Transport, optical properties and quantum ratchet effects for quantum dots and molecules coupled to Luttinger liquids.

A. Komnik$^1$ and A. O. Gogolin$^2$

$^1$Physikalisches Institut, Albert–Ludwigs–Universität, D–79104 Freiburg, Germany
$^2$Department of Mathematics, Imperial College London, 180 Queen’s Gate, London SW7 2BZ, United Kingdom

(Dated: October 31, 2018)

We present non-perturbative solutions for multi-level quantum dot structures coupled to interacting one-dimensional electrodes out of equilibrium. At a special correlation strength the Hamiltonian can be mapped to the Kondo problem which possesses a solvable Toulouse point, where all conductance and noise properties can be calculated exactly. Special attention is paid to the fully asymmetric setup when each dot level is coupled to only one of the leads and the electron transport through the structure is accompanied by photon absorption (emission). A relation between the optical spectra and the energy dependent current noise power is established. Experimental implications of the results, specifically for the Fano factor, the ratchet current, and field emission via localised states, are discussed. In particular, we predict that the peak in the ratchet current as function of the irradiation frequency splits up in two due to correlation effects.

PACS numbers: 73.63.-b, 71.10.Pm, 73.63.Kv

I. INTRODUCTION

Manufacturing of micro- and nano-electronic circuitry based on single molecules represents one prospective way to achieve further miniaturisation as well as efficiency improvement of electronic devices. First successful attempts of contacting single molecules have been reported in a number of recent experimental works [1, 2, 3, 4]. One possible mechanism for the electron transport through them is tunnelling on and off the molecular orbitals (MOs). The smaller the molecule the larger is the energy distance between the MOs so that in some cases the transport occurs through only one electron level even at room temperatures. Hence, the adequate physical description of such systems coincides with that of the single-state quantum dot (QD): a fermionic level coupled to metallic electrodes (we shall also call them ‘leads’ or ‘contacts’).

If one aims at small device dimensions one has to go for one dimensional (1D) electrodes. Promising candidates for such wirings are the carbon nanotubes in their single-wall version (SWNTs) [5, 6]. However, truly one-dimensional electron systems cannot be described by the Fermi liquid (FL) model. No matter how weak the fermion interactions are, they cannot be taken into account perturbatively. It is well known that in the low energy sector the interacting 1D fermions constitute a universality class of Luttinger liquids (LL), which display a completely different physics than the conventional FLs [7]. As a consequence, the electronic degrees of freedom in sufficiently thin SWNTs are also described by a generalisation of the LL model [8, 9]. This has also been confirmed experimentally [10].

In the most of existing experiments the contacts between the molecule and the leads are quite weak. The optimal operation of the future nano-electronic devices is, however, expected in setups with small contact resistances. That can be achieved only in systems where the current-mediating MOs undergo strong hybridisation with the valence bands of the electrodes. In the case when two MOs with different symmetries couple to different leads electronic transport can only take place via emission or absorption of photons. Transport resulting from such ‘optical’ coupling can be distinguished from the background transport by its dependence on the laser irradiation frequency. In spite of vast amount of contributions dealing with QD’s transport and optical properties (see, for example, [11], and references therein), systems with good contacts did not receive much attention. Moreover, to the best of our knowledge, the issue of interacting electrodes has been discussed only in resonant tunnelling context [12, 13, 14, 15, 16]. In this paper we begin to close these gaps and present non-perturbative solutions for multi-level dots contacted by interacting 1D electrodes. We wish to clarify here that these type of correlations are different from ‘on-dot’ couplings (Coulomb or Hubbard terms). The latter coupling gives rise to the Kondo type phenomena [17]. In this paper we neglect the on-dot couplings and effectively deal with spinless electrons. This restricts the validity of our results to the temperatures above the Kondo temperature $T_K$ or to the case when a polarising magnetic field is present. Such an approach appears to be justified for SWNTs, which display strong LL correlations in a wide temperature range from about 5 K to about 100 K [10], typical $T_K$’s being much smaller. The interplay of on-dot and ‘in-lead’ correlation is of theoretical interest but remains outside the scope of this paper.

The outline of the paper is as follows.

In Section II we start with a review of the simplest realisation: a single-state dot coupled to 1D electrodes. These results have been recently announced in our Letter [12]. Contrary to [12] we work in the Green’s function formalism, which is more suited to access the noise prop-
properties. Since the transport in such a setup is completely understood both in the resonant case as well as in the off resonant case, our primary goal in the Section III is the study of the Fano factor, which is a ratio of the zero frequency noise and the transport current, as a function of bias voltage. For the sake of completeness we derive all equations needed to access the full energy dependence of the noise power spectrum.

In Section II we go over to a two-state QD with both levels coupled to 1D leads. Since the non-interacting situation is more or less trivial we restrict our considerations only to the interacting system. It turns out, that, as in the case of the single-state QD, the Hamiltonian can again be brought to a quadratic form in terms of new fermions for a special interaction strength \( \lambda \). In this representation the non-linear transport as well as zero frequency noise properties can be expressed via the transmission coefficient even in case of an additional tunnelling term between the dot levels. Our non-perturbative approach allows then to study all resonant tunnelling effects (known for single-state setups \[13\]) in this situation.

In Section IV we investigate transport in a similar structure where every level is only coupled to one of the electrodes and where the dominant transport mechanism is the photon-assisted tunnelling between the levels. That situation corresponds to coupling of two MOs of different symmetries discussed above. It turns out that a finite current can flow even without any applied voltage. That makes such a system one of the simplest realisation of the so-called ‘quantum ratchet’ effect \[19, 20, 21\]. We concentrate on the analysis of the ‘ratchet’ current as a function of the radiation frequency. Some general results, including an important relation between the current noise power and the absorption and emission spectra are discussed in Section IV A. A treatment of non-interacting systems follows in Section IV B. Transport through a dot coupled to LL electrodes is then analysed in Section IV C.

A short summary of results (Section V) concludes the paper. We stress again that though we model the molecule–electrodes coupling by means of tunnelling Hamiltonians, all our results are non-perturbative in tunnelling amplitudes contrary to the bulk of existing studies \[11\].

II. TRANSPORT THROUGH A SINGLE-STATE QUANTUM DOT

First we briefly review the method of \[12\] and then discuss the noise properties.

A. Scattering states solution and duality

We model the system by the following Hamiltonian (we ignore the spin throughout the paper):

\[
H = H_K + H_t + H_C ,
\]

where \( H_K \) is the kinetic part,

\[
H_K = \Delta d^\dagger d + \sum_{i=R,L} H_0[\psi_i] ,
\]

describing the electrons in the leads \( H_0[\psi_i] \), and the resonant level \( \Delta \), the corresponding electron operators being \( d^\dagger, d \). The dot can be populated from either of the two leads \( (i = R, L) \) via electron tunnelling with amplitudes \( \gamma_i \),

\[
H_t = \sum_i \gamma_i [d^\dagger \psi_i(0) + h.c.] .
\]

In \[11\], \( H_C \) describes the electrostatic Coulomb interaction between the leads and the dot,

\[
H_C = \lambda_C d^\dagger d \sum_i \psi_i(0)^\dagger \psi_i(0) .
\]

This interaction is a new ingredient we have introduced, absent in the related studies \[13\] and \[14\]. It does not, however, affect the universality as we shall show later.

The contacting electrodes are supposed to be one-dimensional half-infinite electron systems. We model them by chiral fermions living in an infinite system: the negative half-axis then describes the particles moving towards the boundary, while the positive half-axis carries electrons moving away from the end of the system. In the bosonic representation \( H_0[\psi_i] \) are diagonal even in presence of interactions (for a recent review see e.g. \[2\]): we set the renormalised Fermi velocity \( v = v_F/g = 1 \), the bare velocity being \( v_F \):

\[
H_0[\psi_i] = (4\pi)^{-1} \int dx [\partial_x \psi_i(x)]^2 .
\]

Here the phase fields \( \phi_i(x) \) describe the slow varying spatial component of the electron density (plasmons),

\[
\psi_i(x) = \partial_x \phi_i(x) / 2\pi \sqrt{\gamma} .
\]

The electron field operator at the boundary is given by \[12\],

\[
\psi_i(0) = e^{i\phi_i(0)}/\sqrt{\gamma}/\sqrt{2\pi a_0} ,
\]

where \( a_0 \) is the lattice constant of the underlying lattice model. Here \( g \) is the conventional LL parameter (coupling constant) connected to the bare interaction strength \( U \) via \( g = (1 + U / \pi v_F)^{-1/2} \). In the chiral formulation the bias voltage amounts to a difference in the densities of the incoming particles in both channels far away from the constriction \[22\]. The current is then proportional to the difference between the densities of incoming and outgoing particles within each channel.

To the best of our knowledge, Hamiltonian \[11\] cannot be solved exactly even in the \( g = 1 \) case as long as \( \lambda_C \) remains finite. However, after a transformation of \( d^\dagger \) and \( d \) operators to the spin representation of the form

\[
\begin{align*}
S_x &= (d^\dagger + d)/2, \\
S_y &= -i(d^\dagger - d)/2, \\
S_z &= d^\dagger d - 1/2, 
\end{align*}
\]
one immediately observes that the $\lambda_C$ term is analogous to the $S_z$-spin density coupling in the Kondo problem. The latter is known to be explicitly solvable at a particular value of the longitudinal coupling: the Toulouse limit (see e.g. Ref. [2]). Let us perform a similar calculation. As a first step we introduce new symmetric and antisymmetric fields

$$\phi_{\pm} = (\phi_L \pm \phi_R)/\sqrt{2},$$  

which still fulfill the bosonic commutation relations. Then we apply the transformation $H' = U^\dagger H U$ with

$$U = \exp(iS_z\phi+(0)/\sqrt{2}g),$$

which changes the kinetic and the Coulomb coupling parts of the full Hamiltonian to [we drop a constant contribution proportional to $S_z\delta(x)$ that does not affect the transport]

$$H'_K + H'_C = H_K + (\lambda_C/\pi \sqrt{2}g - \sqrt{2/g})S_\partial \phi+(0),$$

and the tunnelling part (terms containing $\gamma_i$) to

$$H'_t = (2\pi a_0)^{-1/2} \left[ S_\ell (\gamma_L e^{i\phi_\ell}/\sqrt{2}g + \gamma_R e^{-i\phi_\ell}/\sqrt{2}g) + (\gamma_L e^{-i\phi_\ell}/\sqrt{2}g + \gamma_R e^{i\phi_\ell}/\sqrt{2}g) S_\ell \right],$$

where $S_{\pm} = S_x \pm iS_y = d_\dagger d$. At the point $g = 1/2$ one can re-fermionise the problem by defining new operators

$$\psi_{\pm} = e^{i\phi_{\pm}}/\sqrt{2\pi a_0},$$

which fulfill standard fermionic commutation relations. With the help of the particle density operator $\psi_{\pm}^\dagger \psi_{\pm} = \partial_x \phi_{\pm}/2\pi$ we can immediately write down the refermionised Hamiltonian,

$$H = H_0[\psi_{\pm}] + (\lambda_C - 2\pi)2S_z \psi_{\pm}^\dagger \psi_{\pm} + \Delta S_z$$

$$+ S_\ell (\gamma_L \psi_+ + \gamma_R \psi_+) + (\gamma_L \psi_+ + \gamma_R \psi_-) S_\ell.$$  

(6)

In the case of the symmetric coupling $\gamma_L = \gamma_R$ this Hamiltonian is similar to that of the two-channel Kondo problem and, at the Toulouse point $\lambda_C = 2\pi$, can be solved exactly (out of equilibrium) using the method of Ref. [2]. The novel ingredient in the following analysis is the extension to the asymmetric case. To take advantage of the Toulouse point we set the Coulomb coupling amplitude to $2\pi$ in what follows. This not only removes the four fermion interaction but decouples the ‘$\pm$’ channels making the ‘$+$’ channel free (i.e. decoupled from the dot variables).

At the Toulouse point our Hamiltonian describes free fermions which scatter at the origin. These new fermions are related to the physical electrons in a highly non-local way. As the relations between the particle densities are still linear, in order to access the transport properties it is sufficient to calculate the energy dependent transmission coefficient $1 - T(\omega)$ of the new fermions $[T(\omega)]$ being the transmission coefficient of the physical ones]. The non-linear $I - V$ characteristics is then given by

$$I(V) = G_0 \int d\omega T(\omega) [n_F(\omega - V) - n_F(\omega)],$$  

(7)

where $n_F$ denotes the Fermi distribution function and $G_0 = e^2/h$ is the conductance quantum. The easiest way to identify $T(\omega)$ is the equation of motion method. We calculated $T(\omega)$ in Ref. [2], it is given by (we measure all energies in units of $\Gamma = \gamma_L^2 + \gamma_R^2$):

$$T(\omega) = \frac{4\gamma^2 E^2}{(E^2 + \beta_+^2)(E^2 + \beta_-^2) + 2\gamma^2(E^2 + \beta_- \beta_+ + \gamma^4)},$$

(8)

where $E = \Delta^2 - \omega^2,$

$$\beta_{\pm} = [(1 - 2\alpha)\Delta \pm \omega]/2,$$

$$\gamma = \omega \sqrt{\alpha (1 - \alpha)},$$

and $\alpha = \gamma_L^2/(\gamma_L^2 + \gamma_R^2)$ is the asymmetry parameter.

Using expressions (4) and (5) one can access all conductance properties of the system. We shall not discuss them again (see Ref. [2]) but concentrate instead on the duality property. In the simplest case of the symmetric model on-resonance ($\alpha = 1/2, \Delta = 0$) we obtain

$$T(\omega) = (1 + \omega^2)^{-1}.$$  

(9)

As a consequence, the temperature dependent differential conductance at zero bias (which is the most relevant quantity from the experimental point of view) amounts to

$$G_{\Delta=0}(T)/G_0 = \frac{1}{2\pi T} \psi' \left( \frac{1}{2} + \frac{1}{2\pi T} \right),$$

(10)

where $\psi$ denotes the $\psi$-function. Comparing this result with the conductance $G_{1/2}$ through a single scatterer in an LL with $g = 1/2$, given in Ref. [14], we find that (similar relations are valid for the non-linear $I - V$'s)

$$G_{\Delta=0}(T)/G_0 = 1 - G_{1/2}(T)/G_0,$$  

(11)

where $T$ in $G_{1/2}(T)$ is measured in units of the backscattering strength. According to the duality hypothesis the strong coupling fixed point at $g$ corresponds to the weak coupling one at $1/g$ and vice versa, leading to the relationship of the conductances of the form $[14] [25] [24].$

$$G_2(T)/G_0 = 1 - G_{1/2}(T)/G_0.$$  

(12)

Therefore, Eqs. (11) and (12) suggest that tunnelling between two LLs with $g = 1/2$ via a resonant level is equivalent to direct tunnelling between two LLs with $g = 2$.

In fact, we can demonstrate this equivalence on the Hamiltonian level. We start with (10) and in order to
simplify things introduce new real (Majorana) fermions a, b and ζ, η according to
\[ d = (a + i b)/\sqrt{2} , \psi_- = (\zeta + i \eta)/\sqrt{2} . \] (13)
In this language the Hamiltonian acquires the form
\[ H = H_0[\zeta, \eta, a, b, i \gamma_{-a} \eta(0) + i \gamma_{+b} \zeta(0)] , \] (14)
where \( \gamma_{\pm} = \gamma_L \pm \gamma_R \) and the unperturbed part is defined by
\[
H_0[\zeta, \eta, a, b] = i \Delta a b + i \int dx \left[ \eta(x) \partial_x \eta(x) + \zeta(x) \partial_x \zeta(x) \right] + V \zeta(x) \eta(x) .
\]
For future reference, we give the current operator in this representation:
\[ J = -i/2 \gamma_- a \zeta(0) - i/2 \gamma_+ b \eta(0) . \]

On the other hand, the Hamiltonian for the direct tunnelling between two LLs in terms of the physical fermions is
\[ H = \sum_{i=R,L} H_0[\psi_i] + \gamma \left( \psi_L^\dagger \psi_R + \psi_R^\dagger \psi_L \right) , \]
containing the free part \( H_0 \), which again describes two half-infinite LLs, and tunnelling between them with the amplitude \( \gamma \). We bosonize the above Hamiltonian as in the previous Section using rules \( \ref{boson} \) and \( \ref{boson2} \), and obtain
\[ H = \sum_i H_0[\phi_i] + \frac{\gamma}{2 \pi a_0} \left[ e^{i(\phi_L-\phi_R)/\sqrt{3}} + e^{-i(\phi_L-\phi_R)/\sqrt{3}} \right] . \]

Introducing new fermions \( \ref{fermions} \) at the point \( g = 2 \) (we drop the ‘+’ channel again since it is free)
\[ H = H_0[\psi_-] + \frac{\gamma}{\sqrt{2 \pi a_0}} (\psi_-^\dagger + \psi_-) . \]
Next we take advantage of a trick from Ref. \( \ref{trick} \) and perform a substitution \( \Psi = (d - d^\dagger) \psi_- \), where \( d \) is some local fermionic operator not related in any way to the dot operator of the previous Section. Obviously, such transformation does not change either the commutation relations or the normalisation of the operators. Then
\[ H = H_0[\Psi] + \frac{\gamma}{\sqrt{2 \pi a_0}} (c - c^\dagger)(\Psi^\dagger + \Psi) . \]
The last step is obvious: one introduces the Majorana components according to \( \ref{-majorana} \). This results in
\[ H = H_0[\zeta, \eta] + i \gamma \sqrt{2 \pi a_0} b \zeta(0) , \]
which is precisely the Hamiltonian \( \ref{Hamiltonian} \) of a resonant setup \( (\gamma_\zeta = 0, \Delta = 0 \) and, of course \( V = 0) \) up to the redefinition \( \gamma_\zeta = \gamma \sqrt{2 \pi a_0} \). We have checked that the bias voltage and the current operator of the \( g = 2 \) LL problem and the resonant tunnelling system transform correctly.

### B. Green’s functions solution and noise properties

Although the transport properties can easily be accessed by means of the scattering formalism as shown in Ref. \( \ref{GFK} \), it is not immediately clear (see below though) how the information about the finite frequency noise can be extracted from the transmission coefficient. A more appropriate method to calculate the fluctuations is the Green’s functions (GFs) method in its non-equilibrium (Keldysh) formulation. For further reference we now define all possible non-equilibrium GFs. Let \( \mu, \nu \) stand for either of the electrode Majoranas \( \zeta \) or \( \eta \) (taken at \( x = 0 \)) and \( f, h \) stand for either the dot level operators, \( a \) or \( b \). Then define
\[ D^{ij}_{fh}(t-t') = -i(T_C f(t) h(t')) , \]
\[ G_{\mu \nu}^{ij}(t-t') = -i(T_C \mu(t) \nu(t')) , \]
\[ G_{ij f}^{ij}(t-t') = -i(T_C f(t) f(t')) , \]
\[ G_{ij h}^{ij}(t-t') = -i(T_C f(t) h(t')) , \]
where \( T_C \) is the time ordering operator along the Keldysh contour \( C \), which consists of the forward \( C_- \) and the backward \( C_+ \) paths. The times \( t, t' \) belong to the paths \( C_{i,j} \), respectively. Sometimes we shall omit the Keldysh indices \( i, j \) in what follows adopting matrix notation for the Keldysh GFs. In the above definitions we assumed the system to be in a steady state so that all GFs are translationally invariant in the time domain and therefore depend only on the time differences. There is an obvious relation \( G_{\mu \nu}^{ij}(t-t') = -G_{ij}^{ij}(t'-t) \). However, working with two definitions possesses advantages as we shall see later.

It is, in fact, not difficult to calculate the zero order GFs, when \( \gamma_i = 0 \). For the electrode Majoranas we obtain
\[ G_{\zeta \zeta}^{(0)}(\omega) = G_{\eta \eta}^{(0)}(\omega) = i \left( \frac{H(\omega)}{H(\omega) - 1} \right) , \] (15)
where \( H(\omega) \) contains the information about the Fermi distribution functions \( n_F \) of the original electrons in the leads, \( H(\omega) = n_F(\omega + V) - n_F(-\omega + V) \). Obviously, the cross correlations \( G_{\zeta \eta}^{(0)} \) exist only as long as the applied voltage is finite,
\[ G_{\zeta \eta}^{(0)}(\omega) = -G_{\eta \zeta}^{(0)}(\omega) = \frac{1}{2} F(\omega) \left( \begin{array}{cc} 1 & 1 \\ 1 & 1 \end{array} \right) . \] (16)
This fact is reflected by the function \( F(\omega) = n_F(\omega + V) - n_F(-\omega - V) \) vanishing as \( V \to 0 \). As a consequence of the special forms of \( \ref{cross} \) and \( \ref{cross2} \) the retarded and the advanced components are fairly simple: \( G_{\mu \nu}^{(0) R,A} = 0 \) while \( G_{\mu \nu}^{(0) R,A} = \mp i/2 \). The \( a - b \) subsystem being in equilibrium makes the calculation of the corresponding GFs even simpler. The result is
\[ D_{ff}^{(0) --}(++) = \pm \frac{1}{2} \sum_{p=\pm} |\omega + p(\Delta - i \delta)|^{-1} , \]
In the same way one can derive the corresponding equations for the electrode Majorana GFs:

\[ D_{ff}^{(0)-(+)} = \pm i\delta(\omega + \Delta), \]
\[ D_{ab}^{(0)-(+)} = \mp \frac{i}{2} \sum_{p=\pm} |\Delta - p(\omega + i\delta)|^{-1}, \]
\[ D_{ab}^{(0)-(+)} = \mp \pi\delta(\omega + \Delta). \]

Using these zero order GFs as a starting point we can calculate any correlation function exactly because of the quadratic form of the Hamiltonian. Our goal is to calculate the average of the current operator \( I(V) = \langle J \rangle \) and the power spectrum of current fluctuations (we shall also call this quantity noise spectrum), which is defined as

\[ P(\Omega) = \int dt e^{i\Omega t} \left[ \langle J(t)J(0) \rangle - \langle J(0) \rangle^2 \right]. \tag{17} \]

The averages are calculated using the \( S \) matrix for the coupling of dot and electrode Majoranas, \( \langle \ldots \rangle = \langle \ldots \rangle_S \), which is

\[ S = T_C \exp \left( -\int d\tau\gamma_a(\tau)\eta(\tau) + \gamma_b(\tau)\zeta(\tau) \right). \]

Expanding in powers of \( \gamma \), one can derive for the current the analog of the Meir-Wingreen formula \[ \text{22}, \]

\[ I(V) = \frac{i}{8\pi} \int d\omega F(\omega)[\gamma_+^2 D_{bb}^{(0)}(\omega) - \gamma_-^2 D_{aa}^{(0)}(\omega)]. \tag{18} \]

We choose to split the noise spectrum into two contributions, \( P(\Omega) = P_\parallel(\Omega) + P_\perp(\Omega) \), where both quantities can be expressed in terms of off-diagonal (containing different fermion species) GFs, what different structure,

\[ G_{ab}^{+-} = -\frac{\gamma_+}{2} F D_{bb}^{(0)} - i\gamma_- (D_{ab}^{(0)-} G_{\eta\eta}^{(0)+} + D_{ab}^{(0)+} G_{\eta\eta}^{(0)-}) \tag{22} \]

where the remaining GFs, \( G_{\eta\eta}^{+-}, G_{\zeta\zeta}^{+-} \) and \( G_{\zeta a}^{+-} \) are similar. The remaining off-diagonal GFs cannot be reduced to the diagonal ones. They should rather be found as solutions of a chain of Dyson equations. For the \( a - b \) subsystem one obtains the following system of equations (to simplify notation we ignore here the Keldysh indices):

\[ D_{ab} = D_{ab}^{(0)} + \gamma_+^2 D_{ab}^{(0)+} G_{\zeta\zeta}^{(0)} D_{ab} + \gamma_-^2 D_{ab}^{(0)-} G_{\eta\eta}^{(0)} D_{ab} + \gamma_+ \gamma_- (D_{ab}^{(0)-} G_{\eta\eta}^{(0)+} + D_{ab}^{(0)+} G_{\eta\eta}^{(0)-}) \tag{23} \]
\[ D_{bb} = D_{bb}^{(0)} + \gamma_+^2 D_{bb}^{(0)+} G_{\zeta\zeta}^{(0)} D_{bb} + \gamma_-^2 D_{bb}^{(0)-} G_{\eta\eta}^{(0)} D_{bb} + \gamma_+ \gamma_- (D_{bb}^{(0)-} G_{\eta\eta}^{(0)+} + D_{bb}^{(0)+} G_{\eta\eta}^{(0)-}) \tag{24} \]

In the same way one can derive the corresponding equations for the electrode Majorana GFs:

\[ G_{\zeta\eta} = G_{\zeta\eta}^{(0)} + \gamma_+^2 G_{\zeta\zeta}^{(0)} D_{bb}^{(0)} G_{\zeta\eta} + \gamma_-^2 G_{\zeta\zeta}^{(0)} D_{ab}^{(0)} G_{\eta\eta} - \gamma_+ \gamma_- (G_{\zeta\zeta}^{(0)} D_{bb}^{(0)} G_{\eta\eta} - G_{\zeta\zeta}^{(0)} D_{ab}^{(0)} G_{\eta\eta}), \tag{25} \]
\[ G_{\eta\eta} = G_{\eta\eta}^{(0)} + \gamma_+^2 G_{\eta\zeta}^{(0)} D_{bb}^{(0)} G_{\zeta\eta} + \gamma_-^2 G_{\eta\zeta}^{(0)} D_{ab}^{(0)} G_{\eta\eta} + \gamma_+ \gamma_- (G_{\eta\zeta}^{(0)} D_{bb}^{(0)} G_{\eta\eta} + G_{\zeta\eta}^{(0)} D_{ab}^{(0)} G_{\eta\eta}). \tag{26} \]

In the simplest symmetric case \( \gamma_- = 0 \) we obtain from Eq. \[ \text{24} \] for the advanced dot level GF

\[ D_{bb}^{(0)} = D_{bb}^{(0)+}/(1 + \gamma_+^2 D_{bb}^{(0)} G_{\zeta\zeta}^{(0)}). \]
Plugging this result into the expression for the current results in Eq. \( \mathbf{4} \) with all the energy variables measured in units of \( \Gamma = \gamma^2 / 4 \) we identify

\[
T(\omega) = \omega^2 / [ (\omega^2 - \Delta^2)^2 + \omega^2 ],
\]

as precisely the transmission coefficient \( \mathbf{5} \) at \( \alpha = 1/2 \) found previously by means of the equations of motion method.

As for the noise spectrum, only the parallel component survives. Eq. \( \mathbf{24} \) has the following solution:

\[
\alpha = \gamma \text{ precisely the transmission coefficient (8) at where the current carrying excitations are again of the}
\]

\[
\text{above Dyson equations but can simply use formula (8), even in the asymmetric case we do not have to solve the}
\]

\[
(18) \text{ results in Eq.(7) with all the energy variables measured in units of } \Gamma = \gamma^2 / 4 \text{ we identify}
\]

\[
\text{the correct pre-factors):}
\]

\[
G_{\mathbf{C}_0}^A = G_{\mathbf{C}_0}^{(0)A} / (1 + \gamma^2 D^{(0)A} G_{\mathbf{C}_0}^{(0)A}) ,
\]

which is zero. Therefore we have \( G_{\mathbf{C}_0}^+ = G_{\mathbf{C}_0}^{(0)+} \). The rest of the needed GFs can be read off Eq. \( \mathbf{22} \), \( G_{bb}^+ = i \gamma F D_{bb}^R / 2 \), and \( G_{bb}^- = -i \gamma F D_{bb}^A / 2 \). Collecting all terms in \( \mathbf{19} \) we obtain the following result:

\[
P_{\text{sym}}(\Omega) = e G_0 \int d\omega \left\{ - \frac{F(\omega) F(\Omega - \omega) \omega (\Omega - \omega)}{(\omega^2 - \Delta^2 - i \omega)(\Omega^2 - \Delta^2 + i (\Omega - \omega))} + \frac{H(\omega) - 1}{|H(\omega) - 1|^2} \right\}.
\]

The same formula has been obtained by Schiller and Hershfield (SH) in Ref. \( \mathbf{24} \) in the context of the non-equilibrium Kondo problem, where the magnetic field strength plays the role of our level energy \( \Delta \).

In the following we do not repeat the results for the noise spectrum which are already contained in Ref. \( \mathbf{24} \) but rather concentrate on the aspects which are specific to resonant tunneling, in particular on the asymmetric case and on the calculation of the Fano factor not covered by SH.

In the limit of zero frequency we find the noise spectrum to be given by the formula

\[
P_{\text{sym}}(0) = e G_0 \int d\omega T(\omega)[1 - T(\omega)]
\]

\[
\times \left[ n_F(\omega - V) - n_F(\omega) \right].
\]

identical to the one derived for non-interacting electrons \( \mathbf{24} \). This is somewhat surprising. The reason must be that we map the original Hamiltonian onto a free one, where the current carrying excitations are again of the fermionic nature. That is why in order to access \( P(0) \) even in the asymmetric case we do not have to solve the above Dyson equations but can simply use formula \( \mathbf{18} \) for the transmission coefficient.

Let us pause here to mention that, in non-interacting resonant tunneling systems at zero temperature, the Fano factor \( \nu_V = P(0)/e I(V) \) is suppressed in comparison to the Schottky value \( \nu = 1 \) by the factor \( (\Gamma_L^2 + \Gamma_R^2)/(\Gamma_L + \Gamma_R)^2 \) at high voltages \( V \gg \Gamma \) and by the factor \( (\Gamma_L - \Gamma_R)^2/(\Gamma_L + \Gamma_R)^2 \) in the opposite limit \( V \ll \Gamma \) is related to the cross-correlation \( G_{\mathbf{C}_0}^A \). The latter is a solution to one of the equations in \( \mathbf{26} \).

\[
G_{\mathbf{C}_0}^A = G_{\mathbf{C}_0}^{(0)A} / (1 + \gamma^2 D^{(0)A} G_{\mathbf{C}_0}^{(0)A}) ,
\]

\[
\text{As previously } \Gamma_{L(R)} \text{ denote the dimensionless conductances of the left(right) contact. The suppression is}
\]

\[
\text{maximal value no matter how strong the asymmetry is.}
\]

At zero temperature and on–resonance we find using \( \mathbf{30} \) that the on-resonance shot noise is (we also recover the correct pre-factors):

\[
P_{\text{sym}}(0) = e G_0 \frac{1}{2} \left[ \frac{\tan^{-1} V}{V} - V/(1 + V^2) \right],
\]

Taking into account the formula for current, \( I(V) = G_0 \tan^{-1} V \), one can read off the Fano factor, which has the following limiting forms:

\[
\nu_{\nu \to 0} = \frac{1}{3} V^2 + O(V^4),
\]

\[
\nu_\infty = \frac{1}{2}.
\]

In the general asymmetric situation the transmission coefficient takes the form

\[
T(\omega) = \frac{4 \alpha (1 - \alpha) \omega^2}{[\omega^4 + 1/4 + \alpha(1 - \alpha)]^2 - \alpha(1 - \alpha)}.
\]

The evaluation of the Fano factor now yields

\[
\nu_0 = 1,
\]

\[
\nu_\infty = 2\alpha^2 - 2\alpha + 1 = \frac{\Gamma_L^2 + \Gamma_R^2}{(\Gamma_L + \Gamma_R)^2}.
\]
can see from [28, 32]. The latter is not at all the same in both fermionic representations of the problem, so that we have to solve the full set of Dyson equations [25, 26]. The full solution is rather lengthy. It does not appear to reveal qualitatively new features as compared to what is already known [24] as the effects of asymmetry and finite $\Delta$ do not compete but rather enhance each other. We shall therefore conclude the discussion of the noise spectrum at this point.

III. THE TWO–STATE QUANTUM DOT

In the spirit of the previous Section we model the double QD by two fermionic levels with energies $\Delta_{1,2}$, which are coupled to LL leads, see Fig. 2. As long as the levels do not interact with each other in any other way then by tunnelling, the whole treatment including the solution of the equations of motion can be performed for an arbitrary number of levels. Throughout this Section we are not interested in noise power spectra so that we concentrate only on the conductance properties of the system, which are most easily accessed by means of the equation of motion method. The Hamiltonian of the system is still assumed to be of the form [11] with following changes:

(i) The kinetic part describes two (or more) levels instead of only one,

$$H_K = \sum_{i=1,2} \Delta_i d_i^{\dagger} d_i + \sum_{i=R,L} H_0[\psi_i].$$

(ii) The tunnelling amplitudes are the same for both levels. It is, in fact, not difficult to solve the problem with arbitrary amplitudes. This, however, does not induce new physics, so we restrict our solution to this special case:

$$H_t = \sum_i \sum_{j=R,L} \gamma_{ij}[d_i^{\dagger} \psi_j(0) + h.c.].$$

(iii) The strength of the electrostatic Coulomb interaction is also assumed to be the same for both levels,

$$H_C = \lambda C \sum_i d_i^{\dagger} d_i \sum_j \psi_j(0) \psi_j(0).$$

(iv) There is an additional term in the Hamiltonian that is responsible for the tunnelling processes between the dot levels [11]:

$$H_W = W(d_1^{\dagger} d_2 + h.c.).$$

To describe the electronic degrees of freedom in the electrodes we use the same formalism as in the previous Section, see Eqs. [29]. As in the case of a single level we can introduce symmetric and anti-symmetric components [11] and spin representations for the level operators (they acquire an index $i = 1, 2$) and apply a slightly different transformation to the overall $H$, defined by

$$U = \exp \left( i \sum_i S_i^z \phi_i / \sqrt{2g} \right).$$

[Fig. 1: The Fano factor as a function of the bias voltage for different asymmetry values ($\alpha = 0.5, 0.4, 0.3, 0.2, 0.1$ from the bottom curve upwards).]
where in (6) only by the sums over both spins, and when the Hamiltonian acquires a very convenient quadratic form.

\[ H = H_0[\psi_\pm] + \sum_i (\lambda_C - 2\pi) S^z_i \psi^\dagger_i \psi_i + \Delta_i S^z_i + S^z_i (\gamma_L \psi_- + \gamma_R \psi^\dagger_-) + (\gamma_L \psi_+ + \gamma_R \psi^\dagger_+) S^z_i. \] (33)

In what follows we concentrate on the Toulouse point when \( \lambda_C = 2\pi \), where the `±' channels decouple and when the Hamiltonian acquires a very convenient quadratic form.

In order to calculate the non-linear \( I(V) \) we employ the method of Ref.\[12\], which results in Eq.\[7\] with some modified \( T(\omega) \). As in the case of the single-level dot, the easiest way to find the transmission coefficient is the equations of motion method. Since we have two types of operators: for the electrons of the `−' channel and for the dot levels (we go back to the original \( d_i^\dagger, d_i \) operators), we need two types of equations of motion,

\[ i\partial_t \psi_-(x) = -i\partial_x \psi_-(x) + \sum_i \delta(x)(\gamma_L d_i - \gamma_R d_i^\dagger), \]

\[ i\partial_t d_i = \Delta_i d_i + W d_{-i} + \gamma_L \psi_-(0) + \gamma_R \psi^\dagger_-(0). \] (34)

Integrating the first one around \( x = 0 \) with respect to \( x \) from \(-\epsilon \) to \( \epsilon \) and then sending \( \epsilon \) to zero we obtain

\[ i[\psi_-(0^+) - \psi_-(0^-)] = \sum_i \gamma_L d_i - \gamma_R d_i^\dagger. \] (35)

where \( 0^\pm \) denotes positive (negative) infinitesimal. We define new operators

\[ Y = \prod_i (i\partial_t - \Delta_i) - W^2, \]

\[ L = 2(i\partial_t + W) - \sum_i \Delta_i, \]

\[ Z_\pm = \partial_t^2 + i \sum_i \Delta_i \partial_t \pm (\Delta_0 \Delta_1 - W^2). \] (36)

By acting with \( |Y|^2 \) on both sides of Eq.\[35\] and using the last two equations \[36\] we can eliminate the dot operators. We obtain as a result

\[ |Y|^2[\psi(0^+) - \psi(0^-)] = (\gamma_L^2 Z_+ L + \gamma_R^2 Z_- L^*)\psi(0) + \gamma_L \gamma_R (Z_+ L + Z_- L^*)\psi^\dagger(0). \] (37)

Now we can insert into this relation the momentum decomposition of the field operator \( \psi_- \)

\[ \psi_-(x,t) = \int \frac{dk}{2\pi} e^{ik(t-x)} \left\{ \begin{array}{l} a_k \text{ for } x < 0 \\ b_k \text{ for } x > 0 \end{array} \right. \] (38)

Because the dispersion relation is linear, \( \omega = vk = k \), we can use \( \omega \) as the momentum variable as well as the energy variable. Inserting Eq.\[38\] into Eq.\[37\] and using \( \psi_-(0) = [\psi_-(0^+) + \psi_-(0^-)]/2 \) results in a following equation, which (up to a redefinition of constant factors) is independent of the number of levels,

\[ E(b_\omega - a_\omega) = -i\beta_+ (a_\omega + b_\omega) + i\gamma (a^\dagger_- \omega + b^\dagger_- \omega). \] (39)

We introduced the following objects,

\[ E = \prod_{p=\pm} \left[ \Pi_i (\omega + p\Delta_i) - W^2 \right], \]

\[ \gamma = \omega \sqrt{\alpha(1 - \alpha)} \] \[ \left[ \sum_j \Delta_j^2 - 2\omega^2 \right] - 2W \left[ \sum_i \Delta_i - W \right], \]

\[ \beta_\pm = (1/2 - \alpha) \left[ \sum_j \Pi_{m\neq j} (\Delta_m - \omega)^2 - 2W\omega^2 \right] - W^2 \sum_i \Delta_i \pm \gamma/2\sqrt{\alpha(1 - \alpha)}. \] (40)

Formally Eq. \( [39] \) has exactly the same form as in the single-level case. Therefore the resulting transmission coefficient is still given by formula \[38\] with modified constants contained in \[40\].

The temperature behaviour of \( G \) at the maxima does not differ considerably from that for one single level, which has already been studied in Ref.\[12\]. The conductance \( G(T) \) in the valley between the peaks turns out to be very well described by a superposition of two peaks of the single level problem. Moreover, the presence of the
The couplings to individual levels are subject to change as a consequence of the operator sum transformation
\[ d_1 + d_2 = \sqrt{2}\cos(\alpha + \pi/4)\tilde{d}_1 + \sqrt{2}\cos(\alpha - \pi/4)\tilde{d}_2, \]
which makes the coupling of the upper level decrease with growing tunnelling amplitude \( W \) when \( \alpha \) tends to \( \pi/4 \). The coupling of the lower level, on the contrary, increases. Notice that such renormalisation occurs even in the non-interacting systems. The physical reason is that the conductance via tunnelling through the lower level is enhanced because of the additional depopulation process which transfers electrons to the upper level. The non-trivial interaction effect in our setup is the increasing height of the upper level. The reason is that due to smaller \( \Gamma_{R,L} \) the upper level is at effectively higher temperature, which, in turn, means that the conductance is higher in the asymmetric case. For the same reason the height of the lower peak diminishes.

The coupling symmetry is not affected by the diagonalisation transformation, so that the resonant conductance is increasing monotonically all the way to zero temperature. That is why in this case the upper peak amplitude is lower, see Fig. 3.

\[ 1 \leq \frac{\Gamma}{\gamma} \leq 2 \]

\[ d_{1,2} \]

\[ \Delta_{1,2} \]

\[ \tilde{d}_{1,2} \]

\[ R_{\alpha} \]

\[ \tan 2\alpha = \frac{2W}{\Delta_2 - \Delta_1} \]

\[ \Delta_{1,2} = \frac{\Delta_1 + \Delta_2}{2} \pm \sqrt{\left( \frac{\Delta_1 - \Delta_2}{2} \right)^2 + W^2}. \]

\[ H_l = \gamma_L d_1^\dagger \psi_L(0) + \gamma_R d_2^\dagger \psi_R(0) + h.c. \quad (41) \]

We assume that the localised levels possess different symmetries so that a direct tunnelling between them is forbidden (nevertheless, the Coulomb coupling, being free of selection rules, is still symmetric) while hopping with simultaneous emission or absorption of a photon with energy \( \Omega \) is possible. Then the coupling of the levels can be written as

\[ H_W = W \left( e^{i\Omega t} d_2^\dagger d_1 + e^{-i\Omega t} d_2^\dagger d_1^\dagger \right), \]

where \( W \) denotes the coupling amplitude. We dropped the photon operators since we assume that the dot (or molecule) is subject to intensive laser radiation (so that there is always a phonon which can be absorbed).

Although the full Hamiltonian is now explicitly time dependent it still can be reduced to a system in a steady state via the gauge transformation

\[ d_1 \to d_1 e^{i\Omega t/2}, \]

\[ d_2 \to d_2 e^{-i\Omega t/2}. \quad (42) \]
Thus, the coupling to the radiation effectively results in a finite bias voltage. As a consequence, a finite current can flow without any real applied voltage at all. This is the quantum ratchet effect which has recently been intensively discussed in Refs. [14, 21] in various non-interacting setups with mostly weak couplings to the leads.

The physical explanation of the effect in the original picture prior to the gauge transformation is quite simple. We start with a system where the two dot levels possess two different energies, \( \Delta_2 < \Delta_1 \) as in Fig. The population probability of the lower of level (\( \Delta_2 \)) is higher than that of its counterpart \( \Delta_1 \), so that the photon absorption is the dominant process transferring the electrons to \( \Delta_1 \). They can relax either to the left lead or back to \( \Delta_2 \). However, if the hybridisation of the dot levels is larger than the electromagnetic coupling, the dominant relaxation process is tunnelling into the left lead. This leads to a non-zero net current through the system.

The quantities we are interested in are again the full current \( I(V) \) through the system, the current noise power \( P(\omega) \) and the light absorption (emission) \( A(\omega)[E(\omega)] \) spectra. The first quantity can be defined e.g. via the expectation value of the particle flow between the left lead and \( \Delta_1 \),

\[
\hat{I} = -i\gamma_L[d_2^\dagger \psi_L(0) - \psi_L^\dagger(0)d_1].
\]  

(43)

In the language of non-equilibrium Keldysh diagram technique it is given by the following expression [32, 34, 35],

\[
I = \frac{\gamma_L^2}{2\pi} \int d\omega \left[ G_L^{(0)+}(\omega)D_1^{+}(\omega)
- G_L^{(0)-}(\omega)D_1^{-}(\omega) \right],
\]  

(44)

where \( +, - \) (\( +, - \)) indices stand for lesser and greater Keldysh GFs. For the left (right) lead electrons they are denoted by \( G_{L,R}^{ij}(\omega) \) while for the dot electrons by \( D_{L,R}^{ij}(\omega) \). The additional superscript (0) distinguishes the GFs in the absence of tunnelling couplings. Alternatively one can define the current as the expectation value of the tunnelling operator between the dot states,

\[
\hat{I}' = -iW(d_1^d d_2 - d_1^d d_1). 
\]  

(45)

The third possible method to describe the transport is the transmission coefficient formalism. Using definition [45], we derive an expression for the noise power spectrum of the form:

\[
P(\omega) = W^2 \int dt e^{i\omega t} \left[ \langle d_1^d(t)d_2(t)d_2^d(0)d_1(0) \rangle 
+ \langle d_2^d(t)d_1(t)d_1^d(0)d_2(0) \rangle \right] - 2\pi\delta(\omega)I^2.
\]

The light absorption rate can be evaluated via the Golden Rule and is given by

\[
A(\omega) = 2\pi W^2 |\langle f|d_1^d d_2|0 \rangle|^2 \delta(\omega - \Delta_1 + \Delta_2),
\]

where the vacuum \( |0 \rangle \) is assumed to be the state with the level \( \Delta_1 \) empty and \( \Delta_2 \) full. The final state \( |f \rangle \) is just the opposite. It turns out that the Fourier transform of the function

\[
S^A(t) = -i\langle d_2^d(t)d_1(t)d_1^d(0)d_2(0) \rangle
\]

(46)
directly related to the absorption rate,

\[
A(\omega) = iW^2 S^A(\omega).
\]

(47)

\[
S^A(\omega) = -\frac{i}{2\pi} \int d\epsilon D_2^{+}(\epsilon)D_1^{-}(\epsilon + \omega).
\]

Another interesting aspect of Eq. [40] is the fact that it can be written down in the form

\[
S^A(\omega) = -\frac{i}{2\pi} \int d\epsilon D_2^{+}(\epsilon)D_1^{-}(\epsilon + \omega).
\]

(47)

This formula is exact as long as the GFs are calculated exactly and the level operators participate only in two-particle interaction terms (such as \( H_W \) or \( H_e \)).

Similarly one can show that the emission rate \( E(\omega) \) is proportional to a related function

\[
E(\omega) = iW^2 S^E(\omega),
\]

(48)

where

\[
S^E(t) = -i\langle d_1^d(t)d_2(t)d_2^d(0)d_1(0) \rangle,
\]

(49)

the Fourier transform having the same form as (48), up to the exchange \( 1 \leftrightarrow 2 \). Thus we establish a very convenient expression relating the total optical spectrum with the noise power of our system:

\[
P(\omega) = A(\omega) + E(\omega) - 2\pi\delta(\omega)I^2.
\]

(50)

This relation has far reaching consequences for the field emission (FE) physics [36]. In the case when the chemical potential of the right lead is sent to \( \mu_R = -\infty \) the double dot setup describes tunnelling into vacuum through a sequence of localised states, which is nothing else than FE [37]. Experimentally, it turns out that under certain conditions the FE from carbon nanotubes is accompanied
by luminescence phenomena. One possible explanation has been offered in Ref. However, it was suggested that localised levels on the nanotube tip play the dominant role during the light emission. Therefore, measuring the noise power spectrum along with the luminescence spectrum during FE experiments and comparing them with the prediction would allow one to check the hypothesis put forward in Ref. . However, we would like to postpone the detailed discussion of this very extensive issue to a later publication and rather concentrate here on the ratchet effects. (We shall still give some general formulae for the noise power spectrum.)

\[
D_{1,2}^{(0)}(\omega) = \frac{1}{(\omega - \Delta_{1,2})^2 + \Gamma_{L,R}^2} \left( \omega - \Delta_{1,2} - i\Gamma_{L,R} \text{sgn}(\omega \mp V/2) \right) \begin{pmatrix}
\omega + \Delta_{1,2} - i\Gamma_{L,R} \text{sgn}(\omega \mp V/2) & i2\Gamma_{L,R}(\omega \mp V/2) \\
-i2\Gamma_{L,R} \Theta(\omega \mp V/2) & -\omega + \Delta_{1,2} - i\Gamma_{L,R} \text{sgn}(\omega \mp V/2)
\end{pmatrix},
\]

Equation system is linear and can easily be solved for all GFs. We need only the following ones (we drop the trivial energy argument \(\omega\)):

\[
\begin{align*}
D_{1,2}^{+} & = \frac{D_{1,2}^{(0)} - K_{1,2}^{+} + [1 - K_{1,2}^{-}]D_{1,2}^{(0)} + \text{det}K_{1,2}^{-}}{\text{det}K_{1,2}}, \\
D_{1,2}^{-} & = -\frac{D_{1,2}^{(0)} - K_{1,2}^{-} + [1 + K_{1,2}^{-}]D_{1,2}^{(0)} - \text{det}K_{1,2}^{-}}{\text{det}K_{1,2}},
\end{align*}
\]

where

\[
\text{det}K_{1,2} = (1 - K_{1,2}^{-})(1 + K_{1,2}^{+}) + K_{1,2}^{-}K_{1,2}^{+} = |1 + K_{1,2}^{R}|^2.
\]

Evaluation of the current with help of and yields at zero temperature the following result:

\[
I = G_0 \int_{-V/2}^{V/2} d\omega T(\omega),
\]

where the transmission coefficient is given by

\[
T(\omega) = \begin{pmatrix}
4\alpha_{L}\alpha_{R} \\
[((\omega - \Delta_1)^2 + \alpha_R^2)((\omega - \Delta_2)^2 + \alpha_L^2) - 2(\omega - \Delta_1)(\omega - \Delta_2) - \alpha_R\alpha_L] + 1
\end{pmatrix}.
\]

From now on all energy variables are normalised to \(W\) and dimensionless, \(\alpha_{R,L} = \Gamma_{R,L}/W\). However, it is expected that the optical coupling is much smaller than

FIG. 6: Diagrammatic representation of the Dyson equation. Solid lines represent the GFs of the level \(\Delta_1\) and the dashed lines those of \(\Delta_2\). Thick lines are the exact GFs.

The energies \(\Delta_{1,2}\) are already renormalised by the gauge transformation and \(\Gamma_{R,L} = \rho_{R,L}\gamma_{R,L}^{\gamma}\) where \(\rho_{R,L}\) are the density of states (DOS) in the corresponding lead. In the absence of an external bias voltage \(V = \Omega\). The tunnelling term \(H_W\) is quadratic so that we can write down the exact Dyson equation for the full GFs of the dot levels. In terms of the Keldysh contour ordered GFs it can be written as

\[
D_{1,2}(t - t') = D_{1,2}^{(0)}(t - t') + W^2 \int_C dt_1 dt_2 D_{1,2}^{(0)}(t_1 - t) \times D_{2,1}^{(0)}(t_1 - t_2)D_{1,2}(t_2 - t') .
\]

The corresponding diagrams are depicted in Fig. Disentangling the indices one obtains the following set of equations,

\[
D_{1,2}^{ij}(\omega) = D_{1,2}^{(0)ij}(\omega) - \sum_{m=\pm} mK_{1,2}^{im}(\omega)D_{1,2}^{mj}(\omega) ,
\]

with the kernels

\[
K_{1,2}^{ij}(\omega) = -W^2 \sum_{m=\pm} mD_{1,2}^{(0)im}(\omega)D_{1,2}^{jm}(\omega) .
\]

B. The non-interacting case

We first discuss the case of non-interacting leads (and \(\lambda_C = 0\)). Then the lead electron fields can be integrated out exactly. As a result we are then left with zero-dimensional problem of two fermionic levels. The corresponding Keldysh GFs at zero–order in tunnelling are given by the following matrices:

\[
D_{1,2}^{0} = \frac{1}{(\omega - \Delta_{1,2})^2 + \Gamma_{L,R}^2} \begin{pmatrix}
(\omega - \Delta_{1,2}) - i\Gamma_{L,R} \text{sgn}(\omega \mp V/2) & i2\Gamma_{L,R}(\omega \mp V/2) \\
-i2\Gamma_{L,R} \Theta(\omega \mp V/2) & (\omega + \Delta_{1,2}) - i\Gamma_{L,R} \text{sgn}(\omega \mp V/2)
\end{pmatrix}.
\]
that to the contacting electrodes, so that we can expand \( \Gamma_0 \) for large \( \alpha_{R,L} \). As a result we obtain the transmission coefficient in form of two superimposed Lorentzians at energies \( \Delta_{1,2} \) and widths \( \Gamma_{R,L} \). This implies for the \( I - V \),

\[
I(V) = \frac{G_0 2\Gamma_R\Gamma_L W^2}{\pi} \int_{-V/2}^{V/2} \frac{d\omega}{\pi \omega^2 + \Gamma^2} - \frac{d\omega}{\pi \omega^2 + \Gamma^2 - \omega} .
\]

The latter integral can easily be calculated in a closed form. The results are given in Appendix VI. In the case of symmetric electrode couplings, \( \Gamma_R = \Gamma_L = \Gamma \), the current induced by the photon absorption (we shall call it ‘ratchet’ current) decays according to \( \sim W^2\Gamma/\Omega^2 \) at high frequencies. In the opposite infrared limit it varies linearly:

\[
I \sim \frac{W^2\Omega}{\Delta_1 - \Delta_2} \sum_{i=1,2} \frac{\Delta_i}{\Delta_i^2 + \Gamma^2} .
\]

In the intermediate regime the ratchet current has its maximal value around \( \Omega \sim \Delta_1 - \Delta_2 \), see Fig. 7. A slight shift towards higher frequencies is a result of the mutual level hybridisation due to tunnelling similar to that occurring in the case of a double dot. It turns out that coupling asymmetry does not result in any qualitative change in the induced current. The qualitative picture remains the same for all values of the optical coupling \( W \). Depending on the sign of the external bias voltage the ratchet current enhances or suppresses the transport. This effect might be of immediate experimental relevance since it indicates the way the levels of the quantum dot (or orbitals in the case of molecular dot) are arranged with respect to contacting electrodes.

As already discussed in Section III the knowledge of \( T(\omega) \) enables not only to access the transport but also the zero–frequency noise properties using formula (29). For the calculation of the full frequency dependent noise spectrum the mere knowledge of \( T(\omega) \) is insufficient because one needs the transmission amplitudes \( \tilde{T}(\omega) \). In such a situation one can use the relation (32). Generally the optical coupling is expected to be relatively weak with respect to the lead-dot coupling so that we can calculate the emission spectra at the leading order using Eqs. (51):

\[
S^E(\omega) = 2\Gamma_R\Gamma_L W^2 \pi \Theta(V + \omega) \int_{-V/2}^{V/2} \frac{\pi}{(\epsilon - \Delta_1)^2 + \Gamma^2} \left[ (\epsilon + \omega - \Delta_2)^2 + \Gamma^2 \right]^{-1} (\epsilon + \omega)^{-1} .
\]

This integral can be performed analytically but the result is lengthy. So we shall discuss only the special case of high bias voltage, corresponding to the FE via localised states. The evaluation of (58) then yields

\[
S^E(\omega) = 2\Gamma_R\Gamma_L (\Gamma_R + \Gamma_L) \frac{W^2}{\pi} \frac{(\Delta_1 + \Delta_2 + \omega)^2 + (\Gamma_L - \Gamma_R)^2}{(\omega + \Delta_1 - \Delta_2)^2 + \Gamma_L^2 + \Gamma_R^2 - 2\Gamma_L^2\Gamma_R^2} .
\]

Since the emission process lowers the energy of the radiating system the actual spectrum is \( \Theta(\omega)S^E(\omega) \). Then this result describes a Lorentz-shaped peak around \( \Delta_2 - \Delta_1 \), which is at odds with the experimental finding of Ref. 38, where a superposition of two Gaussian peaks has been detected. There are three possible reasons for this discrepancy; we list them in the order of relevance. First of all, the measuring apparatus can superimpose its own sensitivity curve, which is usually of a Gaussian shape, over the actual luminescence spectrum. Secondly, since the localised states on the nanotube tip could exist on the ends of free dangling \( C - H \) bonds the finite temperature can lead to oscillations of the energy levels \( \Delta_{1,2} \), which then could become normally distributed. Another possible reason could be the electronic correlations inside the nanotube. However, there is no \textit{a priori} argu-
ment why their influence can result in Gaussian-shaped spectra.

C. Interacting LL leads at \( g = 1/2 \)

Now we turn to the case of interacting leads. The Hamiltonian still contains the terms \( H_K, H_C \), the new tunnelling contribution \( H_W \) and the photon coupling term \( H_V \) after the gauge transformation \( \omega \). As usual, we apply the transformation \( \omega \) and in order to access the Toulouse point we again set the Coulomb coupling to \( \lambda_C = 2\pi \). In the language of the new fermions defined by \( \nu \) we obtain then the following Hamiltonian (\( \Delta_{1,2} \) are assumed to be shifted by \( \pm \Omega/2 \)):

\[
H = H_0[\psi_\pm] + \sum_{i=1,2} \Delta_i d_i^\dagger d_i + \left[ \gamma_L d_i^\dagger \psi_-(0) + \gamma_R d_2^\dagger \psi_+(0) + W d_1^\dagger d_2 + \text{h.c.} \right].
\]

We proceed in the spirit of Section 11 and derive the equations of motion for the participating operators,

\[
(i\partial_t - \Delta_1) d_1 = W d_2 + \gamma_L \psi_- (0),
\]

\[
(i\partial_t - \Delta_2) d_2 = W d_1 + \gamma_R \psi_+ (0),
\]

\[
W_{\psi_1} \psi_2 = \gamma_L d_1 + \gamma_R \psi_+(0),
\]

where \( \Delta_1 \) is again positive (negative) infinitesimal. Acting with \( Y[\gamma] \) [see Eq. 29] on both sides of the third of Eqs. 59 we then use the first two ones in order to eliminate the dot operators. As a result of this procedure we obtain an equation containing only \( \psi_- \) operators,

\[
i|Y|^2 \psi_2 (0^+) - \psi_- (0^-)
\]

\[
= \gamma \gamma_R Y W \psi_1 (0) + \gamma_L (i\partial_t - \Delta_2) \psi_-(0)] + \gamma_R Y W \gamma_R \psi_+ (0) + \gamma_R \partial_t - \Delta_2 \psi_- (0).\]

At this stage we again can make use of the decomposition \( \delta \) and reduce the relation 60 to exactly the form given in 59 with the same \( E \) [see definition 10] but different \( \gamma \) and \( \beta_\pm \),

\[
\gamma = W \sqrt{\omega} \alpha_R \alpha_L (\omega^2 + \Delta_1 \Delta_2 - W^2),
\]

\[
\beta_+ = \left\{ \alpha_R (\omega + \Delta_1) (\omega + \Delta_2) - W^2 (\omega - \Delta_1) - \alpha_L (\omega - \Delta_1) (\omega - \Delta_2) - W^2 (\omega + \Delta_2) \right\}/2,
\]

\[
\beta_- (\omega) = \beta_+ (\omega),
\]

where we again normalised all energy variables to \( \Gamma \) and defined \( \alpha_{R,L} = \gamma_{R,L}/\Gamma \). With these conventions the total dot transmission coefficient is still given by the formula 28 and the corresponding \( I - V \) by Eq. 14.

In the case of a weak dot-leads coupling and the symmetric level configuration (\( \Delta_{1,2} = \pm \) const), the ratchet current does not show any significant change in comparison to the non-interacting case, see Fig. 9. The maximum emerges at low energies, see Fig. 8. This effect occurs only in the interacting system. More interesting features arise in a system with weak optical coupling, see Fig. 11. For positive \( \Omega \) (which in our picture corresponds to a situation favouring the ratchet effect) the distinguished peak at \( \Delta_1 - \Delta_2 \) which exists at \( \Gamma \ll W \) splits in two (which is actually a more relevant parameter range from the experimental point of view). This effect does not occur in the non-interacting situation. The origin of the total four peaks can be traced back to the presence of the LL zero-bias anomaly in vicinity of the Fermi energy. Concentrating only on the left half of the system – the left electrode with the level \( \Delta_1 \), one can show in the lowest order in tunnelling that the correction to the level spectral function (which at \( \gamma_L = 0 \) is a delta function) is given by 39,

\[
\delta A(\omega) \sim \frac{\nu(\omega)}{\Gamma} \frac{\nu(\omega)}{\Gamma},
\]

where \( \nu(\omega) \sim |\omega/\omega_c|^{1/g-1} \) is the DOS of a half-open LL with a bandwidth \( \omega_c \). At \( g = 1/2 \) \( \delta A(\omega) \) possesses two maxima at \( \pm \Delta_1 \). This basic structure persists in all orders of \( \gamma_L \) with some corrections to the maxima positions. Exactly the same thing happens in the right part of the system thus accounting for the total of four peaks seen in the ratchet current.

V. SUMMARY

To conclude, we presented exact solutions of the single- and two-state non-interacting QDs coupled to interacting (and non-interacting) electrodes. In both situations the corresponding Hamilton operators can be brought to a quadratic form, and thus solved exactly, at the special interaction strength \( g = 1/2 \) (the Toulouse point).

FIG. 8: The same plot as in Fig. 7 for the symmetric interacting system for different lead-dot coupling strengths.
In the limit of small bias. In the asymmetric case (zero in the
voltages and unity in the asymmetric case (zero in the
interacting value at high
port signatures as long as the couplings are symmetric
expected, the system turns out to show resonant trans-
applying the scattering formalism in this situation. As
This additional feature does not destroy the solubility of
in the case when each of them is coupled to both leads.
light on this, we considered a model of a two-level QD,
the zero frequency noise properties we discussed the details of the voltage
behaviour of the universal Fano factor \( \nu_V \). It turns out
to interpolate between the non-interacting value at high
voltage and unity in the asymmetric case (zero in the
symmetric case) in the limit of small bias. In the asymmetric
symmetry \( \nu_V \) possesses a minimum at some \( V^* \), which
is absent in non-interacting systems.
In the case of many levels on the dot the question of the conductance properties in the valley between the re-
on–bias anomaly in the DOS of the interacting systems.
Spectacular effects make the ratchet current measure-
ments an invaluable instrument for studying interacting
structures. In addition we derived an exact relation
between the absorption (emission) spectra and frequency
dependent noise power spectrum. It has important impli-
cations in the luminescence accompanying field emission,
which is believed to occur during cold electron emission
from carbon nanotubes.
The key quantity, which generates an energy scale at
which most of the predicted effects take place, is the
lead–dot coupling \( \Gamma \). In the typical experiments made
on semiconducting QDs \( \Gamma \) ranges between 0.1 – 1 \( \mu \)eV,
which corresponds to temperatures around 1 – 10 mK
10. In the most current experiments conducted on con-
tacted molecules the coupling strength is expected to be
even smaller \( 2, 3 \). For the results of Sections III and II B
to be accessible in the experiments it is not necessary to
go significantly below the temperatures \( T \sim \Gamma \). On the
contrary, the experimental observability of all other pre-
dicted phenomena, and especially of the ratchet effects,
depends crucially on the ability to either lower the tem-
peratures beyond the \( T \sim \Gamma \) mark or build devices with
high enough \( \Gamma \) 10.

Acknowledgments

The authors would like to thank H. Grabert for many valuable discussions. This work was supported
by the Landesstiftung Baden–Württemberg gGmbH (Germany), by the EC network DIENOW, and the EPSRC
of the UK under grants GR/N19359 and GR/R70309.

VI. APPENDIX A

The result of integration in 157 is
\[ I(V) = G_0 \frac{2\Gamma_R \Gamma_L W^2}{\pi} \]
\[
\times \frac{\sum_{i=1,2(L,R)} \Gamma_i ((\Delta_1 - \Delta_2)^2 - (-1)^i(\Gamma_R^2 - \Gamma_L^2)) \sum_{p=\pm} \tan^{-1} \left( \frac{\sqrt{\frac{V^2 + p\Delta}{\Gamma_i^2}}}{\Gamma_i} \right) + \Gamma_R \Gamma_L \sum_{j=1,2(L,R)} p\Delta_j \ln(\Gamma_j^2 + (\Delta_j - p(-1)^jV/2)^2)}{\Gamma_R \Gamma_L ((\Delta_1 - \Delta_2)^2 + 2(\Gamma_R^2 + \Gamma_L^2)(\Delta_1 - \Delta_2)^2 + (\Gamma_R^2 - \Gamma_L^2)^2)}
\]

[1] M. A. Reed, C. Zhou, C. J. Muller, T. P. Burgin, and J. M. Tour, Science 278, 252 (1997).
[2] H. Park, J. Park, A. K. L. Lim, et al., Nature 407, 57 (2000).
[3] J. Reichert, R. Ochs, D. Beckmann, H. B. Weber, and H. v. Loecheneyen, Phys. Rev. Lett. 88, 176804 (2002).
[4] M. Bockrath, D. H. Cobden, J. Lu, A. G. Rinzler, R. E. M. Blanter, and M. Böttiker, Phys. Rep. 336, 151 (1997).
[5] P. Avouris, Chem. Phys. 281, 429 (2002).
[6] C. Dekker, Phys. Today 52, 22 (1999).
[7] A. O. Gogolin, A. A. Nersesyan, and A. M. Tsvelik, Bosonization and Strongly Correlated Systems (Cambridge University Press, 1998).
[8] R. Egger and A. O. Gogolin, Phys. Rev. Lett. 79, 5082 (1997).
[9] C. Kane, L. Balents, and M. P. A. Fisher, Phys. Rev. Lett. 79, 5086 (1997).
[10] M. Bockrath, D. H. Cobden, J. Lu, A. G. Rinzler, R. E. Smalley, L. Balents, and P. L. McEuen, Nature 397, 598 (1999).
[11] H. Schoeller, in Mesoscopic Electron Transport, edited by L.L. Sohn, L.P. Kouwenhoven, and G. Schön (Kluwer, Dordrecht, 1997).
[12] A. Komnik and A. O. Gogolin, Phys. Rev. Lett. 90, 246403 (2003).
[13] Yu. V. Nazarov and L. I. Glazman, cond-mat/0209090 (2002).
[14] C. L. Kane and M. P. A. Fisher, Phys. Rev. B 46, 15233 (1992).
[15] A. Furusaki, Phys. Rev. B 57, 7141 (1998).
[16] D. G. Polyakov and I. V. Gornyi, cond-mat/0212355 (2002).
[17] S. Huegle and R. Egger, cond-mat/0304158 (2003).
[18] L. Kouwenhoven and L. I. Glazman, Phys. World 14, 33 (2001).
[19] A. Komnik and A. O. Gogolin, cond-mat/0209090 (2002).
[20] Yu. V. Nazarov and L. I. Glazman, cond-mat/0209090 (2002).
[21] C. L. Kane and M. P. A. Fisher, Phys. Rev. B 46, 15233 (1992).
[22] A. Furusaki, Phys. Rev. B 57, 7141 (1998).
[23] D. G. Polyakov and I. V. Gornyi, cond-mat/0212355 (2002).
[24] S. Huegle and R. Egger, cond-mat/0304158 (2003).
[25] L. Kouwenhoven and L. I. Glazman, Phys. World 14, 33 (2001).
[26] A. Komnik and A. O. Gogolin, Phys. Rev. Lett. 90, 246403 (2003).
[27] Yu. V. Nazarov and L. I. Glazman, cond-mat/0209090 (2002).
[28] C. L. Kane and M. P. A. Fisher, Phys. Rev. B 46, 15233 (1992).
[29] A. Schiller and S. Hershfield, Phys. Rev. B 58, 14978 (1998).
[30] P. Fendley, A. Ludwig, and H. Saleur, Phys. Rev. Lett. 75, 2196 (1995).
[31] U. Weiss, Sol. State Comm. 100, 281 (1996).
[32] K. A. Matveev, Phys. Rev. B 51, 1743 (1995).
[33] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. 68, 2512 (1992).
[34] T. Martin and R. Landauer, Phys. Rev. B 45, 1742 (1992).
[35] L. Y. Chen and C. S. Ting, Phys. Rev. B 43, 4534 (1990).
[36] A. Braggio, R. Fazio, and M. Sassetti, cond-mat/0304329 (2003).
[37] Y. M. Blanter and M. Büttiker, Phys. Rep. 336, 1 (2000).
[38] L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1515 (1964).
[39] E. M. Lifshits and L. P. Pitaevskii, Physical Kinetics (Pergamon Press, Oxford, 1981), we are using the notation of this book.
[40] A. Komnik and A. O. Gogolin, Phys. Rev. B 66, 125106 (2002).
[41] J. W. Gadzuk and E. W. Plummer, Rev. Mod. Phys. 45, 487 (1973).
[42] A. Komnik and A. O. Gogolin, Phys. Rev. B 66, 035407 (2002).
[43] J.-M. Bonard, T. Stöckli, F. Maier, W. A. de Heer, A. Chatelain, J.-P. Salvetat, and L. Forró, Phys. Rev. Lett. 81, 1441 (1998).
[44] A. Furusaki and K. A. Matveev, Phys. Rev. Lett. 88, 226404 (2002).
[45] U. Lundin and R. H. McKenzie, Phys. Rev. B 66, 075303 (2002).
[46] D. Goldhaber-Gordon, H. Shtrikman, D. Mahalu, D. Abush-Magder, U. Meirav, and M. A. Kastner, Nature 391, 156 (1998).
[47] S. M. Cronenwett, T. H. Oosterkamp, and L. P. Kouwenhoven, Nature 281, 540 (1998).
[48] Strictly speaking \( \psi(x=0) = 0 \), so we assume that the tunnelling takes place at the second last site of the corresponding lattice model, at \( x = \pm a_0 \). Also, we ignore the Klein factors as they can be absorbed into Eqs. disappearing from the analysis.
[49] If one considers the levels \( \Delta_{1,2} \) to be the molecular orbitals after the contacting process there is no need to include the tunnelling. However, we assume the dot level wave functions to be those of a free molecule prior to contacting. In that case there could be some finite overlap, and hence tunnelling, between them.
[50] Higher \( \Gamma \) would automatically lead to higher Kondo temperatures, which in QD can be as high as several \( K_1 \). The Kondo physics would then obscure all predicted effects. Therefore it is essential to keep the system spin-polarised as assumed throughout the paper.