Excitation transport in a network

Submitted by: Omer Amit

We investigat transfer in sevral different models describing energy transport.
For this purpose we define and look into the maximum output probability.

Excitation - Particle similarity:

Our network compose of N complex mulecules with an interaction between them. If we reduce each mulecule to a two level system such that out whole network can be described by N two level systems coupled to eahc other, the Hemiltonian of the network would be:

$$\mathcal{H} = \sum_{i=1}^{N} E_i a_+^{(i)} a_-^{(i)} + \sum_{i \neq j}^{N} V_{i,j} a_+^{(i)} a_-^{(j)} \tag{1}$$

If we demand that the on site energy for all the sites is the same we can set: $E_i = E = 0$.

We define our initial state as a single excitation in the first site, 'in'. we can then rewrite the Hemiltonian for a system with a single exitation as:

$$\mathcal{H} = \sum_{i \neq j}^{N} V_{i,j} |i\rangle\langle j| \tag{2}$$

Where $|i\rangle$ is the state when the particle is at the ith site.
We can now note taht this Hemiltonian is the same for a system of N sites with one particle moving between them.

Transport efficiency:

In order to talk about transport efficiency we look at the probability to find the excitation at the last site, 'out', $p_{out}(t)$. We consider the transport successful when $p_{out}(\bar{t}) = 1$. The time $\bar{t}$ would be our measure for transport time and we would look for a configuration that minimaize $\bar{t}$. Note that for some configurations $p_{out}(t)$ never reaches the value of 1, for this cases we would consider the maximum value of $p_{out}(t)$ as a second paramater for the transport efficiency.

Two site system:

We start with the simple case of a two sites system. The Hemiltonian is:

$$\mathcal{H} = \begin{pmatrix} 0 & V \\ V & 0 \end{pmatrix} \tag{3}$$

it is clear that the oscillation frequency is then $\Omega = 2V$ and the time $\bar{t}$ to get complete population invertion is:

$$\bar{t} = \frac{\pi}{2|V|} \tag{4}$$

We would use this $\bar{t}$ as a baseline for our future calculations. We should also note that in this simple

configuration we do achieve complete transport

N site network, stochastic approach:

Writing the master equation for the probability of the excitation to be in site i:

$$\frac{\partial p_i}{\partial t} = \sum_j W_{ij} p_i - \Gamma_i p_i \tag{5}$$

where $\Gamma_i = \sum_j W_{ij}$ and $W_{ij} = \frac{c^2}{|x_i - x_j|^{2\alpha}}$ Solving thise equation we get that the probability $p_{out}(t)$ decay in time to a value of $\frac{1}{N}$. However, the decay rate would be of the order of Γ and in the case where Γ is much smaller then the oscillation time of the system we would get a $p_{out}(t) \to 1$ and we can consider the transport successful .

N site network, ballistic estimate:

We assume a potential in the form of:

$$V_{ij} = \frac{c}{|x_i - x_j|^\alpha} \tag{6}$$

where α is a natural number. We define our network as a ring with length L so the distance between two neighbouring sites is $a = \frac{L}{N}$. Our output site would be site number $N/2$, so that it is the furthest away from the input site.
The Hemiltonian is:

$$\mathcal{H} = \begin{pmatrix} 0 & V_1 & V_2 & \cdots \\ V_1 & 0 & V_1 & \cdots \\ V_2 & V_1 & 0 & \cdots \\ \vdots & \vdots & \vdots & \ddots \end{pmatrix} \tag{7}$$

This Hemiltonian can also be rewriten as a sum of translation operators D_x where $x = x_i - x_j$:

$$\mathcal{H} = \sum_{x=0}^{N/2} \frac{c}{x^\alpha}(D_x + D_x^*) = \sum_{x=0}^{N/2} \frac{2c}{x^\alpha} \cos(\hat{p}x) \tag{8}$$

if we go to the continum limit and assume an infinite number of sites we can rewrite the Hemiltonian for the $\alpha = 1$ case as:

$$E(k)_{\alpha=1} = -2cCi(ka) = 2c \int_{ka}^{\infty} \frac{\cos x'}{x'} dx' \tag{9}$$

and for the case of $\alpha = 2$:

$$E(k)_{\alpha=2} = 2c(\cos ka - \frac{1}{2}ka + kaSi(ka)) \tag{10}$$

and the corresponding group velocities are:

$$V(k)_{\alpha=1} = 2c\frac{\cos ka}{k} \tag{11}$$

$$V(k)_{\alpha=2} = 2c(1-a)\sin ka - ca + caSi(ka)/2 \tag{12}$$

It is easy to notice that in both cases showen above our initial wave packet will disperse, there for we do not expact to achieve a high value for $p_{out}(t)$. However, we are able to calculate a the time $\bar{t}$

in which the first and main part of the exitation will arrive to the output site at a distance $\frac{L}{2}$: For the first case of $\alpha = 1$ we would get an infinitly large velocity wich will resualt in $\bar{t}_{\alpha=1} \to 0$, for the case of $\alpha = 2$ one can calculate:

$$\bar{t}_{\alpha=2} = \frac{2}{L} V(0)_{\alpha=2} \approx \frac{L}{4ac} \tag{13}$$

Comparing this resualt to our original two-site benchmark time, $\bar{t}_0 = \frac{\pi L^2}{8c}$, one can easly see that the transport time for a coherent system of many sites is much shorter.

Bloch electron in a constant electric field:

We now look as a nearest neighbour interaction V within a constant electric field ϵ where we would be intrested in the probability distribution $|a_n(t)|^2$. The Schrodinger equation would be:

$$\frac{da_n}{dt} = -i\epsilon n a_n + V(a_{n+1} - a_{n-1}) \tag{14}$$

plugging $a_n = c_n e^{-i\epsilon n t}$ into the equation we get:

$$\frac{dc_n}{dt} = V(c_{n+1} e^{-i\epsilon t} - c_{n-1} e^{i\epsilon t}) \tag{15}$$

fourier transform will show that the equation is diagonal in k space:

$$\frac{dc_k}{dt} = V c_k (e^{-i(\epsilon t + k)} - e^{i(\epsilon t + k)}) = -i2V c_k \sin(\epsilon t + k) \tag{16}$$

$$c_k(t) = c_k(0) e^{-i2V \int_0^t \sin(\epsilon t' + k) dt'} = c_k(0) e^{i \frac{4V}{\epsilon} \sin(\frac{\epsilon t}{2} + k) \sin(\epsilon t)} \tag{17}$$

inverse transform would yeald:

$$c_n(t) = J_{n-n_0}(\frac{4V}{\epsilon} \sin \frac{\epsilon t}{2}) \tag{18}$$

and the probability distribution is:

$$p_t(n) = |J_{n-n_0}(\frac{4V}{\epsilon} \sin \frac{\epsilon t}{2})|^2 \tag{19}$$

we note that a Bessel function of the first kind would only give a value of 1 for $n - n_0 = 0$, since this is the trivial case where no transport had taken place and for any $n > n_0$ $p_{out} < 0.6$ we can conclude that this model does not provide a good transport effiecency.

The FMO model:

The FMO (Fenna-Matthews-Olson) complex carries absorved solar energy to a reaction centre, where it is transformed to chemical energy that fuels the bacterium. With only seven molecules praticipating in the excitation transport, it is the simplest and smallest light-harvesting structure in nature. Transfer of an excitation between two molecules of the light harvesting complex occurs by dipole-dipole interaction in the form:

$$V_{ij} = c|r_i - r_j|^{-3} \tag{20}$$

in this model we assume that the input and output sites are located on opposite poles of a sphere and the rest of the sites are located within this sphere. We neglect any angular dependency and define our potential to depend only on the distance between the sites.

We start by setting a benchmark for our transport time, $\mathcal{T}$, as the oscillation time between only two sites as mantion before:

$$\mathcal{T} = \frac{\pi}{2|V_{ij}|} \tag{21}$$

For the FMO case of only $N = 7$ sites we can solve the Hemiltonian analytically for 1D cases with reflaction symmetry.
for the case of evenly spread sites between the input and output we would get:

$$\bar{t} = 0.156\mathcal{T} \qquad\qquad p_{max} = 0.93 \tag{22}$$

However if we take a different configuration when the five middle sites are located at $(\frac{2}{8}, \frac{3}{8}, \frac{4}{8}, \frac{5}{8}, \frac{6}{8})$ of the way from the input to the output we would get:

$$\bar{t} = 0.038\mathcal{T} \qquad\qquad p_{max} = 0.999 \tag{23}$$

clearly a much more efficient configuration.

References:

[1]Gregory S. Engel, Tessa R. Calhoun, Elizabeth L. Read, Tae-Kyu Ahn, Tom Manal, Yuan-Chung Cheng, Robert E. Blankenship and Graham R. Fleming, Nature 446, 05678 (2007).

[2]A. Stotland and D. Cohen, J. Phys. A 39, 10703 (2006).

[3]T. Scholak, T. Wellens and A. Buchleitner, J. Phys. B 44, 184012 (2011).