

Nonmonotonic charge occupation in double dots

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We study the occupation of two electrostatically coupled single-level quantum dots with spinless electrons as a function of gate voltage. While the total occupation of the double-dot system varies monotonically with gate voltage, we predict that the competition between tunneling and Coulomb interaction can give rise to a nonmonotonic filling of the individual quantum dots. This nonmonotonicity is a signature of the correlated nature of the many-body wave function in the reduced Hilbert space of the dots. We identify two mechanisms for this nonmonotonic behavior, which are associated with changes in the spectral weights and the positions, respectively, of the excitation spectra of the individual quantum dots. An experimental setup to test these predictions is proposed.

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INTRODUCTION

Quantum dots coupled to an electron reservoir can be filled with electrons one by one in a controlled way. With increasing gate voltage, the number of electrons on the dot increases in a steplike manner. As long as Coulomb interaction among the dot electrons is negligible, the dot levels are filled independently from each other. However, small semiconductor quantum dots are usually subject to strong Coulomb interaction: charging energy gives rise to extended plateaus of (almost) fixed dot charge as a function of gate voltage. The aim of the present paper is to demonstrate that more dramatic signatures of Coulomb correlations can be found in studying the filling of the individual levels in the quantum dot. While the total occupation of the dot remains a monotonic function of the gate voltage, we predict that, under circumstances specified below, the individual levels are filled in a nonmonotonic way. The physics proposed here enables one to observe nontrivial many-body correlations in a particularly simple (effective) four-dimensional Hilbert space.

To illustrate the signature and mechanisms of the nonmonotonic filling, we consider the simplest possible model system that allows for a separate access to the individual level occupation: two electrostatically coupled single-level quantum dots with spinless electrons, (see Fig. 1). The double dot is equivalent to a single-quantum dot accommodating two levels. The advantage of the double-dot setup lies in the possibility to read out separately each dot's occupation by electrostatically coupled quantum point contacts. This kind of charge sensing has been experimentally demonstrated for single¹ and double dots² recently. The quantum dots are tunnel coupled to one common or to two separate electron reservoirs.

We model the double-dot system with the standard tunnel Hamiltonian $H = H_{\text{dot}1} + H_{\text{dot}2} + H_{\text{ch}} + H_{\text{lead}1} + H_{\text{lead}2} + H_{T,1} + H_{T,2}$. Quantum dot $i=1,2$, described by $H_{\text{dot},i} = \epsilon_i c_i^\dagger c_i$, accommodates a level with energy ϵ_i , measured relative to the Fermi energy in the leads. The level energies can be tuned by the gate electrodes. Without loss of generality,

we always assume $\epsilon_2 \geq \epsilon_1$. The charging energy is accounted for by $H_{\text{ch}} = U n_1 n_2$. Each dot is coupled to an electron reservoir,^{3,4} $H_{\text{lead},i} = \sum_k \epsilon_{ki} a_{ki}^\dagger a_{ki}$. Tunneling is modeled by $H_{T,i} = \sum_k (t_i c_i^\dagger a_{ki} + \text{h.c.})$, where we assume the tunnel matrix elements to be energy independent. The tunnel coupling introduces a linewidth $\Gamma_i = 2\pi |t_i|^2 \rho_i$, where ρ_i is the density of states of lead i at the Fermi energy.

There are four possible states for the double-dot system: the double dot being empty ($\chi=0$), singly ($\chi=1,2$), or doubly ($\chi=d$) occupied. The corresponding energies are E_χ (Ref. 5). To address the question of how the occupations $\langle n_1 \rangle$ and $\langle n_2 \rangle$ of the dots vary as both levels ϵ_1 and ϵ_2 are simultaneously pulled down, we keep the bare level separation $\Delta = \epsilon_2 - \epsilon_1$ fixed. The fact that tunneling does not commute with the kinetic and charging terms is essential for the physics outlined below.

REGIMES

We begin with identifying the regime at which a nonmonotonic filling of the individual dots is expected. Consider first $U=0$. The spectral density of dot i is a Lorentzian centered around ϵ_i with a width Γ_i . As ϵ_i crosses the Fermi energy of the lead, the occupation $\langle n_i \rangle$ of level i changes

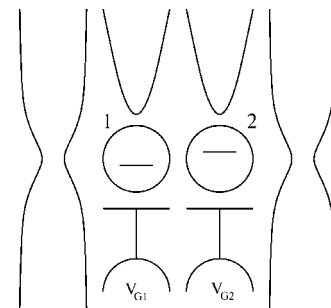


FIG. 1. Two electrostatically coupled single-level quantum dots are attached to electron reservoirs and gated by gate electrodes. The individual occupation is determined by adjacent quantum point contacts.

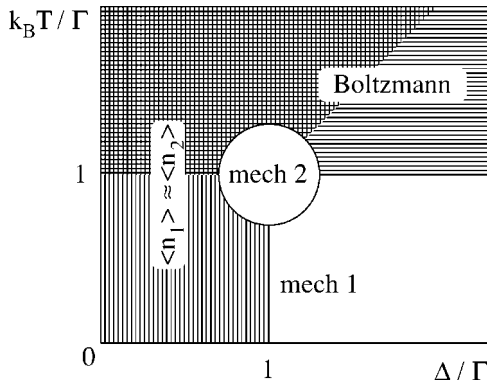


FIG. 2. For $k_B T \gg \Gamma$ or $\Gamma \gg \Delta$, the occupation of the individual dots is a monotonic function of the gate voltage. Nonmonotonicities due to mechanism 1 are strongest for $\Delta \sim \Gamma \gg k_B T$, and mechanism 2 becomes effective for $\Delta \sim \Gamma \sim k_B T$ and $\Gamma_1 \neq \Gamma_2$.

from 0 to 1. The width of this transition is governed by $\max\{\Gamma_i, k_B T\}$. It is obvious that in this case the occupation of each level is a monotonic function of the gate voltage. We thus turn our attention to the limit $U \gg \max\{\Delta, \Gamma, k_B T\}$.

For temperatures larger than the level broadening, $k_B T \gg \Gamma$, the influence of the tunnel couplings on the occupations is negligible and the double-dot system is filled according to the Boltzmann factors $\exp(-E_\chi^{(0)}/k_B T)$ of the corresponding bare energies $E_\chi^{(0)}$ with $\epsilon_i = E_i^{(0)} - E_0^{(0)}$ and $U = E_d^{(0)} - E_1^{(0)} - E_2^{(0)} + E_0^{(0)}$. Now the occupations of the two dots are correlated due to charging energy, $\langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle < 0$, but the individual dot occupations $\langle n_i \rangle$ are still found to depend monotonically on the gate voltage. Therefore, we require $\Gamma \geq k_B T$.

If the level separation is small compared to either the level width or temperature, $\max\{k_B T, \Gamma\} \gg \Delta$, then the double dot will be filled in a symmetric way, $\langle n_1 \rangle \approx \langle n_2 \rangle$, which again yields a monotonic filling. This restricts the regime of interest to $\Delta \geq \Gamma \geq k_B T$.

MECHANISMS FOR NONMONOTONICITY

We identify two different mechanisms by which a nonmonotonic filling of the individual levels can occur. They are both related to the Coulomb interaction, with two different consequences concerning the spectral density. First, as mentioned above, there is a spectral weight of level 2 not only around ϵ_2 (describing filling or depleting of dot 2 while dot 1 is empty) but also around $\epsilon_2 + U$ (describing transitions when dot 1 is filled). The total spectral weight is normalized to 1, with the relative spectral weight between the two peaks being determined by occupation of dot 1. Both peaks of level 2 are broadened by Γ_2 (Ref. 6), which yields a partial filling of the dot once the peak is close to the Fermi energy. As the level in dot 1 passes through the Fermi level and dot 1 is filled, the spectral weight for the level in dot 2 is transferred from the peak around ϵ_2 to the one around $\epsilon_2 + U$, which is further away from the Fermi level, yielding a reduction of the partial occupation of dot 2, giving rise to a nonmonotonic filling. This mechanism will be visible for $\Delta \gg k_B T$ (Fig. 2)

(since otherwise the filling is dominated by thermal fluctuations) and is most pronounced for $\Delta \sim \Gamma \gg k_B T$.

The second mechanism is based on a renormalization of the peak positions in the spectral density due to Coulomb interaction.⁷ As discussed in more detail below, tunneling in and out of dot 2 yields a renormalization of dot level 1, $\epsilon_1 \rightarrow \tilde{\epsilon}_1$. The renormalization is strongest when level 2 is close to the Fermi energy. This renormalization can give rise to a nonmonotonic filling of dot 2 under the condition $\Delta \sim k_B T$ and $\langle n_1 \rangle + \langle n_2 \rangle \approx 1$. In this case, both dots will be partially filled once $\tilde{\epsilon}_1$ and $\tilde{\epsilon}_2$ are below the Fermi level. The relative occupation is approximately given by $\exp(-\Delta/k_B T)$. A renormalization of the level splitting Δ then changes the relative occupation. If level 2 is close to the Fermi level, then level 1 is strongly renormalized towards higher energies. As a consequence, the occupation of dot 2 is increased at the cost of level 1, i.e., there is a reshuffling of the relative occupation. When the gate voltage is increased further, the renormalization becomes weaker, and level 1 gains back some of the occupation it lost to level 2, which leads to a nonmonotonicity of $\langle n_2 \rangle$. It turns out that for symmetric coupling, $\Gamma_1 = \Gamma_2 = \Gamma$, the renormalization of the level separation is negligible in the regime $\Delta \sim \Gamma \sim k_B T$ since both levels are renormalized approximately by the same amount. To get a nonmonotonic dot filling, an asymmetric coupling $\Gamma_1 \neq \Gamma_2$ is required.

PERTURBATION EXPANSION

To substantiate the qualitative ideas outlined above, we perform a perturbation expansion of the dot occupations in the tunnel-coupling strength Γ . For this, we employ a diagrammatic imaginary-time technique that was introduced to study charge fluctuations in a metallic single-electron box,⁸ adjusted to a two single-level-dot system. The central quantity is the partition function $Z = \sum_\chi \exp(-\beta E_\chi)$. The corresponding effective energies $E_\chi = E_\chi^{(0)} + E_\chi^{(1)} + \mathcal{O}(\Gamma^2)$ can be expanded in orders of Γ . Evaluation of $E_\chi^{(1)}$ yields $E_0^{(1)} = -[\sigma_1(\epsilon_1) + \sigma_2(\epsilon_2)]$, $E_1^{(1)} = -[\sigma_1(\epsilon_1) + \sigma_2(\epsilon_2 + U)]$, $E_2^{(1)} = -[\sigma_1(\epsilon_1 + U) + \sigma_2(\epsilon_2)]$, and $E_d^{(1)} = -[\sigma_1(\epsilon_1 + U) + \sigma_2(\epsilon_2 + U)]$, with $\sigma_i(\omega) = \Gamma_i/2\pi [\ln(\beta D/2\pi) - \text{Re} \Psi(\frac{1}{2} + i\beta\omega/2\pi)]$, where the bandwidth D appears as a high-energy cutoff at this intermediate step but drops out for all physical observables since they only depend on differences of E_χ 's. As a result, the dot levels are renormalized according to $\tilde{\epsilon}_i = \epsilon_i + \epsilon_i^{(1)}$ with

$$\epsilon_1^{(1)} = \frac{\Gamma_2}{2\pi} \text{Re} \left[\Psi \left(\frac{1}{2} + i \frac{\beta(\epsilon_2 + U)}{2\pi} \right) - \Psi \left(\frac{1}{2} + i \frac{\beta\epsilon_2}{2\pi} \right) \right], \quad (1)$$

for $\epsilon_2^{(1)}$ the same expression holds with 1 replaced by 2, and U remains unrenormalized in first order in Γ . Here, $\Psi(x)$ is the digamma function. From Eq. (1) we see that the renormalization of the level in dot 1 is proportional to the tunnel coupling of dot 2 (and vice versa). It is maximal when ϵ_2 or $\epsilon_2 + U$ is in resonance with the Fermi level, and vanishes in the absence of Coulomb charging.

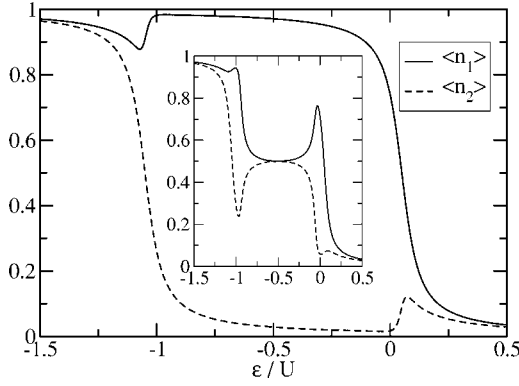


FIG. 3. Nonmonotonic dot filling due to mechanism 1. The parameters are $\Delta=0.1U$, $\Gamma_1=\Gamma_2=0.1U$, and $k_B T=0.01U$ (Ref. 9). Inset: Result for the symmetrized Hartree approximation.

Thermodynamic quantities such as the occupation of the dots are obtained by adequate logarithmic derivatives of the partition function. In particular,

$$\langle n_i \rangle = -\frac{1}{\beta} \frac{\partial}{\partial \epsilon_i} \ln Z, \quad (2)$$

$$\langle n_1 n_2 \rangle = -\frac{1}{\beta} \frac{\partial}{\partial U} \ln Z. \quad (3)$$

The first-order corrections of these quantities are directly obtained from

$$\frac{(\ln Z)^{(1)}}{k_B T} = \{ [1 - \langle n_2 \rangle^{(0)}] \sigma_1(\epsilon_1) + \langle n_2 \rangle^{(0)} \sigma_1(\epsilon_1 + U) + [1 - \langle n_1 \rangle^{(0)}] \sigma_2(\epsilon_2) + \langle n_1 \rangle^{(0)} \sigma_2(\epsilon_2 + U) \}, \quad (4)$$

plus a constant that is independent of ϵ_1 , ϵ_2 , and U . The set of Eqs. (2)–(4) provides the starting point for the subsequent quantitative analysis. For example, the correction to occupation of dot 2 is

$$\begin{aligned} \langle n_2 \rangle^{(1)} = & - [1 - \langle n_1 \rangle^{(0)}] \frac{\partial \sigma_2(\epsilon_2)}{\partial \epsilon_2} - \langle n_1 \rangle^{(0)} \frac{\partial \sigma_2(\epsilon_2 + U)}{\partial \epsilon_2} \\ & + \frac{\partial \langle n_2 \rangle^{(0)}}{\partial \epsilon_1} [\sigma_2(\epsilon_2) - \sigma_2(\epsilon_2 + U)] + \frac{\partial \langle n_2 \rangle^{(0)}}{\partial \epsilon_2} [\sigma_1(\epsilon_1) \\ & - \sigma_1(\epsilon_1 + U)], \end{aligned} \quad (5)$$

where we have used $\partial \langle n_2 \rangle^{(0)} / \partial \epsilon_1 = \partial \langle n_1 \rangle^{(0)} / \partial \epsilon_2$.

RESULTS

In Fig. 3 we show the dot occupations as a function of the mean-level position $\epsilon = (\epsilon_1 + \epsilon_2)/2$ in the regime $\Delta \sim \Gamma \gg k_B T$. We find nonmonotonicities which we relate to mechanism 1 discussed above. For an approximate analytical understanding of the result, we concentrate on the nonmonotonicity of $\langle n_2 \rangle$ on the right step. Since $\epsilon_2 > \epsilon_1$ and $\Delta \gg k_B T$, we can set $\langle n_2 \rangle^{(0)} \approx 0$ for the whole region over which level 1 is filled up, and $\langle n_1 \rangle^{(0)}$ is independent of ϵ_2 . Nevertheless, level 2 is partially filled due to quantum fluctuations,

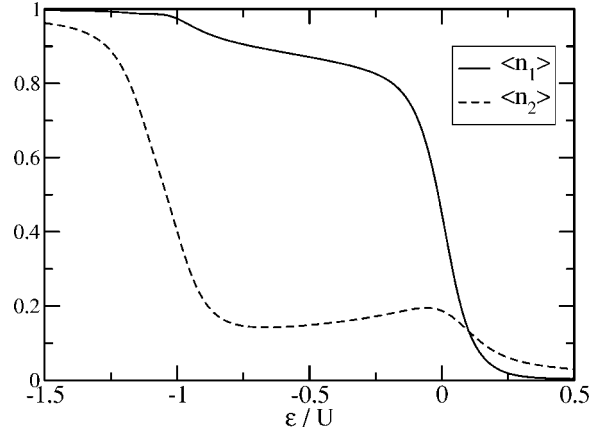


FIG. 4. Nonmonotonic dot filling due to mechanism 2 for $\Delta=0.1U$, $\Gamma_1=0.01U$, $\Gamma_2=0.1U$, and $k_B T=0.05U$.

$$\langle n_2 \rangle^{(1)} \approx [1 - \langle n_1 \rangle^{(0)}] \frac{\Gamma_2}{2\pi\epsilon_2} + \langle n_1 \rangle^{(0)} \frac{\Gamma_2}{2\pi(\epsilon_2 + U)}. \quad (6)$$

As level 1 passes through the Fermi level, filling up quantum dot 1, the occupation of dot 2 drops from $\Gamma_2/(2\pi\epsilon_2)$ down to $\Gamma_2/[2\pi(\epsilon_2 + U)]$. Within our perturbative analysis, the width of this drop is provided by temperature, as a broadening of level 1 due to quantum fluctuations would enter in higher orders only. When going to higher order in Γ , the width is expected to be $\max\{k_B T, \Gamma_1\}$. The amplitude of the drop is approximately

$$\delta \langle n_2 \rangle \approx \frac{\Gamma_2 U}{2\pi\Delta(\Delta + U)}. \quad (7)$$

We now turn to the regime $\Delta \sim \Gamma \sim k_B T$ to discuss mechanism 2. From inspection of Eq. (1) we realize that in the given regime the renormalization of both levels is roughly the same. In order to generate a substantial renormalization of the level separation that can give rise to a nonmonotonicity, we choose different coupling strengths $\Gamma_1 \neq \Gamma_2$. In the following we discuss the case $\Gamma_2 > \Gamma_1$. The result is shown in Fig. 4. The second line of Eq. (5) is associated with the renormalization of level 1. The renormalization (towards higher energies) is strongest when ϵ_2 passes through the Fermi energy. The peak of the nonmonotonicity is therefore located to the left as compared to the one related with mechanism 1. The width is given by Γ_2 . For an estimate of its height $\delta \langle n_2 \rangle$, we neglect both the possibility of the double dot being empty or doubly occupied, i.e., $\langle n_2 \rangle^{(0)} \approx \exp(-\beta\Delta)/[\exp(-\beta\Delta) + 1]$, and we assume $U \gg k_B T$. We find

$$\delta \langle n_2 \rangle \approx \frac{\beta e^{-\beta\Delta}}{(e^{-\beta\Delta} + 1)^2} \frac{\Gamma_2 - \Gamma_1}{2\pi} \ln \frac{\beta U}{2\pi}. \quad (8)$$

If we reverse the asymmetry of the coupling strengths, $\Gamma_1 > \Gamma_2$, we find, by using analogous arguments as above, a nonmonotonicity of $\langle n_1 \rangle$ on the left step, while $\langle n_2 \rangle$ remains monotonic.

In Fig. 5 we show the correlator $\langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle$, expanded up to first order in Γ , for the two sets of parameters

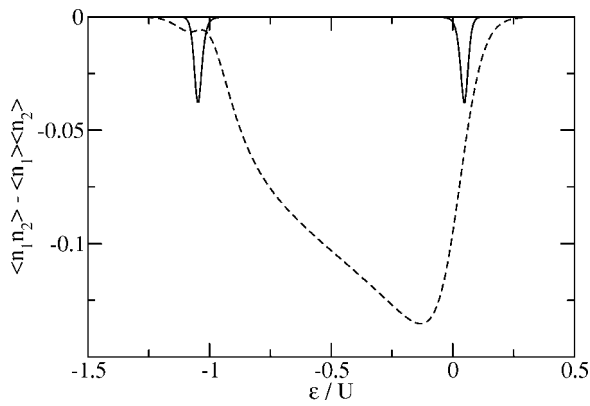


FIG. 5. Correlator $\langle n_1 n_2 \rangle - \langle n_1 \rangle \langle n_2 \rangle$. The parameters are as in Fig. 3 (solid line) and Fig. 4 (dashed line).

chosen in Figs. 3 and 4. We find that the nonmonotonicity of the dot occupation is accompanied with an enhancement of correlation.

Throughout our analysis, we assumed flatbands, i.e., particle-hole-symmetric densities of states, in the leads. As a consequence, $\langle n_1 \rangle$ at ϵ for given Γ_1, Γ_2 is identical to $1 - \langle n_2 \rangle$ at $U - \epsilon$ with the values of Γ_1 and Γ_2 being interchanged. Thus our discussion, which was focused on the right step of $\langle n_2 \rangle$, holds for the left step of $\langle n_1 \rangle$ as well (after interchanging $\Gamma_1 \leftrightarrow \Gamma_2$).

In parallel to this perturbation theory, we have performed an equation-of-motion (EOM) analysis up to the second hierarchy solving a set of 11 coupled equations for the various Green's functions. This approach, however, is not systemati-

cally controllable. Truncating the EOMs on the first hierarchy results in the symmetrized Hartree approximation¹⁰ with the Green's function $G_{11}^{\text{ret}}(\omega)$ given by $[G_{11}^{\text{ret}}(\omega)]^{-1} = [(1 - \langle n_2 \rangle)/(\omega - \epsilon_1) + \langle n_2 \rangle/(\omega - \epsilon_1 - U)]^{-1} + i\Gamma_1/2$ and similar for $G_{22}^{\text{ret}}(\omega)$. The resulting $\langle n_1 \rangle$ and $\langle n_2 \rangle$ tend to be biased towards each other, but the nonmonotonicity is still observed (see the inset of Fig. 3).

SUMMARY

We predict that the competition between tunneling and charging energy can give rise to nonmonotonic filling of individual quantum-dot levels. We identify two different mechanisms leading to nonmonotonicities and determine the regimes at which they occur. Based on a perturbation expansion in the tunnel couplings, we derive analytic expressions for the location and the strength of the signal.

Note added. While performing this work, we became aware of parallel work¹¹ in which nonmonotonic level occupations are discussed as well.

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³To describe a system in which both quantum dots are coupled to one common electron reservoir, one has to replace a_{ki} by a_k and to take into account the relative signs of the dot-level tunnel matrix elements, making a distinction between the “in-phase” and the “out-of-phase” scenario [A. Silva, Y. Oreg, and Y. Gefen, Phys. Rev. B **66**, 195316 (2002)]. On the level of calculations performed in this paper the results for all these models are identical.

⁴Nonmonotonic charge occupation is already found when replacing the reservoirs by single levels [P. Pou *et al.*, Phys. Rev. B **62**, 4309 (2000); A. Levy Yeyati *et al.*, *ibid.* **62**, 7307 (2000)].

⁵Here, E_χ is defined as the expectation value of the Hamiltonian in the subspace χ .

⁶To the order our perturbation theory is carried out, renormalizations of the golden-rule broadenings Γ_1, Γ_2 are not taken into

account [see, however, J. König, Y. Gefen, and G. Schön, Phys. Rev. Lett. **81**, 4468 (1998)].

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⁹The perturbative result for $\langle n_1 \rangle$ breaks down at low temperature when $\epsilon_2 \approx 0$. Therefore, in the plot we replace $\langle n_1 \rangle^{(0)} + \langle n_1 \rangle^{(1)}$ in this region by the occupation of a noninteracting level. This does not affect $\langle n_2 \rangle$, for which the perturbation scheme remains valid. The same procedure is done for $\langle n_2 \rangle$ around $\epsilon_1 \approx 0$.

¹⁰This is to be distinguished from the self-consistent Hartree approximation, in which the bare level energies ϵ_1 and ϵ_2 are replaced by $\epsilon_1 + U\langle n_2 \rangle$ and $\epsilon_2 + U\langle n_1 \rangle$. Within that scheme, the spectral density for each dot has *one* peak that is renormalized *linearly* with U . By contrast, our understanding of the nonmonotonic filling at low temperature (mechanism 1) is based on the fact that the spectral density of each dot level has *two* peaks. Mechanism 2, on the other hand, which is effective at higher temperatures, is related to a renormalization of the level positions that depend *logarithmically* on U .

¹¹M. Sindel, A. Silva, Y. Oreg, and J. v. Delft, cond-mat/0408096 (unpublished).