Quantum Limitation on Coulomb Blockade Observed in a 2D Electron System

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The evolution of the Coulomb blockade of conduction through a small 2D electron island of large density of states, realized in a GaAs/Ga(Al)As heterojunction, is studied from the weak tunnel coupling to the Ohmic regime. Cotunneling arising from quantum charge fluctuations is identified, at low voltage, by its dependence on the product of junction conductances, and the current at threshold voltage is well described by a nonperturbative model. Small Coulomb conductance oscillations are found to persist even when both junctions exceed the conductance quantum.

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Coulomb blockade [1,2] refers to the inhibition of charge transfer to a mesoscopic metallic system weakly coupled to electron reservoirs. The basic ingredients are (a) particle number quantization, (b) the charge quantum carried by an electron, and (c) the energy $\cos z \approx E_c$ to increase the charge of the metallic island of capacitance C. The present work investigates the effect of quantum charge fluctuations which lift the Coulomb blockade by progressively relaxing condition (a) by a controlled increase in the tunnel coupling.

The simplest system showing Coulomb blockade is a metallic island connected to a voltage source by tunnel junctions of conductance $\sigma_{1,2}$. In the limit of vanishing coupling, the island charge is quantized according to (a), and the electrostatic energy of the circuit, including the voltage source, takes discrete values and forms well separated energy levels. The tunnel current through the island results from single electron tunneling (SET) [1-3]. Incoherent sequential tunnel events increase or decrease by e the electrical charge of the island. A thermally activated Coulomb blockade of the current occurs when the circuit energy variation $\sim E_c$ is positive, because the tunneling electrons must find a lower kinetic energy empty state to satisfy energy conservation. Lowering the energy cost until conduction occurs is achieved by increasing the drive voltage V above $\simeq e/C$ or by changing the island potential energy by capacitive coupling to an external charge Q_0 . The SET mechanism explains the threshold voltage in the I-V characteristics (IVC) and the e periodicity of the conductance with Q_0 , and predicts a linear dependence of the current on the series conductance $\sigma_T = \sigma_1 \sigma_2 / (\sigma_1 + \sigma_2)$. At finite coupling, the decay rate $\Gamma \sim (\sigma_1 + \sigma_2)/2C$ of the charged island states broadens the circuit energy levels and introduces quantum corrections according to the ratio $\gamma = \hbar \Gamma / E_c = (\sigma_1 + \sigma_2) / \pi \sigma_Q$, where $\sigma_Q = 2e^2/h$. The first correction is the secondorder tunnel process of cotunneling or quantum macroscopic tunneling of charge: A charge is transferred

through the island by simultaneous tunneling of two electrons and virtual increase of the circuit energy [4,5]. The cotunneling current grows with tunnel coupling as $\sigma_T \gamma \sim \sigma_1 \sigma_2 / \sigma_Q$ and presents an algebraic variation with the temperature T in contrast to the SET thermal activation. At much higher coupling, $\gamma = \hbar \Gamma / E_c \gtrsim 1$, the full broadening of the circuit energy levels leads to Ohmic conduction. However, a physical description of this regime is not simple [6] and the conductance value separating the Coulomb from the Ohmic domain is not well established.

Cotunneling has been quantitatively identified in metal-insulator-metal (MIM) junction arrays below the threshold voltage (V < e/C) for a limited range of discrete coupling values [7]. Its presence has also been indicated in preliminary observation in a 2D electron system [8]. It is thought to limit the accuracy of Coulombblockade-based devices. A good understanding is particularly needed at large coupling and near the threshold voltage. For strong coupling, the few experimental data suggest already washed out Coulomb effects for $\gamma \approx 1.4$ [9].

A 2D electron system is a very suitable tool to study the Coulomb blockade physics. Electrostatically defined quantum point contacts (QPC) provide accurate and in situ adjustable control of the tunnel coupling. If the (2D) metallic island has small energy level spacing $\Delta \ll E_c$, resonant tunneling effects, which otherwise couple to charging effects, can be neglected [10,11], particularly if eV or k_BT exceed Δ . Most tunnel processes then involve several electronic states of the island and conduction is similar to that of MIM systems. In this Letter, Coulomb blockade is studied from the weak coupling to the Ohmic regime, using a 2D system. We give experimental evidence for cotunneling and show that an approximate nonperturbative cotunneling expression, extended to voltage $V \simeq e/C$ [12], quantitatively reproduces the IVCs for a wide range of coupling. While fully developed charging effects only appear for $\sigma_{1,2} < \sigma_Q$, we find that Coulomb conductance modulations persist to $\sigma_1 + \sigma_2 \simeq 3\sigma_Q$.

The device is realized in a high-quality molecularbeam-epitaxy-grown GaAs/Ga(Al)As heterojunction with the Si δ -doping plane at 800 Å from the electron plane and 200 Å from the surface. A 2D island of size $\lesssim 1.15 \times 1.3 \ \mu m^2$ is formed by lateral confinement using pairs of Schottky gates on the sample surface (Fig. 1). Two pairs of gates $G_{1,2}$ (potential $V_{1,2}$) define two 3000-Å-wide QPCs (conductance $\sigma_{1,2}$) and a third pair G_0 (potential V_0) the island size. The electron density $n_s = (1.1-1.2) \times 10^{15} \text{ m}^{-2}$ gives a Fermi energy $E_F \approx 47 \text{ K}$, the mobility 280 V⁻¹s⁻¹m², an elastic mean free path $l_{el} \approx 15 \ \mu m$, and the island size $\Delta \approx 85 \ mK$. The separate QPCs display well quantized conductance plateaus. Their pinch-off voltages $\simeq -760$ mV differ only by $\simeq 20$ mV. In the tunnel regime, $\sigma_{1,2}$ show identical exponential variation with $V_{1,2}$ and equal modulation by V_0 except when $\sigma_{1,2} \gtrsim 0.5 \sigma_0$ (perhaps because the weak disorder then dominates the gate potential). The two-point conductance is measured using a 13-Hz square-wave voltage source whose first harmonic is 20 μ V rms and detecting the first harmonic of the current. A resistance $R_s = 8.3 \text{ k}\Omega$ is in series with each QPC and represents the Ohmic contact, mesa, and rf-filter resistances. When specified, conductances are corrected for this. A large capacitance is in parallel with R_s . For all measurements we add V/2 to all gate voltages for symmetrization [13]. The equivalent circuit is shown in Fig. 1(b). The condition $(eV \text{ or } k_B T) > \Delta$ is always fulfilled.



FIG. 1. (a) Schottky gate geometry. (b) Equivalent circuit. $n_{1,2}$ are the number of electrons having tunneled through each barrier. $Q_0 = \text{const} - C_{G1}V_1 - C_{G2}V_2 - C_{G0}V_0$. (c) Electron and hole kinetic energies and (d) complete circuit energy levels $U_{n1,n2}$ involved for cotunneling from left to right reservoirs. The cotunneling transition amplitude is proportional to the inverse distance between the actual intermediate energy (dashed line) and the virtually excited circuit energy level ($U_{n+1,n}$ or $U_{n,n+1}$). When they coincide (first-order tunneling then contributes) the amplitude diverges if the linewidths $\hbar \Gamma_{1,-1}/2$ of the intermediate states are not considered.

Upon sweeping the gate voltages, well developed conductance oscillations appear for $T \leq 1.5$ K and $\sigma_{1,2}$ just below the lower conductance plateau. Their Coulomb origin has been shown in Ref. [8] for another sample having similar gate geometry. They show best contrast when $\sigma_1 = \sigma_2$ and are very periodic as shown in Fig. 2. The regular variation of the peak height comes from the modulation of $\sigma_{1,2}$ by V_0 . The peaks slightly increase with T and confirm negligible resonant tunneling effects for which a T^{-1} variation would be expected [11]. SET analysis accounts well for the high-temperature behavior of low- σ_T ($\sigma_{1,2} \leq 0.1 \sigma_Q$) conductance peaks (0.25 $\leq Q_0/e \leq 0.75$) and gives an accurate determination of E_c [12]. E_c , found in the range 2.1-3 K depends on gate voltage configuration.

Cotunneling.—IVCs with $\sigma_1 \simeq \sigma_2$ are shown in Fig. 3(a) for two consecutive conductance minima and the intervening maximum and, in Fig. 3(b), for a minimum at higher σ_T and lower T. In (a) the large-voltage asymptote extrapolated to I=0 yields $e/C \simeq 380 \ \mu\text{V}$ ($E_c \simeq 2.2$ K) for the curve with lowest σ_T . This value of E_c gives a good SET fit of the associated conductance peaks except very close to the minima. At the minima, the SET model does not fit the IVCs well. The predicted current is compared with the data using the same $E_c = 2.2$ K and a weak 5% capacitance asymmetry $(c_1 - c_2)/C$ deduced from the IVC dependence on Q_0 [12]. The observed current is much larger for $V \leq e/C$, and does not fall off



FIG. 2. Conductance oscillations with V_0 at T=105 mK. The highest peaks are translated by 5 or 10 μ S. $V_1 = -743$ mV, $V_2 = -752$ mV are tuned to give the best peak-to-valley ratio. As a result of cotunneling, this ensures $\sigma_1 \simeq \sigma_2$ and holds as long as $\sigma_{max} = 0.5\sigma_1\sigma_2/(\sigma_1 + \sigma_2)$ varies exponentially with V_0 . SET (points) and nonperturbative cotunneling (triangles) fits are shown. Inset: The exponential variation of σ_{max} and σ_{min} corrected for R_s . The dashed-line slope is twice the solid-line slope.



FIG. 3. IVCs: (a) for two neighboring conductance minima and the intervening maximum at 105 mK; (b) for a different run at higher σ_T and 55 mK. The SET (dashed lines) and nonperturbative cotunneling (open symbols) currents are calculated using $E_c = 2.2$ K for (a) and 2.1 K for (b). σ_T is deduced from the interpolated value of the nearest conductance maxima obtained when V_0 changes Q_0 by $\pm e/2$.

exponentially nor scale as σ_T . Using reduced units $e\sigma_T/C$ to compare the lower- σ_T curve in (a) with the higher- σ_T , but lower-T, curve in (b) clearly shows that this extra current rises with conductance. High-temperature SET predictions like that shown in (b) do not fit the data using E_c deduced from SET fits of the associated conductance peaks. We found no better agreement using $E_c \simeq 1.8$ K deduced from the asymptote extrapolation of the curve in (b). In Fig. 2 the conductance oscillation minima display a faster increase with σ_T than the SET model predicts. The inset shows that both the maxima and the minima vary exponentially with V_0 , but that $\sigma_{\min} \sim \sigma_{\max}^2$. This quadratic relation reflects a second-order tunneling process and identifies cotunneling. In the Coulombblockaded regime $(Q_0 \simeq 0, V \ll e/C, k_BT \ll E_c)$, the theoretical cotunneling current is [5]

$$I_{\text{cot}} = \frac{h}{6e^2} \sigma_1 \sigma_2 \left(\frac{1}{E_1} + \frac{1}{E_2} \right)^2 \left[(k_B T)^2 + \left(\frac{eV}{2\pi} \right)^2 \right] V \quad (1)$$

and yields $\sigma_{\min} \sim \sigma_1 \sigma_2 \sim \sigma_{\max}^2$ as $\sigma_1 = \sigma_2$. Using the notations of Fig. 1, E_1 (E_2) is the increase of circuit energy for zero voltage above the initial state, $N = n_1 - n_2 = 0$, associated with the two interfering processes where a first electron enters the island from the right (leaves to the left) and virtually increases (decreases) its charge:

$$E_{1,2} = (e^2/C) \left[\frac{1}{2} \pm N \mp Q_0/e - (C \pm c_2 \mp c_1)V/2e \right].$$
(2)

From Fig. 2, the ratio $\sigma_{\min}/\sigma_{\max}^2$ gives $k_B T^*/E_c \simeq 0.075$, where

$$(k_B T^*)^2 = (k_B T)^2 + (eV/2\pi)^2 + 3(eV_n/2\pi)^2 + 3E_c\Delta/8\pi^2$$

includes the bias voltage V, a voltage noise V_n , and the elastic cotunneling contribution due to nonzero Δ [5].

Using $E_c \simeq 2.2$ K, deduced from the IVC threshold voltage at low conductance, one finds $(3E_c\Delta/8\pi^2k_B^2)^{1/2}=84$ mK, $T^* \simeq 165$ mK, and $V_n \simeq 28 \ \mu V$ [14]. The quantitative agreement is good and supported by a T^2 and V^3 variation of the current [8]. The perturbative theory [4,5] captures most of the physics but fails to describe the conductance away from the minima and the IVCs near the threshold voltage. The current diverges at V = e/Cfor T=0 and at any V for finite T because real transitions toward the intermediate state make the second-order tunneling rate infinite. The problem is similar to the resonance effects for light scattering or radiative cascade for a three-level atom [15]. Here the charging energy replaces the atomic levels and the change in the tunneling electron kinetic energy the photon frequency. Introducing the first-order decay rate $\Gamma_{1,-1}$ of the $(N = \pm 1)$ intermediate charged states of the island (see Fig. 1) regularizes the divergence and is equivalent to partial summation of the perturbation series at weak coupling. The derived current is $I = P(0)e/\tau_{if}$ [12] and is found to be accurate for $V \lesssim 1.3e/C$, $|Q_0| < 0.4e$, $k_BT \ll E_c$, and $\gamma \ll 1$ [16]. The decay rate $1/\tau_{if}$ from the state $(n_1 = n, n_2 = n)$ to the lower potential energy state (n+1, n+1) is given by Eq. (9) of Ref. [5] with $E_{1,2}$ replaced by $E_{1,2}$ $+i\hbar\Gamma_{1,-1}/2$ and the bracket by a modulus. The SET master equation gives the probability P(0) of the initial state within this approximation. SET is recovered for $\gamma \rightarrow 0$ and perturbative cotunneling [Eq. (1)] for $(\pi/4) \gamma e/C < V \ll e/C$ and $\gamma E_c/4 < k_B T \ll E_c$. The calculated current agrees well with the data. A wide $(|Q_0| \leq 0.4e)$ region of the minima (3) to (7) of Fig. 2 $(\gamma = 0.06 - 0.5)$ is perfectly reproduced using the same $E_c = 2.2$ K, $T^* = 165$ mK, and assuming $\sigma_1, \sigma_2 = 4\sigma_{\text{max}}$ to have the exponential variation displayed in the inset. The last minimum, not fitted, is $\approx 20\%$ above the predicted value, either because conductance asymmetry increases σ_{\min} , as the deviation from the exponential variation of $\sigma_{\rm max}$ suggests, or because the expression no longer holds as $\gamma \gtrsim 0.5$. In Fig. 3(a), the IVCs ($\gamma = 0.09, 0.18$) are also well described using the bare temperature and the only adjustable parameter $E_c = 2.2$ K. A good fit of the curve in (b) ($\gamma = 0.25$) is obtained with $E_c = 2.1$ K. The E_c values agree with those deduced from SET fits of the conductance peaks. For $\sigma_{1,2} \gtrsim 0.5 \sigma_0$ and $V \gtrsim e/C$, tunnel barrier nonlinearities, observed for single QPC, become important and complicate the quantitative analysis.

High-conductance regime.— From $\sigma_1 = \sigma_2 \approx 0.5 \sigma_Q$ to $\approx \sigma_Q$, the conductance oscillations with gate voltage quickly damp and the maxima $\approx 0.5\sigma_T$ evolve to $\approx \sigma_T$ as expected for the conducting regime. Figure 4 shows conductance traces when V_1 is swept $[\sigma_1 \approx (0.2-6)\sigma_Q]$ for three fixed gate voltages V_2 . The bold lines on the V_1 - V_2 inset diagram represent the experimental paths and the dashed lines define the regions labeled (m,n) for σ_1 and σ_2 on plateaus $m\sigma_Q$ and $n\sigma_Q$. While the QPC separately display flat plateaus, the traces reveal nonperfect Ohmic



FIG. 4. Conductance traces in the conducting regime upon sweeping V_1 at fixed V_2 . The period of the weak modulation can be compared with the Coulomb comb obtained in the tunnel regime. A systematic study of the successive quantization of σ_1, σ_2 , when V_1, V_2 are varied, gives the inset diagram on which the experimental paths are shown. Figures 2, 3(a), 3(b), and 4 correspond to four different runs.

additivity and the $\approx (10-25)\%$ conductance modulation on the $\approx 12-20$ mV scale probably results from weak coherent transmission. More striking are the fine [(2-4)%] conductance oscillations. The fact that their periods with V_1 , V_2 , and V_0 all correspond with those of Coulomb oscillations suggests a Coulomb origin, particularly as the insensitivity to the introduction of up to $\simeq 10$ magnetic flux quanta through the island rules out resonance transmission effects. However, their rapid disappearance with temperature ($\simeq 0.4$ K for $\sigma_{1,2} \simeq \sigma_0$) indicates renormalized E_c . Within our experimental resolution they are detected up to $\sigma_1 + \sigma_2 \approx 3\sigma_0$. This is consistent with recent theoretical estimations of a critical resistance in the range 6-0.6 k Ω for a single junction [6]. However, we must be cautious when comparing 2D junctions having few transmitting channels with MIM junctions having the same conductance but many nearly closed channels.

We have experimentally shown that, for a wide range of tunnel coupling, the conducting properties of a large 2D island are well explained by taking into account the quantum charge fluctuations which lift the Coulomb blockade of tunneling. We think that observation of such coherent processes can be useful in putting a lower bound on the coherence time τ_{φ} of single-electron states τ_{φ} $> 2C/(\sigma_1 + \sigma_2)$. The high-conductance observations suggest a smooth transition from the Coulomb to the Ohmic regime for this system. We are grateful to D. Estève, M. H. Devoret, and P. Lafarge for very helpful discussions and to C. Chaleil and J. Verrier for technical help.

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- [1] K. K. Likharev, IBM J. Res. Dev. 32, 144 (1988).
- [2] For a recent review, see Single Charge Tunneling, NATO ASI Ser. B, edited by H. Grabert and M. H. Devoret (Plenum, New York, 1991).
- [3] I. O. Kulik and R. I. Shekhter, Zh. Eksp. Teor. Fiz. 68, 623 (1975) [Sov. Phys. JETP 41, 308 (1975)].
- [4] D. V. Averin and A. A. Odintsov, Phys. Lett. A 140, 251 (1989).
- [5] D. V. Averin and Yu. V. Nazarov, Phys. Rev. Lett. 65, 2446 (1990).
- [6] Theoretical studies are for single junctions: S. V. Panyukov and A. D. Zaikin, Phys. Rev. Lett. 67, 3168 (1991);
 V. Scalia, G. Falci, R. Fazio, and G. Giaquinta, Phys. Rev. Lett. 67, 2203 (1991); W. Zwerger and M. Scharpf, Z. Phys. B 85, 421 (1991).
- [7] L. J. Geerligs, D. V. Averin, and J. E. Mooij, Phys. Rev. Lett. 65, 3037 (1990); T. M. Eiles, G. Zimmerli, H. D. Jensen, and J. M. Martinis, Phys. Rev. Lett. 69, 148 (1992).
- [8] D. C. Glattli, C. Pasquier, U. Meirav, F. I. B. Williams, Y. Jin, and B. Etienne, Z. Phys. B 85, 375 (1991).
- [9] L. J. Geerligs, V. F. Anderegg, C. A. van der Jeugd, J. Romijn, and J. E. Mooij, Europhys. Lett. 10, 79 (1989).
- [10] For a recent review, see H. van Houten, C. W. J. Beenakker, and A. A. M. Staring, in *Single Charge Tun*neling (Ref. [2]).
- [11] Y. Meir, N. S. Wingreen, and P. A. Lee, Phys. Rev. Lett. 66, 3048 (1991); C. W. J. Beenakker, Phys. Rev. B 44, 1646 (1991).
- [12] D. C. Glattli et al. (to be published).
- [13] This makes the capacitance asymmetry $(c_2 c_1)/C$ weak and the IVCs nearly symmetrical even for $Q_0 \neq 0$. Otherwise $\sum_i C_{G_i}$ adds to c_1 .
- [14] In Ref. [8], a too small value of E_c , deduced from threshold voltage, underestimated elastic cotunneling.
- [15] See, for example, W. Heitler, The Quantum Theory of Radiation (Clarendon, Oxford, 1954), p. 196; L. D. Landau and E. M. Lifshitz, Relativistic Quantum Theory (Pergamon, Oxford, 1971), p. 206.
- [16] Our expression agrees with the T=0 formula derived by D. Esteve and P. Lafarge (unpublished) for $V \leq 2e/C$, and derived by D. V. Averin, in *Single-Electron Tunneling and Mesoscopic Devices*, edited by H. Koch and H. Lubig, Springer Series in Electronics and Photons Vol. 31 (Springer-Verlag, Berlin, 1992), and by Yu. V. Nazarov (to be published) for $V \simeq e/C$.