

Supplementary Information: Nonuniform Screening
Reshapes Collective Excitons in Molecular
Aggregates from Stochastic Bethe-Salpeter Theory

Barry Y. Li^{1*}, Chern Chuang^{2*}, Tim Duong³, Tucker Allen⁴,
Justin R. Caram⁴, Daniel Neuhauser⁴

¹Department of Chemical and Biological Physics, Weizmann Institute
of Science, Rehovot, 7610001, Israel.

²Department of Chemistry and Biochemistry, University of Nevada, Las
Vegas, Las Vegas, NV, 89154, USA.

³Department of Chemistry, Princeton University, Princeton, NJ, 08544,
USA.

⁴Department of Chemistry and Biochemistry, University of California,
Los Angeles, Los Angeles, CA, 90095, USA.

*Corresponding author(s). E-mail(s): yangtao.li@weizmann.ac.il;
chern.chuang@unlv.edu;

Contributing authors: td0303@princeton.edu; tuckerallen27@g.ucla.edu;
jcaram@chem.ucla.edu; dxn@ucla.edu;

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1 Geometries

1.1 Cy7-DiMePh monomer geometry and electronic structure

The Cy7-DiMePh monomer is optimized using the ORCA 6.0 package at the B3LYP/def2-TZVPPD level with the D4 dispersion correction and an implicit methanol environment.^[1–5] The resulting full Cartesian coordinates are listed below. The terminal sulfonate groups of the experimental dye are replaced by methyl groups for simplicity. This choice preserves the electronic structure of the polymethine chromophore. The optimized structure exhibits the characteristic cyanine architecture consisting of a conjugated heptamethine bridge connecting two heterocyclic end groups and a central 3,5-dimethylphenyl substituent.^[6, 7]

Fig. S1 compares the highest occupied molecular orbitals (HOMOs) and lowest unoccupied molecular orbitals (LUMOs) obtained from grid-based local-density approximation (LDA) and the near-gap hybrid (ngH) CAM-LDA0 Hamiltonian used throughout the sBSE calculations. In both cases, the HOMO and LUMO remain strongly delocalized along the polymethine backbone, indicating that the low-energy optical transitions are dominated by the extended π -conjugated framework.

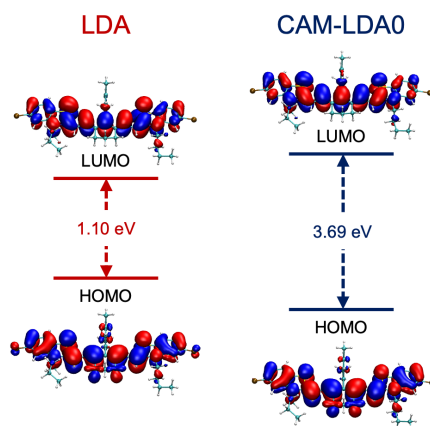


Fig. S1 HOMOs and LUMOs of the Cy7-DiMePh monomer calculated using grid-based LDA and ngH CAM-LDA0. HOMO-LUMO gaps are indicated in eV.

56 89

57 Charge = +1, 2S+1 = 1

58 C -4.80728185959975 0.46082564220115 -0.46772717392124
59 C -3.70840866375433 -0.36051897104414 -0.24607655013954
60 C -2.42012255993716 0.13648605033317 -0.09158759277093
61 C -1.26520115341928 -0.61416521577321 0.12103651738403
62 C -0.01444925006394 0.03123783247986 0.19140907001600
63 C 1.19202143747918 -0.68576967375387 0.31844999297169
64 C 2.40797115235338 -0.00624279241220 0.27297871120446
65 C 3.67369806027698 -0.57764759165306 0.31832729788738
66 C 4.83781845803628 0.17453260844845 0.21561731406180
67 C -1.33747265510561 -2.11730503558827 0.23661051741499
68 C -0.16246504345716 -2.67986817419507 1.02573705044325
69 H -1.35279281447728 -2.56092250514079 -0.76591179191062
70 H -2.27399750535267 -2.40930024504823 0.71323358576695
71 C 1.16002654077568 -2.18894231466377 0.45214258180984
72 H 1.98290825932811 -2.52227365489192 1.08603078140822
73 H 1.32799676528300 -2.64466523284495 -0.53067550012753
74 H -0.19299015498242 -3.77064763761032 1.01977752972267
75 H -0.24405669496418 -2.36233321231579 2.06946269055403
76 H -3.88603325808707 -1.42313033860627 -0.19420022396519
77 H -2.29035457097862 1.20970025611124 -0.15659086338633
78 H 2.35240206548571 1.07053318543034 0.17140756675677
79 H 3.78045341642576 -1.64692101752825 0.41132772407835
80 N -6.08453210225632 0.03421900938819 -0.64377950769699
81 S -4.71602905379388 2.20706645695317 -0.55061082124667
82 C -6.44055486775476 2.32166574555322 -0.79311042935225
83 C -7.01837019630228 1.04776848423675 -0.80714684646302
84 S 4.85310775913875 1.90889388318491 -0.02470640554886
85 C 6.59712571488409 1.92216888282921 -0.08136752600990
86 C 7.10175633902227 0.62809854982213 0.08365495625429
87 N 6.10244728056197 -0.31734829445091 0.27023963641027
88 C -7.20798772725622 3.46332332271830 -0.95373441312774
89 C -8.57864031165134 3.32522874293866 -1.12676695815519
90 C -9.14128795843206 2.05201061986540 -1.13253246665064
91 C -8.39092275530576 0.89765517497672 -0.97395609197586

92 C 7.43877248388427 3.00653727091676 -0.26552780759624
 93 C 8.80964395371908 2.78800638196856 -0.29292108380711
 94 C 9.29796478161648 1.49368350061165 -0.13859704294711
 95 C 8.47284184314394 0.39620719504820 0.04912854623245
 96 H -6.75728145583701 4.44594753761344 -0.94410960868353
 97 H -9.20132559330040 4.19862793139270 -1.25261223538723
 98 H -8.86193934390180 -0.07264396480296 -0.97738403648985
 99 H 8.88644714433339 -0.59398104603571 0.15645349232946
 100 H 7.04458692202433 4.00566351067811 -0.38842981292643
 101 H 9.48925829748808 3.61496264597024 -0.43685506715734
 102 Cl -10.87115593226966 1.89729915555444 -1.34482530871031
 103 Cl 11.02761994780082 1.23485889694570 -0.18859422986061
 104 C 0.03656227669881 1.51957095986949 0.09597994550182
 105 C 0.12236871952047 2.14309839801720 -1.14984723421759
 106 C 0.17166952277762 3.52950264065108 -1.25161528492229
 107 C 0.13286992890058 4.28728804640827 -0.07752794700768
 108 C 0.04663552107245 3.68935757289419 1.17568387213679
 109 C -0.00075698506539 2.29457532382861 1.24804715038281
 110 H 0.17066419130762 5.36886332519866 -0.14583607856734
 111 H 0.15118256163785 1.53664173358963 -2.04675225886761
 112 H -0.06759212251234 1.80971835686268 2.21478503322156
 113 C -6.45875736043259 -1.38286739129817 -0.59136667180163
 114 C -6.73171839329123 -1.84751714474109 0.83639100207938
 115 H -5.65964322146225 -1.95026117583112 -1.05991549268318
 116 H -7.34053145685759 -1.51100630066135 -1.21502747007851
 117 C -7.07620748795327 -3.33561723488253 0.91628554312010
 118 H -7.55566647437482 -1.25527547000223 1.24089641350274
 119 H -5.85465327403164 -1.63811689882530 1.45464216904696
 120 C 6.39705194706212 -1.74302102328901 0.45183611564736
 121 C 6.43439568644155 -2.49358733381537 -0.87644673228570
 122 H 5.64345022434731 -2.15979332177362 1.11669966539433
 123 H 7.34586035674289 -1.80719059633754 0.97743257789342
 124 C 6.69183220515060 -3.99074957407641 -0.70039980128666
 125 H 7.20814749845180 -2.05483386987622 -1.51239185807216
 126 H 5.48033861601305 -2.34495207130620 -1.38715415043395
 127 C 0.26276484886009 4.20611440948891 -2.59172151658453

128 C 0.00621163188003 4.51494382516779 2.43223903773819
 129 C -5.91579691810879 -4.26264191503606 0.55857888074982
 130 H -7.93079399358454 -3.54546225721270 0.26536790059665
 131 H -7.40703410092135 -3.55359542889946 1.93475183768279
 132 H -5.60510091642769 -4.15083466910199 -0.48183419195799
 133 H -6.19549954768606 -5.30828167832688 0.70263941156238
 134 H -5.04535200378314 -4.06262706551462 1.18873161710746
 135 C 8.07560823185203 -4.32782461334039 -0.14868745097970
 136 H 6.56756600502887 -4.47320336322289 -1.67313680015951
 137 H 5.92127256847330 -4.41383482861636 -0.04818955443931
 138 H 8.86253683390529 -3.90754963618954 -0.78033029624626
 139 H 8.22526961473461 -5.40870928204437 -0.10871147227783
 140 H 8.22025532950043 -3.94224369313536 0.86243251787764
 141 H 0.29961320937577 3.47819636221360 -3.40208752612934
 142 H 1.15584977281854 4.83203043511360 -2.65867609269891
 143 H -0.59698993268481 4.85843151544811 -2.76362438902762
 144 H 0.05665653229722 5.58054863803861 2.20904590443838
 145 H 0.84065302786171 4.26810915550027 3.09309026866330
 146 H -0.91152781435571 4.33097558325504 2.99612523968880

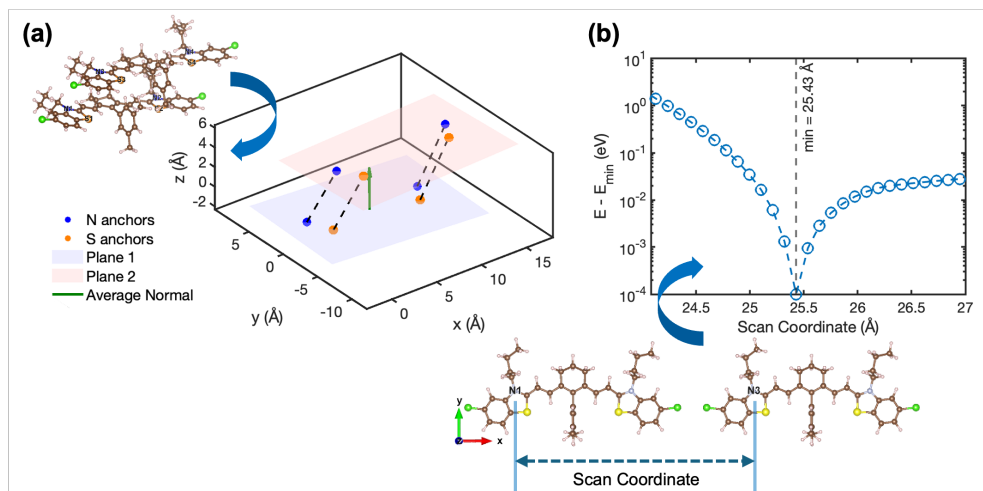


Fig. S2 (a) Definition of intermolecular π - π stacking geometry used to construct aggregate configurations. Molecular planes are defined using sulfur and nitrogen anchor atoms, and average plane normals are used to characterize the inter-layer distance of 4.72 Å. (b) Potential energy surface scan to find the lateral spacing between adjacent molecules. The scanning coordinates are the N–N distance between two molecules along their “long axis”. A logarithmic scale is used for the total energy difference, $E - E_{\min}$. An optimal distance of 25.43 Å is identified.

Aggregate structures are constructed from the optimized monomer geometry. As illustrated in Fig. S2 (a), the intermolecular π - π stacking distance is determined from a constrained dimer optimization. Molecular planes are defined using sulfur and nitrogen anchor atoms, and average plane normals are used to characterize the relative orientation of neighboring chromophores. The optimized dimer yields an equilibrium inter-layer separation of approximately 4.72 Å, which was subsequently adopted for all aggregate constructions.

The lateral displacement along the molecular long (N–N) axis is determined by a potential energy surface scan by translating one monomer relative to the other. As shown in Fig. S2 (b), the total energy exhibits a clear minimum at 25.43 Å. Therefore, this distance was used as the equilibrium lateral spacing for aggregate construction. Both the constrained dimer optimization and the lateral potential energy surface scan

160 are performed in the ORCA 6.0 package using the r^2 SCAN-3c method with an implicit
161 water solvent.[\[4, 5, 8\]](#)

162 Using the optimized π - π separation and lateral spacing, different aggregate clusters
163 are generated by systematically varying the intermolecular slip along the molecular
164 “long axis.” Slip distances of 2.60 Å, 4.60 Å, and 6.60 Å are employed to represent
165 H -, I -, and J -like packing motifs, respectively. These values span the experimentally
166 relevant range reported for cyanine aggregates and provide a controlled structural
167 series.[\[9–12\]](#)

168 2 General Information for Computation

169 The computational parameters used for all systems are summarized in Table [S1](#).

Table S1 Grid and active-space parameters used in this work.

	Monomer			Dimer 2×1			Trimer 3×1			Tetramer 4×1			Aggregate 4×3		
	H	I	J	H	I	J	H	I	J	H	I	J	H	I	J
dx	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
n_x	108	118	132	128	142	158	138	160	182	326	350	372	326	350	372
n_y	66	66	66	66	66	66	66	66	66	66	66	66	66	66	66
n_z	50	68	68	86	86	86	102	102	102	102	102	102	102	102	102
N_v	40	80	80	120	120	120	160	160	160	160	160	160	240	240	240
N_c	120	240	240	360	360	360	480	480	480	480	480	480	720	720	720
$N_{k\text{-low}}$	749	1123	1249	1515	1721	1927	1959	2271	2599	4649	4969	5305	4649	4969	5305

Real-space computational grids, where a uniform grid in Bohr is used, i.e., $dx = dy = dz$. N_v and N_c denote the number of occupied and virtual orbitals included in the near-gap active space, respectively. $N_{k\text{-low}}$ is the number of deterministically treated long-wavelength terms, and the high- k space is represented with a similar number of sparse stochastic vectors. Details in Ref. [13].

170 3 Details of the Parameterized Screened Interaction 171 and sBSE Formalism

172 3.1 Stochastic occupied-pair sampling

173 A stochastic valence occupied orbital is constructed as

$$174 \quad \bar{\beta}(\mathbf{r}) = \sum_{i \in N_v} \bar{\beta}_i \phi_i(\mathbf{r}), \quad \bar{\beta}_i = \pm 1, \quad (\text{S1})$$

175 and another statistically independent orbital is,

$$176 \quad \bar{\bar{\beta}}(\mathbf{r}) = \sum_{j \in N_v} \bar{\bar{\beta}}_j \phi_j(\mathbf{r}), \quad \bar{\bar{\beta}}_j = \pm 1, \quad (\text{S2})$$

177 Together these orbitals form the occupied-pair density

$$178 \quad \beta(\mathbf{r}) = \bar{\beta}(\mathbf{r}) \bar{\bar{\beta}}(\mathbf{r}). \quad (\text{S3})$$

179 Statistical averaging over a large number of stochastic realizations recovers the
180 deterministic occupied-pair density,

$$181 \quad \{\beta(\mathbf{r})\} = \left\{ \bar{\beta}_i \bar{\bar{\beta}}_j \bar{\beta}(\mathbf{r}) \bar{\bar{\beta}}(\mathbf{r}) \right\} = \phi_i(\mathbf{r}) \phi_j(\mathbf{r}) \quad (\text{S4})$$

182 The above expression uses $\{\bar{\beta}_i \bar{\beta}_j\} = \{\bar{\bar{\beta}}_i \bar{\bar{\beta}}_j\} = \delta_{ij}$ in limit $N_\beta \rightarrow \infty$. In practice, a
183 small number of samples is sufficient to converge the screened-exchange kernel.

184 For each stochastic density, the bare perturbing potential is

$$185 \quad v_\beta(\mathbf{r}) = \int v(\mathbf{r} - \mathbf{r}') \beta(\mathbf{r}') d\mathbf{r}', \quad (\text{S5})$$

186 and the induced density, $\Delta n_\beta(\mathbf{r})$, is obtained by short-time linear-response
 187 propagation.[14, 15] The corresponding screened-response potential is

$$188 \quad W_\beta(\mathbf{r}) = \int v(\mathbf{r} - \mathbf{r}') \Delta n_\beta(\mathbf{r}') d\mathbf{r}'. \quad (\text{S6})$$

189 3.2 Connection to the form-free screened interaction

190 In the original optimized attenuated interaction formalism,[7, 15, 16] the screened
 191 interaction is obtained by minimizing the functional

$$192 \quad J = \sum_{ij} (\phi_i \phi_j | (W - v_W)^2 | \phi_i \phi_j), \quad (\text{S7})$$

193 where W denotes the screened Coulomb interaction and v_W is the effective screened
 194 exchange kernel. Replacing the orbital-pair products $\phi_i \phi_j$ by stochastic pair densities,
 195 $\beta(\mathbf{r})$, and moving to reciprocal space yields the form-free estimator,

$$196 \quad v_W(\mathbf{k}) = \left\{ \frac{\beta^*(\mathbf{k}) \langle \mathbf{k} | W | \beta \rangle}{|\langle \mathbf{k} | \beta \rangle|^2} \right\}_\beta, \quad (\text{S8})$$

197 where the braces denote averaging over stochastic samples.

198 The form-free representation is mathematically attractive because each reciprocal-
 199 space grid point acts as an independent degree of freedom. In principle, this allows
 200 an exact representation of the screened interaction on the chosen reciprocal-space
 201 mesh. However, the resulting $v_W(\mathbf{k})$ is noisy in the large- $|\mathbf{k}|$ region and remains
 202 tied to a specific reciprocal-space grid. On the other hand, the previously employed
 203 one-dimensional seven-parameter parameterization combined with isotropic three-
 204 dimensional reconstruction is often too restrictive to reproduce the actual dielectric
 205 screening. Therefore, we develop here a compact isotropic+anisotropic representation
 206 that retains the physically relevant screening information while reducing stochastic
 207 noise and improving transferability.[17]

208 3.3 Polynomial-Gaussian parameterization

209 As shown in the main text, we expand $g(\mathbf{k})$ in a finite polynomial-Gaussian basis,
 210 where $N_{\text{basis}} = 16$. The basis functions are

$$\begin{aligned}
 f_1(\mathbf{k}) &= e^{-k^2/0.1}, f_2(\mathbf{k}) = e^{-k^2/0.3}, f_3(\mathbf{k}) = e^{-k^2}, f_4(\mathbf{k}) = e^{-k^2/3.0}, \\
 f_5(\mathbf{k}) &= k_x^2 e^{-k^2/0.3}, f_6(\mathbf{k}) = k_y^2 e^{-k^2/0.3}, f_7(\mathbf{k}) = k_z^2 e^{-k^2/0.3}, \\
 f_8(\mathbf{k}) &= k_x^2 e^{-k^2}, f_9(\mathbf{k}) = k_y^2 e^{-k^2}, f_{10}(\mathbf{k}) = k_z^2 e^{-k^2}, \\
 f_{11}(\mathbf{k}) &= k_x^4 e^{-k^2}, f_{12}(\mathbf{k}) = k_y^4 e^{-k^2}, f_{13}(\mathbf{k}) = k_z^4 e^{-k^2}, \\
 f_{14}(\mathbf{k}) &= k_x^2 k_y^2 e^{-k^2}, f_{15}(\mathbf{k}) = k_x^2 k_z^2 e^{-k^2}, f_{16}(\mathbf{k}) = k_y^2 k_z^2 e^{-k^2},
 \end{aligned} \tag{S9}$$

212 with $k^2 = k_x^2 + k_y^2 + k_z^2$. The first four basis functions describe isotropic radial screening,
 213 and the remaining terms introduce anisotropy and mixed-direction couplings.

214 3.4 Difference from the form-free representation

215 In the form-free representation, every reciprocal-space grid point acts as an indepen-
 216 dent degree of freedom. For a reciprocal-space mesh of size $2n_x \times 2n_y \times 2n_z$ (the
 217 doubling comes from the use of the Martyna-Tuckerman approach for electron-electron
 218 interactions to reduce periodic images), [18] the screened interaction contains approx-
 219 imately $8n_x n_y n_z$ independent variables. The present parameterization replaces this
 220 by only $N_{\text{basis}} = 16$ coefficients, where $v_W(\mathbf{k}) \rightarrow \{\theta_m\}_{m=1}^{16}$. More importantly, this
 221 parameterized kernel is transferable, i.e., it is no longer grid-dependent. With these
 222 16 parameters, it can be reconstructed on any arbitrary grid for the same or different
 223 molecular systems.

224 3.5 Smooth physical filtering

225 The finite basis expansion may introduce small overfitting artifacts, particularly in
 226 regions where the stochastic signal is weak. As a result, the reconstructed inverse
 227 dielectric function may occasionally exceed the physically allowed interval of $0 <$
 228 $\epsilon^{-1}(\mathbf{k}) \leq 1$. To suppress such unphysical excursions, a smooth error-function filter is
 229 applied. First, the raw parameterization is constructed as

$$230 \quad \epsilon^{-1}(\mathbf{k}) = 1 + g^{\text{param.}}(\mathbf{k}). \quad (\text{S10})$$

231 The upper bound is enforced through

$$232 \quad \epsilon_1^{-1}(\mathbf{k}) = \epsilon^{-1}(\mathbf{k}) - \frac{\epsilon^{-1}(\mathbf{k}) - 1}{2} [1 + \text{erf}(\alpha[\epsilon^{-1}(\mathbf{k}) - 1])], \quad (\text{S11})$$

233 which smoothly suppresses values exceeding unity. The lower bound is then imposed
 234 through

$$235 \quad \epsilon_{\text{filt.}}^{-1}(\mathbf{k}) = \epsilon_1^{-1}(\mathbf{k}) - \frac{\epsilon_1^{-1}(\mathbf{k})}{2} [1 - \text{erf}(\alpha \epsilon_1^{-1}(\mathbf{k}))]. \quad (\text{S12})$$

236 Throughout this work, $\alpha = 16$ is used. Finally,

$$237 \quad g_{\text{filt.}}^{\text{param.}}(\mathbf{k}) = \epsilon_{\text{filt.}}^{-1}(\mathbf{k}) - 1. \quad (\text{S13})$$

238 The filtering is applied only after the least-squares optimization and does not modify
 239 the fitting procedure itself.

240 3.6 sBSE spectra

241 The sBSE calculations are constructed from the ngH Hamiltonian, [1, 13] $H_{\text{ngH}} \phi_p(\mathbf{r}) =$
 242 $\varepsilon_p \phi_p(\mathbf{r})$, where $\phi_p(\mathbf{r})$ and ε_p denote molecular orbitals and orbital energies, respec-
 243 tively. Occupied orbitals are labeled by i, j , virtual orbitals by a, b , and general orbitals

244 by p, q . Neutral excitations are represented in the occupied-virtual transition basis,
 245 $|ia\rangle \equiv \phi_i(\mathbf{r})\phi_a(\mathbf{r})$, with independent-particle excitation energies, $\Delta\varepsilon_{ia} = \varepsilon_a - \varepsilon_i$.

246 The sBSE excitation problem is solved in the occupied-virtual transition basis.[14,
 247 15, 19, 20] We use the full resonant-antiresonant form rather than the Tamm-Dancoff
 248 approximation. The eigenvalue problem is

$$249 \quad \mathcal{L} \begin{pmatrix} X \\ Y \end{pmatrix} = \begin{pmatrix} \mathcal{A} & \mathcal{B} \\ -\mathcal{B} & -\mathcal{A} \end{pmatrix} \begin{pmatrix} X \\ Y \end{pmatrix} = \Omega \begin{pmatrix} X \\ Y \end{pmatrix}, \quad (\text{S14})$$

250 where $\mathcal{L}$ denotes the BSE Liouvillian, Ω is the excitation energy and X and Y are
 251 resonant and antiresonant amplitudes, respectively. The resonant block is

$$252 \quad \mathcal{A}_{ia,jb} = \Delta\varepsilon_{ia}\delta_{ij}\delta_{ab} + K_{ia,jb}^{\text{dir.}} + K_{ia,jb}^{\text{sx.}}, \quad (\text{S15})$$

253 and the coupling block is

$$254 \quad \mathcal{B}_{ia,jb} = K_{ia,bj}^{\text{dir.}} + K_{ia,bj}^{\text{sx.}}. \quad (\text{S16})$$

255 The direct Coulomb term for singlet excitations is

$$256 \quad K_{ia,jb}^{\text{dir.}} = 2 \iint \phi_i(\mathbf{r})\phi_a(\mathbf{r})v(\mathbf{r} - \mathbf{r}')\phi_j(\mathbf{r}')\phi_b(\mathbf{r}')d\mathbf{r}d\mathbf{r}'. \quad (\text{S17})$$

257 The screened-exchange term is

$$258 \quad K_{ia,jb}^{\text{sx.}} = - \iint \phi_i(\mathbf{r})\phi_j(\mathbf{r})v_W^{\text{param.}}(\mathbf{r} - \mathbf{r}')\phi_a(\mathbf{r}')\phi_b(\mathbf{r}')d\mathbf{r}d\mathbf{r}'. \quad (\text{S18})$$

259 The parameterization allows the screened-exchange action to be evaluated without
 260 storing the full four-index interaction tensor.

261 Direct diagonalization of the sBSE Liouvillian is computationally expensive.
 262 Instead, we evaluate spectral quantities through a Chebyshev polynomial expansion of
 263 $\mathcal{L}$. For a given polarization direction, $\hat{r} = x, y, z$, we define the dipole excitation vector,

$$|\chi\rangle = \begin{pmatrix} \chi_{ia}^+ \\ \chi_{ia}^- \end{pmatrix} = \begin{pmatrix} +\langle\phi_a|\hat{r}|\phi_i\rangle \\ -\langle\phi_a|\hat{r}|\phi_i\rangle \end{pmatrix}. \quad (\text{S19})$$

The optical absorption spectrum is obtained from the spectral density associated with this dipole vector,

$$A(\omega) \propto \omega \langle \chi | \delta(\mathcal{L} - \omega) | \chi \rangle. \quad (\text{S20})$$

To evaluate Eq. (S20), the Liouvillian is shifted and scaled to produce a dimensionless operator, $\tilde{\mathcal{L}}$, whose spectrum lies within the interval $[-1, 1]$. The broadened spectral projector is then expanded in Chebyshev polynomials,[20–22]

$$\delta(\mathcal{L} - \omega) = \sum_{l=0}^{N_{\text{Cheb.}}} a_l(\omega) T_l(\tilde{\mathcal{L}}), \quad (\text{S21})$$

where T_l is the l^{th} -order Chebyshev polynomial and $a_l(\omega)$ are the corresponding expansion coefficients. The absorption cross section is calculated from the Chebyshev moments,

$$\sigma(\omega) = \frac{4\pi\omega}{c} \sum_{l=0}^{N_{\text{Cheb.}}} a_l(\omega) \langle \tilde{\chi} | T_l(\tilde{\mathcal{L}}) | \chi \rangle, \quad (\text{S22})$$

where $c \simeq 137$ in atomic units, $|\tilde{\chi}\rangle = \pm|\chi\rangle$, and a simple decaying weight is used for the coefficients, $a_l(\omega)$. In our simulations, $N_{\text{Cheb.}} = 2,000$ is used across all systems. Thus, all spectra are evaluated with the same numerical broadening determined by the Chebyshev expansion; no explicit physical dephasing model is introduced. At a selected energy, ω , the Chebyshev-filtered exciton amplitude is represented as $F_{ia}(\omega) = X_{ia}(\omega) + Y_{ia}(\omega)$.

The filtered amplitudes are normalized according to

$$\sum_{ia} |F_{ia}(\omega)|^2 = 1, \quad (\text{S23})$$

284 and the exciton participation ratio is

$$285 \quad \text{PR}(\omega) = \frac{1}{\sum_{ia} |F_{ia}(\omega)|^4}. \quad (\text{S24})$$

286 The corresponding real-space transition density is

$$287 \quad n_{\text{tr}}(\mathbf{r}; \omega) = \sum_{ia} F_{ia}(\omega) \phi_i(\mathbf{r}) \phi_a(\mathbf{r}). \quad (\text{S25})$$

288 The bare transition electrostatic potential is

$$289 \quad \Phi_{\text{tr}}^{\text{bare}}(\mathbf{r}; \omega) = \int \frac{n_{\text{tr}}(\mathbf{r}'; \omega)}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'. \quad (\text{S26})$$

290 Including the nonlocal dielectric response gives the screened transition electrostatic
291 potential,

$$292 \quad \Phi_{\text{tr}}^{\text{scr}}(\mathbf{r}; \omega) = \iint \epsilon^{-1}(\mathbf{r} - \mathbf{r}') \frac{n_{\text{tr}}(\mathbf{r}''; \omega)}{|\mathbf{r}' - \mathbf{r}''|} d\mathbf{r}' d\mathbf{r}'', \quad (\text{S27})$$

293 where the same screened interaction employed in the sBSE Hamiltonian is used
294 consistently in the post-processing analysis.

295 **3.7 Spectral validation of all- $\mathbf{k}$ and parameterized v_W**

296 A comparison between the form-free all- $\mathbf{k}$ and parameterized v_W representations is
297 shown in Fig. [S3](#).

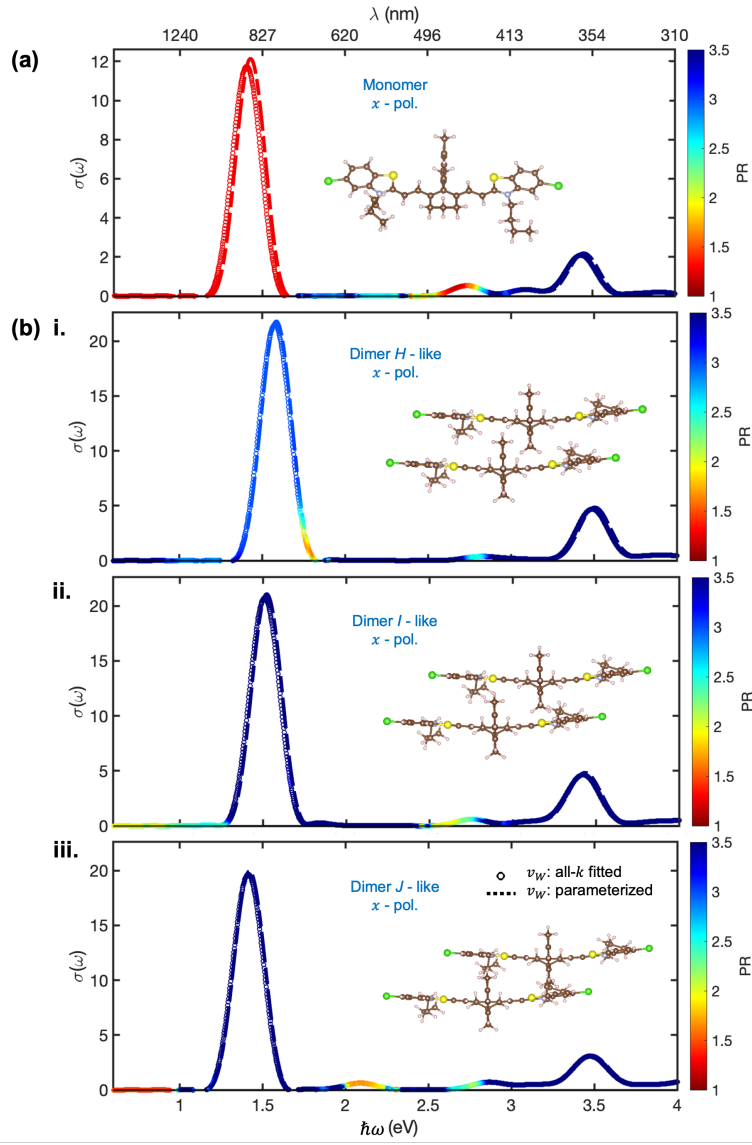


Fig. S3 (a) sBSE absorption spectrum with participation ratio of the Cy7-DiMePh monomer under x -polarization. Open-circle spectra correspond to calculations using the all- k fitted screened interaction v_W , while dashed curves correspond to a compact parameterized v_W . (b) Similar to panel (a), but for i. H -, ii. I -, and iii. J -like 2×1 dimers.

298 4 v_W Parameters

299 The optimized polynomial-Gaussian coefficients for the monomer and 4×1 aggregates
 300 are listed in Table S2.

Table S2 Parameterized polynomial-Gaussian expansion coefficients, θ_m , for the parameterized v_W of the Cy7-DiMePh monomer and 4×1 aggregates.

m	Monomer	H -like 4×1	I -like 4×1	J -like 4×1
1	3.100551	4.943122	5.021161	4.633988
2	23.145005	39.898093	41.043609	36.974514
3	-28.500174	-50.937019	-52.464969	-47.125475
4	2.194692	5.998689	6.314311	5.438713
5	44.143239	78.298876	80.634325	73.618073
6	47.928539	81.272304	83.119569	74.474127
7	52.994778	88.478807	90.022103	81.100479
8	25.934359	42.135131	42.943137	38.427649
9	23.697053	40.298343	41.467467	37.823616
10	20.790429	36.864283	38.078408	34.561447
11	-9.106951	-15.244795	-15.578933	-13.726605
12	-7.700937	-14.169292	-14.687103	-13.388729
13	-5.884088	-12.075102	-12.625344	-11.362575
14	-17.412537	-29.954405	-30.806910	-27.591424
15	-15.961918	-28.525687	-29.292327	-26.128510
16	-14.371624	-26.778774	-27.859911	-25.358416

301 5 Transition Electrostatic Potential Analysis

302 To quantify the spatial organization of the screened transition electrostatic potential,
 303 the positive and negative potential lobes above a common absolute potential threshold,
 304 Φ_{cut} , are analyzed separately. Their weighted centroids are defined as

$$305 \quad R_{\pm} = \frac{\int_{\Omega_{\pm}} \mathbf{r} |\Phi_{\text{tr}}^{\text{scr}}(\mathbf{r})| d\mathbf{r}}{\int_{\Omega_{\pm}} |\Phi_{\text{tr}}^{\text{scr}}(\mathbf{r})| d\mathbf{r}}, \quad \Omega_{\pm} = \{\mathbf{r} : \pm \Phi_{\text{tr}}^{\text{scr}}(\mathbf{r}) \geq \Phi_{\text{cut}}\}, \quad (\text{S28})$$

while the average RMS extent of the positive and negative potential lobes along the polarization direction, i.e., the x -axis, is given by

$$L_x = \frac{1}{2} \sum_{\sigma=\pm} \left[\frac{\int_{\Omega_\sigma} (x - x_\sigma)^2 |\Phi_{\text{tr}}^{\text{scr}}(\mathbf{r})| d\mathbf{r}}{\int_{\Omega_\sigma} |\Phi_{\text{tr}}^{\text{scr}}(\mathbf{r})| d\mathbf{r}} \right]^{1/2}, \quad (\text{S29})$$

where x_σ denotes the x -coordinate of the corresponding centroid R_σ .

Quantitative analysis of the screened transition electrostatic potential for the H -, I -, and J -like 4×3 aggregate clusters is shown in Table S3. The positive and negative potential lobes are analyzed using common absolute potential thresholds ranging from 8 to 16 a.u. At these thresholds, the analyzed isosurfaces are fully enclosed within their native computational boxes for all three aggregate motifs, ensuring that the reported geometric descriptors are free from finite-box truncation effects.

Table S3 For each Φ_{cut} , the centroid separation, d_c , average longitudinal extent, L_x , RMS lobe radius, R_{RMS} , and volume of the high-magnitude potential region, V_{iso} are computed from the positive and negative regions.

Φ_{cut} (a.u.)	Agg. Motif	d_c (Å)	L_x (Å)	R_{RMS} (Å)	V_{iso} (Å ³)
8	H	55.38	11.88	13.23	8373
	I	50.38	12.43	13.58	7338
	J	47.33	11.09	12.35	4713
10	H	54.63	11.40	12.45	4671
	I	47.47	12.44	13.28	3653
	J	41.30	10.10	11.15	1660
12	H	54.86	10.82	11.65	2520
	I	44.52	12.18	12.83	1630
	J	35.23	7.52	8.67	584
14	H	56.04	10.12	10.76	1215
	I	37.76	10.74	11.33	566
	J	33.20	5.26	6.06	229
16	H	58.27	8.51	9.02	457
	I	29.97	8.93	9.55	174
	J	31.94	3.84	4.41	90

316 6 Frenkel Exciton Analysis

317 6.1 Real-space Frenkel projection

318 Using the site-resolved monomer transition densities defined in the main text, the
319 Frenkel reconstruction is

$$320 \quad n_{\text{tr}}^F(\mathbf{r}) = \sum_{i=1}^N c_i n_{\text{tr}}^{(i)}(\mathbf{r}), \quad (\text{S30})$$

321 where N is the number of monomers and $\{c_i\}$ are the Frenkel coefficients that satisfy
322 $\sum_i c_i^2 = 1$. The quality of the reconstruction is quantified by the normalized overlap
323 between $n_{\text{tr}}^F(\mathbf{r})$ and $n_{\text{tr}}^{\text{agg}}(\mathbf{r})$, i.e., the projected overlap.[23, 24]

324 For comparison, an equal-weight Frenkel transition density is also constructed,

$$325 \quad n_{\text{tr}}^{\text{EW}}(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{i=1}^N n_{\text{tr}}^{(i)}(\mathbf{r}), \quad (\text{S31})$$

326 where each monomer contributes with the same amplitude. The equal-weight overlap
327 is therefore defined as the normalized overlap between $n_{\text{tr}}^{\text{EW}}(\mathbf{r})$ and $n_{\text{tr}}^{\text{agg}}(\mathbf{r})$.

328 6.2 Optimized Frenkel coefficients

329 Fig. S4 presents the normalized coefficients obtained from the least-squares projec-
330 tion onto the translated monomer transition-density basis for the H -, I -, and J -like
331 aggregate clusters. Each rectangle represents one monomer in the 12-molecule aggre-
332 gate, with the color intensity and numerical value indicating its normalized expansion
333 coefficient. The coefficients satisfy the normalization condition, i.e., $\sum_i c_i^2 = 1$.

334 Although the coefficient distributions differ among the three packing motifs, the
335 transition densities remain well-represented within the Frenkel basis, consistent with
336 the high projection overlaps reported in the main text. These coefficients therefore
337 provide a real-space visualization of how the aggregate excitation is distributed among
338 the localized monomer excitations.

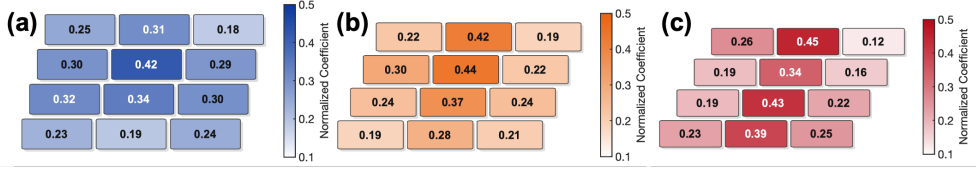


Fig. S4 Normalized Frenkel expansion coefficients for the (a) *H*-, (b) *I*-, and (c) *J*-like aggregate clusters, obtained by least-squares projection of the sBSE transition density onto the translated monomer transition-density basis. The blue, orange, and red color schemes, respectively, are chosen to indicate the characteristic absorption colors of the three aggregate motifs. Each rectangle represents one monomer in the 12-molecule aggregate, with both the color scale and numerical label indicating the normalized coefficient.

6.3 Screening effects in 1D Frenkel excitons

For the transition-charge analysis, the parameterized interaction kernel $v_W(\mathbf{k})$ is transformed to its translationally invariant real-space representation, $v_W(\mathbf{k}) \xrightarrow{\mathcal{F}^{-1}} v_W(\mathbf{r} - \mathbf{r}')$. The corresponding real-space inverse dielectric screening factor is

$$\epsilon^{-1}(\mathbf{r} - \mathbf{r}') = 1 + \frac{v_W(\mathbf{r} - \mathbf{r}')}{v(\mathbf{r} - \mathbf{r}')}.$$
 (S32)

Since the parameterized kernel is translationally invariant, we choose $\mathbf{r}' = 0$ as the reference point when evaluating and visualizing the real-space screening function. Thus, the $\epsilon^{-1}(\mathbf{r}_{kl})$ used in the screened transition-charge couplings is obtained from the real-space interaction kernel and is not the direct Fourier transform of $\epsilon^{-1}(\mathbf{k})$.

Compared with the point-dipole approximation, where the coupling between two selected sites, n and m , separated by r_{nm} , is screened by $\epsilon^{-1}(r_{nm})$, the screening of individual transition-charge pairs is expected to differ, especially when the dimension of the molecule, or the extent of the transition density, is comparable to or larger than the center-to-center distance. Also, for chromophores with a large aspect ratio, we expect the effect of screening to be more different from the point dipole limit. The transition charges corresponding to the ground and the first excited state of molecules are calculated using the ZINDO method, with chlorine atoms swapped with fluorine.^[25, 26]

357 The unusual observation of “anti-screening” discussed in the main text arises from
 358 the combination of the large aspect ratio and strong polarity (extended dipole char-
 359 acter) of the transition charge distribution with range-sensitive screening. In the bare
 360 coupling case, the contributions from the sum over charge pairs largely cancel them-
 361 selves, while some of these terms correspond to those between closely separated pairs
 362 and are consequently strongly screened, leaving a strong nonzero cumulative contribu-
 363 tion. More specifically, at this particular (but highly relevant) slipped geometry, the
 364 nearest-neighbor molecules are offset to a degree such that the negative lobe of the
 365 transition charge distribution of one molecule lines up with the positive lobe of the
 366 other, creating a strong negative contribution to the excitonic coupling. On the other
 367 hand, there are the two “diagonal” contributions, referring to the positive-positive
 368 and negative-negative charge pairs across the two molecules, each contributing posi-
 369 tively at roughly half the amplitude, together with the far-end positive-negative pair,
 370 which gives only a negligible negative contribution. These four parts end up nearly
 371 canceling one another at their unscreened values. With short-range screening, only the
 372 first strong negative contribution is significantly reduced in magnitude, leaving a net
 373 positive excitonic coupling between the two molecules.

374 The above scheme only occurs at a specific value of slip (7.5 Å) for the Cy7-DiMePh
 375 dye as expected. To better understand the full picture of the screening effects in this
 376 context, we need to explore all plausible packing geometries that are experimentally
 377 relevant. In Fig. S5 we examine the dependence of J_1 and J_2 as functions of the slip
 378 offset between successive molecules in a 1D aggregate, and compare the effects of
 379 screening. As anticipated, only minute differences can be found between screened and
 380 unscreened J_2 , while J_1 is significantly altered. For a constant screening scheme the
 381 two curves should be simply related by a multiplicative factor $\epsilon^{-1}(r) = \epsilon_0^{-1}$, where
 382 $r \equiv |\mathbf{r} - \mathbf{r}'|$. Here, instead, the screened J_1 is affected not only in terms of magnitude

383 but also in terms of the onset of sign-flipping as the geometry transitions from an H -
 384 to a J -aggregate.

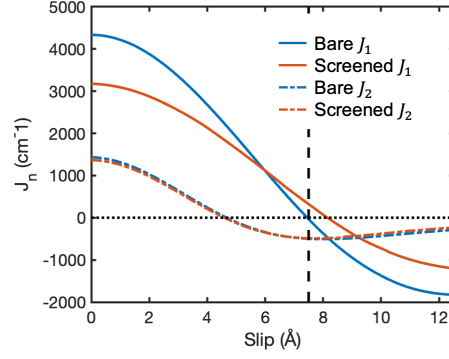


Fig. S5 The dependence of the nearest-neighbor coupling, J_1 , on the slip parameter of the 1D aggregate and that of the next-nearest neighbor, J_2 . Both couplings are shown with and without screening derived from a J -like 4×1 cluster. The 7.5 Å slip used in the main text analysis is indicated by the vertical dashed line.

385 As a result of the screening-induced change in the excitonic band structure, shown
 386 in Fig. S6, the density of states of these systems, while possessing similar van Hove
 387 singularity features due to the double-well geometry of the band, exhibits a reversed
 388 order of these van Hove peaks, with one of them corresponding to the bright state
 389 (vertical dashed lines). Consequently, the unscreened aggregate is predicted to be
 390 emissive while the screened one is not.

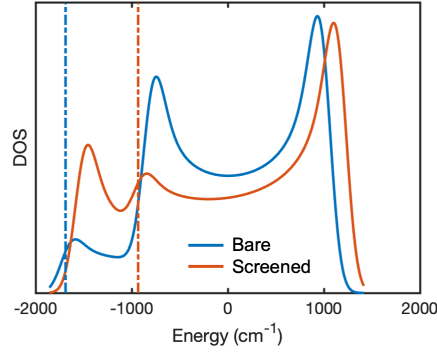


Fig. S6 Density of states (DOS) corresponding to the unscreened and screened excitonic bands shown in the main text. The screening is derived from a J -like 4×1 cluster.

391 7 Active-space Convergence Check

392 The active-space convergence of the 4×3 optical gaps is summarized in Table S4.

Table S4 Active-space convergence of the x -polarized sBSE optical gaps for 4×3 H -, I -, and J -like aggregates. Linear fits were performed against $1/(N_c + N_v)$ using $E_{\text{gap}} = a/(N_c + N_v) + b$, where b gives the extrapolated infinite-active-space limit.

N_v	N_c	H -like gap (eV)	I -like gap (eV)	J -like gap (eV)
40	120	1.72	1.68	1.52
80	240	1.62	1.56	1.36
120	360	1.54	1.48	1.30
160	480	1.50	1.42	1.24
200	600	1.48	1.38	1.20
240	720	1.44	1.36	1.16
∞		1.42	1.33	1.13
Difference		0.02	0.03	0.03

The difference is computed between the largest active-space calculation, $(N_v, N_c) = (240, 720)$, and the extrapolated infinite-active-space limit.

393 8 Additional Spectral and Electronic Structure

394 Results

395 Additional spectra for the 3×1 and 4×1 aggregates are shown in Fig. S7. Key
 396 electronic and optical properties for all systems are collected in Table S5.

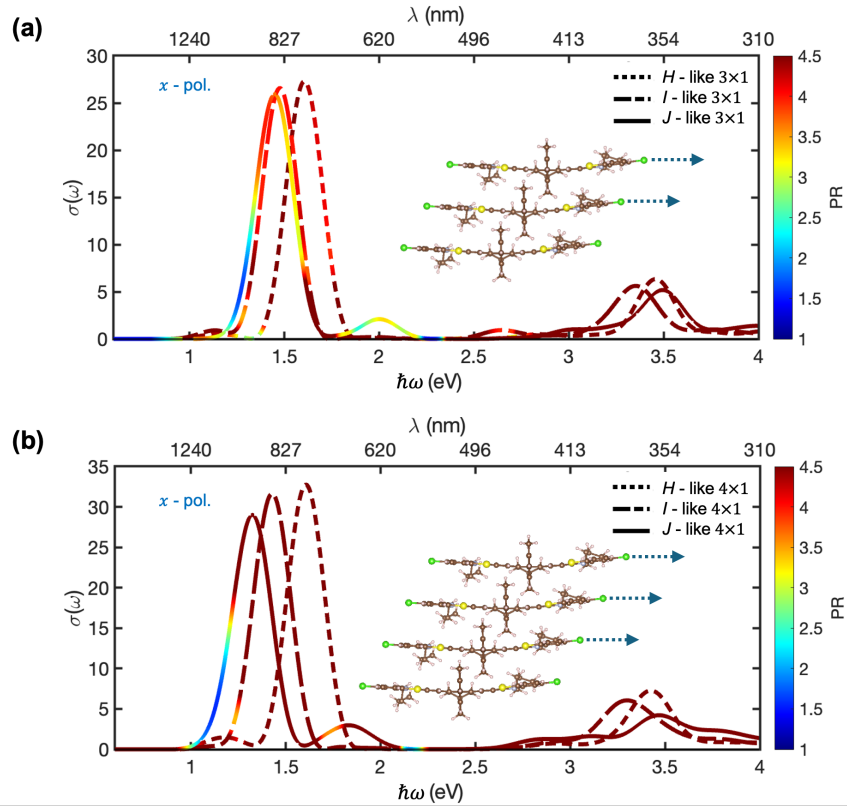


Fig. S7 sBSE absorption spectra with PRs for 1D Cy7-DiMePh aggregates under x -polarization and their individually parameterized v_W kernels. (a) 3×1 H -, I -, and J -like aggregates. (b) 4×1 H -, I -, and J -like aggregates.

Table S5 Ground- and excited-state properties of the Cy7-DiMePh monomer, 1D aggregates (2×1 , 3×1 , and 4×1), and 2D 4×3 aggregates.

Monomer	Dimer 2×1			Trimer 3×1			Tetramer 4×1			Aggregate 4×3		
	H	I	J	H	I	J	H	I	J	H	I	J
LDA gap (eV)	1.102	1.061	1.058	1.027	0.834	0.817	0.782	0.591	0.551	0.442	0.379	0.370
ngH gap (eV)	3.687	3.592	3.607	3.625	3.386	3.283	3.330	3.101	3.063	1.409	1.143	0.795
Bright state (eV)	1.426	1.584	1.521	1.421	1.606	1.477	1.451	1.606	1.429	1.447	1.360	1.173
Bright state (nm)	869	782	815	872	772	839	854	772	867	856	912	1057
PR	1.27	2.99	3.73	5.05	4.38	4.01	3.66	6.11	6.51	15.09	13.54	12.01

LDA and ngH denote the Kohn-Sham and CAM-LDA0 HOMO-LUMO gaps, respectively. Bright-state energies correspond to the most intense x -polarized sBSE excitation. PR denotes the participation ratio of the brightest exciton.

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