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# NEMO-3D based atomistic simulation of a double quantum dot structure for spin-blockaded transport

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**Abstract** This work combines an atomistic electronic structure calculation with many-body rate equations to simulate the current-voltage (I–V) characteristics of a weakly-coupled Double Quantum Dot (DQD) system in the spin-blockade regime. Here we performed a NEMO-3D based, atomistic simulation of the geometry of the DQD to obtain its single electron eigen-states, hopping parameters, and Coulomb integrals followed by the evaluation of I–V characteristics with the many-electron spectrum of the DQD system, derived from this single-electron parameter set. The many-electron spectra and wave-functions are evaluated by exact-diagonalization of the many-electron system. The Hamiltonian is constructed from single electron eigen-states, hopping parameters and Coulomb integrals derived from atomistic NEMO 3-D simulations. Calculated I–V characteristics exhibit multiple regions of prominent Negative Differential Resistances (NDRs) that resemble the experimental trends. Unlike resonant tunnelling devices, however, level crossings in DQDs are negligible, and the NDRs result from a delicate interplay of delocalization, orbital offset and Coulomb interaction.

**Keywords** Double quantum dots · NEMO-3D · Spin blockade

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B. Muralidharan, H. Ryu and Z. Huang contributed equally to this work.

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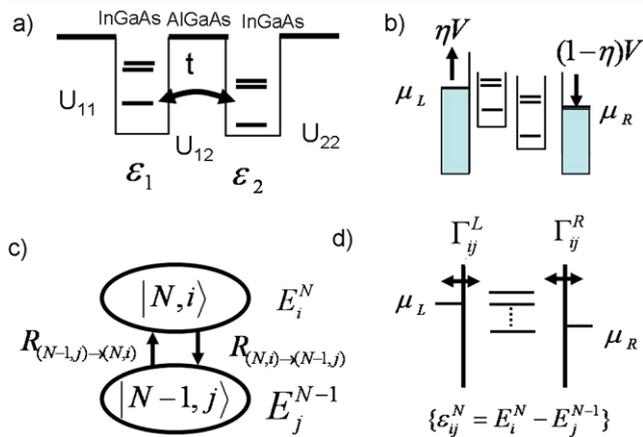
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## 1 Introduction

The recent observation of current suppression due to “Pauli spin blockade” in weakly coupled-double quantum dots (DQDs) represents an important step towards the realization and manipulation of qubits [1, 2]. The basic idea of spin blockade is the formation of “*blocking states*” [3] in the I–V characteristics, which effectively initializes a qubit [2].

A salient feature of this regime is the occurrence of multiple NDR regions in the I–V characteristics, which results from the rich physics of delocalization, Coulomb interaction and orbital offset. The focus of this paper is a reliable atomistic simulation of the DQD parameter set. We find that this calculation is very successful in interpreting multiple NDRs in the spin blockade I–V characteristics. While the physics of multiple NDRs was discussed in an earlier work [3], we focus on the atomistic simulation of the DQD parameter set through atomistic NEMO 3-D simulations [4–6], and briefly summarize our transport results via rate equations. The DQD device considered here are made of AlGaAs-InGaAs-AlGaAs-InGaAs-AlGaAs layers that have been etched to vertical QD tower and side-gated to control the lateral confinement. The typical layer thicknesses are 8-12-6-12-8 nm and the mesa of lateral circular plane is ~600 nm, which imply we have ~13 million atoms in the device of consideration. While NEMO-3D has demonstrated [4–6] successful computation of electronic structure containing 52 million atoms, calculation of Coulomb matrix in such devices is still prohibitively time-consuming. To reduce the computational time requirements a saturated Coulomb potential with Thomas-Fermi screening length for an approximate solution of the Coulomb integral is used and calibrated.



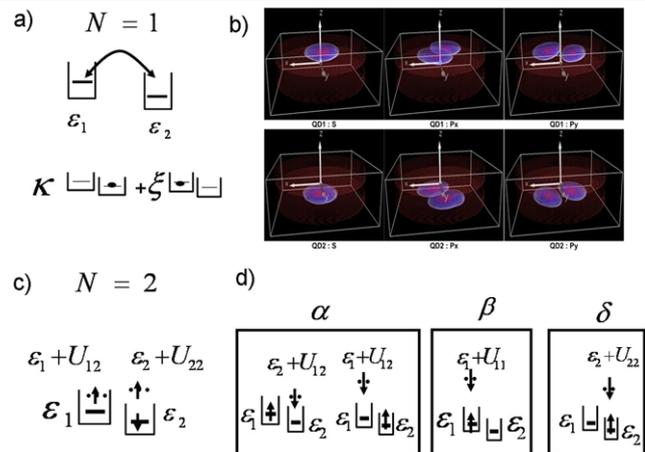
**Fig. 1** Preliminaries: **(a)** Schematic of the DQD structure. The first stage involves the evaluation of the single particle spectrum  $\{\varepsilon_{1,2}\}$ , hopping parameters  $t$ , and Coulomb integrals  $\{U_{ij}\}$ . **(b)** Voltage drop is accounted for by a capacitance divider  $\eta$ . The second stage involves the **(c)** evaluation of many electron energies and transition rates using the many-electron wave-functions. **(d)** Transport is then modeled using a master equation approach [7, 8] involving the previously calculated transition rates

## 2 Preliminaries

The summit of this effort is the evaluation of spin blockade I–V characteristics utilizing the many-electron spectrum and wave-functions of the DQD structure (Fig. 1a) by the exact-diagonalization of its many-electron Hamiltonian:

$$\hat{H} = \sum_{\alpha} \varepsilon_{\alpha} n_{\alpha} + \sum_{\alpha \neq \beta} t_{\alpha\beta} c_{\alpha}^{\dagger} c_{\beta} + \sum_{\alpha} U_{\alpha\alpha} n_{\alpha\uparrow} n_{\alpha\downarrow} + \frac{1}{2} \sum_{\alpha \neq \beta} U_{\alpha\beta} n_{\alpha} n_{\beta} \quad (1)$$

Here  $\varepsilon_i$ ,  $t_{ij}$  and  $U_{ij}$  are what we shall call the “parameter set”, representing orbital energies, hopping parameters, and Coulomb integrals between various single particle eigenstates of the DQD structure (Fig. 1a). These parameters are calculated from the diagonalization of a tight-binding atomistic Hamiltonian [4–6]. The I–V characteristics are evaluated using the standard master equation scheme [2, 3], by calculating the addition and removal transition rates (Fig. 1c) and transition energies  $\{\varepsilon_{ij}\}$  (Fig. 1d). The voltage drop across the system is accounted for using an equal capacitive divider ( $\eta = 0.5$ ) between the device and either contact. The spin blockade current voltage characteristics (I–Vs) arise from transitions between the one-electron ( $N = 1$ ) and two-electron ( $N = 2$ ) wave-functions of the many-electron Hamiltonian described in (1). Given an orbital energy offset between the two quantum dots due to local gating, the one-electron wave-functions tend to localize on one quantum dot (Fig. 2a, b). The two-electron wave-functions are a superposition of various two electron configurations (Fig. 2c, d) and usually depend on the Coulomb



**Fig. 2** Schematic of wave-functions: Spin blockade transport involves transitions between  $N = 1$  and  $N = 2$  subspaces of the many-electron space. **(a)** Given an orbital off-set between the orbital energies, the  $N = 1$  electron spectra is localized on one dot. **(b)** Electron densities of the first six one particle eigen-states show strong localization on either left or right dot. **(c)** The two electron wave-functions, however, are influenced by Coulomb interaction. The presence of one electron localized on either dot raises the orbital energy of the second electron (shown dotted). **(d)** The two electron wave-functions are linear combinations of states in which two electrons on the same dot and those in which two electrons reside on different dots

interaction parameters. The structure of two-electron wave-functions plays an important role in the formation of “blocking states” that cause multiple NDRs. Thus the evaluation of Coulomb integrals is a crucial part of this effort. We thus focus a major part of this paper on the evaluation of the parameter set  $\varepsilon_i$ ,  $t_{ij}$  and  $U_{ij}$  of the many-electron Hamiltonian (1).

## 3 Modelling the parameter set

For a many-body, rate equation-based transport simulation one needs (1) the one-electron conduction band eigenstates (2) hopping parameters between the two dots and (3) Coulomb interactions between different eigenstates which usually requires enormous computation power. Here, we give a detailed description of this process:

(1) *One-electron eigen-states*: We use NEMO-3D [4–6], a MPI/C/C++ based application for multimillion atom simulations for the computation of one-electron eigen spectra. Here, we have used semi-empirical spds\* tight-binding parameters with spin for construction of one-electron Hamiltonian.

The lateral confinement of DQD structure has been approximated as a harmonic potential ( $\sim 4$  meV) [1]. The spatial distribution of this harmonic potential has been calculated analytically and directly added to diagonal components

of one-electron Hamiltonian. This becomes an efficient approach since a full charge self-consistent solution is prohibitively time-consuming. Wave function profiles for first several states of DQD structures have are shown in Fig. 2b.

(2) *Hopping parameters*: Since electron states in conduction band of the DQD structure are known from NEMO-3D simulation, hopping parameters can be easily estimated. For this, we write a new Hamiltonian, using  $(s, p)$  states of single QD as basis:

$$\begin{matrix}
 & s1 & s2 & p1 & p2 \\
 \begin{matrix} s1 \\ s2 \\ p1 \\ p2 \end{matrix} & \begin{pmatrix} E_S & t_{ss} & 0 & t_{sp} \\ t_{ss} & E_S & t_{sp} & 0 \\ 0 & t_{sp} & E_P & t_{pp} \\ t_{sp} & 0 & t_{pp} & E_P \end{pmatrix}
 \end{matrix} \quad (2)$$

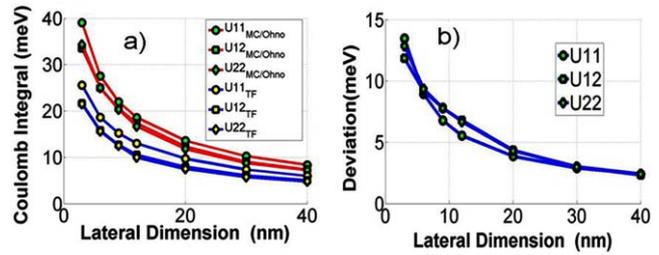
where index  $\{s1, s2, p1, p2\}$  stands for  $(s, p)$  states of two separated quantum dots,  $\{t_{ss}, t_{sp}, t_{pp}\}$  are hopping parameters between states, and  $\{E_S, E_P\}$  are on-site energies of  $(s, p)$  states of a single quantum dot. Note that here we assumed hopping parameter between  $s$  and  $p$  state of a single dot is zero. Hopping parameters can be calculated by matching eigen values of (2) with those calculated for DQD structure by NEMO-3D. This approximation becomes reasonable since two dots of the structure here are weakly coupled therefore off-diagonal components of Hamiltonian (1) will be much smaller than its diagonal elements [1, 9].

(3) *Coulomb Integrals*: Coulomb/Exchange integrals between two eigen states, requires extremely time-intensive computation. Although it has been shown that this can be done using a wave function based on the tight binding models [10, 11], a huge computation cost becomes inevitable when the device structure has more than 1–2 million atoms. Therefore, we used saturated Coulomb potential [12], with Coulomb screening length based on the Thomas-Fermi approximation [13, 14]. The saturated Coulomb potential is computed using:

$$\begin{aligned}
 & \sum_{r_1} \sum_{r_2} \frac{\psi_1(r_1)\psi_1^*(r_1)\psi_2(r_2)\psi_2^*(r_2)}{\sqrt{(r_1-r_2)^2 + \lambda(r_1, r_2)^2}} \\
 &= \sum_{r_1} \sum_{r_2} \frac{|\psi_1(r_1)|^2 |\psi_2(r_2)|^2}{\sqrt{(r_1-r_2)^2 + \lambda(r_1, r_2)^2}}
 \end{aligned} \quad (3)$$

where  $(\Psi_1, \Psi_2)$  are wavefunctions of two electron states, and  $\lambda(r_1, r_2)$  is the screening length between two point charges  $(|\Psi_1(r_1)|^2, |\Psi_2(r_2)|^2)$  given by:

$$\begin{aligned}
 \lambda(r_1, r_2) &= \left( \frac{3e^2 \rho(r_1, r_2)}{\epsilon E_F} \right)^{-1} \\
 \rho(r_1, r_2) &= \frac{\sum_{R=r_1}^{r_2} (|\psi_1(R)|^2 + |\psi_2(R)|^2)}{V(r_1, r_2)}
 \end{aligned} \quad (4)$$



**Fig. 3** (a) Coulomb integrals calculated using two methods—TF approximation and MC/Ohno’s formula. (b) Difference between two results vs. vertical dimension of GaAs/InAs QD

Since transport measurements involved very low temperatures ( $\sim 0.2$  K) [1], we have used the Thomas-Fermi approximation to calculate screening lengths between two points  $r_1$  and  $r_2$ . Details are described in (4), where  $V(r_1, r_2)$  is volume of the box whose diagonal endpoints are  $r_1$  and  $r_2$ . When  $r_1$  equals  $r_2$ ,  $\rho(r_1, r_2)$  is assumed to be an averaged electron density of the unit cell where  $(r_1, r_2)$  belongs.

To test the validity of TF approximation used here, we computed Coulomb interactions using the two lowest conduction band electron states in a InAs/GaAs quantum dot disk [6] with various lateral dimensions, where reference data has been calculated by the method based on Monte Carlo integration and Ohno formula [10]. The QD disks have a constant height of 45 nm. In Fig. 3, we see that results with TF approximation become closer to the reference data as the vertical dimension increases, which validates our approach since the lateral dimension of our DQD structure is  $\sim 600$  nm.

### 4 Results

Coulomb charging energy and hopping parameters used for this simulation are summarized in Table 1. The Coulomb and hopping matrix elements show a clear state and site dependence that can not be predicted in an ad-hoc manner.

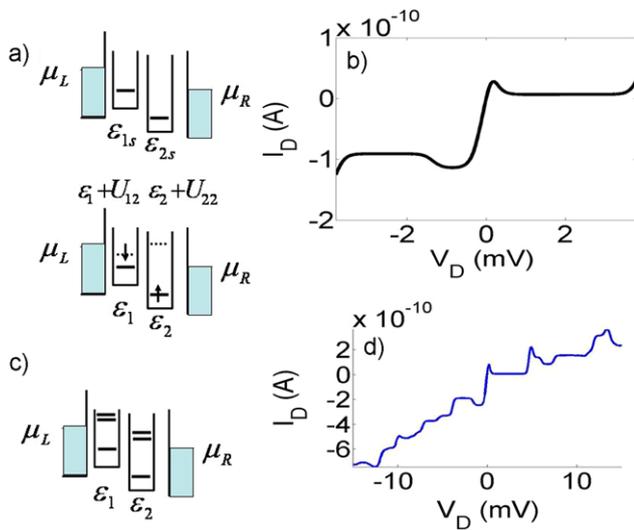
The I–V characteristics evaluated using this parameter set and the many-electron Hamiltonian (1) shown in Fig. 4d, broadly resemble those observed in the spin blockade I–Vs [1].

### 5 Conclusion

We have presented for the first time, a many-body transport simulation of an experimental DQD geometry (Fig. 1a) starting from a NEMO-3D based atomistic calculation of the single particle parameter set. While the NEMO-3D atomistic simulation can be generalized to any geometry, and our many-body transport simulation based on our rate equations can be generalized to Quantum Dot or molecular systems with multiple levels.

**Table 1** Computed Coulomb integral and hopping parameters

Coulomb charge energy	Value (meV)	Normalized Coulomb parameters	Hopping parameters	Value (meV)
$U(sL, sL)$	4.971	1	$t(sL, sR)$	-0.227
$U(pL, pL)$	3.821	0.7687	$t(sL, pR)$	-0.1
$U(sR, sR)$	4.971	1	$t(pL, sR)$	-0.1
$U(pR, pR)$	3.821	0.7687	$t(pL, pR)$	-0.229
$U(sL, pL)$	3.976	0.7998	$t(sL, pL)$	0
$U(sL, sR)$	2.056	0.4136	$t(sR, pR)$	0
$U(sL, pR)$	1.970	0.3963		
$U(pL, pR)$	1.909	0.3840		
$U(sR, pR)$	3.976	0.7998		



**Fig. 4** Spin Blockaded transport shows multiple NDRs. (a) Each pair of NDRs is caused by the interaction of two orbital levels, their offset and Coulomb parameters [3]. For example (b) Low Bias NDR pair is formed due to the  $1s$  levels in both dots. (c) When  $2p$  levels are included one obtains the  $I$ - $V$  characteristics (d) that broadly resemble the experimental trends [1]

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