Evaluation of the Green's function of disordered graphene

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Accurate simulations of Green's function and the self-energy function for noninteracting electrons in disordered graphenes are performed. The fundamental physical quantities such as the elastic relaxation time τ_e , the phase velocity v_p , and the group velocity v_g are evaluated. New features around the Dirac point are revealed, indicating that multiscattering-induced hybridization of Bloch states plays an important role in the vicinity of the Dirac point.

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I. INTRODUCTION

Graphene, a single layer of graphite, has been intensively studied in recent years for its many intriguing transport properties.¹⁻³ Examples include minimum conductivity and linear carrier density dependence of conductivity.^{4,5} Electrons in an ideal graphene are governed by the relativistic massless Dirac equation and exhibit a linear-dispersion relation in the vicinity of the Dirac point, and zero density of state at the Dirac point.⁶ Among many relativistic effects, Klein paradox³ is arguably one of the most important effects that makes Dirac electrons different from the Schrödinger electrons in disordered systems.⁷

An in-depth understanding of Dirac electrons in disordered graphenes requires a better knowledge of the self-energy function to extract such fundamental physical quantities as the phase velocity v_p , the group velocity v_g , and the elastic relaxation time τ_e . However, it is known⁸ that the accurate and reliable calculation is quite difficult and non-trivial, although various approximations have been employed in different theoretical studies. So far, almost all calculations^{9,10} on electron properties in disordered graphenes were performed without fully considering the disorder effects. The wave nature of Dirac electrons is more pronounced near the Dirac point because of very large electron wavelength there. It is known that interference of multiscattering leads to the weak localization and the Anderson localization in conventional disordered electron systems.¹¹ Quantum interference also plays important roles in coherent wave propagation through quasirandom¹² and random media.^{13,14} As it will be shown below, hybridization of Bloch states near the Dirac point in realistic disordered graphenes is essential in understanding the diffusion properties although previous calculations^{9,10} can capture the essential physics far away from the Dirac point.

In this Brief Report, we present a systematic method for the precise computation of Green's function and the self-energy of large size disordered graphenes. We extract accurate τ_e , v_p , and v_g values from the spectral function $A(\mathbf{k}, E)$ derived from the self-energy function. Compared the results

from the self-consistent Born approximation (SCBA), it is found that the τ_e is overestimated by the SCBA in the strong disorder regime. We show that both v_p and v_g deviate significantly from their unrenormalized values and exhibit substantial energy dependence, especially near the Dirac point. The effective group velocity is larger than the effective phase velocity but substantially lower than its unrenormalized value when the mixing of the different Bloch states is dominant. Moreover, we generalize the Einstein relation to calculate the conductivity of the disordered graphene.

II. MODEL AND METHOD

π electrons of undoped graphene can be modeled by a tight-binding Hamiltonian on a honeycomb lattice of two sites per unit cell, $H_0 = t \sum_{\langle ij \rangle} |i\rangle \langle j| + \text{H.c.}$, where $t = -2.7$ eV is the hopping energy. The corresponding eigenvalues and eigenstates of H_0 near the Dirac point are $E_{k,\pm} = \pm \hbar v_F^0 k$ and $|\mathbf{k}\pm\rangle = (|\mathbf{k}A\rangle \pm e^{i\phi(\mathbf{k})} |\mathbf{k}B\rangle)/2$, respectively.^{4,15} Here, v_F^0 is the unrenormalized Fermi velocity and $\hbar$ is the Planck's constant. A and B stand for A and B sublattices. $\phi(\mathbf{k})$ is the polar angle of the momentum $\mathbf{k}$ and $|\mathbf{k}A(B)\rangle = \frac{1}{\sqrt{N_{A(B)}}} \sum_{\mathbf{r}_{A(B)}} e^{i\mathbf{k}\cdot\mathbf{r}_{A(B)}} |\mathbf{r}_{A(B)}\rangle$, where $\mathbf{r}_{A(B)}$ is the position vector of $A(B)$ lattice and $N_{A(B)}$ is the total $A(B)$ -lattice points. The plus (minus) sign denotes the conduction (valence) band. The Green's function of the clean graphene is, in a diagonal basis, $G_0(\mathbf{k}, E) = \frac{1}{E + i0^+ - \hbar v_F^0 k} |\mathbf{k}+\rangle \langle \mathbf{k}+| + \frac{1}{E + i0^+ + \hbar v_F^0 k} |\mathbf{k}-\rangle \langle \mathbf{k}-|$. A weak pointlike disorder is introduced through $V = \sum_i \epsilon_i |i\rangle \langle i|$ with n_{imp} randomly distributed impurity sites where the on-site energy ϵ_i of each impurity can take $-V_0$ or V_0 (measured in the unit of t) with equal probability. In this Brief Report, V_0 is chosen $V_0 < 3$ to guarantee no resonant states around the Dirac point.^{16,17} A dimensionless parameter $\alpha = \frac{n_{\text{imp}} V_0^2 A_c}{2\pi(\hbar v_F^0)^2}$ can be used to characterize disorder strength. Here, A_c is the area of the unit cell. Our calculation shows that physical quantities such as self-energy are only determined by the parameter α .¹⁸

The ensemble-averaged Green's function is defined as $G(\mathbf{k} \pm, E) = \langle \mathbf{k} \pm | \frac{1}{E + i\eta - H_0 - V} | \mathbf{k} \pm \rangle$, where the bar means the en-

FIG. 1. (Color online) (a) Real part of self-energy as a function of energy. (b) Imaginary part of self-energy as a function of energy. The squares and circles are our numerical calculations for the disorder concentration $n_{\text{imp}}/N=10\%$ and $V_0=0.5$ and $V_0=2.0$, respectively. Dashed lines are for the SCBA results for the same disorder. Energy is measured in the units of t .

semble average. It can be calculated by using the well-developed Lanczos recursive method.^{16,19,20} In order to obtain an accurate ensemble-averaged Green's function near the Dirac point with high-energy resolution,²¹ a large sample containing $N=L_x \times L_y \approx 6.0$ millions carbon atoms (2400×2400) and over 1000 ensembles are used in our simulation, where L_x and L_y is the number of atoms in x and y directions, respectively. The large lattice samples guarantee that the calculated Green's function is free of finite-size errors.²² The periodic boundary condition has been used and the wave vectors satisfy $k_x = n_x 4\pi/3aL_x$ and $k_y = n_y 4\pi/\sqrt{3}aL_y$, where $n_{x(y)}$ is an integer and a is the lattice constant.

III. SELF-ENERGY FUNCTION

Figure 1 shows the calculated (a) real and (b) imaginary parts of the self-energy function for $n_{\text{imp}}/N=10\%$ and $V_0=0.5$ (squares) and 2.0 (circles), respectively. The self-energy function is defined based on the Dyson's equation $G(\mathbf{k}, E) = G_0(\mathbf{k}, E) + G_0(\mathbf{k}, E)\Sigma(\mathbf{k}, E)G(\mathbf{k}, E)$, thus $\Sigma(\mathbf{k}, E) = G_0^{-1}(\mathbf{k}, E) - G^{-1}(\mathbf{k}, E)$.⁸ In principle, the self-energy function depends on energy E and wave vector $\mathbf{k}$. However, our simulation shows that the self-energy function is not sensitive to wave vector $\mathbf{k}$ while the one-particle Green's function depends on both E and $\mathbf{k}$. This is not surprising since the scatterer size is much smaller than the electron wavelength near the Dirac point so that the inhomogeneous structure of disordered graphene can be well described by an effective homogeneous medium. This is also why the self-energy function is assumed to be $\mathbf{k}$ independent in many perturbative calculations. Our finding validates these assumptions.²³

Our calculation should be compared with widely used results from the SCBA that predicts^{15,24}

$$\Sigma(E) = \begin{cases} -E/2\alpha - i\Gamma_0, & |E| \ll \Gamma_0, \\ -2\alpha(E + i\pi\alpha|E|)\ln\left|\frac{E_c}{E}\right| - i\pi\alpha|E|, & |E| \gg \Gamma_0, \end{cases}$$

where $\Gamma_0 = E_c e^{-1/2\alpha}$ (E_c is the cut-off energy). As shown in Fig. 1, SCBA results agree well with our exact self-energy

FIG. 2. (Color online) Single-particle spectral function $A(\vec{k} +, E)$ plotted as a function of energy E at several k points (from left to right) $k=0.000, 0.014, 0.028, 0.042, 0.056, 0.070, 0.084$, and 0.098 (or $n_x=0-56$) along the k_x direction. Model parameters are $n_{\text{imp}}/N=10\%$ and $V_0=1.0$. Inset: the energy dependence of single-particle relaxation time τ_e for $V_0=0.5-2.0$ and $n_{\text{imp}}/N=10\%$.

function for very weak disorder ($V_0=0.5$ and $n_{\text{imp}}/N=10\%$).²⁵ When the disorder strength increases several times ($V_0=2.0$ or $\alpha \sim 0.07$), the perturbative results cannot capture the main features, especially near the Dirac point. The discrepancy is obvious for $\text{Im}\Sigma(0)$ as shown in Fig. 1(b). $\text{Im}\Sigma(E)$ at the Dirac point is $\Gamma_0 \sim 10^{-3}t$ [$E_c = \sqrt{3}t$ (Ref. 6)] from the SCBA while our exact value is $\text{Im}\Sigma(0) \geq 10^{-2}t$. Therefore, the true broadening of states at or near the Dirac point is much bigger than what is predicted by SCBA. This difference can be attributed to the mixture of Bloch states caused by the impurities.^{5,26} Furthermore, the level-repulsion effect pushes all energy level toward the Dirac point so that the density of states at the Dirac point increases more in the presence of impurities (see inset of Fig. 4 below). Therefore, the impurities increase the imaginary part of the self-energy function (directly associated with the density of states) at the Dirac point. When the wavelength becomes short, and the quantum interference as well as the Bloch state mixing are less important, $\text{Im}\Sigma(E)$ are determined by the disorder scattering. The difference between our exact simulation and that of the SCBA is small, as shown in Fig. 1(b).

IV. SPECTRAL FUNCTION

The single-particle spectral function relates to the Green's function through $A(\mathbf{k} \pm, E) = -\text{Im} G(\mathbf{k} \pm, E)/\pi$.²³ Figure 2(a) is $A(\mathbf{k} +, E)$ for $k_y=0$, $V_0=1$, $n_{\text{imp}}/N=10\%$, and various k_x (in unit of a^{-1}) (curves from the left to the right in the figure) ranging from 0.0 ($n_x=0$) to 0.098 ($n_x=56$). In the absence of disorders, the spectral function $A_0(\mathbf{k} \pm, E)$ is a delta function, reflecting that the wave vector $\mathbf{k}$ is a good quantum number and has all its spectral weight precisely at the energy $E=E_{k\pm}$. In the presence of disorders, the translational symmetry is broken and the spectral function is broadened, resulting from the disorder scattering effect. The widths of the spectral function is given by $\text{Im}\Sigma(E)$ that measures the elastic relaxation lifetime τ_e , $\tau_e = \hbar/(-2\text{Im}\Sigma(E))$. Therefore, the elastic scattering relaxation time is then akin to

FIG. 3. (Color online) (a) Effective energy dispersion relation for ideal graphene and disordered graphenes with $n_{\text{imp}}/N=10\%$ and $V=0.5$ (filled squares) and $V=2.0$ (filled circles). (b) The ratio v_g/v_F^0 (filled symbols) and v_p/v_F^0 (open symbols) as a function of wave number k (in units of $1/a$).

$\text{Im } \Sigma(E)$, and τ_e around the Dirac point is mainly determined by the Bloch state mixing and level repulsion effect.²⁷ Far away from the Dirac point, the lifetime is mainly attributed to the disorder scattering. As shown in the inset of Fig. 2, lifetime become shorter as the wave vector increases. Physically, this is because the density of states ρ increases linearly with energy and the disorder scattering effects become larger, in qualitative agreement with the prediction of SCBA.^{15,24,28}

V. EFFECTIVE BAND VELOCITY

Dirac electron propagation velocities including the group velocity v_g and phase velocity v_p are also greatly modified by disorder effects. These quantities relate to the shape of the dispersion relation that is the roots of $E - E_0(k) - \text{Re } \Sigma(E) = 0$.^{29,30} One can also extract the dispersion relation from the peak of the spectral function $A(\mathbf{k}, E)$ for a given $\mathbf{k}$. Figure 3(a) is our exact dissipation curve $E_{\text{eff}}(k)$ that is linear for very weak disorder ($\alpha < 0.01$). However, when the disorder strength becomes large enough ($\alpha \sim 0.1$), the $E_{\text{eff}}(k)$ is concave near the Dirac point, indicating the reduction in both the group and phase velocities.

Figure 3(b) is the k dependence of the group and phase velocities that can be obtained from $v_g = \partial E_{\text{eff}}(k) / \partial k$ and $v_p = E_{\text{eff}}/k$. For very weak disorder $\alpha=0.01$, v_g and v_p are not too much different from the unrenormalized velocity v_F^0 with only 5% reduction at the Dirac point as shown in the figure. When the disorder strength increase several times $\alpha=0.07$, the renormalized v_g is higher than v_p , and both v_g and v_p are reduced by a large percentage near the Dirac point. It shows that disorder not only renormalizes the Dirac electron velocities but also changes the linear-dispersion relation.²⁶ The observation that significant reduction in velocities occur at the Dirac point reflects that the Dirac electrons near the Dirac point are more sensitive to the disorder.

One can use the renormalization factor Z defined as $Z = [1 - \frac{\partial \text{Re } \Sigma(E, \mathbf{k})}{\partial E}]^{-1}$ to measure the disorder effect on electronic structure.³¹ Its value equals the ratio v_g/v_F^0 . As shown in Fig. 3(b), Z is very close to 1.0 for very weak disorder. In this regime, the Bloch state is still a good starting point for understanding disordered graphene and the transport properties are expected to be described by the quasiclassical Boltz-

FIG. 4. (Color online) Conductivity as a function of charge density. The model parameters are $n_{\text{imp}}/N=10\%$ and $V_0=0.5, 1.0$, and 2.0 . Inset: density of states for the same parameters.

mann theory. When the disorder strength increases several times, Z is much smaller than 1.0 around the Dirac point. This unusual feature directly reflects that a Bloch state around the Dirac point strongly couples with other nearby Bloch states. These hybridized waves can also be expanded in a complete set of Bloch states $[\phi_{\mathbf{k}}(r) = e^{i\mathbf{k} \cdot \mathbf{r}} u(r)]$: $\Psi(r) = \sum_{\mathbf{k}} c_{\mathbf{k}} \phi_{\mathbf{k}}(r)$, where $c_{\mathbf{k}} = \langle \phi_{\mathbf{k}} | \Psi \rangle$. If Z approaches 1.0, the system is dominated by a single “majority representation” wave vector $\mathbf{k}_0$ while $\Psi(r)$ essentially behaves as a Bloch periodic state $\phi_{\mathbf{k}_0}(r)$. However, $\mathbf{k}_0$ is not a good approximate quantum number anymore when Z is much smaller than 1.0. Therefore, Bloch states would not be a good starting point to perform perturbative calculations. In fact, similar features had been observed in semiconductor alloys and been termed non-Bloch nature of alloy states.³² This is also why the SCBA method cannot produce accurate enough results.²⁶ Furthermore, this nontrivial feature could lead to intriguing transport and optical properties. For example, linear-dispersion relation leads to $E_n = \pm v_F^0 \sqrt{2e\hbar n B} (n=0, 1, 2, \dots)$ Landau-level (LL) series. Since the LLs come from the quenching of electrons’ kinetic energies, lower group velocity near the Dirac point means the energy gap between $n=0$ LL and $n = \pm 1$ LL should be substantially reduced by the disorders while that between $n = \pm 1$ LLs and $n = \pm 2$ LLs will be affected less by disorder. This is indeed what was observed in a recent experiment of cyclotron resonance.³³

VI. CONDUCTIVITY

Assuming that the Einstein relation for the Dirac electrons is valid, one can obtain the conductivity from $\sigma = e^2 D \rho$, where e is the electron charge and $D = v_g^2 \tau_{tr} / 2$ is the diffusion coefficient, and τ_{tr} denotes the transport relaxation time and satisfies the relationship $\tau_{tr} = 2\tau_e$ for the pointlike disorder.⁵ Figure 4 is the conductivity calculated based on the transport relaxation time $\tau_{tr} = 2\tau_e$ in Fig. 2. The group velocity v_g is shown in Fig. 3. The density of states is obtained by $\rho(E) = -\frac{A_c}{\pi} \int_0^{k_c} \frac{d\tilde{k}}{(2\pi)^2} A(\tilde{k}, E)$ as shown in the inset of Fig. 4. For very weak disorder, the conductivity is sublinear in the Fermi energy. While increasing the disorder strength, the conductivity depends weakly on the Fermi energy or the carrier density,

which agrees with the prediction of Boltzmann theory for short-range scatterers.¹⁵ At the Dirac point, when combining the self-energy function with the Kubo formula, the conductivity is predicted to be the universal value $\sigma_{xx}(0) = 4e^2/\pi h$.¹⁵ However, our calculations find that $\sigma_{xx}(0)$ takes the values $8.2e^2/h$, $5.9e^2/h$, and $2.7e^2/h$ for $\alpha = 0.0046$, 0.018 , and 0.07 , respectively and shows nonuniversal behavior.

VII. SUMMARY

In conclusion, we studied pointlike disorder effects on the one-electron properties of graphene. The exact ensemble-averaged Green's function is obtained from a large-scale real-space calculation. Through the analysis of self-energy and spectral functions, we conclude that the single-particle lifetime reduction and the linear-dispersion relation are

modified by the hybridization of the Bloch states. The strong hybridization of Bloch states near the Dirac point indicates the Bloch-Boltzmann theory need to be modified in disordered graphene. Our approach is very general and robust, and is applicable to other disordered systems.

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