Binary Atomic Silicon Logic

Taleana Huff1,3,*, Hatem Labidi1,2, Mohammad Rashidi1, Roshan Acha1,3, Lucian Livadaru3, Thomas Dienel1, Jason Pitters2,3, and Robert A. Wolkow1,2,3,*
1Department of Physics, University of Alberta, Edmonton, Alberta, T6G 2J1, Canada
2National Institute for Nanotechnology, National Research Council of Canada, Edmonton, Alberta, T6G 2M9, Canada
3Quantum Silicon, Inc., Edmonton, Alberta, T6G 2M9, Canada
*email: taleana@walberta.ca; rwolkow@walberta.ca
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The ultimate in miniature circuitry, exactlying crafted of single atom building blocks, can unleash a new basis for electronic devices one that is far more energy efficient, while at once much faster and more compact than todays state of the art. A most enticing goal. While examples of atom-crafting with a suitable level of precision and complexity has existed for 3 decades, that control did not extend to electronically useful and also strongly bonded material systems. The attractive idea of forming circuitry of very robust patterned hydrogen atom terminated silicon surface states had been expressed decades ago, but, sufficient understanding of single atom states, ensemble states, and the interaction of those with dopants and bulk states was lacking and has only very recently been established. That new understanding coupled with greatly improved atomic scale fabrication methods developed just months ago have enabled a great step forward. Here, we report the first example of reversible information transmission through a binary wire a two state wire made of atomic silicon quantum dots. A binary OR gate and its full truth table has also been demonstrated. Because electrons are merely re-arranged and are conserved, and as no conventional current is used, minuscule power consumption is expected. Signal transfer is mediated by electrons confined by and tunneling among constituent dots. The tunneling rate in this strongly coupled field controlled system is of order \(10^{14}\) s\(^{-1}\), indicating the potential for clocking in the THz regime. An atomic force microscope operating in the non-contact mode was deployed to fabricate these active structures. A variant of that technique, atom resolved Kelvin probe force spectroscopy, records individual atom charge state changes to reveal and verify the inner workings of this approach. Spontaneous spatial charge arrangements in ensembles occur in response to imposed charge inputs, allowing information representation and computation. The recently developed atomic white-out editing technique allowed structure and function alteration and changeable inputs. This approach to electronics we call BASIL for Binary Atomic Silicon Logic.

INTRODUCTION

The promise of atom scale computing first became a possibility when Eigler et al. controllably moved atoms on a surface to achieve structures of their design [1]. In a subsequent work, the same lab made molecular cascades where, in analogy to falling dominoes, a terminal molecule was tipped to, in turn, tip over a neighboring molecule, which tipped the next molecule, and so on [2]. Separate branches of the cascade were delicately timed to come together in such a way as to achieve binary logic functions. With these results a new era was begun, however, challenges preventing practical applications remained and those limitations have been very difficult to overcome. Some of those challenges are; 1) the need to have the patterned atoms be robust at practical operation temperatures ideally room temperature. The initial atomic patterns were very delicately bound and would not persist above about -230°C [1,2]. In general, atom fabrication of structures robust enough to withstand relatively high operating temperature are more difficult to make. This is because larger energy inputs from the scanned probe are required to dislodge and move strongly bound atoms, and under such conditions covalent bonds within the probe itself break with some probability comparable to that of the target bond [3]. 2) The patterned atoms need to be electrically distinct from the substrate so as to enable conduction pathways that are not shorted-out or altered by the substrate. Studies performed on metals [4,5], the most common choice, were therefore limited in that regard. Isolation has been achieved in studies of metal atoms and of molecules separated from a metal substrate by a salt layer, but these have their limitations in uniformity of layer thickness and issues with spontaneous loss of charge to the substrate [6,7]. 3) The atomic circuitry must not require mechanical or other reset processes (analogous to standing all the dominoes back up) that would prevent the circuitry being instantly reusable.

The above are necessary, but far from sufficient qualities of a working atomic scale circuitry. Ideally, and to justify the substantial retooling required, a new circuitry should be more than compact. In order to provide a path beyond the established semiconductor roadmap, it must address the particular limitations of todays dominant, and very nearly mature CMOS technology; the primary problem being excessive power density which causes devices to become hot.

By putting aside transistor-based logic that sinks a burst of electrons to ground with every cycle of the clock, to considering field controlled computing schemes such as quantum dot cellular automata [8], it is possible, in principle, to expend only the entropic and electrostatic energy associated with rearranging, but not disposing of electrons. As the load of such circuits present to the clock is essentially reactive, there is the opportunity for very low power and fast resonant clocking also.

These types of low power field controlled binary logic elements formed of quantum dot ensembles have been hypothe-
sized, and even rudimentarily realized, for over a decade now \cite{8-11}. Single electrons, or their absence, serve as 1 and 0 inputs and, likewise, the logic state at the output is represented by a localized charge, the value of which can be read and transmitted by a two-state wire to serve as inputs to subsequent logic elements. Clocking circuitry provides gain by restoring such transmitted signals \cite{8}. It is necessary that electrons exhibit ground state behavior for such quantum dot circuitries to operate. If, as typical, the quantum dots are of a size that requires cryogenic conditions to ensure dominant ground state electronic behavior, circuitry built of such dots would also need to operate at cryogenic conditions; a limiting condition to reduce power consumption for many applications.

In this work we describe a path forward that addresses these various concerns and is based upon the atomic silicon quantum dot (ASiQD) \cite{12}. An ASiQD is created by removing a single hydrogen atom from a H-terminated silicon surface \cite{13}. ASiQDs can be occupied by 0, 1 or 2 electrons depending on dopant type and concentration, local electric fields as well as proximity to other ASiQDs \cite{14}. Crucially too, ASiQDs electronic states exist in the bulk silicon band gap allowing isolation from or spatially and temporally controlled connection to the bulk substrate \cite{15, 16} and eliminating the need for a distinct insulating layer. Diverse ASiQD ensembles can embody passive and active elements for binary, analog and quantum circuitry.

In addition to being small and therefore having relatively large intra-dot energy level spacing, the ASiQDs can be placed very close (within 0.4 nm) allowing large interaction energies, of order 100 meV \cite{17, 18}. ASiQD-based circuitry has a bit energy of this order. This value, somewhat larger than $k_B T$ at room temperature, is desirable as information is held just sturdy enough to withstand environmental noise, while the barrier to purposeful manipulation of data is not so large, as in CMOS today, that excessive energy must be expended to achieve computation, causing inadvertent device heating. The tunnel rate between coupled ASiQDs is of order 10 THz \cite{17} a large increase over more conventional coupled dots where a rate in GHz range is typical \cite{19}. Tunnel period relates to information transmission speed along lines of field-coupled bi-stable tunneling oscillators.

It has been shown that ASiQDs can be lithographically patterned with the sharp tip of a scanning probe microscope tip \cite{20-21}, but placement errors of 5 to 50% are observed, depending greatly on tip character, among other parameters. Most recently a powerful editing process for ASiQDs has been described allowing perfect patterns to be created \cite{22}. As the dots contain only one atom, dispersion of dot size is zero, leading to homogeneity of circuit properties. Moreover, because the dots are naturally restricted to silicon crystalline lattice positions, inhomogeneity related to inter dot spacing is also eliminated.

In this work scanning tunneling microscopy (STM), and non-contact atomic force microscopy (NC-AFM), are used to fabricate and analyze ASiQD-based structures. NC-AFM can discriminate among and can controllably alter charge states of adatoms and molecules \cite{7, 23}. Kelvin probe force microscopy, a variant of NC-AFM, can detect the energy at which charge state transitions occur on single atoms \cite{7, 24}. With these tools it is shown that double, and more generally, multi-well electron confinement potentials can be made of controllably placed ASiQDs. Tunneling among select sites is enabled and as a result spatial arrangements of electrons are automatically adjusted in response to applied fields. In this way 2-state information is rapidly transmitted along a binary wire. As no conventional current is required there is no substantial consumption of energy. Finally, to illustrate the potential of these concepts to realize extremely small, fast and low energy consuming binary logic, an OR gate is demonstrated.

**RESULTS AND DISCUSSIONS**

Earlier experimental and theoretical work established that on a highly doped n-type crystal, isolated silicon surface states spontaneously accept an addition electron for a total of 2 electrons and a net charge of -1 \cite{14, 16, 20, 25, 26}. Hence, we expect an ASiQD to be negatively charged in STM and NC-AFM recorded at zero bias. It is also understood that upon applying a negative sample bias the ASiQD becomes neutral before again becoming negative at larger negative bias (of order -1 V) as the depletion barrier separating bulk and surface is overcome allowing the conduction band electrons to fill the ASiQD \cite{20}. Adjustments dependent upon dopant concentration and the dynamics particular to each variation have been studied \cite{15, 20, 27}.

The transition of the ASiQD from a negatively charge state to zero charge at small negative bias has been inferred, but never seen as this effect is masked by the Si bandgap. No current is available in that range to generate a STM image or current-voltage spectrum. In Figure 1 we show the first direct experimental evidence of this transition in the form of an AFM force vs voltage spectrum.

Figure 1-a shows a constant-current STM image of an ASiQD created by tip-induced desorption of a single hydrogen atom from the H-Si(100) surface. At relatively high negative sample bias (e.g. -1.7 V), the negatively charged ASiQD appears in filled state images as a bright protrusion surrounded by a characteristic small dark halo \cite{15, 20}. In the corresponding frequency shift map at 0 V (Figure 1-b), hydrogen capped silicon atoms appear as bright protrusions arranged in the 2 1 surface reconstruction pattern and the ASiQD shows up as a dark feature in the otherwise bright landscape indicating a larger attractive tip-sample interaction \cite{22}.

Figure 1-c shows current vs. sample bias (I(V)) spectroscopy curves taken above the ASiQD and H-Si surface, with both the surface and ASiQD showing zero current bandgap from around -0.8 to 0.2 V. A detailed description of the observed features can be found in prior studies \cite{15, 20, 27}. In the same plot, the frequency shift vs. sample bias ($\Delta f$ V) or KPFM spectra measured above the ASiQD with a bias sweep range of -0.6 to 0 V is also plotted. Note,
this range is in the bandgap of the material where no STM information would be available. A pronounced step is seen at -250 mV. Prior work examining charged species [6,24], electron transfer between molecules [27], and charge states in quantum dots [28] with NC-AFM has shown this type of step feature to correspond to a dynamic single-electron charge state change. Based on previous works, the step seen in Figure 1-d can be assigned to the charge state transition of the ASiQD from a negative (doubly occupied) to neutral (singly occupied) charge state, respectively right and left of the step in the ∆f (V) curve at -250 mV.

We now examine structures constructed from multiple ASiQDs. When 2 ASiQDs are closely spaced, within about 1 nm or less, Coulombic repulsion causes one of the pair’s extra electrons to delocalize in the conduction band [18,25]. As a result, the double well potential formed of the pair of ASiQDs has a filled symmetric state and half-filled antisymmetric state. The low (∼0.5 eV) and narrow (∼1 nm) barrier enables facile electron tunneling. Figure 2 demonstrates the step-by-step fabrication and characterization of such pairs and in addition shows the preparation of the two possible polarization states of the pair under the influence of a negative charge.

In Figure 2(a), (b) and (c), the STM, constant height AFM, and KPFM spectra respectively are shown for an isolated ASiQD that will become the left member of a pair. To ensure an unbiased view of the right atom, the left ASiQD was erased (capped with a H atom) before creating and characterizing the right atom as shown in (d), (e) and (f). Filled state STM images in (a) and (d) show each isolated ASiQD as a bright white protrusion. Corresponding AFM images in (b) and (e) show, as in Figure 1, a pronounced attractive feature in the form of a dark depression at the ASiQD location. Potential energy sketches of ASiQD wells are included at the bottom of each AFM image. Also as in Figure 1, the KPFM spectra of the isolated ASiQDs, shown in (c) and (f) show sharp steps indicating a charge transition occurring at -60 mV.

The left ASiQD was then recreated to form a pair, with exactly 2 lattice sites separation, which is 0.768 nm, as shown in (g), (h) and (i). Between these tunnel coupled dots there is one intervening surface bound H atom as is evident in the AFM image in (h). The KPFM spectra in (i) of the left and right dots are nearly identical. The higher energy feature, seen in both spectra in (i) is likely due to the doubly negative paired ASiQD species a spectral feature which is not expected and was not observed for the isolated dots.

Figures 2 (j) and (k) show the addition of a third ASiQD, marked with a orange triangle. There are 4 intervening H atoms between the closest of the paired dots and the teal labeled ASiQD. The teal labeled ASiQD is not significantly tunnel coupled to the pair as the tunneling interaction falls off exponentially, nor Coulombically destabilized enough to lose its negative charge. It therefore can be deployed as a negative electrostatic perturber of the pair.

While the STM image in (j) primarily reveals the position of the atoms, the AFM image, (k), taken at a relatively large separation so as to make electrostatic forces dominate the tip-sample interaction, provides a spatial map of the polarizable electron in the double well.

In (k), a stark contrast is seen among the same dots which appeared indistinguishable in Figures 2 (a) through (i). The middle ASiQD (teal) is significantly lighter meaning less negative than the leftmost dot (blue). On examining the KPFM curves for this "2+1" experiment in (l), it is seen that the KPFM transition energies for the perturbing ASiQD (orange) and for the ASiQD furthest left (blue) overlap and occur around -30 mV. However, the KPFM trace of the middle dot (teal) shows a distinctly shifted charge transition energy near +400 mV.

This shift can be attributed to the repulsive electrostatic effect of the perturbing ASiQD. We see that a less negative voltage between probe and sample must be applied to record the negative to neutral charge state transition. Expressed otherwise, we see that the middle ASiQD at zero probe bias, but under the influence of the negative perturbing ASiQD, is already in the neutral state. It is evident that the tunnel coupled ASiQDs form a double well potential that can be polarized by an electric perturbation. In this example, the double well is tilted to the left causing the shared electron to tend to localize there.

For completeness, a fourth ASiQD is added in (m) to re-symmetrize the pair from (k). In (n), the central paired dots
FIG. 2. Polarization Of Coupled Atomic Silicon Quantum Dots. Filled state STM images of the isolated left (a), isolated right (d), coupled (g), polarized left (j), symmetric (m), and polarized right (p) ASiQD assemblies (-1.8 V and 50 pA). Corresponding AFM frequency shift maps for each case are shown in (b),(e),(h),(k),(n),and (q) respectively ($z_{rel}=-300$ p.m, Osc. Amplitude=50 p.m, 0 V). Potential energy well sketches are included at the bottom of each AFM map. Color coded $\Delta f$ (V) spectra taken over the dots in the frequency shift maps are shown in (c),(f),(i),(l),(o), and (r) ($z_{rel}=-300$ p.m, Oscillation Amplitude=50 p.m).

(blue and teal) are light colored and symmetric. Once again, the KPFM at 0 V in (o) gives a result consistent with the emerging model. In this case the entire double well is destabilized and as a result less negative charge, as recorded in these time averaged views, is seen to be resident there.

To show reversibility of polarization state, the right ASiQD is capped in (p). The frequency shift map of (q) now shows a mirror of the tilt of the pair from (k). The middle DB (blue) is light, and the right (teal) dark. KPFM in (r) now shows the transition energy for the (blue) dot has been shifted from -60 mV to +400 mV, and exists in a neutral state at 0 V.

This is our central result. From this we can build all manner of ensembles of diverse function. Information can be expressed as physical spatial charge maps and computation can be performed by allowing latched charges to affect and rearrange partially constrained charges. The fixed charges used here are stand-ins for soon to be available analog wires, again formed of ASiQDs, which interface to conventional much larger lithographic electrodes. We now provide two examples: binary wires and an OR gate.

Figure 3 shows reversible direction information transfer in a binary wire consisting of multiple paired ASiQDs. The main
In Figure 3(c), one ASiQD is added at the far right to create an additional coupled pair and eliminating the input or perturbation. The wire responds by displaying a self polarizing effect. A division in the middle of the line is apparent, and marked with a vertical dashed line. The multiple partially constrained charges come to an optimal arrangement by having pairs to the left of center polarize to the left while right side pairs polarize to the right. Upon seeing this result it becomes hard to avoid imagining myriad tiny computer elements.

In Figure 3(e) and (f), the same line of pairs is polarized from the opposite side with a 19th ASiQD added on the left. The wire responds by polarizing to the right, reversing what was shown initially in (a). These structures are instantly (a small multiple of the tunneling time) and always prepared to respond to inputs. It is notable that no reset step is required and that the scanned probe instrument is not a component of the device, only an observer. Also, beyond the few tenths of an eV required to impose an electrostatic input, little energy is consumed in the process. As no current is used there is no $I^2R$ heating. There will be a small entropic cost associated with rearranging electrons. In simple ground state seeking arrangements as shown here, energy balance requires a minuscule temperature change of the substrate. In this study we do not have access to positive charges and are therefore preventing from pulling in addition to pushing electrons. In a subsequent study bipolar operation will be exhibited we expect, allowing more flexibility in design.

We show one final adaption of the basic principal established in Figure 2. Figure 4 shows a prototype binary logical OR gate. The two topmost branches are inputs, and the lower branch an output. Figure 4(a) shows the central structure comprising the gate: 3 tunnel coupled pairs arranged in a Y. In the absence of electrostatic inputs and in analogy to what was seen for the line in Figure 3(c) and (d), the 2D assembly experiences electrostatic repulsion among the pairs, causing the partially constrained electrons to relax to the outer most points of the structure as indicated by the contrast in the AFM frequency shift map in Figure 4(b).

We note again here the limitation of this first embodiment that permits us only to "push" with the negative charge of a perturbing ASiQD, not "pull". In order to establish an opposite polarization state of the output ASiQD, we make an adaptation and fabricate a weak perturber at the terminus of the lower output branch as demonstrated in (d). This perturber acts as a weak spring”, polarizing the bottom-most pair upward, leaving the ASiQD just above the spring in a neutral state, which we define as the 0 state. This is shown experimentally in (e) and schematically in (f). Consistent with our output definition we use here a neutral state - the absence of an input - as binary 0. The first row of the truth table is established with two 0 inputs giving a 0 output.

When a negative input is in place at either the top left (g),
CONCLUSION

In this paper we have demonstrated the essential capabilities required to build all-silicon, atom-scale electronics for binary computation. Our primary building block is the atomic silicon quantum dot. Ensembles of ASiQDs enable flexible confinement of electrons - allowing electric field driven movement among two or more distinct positions. These capabilities enable information representation and field controlled computation.

Single electron induced switching of paired ASiQDs, and of a longer sequences of pairs, were shown to transmit information bi-directionally. To demonstrate the capacity to build units of higher function a binary OR gate was shown.

While we worked on a highly doped n-type substrate here, low doped and p-type doping together with local Fermi level controls will enable different charge states and modes of operation.

Parallel work, not shown here, will provide environmental protection through encapsulation and connection of the atomic circuitry to ordinary lithographic features, and to CMOS circuitry enabling operation in a normal packaged chip format. Hybrid BASIL-CMOS circuitry of lower power consumption and greater speed is targeted in the near term.

Though detailed studies of power requirements and speed of operation remain to be done, it is evident that the methods outlined here will be attractive in those regards. Because the gates and the binary wires between gates require only single electron-level electrostatic actuation, and because no conventional current is required, power consumption will be extremely low. As the tunnel rate among coupled atomic quantum dots is of the order of femtoseconds, signal transmission and gating action will be fast. THz operation rates are anticipated. The approach described here may enable a beyond
Moore technology combining enormous speed with ultra low power consumption.

While an OR gate and a binary line do not provide a binary basis to construct a general purpose computer, we expect the advancements yet required and desired, such as fanout, NOT and AND among other functions, are more likely to be extensions of the present work than barriers requiring entirely new device principles. The addition of clocking is likewise anticipated to be within reach. With clocking will effectively come gain and periodic restoration of signal a requirement to achieve complex functionality. Though not discussed here, it has not escaped our notice that analog and quantum circuitry can be built of the same fabric and plans in those directions are well advanced.

**METHODS**

Experiments were carried out using a commercial (Omicron) qPlus AFM system operating at 4.5 K. We used highly arsenic-doped (\(\sim 1.5 \times 10^{19} \text{ atom cm}^{-3}\)) silicon (100). Sample preparation involved degassing at \(\sim 600^\circ \text{C}\) for 12 hours in ultra-high-vacuum (UHV), followed by a series of resistive flash anneals reaching 1250°C. Secondary ion mass spectroscopy done in prior work has shown similar heat treatments create a surface regime 60 nm deep where the dopant concentration is reduced near the surface to 40 times less than that of the bulk [20][29]. While holding the Si substrate at 330°C for 2 minutes, molecular hydrogen at \(10^6\) Torr was cracker on a 1600°C tungsten filament above the sample creating the \(2 \times 1\) reconstructed hydrogen atom-terminated Si(100) surface.

qPlus AFM sensors [30] with a separate electrode for tunnel current were used. Focused ion beam (FIB) was used to cut a micro-tip from an electrochemically etched 50 \(\mu\)m polycrystalline tungsten wire, then weld it to the sensor. This fabrication technique ensured high sensitivity and well defined sensor mechanical properties [31].

After being loaded in UHV, tips first had the oxide layer removed by electron beam heating treatments, followed by field evaporation cleaning of the apex in a field ion microscope (FIM). Further sharpening was conducted using a FIM nitrogen etching process to obtain the smallest possible tip radius of curvature [31][32]. Final in-situ tip processing was done through creation of a bare silicon patch through tip induced hydrogen desorption, followed by gentle controlled contacts with the tip on the reactive patch. This procedure usually results in a stable and artifact-free tip [33].

To create an ASiQD, the tip is positioned on top of a surface hydrogen atom at 1.3 V and 50 pA, and pulses of 2.5 V for 10 ms are applied. Mechanically induced covalent bonding of a single H atom at the tip apex to a silicon ASiQD to passivate it was done by following the procedure described in ref [22].

Nanonis control electronics and software were used for both STM and AFM data acquisition. For all frequency shift maps, \(Z=0\) Å corresponds to the relative tip elevation defined by the STM imaging set points 50 pA and -1.8 V. To minimize drift during AFM image acquisition, the tip was left to settle for 12 hours after approach to allow piezo scanner stabilization. All STM and AFM images are raw data.

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