Occupancy calculations for quantum-dot-based memory devices

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Abstract. We investigate the addition spectra of arrays of quantum dots (QDs) under different geometrical distributions. We use a Hubbard Hamiltonian where we include intra- and inter-dot interactions. These parameters, which determine the correlation between the electrons, are varied on different QD array spatial distributions and the effects on the Coulomb staircase and conductance of the system are analysed. Exact diagonalization is used to calculate the eigenstates of arrays containing several QDs and the conductance addition spectrum is calculated using the Beenakker approach for a single dot generalized to an array of QDs. The charging/discharging process of the QDs is theoretically studied when a bias is applied to a metallic gate on top of the structure. The occupancy and conductance as functions of the gate bias are obtained, a crucial feature to the understanding of the memory charging process for non-volatile memories that are based on MOS devices with embedded semiconductor nanocrystals. A memory-device proposal application is given, where the storage of information is based on the charge present on the QDs, readable by means of the threshold voltage necessary to observe a current flowing in a narrow channel underneath the QD array structure.
1. Introduction and motivation

Non-volatile memories based on Si nanocrystals (NC) have attracted significant attention recently because of their potential for memory applications due to their characteristics: ultrafast data access, low power consumption, high endurance and long time storage [1]–[4]. The high compatibility of NC-based devices with integrated circuits makes them a suitable candidate for production on an industrial scale. The operation of Si NC flash memory devices consists of storing (programming), holding (retention) and removing (erase) charges in the NC by means of the gate voltage in a MOS device [5]. The content of the memory is then read by monitoring the source-to-drain current, as the charge of electrons stored in the NC screens the gate voltage, thereby altering the conductivity characteristics of the channel [6]–[8].

The low dimensionality of NC memory cells imposes behaviour almost entirely governed by quantum mechanics because (i) quantum tunnelling of confined electrons between the NC and the channel is the basic mechanism for write/erase operations and (ii) quantum confinement determines the conditions for charge detention in the NC. Such structures are so small that they can contain just one mobile electron; therefore it is also appropriate to refer to those NCs as quantum dots (QDs). By varying controllably the number of electrons applying a bias in these ‘artificial atoms’ and measuring the energy required to add successive electrons with a specialized transistor, a peak reproducible structure in the capacitance versus gate bias plot can be obtained, as will be described in this paper.

The nanocrystal memory is thus a single-transistor memory structure, where changes in threshold voltage are achieved by injecting charge into nanocrystals that act as a floating gate and are placed in between the channel and the metallic gate with an intervening barrier formed by a thin oxide [2, 8].

To understand the memory charging process, it is imperative to study the complex electronic correlation effects in the occupancy of these QD structures. Many theoretical studies have been done for the case of one QD [9]–[12]. Experimental groups have reported that the redistribution of injected electrons in the QDs floating gate proceeds stepwise as a result of the Coulomb interaction among the charged QDs [13, 14].

Here we present a theoretical study on the Coulomb staircase that describes the occupancy as a function of the applied bias, for two-dimensional (2D) arrays of QDs with different geometrical distributions and study the effects of the size and shape of different arrays of QDs, already controllable for self-assembled QDs [15]. The aim of this work is first to predict the role of the...
Figure 1. Schematic representation of a NC-based device: a voltage is applied in a metallic gate on top of the device and the QD layer acts as a floating gate, where different amounts of charge shift the conductance of the channel below the structure: (a) gate and QDs; (b) cross-section including the channel.

Coulomb interaction among the neighbouring charged QDs by studying the addition spectra, which describes the ‘additional’ energy to place an extra electron in a semiconductor QD, in analogy to the electron affinity for a real atom [16]. Secondly, to study the geometrical effects on such devices as self-assembled QD-based devices, where the position of the QDs grown, e.g. using the Stranski–Krastanov [17] method, can vary in different samples.

The simulations were done for arrays of QDs considering a Hubbard Hamiltonian, which includes strong coupling interactions among the QDs and weakly coupled to a lead, namely by resonant tunnelling through an oxide barrier (see figure 1), as in the flash-memory devices we intend to model [18].

2. Theory and model

The system we intend to model consists of an array of a few semiconductor self-assembled QDs. We consider the case where each QD contains only a few electrons and zero magnetic field; then the electron–electron interactions within the parabolic confining potential of a single QD are rather well described by a self-consistent Hartree energy for small variations about some large $n$, and can be parametrized by a capacitive charging energy $U_i$, in accordance with the Coulomb blockade (CB) picture [19] $U_i = n(n - 1)e^2/2C_i$ which works well for tunnelling between metallic electrodes. Here the capacitance $C_i$ is the sum of the capacitances of the top gate and the channel with respect to the QD ‘$i$’ and depends on the size of the QD, which can be controlled in the present day [20, 21]. For a semiconductor NC, size of a few tens of nm, the typical charging energy is in the 10–100 meV range [11, 13, 22]. We model collective phenomena at the meV energy scale in semiconductor QDs of area $\sim (10 \text{nm})^2$.

The Hamiltonian can thus be described using the extended Hubbard model [23] and adopted by many groups [13], [24]–[26], in which the Hamiltonian reads in second quantization language:

$$\hat{H}_0^{\text{dots}} = \sum_{i,\alpha} [\epsilon_i,\alpha - eV_g] \hat{c}^\dagger_{i,\alpha} \hat{c}_{i,\alpha} + \frac{1}{2} \sum_{i,\alpha\neq\alpha'} U_{ij} \hat{n}_{i,\alpha} \hat{n}_{i,\alpha'} + \sum_{i,j} w_i \hat{n}_i \hat{n}_j + \sum_{i,j,\alpha} t_{ij,\alpha} (\hat{c}^\dagger_{i,\alpha} \hat{c}_{j,\alpha} + \hat{c}^\dagger_{j,\alpha} \hat{c}_{i,\alpha}),$$

(1)

where $\hat{c}^\dagger_{i,\alpha}$ and $\hat{c}_{i,\alpha}$ are the creation and annihilation operators, respectively.

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The first term describes the single-particle energy levels of the individual quantum dots, the index \(i\) is the site and \(\alpha\) enumerates the energy levels, so \(\varepsilon_{i,\alpha}\) is the ground energy of this particular dot. The energy separation of the first few levels of the QDs is large \([27, 28]\). The electron addition energies have been measured \([29]\) and it has been noted that the addition energy for the third electron is significantly larger than the addition energy of the second electron, due to the single-particle gap between the s- and p-like states. Thus it seems to be adequate to consider only the first two single-particle energy levels in the confining potential of each QD which are nearest the Fermi level. This energy is shifted by \(-eV_g\), the gate potential, and is assumed to be distance-independent. This term effectively changes the QDs levels so at the end we get \(\varepsilon_i \to \varepsilon_i - eV_g\).

The second term describes the on-site Coulomb interaction, where \(U_i\) is the Coulomb repulsion on the \(i\)th dot and \(\hat{n}_i = \sum_\alpha \hat{c}_{i,\alpha}^\dagger \hat{c}_{i,\alpha}\) is the number operator. The third term gives the Coulomb interaction between electrons in dots \(i\) and \(j\), so \(w_{i,j}\) is the Coulomb repulsion between dots \(i\) and \(j\). The fourth term describes the electron tunnelling between dots, where \(t_{ij,\alpha}\) is the tunnelling matrix element between the \(\alpha\) states on dots \(i\) and \(j\). Tunnelling between non-resonant states is neglected by allowing only tunnelling processes where the spin is conserved; this is the usual tight-binding approximation and is justified for nearly identical dots provided \(t_o\) is not too large. This term gives the exchange correlation, already observed experimentally \([12, 30]\) competing with the CB and giving rise to rich addition spectra. Some groups \([31]\) have measured the effect of the tunnel splitting in the conductance peaks and compared with the additional resonance due to an excited dot state, finding the latter much higher. Thus for the range of voltages within our interest, it should be sufficient to consider a single level per QD.

Because the nanocrystals are located in the devices of interest far away from the metallic gate, the coupling between the wave functions in the control-gate contact and the QDs is negligible. We also assume that there is only weak coupling between the channel and the QDs. Thus, the array of QDs can be solved separately and then add a new perturbative term to (1), the tunnelling Hamiltonian, which describes the transfer of electrons between the lead (channel) and the QDs:

\[
\hat{H}_{1-d} = \sum_k \varepsilon_k \hat{c}_k^\dagger \hat{c}_k + \sum_k V_k (\hat{c}_i^\dagger \hat{c}_k + \hat{c}_k^\dagger \hat{c}_i),
\]

where the first term gives the energy in the different ‘\(k\)’ levels of the leads and the second term gives the tunnelling between state \(i\) in the QDs and \(k\) in the lead (channel). Thus, by lowering (increasing) the bias \(V_g\), the transfer process of one electron from (to) the QDs to (from) the channel is enhanced, as represented in figure 2.

To calculate the conductance of the system we thus add the tunnelling Hamiltonian (2) to that of the dots (1) and treat it in first order in perturbation for single-particle transitions, and the single-particle electron states in the QDs are considered to be sharp compared with the temperature \(k_B T\) and the Coulomb repulsion, \(U\). We evaluate the Hamiltonian \(H_{0,\text{d}}\) describing the decoupled QDs region in the occupation number notation via direct diagonalization. The conductance \(G\) is then calculated using the (single-QD) Beenakker approach \([19]\) generalized to the case of several QDs \([32]\) and adopted here to a situation of a single-channel transfer:

\[
G = G_0 N_{\text{max}} \sum_{n=1}^{N_{\text{max}}} \sum_{i,j} \Gamma_{ni,j}^{\text{C}} P_{n,i}^{\text{eq}} [1 - f(E_{n,i} - E_{n-1,j} - \mu)].
\]
Here $G_0 = e^2/k_BT$, $N_{\text{max}}$ is the maximum possible number of electrons in the sample; the QD array is weakly coupled to the leads in such a way that the temperature $T$ is large enough to neglect the Kondo effect due to correlations of the electrons in the leads with the ones in the central region but small enough to neglect mixing in of higher lateral states [24] and observe quantum effects. $\Gamma_{ni}^C$ is the transmission matrix from the $i$th $n$th-particle state to the $j$th $(n-1)$th-particle state via tunnelling to the channel, given by

$$\Gamma_{ni}^C = 2\pi \sum_{\sigma} |\langle n, i | V_{km\sigma} c_{m\sigma}^\dagger | n-1, j \rangle|^2.$$ (4)

This quantity describes the correlations in the system, and its contribution to the current proves to be dominant in determining the salient features measurable in experiments [33]. To facilitate the calculations, we consider a coupling scheme of the dot array to channel as indicated in figure 2, where the transfer of electrons is carried out by only one mode [34], i.e. $V_{km\sigma} = V_{0m}$, with $m$ representing the GS of the $m$th QD. $P_{eq}^{n,i}$ is the equilibrium probability for occupying the eigenstate $(n, i)$ with eigenvalue $E_{n,i}$ given by

$$P_{eq}^{n,i} = (1/Z) \exp \left[ - (E_{n,i} - n\mu)/k_BT \right].$$ (5)

$Z$ is the partition function, defined as usual:

$$Z = \sum_{n,i} \exp \left[ - (E_{n,i} - n\mu)/k_BT \right]$$ (6)

and $f(E_{n,i} - E_{n-1,j} - \mu)$ is the Fermi–Dirac distribution function. The electronic states in the leads are assumed to be 1D subbands filled according to Fermi–Dirac statistics and $(1 - f)$ indicates the probability of finding an empty state in the lead, which satisfies the energy conservation requirement for the $(n, i) \rightarrow (n-1, i)$ transition. The geometry of the system is crucial in this work, since the parameters describing the Coulomb inter-dot interactions, $t_{i,j}$ and $w_{i,j}$, are distance-dependent, affecting the conductance peaks [35]. The symmetry of the system affects significantly the Coulomb staircase of the array of QDs, as will be shown in this work.

Figure 2. Schematic representation of the band-edge profile model of the QDs.
3. Results

We predict theoretically the behaviour of the response of a few coupled QDs to propose a flash-memory device based on a limited and controlled number of nanocrystals. In this section, we present different simulations for a Coulomb staircase, showing the occupancy in each \( j \) QD, \( Q_j \), versus the gate bias. Note that due to the coupling among the QDs, \( Q \) can be fractional, but the sum \( \sum_j Q_j = n \), with \( n \) being an integer and equal to the number of extra electrons in the system, since the array is weakly coupled with the channel through an oxide layer and the tunnelling will depend on the layer thickness [36].

3.1. One-dimensional arrays

3.1.1. Degenerate case. In figure 4, each plot represents the electronic occupation (vertical axis) as a function of the gate voltage in each dot for 1 (a) to 7 (g) QD systems. The solid lines represent the conductance through the array and to the channel, calculated as in equation (3). As expected, for every trapped electron, there is a transfer of charge so that the conductance is enhanced. Note that the horizontal axis represents the energetic contribution of the gate bias \(-eV_g\) on the QDs so that the sign in the gate voltage is changed.

The QDs are equidistant and have the same single-particle energy. They can be considered here as in a 1D chain, as represented schematically in figure 3. The direction of the 1D chain is parallel to the channel.

In the case of only one QD, figure 4(a), the situation is trivial: the QD traps a charge when the gate bias, \( V_g \), is such that the Fermi level coincides with the eigen-energy in the QD (see figure 2), so that a tunnelling process is favoured (right to left). There is an intermediate state where only one electron occupies the QD, and the length of the plateau depends on the intra-dot Coulomb repulsion, \( U \). When the gate bias is strong enough so that the double occupancy of the QD is favoured, a second electron can be trapped by the QD.

In figure 4(b) with 2 dots, the situation is similar and symmetric: the hopping and matrix elements are symmetric, \( t_{12} = t_{21} \) and \( w_{12} = w_{21} \) (see figure 3). In figure 4(c) with 3 dots, the symmetry is broken since the interactions are distance-dependent, in such a way that the middle QD (dot 2) interacts strongly with the other two, whereas the ‘left’ (dot 1) and ‘right’ (dot 3) QD have a weaker interaction between them, since the distance between them is larger. Therefore, in (c), dots 1 and 3 have a similar behaviour (\( t_{12} = t_{23} \) and \( w_{12} = w_{23} \)) but dot 2 is different. In figure 4(d) again, the QDs at the extremes of the chain (dots 1 and 4) have a similar behaviour, so do the middle ones (dots 2 and 3); therefore two different behaviours are expected and observed. A similar explanation can be given for the cases of 5 (e), 6 (f) and 7 (g) dots.
Figure 4. These plots correspond to simulations with 1 (a) to 7 (g) dots, all of them with the same energy and effectively placed in a uniform and linear chain, with distance-dependent interactions.
Figure 5. These plots correspond to simulations with 2 (a) to 5 (d) dots, all of them with different energies, separated by 0.2 meV: $\varepsilon_{i+1} - \varepsilon_i = 0.2$ meV.

The shift of conductance peaks due to tunnelling has been experimentally measured [37], where the excitation energies of an individual QD were found to be much higher. Thus, the consideration of a single level in the QDs can be justified in the range of $V_g$ we are considering.

For a reduced QD number, $U$ is the most crucial parameter in our calculations, which determines the separation between the two mean sets of peaks. As the number of QDs considered in the system increases we reach a regime where both intra- and inter-dot interactions shift the conductance peaks by the same order of magnitude.

3.1.2. Non-degenerate case. The following simulations (figure 5) were carried out for a system of QDs with different energies, which is the usual case in nature, since the size of self-assembled QDs varies slightly from one QD to another [38]. As expected, the symmetry is broken already in the two-QD case, since the single-particle energies align with the Fermi level at different gate potentials.

The addition spectra is not changed significantly in this results and a number of $2 \times n$ peaks will still be observed independent of the single-particle energy of each QD, where $n$ is the total number of QDs in the array.

3.2. 2D arrays

Self-assembled semiconductor QDs are formed randomly on a substrate; thus a more realistic picture requires consideration of 2D arrays (figure 6). In this section, the model is applied to
uniform and random patterns of 3, 4, 5, 6 and 7 QDs, illustrated in the insets of figure 7, to explore the geometrical effects.

In figure 7(a), the 3 dots are placed equidistant to each other, thus the behaviour is identical: the geometrical symmetry brings identical interactions among all the QDs, so the charging–discharging process takes place in each QD at the same time. In figure 7(b), the inter-dot distance is random, so all the intra-dot interactions are different and the pattern in the addition spectra is not coincident. In figure 7(c), 4 QDs placed in the corners of a perfect square, the symmetry is equivalent as in figure 7(a). In figure 7(d), 5 QDs with such pattern shows that the middle quantum dot (dot 5) has a different addiction spectrum than the rest of the 4 QDs placed as in (c) breaking the symmetry of figure 7(c). In figure 7(e), the symmetry is perfect, with six equidistant and thus symmetric interactions. In figure 7(f), the pattern is logically equal for the QDs in the corners of the figure and independent for QDs 2 and 5. In figure 7(g), the situation is again similar to that in figure 7(d), where the middle QD behaves differently. The dimensionless conductance is also plotted, $\sigma \propto G/G_0$, showing peaks where an electron transfer to (or from) the channel is expected.

Thus, measurements of conductance versus gate voltage can reveal the symmetry of QD arrays. This could be used as a diagnostic tool to determine the accuracy of positioning QDs through lithography [18] or pre-patterning of substrates [20].

3.2.1. Four-QD systems. In figure 7(c), the result of a four-QD array gives a total of eight peaks in the addition spectra. This result could be applied to a memory device with eight different positions if the correct gate voltage could be imposed in the device. Then the threshold voltage $V_{th}$ necessary to get a current in a electrostatically defined channel underneath the sample as in [36] would be a single-valued function of the total charge contained in the system and thus it could be used as a 8-bit memory storage device. The peak structure is completely reproducible as one scans the gate voltage up and down.

Such a system would be possible if a small ac voltage was added to the dc gate voltage as in [16]: when the gate voltage is adjusted to a voltage at which an electron can be added to the QDs, the ac voltage causes an electron to tunnel back and forth between the QD array and the channel and charge appears synchronously with the ac voltage. As a result, a peak in the capacitance signal can be detected. A narrow channel underneath the NC structures will show a shift in the threshold voltage necessary to observe a current.

In figure 8, the relevant parameter to observe such effect, the total charge per QD in the QD array, $\delta = 1/N \sum_{i=1}^{N} q_i$ is represented versus the gate voltage. A total of eight peaks in the capacitance signal is observed for all the different array spatial distribution.
Figure 7. Different geometries for identical QDs: in (a), (c) and (e) the QDs are placed symmetrically; thus the addition spectrum for each QD is equivalent. The symmetry is broken in (b), (d) and (e). In (g), both the occupancy and the dimensionless conductance, $\sigma \propto G/G_0$ are represented.
Figure 8. Different geometries for 4 QDs array; ——, conductance of the system; – – –, charge per QD present in the structure.

4. Conclusions

The calculations shows that the symmetry of one array of QDs is reflected in the quantum staircase. The aim of this work is to predict the occupancy of the QDs in terms of all the possible relevant quantities that may be controlled in a real system.

In general, an array of similar QDs that presents an ordered structure will present a more homogeneous output and the addition spectra, which represent the conductivity versus the gate bias, will show a reduced number of peaks, one every time there is an electron transfer in or out of the system. The shape of such a spectrum is not significantly changed when the QDs present different single-particle energies.

The conductance of a channel below the array can be made to depend on the charge of the array, which gives, in real systems, a means to detect trapped charges and thus read the charge state. We have shown that the subtle combination of symmetries in the array can give a unique output in the Coulomb staircase.

A system containing a total of four self-assembled QDs could be used to store 8 bits of information, since the addition spectra present a total of eight well-defined peaks, each one of which could represent 1 bit. We propose a simple model for a possible memory device application.

The simulations have been done in site representation (spatial coordinates) for an array of QDs, allowing the calculations of the spin of the system, so our model could be used in
future calculations to include the effect of an external magnetic field and measure the tunnel splitting of the states in the system, which could be used for the realization of qbit-based memory devices [39].

The occupancy calculations presented here may be interesting for the realization of the idea of quantum cellular automata and logic elements on the basis of them, where the array of QDs can be considered as one of the cells [40].

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