Time-dependent electron transport through a strongly correlated quantum dot: multiple-probe open-boundary conditions approach

A Pertsova, M Stamenova and S Sanvito

School of Physics and CRANN, Trinity College Dublin, Dublin 2, Ireland

E-mail: pertsova@tcd.ie

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Abstract

We present a time-dependent study of electron transport through a strongly correlated quantum dot, which combines adiabatic lattice density functional theory in the Bethe ansatz local-density approximation (BALDA) to the Hubbard model, with the multiple-probe battery method for open-boundary simulations in the time domain. In agreement with the recently proposed dynamical picture of Coulomb blockade, a characteristic driven regime, defined by regular current oscillations, is demonstrated for a certain range of bias voltages. We further investigate the effects of systematically improving the approximation for the electron–electron interaction at the dot site (going from non-interacting, through Hartree-only to adiabatic BALDA) on the transmission spectrum and the $I$–$V$ characteristics. In particular, a negative differential conductance is obtained at large bias voltages and large Coulomb interaction strengths. This is attributed to the combined effect of the electron–electron interaction at the dot and the finite bandwidth of the electrodes.

(Some figures may appear in colour only in the online journal)

1. Introduction

Electron transport through nanoscale devices is a diverse subject, which is currently the focus of extensive experimental and theoretical research. The fuel of such interest is the expectation that nanoscale objects, such as quantum dots [1] and even single molecules [2], are to become active components in novel electronic devices, which potentially offer unique advantages over existing technologies [3]. At the fundamental level, the physics of such reduced-dimensional systems is dominated by quantum effects. Among them are electron correlations, which strongly affect the electron transport at this level of confinement, giving rise to prototypical quantum phenomena, such as Coulomb blockade (CB) [4, 5] and the Kondo effect [6–8].

While the Landauer formula is the solution to the non-interacting quantum transport problem [9], the interacting case continues to be challenging to the theory. The latter is typically approached with the non-equilibrium Green’s function (NEGF) formalism [10], which allows, in principle, the derivation of an interacting many-body Landauer-type formula for the steady-state current in the case where interaction is limited to a finite region in space [11]. In practice, for the majority of the state-of-the-art $ab$ $initio$ transport calculations and numerical algorithms [12–15], the method of choice for the electronic structure description is the density functional theory (DFT). However, typical steady-state DFT + NEGF transport schemes have a range of limitations, both conceptual and technical [16].

At the fundamental level it has been recently demonstrated, at least for the case of a single Anderson impurity model, that the linear-response conductance calculated from the Kohn–Sham levels for the exact exchange–correlation (XC) functional reproduces closely that computed with
many-body approaches [17–19]. If the same holds true for \textit{ab initio} DFT, then the DFT + NEGF scheme will provide a complete solution for the zero-bias limit. Still, on the practical side, the commonly used approximations to the XC functional, lacking the so-important derivative discontinuity [20], fail to capture essential physics for the transport in molecular junctions, qualitatively mispredicting the conduction regime [21, 22]. Different is the situation at finite bias, where, let alone the implementation, conceptual concerns reflect on the very applicability of a ground-state electronic structure theory to an intrinsically non-equilibrium problem especially if electron correlations are significant [23, 24].

One strategy to avoid some of the shortcomings of using equilibrium DFT has been sought in its natural extension, time-dependent DFT (TDDFT) [25], and practical schemes for time-dependent transport having been already developed [16]. In general, real-time dependent schemes for quantum transport can be roughly divided into two categories based on their assumption for the initial conditions. In one case, the so-called partitioning approach, the electrodes are prepared in equilibrium with the poles of a battery, but not yet connected to the nanoscopic device [26]. The current then starts to flow when the connection is made. In the other, partition-free approach, the system electrodes plus device is initially at equilibrium and subsequently an electric field is applied to the electrodes [27]. The former assumption, where two initial electrochemical potentials are well defined, is more in the spirit of the Landauer transport picture. The latter is instead more DFT-friendly, as the starting point is the ground state of the system [16].

There has been evidence that these two time-dependent transport variants agree in the non-interacting case, i.e. they lead to the same history-independent steady-state current [27, 28]. More recently, the analysis of the two different time-dependent transport settings has been extended to interacting systems in certain limits [29]. In the work by Kurth et al [30] the partition-free approach combined with TDDFT, further equipped with a novel XC functional carrying the physical derivative discontinuity of the DFT potential, has been applied to study the transport through a quantum dot in the CB regime. In particular that work has put forward an important novel description of CB as a dynamical process with rapidly oscillating local currents. This is a regime clearly inaccessible by conventional steady-state transport models.

In this work we adopt another recently proposed time-dependent transport scheme, the so-called, multiple-probe battery (MPB) method [31, 32], to study electron transport through a strongly correlated quantum dot. The MPB scheme was first proposed in the context of correlated electron–ion dynamics and was applied to a wide range of problems, such as current-induced heating in atomic wires [31, 33]. This method belongs to the first of the fore-mentioned categories and enables the realization of an external battery within the finite system of electrodes + device. The external bias is introduced through the difference in the electrochemical potentials of the set of reservoirs, or \textit{probes}, attached individually to each atom in a pair of large but finite metallic electrodes (leads). The scheme is very tractable computationally and has the control knobs to be an arbitrarily close approximation to the non-interacting Landauer transport in the long-time dc limit.

The MPB time-propagation scheme is based on the integration of the Liouville–von Neumann equation of motion for the reduced density matrix of the system, in which the open boundaries are described explicitly by a source and a drain term. For the time-dependent Hamiltonian of the quantum dot, entering the equation of motion, we adopt the description used by Kurth et al [30]. This is based on the adiabatic Bethe ansatz local-density approximation [34] (adiabatic BALDA, or ABALDA) to the XC functional, which exhibits a derivative discontinuity at half-filling.

By investigating the real-time evolution of the current through the quantum dot, we find an agreement with the work contained in [30], i.e. for a certain set of parameters the system does not reach a steady-state but rather remains in a dynamical situation, characterized by oscillations in the current. We note that this non-trivial result, first obtained in the original work by Kurth et al [30], represents an important development in time-dependent quantum transport theory [25]. The question whether this result is universal or is an artifact of the approximations made to the XC functional has in fact stimulated much debate in subsequent literature, with the most recent studies suggesting that approximations beyond ABALDA may restore the tendency to the steady-state [29, 35–37]. We would like to emphasize here that developments beyond ABALDA are outside the scope of the present work and our description of the electronic structure is identical to the one used in [30]. Therefore, in terms of time-dependent transport, the main purpose of our study is to test the stability of the result of Kurth et al [30] against the variation of the time-dependent transport scheme. We present a detailed numerical study of the dynamical CB regime using the MPB method and report on the key similarities and differences to the previous result.

Furthermore we propose an interpretation of the time-dependent results in terms of the more familiar steady-state picture of transport. In particular, we construct the current–voltage, \( I–V \), characteristics of the quantum dot from the long-time average of the current obtained from the time-dependent simulations. This is done for a wide range of parameters, even in the cases when a steady-state is not achieved. Importantly, we observe a drop of the current as a function of the source–drain voltage and, as a consequence, a negative differential conductance above a critical bias voltage. We demonstrate that such an effect is not possible if the derivative discontinuity is not included in the one-particle potential.

This is particularly interesting in the view of some recent contrasting results. On the one hand, a number of studies, based on several distinct many-body approaches [38–40], attribute the negative differential conductance mainly to electron–electron interaction. On the other hand, it has been demonstrated by Băldea and Koppel [41] that in the case of an exactly solvable model for a non-interacting dot within the steady-state formalism, the finite bandwidth
of the electrodes can alone lead to pronounced negative differential conductance for a wide range of parameters. Here we find a numerical proof that this result can be generalized to the interacting case and time-dependent transport. Our calculations suggest, however, that for the system considered here, the current drop is due to a combination of two effects, namely electron–electron interaction on the dot and the finite bandwidth of the electrodes.

Our paper is organized as follows. In section 2 we introduce the model system and our theoretical framework, i.e. the Hamiltonian and the computational scheme for MPB quantum transport. In the first part of section 3 the $I$–$V$ characteristics of a non-interacting quantum dot calculated by using the TD MPB method are compared to analytic NEGF results. We then discuss the finite electrode bandwidth as a source of the negative differential conductance. In the second part of section 3, we present the time-dependent results for a strongly correlated dot in the CB regime. Finally, we propose an explanation for the observed current drop in the $I$–$V$ characteristics.

2. Methods

The model system considered in this work is presented in figure 1. This consists of a central region, which contains the quantum dot surrounded by two $N_L$-site long atomic chains at both sides, and two one-dimensional finite leads, each counting $N_{LR}$ atoms. The physics of the quantum dot connected to two leads is described by the Anderson impurity model [42, 11]. The Hamiltonian of the total system thus reads

$$\hat{H}_S = \sum_{\alpha=L,R} \hat{H}_\alpha + \hat{H}_T + \hat{H}_{QD}. \tag{1}$$

Here the first term is the nearest-neighbors single-orbital tight-binding (TB) Hamiltonian describing respectively the left-hand side ($\alpha = L$) and right-hand side ($\alpha = R$) lead. This is written as

$$\hat{H}_\alpha = \sum_{i,\sigma} \varepsilon_{ia} \hat{c}_{ia}^{\dagger} \hat{c}_{ia} + \sum_{i,\sigma} \gamma_0 \left( \hat{c}_{ia}^{\dagger} \hat{c}_{i+1a}^{\dagger} + h.c. \right), \tag{2}$$

where $\varepsilon_{ia}$ are the on-site energies and $\gamma_0$ is the hopping integral; $\hat{c}_{ia}^{\dagger}$ ($\hat{c}_{ia}$) is the creation (annihilation) operator for an electron with spin $\sigma$ at the atomic site $i$ of the lead $\alpha$ (the index $i = 1, \ldots, N_\alpha$ runs from left to right for $\alpha = R$ and from right to left for $\alpha = L$). Note that two atomic chains on each side of the quantum dot are also described by a TB model with the hopping integral $\gamma_0$ and therefore they are included in the Hamiltonian of the leads.

The second term in equation (1) describes the tunneling between the quantum dot and the two adjacent sites and it is given by

$$\hat{H}_T = \sum_{\gamma} \gamma_\gamma \left( \hat{c}_{0}^{\dagger} \hat{c}_{\gamma L}^{\sigma} + \hat{c}_{0}^{\dagger} \hat{c}_{\gamma R}^{\sigma} + h.c. \right), \tag{3}$$

where $\hat{c}_{0}^{\dagger}$ ($\hat{c}_{0}$) is the creation (annihilation) operator for an electron with spin $\sigma$ on the dot and $\gamma_\gamma$ is the hopping integral between the dot and site $i = 1$ in the lead $\alpha$.

Finally, the Hamiltonian of the quantum dot reads

$$\hat{H}_{QD} = \sum_\sigma V_g \hat{n}_0^\sigma + U \hat{n}_0^{\uparrow}\hat{n}_0^{\downarrow}, \tag{4}$$

where $V_g$ is the on-site energy of the dot, which acts as a local gate voltage; $U$ ($U \geq 0$) is the charging energy, which expresses the strength of the Coulomb repulsion on the dot; $\hat{n}_0^\sigma = \hat{c}_0^{\dagger} \hat{c}_0^\sigma$ is the site-occupation operator.

Within the lattice DFT framework [43] the many-body Hamiltonian in equation (4) is mapped onto an effective single-particle Kohn–Sham Hamiltonian which, in the local-density approximation, reads

$$\hat{H}_0 = \sum_\sigma v_{KS} [n_0] \hat{n}_0^\sigma. \tag{5}$$

Here $n_0$ is the charge density of the dot and $v_{KS}$ is the effective Kohn–Sham potential, which can be written as a sum of three terms

$$v_{KS} [n_0] = V_g + \frac{n_0}{2} U + v_{XC} [n_0]. \tag{6}$$

The second and third terms are respectively the Hartree and the XC potential. The latter is approximated by a modified BALDA potential, specifically tailored to a nonuniform configuration with a weakly coupled dot. Such potential has been introduced in [30] and it is essentially an extension to the Anderson impurity model of the original local-density approximation (LDA) exchange–correlation functional, derived by Lima et al [34] from the exact Bethe ansatz (BA) solution of the one-dimensional Hubbard model. Following [30], we define the BALDA potential for the dot as

$$v_{XC} [n_0] = \theta (1 - n_0) v_{XC}^{(\gamma)} [n_0] - \theta (n_0 - 1) v_{XC}^{(\gamma)} [2 - n_0]. \tag{7}$$

where $\theta (n_0)$ is a Heaviside step function and

$$v_{XC}^{(\gamma)} [n_0] = -\frac{U n_0}{2} - 2 \gamma_\gamma \left[ -\cos \left( \frac{\pi n_0}{2} \right) + \cos \left( \frac{\pi n_0}{\beta} \right) \right]. \tag{8}$$

Here the parameter $\beta$ is determined for each $U$ by solving the transcendental equation [34]. Notably, such $v_{XC}$ exhibits a derivative discontinuity at $n_0 = 1$, i.e. at the phase transition of the 1D Hubbard model. In practice, however, we use a continuous approximation to the BALDA potential [30] where the true discontinuity, expressed through a Heaviside step function $\theta (n_0)$ (see equation (7)), is replaced by a function

![Figure 1](https://example.com/figure1.png)
Hamiltonian for the quantum dot, we use the adiabatic through the instantaneous charge density of the dot clarity.

\[ v = \text{or the Hartree approximation, where the potential on the dot potential of the dot is simply given by} \]

\[ U = \frac{v}{2} + \frac{v_{\text{XC}}[n_0(t)]}{2}, \quad \text{(9)} \]

Here \( n_0(t) \) is the charge density of the dot at time \( t \) and \( v_{\text{XC}}[n_0(t)] \) is evaluated at each time \( t \) using equation (7) with the charge density \( n_0 \) substituted by its instantaneous value \( n_0(t) \) at time \( t \). Such approximation to the XC potential is referred to as adiabatic BALDA (ABALDA).

Whether such adiabatic local approximation to the description of non-equilibrium transport in strongly correlated systems is appropriate, is a question which has been addressed in a number of recent works [29, 35–37]. In particular, a comparative study between the TDDFT approach with ABALDA (TDDFT + ABALDA) and many-body perturbation theory, applied to out-of-equilibrium Anderson impurity model, has been carried out by Uimonen et al [35]. The results obtained with both approaches have been tested against numerically exact results produced by time-dependent density matrix renormalization group theory. It was found that in general the TDDFT + ABALDA approach is in good qualitative agreement with many-body perturbation theory over a wide range of parameters. However, in many cases it overestimates the steady-state currents. This problem was linked to the shortcomings of the local approximation to the XC functional, in particular to the fact that the XC potential vanishes inside the electrodes. Moreover, it was demonstrated by Khosravi et al [36] that the inclusion of dynamical correlations, or memory effects, might eliminate the multistability in the density and the current, which can be found within the TDDFT + ABALDA approach. Possible implications of these results on time-dependent transport though a strongly correlated dot in CB regime will be further discussed in section 3.2.1.

We now discuss, following the work of Todorov and co-workers [31, 32], how the open-boundary conditions are introduced in the MPB setup. In the MPB method, each atom \( i \) of the leads (with the exception of the \( N_d \) atoms at both sides of the quantum dot) is connected to an external probe \( P_i \) (see figure 1). All the probes attached to the sites in the left (right) lead are kept at the electrochemical potential \( \mu_L = \mu_R \) (here \( V_{sd} \) is in units of eV). For symmetrically applied bias \( \mu_L = \epsilon_F + V_{sd}/2 \) and \( \mu_R = \epsilon_F - V_{sd}/2 \), where \( \epsilon_F \) is the Fermi level of the electrodes (assumed identical). The time-dependent equation of motion for the density matrix of the system coupled to the probes reads

\[ i\hbar \dot{\rho}_S(t) = [\hat{H}_S(t), \rho_S(t)] + \hat{\Sigma}^+ \dot{\rho}_S(t) - \dot{\rho}_S(t) \hat{\Sigma}^{-} + \int_{-\infty}^{\infty} \left[ \hat{\Sigma}^{-}(E) \hat{G}_{KS}^{-}(E) - \hat{G}_{KS}^{+(E)} \hat{\Sigma}^{-}(E) \right] dE. \]

The last two terms on the right-hand side are extraction (drain) and injection (source) terms, respectively; \( \hat{G}^{+} \) (\( \hat{G}^{-} \)) is the retarded (advanced) Green’s function of the system and it is given by

\[ \hat{G}^{\pm} = \left( E \hat{I}_S - \hat{H}_S - \hat{\Sigma}^\pm \pm i \hat{\Sigma} \right)^{-1}, \]

where \( \hat{H}_S = \sum_{\sigma=L,R} \hat{H}_\alpha + \hat{H}_T + \sum_{\sigma} \hat{V}_g n_0^\sigma \) is the time-independent part of \( \hat{H}_S(t) \) and \( \Delta \) is a dephasing factor (see later for an exact definition). The self-energies due to the presence of the external probes and the in-scattering self-energy are written as

\[ \hat{\Sigma}^\pm = \mp i \frac{\Gamma}{2} \hat{I}_L \mp i \frac{\Gamma}{2} \hat{I}_R, \]

\[ \hat{\Sigma}^{-} = \frac{\Gamma}{2\pi} f_{\text{L}}(E) \hat{I}_L + \frac{\Gamma}{2\pi} f_{\text{R}}(E) \hat{I}_R, \]

with the broadening \( \Gamma \) defined as \( \Gamma = 2\pi \gamma^2 d \), where \( \gamma_p \) is the coupling to the probes, assumed to be identical for all sites in the leads, and \( d \) is an energy-independent constant, which represents the surface density of states of the probes within the wide-band limit; \( \hat{I}_M \) is the identity operator in region \( M (M = L, R, S) \).

Equation (10) is derived from a general Liouville–von Neumann equation for the total density matrix of the system and the probes combined. It incorporates two main approximations: (i) the wide-band limit in the probes and (ii) the decoherence in the injection process, introduced through the relaxation time \( \tau_\Delta \), with \( \Delta = \hbar/\tau_\Delta \) (see equation (11)). The second approximation essentially decouples, over the time interval \( \tau_\Delta \), the injection of electrons from the probes into the leads and their subsequent scattering from the time-dependent potential inside the central region, provided that the latter is long enough. In other words the dephasing factor imposes a restriction on the size of the central region (2\( N_d \) + 1 sites). Therefore the inclusion of \( N_d \) buffer sites on both sides of the dot is essential within the time-dependent formalism.

The value of \( \Delta \) is determined in such way that the distance traveled by the electrons during the time interval \( \tau_\Delta \) is smaller than the distance between the electrodes and the interior of the central region, i.e. the quantum dot. This condition can be written as \( v_e \tau_\Delta < N_d a \), where \( v_e \) is the electron group velocity and \( a \) the lattice constant (\( a = 1 \)). In practical terms, the introduction of the dephasing factor allows one to write down the injection term, which is in general non-local in time, in a rather simple time-independent form (see equation (10)). This, however, also introduces an additional broadening, proportional to \( \Delta \), in the steady-state \( I-V \) characteristics, which is absent in the standard static
where $E_0$ is the self-energy due to the presence of electrodes, with $E_0^\mathrm{R}$ and $E_0^\mathrm{L}$ representing, respectively, the real and imaginary part of the total energy of an isolated system (not coupled to the probes), constructed from the eigenstates of the Hamiltonian $H_0$. The open-boundary terms are switched on over a short time interval of 5 fs and maintained throughout the simulation. The current through the dot is then calculated as a bond current between the dot and the adjacent site [45]. The typical parameters of the MPB setup used in our simulations, unless specified otherwise, are $N_{L,R} = 90$ and $N_d = 20$. We have tested that further increasing the size of the system does not lead to significant difference in the $I$–$V$ characteristics. In order to have one free parameter instead of two, we use the condition $\Delta = \Gamma/2$, which has been discussed in detail in [31], and $\Gamma = 0.35$ eV in our simulations.

3. Results

3.1. Non-interacting case

As a test of the applicability of the TD MPB method we first examine the non-interacting case ($U = 0$). For this situation, we directly compare the $I$–$V$ characteristics obtained from the time-dependent simulations to the ones calculated by using the standard NEGF-based Landauer solution, which we refer to as exact NEGF [10]. The comparison is presented in figure 2, where the current is plotted as a function of the source–drain voltage for the non-interacting level aligned with the Fermi level in the leads ($V_g = 0$). In the case of the TD MPB approach, the value for the steady-state current is obtained from the time-dependent simulation for the corresponding value of $V_{sd}$ after the steady-state has been established, i.e. when the variation of the current with time becomes negligible. In the case of the exact NEGF method, we use the well-known analytical expression for the non-equilibrium current through a non-interacting resonant level coupled to two semi-infinite electrodes [10, 41]

$$ I_{\mathrm{EN}} = \frac{2e}{h} \int dE \frac{\Gamma_L(E)\Gamma_R(E)}{[E - V_g - \Delta(E)]^2 + [\Gamma(E)/2]^2} \times [\tilde{f}_L(E) - \tilde{f}_R(E)]. $$

(14)

Here $\Delta(E) = \Delta_L(E) + \Delta_R(E)$ and $\Gamma(E) = \Gamma_L(E) + \Gamma_R(E)$ represent, respectively, the real and imaginary part of the total self-energy due to the presence of electrodes, with $\Gamma_L(R)$ and $\Gamma_L(R)$ given by

$$ \Delta_L(R)(E) = \frac{\gamma_R^2}{\gamma_0^2} E_{L(R)}, $$

(15)

$$ \Gamma_L(R)(E) = \frac{\gamma_R^2}{\gamma_0^2} \theta(2\gamma_0 - |E_{L(R)}|) \sqrt{4\gamma_0^2 - E^2_{L(R)}}, $$

(16)

where $E_{\alpha} = E - \varepsilon_{\alpha}$, $\varepsilon_{\alpha}$ being the on-site energy in the lead ($\alpha = L, R$).

We consider two possible limits for the on-site energies in the electrodes: (i) the highly conducting regime with $\varepsilon_{\alpha} = 0$ for all atoms in $\alpha = L, R$ and (ii) the weakly conducting regime for which the on-site energies in L(R) are shifted in accordance with the respective electrochemical potential, $\varepsilon_{L(R)} = \pm V_{sd}/2$. As expected, the difference between the $I$–$V$ curves calculated in these two limits becomes significant at large bias, since the transmission in case (ii) rapidly drops to zero once the bias voltage exceeds the bandwidth of the leads ($4|\gamma_0|$). This high-bias negative differential conductance, stemming entirely from the finite electrode bandwidth, is a well-understood feature of steady-state transport in low-dimensional yet uncorrelated electron systems [41]. We also note that the low-bias agreement between the two transport limits can, in principle, be extended to arbitrarily high biases $V_{sd}$ by increasing $\gamma_0 > V_{sd}/4$.

An encouraging result is that for both the transport limits the TD MPB method reconstructs rather well the exact NEGF $I$–$V$. The agreement is particularly good in the highly conducting limit. The smearing of the abrupt $I$–$V$ features at low bias and again the drop of the current at $V_{sd} \lesssim 4\gamma_0$ for the weakly conducting limit are inherent to the TD MPB method [31]. These are due to the explicit dephasing factor, which simplifies the equation of motion for the density matrix by eliminating temporal non-localities of the injection.

In order to eliminate the drop in the current at large bias voltages and to focus on the electron interaction at the quantum dot, we will use the $\varepsilon_{L(R)} = 0$ limit in all the further

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1 We have established that if $|\gamma_0|$ is increased from 1 eV to 3.88 eV, the result for the current, obtained using two limits for the on-site energies of the leads, differ by at most 3% for $V_{sd} = 4$ eV and for $V_g$ between 0 and 1 eV.
calculations presented. In this case, the saturation current at high voltages is entirely determined by the position of the resonant level, set by the gate voltage $V_g$ (see the inset of figure 2), relatively to the electrodes band center. As the resonant level approaches the band-edge of the leads ($V_g \lesssim 2\gamma_0$), the saturation current decreases. In section 3.2.2 we will recognize the contribution of the latter effect to the drop in the current as a function of the source–drain voltage.

### 3.2. Interacting case

#### 3.2.1. Time-dependent transport.

While in the non-interacting case the time-dependent current through the dot always reaches the steady-state, in the case when electron–electron interaction is considered this is not guaranteed. In fact, it has been shown that for the quantum dot, described at the TDDFT level with the ABALDA XC functional, there exist a range of the source–drain voltages, for which the system is driven into a dynamical state, where the current, the density and the on-site potential oscillate without ever reaching a steady-state [30].

The question we address here is whether such dynamical state can be captured by the MPB method. Before proceeding to fully time-dependent simulations, we have carried out the calculation of the interacting steady-state transport using the steady-state version of the MPB approach. We have found that for a certain range of source–drain voltages the steady-state calculation does not converge due to the discontinuity of the Kohn–Sham potential at unitary occupation. This numerical observation is consistent with analytical calculation by Kurth et al [30], based on the analysis of the self-consistency condition for the steady-state density of the dot for different values of the source–drain voltage. The absence of a stationary solution in a certain region of the parameter space is a clear indication that the system is in a dynamical state. However, the details of this dynamical state are only accessible by time-dependent simulations. We therefore resort to the TD MPB method in order to determine the dynamical trajectories of the transport-related observables over a wide range of source–drain voltages.

The results of our calculations of time-dependent transport through the interacting dot are shown in figure 3. For all values of the source–drain voltage below a critical value $V_{sd}^{cr}$ a steady-state is achieved. However, for source–drain voltages above $V_{sd}^{cr}$, oscillations indeed develop in all transport-related quantities. As shown in figure 3 for this range of $V_{sd}$ the density quickly reaches a critical value of $n_0 = 1$. At the same time the first jump of the on-site potential occurs, followed by a series of almost rectangular pulses (see figure 3(b)). Due to the derivative discontinuity at $n_0 = 1$, the on-site potential reaches an oscillating regime, abruptly alternating in time between two values, one just below and the other just above the discontinuity. This translates into oscillations of the charge density around $n_0 = 1$ (see the inset in figure 3(a)) and also into oscillations in the current (figure 3(b)).

Below, we elaborate on the dynamical features observed for different values of $V_{sd}$. The height of the pulses in $v_{KS}(t)$ is equal to the height of the jump of $v_{KS}[n_0]$ at the derivative discontinuity and it is mainly governed by the value of the charging energy $U$. The width of the pulses increases with increasing $V_{sd}$. This essentially means that for larger $V_{sd}$ the system tends to stay longer in the state with a larger on-site potential, corresponding to the density above 1. Further increasing $V_{sd}$ will finally lead to a steady-state. The exact value of the threshold voltage, $V_{sd}^{cr}$, is difficult to determine since the on-site potential changes with time. From simple considerations, however, we established that $V_{sd}^{cr} \geq v_{KS}[n]$, where $n$ is a value of the charge density just below 1. For the set of parameters used here $V_{sd}^{cr} \approx 1.5$ eV.

As discussed by Kurth et al, the dynamical state of the quantum dot described above is a manifestation of dynamical Coulomb blockade. By applying a large enough source–drain voltage the dot can be charged. However, when the charge reaches the critical value $n_0 = 1$, the on-site potential immediately increases by an amount, determined by Coulomb repulsion $U$, thus preventing further charging. This essentially corresponds to the CB regime. In addition, the time-dependent simulations reveal that in this regime the quantum dot is alternating between two states, separated by an energy barrier determined by $U$. These two states correspond to the fluctuation of the charge on the dot around $n_0 = 1$, which originates from the fact that the ABALDA potential has a derivative discontinuity at $n_0 = 1$ but it is a smoothly varying function of $n_0$ away from this occupation.

It follows from the discussion that the dynamics of the quantum dot in the CB regime, calculated with the TD MPB method, is in a good agreement with the results reported in [30] both qualitatively and quantitatively.
have established numerically that the two different methods reproduce practically identical dynamical trajectories for all the observables in the long-time limit in the case of an interacting system. The remaining differences are limited to the early stage of the time-evolution. A characteristic feature of the on-site potential of the dot, observed in [30], is a transition period just after the start of the oscillations, where the series of rectangular pulses in the time-dependent $V_{KS}$ is preceded by a larger pulse whose width increases with $V_{sd}$. This characteristic transient pulse is not present in our calculations (see figure 3).

In order to establish to what extent the transient pulse is determined by the initial conditions, we performed time-dependent simulations for the same system as shown in figure 1 but without attaching the external probes, i.e. for a closed-boundary finite system. Instead, we applied the source–drain voltage as a rigid shift of the on-site energies in the left lead, i.e. a term $V_{sd} \sum_{i,\sigma} c_{i\alpha}^\dagger c_{i\alpha}^\sigma$ has been added to the Hamiltonian $\hat{H}_\alpha$ for $\alpha = L$ (see equation (2)) at the start of the time-dependent simulation. We used longer leads ($N_{L/R} = 220$) and limited the time of the simulations to 100 fs, which is sufficient to observe the time propagation before the reflections from the finite boundaries start to affect the dynamics. The time-dependence of the charge density, current and on-site potential, obtained from the closed-boundary simulation, is presented in figure 4. In contrast to our open-boundary simulations, we indeed observed qualitatively the same transient regime as in [30]. This is mainly characterized by an earlier onset of the CB oscillations for larger source–drain voltages and by the increase of the width of the first pulse in the time-dependence of the Kohn–Sham potential with increasing $V_{sd}$.

Finally, we would like to emphasize that the dynamical CB state, described in this section, originates directly from the shape of the XC correlation functional, which exhibits a derivative discontinuity at integer occupation [30]. As such this is an important result in the theory of TDDFT for quantum interacting transport. However, it should be noted that several recent works have investigated non-equilibrium transport in interacting nanojunctions using approaches that go beyond the local adiabatic approximation, for example by including dynamical correlations in the framework of the many-body perturbation theory [36, 37]. These studies suggest that the multistabilities and time-dependent solutions with XC effects may be a direct consequence of the common treatments of electron–electron interaction, such as the local adiabatic approximation within TDDFT. An even more general statement has been made by Moldoveanu et al. [29], who demonstrated, within a perturbative approach based on time-dependent scattering theory, that the steady-state regime for open interacting systems is always achieved. These recent evidences do not formally disprove the TDDFT results but rather should be considered as strong indications that non-local approximations to the XC functional both in space and time are necessary to capture all of the subtleties of time-dependent interacting transport within the TDDFT framework.

$3.2.2. \text{Steady-state transport.}$ In section 3.2.1 we demonstrated that, within a certain range of parameters, the derivative discontinuity prevents the quantum dot from evolving towards the steady-state. Outside this range, however, a steady-state is achievable. Here we determine the steady-state current through the dot for various gate voltages and map out the corresponding $I$–$V$ curves. For situations, where the dot is trapped in oscillations, we take as steady-state current its time-average in the long-time limit.

The linear-response conductance as a function of $V_g$ is depicted in figure 5. This is calculated as the finite-difference ratio $\Delta I_0/\Delta V_{sd}$ close to zero-bias (for a very low but finite bias $\Delta V_{sd} = 0.01$ eV) and represents an approximation to the zero-bias differential conductance. In the non-interacting case, the conductance is composed of a single peak centered around $V_g = 1.5$ eV, which corresponds to the Fermi level of the leads. This is expected from the steady-state picture of transport through a non-interacting resonant level. In principle the width of the resonance peak is given by the dot-lead hopping integral $\gamma_C$. In our TD MPB calculations, however, there is an additional resonance broadening factor ($\tau_\Delta$) related to the dephasing condition in the equations of motion. Its corresponding energy unit, $\Delta = h/\tau_\Delta$, can be associated to a fictitious temperature, smearing the electronic energy distributions in the leads [31]. As a result, a suppression of the transmission resonance proportional to $1/\Delta$ is also expected. This is the reason of why the amplitude of non-interacting resonance conductance in figure 5 is below the conductance quantum, $G_0 = 2e^2/h$.

In the interacting case and at finite temperatures, or temperatures higher than the Kondo temperature, $T_K$, the Anderson impurity model predicts two distinct Coulomb peaks [46] in the conductance as a function of the gate voltage. These are manifestation of charge quantization at the dot.
and correspond to each of the two integer electron number states, in which the dot is inhabited by one or two electrons, respectively [47]. However, at very low temperatures ($T < T_K$) the model predicts a conductance plateau, which is a signature of the Kondo effect for a singly-occupied dot [48]. Recently, the possibility of accurately calculating the linear-response conductance of the Anderson impurity model within the DFT framework has been addressed independently by several authors [17–19]. In particular, it has been shown that the static DFT + NEGF approach with a numerically exact XC functional [19] and a novel temperature-dependent XC functional [17], both exhibiting a derivative discontinuity, reproduce the conductance plateau at $T = 0$. Although in the present work we use a less accurate approximation to the XC functional (ABALDA), our results for the linear-response conductance, obtained directly from the time-dependent simulations, qualitatively agree with these findings. Note that, apart from the electron-phase relaxation in the reservoirs, which acts as a fictitious electronic temperature, our model is a zero-temperature one.

As one can see from figure 5, in the case of the full discontinuous potential ($\nu_{KS}$), there exist a special range of gate voltages, roughly between the position of the $U = 0$ resonance level, $V_{res} = E_F$, and the value $V_{res} - U$, for which we observe features in the conductance quite different from the Hartree-only case and the non-interacting ($U = 0$) case. The width of this region increases with increasing $U$ and is given by the value of the jump of the on-site potential $\nu_{KS}[n_0]$ at the discontinuity. In fact, for the calculations with $\nu_{KS}$ and for $V_g$ in this range, no steady-state is achieved. Hence, the conductance curves for these voltages carry some degree of uncertainty, associated with the interpretation of the time-averaged time-dependent current. However, the calculated time-averaged current in this range of gate voltages is clearly nonzero and the corresponding values of conductance fall on a smooth curve, which indeed resembles a plateau for large $U$. This feature is not reproduced with the Hartree potential, for which the conductance displays a broad peak, whose width is also proportional to $U$.

As already pointed out in section 3.2.1, for those voltages, which drive the charge density of the dot close to unity, even the calculation of the ground-state is problematic from a numerical viewpoint, because of the derivative discontinuity. In such cases we used the following iterative procedure. Let $V_g^0$ be the value of the gate voltage, for which the ground-state (initial) density is calculated self-consistently, while $V_g^0 + \delta V_g$ is the value of the gate voltage for which the self-consistent calculation does not converge. In this case, the final density, obtained at the end of the time-dependent simulation with $V_g = V_g^0$, is taken as initial density for the simulation with $V_g = V_g^0 + \delta V_g$.

In the same way, from the time-averages in the long-time limit, we map out the $I$–$V$ characteristics of the interacting dot ($\nu_{KS}$) at a given $V_g$ (see figure 6). A remarkable feature of the $I$–$V$ curves is the drop of the current at large source–drain voltages, which is almost negligible for small $U$ but increases with increasing $U$.

For all values of $U$ the current initially increases with increasing $V_{sd}$ as the dot is charging. It then reaches its maximum value as the charge density approaches $n_0 = 1$. This point corresponds to a threshold source–drain voltage $V_{sd}^{cr}$, which is roughly the same for all values of $U$. Beyond $V_{sd}^{cr}$, the system is driven into a dynamical state (where the steady-state current is calculated by averaging out the oscillations). In the limit of very large $V_{sd}$, the dot recovers its long-time tendency to a steady-state and the average current saturates. At saturation and beyond the dot occupation is above 1 and the on-site potential assumes a value above the discontinuity.

\[ \text{Figure 5. Differential conductance of the dot as a function of the gate voltage ($V_g$) for the Kohn–Sham potential, $\nu_{KS}$, (thick solid lines) and for the Hartree potential, $\nu_H$, (dashed lines) with $U = 1$, 2, and 3 eV, and for the non-interacting case (thin solid line). The inset shows a comparison between the density-dependence of $\nu_{KS}$ (solid lines) and $\nu_H$ (dashed lines) for the same values of $U$. Parameters are the same as those of figure 3 and $V_{ad} = 0.01$ eV.} \]

\[ \text{Figure 6. Current through the dot, } I_0, \text{ as a function of the source–drain voltage, } V_{sd}, \text{ for } \nu_{KS} \text{ and different values of } U. \text{ The horizontal dashed lines represent the corresponding saturation currents } I_s \text{ (see text for the exact definition). The following parameters are used: } \gamma_0 = -3.88 \text{ eV}, \gamma_c = -0.5 \text{ eV}, \epsilon_F = 1.5 \text{ eV}, \ V_g = 2.0 \text{ eV.} \]


Hence, the on-site energy at the dot is proportional to the jump of the $v_{\text{KS}}$ at $n_0 = 1$, i.e. it is proportional to $U$.

As discussed in section 3.1 for the non-interacting case, the saturation current decreases with increasing the dot on-site potential, because of the finite bandwidth of the electrodes. For the same reason here the drop of the current becomes larger when $U$ increases. In fact a large $U$ corresponds to a large value of the steady-state on-site potential, which then approaches the electrodes band-edge. In order to confirm this conjecture, we compare the saturation current $I_S$ calculated at finite $U$, with that for $U = 0$ and $V_g$ equal to the steady-state on-site potential corresponding to that obtained at the same $U$. Indeed $I_S$ matches quite well the value of the current obtained at large source–drain voltages in the $I$–$V$ characteristics of the interacting dot (see horizontal dashed lines next to each curve in figure 6). This argument can obviously be reversed, i.e. the negative differential conductance cannot be observed, if the variation of the on-site potential at the derivative discontinuity, determined by $U$, is much smaller than the electrodes’ bandwidth. For instance, for the same set of parameters used before for the dot + electrodes system, such situation is found for $U = 2$ eV ($U \ll 4|\gamma_0|$ for $\gamma_0 = 3.88$ eV). In this case the drop of the current above $V_{sd}^\text{cr}$ is practically negligible.

Importantly, the current drop displayed in figure 6 is not found in $I$–$V$ curves calculated within the Hartree approximation, even for large values of $U$ (see figure 7). When comparing calculations at the Hartree level with those performed with the complete Kohn–Sham potential we intentionally use different $U$. These are selected in such a way that the value of the potential at $n_0 = 1$ is identical in the two calculations (see the inset in figure 7(c)), i.e. in such a way that the two calculations give the same saturation current. At variance with the complete Kohn–Sham case, in the Hartree-only problem the current, as well as the density and the on-site potential, monotonically increase with $V_{sd}$ until the saturation is reached. Based on these numerical results we can argue that the self-interaction-free shape of the on-site potential $v_{\text{KS}}$ at the dot is a necessary condition for the occurrence of the negative differential conductance in the $I$–$V$. The shallow increase of the on-site potential with the charging, produced by the opening of the bias window, keeps the resonant level away from the electrodes band-edge and allows the current to rise. Once the on-site charge exceeds $n_0 = 1$ and the resonant energy level shoots up towards the band-edge, the currents drops. The averaged dynamical current monotonically approaches its saturation value corresponding to a steady-state solution.

4. Conclusions

We have investigated the electronic transport through a strongly correlated quantum dot by using a recently proposed multiple-probe battery method for time-dependent simulations of open systems. Our aim was two-fold. Firstly, we wanted to assess the outcomes of a time-dependent transport scheme conceptually different from what used so far in the literature, for a problem involving strong electron correlation as in Coulomb blockade. Clearly our MPB-based simulations agree well with previous findings [30]. In particular, we have established numerically that (i) the two practical time-dependent methods, based on qualitatively different initiation of the quantum transport, agree quantitatively in the long-time dynamics, (ii) the differences in the dynamical trajectories obtained with the two methods are limited to the early stages of the time-evolution and are determined by the initial conditions, and (iii) the observed long-time dynamics is characterized by self-sustained oscillations in the current, density and effective on-site potential, originating directly from the derivative discontinuity of the approximate XC potential used (ABALDA).

As a further aspect we have addressed the question of whether the peculiar dynamics obtained from the time-dependent simulations can be related to the more familiar steady-state picture of transport through an interacting quantum dot. In particular, we have calculated the linear-response differential conductance and extracted the time-dependent version of $I$–$V$ characteristics, based on the time-averaged current through the dot in the long-time limit. The resulting $I$–$V$ curves, at a critical voltage, exhibit a drop in the average current through the dot. This drop corresponds to the range of parameters where no steady-state is found and the dot is in the oscillatory Coulomb blockade state. The main reason for the observed current drop is the finite bandwidth in the electrodes. However the effect is present only when the calculation is performed at a DFT level in which the potential includes the derivative discontinuity at unitary occupation.

To summarize, the primary focus of our study has been that to explore some of the methodological issues

![Figure 7. Current (a), density (b) and on-site potential (c) of the dot as a function of the source–drain voltage, $V_{sd}$, for $v_{\text{KS}}$ with $U = 5$ eV and for $v_{\text{KS}}$ with $U = 9$ eV. The inset shows $v_{\text{KS}}$ and $v_{\text{KS}}$ as functions of the dot density for the corresponding values of $U$. The parameters are the same as those in figure 6.](image-url)
in time-dependent quantum transport theory, applied to a specific case of an interacting nanojunction described in the TDDFT framework. Clearly, at the present level of theory, the simulations of realistic nanoscale devices is still quite a remote prospect. Before such simulations become possible, one needs to consider other effects relevant to electron transport, for instance the inelastic interaction between electronic and vibrational degrees of freedom [49]. To date, the development of \textit{ab initio} approaches to electron transport, that take into account electron–phonon effects, has been mostly limited to the steady-state [50]. One way of incorporating such effects into time-dependent transport framework is to combine an efficient method for electronic open boundaries, such as the MPB, with an accurate method for real-time simulations of electron–ion dynamics, where electronic and ionic motions are treated on an equal quantum-mechanical footing [31]. However, before including interactions with additional degrees of freedom in the existing TDDFT-based schemes for electron transport, much work yet needs to be done to improve the description of electronic structure within the TDDFT framework. There are clear indications that in the case of reduced-dimensional strongly correlated systems the development of spatially and temporally non-local approximations to the XC functional is required in order to obtain a better agreement with more accurate many-body methods [35–37]. Such developments are subject of ongoing research.

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