## Supporting Information: Delocalized Two-Exciton States in DNA Scaffolded Cyanine Dimers

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Table S1. DNA Sequences:

Nomenclature	Sequence	T <sub>m</sub> (350mM
		Na <sup>+</sup> ) °C
1bp 2Cy3	5'-CGTCATTG/iCy3/CGGAAAGCT	63.6
	3'-GCAGTAACA/iCy3/CCTTTCGA	60.5
0bp 2Cy3	5'-CGTCATTGC/iCy3/CGACCAGCT	68.2
	3'-GCAGTAACG/iCy3/GCTGGTCGA	68.2
0bp 2Cy5	5'-A488/GGATCAGAGCTGGACGACA/iCy5/TGACGTAGGTCC	-
	3'-CCTAGTCTCGACCTGCTTG /iCy5/ACTGCATCCAGG	-

Note that while the 0bp 2Cy5 DNA duplex is also labeled with Alexfluor A488, that dye is both spectrally and spatially remote from the Cy5 dimer and therefore does influence the photophysics in the work presented here.



**Figure S1. Cyanine Dimer Absorption Spectra.** (a) Measured absorption spectra of Cy3 monomer (black), 1bp Cy3 dimer (blue), and 0bp Cy3 dimer (red) in PBS at 295K. (b) Measured absorption spectra of Cy5 monomer (black), and 0bp Cy5 dimer (red) in PBS at 295K.



**Figure S2. Separation-dependence of Cy3 Dimer Transient Absorption Spectra.** Measured transient absorption spectra at 1ps (black) and 100ps (red) for (a) Cy3 monomer, (b) 1bp Cy3 dimer, (c) 0bp Cy3 dimer in PBS at 295K.



**Figure S3. Viscosity Dependence of Cy3 Dimer Transient Absorption.** Measured transient absorption spectra of 0bp Cy3 dimer at 1ps (black), 10ps (red), 20ps (orange), 50ps (green), and 100ps (blue) in (a) PBS at 295K, (b) 2:1 gly-H<sub>2</sub>O/PBS at 295K, and (c) cooled to ~100K.



Figure S4. Viscosity Dependence of Cy5 Dimer Transient Absorption. Measured transient absorption spectra of 0bp Cy5 dimer in (a) PBS at 295K, (b) 2:1 gly-H<sub>2</sub>O/PBS at 295K, and (c) cooled to ~100K.



Figure S5. Two-exciton Eigensystem. Dependence of the two-exciton energies on (a) J (assuming  $\Delta = 420$  meV and K = 7 meV), (b) K (assuming J = 33 meV and  $\Delta = 420$  meV), (c)  $\Delta$  (assuming J = 33 meV and K = 7 meV). Dependence of the Hopfield coefficients of  $|E_{2-}\rangle$  on (d) J (assuming  $\Delta = 420$  meV and K = 7 meV), (f) K (assuming J = 33 meV and  $\Delta = 420$  meV), (g)  $\Delta$  (assuming J = 33 meV and K = 7 meV). These provide the weighting of the constituent  $|S_1\rangle$  and  $|S_2\rangle$  character of the  $|E_{2-}\rangle$  state.