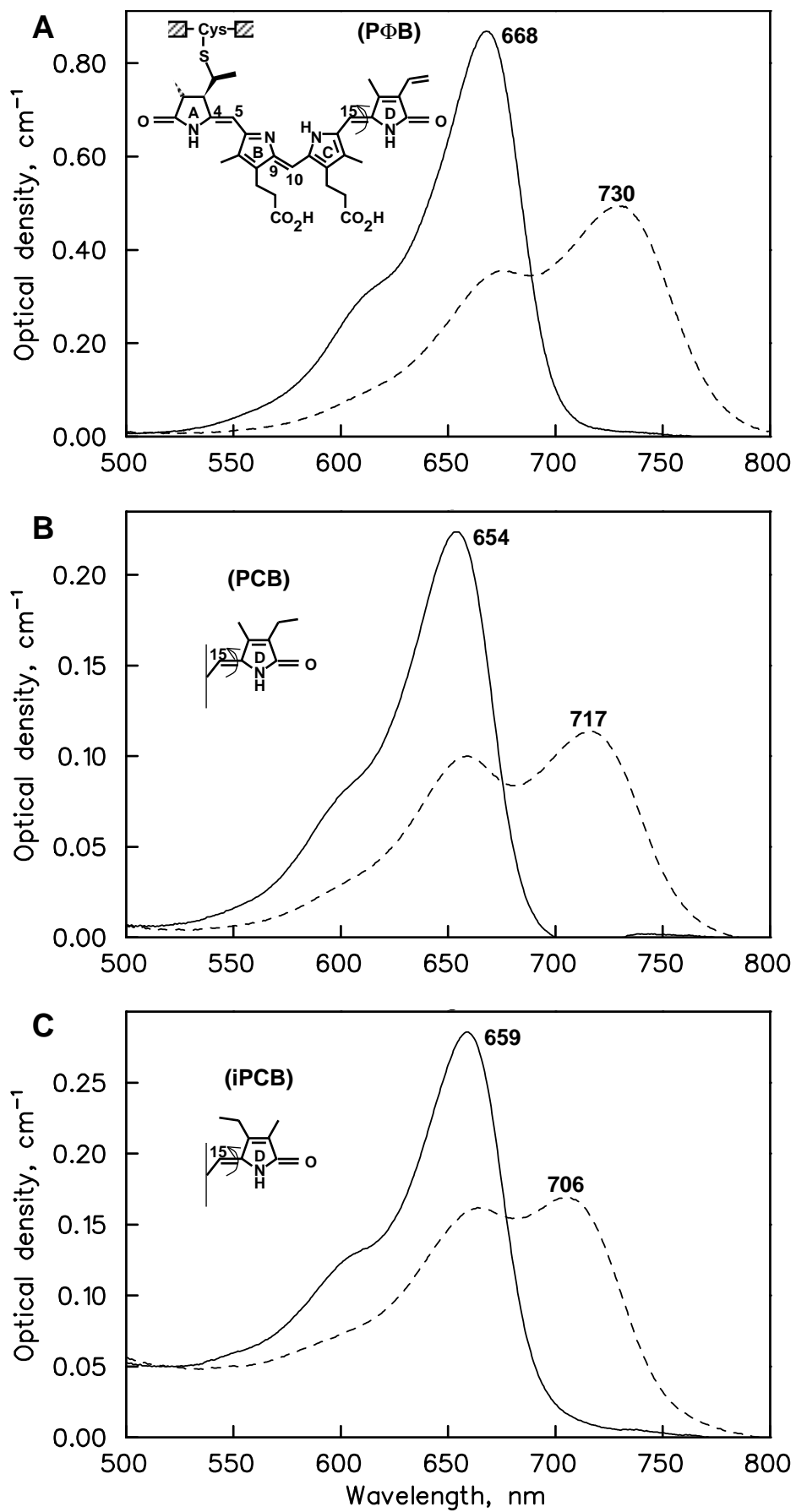


## Supplementary Material

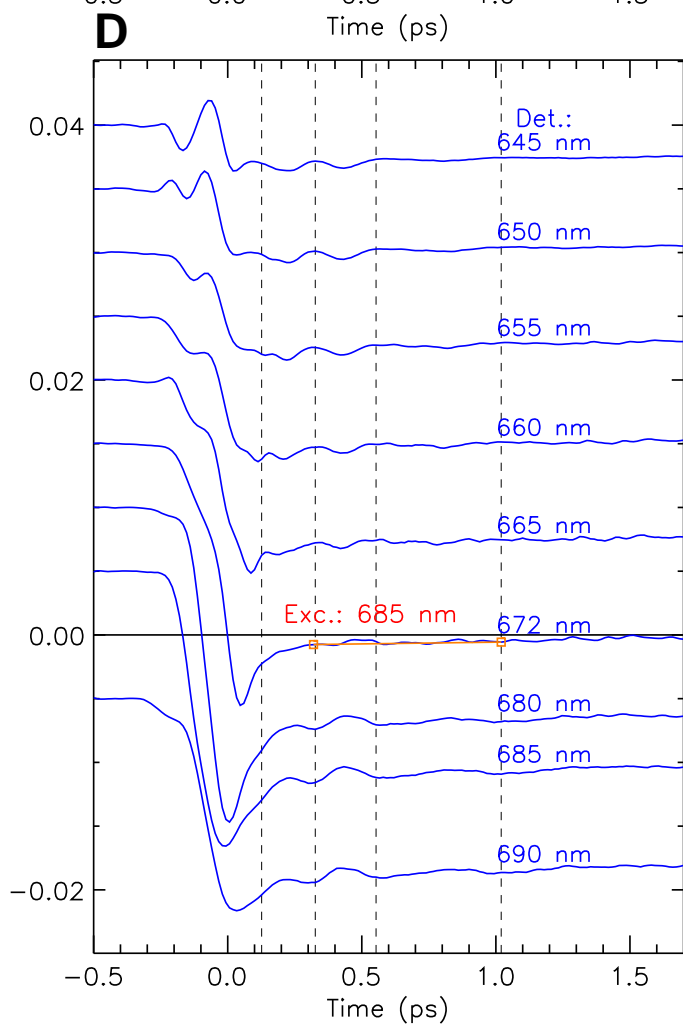
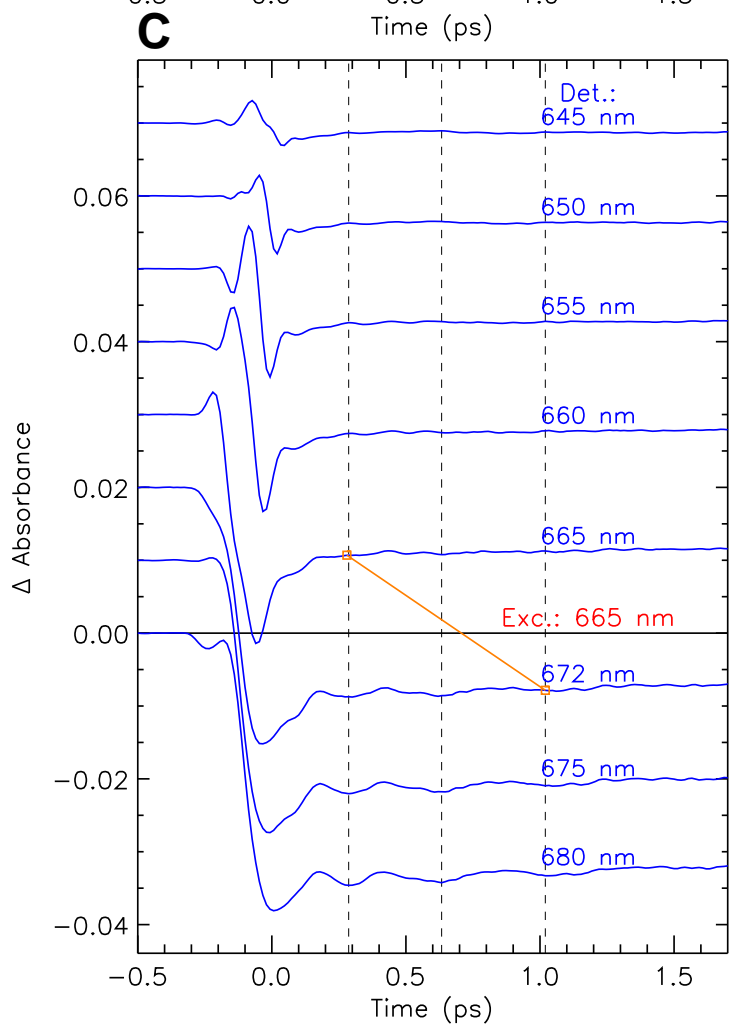
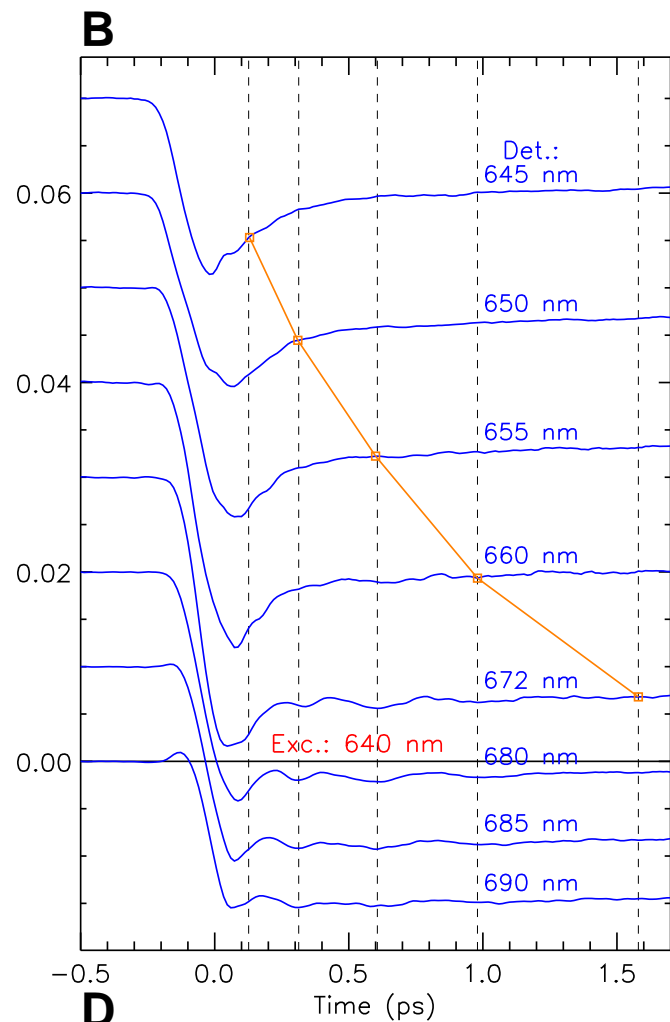
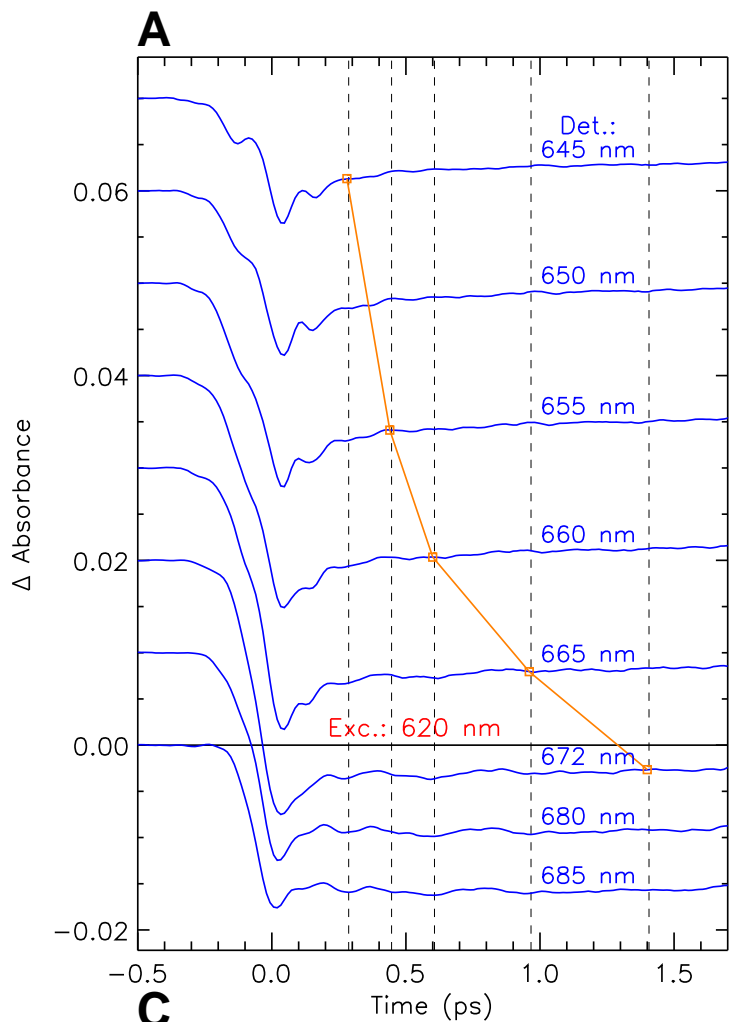
**Fig. S1.** Stationary absorption spectra of (A) native phyA with PΦB and recombinant phytochromes assembled with (B) PCB-65kDa-phy and (C) iso-PCB-65kDa-phy in their P<sub>r</sub> forms (solid lines) and P<sub>fr</sub> forms (dashed lines). Partial structures of the three different tetrapyrrole chromophores, phytochromobilin (PΦB), phycocyanobilin (PCB) and iso-phycocyanobilin (iso-PCB) are also shown at the respective spectra. It is assumed the photoisomerization from Z,Z,Z to Z,Z,E configuration upon illumination with red light occurs along the 15-16 double bond as indicated. Note, all samples can be interconverted to the P<sub>fr</sub> form and back by illumination with red or far-red light, respectively.

**Fig. S2.** Excitation wavelength dependence of the vibrational oscillations observed for the P<sub>r</sub> form of native phyA. Excitation wavelengths are 620 nm (A), 640 nm (B), 665 nm (C) and 685 nm (D).

**Fig. S3.** Strongly overdamped vibrational oscillations observed for the P<sub>r</sub> forms of (A) PCB-65kDa-phy and (B) iso-PCB-65kDa-phy both excited at 660 nm. As compared to native phyA (Fig. S1), the amplitudes of the oscillations are reduced and the damping is more pronounced.



**Fig. S1**



**Fig. S2**

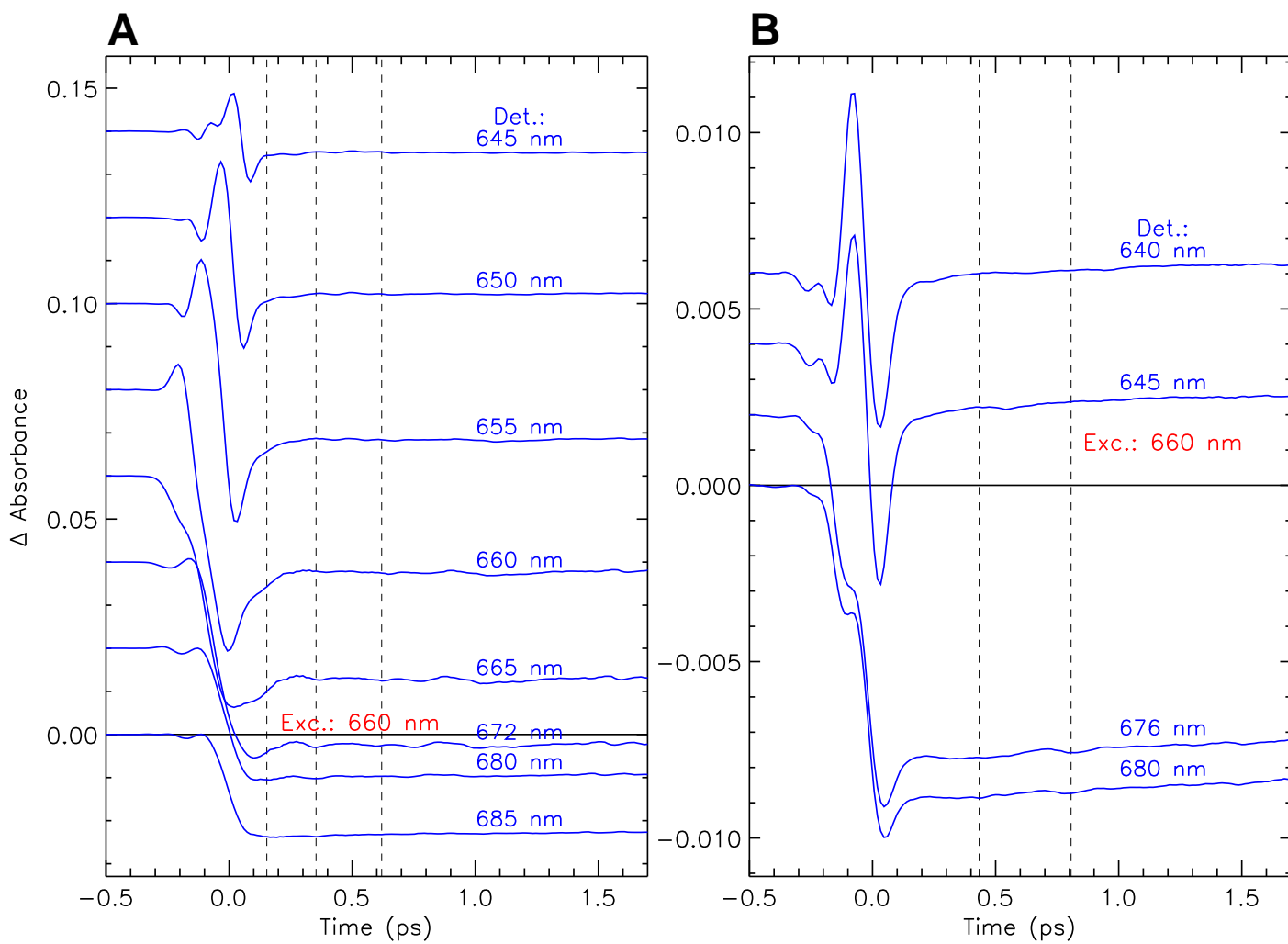


Fig. S3