PhD Thesis

Burkhard Ritter

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Chapter 1

Introduction

Chapter 2

Quantum-dot cellular automata

Lent et al. introduced the concept of quantum-dot cellular automata as an alternative computing paradigm in 1993 [1]. Thus the aim was a novel physical scheme to build digital circuits that would overcome some of the limitations of CMOS technology, promising potentially lower power consumption, higher device density, and faster clocking. As the name alludes to, quantum-dot cellular automata (QCA) is built from quantum-dots which are grouped together in cells. Figure 2.1(a) shows a basic QCA cell. Four quantum dots are arranged on the corners of a square. The dots are idealized as perfectly localized single orbitals on a perfectly decoupled non-intrusive medium. Therefore, each dot can be occupied by up to two electrons. In the QCA scheme, however, each cell is occupied by only two electrons in total. The cell is quarter-filled. The electrons tunnel between different dots in a cell, but the dominant energy scale is set by the Coulomb repulsion between the particles. Simply by virtue of the Coulomb repulsion, and ignoring the comparatively small tunnelling for now, the diagonal states, Fig. 2.1(a), are the two energetically preferred electron configurations. In comparison, edge states or doubly occupied quantum dots are unfavourable higher energy states, Fig. 2.1(b). A priori the two diagonal states are energetically degenerate, but this degeneracy can be lifted by introducing an external Coulomb potential, for example a second nearby QCA cell. Then these two states can be identified with logic 0 and 1, as indicated in the figure.

A single cell by itself is, of course, not very interesting. Thus, multiple cells can be positioned next to each other, for example as a straight line of cells, Fig.2.1(c). The approach now again assumes that Coulomb is the driving force and that electron tunnelling between cells is very small and ideally zero. For a straight line of cells, these long-ranging, unscreened Coulomb forces will tend to align the electron configurations of adjacent cells. If the first cell is in logic state 1 then the second cell will also prefer logic state 1 and so will in turn all the other cells in the line. The situation is the same for logic state 0. Therefore, a straight line of cells is similar to a wire not only in geometry, but also in functionality: It transmits a digital signal. The same is true, with slight modifications, for a diagonal

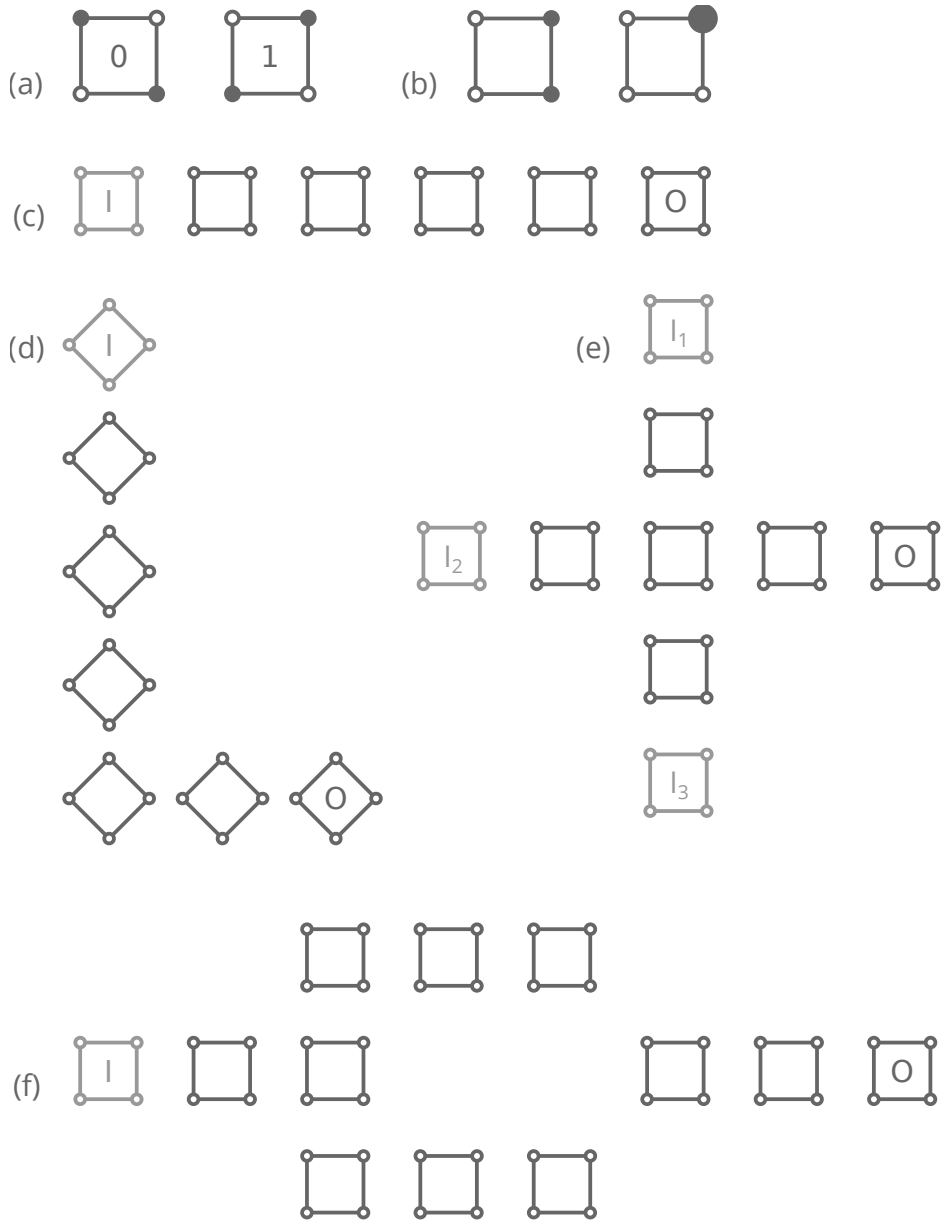

Figure 2.1: ...

line of cells — cells rotated by 45°—, Fig. 2.1(d). In this case the signal alternates from cell to cell, that is, logic 1 will follow logic 0 which followed from logic 0, and this again is simply by virtue of the dominant Coulomb interactions between electrons on different cells. By using an even number of cells the diagonal line of cells works as a wire just as well as a straight line of cells. The pictogram also demonstrates a 90° for the diagonal line of cells which our newly gained intuition for these Coulomb-driven systems expects to pose no problem for signal transmission.

The main idea of the QCA approach becomes apparent: Ideally bistable cells interact with each other solely by Coulomb repulsion. By arranging the cells in clever geometries we can realize interesting functionalities. The idea as such is quite general and does not strictly rely on the two-electron-four-dot cell introduced above. Indeed, a number of variations exist, such as one-electron-two-dot cells, interacting via dipole fields instead of qudrupole fields as for the conventional cells, or four-dot cells with six electrons—two holes—instead of two electrons. Even the interaction need not be Coulombic. For example, magnetic QCA schemes have been explored. While QCA carries "quantum" in its name and is sought to be implemented at the nanoscale, the approach operates close to the classical limit. The Coulomb interaction is absolutely dominating with the tunnelling of electrons a small perturbation, which nonetheless drives the system's dynamics. The approach is insensitive and in fact ignores the spin degree of freedom. Let us finally note that QCA is a not a cellular automata in a strict mathematical sense, but only by analogy to the idea of interacting cells.

One clever geometrical cell arrangement is the majority gate, Fig. 2.1(e). The gate has three inputs which "vote" on the central cell. The majority wins and sets the single output. The device is commonly operated with one fixed input, for example $I_3 \doteq 0$ or $I_3 \doteq 1$. In the first case, $I_3 \doteq 0$, the device functions as an AND gate for the remaining two inputs, $O = I_1 \wedge I_2$. In the second case, $I_3 \doteq 1$, it is an OR gate with $O = I_1 \vee I_2$. Now, the only missing piece for Boolean algebra is negation, $O = \neg I$. We had already seen that simply arranging cells at an 45° angle as in the diagonal line of cells above negates the signal from cell to cell. The inverter, Fig. 2.1(f) recasts this idea into a more robust layout. With that we have, at least in principle, all the necessary building blocks for Boolean algebra and thus digital circuitry.

Conceptually, it is most elegant to set the inputs to a QCA circuit via driver cells — cells that resemble the QCA cell in form, but are made up from static point charges instead of quantum dots. These static charges are thought to be manipulatable to vary the input smoothly from the logic 0 to the logic 1 state. In figure 2.1 these driver cells are represented in light grey. Of course, in practice such driver cells would be difficult if not impossible to implement and the input would more likely be set by leads that provide the necessary perturbative electrostatic field. The output of a QCA device can be directly read from its output cells. In practical implementations this will require a non-trivial charge probing apparatus.

Changing the input for a QCA device throws the system into an excited, non-equilibrium

state. The system will then dissipatively propagate to its new ground state. For the given inputs, this ground state corresponds to the solution of the computational problem the circuit is designed to solve. Let us emphasize this: In QCA, the computational solution maps directly to the physical ground state! At all times during performing a computation, the system is relatively close to its ground state. Only a few charges move locally, in each cell. QCA is a truly current-free approach and consequently inherently low-power, especially when compared with CMOS technology. But the operation close to the ground state also raises concerns for the operational temperature for these devices. It is clear that for applications we would want to engineer the system so that the energy gap between the ground state and the low-lying excited states far exceeds room temperature.

It is difficult to derive general expectations for the clocking speed of QCA circuits. The switching speed of a majority gate, for example, will greatly depend on the system's parameters, but particularly on the nature of the dissipative coupling of the circuit to its environment. A small dissipative coupling will have the output polarization oscillating before it eventually settles to its correct value. A very dissipative system in contrast might get stuck in meta-stable states. Different material systems provide different dissipative channels and modelling them quantitatively or even qualitatively correctly is very challenging.

QCA circuits consist of wires, gates, and other structures arranged on a two-dimensional surface—very similar to conventional electronics devices. However, the structures themselves are quasi-one-dimensional and this poses a challenge for building large-scale QCA circuits. A good example is a single long wire, which is truly one-dimensional. Generally, a one-dimensional system cannot be ordered in the thermodynamic limit except at zero temperature. Therefore, the finite-temperature infinitely long wire will always have at least two different domains, logic 0 and logic 1 (assuming perfectly bistable cells for simplicity), and thus not be able to transmit a signal. When we think about switching the input for the wire, we think of the information being propagated as a charge density wave throughout the wire, or, equivalently, as propagating the domain boundary between logic 0 and logic 1. This domain boundary incurs an energy cost that the system seeks to minimize. For an increasingly longer wire, however, the gain in entropy for moving the domain boundary freely throughout the wire ($S \sim \log N$, N the number of cells) soon exceeds the loss in energy, which is reflected by the free energy of the system, $A = U - TS$. Consequently, the gap between the first excited state and the ground state and the desired operational temperature will determine the maximum system size.

To address this scaling problem we partition large circuits into smaller units. Each unit can be turned "on" and "off" separately: Individual gates allow to control the electrostatic potential for each unit, effectively raising and lowering the tunnelling barriers between quantum dots and thus allowing to freeze or delocalize the electrons. A unit with *frozen* electrons can serve as the input for a unit with more *active* charge carriers, which works like a regular QCA circuit. A unit with completely *delocalized* electrons, in contrast, will not influence adjacent units. By putting each unit through the three phases *delocalized*, *active*,

and *frozen* and synchronizing adjacent units appropriately, we can control the information flow through the system very nicely, as illustrated in Fig. TODO. Therefore, by partition the circuit and introducing a clocking scheme we not only handle the scaling problem, but also arrive at a pipelining architecture that greatly improves the control over information flow in the system. Of course, in practice the QCA circuit units cannot be too small as they must be individually addressable. Gates which turn QCA units "on" and "off" also provide another potential benefit. We are able to control how and especially how fast the gate voltage is changed and should be able to tune it with respect to the inherent time-scales of the QCA system, which are set by the system's parameters and the dissipative coupling to its environment. This should allow a better control over the dynamics of the switching process.

Our objective is the general, not implementation-specific characterization of the QCA approach. Even so it is still important to consider concrete experimental realizations, not only as a motivation for our work, but also to put our modelling and results into context. One of the most promising and recent experimental implementations of QCA is based on atomic silicon quantum dots and we will therefore use them as our experimental reference. Atomic silicon quantum dots were first demonstrated as a possible QCA implementation by Wolkow et al. in 2009, when the group realized one single QCA cell. Fig. 2.2(a) shows a scanning tunnelling microscope (STM) image of this cell. Since then impressive advances have been made both in the understanding of the electronic properties of these quantum dots as well as in the precise fabrication of larger QCA structures. With atomic-scale feature sizes this experimental system promises room temperature operation, while at the same time tapping into the established and highly-sophisticated silicon technology. Being based on silicon should also ease integration with existing CMOS circuitry.

Atomic silicon quantum dots are *dangling bonds* on a hydrogen-terminated silicon (100) surface. Atoms on a (100) silicon surface have two unsatisfied bonds. Pairs of surface atoms form dimers, satisfying one bond. The remaining bond is satisfied by passivating the surface with hydrogen. Fig. 2.2(c) shows a STM image of the reconstructed silicon surface, where the dimer rows are clearly visible and the dimensions are indicated. By applying a relatively large current through the STM tip, individual hydrogen atoms can be removed, with atomic precision. This leaves a *dangling bond* (DB) which acts as a quantum dot: Energetically, electrons on the DB orbital sit in the silicon band gap (see Fig. 2.2(b)) and are therefore decoupled from the silicon substrate. Chemically, DBs have proven to be surprisingly robust with respect to environmental molecules. From *ab initio* calculations it is known that the sp^3 DB orbital extends predominantly into the bulk and only a little into the vacuum. The orbital's lateral extent is on the order of $1nm$ and therefore spans multiple silicon lattice atoms. Due to the orbital overlap, closely spaced DBs are tunnel-coupled. The neutral DB consists of the positive silicon ion and one electron. In the experimentally common strongly n-doped system, the DB accepts one more electron and is therefore $-1e$ negatively charged. Conversely, in a p-doped sytem the DB will donate its electron and become $+1e$ positively charged. The Coulomb repulsion between negatively charged DBs

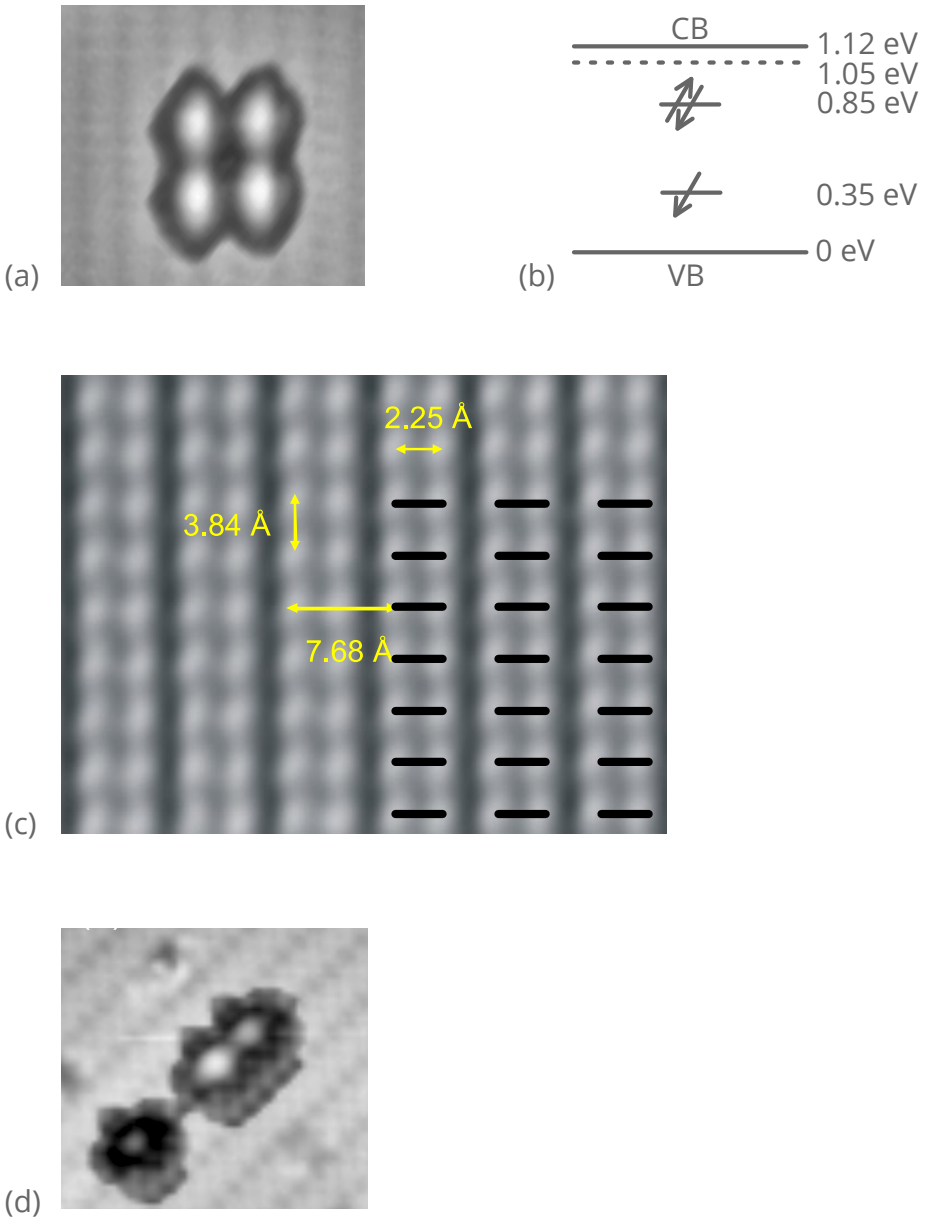

Figure 2.2: ...

can be used to adjust the filling of DB assemblies simply by the DBs' positions. For example, on a n-doped substrate two DBs may eject one electron (which goes back to the bulk) and share the remaining single electron, when placed close enough together. To proof this, a third DB is placed close by, but not close enough to be tunnel-coupled. The effect of the Coulomb repulsion can be seen via STM imaging, Fig. 2.2(d), where the DB farthest from the perturbing external charge is more negatively charged (darker in the STM image) than the closer DB. The observed charge shift is only possible when both closely-spaced DBs share a single electron. To form the previously shown QCA cell, Fig. 2.2(a), four DBs are brought close enough together so that two electrons go back to the bulk, leaving the cell with six electrons in total and a cell net charge of $-2e$—the right charge regime for QCA.

Atomic silicon quantum dots provide some examples of how a real world system might be different from the idealized picture we typically employ to describe the QCA approach. We like to think of quantum dots as perfectly localized orbitals. But in the silicon system the orbitals of the DBs actually span multiple lattice sites and only if the DBs are placed far enough apart, might we still be able to consider them as localized. We do not consider the substrate but treat quantum dots as perfectly isolated entities. Of course, in practice the substrate will certainly influence the QCA device. In the silicon system free charge carriers will screen the long-ranged Coulomb interactions that the QCA scheme relies on. The screening is not necessarily disruptive for QCA and might even be beneficial, for example by minimizing charge-buildup in large systems. But to quantify the screening effect accurately, thorough understanding and precise modelling are necessary, which, for atomic silicon quantum dots which live at the surface, would surely be very challenging. The silicon substrate could also, conceivable, provide a second tunnelling channel between DBs. In addition to electrons hopping directly from DB to DB they could first tunnel from the first DB to the substrate and then back to the second DB. Thus an accurate model for atomic silicon quantum dots might need to accommodate the nature of the DB orbitals, screening, multiple tunnelling channels and other effects.

QCA systems are typically modelled by an extended Hubbard Hamiltonian. The Hubbard model originated in the early 1960s to describe rare-earth systems with highly localized d- and f-electrons and has since then, of course, become one of the most widely studied and successful models in condensed matter physics. In basing our description on the Hubbard model we already put some key assumptions in place. For example, we assume that the quantum dots are similar to the highly localized d-orbitals. As discussed above, depending on the particular QCA implementation this might or might not be a good description. However, our interest is not in the precise details of any particular material system QCA might be implemented on, but our aim is to investigate universal characteristics of QCA systems. An idealized but semi-realistic description is what we want and for that the Hubbard model is indeed an appropriate—and tractable—starting point. Specifically, the

Hamiltonian we use is

$$H = -\sum_{ij\sigma} t_{ij}\, c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_{i} n_{i\uparrow} n_{i\downarrow} - \mu \sum_{i\sigma} n_{i\sigma} + \sum_{i<j} V_{ij}\, (n_{i\uparrow} + n_{i\downarrow} - q)\, (n_{j\uparrow} + n_{j\downarrow} - q)\ , \tag{2.1}$$

where $c^{\dagger}_{i\sigma}$ ($c_{i\sigma}$) creates (annihilates) an electron on quantum-dot i with spin σ and the particle number operator is $n_{i\sigma} = c^{\dagger}_{i\sigma} c_{i\sigma}$; t_{ij} is the overlap integral between dots i and j, U is the Hubbard on-site Coulomb repulsion, μ the chemical potential, and V_{ij} the long-ranged Coulomb interaction, which is characteristic for QCA systems. For simplicity the Coulomb term is chosen to be $V_{ij} = \frac{1}{r_{ij}}$ where r_{ij} is the distance between the two dots i and j. We also introduce the *compensation charge* q which is thought to represent a possible positive ion at each quantum dot site. This constant positive charge allows us to tune the net cell charge. For two electrons per cell, for example, $q = 0$ yields a net cell charge of $-2e$ whereas $q = \frac{1}{2}$ represents zero net cell charge, and here the cell becomes a perfect electrostatic quadrupole.

The geometric layout of the QCA system and therefore its functionality is encoded in the hopping parameter t_{ij} and the long-ranged Coulomb term V_{ij}. For the hopping parameter we usually only consider nearest-neighbour hopping and specifically no hopping between the cells. While this constraint is not strictly necessary for QCA, it is in line with the approach's underlying idea and greatly simplifies calculations. Because the overlap integral decays exponentially with distance, as long as the distance between dots from different cells is larger then the distance between dots within one cell, the assumption will introduce only a small error. Still, this is something to keep in mind if we place cells very close to each other. Note that without inter-cell hopping we can decompose the Hamiltonian into purely Coulombic cell-cell interaction terms H_{kl} and single cell terms H_k which capture the kinetics as well as the inside-cell Coulomb interactions,

$$H = -\sum_{k} H_k + \sum_{k<l} H_{kl}\,, \tag{2.2}$$

where k and l number the cells.

To parameterize the Coulomb term V_{kl} and specifically r_{ij}, the distance between quantum-dots i and j, we introduce the cell edge length a and the cell-cell distance d, as illustrated in Fig. TODO where we have used a short line of cells as an example QCA system. The angle between adjacent cells is denoted by θ. Ideally each cell should be in logic state 0 or logic state 1, but, of course, in practice a cell can be in any superposition of the two states or even in a different state altogether. The *cell polarization* P_k quantifies the state of the cell,

$$P_k = \frac{1}{2}\left(n_{4k+2} + n_{4k+4} - n_{4k-1} + n_{4k-3}\right)\ , \tag{2.3}$$

where the dots in each cell are numbered clockwise as indicated in the figure. The cell polarization is $P_k = -1$ for a logic 0 and $P_k = +1$ for a logic 1 state. Without any external

input the polarization of a cell will be $P_k = 0$. In the example line of cells, the input is set via the driver cell's polarization P_D at the left end. The driver cell's four static point charges are adjusted to reflect the desired polarization P_D. For QCA, the cell polarization really is the observable of utmost interest. It indicates whether a cell is in logic state 0 or logic state 1 and how polarized the cell is, where ideally, of course, the cell should always be fully polarized $|P_k| = 1$. In short, the cell polarizations will indicate how well the QCA approach works for a given system and, unsurprisingly, calculating cell polarizations for various geometric layouts over a wide range of system parameters will be our main focus.

Chapter 3

Approximations

Bibliography

[1] C. S. Lent, P. D. Tougaw, W. Porod, and G. H. Bernstein, "Quantum cellular automata," *Nanotechnology* **4** (1993) 49.