narrow energy intervals. As to other formal solutions by Eq. 3 (analyzed with inclusion of the resting $B_j$), they are mostly invalidated within the common overlapped range of localized states by all the types of impurity centers, that extends up to the main gap edge $\varepsilon_g$ and contributes to its concentrational broadening. The latter value can be also found along the same lines, together with some finer details of the energy spectrum as, e.g., the mobility edge near the extremum $\varepsilon_{g,2} \approx \varepsilon^{(3)}(\xi_0)$ of the impurity subband, but these data are not so relevant for our main practical purpose to be considered below. In summary, the presented results give an important extension and diversification of the general scenario of collective restructuring of spectra of elementary excitations in crystals with impurities under external fields\textsuperscript{21}.

5. CONDUCTION AND BIASED METAL-INSULATOR TRANSITIONS

Now we can pass to the important processes of electric transport in the system with the above described band spectrum. For simplicity, this consideration is restricted to the case of zero temperature and the main attention is paid to the position of Fermi level $\varepsilon_F$ and to the lifetime $\tau_F$ of Fermi states under the applied bias control $V$ at given parameters of impurity perturbations $c$ and $U$. The basic condition for the Fermi level:

$$2 \int_{-\infty}^{\varepsilon_F} \rho(\varepsilon) d\varepsilon = 1 + c', \quad (21)$$

defines its shift from the zero energy position in the unperturbed system, in order to accommodate the total of $c'$ extra carriers per unit cell (brought by impurities themselves and possibly by some external sources). This generally requires knowledge of DOS functions for all the impurity subbands (besides almost non-perturbed ones for main subbands). But our main interest here is in finding possibility for $\varepsilon_F$ to be located within the most dispersive impurity band $\varepsilon^{(1)}(\xi)$, so we focus on the related DOS, especially close to this band termination $\varepsilon_{g,1}$ (Fig. 4). An important simplification of this task is obtained by noting that for this energy range all the self-energies $\Sigma_j$ in Eq. 10 can be taken constants, small enough compared to the gap width, thus the solutions of the dispersion equation, Eq. 3, almost reproduce here the non-perturbed $\varepsilon_2(\xi)$ band within to a constant shift of its edge from $\varepsilon_g$ to $\varepsilon_{g,1}$ (see Fig. 4), just due to the common effect of all $\Sigma_j$. The resulting DOS function:

$$\rho_2(\varepsilon) \approx \frac{2\varepsilon}{\sqrt{\varepsilon^2 - \varepsilon_{g,1}^2}}, \quad (22)$$

at $0 < \varepsilon - \varepsilon_{g,1} \lesssim \varepsilon^{(1)} - \varepsilon_{g,1}$ defines from Eq. 20 the Fermi level $\varepsilon_F$ position by the equation

$$c' \approx \left(\frac{2}{W}\right)^2 \sqrt{(t^2 + V^2)(\varepsilon^2_F - \varepsilon_{g,2}^2)}, \quad (23)$$

and the maximum permitted amount of carriers such that $\varepsilon_F$ stays within the conducting range, e.g., for the case of Fig. 4, results: $c'_{\text{max}} \approx 4 \cdot 10^{-3}$, that is somewhat lower than the proper impurity concentration $c = 10^{-2}$ in this case. Nevertheless, conduction through the impurity band can be realized if $c'$ is brought below the indicated limit of $c'_{\text{max}}$, e.g., by external compensation of a part of charge carriers\textsuperscript{21}. Once this assured, one can then strongly change the conductivity by rising the applied $V$, since the localized range width by Eq. 19 grows with bias faster than $\propto V^{2/3}$ against the almost bias insensitive (at $V \sim V_t$, see Fig. 3) width of the impurity band, while the Fermi level $\varepsilon_F$ goes to the band edge $\varepsilon_{g,1}$ slower than $\propto V^{-2}$. Then the faster advancing mobility edge $\varepsilon_{c,1}$ will finally cross $\varepsilon_F$ at some bias $V_{M-I}$, giving rise to the Mott metal-insulator transition and vanishing conductivity. Thus, for the proposed choice of $U = -W/2 = -10t$ and $c' = 3 \cdot 10^{-3}$, we obtain $V_{M-I} \approx 0.87$ eV. In this course, at $V \rightarrow V_{M-I}$, conductivity can vary by orders of magnitude, at driving the Fermi inverse lifetime $\tau_F^{-1} \sim \Gamma_2(\varepsilon_F)/\hbar$ close to divergence, under very tiny variations (say, some meV) of bias. This indicates a tremendous potentiality of such type of doped semiconducting systems in comparison with traditional materials.

At least, with further growing bias, the collapse of upper and lower mobility edges and the aforementioned Anderson transition to fully localized in-gap spectrum can be realized. From Eq. 20 this bias value estimates as $V_A \sim c^{3/2} W^2 |U|^{-2} t_z^{-4}$, but, with the natural choice of not too high $|U|$, this analytic estimate is only consistent with validity range of Eq. 20 ($V \lesssim t_z$) for as low impurity concentrations as $c \lesssim 10^{-3}$. But numerical analysis with use of full Eq. 6 shows that $V_A$ remains attainable for concentrations up to $c \sim 10^{-2}$ as well.

6. DISCUSSION AND CONCLUSIONS

The above main conclusion about the possibility to get an extensive control on electrical conduction through very slight variations of applied potential implies, of course, many additional factors to be taken into account. They can be indicated both from the theoretical and practical sides. Thus, the used theoretical approach is restricted to a simple model of impurity perturbation by a single on-site parameter, and some its elaboration could be done involving for instance perturbations of hopping parameters. Those kinds of analyses are known for traditional semiconductors with impurities and also demonstrated possibility for similar bands of collective states to be formed near localized impurity levels at high enough
impurity concentrations. Notably, for common materials, it was just the Lifshitz perturbation model that presented biggest theoretical problems for such effects, for instance, by leading to unrealistically high values of critical concentration, of order of unity or even more (unlike that by Eq. 19). This permits to expect that modifications of the present Lifshitz model, as in Ref. 15 for single impurity at gapless spectrum, or using the Anderson hybride model14 as in Ref. 15, if all the technical aspects be arranged, will not change essentially physical behavior of the system. On the other hand, there are yet many properties of this simple model that can be further studied, for instance, the possibilities to realize multiple conducting impurity subbands and subsequent processes of multiple switching between them, including, e.g., optical transitions under electrical biasing. Of course, a more realistic approach should also involve the effects of other defects in the graphene structures (vacancies, boundaries, domain borders, etc.), as well as the above mentioned Coulomb interactions, thermal effects, etc. Generally, this would require to reach the basic scales of the impurity band structure above a certain "background" relaxation level, that could be achieved by variations of either impurity sort (that is, $U$ parameter) and concentration or/and the applied bias $V$. At least, similar impurity multiband effects can be also sought in other atomically multilayered systems, as those mentioned in the Introduction, where a special focus might be on tuned bandgap in silicene bilayers (yet wider than in bigraphene10) or even on single layers of buckled silicene or germanene30.

As to the practical issues, first of all, rather strict conditions on fabrication of the basic doped bilayered system are in order, perhaps mainly aimed to minimize all the "foreign" defects vs the chosen dopants, but the next requirement to keep the levels of dopants (and possibly their compensating species) within to fractions of percent should not be a real problem for modern nanoelectronics. A special attention is also required for precise control and manipulation of the applied bias $V$, in particular, exploring possibilities to realize its near-critical and supercritical regimes, like those indicated in the above analysis. Finally, the practical arrangement of an experimental transistor-type setup based on the suggested conductivity control by tiny impurity subbands would perhaps require some specific technical solutions, but they do not look too difficult to be found in the available engineering depository. Thus, a fair hope exists for this theoretical proposition to be realized in a practical device.

In conclusion, the effects of localized on-site perturbations by rather disperse impurities on bilayered graphene system under the applied electrical bias between the layers are theoretically considered using the Green function techniques adapted for a multiband electronic system, demonstrating the conditions for different types of localized impurity energy levels to appear within the bias-induced bandgap in the electronic spectrum of this system and then extension of these levels into specific narrow energy bands at impurity concentration surpassing certain characteristic values. The analysis on these processes demonstrated their similarities to those known from literature on various crystalline materials with impurities, also some specifics of the present system was shown in considerable bias dependences of impurity bands and of critical concentrations for their formation. These dependences can be further treated to provide some specific phase diagrams in variables "bias-concentration". A practical application of the described electronic band structure is suggested in a form of highly sensitive bias control on the system conductivity through the impurity subband when brought close to a regime of bias-controlled Mott’s metal-insulator transition.

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\[ \frac{1}{2N} \sum_{k} e^{i k \cdot n} \left( G^{(0)}_{k} \right)_{j j} = \frac{2 \cos K \cdot n}{W^2} \int_{0}^{W} J_0 \left( \frac{\xi n}{W} \right) \frac{(N_j(\xi) - \xi^2)\xi d\xi}{(\xi^2 - \xi_j^2)(\xi^2 - \xi_j^2)} \] (A1)

where \( J_0 \) is the zeroth order Bessel function, \( \xi_{1,2}^2 = \varepsilon^2 + \varepsilon_2^2 \pm i \sqrt{(t_2^2 + V^2)(\varepsilon_2^2 - \varepsilon^2)} \) are the complex poles of \( \det G_{k}^{(0)} \) in \( \xi \)-variable and all \( |N_j(\xi)| \sim \varepsilon_2 \) (as seen from Eq. 6). Since this integral is fast converging after \( \xi \gtrsim \varepsilon_2 \), its upper limit can be safely extended to infinity. Then, after expanding the factor besides the Bessel function in simple fractions:

\[ \frac{N_j(\xi, \xi)}{(\xi^2 - \xi_j^2)(\xi^2 - \xi_j^2)} = \frac{N_j(\xi) - \xi_j^2}{\xi^2 - \xi_j^2} - \frac{N_j(\xi) - \xi_j^2}{\xi^2 - \xi_j^2}, \] (A2)

the Hankel-Nicholson integration formula can be applied to each of them:

\[ \int_{0}^{\infty} J_0(\xi) x dx = K_0(\xi), \] (A3)

with the zeroth order MacDonald function \( K_0 \), valid for complex \( \xi \) if \( \text{Re} \xi > 0 \) (Ref. 37). The \( z \)-arguments related to the terms in Eq. A2, can be defined as \( z_{1,2}^2 = -\xi_{1,2}^2(n/hV)^2 \) and the above requirement will read \( \text{Re} \sqrt{-\xi_{1,2}^2} > 0 \). For the relevant energy range \( 0 < \varepsilon_3 - \varepsilon \ll \varepsilon_2 \), we can use the choices \( \sqrt{-\xi_{1,2}^2} = \sqrt{\sqrt{(\varepsilon_1^2 - \varepsilon^2)(\varepsilon_2^2 - \varepsilon^2)} - \varepsilon^2 - \varepsilon_2^2 \mp i \sqrt{(\varepsilon_1^2 - \varepsilon^2)(\varepsilon_2^2 - \varepsilon^2)} + \varepsilon^2 + \varepsilon_2^2} \). At least, for relevant distances \( n \geq r_0 \), the resulting \( K_0(z_{1,2}) \) have big enough arguments, \( |z_{1,2}| = |n\xi_{1,2}/hV| \gtrsim 1 \), to use their asymptotics: \( K_0(z_1) \approx -\sqrt{2/\pi z_1}e^{-z_1} \) and \( K_0(z_2) \approx \sqrt{2/\pi z_2}e^{-z_2} \). Then, taking account of all prefactors besides these expressions, present in Eqs. 15 and A2, we arrive at the final result of Eq. 15.