The Development of Theoretical Quantum-Mechanical Models to Advance Theoretical and Experimental Research in Clocked Molecular Quantum-dot Cellular Automata

Enrique Pacis Blair

United States Naval Academy

(Dated: December 20, 2009)

Abstract

Quantum-dot cellular automata (QCA) is a novel, current-free approach to computing at the nanoscale, which, if realized in computing devices, will have broad-reaching effects in the electronics industry. First, the concept of QCA is outlined. Next, personal research topics are proposed, to include the development of theoretical quantum-dynamic models of switching QCA cells; the development of a robust theoretical model to describe the interaction between a single-electron transistor (SET) and a QCA molecule when the SET is used as an electrometer for the measurement of intracellular charge transport; an examination of Landauer's Principle within the framework of the QCA paradigm; the development of a theoretical model for scanning tunneling microscope imaging of QCA molecules; and the development of simpler theoretical models for more complex calculations of the behaviors of candidate QCA molecules. Finally potential ramifications of realizing computing devices based on QCA are described.

Keywords: Quantum-dot Cellular Automata, QCA, clocked molecular QCA, clocking wires, power dissipation, Landauer's Principle, single-electron transistor electrometer, scanning tunneling microscope imaging, nuclear vibronics, fundamental thermodynamics of computing and power dissipation

I. INTRODUCTION

The transistor, functioning as a current switch, is the basic element in modern computing. It is arguably the most significant technological invention of the twentieth century. The transistor made electronics cheaper and more portable. The downscaling of the transistor has made computational electronics smaller, faster, more powerful, and more affordable than ever before, and it has done so at a consistent rate. Modern computing technology is everywhere, and it is foundational to research and progress in countless other fields of knowledge. Portable electronics have also become an irreplaceable part of our way of life. Phones and computers grow ever smaller, cheaper, and more powerful, and we are now accustomed to having access to unprecedented amounts of knowledge in a few short keystrokes.

Much of what we know and how we live can be attributed to the transistor and our ability to shrink it, but we are reaching fundamental limits to the miniaturization of the transistor. The current flow essential to transistor operation can be scaled only so far before the quantization of charge becomes evident and a steady flow of electrons is recognizable as motion of individual electrons. Also, at the nano-scale, quantum-mechanical effects such as tunneling stymie device operation as we know it. Perhaps the most daunting limitation is the resistive heating problem. As transistors shrink, so must the conductive interconnections between devices. But narrower interconnections imply higher electrical resistances and will generate massive amounts of heat. A chip covered in transistors shrunk to the molecular would melt due to an incredible release of heat [1]. If we are to achieve computing at the molecular scale, a new technology must be developed. In this proposal, I briefly describe quantum-dot cellular automata (QCA) as a promising paradigm for computing at the nanoscale; I describe my research interests in the field of molecular QCA; and I describe some of the far-reaching effects of realizing large-scale computing devices based on molecular QCA.

II. QCA

QCA is a novel current-free paradigm for computing [2, 3] that is naturally suited to computing at the molecular scale; takes advantage of quantum-mechanical effects and the quantization of charge [4]; being independent of current flow, is free of the resistive heating problem; and promises room-temperature operation [5–7].

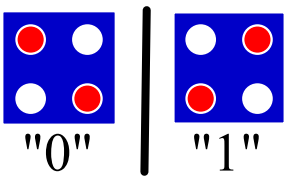

FIG. 1: The two states of a four-dot QCA cell with two mobile charges and four dots. A quantum dot is represented by a white disc, and the red disc within a white disc represents a mobile charge occupying a dot.

The QCA paradigm is built on the use of charge-containing structures known as QCA cells. Cells have multiple sites where mobile charge tends to localize known as "quantum dots." With four dots laid out in a square geometry, and with two excess mobile charges per cell, an individual cell has two degenerate states which can be used to encode a single single binary digit (bit), as depicted in Figure 1.

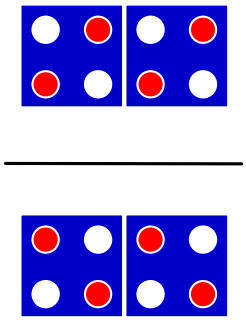

FIG. 2: Broadside coupling causes cells to align.

Neighboring QCA cells interact via simple Coulomb interactions as shown in Figure 2 . QCA cells may be laid out to form circuits as in Figure 3, and circuits may be combined hierarchically for general-purpose computing. Clocking of QCA cells and devices may be achieved by varying the relative energy levels of the charge configurations of individual cells. Clocking enables large circuits [8], power gain for the restoration of weakened signals, and allows quasi-adiabatic switching which minimizes power dissipation [9, 10].

Though device operation has been demonstrated under cryogenic conditions in materials systems [9, 11–34], molecular QCA offer significant advantages over metal-dot and semiconductor-dot QCA. First, molecular QCA offer synthetic regularity between devices based on the lengths of atomic bonds. Additionally, they offer device scaling to the molecular limit, with accompanying ultra-high device densities. Room-temperature molecular QCA operation has also been theorized [35] and demonstrated [36].

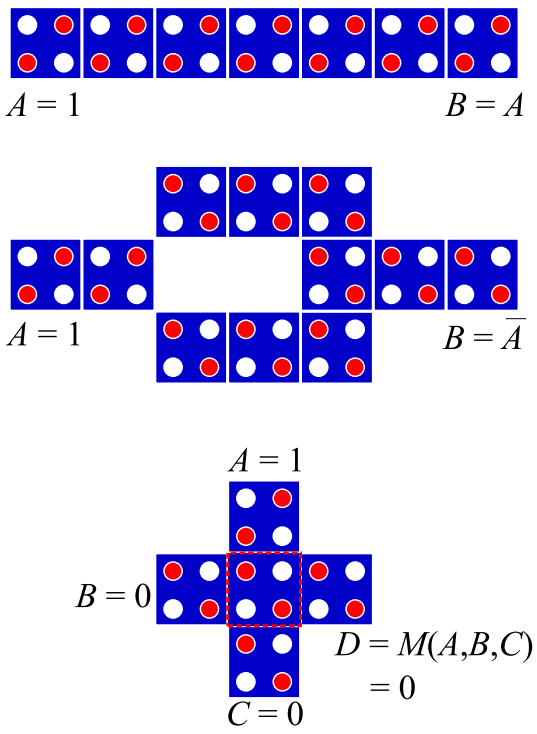

FIG. 3: Basic QCA devices: binary wire (top); inverter (middle); majority gate (bottom). The majority gate has three inputs (A, B, and C). The inputs "vote" at the device cell (in the red dashed box), and the output D is copied from the device cell. Here, two input zeros dominate a single one, and the output is zero.

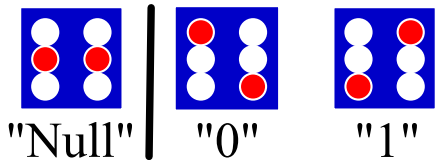

FIG. 4: Three states of a six-dot cell.

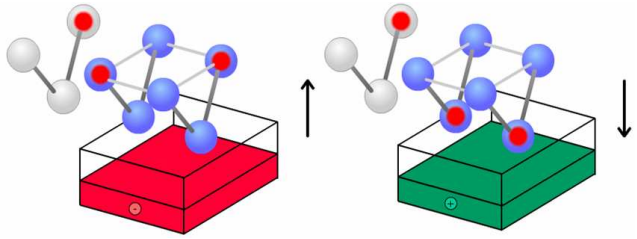

FIG. 5: A six-dot cell can be clocked using a conductor buried below the substrate. When positively charged (right, conductor shown in green) the conductor creates an electric field that draws the mobile charge (in this case, electrons shown as red dots) down to the null dots; when negatively charged (left, conductor colored red), the conductor's electric field drives the mobile electrons up and the cell takes an active state.

Clocked molecular QCA are achieved by the introduction of two additional null dots, yielding an additional state, called the "null state" which conveys no information (depicted in Figure 4). If the additional middle dots, called "null dots," are in a plane below that of the other dots (called "active dots") as in Figure 5, then such a cell can be driven to or from the null state using an electric field generated by a charged conductor buried in the substrate beneath the molecular devices.

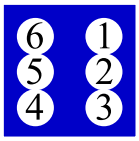

FIG. 6: A six-dot cell with dots numbered for reference.

The state of a six-dot cell can be described by two quantities, known as polarization P and activation A. P and A can be expressed as functions of the charge on the dots of a QCA cell. With dots numbered as in Figure 6, P and A are:

$$P = \frac{q_1 + q_4 - (q_3 + q_6)}{\sum_{i=1}^{6} q_i} \tag{1}$$

$$A = 1 - \frac{q_2 + q_5}{\sum_{i=1}^{6} q_i} \tag{2}$$

where q_i is the charge occupying the i^{th} dot. P and A originate in a three-state quantum-mechanical approximation of a six-dot QCA cell as described in [37].

Computational arrays of molecules can be clocked using arrays of wires charged using a multi-phase, time-varying voltage [38] as shown in Figure 7. The multi-phase nature of the clocking signal will cause cells in certain regions of the array, known as "active domains" to take an active state; other regions, called "null domains," will contain cells driven to the null state. The time-varying nature of the multi-phase signal will cause motion of the active domains about the device plane (Figure 8), and moving active domains will push bits through wires to logic gates where computation occurs (Figure 9) [39].

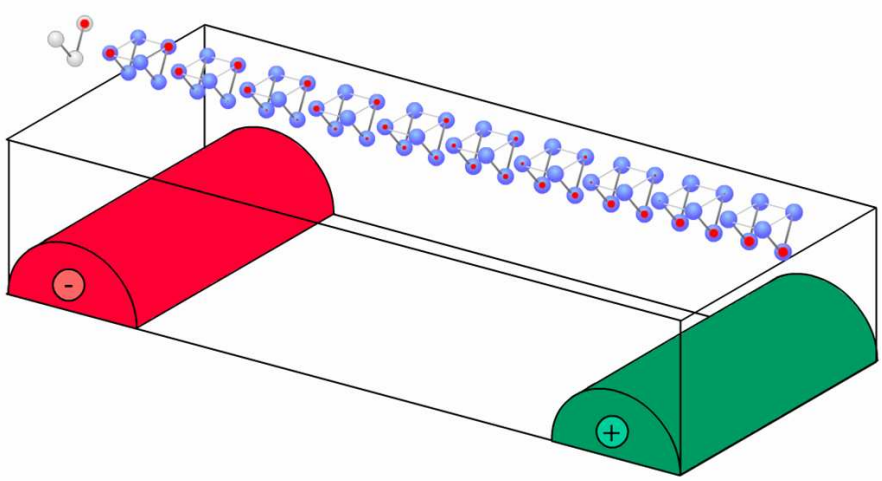

FIG. 7: Charged clocking wires activate some molecular QCA cells on the substrate while driving other cells to the null state, thus forming active and null domains.

III. RESEARCH INTERESTS

Here I outline future research interests, link those interests to the foundation I developed from previous research, and describe the research process.

A. Future Research Interests

I intend to further develop our theoretical of understanding QCA by modeling and simulating individual clocked molecular QCA and QCA circuits. I would need to take additional advanced courses in quantum mechanics to gain the proper understanding of the phenomena I intend to model. My primary research requirements are computing tools and software, and research of this type is well-suited to my proclivity for computer programming.

I intend to advance the theoretical understanding of the use of single electron transistor (SET) electrometers to measure intramolecular charge transfer. Experimental work in this area already has begun, but a detailed theoretical model of the probe fields and the sensitivity of the capacitively-coupled detectors to molecular switching events would be extremely useful. I intend to use finite element modeling software suites such as COMSOL to enable these calculations.

Additionally, the QCA paradigm presents an excellent framework in which I can explore

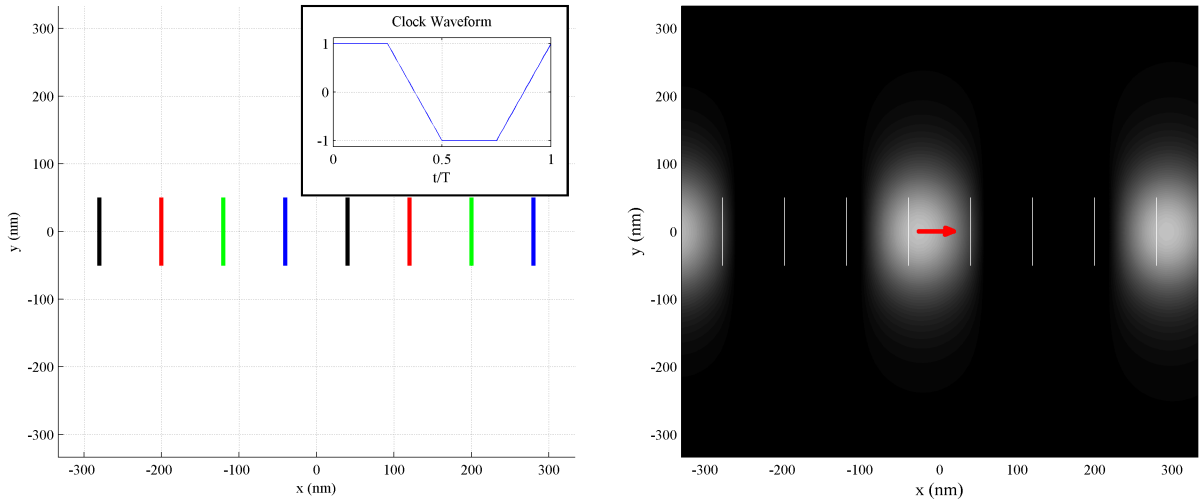

FIG. 8: An plan view of an array of charged wires is shown in (a). Wires are represented as colored lines, and the color of each represents an individual phase of the multi-phase clock voltage. The inset to (a) shows one phase of the clock voltage. A snapshot of the resulting electric field (z-component [out of the page] only) is shown in (b). Light regions represent active domains, and dark regions represent null domains. The position of the wires in (b) is indicated using white lines. The time-varying nature of the clock drives active domains rightward, as indicated by the red arrow.

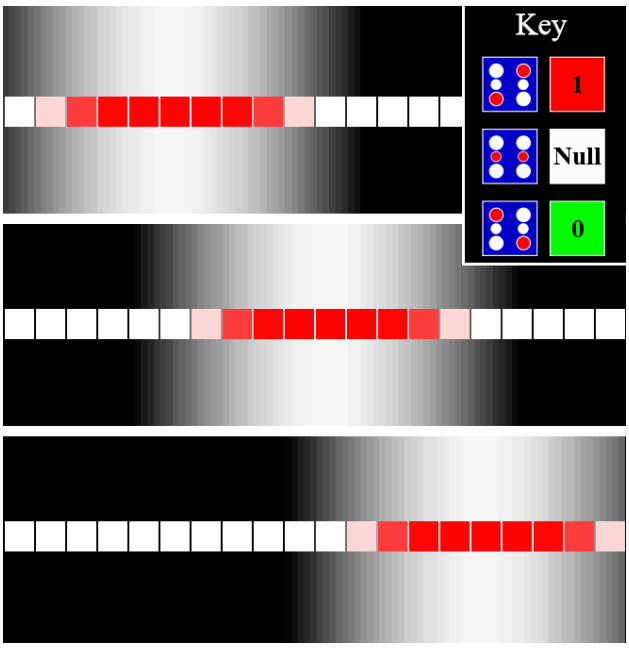

FIG. 9: A moving active domain pushes a bit packet (in this case, a "1") rightward through a binary wire. The color of the background represents the vertical component of the electrical field. The white region is the moving active domain, and the black region is the null domain. Three snapshots in time are shown arranged early to late, from top to bottom.

the fundamental concepts of the thermodynamics of computation. Landauer's principle connects heat dissipation to bit erasure [40], but it does not go unchallenged. A natural question to ask is whether this principle even is correct. If so, I intend to explore how this principle concretely manifests itself in QCA.

In the modeling of single QCA molecules, I intend to explore the relationship between intracellular charge transport and nuclear vibronic motion, as this is the means by which the switching of a QCA cell dissipates heat. I also seek to model circuit-level dynamics of molecular assemblies. Such a model must include phenomenological coupling to the substrate as a heat bath. It will then allow us to "see" energy build-up in a switching molecule and dissipation via coupling to the substrate.

Additional research could have me delve into quantum chemistry (QC) so that I can model the behavior of specific QCA molecular candidates. QC programs such as Gaussian and NWChem can help to calculate the polarization of molecular QCA cells when driven by the electric field from neighboring molecules. QC calculations can also be used to extract only the most relevant Hamiltonian matrix elements, thus allowing me to create simpler Hamiltonian models which can be used to calculate time-independent behavior and open-system thermodynamic behaviors in QCA circuitry. Finally, another project I can undertake will bridge theory and experiment. Much basic work on molecular QCA will be driven by scanning tunneling microscope (STM) imagery of a switching molecule. A solid model of the STM imaging process using the projected local density of states could prove highly useful in guiding the interpretation of experimentally-obtained STM images of switching molecules.

B. Linking Future and Past Research Interests

Past personal research included the simulation and design of clocking wire arrays and arrays of clocked molecular QCA circuits; calculating power dissipation in clocking wire arrays; and calculating the energetics of forming bits in QCA circuits. My future research plans are to further develop our understanding of QCA devices through theoretical and simulation-based research. This means that I will be using the latest models developed by my colleagues and refining them. My future research plans are well-founded upon previous experience I have in building, testing, and verifying models and using those models to make predictions. Additionally, these interests are a natural progression from the theoretical work

I came to enjoy as a senior undergraduate student and continued as a master's student under Dr. Lent.

C. The Research Process

Developing theoretical models is a process which will require me to study a previously designed and validated model and either enhance it to account for additional physical phenomenon or simplify it to ignore processes previously determined to be insignificant. Validation of my model will require testing it to verify that in the limit where the newly added phenomenon is zeroed, my model matches the previously tested, simpler model. In the case that my model simplifies a more complex calculation, the original model should match mine when the effect that was removed is zeroed. Other deviations in my adjusted model from the original should be explained, and new insights gleaned from a newer model are to be written and submitted for publishing so as to advance the body of scientific knowledge in our field. In the case of a model which seeks to explain previous experimental results, the model should be tested by examining it at it limits and by verifying that it accurately predicts experimental results.

The goal of my theoretical research is to develop theory that experimentalists can explore, validate, and build upon, with the ultimate aim of realizing practical computing devices based on molecular QCA.

IV. THE POTENTIAL IMPACT OF QCA

The National Science Foundation Graduate Research Fellowship Program will be a significant enabler in my studies. Study in the field of molecular QCA is an endeavor having both intellectual merit and a broader impact on our nation with the potential for global effects.

A. Intellectual Merit

My development of the theory within the broader field of QCA research will refine and develop current theories on the mechanics of power dissipation in QCA molecules and cir-

cuits. My study will strive for simpler, more tractable models derived from studies of more complex QC calculations. Landauer's principle will be put to the test, and if possible, I will seek to discover its application in QCA. Finally, I seek to provide a bridge between theory and experiment to aid experimentalists in understanding STM images of molecular QCA switches. All this is with the aim of realizing QCA-based computing devices.

B. Broader Impact

My studies in the field of molecular QCA have an educational focus, will lead to increased interdisciplinary collaboration, and, in leading to the realization of QCA-based computing devices, may having tremendous ramifications with global scope.

1. Educational Focus

Teaching and mentoring are my passion, so I am keenly excited that I will be mentored by Dr. Lent in teaching so that I may hone my pedagogical skills. I am particularly interested in taking part in and learning from his outreach programs which target first-year engineering students and high-school students, teaching them problem-solving skills through constructing MATLAB graphical user interface (GUI) tools. As an undergraduate, I recall honing my own problem-solving skills in Dr. Lent's class which required the construction of GUIs; furthermore, I have successfully applied problem-solving skills gained from those experiences in both my previous research and in teaching at the U.S. Naval Academy, and I am looking forward to passing that along to future generations of students.

2. Interdisciplinary Collaboration

Although the preponderance of my graduate work is theoretical and focused on molecular electronic devices, in later stages and beyond, it will lead to more opportunities for interdisciplinary collaboration - specifically, between both theorists and experimentalists in chemistry, physics, electrical engineering, and computer engineering. Eventual interdisciplinary collaboration, then, will allow me to hone my skills and teach in such a way that is more interdisciplinary and holistic.

3. Global industrial and economic impact

The development of the transistor in California made Silicon Valley a nexus of research, development, and education in semiconductor electronics. Similarly, a realization of QCA-based computers as a replacement technology for transistors at the University of Notre Dame in Indiana could make the midwestern states a hotbed for research, development, and education in molecular electronics. This research has the potential to create a whole new industry triggering an avalanche of technology transfer, thereby creating countless further opportunities for employment and research. Increased fervor in research mean more opportunities for collaboration between academic institutions, research institutions, and the U.S. Government. Already, the Notre Dame QCA group has collaborated with several other U.S. institutes, including Ball State University and the U.S. Naval Academy, and funding has come from the Office of Naval Research and NASA's Jet Propulsion Laboratory. Furthermore, if a new molecular electronics industry is to emerge from this research, it is in our nations best interests for that industry to emerge within the United States and for us to maintain leadership in molecular electronics by being at the forefront of continued research and development. This will ensure that the U.S. is the recipient and beneficiary of research and development funds from around the globe; and students will come from around the world to study molecular electronics in the United States.

---

[1] C. Lent, M. Liu, and Y. Lu, Nanotechnology **17**, 4240 (2006).

[2] C. Lent, P. Tougaw, and W. Porod, Appl. Phys. Lett. **62**, 714 (1993).

[3] C. Lent, P. Tougaw, W. Porod, and G. Bernstein, Nanotechnology **4**, 49 (1993).

[4] C. Lent and P. Tougaw, A device architecture for computing with quantum dots, in Proc. IEEE, pages 541–557, IEEE, 1997.

[5] M. Lieberman et al., Ann. N.Y. Acad. Sci. **960**, 225 (2002).

[6] C. Lent, B. Isaksen, and M. Lieberman, J. Am. Chem. Soc. **125**, 1056 (2003).

[7] C. Lent and B. Isaksen, IEEE T. Electron Dev. **50**, 1890 (2003).

[8] M. Niemier and P. M. Kogge, Logic-in-wire: Using quantum dots to implement a microprocessor, in Int. Conf. Elec., Circ. Sys., 1999.

[9] A. Orlov et al., Appl. Phys. Lett. **81**, 1332 (2002).

[10] J. Timler and C. Lent, J. Appl. Phys. **91**, 823 (2002).

[11] A. Orlov, I. Amlani, G. Bernstein, C. Lent, and G. Snider, Science **277**, 928 (1997).

[12] I. Amlani, A. Orlov, G. Snider, C. Lent, and G. Bernstein, Appl. Phys. Lett. **72**, 2719 (1998).

[13] G. Snider et al., J. Vac. Sci. Technol. A **17**, 1394 (1999).

[14] G. Snider et al., Microelectron. Eng. **47**, 261 (1999).

[15] G. Snider et al., J. Appl. Phys. **85**, 4283 (1999).

[16] G. Snider et al., Jpn. J. Appl. Phys. **38**, 7227 (1999).

[17] I. Amlani et al., Science **284**, 289 (1999).

[18] I. Amlani et al., Appl. Phys. Lett. **74**, 2875 (1999).

[19] R. Kummamuru et al., A quantum-dot cellularu atuomata shift register, in Dev. Res. Conf., pages 103–104, 2001.

[20] A. Orlov et al., J. Nanosci. Nanotechno. **2**, 351 (2002).

[21] G. Snider et al., Quantum-dot cellular automata: introduction and experimental overview, in Proc. IEEE-NANO, pages 465–470, IEEE, 2001.

[22] A. Orlov et al., Appl. Phys. Lett. **78**, 1625 (2001).

[23] I. Amlani et al., Appl. Phys. Lett. **77**, 738 (2000).

[24] A. Orlov et al., Clocked quantum-dot cellular automata devices: experimental studies, in Proc. IEEE-NANO, IEEE, 2001.

[25] A. Orlov et al., Clocked quantum-dot cellular automata devices, in Proc. SPIE, pages 441–444, SPIE, 2002.

[26] A. Orlov et al., Surf. Sci. **532**, 1193 (2003).

[27] R. Kummamuru et al., Experimental demonstration of a qca shift register and analysis of errors, in Int. Electron Dev. Meeting, pages 95–98, 2002.

[28] A. Orlov et al., Appl. Phys. Lett. **77**, 295 (2000).

[29] I. Amlani et al., Superlattice Microst. **25** (1999).

[30] G. Snider et al., Semicond. Sci. Tech. **13**, A130 (1998).

[31] G. Snider et al., Experimental progress in quantum-dot cellular automata, in Proc. SPIE, 2002.

[32] A. Orlov et al., Experimental studies of clocked quantum-dot cellular automata devices, in 58th Dev. Res. Conf., pages 157–158, 2000.

[33] K. Yadavalli, A. Orlov, J. Timler, C. Lent, and G. Snider, Nanotechnology **18** (2007).

[34] G. Bernstein, I. Amlani, A. Orlov, C. Lent, and G. Snider, Nanotechnology **10**, 166 (1999).

[35] C. Lent, Science **288**, 1597 (2000).

[36] M. Haider et al., Phys. Rev. Lett. **102** (2009).

[37] J. Timler and C. Lent, J. Appl. Phys. **94**, 1050 (2003).

[38] K. Hennessy and C. Lent, J. Vac. Sci. Technol **19**, 1752 (2001).

[39] E. Blair and C. Lent, An architecture for molecular computing using quantum-dot cellular automata, in IEEE C Nanotechnol., pages 402–405, IEEE, 2003.

[40] R. Landauer, IBM J. Res. Dev. **5**, 183 (1961).