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# Random-Gap Model for Graphene and Graphene Bilayers

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The effect of a randomly fluctuating gap, created by a random staggered potential, is studied in a monolayer and a bilayer of graphene. The density of states, the one-particle scattering rate, and the transport properties (diffusion coefficient and conductivity) are calculated at the neutrality point. All of these quantities vanish at a critical value of the average staggered potential, signaling a continuous transition to an insulating behavior. Transport quantities are directly linked to the one-particle scattering rate. Although the behavior is qualitatively the same in mono- and bilayers, the effect of disorder is much stronger in the latter.

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Graphene, a sheet of carbon atoms, or bilayer graphene are semimetals with good conducting properties [1–3]. In particular, the minimal conductivity at the neutrality point (NP) is very robust and almost unaffected by disorder or thermal fluctuations [3–6]. Recent experiments with hydrogenated graphene [7] and biased bilayer graphene [8–10] have revealed that a staggered potential (SP) can be created in graphene and bilayer graphene which breaks the sublattice symmetry. This opens a gap at the Fermi energy, leading to an insulating behavior. With this opportunity, one enters a new field, where one can switch between a conducting and an insulating regime of a two-dimensional material, either by a chemical process (e.g., oxidation or hydrogenation) or by applying an external electric field [11].

It is clear that the opening of a uniform gap destroys the metallic state immediately. This means that the (minimal) conductivity at the NP drops from a finite value of order  $e^2/h$  directly to zero. In a realistic system, however, the gap may not be uniform. This means that locally gaps open, whereas in other regions of the sample there is no gap. The situation can be compared with a classical random network of broken and unbroken bonds. The conductivity of such a network is nonzero as long as there is a percolating cluster of unbroken bonds. In such a system, the transition from conducting to insulating behavior is presumably a second-order percolation transition [12].

Disorder in graphene has been the subject of a number of recent numerical studies [13,14]. The results can be summarized by the statement that chiral-symmetry-preserving disorder provides delocalized states, whereas a chiral-symmetry-breaking potential disorder leads to Anderson localization, even at the NP.

Conductivity and other transport properties in graphene can be evaluated by solving the Bethe-Salpeter equation for the average two-particle Green's function (Cooperon) [15–19]. Unfortunately, the Bethe-Salpeter equation is usually a complex matrix equation which is difficult to handle. Therefore, a different approach will be employed here that eliminates a part of the complexity by focusing on

continuous symmetries and spontaneous symmetry breaking. This allows us to identify a (massless) diffusion mode in the system with a randomly fluctuating gap. Consequently, diffusion can stop only when the spontaneous symmetry breaking vanishes. It will be discussed in this Letter that this can happen if the average SP approaches a critical value. Moreover, there is no drop of the conductivity but a continuous decay to zero, depending on the fluctuations of the SP. The fact that the density of states does not vanish at the NP of the pure bilayer leads to a much bigger effect of a random SP in comparison with a monolayer.

*Model.*—Quasiparticles in monolayer graphene (MLG) or bilayer graphene (BLG) are described in tight-binding approximation by a nearest-neighbor hopping Hamiltonian

$$\mathbf{H} = -t \sum_{\langle r, r' \rangle} c_r^\dagger c_{r'} + \sum_r m_r c_r^\dagger c_r + \text{H.c.}, \quad (1)$$

where the underlying structure is either a honeycomb lattice (MLG) or two honeycomb lattices with Bernal stacking (BLG).

The sublattice symmetry of the honeycomb lattice is broken by a SP  $m_r$  which is positive (negative) on sublattice A (B) [20,21]. Such a potential can be the result of chemical absorption of other atoms (e.g., oxygen or hydrogen [7]) or of an external gate voltage applied to the two layers of BLG [8]. Neither in MLG nor in BLG are the potential  $m_r$  and, therefore, the gap uniform, because of fluctuations in the coverage of the MLG by additional noncarbon atoms or by the fact that the graphene sheets are not planar [22–24]. Deviations from the planar structure in the form of ripples cause fluctuations in the distance between the two sheets in BLG which results in an inhomogeneous potential  $m_r$  along each sheet. It is assumed that the gate voltage is adjusted at the NP such that on average the SP is exactly antisymmetric:  $\langle m_A \rangle = -\langle m_B \rangle$ .

At first glance, the Hamiltonian in Eq. (1) is a standard hopping Hamiltonian with random potential  $m$ , frequently used to study the generic case of Anderson localization [25]. The dispersion, however, is special in the case of

graphene due to the honeycomb lattice: At low energies, it consists of two valleys  $K$  and  $K'$  [18,23]. It is assumed that weak disorder scatters only at small momentum such that intervalley scattering is not relevant. Then each valley contributes separately to transport, and the contribution of the two valleys to the conductivity  $\sigma$  is additive:  $\sigma = \sigma_K + \sigma_{K'}$ . This allows us to consider for the low-energy properties a Dirac-type Hamiltonian for each valley separately:

$$H = h_1\sigma_1 + h_2\sigma_2 + m\sigma_3, \quad (2)$$

with Pauli matrices  $\sigma_j$  and with  $h_j$

$$h_j = i\nabla_j \text{ (MLG)}, \quad h_1 = \nabla_1^2 - \nabla_2^2, \quad h_2 = 2\nabla_1\nabla_2 \text{ (BLG)}. \quad (3)$$

$\nabla_j$  is the differential operator in  $j$  ( $=1, 2$ ) direction. Within this approximation, the SP  $m_r$  is a random variable with mean value  $\langle m_r \rangle_m = \bar{m}$  and variance  $\langle (m_r - \bar{m}) \times (m_{r'} - \bar{m}) \rangle_m = g\delta_{r,r'}$ . The following transport calculations will be based entirely on the Hamiltonian of Eq. (2). This is justified, since additional terms, which may break the symmetry (nearest-neighbor hopping term, weak coupling between the valleys), are small and can be included as a small energy shift of the NP. The average Hamiltonian  $\langle H \rangle_m$  is translational invariant and can be diagonalized by Fourier transformation:  $k_1\sigma_1 + k_2\sigma_2 + \bar{m}\sigma_3$  for MLG with eigenvalues  $E_k = \pm\sqrt{\bar{m}^2 + k^2}$ . For BLG the average Hamiltonian is  $(k_1^2 - k_2^2)\sigma_1 + 2k_1k_2\sigma_2 + \bar{m}\sigma_3$  with eigenvalues  $E_k = \pm\sqrt{\bar{m}^2 + k^4}$ .

*Symmetries.*—Transport properties are controlled by the symmetry of the Hamiltonian and of the corresponding one-particle Green's function  $G(i\epsilon) = (H + i\epsilon)^{-1}$ . In the absence of sublattice-symmetry breaking (i.e., for  $m = 0$ ), the Hamiltonian  $H = h_1\sigma_1 + h_2\sigma_2$  has a continuous chiral symmetry

$$H \rightarrow e^{\alpha\sigma_3} H e^{\alpha\sigma_3} = H, \quad (4)$$

with a continuous parameter  $\alpha$ , since  $H$  anticommutes with  $\sigma_3$ . The SP term  $m\sigma_3$  breaks the continuous chiral symmetry. However, the behavior under transposition  $h_j^T = -h_j$  for MLG and  $h_j^T = h_j$  for BLG provides a discrete symmetry:

$$H \rightarrow -\sigma_j H^T \sigma_j = H, \quad (5)$$

where  $j = 1$  for MLG and  $j = 2$  for BLG. This symmetry is broken for the one-particle Green's function  $G(i\epsilon)$  by the  $i\epsilon$  term. To see whether or not the symmetry is recovered for  $\epsilon \rightarrow 0$ , the difference of  $G(i\epsilon)$  and the transformed Green's function  $-\sigma_j G^T(i\epsilon) \sigma_j$  must be evaluated:

$$G(i\epsilon) + \sigma_j G^T(i\epsilon) \sigma_j = G(i\epsilon) - G(-i\epsilon). \quad (6)$$

For the diagonal elements, this is the density of states at the NP  $\rho(E = 0) \equiv \rho_0$  in the limit  $\epsilon \rightarrow 0$ . Thus the order parameter for spontaneous symmetry breaking is  $\rho_0$ .

*Conductivity.*—The conductivity can be calculated from the Kubo formula. Here we focus on interband scattering between states of energy  $\omega/2$  and  $-\omega/2$ , which is a major contribution to transport near the NP. The frequency-dependent conductivity then reads [26]

$$\sigma_0(\omega) = -\frac{e^2}{2h} \omega^2 \langle \langle \Phi_{\omega/2} | r_k^2 | \Phi_{-\omega/2} \rangle \rangle_m, \quad (7)$$

where  $|\Phi_E\rangle$  is an eigenstate of  $H$  in Eq. (2) with energy  $E$ . In other words, the conductivity is proportional to a matrix element of the position operator  $r_k$  ( $k = 1, 2$ ) with respect to energy eigenfunctions from the lower and the upper band. The matrix element  $\langle \Phi_{\omega/2} | r_k^2 | \Phi_{-\omega/2} \rangle$  is identical with the two-particle Green's function

$$\sum_r r_k^2 \text{Tr}_2 [G_{r0}(-\omega/2 - i\epsilon) G_{0r}(\omega/2 + i\epsilon)]. \quad (8)$$

This indicates that transport properties are expressed by the two-particle Green's function  $G(i\epsilon)G(-i\epsilon)$ . Each of the two Green's functions  $G(i\epsilon)$  and  $G(-i\epsilon)$  can be considered as a random variable which are correlated due to the common random variable  $m_r$ . Their distribution is defined by a joint distribution function  $P[G(i\epsilon), G(-i\epsilon)]$ . In terms of transport theory, both Green's functions must be included on equal footing. This is possible by introducing the extended Green's function

$$\hat{G}(i\epsilon) = \begin{pmatrix} G(i\epsilon) & 0 \\ 0 & G(-i\epsilon) \end{pmatrix} = \begin{pmatrix} H + i\epsilon & 0 \\ 0 & H - i\epsilon \end{pmatrix}^{-1}.$$

In the present case, one can use the symmetry transformation of  $H$  in Eq. (5) to write the extended Green's function as

$$\begin{pmatrix} \sigma_0 & 0 \\ 0 & i\sigma_j \end{pmatrix} \begin{pmatrix} H + i\epsilon & 0 \\ 0 & H^T + i\epsilon \end{pmatrix}^{-1} \begin{pmatrix} \sigma_0 & 0 \\ 0 & i\sigma_j \end{pmatrix}.$$

This introduces an extended Hamiltonian  $\hat{H} = \text{diag}(H, H^T)$  which is invariant under a global “rotation”

$$\hat{H} \rightarrow e^S \hat{H} e^S = \hat{H}, \quad S = \begin{pmatrix} 0 & \alpha\sigma_j \\ \alpha'\sigma_j & 0 \end{pmatrix} \quad (9)$$

with continuous parameters  $\alpha$  and  $\alpha'$ , since  $\hat{H}$  anticommutes with  $S$ . The  $i\epsilon$  term of the Green's function also breaks this symmetry. For  $\alpha\alpha' = -\pi^2/4$ , the diagonal element of  $\hat{G} - e^S \hat{G} e^S$  is proportional to the density of states  $\rho_0$ , similar to Eq. (6). Thus, the symmetry is spontaneously broken for  $\epsilon \rightarrow 0$  if  $\rho_0$  is nonzero. Since this is a continuous symmetry, there is a massless mode which describes diffusion [27]. Symmetry breaking should be studied for average quantities. Therefore, the average density of states must be evaluated.

*Spontaneous symmetry breaking.*—The average one-particle Green's function can be calculated from the average Hamiltonian  $\langle H \rangle_m$  by employing the self-consistent Born approximation (SCBA) [15,16,21]

$$\langle G(i\epsilon) \rangle_m \approx (\langle H \rangle_m - 2\Sigma)^{-1} \equiv G_0(i\eta, m_s). \quad (10)$$

The self-energy  $\Sigma$  is a  $2 \times 2$  tensor due to the spinor structure of the quasiparticles:  $\Sigma = -(i\eta\sigma_0 + m_s\sigma_3)/2$ . Scattering by the random SP produces an imaginary part of the self-energy  $\eta$  (i.e., a one-particle scattering rate) and a shift  $m_s$  of the average SP  $\bar{m}$  (i.e.,  $\bar{m} \rightarrow m' \equiv \bar{m} + m_s$ ).  $\Sigma$  is determined by the self-consistent equation

$$\Sigma = g\sigma_3(\langle H \rangle_m - 2\Sigma)_{rr}^{-1}\sigma_3. \quad (11)$$

For simplicity, the dc limit  $\omega \sim 0$  is considered here. Then the average density of states at the NP is proportional to the scattering rate:

$$\rho_0 = \frac{1}{\pi} \text{Im} G_{0,rr} = -\frac{1}{g\pi} \text{Im} \Sigma = \frac{\eta}{2g\pi}. \quad (12)$$

This demonstrates that scattering by the random SP creates a nonzero density of states at the NP. It should be noticed that the entire calculation of the scattering rate  $\eta$  is based on the average one-particle Green's function. Therefore, it is not related to the continuous symmetry of Eq. (9). On the other hand,  $\eta > 0$  reflects spontaneous breaking of this symmetry.

Equation (11) can also be written in terms of two equations, one for the one-particle scattering rate  $\eta$  and another for the shift of the SP  $m_s$ :

$$\eta = gI\eta, \quad m_s = -\bar{m}gI/(1 + gI). \quad (13)$$

$I$  is a function of  $\bar{m}$  and  $\eta$  and also depends on the Hamiltonian. For MLG it reads with momentum cutoff  $\lambda$

$$I = \frac{1}{2\pi} \ln \left[ 1 + \frac{\lambda^2}{\eta^2 + (\bar{m} + m_s)^2} \right] \quad (14)$$

and for BLG

$$I \sim \frac{1}{4\sqrt{\eta^2 + (\bar{m} + m_s)^2}} (\lambda \sim \infty). \quad (15)$$

A nonzero solution  $\eta$  requires  $gI = 1$  in the first part of Eq. (13), such that  $m_s = -\bar{m}/2$  from the second part. Since the integrals  $I$  are monotonically decreasing functions for large  $\bar{m}$ , a real solution with  $gI = 1$  exists only for  $|\bar{m}| \leq m_c$ . For both MLG and BLG, the solutions read

$$\eta^2 = (m_c^2 - \bar{m}^2)\Theta(m_c^2 - \bar{m}^2)/4, \quad (16)$$

where the model dependence enters only through the critical average SP  $m_c$ :

$$\begin{cases} \frac{2\lambda}{\sqrt{e^{2\pi/g} - 1}} \sim 2\lambda e^{-\pi/g} & (\text{MLG}) \\ g/2 & (\text{BLG}). \end{cases} \quad (17)$$

$m_c$  is much bigger for BLG (cf. Fig. 1), a result which indicates that the effect of disorder is much stronger in BLG. This is also reflected by the scattering rate at  $\bar{m} = 0$  which is  $\eta = m_c/2$ . This difference can be easily understood by comparing  $\rho_0$  of both models in the absence of disorder, where the scattering rate  $\eta$  vanishes. The spectrum without a SP is linear for MLG and quadratic for BLG. Then the density of states is linear and vanishes at the NP for MLG, whereas it is constant for BLG. [This follows

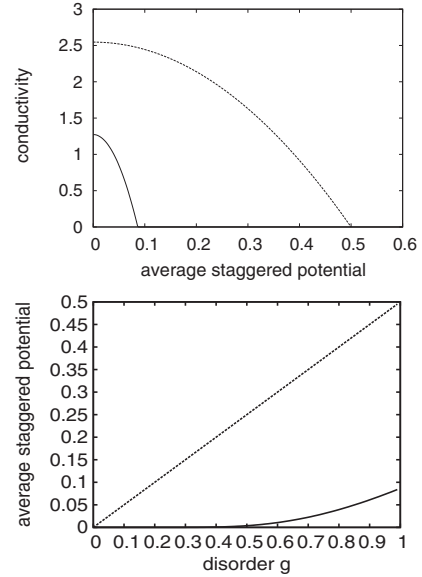


FIG. 1. Upper panel: dc conductivity in units of  $e^2/h$  for ML graphene (full curve) and BL graphene (dashed curve) vs the average staggered potential  $\bar{m}$ , calculated from Eq. (19) for  $g = 1$  and  $\lambda = 1$ . Lower panel: Critical average staggered potential as a function of  $g$  (variance of the staggered potential fluctuations) for ML graphene (full curve) and BL graphene (dashed curve).

also from the expression in Eq. (12) by taking the limit  $g \rightarrow 0$ , since  $\eta = g/4$  for  $\bar{m} = 0$  gives  $\rho_0 = 1/8\pi$  in the case of BLG, and  $\eta = \lambda e^{-\pi/g}$  gives  $\rho_0 \propto e^{-\pi/g}/g \sim 0$  in the case of MLG.] Consequently, there is much more scattering in the presence of disorder (with a much bigger  $\eta$ ) near the NP for BLG than for MLG.

**Diffusion.**—The average matrix element of the position operator  $\langle\langle \Phi_{\omega/2} | r_k^2 | \Phi_{-\omega/2} \rangle\rangle_m$  in the conductivity expression of Eq. (7) can be evaluated from an effective field theory [27]. If  $\eta > 0$ , the corresponding spontaneous breaking of the symmetry in Eq. (9) creates a single massless mode and a divergent matrix element for  $\omega \sim 0$  [28]:

$$\langle\langle \Phi_{\omega/2} | r_k^2 | \Phi_{-\omega/2} \rangle\rangle_m \approx -\frac{\eta^2}{(\omega/2)^2} \langle\Phi_{i\eta}^0 | r_k^2 | \Phi_{-i\eta}^0 \rangle, \quad (18)$$

where  $|\Phi_E^0\rangle$  is eigenfunction of the translational-invariant Hamiltonian  $\langle H \rangle_m$  with eigenvalue  $E$ . This relation is similar to the relation of the average one-particle Green's function in the SCBA of Eq. (10). Like in the latter case, the averaging process leads to a change of energies  $\omega/2 \rightarrow i\eta$  (i.e., a replacement of the frequency by the scattering rate). Moreover, in the relation of the matrix element there is an extra prefactor  $-\eta^2/(\omega/2)^2$ . This is important for the transport properties, since it cancels the factor  $\omega^2$  in the conductivity of Eq. (7). The relation in Eq. (18) can be understood as a factorization of the averaged matrix element into a product of a power law (i.e.,  $\sim \omega^{-2}$ ) and a smooth scaling function  $\eta^2 \langle\Phi_{i\eta}^0 | r_k^2 | \Phi_{-i\eta}^0 \rangle$ . This means that the states  $|\Phi_{\pm\omega/2}\rangle$  are delocalized for  $\omega = 0$  in the pres-



ence of weak SP disorder and localized for  $\omega \neq 0$  with a decreasing localization length as one goes away from the NP. Such a behavior was also found for bond disorder in analytic [26] and in numerical studies [14].

After evaluating  $\langle \Phi_{i\eta}^0 | r_k^2 | \Phi_{-i\eta}^0 \rangle$ , the conductivity in Eq. (7) reads

$$\sigma_0 \approx \frac{a}{\pi} \frac{\eta^2}{\eta^2 + \bar{m}^2/4} \frac{e^2}{h}, \quad (19)$$

where  $a = 1$  ( $a = 2$ ) for MLG (BLG). First, this result indicates that the relevant quantity is the one-particle scattering rate  $\eta$ . The difference between MLG and BLG is only due to the parameter  $a = 1, 2$  and due to the specific  $\bar{m}$  dependence of  $\eta$ . Second, the result reflects a diffusive behavior as long as the scattering rate  $\eta$  does not vanish, with diffusion coefficient  $D = \sigma_0/\rho_0 \propto \eta/(\eta^2 + \bar{m}^2/4)$  due to the Einstein relation.

Equation (16) gives a vanishing scattering rate for  $\bar{m} \geq m_c$ , where the critical value  $m_c$  is twice the scattering rate at  $\bar{m} = 0$ . Moreover, a global gap opens only for  $\bar{m} > m_c$ , since the average density of states at the NP is proportional to  $\eta$ . There is a qualitative difference in the details of the transport properties at  $\bar{m} = 0$ , on the one hand, and for  $\bar{m} \neq 0$ , on the other:

$\bar{m} = 0$ .—A fluctuating SP with variance  $g$  not too large (here we work in the weak disorder regime) has no effect on the minimal conductivity. In terms of the Einstein relation  $\sigma_0 \propto \rho_0 D$ , this means that an increase of  $\rho_0$  due to increased scattering is compensated by a reduction of the diffusion coefficient. Such a behavior was also observed in the chiral-invariant case with random bond disorder which is related to ripples [3,16,22,27].

$\bar{m} \neq 0$ .—The conductivity decreases with  $\bar{m}$  and eventually goes to zero at  $\bar{m} = m_c$ . This is due to two effects, namely, the reduction of the density of states and the reduction of the diffusion coefficient with  $\bar{m}$ , caused by a fluctuating gap.

The different spectra of MLG and BLG have quantitative consequences for the conductivity through  $\eta$ : For BLG it is twice as big as for MLG at  $\bar{m} = 0$  (this was also found earlier for  $g = 0$  [29]) and also decays on a larger scale for  $0 < \bar{m} \leq m_c$ , since the critical value is  $m_c = g/2$  for BLG, whereas it is  $m_c \sim \exp(-\pi/g)$  for MLG. As shown in Fig. 1, the conductivity of MLG vanishes for a given  $g$  at much lower values of  $\bar{m}$ .

Our result of the random SP represents a case that is different from random bond disorder (i.e., for a system with chiral symmetry) and random scalar potential (which breaks the chiral symmetry but not the sublattice symmetry). The former does not localize states at the NP, whereas the latter has presumably always localized states, with a very large localization length though. In a recent paper, Zhang *et al.* suggested a Kosterlitz-Thouless (KT) transition for a long-range random scalar potential [13]. The KT transition is a phase transition that has no spontaneous symmetry breaking but a single massless mode in the

ordered phase due to  $U(1)$  phase fluctuations. In the case of the random SP, the situation is very different: There is spontaneous symmetry breaking in the diffusive phase due to  $\eta > 0$ . Moreover, the symmetry of the fluctuations in Eq. (9) has two components rather than one. Therefore, the transition to the insulating behavior in the case of a random SP cannot be linked to the conventional KT transition.

A possible experimental realization of a random gap was recently observed by Adam *et al.* [30]. It still remains to be seen whether or not the observed transition, which was studied by varying the gate voltage at a fixed gap, can be related to a nonzero average SP. This would require a tuning of the gap fluctuations and measurement of the local density of states.

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