Distinguishing Quantum and Classical Transport through Nanostructures

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We consider the question of how to distinguish quantum from classical transport through nanostructures. To address this issue we have derived two inequalities for temporal correlations in nonequilibrium transport in nanostructures weakly coupled to leads. The first inequality concerns local charge measurements and is of general validity; the second concerns the current flow through the device and is relevant for double quantum dots. Violation of either of these inequalities indicates that physics beyond that of a classical Markovian model is occurring in the nanostructure.

PACS numbers: 73.63.−b, 03.65.−w

Quantum coherence of electrons is the essential ingredient behind many interesting phenomena in nanostructures (e.g., [1,2]). Considerable progress has recently been made in the investigation of coherent effects in nanostructures with both charge and transport measurements, (e.g., [2–7]). Typically, Rabi oscillations in the current are taken as a distinctive signature of quantum coherence. However, since even classical autonomous rate equations can admit oscillatory solutions (e.g., [8]), oscillations by themselves cannot be considered as a definitive proof of the existence of quantum coherent dynamics.

In this Letter we formulate a set of inequalities that would allow an experimentalist to exclude the possibility of a classical description of transport through a nanostructure. The inspiration for this comes from the Leggett-Garg inequality [9], which has been described as a single-system temporal version of the famous Bell inequality, also a topic of interest in nanostructures at the present (e.g., [10]). The Leggett-Garg inequality [9] can be summarized as follows. Given an observable $Q(t)$, which is bound above and below by $|Q(t)| \leq 1$, the assumption of: (A1) macroscopic realism and (A2) noninvasive measurement implies the inequality,

$$\langle Q(t_1)Q\rangle + \langle Q(t_1 + t_2)Q(t_1)\rangle - \langle Q(t_1 + t_2)Q\rangle \leq 1, \quad (1)$$

where $Q \equiv Q(t = 0)$. The question of (A1) "realism" [9] can be phrased as *before* we perform the measurement Q on the system [11], does it have a well-defined value? A classical system does, but a quantum system does not.

In the context of nanostructures weakly coupled to contacts, such that a generalized master equation description (e.g., [1,12,13]) is appropriate, we derive and study two inequalities. The first concerns correlations between local charge measurements performed, e.g., by a quantum point contact (QPC) (e.g., [7]). We formulate this inequality in quite general terms, applicable to a range of nanostructures. The charge measurements we consider here are

related, in spirit, to recent work (e.g., [14]) on violations of the Leggett-Garg inequality, using continuous weak measurements on closed systems. However, in contrast to their work, here we are considering a very different situation: ensemble averages of *strong* (i.e., projective) noncontinuous measurements on *open* transport systems. Moreover, our second inequality explicitly focuses on DQDs, providing an inequality for the correlation functions of the current flowing through this widely-studied nanostructure. This second inequality is of particular relevance to DQD experiments along the lines of those of Refs. [3,4], where we predict that violations of both inequalities should occur.

Systems.—We begin by outlining the class of systems studied in this work. We consider nanostructures (Fig. 1)
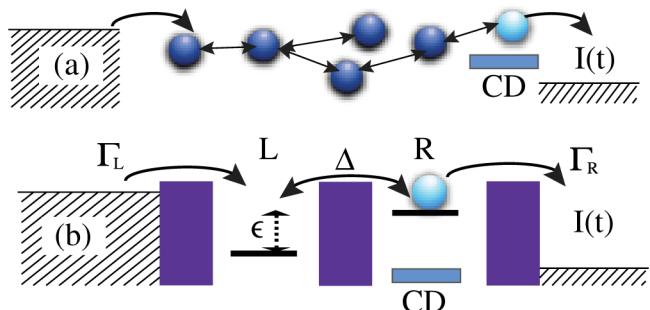
FIG. 1 (color online). (a) A generic single-charge nanostructure. The single-charge can occupy one of N internal states. Local charge occupation of one or more sites is measured using a charge detector (CD) Eq. (2), e.g., a quantum point contact. (b) Schematic of a double quantum dot, the main example we discuss here. $\Gamma_{L/R}$ describe the left/right tunneling rates, Δ is the coherent tunneling amplitude between the left and right dots, and ϵ is the energy difference between the left and right occupation states. Local charge measurements are done on the right (R) state for the charge inequality of Eq. (2), or transport is measured into the collector reservoir for the current inequality of Eq. (4).

weakly coupled to leads such that transport proceeds via sequential tunneling, and we assume a large bias such that higher-order tunneling, level-broadening, and non-Markovian effects can be neglected [12]. We assume strong Coulomb blockade such that the system admits at most one excess electron. In these limits the master equation formalism we apply here, while simple, has been shown to be very accurate by a variety of experiments [2–7]. In general, non-Markovian effects might lead to a violation of these inequalities, so care must be taken to verify one is in these limits. Our system comprises of $(N + 1)$ states: the "empty" state, $|0\rangle$, with no excess electron, and states $|n\rangle$ with a single excess electron in state $n = 1 \ldots N$. The dynamics is described by the generalized master equation $\dot{\rho}(t) = \mathcal{W}[\rho(t)]$, where $\rho(t)$ is the density matrix and the superoperator $\mathcal{W}$ is the Liouvillian. Within a quantum-mechanical description, the density matrix $\rho(t)$ contains coherences, and the Liouvillian has a Lindblad form $\mathcal{W} = \mathcal{W}_0 + \Sigma$, where the coherent evolution of the system is given by $\mathcal{W}_0^{qm} = -i[H, \cdot]$ with H the internal system Hamiltonian, Σ is the self-energy induced by the contact with reservoirs (both electronic and otherwise), and throughout we set $e = \hbar = 1$.

The analogous classical description is a rate equation for the probabilities $P_n(t)$ of finding the system in state n at time t. This rate equation can be written in the same form as above: $\dot{\rho} = \mathcal{W}^{cl}[\rho]$, but now the density matrix ρ only includes diagonal elements [the probabilities $P_n(t) = \rho_{nn}(t)$]. This ρ can be represented as a vector such that the Liouvillian $\mathcal{W}^{cl}$ is a classical rate matrix with $\mathcal{W}_{ij}^{cl} > 0$; $i \neq j$ and $\mathcal{W}_{ii}^{cl} = -\sum_{j \neq i} \mathcal{W}_{ji}^{cl}$. Our general strategy is to explore the behavior allowed by this classical rate equation and use this to derive our inequalities.

Charge inequality.—The first inequality we derive is for localized state measurements. Consider a charge detector which registers the value $Q_n \geq 0$ when the system is in state n (see Fig. 1). Furthermore, let us designate as state N the state for which Q has maximum value: $Q_N = Q_{max}(i)$ *Classical regime*: We assume that for a classical system the charge measurement can be performed noninvasively. An initial state, described by a set of probabilities $P_n(0)$, is fixed and known (actually one only requires knowledge of the relevant expectation values in this state). When noninvasively measuring the charge of a classical Markovian system, we posit that the following inequality holds:

$$|L_Q(t)| \equiv |2\langle Q(t)Q\rangle - \langle Q(2t)Q\rangle| \leq Q_{max}\langle Q\rangle, \quad (2)$$

where $\langle Q\rangle = \sum_k P_k(0)Q_k$ is the expectation value of $Q \equiv Q(t = 0)$, and $\langle Q(t)Q\rangle$ is the charge-charge correlation function. This inequality holds in two regimes: (i) stationarity, where it follows from the original Leggett-Garg [Eq. (1)] by defining the normalized operator $Q = 2Q/Q_{max} - 1$ [15] and taking the stationary expectation value; and (ii) if only a single state contributes to the detection process, i.e., $Q_n = Q_{max}\delta_{nN}$, then Eq. (2) holds for an arbitrary initial state (defined by the

set of probabilities P_k), and not just the stationary state. This latter can be seen as follows. Within both classical and quantum stochastic theory, the charge-charge correlation function can be written as $\langle Q(t)Q\rangle = Q_{max}\Omega_{NN}(t)Q_{max}P_N(0)$, where the "propagator" $\Omega_{NN}(t)$ is an element of the stochastic matrix giving the probability of finding the system in local charge state N a time t after it is in state N. The quantity L_Q can thus be written as (for this single state measurement) $L_Q(t) = Q_{max}^2 P_N(0) \times [2\Omega_{NN}(t) - \Omega_{NN}(2t)]$. If the behavior is classical and Markovian, then the Chapman-Kolgomorov equation for classical rate equations applies [13], and we can write the propagator with argument $2t$ as a decomposition over intermediate states $\Omega_{NN}(2t) = \sum_k \Omega_{Nk}(t)\Omega_{kN}(t)$ to obtain $L_Q(t) = Q_{max}^2 P_N(0)[\Omega_{NN}(2 - \Omega_{NN}) - \sum_{k \neq N}\Omega_{Nk}\Omega_{kN}]$. Hereafter, we suppress the time argument, $\Omega = \Omega(t)$. $L_Q(t)$ is then *maximized* by choosing the propagators such that the system always ends up in state N, i.e., $\Omega_{NN} = 1$, which gives $\max\{L_Q(t)\} = Q_{max}^2 P_N(0) = Q_{max}\langle Q\rangle$. The lower bound is $\min\{L_Q(t)\} = -Q_{max}\langle Q\rangle$. For this single state measurement the inequality holds independent of the initial state, as the dynamics are sufficiently constrained by the Chapman-Kolgomorov equation alone. We first illustrate the charge inequality violation with an example, before continuing to derive the inequality for current measurements. *(ii) Quantum regime*: The transport DQD consists of a dot L, connected to the emitter, and dot R, connected to the collector (see Fig. 1). Assuming weak coupling, large bias, and Coulomb Blockade, the basis of electron states is $\{|0\rangle, |L\rangle, |R\rangle\}$. Its Hamiltonian becomes

$$H = \epsilon(|L\rangle\langle L| - |R\rangle\langle R|) + \Delta(|L\rangle\langle R| + |R\rangle\langle L|), \quad (3)$$

with ϵ the level splitting, and Δ the coherent tunneling amplitude betweens the dots, and with self-energy

$$\Sigma[\rho] = -\frac{1}{2}\sum_{\alpha=L,R}\Gamma_\alpha[s_\alpha s_\alpha^\dagger\rho - 2s_\alpha^\dagger\rho(t)s_\alpha + \rho s_\alpha s_\alpha^\dagger],$$

where $s_L = |0\rangle\langle L|$, $s_R = |R\rangle\langle 0|$, and Γ_L and Γ_R are the left/right tunnelling rates (throughout we set $e = \hbar = 1$). The influence of phonons can also be included in Σ in the standard way [1,16]. The corresponding classical Liouvillian is a 3×3 matrix with elements $\mathcal{W}_{\alpha\beta}^{cl}$; α, $\beta = 0, L, R$. For illustrative purposes, we consider a charge measurement in which the detector only registers when there is an electron in the right-hand QD: $Q = |R\rangle\langle R|$ for which $Q_{max} = 1$. The correlation functions are then calculated from $\langle Q(t)Q\rangle = \text{Tr}[Qe^{\mathcal{W}t}Q\rho_0]$, with ρ_0 the stationary density matrix of the system.

In Fig. 2 we plot $|L_Q(t)|/(Q_{max}\langle Q\rangle)$ as a function of time for a DQD. The behavior is oscillatory, but also damped due to coupling to both the collector and the phonon bath. The shaded region (>1) indicates where $|L_Q(t)|$ violates the inequality of (2). The most prominent violation occurs at the maximum closest to $t = 0$. For these parameters then, no classical Markov description of the system is possible and here, it is quantum oscillations between L and

$|L_Q(t)| / (Q_{max}\langle Q \rangle)$
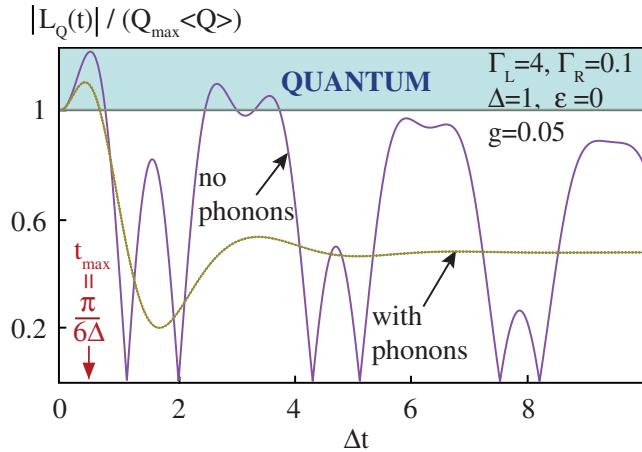
FIG. 2 (color online). The charge inequality $|L_Q(t)|/$ $(Q_{max}\langle Q \rangle)$ Eq. (2) for QPC charge measurements on the R state of a double quantum dot, as a function of dimensionless time Δt. The parameters are shown on the figure, and we have set $Q_{max} = 1$. We chose a dimensionless coupling constant $g = 0.05$ for the bulk phonons [1]. The solid line represents the case of no phonons, and the dashed line includes a phonon bath at temperature $T = 10\hbar\Delta/k_B$, to illustrate how an invasive environment masks the violation. The colored region marks the area of violation of the inequality in Eq. (2).

R states that are responsible for the violation. As we discuss later, the degree of violation can be increased by decreasing Γ_R, which permits the electron to spend a longer time in the DQD. In the limit $\Gamma_R \to 0$ and with $\Gamma_L \to \infty$, such that the empty state may be eliminated, we find the analytic form $L_Q(t)/(Q_{max}\langle Q \rangle) = [\cos(2\Delta t) + \sin^2(2\Delta t)]$, for $\epsilon = 0$. The coherent tunnelling Δ defines the time when the violation is maximum, $t_{max} = \pi/6\Delta$, such that $L_Q(t_{max})/(Q_{max}\langle Q \rangle) = \frac{5}{4}$. The time t_{max} is in agreement with that observed for the Leggett-Garg inequality [9] for a single free qubit with level coupling $\Delta/2$. The effects of a phonon bath are also apparent in Fig. 2, where we have used reasonable bath parameters [1]. Although the oscillations of $L_Q(t)$ are damped, the first and most significant maximum remains.

Current Inequality.—Our second inequality concerns the current $I(t)$ flowing through the transport DQD:

$$|L_I(t)| \equiv |2\langle I(t)I \rangle - \langle I(2t)I \rangle| \leq \Gamma_R \langle I \rangle, \qquad (4)$$

where Γ_R is the coupling to the collector, $I \equiv I(t = 0)$ and $\langle I \rangle$ is the average current of the initial state. Although this second inequality resembles the first one, in Eq. (2) (Γ_R is the maximum instantaneous collector current), its derivation and significance are somewhat different. This is because, in the master equation approach, the current operator translates into a "jump" superoperator and Eq. (4) thus represents an inequality concerning transitions in the system, and not static properties such as the charge under the noninvasive measurements of Eq. (2). For the DQD model in the infinite bias limit, the current superoperator acts as $\mathcal{J}[\rho] = \Gamma_R|0\rangle\langle R|\rho|R\rangle\langle 0|$, such that

the average current is $\langle I \rangle = Tr\{\mathcal{J}\rho\}$ and the correlation function of interest is obtained as $\langle I(t)I \rangle = Tr\{\mathcal{J}e^{\mathcal{W}t}\mathcal{J}\rho_0\}$, where again the stationary distribution is chosen as the initial state. Thus, while the stationary current depends on the occupation of the right dot, the correlation function illustrates the invasive nature of $\mathcal{J}[\rho]$; immediately after the first measurement, the state of the system has changed. In these terms, Eq. (4) can be written as $L_I(t) = Tr\{\mathcal{J}(2e^{\mathcal{W}t} - e^{2\mathcal{W}t})\mathcal{J}\rho_0\}$. In the classical description of the DQD, $\mathcal{J}$ is the 3×3 matrix with elements $\mathcal{J}_{\alpha\beta} = \Gamma_R\delta_{\alpha,0}\delta_{\beta,R}$. Thus using Chapman-Kolgomorov again, we have $L_I(t) = \Gamma_R^2 P_R(0)(\Omega_{R0}(2 - \Omega_{00} - \Omega_{RR}) - \Omega_{RL}\Omega_{L0})$. For a general Markov stochastic matrix, Ω, the maximum of L_I is $2\Gamma_R^2 P_R(0)$. However, the rate equation form $\Omega(t) = e^{\mathcal{W}t}$ furnishes us with a further constraint. Maximizing $L_I(t)$ with respect to time, from $\dot{L}_I = 0$ and $\dot{\Omega} = \mathcal{W}\Omega$, we find that the maximum of L_I occurs when $\Omega_{00} + \Omega_{RR} = 1$ and $\Omega_{R0} = 1$, giving $\max\{L_I\} = \Gamma_R^2 P_R(0) = \Gamma_R\langle I \rangle$. This result relies on the geometry of the DQD, and, in particular, the form of the jump operator and the absence of direct tunnelling from emitter to dot R, i.e., $\mathcal{W}_{R0} = 0$.

Figure 3 illustrates the violation of the current inequality Eq. (4) for the DQD. As with the charge measurement, the quantity $L_I(t)$ is oscillatory and violates the respective inequality with the strongest violation occurring at the first maximum, which is here at a time $t_{max} = \pi/(2\Delta)$. The degree to which this current inequality is violated is of greater magnitude than that for the charge measurement. This is because the current correlator relies on a different propagator from the charge one. Again, in the limit $\Gamma_L \to \infty$, one can eliminate the empty state and find an analytical form. In addition, the $\Gamma_R \to 0$ limit gives
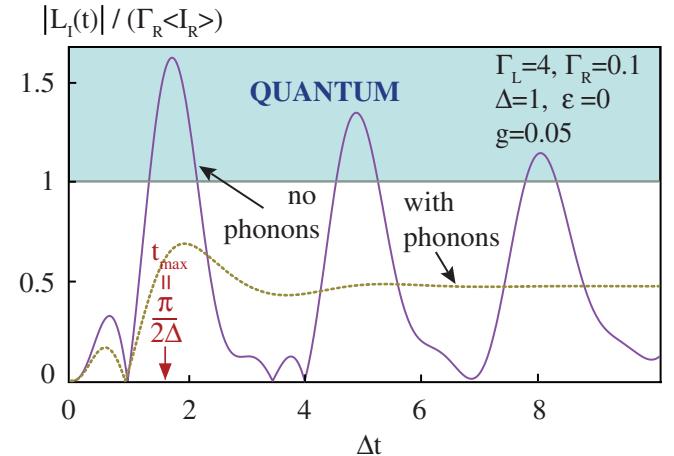
$|L_I(t)| / (\Gamma_R\langle I_R \rangle)$

FIG. 3 (color online). The current inequality $|L_I(t)|/(\Gamma_R\langle I_R \rangle)$ Eq. (4) for current measurements on a double quantum dot, as a function of dimensionless time Δt. The solid line represents the case of no phonons, and the dashed line includes a phonon bath at $T = 10\hbar\Delta/k_B$. The colored region marks the area of violation of the inequality in Eq. (4). The violation of this inequality is more pronounced than that of the charge inequality. The effect, however, is more sensitive to the phonon bath.
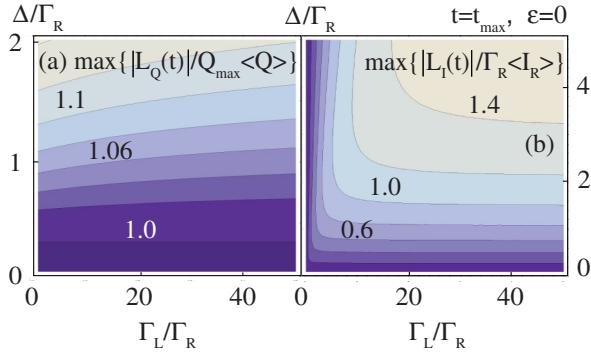
FIG. 4 (color online).   Contour plot of (a) $\max\{L_Q(t)/Q_{\max}\langle Q\rangle\}$ and (b) $\max\{L_I(t)/\Gamma_R\langle I_R\rangle\}$ as a function of the tunneling amplitude Δ/Γ_R and relative rates Γ_L/Γ_R. As discussed in the text, violation requires $\Delta \gtrsim 0.3\Gamma_R$ in (a) and, in (b), $\Delta \gtrsim 1.5\Gamma_R$, $\Gamma_L/\Gamma_R \gg 1$. The phonon bath is excluded.

$L_I(t)/(\Gamma_R\langle I\rangle) = -2\sin^2(\Delta t)\cos(2\Delta t)$. Thus the violation has a maximum of $L_I(t_{\max})/(\Gamma_R\langle I\rangle) = 2$.

Under the above assumptions, the three-state classical DQD Liouvillian cannot produce a violation. However, if these assumptions are relaxed (e.g., allowing $\mathcal{W}_{R0} \neq 0$), a small violation (on the order of $0.003\%\Gamma_R\langle I\rangle$) of the inequality can be observed in extreme parameter regimes. This is in contrast to the L_Q inequality, where no further constraints are required of the Liouvillian. This difference reflects the fact that here the current measurement is essentially a destructive measurement of the state of the nanostructure. An infinitesimal time interval after a positive current measurement is obtained, the electron has left the system, leaving it in the "empty" state. This behavior is implicit in the jump superoperator form of the current measurement.

Figure 4 shows how the maximum degree of violation of both inequalities depends on the parameters of the DQD with no phonons. A violation of the current inequality Eq. (4) requires $\Delta \gtrsim 1.5\Gamma_R$, $\Gamma_L \gg \Gamma_R$, and small detuning ($\epsilon < \Delta$). The violation of the charge inequality Eq. (2) is more resilient, and always occurs unless there is strong over-damping from the reservoir $\Gamma_R \gg \Delta$.

Finally, we note that in practice one needs to measure the correlation functions in Eq. (2) or Eq. (4) on very short time scales (e.g., [7]). Alternatively, one can obtain either correlator from the inverse Fourier transform of the appropriate noise power function. In the transport case [Eq. (4)], one must consider contributions from both particle- (as considered here) and displacement-currents. In principle, one can either choose appropriate gate/junction capacitances to neglect the displacement current contribution, or include them in the definition of Eq. (4), and its subsequent maximization.

Conclusions.—In summary, we have derived two inequalities for nonequilibrium transport in nanostructures: one concerning local charge measurements and the other for current flow through the device. The first is of general validity; the second of relevance to the usual DQD geometry

found in numerous experiments. Violation of either of these inequalities indicates that physics beyond that of a classical Markovian model is occurring in the nanostructure. This may be taken as evidence for quantum oscillations of the electron within the device; or it may indicate a non-Markovian interaction with previously unappreciated degrees of freedom. Finally, we point out that these ideas can be expanded in a number of different directions: to other types of measurements; to inequalities with different time dependencies, as in the original Leggett-Garg work [9]; and to different physical situations for which master equations are appropriate, such as atom-field interactions in quantum optics. This work can also be applied to networks of quantum dots, Cooper pair boxes, and molecules.

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[15] We are grateful to S. Hoyer on this point.
[16] The rates for the electron-phonon interaction are, $\gamma_\pm = [-\epsilon(T/\Delta^2)\mathrm{Coth}[\beta\Delta/2] \pm T/\Delta]\pi/2J[\Delta]$, $\gamma = (T^2/\Delta^2)\mathrm{Coth}[\beta\Delta/2]2\pi J[\Delta]$, where $\Delta = \sqrt{\epsilon^2 + 4T^2}$, $J[\Delta] := g\Delta e^{[-\Delta/\Omega_c]}$, g is the dimensionless coupling strength, $\beta = (k_b T)^{-1}$ and Ω_c is an ultraviolet cutoff.