Effect of weak disorder on the density of states in graphene

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The effect of weak potential and bond disorder on the density of states of graphene is studied. By comparing the self-consistent noncrossing approximation on the honeycomb lattice with perturbation theory on the Dirac fermions, we conclude that the linear density of states of pure graphene changes to a nonuniversal power law whose exponent depends on the strength of disorder like $1 - 4g/\sqrt{3}\pi^2$, with g the variance of the Gaussian disorder and t the hopping integral. This can result in a significant suppression of the exponent of the density of states in the weak-disorder limit. We argue that even a nonlinear density of states can result in a conductivity that is proportional to the number of charge carriers, in accordance with experimental findings.

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I. INTRODUCTION

Graphene is a single sheet of carbon atoms with a honeycomb lattice, exhibiting interesting transport properties. These are ultimately connected to the low-energy quasiparticles of graphene, i.e., two-dimensional Dirac fermions. Its conductivity depends linearly on the carrier density and reaches a universal value in the limit of vanishing carrier density. The former has been explained by the presence of charged impurities, while the latter does not allow a charged particle-hole symmetric filling of the half-integer quantum Hall effect is explained in terms of nonlinear density of states, we evaluate the conductivity away from the Dirac point by using the Einstein relation. We show that based on the specific form of the diffusivity coefficient, this can result in a conductivity, depending linearly on the carrier concentration, and in a mobility, decreasing with increasing disorder. These are in accord with a recent experiment on K adsorbed graphene. By varying the K doping time, the impurity strength was controlled. The conductivity away from the Dirac point still depends linearly on the charge carrier concentration, but its slope, the mobility, decreases steadily with doping time.

Our results apply to other systems with Dirac fermions such as the organic conductor α-(BEDT-TTF)$_2$I$_3$.

II. HONEYCOMB DISPERSION

We start with the Hamiltonian describing quasiparticles on the honeycomb lattice, given by

$$H_0 = h_1\sigma_1 + h_2\sigma_2,$$

where $\sigma_i$'s are the Pauli matrices, representing the two sublattices. Here,

$$h_1 = -t \sum_{j=1}^{3} \cos(a_jk), \quad h_2 = -t \sum_{j=1}^{3} \sin(a_jk),$$

with $a_1 = a(-\sqrt{3}/2, 1/2)$, $a_2 = a(0, -1)$, and $a_3 = a(\sqrt{3}/2, 1/2)$ pointing toward nearest neighbors on the honeycomb lattice,
\(a\) is the lattice constant, and \(t\) is the hopping integral. The resulting honeycomb dispersion is given by \(\pm \sqrt{h_1^2 + h_2^2}\), which vanishes at six points in the Brillouin zone. To take scattering into account, we consider the mutual coexistence of both Gaussian potential (on-site) disorder (with matrix element \(V_{o,r}\), satisfying \(\langle V_{o,r} \rangle = 0\) and variance \(\langle V_{o,r} V_{o,r}' \rangle = g_o \delta_{rr}\)) and bond disorder in only one direction (in addition to the uniform hopping with matrix element \(V_{b,r}\), satisfying \(\langle V_{b,r} \rangle = 0\) and variance \(\langle V_{b,r} V_{b,r}' \rangle = g_b \delta_{rr}\)), which is thought to describe reliably the more complicated case of disorder on all bonds, and is shown in Fig. 1. In graphene, ripples can represent the main source of disorder and are approximated by random nearest-neighbor hopping rates, while potential disorder might only be relevant close to the Dirac point. The corresponding term in the Hamiltonian is
\[
V = V_{o,r} \sigma_0 + V_{b,r} \sigma_1,
\]
which results in \(H = H_0 + V\).

Without magnetic field, the self-energy for the Green’s function, which takes all noncrossing diagrams to every order into account (noncrossing approximation, NCA), can be found self-consistently from
\[
\Sigma(i\omega_n) = \frac{1}{1 - (g_o + g_b)G^2} - G,
\]
where \(i\omega_n\) is the fermionic Matsubara frequency and
\[
G = G_0[i\omega_n - \Sigma(i\omega_n)].
\]
Here, \(G_0\) is the unperturbed local Green’s function on the honeycomb lattice, given by
\[
G_0(z) = \frac{A_c}{(2\pi)^2} \int \frac{zd^2k}{z^2 - t^2[4 \cos(\sqrt{3}k_x/2) \cos(3k_y/2) + 2 \cos(\sqrt{3}k_y) + 3]},
\]
where \(A_c = 3\sqrt{3}a^2/2\) is the area of the unit cell, and the integral runs over the hexagonal Brillouin zone with corners given by the condition \(h_1^2 + h_2^2 = 0\). This can further be brought to a closed form using the results of Ref. 27. On the other hand, in the continuum representation, using the Dirac Hamiltonian, the above Green’s function simplifies to
\[
G_0(z) = \frac{2A_c \sigma_0}{(2\pi)^2} \int \frac{d^2k}{z + v(k_x \sigma_1 + k_y \sigma_2)} \cdot - \frac{A_c}{2 \pi v^2 \rho} \ln \left(1 - \frac{\lambda^2}{z^2}\right),
\]
and \(v = 3ta/2\). The cutoff \(\lambda\) can be found by requiring the number of states in the Brillouin zone to be preserved in the Dirac case as well. This leads to \(\lambda = \sqrt{3t}\). Another possible choice relies on the comparison of the low frequency parts of the Green’s function in the lattice and in the continuum limit, which reveals the presence of \(\ln(\omega/3t)\) terms. This leads to \(\lambda = 3t\), which coincides with the real bandwidth on the lattice. We are going to use this form in the following. The difference of the variances becomes important when calculating the second order correction (in variance) to the self-energy. It is clear from Eq. (5) that the same self-energy is found for pure potential or unidirectional bond disorder. The effect of their coexistence is strongest when they possess the same variance. From this, the density of states follows as
EFFECT OF WEAK DISORDER ON THE DENSITY OF...

\[ G(z) = G_0 + G_0 V G_0 + G_0 V G_0 V G_0 + \ldots, \]

where \( V = V_{o,r} \sigma_0 + V_{b,r} \sigma_1 \) describes both Gaussian potential and unidirectional bond disorder, \( G = (z-H_0-V)^{-1} \) and \( G_0 = (z-H_0)^{-1} \), and \( H_0 = \nu(k, \sigma_1 + k, \sigma_2) \) is the Dirac Hamiltonian. After averaging over disorder, we get

\[ \langle G_{rr} \rangle = G_{0,rr} + g \sum_{r'} G_{0,rr} G_{0,r'r} G_{0,r'r} + \ldots \]

with \( g = g_o + g_b \). The Green’s function \( G_0 \) is translationally invariant and reads from Eq. (8) at real frequencies as

\[ G_{0,rr} = \frac{A_r E}{2 \pi \nu^2} \ln \left( \frac{\lambda^2}{E^2} - 1 - i \pi \right). \]

This implies

\[ \langle G_{rr} \rangle = G_{0,rr} + g (G_{0,rr})_r G_{0,rr} + o(g^2) \]

\[ = G_{0,rr} [1 + g (G_{0,rr})^2] + o(g^2). \]

Moreover, we have, with Eq. (14),

\[ (G_{0,rr})^2 = \frac{A_r}{(2\pi)^2} \int \frac{d^2k}{[z + \nu(k, \sigma_1 + k, \sigma_2)]^2} \]

\[ = - \frac{\partial G_{0,rr}}{\partial z} \]

\[ = \frac{A_r}{2 \pi \nu^2} \left[ \ln \left( 1 - \frac{\lambda^2}{z^2} \right) - \frac{2\lambda^2}{z^2 - \lambda^2} \right] \sigma_0 \]

\[ \approx \frac{A_r}{2 \pi \nu^2} \left[ -2 \ln \left( \frac{|E|}{\lambda} \right) - i \pi \right] \sigma_0 \]

for \( \lambda \gg |E| \gg \epsilon \). Therefore, we obtain

\[ \langle G_{rr} \rangle = G_{0,rr} \left[ 1 - \frac{A_r g}{\pi \nu^2} \ln \left( \frac{|E|}{\lambda} \right) - \frac{i g A_r}{2 \nu^2} \ln \left( \frac{|E|}{\lambda} \right) \right] + o(g^2). \]

From this, the density of states follows as

\[ \rho(E) = - \frac{1}{\pi} \text{Im} \langle G_{rr} \rangle = \frac{A_r E}{2 \pi \nu^2} \left[ 1 - 2 \frac{g A_r}{\pi \nu^2} \ln \left( \frac{|E|}{\lambda} \right) \right]. \]

If we further assume that: (i) the density of states as a function of \( E \) satisfies a power law (inspired by Refs. 16–18) and (ii) the disorder strength \( g \) is small, we can formally consider
Eq. (18) as the lowest order expansion in disorder and sum it up to a scaling form as

$$\rho(E) = \frac{A \lambda}{2 \pi v^2} \left( \frac{E}{\lambda} \right)^{1-(2gA/\pi v^2)}.$$  \hspace{1cm} (19)

Hence, this suggests that the linear density of states of pure graphene changes into a nonuniversal power law depending on the strength of the disorder as $\gamma = 1 - (4g/\pi v^2 r)^2$. Note that the exponent does not depend on the ambiguous cutoff $\lambda$. We mention that Eq. (18) might suggest closed forms other than Eq. (19). By using the renormalization group procedure to select the most divergent diagrams at a given order $g$ (similar to parquet summation in the Kondo problem), one can sum it up as a geometrical series. \cite{25,29} However, the resulting expression contains a singularity around $|\Sigma(0)|$ [Eq. (10) plays the role of the Kondo temperature here], and is valid at high energies compared to $|\Sigma(0)|$ as Eq. (18). To avoid such problems, we use a different scaling function, suggested by the results of Refs. 16–18. A similar relation between the Kondo problem and disordered graphene has been highlighted in Ref. 28 with a Kondo scale in Eq. (10).

We compare this expression to the numerical solution of the self-consistent noncrossing approximation in Fig. 3. To extract the exponent, we fit the data with $\rho(E) = \rho_0 + 2 \rho_1 (E/\lambda)^\gamma$ and extract $\rho_{0,1}$ and the exponent $\gamma$. As can be seen in the left panel, the power-law fits are excellent in an extended frequency window up to $t/4$. This suggests that this effect should also be observable experimentally as well. The obtained value of $\rho_0$ is negligibly small, as follows from Eq. (11). From the fits, we deduce the exponent and its coefficient, which is shown in the right panel. It agrees well with the result of perturbation theory [Eq. (19)] in the limit of weak disorder. The suppression of the exponent is significant and can be as big as $30\%–35\%$ around $(g_o + g_b)/t^2 \sim 0.4$. A similar phenomenon has been observed for Dirac fermions on a square lattice in the presence of random hopping, \cite{17,18} where disordered systems were studied by exact diagonalization. The decreasing exponent with disorder agrees with our results.

Similar structures in the density of states have been revealed around isolated impurities, \cite{30} as well as in the case of (non-Gaussian) substitutonal potential disorder using the coherent potential approximation. \cite{31,32}

IV. CONDUCTIVITY FOR NONLINEAR DENSITY OF STATES

Now we turn to the discussion of the conductivity in graphene. A possible starting point is the generalized Einstein relation, which is a generally valid relation of nonequilibrium statistical mechanics, first derived by Kubo. \cite{33,34,35} It states that the conductivity

$$\sigma = e^2 \rho D,$$  \hspace{1cm} (20)

where $\rho$ is the density of states and $D$ is the diffusion coefficient, both at the Fermi energy $E_F$. Assuming a general power-law density of states, as found above, we have $\rho(E) = \rho_1 (E/\lambda)^\gamma$. In the weak-disorder limit and away from the Dirac point $E=0$, we can safely neglect any tiny residual value. Moreover, the diffusion coefficient in this case is of the form $D = D_1 E$, which is validated from the Boltzmann approach in the presence of charged impurities. \cite{11} On the other hand, at the Dirac point, there is an exponentially small density of states and a finite nonzero diffusion coefficient $D \propto g/\rho$ in the presence of uncorrelated bond disorder \cite{36} such that the conductivity is of order 1 in units of $e^2/h$.

Recently, the validity of the Boltzmann equation for two-dimensional Dirac fermions was discussed in detail, \cite{28} which took into account Zitterbewegung corrections. Our approach starts from the generally valid expression for $\sigma$ given by the Einstein relation Eq. (20), which avoids certain ambiguities.
hidden in the Kubo or Landauer formulation. So far, we have determined the density of states away from the Dirac point for impure graphene in accordance with exact diagonalization studies, taking into account interband transitions due to impurity scattering as well, which might correspond to Zitterbewegung. Then we have used the diffusion coefficient obtained from the Boltzmann approach without a further justification of $D_1$. The aim of our semiphenomenological approach for the conductivity is only to reach a qualitative understanding, and not a complete derivation from microscopic analysis.

The self-consistent noncrossing approximation is expected to give reliable results in the case of a weak disorder, as was shown in Ref. 25 by a direct comparison with the more sophisticated renormalization group analysis. Moreover, numerically exact studies on the DOS also reported about a similar evolution of the slope of the DOS at low energies and the rounding of the van Hove singularity at $\omega = t$. On the other hand, corrections to the noncrossing approximation may play a role in the minimal conductivity at the Dirac point. This is indicated by the fact that the two-particle Green’s function decays exponentially in the noncrossing approximation, whereas it has a power law according to the saddle-point integration.

In the following, however, we will concentrate on the regime away from the Dirac point. Putting these results together, we find

$$\sigma = e^2 \rho_1 D_1 \frac{E_p^{\gamma+1}}{\gamma}. \tag{21}$$

This can be simplified further by expressing the total number of charge carriers participating in electric transport as

$$n = \int_0^{E_F} \rho(E) dE = \rho_1 \frac{E_p^{\gamma+1}}{(\gamma+1)\gamma}. \tag{22}$$

By inserting this back to Eq. (21), we can read off the conductivity as

$$\sigma = e^2 D_1 (\gamma + 1) n. \tag{23}$$

From this we can draw several conclusions. First, it predicts that away from the Dirac point, where our approach predicts a general power-law density of states, the conductivity varies linearly with the density of charge carriers, in agreement with experiments. Second, the mobility of the carriers, which is the coefficient of the $n$ linear term in the conductivity, behaves as

$$\mu = e \left(2 - \frac{4g}{2\sqrt{3}r^2}\right) D_1, \tag{24}$$

where we used our approximate expression for the exponent in the density of states [Eq. (19)]. This means that with increasing disorder ($g$), the mobility decreases steadily, in agreement with recent experiments on K adsorbed graphene. There, the graphene sample was doped by K, representing a source of charged impurities. Nevertheless, these centers also distort the local electronic environment and act as bond and potential disorder as well. The observed conductivity varied linearly with the carrier concentration $n$, similar to Eq. (23). Moreover, the mobility (the slope of the $n$ linear term) decreased steadily with the doping time (and, hence, the impurity concentration), which, in our picture, corresponds to a reduction of the exponent $\gamma$ as well as the mobility [Eq. (24)].

To study the properties close to the Dirac point, we have to go beyond the perturbative regime. Then we realize that the density of states does not vanish at $E=0$. As an approximation, we add a small contribution near the Dirac point

$$\rho(E) = \rho_0 \delta_\gamma(E) + \rho_1 \left(\frac{E}{\lambda}\right)^\gamma,$$

in the form of a soft Dirac delta function

$$\delta_\gamma(E) = \frac{1}{\pi E^2 + \eta^2} \quad (\eta > 0).$$

This implies a particle density $n$, which does not vanish at the Dirac point, as follows:

$$n(E_F) = \int_0^{E_F} \rho(E) dE = \rho_0 + \frac{\rho_1}{(\gamma + 1)\lambda^\gamma} E_F^{\gamma+1}. \tag{25}$$

Moreover, the diffusion coefficient does not diverge or vanish at the Dirac point $\rho_0 \neq 0$ such that we can assume

$$D(E) = D_0 \delta_\gamma(E) + D_1 E.$$

From the Einstein relation, we get the conductivity which provides an interpolation between a behavior linear in $n$ away from the Dirac point and a minimal conductivity at the Dirac point as follows:

$$\sigma = e^2 \left(\frac{D_0 \rho_0 \delta_\gamma(E_F)}{\rho_1} \right) \quad \text{for } E_F \sim 0$$

$$\sigma \sim \frac{e^2}{R} \left(\frac{D_0 \rho_0 \delta_\gamma(E_F)}{\rho_1 E_F^{\gamma+1}}\right) \sim (1 + \gamma)n \quad \text{for } E_F \gg 0. \tag{26}$$

This, together with Eq. (25), implies for $E_F \gg 0$ the same behavior as in Eq. (23) with the mobility of Eq. (24). The value of the minimal conductivity can be adjusted by choosing the parameter $\eta$ properly. Therefore, Eq. (26) provides us with a qualitative understanding of the conductivity in graphene for arbitrary carrier density.

**V. CONCLUSIONS**

We have studied the effect of weak on-site and bond disorders on the density of states and conductivity of graphene. By using the honeycomb dispersion, we determine the self-energy due to disorder in the self-consistent noncrossing approximation. The density of states at the Dirac point is filled near a similar evolution of the slope of the DOS at low energies and the rounding of the van Hove singularity at $\omega = t$. The mobility of the carriers, which is the coefficient of the $n$ linear term in the conductivity, behaves as

$$\mu = e \left(2 - \frac{4g}{2\sqrt{3}r^2}\right) D_1, \tag{24}$$

where we used our approximate expression for the exponent in the density of states [Eq. (19)]. This means that with increasing disorder ($g$), the mobility decreases steadily, in agreement with recent experiments on K adsorbed graphene. There, the graphene sample was doped by K, representing a source of charged impurities. Nevertheless, these centers also distort the local electronic environment and act as bond and potential disorder as well.
fermions. The mobility decreases steadily with increasing disorder. This can also be relevant for other systems with Dirac fermions.

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