Non-equilibrium Transport in dissipative one-dimensional Nanostructures

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We study the non-equilibrium transport properties of a one-dimensional array of dissipative quantum dots. Using the Keldysh formalism, we show that the dots’ dissipative nature leads to a spatial variation of the chemical potential, which in disordered arrays, breaks the invariance of the current, \(I\), under bias reversal. Moreover, the array’s nanoscopic size results in an algebraic low-temperature dependence of \(I\). Finally, we show that a local Coulomb interaction splits the dots’ electronic levels, resulting in a Coulomb blockade, which is softened with increasing dissipation and array size.

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Arrays of metallic \(^1\) or semiconducting \(^3\) quantum dots (QDs) have attracted significant interest due to the unprecedented experimental control in assembling such arrays, and the resulting ability to custom-design their electronic structure and transport properties \(^1\). The continued miniaturization of such arrays, important for many applications in quantum computation \(^7\) and nanoelectronics \(^5\), raises the important question of how the non-equilibrium transport properties in nanoscopic arrays differ from those in mesoscopic ones and how they evolve across different length scales. Nanoscale arrays differ from those in mesoscopic arrays due to the exponential scaling observed in mesoscopic arrays \(^4\) which is ascribed either to some variable-range hopping (VRH) \(^2\) or cotunneling \(^1\) mechanism.

In this Letter, we address the above questions by studying the non-equilibrium transport properties of one-dimensional (1D), nanoscopic arrays of dissipative QDs, as schematically shown in Fig. 1(a). Using the Keldysh Green’s function formalism \(^1\),\(^1\),\(^1\),\(^1\), we demonstrate that the interplay between dissipation, Coulomb interaction, and disorder gives rise to a series of novel non-equilibrium quantum effects. In particular, the dots’ dissipative nature leads to a spatial variation of the chemical potential, \(\mu\), reflecting the array’s resistance, which changes qualitatively with increasing disorder, and breaks the invariance of the current under bias reversal. Moreover, the array’s nanoscopic size yields an algebraic low-temperature dependence of the current in disordered arrays, in contrast to the exponential scaling observed in mesoscopic arrays \(^1\). Finally, we demonstrate that a local Coulomb interaction results in a splitting of a dot’s energy level, which continuously varies with the level’s occupation. This splitting gives rise to a Coulomb blockade in the charge transport, which is softened with increasing dissipation and/or size of the array. These results provide important new insight into the complex non-equilibrium transport properties of nanoscale systems.

We consider a one-dimensional array of \(N\) quantum dots connected to a left and right lead [Fig. 1(a)] whose Hamiltonian is given by \(\mathcal{H} = \mathcal{H}_c + \mathcal{H}_{ph} + \mathcal{H}_{leads}\) where

\[
\mathcal{H}_c = \sum_{j,\sigma} \varepsilon(j) c_{j,\sigma}^\dagger c_{j,\sigma} + U \sum_j \hat{n}_{j,\uparrow} \hat{n}_{j,\downarrow} - \sum_{j,\sigma} t c_{j,\sigma}^\dagger c_{j+1,\sigma} - \sum_{\sigma, \nu = R,L} t_{\nu} c_{\nu,\sigma}^\dagger d_{\nu,\sigma} + \mathcal{H}_{c.c.}\quad (1)
\]

Here \(c_{j,\sigma}^\dagger\) and \(d_{\nu,\sigma}^\dagger\) create an electron with spin \(\sigma\) on dot \(j\) and lead \(\nu\) \((\nu = R, L)\), respectively. We assume for simplicity that on each dot, there is only a single electronic state with energy \(\varepsilon(j)\) that is relevant for charge transport, as is the case in semiconducting QDs \(^3\). \(U\) represents the strength of the local (intra-dot) Coulomb interaction, and \(t\) is the hopping matrix element between neighboring dots. \(t_{\nu}\) describes the coupling of the array to the leads with \(n_{\nu,(R,L)} = 1(N)\). Finally, \(\mathcal{H}_{ph}\) and \(\mathcal{H}_{leads}\) represent the local electron-phonon interaction on each dot and the leads’ electronic structure, respectively.

In order to study the array’s non-equilibrium transport properties, we consider a (symmetric) bias, \(V = \mu_L - \mu_R\),

\[
\text{FIG. 1: (a) One-dimensional array of quantum dots coupled to two leads. (b) First and (c) second order fermionic self-energy corrections due to a local Coulomb interaction.}
\]
across the array arising from different chemical potentials
in the left and right leads with $\mu_R = -\mu_L$. The resulting
current between dots $j$ and $(j + 1)$ is given by:

$$ I_{j,j+1} = -\frac{e}{\hbar} \int_{-\infty}^{+\infty} \frac{d\omega}{2\pi} \text{Re} \left[ \hat{F}_{j,j+1}(\omega) \right]. $$

Here, $\hat{F}$ is the full Keldysh Green’s functions matrix of
the entire array, which accounts for (electron-phonon and
Coulomb) interactions, electronic hopping between dots
and the coupling to the leads. Since scanning tunneling
spectroscopy (STS) on QD arrays \cite{9} reported that intra-
dot interactions are larger than the inter-dot electronic
hopping \cite{9}, we include the former first in the perturba-
tive calculation of $\hat{F}$, in contrast to the limit considered
previously \cite{14,15}. Moreover, STS experiments \cite{8,9}
also reported significant energy broadening (i.e., dissi-
pation) of the dots’ electronic states (likely arising from
electron-phonon coupling) which is only weakly tempera-
ture and energy dependent, and remains substantial even
for $T \to 0$ \cite{9}. We account for the dissipation by intro-
ducing a (phenomenological) lifetime, $\Gamma^{-1}$, of the dots’
electronic states (we take $\epsilon$ to include a possible shift due
to the electron-phonon interaction) \cite{14,15}. The Green’s
function of a single (isolated) dot is then (the effects of
a Coulomb interaction are discussed below)

$$ g_r^{-1}(\omega) = \omega - (\epsilon - \mu) + i\Gamma. $$

Including next the electronic hopping between the dots
and the coupling to the leads, we obtain

$$ \hat{F} = (1 - \hat{g}_r\hat{t})^{-1} \hat{f} (1 - \hat{t}\hat{g}_a)^{-1}. $$

Here, $\hat{g}_{r,a}$ and $\hat{f}$ are the (diagonal) retarded, advanced
and Keldysh Greens function matrices describing the de-
coupled ($t = t_\nu = 0$) dots and leads, respectively, with

$$ \hat{f}(\omega) = 2i \left[ 1 - 2\hat{n}_F(\omega) \right] \text{Im} \hat{g}_r(\omega). $$

$\hat{n}_F$ is a diagonal matrix containing the dots’ and leads’
Fermi-distribution functions, and $\hat{t}$ is the symmetric hop-
ping matrix. While the results shown below were ob-
tained in arrays with $N = 20$ dots, we found qualitatively
similar behavior up to the largest arrays ($N = 150$)
we studied. For concreteness, we take $t = t_{\nu,L} = 0.3E_0$
and $\Gamma = 0.05E_0$, unless otherwise specified.

In order to understand an array’s transport properties,
it is instructive to first consider its (equilibrium) elec-
tronic structure. To this end, we present in Fig. 2(a) the
local density of states (LDOS), $N_j(\omega)$, where $\hat{G}_r = (1 - \hat{g}_r\hat{t})^{-1} \hat{g}_r$ of a non-disordered array
with $N = 20$ dots. The array’s nanoscopic 1D structure gives
rise to a significant curvature of the band (see discussion
below), with the LDOS exhibiting the precursors of the

![FIG. 2: QD array with $N = 20$. (a) $N_j(\omega)$ in equilibrium with $\epsilon(j) = \mu(j) = 0$. (b) $\mu(j)$, $\epsilon(j)$ for $V = 0.4E_0/e$ and different $\Gamma$. $L$ and $R$ refer to the left and right leads, respectively. (c) Resistance $R = V/I$ as a function of $N$. Inset: $R_d$ as a function of $\Gamma$. (d) IV-curve for a non-disordered array and two different disorder realizations with $s = 0.2E_0$.](image-url)
FIG. 3: \(\varepsilon(j), \mu(j)\) for (a) a non-disordered array and disorder case I, and (b) for a strongly disordered array with \(t = \Gamma = 0.01E_0\), and \(s = 0.5E_0\). (c) \(\varepsilon(j), \mu^+_{d}(j)\) for a disordered array under bias reversal. (d) \(I(T)\) for an array with \(N = 150\) at intermediate disorder with \(s = t = 0.3E_0\) and \(\Gamma = 0.05E_0\). Inset: \(I(T)\) for the strongly disordered array of (b).

...port properties, and take \(\varepsilon(j)\) to be Gaussian (disorder-) distributed with average value \(\langle\varepsilon(j)\rangle = 0\) and standard deviation \(s\). The IV-curve for two realizations of intermediate disorder \(t \sim s\), case I and II with \(s = 0.2E_0\), are plotted in Fig. 2(d) \(\varepsilon(j)\) for case I is shown in Fig. 3(a)]. While in both cases, the current is suppressed by disorder, the extent of the suppression depends on the specific disorder realization. The microscopic origin of this suppression lies in the diminished hybridization of electronic states between neighboring dots due to the differences in their energies, \(\varepsilon(j)\). In Fig. 3(a) we present \(\varepsilon(j)\) and \(\mu_d(j)\) for disorder case I together with \(\mu(j)\) for the non-disordered array. Surprisingly, \(\mu_d(j)\) does not differ significantly from \(\mu(j)\), and in particular, does not track the variations in \(\varepsilon(j)\). The reason for this robustness of \(\mu_d\) is that in the intermediate disorder case, electrons can tunnel even through those dots where \(|\varepsilon|\) is large. In contrast, in the strong disorder limit, \(t, \Gamma \ll s\), shown in Fig. 3(d), \(\mu_d(j)\) exhibits step-like changes between those neighboring dots where the variation in \(\varepsilon(j)\) is the largest. This is expected since large spatial variations of \(\varepsilon(j)\) indicate the regions of largest resistance in the array, with concomitant large (potential) changes in \(\mu_d(j)\).

The interplay between disorder and dissipation gives rise to an interesting novel quantum phenomenon in which the variance of the current’s magnitude is broken under bias reversal. To demonstrate this effect, we present in Fig. 3(c) the chemical potentials, \(\mu^+_{d}(j)\), in a dissipative, disordered array for two different bias \(V_\pm = \pm E_0/\epsilon\). Due to the combination of disorder and dissipation, \(\mu^+_{d}\) and \(\mu^-_{d}\) are not related by a spatial symmetry, implying that the corresponding currents are in general different: in Fig. 3(c), the current for positive bias \(V_+\) is \(I_+ = 0.0089eE_0/h\), while for \(V_-\) one finds \(I_- = -0.0261eE_0/h\). We note that the invariance is restored either in a non-disordered dissipative array, or in a non-dissipative \((\Gamma = 0^+)\) disordered array. In the latter case, the current is independent of \(\mu\) and \(\mu^0_d(j) = 0\) again satisfies the spatial symmetry.

The temperature dependence of \(I\) is generally determined by both disorder and interaction effects, with the latter being represented by \(\Gamma\). However, when disorder becomes stronger than dissipation (i.e., \(s \gtrsim \Gamma\)), we find that the current is predominantly determined by the former, and becomes essentially independent of \(\Gamma\). Thus, in calculating \(I(T)\), we can neglect the temperature dependence of interaction effects, i.e., \(\Gamma(T)\), as was previously assumed in Refs. 17, 18. In the limit of intermediate disorder, \(\Gamma \ll t \sim s\), and for \(k_BT \ll s\), \(I(T)\) possesses an algebraic temperature dependence, \(I(T) = I_0 + BT^2\), as shown in Fig. 3(d) for an array with \(N = 150\) dots. This algebraic dependence is the lowest order (in \(T\)) contribution arising from the frequency dependence of the Green’s functions entering Eq. (4), which in turn arises from the finite 1D structure of the array [this frequency dependence is also present in the LDOS of Fig. 2(a)]. In the strong disorder limit, \(\Gamma, t \ll s\) (see inset of Fig. 3[d]), the algebraic temperature dependence of \(I(T)\) at small \(T\) is followed by a more rapid increase at larger temperatures, which can be described by an activated behavior over some limited temperature range. We note that the absence of an exponential VRH temperature scaling 17, 18, is one of the characteristic hallmarks of nanoscale systems. Not only would the algebraic contribution to \(I(T)\) dominate any exponential scaling for \(T \rightarrow 0\), but VRH scaling also assumes that a disordered system is characterized by a single parameter, the localization length, and thus neglects spatial fluctuations in the disorder. These fluctuations are, however, highly relevant in nanoscale systems due to the lack of disorder self-averaging, resulting in different IV-curves for different disorder realizations with the same \(s\) [Fig. 2(d)]. Work is currently under way to identify the characteristic size of the QD array at which disorder self-averaging will lead to a crossover from algebraic to VRH scaling of \(I(T)\).

We next study the effects of a local Coulomb interaction by including the first [Fig. 4(b)] and second order [Fig. 4(c)] fermionic self-energy corrections in the local Green’s functions entering Eq. (4). Based on the experimental STS results of Ref. 19, we take the Coulomb interaction to be weaker than the electron-phonon interaction, and thus take the internal fermionic lines of the diagrams in Figs. 1(b) and (c) to be given by the dissipative Green’s function of Eq. (4). The first order diagram of Fig. 4(b) leads to an energy shift \(\varepsilon_\sigma(j) = \varepsilon(j) + U n_{j,\sigma}\) with \(n_{j,\sigma}\) being the electron occupation and \(\sigma, \bar{\sigma}\) being opposite spin quantum numbers. In contrast, the second order di-
agam, for which an analytical form can be derived, leads to an energy splitting which depends on \( n_{j,\sigma} \); for a completely empty or filled state, the splitting vanishes, while it is the largest for a half-filled state. To demonstrate the evolution of the dot’s electronic structure with \( n_{j,\sigma} \), we present in Fig. 4(a) the LDOS of a single dot [including the self-energy corrections of Figs. 4(b),(c)] for \( n_{j,\sigma} = 0, 0.4, 0.5, \) and \( U = 2E_0 \) (the LDOS for occupation \( 1 - n_{j,\sigma} \) is obtained via \( \omega \leftrightarrow -\omega \)). For \( n_{j,\sigma} = 0 \) \( (n_{j,\sigma} = 1) \), the dot possesses a single, doubly degenerate energy level at \( \omega = -E_0 \) \( (\omega = +E_0) \). As \( n_{j,\sigma} \) increases, this level shifts to larger energies, and a splitting emerges. For \( n_{j,\sigma} = 0.5 \), the state’s spectral weight is predominantly located at \( \omega = \pm E_0 \), i.e., separated by \( U = 2E_0 \), while a small contribution of spectral weight is found at \( \omega = 0 \), which is a direct consequence of \( \Gamma \neq 0 \), and hence vanishes for \( \Gamma \to 0 \). This evolution of the LDOS with \( n_{j,\sigma} \) is identical to that of Ref. 10 for \( \Gamma = 0 \), but qualitatively differs from it for \( \Gamma \neq 0 \). The state’s splitting, which is analogous to the Coulomb interaction driven suppression of the LDOS near the Fermi energy discussed by Efros and Shklovskii 18, is the origin of the Coulomb blockade observed in IV curves. For a single QD with \( \mu = 0 \) and \( n_{j,\sigma} = 0.5 \) [Fig. 4(b)], the dot’s energy level is maximally split, and \( I \) is strongly suppressed until \( V \) exceeds \( U \), thus manifesting the Coulomb blockade. The state’s spectral weight at \( \omega = 0 \) (due to \( \Gamma \neq 0 \)) leads to a non-zero \( I \) even for a \( V < U \). The Coulomb blockade is softened both with increasing \( \Gamma \) [Fig. 4(b)] as well as with increasing number of dots \( N \) in the array, as shown in Fig. 4(c) for \( N = 5 \). In addition, in the latter case, new features appear for \( V < U \) which arise from changes in \( \Delta(j) = \mu(j) - \varepsilon(j) \), where \( \Delta(j) = 0 \) corresponds to \( n_{j,\sigma} = 0.5 \) and thus a maximum splitting of the energy level. As \( \Delta(j) \) increases, this splitting is reduced, and \( I \) increases (and vice versa) as follows from a comparison of Figs. 4(c) and (d). Therefore, the local maximum of \( I \) at \( V_1 \approx 0.7E_0/e \) coincides with that of \( \Delta(j) \), while its sharp increase at \( V_2 \approx 1.2E_0 \) reflects that of \( \Delta(j) \). For \( V > V_2 \), \( I \) decreases slightly due to an increasing separation between the dots’ energy levels, but then increases again as \( V \) approaches the Coulomb gap, \( U \).

In summary, we studied the non-equilibrium transport properties of a 1D array of dissipative QDs. We showed that the QDs’ dissipative nature leads to a spatial variation of \( \mu(j) \), reflecting the resistance of the array. In disordered arrays, this variation breaks the invariance of the current under bias reversal while the array’s nanoscopic size results in an algebraic low-temperature dependence of \( I(T) \). Finally, we showed that a local Coulomb interaction gives rise to a splitting of the dots’ energy levels, and the emergence of a Coulomb blockade, which is softened with increasing dissipation and size of the array.

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[1] T. B. Tran et al., Phys. Rev. Lett. 95, 076806 (2005); T. B. Tran et al., Phys. Rev. B 78, 075437 (2008).
[2] A. Zabet-Khosousi et al., Phys. Rev. Lett. 96, 156403 (2006).
[3] D. Yu et al., Phys. Rev. Lett. 92, 216802 (2004); D. Yu et al., Science 300, 1277 (2003).
[4] A. J. Houtepen, D. Kockmann, and D. Vanmaekelbergh, Nano Letters 8, 3516 (2008).
[5] A.N. Shipway et al., Chemphyschem 1, 18 (2000). (2008).
[6] I.S. Beloborodov et al., Rev. Mod. Phys. 79, 469 (2007).
[7] D. Loss and D.P. DiVincenzo, Phys. Rev. A 57, 120 (1998).
[8] T. Maltezopoulos et al., Phys. Rev. Lett. 91, 196804 (2003).
[9] L. Jdira et al., Phys. Rev. B 77, 205308 (2008).
[10] I.S. Beloborodov et al., arXiv:cond-mat/0501094, unpublished.
[11] J. Schwinger, J. Math. Phys. (N.Y.) 2, 407 (1961); L.P. Kadanoff and G. Baym, Quantum Statistical Mechanics (Benjamin, New York, 1962); J. Rammer and H. Smith, Rev. Mod. Phys. 58, 323 (1986).
[12] L.V. Keldysh, 1964, Zh. Eksp. Teor. Fiz. 47, 1515 (1964) [Sov. Phys.—JETP 20, 1018 (1965)];
[13] C. Caroli et al., J. Phys. C: Solid St. Phys 4, 916 (1971).
[14] A. Mitra et al., Phys. Rev. B 69, 245302 (2004).
[15] Z. Bihary and M. A. Ratner Phys. Rev. B 72, 115439 (2005).
[16] M. Büttiker, Phys. Rev. B 32, R1846 (1985).
[17] N. F. Mott, J. Non-Cryst. Solids 1, 1 (1968).
[18] A. L. Efros and B.I. Shklovskii, J. Phys. C: Solid State Phys. 8, L49 (1975).
[19] Y. Meir et al., Phys. Rev. Lett. 66, 3048 (1991).