NANO 705-Notes

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 $\tilde{\varepsilon}$ for each electron with the total electrostatic repulsion between each pair of electrons and the potential energy $U_L = qV_g$ due to an applied gate voltage with respect to the source:

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

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

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

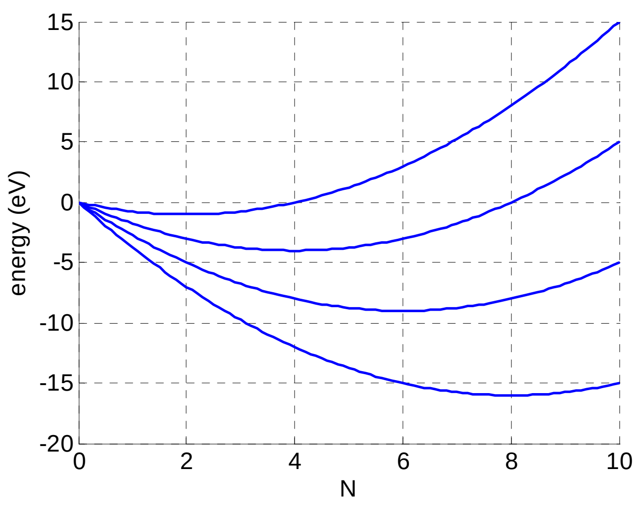
where

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

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 $U_L > \tilde{\varepsilon} - U_0/2$.



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

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

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

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

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

$$\cancel{\gamma} \cdot f_0(\varepsilon_N - \mu) \cdot P_{N-1} = \cancel{\gamma} \cdot [1 - f_0(\varepsilon_N - \mu)] \cdot P_N$$

Now we can find the ratio

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

Define

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

Then

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

We see that

$$\frac{P_N}{P_0} = \frac{P_N}{P_{N-1}} \cdot \frac{P_{N-1}}{P_{N-2}} \cdots \frac{P_1}{P_0} = S_N \cdot S_{N-1} \cdots S_1$$

Then

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

Observe that

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

So

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

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

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

Now we can write

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

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

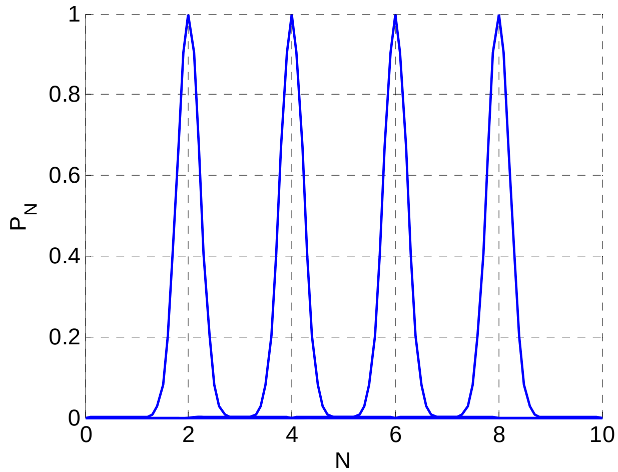
$$\langle N \rangle = N_\varepsilon \cdot f_0(\varepsilon - \mu)$$

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 $\exp\left\{-\left[E\left(N^*\right) - N^* \cdot \mu\right]/kT\right\}$ 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

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

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

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

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

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

In particular,

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

which gives the estimate

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

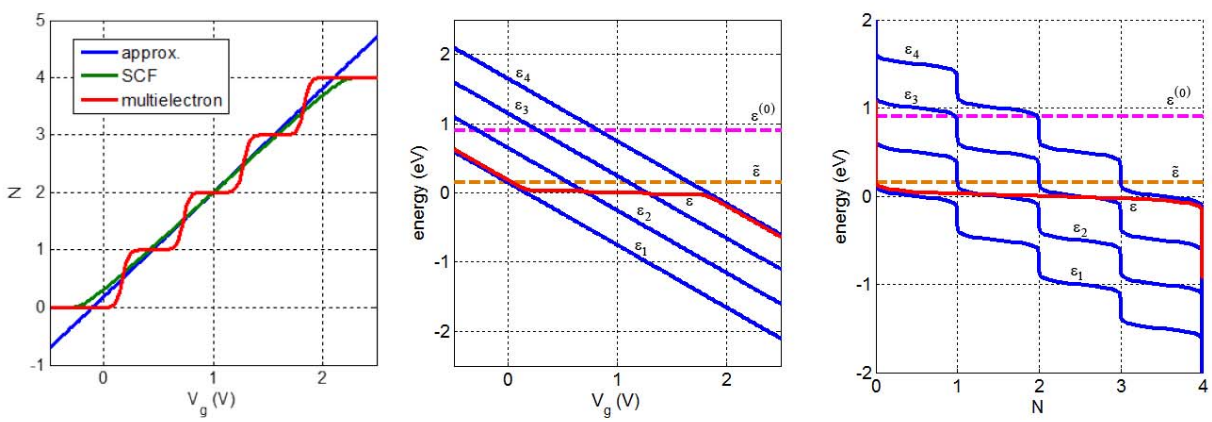
Let's say that when the channel is neutral, the level contains $N^{(0)}$ electrons.

$$\mu \approx \varepsilon'^{(0)} = \tilde{\varepsilon} + U_{SCF}\left(N^{(0)}\right) - U_L^{(0)}$$

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 $\varepsilon^{(0)} \approx \mu^{(0)}$. We then know the energy level in the isolated, neutral channel

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

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 ($N_\varepsilon = 4$, $N^{(0)} = 2$ $U_0 = 0.50$ 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 can calculate the occupancy of each using the Fermi function. Then

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

Notice that

$$f_0(\varepsilon_N - \mu) = \frac{1}{1 + \frac{1}{S_N}}$$

Then

$$\langle N\rangle = \sum_{N=1}^{N_\varepsilon}\left(\frac{1}{1+\frac{1}{S_N}}\right) = \frac{\sum_{N=1}^{N_\varepsilon}\left(1+\frac{1}{S_N}\right)}{\Pi_{N=1}^{N_\varepsilon}\left(1+\frac{1}{S_N}\right)}$$

In the multielectron picture, we had

$$\langle N\rangle = \frac{\sum_{N=0}^{N_\varepsilon}\left(N\cdot\Pi_{n=0}^{N}S_n\right)}{\sum_{N=0}^{N_\varepsilon}\left(\Pi_{n=0}^{N}S_n\right)}$$

At some level of filling N, we have $S_N \gg 1$ and $S_{N+1} \ll 1$ (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.