Disorder and Interaction-Induced Pairing in the Addition Spectra of Quantum Dots

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We have numerically investigated the electron addition spectra in quantum dots containing a small number ($N \leq 10$) of interacting electrons, in the presence of strong disorder. For a short-range Coulomb repulsion, we find regimes in which two successive electrons enter the dot *sequentially* but almost at the same value of the chemical potential. In the strongly correlated regime these close additions, or pairings, are associated with electrons tunneling into distinct electron puddles within the dot. We discuss the tunneling rates at pairing and argue that the results are related to a recently observed phenomenon known as *bunching*.

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In a small metallic island weakly coupled to the environment the number of electrons is quantized at low temperatures. Because of the Coulomb repulsion from the electrons already on the island, it takes a finite energy to add one more electron to the island. This can be achieved with the help of an external gate voltage, coupled capacitively to the island. Additions of single electrons occur roughly periodically as a function of the gate voltage. This is the essence of Coulomb blockade, one of the most robust facts in mesoscopic physics.

By means of an experimental technique known as single-electron capacitance spectroscopy (SECS) [1,2], one can study electron additions from a metallic electrode to a semiconductor quantum dot. In these experiments the electrons tunnel into localized states of the dot one by one, starting from the very first electron; tunneling events are recorded as a function of the gate voltage.

A few years ago a SECS experiment [1,3] showed that *pairs* of electrons sometimes entered the dot at the same gate voltage, thus violating all of the common wisdom that we have on Coulomb blockade. A more recent and systematic investigation [4] has shown that quite generally, in dots containing $N < 200$ electrons, electron additions are not evenly spaced in gate voltage. Rather, in some cases, they group in *bunches* of up to 6 electrons. The first bunching already occurs when the number of electrons $N \sim 7$, and with increasing N it evolves from occurring randomly to periodically at about every 5th electron. To explain this puzzling effect two different theories have been proposed [5,6], in which the paired electrons enter the dot *coherently*. In contrast, the experiments suggest that the paired electrons tunnel into the dot *sequentially* [4]. Notice that, in dots with lithographic diameters <1 μm, paired traces do not coincide exactly. *Classical* simulations [7–9] reproduce features observed in the presence of a strong magnetic field. However, the mechanism behind the pairing effect is still unclear.

In this paper we carry out a full quantum mechanical calculation of the addition spectra of dots containing a small number of particles $N < 10$. Our goal is to investigate the first appearance of aperiodic pairing. We consider the limit of strong disorder generating localized states that can be occupied by two electrons of opposite spin [10]. When the Coulomb repulsion is *short range*, we find two different regimes where two successive additions sometimes almost coincide, yet still occur *sequentially*. We denote such occurrences as *pairings*. The first case takes place for intermediate values of the direct Coulomb interaction but strong on-site repulsion, which favors the appearance of a dense droplet with no holes or doubly occupied states. Both electrons participating in the pairing tunnel into the edges of the dot but in spatially distinct regions. The second situation occurs in the strongly correlated regime, with strong values of the direct Coulomb interaction competing with the on-site repulsion and the disorder. In this case pairing is characterized by the formation of distinct puddles of electrons; doubly occupied states appear in the center of the dot, where one of the two electrons tunnels. The close additions found in this regime bear strong similarities with the pairing seen in Ref. [4] for smaller dots.

We model the quantum dot with a tight-binding Hamiltonian on a square lattice with $\mathcal{N}_x \times \mathcal{N}_x$ sites:

$$H = \sum_{i,\sigma} (\epsilon_i - eV_g) c^\dagger_{i,\sigma} c_{i,\sigma} + t \sum_{\langle ij \rangle,\sigma} (c^\dagger_{i,\sigma} c_{j,\sigma} e^{i\varphi_{ij}} + \text{H.c.}) + H_{\text{int}}, \quad (1)$$

where the ϵ_i's are site energies ϵ_i randomly distributed with uniform probability between $-W$ and W; V_g is a gate voltage. The hopping term, connecting nearest-neighbor sites, is proportional to the matrix element t modified by Peierls phases φ_{ij}, due to the possible presence of a magnetic field. $\sigma = \uparrow, \downarrow$ is a spin variable. The interaction part is given by

$$H_{\text{int}} = \sum_{i>j;\sigma,\sigma'} v(\mathbf{r}_i - \mathbf{r}_j) n_{i,\sigma} n_{j,\sigma'} + U \sum_i n_{i,\uparrow} n_{i,\downarrow}, \quad (2)$$

where $n_{i,\sigma}$ is the number operator, $v(\mathbf{r}_i - \mathbf{r}_j)$ is the matrix element of the direct part of the Coulomb interaction, and U is the on-site interaction constant. Experimentally, the Coulomb interaction is screened by the external

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gate. For simplicity we will model the interaction as a nearest-neighbor repulsion, $v(\mathbf{r}_i - \mathbf{r}_j) = V\delta_{\mathbf{r}_j,\mathbf{r}_i+\boldsymbol{\delta}} = \frac{e^2}{a}\delta_{\mathbf{r}_j,\mathbf{r}_i+\boldsymbol{\delta}}$, where a is the lattice constant [11]. The important parameter describing the strength of the average Coulomb interaction relative to the Fermi energy is $r_s = 1/\sqrt{\pi n}\, a_B$, where n is the electronic density and a_B is the Bohr radius. For our lattice model $r_s = \frac{2}{\sqrt{\pi\nu}}\,(V/4t)$, where $\nu = \langle\hat{N}\rangle/[(\mathcal{N}_x - 1)\,(\mathcal{N}_y - 1)]$ and $\langle\hat{N}\rangle$ is the average number of electrons on the dot.

By using Lanczos techniques, we have diagonalized the Hamiltonian equation (1) on a 3×4 dot, within the Hilbert subspaces corresponding to a fixed total number of electrons between 1 and 10. The ground-state energy and wave function, the total number of electrons $N = \langle\hat{N}\rangle$, and site occupation $\langle n_{i,\sigma}\rangle$ can be obtained for different values of the parameters entering Eq. (1). N is controlled by the voltage V_g. At a specific value V_g^{N+1} given by

$$eV_g^{N+1} = E_0^{N+1} - E_0^N \equiv \mu(N + 1)\,, \qquad (3)$$

the number of electrons on the dot jumps from N to $N + 1$. Here E_0^N is the ground-state energy for N particles at $V_g = 0$; $\mu(N)$ is the chemical potential, which is the quantity measured experimentally.

In Fig. 1 we plot N as a function of V_g for one random configuration in the regime of strong disorder ($W = 7.5t$) and intermediate values of Coulomb interaction $V = 2t$, corresponding to $r_s \approx 0.5$ when $N = 6$. For on-site repulsion $U = 8t$ (dashed line), the two additions $6 \Rightarrow 7$ and $7 \Rightarrow 8$ occur at close gate voltages; in contrast, all other electrons enter the dot at well-spaced voltages. Moreover, the tendency of electrons 7 and 8 to pairing is strongly enhanced by increasing U. This is clearly shown by the solid line in Fig. 1, where we consider the special case of *spin-polarized* electrons, which automatically enforces $U = \infty$. In this case the pairing particles still enter the dot *sequentially* but almost at the same gate voltage. Since this result refers to one particular disorder realization, one might think that it represents an extremely rare episode. Remarkably, however, this is not the case, as the inset of Fig. 1 shows: out of 8 random realizations that we have tried, half of them displayed pairing between the additions $6 \Rightarrow 7$ and $7 \Rightarrow 8$, when the system is polarized.

In order to understand how pairing takes place, we look at how the site occupation $\langle n_i\rangle$ changes when the two participating particles tunnel into the dot. Figure 2 displays $\langle n_i\rangle$ for the 3×4 sites of the dot, corresponding to the polarized case of Fig. 1. Because no double occupancy is allowed, the electrons tend to form one rather compact puddle (see the state $N = 6$ in Fig. 2). This is reminiscent of the effect of the exchange interaction studied within the Hartree-Fock approximation [12] for a dot in a strong magnetic field: exchange generates a local attraction between the electrons, causing the formation of a dense droplet. The 7th electron, the first involved in the pair, tunnels mainly into two almost empty sites at the right edge of this compact puddle, as shown by the grey circles of the $N = 7$ state. The 8th electron, the second involved in the pairing, fills up two already partially filled sites on the bottom edge of the dot. Thus, the pairing of Fig. 1 is associated with electron additions into spatially distinct regions of a compact electron puddle. Because the Coulomb interaction is short range, the energy costs of these two additions can be almost equal.

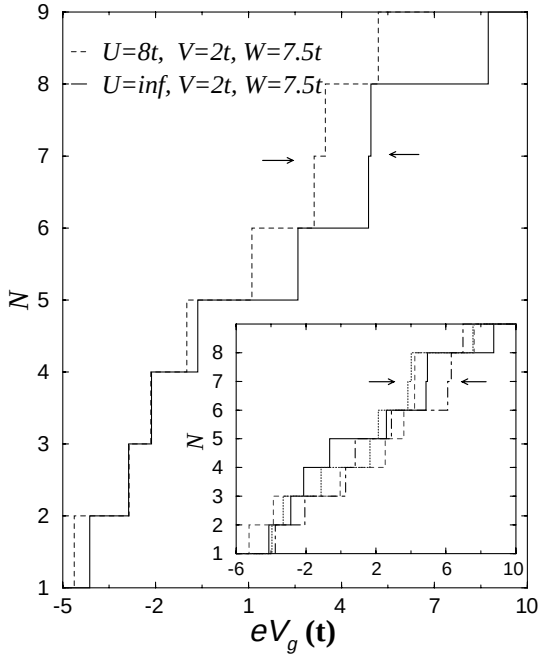

FIG. 1. Number of electrons N on a 3×4 dot vs gate voltage for one particular disorder realization ($W = 7.5t$) and nearest-neighbor Coulomb interaction ($V = 2t$). Dashed line: on-site interaction strength $U = 8t$. Solid line: spin-polarized electrons ($U = \infty$). The arrows point to the *pairing* between the additions $6 \Rightarrow 7$ and $7 \Rightarrow 8$. Inset: N vs V_g for four disorder realizations of polarized electrons, displaying a similar pairing.

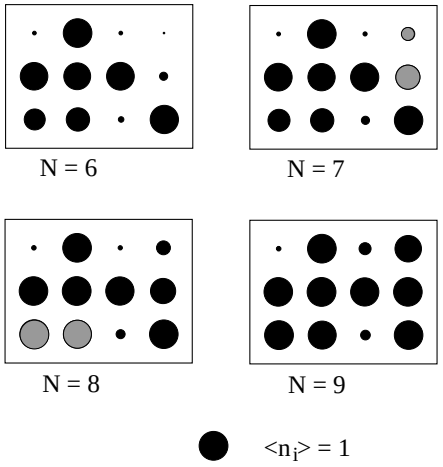

FIG. 2. Ground-state site occupation $\langle n_i\rangle$ for different values of N, for the dot of Fig. 1 (solid line). The area of the circles is proportional $\langle n_i\rangle$. Pairing occurs between the $N = 7$ and $N = 8$ states. The grey circles indicate the sites where the largest portion of the incoming electron, participating in the pairing, is distributed.

The results shown above hold for intermediate values of the Coulomb interaction ($V \approx 2t \Leftrightarrow r_s < 1$ for $N \sim 6$), much smaller than the strength of disorder and on-site repulsion. Experimentally, however, pairing occurs at larger values of Coulomb repulsion ($r_s \geq 2$). If we increase the strength of the direct Coulomb interaction relative to the on-site repulsion and the disorder, we reach the limit where the dot finds it more advantageous to generate local singlets of doubly occupied localized states. Instead of grouping into one compact puddle, the electrons can now form distinct puddles. Electron pairing can also take place in this strongly correlated regime.

An example is shown in Fig. 3, where we plot N vs V_g for a strength of the direct Coulomb repulsion $V = 9t$ ($r_s \approx 2$ for $N = 8$), close to the disorder strength, $W = 8t$. The two curves correspond to two values of the on-site repulsion, $U = 30t$ and $U = 29.06t$. We focus on the additions $7 \Rightarrow 8$ and $8 \Rightarrow 9$, where pairing takes place [13]. The two curves differ only in this region, indicating that the on-site repulsion U plays a crucial role in the pairing. We can again understand how this comes about by looking at the site occupation $\langle n_i \rangle$ for the states involved in the pairing. This is shown in Fig. 4 for $U = 30t$. The 8th electron, the first involved in the pair, enters the dot at the top left corner of the dot. The 9th electron jumps mainly into one of the central sites, which becomes partially *doubly occupied*. The dot shows a tendency to create two separate puddles of electrons, one composed of the 5 occupied sites along the left edge, and the second made up of the 3 sites of the right edge plus the doubly occupied site. Each electron involved in the pair tunnels into one of these spatially distinct regions. Finally, when the 10th electron enters the dot, the gap between the two regions is filled and the dot is occupied more uniformly. This example supports the suggestion, borne out of the experiment, that pairing is associated with electron localization in distinct puddles of the dot [4]. The merging of the two puddles upon increasing N corresponds to a sort of localization-delocalization transition.

If U is slightly smaller, e.g., $U = 28t$, the roles of the 8th and the 9th electrons are interchanged: the first pairing electron tunnels into the central site, creating the spin singlet; next, the second electron occupies the top edge site. By tuning U between $30t$ and $28t$, we have found that the intermediate value $U = 29.06t$ gives rise to the closest additions $7 \Rightarrow 8$, $8 \Rightarrow 9$. These two events still take place *sequentially* (see Fig. 3). For this value of U, an analysis of $\langle n_i \rangle$ reveals that, in each tunneling event participating in the pairing, half an electron goes into the central site and the other half goes into the top-left corner site. We have checked that some other disorder realizations display similar behavior.

The inset of Fig. 3 displays the magnetic flux ϕ dependence of the chemical potential [Eq. (3)] for the system corresponding to the "best pairing" ($U = 29.06t$). For such strong values of the interaction and the disorder, the traces of the addition spectrum are only very weakly dependent on ϕ. This implies that the pairing states $N = 8$ and $N = 9$ remain close to each other for the entire range of ϕ. This is also what happens experimentally for bunchings occurring at low $N < 10$.

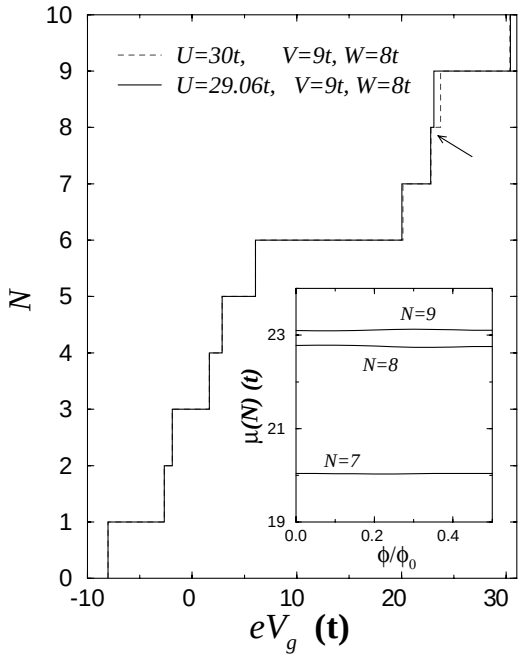

FIG. 3. Number of electrons N on a 3×4 dot vs gate voltage for one particular disorder realization ($W = 8t$) and nearest-neighbor Coulomb interaction ($V = 9t$). The two lines correspond to an on-site interaction strength $U = 30t$ (dashed line) and $U = 29.06t$ (solid line), respectively. The arrow points to the additions $7 \Rightarrow 8$ and $8 \Rightarrow 9$, where pairing occurs. The two lines differ perceptibly only at pairing. For the system relative to the solid line, the inset displays $\mu(N)$ vs the magnetic flux ϕ for $N = 7, 8, 9$.

So far we have discussed pairing only from the point of view of energy balance. We conclude by studying the *rates* at which the two electrons, involved in almost coincident

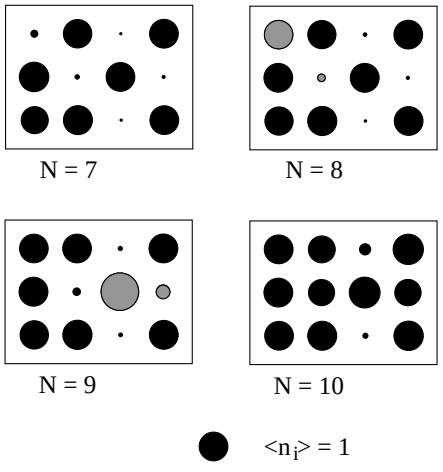

FIG. 4. Ground-state site occupation $\langle n_i \rangle$ of the 3×4 dot in Fig. 3 (dashed line, $U = 30t$). Pairing occurs between the states $N = 8$ and $N = 9$. Most of the second electron participating in the pair ($N = 9$) tunnels into a central site, which becomes partially doubly occupied, $\langle n \rangle = 1.70$.

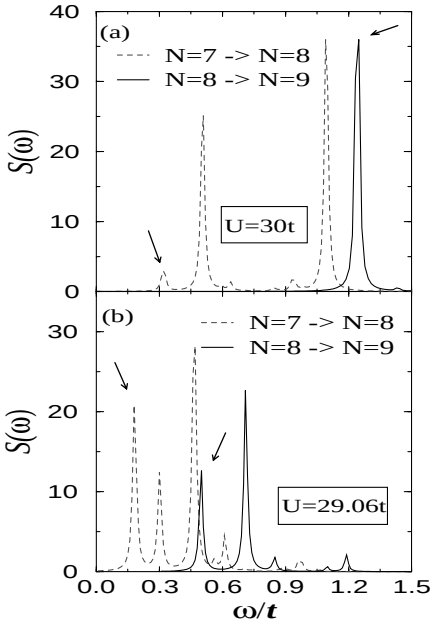

FIG. 5. The addition spectral function for the two particles forming a pair, as in Fig. 3. The dashed and solid lines refer to the first and second particles of the pair, respectively. The peaks indicated by the arrows are proportional to the tunneling rate, $1/\tau_{N+1}$, for the N- to the $(N+1)$-ground-state transition. (a) and (b) refer to two cases of Fig. 3.

additions, tunnel into the dot, when $r_s \geq 2$. Since the two events are independent, the rates can be obtained from the *one-particle* addition spectral function:

$$S(\omega) = \sum_n \left| \langle \Phi_n^{N+1} | \sum_{i,\sigma} c_{i,\sigma}^\dagger | \Phi_0^N \rangle \right|^2 \times \delta[\omega - (E_n^{N+1} - E_0^N)], \quad (4)$$

where $|\Phi_n^{N+1}\rangle$ is the eigenstate of the $(N+1)$-particle system corresponding to the energy E_n^{N+1}. The rate of tunneling, $1/\tau_{N+1}$, from the ground state of the N- to the $(N+1)$-particle system is proportional to the $n=0$ term in Eq. (4): $1/\tau_{N+1} \propto |\langle \Phi_0^{N+1} | \sum_{i,\sigma} c_{i,\sigma}^\dagger | \Phi_0^N \rangle|^2$. Equation (4) relies on the assumption that electrons tunnel with equal probability amplitude into all sites of the dot.

Figure 5 displays the spectral functions for the additions of the two pairing electrons in the strongly correlated regime of Fig. 3. The heights of the lowest-energy peaks of the dashed and solid curves (pointed out by the arrows) are proportional to $1/\tau_8$ and $1/\tau_9$, respectively. In Fig. 5(a) we can see that for $U = 30t$ the tunneling rate of the first particle of the pair is *much smaller* than the second. As discussed above, in this case the first particle is mainly localized into one corner site of the dot (see Fig. 4). The presence of fast tunneling in one state of the pair is also seen experimentally, albeit starting at strong magnetic field and at higher N. At low N the experiment shows that the tunneling rates of *both* particles are large for all magnetic fields. As shown in Fig. 5(b) the two tunneling rates are *both large* when U takes on the value that ensures the *closest additions*. This can be understood if we remember that in this case the electronic densities of the pairing particles are distributed among several sites of the dot. Thus, even if the system is strongly correlated, the tunneling rates for the pairing states are not negligible, in agreement with the experiment. In contrast, the two models in Refs. [5] and [6] predict a dramatic suppression of the tunneling rate at pairing.

In conclusion, our numerical simulations show evidence of pairing in the addition spectra of disordered quantum dots with strong on-site repulsion. Specific features of the pairing observed in the strongly correlated regime $r_s \geq 2$ suggest that our results may be related to the analogous phenomenon seen experimentally in the regime of a small number of electrons.

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