Supplementary material to the paper:
Room-Temperature Biological Quantum Random Walk in Phycocyanin Nanowires

Table of Content:

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Dual Probe measurements</td>
<td></td>
</tr>
<tr>
<td>Scan 1</td>
<td>2</td>
</tr>
<tr>
<td>Scan 2</td>
<td>4</td>
</tr>
<tr>
<td>Scan 3 – detailed explanation about the measuring process</td>
<td>6</td>
</tr>
<tr>
<td>• Time resolved measurements</td>
<td></td>
</tr>
<tr>
<td>Comparison between several samples</td>
<td>10</td>
</tr>
<tr>
<td>Comparison between several Substrates</td>
<td>12</td>
</tr>
<tr>
<td>• Protein stability</td>
<td></td>
</tr>
<tr>
<td>Solution measurements</td>
<td>14</td>
</tr>
<tr>
<td>Dried sample measurement</td>
<td>15</td>
</tr>
</tbody>
</table>
Dual Probe measurements

Scan 1:
At this scan we have scanned a branch of Phycocyanin (PC) that goes from (15,10) to (12,0). The excitation probe was placed around (15,10.5). The sample was illuminated through 150 nm diameter tip with laser of 532 nm wavelength and 15 mW power. The actual power that excites the sample is much lower due to many losses along the fiber and an optic attenuator that was placed along the optic axis. Scanning was done with 250 nm diameter probe two times, first with no filter, to measure the excitation signal (left) and then with low pass filter of 600 nm to measure the luminescence (right). The excitation map is not uniform due to scattering of the light. The presented results are filtered with low pass filter to reduce the measurement fluctuations. In such scans there may be small movement between two scans and therefore we assume that the peaks should be coincided.

![Excitation at 532nm and Luminescence above 600nm](image)

A microscope photo of the scanned area:

![Microscope photo](image)

The black rectangle is the scanned area and the green spot is the place of the excitation.
4 cross-sections of these graphs are shown below:

$y$ – $y$ value of cross-section

$\Delta e$ – Full-width-half-maximum (FWHM) of excitation signal of maximum peak

$\Delta l$ – FWHM of luminescence signal of maximum peak

Comparison between the FWHM of excitation peak (green) and the FWHM of the luminescence peak (red). The cross-sections are parallel to the x-axis and their $y$ value is expressed by the horizontal axis of this graph. It can be seen that the excitation FWHM is around 1.5 $\mu$m and that the FWHM of the luminescence is around 3-4 $\mu$m.
Scan 2:
At this scan we have scanned a branch of PC that going diagonally from (10,10) towards (5,5). The excitation probe is placed around (10.5,10.5) and the excitation map (left) is scattered light. The luminescence map (right) shows clear broadening of the signal. In this measurement we should take into account that the branch direction is diagonally and that the cross-sections are parallel to the Y axis and therefore the broadening should be greater in squared two.
4 cross-sections of these graphs are shown below:

\[ x \text{ – x value of cross-section} \]

\[
\begin{align*}
\text{Intensity (normalized)} \\
\text{Y (um)} & \\
0 & 2 & 4 & 6 & 8 & 10
\end{align*}
\]

\[
x=9.97\mu m \Delta e=0.77\mu m \Delta l=2.85\mu m
\]

\[
x=9.71\mu m \Delta e=0.64\mu m \Delta l=2.9\mu m
\]

\[
x=9.07\mu m \Delta e=1.32\mu m \Delta l=2.54\mu m
\]

\[
x=8.69\mu m \Delta e=1.07\mu m \Delta l=2.92\mu m
\]

In this case the FWHM of the luminescence contain more than one peak and therefore the comparison is not trivial, nevertheless the luminescence signal is much wider.
Scan 3:
Description of the dual probe measurement process:

1. A microscope image of dried sample of PC that contain many PC branches:

2. The right probe is the excitation probe (in this case 200 nm diameter) and the left probe is the detection probe (in this case 300 nm diameter):
3. The two probes are brought into contact with the sample and each other:

![Image 1](image1.png)

4. The excitation probe is placed over a branch of PC:

![Image 2](image2.png)

5. Sample scan without any filter. Such scanning gives us two maps: optical map and topography map (AFM). Raw data of topography in 2D:

![Image 3](image3.png)
6. The data is flattened to reduce scanning noises. 3D image of the topography after flattening:

![3D image of topography after flattening]

7. Similar process is done with the optic map:

![3D image of optic map after flattening]
8. Than we perform second scan with low pass filter that pass wavelengths above 600 nm to collect the luminescence. In this scan we scanned only 0.35 μm at the y-axis because we wanted to measure the broadening along the x-axis. We used special low pass filter to filter out the noise:

9. Now we take a cross-section both in figure 7 and 9 and compare the signal broadening:
Time-Resolved measurements:

Comparison between several samples

Time-Resolved measurements show the difference in life-time of four different samples. A solution of PC (red curve) shows life-time of 1.15 ns while two dried samples (blue curve) show life-time of 0.4 ns. Semi-dried sample (purple curve) exhibits an intermediate result of 0.6 ns. Additional measurement was done using PC solution that contained salts. We believe that the salts change the distance between the proteins and therefore it changes the quantum coupling. Indeed the life-time of the samples with the salt is around 0.85 ns (green curve).
This graph shows the same results where the vertical axis starts at $10^5$. Below $10^{-3}$ the noise is high and there is an artificial peak at rise time. This peak is caused by the excitation laser and is three orders of magnitude lower than the main peak. At low intensity below $10^{-3}$ some multi-exponential behavior may be present.
Comparison between several substrates

The life-time shortening of the ordered samples is presented for different substrates. Five samples are presented; wet PC (in cuvette) and four dried samples over four different substrates, glass, silicon, gallium-arsenide and gold. The graph shows a big difference in the life time between the first sample and the other four. All four dried ordered samples life times are similar ruling out the quenching mechanism as the explanation for the life time shortening. In case of quenching we would except to see different life-time for different substrates. The factor of 3 shortening in the life time fit the expected super-radiance factor for nine nearest neighbors. Nine nearest neighbors are achieved in a closely packed two dimensional sheet of a rectangles that reasonable in a top view our system (see image below).
Microscope images show the difference between PC dendrite with salts and without:

Sample of PC with salts  
Sample of PC without salts

The images above show that sample with salts appears different than sample without salts. With salt the dendrites are less curvy and organize in a straight line. The image below shows semi-dried sample of PC (without salts). In this sample curve dendrites can be seen and wet PC fills the gap in between.
Protein stability

Solution measurements

Spectral measurements of the PC solution were done in different laser intensities (532 nm wavelength). The normalized spectrums are identical even at high power above 100 mW. The laser spot diameter is ~1 mm.
Additional measurements with dried sample show similar results. The sample is not affected by high power of laser.