

## Description of Additional Supplementary Files

**File name:** Supplementary Data 1

**Description:** Figure\_1.xlsx. Normalized absorption spectra of different OCP variants in 0.8 M phosphate, presented in Figure 1. The tabs are labeled according to the panels in the figure.

**File name:** Supplementary Data 2

**Description:** Figure\_2.xlsx. Time-courses of the dihedral angle ( $\chi_2$ ) between the peptide backbone and the indole ring of Trp-201 as revealed by molecular dynamics simulations of OCP<sup>WW</sup> with different initial conformation of Trp-201.

**File name:** Supplementary Data 3

**Description:** Figure\_3.rar. Archive contains transient absorption spectra of WT OCP and OCP<sup>WW</sup> at different time delays.

**File name:** Supplementary Data 4

**Description:** Figure\_5.xlsx. Calculated and measured Raman spectra of ECN in regular and protonated form. Normalized steady-state absorption of ECN and CAN oxocarbenium ions obtained by protonation of carotenoids using 1 M trifluoroacetic acid (TFAA) in chloroform. The tabs are labeled according to the panels in the figure.