# Direct Visualization of Exciton Reequilibration in the LH1 and LH2 Complexes of *Rhodobacter sphaeroides* by Multipulse Spectroscopy

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## Supporting Information

## Model description

A simple description of the disordered exciton model for LH1 and LH2 rings is depicted in figure 7, together with a schematic interpretation of the hopping mechanism of wavepackets short excitation pulses can generate in these ring systems.

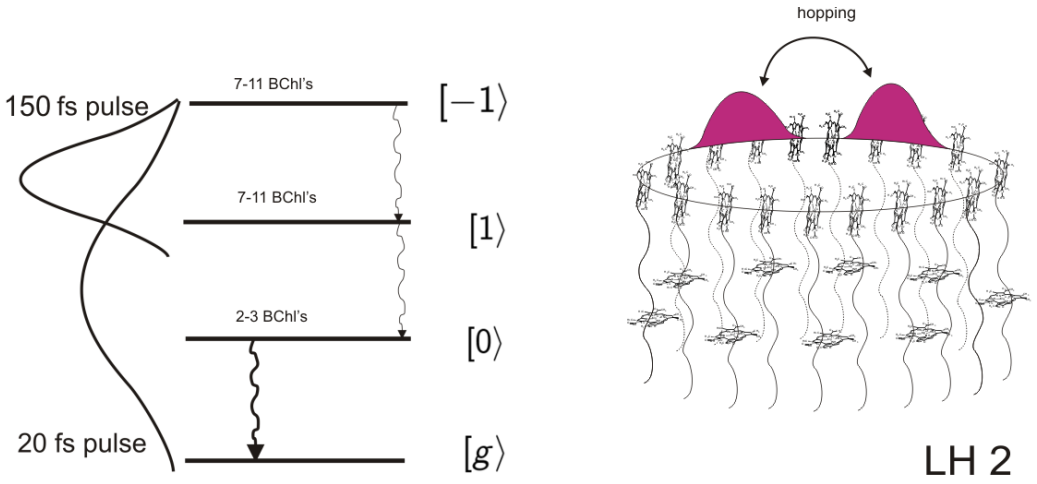


*Figure 7: The lowest exciton level (k = 0) is a superradiant excitonic level, involving 2-3 closely coupled bacteriochlorophylls. The higher lying exciton levels, (k = 1 or -1) normally involve 7-11 bacteriochlorophylls. Depending on the pulse length one typically excites the complete exciton manifold with ultrashort pulses and only one or two levels with longer (~150 fs) pulses. The model includes a hopping type energy transfer of excitonic wavepackets*

*across the ring. This is schematically drawn on the right, where hopping occurs from one cluster of 3-5 bacteriochlorophyll to another on a few 100 fs time scale.*

In figure 8 a schematic representation is given of the pump dump probe experiment.

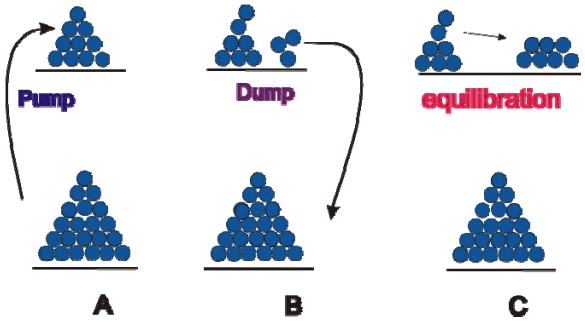


*Figure 8: A simple schematic representation of a pump-dump experiment. (A) The pump pulse excites population from the ground state to the excited state manifold. (B) The dump pulse takes away a part of the excited state population. (C) A re-equilibration of the excited state takes place.*

**Pump-probe spectroscopy**

Transient pump-probe spectra for LH1 and LH2 are represented in figure 9.

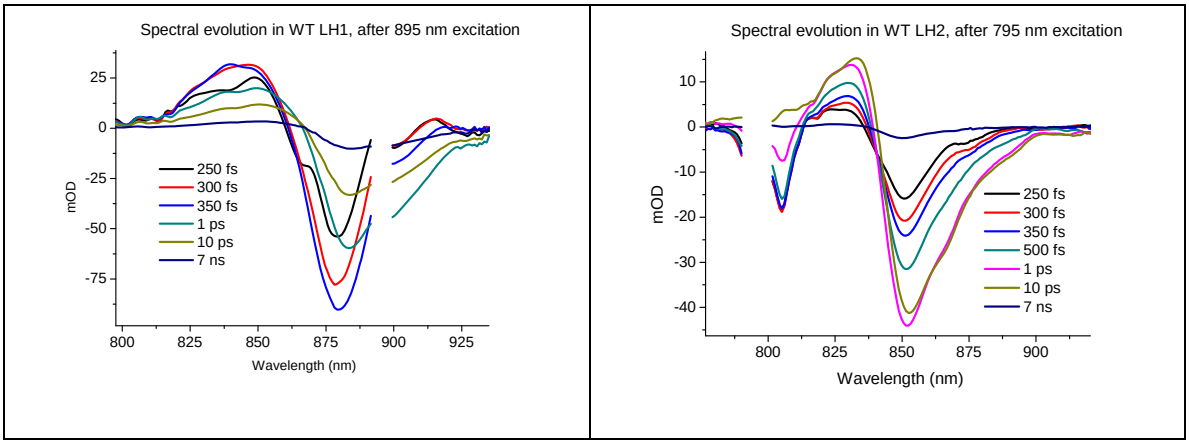


*Figure 9: Time-gated transient pump-probe spectra of LH1 and LH2. These spectra are in good agreement with earlier measurements [16, 50, 61] by and primarily shown for reference purposes. The pump-scatter is removed around 895 and 795 nm.*

The pump-probe spectra shown in figure 9 are in comparison with spectra measured previously by other authors [16, 50, 61] The powers used (10-20 nJ) produce a high amount of excited state population in both LH1 and LH2, needed to be able to dump enough of this excited state population. As a consequence however, the measurements are not annihilation-free and estimations of the excited state lifetimes cannot be made. Annihilation has strong implications on the lifetime of the excited states involved, especially in these well-coupled ring systems [62].