Linear Conductances of Gated Graphene Structures with Selected Connectivity

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In memory of prof. dr. Janko Jamnik.

Abstract

The ratio of conductances through carbon-ring based molecules are calculated for various positions of source-drain electrode leads on the molecule. These ratios are usually integers the so-called magic numbers. We find that deviations of the magic number ratios are either zero or quadratic in ratios of tight-binding model parameters.

Keywords: Conductance, Landauer formalism, quantum dots, interference effects

1. Introduction

Charge transport through nanostructures represents a challenge from the experimental point of view as well as for theoretical approaches.[1] Most of experimental work so far has been done for semiconducting structures[2] as promising candidates for tailoring various electronic devices, such as single electron transistors[3] and charge or spin quantum bits.[4] One of the advantages of semiconductor technology is its versatility in formatting structures on demand, with reliable and reproducible gating and connectivity to external leads. A disadvantage of these structures is their relatively large spatial extent limiting functional operation to low temperatures. For possible applications to sensors or quantum information processing devices room-temperature operation is desired, which demands reduction of device size leading to larger energy scale. Molecules connected to metallic leads therefore represent ideal candidates for devices where phase-coherent transport between attached electrodes is required at moderate temperatures.[5,6]

In general, nanostructures exhibit an extremely rich spectrum of quantum phenomena. In particular, in conductance measurements strong electron-electron interaction leads to Coulomb blockade,[7] the Kondo effect,[8] various spin dependent anomalies[9,10] or instabilities due to vibrational degrees of freedom, where different electron-phonon interactions can play an important role.[11,12] Due to large coherence lengths in clean structures, interference effects also play an essential role in transport through the nano-device, as was recently studied experimentally even at room temperature.[13–21] Small graphene based nanostructures are well defined since they are nearly defect-free which enables reproducible conductance measurements exhibiting subtle interference effects.[22–24]

Here we concentrate on "magic" ratios, found recently in connectivity driven electrical conductance of graphene-like aromatic molecules.[25] Theoretical analysis of experiments using mechanically controlled break junctions to measure electrical conductance of such molecules reveals specific ratios between different connectivity geometry of external leads. These magic ratios appear in the regime of particle-hole symmetrically filled molecules, where the chemical potential is located at the HOMO-LUMO mid-gap. Numerical analysis has been performed for a tight-binding approximation of a molecule weakly coupled to charge reservoirs connected to the graphene molecule via linear chains referred to as source and drain leads. In this paper we analyze the stability of magic ratios with respect to changes of the coupling to the leads and also due to changing the potential of top gates which in turn change the electron occupation of the molecule.

2. Model and Methods

We consider polycyclic aromatic hydrocarbon-like graphene structures, coupled to two metallic electrodes via source and drain leads. This is shown schematically in Figure 1 for the case of a benzene molecule.
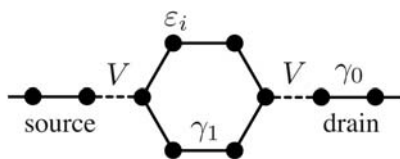
Figure 1. A benzene-like structure (molecule) attached to the leads.

To model such a system we adopt the effective Hamiltonian

$$H = H_{\text{molecule}} + H_{\text{leads}} + H_{\text{coupling}}, \tag{1}$$

where the molecule is described in terms of a tight-binding Hamiltonian

$$H_{\text{molecule}} = \sum_{i,\sigma} \varepsilon_i n_{i,\sigma} - \sum_{i,j,\sigma} \gamma_{ji} c_{j,\sigma}^{\dagger} c_{i,\sigma}. \tag{2}$$

Here i and j run over the sites of the molecule, i.e., the p_z orbitals on each of the carbon atoms, $c_{i,\sigma}^{\dagger}$ and $c_{i,\sigma}$ are the electron creation and anihilation operator, respectively, for site i and spin σ, and $n_{i,\sigma} = c_{i,\sigma}^{\dagger} c_{i,\sigma}$ is the electron number operator. ε_i are the on-site energies controlled by the top gate voltage with energy zero being the Fermi-energy in the leads in the limit of zero source-drain bias. They may also be influenced by the electrodes attached to the leads. To be specific in what follows, we assign a uniform on-site energy ε_0 to all molecular sites, which includes the effect of the top gate voltage. However, we allow on-site energies on the two sites where the leads are attached to the molecule to take a different value of ε_1. γ_{ji} are hopping integrals for which we take a value depending only on the distance on a lattice between the two sites, i.e., $\gamma_{ji} = \gamma_1$ if atoms i and j are nearest neighbors, $\gamma_{ji} = \gamma_2$ if atoms i and j are next nearest neighbors, etc. Taking into account that next nearest neighbor hopping integrals in graphene are at least an order of magnitude smaller than nearest neighbor hopping integrals,[26] in what follows we neglect all but nearest neighbor hopping integrals γ_1.

The leads, modeled as chains with sites connected by nearest neighbor hopping integrals γ_0, have a Hamiltonian

$$H_{\text{leads}} = \sum_{\alpha,i,\sigma} \varepsilon_{\alpha,i} n_{\alpha,i,\sigma} - \gamma_0 \sum_{\alpha,i,\sigma} c_{\alpha,i+1,\sigma}^{\dagger} c_{\alpha,i,\sigma} + \text{H.c.} \tag{3}$$

Here $\alpha \in \{s,d\}$ labels the source and drain leads, respectively, and i runs over the sites of a lead. $c_{\alpha,i,\sigma}^{\dagger}$, $c_{\alpha,i,\sigma}$ and $n_{\alpha,i,\sigma} = c_{\alpha,i,\sigma}^{\dagger} c_{\alpha,i,\sigma}$ are the electron creation, anihilation operator and the electron number operator for lead sites, respectively.

The couplings between leads and the molecule are

$$H_{\text{coupling}} = -V \sum_{\alpha,\sigma} c_{\alpha,1,\sigma}^{\dagger} c_{i_\alpha,\sigma} + \text{H.c.} \tag{4}$$

Here V is the hopping integral between the lead site closest to the molecule and the molecular site i_α to which lead α is attached.

The electron-electron and the electron-phonon interactions are not considered here – the systems are not in the Coulomb blockade regime. However, some interesting features due to the electron correlations in benzene were found recently.[27,28]

In the absence of many-body effects, the conductance of such a molecule, i.e., the proportionality coefficient between the current through the molecule and the voltage V_{sd} applied between the source and the drain electrode, is, in the limit of vanishing V_{sd} given by the Landauer-Büttiker formula,[29,30]

$$G = G_0 \int \mathcal{T}(\varepsilon) \left(-\frac{\partial f(\varepsilon)}{\partial \varepsilon} \right) d\varepsilon, \tag{5}$$

where $G_0 = 2e^2/h$ is the conductance quantum, with e and h being the electron charge and the Planck constant, respectively. $\mathcal{T}(\varepsilon)$ is the transmission probability through the molecule at energy ε. $f(\varepsilon) = \left(\exp \frac{\varepsilon - \mu}{k_B T} + 1 \right)^{-1}$ is the equilibrium Fermi function of the leads with μ and T being their chemical potential and temperature, respectively, and k_B being the Boltzmann constant. For the sake of convenience we set the chemical potential to the middle of the band in the leads and vanishing on-site energies in the leads, $\mu = \varepsilon_{\alpha,i} = 0$.

To calculate the transmission probability $\mathcal{T}(\varepsilon)$ we need to find the scattering eigenstate $|\psi\rangle$ of the Hamiltonian for an electron with energy ε and spin σ, incoming from the source electrode,

$$H|\psi\rangle = \varepsilon|\psi\rangle. \tag{6}$$

We expand such an eigenstate in the local basis states of the molecule $c_{j,\sigma}^{\dagger}|0\rangle$ and the leads $c_{\alpha,j,\sigma}^{\dagger}|0\rangle$

$$|\psi\rangle = \sum_j \psi_j c_{j,\sigma}^{\dagger}|0\rangle + \sum_{\alpha,j} \psi_{\alpha,j} c_{\alpha,j,\sigma}^{\dagger}|0\rangle. \tag{7}$$

The wavefunction in the source electrode is a linear combination of an incoming and a reflected plane wave, $\psi_{s,j} = e^{-ikj} + re^{ikj}$, while the wavefunction in the drain elec-

trode consists of a transmitted wave, $\psi_{d,j} = te^{ikj}$. The wavevector k can be calculated from the dispersion relation of the leads, $\varepsilon = -2\gamma_0 \cos k$. r and t are the reflection and the transmission amplitude, respectively. The transmission probability is $\mathcal{T}(\varepsilon) = |t|^2$.

Here we demonstrate the method of calculating the transmission probability $\mathcal{T}(\varepsilon)$ for the case of the simplest possible molecule, namely a single site with the on-site energy of ε_0 coupled to two leads. The Schrödinger equation for such a system reads as a set of linear equations for t, r and ψ_0

$$-\gamma_0 \left(e^{-2ik} + re^{2ik} \right) - V\psi_0 = \varepsilon \left(e^{-ik} + re^{ik} \right),$$
$$-V \left(e^{-ik} + re^{ik} \right) + \varepsilon_0 \psi_0 - Vte^{ik} = \varepsilon \psi_0, \qquad (8)$$
$$-V\psi_0 - \gamma_0 te^{2ik} = \varepsilon te^{ik}.$$

By eliminating r and ψ_0 we find the transmission amplitude,

$$t = \frac{i \frac{2V^2}{\gamma_0} \sin k}{\varepsilon - \varepsilon_0 + \frac{2V^2}{\gamma_0} e^{ik}}. \qquad (9)$$

A Breit-Wigner resonance of width $\Gamma_0 = \Gamma_s + \Gamma_d$, where $\Gamma_s = \Gamma_d = \frac{2V^2}{\gamma_0}$ are the partial widths due to coupling to the source and the drain lead, respectively, forms at energy ε_0 in the transmission probability in the wide band limit where $\gamma_0 \gg \varepsilon, \varepsilon_0 \Gamma_0$,

$$\mathcal{T}(\varepsilon) = \frac{\left(\frac{\Gamma_0}{2}\right)^2}{(\varepsilon - \varepsilon_0)^2 + \left(\frac{\Gamma_0}{2}\right)^2}. \qquad (10)$$

Generalization to more general molecules with arbitrary topology is straightforward. In the limit of weak coupling to the leads, $\Gamma = \frac{2V^2}{\gamma_0} \gg \gamma_1$, the transmission probability consists of similar resonances, positioned at eigenenergies of the molecule.

3. Results

As shown in Figure 2(a–c), sites of graphene-like molecules we consider in this work form a bipartite lattice, i.e. they break up into two sublattices in such a way that unprimed sites 1, 2, 3 …, forming one sublattice, are connected only to primed sites 1′, 2′, 3′ …, forming the other sublattice. The Hamiltonian of such a system possesses the particle-hole symmetry.[31] In molecules considered here, this symmetry is actually weakly broken due to next nearest neighbor hopping integrals γ_2. Since, as discussed in Section 2, $\gamma_2/\gamma_1 \ll 1$ for structures considered in this work, we neglect such terms in what follows. Therefore, the conductance as a function of the top gate voltage ε_0 is even with respect to the particle-hole symmetric point $\varepsilon_0 = 0$, where the Fermi energy of the leads coincides with the center of the HOMO-LUMO gap. This is shown in Figure 2 where the zero temperature and room temperature conductances are plotted as a function of top gate voltage for benzene, naphthalene and anthracene molecules for a particular choice of sites to which electrodes are attached. The zero temperature conductance curves consist of a set of resonances, each corresponding to a molecular level being at the Fermi energy of the electrodes. The width of a resonance measures the coupling of the molecular level to the leads. Note that some of the re-
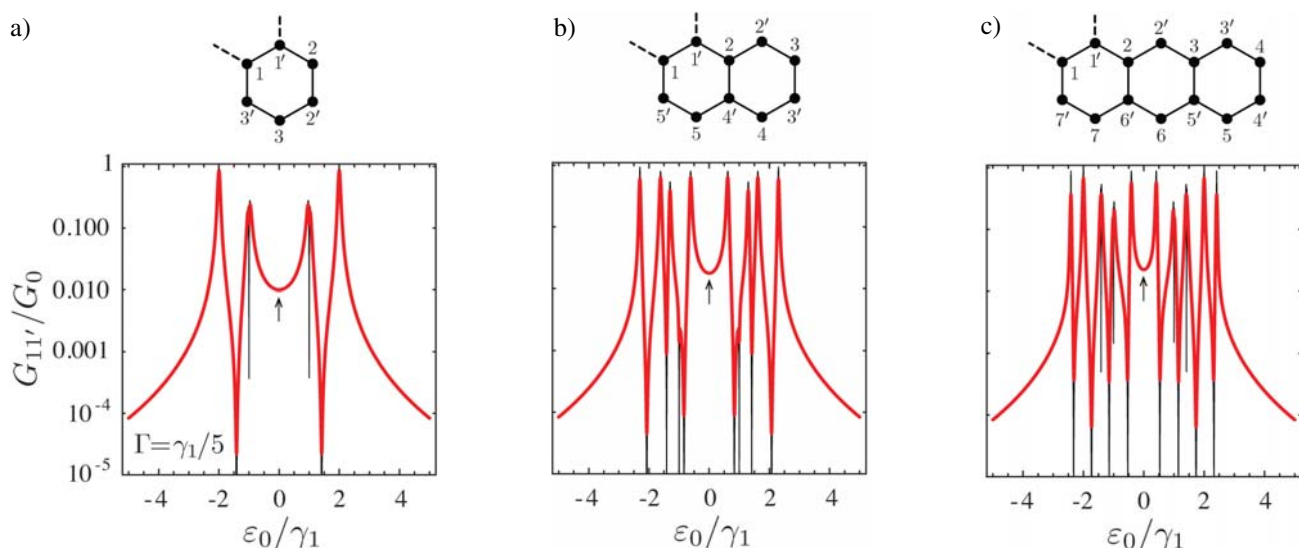
Figure 2. Conductance as a function of top gate voltage of the (a) benzene, (b) naphthalene, and (c) anthracene molecule when one electrode is connected to site 1 and the other is connected to site 1' of the molecule, at $T = 0$ (black lines) and at the room temperature (red lines). The coupling of an electrode to the molecular site is $\Gamma = \gamma_1/5$. Arrows indicate the position of the center of the HOMO-LUMO gap.

sonances are split due to degeneracy of molecular orbitals. At room temperature, i.e., $T = 300$ K $\sim 0.01\gamma_1$, with $\gamma = 2.5$ eV as appropriate for graphene,[32] thermal broadening only slightly lowers the peak heights, increases their widths and broadens minima. Within the HOMO-LUMO gap the effect of finite temperature is negligible at room temperature. As we will concentrate on the vicinity of the center of the HOMO-LUMO gap in the rest of this work, the calculations will be done at zero temperature in what follows.

We now study the dependence of the conductance on the on-site energy ε_0 incorporating the top gate voltage and the coupling Γ of a lead to a molecular site, for different combinations of molecular sites to which the electrodes are attached. Let us first discuss the situation at the particle-hole symmetric point (indicated by arrows in Figure 2), when the coupling to the leads Γ is weak.[25] If the leads are connected to two sites in the same sublattice the conductance is zero due to destructive interference. On the other hand, if the leads are attached to two sites in distinct sublattices, a "magic integer" can be associated with such a system. The ratio of conductances of two such systems is the so-called "magic ratio" which is a square of the ratio of the corresponding magic integers.

To prove this, we follow Ref. 25. Provided the molecule is weakly coupled to the leads, all the on-site energies of the molecular sites are equal and the Fermi energy of the leads coincides with the center of the HOMO-LUMO gap, the conductance is proportional to the absolute square of the Green's function of an isolated molecule between sites to which the source and drain leads are attached $G_{i_d, i_s}(0)$. The Green's function is determined by the molecular Hamiltonian, $G(0) = (0 - H_{\text{molecule}})^{-1}$. If only nearest neighbor hopping is included in the Hamiltonian and all the corresponding hopping integrals are equal, such a Hamiltonian can be written in a block form: the diagonal blocks representing Hamiltonians of each sublattice are zero while the off-diagonal blocks are proportional to the adja-

cency matrix of the graph of the molecule. The inverse of the adjacency matrix, multiplied by its determinant, is a matrix containing integer matrix elements, the "magic integers". Comparing conductances of the same molecule with different connectivities, the ratio of those conductances is equal to the square of the ratio of the corresponding magic integers, provided the coupling to the leads is the same for both connectivities.

In Figure 3 we show how magic ratios evolve with the coupling Γ increasing both at the particle hole symmetric point and away from it at $\varepsilon_0 = \gamma_1/5$, which is still within the HOMO-LUMO gap of all the molecules considered in this work. At the particle-hole symmetric point a magic ratio, provided its weak coupling value is different from one, starts to deviate from its weak coupling value when Γ becomes of the order of γ_1. A magic ratio increases with Γ if its weak coupling value is less than one and it decreases with Γ if its weak coupling value is larger than one. At the particle-hole symmetry point $\varepsilon_0 = 0$ the deviation of magic ratios greater than one is quadratic in Γ for $\Gamma \ll \gamma_1$. Away from the center of the HOMO-LUMO gap magic ratios deviate from a square of the ratio of magic integers even in the weak coupling limit. The deviation is again quadratic in ε_0 for $|\varepsilon_0| \ll \gamma_1$. At $\varepsilon_0 \neq 0$ a molecule conducts even if electrodes are attached to sites in the same sublattice. Compared to the conductance when electrodes are attached to sites in different sublattices it is smaller by a factor of $(\varepsilon_0/\gamma_1)^2$ for $|\varepsilon_0| \ll \gamma_1$.

An electrode may shift the on-site energy at the molecular site to which it is attached. This turns out to be another cause of deviation of a magic ratio from a square of the ratio of the magic integers. Figure 4 displays that the departure of on-site energies ε_1 at these molecular sites from on-site energies $\varepsilon_0 = 0$ at other molecular sites causes a magic ratio to increase quadratically with ε_1 if it is larger than one for $\varepsilon_1 = 0$. A magic ratio is independent of ε_1 if it is equal to one for $\varepsilon_1 = 0$.
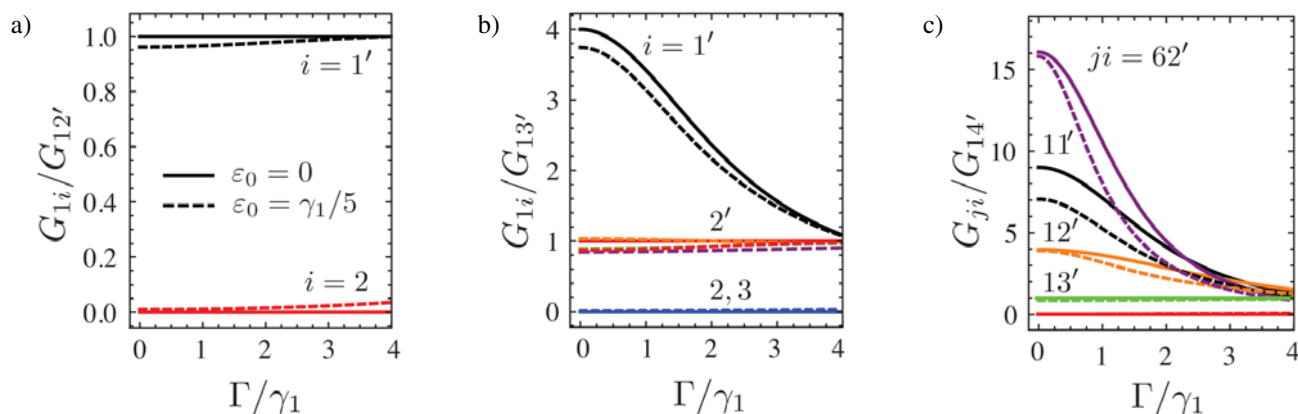
Figure 3. Magic ratios at $T = 0$ of the (a) benzene, (b) naphthalene, and (c) anthracene molecule at the particle-hole symmetric point (full lines) and for $\varepsilon_0 = \gamma_1/5$ (dashed lines) as a function of the coupling Γ of an electrode to a molecular site. Molecular sites to which electrodes are attached are indicated next to each curve. The other combination of electrode attachment sites appearing in the conductance ratio corresponds to the most distant sites of a particular lattice.
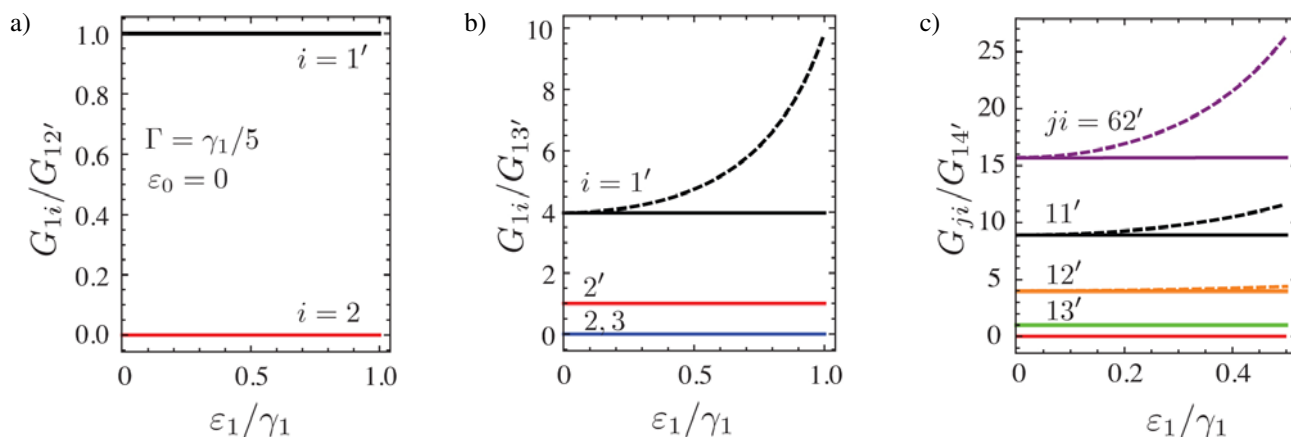
Figure 4. Magic ratios at *T = 0* of the (a) benzene, (b) naphthalene, and (c) anthracene molecule when the on-site energies ε_1 at molecular sites where the electrodes are attached differ from those at other molecular sites where $\varepsilon_0 = 0$ (dashed lines). Full lines show magic ratios for $\varepsilon_1 = 0$. Here $\Gamma = \gamma_1/5$.

4. Conclusions and Outlook

In conclusion, we have calculated the ratios of conductances of graphene-like structures for different combinations of sites to which leads are attached away from the regime where those ratios can be expressed in terms of magic integers. The deviations were due to top gate voltage pushing on-site energies ε_0 away from the center of the HOMO-LUMO gap, due to the coupling to leads Γ being non-negligible and due to the electrodes causing on-site energies ε_1 on atoms to which leads are attached to deviate from on-site energies on other atoms. The deviation from the ratio given by magic integers was found to become important when those parameters become of the order of the hopping integral γ_1 of the molecule. For small values of those parameters, the deviation was found to increase proportionally to $(\varepsilon_0/\gamma_1)^2$, $(\Gamma/\gamma_1)^2$ or $(\varepsilon_1/\gamma_1)^2$. Furthermore, if the top gate voltage is non-zero, the molecule conducts even when both leads are attached to sites in the same sublattice, which for other perturbations is not the case. What remains to be done is to study the robustness of magic ratios to Coulomb interaction and to perturbations breaking the particle hole symmetry, i.e., the second neighbor hopping within the molecule.

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6. References

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Povzetek

Razmerja prevodnosti skozi molekule, sestavljene is obročev ogljikovih atomov, smo izračunali za različne konfiguracije priključkov elektrod med izvorom in ponorom. Običajno so ta razmerja cela števila, t.i., magična števila. Ugotovili smo, da s spreminjanjem parametrov v okviru modela tesne vezi ostanejo magična števila ali nespremenjena, ali pa so od parametrov odvisna kvadratično.