Resonant Photon-Assisted Tunneling Through a Double Quantum Dot: An Electron Pump From Spatial Rabi Oscillations

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(1 September 1995)

The time average of the fully nonlinear current through a double quantum dot, subject to an arbitrary combination of ac and dc voltages, is calculated exactly using the Keldysh nonequilibrium Green function technique. When driven on resonance, the system functions as an efficient electron pump due to Rabi oscillation between the dots. The pumping current is maximum when the coupling to the leads equals the Rabi frequency.

The spatial coherence of the electronic states in mesoscopic systems is fundamental to understanding their dc transport properties [1]. Recently, it has become possible to experimentally investigate coherent effects in time-dependent transport through mesoscopic systems [2], opening the possibility to study qualitatively new effects which depend in a crucial way on the spatio-temporal coherence of the electronic states of a time-dependent system. While the phenomena of Ref. [2] found a natural explanation within linear response theory [3,4], many time-dependent phenomena, such as electron pumps [5], photon-assisted tunneling [6–10], and lasers [11], necessitate a nonlinear analysis.

In this Letter, we present a fully nonlinear treatment of a novel electron pump based on a spatio-temporal coherence effect: Rabi oscillation between states of a double quantum dot. The double dot is modeled as two spatially separated nondegenerate electronic orbitals, each connected via a tunnel barrier to an electron reservoir (Fig. 1). If the tunneling matrix element w between the orbitals is small compared to their energy difference $\Delta \epsilon = \epsilon_2 - \epsilon_1$, the electrons are highly localized on one orbital or the other, inhibiting transport. However, if the system is driven at a frequency (or subharmonic) corresponding to the energy difference $(\Delta \epsilon^2 + 4w^2)^{1/2}$ between the time-independent eigenstates, the electrons become completely delocalized due to spatial Rabi oscillations. If, as shown in Fig. 1, the reservoirs are biased in such a way that one reservoir can donate electrons to the low-energy orbital ($\mu_L > \epsilon_1$) and the other can accept electrons from the high-energy orbital ($\mu_R < \epsilon_2$), the system will then pump electrons from $\mu_L$ to $\mu_R$ with an efficiency that can be made arbitrarily close to unity, i.e., one electron transferred for each photon absorbed.

We employ the Keldysh nonequilibrium Green function technique [12] to calculate the time-averaged current in response to an arbitrary combination of ac and dc driving voltages, including finite coupling to the leads. The pumping current is found to be a maximum when the coupling to the leads is equal to the Rabi frequency. Furthermore, resonant features in the current are broadened by the coupling to the leads, implying that additional levels of the dot contribute only to a homogeneous background. Importantly, these results for the double-dot system also apply to transport through a double quantum well with negligible interface scattering.

The Hamiltonian of the double-dot system can be expressed as $H(t) = H_{\text{dots}}(t) + H_1$, where

$$H_{\text{dots}}(t) = \sum_{i=1}^{2} \epsilon_i(t)d_i^\dagger d_i + w(d_2^\dagger d_1 + H.c.),$$

$$H_1 = \sum_{\mathbf{k},\ell \in L,R} \epsilon_{\mathbf{k}\ell}c_{\mathbf{k}\ell}^\dagger c_{\mathbf{k}\ell} + \sum_{\mathbf{k},\ell \in \{L,i=1,R,i=2\}} \left(V_{\mathbf{k}\ell} c_{\mathbf{k}\ell}^\dagger d_i + H.c.\right).$$

Here $d_i^\dagger$ creates an electron in the $i$th quantum dot and $c_{\mathbf{k}\ell}^\dagger$ creates an electron of momentum $\mathbf{k}$ in reservoir $\ell$. For simplicity, spin is neglected and the external time dependence is applied only to the dots [13], $\epsilon_{\mathbf{p}}(t) = (-1)^i(\Delta \epsilon + V \cos \omega t)/2$. We do not explicitly treat interactions, but assume that $\Delta \epsilon$ includes contributions both from electrostatic confinement and Coulomb interaction within each dot.

Before discussing transport in a system connected to leads, it is useful first to consider the eigenstates of the closed system of two quantum dots coupled capacitively to an ac voltage source, as described by Eq. (1). The relevant eigenstates of a system such as (1) for which $H(t + 2\pi/\omega) = H(t)$ is a periodic function of time, are the eigenstates

\[ \text{Eqs. (1-2)} \]
of the one-period evolution operator \( U(t + 2\pi/\omega, t) = T \{ \exp[ -\frac{i}{\hbar} \int_t^{t+2\pi/\omega} dt' H(t')] \} \). For the double-dot system, these states have the form [14]

\[
\psi_{i}^{(j)}(t) = \exp(-iE_j t/\hbar) \varphi_{i}^{(j)}(t),
\]

where \( E_j \) is the \( j \)th quasienergy, and \( \varphi_{i}^{(j)}(t + 2\pi/\omega) = \varphi_{i}^{(j)}(t) \) is a Bloch function whose components, \( i = 1, 2 \), give the time-dependent amplitudes on the two quantum dots. The eigenvalue problem defined by Eqs. (1) and (2) must in general be solved numerically [15] because \( [H(t), H(t')] \neq 0 \). However, in the experimentally interesting case of strongly localized dc eigenstates, \( w \ll \Delta \epsilon \), Eqs. (1) and (2) can be solved analytically by expanding \( U(t, t') \) to linear order in \( w \): At the \( N \)-photon resonance, \( N\hbar \omega = \sqrt{\Delta \epsilon^2 + 4w^2} \approx \Delta \epsilon \), one finds for the quasienergies

\[
E_{\pm} = \Delta \epsilon/2 \pm wJ_N(V/\hbar \omega),
\]

where \( J_N \) is the Bessel function of order \( N \). For a small detuning \( \delta \omega \) away from the \( N \)-photon resonance, the occupancy of dot 1 in state \( E_{\pm} \) is \( |\psi_{1,\pm}^{(j)}|^2 = [1 + (m^2 - \pi^2 + \Gamma)^2]^{-1} \), where \( m = \hbar \omega \sin(\pi N \delta \omega/\omega)/[2\pi w J_N(V/\hbar \omega)] \). The quasienergy eigenstates are thus completely delocalized on resonance \( (|\psi_{1,\pm}^{(j)}|^2 = 1/2) \).

Qualitatively, the behavior near resonance for \( w \ll \Delta \epsilon \) can be understood in terms of the hybridization of the electronic orbital on one dot with the \( N \)th sideband of the electronic orbital on the other dot (Fig. 2). For example, in the voltage frame in which \( \epsilon_2 = \Delta \epsilon \) is independent of time, the energy spectrum of the first dot in the absence of tunneling has peaks at \( E = \hbar \omega \) with amplitudes \( J_N(V/\hbar \omega) \). As discussed in Refs. [12,14], when the energy of one of these sidebands coincides with \( \epsilon_2 \), interdot tunneling will hybridize the two orbitals into two delocalized combinations. The effective coupling between orbitals is the product of \( w \) and the sideband amplitude, leading to the energy splitting in [14]. An electron placed on one of the dots at resonance will therefore oscillate back and forth between the dots at the Rabi frequency \( \Omega_{R}/\hbar = 2(w/\hbar)J_N(V/\hbar \omega) \). It should be emphasized that although the quasienergy states are delocalized on resonance, their energy spectrum remains spatially asymmetric, centered near \( \epsilon_1 = 0 \) on dot 1 and near \( \epsilon_2 = \Delta \epsilon \) on dot 2; the delocalized states must be thought of as coherent superpositions of states of the coupled electron-photon system.

The coupling of the double-dot system to the reservoirs is characterized by the parameters \( \Gamma^{LR}(\epsilon) = 2\pi \sum_{k,\ell \in L/R} |\langle \ell | e^{i\delta} \rangle|^2 (\epsilon - \epsilon_\ell) \). In order to obtain an analytic solution for the nonequilibrium time-dependent transport, we consider the case where \( \Gamma^{L}(\epsilon) = \Gamma^{R}(\epsilon) = \Gamma \) is independent of energy. The expectation value of the current through the left barrier can then be expressed using the formalism of Ref. [12] as

\[
J_L(t) = \frac{-2e\Gamma}{\hbar} \int_{-\infty}^{t} dt' \frac{d\epsilon}{2\pi} \text{Im} \left\{ e^{-i\epsilon(t'-t)} \left[ G_{11}^< (t, t') + f_L(\epsilon) G_{11}^> (t, t') \right] \right\},
\]

where \( G_{i\ell}^<(t, t') = \langle \epsilon_i | \langle t' | c_{\ell}(t) \rangle \rangle \) and \( G_{i\ell}^>(t, t') = -i\theta(t-t') \langle \{ c_i^\dagger (t'), c_{\ell}(t) \} \rangle \) are Green functions describing propagation within the double-dot system in the presence of coupling to the leads. The retarded Green function can be expressed simply in terms of the quasienergy eigenstates as \( G_{1\ell}^<(t, t') = -i\theta(t-t') \exp[-\Gamma(t-t')/2] \sum_j \varphi_{i,j}^{(j)}(t) \varphi_{j\ell}^{(j)*}(t') \). Given \( G^r \), the other Green function \( G^c \) can be determined via the Keldysh relation [12], which allows the time-average of \( J_L(t) \) to be expressed in terms of the Fourier components of the quasienergy Bloch functions as

\[
\bar{J} = \frac{e\Gamma}{\pi\hbar} \int df_{L}(\epsilon) \sum_{j,n} \text{Im} \left\{ \frac{|\varphi_{i,j}^{(j)}|^2}{n\hbar \omega + E_j - \epsilon - i\Gamma/2} \right\} \\
- \frac{\Gamma}{2} \sum_{\ell \in L,R} \int df_{L}(\epsilon) \sum_{i',j',n,m} (n\hbar \omega + E_j - \epsilon - i\Gamma/2)[(n + m)\hbar \omega + E_j - \epsilon + i\Gamma/2],
\]

where

\[
\varphi_{i'n}^{(j)} = \frac{\omega}{2\pi} \int_{-\pi/\omega}^{\pi/\omega} dt e^{i\omega t} \varphi_{i'}^{(j)}(t).
\]

Eq. (3) is an exact result for the time-averaged current, valid for arbitrary gate and bias voltages.

Figure 3 shows the time-averaged current for the case \( \mu_L = \mu_R = 0 \) for several ac driving voltages, calculated via Eq. (3) in the limit of zero temperature. A series of peaks in \( \bar{J} \) are evident, occurring at the frequencies \( \omega_N = \Delta \epsilon $$
\((\Delta^2 + 4\omega^2)^{1/2}/N\hbar\), corresponding to the delocalization transitions. At the \(N\)th peak, the dc current flows in response to resonant \(N\)-photon-assisted tunneling: When the electron is on dot 1, it has an energy \(\simeq -\Delta \epsilon/2 < \mu_L\), and can not tunnel into reservoir \(L\). In performing a Rabi oscillation to dot 2, the electron absorbs \(N\) photons, giving it an energy \(\simeq -\Delta \epsilon/2 + N\hbar \omega \simeq \Delta \epsilon/2 > \mu_R\); the electron can thus tunnel from dot 2 to reservoir \(R\). Subsequently, another electron can tunnel from reservoir \(L\) onto dot 1 and the process is repeated, leading to a dc current. Each of the two delocalized quasienergy states contributes independently to this current. Consequently, one can resolve the Rabi splitting between these states by sweeping one of the chemical potentials. For example, the inset to Fig. 3 shows sharp jumps in \(J\) when \(\mu_R\) crosses the two quasienergies for the one-photon resonance of the system, indicated as vertical dashed lines. The spacing \(\delta \mu_R\) between the two jumps is equal to the Rabi splitting \(\Omega_R \simeq 2\omega J_1/(\hbar \omega)\) between the quasienergy eigenstates. The Rabi splitting can thus be resolved by examining the \(I-V\) characteristic of the electron pump.

In order to understand the heights and widths of the resonances in \(J\), it is useful to consider the limit of strongly localized orbitals \(w \ll \Delta \epsilon\) with weak driving \(V \ll \hbar \omega\), so that only resonant processes contribute to the current. Using Eqs. (4) and (6) at zero temperature and the fact that the quasienergy eigenstates are completely delocalized on resonance, one obtains the time-averaged current at the \(N\)-photon resonance,

\[
\bar{J}_{\text{res}} = \frac{e\Gamma}{2\hbar} \frac{\omega_R^2}{\omega_R^2 + \Gamma^2} \sum_{\sigma = \pm 1} \left[ \tan^{-1} \left( \frac{\mu_L - \epsilon_1 + \sigma \omega_R/2}{\Gamma/2} \right) - \tan^{-1} \left( \frac{\mu_R - \epsilon_2 + \sigma \omega_R/2}{\Gamma/2} \right) \right] + \frac{\sigma \Gamma}{2\omega_R} \ln \left( \frac{(\mu_L - \epsilon_1 + \sigma \omega_R/2)^2 + (\Gamma/2)^2}{(\mu_R - \epsilon_2 + \sigma \omega_R/2)^2 + (\Gamma/2)^2} \right). \tag{8}
\]

Eq. (8) shows explicitly that each of the two delocalized quasienergy states contributes independently to the resonant current. For \(\Gamma < \Omega_R\), the logarithmic term in Eq. (8) is negligible, and the arctangents jump rapidly from \(-\pi/2\) to \(\pi/2\) when \(\mu_{L,R}\) cross one of the quasienergies, leading to the sharp jumps in \(J\) shown in the inset to Fig. 3. Eq. (8) predicts that \(\bar{J}_{\text{res}}\) is not a monotonically increasing function of the ac amplitude \(V\), but reaches a maximum for \(V \sim \Delta \epsilon\) then decreases, due to the oscillatory character of the Bessel function in \(\Omega_R\). This behavior is borne out in the exact solution. The inhibition of transport at large ac amplitudes is one feature which distinguishes true photon-assisted tunneling from adiabatic electron transfer [4].

It is instructive to consider several limits of Eq. (8) with regard to the coupling to the leads \(\Gamma\). For \(\mu_L - \epsilon_1, \epsilon_2 - \mu_R \gg \Gamma\), one finds

\[
\bar{J}_{\text{res}} = \frac{e\Gamma}{2\hbar} \frac{\omega_R^2}{\omega_R^2 + \Gamma^2}. \tag{9}
\]

For \(\Gamma \ll \Omega_R\), \(\bar{J}_{\text{res}}\) increases linearly with \(\Gamma\), and the resonances in \(J\) have an intrinsic width of \(\delta \omega_{\text{FWHM}} = 2\Omega_R/\hbar\). \(\bar{J}_{\text{res}}\) obtains a maximum of \(e\Omega_R/4\hbar\) when the tunneling rate to the leads is equal to the Rabi frequency. In the limit \(\Gamma \gg \Omega_R\), \(\bar{J}_{\text{res}} \simeq \omega_R^2/2\hbar\Gamma\) and the resonances are broadened in energy by \(\Gamma\) (and hence in frequency by \(\delta \omega_{\text{FWHM}} = \Gamma/\hbar N\)). In this limit, the photon-assisted tunneling is incoherent because the phase of the electron is randomized on a time-scale short compared to the Rabi oscillations. It is only in this limit, \(\Gamma \gg \Omega_R\), that \(J\) can be calculated via Fermi’s golden rule using the lifetime broadened density of states of the \(N\)th sideband, as in the original calculation of Tien and Gordon [5]. Eq. (4) also implies that for \(V < \hbar \omega\) the current at very high-order resonances is exponentially suppressed compared to the current at the 1-photon resonance, because \(\Omega_R \sim J_N(x) \sim (x/2)^N/N!\). One can therefore generally neglect additional energy orbitals within each quantum dot, since even when in resonance the contribution to the current of an orbital spaced by \(\Delta E\) will be exponentially small in \(N = \Delta E/\hbar \omega\).

We find that the resonances in \(J\) are not broadened at finite temperatures, provided \(k_B T \ll \min |\mu_L - E_j|\). A similar phenomenon in dc resonant tunneling through a double quantum dot was recently observed by van der Vaart et al. [6], underlining the analogy between resonant photon-assisted tunneling between nondegenerate orbitals and dc resonant tunneling through degenerate hybridized orbitals. We find a reduction of \(\bar{J}\) when \(k_B T \gtrsim \min |\mu_L - E_j|\), indicating that a minimum energy \(\sim k_B T\) must be dissipated in order for the electron pump to operate at a maximal rate.

The limit \(\Gamma \ll \Omega_R\) is of particular interest because it allows us to estimate the intrinsic efficiency \(\mathcal{E}\) of the electron pump, defined as the number of electrons transferred per photon absorbed within the nanostructure, neglecting any losses within the external ac voltage source. For general \(\Gamma_L, \Gamma_R \ll \Omega_R\), Eq. (4) becomes \(\bar{J}_{\text{res}} = e\Gamma_L \Gamma_R/(\hbar(\Gamma_L + \Gamma_R)) \equiv J_{\text{MAX}}\). This is the maximum dc current which can be passed through a single pair of hybridized orbitals, and signifies that no electrons traverse the system in the opposite direction. The intrinsic efficiency of the electron pump at the one-photon resonance is thus unity in this limit, since processes involving the net absorption of \(N \neq 1\) photons are
negligible. A lower bound on this efficiency is $\mathcal{E} \geq \bar{J}/J_{\text{max}}$, which becomes an equality as $\Gamma \to 0$. One thus obtains $\lim_{\Gamma \to 0} \mathcal{E} = 1 - \mathcal{O}(w/\Delta \epsilon)^2$ at the one-photon resonance, where the deviation from unity stems from the fact that the dc eigenstates are not completely localized. For the parameters of the inset to Fig. 3, one finds $\mathcal{E} \geq 0.93$, even when pumping up potential gradients $\mu_R - \mu_L \sim \Delta \epsilon$. This remarkably high efficiency stems from the coherent character of photon-assisted tunneling in this system, in which resonant absorption necessarily involves charge transfer. Other electron pumps based on photon-assisted tunneling in single dots [7,8] or intrawell optical excitation [9] do not share this feature.

Our results for the double-dot system can also be applied to vertical transport through double quantum wells. If interface scattering is negligible, each transverse mode is independent and can be modeled by the same Hamiltonian used for the double-dot system. A mode of transverse momentum $k_\perp$ will contribute a current on resonance given by Eq. (8) with $\epsilon_i \to \epsilon_i(0) + \hbar^2 k_\perp^2 / 2m^*$, where $m^*$ is the effective mass. Integrating over transverse modes, including spin, one obtains, in the limit $\mu_L - \epsilon_1(0), \epsilon_2(0) - \mu_R \gg \Gamma$,

$$\bar{J}_{2D} = \frac{e\Gamma}{2\hbar} \left( \frac{\Omega_R^2}{\Omega_R^2 + \Gamma^2} \right) \frac{A m^* [\mu_L - \epsilon_1(0)]}{\pi \hbar^2},$$  

(10)

where $A$ is the area of the 2D electron gas.

In conclusion, we have obtained an exact solution for the time-average of the fully nonlinear current driven through two quantum dots, each coupled to an electron reservoir. The system is found to function as an electron pump capable of transporting electrons up large potential gradients with an efficiency near unity due to resonant photon-assisted tunneling. The pumping current is maximized when the coupling to the leads $\Gamma$ equals the Rabi frequency $\Omega_R$. Since resonances in the current are broadened by the coupling to the reservoirs, the presence of additional energy orbitals contributes only to a homogeneous background. These results also apply to transport through double quantum wells with negligible interface scattering.

We thank Leo Kouwenhoven for raising our interest in this problem, and Peter Wolff for valuable suggestions. One of us (C. A. S.) acknowledges support from the Swiss National Science Foundation.

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FIG. 1. Schematic diagram of the double-quantum-dot electron pump.

FIG. 2. Exact quasienergies of two coupled quantum dots vs. detuning $\epsilon_2$. Here $\epsilon_1 = V \cos \omega t$, with $V = h\omega = 10w$. Note that the quasienergies are defined mod($h\omega$). The electronic states on the dots hybridize and split by $\simeq 2wJ_N(V/h\omega)$, becoming delocalized, when $\epsilon_2$ crosses the $N$th sideband of $\epsilon_1$. 
FIG. 3. Time-averaged current $\bar{J}$ (in units of $J_{\text{max}} = e\Gamma/2\hbar$) through a double quantum dot with $\epsilon_1 = -5$, $\epsilon_2 = 5$, $\Gamma = 0.5$, and ac amplitude $V = 2, 4, 6$ (increasing $J$). Energies are given in units of $w$, the tunneling matrix element between the dots. With $\mu_L = \mu_R = 0$, the system functions as an electron pump due to coherent $N$-photon-assisted tunneling, with resonances at $\omega_N = (\Delta \epsilon^2 + 4w^2)^{1/2}/N\hbar$. Inset: Time-averaged current at the one-photon resonance $\omega = \omega_1$ versus dc bias $\mu_R$, with $\epsilon_1 = -5$, $\epsilon_2 = 5$, $V = 6$, and $\Gamma = 0.05$. The vertical dashed lines indicate the values of the quasienergies, $E_{\pm}$. The jumps, of width $\Gamma$, of $\bar{J}$ at $E_{\pm}$ allow one to resolve the Rabi splitting $|E_+ - E_-| \approx 2wJ_1(V/\hbar\omega) = 0.563$. 