## **Supporting Information for Publication**

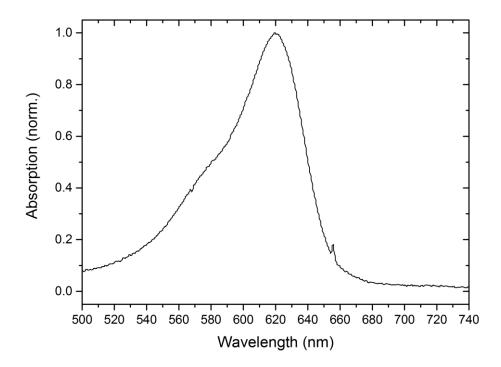
## Phycocyanin: One Complex, Two States, Two Functions

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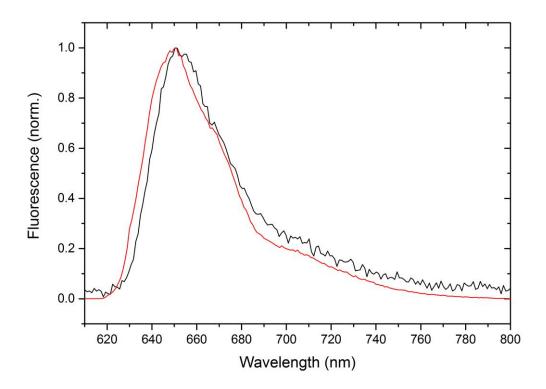
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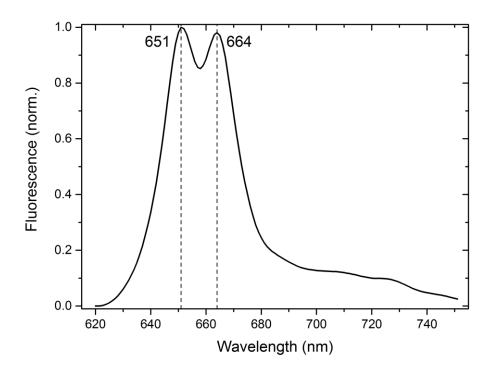
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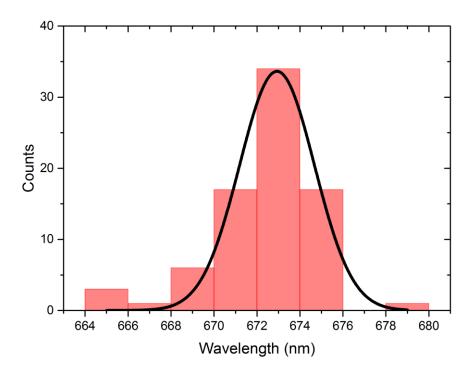
**Figure S1.** Absorption spectrum of PC rods used in this study, characterized by the main peak position below 620 nm and a blue shoulder.



**Figure S2.** Normalized bulk fluorescence emission spectrum of PC rods corrected with the wavelength-dependent transmission efficiency of the optical components used between the microscope and detector in the SMS setup (red). Normalized, weighted average of single molecule fluorescence spectra (black) collected in the same setup. The blue, red and double spectra from Figure 2 were weighted by their frequency of occurrence, i.e. 74.2%, 11.3% and 14%, respectively. The total number of complexes considered was 191.



**Figure S3.** Steady-state fluorescence emission spectrum at 77 K of PC isolated from the ΔAB mutant of *Synechocystis* upon excitation at 590 nm. Spectrum was normalized to 1 at the emission maximum. Peak maxima are indicated (in nm). PC complexes were solubilized in 0.8 M K-phosphate at pH 7.5, which contained ~0.5 M sucrose from a sucrose gradient.



**Figure S4.** Distribution of peak positions like in Figure 4 for the red band, fitted with a Gaussian (black). Bands of the fluorescence intensity falling within the top 70% were included. The average peak position was at 672.9 nm (SE = 0.1 nm). For details see the main text.