Room-Temperature Charge Stability Modulated by Quantum Effects in a Nanoscale Silicon Island

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ABSTRACT: We report on transport measurement performed on a room-temperature-operating ultrasmall Coulomb blockade devices with a silicon island of sub-5 nm. The charge stability at 300K exhibits a substantial change in slopes and diagonal size of each successive Coulomb diamond, but remarkably its main feature persists even at low temperature down to 5.3K except for additional Coulomb peak splitting. This key feature of charge stability with additional fine structures of Coulomb peaks are successfully modeled by including the interplay between Coulomb interaction, valley splitting, and strong quantum confinement, which leads to several low-energy many-body excited states for each dot occupancy. These excited states become enhanced in the sub-5 nm ultrasmall scale and persist even at 300K in the form of cluster, leading to the substantial modulation of charge stability.

KEYWORDS: Single-electron transport, nanoscale silicon dot, Coulomb blockade, room-temperature charge stability, quantum effects

An ultrasmall Coulomb blockade device can be regarded as a mesoscopic artificial atom system. Tunneling through it can provide a rich experimental environment for studying quantum transport phenomena.1 Previously, these quantum effects have been investigated using relatively large devices at low temperatures, where they give rise to additional fine structures on the Coulomb oscillations.2–9 However, as temperature increases up to 300 K such fine structures observed at low temperature normally vanish together with Coulomb peaks themselves because of the weak Coulomb charging energy due to the relatively large dot size. However, as the dot size is reduced below 5 nm, the very small number of electrons on the dot is expected to ensure that electron-electron interactions with Pauli spin exclusion strongly influence the electron transport characteristics. Here, we report on an extensive transport measurement performed on a room-temperature-operating Coulomb blockade device with an ultrasmall silicon island of sub-5-nm size. Transport data exhibit the striking feature that the main room-temperature characteristics of the Coulomb peaks persist even at ultralow temperature down to 5.3 K. Substantial change in slopes and diagonal size of the room-temperature Coulomb diamond and bias-dependent peak splitting must reflect low-energy many-body excited states associated with total spin for each dot occupancy N. This quantum effects become enhanced in our ultrasmall Coulomb island and persist even at room temperature, leading to the substantial modulation of the charge stability for finite bias window.

The Coulomb blockade device used in transport measurement has been fabricated by scaling a state-of-the-art finFET structure down to an ultimate form, by using deep-trench and subsequent oxidation-induced strain, which can be used to form a single electron transistor (SET) with a Coulomb island of sub-5-nm size.10 Figure 1a shows a scanning electron microscopy (SEM) image of the SET device whose active channel is detailed as a schematic 3-D layout (Figure 1b). Note how the top Si nanowire, exposed by the nanogap between the source and the drain, is further etched down to 30 nm in depth by dry etching and gate oxidation. This key process, different from the conventional nanoscale finFET, enables a Coulomb island to be formed with nearly identical tunnel barriers in a self-aligned manner. Moreover, by wrapping a fin-gate almost completely around the Coulomb island, good control of the local electron potential is maintained. Figure 1b also shows a transmission electron microscopy (TEM) cross-sectional image of the etched top-Si nanowire along the channel, exhibiting an island diameter of ~2 nm. Good control over the island size was achieved through the

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oxidation process. Figure 1c shows the drain current measured at 300 K as a function of the fin-gate voltage $V_G$ for SET, which is compared with that of the conventional finFET that was fabricated by a similar process in the same wafer, but without the deep-trench process on the silicon wire channel. Note that the first Coulomb peak of the SET appears just above the threshold voltage of the finFET that occurs at $V_G \sim 2.5$ V for drain bias voltages up to 50 mV. The onset of the Coulomb oscillations was not detected below the threshold voltage, indicating the first peak to be associated with the first electron tunneling.

Figure 2 shows temperature dependence of the $I-V$ characteristics of the SET measured for various temperatures down to 5.3 K for a bias of 50 mV. As seen in Figure 2a, the main feature of the Coulomb oscillations of 300 K persists even at low temperature down to 5.3 K, except for additional splitting observed in each Coulomb peak. We point out that this temperature-dependent feature is quite new and remarkable because the peak splittings so far observed at low temperatures have been reported to vanish together with Coulomb peaks themselves with increasing temperature. This strongly indicates more evidence that the Coulomb island of our device is a quite small well-defined single dot (of sub-5-nm size), providing its charging energy to be large enough to get over the room-temperature thermal energy. If the main four Coulomb peaks were due to some multiple defects at the Si/SiO$_2$ interface, some of the peaks should be randomly created or disappear depending on their thermal activation energies as temperature changes. Note also that the magnitude of the Coulomb peak decreases for low temperatures, as seen in Figure 2e which shows the temperature-dependent magnitude of the third Coulomb peak for each bias up to 50 mV. This can be attributed to the possible decrease of the carrier concentration (or, carrier freeze-out) in the S/D wires as temperature decreases down to 5.3 K.

Charge stability plots (displayed for down to 100 K) are seen in panels b, c, and d of Figure 2, respectively, where successive Coulomb diamonds are clearly seen. Each diamond corresponds to a stable charge configuration state with fixed electron occupancy $N$. Coulomb peak splittings are seen for diamonds of $N = 3$ and 4 in the charge stability even at 100 K. Note that Coulomb diamonds for each $N$ are very symmetric with respect to the positive and negative drain biases, strongly indicating that a single ultrasmall Coulomb island is formed at the middle point of the channel and that its tunnel barriers with source and drain are nearly identical. This rules out a possibility that the observed Coulomb oscillations may be related to the possible dopant or defect which must be randomly formed. The charge stability data also show that as the gate voltage is made less positive, the slope of each Coulomb diamond steeply increases, and the Coulomb diamond (for $V_G < 3$ V) does not close. This feature is consistent with the lack of any Coulomb peaks below the threshold (Figure 1c), indicating that the island is unpopulated by electrons for $V_G \leq 3$ V. To further ensure the assignment of the dot occupancy, a charge sensing device by means of a separated circuit (such as an additional SET or quantum point contact) must be installed next to the Coulomb island. In this case, however, an additional sensing bridge gate should be designed to be located very close to the dot to maximize mutual charge coupling. This, without doubt, yields a substantial increase of the total capacitance of the Coulomb island, leading to the SET operating only at ultralow temperatures. Any room temperature features will vanish. This is why most of experiments based on these devices including charge sensor have shown Coulomb oscillation behavior only at dilution refrigerator temperatures of $10−100$ mK. We, therefore, addressed the assignment of the dot occupancy by somewhat indirect ways (mentioned above) without using an additional charge sensor.

It is noted that substantial change in slopes and diagonal size of each successive diamond is observed, implying that the charging energy is not constant over the gate voltage range studied. This behavior could be accounted for by strong interplay of the Coulomb interaction and additional quantum effects associated with very low electron number on the island. The size of the island and its Coulomb charging energy can be roughly estimated using the first diamond associated with the lowest dot occupancy, $N = 1$, that is determined mainly by the Coulomb charging energy. Values for the gate and junction capacitances can be directly obtained from the Coulomb peak spacing $\Delta V_G$ and the slopes of the first diamond. This yields the total capacitance $C_0 \sim 0.42$ aF, which corresponds to a 1.94 nm diameter spherical silicon dot, in good agreement with the TEM image in Figure 1b. The charging energy of a dot of this size is thus $e^2/C_0 \sim 0.38$ eV, which is more than 1 order magnitude larger than the thermal energy at 300 K.
The fine structure with decreasing temperature must reflect low energy excited levels associated with each dot occupancy $N_i$ and can be explored in more detail with increasing bias window. Panels a and b of Figure 3 are charge stability data at 5.3 K. They illustrate the fine structure of the bias dependence of the Coulomb oscillations, showing typical behavior of increasing splitting with bias window. For more clarity, we present panels c and d of Figure 3, reproducing $I_{dc}$ vs. $V_g$ for some specific bias voltages in the charge stability data. As seen in Figure 3a with Figure 3c, when bias voltage increases up to 100 mV, the first peak starts to split into two subpeaks and persists even at high bias, while the second peak splits into four subpeaks. Note that
for the first main peak, the valley between two subpeaks is raised up with bias voltage, indicating the increase in tunneling current as bias window becomes wide. This is not due to a peak broadening effect because the heating energy by increasing bias is only about \( \sim 0.05 \, \text{nW} \), negligible compared to the thermal energy of 5.3 K. Similarly, Figure 3b with Figure 3d illustrate that the number of splittings of the third Coulomb peaks rapidly increase from 3 to more than 12, while that of the fourth peak increases from 2 to more than 8. This strong bias dependence of peak splitting demonstrates the evident transition of the transport behavior of our device from linear to nonlinear transport regime where single-electron tunneling can be made through many excited levels lying within the bias window.\(^{17,18}\) It is thus important to know the low energy level spectrum associated with each dot occupancy \( N \).

Figure 3. Charge stability plot at 5.3 K and specific bias dependence of each main Coulomb peak. (a, b) Charge stability plot at 5.3 K, showing typical behavior of increasing splitting with bias window. (c, d) \( I-V \) characteristics for some specific bias voltages, which are reproduced from the charge stability data. Strong bias dependence on peak splitting is clearly seen, which can be accounted for by the nonlinear transport made through many excited levels associated with each dot occupancy \( N \).
In such a wire, valley splitting lifts the 2-fold and 4-fold degeneracies into two, and the energy levels of $\Gamma$ valleys are lower than those of off-$\Gamma$ valleys. In Si valley splitting is a main source of spin decoherence. It has been observed even in zero external electric field in strongly confined nanostructures and could be further enhanced by the application of bias, strain, or magnetic field. In the presence of a confinement potential along the wire, the energy levels of valleys are quantized, as shown schematically in inset (a) of Figure 4. With the inclusion of the valley splitting, the four lowest energy levels of the dot are $\varepsilon_1 = E_{v2,1}$, $\varepsilon_2 = E_{v2,1} + \Delta$, $\varepsilon_3 = E$, and $\varepsilon_4 = E + \Delta'$, where $E_{v2,1}$ is the lowest energy quantized with valley splitting $\Delta$ and $E$ is the second lowest energy quantized with valley splitting $\Delta'$. While $E_{v2,1}$ and $E_{v2,1} + \Delta$ originate from the $\Gamma$ valley, the energy $E$ may originate from either $\Gamma$ or off-$\Gamma$ valleys.

On the basis of the above information on single electron levels, we model the many-body Hamiltonian of the dot by

$$H = \sum_i \varepsilon_i n_i + \sum_{i<j} V_{ij} n_i n_j + \sum_i U_i n_i n_i - \sum_{i<j} J_{ij} \left( \vec{S}_i \cdot \vec{S}_j + \frac{1}{4} n_i n_j \right)$$

Here the label $i = 1, 2, 3, 4$ denotes the four single electron states. For each single electron level $i$ we define the quantities $\vec{S}_i$, $n_{i\sigma}$, $n_i$, and $U_i$ as the spin operator, number operator of electrons with spin $\sigma$, number operator of occupied electrons, and the intralevel Coulomb repulsion, respectively. The Coulomb repulsion (exchange) energy between an electron in the $i$th and an electron in $j$th levels is $V_{ij}(J_{ij})$. Each many-body eigenstate can be represented by a ket state $|\{n_i\}, S, S_z\rangle$. Level occupation numbers $\{n_i\}$ and the total spin quantum number $S$ of some of the lowest energy many-body states are analyzed and displayed in Figure 5. (Note that since the Hamiltonian is spin rotationally invariant, eigenvalues are independent of the $z$-component total spin $S_z$.)

In this classification of eigenstates, it is useful to exploit the fact that when the $i$th level is doubly occupied, its spin state is necessarily a singlet state with $S_i = 0$. In addition, according to the quantum rules of spin addition, the total spin state of three electrons can be constructed by adding spin 0 and 1/2 or by subtracting spin 1/2 from 1. This implies that, when adding three electron spins, there are two different spin wave functions for the same $S = 1/2$ and $\{n_i\}$. Due to many-body exchange interactions, states with the same $\{n_i\}$ but with different $S$ do not have the same energy. For example, for $N = 2$ the $S = 0$ singlet and $S = 1$ triplet states are split; see Figure 5b. From our observed data,
Figure 3c, the magnitude of the exchange interaction is estimated to be about 0.01 eV. Using these rules, we find 2, 4, 12, and 8 lowest energy states for \( N = 1, 2, 3, \) and 4, respectively, that can be formed from the energies \( \varepsilon_1, \varepsilon_2, \varepsilon_3, \) and \( \varepsilon_4; \) see Figure 5. We stress that these numbers are independent of model parameters. For a given \( N, \) the estimate of energies shows that there is an energy gap to the next excited states from the group of lowest energy states mentioned above.

For \( N = 1 \) and 2, the theoretical number of lowest energy levels agrees with experimentally observed peak values of 2 and 4. According to our model for \( N = 2 \) there are three singlet and one triplet levels; see Figure 5b. The three singlet states have different energies because their occupation number configurations \( \{ n_i \} \) are different. For \( N = 3 \) we displayed 12 lowest energy states in Figure 5c. In near agreement with this value, the observed number low energy excited states is about 12–14 (when small noiselike peaks are included, there are 14 peaks). For \( N = 4, \) some of the lowest excited energy states have the same \( \{ n_i \} \) but different spin values \( S = 0 \) and 1; see Figure 5d. Their energies are again different because these states have different spin wave functions and, therefore, have different exchange energies. For \( N = 4, \) the observed number of excited states is between 8 and 10, which is close to predicted value of 8; see Figure 5d. Since there may be other single electron energy levels close to \( \varepsilon_3 \) and \( \varepsilon_4 \), different many-body excited energies may be present near those 12 and 8 states shown in panels c and d of Figure 5. Moreover, the effect of quantum fluctuations of occupation numbers, which is absent in Hartree–Fock approximations, may give rise to additional excited states.\(^{14}\) These factors suggest a possible explanation for why more than 12 and 8 peaks may have been observed for \( N = 3 \) and 4, respectively, when small noiselike peaks are included.

The Coulomb interaction energy between two electrons in the level 1 is \( U_1 \approx \varepsilon^2/2R \approx 0.38 \text{ eV}, \) where \( \varepsilon \) and \( R \) are the dielectric constant and the radius of the dot. The energy separation between different Coulomb peaks can be fitted by choosing two parameters \( V_{13} \) and \( \varepsilon_3 - \varepsilon_2 \); judiciously: \( V_{13} \approx 0.3 \text{ eV} \) is the Coulomb interaction energy between two electrons in the levels 1 and 3 and \( \varepsilon_3 - \varepsilon_1 \approx 0.3 \text{ eV} \) is the energy separation between them. The fit value of \( V_{13} \) is reasonable because the interlevel Coulomb interaction is comparable to the intralevel \( U_0 \), but must be smaller than it. The fit value of the quantum confinement of \( \varepsilon_3 - \varepsilon_1 \) is also in the range of the expected value for our dot size of 2 nm by theoretical calculations.\(^{15,20}\) Using these values, we find that the addition charging energies of \( N = 1 \to 2, 2 \to 3, \) and \( 3 \to 4 \) are approximately \( U_1 \approx 0.38 \text{ eV}, \) \( 2V_{13} + \varepsilon_4 - \varepsilon_2 \approx 0.9 \text{ eV}, \) and \( V_{13} + 2(U_1 - V_{13}) \approx 0.46 \text{ eV}, \) which are illustrated in Figure 4.

Using the energy coupling factor, defined by \( \alpha_C = C_C/C_Z \approx 0.22, \) the corresponding observed values 0.37, 0.84, and 0.50 eV (see inset (b) in Figure 4) are in the same range. For \( N = 1 \) the observed Coulomb oscillations exhibit splitting of the first peak is due to the valley splitting; see Figure 5a. The measured value of valley splitting \( \Delta \) is 16 meV, whose order of magnitude is consistent with recently reported theoretical values of sub-3-nm Si nanostructures.\(^{15,20}\) Note that its value is much smaller than Coulomb charging and quantum confinement energies. These approximate agreements between experimental and theoretical values suggest that our model can account consistently for several features of excited states in the ultrasmall Si dot formed along the (110) direction.

In summary, we report on extensive transport measurements performed on the room-temperature-operating ultrasmall silicon SET devices with a Coulomb island of sub-5-nm size. The room-temperature feature of \( I-V_g \) persists even at low temperature down to 5.3 K, where additional fine structures of Coulomb peaks appear. The unusual energy separation between Coulomb diamonds and the fine splitting of each Coulomb peak is accounted for by including quantum many-body interactions, leading to the substantial modulation of Coulomb diamonds at 300 K. It further supports the reliability in our CMOS-compatible implementation of the ultrasmall SET operating at room temperature.

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The vertical indexing of the current grayscale in inset (b) of Figure 4 is incorrect in the original article; 0 < V_G < 12 V should be corrected to 6 < V_G < 12 V. In addition, the value of spin S = 1/2 in the legend of Figure 5b is incorrect; S = 1/2 should be corrected to S = 0. Finally, electronic occupation configurations of lower panels in Figure 5c are incorrect. Their correct versions are reproduced below. No changes to the figure captions and their respective text are required. None of the results and conclusions of the article are affected by these corrections.

Figure 4. inset (b): The calculated addition charging energies, approximately U_1 ≈ 0.38 eV, 2V_{13} – Ω_a ≈ 0.9 eV, and V_{13} + 2(U_1 – V_{13}) ≈ 0.46 eV (for N = 1 → 2, 2 → 3, and 3 → 4, respectively) are denoted by arrows in inset (b), which are in the same range as those of the charge stability data observed at 300 K.

Figure 5. (b and c) Electronic occupation configurations illustrating the number of lowest energy states for (b) N = 2 and (c) N = 3. Note that due to many-body exchange interactions states with the same \{n_i\} but with different S do not have the same energy.

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