Thermoelectric conversion in Silicon quantum-dots

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Abstract. Quantum dot-based devices have specific thermoelectric properties. Thanks to their delta-like density of states, they are expected to exhibit high Seebeck coefficient, nearly zero electronic thermal conductance and ultra-low phononic thermal conductance if embedded in an oxide matrix. Using a physical simulator dedicated to the sequential transport through quantum dots (QDs), the thermoelectric properties of devices based on Silicon QDs embedded in silicon oxide are assessed. Fully self-consistent 3D Poisson/Schrödinger simulation is performed. From the accurate computation of tunneling rates, a Monte-Carlo algorithm is used to solve the master equation and to extract the current-voltage characteristics for different temperature gradients applied between the electrodes. The evolution of both the Seebeck coefficient and the electronic conductivity resulting from a temperature bias are investigated for dissymmetric spherical and cubic quantum-dot-based (QD) single-electron transistors (SETs). Finally, the validity of the linear regime and the potentiality of semiconducting SETs in the field of Seebeck nanoscale metrology are discussed.

1. Introduction

Some specific properties of nanostructures have generated a recent revival of interest in thermoelectric devices. Indeed, it has been proved that in low-dimensional systems the "sharp" density-of-states can increase the Seebeck effect [1]. Additionally, in nanostructures electronic transport and thermal transport may be decoupled [2], i.e. the Wiedemann-Franz Law which governs conventional solids can be violated. This opens the way of designing a thermoelectric "Holy Grail" which would behave at the same time as a phonon glass and an electron crystal. Thus, much theoretical and experimental efforts have been focused on thermoelectric properties of nanostructures such as superlattices [3], graphene nanoribbon [4], nanowires [5]. PbSeTe-based Quantum Dot (QD) superlattices have been shown to exhibit thermoelectric figure of merit ZT as high as 1.6 [6] and the enhancement of the thermoelectric properties of a weakly coupled QD has been predicted [7].

Recently, Single Electron Transistors (SETs) with an Si QD of diameter smaller than 5 nm exhibiting typical Coulomb oscillations at room temperature have been demonstrated [8]. The single-electron tunnelling across discrete levels in the QD occurring in such devices can be used as a quasi ideal energy filter providing incomparable thermoelectric properties. First, the electronic thermal

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conductance is suppressed in a SET as no heat flux can be transported by electrons between the electrodes if there is no net electron flux. Second, the Seebeck coefficient in a SET thanks to its material-independent characteristics has been proposed to become a standard in nanoscale metrology [9].

To our knowledge, very few theoretical studies have investigated the thermoelectric properties of SETs in sequential transport regime by taking into account properly the confinement effects inherent in a semiconducting quantum dot. We have previously developed the simulator SENS (Single-Electron Nanostructure Simulation) which is well suited for studying the electrical behaviour and the temperature effects of a SET and thus to investigate its thermoelectric properties. It is the aim of this paper which is organized as follows. The model is described in Section 2. The thermoelectric properties of spherical and cubic quantum dots are presented and discussed in Section 3. In particular, we focus on the potentialities of single electron transistors in the field of Seebeck coefficient metrology.

2. Model
The method used in the code SENS to simulate NC-based single-electron devices has been described elsewhere for single- [10, 11] and double-dot [12] tunnel junctions. It has been recently extended to single-electron transistors [13]. Here, we just summarize briefly the main stages of the calculation in the case of SET, including the extraction of thermoelectric parameters.

The first stage consists in calculating the electronic structure of the NC by solving self-consistently the 3D Poisson and Schrödinger equations within the effective mass and the Hartree approximations which have been proved to be correct for Si NCs of radius greater than 1.5 nm [14, 15]. With a view to calculating the tunnelling rates to be implemented in sequential transport simulation, the Hartree method offers the advantage of providing the wave function \( \psi_i(r) \) and the energy level \( E_i \) of each electron labelled \( i \). For each possible bias condition, the Poisson-Schrödinger system to be solved is thus

\[
\begin{bmatrix}
-\frac{\hbar^2}{2m} \nabla \left( \frac{1}{m} \nabla \right) + V^{\text{conf}}(r) + V^{\text{bias}}(r) + V^{\text{coul}}(r)
\end{bmatrix} \psi_i(r) = \mu_i \psi_i(r)
\]

(1)

Where \( m \) is an isotropic effective mass, \( \varepsilon \) is the dielectric constant, \( V^{\text{conf}} \) is the confinement potential and \( V^{\text{bias}} \) is the bias potential, obtained by solving the 3D Poisson equation without charge. The Coulomb potential \( V^{\text{coul}}_i \) for the \( i \)-th electron is obtained from the charge density \( \rho_i \) given by

\[
\rho_i(r) = -e \sum_{j \neq i} |\psi_j(r)|^2
\]

(2)

The wave functions are then used to calculate the tunnelling rates between the dot \( d \) and the lead \( L \) from the Fermi golden rule (weak coupling limit) as

\[
\begin{align*}
\Gamma_{ld} (\mu_i, T_L) &= \sum_{\mu_i} \frac{2\pi}{\hbar} |M| \rho_L f_L (\mu_i, T_L) \\
\Gamma_{dl} (\mu_i, T_L) &= \sum_{\mu_i} \frac{2\pi}{\hbar} |M| \rho_L g_L \left[ 1 - f_L (\mu_i, T_L) \right]
\end{align*}
\]

(3)
where $g_\mu$, $l_\mu$, $T_\mu$, $f_\mu(\mu, T_L)$, and $\rho_L$ are the number of electrons on the energy level $\mu$, the number of free states on this level, the temperature of the lead, the Fermi function at lead Fermi level $E_{FL}$ and the density of states of the lead, respectively. The tunnelling matrix element $M$ is given by the Bardeen formula [10, 16]

$$M = \frac{\hbar^2}{2m_{ox}} \int \int \left[ \psi_L(r) \nabla \psi_d(r) - \psi_d(r) \nabla \psi_L(r) \right] d\mathbf{S}$$

(4)

where $S_{barr}$ is a surface inside the tunnel barrier which separates arbitrarily the dot domain from the electrode domain [10,17], $\psi_d$ and $\psi_L$ are the electronic wave functions in the QD and the lead, respectively, and $m_{barr}$ is the electron effective mass in the barrier. The wave function $\psi_L$ is deduced from an analytical expression derived within the Wentzel-Kramer-Brillouin approximation, which has been proven correct by comparing the tunnelling current obtained for the simple case of a triangular barrier using our approach and an exact calculation [10].

The tunneling rates are then used as input data in a Monte-Carlo (MC) algorithm to compute the tunnel transitions as a function of time. This MC simulation provides the probabilities $P(N)$ of finding $N$ electrons in the dot and the current at the dot-lead interface is deduced from the master equation

$$I = -e \sum_N P(N) \left[ \Gamma_{d\ell}(N) - \Gamma_{Ld}(N) \right]$$

(5)

Assuming the source and drain leads to be connected to heat reservoirs of temperature $T_h$ and $T_c$, the source-drain current is expressed in the linear regime as

$$I = \alpha G_e \Delta T + G_e \Delta V$$

(6)

where $G_e$ is the electronic conductance, $\alpha$ is the Seebeck coefficient, $\Delta T = T_h - T_c$, and $\Delta V = V_{DS}$. The parameters $G_e$ and $\alpha$ may be deduced from the simulation data as

$$\left\{ \begin{array}{l}
G_e = \frac{dI}{dV} \bigg|_{\Delta T=0} \\
\alpha = -\frac{V_{DS}}{G_e} \frac{dI}{d\Delta T} \bigg|_{V_{GS}=0}
\end{array} \right.$$ 

(7)

3. Results and discussion

The simulated SET consists in a cubic Si-QD of 50-nm edge with 1.2-nm (1.5 nm)-thick source (drain) tunnel barriers and a 5-nm-thick gate oxide, as schematized in figure 1. However, it is also possible to obtain Si QDs of quasi-cubic shape by top-down techniques on SOI wafer [18]. The simulator SENS is able to describe any 3D shape of QD and we have considered also a SET with a cubic QD of 50-nm edge length. The results presented will focus on those obtained for the latter SET which gives the best performance in terms of conductance and power factor. To investigate the thermoelectric behaviour of

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temperature gradient was introduced by fixing the temperature of the source (drain) lead at \( T + \Delta T/2 \) or \( T - \Delta T/2 \), where \( \Delta T \) can be either negative or positive to exchange the position of hot and cold electrodes.

The Figure 2 shows the drain-current stability diagram of the SET with cubic QD at 77 K. We observe the typical diamonds that are commonly obtained with experimental SETs, either for metallic or semiconducting QDs.
To use the SET as a thermoelectric device, the bias point must be chosen such that the Coulomb blockade vanishes and the electronic conductance is maximum. The conductance $G_e = d/dV_{DS}$ at $V_{DS} = 0$, the Seebeck coefficient $\alpha = I/(\Delta T G_e)$ at $V_{DS} = 0$ and the power factor $\alpha^2 G_e$ of SETs have been numerically calculated as a function of the gate voltage and plotted in figures 3 (spherical QD) and 4 (cubic QD). The values of Seebeck coefficient have been extracted from the slope of the $I$-$\Delta T$ characteristics at $V_{DS} = 0$. For instance, Figure 5 shows the $I$-$\Delta T$ curve at $V_G = 3.4$ V for the cubic dot, which gives $\alpha = 2.57$ mV/K.

As expected, the evolution of $G_e$ in Figures 3-4 exhibit several peaks that correspond to the alignment of the Fermi level in the leads $E_{F_L}$ and the $i$-th energy levels in the dot $\mu_i$. The Seebeck coefficient decreases linearly with the gate voltage and equals zero when $E_{F_L} = \mu_i$. Besides, it switches abruptly from positive to negative values when an upper quantum level is activated, which oscillations, as predicted by Beenaker et al. [19].
Figure 6. Drain current-bias voltage characteristics for different temperature gradients around $T = 77$ K at $V_G = 3.4$ V.

Hence, to get a maximum of power factor, a trade-off between the conductance (maximum when $E_{F_L} = \mu_i$) and the Seebeck coefficient (equal to 0 if $E_{F_L} = \mu_i$) has to be found. It explains why the first maximum of power factor at $V_{GS} = 1.94$ V in figure 4 for the cubic dot occurs before the maximum of conductance at $V_{GS} = 2.06$ V (for $E_{F_L} = \mu_i$). The second peak at $V_{GS} = 3.4$ V gives the maximum absolute value of power factor. The first peak of the spherical structure occurs at a higher gate voltage ($V_G = 2.4$ V) than in the cubic structure. This is a consequence of the smaller volume of the sphere, which yields larger energy spacing between the QD levels. If the Seebeck coefficient reaches very comparable values in cubic and spherical QD structures, the conductance and thus the power factor are about two orders of magnitude higher in the cubic QD-based SET. This strong influence of the shape on the current has been widely discussed in the case of the double-tunnel junction [11].

The influence of the temperature gradient $\Delta T$ applied to the SET is shown in figure 6. The drain current $I_D$ vs drain bias $V_{DS}$ characteristics are plotted for the cubic dot at the maximum of the power factor ($V_{GS} = 3.4$ V). In accordance with (3), the influence of temperature is taken into account in the tunnelling rate expressions through the Fermi distribution function in the leads. Since at $V_{GS} = 3.4$ V, $E_{F_L} < \mu_i$ (2), for $\Delta T > 0$ we have $f_L(T + \Delta T/2) > f_L(T) > f_L(T - \Delta T/2)$. Thus, as illustrated in figure 7, we obtain $\Gamma_{sd}(T + \Delta T/2) > \Gamma_{sd}(T)$ and $\Gamma_{dd}(T - \Delta T/2) > \Gamma_{dd}(T)$. As a consequence, the drain current increases when increasing the temperature gradient, and a finite current takes place at $V_{DS} = 0$ V and finite $\Delta T$. 
Figure 7. In-and-out tunnelling rates as a function of drain bias at $V_G = 3.4$ V, with and without temperature gradient, with $T = 77$ K and $\Delta T = 10$ K.

Figure 8. Seebeck coefficient $\alpha$ as a function of “ideal Seebeck coefficient” $(\mu_i - E_{F_L})/eT$ for 1, 2 and 3 electrons in the dot.

The $I$-$V$ curves of figure 6 are of great importance as they clearly illustrate that for all temperature gradients considered here the thermoelectric generators work in the linear regime. Besides, they give directly access to both the conductance $G_e$, given by the slope of $I$-$V_{DS}$ characteristics and the Seebeck coefficient $\alpha$ via the expression $V_{DS} = -\alpha \Delta T$ at $I_D = 0$. These evaluations are in full agreement with the results of figures 3-4 as long as the device operates in the linear regime.

Finally, we have plotted in figure 8 the Seebeck coefficient $\alpha$ as a function of $(\mu_i - E_{F_L})/eT$. In our semi-conducting SET simulated in sequential transport regime, $\alpha$ really acts as the “ideal Seebeck coefficient” described in [9] in the [-1 mV – +1 mV] range. Our self-consistent multi-level model reproduces well this $\alpha_{\text{ideal}}$ for 1, 2 or 3 electrons in the dot even at $T = 77$ K.

4. Conclusion
Using our 3D self-consistent Monte Carlo SET simulator, the thermoelectric properties of dissymmetric spherical and cubic Silicon QDs SETs embedded in silicon oxide have been investigated. The evolutions of the Seebeck coefficient and the electronic conductance as a function of the gate bias exhibit an interesting regime where the power factor reaches some fW/K² for the single cubic dot of 50-nm edge length. Next, the influence of a temperature gradient applied to the SETs on their current-voltage characteristics has validated the linear assumption to study the SET as a thermoelectric generator. Finally, the interest of the SETs in the field of Seebeck nanoscale metrology has been highlighted. In the near future, to evaluate accurately the thermoelectric figure of merit ZT, the effect of collisional energy level broadening [12] and the computation of both electronic and phononic thermal conductance will be incorporated in the simulation.

5. References

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