Supplementary Information

Analytical data of SiPc oligomers

(a) SiPc dimer, HO(SiPcO)₂H:

Anal. Calcd for C₉₆H₉₈N₁₆O₃Si₂: C, 72.97; H, 6.25; N, 14.18. Found: C, 73.95; H, 6.75; N, 13.19.

MALDI-TOF-mass m/z = 1579.1 (M⁺), UV-vis (Toluene) λ /nm (log ϵ) 639.5 (5.5), 342.0 (5.1)

(b) SiPc trimer, HO(SiPcO)₃H:

Anal. Calcd for C144H146N24O4Si3: C, 73.25; H, 6.23; N, 14.24. Found: C, 73.32; H, 6.65; N, 13.24(4).

MALDI-TOF-mass m/z = 2359.6 (M⁺), UV-vis (Toluene) λ /nm (log ϵ) 626.5 (5.5), 335.5 (5.4)

(c) SiPc tetramer, HO(SiPcO)₄H:

Anal. Calcd for C₁₉₂H₁₉₄N₃₂O₅Si₄: C, 73.39; H, 6.22; N, 14.26. Found: C, 74.06; H, 6.27; N, 13.32.

MALDI-TOF-mass m/z = 3139.1 (M⁺), UV-vis (Toluene) λ /nm (log ϵ) 621.5 (5.6), 330.5 (5.3)



Fig. S1 Ground-state absorption spectra of SiPc oligomers (dimer: red line, trimer: green line, tetramer: blue line) in toluene in addition to a spectrum of the corresponding monomer (black line). The Q band peak (monomer: 678 nm, dimer: 639.5 nm, trimer: 626.5 nm, tetramer: 621.5 nm) shifts to the blue-side with increasing the number of SiPc units, which is explained by the exciton interaction. The ε values of the Soret and Q band increase due to oligomerization reflecting an increase in the number of SiPc units, whereas the ε value of the Q band is slightly smaller in the trimer than the dimer because of the broad bandwidth of the trimer.



Fig. S2 Transient difference absorption spectra of SiPc oligomers (a: dimer, b: trimer, c: tetramer) in toluene.



Fig. S3 Time-profiles of transient absorption signals of the dimer (**a**), trimer (**c**), and tetramer (**d**), respectively. The T₁ lifetime decreases in the order monomer (500 μ s) > dimer (150 μ s) > trimer (73 μ s) > tetramer (40 μ s).



Fig. S4 Relationships between the incident light (I_{in}) and $\alpha (= \alpha_0 + \beta I_{in})$ at 532 nm (\blacktriangle) and for the excited-state Q band (\bigcirc : 720nm, 670nm, and 660nm for the dimer, trimer, and tetramer, respectively). The linear (α_0) and non-linear absorption (β) coefficients were evaluated from the gradients and y-intercepts in these figures, respectively.