Density matrix to quantum master equation (QME) model for arrays of Coulomb coupled quantum dots in the sequential tunneling regime

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Coulomb coupled quantum dot arrays with staircase ground state configuration have been proposed in literature for enhancing heat-harvesting and refrigeration performance [1–7]. Due to their mutual Coulomb interaction, a performance analysis of such systems remains complicated and necessitates consideration of microscopic physics using density matrix formulation. However the path of transport analysis starting from the system Hamiltonian to density matrix formulation is complicated and lacks the simplicity and intuitive aspect of sequential electron transport conveyed by the quantum master equation (QME) approach. In this paper, starting from the system Hamiltonian and employing the density matrix formulation, I derive the QME of a system of three quantum dots, two of which are electro-statically coupled. The framework formulated in this paper can be further extended to derive QME of systems with higher number of Coulomb coupled quantum dots.

Recently with the progress of fabrication technology, a lot of effort has been geared towards nanoscale solid state quantum dot devices which, due to their discrete energy spectrum, form ideal beds of quantum computation, heat harvesting and refrigeration, etc. Due to their small size, quantum dots that are separated in space often exhibit capacitive charge coupling which offers an attractive coupling between spatially separated quantum dots and spin. This phenomenon of electrostatic or charge-based coupling between spatially separated quantum dots is known as Coulomb coupling, which gives rise to a well known phenomena known as Coulomb blockade. Quantum dots that are spatially separated, may be bridged to obtain strong electrostatic coupling between them [8, 9]. In addition, the bridge may be fabricated between two desired dots to radically increase their mutual electrostatic coupling, without affecting the other dots [8–10]. The effect of Coulomb coupling on the current spectra as well as methods to enhance Coulomb coupling between quantum dots has been well explored via experiments [8–10]. However, a theoretical analysis of such Coulomb coupled dots is complicated and requires a full analysis of the microscopic physics starting from the system Hamiltonian. This is particularly true when a few or all of the dots in the system are Coulomb coupled to one or more adjacent dots. In addition, the analysis approach of such set-ups, starting from the system Hamiltonian, often masks the intuitive aspect of sequential transport physics, which is generally beneficial to the experimental community to further refine device characteristics.

In this paper, starting from the microscopic physics, I methodically derive the quantum master equations (QME) of Coulomb-coupled dot arrays in the sequential tunneling limit. In addition to being a simpler framework, the QME approach [11–16] bears the intuitive aspect of sequential electron transport and have been extensively used in literature to analyze the properties of single electron transistors and quantum dots in the Coulomb blockade regime. However, to the best of my knowledge, such approach has not yet been used to analyze arrays of quantum dots in which one or more dots may be Coulomb coupled with some others. Here, starting from the system Hamiltonian and the density matrix formulation, I derive the quantum master equations of the Coulomb coupled system demonstrated in Fig. 1. Such type of Coulomb coupled systems have already been proposed for the optimal non-local refrigeration [17]. The system consists of three dots $S_1$, $S_2$, and $G_1$ which are electrically coupled to the reservoirs $L$, $R$ and $G$ respectively. $S_1$ and $S_2$ are tunnel coupled to each other, while $G_1$ is capacitively coupled to $S_1$. The ground states of $S_1$ and $S_2$ form a stair-case configuration with $\varepsilon_2^s \approx \varepsilon_1^s + U$.

To derive the quantum master equations of the system, I start from the device Hamiltonian. The total electrostatic energy $E_C$ of the system consisting of three dots can be given by:

$$
E_C(n_{S_1}, n_{G_1}, n_{S_2}) = \sum_{x \in \{S_1, G_1, S_2\}} E_x^{\text{self}} \left(n_x - \frac{V_x C_x^g}{q}\right)^2 + \sum_{\{x_1, x_2\} \in \{S_1, G_1, S_2\}} \sum_{\{n_{x_1}, n_{x_2}\}} E^{x_1, x_2}_{x_1, x_2} \left(n_{x_1} - \frac{V_x C_x^g}{q}\right) \left(n_{x_2} - \frac{V_x C_x^g}{q}\right)
$$

where $n_x$ is the electron number, $C_x^g$ is the gate-capacitance and $E_x^{\text{self}} = \frac{q^2}{C_x^g}$ is the electrostatic energy due to self-capacitance $C_x^g$ of quantum dot ‘$x$’ with its surrounding terminals. $E^{x_1, x_2}_{x_1, x_2}$ is the electrostatic en-
energy arising out of interdot Coulomb interaction between two different quantum dots that are separated in space. Here, a minimal physics based model is used to derive the rate equations. I assume that the electrostatic energy due self-capacitance is much greater than the average thermal voltage $kT/q$ or the applied bias voltage $V$, that is $E_{self}^c = \frac{q^2}{C_G} \gg (kT, qV)$, such that electron occupation probability or transfer rate via the Coulomb blocked energy level, due to self-capacitance, is negligibly small. The analysis of the entire system of dots may hence be approximated by limiting the maximum number of electrons in each dot to one. Thus the analysis of the entire system may be limited to eight multi-electron levels, which I denote by the electron occupation number in the ground state of each quantum dot. Hence, a possible state of interest in the system may be denoted as $|n_{S_1}, n_{G_1}, n_{S_2}\rangle = |n_{S_1}\rangle \otimes |n_{G_1}\rangle \otimes |n_{S_2}\rangle$, where $(n_{S_1}, n_{G_1}, n_{S_2}) \in (0, 1)$. To proceed further from here, with a slight abuse of notation, I simply denote the eight multi-electron states as $|0, 0, 0\rangle \rightarrow |0\rangle$, $|0, 0, 1\rangle \rightarrow |1\rangle$, $|0, 1, 0\rangle \rightarrow |2\rangle$, $|0, 1, 1\rangle \rightarrow |3\rangle$, $|1, 0, 0\rangle \rightarrow |4\rangle$, $|1, 0, 1\rangle \rightarrow |5\rangle$, $|1, 1, 0\rangle \rightarrow |6\rangle$, and $|1, 1, 1\rangle \rightarrow |7\rangle$. The Hamiltonian of the system consisting of these three quantum dots without any reservoir coupling may be written as:

$$H = \sum_{\beta} \epsilon_{\beta} |\beta\rangle \langle \beta| + \sum_{\{3\}} |3\rangle \langle 6| + |1\rangle \langle 4|$$

$$+ U\{6\} \langle 6| + |7\rangle \langle 7| \rangle + h.c.,$$

(1)

where $U = E_{S_1,G_1}^c$ is the Coulomb coupling energy between the dots $S_1$ and $G_1$ in Fig. 1 and $t$ is the electron hopping amplitude between the adjacent dots $S_1$ and $S_2$. Under the assumption of weak reservoir to system coupling and small hopping amplitude $t$, the temporal dynamics of the system density matrix can be evaluated by the partial trace over the density matrix of the entire set-up of the reservoirs and the dots [11–16]. Taking the partial trace of the combined density matrix over the reservoir states, the diagonal and the non-diagonal elements of the density matrix $\rho$ of the system of quantum dots may be given as a set of modified Liouville equations [11–16]:

$$\frac{\partial \rho_{\eta\eta}}{\partial t} = -i[H, \rho_{\eta\eta}] - \sum_{\nu} \Gamma_{\eta\nu} \rho_{\eta\nu} + \sum_{\delta} \Gamma_{\delta\eta} \rho_{\delta\delta}$$

$$\frac{\partial \rho_{\eta\beta}}{\partial t} = -i[H, \rho_{\eta\beta}] - \frac{1}{2} \sum_{\nu} (\Gamma_{\eta\nu} + \Gamma_{\beta\nu}) \rho_{\eta\beta},$$

(2)

where $[A, B]$ denotes the commutator of $A$ and $B$ and $\rho_{\eta\beta} = \langle \eta | \rho | \beta \rangle$. The elements $\rho_{\eta\eta}$ and $\rho_{\eta\beta}$ in the above equation denote any diagonal and non-diagonal element of the system density matrix respectively. The parameters $\Gamma_{ij}$ take into account the transition between system states due to electronic tunneling between the system and the reservoirs and are only non-zero when the system can transit from state $|i\rangle$ to $|j\rangle$ (or vice-versa) due to tunneling of electrons in and out of the system from the reservoir. In our derivation, assuming a statistical quasi-equilibrium distribution of electrons inside the reservoirs, we can express $\Gamma_{ij}$ as:

$$\Gamma_{ij} = \gamma_c f_{\lambda}(\epsilon_i - \epsilon_j).$$

(3)

$f_{\lambda}(\epsilon)$ being occupancy probability of the corresponding reservoir $\lambda$ at energy $\epsilon$ and $\epsilon_{(i,j)}$ is the total electronic energy of the system in the state $i \langle j \rangle$. To derive the quantum master equations for the entire system, it is essential to derive the inter-dot tunneling rates. For the particular system schematic demonstrated in Fig. 1, interdot tunneling changes the system states as: $|4\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |6\rangle$. Taking the time derivative of the density matrix to be zero in steady state, I use the second equation of (2), to get,

$$\rho_{4,1}^t = \rho_{4,1}^4 = \frac{\rho_{4,4} - \rho_{1,1}}{\epsilon_4 - \epsilon_1 - i\Omega_{1,4}}$$

$$\rho_{6,3}^t = \rho_{5,6}^5 = \frac{\rho_{6,6} - \rho_{3,3}}{\epsilon_6 - \epsilon_3 - i\Omega_{3,6}},$$

(4)

(5)

where $\Omega_{i,j}$ is the combination of all the reservoir-to-system tunneling events (or vice-versa) leading to the decay of the states $i$ and $j$. For the system under consideration, $\Omega_{4,1}$ and $\Omega_{6,3}$ can be given by:

$$\Omega_{4,1} = \Gamma_{4,0} + \Gamma_{4,6} + \Gamma_{4,5} + \Gamma_{1,0} + \Gamma_{1,6} + \Gamma_{1,3}$$

$$\Omega_{6,3} = \Gamma_{6,4} + \Gamma_{6,2} + \Gamma_{6,7} + \Gamma_{3,1} + \Gamma_{3,2} + \Gamma_{3,7}$$

(6)

The time derivative of diagonal density matrix elements $\rho_{n,n}$ and $\rho_{3,3}$ can be written as (using the first equation of (2)):

$$\dot{\rho}_{6,6} = i(t(\rho_{6,3} - \rho_{3,6}) - (\Gamma_{4,6} + \Gamma_{6,2} + \Gamma_{6,7}) \rho_{6,6}$$

$$+ \Gamma_{4,6} \rho_{4,4} + \Gamma_{6,2} \rho_{2,2} + \Gamma_{7,6} \rho_{7,7})$$

$$\dot{\rho}_{4,4} = i(t(\rho_{4,1} - \rho_{1,4}) - (\Gamma_{4,0} + \Gamma_{4,6} + \Gamma_{4,5}) \rho_{4,4}$$

$$+ \Gamma_{0,4} \rho_{0,0} + \Gamma_{4,6} \rho_{6,6} + \Gamma_{4,5} \rho_{5,5},$$

(7)
I next substitute, in Eq. (7), the expressions for \( \rho_{6,3} \), \( \rho_{3,6} \), \( \rho_{4,1} \) and \( \rho_{1,4} \) from Eq. (4), to get the time evolution of the density matrix elements of \( \rho_{6,6} \) and \( \rho_{4,4} \) as:

\[
\dot{\rho}_6 = \dot{\rho}_{6,6} = \sum_{\alpha} (-\Gamma_{6,\alpha} p_6 + \Gamma_{\alpha,6} p_\alpha) - \tau_{6,3} p_6 + \tau_{3,6} p_3
\]

\[
\dot{\rho}_4 = \dot{\rho}_{4,4} = \sum_{\alpha} (-\Gamma_{4,\alpha} p_4 + \Gamma_{\alpha,4} p_\alpha) - \tau_{4,1} p_4 + \tau_{1,4} p_1,
\]

where \( p_\eta = \rho_{\eta,\eta} \) and

\[
\tau_{6,3} = \tau_{3,6} = \frac{\Omega_{6,3}}{\epsilon_6 - \epsilon_3} = \frac{\Omega_{3,6}}{\epsilon_3 - \epsilon_6},
\]

\[
\tau_{4,1} = \tau_{1,4} = \frac{\Omega_{4,1}}{\epsilon_4 - \epsilon_1} = \frac{\Omega_{1,4}}{\epsilon_1 - \epsilon_4}.
\]  

(8)

In the set of Eqs. (8), \( \tau_{4,1} \) and \( \tau_{6,3} \) correspond to the interdot tunneling rates when the number of electrons in dot \( G_1 \) is 0 and 1 respectively. When \( U >> |\Omega_{4,1}| \), by an appropriate choice of \( \epsilon_4 = \epsilon_1 \) and \( \epsilon_6 = \epsilon_3 \), such that, \( \epsilon_6 = \epsilon_0 + \epsilon_4 + U = \epsilon_0 + \epsilon_3 + U, \) that is by making \( \epsilon_2^s = \epsilon_1^s + U \), we may arrive at a condition where \( \tau_{6,3} >> \tau_{4,1} \). Such a condition implies that the tunneling probability between the dots is negligible in the absence of an electron in \( G_1 \), which is the combined impact of the capacitive coupling between \( S_1 - G_1 \) and staircase ground-state configuration of \( S_1 - S_2 \).

Next, I proceed towards deriving the QME of the system demonstrated in Fig. 1. Since, the electronic transport and ground states in \( S_1 \) and \( G_1 \) are mutually coupled, I treat the pair of dots \( S_1 \) and \( G_1 \) as a sub-system \((\varsigma_1)\), \( S_2 \) being the complementary sub-system \((\varsigma_2)\) of the entire system consisting of three dots. I assume that \( U >> |\Omega_{4,1}| \) and \( \epsilon_2^s = \epsilon_1^s + U \), such that \( \tau_{4,1} \ll \tau_{6,3} \). For all practical phenomena relating to electron transport, it can hence be assumed that \( \tau_{4,1} \approx 0 \). The state probability \( \varsigma_1 \) is denoted by \( \rho_{i,j} \), \( i \) and \( j \) being the number of electrons in the dot \( S_1 \) and \( G_1 \) respectively. \( \rho_{k,0} \), on the other hand, denotes the probability of occupancy of the dot \( S_2 \) in the sub-system \( \varsigma_2 \). Note that breaking down the entire system into two sub-systems in this way is possible only in the limit of weak tunnel and Coulomb coupling between the two sub-systems, as such the state of one sub-system remains unaffected by a change in state of the other sub-system. In such a limit, the diagonal elements of the density matrix can be written as:

\[
\rho_{0,0} = \rho_{0,0}^S, \quad \rho_{1,1} = \rho_{1,1}^S \quad \text{and} \quad \rho_{2,2} = \rho_{2,2}^S, \quad \rho_{3,3} = \rho_{3,3}^S, \quad \rho_{4,4} = \rho_{4,4}^S, \quad \rho_{5,5} = \rho_{5,5}^S, \quad \rho_{6,6} = \rho_{6,6}^S.
\]

The QME for the sub-system \( \varsigma_1 \) and \( \varsigma_2 \) can, hence, be derived by expressing the sub-system state probabilities as the sum of two or more diagonal elements of the density matrix:

\[
\rho_{0,0} = \rho_{0,0}^S + \rho_{0,0}^F, \quad \rho_{1,1} = \rho_{1,1}^S + \rho_{1,1}^F, \quad \rho_{2,2} = \rho_{2,2}^S + \rho_{2,2}^F, \quad \rho_{3,3} = \rho_{3,3}^S + \rho_{3,3}^F, \quad \rho_{4,4} = \rho_{4,4}^S + \rho_{4,4}^F, \quad \rho_{5,5} = \rho_{5,5}^S + \rho_{5,5}^F, \quad \rho_{6,6} = \rho_{6,6}^S + \rho_{6,6}^F
\]

The QME for the QM system can be expressed as:

\[
\frac{d}{dt}(\rho_{0,0}) = \frac{d}{dt}(\rho_{0,0}^S) + \frac{d}{dt}(\rho_{0,0}^F) = \gamma \c \left\{ -\rho_{0,0}^S f_L(\epsilon_0^s) + f_L(\epsilon_0^s) - \rho_{0,1}^S f_L(\epsilon_1^s) + f_L(\epsilon_1^s) + 1 \right\}
\]

where \( \gamma = \tau_{6,3} = \tau_{3,6} \). The sets of Eqs. (9) and (10) coupled to each other. To calculate the values of the state probabilities, these sets of equations may be solved numerically using any iterative method. On solution of the state probabilities given by Eqs. (9) and (10), the charge current \( I_{L(R)} \) through the system can be calculated using the equations:

\[
I_L = q \gamma_{c} \times \left\{ \rho_{0,0}^S f_L(\epsilon_0^s) + \rho_{0,1}^S f_L(\epsilon_1^s) \right\}
\]

\[
I_R = -q \gamma_{c} \times \left\{ \rho_{0,0}^S f_R(\epsilon_0^s) + \rho_{0,1}^S f_R(\epsilon_1^s) \right\}
\]

Next, I use the set of Eqs. (9)-(11), to characterize the
set-up demonstrated in Fig. 1. Without loss of generality, I assume that \(\gamma_c = 5 \times 10^{-7}\) and \(\gamma = 5 \times 10^{-6}\). In particular, I show the characteristics of the set-up, as captured by the proposed QME for three different cases: (i) fixed voltage bias, (ii) fixed temperature bias and (iii) varying voltage bias and capacitive coupling energy.

Fig. 2(a) demonstrates the regime of current flow through the system at \(T_R = T_L = T_G = T = 5K\), \(U = 2meV\) \((\approx 4.63\, \text{meV})\) a voltage bias \(V = 1mV\) \((\approx 2.3\, \text{meV})\) for a range of positions of \(\varepsilon_g\) and \(\varepsilon_L\). We note that the maximum current flow occurs when \(\varepsilon_g\) goes a few \(kT\) below the equilibrium Fermi energy \(\mu_0\), that is when the level \(\varepsilon_g\) is always occupied with an electron, as expected. Similarly, the current flow occurs when \(\varepsilon_L + U - \mu_0\) lies in the bias window, that is from \(-qV/2 < \varepsilon_L + U - \mu_0 < qV/2\). Fig. 2(b) demonstrates the regime of short-circuited thermoelectric current flow through the system for a temperature bias given by \(T_R = T_L = T_G = T = 5K\) and \(U = 2meV\) \((\approx 3.1\, \text{meV})\) for a range of position of \(\varepsilon_g\) and \(\varepsilon_L\). This short-circuited current flows by absorbing heat energy from the reservoir \(G\) and constitutes the non-local thermoelectric action proposed in the Refs. [1–6]. Fig. 2(c) shows the regime of current flow (absolute value) with variation of the Coulomb coupling energy \(U\) and the voltage bias \(V\) at \(T_R = T_L = T_G = T = 5K\), \(\varepsilon_L + U - \mu_0 = -1.5\, \text{meV}\) \((\approx -3.5\, \text{meV})\) and \(\varepsilon_g - \mu_0 = -2\, \text{meV}\) \((\approx -4.63\, \text{meV})\). As expected, the current magnitude increases to saturation with increase in magnitude of the applied bias \(V\) and decreases with the increase in \(U\) (since the electron occupancy in probability in \(\varepsilon_g\) decreases with increase in \(U\)). In addition the energy level \(\varepsilon_L + U\) moves outside the bias window with an increase in \(U\).

To conclude, in this paper, I have methodically derived the QME for a Coulomb coupled system of quantum dots. The proposed QME has been derived from the system Hamiltonian using density matrix formulation and captures the intuitive aspects of the sequential electron transport and current flow. The derivation proposed in this paper can be generalized and extended to an array of larger number of Coulomb coupled quantum dots and can be used for theoretical analysis of such systems in the limit of sequential electronic transport.

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