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Supporting Information

Exciton Coherence Length and Dynamics in Graphene Quantum Dot Assemblies

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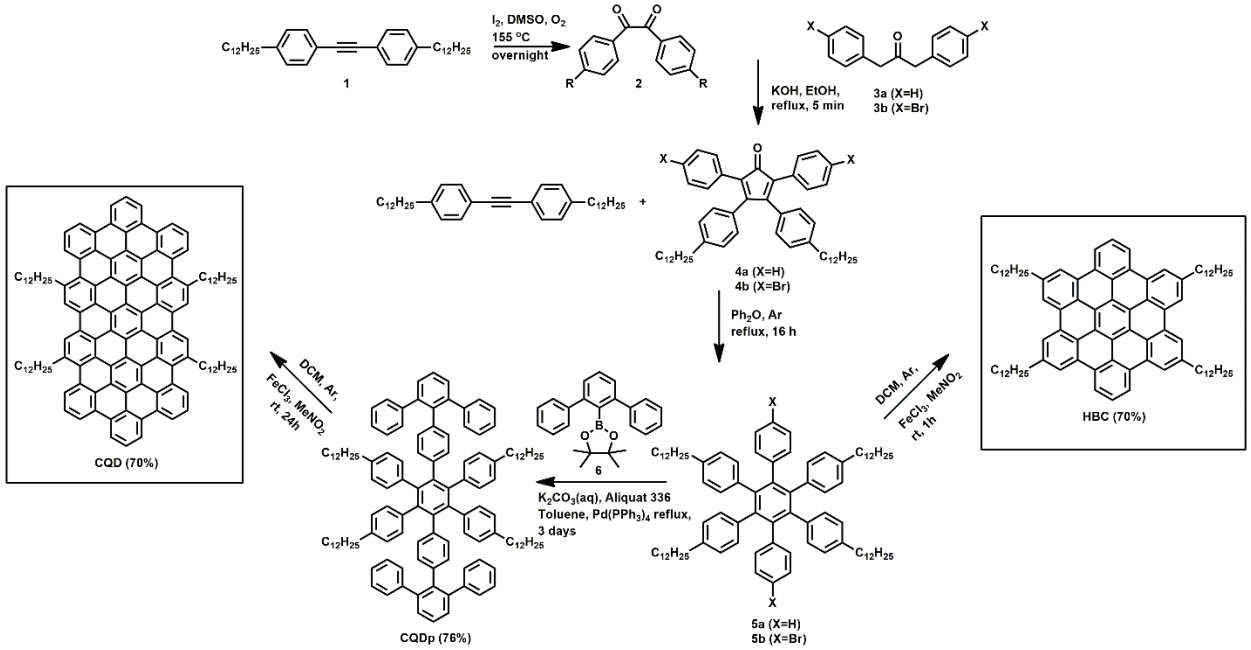
S1. General methods

All chemicals and solvents were purchased from commercial sources and used without further purification. ^{1}H and ^{13}C NMR were recorded on either Bruker Avance III HD 500 MHz or Bruker DPX 400 MHz. MALDI was recorded on Bruker Ultraflextreme MALDI-Tof-Tof. UV-vis spectra were recorded on Cary 300 Bio spectrometer. Fluorescence spectra were recorded on Horiba Fluoromax-4 spectrometer. Scanning electron microscopy (SEM) was recorded on Hitachi S-3000N. Transmission electron micrographs (TEM) were taken on 300 kV JEM-3010 instrument. Samples were drop-casted onto copper/carbon grids from their suspensions. Support grids were purchased from Electron Microscopy Sciences model HC200-Cu (Holey Carbon Film 200 mesh). SEM and TEM instruments used in this study are part of Electron Microscopy Service (Research Resources Center, UIC).

S2. Synthesis

The graphene quantum dots (GQDs) were synthesized using the reaction sequence shown in Scheme S1. Compounds 5a,[1] 5b[2] and 6[3] were synthesized according to published procedures.



Scheme S1: Synthesis of the two graphene quantum dots: HBC and CQD.

CQDp: A mixture of compound 6 (55 mg, 0.154 mmol), 5b (100 mg, 0.073 mmol), K_2CO_3 (2M/H_2O, 547 mg, 3.95 mmol) and Aliquat 336 (0.6 mg, 0.0015 mmol) in toluene (5 ml) was degassed three times via freeze-pump-thaw cycles. $Pd(PPh_3)_4$ (10 mg, 0.009 mmol) was quickly added and the reaction mixture was degassed again via three freeze-pump-thaw cycles. The reaction mixture was refluxed under argon for 3 days, then cooled to room temperature, quenched with water and extracted using dichloromethane (DCM). The organic fractions were collected, dried over $MgSO_4$ and solvent was removed under vacuum. Silica gel column chromatography of the reaction mixture with hexane/DCM (9:1) yielded 92 mg of product (76% yield). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 7.35 (m, 6H), 7.19 (m, 12H), 6.94 (m, 8H), 6.66 (d, J = 7.96 Hz, 8H), 6.57 (d, J = 7.96 Hz, 4H), 6.35 (d, J = 8.12 Hz, 4H), 6.24 (d, J = 8.12 Hz, 4H), 2.38 (t, J = 7.28 Hz, 8H), 1.41-1.11 (b, 80H), 0.89 (t, J = 6.52 Hz, 12H) ppm. ^{13}C NMR (100 MHz, $CDCl_3$): δ = 142.32, 141.76, 140.45, 139.58, 139.42, 138.67, 138.45, 138.28, 135.21, 131.83, 131.17, 130.44, 130.35, 130.03, 127.55, 127.23, 126.62, 126.19, 35.62, 32.13, 31.54, 29.91, 29.88, 29.83, 29.76, 29.58, 29.07, 22.90, 14.34 ppm. MALDI MS: 1663.29 Calculated: 1663.17 (72.5%)

CQD: CQDp (50 mg, 0.030 mmol) in dry DCM (10 ml) was purged with argon for 15 minutes. $FeCl_3$ (204 mg, 1.26 mmol) in dry nitromethane (1 ml) was added dropwise under argon. The reaction mixture was kept under argon and stirred at room temperature for 24 h. The reaction was quenched with methanol (5 ml) and solvent was removed under vacuum. The black-colored solid was suspended in methanol, filtered and dried under vacuum to yield 35 mg of the product (70% yield). MALDI MS: 1635.23 Calculated: 1634.95 (73%)

HBC: A solution of 5a (15 mg, 0.012 mmol) in dry DCM (5 ml) was purged with argon for 15 minutes. $FeCl_3$ (36 mg, 0.22 mmol) in dry nitromethane (0.5 ml) was added dropwise under argon. The reaction mixture was kept under argon and stirred at room temperature for 1 h. The reaction was quenched with methanol (2 ml) and solvent was removed under vacuum. The yellow-colored solid was suspended in methanol, filtered and dried under vacuum to yield 11 mg of the product. MALDI MS: 1195.14 Calculated: 1194.89 (100%).

S3. Steady-state spectroscopy

The aggregation of HBC and CQD was monitored using concentration dependent UV-vis spectroscopy.

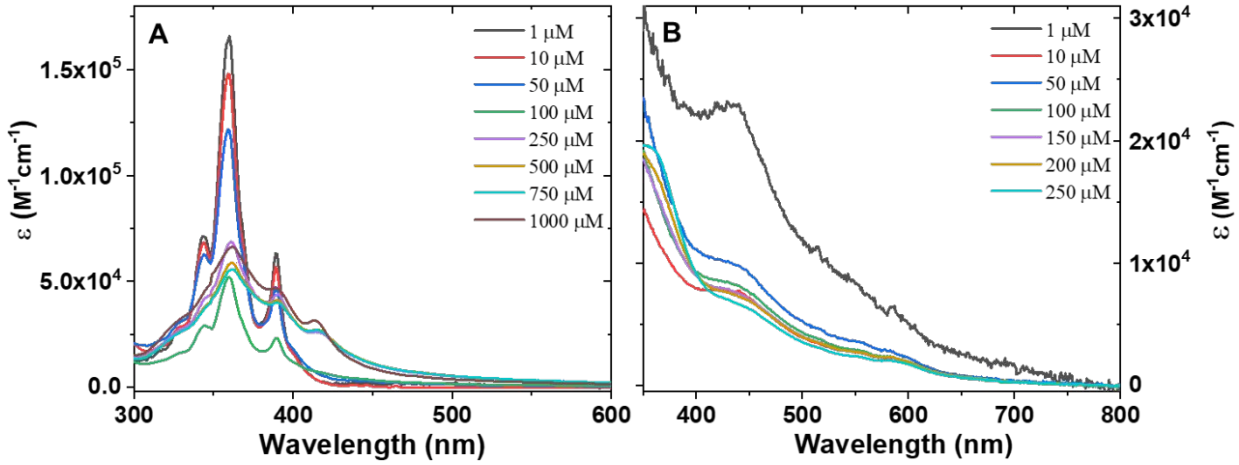


Figure S1: Concentration-dependent UV-Vis absorption spectra for A) HBC in toluene and B) CQD in 1-cyclohexyl-2-pyrrolidone.

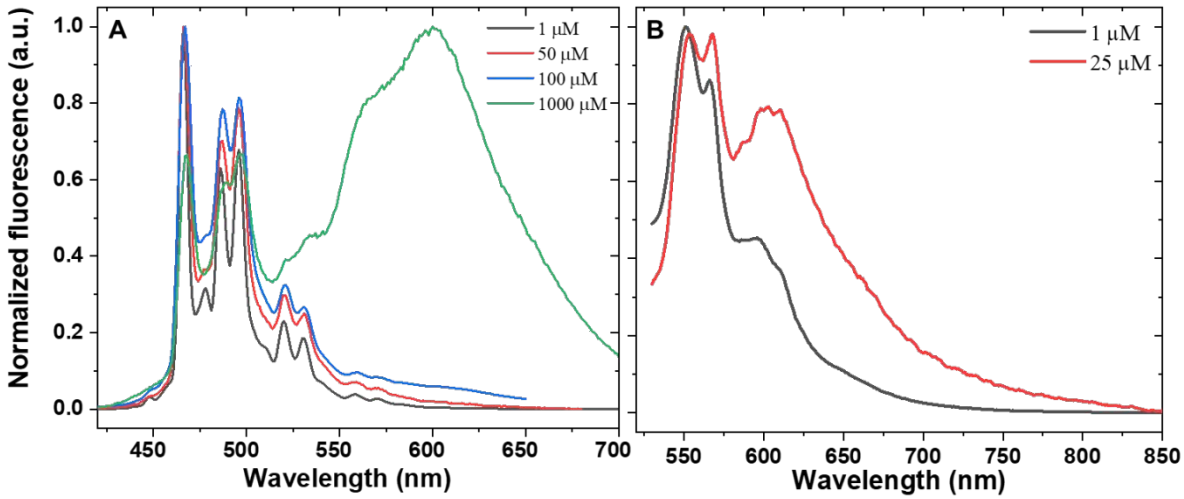


Figure S2: Concentration-dependent fluorescence spectra for A) HBC in toluene (at 360 nm excitation) and B) CQD in 1-cyclohexyl-2-pyrrolidone (at 440 nm excitation).

Table S1: Photophysical properties of HBC and CQD

Sample		**λ_{abs}/nm (ε/$M^{-1}cm^{-1}$)**	**λ_{em}/nm**	**Stokes shift (eV)**	**τ_{fl}/ns**	**Q.Y.(%)**
HBC	1 μM in toluene	360 ($1.5 \cdot 10^5$)	466	0.78	15.4	7.9
	Thin film	367	611	1.35	τ_1=1.65 (0.67) τ_2=7 (0.33)	12.1
CQD	1 μM in 1-cyclohexyl-2-pyrrolidone	440 ($2.3 \cdot 10^4$)	551	0.57	τ_1=1.87 (0.64) τ_2=11.2 (0.36)	8.6
	Thin film	573	690	0.37	τ_1=0.25 (0.44) τ_2= 3.27 (0.56)	0.5

S4. Small angle X-ray scattering (SAXS)

Small angle X-ray scattering data were collected at Advanced Photon Source (APS), Argonne National Laboratory at the beamline 12-ID-B using 14 keV source. The HBC and CQD samples were prepared in toluene and 1-cyclohexyl-2-pyrrolidone respectively with concentration between 0.1 and 1 mM. Solvent background was subtracted from the data before plotting. Powder samples were mounted on a transparent adhesive tape.

The data in low q region is analyzed using Porod analysis which relates signal intensity, I(q), with the scattering vector, q, by the following relation: $I(q) \propto q^{-\alpha}$, where α is the slope of the graph between log of intensity and log of q. The α values for HBC and CQD are close to 3.5 which signifies that the aggregates are coiled like a polymer in solution.

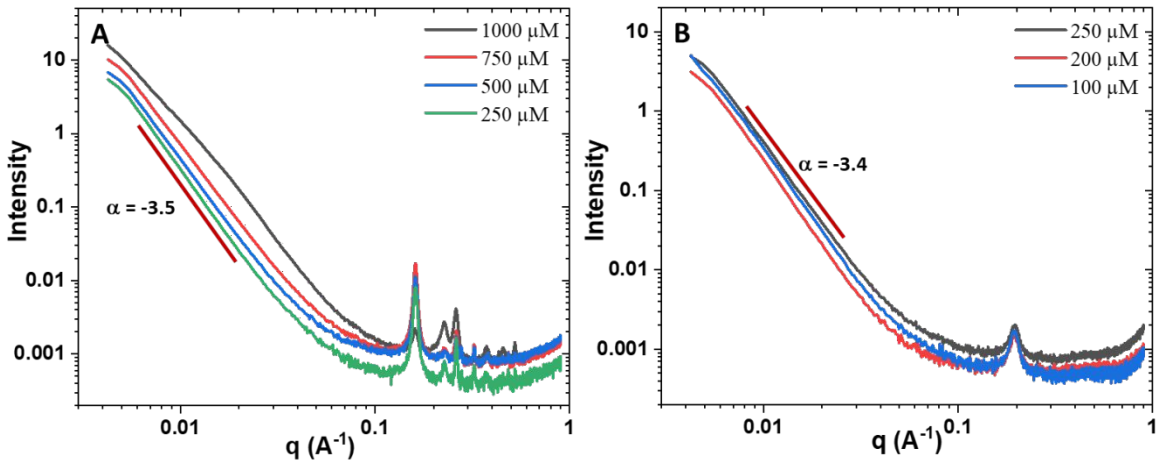


Figure S3: Porod analysis plot for A) HBC and B) CQD.

The Scherrer formula was used to calculate the size (τ) of the crystalline domains:

$$\tau = \frac{k\lambda}{\beta \cos\theta} \tag{1}$$

where *k* is the Scherrer constant (approximate value 0.9), λ is the wavelength of the incident X-ray (0.8856 Å), θ is half of the scattering angle and β is the FWHM of diffraction peak in radians. The scattering vector q and distance d (in Å) were determined using equations 2 and 3:

$$q = \frac{4\pi \sin\theta}{\lambda} \tag{2}$$

$$d = \frac{2\pi}{q} \tag{3}$$

S5. Transient absorption (TA) measurements

A. Sample Preparation: The thin film samples were prepared by mixing 1 mM solutions of HBC and CQD in chloroform with poly methyl-methacrylate (PMMA, added in 1:1 weight ratio with respect to HBC or CQD) to reduce sample scattering. The solutions were drop-casted on a quartz slide to achieve optical density of 0.3-1 at the excitation wavelength (405 nm).

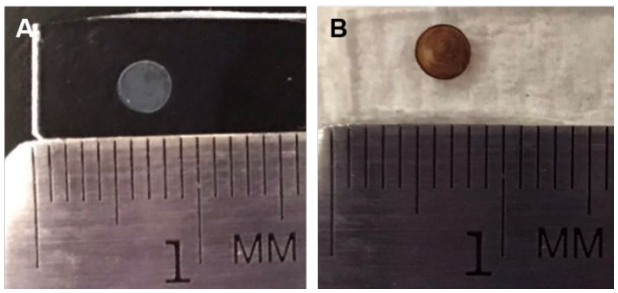


Figure S4: Thin film sample of A) HBC and B) CQD.

Grazing-incidence wide-angle X-ray scattering (GIWAXS) spectra were collected on beamline 8-ID-E at the Advanced Photon Source, Argonne National Laboratory. The samples were drop-casted on a quartz slide. GIWAXS measurements were used to extract the π-π stacking distance in the GQD assemblies.

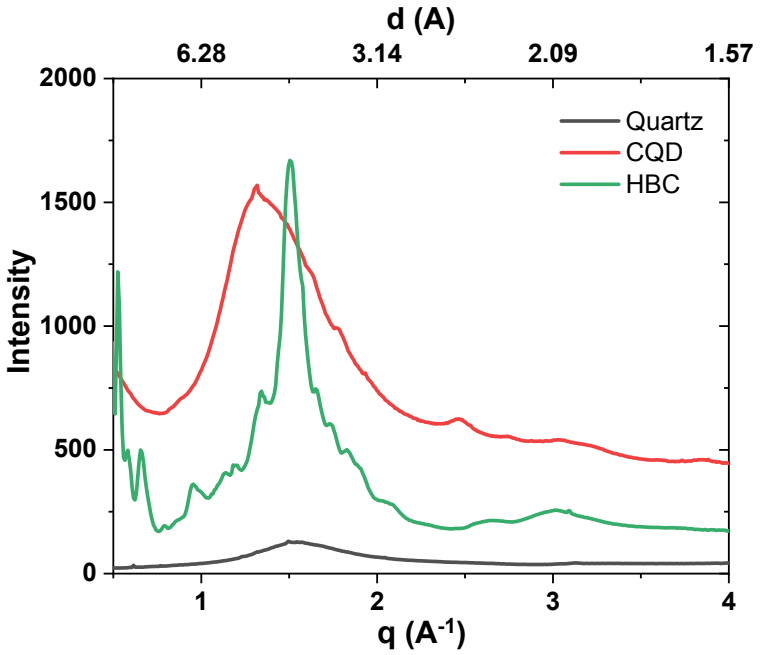


Figure S5: GIWAXS of thin film samples of HBC and CQD

B. Instrument Description: The setup used to perform transient absorption measurements was described previously.[4] In brief, 800 nm laser pulses of 100 fs duration were produced at a 1 kHz repetition rate by a mode-locked Ti:sapphire laser and regenerative amplifier (Astrella, Coherent

Inc.). The output from the Astrella was split into pump and probe beams. The pump beam was sent into an optical parametric amplifier (OPerA Solo, Coherent Inc.) to obtain the excitation pulse. The probe beam was focused into a 4 mm CaF_2 crystal that was continuously translated with a linear stage (Newport MFA-CC), to generate the white light continuum between 350 and 750 nm, which was focused into the sample. Care was taken to ensure the CaF_2 crystal axis matched the polarization of the incident light, to avoid any wavelength polarization dependence. Thin film samples were excited by 405 nm light at pump intensities between 100-500 nJ per pulse in a nitrogen-purged 1 cm quartz cuvette, and a high angle of incidence between pump and probe (15^o) was used to reduce pump scattering. Thin film measurements were performed at magic angle, parallel, and perpendicular polarization between pump and probe. After passing through the sample, the probe continuum was coupled into an optical fiber and input into a CCD spectrograph (Ocean Optics, Flame-S−UV−VIS−ES). The data acquisition was achieved using in-house LabVIEW (National Instruments) software routines. The group velocity dispersion of the probing pulse was determined using nonresonant optical Kerr effect measurements.[5]

S6. Exciton size and dynamics modeling:

A. <u>Exciton size in thin film samples:</u> The exciton size was calculated using following equation:

$$\Delta A^{ani} = -\frac{\epsilon_e \cdot \sigma_m \cdot r_a \cdot \gamma}{2.3 \cdot \epsilon_m} \cdot I_{abs} \qquad \text{(Equation 1 from manuscript)}$$

Here, absorption cross section per molecule (σ_m) was calculated using the following equation:

$$\sigma_m = \frac{2.3 \cdot A \cdot Area}{N_{Total}} \qquad (4)$$

Here, A is the ground state absorption of the sample obtained at the wavelength at which the transient signal was probed (367 nm for HBC and 467 nm for CQD). The area of the thin film was calculated using the ImageJ software[6]. The total number of molecules (N_{Total}) in the thin film was obtained from the molarity and volume of the solution used to drop cast the sample. The number of HBC and CQD molecules per unit length of the chromophore assembly (ϵ_m) was experimentally obtained using the π-stacking distances observed in grazing-incidence wide-angle X-ray

scattering (GIWAXS) measurements (Figure S5, SI). The wavefunction envelope factor γ was obtained assuming that the excitonic wavefunction adopts a rectangular shape.

The anisotropy correction factor (r_a) was calculated from polarization-dependent TA measurements at time t=0 using the following equation:

$$r_a = 1 + \frac{r}{0.4} \cdot 0.8 \text{ ; where } r = \frac{\Delta A_z - \Delta A_x}{\Delta A_z + 2\Delta A_x} \quad (5)$$

Here, ΔA_z is the transient signal at the probing bleach wavelength (367 nm for HBC and 467 nm for CQD) obtained with parallel orientation of the pump and probe beams. ΔA_x is the same signal obtained for perpendicular orientation of the pump and probe beams.

The slope of equation 1 in the manuscript was obtained from a linear fit of the graph between ΔA^{ani} and I_{abs}. Here, ΔA^{ani} is the transient signal at the probing bleach wavelength (367 nm for HBC and 467 nm for CQD) with parallel orientation of pump and probe beams. I_{abs} is the number of absorbed photons per unit area which is calculated from the energy of the pump beam before (I_o) and after (I) hitting the thin film sample using the following equation:

$$I_{abs} = \frac{I_o - I}{\pi \cdot r_{pump}^2} \quad (6)$$

Here, r_{pump} is radius of the pump beam (262 ± 18 μm) which is obtained using the knife edge method.[7] The thin film samples were excited at a wavelength of 405 nm.

Finally, equating the slope of the graph between ΔA^{ani} and I_{abs} with the calculated parameters yields the exciton size (ϵ_e) of 0.8 ± 0.2 nm for HBC and 0.4 ± 0.1 nm for CQD. The parameters and results are also summarized in Table S2. The error analysis is discussed in section S7.

Table S2: Exciton size calculation summary for HBC and CQD thin films

	Parameter	**HBC**	**CQD**
1	N_{Total} (molecules in thin film)	$(1.9 \pm 0.1) \cdot 10^{15}$	$(3.4 \pm 0.3) \cdot 10^{15}$
2	Area of thin film (cm^2)	0.08 ± 0.002	0.09 ± 0.003

3	A (absorption at the probe wavelength, 367 nm for HBC and 467 nm for CQD)	0.32 ± 0.01	0.97 ± 0.04
4	σ_m (absorption cross-section, cm^2)	$(3.1 \pm 0.7)\cdot 10^{-17}$	$(5 \pm 1)\cdot 10^{-17}$
5	Slope of ΔA^{ani} vs I_{abs} fit (cm^2)	$-(4 \pm 0.5)\cdot 10^{-17}$	$-(3.1 \pm 0.2)\cdot 10^{-17}$
6	r_a (anisotropy enhancement factor)	1.6 ± 0.1	1.4 ± 0.01
7	ϵ_m (stacking distance, nm)	0.4 ± 0.02	0.5 ± 0.02
8	**ϵ_e (exciton size, nm)**	**0.8 ± 0.2**	**0.4 ± 0.1**
9	**ϵ_e (exciton size, molecules)**	**2 ± 0.5**	**0.8 ± 0.2**

Exciton size for HBC in solution:

To evaluate the accuracy of the experiment in determining the exciton size, experiments were performed for HBC in solution, where the exciton size is expected to be one molecule. Transient absorption spectra (Figure S6) show that the bleach develops during the first several hundred femtoseconds, likely due to the internal conversion from higher excited states to the S_1 state. Thus, the bleach signal for exciton size calculations was obtained at the time delay of 0.5 ps. The exciton size was obtained using an expression analogous to equation 1, but adopted for molecules in solution, as follows:

$$\Delta A^{ani} = -\frac{A \cdot \epsilon_e \cdot r_a \cdot \gamma}{l \cdot c_{HBC} \cdot N_a} I_{abs} \quad (7)$$

Where A is the ground-state absorption of the sample at the probe wavelength (A=0.33 at 365 nm), r_a is the anisotropy factor (which was found to be 1), γ is the wavefunction overlap factor (which was set to a value of 1), l is the cuvette pathlength (2 mm), c_{HBC}=20 uM is the concentration of HBC in solution and Na is the Avogadro's number. Using the slope of $-1.3\cdot 10^{-16}$ cm^2 obtained in Figure S6C, we derived an exciton size of **0.96 ± 0.06**, which is within the error of the exact value of 1 molecule. The error analysis is discussed in section S7.

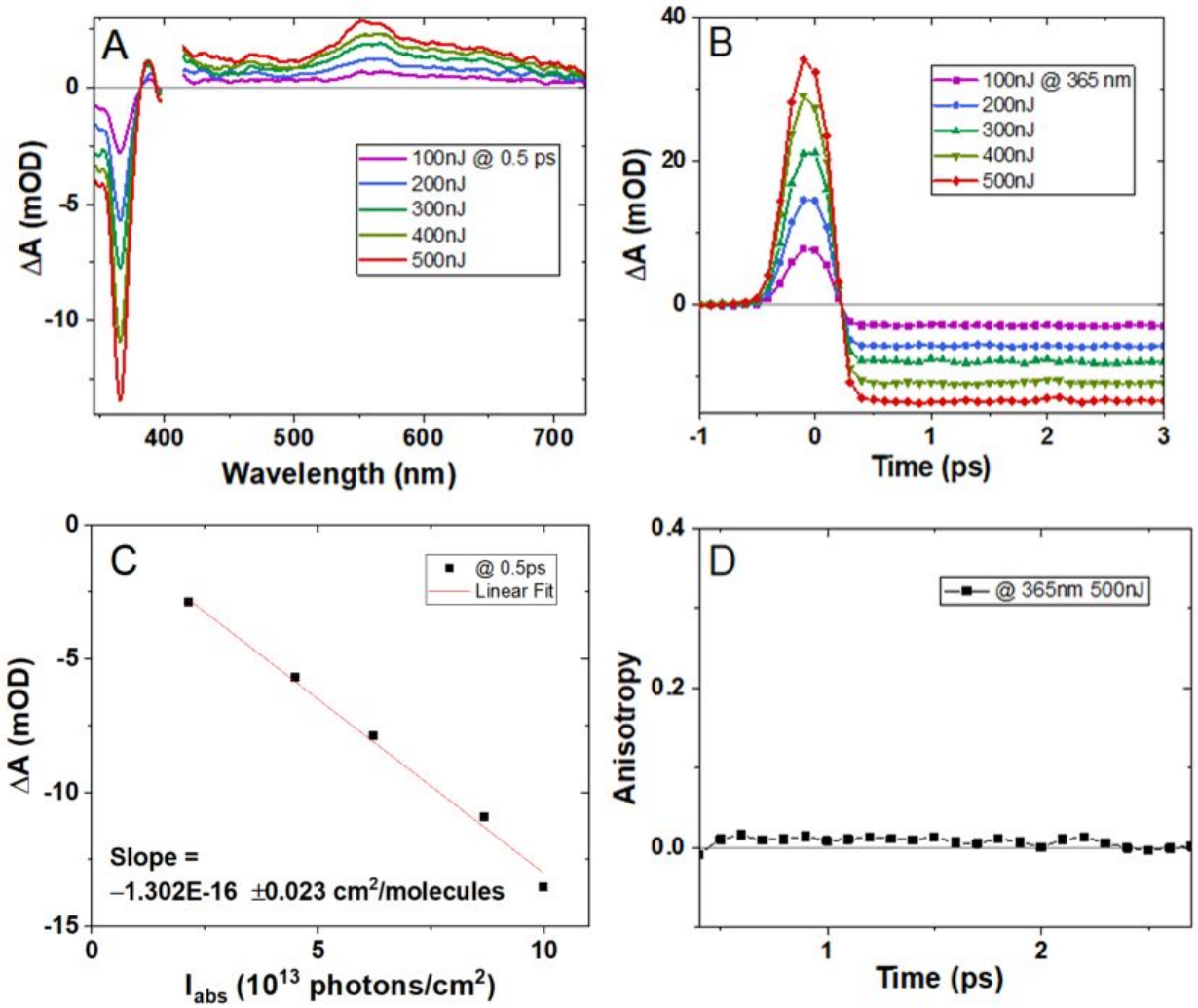


Figure S6. Experimental determination of exciton size for HBC in solution: A and B) Transient absorption spectra (at time t=0.5 ps) and kinetics (at probe wavelength λ=365 nm) obtained for 20 μM solution of HBC in toluene. Sample was excited with 400 nm pump beam with fluences varied in the 100 – 500 nJ range. C) The dependence of bleach signal as a function of pump intensity. The slope of the fit was used to determine the exciton size. D) Anisotropy of the sample was used to determine the r_a factor.

B. <u>Exciton dynamics:</u> The exciton dynamics for quantum dot assemblies was evaluated using exciton-exciton annihilation approach, by following the methodology developed for perylene bisimide assemblies.[8] Experimentally, a series of magic angle transient absorption spectra were collected at varying pump energies (100-500 nJ) and the kinetic profiles of the bleach signal (at

367 nm for HBC and 467 nm for CQD) were used to extract the exciton diffusion length. Data were fit to a set of first order differential equations as follows:

$$N(t) = N_1(t) + N_2(t) + N_{offset} \tag{8}$$

$$\frac{dN_1(t)}{dt} = -\frac{N_1(t)}{\tau_1} - \frac{N_1(t)}{\tau_{tr}} - \sqrt{\frac{2D}{\pi t}} \cdot N_1(t)^2 \tag{9}$$

$$\frac{dN_2(t)}{dt} = \frac{N_1(t)}{\tau_{tr}} - \frac{N_2(t)}{\tau_2} \tag{10}$$

Here, N(t) is the total density of excitons, N_1 and τ_1 are the density and lifetime of mobile excitons, while N_2 and τ_2 are the density and lifetime of trapped excitons. The N_{offset} parameter was used to account for the long-lived signal observed in the case of CQD. The τ_{tr} is the lifetime associated with the trapping of initially formed excitons. D is the diffusion coefficient of mobile excitons. The initial conditions for the above equation, expressed as the number of excitons per volume formed at time zero, n(t=0), were obtained for each pump intensity measurement as follows:

$$n(t=0) = \frac{n_{vol}}{C} \tag{11}$$

Here, C is the chromophore concentration (cm^{-3}), while n_{vol} is the exciton density in the volume sampled by the probe beam. The value for n_{vol} was obtained using the following expression:

$$n_{vol} = \frac{N_{pump}}{\pi r_{probe}^2 l}\left[1 - 10^{-OD(\lambda_{pump})}\right] \tag{12}$$

Here, r_{probe} is radius of the probe beam (112 μm) which is obtained using the knife edge method.[7] The λ_{pump} is the wavelength of the pump beam (405 nm) and OD is the optical density of the sample at λ_{pump}. N_{pump} is the number of pump photons that was calculated using the following equation:

$$N_{pump} = \frac{E_{pump}\lambda_{pump}}{hc} \frac{\int_0^{r_{probe}} r e^{\frac{-2r^2}{r_{pump}^2}} dr}{\int_0^{\infty} r e^{\frac{-2r^2}{r_{pump}^2}} dr} \tag{13}$$

Here, E_{pump} is energy of the pump pulse obtained using a power meter, r_{pump} is radius of the pump beam (262 μm) obtained by using the knife edge method, h is the Planck constant and c is the speed of light in vacuum.

Once the initial condition values were obtained, the variable pump power data were fit to the equation 8 using MATLAB, by varying several parameters: D, τ_1, τ_2, τ_{tr} and N_{offset}. The obtained D and τ_1 values were used to obtain the exciton diffusion length (L_D) as follows:

$$L_D = \sqrt{2D\tau_1} \tag{14}$$

S7. Error analysis for exciton size

To estimate the error in exciton size measurements, we use Taylor-series based error propagation method. If

$$\epsilon = f(x_1,\ldots,x_n) \tag{15}$$

The uncertainty in ϵ is then

$$\delta\epsilon = \sqrt{\left(\frac{\partial\epsilon}{\partial x_1}\delta x_1\right)^2 + \ldots + \left(\frac{\partial\epsilon}{\partial x_n}\delta x_n\right)^2} \tag{16}$$

S8. DFT calculations

A. **Structure optimization:** All calculations were performed using Gaussian 09[9] package with resources of Ohio Supercomputer Center and Extreme cluster at University of Illinois at Chicago. Structure optimization and frequency calculations were performed using B3LYP/6-31 g(d,p) level of theory [10-11] with integral equation formalism variant of polarizable continuum model (iefpcm)[12] for acetonitrile solvation. Frequency calculations were performed to ensure no imaginary frequency. Time-dependent DFT calculations were performed at the same level of theory. To decrease the computational time, dodecyl chains in HBC and CQD were replaced with methyl groups. Tables S3 to S6 lists the coordinates of optimized HBC and CQD monomer and dimer.

Table S3: Coordinates for optimized HBC monomer

C	-1.25404634	0.73156693	-0.00326467
C	-1.30878776	-0.62858044	0.00004766
C	-0.01075216	-1.44961747	0.00429542
C	1.19458758	-0.81688602	0.00440802
C	1.25663102	0.71774471	0.00014135
C	0.10608170	1.44523376	-0.00315334
C	0.11589758	2.98687119	-0.00684574
C	2.62053977	1.43376715	-0.00040123
C	2.52479764	-1.59616672	0.00868658
C	-0.07256854	-2.98878185	0.00845554
C	-2.64877710	-1.39092129	-0.00037335
C	-2.55605981	1.55466583	-0.00714973
C	1.27538922	3.69719800	-0.00747012
C	1.23588211	5.23617588	-0.01070766
C	0.04494129	5.88204240	-0.01280205
C	-1.26776731	5.07654574	-0.01251131
C	-1.23353706	3.72130638	-0.00989067
C	3.80114938	0.74937579	0.00300722
C	5.13053868	1.52997192	0.00158820
C	5.12532638	2.88587499	-0.00354012
C	3.78921503	3.64977733	-0.00660788
C	2.62276873	2.96292226	-0.00492030
C	2.56015360	-2.95551060	0.01282624
C	3.91264556	-3.69093549	0.01741295
C	5.06752560	-2.98260401	0.01724737
C	5.02648652	-1.44297535	0.01237581
C	3.83568397	-0.79479980	0.00823377

C	-1.25546354	-3.66901065	0.00820897
C	-1.24409203	-5.21052752	0.01276276
C	-0.06727663	-5.88397731	0.01678369
C	1.26233993	-5.10877345	0.01678383
C	1.25058894	-3.75520231	0.01289029
C	-3.84377360	-0.74194221	-0.00404829
C	-5.15678467	-1.54578679	-0.00449691
C	-5.12052373	-2.90010521	-0.00151908
C	-3.76658114	-3.63412717	0.00272545
C	-2.61003737	-2.92685414	0.00346769
C	-2.55354208	2.91932373	-0.01005774
C	-3.89410929	3.68042745	-0.01361121
C	-5.06577111	2.99810214	-0.01457555
C	-5.05955384	1.45898078	-0.01148068
C	-3.88153201	0.79206984	-0.00773699
C	6.45582955	3.66141742	-0.00636281
C	6.42119375	-3.71702070	0.02190559
C	-6.43306999	-3.70550058	-0.00241914
C	-6.40234697	3.76292167	-0.01876250
H	2.14822609	5.79523466	-0.01129132
H	0.01640471	6.95164921	-0.01464486
H	-2.20810560	5.58713233	-0.01437289
H	6.06016998	1.00013561	0.00452766
H	3.78245006	4.71974667	-0.01012031
H	3.94052793	-4.76064300	0.02072792
H	5.93895933	-0.88414504	0.01225581
H	-2.16774062	-5.75066530	0.01273895
H	-0.06298019	-6.95396687	0.01997286
H	2.19253857	-5.63769129	0.01980165
H	-6.09724164	-1.03533227	-0.00719405

H	-3.73865629	-4.70373747	0.00508741
H	-3.90000650	4.75047726	-0.01542531
H	-5.98286880	0.91825595	-0.01227843
H	6.32386467	4.59780761	0.49434838
H	7.20389336	3.08831916	0.50034692
H	6.76406740	3.83707304	-1.01583001
H	7.15046996	-3.11765905	0.52572886
H	6.31709706	-4.65347395	0.52903268
H	6.73644335	-3.89032406	-0.98577901
H	-7.19507380	-3.14623545	0.49909367
H	-6.28122196	-4.63553597	0.50438852
H	-6.73493903	-3.89432856	-1.01141444
H	-6.27682338	4.69755864	-0.52436705
H	-6.71417381	3.94162289	0.98905469
H	-7.14472028	3.18104903	-0.52393365

Table S4: Coordinates for optimized HBC dimer

C	-1.31356200	0.52135000	-1.71742700
C	-1.08936500	-0.87795500	-1.71955700
C	0.23479000	-1.38350900	-1.72069800
C	1.33487100	-0.48992600	-1.71687800
C	1.11067300	0.90936100	-1.71205800
C	-0.21349200	1.41493100	-1.71397900
C	-0.43765100	2.81433900	-1.70753500
C	2.21079100	1.80290400	-1.69782300
C	2.65902100	-0.99556700	-1.70969200
C	0.45890200	-2.78294100	-1.72066200
C	-2.18948100	-1.77160300	-1.71337400
C	-2.63772200	1.02696400	-1.71046100

C	0.66388800	3.71054100	-1.69674700
C	0.41574200	5.10430000	-1.69342900
C	-0.88368800	5.60446800	-1.69044800
C	-1.96242200	4.72419800	-1.69394000
C	-1.76385000	3.32233100	-1.70524800
C	3.53774800	1.29805700	-1.68837300
C	4.62038400	2.21018600	-1.66546000
C	4.40647700	3.58910200	-1.63505800
C	3.10038300	4.07844500	-1.64820400
C	1.98751600	3.20469800	-1.68523900
C	2.88480800	-2.39719700	-1.70977600
C	4.21508100	-2.88040700	-1.69156200
C	5.30308100	-2.00704000	-1.66600600
C	5.07485400	-0.63081100	-1.66525500
C	3.76186800	-0.10168400	-1.69279700
C	-0.64260900	-3.67922400	-1.71779500
C	-0.39439000	-5.07297500	-1.72054700
C	0.90507400	-5.57310400	-1.71667600
C	1.98377000	-4.69277700	-1.71303900
C	1.78506900	-3.29091300	-1.71728500
C	-3.51643700	-1.26681500	-1.70480200
C	-4.59918700	-2.17896400	-1.68999500
C	-4.38538000	-3.55808600	-1.66793800
C	-3.07925500	-4.04736200	-1.68004000
C	-1.96626200	-3.17344700	-1.70800600
C	-2.86367200	2.42859900	-1.70454000
C	-4.19406800	2.91177900	-1.68694300
C	-5.28191200	2.03780200	-1.66266100
C	-5.05342800	0.66185900	-1.67351700
C	-3.74047500	0.13291100	-1.70134500

C	5.57593900	4.55393200	-1.58162100
C	6.72111900	-2.54362900	-1.61491100
C	-5.55477100	-4.52347700	-1.62377800
C	-6.70234600	2.56844500	-1.61603400
H	1.18282100	5.85732300	-1.69001000
H	-1.05412400	6.67110500	-1.68275900
H	-2.92555100	5.20139900	-1.68747100
H	5.65194300	1.90925700	-1.65810200
H	3.01811300	5.15004500	-1.62168400
H	4.46856800	-3.92480700	-1.68175800
H	5.96313000	-0.02633900	-1.63486400
H	-1.16145100	-5.82602200	-1.72274300
H	1.07554800	-6.63975700	-1.71399700
H	2.94703200	-5.16974400	-1.70709600
H	-5.63068800	-1.87780400	-1.68312300
H	-2.99712000	-5.11911600	-1.65991900
H	-4.44650000	3.95635500	-1.67765700
H	-5.94244800	0.05795300	-1.65248300
H	5.40423700	5.29348500	-0.79886800
H	6.50257000	4.02217000	-1.36392200
H	5.67181600	5.06139900	-2.54170200
H	7.07682500	-2.52777500	-0.58449900
H	6.75356700	-3.56700500	-1.98983200
H	7.37437900	-1.92430000	-2.23028400
H	-6.48430200	-3.99165400	-1.41900500
H	-5.39125400	-5.25954700	-0.83600900
H	-5.63930800	-5.03514800	-2.58269700
H	-7.19337900	2.37534500	-2.57001900
H	-6.70544500	3.64155400	-1.42312600
H	-7.25330300	2.06731200	-0.81951100

C	-1.11093300	0.87779300	1.71672700
C	-1.33482200	-0.52157300	1.71380500
C	-0.23457700	-1.41494900	1.71398900
C	1.08944600	-0.90908200	1.71563400
C	1.31332700	0.49029400	1.72004400
C	0.21309300	1.38365500	1.72155700
C	0.43695100	2.78313900	1.72306900
C	2.63734500	0.99627600	1.71473500
C	2.18975200	-1.80245200	1.70560700
C	-0.45837700	-2.81442200	1.70736400
C	-2.65883700	-1.02746400	1.70254200
C	-2.21120900	1.77120500	1.70744800
C	1.76303000	3.29143500	1.72328900
C	1.96131700	4.69338600	1.72100600
C	0.88239600	5.57342700	1.72331300
C	-0.41692900	5.07296600	1.72315900
C	-0.66477200	3.67916100	1.71797700
C	3.74025300	0.10253800	1.70018900
C	5.05310100	0.63178400	1.67410000
C	5.28128500	2.00783400	1.66942300
C	4.19330100	2.88146800	1.69937300
C	2.86302000	2.39795900	1.71619600
C	1.96698700	-3.20437200	1.69378400
C	3.08025700	-4.07797900	1.66236300
C	4.38639400	-3.58798700	1.65702200
C	4.59933700	-2.20898700	1.67873600
C	3.51649100	-1.29718800	1.69796200
C	-1.78447300	-3.32265900	1.70095000
C	-1.98278100	-4.72455600	1.68967900
C	-0.90390800	-5.60463800	1.69037200

C	0.39541700	-5.10425500	1.69759100
C	0.64338200	-3.71044300	1.70075200
C	-3.76174300	-0.13364600	1.68957700
C	-5.07447000	-0.66286000	1.65657300
C	-5.30258900	-2.03887400	1.64472800
C	-4.21468100	-2.91259300	1.67368800
C	-2.88445800	-2.42914600	1.69607800
C	-1.98827900	3.17310200	1.70347200
C	-3.10136100	4.04679700	1.67198200
C	-4.40733700	3.55720700	1.65530800
C	-4.62089400	2.17805500	1.67643100
C	-3.53801300	1.26612700	1.69432300
C	6.70145600	2.53925200	1.62385700
C	5.56010900	-4.54768700	1.60445900
C	-6.72255200	-2.57026400	1.59281400
C	-5.57708400	4.52200900	1.60796300
H	2.92436200	5.17079300	1.71762100
H	1.05260000	6.64012700	1.72236400
H	-1.18417600	5.82582500	1.72449400
H	5.94207500	0.02791100	1.64982500
H	4.44555700	3.92612800	1.69501400
H	2.99731300	-5.14938600	1.63327600
H	5.63115100	-1.90871700	1.66736400
H	-2.94580600	-5.20191800	1.68028400
H	-1.07414100	-6.67130800	1.68280900
H	1.16250800	-5.85726400	1.69752800
H	-5.96346800	-0.05906200	1.63201700
H	-4.46698500	-3.95720400	1.66401500
H	-3.01932700	5.11856300	1.65233800
H	-5.65234500	1.87678100	1.66668400

H	7.18996900	2.35163800	2.58022200
H	7.25510200	2.03447700	0.83150500
H	6.70409800	3.61130600	1.42514900
H	6.45867800	-4.06943700	1.99533900
H	5.34430200	-5.43157500	2.20543400
H	5.73470700	-4.84922300	0.57146500
H	-7.27703500	-2.05526400	0.80765500
H	-7.21053000	-2.39582000	2.55193800
H	-6.72467500	-3.63953000	1.37959700
H	-5.40323500	5.26976800	0.83352000
H	-5.67644700	5.01938700	2.57295400
H	-6.50265500	3.99203100	1.38159900

Table S5: Coordinates for optimized CQD monomer

C	-0.71231700	1.22393500	0.16316900
C	0.71233200	1.22393800	0.16316000
C	1.42477900	0.00001600	0.11058400
C	0.71233500	-1.22389400	0.16346500
C	-0.71231400	-1.22389700	0.16345300
C	-1.42476000	0.00001200	0.11057800
C	-1.43346900	2.46137200	0.24096900
C	-2.86411100	0.00000200	0.04920600
C	2.86412800	0.00001100	0.04921500
C	-1.43346100	-2.46132100	0.24150900
C	1.43348500	-2.46131300	0.24157700
C	1.43347900	2.46138100	0.24092600
C	-2.85789000	2.48301700	0.13761600
C	-3.56454600	3.71962000	0.18381100
C	-2.85317500	4.89357400	0.57838100

C	-1.47239800	4.83504500	0.67773400
C	-0.72650900	3.66960300	0.44732900
C	-3.57391700	-1.23387400	0.01836600
C	-4.99282700	-1.23096700	-0.07566300
C	-5.70080200	-0.00002500	-0.14379800
C	-4.99283600	1.23093500	-0.07587900
C	-3.57392500	1.23386700	0.01813400
C	-0.72650200	-3.66950200	0.44816400
C	-1.47239000	-4.83491200	0.67872600
C	-2.85316200	-4.89347600	0.57930300
C	-3.56452600	-3.71959800	0.18449800
C	-2.85787700	-2.48299700	0.13810100
C	2.85790700	-2.48298800	0.13823200
C	3.56456300	-3.71958500	0.18474400
C	2.85318300	-4.89341500	0.57970100
C	1.47240600	-4.83485600	0.67901200
C	0.72652100	-3.66949000	0.44824100
C	3.57393400	1.23388200	0.01807700
C	4.99284200	1.23095100	-0.07597200
C	5.70081200	-0.00001100	-0.14383600
C	4.99284900	-1.23095600	-0.07560100
C	3.57394000	-1.23386500	0.01843600
C	0.72651300	3.66960900	0.44728800
C	1.47239800	4.83505700	0.67764400
C	2.85316900	4.89360200	0.57824600
C	3.56454700	3.71965400	0.18363400
C	2.85789700	2.48303700	0.13752000
C	-7.14040800	-0.00004000	-0.25979900
C	7.14041100	-0.00002400	-0.25991500
C	-3.48673700	-6.20928600	0.99344500

C	3.48670300	-6.20913500	0.99421100
C	3.48668000	6.20946700	0.99230900
C	-3.48674800	6.20944300	0.99234100
C	-7.85545100	-1.22984200	-0.31905400
C	-9.25851700	-1.20424300	-0.32460700
C	-9.95149700	-0.00005800	-0.30927700
C	-9.25852900	1.20413200	-0.32480200
C	-7.85546200	1.22974500	-0.31925200
C	-7.11406400	2.48752600	-0.38029100
C	-7.11404200	-2.48762500	-0.37988600
C	-5.70282100	-2.48119100	-0.18129500
C	-4.98700100	-3.71624900	-0.14880900
C	-5.67402300	-4.89045300	-0.50660700
C	-7.03787500	-4.88002600	-0.77021500
C	-7.75973000	-3.69863500	-0.66943500
C	-7.75976700	3.69848400	-0.67002200
C	-7.03792500	4.87986500	-0.77100400
C	-5.67406800	4.89034600	-0.50742500
C	-4.98703000	3.71620400	-0.14945100
C	-5.70284100	2.48113500	-0.18171800
C	7.85545100	1.22976000	-0.31952300
C	9.25851700	1.20416000	-0.32519400
C	9.95149800	-0.00002200	-0.30963500
C	9.25852600	-1.20421300	-0.32481300
C	7.85546000	-1.22982300	-0.31913600
C	7.11404000	2.48752600	-0.38063100
C	7.11405900	-2.48761400	-0.37984800
C	5.70283100	2.48114900	-0.18195000
C	4.98701300	3.71622000	-0.14975600
C	5.67401500	4.89030400	-0.50800000

C	7.03785000	4.87979900	-0.77168700
C	7.75971400	3.69844600	-0.67057300
C	7.75974500	-3.69862400	-0.66938900
C	7.03789100	-4.88001600	-0.77012300
C	5.67405300	-4.89044600	-0.50645300
C	4.98703700	-3.71624800	-0.14860900
C	5.70284700	-2.48118300	-0.18118500
H	-0.96119300	5.74798300	0.95962400
H	-0.96119000	-5.74780100	0.96078100
H	0.96120200	-5.74770000	0.96120900
H	0.96119500	5.74799000	0.95955400
H	-3.62476500	-6.90231800	0.15438800
H	-2.82905400	-6.71177500	1.70946500
H	-4.46008300	-6.07101900	1.47117100
H	3.62484500	-6.90234700	0.15532200
H	4.45997100	-6.07077600	1.47206300
H	2.82890600	-6.71146900	1.71023700
H	3.62481100	6.90239800	0.15318700
H	2.82888200	6.71203700	1.70817000
H	4.45995200	6.07128000	1.47020400
H	-3.62481500	6.90234300	0.15318200
H	-4.46007400	6.07124100	1.47012600
H	-2.82904200	6.71205100	1.70825700
H	-9.82316100	-2.12870200	-0.31612900
H	-5.13720400	-5.81799700	-0.62741300
H	-7.53844600	-5.79974300	-1.05867300
H	-8.82380200	-3.71853100	-0.87100400
H	-8.82384300	3.71834000	-0.87157500
H	-7.53850900	5.79953200	-1.05959900
H	-5.13725700	5.81787400	-0.62839200

H	9.82316300	2.12862000	-0.31700300
H	9.82317900	-2.12866700	-0.31632700
H	5.13719300	5.81780000	-0.62911700
H	7.53839800	5.79942400	-1.06048200
H	8.82377200	3.71828000	-0.87222500
H	8.82380500	-3.71851600	-0.87102200
H	7.53845200	-5.79973200	-1.05860600
H	5.13723900	-5.81798600	-0.62726800
H	-9.82318200	2.12858700	-0.31647600
H	-11.03743900	-0.00006100	-0.29038600
H	11.03744200	-0.00001500	-0.29084700

Table S6: Coordinates for optimized CQD dimer

C	-0.72229000	1.21337200	1.81089800
C	0.68677400	1.23773400	1.76401400
C	1.40845000	0.02417100	1.77384600
C	0.72393400	-1.21137800	1.81138200
C	-0.68513000	-1.23576200	1.76463400
C	-1.40680700	-0.02219500	1.77402500
C	-1.44325300	2.42055900	1.93755000
C	-2.81743900	-0.04553200	1.78664900
C	2.81908400	0.04751100	1.78623400
C	-1.37135200	-2.46967700	1.76079000
C	1.44489900	-2.41851300	1.93852900
C	1.37301100	2.47164000	1.75960400
C	-2.85357800	2.37904300	2.04535800
C	-3.57479300	3.55738300	2.34173700
C	-2.82554100	4.60813100	2.91156000
C	-1.52710900	4.79189600	2.40338300

C	-0.76523900	3.66055100	2.04058900
C	-3.50807100	-1.27414300	1.73062000
C	-4.91520000	-1.28865500	1.62887400
C	-5.63741900	-0.07787700	1.72513200
C	-4.94823600	1.15338300	1.78636800
C	-3.54083200	1.16177500	1.86954200
C	-0.65098500	-3.68851400	1.82424600
C	-1.40213300	-4.87454100	1.72056300
C	-2.72020800	-4.84675200	2.20315100
C	-3.48352800	-3.71911300	1.82954400
C	-2.78891800	-2.48616900	1.77727200
C	2.85524500	-2.37696100	2.04594600
C	3.57650900	-3.55517200	2.34268900
C	2.82746300	-4.60560300	2.91336600
C	1.52872500	-4.78953500	2.40601500
C	0.76688200	-3.65843300	2.04239500
C	3.50970800	1.27610400	1.72965500
C	4.91681600	1.29056100	1.62767900
C	5.63904700	0.07980200	1.72423500
C	4.94987100	-1.15143100	1.78604800
C	3.54247900	-1.15977200	1.86948100
C	0.65263000	3.69049600	1.82249900
C	1.40377600	4.87647900	1.71839400
C	2.72180200	4.84887700	2.20113300
C	3.48520000	3.72112600	1.82801800
C	2.79058500	2.48815900	1.77607800
C	-7.04873200	-0.09638300	1.75381900
C	7.05037200	0.09828700	1.75259800
C	-3.15488000	-5.85634000	3.19320700
C	3.27057600	-5.37762900	4.09538300

C	3.15628700	5.85881100	3.19092500
C	-3.26803700	5.38053400	4.09355500
C	-7.73570400	-1.33329500	1.68674600
C	-9.10647400	-1.34844700	2.00995900
C	-9.83315500	-0.15055300	2.00146200
C	-9.14245100	1.05969800	2.14856000
C	-7.76866600	1.12095400	1.84301800
C	-7.06621600	2.35486200	1.65194700
C	-7.00510700	-2.51041500	1.32260300
C	-5.59722800	-2.51513700	1.47052800
C	-4.87926600	-3.73827300	1.48470500
C	-5.56849800	-4.91869500	1.13962600
C	-6.94176600	-4.89481000	0.86057500
C	-7.61896400	-3.67013400	0.81442600
C	-7.70138600	3.56368400	1.31305000
C	-7.06963600	4.78137700	1.59598400
C	-5.69794400	4.79952400	1.88573200
C	-4.97563600	3.59689700	2.02482600
C	-5.66059800	2.37259300	1.81275300
C	7.73734500	1.33516000	1.68482400
C	9.10822100	1.35047500	2.00756400
C	9.83487600	0.15256200	1.99950000
C	9.14420100	-1.05762600	2.14726200
C	7.77032400	-1.11901800	1.84214400
C	7.00664500	2.51211300	1.32045100
C	7.06781900	-2.35301700	1.65176400
C	5.59881900	2.51699400	1.46886200
C	4.88100000	3.74023100	1.48330000
C	5.57049100	4.92066000	1.13865200
C	6.94391800	4.89670300	0.86034700

C	7.62031400	3.67169100	0.81178400
C	7.70293500	-3.56199600	1.31329200
C	7.07103000	-4.77960000	1.59627300
C	5.69939500	-4.79756800	1.88627600
C	4.97724000	-3.59485400	2.02534200
C	5.66221700	-2.37064800	1.81275900
H	-1.16797200	5.74046900	2.27857100
H	-1.03324100	-5.70706000	1.25609500
H	1.16924000	-5.73815300	2.28254600
H	1.03489700	5.70878500	1.25354100
H	-3.17087200	-6.84384600	2.72889800
H	-2.46000200	-5.87334000	4.03515300
H	-4.15025000	-5.63795700	3.58377300
H	3.32941200	-6.43746900	3.84181100
H	4.24953700	-5.05018300	4.45010500
H	2.55625900	-5.24855200	4.91080500
H	3.17231900	6.84616900	2.72630200
H	2.46129000	5.87605700	4.03276900
H	4.15159800	5.64058800	3.58171900
H	-3.32684500	6.44031600	3.83973000
H	-4.24688800	5.05332900	4.44879800
H	-2.55337100	5.25156800	4.90868700
H	-9.57258000	-2.21454900	2.28995100
H	-5.08129700	-5.81428000	1.08777300
H	-7.44820200	-5.76721400	0.69338000
H	-8.54375300	-3.62825700	0.38109400
H	-8.60337300	3.56676200	0.83236400
H	-7.60766100	5.65079600	1.59504800
H	-5.23464100	5.70460800	1.98617800
H	9.57443700	2.21674400	2.28686500

H	9.64019400	-1.87399100	2.51239200
H	5.08342300	5.81631200	1.08653800
H	7.45104000	5.76918700	0.69568800
H	8.54407100	3.62942300	0.37630400
H	8.60504200	-3.56528100	0.83284000
H	7.60887200	-5.64913300	1.59504000
H	5.23601200	-5.70257700	1.98692300
H	-9.63832300	1.87619900	2.51354900
H	-10.84747600	-0.15860800	1.87554300
H	10.84918300	0.16055600	1.87345500
C	-0.69583600	1.23008000	-1.61559100
C	0.71471500	1.21477800	-1.67329500
C	1.40953900	-0.01418500	-1.63662000
C	0.69417600	-1.23192000	-1.61500200
C	-0.71637400	-1.21665800	-1.67266200
C	-1.41119000	0.01232300	-1.63653100
C	-1.38934300	2.46092500	-1.57411600
C	-2.82216200	0.02413300	-1.65440400
C	2.82051400	-0.02602900	-1.65462100
C	-1.43066800	-2.42884600	-1.78836500
C	1.38767500	-2.46274200	-1.57293600
C	1.42901800	2.42690300	-1.78954800
C	-2.80294600	2.47475200	-1.55475700
C	-3.49574200	3.71009500	-1.55688600
C	-2.78253500	4.87074400	-1.17414900
C	-1.38280000	4.87190400	-1.32965200
C	-0.67565700	3.68150600	-1.58995800
C	-3.53661100	-1.18995800	-1.77033000
C	-4.93986200	-1.16319200	-1.91892600
C	-5.63630100	0.05759000	-1.79789300

C	-4.91830100	1.27096200	-1.76377400
C	-3.51423000	1.25548500	-1.62402700
C	-0.72809000	-3.65278300	-1.88038900
C	-1.46197600	-4.77998900	-2.28909400
C	-2.82807400	-4.83949400	-1.97207300
C	-3.55125900	-3.62860800	-2.07949700
C	-2.84622800	-2.42080000	-1.83991500
C	2.80127100	-2.47658700	-1.55382000
C	3.49401600	-3.71195300	-1.55533800
C	2.78088100	-4.87233600	-1.17163300
C	1.38108000	-4.87351300	-1.32660300
C	0.67397800	-3.68331700	-1.58791300
C	3.53498700	1.18799500	-1.77104900
C	4.93822300	1.16114600	-1.91985000
C	5.63465300	-0.05963700	-1.79863600
C	4.91661400	-1.27297100	-1.76388300
C	3.51257100	-1.25737600	-1.62385200
C	0.72645100	3.65080100	-1.88230600
C	1.46045700	4.77777200	-2.29147500
C	2.82638500	4.83743700	-1.97380100
C	3.54964300	3.62656100	-2.08067500
C	2.84459300	2.41881500	-1.84089900
C	-7.04699800	0.07131400	-1.79592400
C	7.04535800	-0.07343900	-1.79702300
C	-3.37682600	-6.12358700	-1.47925400
C	3.41240600	-6.07647700	-0.57366500
C	3.37499800	6.12152700	-1.48087900
C	-3.41390700	6.07526200	-0.57677400
C	-7.76121400	-1.14643500	-1.90565700
C	-9.13216100	-1.13306900	-1.58253400

C	-9.83069800	0.08179700	-1.57117400
C	-9.11120200	1.27599300	-1.42891900
C	-7.73922500	1.30603900	-1.74627100
C	-7.01503900	2.51429300	-2.01809300
C	-7.05854600	-2.32178400	-2.33353500
C	-5.64364400	-2.34397800	-2.23756800
C	-4.92125800	-3.53395300	-2.49792800
C	-5.58737000	-4.54654700	-3.21247700
C	-6.98501100	-4.61451500	-3.15214300
C	-7.70461100	-3.43694000	-2.90585600
C	-7.63487600	3.70513800	-2.45057600
C	-6.88517600	4.88365000	-2.57736200
C	-5.48800300	4.79283500	-2.62746900
C	-4.85525200	3.69463600	-2.01784500
C	-5.60107500	2.49579400	-1.91749900
C	7.75958200	1.14426900	-1.90716800
C	9.13062400	1.13094700	-1.58446700
C	9.82917500	-0.08390300	-1.57347100
C	9.10974600	-1.27807200	-1.43069400
C	7.73758700	-1.30817200	-1.74730000
C	7.05687600	2.31953900	-2.33510900
C	7.01322600	-2.51652800	-2.01825300
C	5.64200100	2.34186300	-2.23877700
C	4.91968600	3.53193200	-2.49895400
C	5.58589100	4.54472100	-3.21315700
C	6.98353000	4.61252000	-3.15276600
C	7.70288400	3.43453700	-2.90779000
C	7.63284200	-3.70766200	-2.45028400
C	6.88307500	-4.88626700	-2.57579200
C	5.48591500	-4.79533000	-2.62589800

C	4.85338800	-3.69678200	-2.01668400
C	5.59929100	-2.49790800	-1.91721500
H	-0.87252300	5.75375400	-1.23006600
H	-1.03031000	-5.51179300	-2.85915600
H	0.87069400	-5.75516700	-1.22579500
H	1.02909700	5.50918600	-2.86226200
H	-3.26242500	-6.89146000	-2.24651000
H	-2.83957100	-6.43974600	-0.58399400
H	-4.43345700	-6.05059000	-1.23185100
H	3.28035100	-6.92780900	-1.24383800
H	4.47781600	-5.94612600	-0.39799000
H	2.94637100	-6.31358600	0.38252700
H	3.26230600	6.88898400	-2.24879700
H	2.83654000	6.43862700	-0.58668900
H	4.43115100	6.04795300	-1.23166700
H	-3.28205900	6.92614900	-1.24755500
H	-4.47925300	5.94503200	-0.40065300
H	-2.94757300	6.31298500	0.37912000
H	-9.61972700	-1.99137300	-1.31742400
H	-5.07228800	-5.19934800	-3.80763000
H	-7.46830300	-5.50716800	-3.27167000
H	-8.68809100	-3.39329100	-3.18195600
H	-8.62373800	3.72078700	-2.70979500
H	-7.34781800	5.79411500	-2.61940500
H	-4.94825900	5.49602700	-3.13729100
H	9.61822500	1.98923400	-1.31937400
H	9.58372300	-2.10454800	-1.06081200
H	5.07090100	5.19773600	-3.80815500
H	7.46694200	5.50525500	-3.27116600
H	8.68598000	3.39042400	-3.18518700

H	8.62154200	-3.72348900	-2.71009800
H	7.34562600	-5.79682500	-2.61677200
H	4.94605700	-5.49858900	-3.13550300
H	-9.58492800	2.10251600	-1.05881300
H	-10.84762300	0.09711700	-1.67297200
H	10.84602700	-0.09923200	-1.67598400

Table S7: Electronic transitions for the first 10 singlet states of HBC and CQD monomers

State	**HBC**			**CQD**		
	Orbitals	Wavelength	Osc. Stre.	Orbitals	Wavelength	Osc. Stre.
S1	150 → 153 151 → 152	395.62	0.006	262 → 265 263 → 264	552.84	0.8042
S2	150 → 152 151 → 153	380.49	0.0012	262 → 264 263 → 265	536.29	0.0068
S3	149 → 153 151 → 154	359.67	0	261 → 264 263 → 266	474.41	0
S4	149 → 152 150 → 154	356.31	0.0036	262 → 264 263 → 265	450.88	1.0147
S5	150 → 152 150 → 153 151 → 152 151 → 153	354.06	0.8151	262 → 265 263 → 264	439.51	1.413
S6	150 → 152 150 → 153 151 → 152 151 → 153	353.82	0.7616	261 → 264 263 → 266	435.1	0
S7	149 → 152 150 → 154	335.62	0.0279	260 → 264 261 → 265 262 → 266 263 → 268	424.72	0.0003
S8	149 → 153 151 → 154	333.75	0.0233	260 → 264 261 → 265 262 → 266 262 → 269	408.8	0.0004
S9	149 → 154	316.94	0.0001	259 → 264 263 → 267	402.78	0.0044
S10	147 → 153 147 → 154	300.95	0.0074	261 → 264 263 → 269	402.03	0

	148 → 153 151 → 155 151 → 157					

Table S8: Electronic transitions for the first 10 singlet states of HBC dimer (at the optimized geometry) and HBC monomer frozen in the dimeric state (at the frozen geometry of the dimeric state). Calculations were done at the B3LYP/6-31g(d,p), IEFPCM=acetonitrile level of theory.

State	**HBC Dimer**			**Monomer frozen**		
	Orbitals	Wavelength	Osc. Str.	Orbitals	Wavelength	Osc. Str.
S1	301 → 303 302 → 303 302 → 304	495.32	0.0012	150 → 152 150 → 153 151 → 152 151 → 153	457.03	0.0037
S2	300 → 303 300 → 304 302 → 306 302 → 307	439.25	0.0016	150 → 152 150 → 153 151 → 152 151 → 153	434.19	0.0025
S3	297 → 304 298 → 303 299 → 305	415.12	0	149 → 152 149 → 153 151 → 154	410.93	0
S4	299 → 306 300 → 307 301 → 307	406.31	0.0079	149 → 152 149 → 153 150 → 154	405.77	0
S5	299 → 304 300 → 303 300 → 307 301 → 307 302 → 306	385.33	1.1701	150 → 152 150 → 153 151 → 152 151 → 153	385.4	1.0881
S6	297 → 303 298 → 306 299 → 303	385.12	0.8122	150 → 152 150 → 153 151 → 152	384.87	0.9932

	299 → 308 300 → 304 301 → 306 302 → 307			151 → 153		
S7	297 → 303 297 → 304 298 → 307 300 → 308	369.81	0.0001	149 → 152 150 → 154 151 → 154	366.20	0.0001
S8	297 → 305 298 → 308	366.24	0.0004	149 → 153 150 → 154 151 → 154	363.82	0.0001
S9	296 → 305 298 → 308	354.01	0	149 → 154	351.96	0.0001
S10	296 → 304 297 → 308 301 → 310 301 → 311	326.14	0.0356	145 → 153 151 → 155	320.66	0.0031

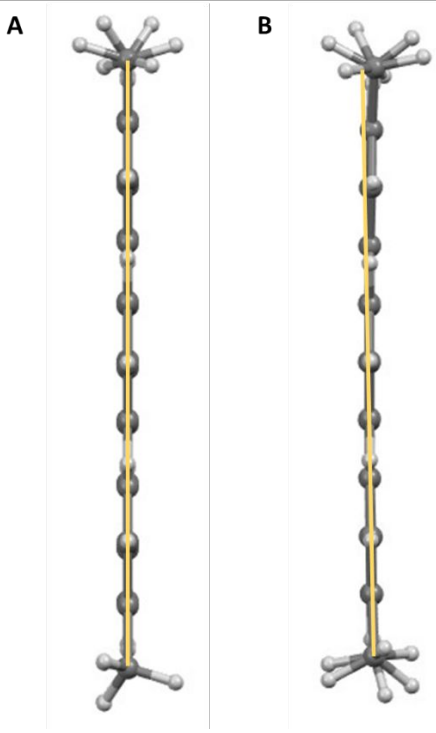


Figure S7: Comparison of a side view of calculated structures for HBC model at A) global minimum; B) geometry adopted upon dimer formation. Calculations were done at the B3LYP/6-31g(d,p), IEFPCM=acetonitrile level of theory.

B. Exciton size calculations: All calculations were performed using a locally modified copy of the development version of GAUSSIAN16.[13] All calculations used the ωB97X-D density functional[14] and the 6-31g(d) basis set. The ground-state geometry of the HBC monomer was fully optimized at the ωB97X-D/6-31g(d) level of theory. The dimer geometry was found by freezing the monomer geometries and varying the distance R and twisting angle θ until a local minimum was found. All calculations subsequently used an intermonomer separation of R = 3.45 Å and a twisting angle of $\theta = 79^{\circ}$. For the trimer and higher n-mers, the twisting angle alternated, producing a staggered formation in order to maximize point-group symmetry. Excited-state calculations were performed with time-dependent density functional theory (TDDFT) using the Tamm-Dancoff approximation (TDA).[15]

To find and analyze excitonic states, we utilized the procedure outlined in Ref. [16]. The exciton size d_{exc}, measured as the root-mean-square separation between electron and hole, was evaluated by

$$d_{exc} = \sqrt{\langle |\vec{x}_h - \vec{x}_e|^2 \rangle_{exc}} \tag{17}$$

As shown in Ref. 16, using transition density matrices $\mathbf{D}^I$ for each excited state I and multipole matrices $\boldsymbol{M}_{\xi}^{(k)}$, where ($k$) is the order of the multipole and $\xi \in$ (x, y, z), this quantity may be evaluated as

$$\begin{aligned} d_{\text{exc}}^2 = \frac{1}{\Omega} \sum_{\xi} \Big[& \text{Tr}\Big((\mathbf{D}^I)^{\dagger} \mathbf{M}_{\xi}^{(2)} \mathbf{D}^I \mathbf{S} \Big) \\ & + \text{Tr}\Big((\mathbf{D}^I)^{\dagger} \mathbf{S} \mathbf{D}^I \mathbf{M}_{\xi}^{(2)} \Big) - 2\text{Tr}\Big((\mathbf{D}^I)^{\dagger} \mathbf{M}_{\xi}^{(1)} \mathbf{D}^I \mathbf{M}_{\xi}^{(1)} \Big) \Big] \end{aligned} \tag{18}$$

where $\Omega = \text{Tr}\left((\mathbf{D}^I)^{\dagger} \mathbf{S} \mathbf{D}^I \mathbf{S} \right)$ is normalization constant. As the exciton size along the intermonomer axis is principally of interest, ξ is restricted to sum only over the z-axis. In addition to measuring the exciton size, the degree of charge-transfer character may similarly be expressed[17] in a Mülliken-like sense as

$$\Omega_{\mathrm{AB}} = \frac{1}{2}\sum_{\mu\in\mathrm{A}}\sum_{\nu\in\mathrm{B}}\left[(\mathbf{D}^I\mathbf{S})_{\mu\nu}(\mathbf{S}\mathbf{D}^I)_{\mu\nu} + (\mathbf{D}^I)^{\dagger}_{\nu\mu}(\mathbf{S}\mathbf{D}^I\mathbf{S})_{\mu\nu}\right] \tag{19}$$

where Ω_{AB} is the degree of charge transfer between monomers A and B, and where μ and ν denote atomic orbitals. The above quantities were evaluated for the lowest-energy charge-transfer state for all systems from the dimer to hexamer, with Ω_{AB} broken into charge-transfer character between monomers that were nearest neighbors, next-nearest neighbors, etc. As can be seen, total energy, exciton size, and charge-transfer character all begin to converge by the hexamer, with both of the latter metrics suggesting the exciton can delocalize over three HBC units.

Table S9: Exciton size and charge transfer character for first 20 states in the hexamer.

State	Energy (eV)	Osc. Str.	d_{exc} (z)	d_{exc}(total)	Ω_{AB} (T)	NN	N+1	N+2	N+3	N+4
1	3.173	0.001	3.435	5.262	0.22	0.2123	0.0102	0.0003	0	0
2	3.177	0.001	3.424	5.285	0.23	0.2159	0.0106	0.0004	0	0
3	3.21	0	3.426	5.199	0.19	0.1789	0.0061	0.0002	0	0
4	3.213	0	3.422	5.21	0.19	0.1802	0.0062	0.0001	0	0
5	3.295	0	3.426	5.151	0.15	0.1417	0.0036	0.0001	0	0
6	3.296	0	3.427	5.153	0.15	0.1414	0.0035	0.0001	0	0
7	3.449	0	3.543	5.328	0.18	0.1688	0.0061	0.0002	0	0
8	3.449	0	3.512	5.307	0.17	0.1624	0.0054	0.0002	0	0
9	3.474	0	3.577	5.337	0.16	0.1524	0.008	0.0003	0	0
10	3.477	0.001	3.497	5.401	0.19	0.1799	0.0104	0.0004	0	0
11	3.532	0.001	3.573	5.472	0.24	0.2279	0.0131	0.0005	0	0
12	3.533	0	3.559	5.536	0.28	0.2594	0.0159	0.0006	0	0
13	3.559	0.015	3.497	5.377	0.19	0.1776	0.0111	0.0005	0	0
14	3.566	0.014	3.579	5.369	0.19	0.1764	0.011	0.0004	0	0
15	3.633	0.005	3.648	5.846	0.45	0.4283	0.0248	0.0008	0	0
16	3.634	0.005	3.653	5.825	0.44	0.4194	0.0242	0.0008	0	0
17	3.785	0.001	3.673	5.977	0.54	0.5126	0.0221	0.0006	0	0

18	3.786	0.002	3.723	5.991	0.55	0.5221	0.0222	0.0006	0	0
19	3.859	0	3.575	5.569	0.28	0.2729	0.0104	0.0002	0	0
20	3.88	0	3.652	5.576	0.29	0.2789	0.0104	0.0002	0	0

References

1. Yang, X.; Dou, X.; Müllen, K. Efficient Synthesis of Symmetrically and Unsymmetrically Substituted Hexaphenylbenzene Analogues by Suzuki–Miyaura Coupling Reactions. *Chem. Asian J.* **2008**, *3* (4), 759-766.

2. Ito, S.; Wehmeier, M.; Brand, J. D.; Kübel, C.; Epsch, R.; Rabe, J. P.; Müllen, K. Synthesis and Self-Assembly of Functionalized Hexa-Peri-Hexabenzocoronenes. *Chem. Eur. J.* **2000**, *6* (23), 4327-4342.

3. Schlütter, F.; Nishiuchi, T.; Enkelmann, V.; Müllen, K. Π-Congested Poly(Paraphenylene) from 2,2′,6,6′-Tetraphenyl-1,1′-Biphenyl Units: Synthesis and Structural Characterization. *Polym. Chem.* **2013**, *4* (10), 2963-2967.

4. Hargenrader, G. N.; Weerasooriya, R. B.; Ilic, S.; Niklas, J.; Poluektov, O. G.; Glusac, K. D. Photoregeneration of Biomimetic Nicotinamide Adenine Dinucleotide Analogues via a Dye-Sensitized Approach. *ACS Appl. Energy Mater.* **2019**, *2* (1), 80-91.

5. Yamaguchi, S.; Hamaguchi, H.-O. Convenient Method of Measuring the Chirp Structure of Femtosecond White-Light Continuum Pulses. *Appl. Spectrosc.* **1995**, *49* (10), 1513-1515.

6. Schneider, C. A.; Rasband, W. S.; Eliceiri, K. W. NIH Image to ImageJ: 25 years of image analysis. *Nat. Methods* **2012**, *9* (7), 671-675.

7. González-Cardel, M.; Arguijo, P.; Díaz-Uribe, R. Gaussian Beam Radius Measurement with a Knife-Edge: A Polynomial Approximation to the Inverse Error Function. *Appl. Opt.* **2013**, *52* (16), 3849-3855.

8. Marciniak, H.; Li, X.-Q.; Würthner, F.; Lochbrunner, S. One-Dimensional Exciton Diffusion in Perylene Bisimide Aggregates. *J. Phys. Chem. A* **2011**, *115* (5), 648-654.

9. M. J. Frisch, G. W. T., H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, B. Mennucci, G. A. Petersson, H. Nakatsuji, M. Caricato, X. Li, H. P. Hratchian, A. F. Izmaylov, J. Bloino, G. Zheng, J. L. Sonnenberg, M. Hada, M. Ehara, K. Toyota,

R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, J. A. Montgomery, Jr., J. E. Peralta, F. Ogliaro, M. Bearpark, J. J. Heyd, E. Brothers, K. N. Kudin, V. N. Staroverov, R. Kobayashi, J. Normand, K. Raghavachari, A. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, N. Rega, J. M. Millam, M. Klene, J. E. Knox, J. B. Cross, V. Bakken, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, R. L. Martin, K. Morokuma, V. G. Zakrzewski, G. A. Voth, P. Salvador, J. J. Dannenberg, S. Dapprich, A. D. Daniels, Ö. Farkas, J. B. Foresman, J. V. Ortiz, J. Cioslowski, and D. J. Fox,. *Gaussian 09, Revision A.02, Gaussian, Inc., Wallingford CT, 2009*.

10. Becke, A. D. Density-Functional Exchange-Energy Approximation with Correct Asymptotic Behavior. *Phys. Rev. A* **1988**, *38* (6), 3098-3100.

11. Lee, C.; Yang, W.; Parr, R. G. Development of the Colle-Salvetti Correlation-Energy Formula into a Functional of the Electron Density. *Phys. Rev. B* **1988**, *37* (2), 785-789.

12. Tomasi, J.; Persico, M. Molecular Interactions in Solution: An Overview of Methods Based on Continuous Distributions of the Solvent. *Chem. Rev.* **1994**, *94* (7), 2027-2094.

13. M. J. Frisch, G. W. T., H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, G. A. Petersson, H. Nakatsuji, X. Li, M. Caricato, A. V. Marenich, J. Bloino, B. G. Janesko, R. Gomperts, B. Mennucci, H. P. Hratchian, J. V. Ortiz, A. F. Izmaylov, J. L. Sonnenberg, D. Williams-Young, F. Ding, F. Lipparini, F. Egidi, J. Goings, B. Peng, A. Petrone, T. Henderson, D. Ranasinghe, V. G. Zakrzewski, J. Gao, N. Rega, G. Zheng, W. Liang, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, K. Throssell, J. A. Montgomery, Jr., J. E. Peralta, F. Ogliaro, M. J. Bearpark, J. J. Heyd, E. N. Brothers, K. N. Kudin, V. N. Staroverov, T. A. Keith, R. Kobayashi, J. Normand, K. Raghavachari, A. P. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, J. M. Millam, M. Klene, C. Adamo, R. Cammi, J. W. Ochterski, R. L. Martin, K. Morokuma, O. Farkas, J. B. Foresman, and D. J. Fox. *Gaussian 16, Revision B.01, Gaussian, Inc., Wallingford CT, 2016*.

14. Chai, J.-D.; Head-Gordon, M. Long-Range Corrected Hybrid Density Functionals with Damped Atom–Atom Dispersion Corrections. *Phys. Chem. Chem. Phys.* **2008**, *10* (44), 6615-6620.

15. Hirata, S.; Head-Gordon, M. Time-Dependent Density Functional Theory within the Tamm–Dancoff Approximation. *Chem. Phys. Lett.* **1999**, *314* (3), 291-299.

16. Bäppler, S. A.; Plasser, F.; Wormit, M.; Dreuw, A. Exciton Analysis of Many-Body Wave Functions: Bridging the Gap Between the Quasiparticle and Molecular Orbital Pictures. *Phys. Rev. A* **2014**, *90* (5), 052521.

17. Luzanov, A. V.; Zhikol, O. A. Electron Invariants and Excited State Structural Analysis For Electronic Transitions within Cis, Rpa, and Tddft Models. *Int. J. Quantum Chem.* **2010**, *110* (4), 902-924.