

## Periodic and Aperiodic Bunching in the Addition Spectra of Quantum Dots

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We study electron addition spectra of quantum dots in a broad range of electron occupancies starting from the first electron. Spectra for dots containing  $<200$  electrons reveal a surprising feature. Electron additions are not evenly spaced in gate voltage. Rather, they group into bunches. With an increasing number of electrons the bunching evolves from occurring randomly to periodically at about every 5th electron. The periodicity of the bunching and features in electron tunneling rates suggest that the bunching is associated with electron additions into spatially distinct regions within the dots. [S0031-9007(97)04028-3]

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Coulomb blockade (CB) is one of the most fundamental and robust concepts in mesoscopic physics. Lambe and Jaklevic first made this point clear in a seminal experiment nearly 30 years ago [1]. For a metallic island poorly coupled to its surroundings, the number of electrons is quantized at low temperatures. Because of the repulsive Coulomb potential created by electrons already on the island, the energy required to add an electron to the island increases by a fixed amount  $\Delta$  with each electron added. An external gate electrode capacitively coupled to the island through a capacitance  $C_g$  can be used to cause electrons to transfer on and off the island. Additions of single electrons occur periodically in gate voltage with a period  $e/C_g$ . Physical phenomena in the system with characteristic energy scales on order  $\Delta$  can disrupt the periodicity. For instance, superconductivity in the system can cause electrons to be added as periodically occurring pairs [2]. One does not expect such results in a semiconductor or a normal metal sample.

In a semiconductor system containing enough electrons to be considered metallic, the deviations from exact periodicity in gate voltage are expected to be rather weak. The corresponding small parameter is  $r_s/R$ , where  $r_s$  is the screening length (about 100 Å), and  $R$  is a characteristic size of the system (0.2–1  $\mu\text{m}$ ) [3–6]. In the opposite limit of a disordered mesoscopic system containing a few electrons localized at spatially distinct sites, significant fluctuations in the addition spectrum are predicted [6].

Several years ago, one experiment on a semiconductor system displayed results which appeared to violate CB [7]. Electrons were seen to enter a quantum dot in pairs rather than individually. The system was a two-dimensional dot with a 1  $\mu\text{m}$  diameter. It was somewhat atypical of quantum dot experiments: the first electrons to enter this system occupy random potential minima created by disorder, and the different minima were screened from each other by a metallic electrode ( $\sim 350$  Å away). Nonetheless, the *exact* coincidence of electron additions into the system is quite

a surprise. The experiment used a method called single electron capacitance spectroscopy (SECS) and was unique in allowing study of electron additions into separate localized sites.

This Letter describes results from a systematic SECS study of different sized two-dimensional dots to help determine the origin of the strange correlation in electron additions. We found that in dots containing small numbers of electrons, electron additions are sometimes grouped in bunches comprising from 2 to 6 electrons. Here, we describe a new startling pattern of the addition spectra. We observe pairing of electrons additions occurring nearly periodically with electron number. Every 5th electron addition peak pairs with a neighboring peak. The details of the addition spectra yield critical clues about the nature of the bunching.

A schematic of our samples is shown in the inset of Fig. 1(a). They are similar to the ones described in Refs. [7,8]. The AlGaAs/GaAs wafer contains the following layers (from the bottom to the top): 3000 Å  $n^+$  GaAs, 400 Å GaAs spacer layer, 136 Å AlGaAs/GaAs superlattice tunnel barrier, 175 Å GaAs quantum well, 500 Å AlGaAs blocking barrier, 300 Å GaAs cap layer. A mesa with deep Ohmic contacts down to  $n^+$  GaAs is initially defined. Then a circular Cr/Au gate electrode is fabricated on the top of the mesa. Eight dots were studied with gate diameters ranging from 1.6 to 0.2  $\mu\text{m}$ . Plasma etching produces a short pillar (300 Å tall) using the gate electrode as a mask. Electrons remain in the quantum well only in the region below the pillar [7]. The measurements are carried out using an on-chip bridge circuit described in [7].

Figure 1(a) displays the electron addition spectrum at zero magnetic field for a dot of 500 nm lithographic diameter. For gate biases below  $-500$  mV, the quantum dot is empty. Peaks occur in the capacitance at gate voltages for single electron additions to the quantum dot [7]. Remarkably, some of the peaks shown are of double

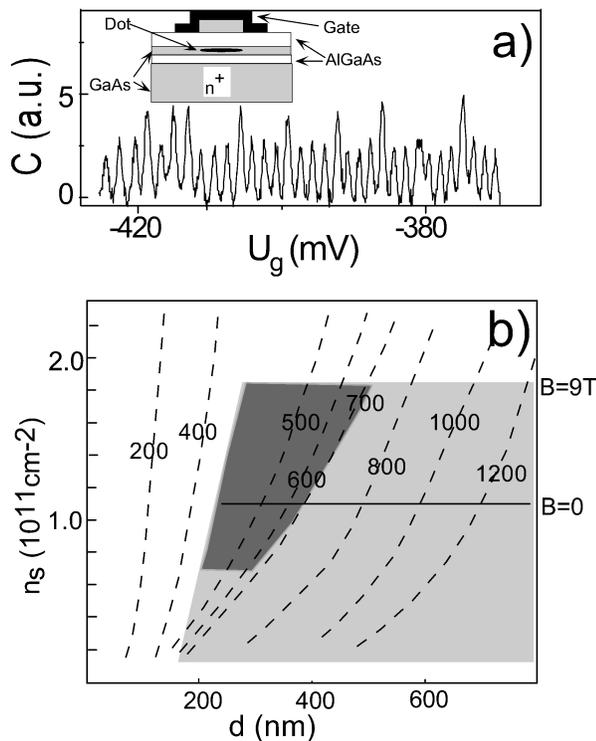


FIG. 1. (a) Quantum dot capacitance as a function of gate voltage. Each peak denotes the appearance of an electron in the dot. Double height peaks indicate the addition of two electrons.  $T = 0.3$  K. Inset: Schematic of the dot. (b) Experimental phase diagram for  $B = 9$  T. Dashed curves show the variation of lateral diameter  $d$  of electron puddles with the electron density for different quantum dots. Shaded area denotes the range of bunching; dark shading indicates the range of periodic bunches. The bunching ceases at an electron density which increases with magnetic field (see text). The density limit at  $B = 0$  is demarcated by a horizontal line.

height indicating the tunneling of two electrons in the dot at the same gate voltage.

Altogether, we can resolve the first 600 electron additions into this dot. The gate voltage scale can be directly converted to an energy scale  $\Delta E = \alpha \Delta V_g$  with the lever arm  $\alpha \sim 0.5$  for these structures determined from the geometry of the dot [7]. The gate voltage position of the  $N$ th capacitance peak, when multiplied by the lever arm, directly measures the chemical potential  $\mu_N$  of the dot containing  $N$  electrons [3].

The magnetic field evolution of a portion of the electron addition spectrum is shown in Fig. 2(a). The gray scale map displays the first 150 additions, with capacitance peaks visible as black traces. Examination of the bottom of Fig. 2(a) shows that the first 7 electrons enter the dot at widely spaced voltages. Beyond the 7th electron trace, something extraordinary occurs. Three electrons enter the dot in very rapid succession in gate voltage over the full range of magnetic fields. The next two electrons also join in a bunch (pair). For higher  $N$ , other bunches can be seen. We note that the experiment shows no hysteretic effects. The bunching is a phenomenon which occurs with the dot in *equilibrium* with its surroundings.

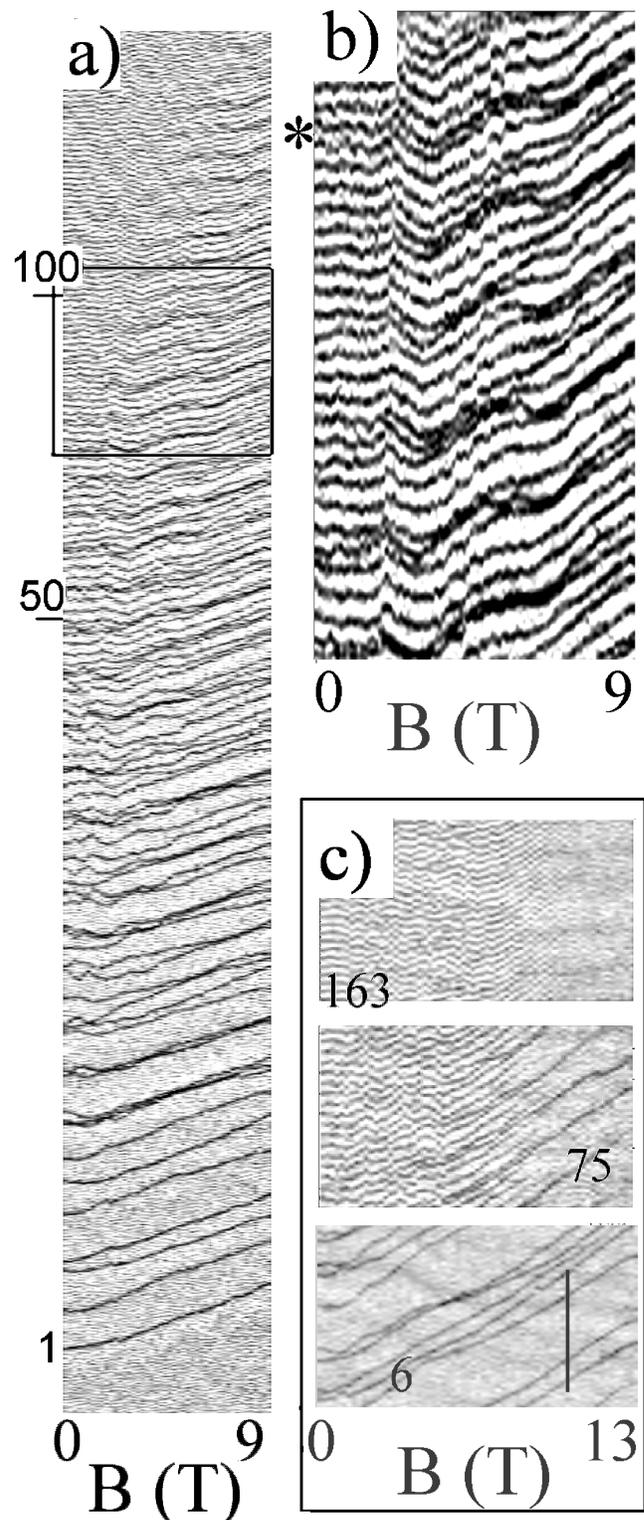


FIG. 2. Gray scale image of the measured capacitance. Black denotes capacitance peaks. Electron occupancies are indicated as numbers. (a) Vertical axis—gate voltage ranging from  $-511$  mV (bottom) to  $-328$  mV (top).  $T = 0.3$  K. (b) Zoom-in of spectrum surrounded by box in (a). (c) Segments of the addition spectrum measured after thermally cycling the dot to room temperature.  $T = 50$  mK. Vertical bar corresponds to energy change of  $5$  meV [common for all images in (c)].

After about 40 electrons are added to the dot, the bunching develops into a *periodic* pattern, with one bunch appearing for each 4–6 electrons added to the dot. As  $N$  is increased beyond about 80, the bunching ceases for zero magnetic field. Instead, the electron additions occur with nearly perfect periodicity, as is typical of CB. However, for nonzero magnetic field strengths, the bunching phenomenon returns. Bunches again occur periodically in gate voltage, and the period is about the same as that for the zero field bunches. A zoom-in of this behavior is shown in Fig. 2(b). The onset of bunching shifts to larger magnetic fields with increasing  $N$ , and the bunches are no longer observable at fields up to 13 T for more than about 200 electrons in the dot.

The behavior of each electron trace can be described roughly as follows. The magnetic field at which all electrons fall into the lowest Landau level,  $\nu = 2$ , can be readily identified as a maximum in the traces at around  $B = 2$  T [9]. As in two-dimensional systems the chemical potential peaks just as higher Landau levels depopulate completely. Jumps in the traces at higher magnetic fields, where both spin levels of the lowest Landau level are filled, are usually interpreted as single electron spin flips [9,10]. The flatness of the traces around  $B = 6$  T demarcates total spin polarization of the dot. We refer to this range as the vicinity of  $\nu = 1$ . For higher fields, the traces rise nearly linearly with magnetic field.

A bunched pair of traces in Fig. 2(b) is marked with an \*. These traces are fairly representative of all of the other traces which appear as electron pairs. Starting at some nonzero magnetic field the two traces are seen to stick together but then they split as the field approaches that which yields  $\nu = 1$ . Passing through  $\nu = 1$ , the lower trace of the bunched pair splits from the trace above it, only to join with the trace below it.

The bunching phenomenon is reflected in the rate at which the electrons tunnel into the dot. At zero magnetic field, the rate of electron tunneling between the  $n^+$  substrate and the quantum well is about 5 MHz. Measurements at a much lower frequency of  $f = 200$  KHz are sensitive only to the tunneling resistance if the tunneling is strongly suppressed by electron correlations within the dot [8,9]. At very low temperatures ( $T < 0.1$  K) the tunneling rate drops substantially in particular regions of magnetic field and electron occupancy.

Figure 2(c) shows a measurement of the addition spectrum of the same dot at base temperature  $T = 50$  mK. For low  $N$ , shown in the bottom part of Fig. 2(c), contrast in all electron traces is the same over the entire range of magnetic field, indicating that the electron tunneling rate is much larger than the measurement frequency. The middle segment of Fig. 2(c) displays the capacitance spectrum in a range of larger  $N$  (75–95 electrons in the dot). Notice here that some of the traces extinguish as the magnetic field increases. As the peaks diminish in strength, the phase of the electron tunneling signal lags relative to the ac excitation [8]. This detectable decay of the tunnel-

ing rates begins in the vicinity of  $\nu = 1$ , for a sufficiently large number of electrons in the dot.

The only traces observable at the highest magnetic field of  $B = 13$  T in Fig. 2(c) extend from paired traces. Examination of the intensity and phase of these unextinguished traces shows that they typically result from only a single electron rather than two electrons tunneling. We note that the dc bias in the experiment is adjusted very slowly so that the electron occupancy in the dot changes even though peaks are not seen in the capacitance experiment. Finally, at higher  $N$  [Fig. 2(c), upper part], the bunching disappears, and all traces extinguish equally.

The boundary for the onset of the bunching is remarkably similar for all dots in which bunches are observed, regardless of their size. This boundary moves to higher magnetic fields as the average electron density (note, *not*  $N$ ) in the dot is increased roughly according to the linear relation

$$n_{\text{onset}} = (1.1 + 0.08 \times B[\text{T}]) \times 10^{11} \text{ cm}^{-2}. \quad (1)$$

Our findings are summarized on an approximate phase diagram shown in Fig. 1(b). Each dashed curve represents the evolution of one measured quantum dot sample: As we increase the electron density  $n_s$  within the dot, the lateral diameter of electron pool  $d$  also grows.  $d$  is determined from the average spacing between addition peaks using a simple parallel plate capacitor approximation. The bunching is absent *either* for very small dots at arbitrary electron density [8,9,11] *or* at sufficiently large electron density in arbitrary large dots (nonshaded area). The nearly periodic bunching (pairing) pattern is observed for dots created with lithographic diameters of about  $0.5 \mu\text{m}$  (dark shaded). The details of the random bunching pattern at small electron numbers vary with thermal cycling of the sample to room temperature. In sharp contrast, the *periodic* bunching behavior remains qualitatively unchanged. The same 5-electron period is consistently detected for different thermal cycling and different samples. For larger dots the bunching still occurs, but the bunches appear to occur randomly with gate voltage rather than periodically (shaded).

We believe that pairs of electrons in the quantum dot observed previously by Ashoori *et al.* [7] are a special case of the bunches in the regime of electrons strongly localized within a large ( $1 \mu\text{m}$  lithographic diameter) dot. In dots of similar size, we have seen more examples of bunches with the traces of two and sometimes three electrons that exactly overlap over a range of magnetic fields. In general, paired traces from dots with smaller lithographic diameters do not coincide exactly. Two theoretical models [12,13], have been suggested to explain the origin of the exact pairs. Both models predict a dramatic suppression of the tunneling rate as soon as two electrons are joined into a pair, since both electrons must be added into the dot in a coherent fashion. Having studied a large number of exact pairs in the frequency range 50 KHz–1 MHz we have never observed

a significant drop of the tunneling rate when the traces merge. This suggests that the paired electrons tunnel into the dot *independently*, though they are added to the system at precisely the same gate voltage. Remarkably, the data indicate that filling one state of a pair has no effect on the energy of the other state in the pair.

The phase diagram shown in Fig. 1(b) suggests that the bunches are intrinsically associated with electron localization within the quantum dot. Small dots likely consisting of one electron puddle do not display the bunching effect, while the effect appears in larger dots at low densities when distinct electron puddles may exist. In the case of large dots whose lateral size significantly exceeds the effective screening length, the direct Coulomb repulsion between different electron droplets may be strongly suppressed. The addition of one electron in one region may not inhibit the addition of a 2nd electron in a remote location. Such localization is caused either by fluctuating potential or may arise intrinsically within a single dot due to interactions.

We speculate that the latter phenomenon gives rise to the periodic bunches. Indeed, the reproducibility of the periodic bunching pattern in several different dots and upon different thermal cycling of the same dot cannot be ascribed to a peculiarity of the disorder potential. The most plausible scenario for spatial segregation of electrons is the separation of an annulus of electrons at the circumference of the dot. To examine this idea, we use the results of classical modeling of the electron distribution within the dot [14,15]. Such a model is justified in the limit of infinite magnetic field. Electrons are then considered as classical point charges that form a nearly triangular lattice with significant lattice deformation only at the dot edges. The sequence of the electron entrances into a classical dot can be calculated. Levitov [15] demonstrated recently that for a classical dot containing 50–150 electrons, 4 electrons enter the center of the dot in succession, and the 5th enters the outer row (circumference).

The idea that one of the bunched electrons appears at the edge of the electron droplet is consistent with the contrast observed in the tunneling rate. The drop of the tunneling rate for  $\nu < 1$  illustrated on Fig. 2(c) can be considered as a special case of the Coulomb gap observed for larger systems [16]. The origin of the tunneling suppression can be understood semiclassically. The tunneling process suddenly adds one more electron into a dot. Until the system relaxes to its new ground state, the tunneling process is not finished. Hence, the effective tunnel barrier depends on the disturbance of the density distribution and its relaxation rate. The higher tunneling rate observed for one trace in each bunch can be explained if that electron is introduced into the edge of the dot. That electron has fewer and more distant neighbors compared with an electron introduced into the bulk of the dot.

The above model may explain the periodicity seen in our data and its increasing prominence at large magnetic field. However, it still does not explain the *pairing*. Stud-

ies of double dot systems show that spatial segregation does not directly lead to paired electron additions [17]. In fact, the residual Coulomb interaction between dots inhibits pairing even when the two dots are tuned to individually add electrons at the same gate bias. However, significant differences exist between these dots and ours, e.g., our dots contain a much lower electron density.

What can compete with the usually dominant Coulomb addition energy to disturb the addition spectrum so profoundly? Hartree-Fock calculations demonstrate [18] that exchange can mediate a local attraction between electrons, tending to keep the system compact. The  $\nu = 1$  state is believed to be fully spin polarized, and exchange maintains  $\nu = 1$  as the lowest energy state of the system over a range of magnetic fields [18]. The switching of the bunches at  $\nu = 1$  [Fig. 2(b)] effectively broadens this range for some of the traces suggesting the involvement of exchange in bunch formation.

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- [1] J. Lambe and R.C. Jaklevic, Phys. Rev. Lett. **22**, 1371 (1969).
  - [2] M.T. Tuominen *et al.*, Phys. Rev. Lett. **69**, 1997 (1992); P. Lafarge *et al.*, Phys. Rev. Lett. **70**, 994 (1993).
  - [3] H. van Houten, C.W.J. Beenakker, and A.A.M. Staring, in *Single Charge Tunneling*, edited by H. Grabert and M.H. Devoret (Plenum, New York, 1992), pp. 167–216.
  - [4] U. Sivan *et al.*, Phys. Rev. Lett. **77**, 1123 (1996).
  - [5] R. Berkovits and B.L. Altshuler, Phys. Rev. B **55**, 5297 (1997).
  - [6] A.A. Koulakov, F.G. Pikus, and B.I. Shklovskii, e-print cond-mat/9612236.
  - [7] R.C. Ashoori *et al.*, Phys. Rev. Lett. **68**, 3088 (1992).
  - [8] R.C. Ashoori *et al.*, Phys. Rev. Lett. **71**, 613 (1993).
  - [9] R.C. Ashoori, Nature (London) **379**, 413 (1996).
  - [10] P.L. McEuen *et al.*, Phys. Rev. B **45**, 11419 (1992); O. Klein *et al.*, Phys. Rev. Lett. **74**, 785 (1995).
  - [11] S. Tarucha *et al.*, Phys. Rev. Lett. **77**, 3613 (1996).
  - [12] Y. Wan, G. Ortiz, and P. Phillips, Phys. Rev. Lett. **75**, 2879 (1995).
  - [13] M.E. Raikh, L.I. Glazman, and L.E. Zhukov, Phys. Rev. Lett. **77**, 1354 (1996).
  - [14] V.M. Bedanov and F.M. Peeters, Phys. Rev. B **49**, 2667 (1994).
  - [15] L.S. Levitov (private communication).
  - [16] R.C. Ashoori *et al.*, Phys. Rev. B **48**, 4616 (1993); R.C. Ashoori *et al.*, Phys. Rev. Lett. **64**, 681 (1990); J.P. Eisenstein, L.N. Pfeiffer, and K.W. West, Phys. Rev. Lett. **69**, 3804 (1992).
  - [17] S.R. Waug *et al.*, Phys. Rev. Lett. **75**, 705 (1995).
  - [18] A.H. MacDonald, S.R.E. Yang, and M.D. Johnson, Aust. J. Phys. **46**, 345 (1993); C. de Chamon and X.G. Wen, Phys. Rev. B **49**, 8227 (1994).