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Abstract

We analyse a picture of transport in which two large but finite charged electrodes discharge across a nanoscale junction. We identify a functional whose minimization, within the space of all bound many-body wavefunctions, defines an instantaneous steady state. We also discuss factors that favour the onset of steady-state conduction in such systems, make a connection with the notion of entropy, and suggest a novel source of steady-state noise. Finally, we prove that the true many-body total current in this closed system is given exactly by the one-electron total current, obtained from time-dependent density-functional theory.

When a metallic nanojunction between two macroscopic electrodes is connected to a battery, electrical current flows across it[3]. The battery provides, and maintains, the charge imbalance between the electrode surfaces needed to sustain steady-state conduction in the junction. This static non-equilibrium problem is usually described according to the Landauer picture [1]. In this picture, the junction is connected to a pair of defect-free metallic leads, each of which is connected to its own distant infinite heat-particle reservoir. The pair of reservoirs represents the battery. Each reservoir injects electrons into its respective lead with the electrochemical potential appropriate to the bulk of that reservoir. Each injected electron then travels undisturbed down the respective lead to the junction, where it is scattered and is transmitted, with a finite probability, into the other lead. From there it flows, without further disturbance, into the other reservoir. The reservoirs are conceptual constructs which allow us to map the transport problem onto a truly stationary scattering one, in which the time derivative of the total current, and of all other local physical properties of the system, is zero. By doing so, however, we arbitrarily enforce a specific steady state whose microscopic nature is not, in reality, known *a priori*. The Landauer construct is highly plausible in the case of non-interacting electrons. In the case of interacting electrons, however, it is not at all obvious

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[3] Here we consider DC bias conditions. Similar considerations, however, apply to the AC case, that is, periodic time-varying disturbances of the charge.
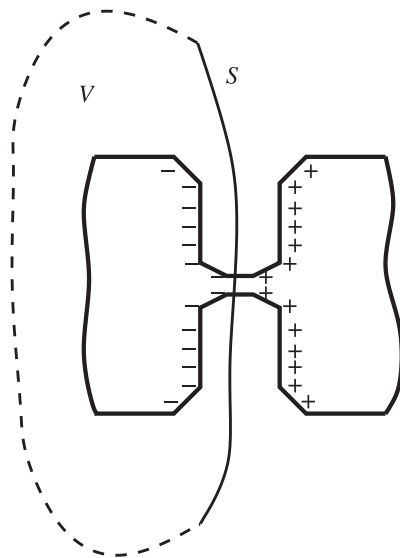
Figure 1. A nanoscale junction between two large but finite metallic charged electrodes. The details are discussed in the text.

that the steady state in the Landauer picture is the same as that which would be established *dynamically* by the electrons originating from the battery and flowing across the junction. It is also not obvious whether the same steady state can be reached with different initial conditions or if it can be realized with microscopically different many-body states.

There is, however, an alternative picture of DC conduction. In that picture, we dispense with the battery and we think of the current as a long-lived, but ultimately transient, discharge of a macroscopic, but finite, capacitor [2–5]. This view has great appeal. We now have a finite system, with a finite number of electrons and nuclei. This system can be visualized and realized practically. It can be described dynamically, at least in principle, by solving a finite, closed set of equations of motion for the particles in the system. In this picture, the system is allowed to find its own electronic structure during the discharge, without the imposition of *a priori* assumptions about what this electronic structure should look like. This 'microcanonical' picture, in which the conventional notion of transport as an open-boundary, 'grand-canonical' problem is replaced by the idea of a long-lived discharge of an isolated finite system, is represented schematically in figure 1. In keeping with the microcanonical framework that we are interested in, we do not introduce dissipative effects in this system [5–7].

The purpose of this paper is twofold. First, we seek to establish a mapping of the conventional steady-state transport problem onto the present microcanonical one. To this end, we define in variational terms a class of dynamical states for the closed geometry in figure 1 that we call instantaneous global quasi-steady states. We then give arguments for their robustness. Second, we demonstrate that the exact total current (as opposed to current density) in the true interacting many-electron system is identical to the one-electron current, obtained from time-dependent density-functional theory (TDDFT) [8]. The closed, finite nature of the system is a requirement for this demonstration. The result is valid throughout the time evolution of the electronic system, regardless of whether or not the system gets anywhere near a steady state. This result places on a rigorous footing the application of time-dependent one-electron methods to transport in nanoscale systems.

Our motivation is the tremendous physical and computational appeal of the resultant microcanonical picture of conduction: it gives us, in principle, a formally exact, dynamical description of many-electron transport within a one-electron picture. It does so, furthermore, in a way that eliminates the numerically cumbersome implementation of scattering boundary conditions and the unphysical notion of infinite systems that continually plague one in the traditional static approach to transport.

Before discussing the closed system in figure 1, let us return to the transport problem in the open-boundary approach. In the self-consistent steady state, a net electron current from one electrode into another across a nanojunction is accompanied by an excess of electrons in one electrode and a deficit of electrons in the other [9]. These charges take the form of surface charge densities, present within a screening length of the electrode surface. Thus, steady-state conduction goes hand in hand with surface charges on each side of the junction: a negative sheet of charge on one side and a positive sheet on the other, as if two infinite capacitor plates were present. The transport problem can then be viewed as a continuous attempt by the electrons to 'passivate' the surface charges on each side of the junction. In other words, the steady-state transport problem in the traditional open-boundary approach *is*, effectively, the continuous discharge of an *infinite* capacitor.

In real life there is no such thing as an infinite capacitor. We must therefore consider the discharge of a finite, though possibly very large, capacitor. If C is its capacitance and R is the resistance across which it discharges, then, according to classical circuit theory, the discharge will take place over a characteristic time RC. The larger C, for a given R, the more stationary the properties of the system will appear, in a temporally local sense, at any one stage of the discharge. This ultimately transient, but very slowly varying, conducting state indeed encapsulates the intuitive picture of a DC steady state that we all have.

There are standard *time-dependent* approaches that allow the DC steady state, in the limit of infinite system size, to be described from a microscopic point of view. One such approach is linear response theory. Here, we imagine that the electrons in the electrode–junction–electrode system start off in the ground state. Then, at some initial time $t = 0$, a static external electric field (which need not be uniform) is applied, and the electronic response for $t > 0$ is calculated to first order in the applied field. If we take the size of the electrodes to infinity (at least in the longitudinal direction) and then take the limit $t \rightarrow \infty$, with the further assumption that the electrons are non-interacting, we may follow the arguments of Stone and Szafer [10] to obtain the two-probe one-electron Landauer formula $G = (2e^2/h) \operatorname{Tr}\{\mathbf{t}\mathbf{t}^\dagger\}$, where G is the conductance and $\mathbf{t}$ is the transmission matrix. The two-probe Landauer formula can therefore be thought of as describing the steady state of a system of non-interacting electrons flowing between two infinitely large electrodes, under an external field. However, this approach is of no use to us here because it explicitly requires the system size to be taken to infinity, which is specifically what we do not wish to do. We observe also that this approach does not take account of electron–electron interactions. Of course, formally, one may write down the linear response calculation in many-body form, but the result is in general intractable. If, on the other hand, we choose the one-electron route of TDDFT, then in the linear response calculation, on top of the external field, we must include the additional time-varying one-electron potential due to the dynamical evolution of the electron density itself. It then stops being obvious whether, and in what form, the one-particle Landauer formula would survive. While the possible resultant corrections to the open-boundary Landauer formula constitute a very interesting line of work, the above approach to the steady state, once again, falls beyond our goals here, because of our specific concern with *finite* systems.

An alternative dynamical approach, in a minimal discrete real-space basis set [4], would be to start with two charged but electronically decoupled electrodes, connect them by a junction

and follow the ensuing discharge [11]. If once again we take the system size to infinity and then take the limit $t \to \infty$, while treating the electrons as non-interacting, then we would end up solving the usual stationary Lippmann–Schwinger equation for the one-electron wavefunctions in an open-boundary system [9, 12]. This approach would once again result in the standard one-electron transport formulae [4, 11]. However, this approach does not help us here for the same reason as above: it relies on taking the system size to infinity. By analogy with the earlier remarks, we note that if we include electron–electron interactions in the *time-dependent* calculation in the form of an additional dynamical one-electron potential that depends on the time-evolving electron density, then it is not obvious that the resultant infinite-system, long-time behaviour would be the same as the self-consistent solution in the standard *time-independent* one-electron scattering open-boundary approach, in which one solves the one-electron static Lippmann–Schwinger equation iteratively and self-consistently, with a given functional relation between density and one-electron potential, and with given fixed incoming one-electron distribution functions [4].

We now return to our finite isolated system in figure 1. We release the electrons from an arbitrary but definite initial state, characterized by a charge imbalance between the electrodes. We thenceforth allow the electrons to propagate dynamically. After an initial transient time (related to the initial state of the electrons and to the electron–electron relaxation time inside the capacitor plates) during which electrons and holes first start to traverse the nanojunction from opposite sides, we expect a quasi-steady state to be established. We expect it to persist until the time when multiple electron reflections off the far boundaries of the system begin to develop. From then on, the electronic system will oscillate in time among several many-body states forever, if we neglect dissipative effects, as we do here[4]. We are interested in the intermediate quasi-steady state.

Our first task is to define this quasi-steady state in variational terms. By appealing to the intuitive concept of a steady state in the case of a macroscopic classical capacitor, considered earlier, we adopt the view that a steady state is one in which the temporal variation of local properties is minimal. A measure of temporal variations is provided by the functional

$$A[\rho] = \int_{t_1}^{t_2} \mathrm{d}t \int_{\text{all space}} \mathrm{d}\mathbf{r} \left(\frac{\partial \rho(\mathbf{r}, t)}{\partial t} \right)^2 \tag{1}$$

where $\rho(\mathbf{r}, t)$ is the density of the electron gas in the system depicted in figure 1 and (t_1, t_2) is some time interval of interest. Let us first perform an unconstrained variational minimization of A with respect to ρ, for a given $\rho(\mathbf{r}, t_1)$ and $\rho(\mathbf{r}, t_2)$. The result is

$$\frac{\partial^2 \rho(\mathbf{r}, t)}{\partial t^2} = 0, \qquad \forall \mathbf{r}, \quad \forall t \in (t_1, t_2). \tag{2}$$

To interpret this result, let us consider the total current, I_S, through an open surface S across the electrode–junction–electrode system, as shown in figure 1:

$$I_S = I_S(t) = \int_S \mathbf{j}(\mathbf{r}, t) \cdot \mathrm{d}\mathbf{S} \tag{3}$$

where $\mathbf{j}(\mathbf{r}, t)$ is the current density of the electrons. We close S in the vacuum, as is indicated by the dashed part of the curve in figure 1, sufficiently far from the boundaries of the system to enable us to ignore any contributions to the surface integral over the dashed part of the curve[5].

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[4] If we do allow dissipative effects [5], then a finite system is expected to reach a truly time-indepedent steady state, in the course of its evolution. If the system is infinite, then this could be a current-carrying steady state.

[5] Here we assume, as we will do again later, that all electrons are bound within the electrode–junction–electrode system, so that the charge and current densities are exponentially small into the vacuum, at all times.

By using the continuity equation

$$\nabla \cdot \mathbf{j}(\mathbf{r}, t) + \frac{\partial \rho(\mathbf{r}, t)}{\partial t} = 0 \tag{4}$$

and by invoking Gauss's theorem, we find

$$\frac{\mathrm{d} I_S(t)}{\mathrm{d}t} = -\int_V \mathrm{d}\mathbf{r} \, \frac{\partial^2 \rho(\mathbf{r}, t)}{\partial t^2} \tag{5}$$

where V is the volume bounded by S, which we take to completely envelop one of the electrodes, as shown in the figure. From equation (5) we see that, under the conditions expressed by equation (2),

$$\frac{\mathrm{d} I_S(t)}{\mathrm{d}t} = 0, \qquad \forall S. \tag{6}$$

Equation (6) by itself says nothing about the actual value of $I_S(t)$ or about the dependence of $I_S(t)$ on S. Equation (6) simply describes a *generic* type of conducting state in which matter (in this case electrons) is being transferred from region to region *at a steady rate*. We *define* such a state as a *true* global steady state. We have seen that such states are minima of the quantity A in equation (1). If we write $t_2 - t_1 = \delta t > 0$ and use the above procedure for smaller and smaller δt, then in the limit $\delta t \rightarrow 0$ we would obtain a weaker version of equation (2), with reference to a single instant in time. This instantaneous form of equation (2) defines a type of conducting state that we call a true *instantaneous* global steady state.

We observe that we could *start* from equation (6) as the mathematical definition of a true steady state, and seek a density functional whose minimization generates such steady states. We would then work our way back to equation (2), look for a functional such that equation (2) is its corresponding Euler–Lagrange equation of motion, and arrive at equation (1).

The minimization procedure applied to A above treats $\rho(\mathbf{r}, t)$ as a freely adjustable function of time, without regard for the actual physical dynamical laws governing the electrons. We did invoke the continuity equation, which is a physical equation, but we did so in interpreting the *results* of the minimization of A, not in the minimization itself. A real gas is governed by microscopic dynamical equations, which may not permit the system to attain a true instantaneous global steady state, as defined above. Our next task, therefore, is to seek the dynamical state *closest* to a true instantaneous global steady state, permitted by the laws of motion that govern our system.

We choose quantum mechanics as the dynamical law in question. Our electrons then are described by a many-body state vector $|\psi(t)\rangle$, governed by the time-dependent Schrödinger equation

$$i\hbar \frac{\mathrm{d}|\psi(t)\rangle}{\mathrm{d}t} = H|\psi(t)\rangle, \qquad |\psi(0)\rangle = |\psi_0\rangle \tag{7}$$

where H is the many-body electron Hamiltonian. For the moment, we regard the initial condition $|\psi(0)\rangle = |\psi_0\rangle$ as a parameter. The electron density and current density are given by

$$\rho(\mathbf{r}, t) = \langle \psi(t)|\hat{\rho}(\mathbf{r})|\psi(t)\rangle \tag{8}$$

$$\mathbf{j}(\mathbf{r}, t) = \langle \psi(t)|\hat{\mathbf{j}}(\mathbf{r})|\psi(t)\rangle \tag{9}$$

where $\hat{\rho}(\mathbf{r})$ and $\hat{\mathbf{j}}(\mathbf{r})$ are the many-body electron number-density and current-density operators, respectively. The time-dependent Schrödinger equation—our chosen dynamics—guarantees equation (4).[6] We may build this dynamical property of the system into the functional that

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[6] In fact, the current-density operator in quantum mechanics is specifically constructed so that the continuity equation is obeyed, but, with quantum mechanics already in place, this does not affect our logic here.

we have chosen as a measure of how close our system is to a steady state, by replacing $(\partial\rho(\mathbf{r},t)/\partial t)^2$ in equation (1) by $(\nabla\cdot\mathbf{j}(\mathbf{r},t))^2$.

We now define the instantaneous dynamical state closest to a true steady state by the following variational procedure. Consider all possible evolutionary paths, allowed by the dynamics of the system, in a small time interval (t_1, t_2). Of these, consider those that have a given total energy E and a given total number of electrons N. (In our microcanonical picture E and N are constants of the motion.) Of those, consider the evolutionary paths that produce a given average current I_S, during the interval (t_1, t_2), across a given open surface S. Of those, we seek that evolutionary path that minimizes A in equation (1).

Let B be defined by $A = B\delta t$, $\delta t = t_2 - t_1$. Then, in the limit of vanishing δt, the above search is equivalent to minimizing (with the same constraints) the instantaneous functional, at time $t = t_1 = t_2$,

$$B = B[|\psi(t)\rangle] = \int_{\text{all space}} \mathrm{d}\mathbf{r}\ (\nabla\cdot\mathbf{j}(\mathbf{r},t))^2 \tag{10}$$

with $\mathbf{j}(\mathbf{r},t)$ given by equation (9). Thus, we write $E = \langle\psi(t)|H|\psi(t)\rangle$, and we put

$$|\psi(t)\rangle = \sum_i c_i|\psi_i\rangle \tag{11}$$

where $\{|\psi_i\rangle\}$ are all N-electron *bound* states of the system in figure 1, with eigenenergies $\{E_i\}$, and $\{c_i\}$ are expansion coefficients. The reason for restricting the expansion to the bound part of the spectrum of H is that we do not wish to allow ionization of the electron–junction–electrode system. Ionization would correspond to the escape, through the vacuum, of some finite electronic charge to infinity and, therefore, would not correspond to the experimental realization of DC transport. We substitute the expansion in equation (11) into (9), and thence into (10). We then seek minima, with respect to $\{c_i\}$, of

$$B[\{c_i\}] = \sum_{i,i',i'',i'''} c_i^* c_{i'} c_{i''}^* c_{i'''} \int_{\text{all space}} \mathrm{d}\mathbf{r}\ (\nabla\cdot\mathbf{j}_{ii'}(\mathbf{r}))(\nabla\cdot\mathbf{j}_{i''i'''}(\mathbf{r})) \tag{12}$$

where $\mathbf{j}_{ii'}(\mathbf{r}) = \langle\psi_i|\hat{\mathbf{j}}(\mathbf{r})|\psi_{i'}\rangle$, subject to the constraints

$$I_S = \sum_{i,i'} c_i^* c_{i'} \int_S \mathbf{j}_{ii'}(\mathbf{r})\cdot\mathrm{d}\mathbf{S} \tag{13}$$

$$E = \sum_i c_i^* c_i E_i \tag{14}$$

$$\sum_i c_i^* c_i = 1. \tag{15}$$

Each solution for $|\psi\rangle = \sum_i c_i|\psi_i\rangle$ is an *instantaneous* many-body state, call it $|\psi(E, I_S, t)\rangle$, containing only electrons bound within the electrode–junction–electrode system in figure 1, that produces a given current I_S and a given total energy E, while globally minimizing the divergence of the current density. The time t is not part of the actual variational procedure that generates the state $|\psi(E, I_S, t)\rangle$. This state is just a snapshot. We *assign* a time t to the snapshot, for the sake of being able to relate the steady state in the snapshot to a definite initial condition, at an arbitrary but definite initial time $t = 0$.

We call the solution $|\psi(E, I_S, t)\rangle$ an instantaneous *quasi*-steady state: the best our finite system can do to mimic a true instantaneous global steady state. We may now explicitly write the initial condition required for the given instantaneous quasi-steady state as

$$|\psi_0(E, I_S)\rangle = \mathrm{e}^{\mathrm{i}Ht/\hbar}|\psi(E, I_S, t)\rangle. \tag{16}$$

This minimization procedure may lead to more than one quasi-steady-state solution $|\psi(E, I_S, t)\rangle$ for a given E and I_S; in other words, there may be different (in terms of charge and current densities) microscopic realizations of the *same* steady-state current. There may be combinations of E and I_S for which no solution for $|\psi(E, I_S, t)\rangle$ exists. Finally, there may be initial conditions $|\psi_0\rangle$ that do not ever lead to a quasi-steady state, i.e. that cannot be reached by back-propagation from any $|\psi(E, I_S, t)\rangle$.

The variational nature of the quasi-steady state enables us to draw the following conclusion. At a quasi-steady state $|\psi(E, I_S, t)\rangle$, B is, by construction, stationary against variations of $|\psi\rangle$ about $|\psi(E, I_S, t)\rangle$, compatible with the constraints. But B is a measure of the magnitude of the divergence of the current density, at least in a macroscopically averaged sense. Thus, we may expect the quasi-steady-state flow pattern itself to be relatively insensitive to variations about $|\psi(E, I_S, t)\rangle$, at least on a coarse-grained scale. However, after back-propagation to $t = 0$, the corresponding spread of initial conditions, about $|\psi_0(E, I_S)\rangle$, may contain large variations in microscopic quantities such as the charge density. In other words, there may be 'pockets' of initial conditions (in Hilbert space), which we denote symbolically by $P_0(|\psi(E, I_S, t)\rangle)$, that differ in their microscopic properties but that produce the same, or nearly the same, quasi-steady-state flow pattern at some later time t. This conclusion supports the intuitive notion that the steady state should be relatively insensitive to the microscopic detail in the initial conditions. This conclusion also suggests a link with the notion of entropy. The likelihood of a system with a given total energy E attaining a steady state, $|\psi(E, I_S, t)\rangle$, with a given total current I_S at time t, and the stability of this steady state against small perturbations, is measured by the relative weight (in Hilbert space) of the 'pocket' $P_0(|\psi(E, I_S, t)\rangle)$, among all initial conditions that lead to the current I_S at t.

To develop this idea further, suppose that, for a given E and I_S, we found several distinct quasi-steady states $|\psi(E, I_S, t)\rangle$. We *postulate* that the quasi-steady state $|\psi(E, I_S, t)\rangle$, which would be observed, or preferentially observed, in a macroscopic experiment on the system at time t, is that whose 'pocket' $P_0(|\psi(E, I_S, t)\rangle)$ has the largest statistical weight. In other words, the system is driven towards a specific microscopic quasi-steady state at time t by a 'maximum-entropy principle', where the 'entropy' measures the number of different initial conditions that realize the given steady state. The 'entropy' introduced here both has a classical thermodynamic meaning and contains the system dynamics through the minimization procedure. The present maximum-entropy principle is a reinterpretation, in the terms of the present microcanonical picture, of the approach used in reference [13] in the grand-canonical case, in a mean-field one-electron picture (see also reference [14]).

There may exist quasi-steady-state solutions that have the same E but different I_S. The simplest interpretation of such solutions is that they represent different degrees of the discharge of the system. Thus, if we assign the same t to them, their respective initial conditions would correspond to different initial voltage drops in the system. Alternatively, if we insist that their initial conditions have the same, or comparable, voltage drops, then steady-state solutions with different I_S would correspond to different t. This does not mean, however, that steady states with the same E but with different I_S are necessarily connected by a single continuous evolutionary path: one solution may or may not occur as a dynamical evolution of another. However, there may in fact be cases where solutions with the same E but different I_S describe steady states of different currents at the same time t corresponding to the *same* voltage drop. Such instances correspond to chaotic transport and may occur in systems with intrinsic nonlinear dynamics [15].

Finally, there is a further, highly speculative but intriguing, possibility. If we consider the quasi-steady-state solutions in an ensemble $\{I_S\}$ of currents and form a linear combination of many-body wavefunctions out of their respective pockets of initial conditions, the system

with this new initial condition could possibly evolve in time into the quasi-steady state of yet *another* I_S, that does not belong to the original ensemble $\{I_S\}$. If this happens, then the system can fluctuate coherently between microscopic quasi-steady states with *different* currents and steady-state noise is produced. This additional noise has nothing to do with the ordinary (shot) noise due to charge quantization [16]; instead, it would be due to possible realizations of a steady state as a linear combination of microscopic steady states corresponding to different currents.

Let us now consider the quasi-steady state from the point of view of a practical measurement or a time-dependent calculation. We let the system go from some initial state that we assume belongs to a 'pocket' $P_0(|\psi(E, I_S, t)\rangle)$, such that, at some later time t, a quasi-steady state with a total current I_S, across a chosen surface S, is established. Let us then consider how we can define a conductance in this finite-system approach. Now that we have dispensed with the infinite reservoirs, we may no longer appeal to Büttiker's definition of conductance, with respect to bulk electrochemical potentials of reservoirs in a multiprobe measurement [17]. We thus fall back on Landauer's non-invasive definition of conductance, with respect to the electrostatic potential drop in the system [18]. The electrostatic potential $\phi(\mathbf{r}, t)$, subject to the boundary condition $\phi(\mathbf{r}, t) \to 0$ as $r \to \infty$ appropriate to our isolated finite system, is a unique functional of the electron density $\rho(\mathbf{r}, t)$ and is thus unambiguously known in the quasi-steady state, or in any other state for that matter. We take it as a physically plausible stipulation that, for large enough electrodes, in a quasi-steady state ϕ will tend (possibly within microscopic Friedel-like oscillations) to well defined values ϕ_L and ϕ_R in the interior of the left and right electrodes of figure 1, respectively, enabling us to define a potential difference $W = \phi_L - \phi_R$, with respect to which we may then define conductance.

Let us now briefly consider processes that would help the system establish a steady state in the present phonon-free, microcanonical picture. The obvious ones are electron–electron interactions. Screening keeps the electron density macroscopically constant, a condition often referred to as charge neutrality. This effective 'incompressibility' of the electron gas in the metal makes current flow somewhat analogous to water flow: local disturbances in the density are not tolerated and heal fast. A further effect of electron–electron interactions, and electronic U-processes in particular, is to produce relaxation of the total electron momentum in the electrodes, a quantity which is not a constant of the motion in our finite system. We suggest, however, an additional intrinsic mechanism that facilitates relaxation in the crucial region of the junction. This mechanism is provided simply by the geometrical constriction experienced by electron wavepackets as they approach the nanojunction [19]. This relaxation mechanism is due to the wave properties of the electron wavefunctions and the resultant uncertainty principle, and has nothing to do with electron–electron interactions. Let us assume that the nanojunction has width w and an electron wavepacket moves into it. The wavepacket has to adjust to the motion appropriate to the given junction geometry in a time $\Delta t \sim \hbar/\Delta E$, where ΔE is the typical energy spacing of lateral modes in the constriction. With $\Delta E \sim \pi^2\hbar^2/m_e w^2$ we find $\Delta t \sim m_e w^2/\pi^2\hbar$. For a nanojunction of width $w = 1$ nm, Δt is of the order of 1 fs. In other words, even in the absence of inelastic effects, the mere presence of the nanojunction would contribute to relaxation of electron momentum. Thus, this effect would seem to suggest that even *without* electron–phonon or electron–electron inelastic scattering a steady state could be reached in a nanoconstriction, with at most mean-field interactions.

All of this analysis would be worthless if in order to do time-dependent transport calculations one had to solve the many-body time-dependent Schrödinger equation. We conclude by showing that in the closed system of figure 1 the true many-body total current is given *exactly* by the total current obtained with TDDFT [8]. This rigorous connection is independent of whether the system has reached a steady state or not. We first note that for a

given initial condition on the many-body wavefunction, a one-to-one correspondence between time-evolving charge density and external potential for the electron gas has been proven only when the density goes to zero at infinity (which is the case for a finite closed system with bound electrons) [8] or for an infinite but perfectly periodic system [20]. Let us then assume that we have solved the time-dependent Kohn–Sham (KS) equations of TDDFT and obtained a set of KS time-dependent one-electron orbitals [8]. Referring to figure 1, we define the KS total current $I_S^{(KS)}$ by

$$I_S^{(KS)}(t) = \int_S \mathbf{j}^{(KS)}(\mathbf{r}, t) \cdot d\mathbf{S} = \int_V d\mathbf{r} \, \nabla \cdot \mathbf{j}^{(KS)}(\mathbf{r}, t) \tag{17}$$

where $\mathbf{j}^{(KS)}(\mathbf{r}, t)$ is the sum of expectation values of the one-electron current-density operator in the populated KS orbitals. For densities that are non-interacting v-representable [21], the charge density of the true many-body system, $\rho(\mathbf{r}, t)$, is the same as the charge density obtained from the KS orbitals, $\rho^{(KS)}(\mathbf{r}, t)$. Furthermore, $\rho^{(KS)}(\mathbf{r}, t)$ and $\mathbf{j}^{(KS)}(\mathbf{r}, t)$ satisfy the continuity equation, just like the many-body density $\rho(\mathbf{r}, t)$ and current density $\mathbf{j}(\mathbf{r}, t)$ in equation (4). Since $\rho^{(KS)}(\mathbf{r}, t) = \rho(\mathbf{r}, t)$, we then have $\nabla \cdot \mathbf{j}^{(KS)}(\mathbf{r}, t) = \nabla \cdot \mathbf{j}(\mathbf{r}, t)$ (even though $\mathbf{j}^{(KS)}(\mathbf{r}, t)$ and $\mathbf{j}(\mathbf{r}, t)$ need not be equal). Hence,

$$I_S^{(KS)}(t) = \int_V d\mathbf{r} \, \nabla \cdot \mathbf{j}^{(KS)}(\mathbf{r}, t) = \int_V d\mathbf{r} \, \nabla \cdot \mathbf{j}(\mathbf{r}, t) = I_S(t) \tag{18}$$

where $I_S(t)$, once again, is the true total many-body electron current, across an arbitrary surface S. The above proof is valid only for a *finite* system because, for an infinite system, we could not have made the transitions between surface and volume integrals above [22]. We note also that in practical calculations, due to the non-local nature of the exchange–correlation kernel in TDDFT, a formulation that relates the external potential directly to the current density may be numerically more efficient. (This formulation is known as time-dependent current-density-functional theory [23].)

In conclusion, we have analysed an alternative point of view of transport in nanoscale systems, in which two large but finite charged electrodes discharge across a nanoscale junction. This microcanonical formulation has key advantages. It permits the notion of a steady state to be expressed in variational form. This variational procedure suggests a link with the notion of entropy and a new source of steady-state noise. It also allows one to show rigorously that the total current in a many-body system is given exactly by the corresponding quantity in TDDFT. This correspondence puts on a rigorous footing the calculation of dynamical transport properties in nanoscale systems by the use of effective one-electron time-dependent Schrödinger equations, without the need to implement scattering boundary conditions. In particular, it allows the investigation of the onset, microscopic nature and dependence on the initial conditions of steady states, and may help tackle open questions about the assumptions of the standard static approach to steady-state conduction.

Acknowledgments

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