1. Introduction

Semiconductor-based quantum computing architectures have been extensively studied for decades because of their high potential to fulfill the five DiVincenzo criteria for realizing a scalable quantum computer. Notably, silicon-based approaches, both photonics-based as well as electron and hole spin-based, have been considered especially promising in the race to the quantum advantage and beyond, thanks to the strong scaling and integration capabilities enabled by the mature complementary metal–oxide–semiconductor (CMOS) industry. Furthermore, benefiting from isotopically enriched $^{28}\text{Si}$ substrates and advanced device architectures, long coherence times, high-speed operation, and high-fidelity universal one- and two-qubit logic gates have been successfully demonstrated in Si-based spin quantum bits (qubits).

In addition to the five DiVincenzo criteria, large-scale quantum chip integration requires that the quantum information can a) be transferred between the computing qubit and the transferring qubit, and b) be conserved during the transportation. To this end, some prospective strategies for semiconductor-based quantum devices have already been proposed in theory. As preliminary attempts to implement these proposals, multi-dot interaction or short distance spin-transfer has been demonstrated in both GaAs and Si-based devices. A promising alternative to having chains of dots is to integrate a single, elongated coupler, known as a jellybean dot, which enables coherent spin transport between two quantum dots located some distance apart. Since a single jellybean dot can potentially replace a chain of multiple dots, the complexity of the device architecture is reduced and thus favors scalability. These jellybean couplers have been demonstrated in the GaAs material system, but not yet in Si-based devices. Furthermore, the elongated jellybean dot can also act as a distributed charge sensor for qubit readout, as recently demonstrated in ref. [34].

Maybe not surprisingly, within a jellybean dot, the electron wavefunction is more spread out than in a typical,
more symmetric dot, which means that disorder effects and electron–electron interactions may dominate the electron distribution, possibly even leading to the formation of a Wigner molecule.[35–41] These effects significantly impact the charge characteristics and spin-transfer mechanism of the elongated dot. On the other hand, the electrical tunability of the atom–atom bonds inside such an artificial molecule is another useful feature that can enable applications in quantum chemistry, where a tunable bond between atoms can be used to simulate chemical phenomena of interest, such as femtochemistry,[42] molecular photochemistry,[43–45] and reactions in biochemistry.[46,47]

In addition to providing deep insights into the mesoscopic physics of solid state systems, the formation of a Wigner molecule within a jellybean dot could also be utilized to effectively simulate the quantum dynamics of a molecule or even a polymer.[48] As such, the characteristics and properties of jellybean dots need to be understood and experimentally studied.

In this paper, we investigate the charge characteristics of a jellybean dot fabricated on a silicon chip. We find that the charging pattern of our jellybean dot differs from that of a typical round dot in that it initially tends to form three distinct smaller dots (artificial atoms) that eventually merge into a larger one. The formation of this artificial molecule may be explained with either disorder effects or electron–electron interactions. Experimentally, the signatures of these two effects are very difficult to separate, and we perform theoretical calculations to understand the different types of electronic states formed at different dot occupations. The experiments show that we can tune the distance between two artificial atoms of this artificial molecule using a side barrier gate (\(J\)). Using field-effect model simulations, we confirm that the side barrier gate \(J\) can be used to tune the distance between the two leftmost artificial atoms by up to 33 nm, more than 40% of the minimum separation. Finally, magneto-spectroscopy measurements show that for low electron occupancies, the total spin behavior does not obey the shell filling pattern expected for a round dot, however, after the atoms of the artificial molecule merge into a single dot, the spin filling follows a distinguishable pattern. These results indicate that the jellybean dot is a promising platform to understand mesoscopic physics, and to perform spin-based quantum operations such as Pauli spin blockade, electron-spin resonance and coherent, long-distance spin transport in a Si-based quantum chip.

2. Charge Characterization of a Jellybean Dot

We start by characterizing the electrical properties of the device. Figure 1a shows a false-colored scanning electron microscope (SEM) image of a device nominally identical to the one measured here. The devices are fabricated on a natural silicon substrate with a stack of palladium gate electrodes that are

Figure 1. Device architecture and jellybean transport measurement. a) False-colored scanning electron microscope (SEM) image of a nominally identical device with gate labels. The location of the jellybean dot is indicated by the dotted ellipse. b) Schematic cross-section of the device along the dot channel, and dot filling in transport mode. c) Coulomb diamond measurement of P2 dot, where two types of the dots with different capacitive coupling to P2 are outlined.
separated by atomic-layer-deposition grown aluminum oxide.[49] The different colors in Figure 1a indicate the separate fabrication layers (first layer: gold; second layer: green; third layer: blue). Gate P2 controls the electrostatic potential of the jellybean dot (width: ≈30 nm; length: ≈150 nm), while gate RESB controls the tunnel rates between the jellybean and an electron reservoir formed by an ohmic contact (D) which runs underneath the metallic gate RG. A single-electron transistor (SET) can be used as a charge sensor or act as a second electron reservoir for transport measurements by accumulating the electrons from one of the SET ohmics (S) underneath the metallic gate TG. The tunnel rate between the jellybean dot and the second reservoir is controlled by gate J. Gates LCB and RCB provide lateral confinement of the quantum dot channel.

Figure 1b shows a schematic cross-section of the device and an illustration of the distribution of electrons and conduction band energies during the transport measurement. The gates SETB and P1 were fully turned-on ($V_{SETB} = V_{P1} = V_{RG} = 3$ V) to extend the electron reservoir toward the P2 dot. The J and RESB gates were tuned to the sub-threshold regime ($V_J = 2.6$ V, $V_{RESB} = 2.7$ V) to create the barriers confining the dots under the P2 gate. The transport measurement then detects the electron current tunneling through the elongated jellybean region under gate P2.

Figure 1c shows the transport current measurement through the dot channel as a function of the potential on P2. Different dot sizes and shapes lead to different charging energies and capacitive couplings to the surrounding electrostatic gates. Here we can identify two distinct charging behaviors, which indicate the existence of at least two types of dots under P2: one dot with stronger capacitive coupling (lever arm of 0.052 eV V$^{-1}$) and charging energy of 0.72 meV (grey dashed lines), and another dot with weaker capacitive coupling (lever arm of 0.008 eV V$^{-1}$) and charging energy of 0.89 meV (red dashed lines). Note that the grey and red dashed lines are a guide to the eye only. The apparent shapes of the diamonds may vary with different color scales.

The evidence of the multi-dot molecule can also be seen in Figure 2a,b, where different slopes of the charging lines indicate that more than one charging pattern was measured, suggesting that there are multiple dots under P2. This twofold charging behavior is a first indication that the jellybean dot supports separate electronic structures that can be interpreted as an artificial molecule.

In order to confirm that the two types of Coulomb diamonds correspond to two different types of dots under P2, we measure charge stability maps using the SET charge sensor. We can determine the location of the dots based on the relative intensities of the SET signal and the capacitive coupling to each of the gates. The map in Figure 2a shows two distinct charge transitions with different capacitive couplings to gate J, suggesting the presence of a dot weakly coupled to J (vertical transitions), and a dot closer to J (tilted transitions). A third dot closer to the RESB gate can be identified from the charge transitions induced by RESB (almost horizontal transitions in Figure 2b,c). We therefore conclude that there are three dots under gate P2: left and

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**Figure 2.** Charge occupation measurements using SET charge sensing. a,b) Charge transitions controlled by P2 together with a) the J gate and b) the RESB gate. c) Zoomed-in structure of the transitions in (b). d) Charging voltage of each of the dots with respect to each of the gates. e) Schematic sketch of the capacitive couplings and their strengths of the dots under P2.
right dots which are closer to the side gates J and RESB, respectively, and a dot located in the middle. At sufficiently high $V_{P2}$ in Figure 2a,b, the three dots merge into one larger dot.

In Figure 2d, the charging voltages of the different dots under gate P2 around the biasing point $V_{P2} = 1.9$ V versus various electrostatic gates are plotted (i.e., how large is the voltage change on a particular gate to load an extra electron onto a specific dot). Here, we extract the charging voltage of a particular gate by determining the voltage difference of two charging lines along the axis representing this gate. Gate P2 dominates the transitions of all three dots compared to other gates, which confirms that the three dots are all under gate P2. However, the P2 gate is more strongly coupled to the left dot than to the right dot, resulting from the asymmetric biasing and different widths of the J and RESB barriers. Figure 2e depicts a schematic of the capacitive model of the three-dot system and indicates the relative couplings of each of the dots to each of the gates. This system, which is composed of one dot in the middle and one dot on each side, can be interpreted as an artificial molecule. These considerations together with electrostatic simulations (see Section 7) allow us to gain an understanding of the experimental system, and how transition lines with different slopes are possible because of the different couplings to each gate. The different couplings are a natural consequence of the P2 dot gate being elongated and three times as long as a regular dot gate. The long gate allows for dots to be formed at both edges of the P2 gate that have a stronger coupling to the neighboring RESB or J gates as compared to the dot that is centered under P2. Furthermore, as shown in the next section, the different gate couplings allow the dots’ relative positions to be shifted by biasing those gates. In other words, the distance between the artificial atoms of the molecule can be tuned.

3. Tuning of the Artificial Molecule

We have identified an artificial molecule with three artificial atoms, each coupled to three electrostatic gates with different coupling strengths in the previous section (see Figure 2). We now show how these differences in coupling strengths can be leveraged to tune the separation between the artificial atoms and, effectively, change the molecular bonds of the artificial molecule.

In Figure 3a, we observe a bending of the transition lines for both the left dot (square markers) and the middle dot

![Figure 3](image_url)

**Figure 3.** Experimentally tuning the distance between artificial atoms. a) Charge stability map showing the transitions of the dots with bending slopes at high $V_J$ (with white dashed line in (a) as a guide to the eye). b) Schematic showing the shift in dot positions when $V_J$ is increased. Relationship between the slope and the position of the dots for c) the left dot, d) the middle dot, and e) the interdot distance.
(circular markers) for $V_j > 2.2$ V. This curvature indicates a change in capacitive coupling to the gates, and therefore a shift of the dot positions in a way that is illustrated in Figure 3b. Assuming the charging energy of a dot with a given number of electrons is fixed, the slope of the transition in gate space represents the ratio of the lever arms of the two electrostatic gates and allows us to determine the ratio of the effective coupling capacitances.\[50\] Then, we can model the relationship between the slopes of the lines (note we use $\tan(\theta)$ here for simplicity) and the dot distance, and plot the extracted distances in Figure 3c.d.

With this model, we can determine the positions of the dots from the values of $\tan(\theta)$. Taking three different voltage levels (I, II, and III) as examples, the left dot shifts from 48.3 nm (I) to 41.6 nm (II) to 80 nm (III) distance to the J gate (see Figure 3c), while the middle dot shifts from 120 nm (I) to 80 nm (II) to 80 nm (III) (see Figure 3d). Both dots shift closer to the J gate, when its bias is increased. The distance between the two dots is then estimated to change from $\approx 70$ to $\approx 40$ nm, at a rate of $-109$ nm V$^{-1}$ (see Figure 3e). This change in interdot distance corresponds to a change in the bond length of the artificial molecule, making our system a tunable artificial molecule.

This tunability of the bond distance can potentially benefit the future design of re-configurable quantum circuits, where in situ tunable couplings are required to create flexible connectivity between qubits without changing the hardware. Furthermore, the presented result can also enable an on-chip electronic platform of chemical reactions for the research of modern chemistry.

4. Spin Structure of the Jellybean Dot

Finally, we conduct a magneto-spectroscopy measurement to show that the jellybean dot has the potential to conserve the spin states while holding or transferring them in future quantum computing architectures.

In the experiment, we carefully measure the charging voltages as a function of applied magnetic field $B_0$. The voltage differences $\Delta$ between the transitions in Figure 4a are extracted and plotted in Figure 4b.\[51,52\] Before the 16th-to-17th transition, there is no clear spin structure visible, in contrast to what we have previously observed in a circular quantum dot.\[53\] This lack of structure is explained by disorder in the device and the complicated interactions between the electrons of the three artificial atoms. In addition, the dot itself is deforming in shape when $V_j$ is increased.

One sign of a robust spin state is that the differences in charging energies depend linearly on the magnetic field.

![Figure 4](image.png)

Figure 4. Magneto-spectroscopy on the jellybean dot. a) Transition map of the jellybean dot when sweeping $V_{P2}$ vs $B_0$. b) Extracted charging voltage differences $\Delta$ of the first 32 transitions.
through the Zeeman effect. Changes in magnetic field dependence are indicative of changes in the total spin quantum number, in general associated with the presence of degeneracies (in close analogy with Hund’s rules of atomic physics). We can see that the first 16 electron states have inconsistent field dependencies, which leads to the interpretation that the quantum dot is not forming homogeneously. However, from the 16th electron onward, we observe that the charging energies follow clear linear trends with occasional kinks. As understood from previous studies,[33] these kinks are where a crossing with an orbital or valley state results in a different spin state being loaded. While this is not definite proof for the formation of spin states in an elongated quantum dot, it does indicate that there may be some spin character in the states formed in such a quantum dot. We will next examine some of these transitions in greater detail.

After the 17th transition, the artificial atoms start to merge, leading to a single large dot when the electrostatic potential is dominating over disorder and electron–electron interactions. In this case, the addition of extra electrons follows a more systematic pattern and the magneto-spectroscopy is easier to analyze. While we cannot assign the spin state of the last electron loaded, while this is not definite proof for the formation of spin states in an elongated quantum dot, it does indicate that there may be some spin character in the states formed in such a quantum dot. We will next examine some of these transitions in greater detail.

Overall, the systematic spin filling observed here, promises that a well-defined spin state can be achieved for higher electron occupancies. This should allow for spin shuttling for a spin-1/2 state,[25–30] and jellybean coupling for a spin-0 state. [32]

5. Theoretical Modeling of Spin States in a Jellybean Dot

We have experimentally shown the charge characteristics of the jellybean dot (Figures 1 and 2), and possible signatures of a spin structure (Figure 4). In this section, we make use of theoretical modeling to understand the possibility of using such a quantum dot as a mediator for long-range coupling of spin qubits. One of the critical questions with regard to doing so is whether the electronic structure of such an elongated dot is stable and reproducible. In GaAs, the coherent coupling of qubits with mediator dots was successfully demonstrated,[31–33] which encourages the investigation of a similar device in MOS silicon. The main scientific questions behind the translation of this technology between platforms are whether 1) the disorder imposed by the SiO₂ interface will deter the dot from forming electronic states that extend across the complete dot, and 2) the electron-electron repulsion and quantum correlations in silicon will create significant departures in performance compared to the results in GaAs.

Experimentally, it is hard to distinguish the signatures of 1) and 2). Therefore, we outline below our theoretical model for understanding the different types of electronic states formed at different dot occupancies, highlighting the interaction effects in silicon. We highlight that silicon quantum dots have more potential to form non-trivial quantum states due to electronic interactions than GaAs. This is because silicon has a lower dielectric constant than GaAs, and its conduction band electrons have a larger in-plane effective mass than in GaAs, besides their valley degeneracy (or in general small valley splitting).

Modeling such a multi-electron dot is often difficult because of the large computational load necessary to simulate more than a couple of electrons. In the context of this device, where we have essentially created a multi-dot system under a single large gate (P2), we are able to use the Hartree–Fock method to simulate the system of many electrons in a single potential well. The key advantage of the Hartree–Fock method lies in its ability to simulate high electron numbers in our system within reasonable times, which is not always possible using other methods like full configuration interaction.[34,35] The trade off here is in the quantitative accuracy of the results, where one of the assumptions central to the method is that the interaction between the electrons is treated as a mean field such that the interaction of each electron with the other electrons depends only on where the electron is located in this field.[56] Regardless, we find that the method is satisfactory for the purpose of achieving a qualitative understanding of the electron system.

We describe here a brief overview of how the method works, and the details will be contained in Experimental Section. There are broadly two separate steps in the Hartree–Fock method: first, obtain the approximate potential of the quantum dot under the gates; second, solve for the Hartree–Fock Hamiltonian iteratively such that the energies and wavefunctions obtained from the final solution are consistent with the initial guess. To optimize the electronic potential of the system we expect from our devices, we convert the geometry of the device into a 3D model in COMSOL, upon which we can perform electrostatic calculations. This generates a 3D potential profile of the device, which will be used to model the confinement potential under which the electrons sit in the system. This process is elaborated in more detail in the Experimental Section.

The surface electric potential of the simulated device is shown in Figure 5a at a particular gate configuration of $V_l = 0.6$ V and $V_{RESB} = 1.0$ V. Of the general potential profile that is generated, we are most concerned with the potential profile within the boundaries of the P2 gate which is the primary gate under which we performed the experiments as indicated in Figure 1. Isolating the relevant part of the potential, we have effectively an elongated single dot quantum well. The profile of this confinement potential, $V_{confinement}$, is shown in Figure 5b. It can be observed that the potential is approximately parabolic along the $x$ direction but is more like a box potential along the $y$ direction. To highlight the profile of the confinement potential in the $y$ direction, we plot a cut along $y$ at $x = 0$ nm in Figure 5c. The obtained potential is then used to perform the Hartree–Fock algorithm.

The Hartree–Fock method begins by guessing an initial value for the energy and wavefunctions, which can be calculated based on the single electron Hamiltonian without interaction or we can use a previous solution. We define the system with a specific number of electrons and construct the Ham-
The Hamiltonian for the system by discretizing the 3D simulation cell defining the quantum dot, similar to a tight-binding model. We are essentially performing general unrestricted Hartree–Fock theory where we are minimizing the energy of a single Slater determinant in the most energetically favorable configuration, which does not restrict the spin degree of freedom. With each iteration of the method, there is a small rearrangement of the charge density of the electrons in the system as a function of position and the total energy also changes. Eventually, we consider the solution to be converged when the change in total energy, charge density, and exchange energy are respectively smaller than $10^{-4}$, $10^{-4}$, and $10^{-3}$ meV.

After obtaining a converged solution to the Hartree–Fock Hamiltonian, the two main outputs of interest are the total energies of the multi-electron system and the wavefunctions of the electrons in the system. We first examine the wavefunction outputs by looking at specifically the results obtained from the confinement potential defined by $V_J = 0.6$ V, $V_{B3SB} = 1.0$ V, and $V_{P2} = 1.8$ V. Using the output wavefunctions, we are able to calculate the spin densities of the highest occupied molecular orbital

$$
\rho_{\text{spin}}(x,y) = \sum_{i} \rho_{i,\uparrow}(x,y,z) - \rho_{i,\downarrow}(x,y,z)
$$

where these densities are summed along the z direction. The spin density informs us of the spin orbital shapes and the spin state of the electron in the highest occupied molecular orbitals, where a positive and negative density indicate spin up and spin down states, respectively. We note that because we are using an unrestricted Hartree–Fock algorithm, which does not place constraints on the spin states of the solution, the spin up and down wavefunctions are minimized separately.[56]

In Figure 5d, we plot the spin densities, extracted for the jellybean quantum dot being occupied with 2 to 20 electrons. The results show that some of the occupation numbers form non-trivial states where charge localizes in a chain, while other occupations reveal a smoother charge density distribution spanning the full quantum dot. The characteristics of these states are not typically expected in a quantum dot with regular shell structure[53] but they are also not identifiable as a fully formed Wigner molecule. Therefore, they are referred to as non-trivial states in the context of these results.

From the experimental results obtained, we developed a model of localized states under the same elongated gate and determined that the wavefunctions of the electrons under the P2 gate must be inhomogeneous, compared to typical formation of electron wavefunctions.[53] Our results corroborate theoretically this earlier hypothesis about the physics of such a jellybean dot in silicon, and the non-negligible, strong
electron–electron interactions. In these simulations, we consider primarily the effect of electron–electron interaction to attain an understanding of the system and to show that electron–electron interaction alone would be capable of causing this formation of non-trivial states. While these states do not match quantitatively with what was observed in the experiment, we show qualitative evidence of wavefunction localization into segmented chains as well as into more continuous shapes depending on the number of electrons, motivating the possibility of non-trivial state formation. In further studies, we could also account for effects of surface roughness and explicit spin-orbit coupling terms.[57]

6. Conclusion

In this article, we demonstrated charge transport through and charging of an elongated quantum dot—a so-called jellybean quantum dot—where the gate is about five times longer than for a typical quantum dot. The data shows that disorder and electron–electron interaction lead to the formation of one dot under the middle of the gate and two dots at the ends of the gate, corresponding to the formation of a tunable, artificial molecule. This interpretation is corroborated with electrostatic simulations in COMSOL and Hartree–Fock multi-electron simulations for up to twenty electrons. Finally, we presented magneto-spectroscopy results, which are indicative of well-defined states of the jellybean quantum dot formed in the device.

Our results are a first demonstration of the jellybean structure in SiMOS architectures, and will be instrumental in understanding the physics of jellybean couplers in silicon as we move toward using the jellybean as a qubit mediator of exchange coupling.

7. Experimental Section

**Measurement Setup:** The device was measured in a Bluefors LD400 dilution refrigerator with a base temperature of Tmin = 22 mK. The DC magnetic field was applied with an American Magnetics 5 T superconducting split-coil magnet. DC bias voltages were generated from a QDAC high-precision, low-noise, computer-controlled voltage source. The SET current and the charge transition of device were measured by a double lock-in technique.[18]

**Hartree–Fock Method:** In Figure 5, results obtained from electrostatic simulations performed were shown using the Hartree–Fock method. In order to perform these simulations, there were several input parameters that will need to be included. The method began with the confinement potential, Vconf, which defines the potential in which the electrons sit. Due to the size of the dot gate for the jellybean dot, it was expected that the potential may not be defined by a typical harmonic potential well. Therefore, COMSOL was used to simulate the potential of the jellybean dot device. Construction of a realistic 3D model of the device as shown in Figure 1a began using COMSOL, with one of the outputs being a realistic potential that described accurately the structure of the device, including but not limited to the substrate and the expected formation of the different gate layers during the fabrication process.

Taking this model, it was able to generate the potential profile in COMSOL that was formed under the gates given a particular set of gate voltages using the electrostatics module. It was able to emulate the voltage configuration used in the experiment and approximate define the shape of the potential, and also generate potentials while simulating accurately the effects of the gates on the device. These simulations were used to obtain a set of confinement potentials that were defined by the P2 gate, the J gate, and the RESB gate, all of which were labeled in Figure 1a.

The Hartree–Fock method is now detailed as it was performed in the simulations, which more commonly would be considered as the unrestricted Hartree–Fock method.[56] At its core, the simulation works by considering all electrons other than the electron being studied as a static electric field, and considered the effect of that field on the electron. The system in which the electrons interact was defined by two properties in the simulations. The first was the confinement potential, which was just described. The second was the simulation cell in which the simulations were performed. The size of the simulation cell was strongly correlated with the potential profile obtained from the COMSOL simulations, and was defined such that only the area defined by the P2 dot gate was included, and did not extend into neighboring gates.

In the calculations, the charge, spin, and valley degrees of freedom was also considered. The charge degree of freedom was defined by two quantities; one was the number of electrons in the system, and the other was the location of the electrons within the simulation cell. The spin degree of freedom was defined such that two separate wavefunctions described the spin up and down states were obtained. Finally, the valley degree was defined by modeling different hopping parameters along the interface direction.[59] In the following paragraphs, the mathematical model for the Hartree–Fock method was described.

The complete Hartree–Fock Hamiltonian was given by

\[ H_{\text{HF}} = \sum_{i=1}^{N} \left( H_{\text{single}}(r_i) + H_{\text{conf}}(r_i) \right) \]

where \( H_{\text{single}}(r_i) \) is the single electron Hamiltonian, without considering electron-electron interaction, and can be written as follows

\[ H_{\text{single}} = H_k + V_{\text{conf}} \]

where \( H_k \) can be interpreted as the kinetic energy of the system and \( V_{\text{conf}} \) is the confinement potential of the system, including both the quantum well potential and the vertical confinement electric field.

The Hamiltonian was constructed by considering a cubic simulation cell in three dimensions, with a fourth dimension accounting for spin, effectively representing the Hamiltonian using a 4D matrix. The cuboid was divided into grids along each of the Cartesian directions, and grid sizes of 0.4 nm, 0.8 nm, and 0.1 nm were chosen along the x, y, and z directions. The grid sizes were chosen such that they were small enough to account accurately for changes in the wavefunction in space, while also not too small that it became computationally difficult. This was because the size of the Hamiltonian was defined by the size of the cuboid grid along the Cartesian directions, and the last dimension of spin having a size of 2.

In essence, the Hartree–Fock algorithm takes into account electron–electron interactions in addition to the Fock states (comprising the kinetic and potential terms) in constructing the Hamiltonian. A self-consistent field method was used to solve the system Hamiltonian iteratively to minimize the total energy of the system. This method allowed to simulate large numbers of electrons in the system, as demonstrated in the main result of the paper.

The method was designed following a similar vein to that in ref. [55], which considered a tight-binding model of electrons and accounts for the valley degree of freedom by having different hopping coefficients for different sites along the z direction. Accounting for the valley degree of freedom was a key ingredient that allowed to account for the distribution of electrons accurately in silicon-based devices.

One of the main factors affecting the accuracy and efficiency of the method was the starting point of the algorithm. In order to estimate a starting point close to the final solution as possible without having to actually perform the algorithm, the single particle states of the system were calculated. An analytical potential model that is close to the expected potential extracted was considered from COMSOL simulations and it was used to calculate the single particle states. The single particle states will be used as the initial point of the simulation.
As a starting point for the simulations, we solve for the single particle Hamiltonian

\[ H_{\text{single}} = H_{\text{K}} + V_{\text{confinement}} \]  

(4)

where \( H_{\text{K}} \) is the kinetic energy Hamiltonian and \( V_{\text{confinement}} \) refers to the confinement potential as described. Without taking into account the electron-electron repulsion, the single particle state energies were able to be calculated and that it serves as the starting point of the simulation. This ensured that the initial state was sufficiently close to the final state.

During the method, the total energies of the electrons in the system will be minimized and is given by

\[ E = \sum_{i} \varepsilon_{i} - \frac{1}{2} \sum_{j} V_{ij} \sum_{i} |\psi_{i}|^{2} - V_{\text{ex}} \]  

(5)

where sums over \( i \) are over the total number of electrons while sums with respect to \( j \) are over the total space. Here it is noted that these were essentially the Fock terms, the Hartree terms and the exchange term, respectively. The change in the total energy was calculated by comparing it with the result obtained in the previous iteration. The energy was considered as converged when the change in total energy, \( \Delta E \), is less than \( 10^{-4} \) meV. This was the first check for convergence as part of a three-step check before the algorithm deems the solution to be converged. Should the total energy, \( E \), be converged, the algorithm will then check for the change in wavefunction at this iteration step, \( \Delta \psi \), and will be deemed as converged if it is less than \( 10^{-4} \) meV. Finally, if the wavefunction was converged, the algorithm will re-calculate the exchange energy for the new electronic configuration and check for the change in the exchange energy, \( \Delta E_{\text{ex}} \), and the solution will be considered overall converged, if this change is less than \( 10^{-3} \) meV. This was based on the idea that the initial calculation of exchange may be wrong and that would also result in an erroneous result for the wavefunction and energy, and if that is not the case, the wavefunction and the exchange energy would be consistent and therefore leading to a small \( \Delta E_{\text{ex}} \). By defining convergence in this way, it can be afforded to set looser bounds on the convergence criteria, and even though there were three different sets of convergence criteria that need to be met, there exists a speed-up in the algorithm due to the looser bounds. If the energy and wavefunction convergences were not achieved, the wavefunction will be mixed to include a proportion of the newly calculated wavefunction in the following way

\[ \psi_{\text{new}} = (1 - \beta)\psi_{i-1} + \beta\psi_{i}, \]  

(6)

where \( \beta \) typically varies from \( 10^{-3} \) to \( 10^{-1} \). Should \( \Delta E \) and \( \Delta \psi \) both be \( <10^{-4} \) meV, but the re-calculated exchange energy differs from its initial value by a significant amount larger than \( 10^{-3} \) meV, the algorithm re-approaches the converged solution with this new value for exchange energy. This method of converging the exchange energy first reduced the overall time required to perform the algorithm as the calculation for the exchange operator can take up a significant portion of the total run time. Second, this stabilized the algorithm as it ensures that not too many parameters in the simulation are being changed at each stage of the simulation.

After convergence was achieved, the two primary outputs of the Hartree–Fock method were stored, namely the total energy of the system, and the wavefunctions of the electrons in the system. The spin densities can be calculated by taking the difference between the highest occupied molecular orbital spin up and down densities as shown in Equation (1).

It was also emphasized that the simulations were only accurate up to the accuracy of the set convergence criteria. There remained error on the order of magnitude smaller than \( 10^{-4} \) meV for the energy and wavefunction, as well as errors smaller than \( 10^{-3} \) meV for the exchange energy. Therefore, the accuracy of the wavefunctions can vary and some inconsistencies as well as artifacts in the wavefunctions can remain at this level of accuracy. The wavefunction was carefully examined only on its general shape and not on its detailed structure at every grid position.

**Dot Distance Modeling:** For the dot distance calculations, a simplified field-effect model was employed, which is depicted in Figure 6. It was assumed that there was only one main electric field line that represents the real influence of the complicated electric field distributions, shown as solid dark color arrows in Figure 6a,b. If the displacement distance of the dot was small enough, the electric field line's trace can be then simplified into two straight lines as Figure 6c, which corresponded to the so-called effective path approximation. This simplification was valid if the P2 voltage was similar to the J gate voltage as in Figure 3. However, owing to the traps that would disperse the electric field, the effective electric field strength will decrease along the direction it extends. It was, therefore, a need to include some compensation to reduce the error further. The detailed expression of the model can be found in the following paragraphs.

![Figure 6](image-url)
To simplify the calculation, the dot was treated under P2 as a point charge. The capacitive coupling between a specific gate and the dot can then be depicted by

\[ C_{\text{tot}} = \sum_{i,j} \frac{p_{C,ij}}{\varepsilon_{C,ij}(r)} \]  

(7)

where \( \vec{r} \) is the unit vector along the electric field line \( C \), and \( p_{C,ij} \) and \( \varepsilon_{C,ij} \) are the weight of the coupling strength and the local permittivity of the path \( C \), respectively. When the P2 voltage was high, the electric field lines from the J gate were likely to be concave, and those from the P2 gate were likely to be straight lines because of the relative location between the dot and the P2 gate, as schematically shown in Figure 6a,b. In this case, the main part of the electric field lines emitted from a specific gate can be condensed into one representative path, for example, \( L' \) and \( L'' \) for the J-dot and P2-dot path, respectively. Then, the capacitance between the gate and the dot can be expressed as

\[ C_{\text{J-dot}} = \sum_{i,j} \frac{\varepsilon_{C,ij}(r)}{\varepsilon_{C,ij}} \]  

(8)

and

\[ C_{\text{P2-dot}} = \int \frac{\varepsilon_{C}(r)}{\varepsilon_{C}} dr \]  

(9)

When sweeping the J (P2) gate by the amount of the charging voltage \( \Delta V_J (\Delta V_{P2}) \), as is done in the measurement in Figure 3a, the charge of the dot changes by one electron. So, the ratio of charging voltages \( \tan \theta \) also equals

\[ \frac{\Delta V_J}{\Delta V_{P2}} = \frac{\int_{C_{\text{J-dot}}} \varepsilon_{C}(r) dr}{\int_{C_{\text{P2-dot}}} \varepsilon_{C}(r) dr} \]  

(10)

Here, the influence from the anisotropy of the dielectrics can be reasonably neglected for simplification. Additionally, by assuming the paths were polylines instead of curves, as shown in Figure 6c, the ratio can be further simplified as

\[ \tan \theta = \frac{\sum_{i,j} \varepsilon_{\text{eff},ij}}{\sum_{i,j} \varepsilon_{i,j}} \]  

(11)

where \( i \) and \( j \) represent the different dielectrics along the path and \( t_{\text{eff}} \) is the effective thickness of the dielectric along the path. For the P2 gate, the dot was near so that the attenuation imposed by the charge traps can be neglected. However, for the J gate, which was far from the dot, the attenuation was no longer negligible, as shown in Figure 6d. In this work, it was assumed to be a factor that increases the intrinsic thickness of the dielectric \( t_{\text{eff}} \) and was calculated in the following form\(^{[25,60]} \)

\[ t_{\text{eff}} = t_{\text{eff}}^{\text{J}}, e^{-z_f/l_z} \]  

(12)

where \( f \) represents the current position of the dot and \( l_z \) is the initial distance between the dot and the J gate. Referring to the fabrication layout design, the left dot and the middle dot was estimated to be 45 and 130 nm away from the middle of the J gate, respectively. Then, given the thickness of each ALD-grown dielectric layer, the relationship between the dot’s location and the charging voltage ratio can be achieved, as articulated in the main text.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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quantum computation, quantum devices, quantum dots, silicon nanostructures, spin qubits

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