# Supporting Information

# Ultrafast Energy Transfer in Chlorosome Probed by Femtosecond Pump-Probe Polarization Anisotropy†

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## Experimental

The 800 nm pulses of 50 fs duration were generated from the 1-kHz regenerative amplified Ti:sapphire laser (Coherent Legend Elite seeded by Vitesse) and were converted to the visible pulses of 720 nm center wavelength and 60 nm bandwidth (see Figure S1) by a home-built, noncollinear optical parametric amplifier (NOPA) based on all-reflective optics design. A prism-pair compressor compensated the dispersion of the transmissive optics in the NOPA and the pump-probe spectroscopy setup, giving the pulse duration of 15 fs at the sample position. The output from NOPA was sent to an experimental setup that was originally built for two-dimensional electronic spectroscopy but is able to perform wavelength-resolved pump-probe spectroscopy as well. An input beam to the setup was focused on a diffractive optic element (DOE; Holoeye DE228) and split into four beams, which are the ± 1 orders of diffracted light. Two of the four beams were blocked by a mask and the remaining two beams were used as pump and probe pulses. The two beams were focused into the sample by a concave spherical mirror. The time delay between the two pulses, called population time $T$, was controlled by a pair of wedges inserted in antiparallel orientation. By translating only one of the wedge pair using a motorized stage, the time delay can be changed with the resolution of 2.7 attoseconds. The polarization of each pulse was controlled by a half-wave plate. The polarization pump-probe signals were measured with the polarizations of the pump and probe pulses parallel ($S_{\parallel}$) and perpendicular ($S_{\perp}$) to each other. The signal was dispersed by a spectrograph and detected using a charge-coupled device (CCD) detector with $1600 \times 400$ pixels (Andor, Newton). The population time $T$ was scanned from –50 fs to 400 fs. From the measured polarization pump-probe signals, $S_{\parallel}$ and $S_{\wedge}$, we obtained the transient polarization anisotropy, $r(T)$, using Eq. (1) in the manuscript.

Chlorosome was extracted from *Cba. Limnaeum*, which consists of BChl *e* molecules, and prepared in solution using 50 mM Tris–HCl buffer with $Na_2S_2O_4$ added as reductant. The optical density of the sample solution was 0.3 in a cell of 1 mm optical path length. During the measurement, the sample was sealed under nitrogen and kept at room temperature. The absorption spectrum of the sample is shown in Figure S1, where the band centered at ~720 nm corresponds to $Q_y$ band of the BChl *e* aggregate.

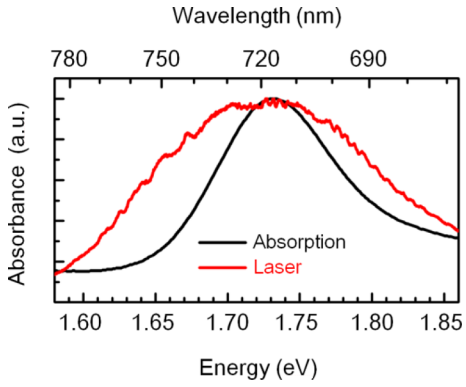


**Figure S1.** Absorption spectrum (black line) of chlorosome from *Cba. limnaeum* measured at room temperature, and the spectral profile of the laser pulse (red line) used for the measurement. The absorption band centered at ~720 nm is ascribed to the $Q_y$ transition of the BChl *e* chlorosome.

†This paper is to commemorate Professor Myung Soo Kim's honourable retirement.