Electron transport through silicon serial triple quantum dots

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1. Introduction

Silicon double quantum dots (DQDs) are promising candidates for developing quantum information devices based on electronic state or electron spin quantum bits (qubits). So far, coherent manipulation of the qubits has been demonstrated mainly by DQDs in GaAs systems [1–3]. However, several deleterious effects, such as the hyperfine interaction, the spin–orbit interaction, and the piezoelectric electron–phonon interaction, severely limit coherence times [1–4]. Longer coherence times are expected in silicon DQDs because of the predominantly spin-zero nuclei, weak spin–orbit coupling, and the lack of piezoelectric electron–phonon coupling [5]. In addition, silicon qubits have a great advantage in integration because of the compatibility with conventional silicon complementary metal–oxide–semiconductor processes.

In order to investigate electron transport through silicon DQDs, several experimental studies have been carried out, including studies on gate-defined DQDs [6,7], ion implanted Si:P DQDs [8], and Ge/Si core/shell nanowires [9]. Furthermore, the spin dependent electron transport referred to as spin blockade due to the Pauli exclusion principle in the tunneling characteristics [10,11] has been demonstrated in DQDs in Si/Ge two-dimensional electron gases [12] and in naturally-formed DQDs in silicon nanowires [13]. This phenomenon is a valuable tool for the measurement and manipulation of individual electron spins [2,3]. Additionally, the multivalley conduction band structures of silicon in DQDs [14] have been highlighted for further investigation.

A triple quantum dot (TQD) system is the natural extension of the DQDs toward scalable multiqubit systems. Theoretical studies of TQDs indicate strong potential for applications in quantum information technology. To improve the coherence time, coded qubits with TQDs have been studied [15,16]. Spin cluster qubits, where no local control of spins is required, have also been proposed with a TQD system as the minimum system [17]. Furthermore, entangled electron spins are expected to be created by TQDs as spin-entangled currents [18]. In addition to applications in quantum information processing, charging rectifiers using TQDs have been proposed and demonstrated [19,20]. Recently, experimental electron transport of TQDs has been investigated in AlGaAs/GaAs heterostructures [21–23], a single-wall carbon nanotube [24], and self-assembled InAs quantum dots [25]. However, clear electron transport characteristics for silicon TQDs have not been reported yet.

In this paper, we demonstrate electron transport through silicon serial TQDs formed effectively in a lithographically-defined multiple quantum dot system on a silicon-on-insulator substrate at a temperature of 4.2 K. Our serial TQDs are composed of two lithographically-patterned QDs and another one in-between formed by stress during the pattern-dependent oxidation process. The TQDs formation is confirmed by equivalent circuit simulations, which show an excellent agreement with the experimental results. With detailed analysis of the charge configurations in the TQDs, we discuss the distinct properties of the TQDs, including electron transport at the charge quadruple points. In addition, we discuss higher order tunneling processes of the TQDs. The analysis of electron states in the silicon TQDs is a crucial step toward the future implementation of integrated silicon quantum information devices.

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formation, by analyzing the electron transport characteristics. In Section 4, we show a detailed analysis of electron transport through the TQD system with calculated charge configurations.

2. Device structure and fabrication process

We fabricated a lithographically-defined silicon multiple QD system, in which TQDs are effectively formed. Fig. 1a shows the scanning electron microscope (SEM) image of the silicon multiple QD system, which has the eight constrictions, defined on a silicon-on-insulator (SOI) substrate with a silicon layer thickness of about 20 nm on a 200-nm-thick buried oxide layer. The multiple QDs are connected to the four terminals (T1, T2, T3, and T4), where bias voltages are applied. The electrochemical potentials in the QDs can be modulated with six side gates (G1, G2, G3, G4, G5, and G6).

First, the 40-nm-thick SOI film, whose thickness was reduced via thermal oxidation, was deeply doped by ion implantation (n-type, phosphorous, doping concentration $\sim 1 \times 10^{19}$ cm$^{-3}$). Multiple QDs were then patterned using high-resolution electron beam lithography with the positive resist ZEP520A. Electron cyclotron resonance reactive ion etching was used to transfer the resist pattern into the SOI layer with CF$_4$ used as the etching gas. Thermal oxidation was then performed for 30 min at 1000 °C in order to passivate the surface states and reduce the size of the QDs. The thickness of the SOI layer was also reduced to about 20 nm during this process. Finally, 300-nm-thick Al electrodes were formed by evaporation.

3. Discussion for the formed QDs

Fig. 1b shows the effectively formed QDs in the multiple QD system with the equivalent circuit, where the dotted blue$^1$ lines indicate the outer shape of the multiple QD system. From the detailed analysis of the experimental and simulated results discussed below, it was concluded that the four QDs (QD1, QD2, QD3, and QD2$^x$) illustrated by the four red circles in Fig. 1b were formed effectively with the seven tunnel junctions $t_i$ ($i = 1 - 7$). In this paper, we focus on electron transport through the serial TQDs formed on the left side of the multiple QDs as indicated by the green square in Fig. 1b.

All measurements were performed using the Agilent 4156A parameter analyzer at a temperature of 4.2 K in liquid helium. Since capacitive coupling between the gate G1(G4) and QD1(QD2) is expected to be strong, the voltages applied to G1 and G4 ($V_{G1}$ and $V_{G4}$) were used to efficiently control the electrochemical potentials in the TQDs. Fig. 2a–d shows the contour plots of the currents $I_{T1}$, $I_{T2}$, $I_{T3}$, and $I_{T4}$ measured at the terminals T1, T2, T3, and T4, respectively, as a function of the gate voltages $V_{G1}$ and $V_{G4}$, where the voltages applied to T1, T2, and T3 ($V_{T1}$, $V_{T2}$, and $V_{T3}$) are $-6$ mV and the voltage to T4 ($V_{T4}$) is 0 mV. The electron configurations of the multiple QDs strongly influence these characteristics, which are referred to as charge stability diagrams [29].

Then we simulated these properties using a Monte Carlo technique with the equivalent circuit (Fig. 1b). We carefully extracted the gate capacitances from the experimental results and optimized the tunnel junction parameters with more than 200 iterations. The simulated charge stability diagrams as shown in Fig. 2e–f, corresponding to the experimental results in Fig. 2a–d, respectively, show an excellent agreement with the experimental results. In particular, the charge configurations are almost perfectly reproduced. Therefore, the simulations support the assumed QDs formation. In addition, since $V_{T1}$, $V_{T2}$, and $V_{T3}$ have the same potentials and the current paths are from T1, T2, and T3 to T4, the current peaks of these charge stability diagrams, particularly $I_{T2}$, strongly reflect the electron configurations of the serial TQDs (QD1, QD2, and QD2$^x$). To the best of our knowledge, this is the first demonstration of electron transport through silicon serial TQDs.

We will now discuss the validity of the absence of the right side tunnel junction as indicated by A in Fig. 1b. Fig. 3a shows the electrical characteristics of $I_{T1}$, $I_{T2}$, and $I_{T4}$ as a function of $V_{T1}$, where $V_{T2}$, $V_{T3}$, and $V_{T4}$ are 0 mV. In the low biased region, which is the inside of the two red dotted lines, only $I_{T1}$ and $I_{T4}$ flow, indicating the electron transport only through QD1 and QD3. In the high biased region, $I_{T2}$ and $I_{T4}$ start to flow indicated by red ovals in Fig. 3a because Coulomb blockade among QD1, QD$x$, and QD2 is lifted. However, when we apply just the voltage $V_{T3}$ as shown in Fig. 3b, only $I_{T1}$ and $I_{T3}$ are detected. In this case, the conduction path is just QD1 and QD3. This is attributed to the weak or almost disconnected right side interconnection. Additionally, in Fig. 2a and c, the current peaks of $I_{T2}$ are very weak compared to those of $I_{T1}$, in spite of the same bias voltages being applied to T1 and T3. As the connection A is absent, the four QDs exist in the conduction path from T3 to T4, resulting in the low current amplitude.

Next, the connection B indicated in Fig. 1b is discussed. At the lower left region of Fig. 2b ($I_{T2}$), the electron configurations of the TQDs have a strong influence on the current peaks, whereas parallel current peak lines are observed at the upper right region of Fig. 2b, in which the currents through the TQDs are blocked [see Fig. 2a], indicating that a single QD is responsible for the current oscillation. In addition, $I_{T2}$ and $I_{T4}$ have almost the same current levels in Fig. 3a. If there had been a QD between $t_5$ and $B$ in Fig. 1b, $I_{T2}$ would be different from $I_{T4}$. Therefore, only QD2 is formed in the left bottom region of the multiple QD system.

Here, we discuss the possible formation mechanism of the effectively formed four QDs. Since the bottom of the conduction bands rises due to the quantum-mechanical size effects in the lithographically-defined strong constrictions, the seven tunnel barriers are formed. However, at the two constrictions indicated by A and B in Fig. 1b, the tunnel junctions are not effective as discussed previously; the coupling at the constriction A is weak because of the very strong constriction, whereas the coupling at the constriction B is strong because of the very weak constriction. In the re-
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4.1. Charge configurations in the charge stability diagram
serial TQDs
4. Detailed analysis for electron transport through the silicon
at a relatively high temperature (T = 4.2 K).
Therefore single-electron tunneling in the TQDs can be observed
very small QD is formed in silicon by the PADOX mechanism.
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patterns of QD1, QD2, and QD3, the bottoms of the conduction band
regions of QD1, QD2, and QD3, the bottoms of the conduction band
are lower than those at the constrictions, resulting in the formation
of the QDs. However, the region of QDx is just on a constriction.
Therefore, the mechanism of quantum confinement for QDx is different
from that of the other QDs. Quantum confinement in QDx is
presumably due to the compressive stress generated during the
pattern-dependent oxidation\cite{30} (PADOX) after electron beam
lithography. If a connection sandwiched with two large areas is
very narrow, compressive stress arises in the connection, lowering
the bottom of the conduction band. As a results, QDx is formed at
the constrictions. In contrast to the gate-defined TQDs\cite{21}, the
very small QD is formed in silicon by the PADOX mechanism.
Therefore single-electron tunneling in the TQDs can be observed
at a relatively high temperature (T = 4.2 K).

4. Detailed analysis for electron transport through the silicon
serial TQDs
4.1. Charge configurations in the charge stability diagram
Fig. 2. (a)–(d) The contour plots of $I_{t1} , I_{t2} , I_{t3} , \text{ and } I_{t4}$ as a function of $V_{G1}$ and $V_{CA}$, where $V_{G1} , V_{T2} , \text{ and } V_{T3}$ are $-6 \, \text{mV}$ and $V_{T4}$ is $0 \, \text{mV}$ (T = 4.2 K). (e)–(h) The simulated
contour plots of $I_{t1} , I_{t2} , I_{t3} , \text{ and } I_{t4}$, respectively, as a function of $V_{G1}$ and $V_{CA}$, where $V_{G1} , V_{T2} , \text{ and } V_{T3}$ are $-6 \, \text{mV}$ and $V_{T4}$ is $0 \, \text{mV}$ (T = 4.2 K). The circuit parameters are as
follows: $C_1 = C_2 = C_3 = 16 \, \text{aF}$, $R_1 = R_2 = R_3 = 500 \, \text{kA}$ ($t_1 , t_2$ \text{ and } $t_3$), $C_4 = C_5 = 5.5 \, \text{aF}$, $R_4 = R_5 = 500 \, \text{kA}$ ($t_4$ and $t_5$), $C_6 = 1.0 \, \text{aF}$, $R_6 = 1 \, \text{M} \, \Omega$ ($t_6$), $C_7 = 0.5 \, \text{aF}$, $R_7 = 1 \, \text{M} \, \Omega$ ($t_7$), $C_8 = 1.258 \, \text{aF}$, $C_{12} = 0.409 \, \text{aF}$, $C_{11} = 0.05 \, \text{aF}$, $C_{10} = 0.018 \, \text{aF}$, $C_{9} = 0.546 \, \text{aF}$, $C_{6} = 1.097 \, \text{aF}$, $C_{4} = 0.014 \, \text{aF}$, $C_{2} = 0.5 \, \text{aF}$, $C_{3} = 0.1 \, \text{aF}$, $C_{1} = 2 \, \text{aF}$, $C_{5} = 2 \, \text{aF}$, and $C_{0} = 2 \, \text{aF}$.
The gate capacitances are estimated from the experimental results. The parasitic capacitances are assumed to have the same order value of the other gate capacitances.
Typical values of tunnel resistances for silicon systems are used\cite{6,7,28}. The offset gate voltages of $1.6 \, \text{V}$ for $V_{G1}$ and $1.7 \, \text{V}$ for $V_{CA}$ are used to correct the effects of the background charges.

Fig. 3. (a) and (b) Electrical characteristics $I_{t1} , I_{t2} , I_{t3} , \text{ and } I_{t4}$ as a function of (a) $V_{T2}$ and (b) $V_{T3}$, where $V_{T2}$ and $V_{T4}$ are grounded and the all gates are floating (T = 4.2 K).

Fig. 4 shows the calculated charge stability diagram from the equivalent circuit simulation, corresponding to the inside of the white square in Fig. 2a. The gray lines mark the boundaries of the charge stable cells in which the stable charge configurations ($N_1 , N_2 , N_3$) are shown, where $N_1 , N_2$, and $N_3$ correspond to the charge numbers of QD1, QD2, and QD3, respectively. For simplicity, we assume that ($N_1 , N_2 , N_3$) = (0, 0, 0) at the hatched region, although there are actually many electrons in the system because of the high doping concentration.

At the boundaries of the charge stable cells, the total electrostatic energies of the neighboring charge states are degenerate. There are three types of boundary in the charge stability diagram of the TQDs. Firstly, since a charging event with a single charge in a QD of the TQDs occurs across, for example, the red lines in
charge configurations in DQDs, a three-dimensional
i.e. the size of the QD
lation is discussed below) compared with those of QD1 and QD2,
the SEM observation (Fig. 1a).
chemical potentials of QD1, QD2, and QD
namely with relatively high bias voltages, and the three electro-
electrochemical potential of the QD
is far from the bias window
is very small, which is also supported by
indicates that the charging energy of QD
A charge triple and quadruple points
Fig. 4. The simulated charge configurations in the stability diagram of the TQDs. In order to show the sharp boundaries of the charge stable regions, almost zero bias voltages and the temperature of 10 mK are used in the simulation. The stable charge configuration in each cell is denoted by \( N_1, N_2, N_3 \), where \( N_1 \), \( N_2 \), and \( N_3 \) correspond to the electron numbers of QD1, QDx, and QD2, respectively. These electron numbers are the relative values from those of the hatched region, where \( N_1, N_2, \) and \( N_3 \) are assumed to (0,0,0) for simplicity.

Fig. 4, these lines are referred to as the charging lines. These three red lines correspond to the charging lines of QD1, QD2, and QDx as indicated in Fig. 4. Many charging lines exist in the stability diagram. Second, at the blue line with the mark R in Fig. 4, a charge at QD2(x) moves to QDx(2), but the total numbers of charges in the TQDs stay constant. This type of line is the charge reconfiguration line, which arises from the electrostatic coupling (and also quantum-mechanical tunnel coupling which is neglected in the simulation). In the charge stability diagram of DQDs, these two types of line also appear [29]. However, the other kind of line indicated by the green line in Fig. 4 is identified in the TQDs, where both a charging event and a charge reconfiguration event occur simultaneously. Since the two charges simultaneously move in the TQDs at this line, in a similar way to quantum cellular automata (QCA) processes [32], this line is referred to as the QCA line [21]. In the experimental results, a strong current peak at the QCA line is observed as indicated by the white arrow in Fig. 2d.

4.2. Charge triple and quadruple points

The meeting points of two charging lines or a charging line and a QCA line are charge triple points, where three charge configurations are degenerate. Although sequential tunneling is allowed at the charge triple points in the case of the serial DQDs [29], electron transport at the charge triple points in the serial TQDs is a second order tunneling process. However, almost all the current peaks of \( I_{T1} \) (Fig. 2a) correspond to the charge triple points because our measurement is performed in the non-linear transport region, namely with relatively high bias voltages, and the three electrochemical potentials of QD1, QD2, and QDx located in the bias window. In the region where the current peaks disappear in Fig. 2a, the electrochemical potential of the QDx is far from the bias window due to the very wide spacing of the charging lines of QDx. This indicates that the charging energy of QDx is very large (the calculation is discussed below) compared with those of QD1 and QD2, i.e. the size of the QDx is very small, which is also supported by the SEM observation (Fig. 1a).

Although a two-dimensional charge stability diagram is enough to describe charge configurations in DQDs, a three-dimensional charge stability diagram spanned by three gates is necessary in order to obtain full charge configurations in TQDs, because there are three discrete charges in TQDs. This is referred to as a beehive diagram [21]. In three-dimensional space, charging lines and triple points turn into charging plains and triple lines, respectively, and the meeting points of the two triple lines form charge quadruple points, where the four electron configurations are degenerate. Sequential tunneling in the linear transport region for the serial TQDs is allowed only at the charge quadruple points. At the electron configurations from (0,0,0) to (1,1,1) in a three-dimensional charge stability diagram of the TQDs, there are the four charge quadruple points [21]:

At Qx and Qy, a single charge is transferred through the TQDs, whereas two charges are involved in the transport at Qz and QDx. These two charge transport processes can only occur in QD systems comprising more than two QDs.

At the black closed circle indicated by Q in Fig. 4, the charge quadruple point which involves three electrons appears. The four charge configurations (0,0,2), (1,0,2), (0,1,2), and (0,1,1) are degenerate at Q, corresponding to Qy with an extra single charge on QD2. Fig. 5a shows the schematic potential diagrams at Q, where the left, center, and right potential wells correspond to those of QD1, QDx, and QD2, respectively. The electrochemical potentials \( \mu_1, \mu_2, \mu_3, \mu_4, \mu_x, \) and \( \mu_y \) are those of QD1, QD2, QDx, the terminal T1, and T4, respectively. In the (0,1,1) configuration, the electron on QDx can move to QD2 because \( \mu_x(0,1,1) = \mu_y(0,0,2) \). This transfer occurs below the electrochemical potentials of T1 and T4. Then one electron fills
\( \mu_1(1, 0, 2) \) from T1, resulting in the \((1,0,2)\) configuration. The configuration of \( \mu_1(1, 0, 2) = \mu_2(0, 1, 2) \) allows the tunneling of the electron from QD1 to QD2. Finally, the electron on QD2 escapes to T4 and the configuration returns to the initial state \((0,1,1)\). Note that the sequential tunneling at Q is different from that at Q4 or Q5 because of the different number of charges. In addition, spin blockade between QDx and QD2 is expected to appear in this charge configuration because the two specific conditions \((0,1,1)\) and \((0,0,2)\) are involved, where the two electron spins can form the spin singlet or triplet. The study of spin blockade in TQDs remains as a matter to be discussed further.

### 4.3. Charging energies of the TQDs

From the parameters used for the equivalent circuit simulation, the charging energies of QD1, QD2, and QD3 (\( E_{QD1}, E_{QD2}, \) and \( E_{QD3} \)), and the electrostatic coupling energies between QD1(2) and QDx \( |E_{1(2)x}| \) are calculated from the electrostatic energies of the system (a full derivation is shown in Appendix A). These energies obtained are \( E_{QD1} \approx 5.6 \) meV, \( E_{QD2} \approx 10.4 \) meV, \( E_{QD3} \approx 101 \) meV, \( E_{1x} \approx 3.6 \) meV, and \( E_{2x} \sim 3.3 \) meV. Note that \( E_{QDx} \) is very large, resulting in the very wide spacing of the charging lines of QDx, corresponding to a very small size for QDx (\( \approx \) few nm radius). The PADOX process in the silicon nanostructures allows us to realize such a small QD.

In contrast, for the estimation of the charging energies from the experimental results, the conversion factors by which gate voltages are converted into energies, must be extracted from the non-linear transport results [29]. Fig. 5b shows the schematic image of the non-linear transport of the charge quadruple point \( Q_3 \) in the three-dimensional stability diagram spanned by the three gate voltages \( V_{Gx}, V_{Gy}, \) and \( V_{Gz} \) for TQDs. In non-linear transport, the charge quadruple point turns into the triangular pyramid (the black bold and dotted lines). On the \( V_{Gx}=V_{Gy} \) plain, a two-dimensional slice appears as illustrated by the red triangle in Fig. 5. The configurations of the electrochemical potentials at the four vertices A, B, C, and D are also shown. The line AD corresponds to the charge reconfiguration line for the three QDs. To extract the accurate conversion factors for three QDs, the two side of the triangular pyramid must be identified.

In the experimental results (Fig. 2a), several triangular current peaks appear, corresponding to just the two-dimensional slice of the triangular pyramids. However, the charging energies can be roughly estimated from the triangles in two-dimensional space. Since \( G1(G4) \) is strongly coupled to QD1(QD2), we obtain the conversion factors of the QD1 and QD2 \( \gamma_1(2) \) from the triangle of the \( V_{Gx}=V_{Gy} \) plain. We chose the clearest triangle indicated by the white arrow in Fig. 2a. If we assume that the bias voltages drop only at the tunnel junctions, \( \gamma_{1(2)} \) is calculated as 0.061 (0.071) eV/V. From the conversion factor and the current peak spacing of QD1(2), the charging energy of QD1(2) is estimated as 7.7 (10.3) meV. These values are consistent with the charging energies calculated from the equivalent circuit parameters, probably because the triangle shape is close to the side of the triangular pyramid due to the small capacitive couplings between QDx and G1 or G4.

### 4.4. Higher order tunneling processes

Finally, we explore the higher order tunneling processes of the TQDs. In the transport of the TQDs, several kinds of tunneling processes appear. The charge quadruple points and the charge triple points correspond to first and second order tunneling processes, respectively. The leakage currents of the Coulomb blockade region are due to fourth order tunneling processes. On the QCA lines (the green line in Fig. 4), where two charges move simultaneously, another higher order tunneling process occurs, which is referred to as QCA cotunneling processes [21].

Here, we compare the other two kinds of higher order tunneling processes. Fig. 6a shows the extended figure corresponding to the blue square in Fig. 2b, where the electron configurations assumed in Fig. 4 are denoted. In this gate voltage region, the three electron configurations \((0,0,1), (0,1,1)\), and \((1,1,1)\) are distinguished. Between \((0,0,1)\) and \((0,1,1)\), only the electrochemical potential of QDx locates in the bias window. As a result, two successive second order tunneling processes with a tunneling rate of \( I_{2-2} \) are required in the electron transport as schematically shown in Fig. 6b. Conversely, at the boundary of \((0,1,1)\), and \((1,1,1)\), there is only the electrochemical potential of QD1 in the bias window and the electron transport is dominated by the third order tunneling processes with a tunneling rate of \( I_{3} \) shown in Fig. 6c. The black broken lines in Fig. 6a indicate these two tunneling processes. Note that the tunneling rate of the two successive second order tunneling process is higher than that of the third order processes, leading to the different current amplitudes in Fig. 6a. This fact is consistent with other TQD systems [21,24].

### 5. Conclusion

Experimental electron transport through the silicon serial TQD system has been successfully observed for the first time at a temperature of 4.2 K. The TQD system is composed of two lithographically-patterned QDs and another one in-between formed by compressive stress generated during the PADOX process [30]. The equivalent circuit simulation shows an excellent agreement with the experimental results, which support the assumed formation of the TQDs. The calculated charge configurations of the TQDs, which are defined by three types of lines (the charging lines, the charge reconfiguration lines, and the QCA lines), provide a detailed analysis of the electron transport for the TQD system, including a discussion of the charge quadruple point which involves three electrons. We also discussed the charge quadruple point at the non-linear transport region for the estimation of the charging energies. From the experimental data, we can roughly estimate the charging energies, which are consistent with the calculated values from the simulation. In addition, we discussed the several kinds of higher order tunneling processes, including the comparison between the two successive second order tunneling processes and the third order tunneling processes. The TQD system provides an essential prerequisite for the future implementation of silicon-based quantum information processing devices because of its high functionality.
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Appendix A. Charging energies for the multiple QDs

The charging energies for the multiple QDs can be derived from the electrostatic energy of the system [29]. In a $N$ conductors system, the node $j$ is connected to the other nodes with the capacitances $c_{ij}$ and the stored charge $Q_j$ is given by:

$$Q_j = \sum_k c_{jk} (V_j - V_k),$$

where $V_j$ is the electrostatic potential of the node $j$. This equation can be expressed in the matrix form

$$\vec{Q} = \mathbf{C} \vec{V}.$$  

Here $\mathbf{C}$ is the capacitance matrix, which consists of the diagonal matrix elements $c_{ii} = \sum_{k \neq j} c_{ik}$ and the off-diagonal matrix elements $c_{ij} = -c_{ji}$. To consider the voltage sources in the network, the matrix equation is separated as

$$\begin{pmatrix} \vec{Q}_N \\ \vec{Q}_S \end{pmatrix} = \begin{pmatrix} \mathbf{C}_{NN} & \mathbf{C}_{NS} \\ \mathbf{C}_{SN} & \mathbf{C}_{SS} \end{pmatrix} \begin{pmatrix} \vec{V}_N \\ \vec{V}_S \end{pmatrix},$$

where $\vec{Q}_{NS}$ and $\vec{V}_{NS}$ are the charges and the voltages on the charge nodes (the voltage sources), respectively, and $\mathbf{C}_{NN}$, $\mathbf{C}_{NS}$, $\mathbf{C}_{SN}$, and $\mathbf{C}_{SS}$ are the four capacitance sub-matrices derived from $\mathbf{C}$. In this expression, the voltage sources are treated as nodes with large capacitances to the ground. Although many voltage sources are set in our systems, it is not necessary to obtain $\mathbf{C}_{NS}$ and $\mathbf{C}_{SN}$ because the values of the voltage sources are already known. Therefore, from the voltages on the charge nodes

$$\vec{V}_N = -\mathbf{C}_{NN}^{-1} \left( \vec{Q}_N - \mathbf{C}_{NS} \vec{V}_S \right),$$

the electrostatic energy of the charge nodes can be determined as follows:

$$E = \frac{1}{2} \vec{Q}_N \cdot \mathbf{C}_{NN} \vec{Q}_N.$$  

From the equivalent circuit for our multiple QDs (Fig. 1b), the charges on QD1 ($i = 1, 2, 3, a$), which are expressed such that $Q_i = -N_0 |e|$, where $N_0$ is the number of electrons on QD1 and $-|e|$ is the electron charge, are given by

$$\begin{align*}
Q_1 &= C_1 (V_1 - V_{11}) + C_2 (V_1 - V_2) + C_3 (V_1 - V_3) + C_4 (V_1 - V_4) + C_{1p} V_1, \\
Q_2 &= C_2 (V_2 - V_{12}) + C_3 (V_2 - V_4) + C_4 (V_2 - V_3) + C_{2p} V_2, \\
Q_3 &= C_3 (V_3 - V_{13}) + C_4 (V_3 - V_2) + C_1 (V_3 - V_1) + C_{3p} V_3, \\
Q_a &= C_4 (V_a - V_4) + C_1 (V_a - V_1) + C_2 (V_a - V_2) + C_{1a} (V_a - V_{1a}) + C_{2a} (V_a - V_{2a}).
\end{align*}$$

These equations can be written in the matrix form as

$$\begin{pmatrix}
Q_1 \\
Q_2 \\
Q_3 \\
Q_a
\end{pmatrix} = \begin{pmatrix}
C_{QQ1} & 0 & -C_2 & -C_3 \\
0 & C_{QQ2} & -C_2 & -C_3 \\
-C_2 & -C_3 & C_{QQ3} & -C_3 \\
-C_3 & -C_3 & -C_3 & C_{QQa}
\end{pmatrix} \begin{pmatrix}
V_1 \\
V_2 \\
V_3 \\
V_a
\end{pmatrix},$$

where $C_{QQ1} = C_1 + C_2 + C_3 + C_4 + C_{1p}$, $C_{QQ2} = C_2 + C_3 + C_4 + C_{2p}$, $C_{QQ3} = C_3 + C_4 + C_{3p}$, and $C_{QQa} = C_4 + C_{1a} + C_{2a}$.

From the matrix equation Eq. (10), we obtain $\vec{V}_S$. As a result, the electrostatic energy $U(N_1, N_2, N_3, N_a)$ can be calculated with Eq. (5).

The electrochemical potentials $\mu_i$ which are defined as the energy required to add the $N_i$th electron to QDI, are given by

$$\begin{align*}
\mu_1(N_1, N_2, N_3, N_a) &= U(N_1, N_2, N_3, N_a) - U(N_1 - 1, N_2, N_3, N_a), \\
\mu_2(N_1, N_2, N_3, N_a) &= U(N_1, N_2, N_3, N_a) - U(N_1, N_2 - 1, N_3, N_a), \\
\mu_3(N_1, N_2, N_3, N_a) &= U(N_1, N_2, N_3, N_a) - U(N_1, N_2 - 1, N_3 - 1, N_a), \\
\mu_a(N_1, N_2, N_3, N_a) &= U(N_1, N_2, N_3, N_a) - U(N_1, N_2, N_3 - 1, N_a).\end{align*}$$

The charging energies (the addition energies) for the QD are

$$\begin{align*}
E_{q11} &= \mu_1(N_1 + 1, N_2, N_3, N_a) - \mu_1(N_1, N_2, N_3, N_a), \\
E_{q22} &= \mu_2(N_1, N_2 + 1, N_3, N_a) - \mu_2(N_1, N_2, N_3, N_a), \\
E_{q33} &= \mu_3(N_1, N_2, N_3 + 1, N_a) - \mu_3(N_1, N_2, N_3, N_a), \\
E_{qaa} &= \mu_a(N_1, N_2, N_3, N_a + 1) - \mu_a(N_1, N_2, N_3, N_a),
\end{align*}$$

giving

$$\begin{align*}
E_{q11} &= |e|^2 / (2C_{23} C_4 C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a}), \\
E_{q22} &= |e|^2 / (2C_2 C_3 C_4 + C_{4a} C_{3a} + C_{4a} C_{3a} + C_{4a} C_{3a}), \\
E_{q33} &= |e|^2 / (C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a}), \\
E_{qaa} &= |e|^2 / (C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a}).
\end{align*}$$

In addition, the electrostatic coupling energies between QD1(2) and QD3(4) $|E_{12}|$ are determined by

$$\begin{align*}
E_{12} &= \mu_1(N_1, N_2, N_3, N_a) - \mu_1(N_1, N_2, N_3, N_a) \\
&= |e|^2 / (C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a} + C_{2a} C_{3a}),
\end{align*}$$

References


