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

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(Received 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.

PACS numbers: 73.50.Pz, 73.20.Dx, 73.20.Jc, 73.40.Gk

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 spatiotemporal coherence of the electronic states of a time-dependent system [3]. While the phenomena of Ref. [2] found a natural explanation within linear response theory [4], many time-dependent phenomena, such as electron pumps [5], photon-assisted tunneling [6–12], and lasers [13], necessitate a nonlinear analysis.

In this Letter, we present a fully nonlinear treatment of a novel electron pump based on a spatiotemporal coherence effect: Rabi oscillation between states of a double quantum dot. The double dot system [9] 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 uphill from μ_L to μ_R.

We employ the Keldysh nonequilibrium Green function technique [14] 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 $H(t) = H_d(t) + H_r$, where

$$H_d(t) = \sum_{i=1}^{2} \epsilon_i(t)\mathbf{d}_i^\dagger\mathbf{d}_i + w(\mathbf{d}_2^\dagger\mathbf{d}_1 + \text{H.c.}) + U_{12}\mathbf{d}_1^\dagger\mathbf{d}_1\mathbf{d}_2^\dagger\mathbf{d}_2\,, \tag{1}$$

$$H_r = \sum_{\mathbf{k},\ell\in L,R} \epsilon_{\mathbf{k}\ell}\mathbf{c}_{\mathbf{k}\ell}^\dagger + \sum_{\mathbf{k},\ell\in\begin{cases} L,i=1 \\ R,i=2 \end{cases}} (V_{\mathbf{k}\ell}\mathbf{c}_{\mathbf{k}\ell}^\dagger\mathbf{d}_i + \text{H.c.})\,. \tag{2}$$

Here, $\mathbf{d}_i^\dagger$ creates an electron in the ith quantum dot and $\mathbf{c}_{\mathbf{k}\ell}^\dagger$ creates an electron of momentum $\mathbf{k}$ in reservoir ℓ. For simplicity, spin is neglected and the external time dependence is applied only to the dots [15], $\epsilon_{i'}(t) = (-1)^{i'}(\Delta\epsilon + V\cos\omega t)/2$. On each dot just one level is active, so the static, on-site interactions can be absorbed into $\Delta\epsilon$ [16]. In contrast, the interdot interaction U_{12}, which is proportional to the interdot capacitance, cannot be absorbed into the single-particle energies.

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

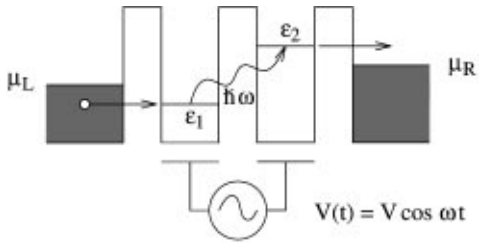

FIG. 1. Schematic diagram of the double-quantum-dot electron pump.

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$H(t + 2\pi/\omega) = H(t)$ is a periodic function of time, are the eigenstates of the one-period evolution operator $U(t + 2\pi/\omega, t) = T\{\exp[-(i/\hbar)\int_t^{t+2\pi/\omega} dt' H(t')]\}$. For the double-dot system, these states have the form [17]

$$\psi_i^{(j)}(t) = \exp(-iE_j t/\hbar)\varphi_i^{(j)}(t), \tag{3}$$

where E_j is the jth quasienergy, and $\varphi_i^{(j)}(t + 2\pi/\omega) = \varphi_i^{(j)}(t)$ is a Floquet function whose components $i = 1, 2$ give the time-dependent amplitudes on the two quantum dots. The eigenvalue problem defined by Eqs. (1) and (3) must in general be solved numerically [18] 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 (3) can be solved analytically by expanding $U(t, t')$ to linear order in w: At the N-photon resonance, $N\hbar\omega = \sqrt{\Delta\epsilon^2 + 4w^2} \simeq \Delta\epsilon$, one finds for the quasienergies

$$E_\pm = \Delta\epsilon/2 \pm wJ_N(V/\hbar\omega), \tag{4}$$

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 $|\psi_1^{(\pm)}|^2 = [1 + (x \mp \sqrt{x^2 + 1})^2]^{-1}$, where $x = \hbar\omega \sin(\pi N\delta\omega/\omega)/2\pi wJ_N(V/\hbar\omega)$. The quasienergy eigenstates are thus completely delocalized on resonance ($|\psi_{1,2}^{(\pm)}|^2 = 1/2$).

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 Nth 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 $E = N\hbar\omega$ with amplitudes $J_N(V/\hbar\omega)$, as discussed in Refs. [6,14]. When the energy of one of these sidebands coincides with ϵ_2, interdot tunneling will hybridize the two orbitals into two delocalized combinations. The effective coupling between orbitals is the product of

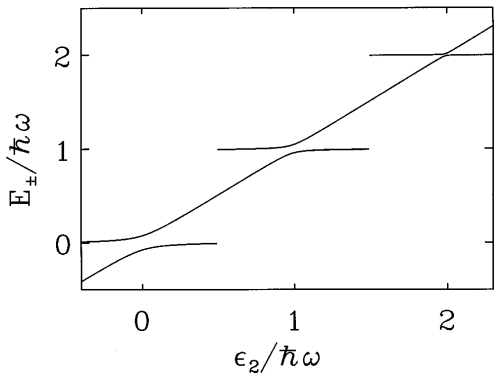

FIG. 2. Exact quasienergies of two coupled quantum dots vs detuning ϵ_2. Here $\epsilon_1 = V\cos\omega t$, with $V = \hbar\omega = 10w$. Note that the quasienergies are defined mod($\hbar\omega$). The electronic states on the dots hybridize and split by $\simeq 2wJ_N(V/\hbar\omega)$, becoming delocalized, when ϵ_2 crosses the Nth sideband of ϵ_1.

w and the sideband amplitude, leading to the energy splitting in (4). 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 $\Gamma^{L/R}(\epsilon) = 2\pi\sum_{\mathbf{k},\ell\in L/R}|V_{\mathbf{k}\ell}|^2\delta(\epsilon - \epsilon_{\mathbf{k}\ell})$. In order to obtain an analytic solution for the nonequilibrium time-dependent transport, we first consider the case where $U_{12} = 0$ and $\Gamma^L(\epsilon) = \Gamma^R(\epsilon) = \Gamma$ is independent of energy. The expectation value of the particle current [19] through the left barrier can then be expressed using the formalism of Ref. [14] as

$$J_L(t) = \frac{-2e\Gamma}{\hbar}\int_{-\infty}^{t} dt' \int \frac{d\epsilon}{2\pi}\,\mathrm{Im}\{e^{-i\epsilon(t'-t)}[G_{11}^<(t,t') + f_L(\epsilon)G_{11}^r(t,t')]\}, \tag{5}$$

where $G_{ii'}^<(t,t') \equiv i\langle \mathbf{c}_{i'}^\dagger(t')\mathbf{c}_i(t)\rangle$ and $G_{ii'}^r(t,t') \equiv -i\theta(t - t')\langle\{\mathbf{c}_{i'}^\dagger(t'), \mathbf{c}_i(t)\}\rangle$ 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 $G_{ii'}^r(t,t') = -i\theta(t - t')\exp[-\Gamma(t - t')/2]\sum_j \psi_i^{(j)}(t)\psi_{i'}^{(j)*}(t')$. Given G^r, the other Green function $G^<$ can be determined via the Keldysh relation [14], which allows the time average of $J_L(t)$ to be expressed in terms of the Fourier components of the Floquet functions as

$$\overline{J} = \frac{e\Gamma}{\pi\hbar}\left[\int d\epsilon\, f_L(\epsilon)\sum_{j,n}\mathrm{Im}\left\{\frac{|\varphi_{1n}^{(j)}|^2}{n\hbar\omega + E_j - \epsilon - i\Gamma/2}\right\} - \frac{\Gamma}{2}\sum_{\ell\in L,R}\int d\epsilon\, f_\ell(\epsilon)\sum_{\substack{i',j,j'\\ n,n',m}}\frac{\varphi_{i'n}^{(j)*}\varphi_{1n'}^{(j)}\varphi_{1,n'+m}^{(j')*}\varphi_{i',n+m}^{(j')}}{(n\hbar\omega + E_j - \epsilon - i\Gamma/2)[(n+m)\hbar\omega + E_{j'} - \epsilon + i\Gamma/2]}\right], \tag{6}$$

where

$$\varphi_{i'n}^{(j)} = \frac{\omega}{2\pi}\int_{-\pi/\omega}^{\pi/\omega} dt e^{inwt}\varphi_{i'}^{(j)}(t). \tag{7}$$

Equation (6) is an exact result for the time-averaged current at $U_{12} = 0$, 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

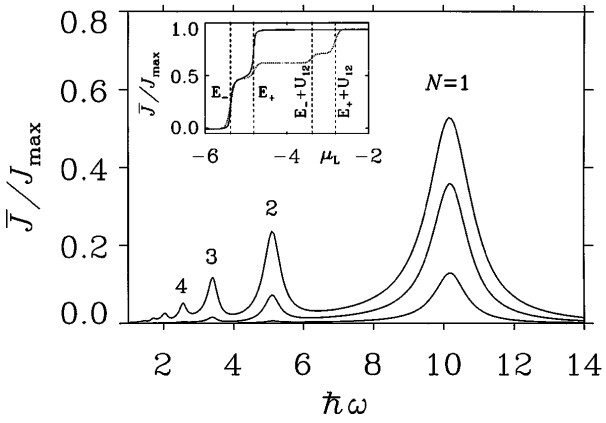

FIG. 3. Time-averaged current $\bar{J}$ (in units of $J_{\max} = e\Gamma/2\hbar$) through a double quantum dot with $\epsilon_1 = -5$, $\epsilon_2 = 5, \Gamma = 0.5$, and ac amplitude $V = 2, 4, 6$ (increasing $\bar{J}$). Energies are given in units of w, the tunneling matrix element between the dots. 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 $\omega = \omega_1$ vs dc bias μ_L, with $V = 6$. Solid curve: $U_{12} = 0, \Gamma = 0.05, k_BT = 0$; dotted curve: $U_{12} = 2, k_BT = 0.05 \gg \Gamma$. The jumps, of width $\max(k_BT, \Gamma)$, of $\bar{J}$ at $E_\pm$ and $E_\pm + U_{12}$ allow one to resolve the Rabi splitting $|E_+ - E_-| \simeq 2wJ_1(V/\hbar\omega) = 0.563$ and the interdot interaction U_{12}.

via Eq. (6) in the limit of zero temperature. A series of peaks in $\bar{J}$, occurring at the frequencies $\omega_N = (\Delta\epsilon^2 + 4w^2)^{1/2}/N\hbar$ corresponding to the delocalization transitions, is evident. At the Nth 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 cannot 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. For $U_{12} = 0$, 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 while keeping $\epsilon_{1,2}$ fixed. For example, the inset to Fig. 3 (solid curve) shows jumps of width Γ in $\bar{J}$ when μ_L crosses the two quasienergies for the 1-photon resonance of the system. The spacing $\delta\mu_L$ between the two jumps is equal to the Rabi splitting $\Omega_R \simeq 2wJ_1(V/\hbar\omega)$.

For $U_{12} > 0$, the two quasienergy states are no longer independent, and the time-dependent transport can no longer be solved exactly. However, one can still calculate $\bar{J}$ in the limit $\Gamma \ll \Omega_R$ using a rate-equation approach [20], and the results for $U_{12} = 2$ are shown for comparison as a dotted curve in the inset to Fig. 3. The current at the one-photon resonance is now reduced by 1/3 for $E_+ < \mu_L < U_{12} + E_-$ due to the inability to populate both quasienergy states simultaneously. There is also a second series of jumps in $\bar{J}$ when μ_L crosses the thresholds $E_\pm + U_{12}$ to inject electrons into state $E_\pm$ when state $E_\mp$ is occupied. Thus both the Rabi splitting and the interdot interaction can be resolved by examining the I-V characteristic of the electron pump.

In order to understand the heights and widths of the resonances in $\bar{J}$, 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 for $U_{12} = 0$,

$$\bar{J}_{\rm res} = \frac{e\Gamma}{2h}\left(\frac{\Omega_R^2}{\Omega_R^2 + \Gamma^2}\right)\sum_{\sigma=\pm1}\left[\tan^{-1}\left(\frac{\mu_L - \epsilon_1 + \sigma\Omega_R/2}{\Gamma/2}\right) - \tan^{-1}\left(\frac{\mu_R - \epsilon_2 + \sigma\Omega_R/2}{\Gamma/2}\right) + \frac{\sigma\Gamma}{2\Omega_R}\ln\left(\frac{(\mu_L - \epsilon_1 + \sigma\Omega_R/2)^2 + (\Gamma/2)^2}{(\mu_R - \epsilon_2 + \sigma\Omega_R/2)^2 + (\Gamma/2)^2}\right)\right]. \quad (8)$$

Equation (8) shows explicitly that each of the two delocalized quasienergy states contributes independently to the resonant current. For $\Gamma \ll \Omega_R$, 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 $\bar{J}$ shown in the inset to Fig. 3. Equation (8) predicts that $\bar{J}_{\rm res}$ 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 Ω_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 [5].

It is instructive to consider several limits of Eq. (8) with regard to the coupling to the leads Γ. For $\mu_L - \epsilon_1 - U_{12}, \epsilon_2 - \mu_R \gg \Gamma$, one finds

$$\bar{J}_{\rm res} = (e\Gamma/2\hbar)\,\Omega_R^2/(\Omega_R^2 + \Gamma^2). \quad (9)$$

For $\Gamma \ll \Omega_R$, $\bar{J}_{\rm res} = e\Gamma/2\hbar$, which is the largest current possible for coupling Γ and signifies that *no* electrons traverse the system in the opposite direction; the resonances in $\bar{J}$ have an intrinsic width of $\delta\omega_{\rm FWHM} = 2\Omega_R/N\hbar$. $\bar{J}_{\rm res}$ 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 $\Gamma \gg \Omega_R$, $\bar{J}_{\rm res} \simeq e\Omega_R^2/2\hbar\Gamma$ and the resonances are broadened in energy by Γ (and hence in frequency by $\delta\omega_{\rm FWHM} = \Gamma/N\hbar$). 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 $\bar{J}$ can be calculated via Fermi's golden rule using the lifetime

broadened density of states of the Nth sideband, as in the original calculation of Tien and Gordon [6]. Equation (9) also implies that for $V < \hbar\omega$ the current at very high-order resonances is exponentially suppressed compared to the current at the one-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 spread by ΔE will be exponentially small in $N = \Delta E/\hbar\omega$.

We find that the resonances in $\bar{I}$ are not broadened at finite temperatures, provided $k_B T \ll \min|\mu_\ell - E_j|$. A similar phenomenon in dc resonant tunneling through a double quantum dot was recently observed by van der Vaart *et al.* [21], underlining the analogy between resonant photon-assisted tunneling between nondegenerate orbitals and dc resonant tunneling through degenerate hybridized orbitals.

The efficiency $\mathcal{E} = (\bar{I}\Delta\mu/e)/(\text{power absorbed})$ of the electron pump is similarly not degraded at finite temperature. Neglecting external losses, the maximal efficiency (obtained as $\Delta\mu \to \Delta\epsilon$) is $\mathcal{E} = 1 - \mathcal{O}(w/\Delta\epsilon)^2$ as $\Gamma \to 0$. The near ideal efficiency stems from the coherent character of photon-assisted tunneling in this system, where resonant absorption necessarily involves charge transfer. Other electron pumps based on adiabatic electron transfer [5], photon-assisted tunneling in single dots [7–9], or intrawell optical excitation [10] 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 $\mathbf{k}_\perp$ will contribute a current on resonance given by Eq. (8) with $\epsilon_i \to \epsilon_i(0) + \hbar^2\mathbf{k}_\perp^2/2m^*$, where m^* is the effective mass. Integrating over transverse modes, including spin, one obtains, in the limit $\mu_L - \epsilon_1(0),\ \epsilon_2(0) - \mu_R \gg \Gamma$,

$$\bar{I}_{2D} = \frac{e\Gamma}{2\hbar}\left(\frac{\Omega_R^2}{\Omega_R^2 + \Gamma^2}\right)\frac{Am^*[\mu_L - \epsilon_1(0)]}{\pi\hbar^2}, \qquad (10)$$

where A is the area of the 2D electron gas. Such an electron pump has recently been experimentally realized by Drexler *et al.* [11], and an analogous effect observed in a multiple-quantum-well structure by Keay *et al.* [12].

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 a potential gradient via resonant photon-assisted tunneling. The pumping current is maximized when the coupling to the leads Γ equals the Rabi frequency Ω_R. Sharp jumps are predicted in the pump I-V characteristic which directly reveal both the Rabi splitting and the interdot Coulomb energy. These results also apply to transport through double quantum wells with negligible interface scattering.

We thank Leo Louwenhoven 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.

Note added.—After submission of this manuscript, we became aware of independent work of Stoof and Nazarov [22], who use a complementary technique to investigate the case $U_{12} = \infty$ in the limit of large bias, and of previous work by Sumetskiĭ and Fel'shtyn [23] on resonant inelastic tunneling through a double quantum well, who independently derived Eq. (10).

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