

Tunneling spectroscopy of quantum charge fluctuations in the Coulomb blockade

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We present a theory of Coulomb blockade oscillations in tunneling through a pair of quantum dots connected by a tunable tunneling junction. The positions and amplitudes of peaks in the linear conductance are directly related, respectively, to the ground-state energy and to the dynamics of charge fluctuations. We study analytically both strong and weak interdot tunneling. As the tunneling decreases, the period of the peaks doubles, as observed experimentally. In the strong tunneling limit, we predict a striking power-law temperature dependence of the peak amplitudes.

The charge of an isolated conductor is quantized in units of the elementary charge e . Surprisingly, even if the conductor is connected to a particle reservoir by a tunnel junction, its charge can still be almost quantized at low temperatures, a phenomenon known as the Coulomb blockade.^{1,2} The simplest system which shows a Coulomb blockade consists of a small metallic grain separated from a bulk lead by a thin dielectric layer. An electron tunneling through the layer inevitably charges the grain, thus increasing its energy by $E_C = e^2/2C$, where C is the capacitance of the grain. At temperatures $T \ll E_C$ a negligible fraction of the electrons in the lead have an energy of order E_C , and one might expect that no tunneling into the grain is possible. More careful consideration shows, however, that even at $T=0$ the electrons can tunnel to the virtual states in the grain, thus lowering the ground-state energy of the system.³ Due to this virtual tunneling, the average charge of the grain is no longer quantized and acquires a correction proportional to the conductance of the barrier. Charge quantization is completely destroyed when the conductance of the barrier approaches e^2/h .⁴⁻⁶ Unfortunately a direct measurement of the equilibrium properties, such as the average grain charge, comprises a challenging, though not impossible,⁷ experiment.

Several recent experiments⁸⁻¹² have probed some equilibrium properties of a Coulomb blockade system by measuring the tunneling conductance through a pair of coupled quantum dots. Focusing on the experiment of Waugh *et al.*,⁸ we develop in this paper a quantitative theory of the linear conductance in such a system; in particular, we predict the gate voltage and temperature dependence of the conductance.

We will study the properties of a two-dot system which is schematically shown in Fig. 1. The electrostatic energy of this system is a quadratic form of three variables: the charges of each dot, eN_1 and eN_2 , and the gate voltage V_g . In the general case, this energy can be written as

$$U(N_1, N_2) = E_C(N_1 + N_2 - 2X)^2 + \tilde{E}_C[N_1 - N_2 + \lambda(N_1 + N_2) - \alpha X]^2. \quad (1)$$

Here X is a dimensionless parameter proportional to V_g , and the constants E_C , \tilde{E}_C , λ , and α are determined by the geometry of the system.¹³ In Eq. (1) we expressed the energy in terms of the total number of particles in the two dots $N_1 + N_2$ and the relative number $N_1 - N_2$. These variables are convenient because the former is constant in the absence of tunneling into the leads, and the latter describes charge fluctuations between the dots. In this paper we will concentrate on the case of symmetric geometry of the system, corresponding to $\lambda = \alpha = 0$, which is apparently the case in the experiment.⁸

We first discuss the location of the peaks in the conductance G of the double-dot system. In the limit of very small interdot conductance, $G_0 \ll e^2/h$, the peaks in G occur when

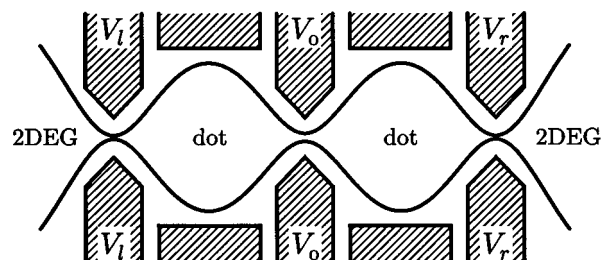


FIG. 1. Schematic view of the double quantum dot system. The dots are formed by applying negative voltage to the gates (shaded); the solid line shows the boundary of the 2D electron gas (2DEG). V_l and V_r create tunnel barriers between the dots and the leads while V_0 controls the transmission coefficient through the constriction connecting the dots.

the electrostatic energy is degenerate; that is, when $U(n+1, n)$ equals either $U(n, n)$ or $U(n+1, n+1)$, where n is an arbitrary integer. As a result we find peaks at the following sequence of gate voltages:

$$X^* = n + \frac{1}{2} \pm \frac{1}{4} \left(1 - \frac{\tilde{E}_C}{E_C} \right). \quad (2)$$

Weak electrostatic coupling between the dots corresponds to $E_C - \tilde{E}_C \ll E_C$. In this case the two peaks with the same n in sequence (2) merge. This limit is observed in the experiment.⁸

If all the junctions shown in Fig. 1 have small conductances, then the charges of both grains are well defined. In order to study the effect of quantum fluctuations on the ground-state energy of the system, it is enough to increase only the interdot conductance G_0 , keeping all other conductances small. Under these conditions, the sum $\mathcal{N} \equiv N_1 + N_2$ is constant and can still be treated as a c number, but $N_1 - N_2$ starts to fluctuate. These fluctuations change the ground-state energy, denoted $E_{\mathcal{N}}$, from the electrostatic estimate. The peaks in G now occur at gate voltages X , where $E_{\mathcal{N}}(X) = E_{\mathcal{N}+1}(X)$. In order to find $E_{\mathcal{N}}$, one should consider the quantum mechanical problem with the Hamiltonian,

$$H = H_0 + H_T + E_C(\mathcal{N} - 2X)^2 + 4\tilde{E}_C \left(\hat{N}_1 - \frac{\mathcal{N}}{2} \right)^2. \quad (3)$$

Here the terms H_0 and H_T describe, respectively, free electrons in the dots and tunneling between the dots; we have replaced N_2 by $\mathcal{N} - N_1$, and N_1 should be treated as a quantum operator \hat{N}_1 . Typically the size of the dots exceeds the effective Bohr radius ($\sim 100 \text{ \AA}$ for GaAs), and therefore the level spacing for electrons in the dots is much smaller than the charging energy. We will neglect the level spacing and assume a *continuous* spectrum in H_0 , in contrast to Refs. 14 and 15. In the continuous model, the noninteracting part of the Hamiltonian, $H_0 + H_T$, does not depend on the total number of particles \mathcal{N} .

In the Hamiltonian (3) the parameter \mathcal{N} is an integer. However, formally we can consider Eq. (3) at any \mathcal{N} , enabling us to relate the ground-state energy $E_{\mathcal{N}}$ to the average value $\bar{N}_1(\mathcal{N})$ of the first dot's charge:

$$\frac{\partial E_{\mathcal{N}}}{\partial \mathcal{N}} = 2E_C(\mathcal{N} - 2X) - 4\tilde{E}_C \left[\bar{N}_1(\mathcal{N}) - \frac{\mathcal{N}}{2} \right]. \quad (4)$$

The condition for the peak position, $E_{\mathcal{N}+1} - E_{\mathcal{N}} = 0$, can now be obtained by integration of Eq. (4). The result is

$$X^* = \frac{\mathcal{N}}{2} + \frac{1}{4} - \frac{\tilde{E}_C}{E_C} \int_{\mathcal{N}}^{\mathcal{N}+1} \left[\bar{N}_1(\mathcal{N}') - \frac{\mathcal{N}'}{2} \right] d\mathcal{N}', \quad (5)$$

where now \mathcal{N} is an integer again. In the limit $G_0 \rightarrow 0$, the average $\bar{N}_1(\mathcal{N}')$ is the integer closest to $\mathcal{N}'/2$, and taking $\mathcal{N} = 2n$ and $\mathcal{N} = 2n+1$ in Eq. (5) reproduces Eq. (2). The advantage of Eq. (5) is that it is valid at any G_0 . However, to make use of it, one has to evaluate $\bar{N}_1(\mathcal{N})$ at any \mathcal{N} , which is a challenging quantum mechanical problem because of the Coulomb interaction in Eq. (3). Fortunately, in the limit of a continuous spectrum, the Hamiltonian (3) coincides with the one for a single dot connected by a tunnel junction to a

massive lead. The latter problem has been extensively studied in the limits of weak and strong tunneling into the dot.^{3-6,16}

For weak tunneling, $G_0 \ll e^2/\hbar$, the deviation of $\bar{N}_1(\mathcal{N})$ from an integer is small and can be found³ from second-order perturbation theory in H_T . For \mathcal{N} in the interval $(2n-1, 2n+1)$, we get

$$\bar{N}_1(\mathcal{N}) = n + \frac{\hbar G_0}{2\pi e^2} \ln \frac{\mathcal{N} - 2n + 1}{2n + 1 - \mathcal{N}}. \quad (6)$$

Substituting Eq. (6) into Eq. (5) and using the periodicity of $\bar{N}_1(\mathcal{N})$, we find peak positions shifted by tunneling:

$$X_{\pm}^* \approx n + \frac{1}{2} \pm \frac{1}{4} \left[1 - \frac{\tilde{E}_C}{E_C} \left(1 - \frac{4 \ln 2}{\pi} \frac{\hbar G_0}{e^2} \right) \right]. \quad (7)$$

The splitting of the two peaks with the same n grows linearly with G_0 . The splitting here results from quantum charge fluctuations between the dots, not changes in geometric capacitances.

Clearly, the charge fluctuations grow with G_0 . In the limit of strong tunneling the discreteness of charge N_1 is no longer important,⁴⁻⁶ and $\bar{N}_1(\mathcal{N}) \rightarrow \frac{1}{2}\mathcal{N}$ [see Eq. (3)]. As a result the peaks are equidistant, $X^* = (2\mathcal{N} + 1)/4$, which is expected because in this limit the two dots form a single conductor. The doubling of the period of the peaks from this regime to that of Eq. (2) is one of the main observations of the experiment of Waugh *et al.*⁸

To find the peak positions as the system approaches the strong-tunneling limit, one has to specify a model of the junction between the dots. For electrostatically controlled dots in semiconductor heterostructures, the junction is known to be a microconstriction with smooth boundaries.¹⁷ The number of transverse modes in the constriction can be readily controlled;¹⁷ when the constriction is nearly open, transport proceeds by the single lowest transverse mode. The ground-state energy near the strong-tunneling limit, when the reflection coefficient for the single propagating mode is small $\mathcal{R} = 1 - \pi\hbar G_0/e^2 \ll 1$, was found in Ref. 5 [Eq. (48)]. Using that result, we get

$$X^* \approx \frac{2\mathcal{N} + 1}{4} + (-1)^{\mathcal{N}} \frac{4e^C}{\pi^3} \frac{\tilde{E}_C}{E_C} \mathcal{R} \ln \frac{1}{\mathcal{R}}, \quad (8)$$

where $C \approx 0.5772$ is Euler's constant.

Figure 2 compares our results (7) and (8) to the observations of Waugh *et al.*⁸ Because the different gates are not independent, the relation between the gate voltage V_0 at which the interdot conductance is measured and the V_0 at which the splitting is measured is not known. We take this relation to be a rigid shift⁸ and have used the data of Ref. 8 with a shift of -0.005 V. The agreement between theory and experiment is very good indeed.

As we have seen, the *positions* of peaks in the linear conductance carry information only about the ground-state energy of the two-dot system. To study the excitations, one can analyze the *heights and shapes* of the peaks. If the interdot tunneling is weak, $G_0 \ll e^2/\hbar$, the excitation spectrum consists of two independent quasiparticle spectra of the two dots. This enables us to apply the standard master-equation technique¹ and find

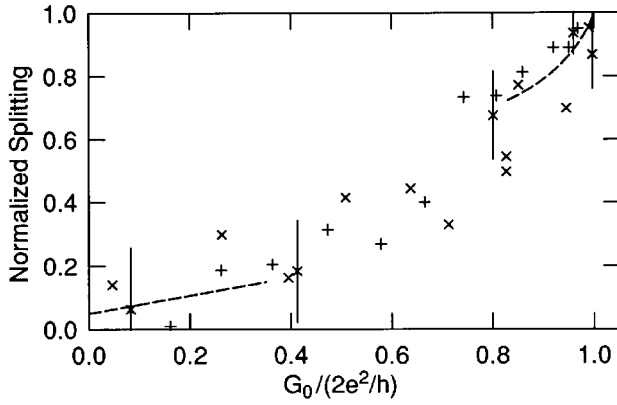


FIG. 2. The normalized splitting of the Coulomb blockade peaks as a function of the interdot conductance. Our theoretical results (dashed lines) are in good agreement with the experiment of Ref. 8 [\times , $+$ for data of their Figs. 3(a) and 3(b), respectively]. The splitting is normalized by the period of the peaks in the strong-tunneling limit. A small asymmetry, $\alpha=0.05$, has been included based on the experimental parameters.

$$G = \frac{G_l G_r}{G_l + G_r} \frac{\pm \beta(X_{\pm}^* - X)}{e^{\pm \beta(X_{\pm}^* - X)} - 1} \frac{1}{e^{\mp \beta(X_{\pm}^* - X)} + 2}, \quad (9)$$

where $\beta = 4E_C/T$ and X_{\pm}^* is a value in the sequence (7). In deriving Eq. (9) we assumed that the tunneling into the leads is much weaker than between the dots, $G_l \sim G_r \ll G_0$, and that the temperature is sufficiently low for the width of a peak ($\sim T/E_C$) to be much smaller than the spacing between the peaks. For small interdot capacitance ($E_C \approx \tilde{E}_C$), the latter constraint implies $T \ll (\hbar G_0/e^2)E_C$. Equation (9) is slightly different from the single-dot result¹⁸ in which the 2 in the last factor is a 1. This difference results from the degeneracy of the states with a charge on the left or right dot in the symmetric double-dot case, and causes the split peaks to be skewed towards each other.

In contrast to weak tunneling, at $G_0 \sim e^2/\hbar$ the excitation spectra of the two dots are not independent. In this regime an electron tunneling into the left dot shakes up the quantum state of the whole two-dot system, leading to a suppression of the conductance at low temperature. To illustrate this phenomenon, we calculate the temperature dependence of the peak heights in the case of perfect interdot transmission, $G_0 = e^2/\pi\hbar$.

As we have seen, the conductance peaks in the strong-tunneling limit are equally spaced, as if the double-dot system were a single dot. In fact, this remains true even for asymmetric double dots [$\lambda, \alpha \neq 0$ in Eq. (1)], because when $\mathcal{N} \rightarrow 0$ the energy is simply given by the first term in Eq. (1). However, the specific geometry of the system—whether it is one or two dots, and the degree of asymmetry—will show up in the peak heights. Unlike in a single dot, the single-mode constriction impedes charge propagation between the two dots, thus producing effects similar to those for a single junction coupled to an environment.¹ When an electron tunnels from the left lead into the left dot, a new value of the uniform potential within the dot is established almost instantaneously. However, in order to minimize the electrostatic energy after this shakeup, the other electrons in both dots must redistribute: a charge of $(1+\lambda)/2$ electrons must pass

through the constriction. As a result, the overlap of the two ground states, before and after the tunneling, vanishes, as in Anderson's orthogonality catastrophe.

At nonzero temperature, the tunneling density of states is suppressed as T^γ , where the exponent is related¹⁹ to the scattering phase shifts δ_m in each one-dimensional channel m by $\gamma = \sum_m (\delta_m/\pi)^2$. According to the Friedel sum rule, δ_m/π is the average charge transferred into each channel. A single-mode constriction provides two channels for each dot (two spins), yielding four channels in total. In our case, $\delta_m/\pi = \pm(1+\lambda)/4$, where the plus (minus) sign is for the channels in the right (left) dot. Thus, the rate of tunneling into the left dot is suppressed by the factor $T^{(1+\lambda)^2/4}$. For the rate between the right dot and lead, one should replace λ by $-\lambda$. Because the junctions are connected in series, the smaller of the two rates determines the conductance,

$$G = G_b \left(\frac{T}{\tilde{E}_C} \right)^\gamma F_\gamma \left(\frac{4E_C(X - X^*)}{T} \right), \quad \gamma = \frac{(1+|\lambda|)^2}{4}. \quad (10)$$

Here the coefficient G_b is of order $G_l \sim G_r$, and the peaks are centered at $X^* = (2\mathcal{N} + 1)/4$. For a symmetric system, $\lambda = 0$, the peak conductance obeys $G \propto T^{1/4}$. The temperature dependence in Eq. (10) can be obtained analytically²⁰ in the spirit of the bosonization approach;^{5,19} this technique also yields for the peak shapes

$$F_\gamma(x) = \frac{1}{\cosh(x/2)} \frac{\left| \Gamma \left(1 + \frac{\gamma}{2} + \frac{ix}{2\pi} \right) \right|^2}{\Gamma(2+\gamma)}. \quad (11)$$

At $\gamma=0$ the shape given by Eq. (11) is identical to that for weak tunneling Eq. (9): in both cases there is no charge transfer between the dots during the act of tunneling, and the spectra of the two dots are decoupled.

Comparing our results in the weak- and strong-tunneling limits [Eqs. (9) and (10)], we see that when G_0 grows the conductance of the system *decreases* due to the orthogonality catastrophe. For intermediate interdot conductances, the system must cross over from temperature independent peak heights for weak tunneling to the power-law suppression for strong tunneling. The theory of this crossover will be reported elsewhere.²⁰

We have seen that at $G_0 = e^2/\pi\hbar$ an asymmetry of capacitances, $\lambda \neq 0$, affects only the temperature dependence of the peak heights (10). In this limit, the peaks are equidistant and have the same height. We will now show that for weak tunneling even a small asymmetry dramatically affects the whole pattern of peaks. Below we study the asymmetry related to a small nonzero α in the electrostatic energy (1); the results are easily generalized to include a small λ by replacing $\alpha \rightarrow \alpha + 2\lambda$. Motivated by experiment,⁸ we will assume $E_C = \tilde{E}_C$.

First, let us determine the positions of the peaks in conductance. The condition for a peak is the degeneracy of the energy (1) with respect to adding an electron to either of the dots; this yields the two sequences of peaks,

$$X_1^* = \frac{n+1/2}{1-\alpha/2}, \quad X_2^* = \frac{n+1/2}{1+\alpha/2}. \quad (12)$$

Since $\alpha \ll 1$, the two periods are very close, and one observes beats with an approximate superperiod of α^{-1} .

The asymmetry lifts the degeneracy of the two states with an extra electron on either the left or right dot. The energy gap between these two states is $\Delta(X) = 4\tilde{E}_C|\alpha X - (N_1 - N_2)|$ with N_1 and N_2 for the lowest energy configuration at X . If the temperature is much lower than Δ , tunneling between two real states is suppressed. Instead, an extra electron in the left dot can escape into the right lead through a virtual state in the right dot. Because we assume that the level spacing is small compared to temperature, the dominant escape mechanism is *inelastic* cotunneling,²¹ i.e., an extra electron-hole pair is created in the right dot. The total rate²¹ of such processes is proportional to $(T/\Delta)^2$ and limits the conductance at sufficiently low temperatures. The calculation of peaks in conductance²⁰ leads to

$$G \propto \left[\frac{T}{\Delta(X)} \right]^2 F_2 \left(\frac{4E_C(X - X^*)}{T} \right). \quad (13)$$

Here the peak positions X^* are defined by Eq. (12). The

oscillations of the energy gap $\Delta(X)$ modulate the peak heights with the period α^{-1} in gate voltage X .

In conclusion, the fluctuation of electron charge between two quantum dots strongly affects tunneling through such a structure. First, the conductance peaks are split because of the lowering of the *ground-state energy* by charge fluctuations. This splitting, and eventual halving of the period of the conductance peaks as the interdot conductance grows, is a dramatic feature of the experimental data⁸ which is fully confirmed in our theory. Second, the temperature dependence of the peak height and shape is directly related to the *dynamics* of the quantum charge fluctuations. For a double dot connected by a reflectionless constriction, this produces a striking fractional power-law temperature dependence. Our theory is valid for a wide range of temperatures, limited only by the charging energy from above, and by the discrete energy level spacing from below.

Note added in proof. After submission of this paper, we received a manuscript from J. M. Golden and B. F. Halperin which treats the splitting of the conductance peaks in a similar way.

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- ¹ *Single Charge Tunneling*, edited by H. Grabert and M. H. Devoret (Plenum Press, New York, 1992).
- ² M. A. Kastner, *Rev. Mod. Phys.* **64**, 849 (1992).
- ³ See Sec. 4 of L. I. Glazman and K. A. Matveev, *Zh. Éksp. Teor. Fiz.* **98**, 1834 (1990) [*Sov. Phys. JETP* **71**, 1031 (1990)]; K. A. Matveev, *ibid.* **99**, 1598 (1991) [*ibid.* **72**, 892 (1991)].
- ⁴ K. Flensberg, *Phys. Rev. B* **48**, 11 156 (1993); *Physica B* **203**, 432 (1994).
- ⁵ K. A. Matveev, *Phys. Rev. B* **51**, 1743 (1995).
- ⁶ G. Falci, G. Schön, and G. T. Zimanyi, *Physica B* **203**, 409 (1994).
- ⁷ R. C. Ashoori, H. L. Stormer, J. S. Weiner, L. N. Pfeiffer, K. W. Baldwin, and K. W. West, *Phys. Rev. Lett.* **71**, 613 (1993); D. Esteve, P. Lafarge, P. Joyez, C. Urbina, and M. H. Devoret, *Physica B* **197**, 500 (1994).
- ⁸ F. R. Waugh, M. J. Berry, D. J. Mar, R. M. Westervelt, K. L. Campman, and A. C. Gossard, *Phys. Rev. Lett.* **75**, 705 (1995).
- ⁹ L. W. Molenkamp, K. Flensberg, and M. Kemerink (unpublished); L. W. Molenkamp and M. Kemerink, *Superlatt. Microstruct.* **16**, 275 (1994).
- ¹⁰ N. C. van der Vaart, S. F. Godijn, Y. V. Nazarov, C. J. P. M. Harmans, J. E. Mooij, L. W. Molenkamp, and C. T. Foxon, *Phys. Rev. Lett.* **74**, 4702 (1995).
- ¹¹ R. H. Blick, R. J. Haug, J. Weis, D. Pfannkuche, K. v. Klitzing,

- and K. Eberl, *Phys. Rev. B* (to be published); R. J. Haug, R. H. Blick, and T. Schmidt, *Physica B* **212**, 207 (1995).
- ¹² F. Hofmann, T. Heinzel, D. A. Wharam, J. P. Kotthaus, G. Böhm, W. Klein, G. Tränkle, and G. Weimann, *Phys. Rev. B* **51**, 13 872 (1995).
- ¹³ To be specific, the relations between our constants and the experimental ones of Ref. 8 are $X = V_a(C_{2a} + C_{3a})/2e$, $E_C = \tilde{E}_C = e^2/4C_\Sigma$, $\lambda = (C_{3a} - C_{2a})/2C_\Sigma$, and $\alpha = 2(C_{2a} - C_{3a})[(C_{2a} + C_{3a})^{-1} - (2C_\Sigma)^{-1}]$, where $a = 5$ or 6 throughout.
- ¹⁴ C. A. Stafford and S. Das Sarma, *Phys. Rev. Lett.* **72**, 3590 (1994).
- ¹⁵ G. Klimeck, G. Chen, and S. Datta, *Phys. Rev. B* **50**, 2316 (1994).
- ¹⁶ A. D. Zaikin, D. S. Golubev, and S. V. Panyukov, *Physica B* **203**, 417 (1994); H. Schoeller and G. Schön, *ibid.* **203**, 423 (1994); H. Grabert, *Phys. Rev. B* **50**, 17 364 (1994).
- ¹⁷ See, e.g., C. W. J. Beenakker and H. van Houten, in *Solid State Physics*, edited by H. Ehrenreich and D. Turnbull (Academic Press, New York, 1991), Vol. 44, pp. 109–125.
- ¹⁸ L. I. Glazman and R. I. Shekhter, *J. Phys. Condens. Matter* **1**, 5811 (1989).
- ¹⁹ K. Schotte and U. Schotte, *Phys. Rev.* **182**, 479 (1969).
- ²⁰ K. A. Matveev, L. I. Glazman, and H. U. Baranger (unpublished).
- ²¹ D. V. Averin and Yu. V. Nazarov, *Phys. Rev. Lett.* **65**, 2446 (1990).