Disorder and interaction induced pairing in the addition spectra of quantum dots

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

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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 (CB), one of the most robust facts in mesoscopic physics.

By means of an experimental technique known as single-electron capacitance spectroscopy (SECS), 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; each tunneling event is recorded as a peak in the differential capacitance, measured as a function of the gate voltage.

A few years ago a SECS experiment showed that electrons sometimes entered the dot in pairs rather than individually, thus violating all the common wisdom that 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 (CB), one of the most robust facts in mesoscopic physics.

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Interaction as a nearest-neighbor repulsion, and nearest-neighbor Coulomb interaction (\(V = 2t\)). The on-site-interaction strength is \(U = 8t\) (dashed line) and \(U = \infty\) (solid line) respectively. 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 (\(U = \infty\)), displaying pairing between the \(N = 7\) and \(N = 8\) states.

Experimentally the Coulomb interaction is screened by the number of electrons on the dot jumps from \(N\) to \(\infty\) as the quantity measured experimentally. The important parameter describing the strength of pairing is \(V\), the total number of electrons.

By using Lanczos techniques, we have diagonalized the Hamiltonian Eq. (1) on a \(3 \times 4\) 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 \(\langle n_i \rangle\), 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

\[
ed V_g^{N+1} = E_0^{N+1} - E_0^N = \mu(N+1)\tag{3}
\]

de 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 the dot occupation \(N\) as a function \(V_g\) for one random configuration in the regime of strong disorder \((W = 7.5t)\) and intermediate values of Coulomb interaction \(V = 2t\), corresponding to \(r_s = 0.5\). For onsite 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 of Fig. 1 where \(U = \infty\), corresponding to spin-polarized electrons. In this case the two 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 represent 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 displays pairing between the additions \(6 \Rightarrow 7\) and \(6 \Rightarrow 8\), when the system is polarized, i.e. \(U = \infty\).

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. Fig. 2 displays \(\langle n_i \rangle\) for the \(3 \times 4\) sites of the dot, corresponding to the polarized case of Fig. 1. Because \(U = \infty\), no double occupancy is allowed and 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 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 \(3 \times 4\) dot vs. gate voltage for one particular disorder realization \((W = 7.5t)\), and nearest-neighbor Coulomb interaction \((V = 2t)\). The on-site-interaction strength is \(U = 8t\) (dashed line) and \(U = \infty\) (solid line) respectively. 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 \((U = \infty)\), displaying pairing between the \(N = 7\) and \(N = 8\) states.](image)

![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.](image)
Each electron involved in the pair tunnels into one of sites along the left edge, and the second made up by the rate puddles of electrons, one composed of the 5 occupied occupied one of the central sites, which becomes partially corner of the dot. The 9th electron jumps mainly into arrows points to the additions 7

\[ R \Rightarrow 7 \]

This is shown in Fig. 4 for two curves corresponds to two values of the on-site repulsion. Experimentally however, pairing occurs at larger dependence of the chemical potential, Eq. 3, for the system relative to the solid line, the inset displays \[ \langle n_i \rangle \text{ vs the magnetic flux \( \phi \) for } N = 7, 8, 9. \]

The results shown above hold for intermediate values of the Coulomb interaction \( V \approx 2t \Leftrightarrow r_s \leq 1 \), much smaller than the strength of disorder and on-site repulsion. Experimentally however, pairing occurs at larger values 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 more advantageous to generate local singlets of doubly occupied localized states. Instead of grouping into one compact puddle, the electrons now can form distinct puddles. Electron pairing can also take place in this strongly correlated regime.

An example is shown in Fig. 3, where we plot \( N \text{ vs } V_g \) for a strength of the direct Coulomb repulsion \( V = 9t \) \( \Leftrightarrow r_s = 2 \), close to the disorder strength, \( W = 8t \). The two curves corresponds 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. Note that the two curves differ only in this region, indicating that the on-site repulsion \( U \) plays a crucial role. 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 by 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 electron of the pairing 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 electron goes into the central site and the other half goes into the top-left corner site. We have checked that other disorder realizations display a similar behavior.

The inset of Fig. 3 displays the magnetic flux \( \phi \) 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 \( \phi \). This implies that the pairing states \( N = 8 \) and \( N = 9 \) remain close to each other for the entire range of \( \phi \). Experimentally, in bunchings occurring at low \( N < 10 \), the traces also stick together for the entire range of \( \phi \), but at the same time they rise nearly linearly, showing that the field dependence of the localized states approaches the one of the lowest Landau level.

So far we have discussed the occurrence of pairing only
The rate of tunneling from the ground state of the $N$-dot involves the sites of the dot with equal probability. The term in Eq. 4 implies that electrons tunnel coherently into all corresponding to energy $|\phi|^2$. To this purpose we have computed the addition spectral function, defined by

$$S(\omega) = \sum_n \left( \langle \Phi_n^{N+1} | \sum_{i,\sigma} c_{i,\sigma}^{\dagger} | \Phi_0^N \rangle \right)^2 \delta[\omega - (E_{n}^{N+1} - E_{0}^{N})]$$

(4)

where $|\phi_n^{N+1}\rangle$ are the eigenstates of the $(N+1)$-system corresponding to energy $E_{n}^{N+1}$. The operator $\sum_{i,\sigma} c_{i,\sigma}^{\dagger}$ in Eq. 4 implies that electrons tunnel coherently into all the sites of the dot with equal probability. The term in Eq. 4 involving $\langle \Phi_n^{N+1} | \sum_{i,\sigma} c_{i,\sigma}^{\dagger} | \Phi_0^N \rangle$ is proportional to the rate of tunneling from the ground state of the $N$- to the $(N+1)$-particle system.

Fig. 3 displays the spectral function for the addition of the two pairing electrons, corresponding to the situation of Eq. 3. The peaks pointed by the arrows are proportional to the tunneling rates. In Fig. 3(a) we can see that for $U = 30t$ the tunneling of the first particle forming 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. 3). 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. 3(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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