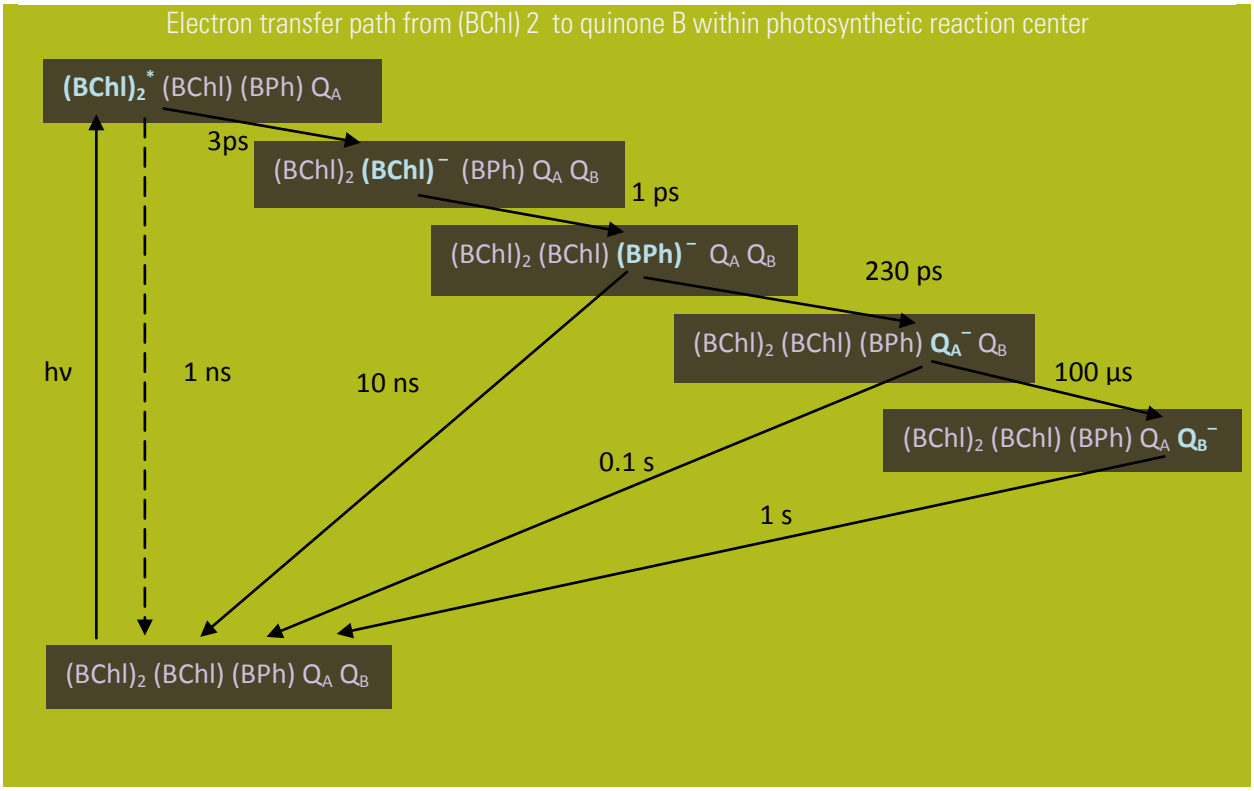
Electron transfer may happen in two different scenarios: an excited electron 'jumps' from a donor, D, to an acceptor, A, within one molecule or between two different molecules. In the first case, D and A are two parts of the same molecule, bound together in a 1-to-1 complex. In the second case, D and A are not bound, whose concentrations may vary with respect to each other.

Electron transfer reactions are important for converting light into electrical energy. They are typically described by electron transfer of the first type and the examples include photosynthetic reaction centers in plants and semiconductor photovoltaic devices.

SimphoSOFT is capable of modeling electron transfer of the fist type by using absorption and relaxation transition modules. It helps to gain more insight on processes happening during electron transfer on a macroscopic level.

Example SimphoSOFT simulation of intramolecular electron transfer: photosynthetic reaction centers

There are many types of photosynthetic reaction centers utilized by plants and bacteria for photosynthesis. The reaction center of the bacteria *Rhodopseudomonas viridis* has been well studied experimentally.[1] The electron transfer path is shown in figure below.



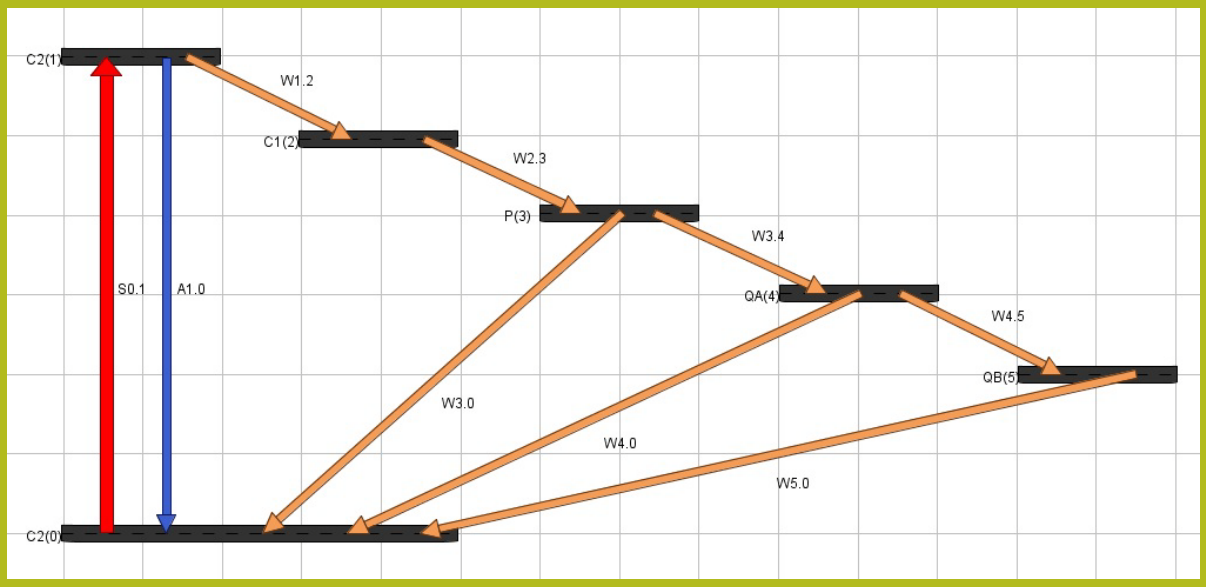
An electron in a bacterial chlorophyll dimer, labeled $(BChl)_2$ in figure above, is excited by approximately 960 nm light to an excited state of the dimer. In approximately 3 ps, the excited electron transfers to a bacterial chlorophyll

[1] Feher, G.; Allen, J.; Okamura, Y.; Rees, D. "Structure and function of bacterial photosynthetic reaction centres", Nature 339, 111-116 (1989).

monomer, BChl, followed by transfer to a bacterial pheophytin, BPh or P, in approximately 1 ps. From pheophytin, the electron transfers to quinone A (QA) in 230 ps and then to quinone B (QB) in approximately 100 µs. There are several possible back reactions to the ground state that are also shown in the figure.

SimphoSOFT can model electron transfer when acceptor and donor are bound together in a single complex. The complex can be simulated as one type of molecule with one concentration (number of complexes per unit volume). The relaxation transition modules can then be used for electron transfers within the complex. The sum of the electronic populations of the filled and unfilled states involved in the electron transfer within the complex is a constant that can be normalized to 1 in the simulation.

Screenshot of SimphoSOFT® M-CAD with energy level diagram of photosynthetic reaction center containing relaxation transition modules which mimic electron transfer



SimphoSOFT user creates 6 energy levels, each representing an energy state which the photosynthetic reaction center may occupy according to the current location of the excited electron: ground state [$(BChl)_2$ (BChl) (BPh) Q_A Q_B] is labeled C2(0), the state [$(BChl)_2^*$ (BChl) (BPh) Q_A Q_B] with an excited electron in $(BChl)_2$ is labeled C2(1) etc (compare it with the electron transfer path figure for the rest). Electron traveling is modeled by relaxation transitions (W1.2, W2.3, W3.4, W4.5).

Cross-sections and relaxation times:

6-level model

From level:	To level:	Transition parameter:
		Single-photon absorption cross-section:
C2(0)	C2(1)	1×10^{-16} cm^2

		Electronic transfer times:
C2(1)	C1(2)	3 ps
C1(2)	P(3)	1 ps
P(3)	QA(4)	230 ps
QA(4)	QB(5)	100 µs

		Relaxation times:
C2(1)	C2(0)	1 ns
P(3)	C2(0)	10 ns
QA(4)	C2(0)	0.1 s
QB(5)	C2(0)	1 s

Numerical Setup parameters		
Temporal domain	$10 \times T_0$	
Radial domain	$5 \times R_0$	
Propagation domain	1 mm	0.11 L_{df}
# Time samples	1024	
# Radial samples	32	
# Propagation samples	128	

Diffraction is turned OFF as the propagation length is much less than a diffraction length, L_{df}.

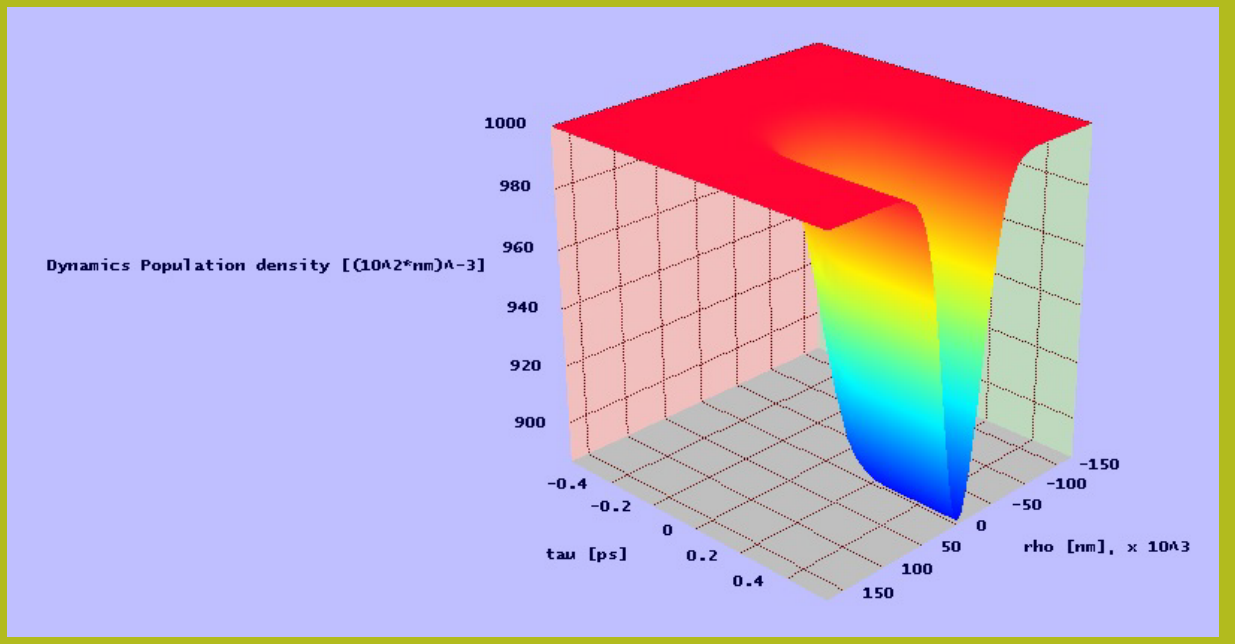
Other sample properties	
Complex density (concentration) in the host material	1.0×10^{18} complexes/cm^3 (1.0×10^{-3} / nm^3)
The host material linear refractive index	$n_0 = 1.4$
Host material linear absorption	$\alpha = 0.075$ cm^{-1}
Sample length	1 mm

Laser beam properties	
Pulse energy	1 µJ
Pulse radius (HW1/e^{2}M)	50 µm
Pulse FWHM	0.2 ps
Wavelength	960 nm

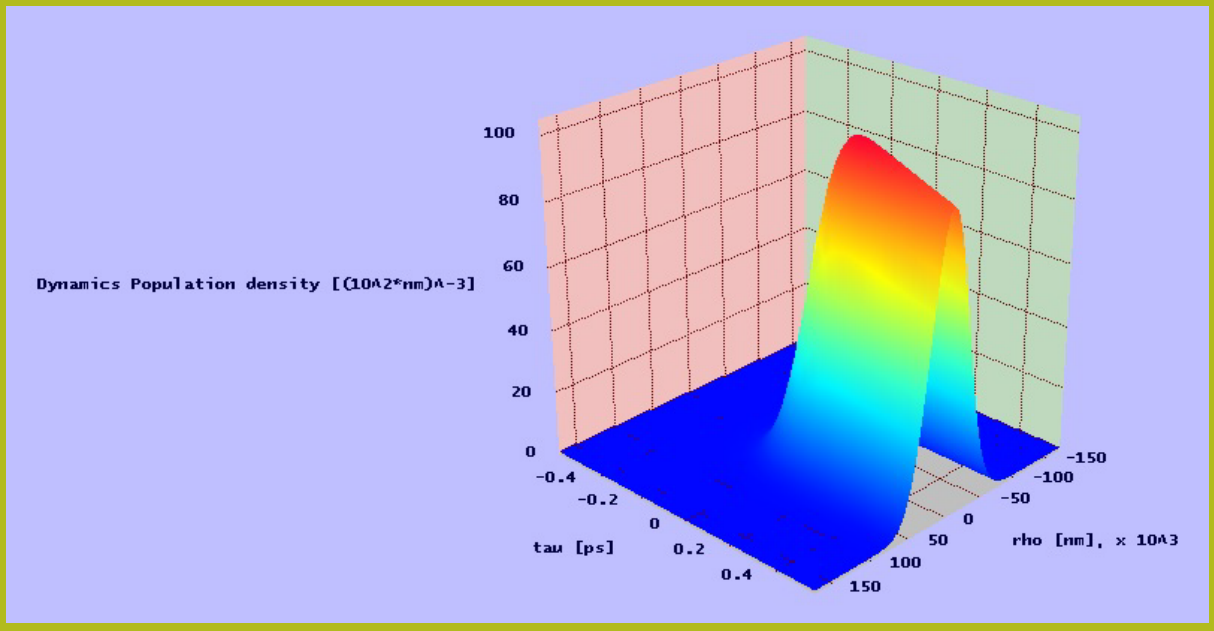
Results of SimphoSOFT simulations of photosynthetic reaction: within 1 ps time interval

Energy level populations are shown below during a 1 ps time interval that includes the 0.2 ps laser pulse. All the figures contain populations densities calculated across the radial domain 'rho [nm]' (incident beam is Gaussian), so that the user can see where electron transfer has the biggest impact.

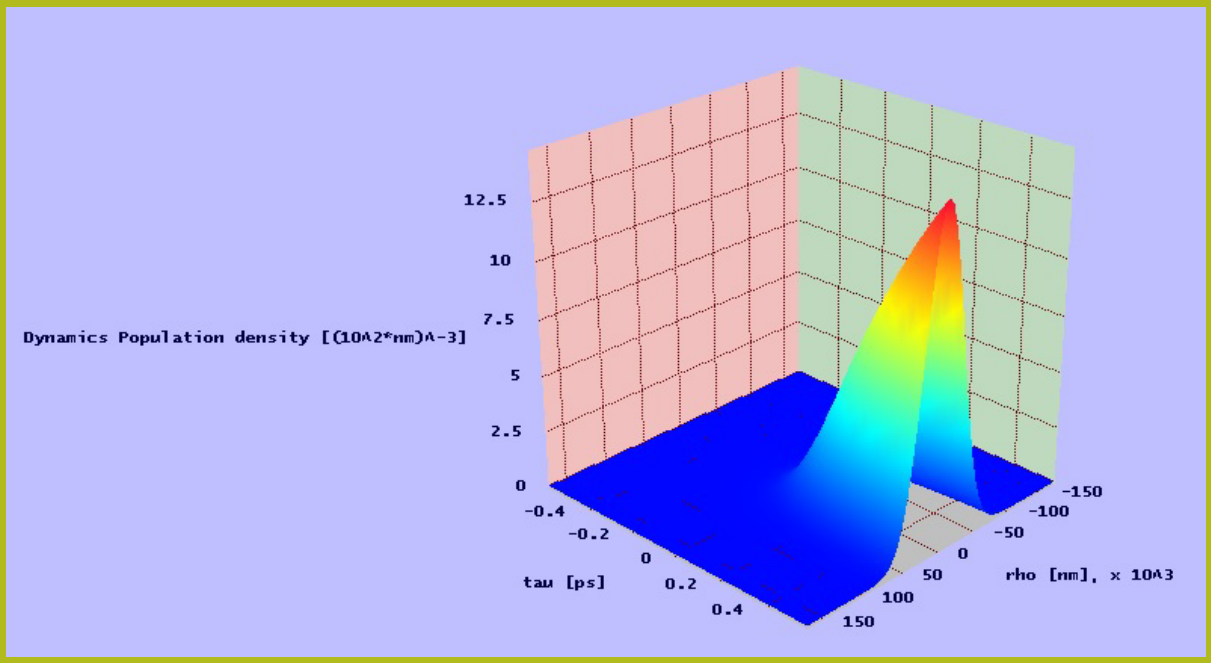
Ground state energy level C2(0) is partially depopulated.



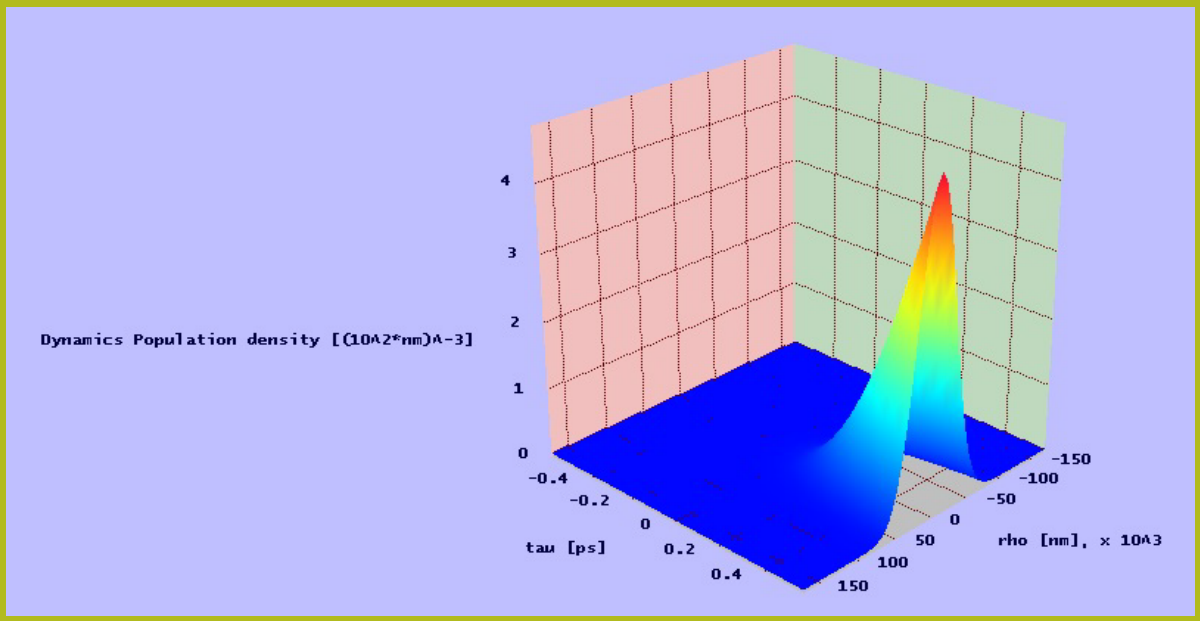
Most of the excited electrons are in energy level C2(1) during and immediately after the 0.2 ps laser pulse.



A few electrons have started to transfer to the chlorophyll monomer, which includes energy level C1(2).

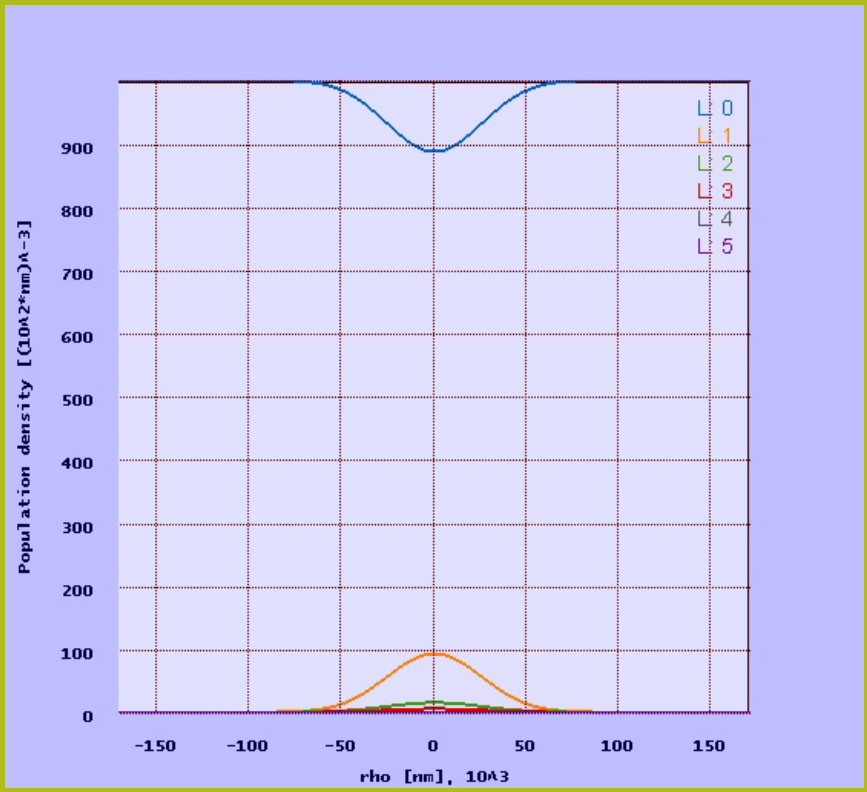


Very few electrons have made it to the pheophytin, energy level P(3).



The energy level populations at the sample input surface immediately after the laser pulse has passed through the sample are shown below as a function of the radial distance from the center of the pulse. Level 0 of the chlorophyll dimer is partially depleted at the center of the pulse and most of the electrons are still in the excited energy level 1 of the chlorophyll dimer and have not had time to transfer to the other energy levels of the reaction center complex.

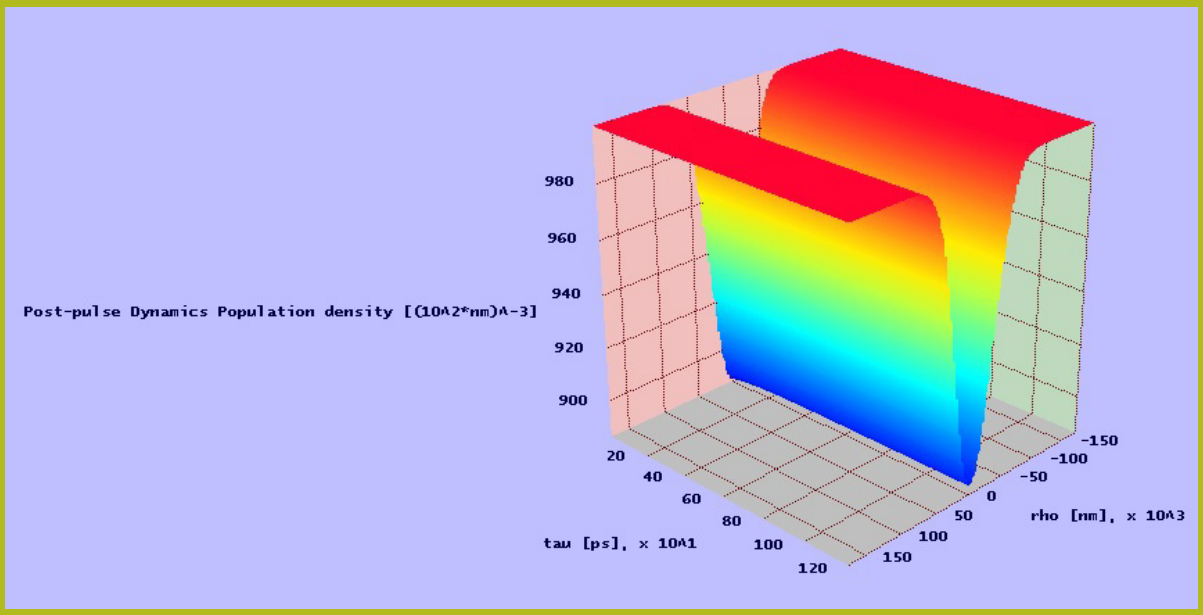
Most of the molecules in the center of the beam are in the excited energy state C2(1)
right after 0.2 ps pulse passes the sample



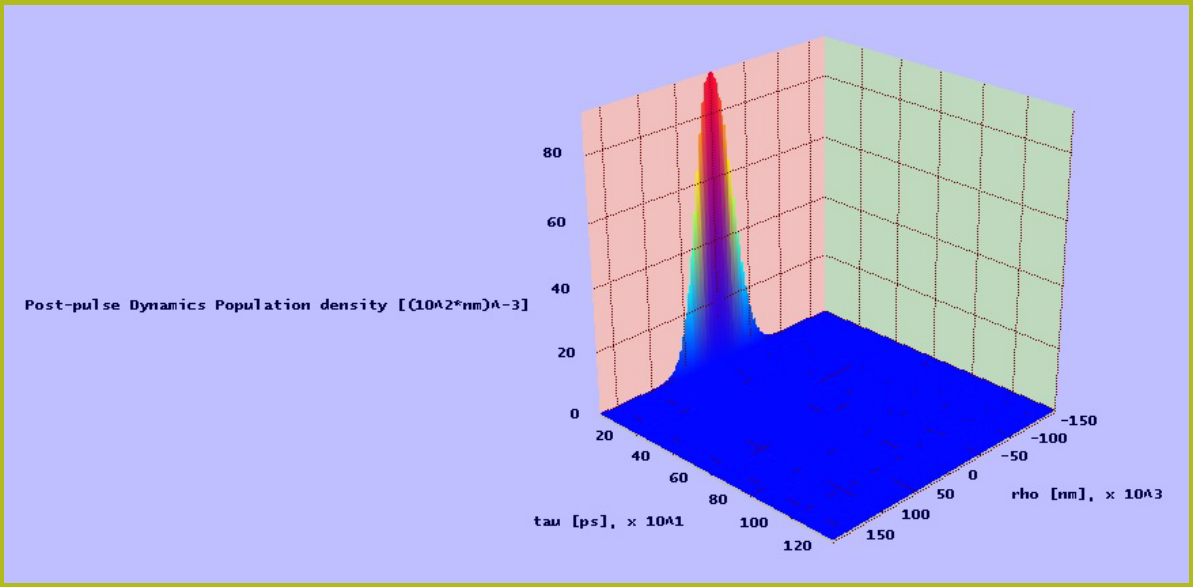
Results of SimphoSOFT simulations of photosynthetic reaction: within 1,200 ps time interval

One can also examine the kinetics of electron transfer after passage of the laser pulse. Below are illustrated the energy level populations for a time period covering the first 1200 ps after the laser pulse has passed through the sample.

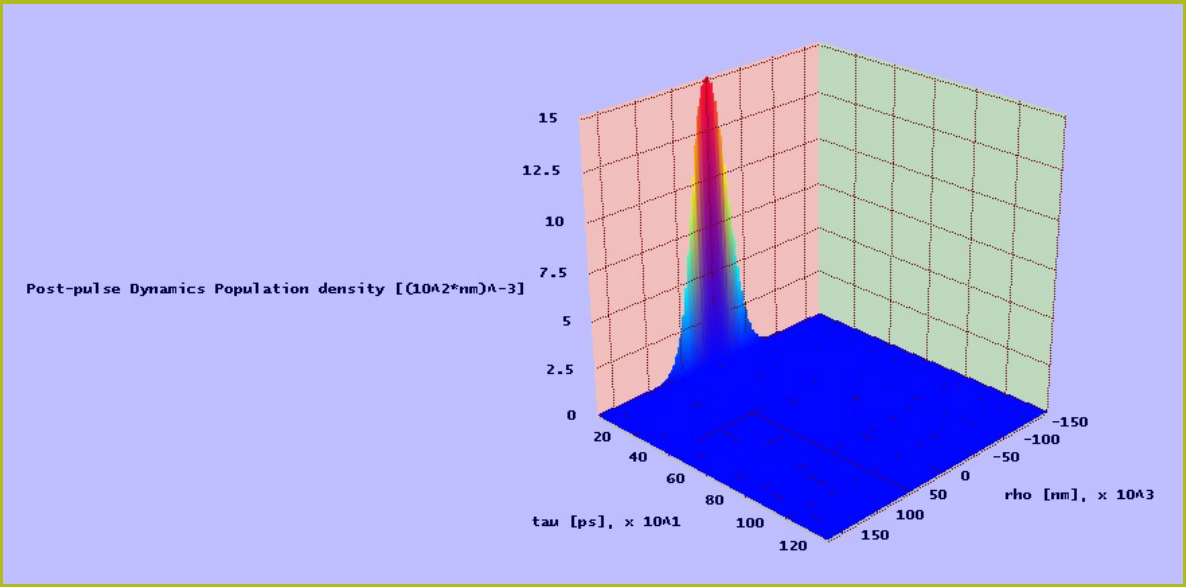
Energy level C2(0): remains partially depleted in the center of the laser pulse.



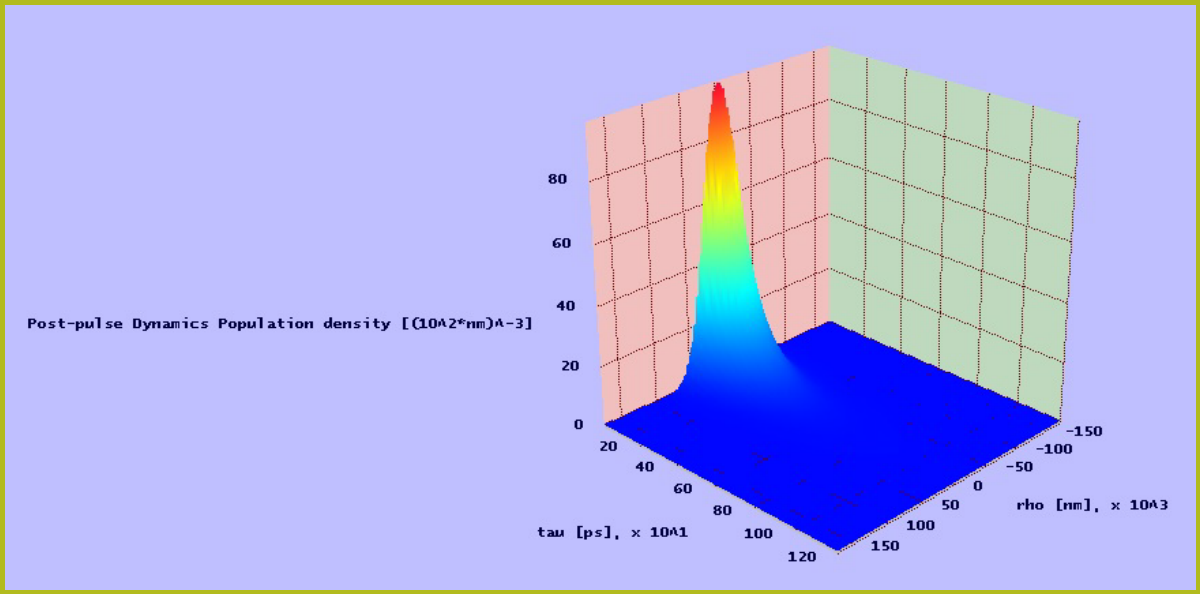
Energy level C2(1): Electrons that were excited to energy level C2(1) have transferred to levels C1(2) through QB(5).



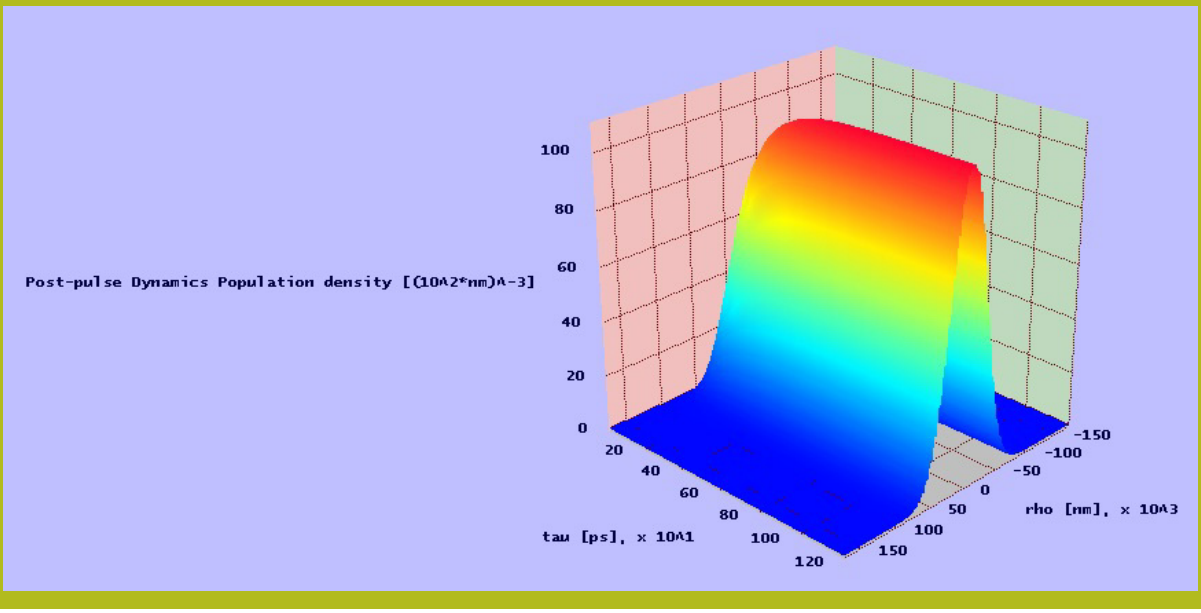
Energy level C1(2): Electrons that were transferred to energy level C1(2) have moved on to levels P(3) through QB(5).



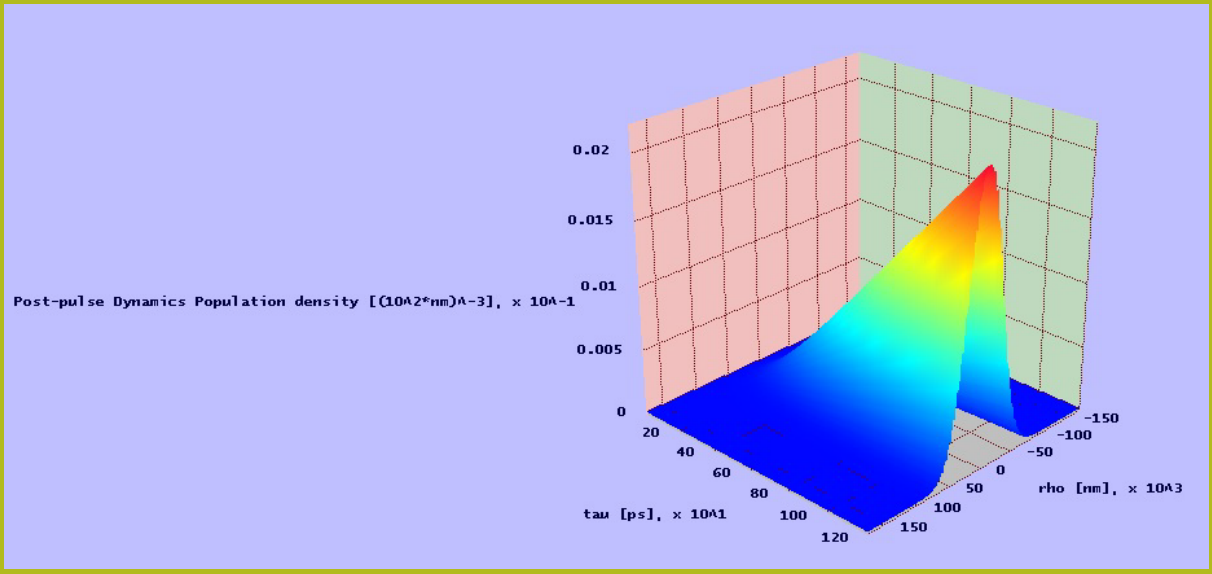
Energy level P(3): Electrons that were transferred to energy level P(3) have moved to levels QA(4) and QB(5).



Energy level QA(4): Most of the electrons that arrived at energy level QA(4) are still in energy level QA(4).



Energy level QA(5): At the end of the first 1200 ps after the laser pulse, energy level AQ(5) is just starting to fill.



The above example and figures illustrate how SimphoSOFT can simulate electron transfers in complicated systems such as photosynthetic reaction centers and provide very informative graphical outputs illustrating the electron transfer processes.

211 Warren Street, Newark, NJ 07103 support@simphotek.com (973) 621-2340

www.simphotek.com

simphotek

Light Interactions Matter