Nonadiabatic Two-Parameter Charge and Spin Pumping in a Quantum Dot

Matthias Braun and Guido Burkard

Institute of Theoretical Physics C, RWTH Aachen University, D-52056 Aachen, Germany

(Received 1 February 2008; published 15 July 2008)

We study dc charge and spin transport through a weakly coupled quantum dot, driven by a nonadiabatic periodic change of system parameters. We generalize the model of Tien and Gordon to simultaneously oscillating voltages and tunnel couplings. When applying our general result to the two-parameter charge pumping in quantum dots, we find interference effects between the oscillations of the voltage and tunnel couplings. We show that these interference effects may explain recent measurements in metallic islands. Furthermore, we discuss the possibility to electrically pump a spin current in presence of a static magnetic field.

DOI: 10.1103/PhysRevLett.101.036802 PACS numbers: 73.23.Hk, 72.10.Bg, 72.25.Pn, 73.63.Kv

A periodic perturbation of the parameters that determine a quantum dot (QD) and its coupling to external leads can lead to an electric (dc) current from one of the attached leads to the other. This phenomenon is known as charge pumping. For perturbations that are slower than the characteristic charge dynamics of the QD, given by the tunnel rates, the pumping process is adiabatic [1–7]. A remarkable property of adiabatic pumping is that the pumped charge is independent of any details of the pumping cycle, making it possible to realize a current standard for metrology.

Fast perturbations, with frequencies exceeding the tunnel rates, can still lead to pumping effects which in this case are nonadiabatic. Nonadiabatic pumping in QDs has broad applications, reaching from photovoltaic power generation [8] to fundamental studies of fast manipulations of quantum systems, as required, for example, in quantum information processing. The driving force behind nonadiabatic pumping is the absorption of quantized photon energy. Therefore, nonadiabatic pumping [9–16] is often studied as a side-effect of boson-assisted tunneling [17,18]. Operated at frequencies of 1–100 GHz, currents of the order pA to nA can be generated.

Although nonadiabatic pumping is observed in different QD realizations, such as carbon nanotubes [14], or self-assembled dots, the most common realization of a QD is in a two-dimensional electron gas (2DEG). By charging gates, the electron gas beneath can be repelled, and a QD and tunnel barriers can be formed; see Fig. 1.

In nonadiabatic pumping experiments, it is usually assumed that pumping originates from an oscillating voltage of the leads or the energy of the electronic levels in the QD [9–13,15,16,19], while a variation of the tunnel-barrier height is less discussed (see, however, Refs. [20,21]). As in 2DEG QDs, tunnel couplings are exponentially sensitive to voltage changes, an oscillating tunnel barrier may as well be the source of photon energy. In the following, we present an extension of the well-known model by Tien and Gordon [19] that also includes tunnel-barrier oscillations. We find that in the case of two oscillating parameters, one can observe interference between the different sources of pumping, which possibly explains the observed asymmetry of forward and backward currents in Ref. [10].

As a model system, we consider a single-level QD contacted with two leads, described by the Hamiltonian

$$H(t) = \sum_{\sigma} \varepsilon_\sigma a_\sigma^\dagger a_\sigma + U a_\uparrow^\dagger a_\uparrow a_\downarrow^\dagger a_\downarrow + \sum_{r,k,\sigma} \varepsilon_{rk}(t) c_{rk\sigma}^\dagger c_{rk\sigma} + \sum_{r,k,\sigma} (T_{rk}(t) c_{rk\sigma}^\dagger a_\sigma + T_{rk}^\star(t) a_\sigma^\dagger c_{rk\sigma}). \qquad (1)$$

The fermionic operators $a_\sigma^\dagger$ (a_σ) create (annihilate) electrons with spin σ on the dot, while $c_{rk\sigma}^\dagger / c_{rk\sigma}$ act on electrons with orbital quantum number k in the left ($r = L$) and right ($r = R$) lead. Because of its low electrostatic capacity, we assume that double occupation of the dot is suppressed by the associated charging energy U. The third term in Eq. (1) models the contacting leads. The two lead reservoirs are assumed to be noninteracting and in equilibrium at the same temperature; thus, they are characterized by the same Fermi distribution $f(\varepsilon_{rk})$. The last part of the Hamiltonian describes spin-conserving tunneling.

We consider harmonically oscillating voltages at the left and right electrode, leading to a time-dependence of the electron energy $\varepsilon_{rk}(t) = \bar{\varepsilon}_{rk} + eV_r \cos(\omega t)$ in the respective lead. Furthermore, we allow for a time-dependent

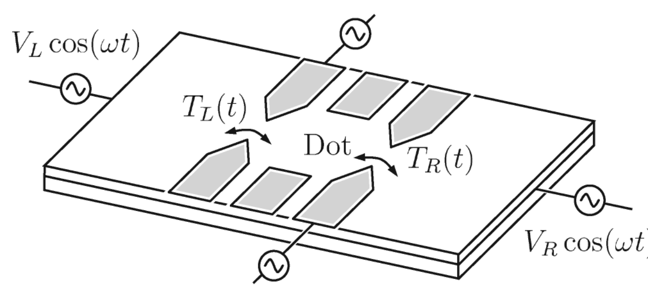

FIG. 1. Typical realization of a quantum dot pump in a two-dimensional electron gas patterned with gates. By a sufficiently fast periodic perturbation of voltages $V_r \cos(\omega t)$ or tunnel couplings $T_{rk}(t) = \bar{T}_{rk}[1 + \alpha_r \cos(\omega t + \eta_r)]$ at the left ($r = L$) or right ($r = R$) contact, nonadiabatic processes can pump charge from one to the other lead.

tunneling amplitude $T_{rk}(t) = \bar{T}_{rk}[1 + \alpha_r \cos(\omega t + \eta_r)]$ [22]. The relative phase of the tunneling amplitude and voltage oscillations on the same side is given by η_r. The tunnel coupling leads to an (time-averaged) intrinsic linewidth $\gamma_r(\varepsilon) = 2\pi \sum_k |\bar{T}_{rk}|^2 \delta(\varepsilon_{rk} - \varepsilon)$ of the QD levels. For simplicity, we will neglect the energy dependence of the linewidth in the following. This single-level model is sufficient as long as the electronic spectrum of the dot is discrete with a level separation exceeding $k_B T$, $\hbar\omega$, γ_r and eV_r. Otherwise, the external driving can lead to dynamics within the QD structure, such as a coherent oscillation of the level populations [23].

Following the standard approach to tunneling in nanostructures [24], we trace out the lead degrees of freedom, and describe the QD by the reduced density matrix $\mathbf{p} = \mathrm{diag}(p_0, p_\uparrow, p_\downarrow)$. The diagonal elements p_χ describe the probability to find the dot empty or occupied with one electron with spin σ. The time evolution of the density matrix is given by a Master equation $\frac{d}{dt}\mathbf{p}(t) = \int_{-\infty}^{t} dt' \mathbf{\Gamma}(t, t'; \varepsilon)\mathbf{p}(t')$, where the elements $\Gamma_{\chi,\chi'}(t, t'; \varepsilon)$ of the kernel describe the tunnel rates from the state χ' at time t' to a state χ at time t.

If the system parameters change faster than the typical time scale of the QD charge evolution, one can assume that the dot density matrix adapts a steady state $\bar{\mathbf{p}} = \mathbf{p}(t') = \mathbf{p}(t)$, which satisfies the equation $0 = \bar{\mathbf{\Gamma}}\,\bar{\mathbf{p}}$, with the end-time-averaged kernel $\bar{\mathbf{\Gamma}}(\varepsilon) = \int_0^{2\pi} d(\omega t)/2\pi \times \int_{-\infty}^{t} dt' \mathbf{\Gamma}(t, t'; \varepsilon)$. In contrast to more sophisticated approaches like Floquet theory [25], the approximation of separating time scales [15,16] covers only the highly nonadiabatic regime. In the following we will only discuss a QD weakly coupled to the external leads; i.e., we expand the kernel in first order in γ_r. This approximation is valid at resonance [26] for $\gamma_r \ll k_B T$. The condition for nonadiabaticity is then directly given by $\hbar\omega > (\gamma_L + \gamma_R)$. The kernel can be decomposed into a left and right part $\mathbf{\Gamma}(t, t'; \varepsilon) = \mathbf{\Gamma}^L(t, t'; \varepsilon) + \mathbf{\Gamma}^R(t, t'; \varepsilon)$, which contain only tunneling processes from and to the respective lead.

The tunnel rates can be calculated by second order perturbation theory, yielding typical expressions as $\Gamma^r_{0,\sigma}(t, t'; \varepsilon) = \sum_k T_{rk}(t) T^\star_{rk}(t') f(\bar{\varepsilon}_{rk}) \exp\{-i \int_{t'}^{t} d\tau[\varepsilon_\sigma - \varepsilon_{rk}(\tau) + i0^+]\} + \mathrm{c.c.}$. As the phase term depends only on the voltage difference between dot and lead, a time dependence of the QD level $\varepsilon \to \varepsilon(t)$ is not qualitatively different from a time-dependent voltage applied to the lead(s). Note that since both the chemical potential and ε_{rk} are shifted by an applied voltage, the argument of the Fermi function is $\bar{\varepsilon}_{rk}$ not influenced by voltage. After the phase averaging, the lowest-order rates transform into

$$\bar{\Gamma}^{r}_{\chi,\chi'}(\varepsilon) = \sum_{n=-\infty}^{\infty} \left| J_n + \frac{\alpha_r e^{i\eta_r}}{2} J_{n+1} + \frac{\alpha_r e^{-i\eta_r}}{2} J_{n-1} \right|^2 \Gamma^{0,r}_{\chi,\chi'}(\varepsilon + n\hbar\omega), \tag{2}$$

where $J_n \equiv J_n(eV_r/\hbar\omega)$ is the Bessel function of the first kind, and $\Gamma^{0,r}_{\chi,\chi'}(\varepsilon)$ is the well-known Golden Rule rate for the time-independent problem [24]. With these modified rates the static occupation probabilities of the QD as well as the current can be calculated [27]. Equation (2) generalizes the result of Tien and Gordon [19] for an oscillating bias voltage, to take into account an oscillating barrier strength.

In the following, we use this general result to discuss charge and spin pumping in QDs in the absence of a bias voltage. Let us first focus on the situation where an electrical dc current is generated by the oscillation of one or more system parameters, in absence of a magnetic field, thus $\varepsilon_\uparrow = \varepsilon_\downarrow = \varepsilon$. For functional clarity we consider only small system parameter changes $\alpha_r \ll 1$ and $eV_r/\hbar\omega \ll 1$; i.e., we calculate only the quadratic response to the system parameter change. The total current through the QD can be written as

$$I = I_{V_L^2} + I_{\alpha_L^2} + I_{V_L \cdot \alpha_L} - (L \to R). \tag{3}$$

If the tunnel amplitudes are constant in time ($\alpha_r = 0$), an applied ac-bias voltage at the QD structure generates the pumped current $I = I_{V_L^2} - I_{V_R^2}$, thus

$$I = I_0 \frac{(eV_L)^2 - (eV_R)^2}{1 + f(\varepsilon)} \times \frac{f(\varepsilon + \hbar\omega) - 2f(\varepsilon) + f(\varepsilon - \hbar\omega)}{(\hbar\omega)^2}, \tag{4}$$

with $I_0 = (e/\hbar)\gamma_L\gamma_R/(\gamma_L + \gamma_R)$. As in real experiments, the capacitances of the left and right tunnel barrier always differ, the voltage drop over the two tunnel barriers will be asymmetric, $V_L \neq V_R$, and a net current is pumped. For oscillation frequencies $\hbar\omega$ much larger or much smaller than $k_B T$, the maximally pumped current scales as $(eV_r/\max[\hbar\omega, k_B T])^2$.

In the absence of an ac voltage applied at the leads ($V_r = 0$), a current $I = I_{\alpha_L^2} - I_{\alpha_R^2}$ can also be driven by an ac signal on one of the gates leading to an oscillation of the left and/or right tunneling amplitudes. The functional form of the current is also given by Eq. (4), where $(eV_r)^2$ is replaced by $(\alpha_r \hbar\omega)^2$.

Nonadiabatic pumping arises from the possibility of the electrons to absorb the photon energy $\hbar\omega$ when tunneling from or to the driven lead [11]. If the QD level lies above the Fermi energy ($\varepsilon > 0$), a lead electron can absorb a photon, and tunnel onto the dot. For $\varepsilon < 0$, the absorption of a photon enables the dot electron to tunnel to the respective lead. A successive tunnel event to (from) the other lead creates a positive (negative) particle current.

The fact that both, pumping via voltage and barrier height, lead to very similar current responses raises the question whether the observed current in the experiments [9–13] is driven by an oscillating voltage or by an oscillation of the tunnel barrier. To discriminate if an oscillating barrier or the oscillating voltage drop causes the current, one needs to look for multiphoton absorption processes. For $eV_r > \hbar\omega$, multiphoton absorption processes become possible, as observed, for example, in photon-assisted tunneling measurements [9]. In contrast to an oscillating voltage, the oscillation of a weak tunnel barrier can (in the lowest-order expansion) lead only to the absorption of a single photon. Two-photon absorption would require a cotunneling event.

Because of mutual cross capacitances, it is rather unlikely that an experimentally applied gate or bias voltage will change only one system parameter. It is more likely that the voltage at the contacting lead as well as the tunnel coupling strength will start to oscillate. As can be seen in Eq. (3), the two pumping processes do not only coexist, but give rise to a mixed two-parameter pumping term

$$I_{V_L\cdot\alpha_L} = 4I_0\cos(\eta_L)\frac{eV_L\alpha_L}{1+f(\varepsilon)}\frac{f(\varepsilon+\hbar\omega)-f(\varepsilon-\hbar\omega)}{2\hbar\omega}. \tag{5}$$

Several differences to the one-parameter pumping currents appear. First, the mixed term depends on the relative phase η_L of the voltage and tunnel amplitude oscillation on one side. This is an indication of interference of the two sources of photon energy $\hbar\omega$. Such an interference between different pumping parameters is also expected in the adiabatic regime [28]. Second, the last term in Eq. (5) resembles the first derivative of the Fermi function, not the second derivative as in the single-parameter pumping case. Therefore, in particular, in the limit of high temperatures $k_BT \gg \hbar\omega$ the maximal pumped current $4I_0eV_L\alpha_L/\max[\hbar\omega, k_BT]$ can significantly exceed the one-parameter pumping currents. Furthermore, the pumped current is no longer (up to a factor 2 due to spin) antisymmetric with respect to the gate voltage. This change of symmetry is probably the most obvious difference.

In Fig. 2 the current I is plotted for an oscillating source lead voltage with amplitude $eV_L = 0.3\hbar\omega$ and $\hbar\omega = 10k_BT$, while the tunneling amplitude of the left barrier is oscillating in phase ($\eta_L = 0$) with an amplitude of 0%, 5%, 25%. Even a tunnel amplitude change of few percent already leads to a noticeable change of the pumped current. Interestingly, for $\eta_L = 0$, which one would expect for an unintentional parameter change, the interference of the two pumping possibilities is constructive if an electron tunnels out of the dot onto the lead and destructive if tunneling from the lead to the dot. Therefore the negative current is enhanced, while the positive one is suppressed.

In the nonadiabatic pumping experiment by Dovinos and Williams [10], an asymmetry in the forward and backward pumping direction was observed, which can be an indication of such a two-parameter pumping situation. In their experiment, a metallic island with a continuous electronic spectrum was exposed to radiation with frequency $\omega = 2\pi \times 2.8$ GHz and a power of 15 μW (stars), 10 μW (diamonds), and 6 μW (triangles), and the pumped current recorded, see Fig. 3. With an analogous calculation one can show, that Eq. (2), with modified golden rule rates $\Gamma^{0,r}_{\chi,\chi'}(\varepsilon)$, also holds in the case of a metallic island. The measured currents in Fig. 3 can be fitted (solid lines) with an electron temperature of 2.5/2.2/2.0 K, a source voltage oscillation amplitude $eV_L = 57/43/30\hbar\omega$ and a left tunnel-barrier variation of 12/7.1/5.5%, respectively. The noticeable asymmetry of the pumped currents in forward and backward direction can be explained by a few-percent variation of the tunnel-barrier strength, making this scenario quite plausible.

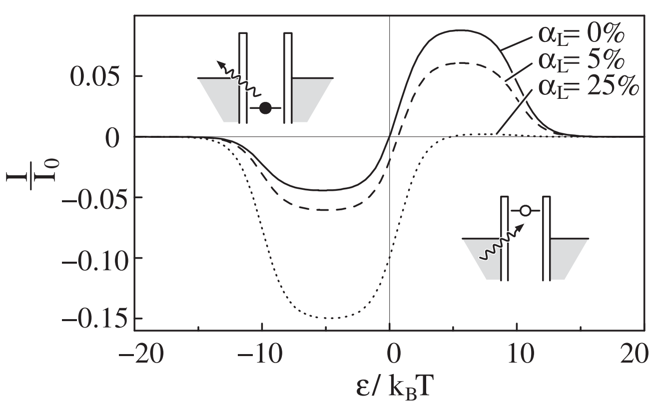

FIG. 2. Current pumped by an ac-voltage $eV_L = 0.3\hbar\omega$, and an in-phase ($\eta_L = 0$) tunneling-amplitude oscillating α_L of 0% (solid), 5% (dashed), and 25% (dotted). Because of interference between these two-photon sources, boson-assisted tunneling onto the dot gets suppressed while tunneling out of the quantum dot is enhanced.

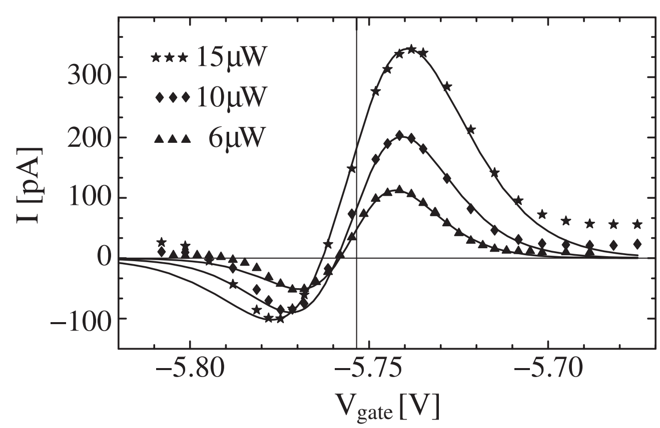

FIG. 3. Pumped currents through a metallic island, exposed to radiation with frequency $\omega = 2\pi \times 2.8$ GHz and a power of 15 μW (stars), 10 μW (diamonds), and 6 μW (triangles) as measured by Dovinos and Williams [10]. The data can be fitted with the parameters temperature 2.5/2.2/2.0 K, source voltage oscillation amplitude $eV_L = 57/43/30\hbar\omega$ and tunnel-barrier variation of 12/7.1/5.5%, respectively.

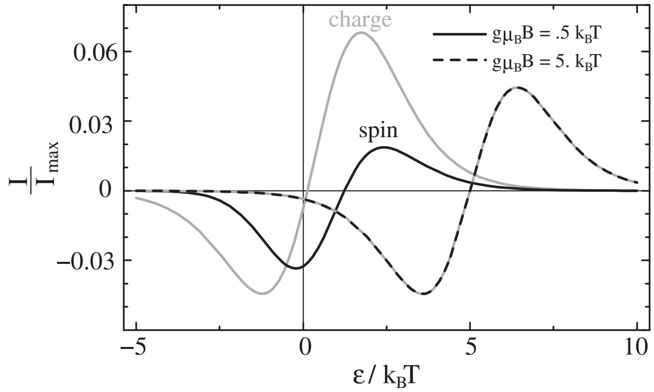

FIG. 4. Particle current (gray) and spin current I_s (black) pumped by oscillating tunnel barrier at the left contact for $\hbar\omega = k_B T$. For low magnetic fields $(g\mu_B B = 0.5 k_B T)$ a net spin current can be generated even in absence of charge current.

Finally, we point out that by applying a static magnetic field, it is also possible to nonadiabatically pump a spin current through a QD in absence of a charge current. The particle current driven by an oscillating left tunnel barrier, carried by electrons with spin σ only, is for $\hbar\omega \ll k_B T$ given by

$$I^{\sigma}_{\alpha_L^2} = (\hbar\omega\alpha_L^2)\frac{I_0}{2}\frac{1 - f(\varepsilon_{\bar\sigma})}{1 + f(\varepsilon_{\bar\sigma})f(\varepsilon_\sigma)}\frac{d^2 f(\varepsilon)}{d\varepsilon^2}\bigg|_{\varepsilon=\varepsilon_\sigma}. \quad (6)$$

For currents of order pA to nA spin-flip processes on the dot can be neglected. In Fig. 4, the particle current (gray) and the spin current $I_s = I^{\sigma}_{\alpha_L^2} - I^{\bar\sigma}_{\alpha_L^2}$ (black) is plotted for different magnetic fields $g\mu_B B = \varepsilon_{\bar\sigma} - \varepsilon_\sigma$ in units of $I_{\max} = \alpha_L^2 I_0$. For the magnetic field $g\mu_B B = 0.5 k_B T$ the charge or particle current shows a node around the Fermi energy, while the spin current bears a maximum. Therefore, a nonadiabatic one-parameter pump can drive a pure spin current without charge current. For larger magnetic fields, $g\mu_B B = 5 k_B T$ for example, only one spin component still participates to transport; therefore, the charge and spin currents become equal. In contrast to spin pumping schemes relying on electron spin resonance in QDs [29], this proposal does not require a strong and fast oscillating magnetic field. Instead, the current is purely driven by an oscillating electric field, and only a static magnetic field is needed for breaking the spin symmetry. With similar requirements electrical spin current generation was demonstrated via adiabatic pumping [7] or by using spin-orbit coupling [30].

In conclusion, we have analyzed an extension of the Tien and Gordon model [19], taking into account simultaneously oscillating tunneling barriers and voltages. By discussing two-parameter charge pumping in QDs and metallic islands, we observed a quantum interference of the tunneling transitions driven by the different pumping parameters. Furthermore, we have discussed the possibility to electrically drive a spin current in absence of a charge current.

This work was supported by DFG SPP Spintronics.

[1] B.L. Altshuler and L.I. Glazman, Science **283**, 1864 (1999); D.J. Thouless, Phys. Rev. B **27**, 6083 (1983).
[2] J. Splettstoesser *et al.*, Phys. Rev. Lett. **95**, 246803 (2005).
[3] P.W. Brouwer, Phys. Rev. B **58**, R10 135 (1998).
[4] H. Pothier *et al.*, Europhys. Lett. **17**, 249 (1992).
[5] M. Switkes *et al.*, Science **283**, 1905 (1999).
[6] L. DiCarlo, C.M. Marcus, and J.S. Harris, Jr., Phys. Rev. Lett. **91**, 246804 (2003).
[7] S.K. Watson *et al.*, Phys. Rev. Lett. **91**, 258301 (2003).
[8] A.J. Nozik, Physica (Amsterdam) **14E**, 115 (2002).
[9] W.G. van der Wiel *et al.*, *Proceedings of the NATO Advanced Study Institute on Strongly Correlated Fermions and Bosons in Low-Dimensional Disordered Systems* (Kluwer Academic, Dordrecht, 2001), p. 43.
[10] D. Dovinos and D. Williams, Phys. Rev. B **72**, 085313 (2005).
[11] L.P. Kouwenhoven *et al.*, Phys. Rev. B **50**, 2019 (1994).
[12] L.P. Kouwenhoven *et al.*, Phys. Rev. Lett. **73**, 3443 (1994).
[13] H. Qin *et al.*, Phys. Rev. B **63**, 035320 (2001).
[14] Y. Shin *et al.*, Phys. Rev. B **74**, 195415 (2006).
[15] T.H. Stoof and Yu.V. Nazarov, Phys. Rev. B **53**, 1050 (1996).
[16] B.L. Hazelzet *et al.*, Phys. Rev. B **63**, 165313 (2001).
[17] T.H. Oosterkamp *et al.*, Phys. Rev. Lett. **78**, 1536 (1997).
[18] R.H. Blick *et al.*, Appl. Phys. Lett. **67**, 3924 (1995).
[19] P.K. Tien and J.P. Gordon, Phys. Rev. **129**, 647 (1963).
[20] M.M. Mahmoodian, L.S. Braginsky, and M.V. Entin, Phys. Rev. B **74**, 125317 (2006).
[21] V. Moldoveanu, V. Gudmundsson, and A. Manolescu, Phys. Rev. B **76**, 165308 (2007).
[22] Only real α_r are considered. A time-dependent phase is equivalent to an applied bias voltage; see Ref. [15].
[23] T. Hayashi *et al.*, Phys. Rev. Lett. **91**, 226804 (2003); J. Gorman, D.G. Hasko, and D.A. Williams, Phys. Rev. Lett. **95**, 090502 (2005).
[24] *Single Charge Tunneling*, NATO ASI Series B: Physics 294, edited by H. Grabert and M.H. Devoret (Plenum, New York, 1992); *Mesoscopic Electron Transport*, edited by L.L. Sohn, L.P. Kouwenhoven, and G. Schön (Kluwer, Dordrecht, 1997).
[25] C.A. Stafford and Ned S. Wingreen, Phys. Rev. Lett. **76**, 1916 (1996); G. Platero and R. Aguado, Phys. Rep. **395**, 1 (2004); S. Kohler, J. Lehmann, and P. Hänggi, *ibid.* **406**, 379 (2005).
[26] Driving softens the resonance condition to the energy range $\pm \max[eV_r, k_B T]$.
[27] A. Thielmann *et al.*, Phys. Rev. B **68**, 115105 (2003).
[28] M. Moskalets and M. Büttiker, Phys. Rev. B **69**, 205316 (2004).
[29] H.-A. Engel and D. Loss, Phys. Rev. Lett. **86**, 4648 (2001); F.H.L. Koppens *et al.*, Nature (London) **442**, 766 (2006).
[30] M. Duckheim and D. Loss, Nature Phys. **2**, 195 (2006); K.C. Nowack *et al.*, Science **318**, 1430 (2007).