Diffusion and localization in carbon nanotubes and graphene nanoribbons

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Abstract. We study transport length scales in carbon nanotubes and graphene ribbons under the influence of Anderson disorder. We present generalized analytical expressions for the density of states, the elastic mean free path and the localization length in arbitrarily structured quantum wires. These allow us to analyze the electrical response over the full energy range, including the regions around van Hove singularies, traditionally difficult to access by alternative approaches. Comparing with the results of numerical simulations, we demonstrate that both the diffusive and the localized regime are well represented by the analytical approximations over a wide range of the energy spectrum. The approach works well for both metallic and semiconducting nanotubes and nanoribbons but breaks down near the edge states of zigzag ribbons.

The exceptionally high electrical conductivity of carbon nanotubes (CNTs) can be attributed to a combination of various factors. The quasi-one-dimensional (quasi-1D) crystalline structure allows the preparation of samples with a very low defect density [1]. The rigidity of the sp^2-hybridized carbon lattice minimizes the effects of thermal vibrations [2]. One truely unique feature of metallic CNTs, however, is their electronic structure [3]: independent of the tube diameter, there are exactly two massless bands of high velocity crossing at the Fermi energy, resulting in a very low density of states (DOS) which effectively suppresses scattering even in the presence of disorder [4] and allows ballistic transport over hundreds of nanometers [5, 6].

Planar, finite-width graphene nanoribbons (GNRs) display many similarties to their rolled-up counterparts. The band gap of armchair-edge GNRs depends on the width just as it depends on the circumference in zigzag CNTs. Zigzag-edge GNRs, on the other hand, are always metallic just as armchair CNTs. A remarkable difference, however, lies in the edge states generally found at zigzag edges in graphene [7, 8] that are nearly localized by their extremely low dispersion and exist beside the conducting channels at the band center [9]. Energetically, these states lie exactly around the Fermi energy, so it would be expected to have an important impact on the low-energy transport properties of GNRs. The low velocity of the edge channels results in a large DOS, which should by itself lead to strong scattering. However, the local DOS (LDOS) is strongly concentrated on the edge atoms and protecting the conduction channel from scattering into the edge states.

The theory of quantum transport in disordered systems has been studied for many decades [10]. Typically, the underlying model for such studies is that of free electrons or, if a discretization is desired, electrons on a (square) lattice. Quantities such as effective electron mass, density of states or Fermi velocity are then viewed as free parameters that can be adjusted to fit the properties of metals [11]. Graphene and CNTs, however, have an electronic structure that is very different from any effective-mass free-electron approximation. In two-dimensional graphene, the electrons have a Dirac-like massless dispersion [12, 13], which results in equally massless bands of high velocity in metallic CNTs [3]. Tuning the Fermi energy away from the charge neutrality point (CNP) via electronic gating or chemical doping, CNTs of all chiralities go through a sequence of additional bands that lead to van-Hove singularities in the DOS which are typically not covered by simple models based on a smooth or fixed DOS and number of conductance channels.

In this paper, we will present a generalized expression for the DOS in arbitrarily structured, Anderson-disordered quantum wires that allows to access the whole energy range including the regions around the van Hove singularities. Using this representation of the DOS, we then show that a general expression, originally devised for metallic quantum wires with a fixed number of channels still holds true near band edges if used correctly. The various expressions will be applied to both CNTs and GNRs, including the special case of pure edge disorder in the latter case.

For 1D systems such as CNTs, it was argued by Mott and Twose [11] that all quantum states should become localized at arbitrarily low disorder, leading to an exponential suppression of the conductance at zero-temperature. For truly 1D quantum wires, Thouless demonstrated that the localization length ℓ_{loc} is identical to the mean free path ℓ_{el} [14]. Later, he derived a generalized expression for metallic quantum wires of finite diameter [15],

$$\ell_{\mathrm{loc}} \approx \frac{2Ak_{\mathrm{F}}^2}{3\pi^2}\ell_{\mathrm{el}}, \tag{1}$$

where the prefactor consisting of cross section A and Fermi wave vector k_{F} can be identified as the number of conduction channels N_{ch} in the modern language of quantum transport theory. A more accurate expression derived by Beenakker from random matrix theory reads [16]

$$\ell_{\mathrm{loc}} \approx \left[\beta\left(N_{\mathrm{ch}} - 1\right)/2 + 1\right]\ell_{\mathrm{el}}, \tag{2}$$

where $\beta = 1$ for time-reversal invariant systems and $\beta = 2$ otherwise. In either case, however, ℓ_{el} and N_{ch} are viewed as fixed parameters and it is not at all clear, how these relations should be applied to systems where these quantities vary strongly with the energy and may not even be well defined near band edges where N_{ch} is discontinuous. An alternative approach based on a perturbative expression for the Lyapunov exponents takes into account the true band structure of a quantum wire, but it still fails to reproduce the correct behavior near band edges [17].
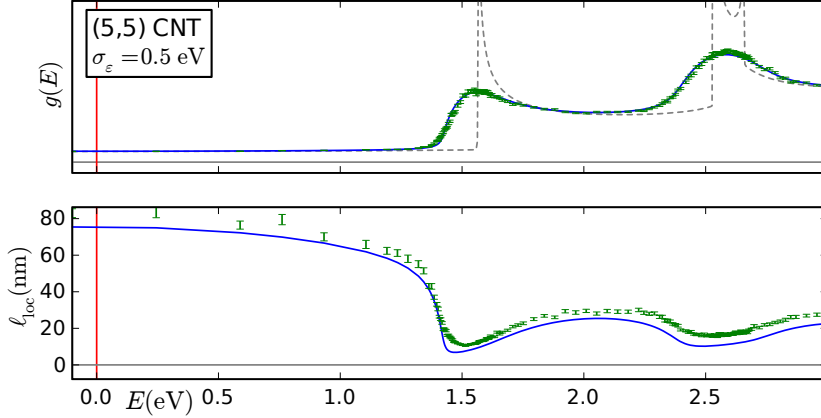
Figure 1. Density of states $g(E)$ and localization length ℓ_{loc} of an armchair (5,5) CNT under the influence of Anderson disorder. Dashed line: $g(E)$ of the clean system displaying the van Hove singularities. Solid lines: $g(E)$ from Eq. (11) and ℓ_{loc} obtained from it via Eqs. (2) and (9). Data with errorbars: values obtained numerically by averaging over 100 samples of length 1000 to $5000\ell_{\mathrm{uc}}$.

We model CNTs based on an orthogonal first-nearest-neighbor tight-binding approximation [3] with the hopping parameter $\gamma_0 = 2.66\,\mathrm{eV}$, which accurately reproduces the Fermi velocity of armchair CNTs and gives reasonable agreement with the correct band structure for several low energy bands.

For zigzag GNRs, this approximation is insufficient, as it results in a completely dispersion-free edge state while precise calculations show that this state in fact has a small but non-vanishing band width [18, 9]. Likewise, this approximation predicts every third armchair GNR to be metallic, while precise calculations predict the opening a small gap [19, 20]. The true electronic structure of a GNR would have to take into account the relaxed electronic structure at the edges [21]. As it turns out, a third-nearest neighbor parameterization [22] of an unrelaxed GNR gives a good approximation of the bands near the Fermi energy and will thus be used here.

Anderson disorder is defined as uncorrelated potential fluctuations [10]:

$$\langle \varepsilon_i \varepsilon_j \rangle - \langle \varepsilon_i \rangle \langle \varepsilon_j \rangle = \delta_{ij} \sigma_{\varepsilon_i}^2, \tag{3}$$

usually in terms of a uniform random distribution $\varepsilon_i - \varepsilon_0 \in [-W/2, W/2]$, which has a standard deviation $\sigma_\varepsilon = W/\sqrt{12}$. For perturbatively weak disorder, however, the exact shape of the distribution is irrelevant. Numerically, we use a Gaussian distribution for practical reasons. Experimentally measured values of ℓ_{el} are of the order of $10\,\mu\mathrm{m}$ in CNT of $1.5\,\mathrm{nm}$ diameter [6], which translates to an equivalent model disorder of strength $\sigma_\varepsilon \sim 0.05\,\mathrm{eV}$. The values used in Figs. 1 and 2 are chosen significantly larger to enhance the visibility of the qualitative effects.

The diffusive transmission through a quantum wire with a disordered section of length L, embedded in an otherwise disorder-free, infinite quantum wire, is defined as

$$T_{\mathrm{diff}} = N_{\mathrm{ch}} \left(1 + \frac{L}{\ell_{\mathrm{el}}} \right)^{-1}, \tag{4}$$

with the elastic mean free path ℓ_{el}. For armchair CNTs at the Fermi energy, this length ℓ_{el} was first derived by White and Torodov [4]. We have previously given a generalized derivation that is valid for arbitrary energies [23], which can be further generalized to also cover the case of inequivalent atoms in the unit cell:

Within a statistical ensemble $P(\mathcal{W})$ of the perturbation Hamiltonian $\mathcal{W}$, the scattering rate between two bands s and d at a fixed energy E is given by the Fermi golden rule

$$\tau_{E, s \to d}^{-1} = \frac{2\pi}{\hbar} g(E, d) \int \mathrm{d}\mathcal{W} P(\mathcal{W}) |\langle E, d | \mathcal{W} | E, s \rangle|^2, \tag{5}$$

which depends on the partial DOS of the destination band d. In the atomic basis $|i\rangle$, the disorder Hamiltonian is diagonal and uncorrelated [Eq. (3)], allowing for the direct evaluation of the integral
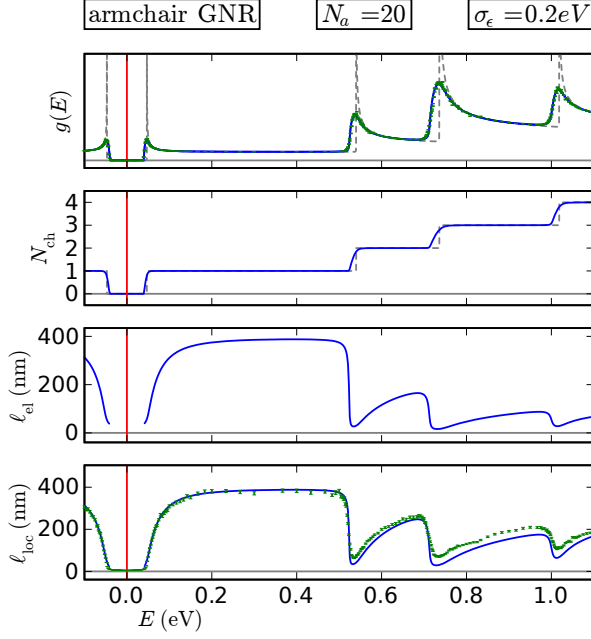
Figure 2. Density of states $g(E)$, number of channels N_{ch}, elastic mean free path ℓ_{el} and localization length ℓ_{loc} of an armchair GNR of width $N_a = 20$ under the influence of Anderson disorder ($\sigma_\varepsilon = 0.2\,\text{eV}$). Dashed lines: $g(E)$ and N_{ch} of the clean system and lengths obtained from these via Eqs. (2) and (9). Solid lines: $g(E)$ from Eq. (11), N_{ch} from Eq. (13) and lengths obtained from these. Data with errorbars: values obtained numerically by averaging over ~ 180 samples of length 2000 to $20000\ell_{\text{uc}}$.

as

$$\tau_{E,s\to d}^{-1} = \frac{2\pi}{\hbar} g\left(E,d\right) \left|\langle E,d|i\rangle\right|^2 \sigma_{\varepsilon_i}^2 \left|\langle i|E,s\rangle\right|^2 . \tag{6}$$

The elastic mean free path ℓ_{el}^s within a band s is related to the scattering rate $\tau_{E,s}^{-1}$ from this band via its electron velocity which is proportional to the inverse of the DOS as

$$\ell_{\text{el}}^s = v_s / \left(\sum_d \tau_{E,s}^{-1} \right)$$

$$= \left(hg\left(E,s\right) \sum_d \tau_{E,s\to d}^{-1} \right)^{-1} . \tag{7}$$

Based on the definition of ℓ_{el} via the diffusive transmission in Eq. (4), the elastic mean free path ℓ_{el} of a multichannel quantum wire is found by inverse averaging

$$l_{\text{el}}^{-1} = \frac{1}{N_{\text{ch}}} \sum_s \left(\ell_{\text{el}}^s\right)^{-1} . \tag{8}$$

At this point, the sums over the bands s and d can be reduced, introducing the LDOS on individual orbitals $g_i\left(E\right)$ and resulting in the final expression

$$\ell_{\text{el}} = \ell_{\text{uc}} N_{\text{ch}} \left(\pi^2 \sum_i^{\text{uc}} \left(\sigma_{\varepsilon_i}^2 g_i^2\left(E\right)\right) \right)^{-1} \tag{9}$$

with the length of the unit cell ℓ_{uc} and the sum running over all orbitals i within one unit cell. In this form, the expression can be applied to arbitrary quantum wires, including GNRs, where it also covers the special case of edge disorder by making $\sigma_{\varepsilon_i}^2$ dependent on the orbital number i.

Neglecting multiple scattering, the elastic mean free path ℓ_{el} and the diffusive transmission T_{diff} are defined in term of the LDOS $g_i(E)$ of the disorder-free system. Likewise, N_{ch} is defined by the leads, where it follows an exact integer step function. Near band edges, this diffusive transmission is discontinuous, as can be confirmed numerically to arbitrary precision, computing it as the sample average $\langle T \rangle$ of the transmission of many disorder configurations [23].

The diffusion coefficient, as it can be obtained via the time evolution of a wave packet within a long, disordered quantum wire, also allows the extraction of the elastic mean free path [24]. In this case, however, it does not depend on the DOS not of the clean, but of the disordered system. Near band edges, the DOS depends on the disorder strength non-perturbatively, causing the van-Hove singularities to broaden and to shift [25]. This effect has to be taken into account when describing diffusion near band edges or around the edge state in GNRs.

The density of states (DOS) of a quantum wire under the influence of Anderson disorder can be obtained via an algorithm based on diagrammatic perturbation theory that takes into account localization effects by including multiple scattering [25]. Dropping the crossing diagrams within the noncrossing approximation (NCA) [26], allows to write the self energy $\Sigma(E)$ to all orders as a recursive expression, which can then be iterated numerically until self-consistency is reached. Though the applicability of the NCA is not obvious, it be justified by comparing the contribution of various terms at low orders [25].

The original formulation of this approach is restricted to the special case of CNTs where all atoms are equivalent through symmetry and the self energy takes the same value for all atoms. It can, however, be generalized to arbitrarily structured quantum wires using matrix notation. The self energy $\Sigma(E)$ caused by the disorder is a diagonal matrix obeying the recursive relation

$$[\Sigma(E)]_{i,j} = \delta_{ij}\sigma_{\varepsilon_i}^2 \left[\left(E + \mathrm{i}0^+ - \mathcal{H}_0 - \Sigma(E)\right)^{-1}\right]_{ij}. \tag{10}$$

For a periodic system, the self energy has the same periodicity as the Hamiltonian. The block-tridiagonal matrix $(E + \mathrm{i}0^+ - \mathcal{H}_0 - \Sigma(E))$ can therefore be inverted numerically using a highly convergent renormalization-decimation algorithm [27, 23], allowing us to go beyond the energy range near the Fermi energy, where the special band structure allows analytic inversion.

Starting with $\Sigma = 0$, each numerical iteration of this recursive relation is equivalent to one additional perturbative order. Typically, convergence is achieved after less then ten iterations, except for energies near a van Hove singularity, where hundreds of iterations may be necessary. This clearly indicates that low-order perturbation theory breaks down near band edges.

The LDOS of each orbital i in the unit cell can now be obtained directly from the imaginary part of the Green function

$$g_i(E) = -\frac{1}{\pi}\mathrm{Im}\left[\left(E + \mathrm{i}0^+ - \mathcal{H}_0 - \Sigma(E)\right)^{-1}\right]_{i,i}. \tag{11}$$

Figs. 1 and 2 show this quantity in direct comparison with the numerically exact value obtained by sample averaging. The slight deviation visible at the flanks of the van Hove singularites is caused by the NCA [25]. The elastic mean free path ℓ_{el} based on the DOS of a disordered system, as it is displayed in Fig. 2 is no longer a purely perturbative quantity, but it takes into account the scattering into localized states present at any given energy.

The number of channels N_{ch} in Eqs. (2) and (9) is another quantity that has to be reconsidered in the vicinity of band edges. In a disorder-free quantum wire, N_{ch} is an integer valued step function that has discontinuities at band edges. The numerically obtained localization length, on the other hand, does not display any discontinuities, so the discontinuities of N_{ch} have to be somehow smoothed out by the disorder.

In any periodic system, the number of channels is equivalent to the transmission through a cross section. For a Hermitian Hamiltonian, this quantity will always take on integer values. Adding a complex self energy may, however, result in non-integer values and smooth out the discontinuities of the transmission function. Using the self-energy obtained from Eq. (10) allows thus to define an equivalent of the number of channels N_{ch} for an Anderson disordered quantum wire.
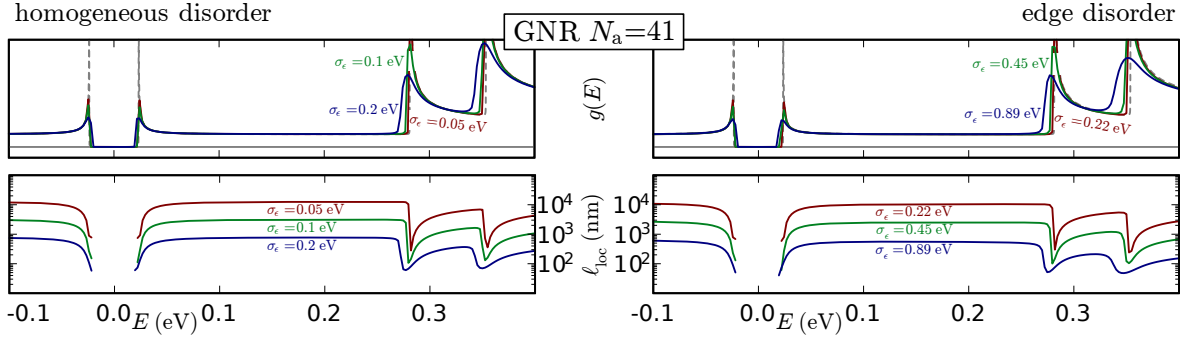
Figure 3. Analytically obtained quantities for an armchair edge GNR of width $N_z = 41$ with Anderson disorder of various strengths. Dashed lines: Density of states $g(E)$ of the clean system. Solid lines: $g(E)$ and localization length ℓ_{loc} obtained analytically. Left: homogeneous disorder over all atoms across the GNR. Right: disorder concentrated on the outermost atoms along both edges. The disorder strength σ_{ε_i} per atom is scaled such that the square total $\Sigma_i \sigma_{\varepsilon_i}^2$ is the same for both types of disorder.

The definition of the transmission through a cross section of a quantum wire in the presence of a complex self-energy requires some care. Unlike a Hermitian periodic wire, where the transmission through a cross section is equal to that through a finite-length section, the imaginary part of the self-energy would act as dissipative term in a conducting region and make the result dependent on its length. To actually obtain the transmission through a cross section, we should rather split the periodic quantum wire in only two parts – similar to the setup of a tunneling junction – writing

$$\mathcal{H} = \begin{pmatrix} \mathcal{H}_{\text{L}} & 0 \\ 0 & \mathcal{H}_{\text{R}} \end{pmatrix} + \begin{pmatrix} 0 & \mathcal{T} \\ \mathcal{T}^\dagger & 0 \end{pmatrix} = \mathcal{H}_0 + \mathcal{V} \qquad (12)$$

with two semi-infinite parts $\mathcal{H}_{\text{L}}$ and $\mathcal{H}_{\text{R}}$ and the hopping matrix $\mathcal{T}$ that connects both parts. Let us denote the Green functions of the two isolated semi-infinite leads by $\mathcal{G}_{\text{L}}$ and $\mathcal{G}_{\text{R}}$. The corresponding spectral functions are $\mathcal{A}_{\text{L/R}} = i\mathcal{G}_{\text{L/R}}^{\text{r}} - i\mathcal{G}_{\text{L/R}}^{\text{a}}$ and the Green functions of the complete system $\mathcal{G}^{\text{r/a}} = (E \pm i0^+ - \mathcal{H})^{-1}$. If $\mathcal{V}$ were a small perturbation like in a tunneling junction its matrix elements $\langle l|\mathcal{V}|r \rangle$ would directly give the transition amplitude from an eigenmode $\mathcal{H}_{\text{R}}|r\rangle = E|r\rangle$ on the righthand side to one $\mathcal{H}_{\text{L}}|l\rangle = E|l\rangle$ on the lefthand side. In a periodic system, however, $\mathcal{V}$ is of the same magnitude as $\mathcal{H}_0$, so we need to consider the full expansion $\langle l|\mathcal{V} + \mathcal{V}\mathcal{G}^{\text{r}}\mathcal{V}|r\rangle$, generalizing the tunneling-transmission $\text{tr}(\mathcal{A}_{\text{L}}\mathcal{V}\mathcal{A}_{\text{R}}\mathcal{V})$ to the nonperturbative expression [23]:

$$T = \text{tr}\left[\mathcal{A}_{\text{L}}\left(\mathcal{V} + \mathcal{V}\mathcal{G}^{\text{r}}\mathcal{V}\right)\mathcal{A}_{\text{R}}\left(\mathcal{V} + \mathcal{V}\mathcal{G}^{\text{a}}\mathcal{V}\right)\right]. \qquad (13)$$

Note that in the case of a finite self-energy Σ all along the periodic wire, this expression differs substantially from more commonly used approaches that depend on the definition of a finite-length conductor between two leads. Numerically, we can exploit the finite support of $\mathcal{V}$ by computing only the contacted regions of $\mathcal{G}_{\text{L}}$ and $\mathcal{G}_{\text{R}}$ [27, 23].

The localization length ℓ_{loc} of armchair CNTs at E_{F} can be derived analytically directly from ℓ_{el} and N_{ch} [28]. For general energies, ℓ_{loc} has so far only been accessible by numerical means [29, 30]. Based on our expressions for the DOS and the number of channels in a homogeneously disordered quantum wire, it is now possible to obtain an approximate value for ℓ_{loc} in the whole energy range. As can be seen in Fig. 1, the analytical value for ℓ_{loc} agrees with the numerically exact results fairly well. The remaining deviation can be understood in view of the oversimplified nature of Eq. (2), which is based on a model with a fixed number of equivalent channels with a single value ℓ_{el} for all scattering processes.

Armchair edge GNRs can be understood as unrolled zigzag CNTs and are indeed physically very comparable. Similar to semi-metallic $(3N, 0)$ nanotubes, which are metallic in zone-folding approximation and develop a small gap due to their curvature, ribbons of width $N_a = 3M - 1$ (counted in rows of carbon dimers) have a tiny gap that is caused by edge effects only. In Fig. 2

one can see that the DOS is smoothed out and the van Hove singularities shifted in the same way as it was observed in CNTs. Near E_{F}, this leads to a narrowing of the gap which is well reproduced by the data based on Eq. (11). The states near the gap exhibit a short localization length, as it is expected from the fact that they are caused by the disorder within the forbidden energy range of the clean system. The analytically obtained localization length coincides with the numerical value to a procesion comparable to the case of CNTs. Again, the true value is slightly underestimated when several inequivalent channels are present at the same energy.

Reducing the strength of the disorder to realistic values leads to a significant reduction of the smoothing effect on van Hove singularieties. Besides this effect, the localization lengths simply scale with $1/\sigma_{\varepsilon}^2$ wherever band edges have no influence, as can be seen in Fig. 3. Assuming the amount of intrinsic disorder in GNRs to be similar to that measured in CNTs at $\sigma_{\varepsilon} \sim 0.05\,\text{eV}$, we find a localization length of $\ell_{\text{loc}} \sim 13\,\mu\text{m}$ in armchair GNRs of 5 nm width within the plateau of only two transmitting channels at sufficient distance to the gap.

The edges of a GNR must be considered to be especially sensitive to disorder due to the lower mechanical rigidity and higher chemical activity. We therefore studied the effect of pure edge disorder as depicted in the right panels of Fig. 3. As can be immediately seen, the overall effect of the disorder scales with the square total $\Sigma_i \sigma_{\varepsilon_i}^2$ over the disorder strength of all individual atoms. When scaling the disorder strength in this way, the effects of edge disorder are identical to the case of homogeneous disorder except for the band edges, where the van Hove singularities are smeared out stronger in the case of pure edge disorder.

In zigzag edge GNRs the edge states at the Fermi energy add a considerable complication. Numerical studies have been done before on these systems using both Anderson-type disorder of short or long range [31, 32] or edge defects [33, 34, 35]. We found that the DOS obtained by Eq. (11) shows a considerable deviation from the true value obtained numerically. Possibly, this effect is related to the highly anomalous behavior found in two-dimensional sheets of graphene [36]. Trying to obtain ℓ_{loc} via Eq. (2) is bound to fail even more seriously as this approach completely neglects the different mean free paths and the spatial separation between the edge channels and the bulk channel. In fact, though this approach correctly describes a very short localization length around the edge states, it failes to reproduce the correct scaling with the ribbon width.

To conclude, we have demonstrated that our generalized expression for the density of states in Anderson-disordered quantum wires in combination with a highly convergent numerical evaluation scheme does reproduce to good precision the true data obtained by performing a numerical sample-average. The method allows to efficiently explore of the full energy range and can be applied to arbitrary quantum wires, including CNTs and armchair-edge GNRs. We further demonstrated how to obtain the localization length of those systems from the density of states in the full energy range, including the vicinity of van Hove singularities which are notoriously hard to access by analytical means. Again, the resulting data shows good agreement with statistically obtained values. The case of zigzag-edge GNRs states has to be excluded due to the extreme inequivalence of the different channels close to the Fermi energy and remains a problem for the future.

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