

Multielectron picture

Multielectron energy levels

Let's assume the total electron energy for N electrons in a particular energy level of a nanostructure is the sum of a core energy $\tilde{\epsilon}$ for each electron with the total electrostatic repulsion between all pairs of electrons and the potential energy $U_L = qV_g$ due to an applied gate voltage with respect to the source:

$$E(N) = N \cdot \tilde{\epsilon} + U_{ee}(N) - N \cdot U_L$$

Let's assume the electrostatic repulsive energy among electrons in a nanostructure is the same value U_0 for every electron pair. Summing over all pairs gives

$$U_{ee}(N) = U_0 \cdot \sum_{n=1}^N (n-1) = U_0 \cdot S_N$$

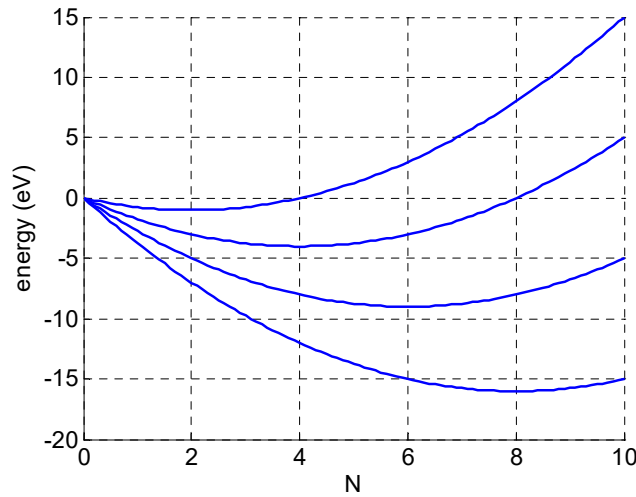
where

$$\begin{aligned} S_N &= \sum_{n=1}^N (n-1) = \sum_{n=1}^N N - \sum_{n=1}^N (N-n+1) \\ &= N^2 - \sum_{n=1}^N n = N^2 - \left\{ \left[\sum_{n=1}^N (n-1) \right] + N \right\} \\ S_N &= N^2 - N - S_N \end{aligned}$$

So $2S_N = N^2 - N$ and $S_N = N \cdot (N-1)/2$. Now

$$U_{ee}(N) = \frac{N \cdot (N-1)}{2} \cdot U_0$$

The function $E(N)$ has a minimum at a positive value of N if $U_L > \tilde{\epsilon} - U_0/2$.



The energy to decrease the number of electrons from N to $N-1$, called the ionization level, is

$$\epsilon_N^{(-)} = E(N) - E(N-1) = \tilde{\epsilon} + (N-1) \cdot U_0 - U_L$$

The energy to increase the number of electrons from N to $N+1$, called the affinity level, is

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$$\varepsilon_N^{(+)} = E(N+1) - E(N) = \tilde{\varepsilon} + N \cdot U_0 - U_L$$

Clearly $\varepsilon_N^{(-)} = \varepsilon_{N-1}^{(+)}$. Let us then refer only to the ionization levels for simplicity, i.e.

$$\varepsilon_N = \varepsilon_N^{(-)} = \varepsilon_{N-1}^{(+)}$$

Solution

Equilibrium between a source contact and the channel can be considered a microscopic steady-state condition. That is, the rate at which electrons enter the channel from the source is proportional to the fraction of filled source levels at energy ε_N (the $N-1$ affinity level) times the probability that the channel contains only $N-1$ electrons, while the rate at which electrons enter source from the the channel is proportional to the fraction of empty source levels at energy ε_N (the N ionization level) times the probability that the channel contains N electrons. Assuming the electron transfer process is reversible, the rate constants for either process should be equal, so

$$\not\leftarrow \cdot f_0(\varepsilon_N - \mu) \cdot P_{N-1} = \not\rightarrow \cdot [1 - f_0(\varepsilon_N - \mu)] \cdot P_N$$

Now we can find the ratio

$$R_N = \frac{P_N}{P_{N-1}} = \frac{f_0(\varepsilon_N - \mu)}{1 - f_0(\varepsilon_N - \mu)}$$

Define

$$x_N = e^{(\varepsilon_N - \mu)/kT}$$

Then

$$R_N = \frac{\frac{1}{x_N + 1}}{1 - \frac{1}{x_N + 1}} = \frac{1}{x_N} = e^{-(\varepsilon_N - \mu)/kT}$$

We see that

$$\frac{P_N}{P_0} = \frac{P_N}{P_{N-1}} \cdot \frac{P_{N-1}}{P_{N-2}} \cdots \frac{P_1}{P_0} = R_N \cdot R_{N-1} \cdots R_1$$

Then

$$\frac{P_N}{P_0} = \exp\left[-\sum_{n=1}^N (\varepsilon_n - \mu)/kT\right]$$

Observe that

$$\begin{aligned} \sum_{n=1}^N (\varepsilon_n - \mu)/kT &= \sum_{n=1}^N [\tilde{\varepsilon} + (n-1) \cdot U_0 - U_L - \mu]/kT \\ &= -[E(N) - N \cdot \mu]/kT \end{aligned}$$

So

$$\frac{P_N}{P_0} = e^{-[E(N)-N\cdot\mu]/kT}$$

This allows us to find the average number of electrons in the channel

$$\langle N \rangle = \sum_{N=0}^{N_\varepsilon} N \cdot P_N$$

We know that

$$\sum_{N=0}^{N_\varepsilon} P_N = 1$$

So

$$\sum_{N=0}^{N_\varepsilon} \frac{P_N}{P_0} = \frac{1}{P_0}$$

Which gives

$$P_0 = \frac{1}{\sum_{N=0}^{N_\varepsilon} \frac{P_N}{P_0}}$$

So

$$P_N = \frac{e^{-[E(N)-N\cdot\mu]/kT}}{\sum_{n=0}^{N_\varepsilon} e^{-[E(n)-n\cdot\mu]/kT}}$$

Now we can write

$$\langle N \rangle = \sum_{N=0}^{N_\varepsilon} N \cdot P_N = \frac{\sum_{N=0}^{N_\varepsilon} N \cdot e^{-[E(N)-N\cdot\mu]/kT}}{\sum_{N=0}^{N_\varepsilon} e^{-[E(N)-N\cdot\mu]/kT}}$$

This is suitable for exact computation of $\langle N \rangle$ within our model. Once we have determined $\langle N \rangle$, we can find the energy level using

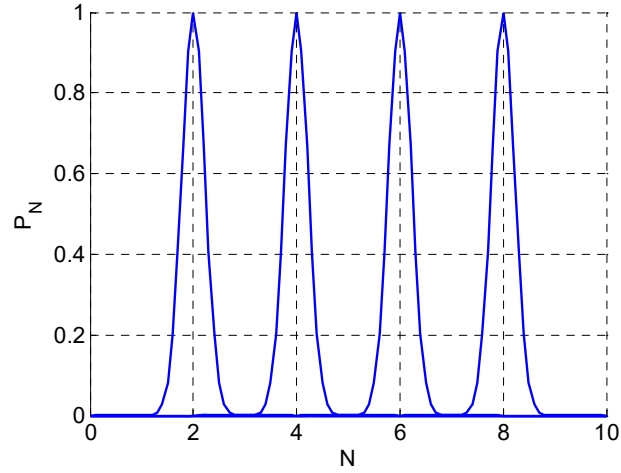
$$\langle N \rangle = N_\varepsilon \cdot f_0(\varepsilon - \mu)$$

which gives

$$\varepsilon - \mu = kT \cdot \ln\left(\frac{N_\varepsilon}{\langle N \rangle} - 1\right)$$

Interpretation

At typical temperatures and biases, when the level is partially filled, the difference $\varepsilon - \mu$ will be quite small. A simple estimate of $\langle N \rangle$ in these conditions be obtained by finding the most probable value of N , that is, find N^* where $\exp\{-[E(N^*) - N^* \cdot \mu]/kT\}$ is a maximum. If $U_0 \gg kT$, the probability distribution will be sharply peaked.



Notice that

$$\frac{\partial}{\partial N} e^{f(N)} = \left[\frac{\partial}{\partial N} f(N) \right] \cdot e^{f(N)}$$

Applying this gives

$$\frac{\partial}{\partial N} e^{-[E(N)-N\mu]/kT} \Big|_{N=N^*} = \left\{ \left[\frac{\partial E(N)}{\partial N} - \mu \right] \cdot e^{-[E(N)-N\mu]/kT} \right\} \Big|_{N=N^*} = 0$$

We can write with some generality

$$\frac{\partial E(N)}{\partial N} = \tilde{\varepsilon} + \frac{\partial U_{ee}(N)}{\partial N} - U_L$$

We define a self-consistent-field energy as

$$U_{SCF}(N) = \frac{\partial U_{ee}}{\partial N} \Big|_N = \left(N - \frac{1}{2} \right) \cdot U_0$$

This gives

$$\tilde{\varepsilon} + U_{SCF}(N^*) - U_L - \mu = 0$$

Under the conditions where this approximation is valid, the energy level is very near the Fermi level, so we can estimate the energy level as

$$\varepsilon^* = \tilde{\varepsilon} + U_{SCF}(N^*) - U_L$$

In particular,

$$\varepsilon^* = \tilde{\varepsilon} + \left(N^* - \frac{1}{2} \right) \cdot U_0 - U_L$$

which gives the estimate

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$$\langle N \rangle = N^* = \frac{1}{2} - \left[\frac{(\tilde{\epsilon} - \mu) - U_L}{U_0} \right]$$

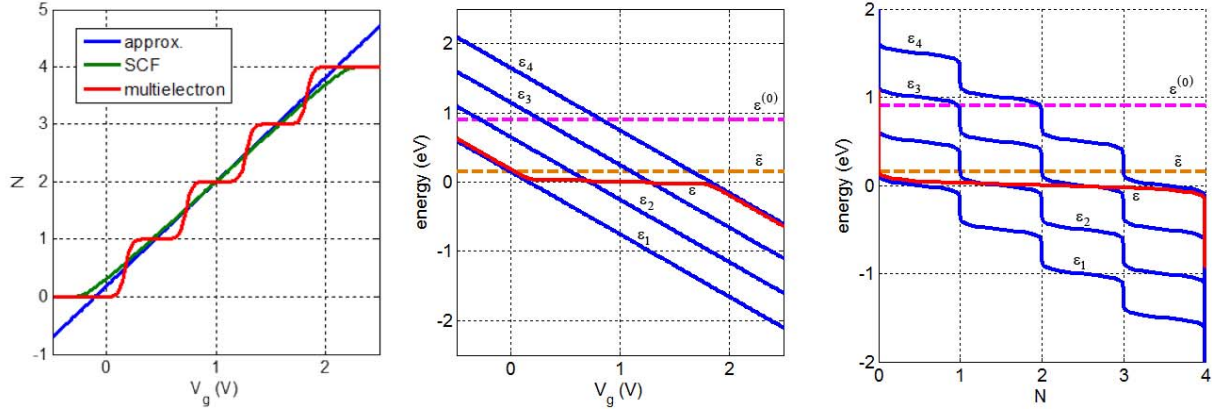
Let's say that when the channel is neutral, the level contains $N^{(0)}$ electrons.

$$\mu \approx \epsilon^{(0)} = \tilde{\epsilon} + U_{SCF}(N^{(0)}) - U_L^{(0)}$$

When we decouple the channel from the source (without changing its charge) and remove the gate voltage, the chemical potential in the channel moves to $\mu^{(0)} = \mu + U_L^{(0)}$. But the channel charge stays the same, so the energy level is at $\epsilon^{(0)} \approx \mu^{(0)}$. We then know the energy level in the isolated, neutral channel

$$\epsilon^{(0)} = \tilde{\epsilon} + U_{SCF}(N^{(0)}) = \tilde{\epsilon} + \left(N^{(0)} - \frac{1}{2} \right) \cdot U_0$$

Although we calculated this in the continuous limit, it is a useful reference for analyzing the energy levels in the channel in the discrete case. A comparison of the number of channel electrons with varying gate voltage is plotted below ($N_\epsilon = 4$, $N^{(0)} = 2$, $U_0 = 0.50$ eV, $C_L = 0.9$, $\mu^{(0)} - \mu_s = 0.9$ eV, $\mu_s = 0$). The variation of the energy levels with gate voltage is also shown. Plotting these energies vs. the number of channel electrons shows that the single-electron energy levels computed in the multielectron picture shift each time the number of electrons in the channel changes. However, the energy level ϵ is pinned very close to the chemical potential during charging.



The single-electron energy levels also allow a simpler description of the above behavior that provides a good estimation of quantities in the full multielectron picture, assuming $U_0 \gg kT$. In this case, the separation of the ϵ_N is much greater than kT , so we can calculate the occupancy of each using the Fermi function. Then

$$\langle N \rangle = \sum_{N=1}^{N_\epsilon} f_0(\epsilon_N - \mu)$$

Notice that

$$f_0(\epsilon_N - \mu) = \frac{1}{1 + \frac{1}{R_N}}$$

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Then

$$\langle N \rangle = \sum_{N=1}^{N_e} \left(\frac{1}{1 + \frac{1}{R_N}} \right) = \frac{\sum_{N=1}^{N_e} \left(1 + \frac{1}{R_N} \right)}{\prod_{N=1}^{N_e} \left(1 + \frac{1}{R_N} \right)}$$

In the multielectron picture, we had

$$\langle N \rangle = \frac{\sum_{N=0}^{N_e} (N \cdot \prod_{n=0}^N R_n)}{\sum_{N=0}^{N_e} (\prod_{n=0}^N R_n)}$$

At some level of filling N , we have $R_N \gg 1$ and $R_{N+1} \ll 1$ (assuming $U_0 \gg kT$), and the two expressions above become equal. Notice that, if $\varepsilon_N - \mu = 0$, then

$$N = 1 - \left(\frac{\tilde{\varepsilon} - \mu - U_L}{U_0} \right)$$

But $f_0(\varepsilon_N - \mu) = 1/2$, so we expect

$$\langle N \rangle = N - \frac{1}{2}$$

This gives

$$\langle N \rangle = \frac{1}{2} - \left(\frac{\tilde{\varepsilon} - \mu - U_L}{U_0} \right)$$

in agreement with the multielectron picture. A graphical depiction of the energy levels with increasing gate voltage is shown below.

