

# Approach to Steady-State Transport in Nanoscale Conductors

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## ABSTRACT

We show, using a tight-binding model and time-dependent density-functional theory, that a quasi-steady-state current can be established dynamically in a finite nanoscale junction without any inelastic effects. This is simply due to the geometrical constriction experienced by the electron wave packets as they propagate through the junction. We also show that in this closed nonequilibrium system two local electron occupation functions can be defined on each side of the nanojunction which approach Fermi distributions with increasing number of atoms in the electrodes. The resultant conductance and current–voltage characteristics at quasi-steady state are in agreement with those calculated within the static scattering approach.

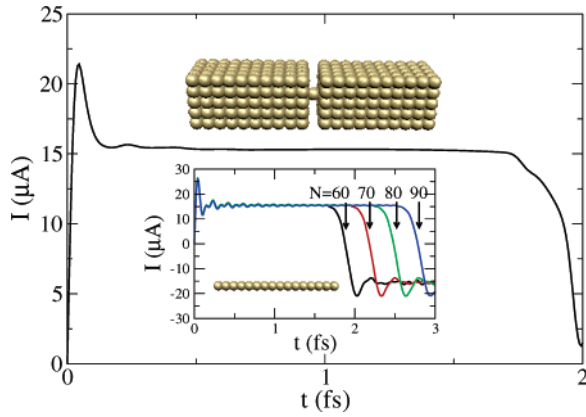
The static scattering approach has been extensively used to treat steady-state transport in mesoscopic and nanoscopic conductors. The approach, as originally introduced by Landauer, treats the sample as a scatterer between two leads, which are connected adiabatically to two infinite electron reservoirs at different local electrochemical potentials.<sup>1,2</sup> The reservoirs are just conceptual constructs which enable one to map the nonequilibrium transport problem onto a static scattering one.<sup>3,4</sup> However, the ensuing steady state may not necessarily be the “true” steady state that is reached dynamically when a battery discharges across the sample. In addition, the static picture says nothing about the dynamical onset of steady states, their microscopic nature, or their dependence on initial conditions. These issues are particularly relevant in nanoscale structures where some of the assumptions of the static approach may hide important physical properties pertaining to the true charge dynamics.

In this paper we employ an alternative picture of transport in nanoscale systems in which we abandon the infinite reservoirs invoked by Landauer. Instead, as recently suggested by Di Ventra and Todorov,<sup>5</sup> we consider the current that flows during the discharge of two large but *finite* oppositely charged electrodes connected by a nanojunction. Unlike the static, open boundary approach, the present approach permits one to describe the current within a microcanonical formalism where both energy and particle numbers are conserved quantities. In addition, due to the finite and isolated nature of the system, it can be demonstrated<sup>5</sup> that the *total* current flowing from one electrode to the other can be calculated *exactly* using time-dependent density-functional theory (TDDFT)<sup>6</sup> provided that one knows

the exact functional, regardless of whether the system reaches a steady state or not.

We find that a quasi-steady-state current, though lasting only for a limited period of time, can be established in the neighborhood of the nanojunction without any dissipation. This is simply due to the change in the spread of momentum of wave packets as they move into a nanojunction and adapt to the given junction geometry. This effect occurs roughly in a time  $\Delta t \sim \hbar/\Delta E$ , where  $\Delta E$  is the typical energy spacing of lateral modes in the junction. For a nanojunction of width  $w$ ,  $\Delta E \sim \pi^2 \hbar^2/m_e w^2$  and  $\Delta t \sim m_e w^2/\pi^2 \hbar$ . If  $w = 1$  nm,  $\Delta t$  is of the order of 1 fs, i.e., orders of magnitude smaller than typical electron–electron or electron–phonon scattering times.<sup>7</sup> We indeed focus on the electron dynamics after the quasi-steady state has been established and make a connection between this dynamical picture and Landauer’s static approach. To this end, we consider a finite three-dimensional (3D) model gold nanojunction and a finite quasi-one-dimensional gold wire (see schematics in Figure 1). These are the simplest structures for which the quantized conductance and current–voltage characteristics have been computed using the static scattering approach<sup>8,9</sup> and have been measured experimentally for similar gold quantum point contacts.<sup>10</sup> Recently, Horsfield et al. have shown that a steady current is generated in a similar finite atomic chain.<sup>11</sup> Nevertheless the question of whether a steady state can be reached without including any electron–ion interactions remains unanswered in their work.<sup>12</sup> In addition to answering this question, we show that one can define two local electron occupation functions on each side of the nanojunction. These are shifted in energy by an amount which can be interpreted, in the limit of large electrodes, as the “bias” of the

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**Figure 1.** Current passing through the junction as a function of time for a 3D nanojunction (schematic is shown) calculated with a noninteracting TB model. Each of the electrodes consists of  $5 \times 5 \times 30$  atoms arranged in a simple cubic geometry. The inset shows the corresponding current for a linear chain of  $N = 60, 70, 80,$  and  $90$  gold atoms. In both cases  $E_B = 0.2$  eV (see text).

corresponding open system. These functions depart from the equilibrium Fermi distributions by an amount which decreases with increasing electrode size. This verifies Landauer’s hypothesis that “geometrical dilution” of wave functions is the most important aspect of a reservoir.<sup>13</sup> However, contrary to previous conclusions,<sup>14,15</sup> we show that finite but long one-dimensional (1D) leads do not need to widen to constitute good “reservoirs”, as long as one considers the electron dynamics in the junction before the electrons reach the edge of the system.

We now begin our study by using a simple time-dependent tight-binding (TB) model for noninteracting electrons where Coulomb interactions and correlation effects are absent. Later, we treat the problem using a fully self-consistent TDDFT approach in the adiabatic local density approximation (ALDA).<sup>16</sup>

Consider the  $N$ -site single-orbital TB Hamiltonian

$$H^{\text{TB}} = \sum_{i=1}^N \epsilon_i |r_i\rangle\langle r_i| + \iota \sum_{i=1}^N |r_i\rangle\langle r_{i+1}| + \text{Hc} \quad (1)$$

where there is one orbital state  $|r_i\rangle$  per atomic site with energy  $\epsilon_i$  and transfer matrix element  $\iota$  connecting nearest-neighbor sites.<sup>17</sup> We then prepare the system such that half of the system has a deficiency of electrons and the other half has a surplus. This can be done by increasing  $\epsilon_i$  of the sites on one side of the system by an energy “barrier”  $E_B$ . For the 1D wire (see inset of Figure 1), the interface between the two regions separated by the barrier defines the nanoscale “junction”. Taking this state as the initial state of the system, we then remove  $E_B$ , and let the electrons propagate according to the time-dependent Schrödinger equation (TDSE) with the time-independent Hamiltonian  $H^{\text{TB}}$ .<sup>18</sup>

Due to the closed and finite nature of the system, the total current can be calculated by time differentiating the charge accumulated on one side of the system, i.e.

$$I(t) = -e \frac{d}{dt} \sum_{n=1}^{N/2} \sum_{i=1}^{N_L} \langle \psi^n(t) | r_i \rangle \langle r_i | \psi^n(t) \rangle \quad (2)$$

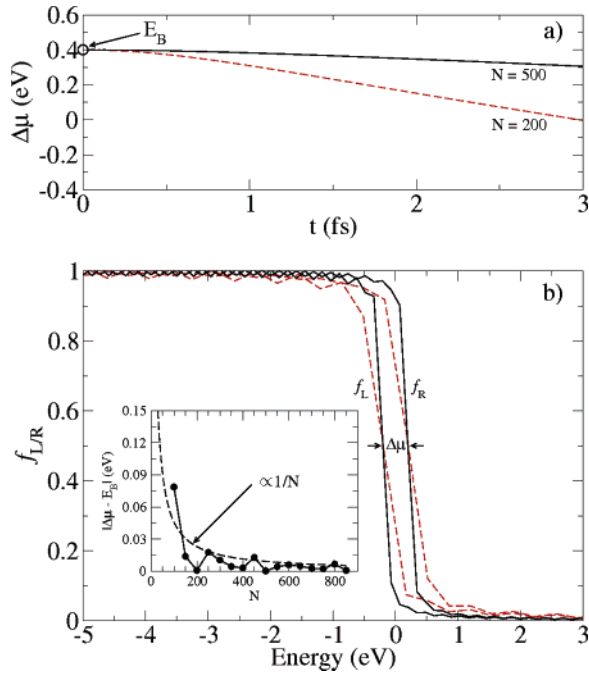
Here  $\psi^n(t)$  denotes the occupied single-electron states that are the solutions of the TDSE, and  $N_L$  is the number of sites on the left of the junction interface. Summation over spin degrees of freedom is implied.

The onset of a quasi-steady state for a 3D nanojunction is shown in Figure 1, where we plot the current eq 2 as a function of time for  $E_B = 0.2$  eV. In the inset, we show that a similar quasi-steady-state current develops in 1D wires of different lengths, where the initial time energy barrier forces electrons to change the spread of electron momentum, and hence plays a role similar to that of the geometric constriction in the 3D case. In all cases, the current initially rises rapidly but quickly settles in a quasi-constant value  $I_{\text{ss}}$ .<sup>19</sup> In the 1D structures, small oscillations are observed which decay in time. The quasi-steady state lasts for a time  $t_d$  during which the electron waves propagate to the ends of the wire and back. The time  $t_d$  is a few femtoseconds for the considered cases and can be made longer by increasing the length of the wires (see Figure 1 inset). We have thus demonstrated numerically our initial conjecture: in a closed and finite nanoscale system, a quasi-steady-state current with a finite lifetime can develop even in the *absence* of dissipative effects. The steady state is a direct consequence of the geometrical constriction experienced by the electron wave packets as they propagate through the junction. This is in contrast with the conclusion of ref 20, where the establishment of a steady state is attributed to a “dephasing mechanism” of the electrons spreading in infinite electrodes.

To calculate the conductance of this closed system, we need to define a “bias”. In this noninteracting electron problem, the energy barrier  $E_B$  seems a natural choice. However, that is not completely satisfying as it relates to the initial conditions and not to the electron dynamics. Let us instead define local occupation numbers for electrons in the left and right regions of the system. This concept is typically introduced as a starting point in the static approach to transport. Here we would like to define it dynamically. We then project the occupation for each eigenstate  $|E_j\rangle$  of the Hamiltonian  $H^{\text{TB}}$ , i.e.,  $f(E_j, t) = \sum_n |\langle E_j | \psi^n(t) \rangle|^2$  onto the left- and right-hand side of the system

$$f(E_j, t) = \sum_n \left| \sum_{i \leq N/2} \langle E_j | r_i \rangle \langle r_i | \psi^n(t) \rangle \right|^2 + \sum_n \left| \sum_{i > N/2} \langle E_j | r_i \rangle \langle r_i | \psi^n(t) \rangle \right|^2 + \sum_n 2\text{Re} \left\{ \sum_{i \leq N/2} \langle E_j | r_i \rangle \langle r_i | \psi^n(t) \rangle \sum_{i > N/2} \langle \psi^n(t) | r_i \rangle \langle r_i | E_j \rangle \right\} \quad (3)$$

We denote the first, second, and third term  $f_L(E_j, t)$ ,  $f_R(E_j, t)$ , and  $f_C(E_j, t)$ , respectively. The quantity  $f_C(E_j, t)$  is the sum of cross terms between the occupation on the left and on the right region. Note also that because the set  $\{|E_j\rangle\}$  forms a complete orthonormal basis,  $\sum_j f_L(E_j, t) = n_L$ ,  $\sum_j f_R(E_j, t) = N - n_L$ , and  $\sum_j f_C(E_j, t) = 0$ , where  $n_L$  is the total charge left to

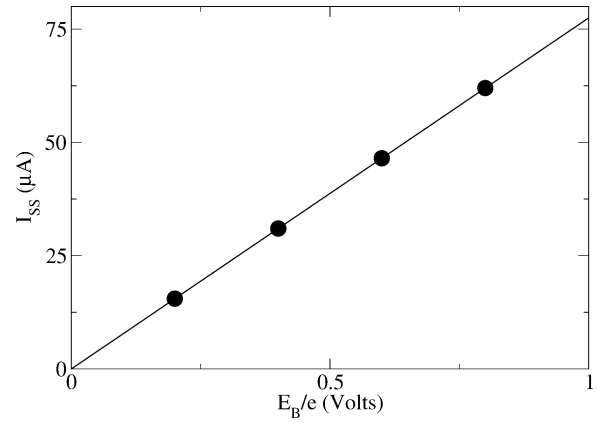


**Figure 2.** Panel a illustrates  $\Delta\mu(t)$  for a linear chain of  $N = 200$  (dashed line) and  $N = 500$  (solid line) atoms. Panel b shows their local occupation functions  $f_L(E_j, t)$  and  $f_R(E_j, t)$  at a small time  $t$  after the onset of current. The inset shows the absolute difference between  $\Delta\mu(t=0)$  and the initial energy barrier  $E_B = 0.4$  eV as a function of  $N$ . The dashed curve is proportional to the function  $1/N$ .

the junction at a given time. The quantities  $f_L(E_j, t)$  and  $f_R(E_j, t)$ , normalized to two electrons per state, are plotted in Figure 2b for two wires of  $N = 200$  and  $N = 500$  atoms immediately after the onset of the current.

One might naively think of these functions as broadened Fermi distributions centered at different “chemical potentials”, separated by an energy  $\Delta\mu(t)$ . A closer examination, however, reveals that they cannot be simply fitted to Fermi functions with just an effective thermal broadening. Instead, the very functional form of these nonequilibrium functions is different from a Fermi distribution. This is not surprising, as in this finite dynamical system electrons spread on each side of the junction and are not in any sort of local equilibrium in the electrodes. In addition,  $\Delta\mu(t)$  decreases with time because of the transport of electrons from one side of the system to the other (see Figure 2a). However, as long as we evaluate  $f_L(E_j, t)$  and  $f_R(E_j, t)$  at times much less than  $t_d$ , these functions approach two zero-temperature Fermi distribution functions centered at two different energies  $\mu_L$  and  $\mu_R$ , and  $f_C(E_j, t)$  tends to zero for every  $E_j$  with increasing number of sites  $N$  in the electrodes. These energies can be interpreted as two local “chemical potentials” on the left and right sides of the system, with  $\Delta\mu = \mu_R - \mu_L$  approaching the initial energy barrier  $E_B$  with increasing  $N$ .

This asymptotic behavior as  $N \rightarrow \infty$  is illustrated in Figure 2b where  $f_L(E_j, t)$  and  $f_R(E_j, t)$  at a small  $t$  ( $0 < t \ll t_d$ ) are plotted for different values of  $N$ . The inset shows the absolute difference between  $\Delta\mu(t=0)$  and  $E_B$  as a function of  $N$ . The difference scales with  $N$  in the same way as does the separation of eigenstates of  $H^{\text{TB}}$  close to  $\mu_L$  and  $\mu_R$ . In three dimensions  $N = N_x \times N_y \times N_z$ , and it is then easy to prove,



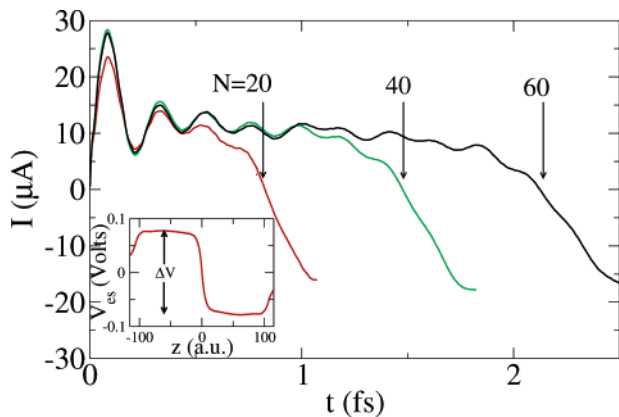
**Figure 3.** Current–voltage ( $I$ – $V$ ) characteristics at quasi-steady state of a finite 1D gold wire obtained using the TB approach. The corresponding conductance is  $1.0G_0$ . The TDDFT calculation yields a conductance of  $(0.99 \pm 0.03)G_0$ .

in this simple TB model, that  $\Delta\mu(t=0)$  approaches (albeit “nonvariationally”)  $E_B$  as  $1/N_x + 1/N_y + 1/N_z$  with increasing number of atoms in the three different directions (see inset of Figure 2b for the 1D case). It is therefore evident that with increasing  $N$ , local equilibrium distributions can be effectively achieved in the two electrodes without inelastic effects: the electron waves moving into these regions are geometrically “diluted” in a practically infinite region of space and therefore do not “disturb” the local electron occupation. This is the equivalent of Landauer’s definition of reservoirs.<sup>14</sup> Our results, however, show that this definition can be extended to one-dimensional electrodes as well.

All this discussion allows us to define a conductance in this closed system in terms of the current at steady state  $I_{ss}$  and the value of  $\Delta\mu(t)/e$  for  $N \rightarrow \infty$ . The former converges very fast with increasing number of atoms, whereas the latter is the desired “bias” which, in turn, is simply the potential “barrier”  $E_B/e$  at  $t = 0$ . The current  $I_{ss}$  as a function of  $E_B/e$  is plotted in Figure 3. The corresponding differential conductance is about  $1.0G_0$  ( $G_0 = 2e^2/h$ ) at all voltages, in good agreement with values obtained from the static approach<sup>8,9</sup> and experimental observations for similar systems.<sup>10</sup>

Finally, we study the onset of quasi-steady states in the presence of electron interactions that we describe at the mean-field level. We illustrate this point using TDDFT within the ALDA<sup>21</sup> for 1D wires. The corresponding current is plotted in Figure 4 for different lengths of a finite chain of gold atoms kept a fixed distance of  $2.8 \text{ \AA}$  apart.<sup>22</sup> The lifetime of the quasi-steady state is short due to the limited system size but clearly increases with increasing length of the wire. What is more interesting, however, is the time for the quasi-steady state to set in. The initial transient time is found to be less than 1 fs, consistent with our original estimate.

The single-particle Kohn–Sham states<sup>23,6</sup> have no explicit physical meaning, so the interpretation of the corresponding functions in eq 3 is less clear. On the other hand, the charge density and, thus, the electrostatic potential are well-defined quantities. We therefore define the conductance in this closed system in terms of the electrostatic potential drop between two points inside each electrode.<sup>5</sup> As in the case of  $\Delta\mu$ , the



**Figure 4.** Current in the middle of the junction as a function of time for a linear chain of  $N = 20, 40, 60$  atoms calculated using TDDFT-ALDA. The inset shows the electrostatic potential drop along the wire at  $t = 0$  for a wire of 40 atoms.

potential drop converges to the  $t = 0$  value (plotted in the inset of Figure 4) with increasing number of atoms.<sup>24</sup> The corresponding differential conductance is about  $(0.99 \pm 0.03)G_0$ , where the average value has been determined from the current in the wire with  $N = 60$  atoms at  $t = 1$  fs.<sup>25</sup> It is worth pointing out that when the hopping parameter in the tight-binding calculation is chosen to match the drop-off time  $t_d$  in the TDDFT calculation for the same number of atoms, the initial transient time during which the quasi-steady state establishes itself in the tight-binding calculation is also less than 1 fs. This observation reinforces the notion that the geometric constriction effect is present irrespective of the inclusion of mean-field interactions.

Finally, one can obtain an order-of-magnitude estimate of the electrode size necessary to observe the quasi-steady state. The drop-off time  $t_d$  is roughly given by the time it takes for an electron to travel at the Fermi velocity of the underlying lattice along the length of the electrode and back, i.e.,  $t_d \approx L/v_f$ , where  $L/2$  is the linear length of one of the electrodes. Therefore, one should observe a quasi-steady state if the time necessary for the quasi-steady state to be established  $\Delta t$  is less than  $t_d$ , i.e.,  $L > v_f m_e v^2 / \pi^2 \hbar$ . The results illustrated in Figures 1 and 4 are consistent with these crude estimates and show that a relatively small number of atoms is necessary to represent the electrodes, thus making the present approach a practical alternative to standard open-boundary calculations of transport.

In conclusion, we have shown numerically that a quasi-steady state can be achieved in a nanoscale system without dissipative effects, simply owing to the geometrical constriction experienced by electron wave packets as they approach the nanojunction. We have also provided a practical scheme for dynamical conductance calculations in finite nanoscale systems that sheds new light on the assumptions of the standard static approach to steady-state conduction. The approach is also suited to study relatively unexplored effects such as transient phenomena, time-dependent charge disturbances, uniqueness of steady states, and their dependence on initial conditions.

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- (17) The matrix element  $\epsilon = -11$  eV is chosen so that the time scales of the TB calculation are comparable to those in the TDDFT calculation for the same number of atoms in the system.
- (18) This is just one of the many (essentially infinite) initial conditions one can choose to initiate current flow. However, note that some initial conditions may not lead to a quasi-steady state. The question of the dependence of steady states on initial conditions has been addressed in refs 5 and 20 and will be analyzed in more detail in a future publication.
- (19) Note that, in the 3D case, a quasi-steady state can be established only if the typical energy spacing of lateral modes of the electrodes is much smaller than the corresponding one in the junction.
- (20) Kurth S.; Stefanucci, G.; Almladh, C.-O.; Rubio, A.; Gross, E. K. U. *Phys. Rev. B* **2005**, *72*, 035308.
- (21) The calculations reported here have been done using the socorro package (<http://dft.sandia.gov/Socorro/mainpage.html>), adapted by Ryan Hatcher to perform time-dependent calculations.
- (22) As in the TB case, we construct the initial state of the system such that the left-hand side of the gold chain has a deficiency of charge and the right-hand side has a surplus; we use a steplike potential to create this imbalance at  $t = 0$ . The current is similarly determined by differentiating in time the charge accumulating on the left side.
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- (24) Note that in the static formulation of transport there is a conceptual (albeit numerically small, see, e.g.: Di Ventra, M.; Lang, N. D. *Phys. Rev. B* **2002**, *65*, 045402) distinction between the chemical potential difference and the electrostatic potential drop, the conductance being usually defined in terms of the former.
- (25) We have recently shown that there exist dynamical corrections to the electron conductance obtained using ALDA which exists even in the limit of zero frequency (Sai, N.; Zwolak, M.; Vignale, G.; Di Ventra, M. *Phys. Rev. Lett.* **2005**, *94*, 186810). These corrections depend on the gradient of the electron density and are therefore negligible for a gold quantum point contact. Combined with the theorem in ref 5 on the exact value of the TDDFT total current in a closed and finite system, this result shows that the TDDFT-ALDA current of a gold junction at steady state is very close to the exact many-body value.

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