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Local spectrum rearrangement in impure graphene

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The evolution of the averaged local density of states at the impurity site with increasing the impurity concentration is studied by the cluster expansion method for the self-energy. It is demonstrated that the general shape of the local density of states and the decrease rate of the resonance peak height undergo a qualitative change at a certain critical impurity concentration. As a result, the distinctive features in the single-impurity local spectrum are completely smeared out when this critical concentration is exceeded. Within the Lifshitz impurity model, this local spectrum rearrangement is described in detail for the two-dimensional system with the Dirac dispersion of electrons, which is characteristic of graphene. The correspondent critical impurity concentration is related to the spatial overlap of individual impurity states.

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I. INTRODUCTION

The phenomenon of spectrum rearrangement has been extensively studied theoretically and verified experimentally for a large variety of systems over the past few decades.[1–3] The essence of this phenomenon lies in a radical change in the spectral properties of a disordered system, which takes place when the impurity concentration exceeds a certain critical value. This characteristic concentration of the spectrum rearrangement corresponds to a spatial overlap of individual impurity states. If the impurity states are of the long-range type, the spectrum rearrangement occurs at the low impurity concentration. Usually, the effective radius of a single-impurity state is considerably larger than the lattice constant when the impurity state energy is close to any of the van Hove singularities in the host spectrum, e.g., to the band edge.

Until now, the term "spectrum rearrangement" has been applied to address a qualitative modification of the total density of states, or the behavior of the renormalized dispersion relation in the disordered system, which characterizes the disordered system as a single whole. However, with recent advances in the scanning tunneling spectroscopy (STS), it became possible to observe directly the local density of states (LDOS).[4–6] The LDOS is not self-averaging, and *minutiae* of its shape depend on the specific distribution of impurities around the selected site. Thus, in actual experiments, the LDOS is frequently averaged over several sites.[7] It is known that this averaging significantly reduces irregularities in the shape of the LDOS.[8–10] Such averaged LDOS is, indeed, self-averaging and does not depend on the particular impurity distribution. Since the change in the spectrum is mostly pronounced close to the impurity state energy, it seems reasonable to expect that the spectrum rearrangement might be reflected in the impurity LDOS.

Below, we argue that qualitatively different regimes in the concentrational dynamics of the averaged LDOS at the impurity site do exist, and the corresponding critical concentration is tightly connected with the overlap of the impurity states. We will try to demonstrate that, on the qualitative level, the shape of the LDOS is determined by the ratio of the actual impurity concentration to the critical one, and thus, it is fully justifiable to speak about the *LDOS rearrangement* as such.

Because graphene[11] is the first purely two-dimensional (2D) object, STS measurements of the electronic LDOS in this material are decidedly natural. Moreover, the required conditions for the resonance state formation and the respective features in the single-impurity LDOS at the impurity site[12,13] and in the near field of the impurity[14,15] have been discussed recently, as well as possibilities to observe them experimentally,[15] and reports on similar STS measurements in graphene have appeared in print already.[7] Therefore, we think that impure graphene is the most suitable object for testing whether the introduced concept of the LDOS rearrangement is productive.

II. MODEL

A substitutional 2D binary alloy with a diagonal disorder in the tight-binding approximation is adopted as the model of the disordered system. This model, which is attributed to Lifshitz,[16] features absolute randomness in the space distribution of impurities. According to the asymmetric definition of the impurity perturbation, the on-site potentials are V_L with the probability c, or 0 otherwise. The corresponding Hamiltonian reads

$$\boldsymbol{H}=\boldsymbol{H}_0+\boldsymbol{H}_{imp}, \quad \boldsymbol{H}_{imp}=V_L\sum_{\boldsymbol{n},\alpha} c^{\dagger}_{\boldsymbol{n}\alpha}c_{\boldsymbol{n}\alpha}, \tag{1}$$

where $\boldsymbol{n}$ refers to lattice cells, α enumerates sublattices, $c^{\dagger}_{\boldsymbol{n}\alpha}$ and $c_{\boldsymbol{n}\alpha}$ are electron creation and annihilation operators, and the summation is restricted to those sites that are occupied by impurities. Because the single-impurity perturbation is local, the host Hamiltonian $\boldsymbol{H}_0$ containing only one Dirac cone can adequately model the linear spectrum of graphene,

$$\boldsymbol{H}_0=\sum_{\boldsymbol{k}}\left[f(\boldsymbol{k})c^{\dagger}_1(\boldsymbol{k})c_2(\boldsymbol{k})+f^{*}(\boldsymbol{k})c^{\dagger}_2(\boldsymbol{k})c_1(\boldsymbol{k})\right],$$

$$c_\alpha(\boldsymbol{k}) = \frac{1}{\sqrt{N}}\sum_{\boldsymbol{n}} e^{i\boldsymbol{k}\boldsymbol{n}} c_{\boldsymbol{n}\alpha}, \quad f(\boldsymbol{k}) = \frac{a}{2\sqrt{\pi}}(k_x + ik_y), \tag{2}$$

where the magnitude of the hopping parameter is chosen so that the bandwidth is unity, when the Brillouin zone is approximated with a circle. Then, the diagonal element of the host Green's function (GF) $\boldsymbol{g}=(\epsilon-\boldsymbol{H}_0)^{-1}$ in the vicinity of the Dirac point is given by[13,17]

$$g_{\boldsymbol{n}\alpha\boldsymbol{n}\alpha}(\epsilon) \equiv g_0(\epsilon) \approx 2\epsilon \ln|\epsilon| - i\pi|\epsilon|, \quad |\epsilon| \ll 1. \tag{3}$$

III. SINGLE-IMPURITY APPROXIMATION

In a case when only a single impurity is present in the lattice, the diagonal element of the GF $\boldsymbol{\mathcal{G}}=(\epsilon-\boldsymbol{H})^{-1}$ at the impurity site becomes

$$\mathcal{G}_0(\epsilon) = \frac{g_0(\epsilon)}{1 - V_L g_0(\epsilon)}. \tag{4}$$

Like in unconventional superconductors,[4] the respective LDOS $\rho_{imp}(\epsilon)=-\pi^{-1}\,\mathrm{Im}\,\mathcal{G}_0(\epsilon)$ manifests a resonance peak at a sufficiently large V_L,[13] the energy ϵ_r of which is specified by the Lifshitz equation,

$$1 = V_L\,\mathrm{Re}\,g_0(\epsilon_r) \approx 2V_L\epsilon_r \ln|\epsilon_r|. \tag{5}$$

The resonance energy is located above the Dirac point in the spectrum at $V_L<0$, and vice versa. This property of the resonance holds valid for any two symmetric bands that are touching each other. Grouping together Eqs. (3) and (4) renders

$$\rho_{imp}(\epsilon) = \frac{|\epsilon|}{(1 - 2V_L\epsilon\ln|\epsilon|)^2 + (\pi V_L\epsilon)^2}. \tag{6}$$

The denominator expansion about the resonance yields

$$\rho_{imp}(\epsilon) \approx \frac{|\epsilon|\Gamma_r^2}{[\pi V_L\epsilon_r]^2[(\epsilon-\epsilon_r)^2 + \Gamma_r^2]},$$

$$\Gamma_r = \frac{\pi|\epsilon_r|}{2|1 + \ln|\epsilon_r||}. \tag{7}$$

Thus, the LDOS has the Lorentz shape when the resonance state is well defined,

$$\gamma_r \equiv \frac{\Gamma_r}{|\epsilon_r|} \approx \frac{\pi}{2|1 + \ln|\epsilon_r||} \ll 1. \tag{8}$$

IV. FINITE-IMPURITY CONCENTRATION

It is convenient to introduce the conditional GF with the impurity-occupied first site,

$$\boldsymbol{\mathcal{G}}^{(imp,host)} = V_L^{-1}\boldsymbol{H}_{imp}\boldsymbol{\mathcal{G}}. \tag{9}$$

At a finite concentration, it can be related to the self-energy after averaging over impurity distributions,[18,19]

$$\boldsymbol{G}^{(imp,host)} = V_L^{-1}\boldsymbol{\Sigma}\boldsymbol{G}, \quad \boldsymbol{G} = \boldsymbol{g} + \boldsymbol{g}\boldsymbol{\Sigma}\boldsymbol{G}, \tag{10}$$

where $\boldsymbol{G}=\langle\boldsymbol{\mathcal{G}}\rangle$ is the averaged GF. While the concentration c is sufficiently small to neglect multiple occupancy corrections, the self-energy can be taken in the modified propagator approximation,[20]

$$\boldsymbol{\Sigma} \approx \sigma\boldsymbol{I}, \quad \sigma = \frac{cV_L}{1 - V_L g_0(\epsilon-\sigma)}. \tag{11}$$

Thus, the self-energy, being identical on both sublattices due to the symmetry, is also site diagonal. In order to obtain a quantity that can be compared to the above $\mathcal{G}_0(\epsilon)$, the diagonal in lattice index element of the conditional GF should be properly scaled,

$$c^{-1}G_0^{(imp,host)} \approx \frac{g_0(\epsilon-\sigma)}{1 - V_L g_0(\epsilon-\sigma)}. \tag{12}$$

This expression resembles Eq. (4), in which the host GF $\boldsymbol{g}$ is replaced by the GF of the disordered system, $\boldsymbol{G}(\epsilon)\approx\boldsymbol{g}(\epsilon-\sigma)$. Finally, the averaged LDOS at the impurity site can be written as follows:

$$\rho_{loc}(\epsilon) \approx -\frac{1}{\pi}\,\mathrm{Im}\left[\frac{g_0(\epsilon-\sigma)}{1 - V_L g_0(\epsilon-\sigma)} + \frac{1}{V_L}\right]$$

$$= -\frac{1}{\pi V_L}\,\mathrm{Im}\,\frac{1}{1 - V_L g_0(\epsilon-\sigma)} = -\frac{1}{\pi}\frac{\mathrm{Im}\,\sigma}{cV_L^2}. \tag{13}$$

With the help of the standard substitution,

$$\epsilon - \sigma = \varkappa\exp(i\varphi), \quad \varkappa > 0, \quad 0 < \varphi < \pi, \tag{14}$$

the imaginary part of the self-consistency condition in Eq. (11) becomes

$$\begin{aligned} &cV_L^2[2\ln\varkappa + (2\varphi-\pi)\cot\varphi] \\ &\quad + \{1 - V_L\varkappa[2\ln\varkappa\cos\varphi - (2\varphi-\pi)\sin\varphi]\}^2 \\ &\quad + \{V_L\varkappa[2\ln\varkappa\sin\varphi + (2\varphi-\pi)\cos\varphi]\}^2 = 0, \end{aligned} \tag{15}$$

where the particular form of the host GF [see Eq. (3)] has been taken into account. At the given V_L and c, this equation always has two solutions for the phase φ when $\varkappa$ exceeds some threshold value. The corresponding energies can be obtained from the real part of Eq. (11),

$$\begin{aligned} \epsilon = {}&\varkappa\cos\varphi + cV_L\{1 - V_L\varkappa[2\ln\varkappa\cos\varphi - (2\varphi-\pi)\sin\varphi]\}/ \\ &(\{1 - V_L\varkappa[2\ln\varkappa\cos\varphi - (2\varphi-\pi)\sin\varphi]\}^2 \\ &+ \{V_L\varkappa[2\ln\varkappa\sin\varphi + (2\varphi-\pi)\cos\varphi]\}^2). \end{aligned} \tag{16}$$

Expressed in the same variables, LDOS reads

$$\rho_{loc}(\epsilon) = \frac{\varkappa\sin\varphi}{\pi cV_L^2}. \tag{17}$$

Several examples of the LDOS calculated by Eqs. (15)–(17) are depicted in Figs. 1 and 2. Insets show the bottom left-hand corner of the plot enlarged.

V. REARRANGEMENT OF THE RESONANCE

The resonance energy that corresponds to the given impurity concentration should be a solution of the equation [cf. Eq. (5)]

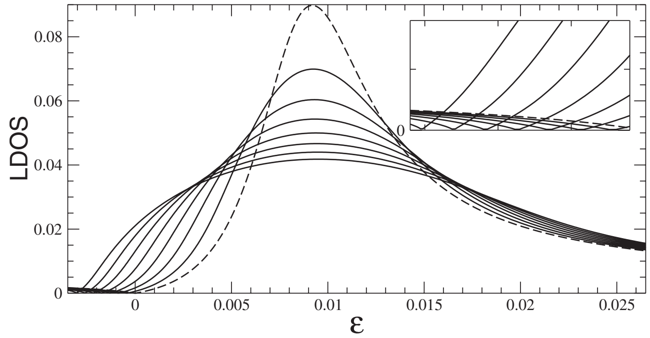

FIG. 1. The LDOS at the impurity site for $\epsilon_r = 0.01$ at $c = 4n \times 10^{-5}$, $n = 1, 2, \ldots, 7$. The single-impurity LDOS is displayed for comparison by the dashed line. The peak height decreases with increasing concentration.

$$1 = V_L \operatorname{Re} G_0(\epsilon_r(c)). \tag{18}$$

By using Eq. (3) and the substitution (14), this equation can be recast as follows:

$$1 = 2V_L\epsilon_r(c)\left\{\ln\left|\frac{\epsilon_r(c)}{\cos\varphi_r(c)}\right| + \left[\frac{\pi}{2} - \varphi_r(c)\right]\tan\varphi_r(c)\right\}. \tag{19}$$

Hereafter, we will assume without any loss of generality that $\epsilon_r > 0$. When the concentration is sufficiently low, an approximate solution of Eq. (19) is

$$\epsilon_r(c) \approx \epsilon_r[1 + \gamma_r\varphi_r(c)], \quad \varphi_r(c) \ll 1. \tag{20}$$

Because the resonance width parameter γ_r [see Eq. (8)] should be small, the resonance energy varies slowly with $\varphi_r(c)$. The concentration dependence of $\varphi_r(c)$ can be obtained from the self-consistency condition (15). The second term in Eq. (15) is zero by the definition of $\epsilon_r(c)$, and the remaining two lead to the relation

$$c = -2\epsilon_r^2(c)\tan\varphi_r(c)\left[\ln\left|\frac{\epsilon_r(c)}{\cos\varphi_r(c)}\right|\tan\varphi_r(c) + \varphi_r(c) - \frac{\pi}{2}\right]. \tag{21}$$

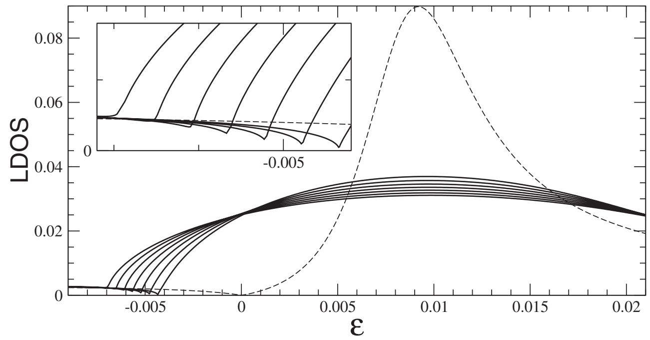

FIG. 2. The LDOS at the impurity site for $\epsilon_r = 0.01$ at $c = (36 + 4n) \times 10^{-5}$, $n = 1, 2, \ldots, 7$. The single-impurity LDOS is displayed for comparison by the dashed line. The dip moves leftward with increasing concentration.

At low concentrations, Eq. (21) considerably simplifies to

$$c \approx \pi\epsilon_r^2(c)\varphi_r(c)\left[1 + \frac{\varphi_r(c)}{\gamma_r}\right], \quad \varphi_r(c) \ll 1. \tag{22}$$

Thus γ_r is, in fact, acting as the characteristic phase of the resonance state, which reveals its hidden physical meaning. The corresponding concentration is given by Eq. (22) with $\varphi_r(c) = \gamma_r$,

$$c_r = -\frac{\pi^2\epsilon_r^2(c_r)}{1 + \ln\epsilon_r} \approx -\frac{\pi^2\epsilon_r^2}{1 + \ln\epsilon_r} \approx -\frac{\pi^2\epsilon_r^2}{\ln\epsilon_r}. \tag{23}$$

At the critical concentration c_r, the resonance damping induced by the disorder becomes equal to the damping of the single-impurity state, $-\operatorname{Im}\sigma \approx \epsilon_r(c_r)\varphi_r(c_r) \approx \Gamma_r$, and, respectively, the LDOS magnitude at the resonance decreases to one-half of the single-impurity case,

$$\rho_{loc}(\epsilon_r(c_r)) \approx \frac{\epsilon_r(c_r)\varphi_r(c_r)}{\pi c_r V_L^2} = \frac{1}{2\pi^2 V_L^2\epsilon_r} = \frac{\rho_{imp}(\epsilon_r)}{2}. \tag{24}$$

The qualitative change in the spectral properties of the system, which occurs at $c \sim c_r$, have been overlooked in Ref. 21. As a result, the estimations provided there for the impurity bandwidth and its shift from the Dirac point are proved to be inadequate at the low impurity concentrations.

Neglecting the concentration offset in the resonance position (20), Eq. (22) can be easily solved for the phase,

$$\varphi_r(c) \approx \frac{\gamma_r}{2}\left(\sqrt{1 + 8\frac{c}{c_r}} - 1\right). \tag{25}$$

Then, the concentration dependence of the resonance peak height immediately follows from Eqs. (17) and (25):

$$\rho_{loc}(\epsilon_r(c)) \approx \frac{\sqrt{1 + 8\frac{c}{c_r}} - 1}{4\frac{c}{c_r}}\rho_{imp}(\epsilon_r). \tag{26}$$

Initially, it decreases linearly with the concentration,

$$\rho_{loc}(\epsilon_r(c)) \approx \left[1 - 2\frac{c}{c_r}\right]\rho_{imp}(\epsilon_r), \quad c \ll c_r, \tag{27}$$

but then the decrease rate considerably slows down,

$$\rho_{loc}(\epsilon_r(c)) \approx \sqrt{\frac{c_r}{2c}}\rho_{imp}(\epsilon_r), \quad c \gg c_r. \tag{28}$$

The qualitative difference between these two regimes is clearly evident from the comparison of Fig. 1 with Fig. 2. Since $\gamma_r = \varphi_r(c_r) \ll 1$, the above treatment is fully justified for all concentrations up to c_r and somewhat higher, according to the known applicability criterion.[22] We suppose that this change of the decrease rate, not to mention the halving of the resonance peak height, could be captured by the actual STS measurements.

VI. REARRANGEMENT OF THE ANTIRESONANCE

Besides the resonance, there is another noticeable feature in the LDOS—the dip, or *antiresonance*—near the Dirac point. At the low impurity concentrations, its position virtually corresponds to that energy in the spectrum at which $\varphi = \pi/2$. Strictly speaking, our approach is not valid at this spectral point.[22] Nevertheless, following estimations will duly serve for a rough guide, especially taking into account that we are mostly looking for qualitative answers. At $\varphi = \pi/2$, the imaginary part of Eq. (11) takes on the form

$$-\mathrm{Im}\,\sigma = \varkappa_{dip} = -\frac{2cV_L^2\varkappa_{dip}\ln\varkappa_{dip}}{1+(2V_L\varkappa_{dip}\ln\varkappa_{dip})^2}, \tag{29}$$

and the effective shift due to impurities is given by

$$\epsilon_{dip}(c) \equiv \mathrm{Re}\,\sigma = \frac{cV_L}{1+(2V_L\varkappa_{dip}\ln\varkappa_{dip})^2}. \tag{30}$$

The characteristic concentration for this spectral point should correspond to $|\mathrm{Re}\,\sigma| = |\mathrm{Im}\,\sigma|$. This yields

$$c_{dip} = -4\epsilon_r^2\ln|\epsilon_r|. \tag{31}$$

For $c \ll c_{dip}$, the magnitude of the LDOS at this specific energy rapidly increases with impurity concentration,

$$\rho_{loc}(\epsilon_{dip}(c)) \approx \frac{2}{\pi}\frac{\exp\left(-\dfrac{1}{2cV_L^2}\right)}{2cV_L^2}, \tag{32}$$

and then reaches its maximum value at $c = c_{dip}$,

$$\rho_{loc}(\epsilon_{dip}(c_{dip})) = \frac{1}{2\pi|V_L|}. \tag{33}$$

According to Eq. (30), the dip position gradually shifts by approximately cV_L with increasing concentration for $c \ll c_{dip}$, and, finally, this dip totally disappears at $c \sim c_{dip}$. The separation of the LDOS curve from the energy axis can be distinctly seen in Fig. 2. As mentioned above, the modified propagator method is not applicable at $\varphi = \pi/2$. However, since the spectral domain, where this approach is expected to fail, is exponentially narrow for $c \lesssim c_{dip}$,[13] and the dip is not present in the LDOS for $c \gtrsim c_{dip}$, the above estimations seem to be fully justified.

VII. DISCUSSION

The characteristic scale of the spatial variations of the GF in a system with the linear dispersion is proportional to $1/|\epsilon|$. Since in a 2D system the average distance between impurities is proportional to $1/\sqrt{c}$, the critical concentration of the spectrum rearrangement should be around ϵ_r^2. The narrow interval between obtained above critical concentrations (23) and (31) covers this rough estimation. Thus, there is an intimate correlation between the LDOS rearrangement and the overlap of the impurity states. Within the adopted impurity model, a well-defined resonance state is possible only at a strong impurity perturbation. However, this restriction on the impurity potential is more relaxed for the double impurity.[15,23,24] In large, the main physics of the LDOS rearrangement for the double impurity should remain intact. The condition on the impurity potential magnitude is also eased when the impurity-host hopping is allowed to vary. In addition, the possibility to adjust the gap width in the bilayer graphene by a gate bias[25] offers the challenge of LDOS measurements with a control over the critical concentration c_r achieved by an applied electric field. In some cases, the impurity concentration can be monitored by measuring the carrier density.[26] Thus, we expect that the task of pinpointing the magnitude of the critical concentration of the spectrum rearrangement, which is a valuable parameter of the system, is accessible by actual experiments.

VIII. CONCLUSION

In summary, we have demonstrated that the concept of the LDOS rearrangement has sound grounds. The spatial overlap of individual impurity states is the true physical reason of this phenomenon. This overlap, which occurs at the critical concentration of impurities, leads to a qualitative change in the shape of the LDOS. Thus, the resonance peak and the antiresonance dip remain discernible in the LDOS at the impurity site only when the respective critical concentration of impurities is not exceeded. This fact is worth considering for the resonance state observation in STS experiments, because the sharper is the resonance, the lower is the critical concentration of the LDOS rearrangement, which imposes the restriction on the amount of doping. We would like to emphasize that not only the way the single-impurity features are gradually erased from the LDOS was described above, but it was also shown that there is a possibility to foresee the general shape of the local spectrum provided the impurity concentration is known. Furthermore, it can be speculated that the effect of the LDOS rearrangement is not specific to graphene or related systems with the Dirac dispersion, and should occur in practically any system that manifests impurity states close to one of its van Hove singularities.

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