Charging Effects in Ultrasmall Quantum Dots in the Presence of Time-Varying Fields

C. Bruder and H. Schoeller
Theoretische Festkörperphysik, Universität Karlsruhe, 76128 Karlsruhe, Germany
(Received 14 June 1993)

The influence of charging effects on time-dependent transport in small semiconductor quantum dots with arbitrary level spectra is studied. Starting from an explicit time-dependent tunneling Hamiltonian, a non-Markovian master equation is derived which is also valid in the nonlinear response regime. The many-body nonequilibrium distribution functions of the dot are calculated and the I-V characteristic of the structure including the displacement currents is obtained. New resonant features show up in the Coulomb oscillations and in the Coulomb staircase, and a new possibility to realize electronic pumps is described.

PACS numbers: 73.40.Gk, 73.20.Dx, 73.50.Fq, 73.50.Mx

The influence of the Coulomb interaction on low-temperature quantum transport through metallic or semiconducting islands (quantum dots) has been the subject of many theoretical and experimental papers (see, e.g., [1] and references therein). It shows up in a variety of interesting effects like, e.g., Coulomb blockade and resonant tunneling phenomena. However, the investigation of time-dependent perturbations and of fluctuations of the electrodynamic environment has started only recently, either neglecting charging effects [2–11] or considering the metallic case for time translational invariant systems [12,13]. An explicit time dependence of the applied voltages introduces a new energy scale $\hbar\omega$ in the problem, and a multitude of new effects is expected to occur, some of them relevant to device applications such as high-frequency oscillators.

In this Letter, we consider an interacting quantum dot with an arbitrary level spectrum subject to an explicit time-dependent classical field. It can describe a periodic modulation of the Fermi energy in the leads (i.e., time-dependent bias voltages) or time-dependent perturbations for the quantum states in the dot. We are especially interested in the effects of the Coulomb interaction in the limit of low tunneling rates but finite level spacing, so that resonant tunneling with thermally broadened line shapes will occur. In contrast to Refs. [12,13], we calculate the complete time-dependent (many-body) distribution function of the dot using a non-Markovian master equation approach. As a consequence we obtain new features in the Coulomb oscillations which cannot be seen in the time-independent case or for continuous level spectra like in metals. Furthermore, for certain asymmetric level structures in the nonlinear response regime, a new mechanism to realize an electronic pump is described.

An important consistency check for our formalism is the fact that the sum of all currents into the system (including the displacement currents) is conserved [9]. We have included the displacement currents in our formalism within the Coulomb-blockade model to make our calculations more realistic. However, since we are mostly interested in the case of low capacitances (where charging effects are important), we do not obtain a significant influence of the displacement currents on the I-V characteristics.

As a model for an interacting quantum dot coupled to two reservoirs by tunnel junctions with capacitances C_L and C_R, we will use the time-dependent tunneling Hamiltonian $H(t) = H_0(t) + V(t) + H_T$. Here,

$$H_0(t) = \sum_{\alpha=L,R} \sum_k \epsilon_{k\alpha}(t) a^\dagger_{k\alpha} a_{k\alpha} + \sum_l \epsilon_l c^\dagger_l c_l \qquad (1)$$

describes noninteracting electrons in the reservoirs $\{\alpha\}$ and in the dot. A time-dependent shift of the Fermi energy of the electrons in the leads has been included in $\epsilon_{k\alpha}(t) = \epsilon^0_{k\alpha} + \Delta_\alpha(t)$. The tunneling part is given by $H_T = \sum_\alpha \sum_{kl} (T^\alpha_{kl} a^\dagger_{k\alpha} c_l + \text{c.c.})$, where T^α_{kl} denotes the tunneling matrix element, and the Coulomb interaction is approximated by the Coulomb-blockade model [14–16],

$$V(t) = E_C[\hat{N}_D + n_0(t)]^2, \qquad (2)$$

corresponding to the equivalent circuit (SET transistor) schematically drawn in Fig. 1. The particle number in the dot is denoted by $\hat{N}_D = \sum_l c^\dagger_l c_l$, $E_C = e^2/2C$ is the charging energy with $C = C_L + C_R + C_g$, whereas $en_0(t) = C_L V_L(t) + C_R V_R(t) + C_g V_g(t)$ is related to the polarization charges produced by the time-dependent voltages of the left and right reservoirs $eV_{L/R}(t) = \mu_{L/R} + \Delta_{L/R}(t)$ as well as a time-dependent gate voltage $eV_g(t) = \mu_g + \Delta_g(t)$ applied to the quantum dot by the capacitance C_g.

The tunneling currents from the left/right reservoir into the quantum dot are given by

$$I^{\text{Tun}}_{L/R}(t) = e\frac{d}{dt}\langle \hat{N}_{L/R}\rangle_{\hat{\rho}(t)}, \qquad (3)$$

where $\hat{N}_\alpha = \sum_k a^\dagger_{k\alpha} a_{k\alpha}$ is the particle number in reservoir α and $\hat{\rho}(t)$ denotes the density matrix of the system at time t. The expectation value $\langle \hat{N}_\alpha \rangle$ can only change due to tunneling processes; therefore, the capacitive currents $\dot{Q}_{L/R}$ ($Q_{L/R}$ is the charge on the left/right tunnel junction; see Fig. 1) are not included in (3) and have to be added separately. The displacement currents are fixed by electrostatic considerations. The total current is

$$I_L = \frac{C_R C_L}{eC}(\dot{\Delta}_L - \dot{\Delta}_R) + \frac{C_g C_L}{eC}(\dot{\Delta}_L - \dot{\Delta}_g) + \frac{C_R + C_g}{C} I^{\text{Tun}}_L - \frac{C_L}{C} I^{\text{Tun}}_R \qquad (4)$$

0031-9007/94/72(7)/1076(4)$06.00
© 1994 The American Physical Society

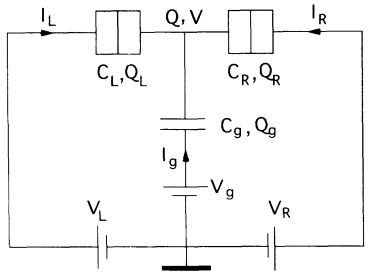

FIG. 1. Equivalent circuit of the double-barrier structure in the static case. C_L, C_R are the capacitances between the dot and the leads, and C_g is the capacitance between the dot and the gate electrode. Q is the excess charge on the dot, and the Q_i are the charges at the capacitor plates.

and I_R is obtained by interchanging L and R. The capacitive current in the gate electrode is denoted by I_g. A direct consequence is the conservation of all currents $I_R + I_L + I_g = 0$, as emphasized in recent works by Büttiker *et al.* [9]. For $C_g = 0$, we obtain at each t $I_L(t) = -I_R(t)$, but the tunneling currents (3) do not have such a property since the charge $Q(t)$ on the island is a function of time [17]. Generally, Eq. (3) gives the total current only if it is used to calculate the time-averaged current [10].

To calculate the tunneling currents (3), we perform a standard time-dependent unitary transformation defined by $U(t) = \exp\{i\int_{-\infty}^{t} d\tau[\sum_\alpha \Delta_\alpha(\tau)\hat{N}_\alpha + \Delta_D(\tau)\hat{N}_D]/\hbar\}$ where $\Delta_D(t) = C^{-1}\sum_{i=L,R,g} C_i\Delta_i(t)$ describes an effective time-dependent perturbation on the dot. The result is $\bar{H}(t) = \bar{H}_0 + \bar{V} + \bar{H}_T(t)$ where $\bar{H}_0$ is the time-independent part of (1) (a constant shift of all energies has been included in ϵ_l), $\bar{V} = E_C\hat{N}_D(\hat{N}_D - 1)$, and $\bar{H}_T(t)$ contains now time-dependent tunneling matrix elements $\bar{T}^\alpha_{kl}(t) = T^\alpha_{kl}\exp\{\frac{i}{\hbar}\int_{-\infty}^{t} d\tau[\Delta_\alpha(\tau) - \Delta_D(\tau)]\}$, and for an arbitrary interaction $\bar{V}$ on the dot the current $I_{L/R}(t)$ will depend only on the difference of the external voltages $\Delta_{L/R}(t) - \Delta_g(t)$. The differences that appear in the Hamiltonian, $\Delta_\alpha - \Delta_D$, are linear functionals of $\Delta_\alpha - \Delta_g$, viz., $\Delta_L - \Delta_D = [(C_R + C_g)(\Delta_L - \Delta_g) - C_R(\Delta_R - \Delta_g)]/C$, and analogously for $\Delta_R - \Delta_D$. Equation (3) is unchanged by the unitary transformation since $\hat{N}_{L/R}$ commutes with $U(t)$. A systematic perturbation expansion in $\bar{H}_T$ then yields

$$I^{\mathrm{Tun}}_\alpha(t) = e\sum_{rr'}\int_{-\infty}^{t} dt'\Gamma_{rr'}(t,t')[N_\alpha(r') - N_\alpha(r)]P_r(t'), \tag{5}$$

where $|r\rangle$ are the eigenstates of $\bar{H}_0 + \bar{V}$ with energy E_r, $P_r(t) = \langle r|\hat{\rho}(t)|r\rangle$ are the diagonal elements of the density matrix, $N_\alpha(r) = \langle r|\hat{N}_\alpha|r\rangle$ is the particle number in reservoir α for the state $|r\rangle$, and $\Gamma_{rr'}(t,t')$ are the time-dependent matrix elements

$$\Gamma_{rr'}(t,t') = \frac{2}{\hbar^2}\mathrm{Re}\left\{\langle r|\bar{H}_T(t)|r'\rangle\langle r'|\bar{H}_T(t')|r\rangle\exp\left[\frac{i}{\hbar}\left(E_r - E_{r'} + i\frac{\Gamma}{2}\right)(t-t')\right]\right\} \tag{6}$$

for the transition r' (at time t') $\to r$ (at time t). Nondiagonal elements of $\hat{\rho}(t)$ are of higher order in $\bar{H}_T$ compared with the diagonal part $P_r(t)$; they can be neglected in the limit of small tunneling rates $\Gamma \ll kT, \Delta E, \omega$. This is equivalent to neglecting initial correlations between the dot and the reservoirs. Furthermore, we have included a finite lifetime $\tau = \frac{2}{\Gamma}$ in (6) due to tunneling, where $\Gamma = \sum_\alpha \Gamma_\alpha$ and $\Gamma_\alpha = 2\pi\sum_k |T^\alpha_{kl}|^2\delta(E - \epsilon_{k\alpha})$ is the tunneling rate into reservoir α (for simplicity, we assume Γ_α to be independent of the state l and the energy). This is equivalent to the well-known inclusion of collisional broadening effects in kinetic equations [18,19].

In the same way, we expand $\dot{P}_r(t)$ in powers of $\bar{H}_T$ and obtain the following non-Markovian master equation

$$\dot{P}_r(t) = \sum_{r'}\int_{-\infty}^{t} dt'\Gamma_{rr'}(t,t')[P_{r'}(t') - P_r(t')]. \tag{7}$$

Equations (5) and (7) are the result of our work in general form. They allow us to treat time-dependent, nonlinear transport problems in the Coulomb blockade regime. We will now approximate the interaction by the Coulomb-blockade model (2), which has turned out to be sufficient to describe the charging effects in small quantum dots qualitatively, provided that the number of electrons is not too small [20].

For weak tunneling and large reservoirs, we can factorize $P_r(t) = P^{\mathrm{eq}}_\phi P_s(t)$ into an equilibrium part P^{eq}_ϕ for the reservoirs (described by the chemical potentials μ_α according to the time-independent part $\bar{H}_0$) and a part $P_s(t)$ for the dot. In order to find the stationary value for $P_s(t)$, we take a periodic modulation $\Delta^\alpha(t) = \Delta^\alpha_0\sin\omega t$ ($\Delta^\alpha = \Delta_\alpha - \Delta_D$) and perform a Fourier transformation $P_s(t) = \sum_{m=-\infty}^{\infty}\tilde{P}_s(m)\exp(-im\omega t)$ with the result

$$\left(N_s - i\frac{m\hbar\omega}{\Gamma}\right)\tilde{P}_s(m) = \sum_l n_l(s_l)\tilde{P}_{s_l}(m) + \left[\sum_n\sum_{\alpha l}\frac{\Gamma_\alpha}{\Gamma}F^\alpha_{nm}(E^{\alpha l}_{N_s - n_l(s)})\,[2n_l(s) - 1][\tilde{P}_s(n) + \tilde{P}_{s_l}(n)] + (m \to -m)^*\right], \tag{8}$$

where $s = |\{n_l(s)\}\rangle$ denotes a state of the dot characterized by the occupation numbers n_l of its single particle levels l, s_l is the state obtained from s by reversing the occupation of level l, i.e., $n_l(s_l) = 1 - n_l(s)$, $N_s = \sum_l n_l(s)$, and

$E_N^{\alpha l} = \epsilon_l + U_N^+ - \mu_\alpha + i\frac{\Gamma}{2}$ where $U_N^+ = 2NE_c$ is the change of the Coulomb energy due to the addition of one particle to the dot. Furthermore, with $\beta = 1/kT$ and $f(E) = (\exp\beta E + 1)^{-1}$, we have defined

$$F_{nm}^{\alpha}(E) = i^{m-n}\sum_k J_{k+n}\left(\frac{\Delta_0^\alpha}{\hbar\omega}\right) J_{k+m}\left(\frac{\Delta_0^\alpha}{\hbar\omega}\right) Y(E + k\hbar\omega), \tag{9}$$

$$Y(E) = \frac{1}{2}f(E) + \frac{1}{4\pi i}\left[\psi\left(\frac{1}{2} + i\frac{\beta E}{2\pi}\right) + \psi\left(\frac{1}{2} - i\frac{\beta E}{2\pi}\right)\right], \tag{10}$$

where J_k and ψ denote the Bessel and digamma functions. Finally, the tunneling current reads

$$\tilde{I}_\alpha^{\mathrm{Tun}}(m) = \frac{e}{\hbar}\Gamma_\alpha\left\{\sum_s N_s P_s(m) - \left[\sum_n\sum_{sl} F_{nm}^{\alpha}(E^{\alpha l}_{N_s - n_l(s)})\,\tilde{P}_s(n) + (m \to -m)^*\right]\right\}. \tag{11}$$

Equation (8) is a linear set of equations for the Fourier components of the probability function $P_s(t)$. It can be solved numerically in a straightforward way and gives directly the time dependence of all many-body distribution functions of the dot together with the tunneling currents (11). It can be applied to the nonlinear response regime (i.e., high bias voltages or high time dependent perturbations), small or high frequencies, arbitrarily strong Coulomb interaction, and discrete level spectra. Furthermore, no assumptions are necessary for the distribution functions of the dot which can differ considerably from the equilibrium value. For the time-independent case, Eq. (8) reduces to the Pauli master equation which has been used in Refs. [20–23] to describe Coulomb oscillations and the Coulomb staircase in small quantum dots. The effect of time-dependent perturbations shows up clearly in the argument $E + k\hbar\omega$ (with $E = E_N^{\alpha l}$) in (9). It coincides with the one-particle excitation (tunneling) spectrum $\epsilon_l + U_N^+ + k\hbar\omega$ of the dot since the electrons can now absorb or emit energy quanta $\hbar\omega$.

We will now discuss several applications of the formalism presented above. As a simple example we consider the dc component of the current through a double-barrier quantum well with two degenerate states ($\epsilon_1 = \epsilon_2 = \epsilon$). Applying an ac gate voltage with frequency ω to the dot ($\Delta_L = \Delta_R = 0$, $\Delta_g \neq 0$), the Coulomb staircase reveals a fine structure (Fig. 2) with additional steps at $\epsilon + 2NE_c + k\hbar\omega$ ($k \neq 0$; $N = 0, 1$) due to the new excitations from the time-dependent perturbation. The dashed line shows the static case. Even for many levels in the dot with a continuous spectrum (where no fine structure due to the finite level spacing can be observed), these side steps will remain. In the same way, the Coulomb oscillations will also show additional resonances at a distance $n\hbar\omega$ ($n = 0, \pm1, \pm2, \ldots$) from the conventional main peaks at $\epsilon + 2NE_c$ ($N = 0, 1$). However, the situation changes drastically if the two single particle levels are no longer degenerate ($\epsilon_1 < \epsilon_2$). In the static case, we get two Coulomb peaks at ϵ_1 and $\epsilon_2 + 2E_c$ (inset Fig. 2, dashed curve); i.e., the excitations ϵ_2 and $\epsilon_1 + 2E_c$ are hidden without time-dependent perturbations. The reason is that on lowering the gate voltage (for small bias voltage and particle number $N = 0$), the first excitation will be $N = 0 \to N = 1$, occupying the level $l = 1$; i.e., the first peak is at ϵ_1. After this transition, the dot contains exactly one particle which cannot escape due to the Coulomb blockade. The next possible excitation for $N = 1 \to N = 2$ is at energy $\epsilon_2 + 2E_c$ and the ones with ϵ_2 and $\epsilon_1 + 2E_c$ are not visible. In the time-dependent case, the particle in the dot can absorb modulation quanta of energy $\hbar\omega$ and thus it can leave the dot or occupy the level $l = 2$. Therefore, the complete tunneling spectrum will show up in the Coulomb oscillations (see inset Fig. 2) in contrast to the static case. In other words, the effect of a finite charging energy in the presence of time-dependent perturbations is not only a simple shift of the resonances at ϵ_l ($l = 1, 2, \ldots$) by $(l-1)2E_c$ but instead a splitting into many peaks at $\epsilon_l + 2NE_c$ ($N = 0, 1, 2, \ldots$) where the amplitude is maximal at $N = l - 1$ and the peaks with $N = l - 1 + k$ correspond to absorption (emission) of energy quanta $k\hbar\omega$. As in the case with the Coulomb staircase, these fine structures are not observable for a

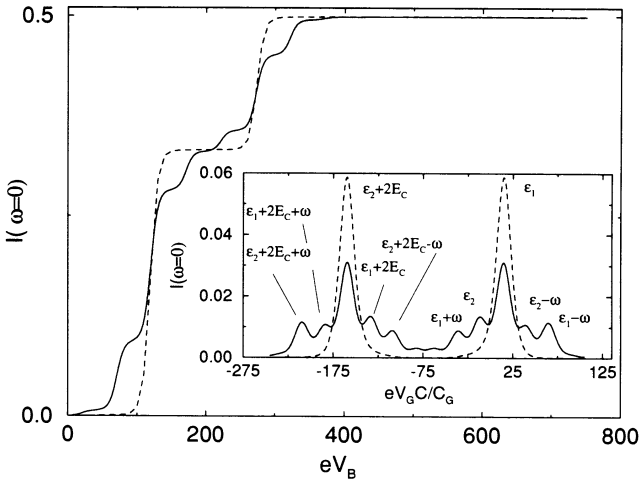

FIG. 2. dc current versus eV_{Bias} for $T = 5$, $E_C = 75$, $\omega = 50$, $\Delta^L = \Delta^R = 0$ (dashed line), $\Delta^L = \Delta^R = 50$ (solid line), and $C_L = C_R = 10^{-4}$. Energies are measured in units of the total tunneling rate, $\Gamma = \Gamma_L + \Gamma_R$, where $\Gamma_L = \Gamma_R$, currents in units of $e\Gamma/\hbar$, frequencies in units of $\Gamma/\hbar$, and capacities in units of e^2/Γ. Inset: dc current versus eV_gC/C_g for two nondegenerate levels with $\epsilon_{1,2} = \mp 12.5$; other parameters as before.

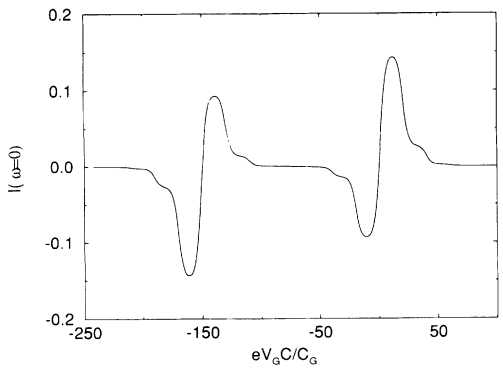

FIG. 3. dc current versus eV_gC/C_g at zero dc bias voltage and $\Delta^L = 30$, $\Delta^R = 0$. Here, $T = 2$, $\omega = 20$, and the other parameters are as before. The structure acts as an electron pump.

continuous spectrum in the dot where only the satellite peaks shifted by $k\hbar\omega$ will remain.

Another possibility is to apply an ac signal to the reservoirs such that $\Delta_L - \Delta_D \neq 0, \Delta_R - \Delta_D = 0$. Even for zero dc-bias voltage, we obtain a positive or negative dc current through the quantum well due to the absorption or emission of energy quanta in the left reservoir. The effect is possible only if there is an asymmetry of level(s) in the dot with respect to the chemical potential ($\mu = \mu_L = \mu_R$) of the reservoirs: if the level closest to μ is above μ, there will be a positive current, and vice versa. Consequently, the system acts like an electron pump in this case and the form of the Coulomb oscillation peaks are changed dramatically as illustrated in Fig. 3. This example of a nonlinear response is a generic result as can be seen by taking the $m = 0$ component of (11) for the case of a single nondegenerate level in the dot and expanding to second order in Δ^α. For $\omega \to 0$, we obtain $I_L^{\rm dc} \sim f''(\epsilon_0 - \mu)(\Delta^L)^2$. The second derivative of the Fermi function will produce the positive and negative regions seen in Fig. 3.

In conclusion, we have presented a formalism that allows us to calculate the frequency-dependent I-V characteristics of strongly interacting quantum dots. It takes into account nonequilibrium many-body distribution functions of the dot, it is not restricted to the linear response regime, and it is exact in the limit of small tunneling rates. We have also included the contribution of the displacement currents caused by the capacities and gate electrodes. The presence of five energy scales leads to a multitude of physical situations that will be studied in future work.

It is a pleasure to thank G. Schön for initiating this work and for many helpful discussions. We would also like to acknowledge stimulating discussions with A. Groshev, F.W. Hekking, Yu.V. Nazarov, A.A. Odintsov, and especially with M. Büttiker. This work was supported by the Swiss National Science Foundation (H.S.) and by the "Deutsche Forschungsgemeinschaft" as part of "Sonderforschungsbereich 195."

[1] *Single Charge Tunneling*, edited by H. Grabert and M. Devoret, NATO ASI Ser. B, Vol. 294 (Plenum, New York, 1991).
[2] F. W. Hekking, Yu. V. Nazarov, and G. Schön, Europhys. Lett. **14**, 489 (1991); **20**, 255 (1992).
[3] P. Johansson, Phys. Rev. B **41**, 9892 (1990).
[4] L. Y. Chen and C. S. Ting, Phys. Rev. Lett. **64**, 3159 (1990).
[5] H. C. Liu, Phys. Rev. B **43**, 12 538 (1991).
[6] E. Runge and H. Ehrenreich, Phys. Rev. B **45**, 9145 (1992); E. Runge and H. Ehrenreich, Ann. Phys. (N.Y.) **219**, 55 (1992).
[7] H. M. Pastawski, Phys. Rev. B **46**, 4053 (1992).
[8] Y. Fu and S. C. Dudley, Phys. Rev. Lett. **70**, 65 (1993).
[9] M. Büttiker and H. Thomas, in *Quantum-Effect Physics, Electronics and Applications*, edited by K. Ismail *et al.*, Institute of Physics Conf. Proc. No. 127 (Institute of Physics, Bristol, 1992); M. Büttiker, A. Prêtre, and H. Thomas, Phys. Rev. Lett. **70**, 4114 (1993).
[10] N. S. Wingreen, A. Jauho, and Y. Meir, Phys. Rev. B **48**, 8487 (1993).
[11] S. Datta and M. P. Anantram (unpublished).
[12] G. Ingold and Yu. V. Nazarov, in *Single Charge Tunneling* (Ref. [1]), p. 21.
[13] A. A. Odintsov, G. Falci, and G. Schön, Phys. Rev. B **44**, 13 089 (1991).
[14] I. O. Kulik and R. I. Shekhter, Zh. Eksp. Teor. Fiz. **68**, 623 (1975) [Sov. Phys. JETP **41**, 308 (1975)].
[15] D. V. Averin and K. K. Likharev, in *Mesoscopic Phenomena in Solids*, edited by B. Altshuler, P. A. Lee, and R. A. Webb (North-Holland, Amsterdam, 1991).
[16] L. I. Glazman and K. A. Matveev, Zh. Eksp. Teor. Fiz. **98**, 1834 (1990) [Sov. Phys. JETP **71**, 1031 (1990)].
[17] For $C_L = C_R + C_g$ the tunneling part of (4) reduces to $(I_L^{\rm Tun} - I_R^{\rm Tun})/2$, an expression sometimes referred to as the Ramo-Shockley theorem [4].
[18] H. Schoeller (to be published).
[19] P. Lipavsky, F. S. Khan, and J. W. Wilkins, Phys. Rev. B **43**, 6665 (1991).
[20] W. Pfaff, D. Weinmann, W. Häusler, B. Kramer, and U. Weiss (to be published).
[21] C. W. J. Beenakker, Phys. Rev. B **44**, 1646 (1991).
[22] H. van Houten, C. W. J. Beenakker, and A. A. Staring, in *Single Charge Tunneling* (Ref. [1]).
[23] D. V. Averin, A. N. Korotkov, and K. K. Likharev, Phys. Rev. B **44**, 6199 (1991).