## Supporting Information

## Molecularly Isolated Perylene Diimides Enable Both Strong Exciton-Photon Coupling and High Photoluminescence Quantum Yield

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**Figure S1: Lifetime data.** Kinetic traces of bPDI-2 (120 mM), bPDI-3 (120 mM), and bPDI-4 (60 mM). Due to varying degrees of aggregation, multiple time constants are needed to fit the data. Current work is focused on investigating the large lifetime difference between aggregates of bPDI-4 and the other perylene diimides.



**Figure S2: PMMA control device.** (a) Angle-dependent transmission spectra of 100 mg/mL PMMA device. Difference in amplitudes are due to the spectral shape of the incident white light source. (b) Mode resonances of the different PMMA control devices. (c) Cavity thickness of different PMMA control devices, tested by both surface profilometry and via the resonances. (d) Q-factor of different PMMA control devices with fit showing a linear dependence.



**Figure S3: Absorbance length.** Rabi splitting value as a function of Absorption<sup>1/2</sup> for bPDI-3 devices at different dye loadings. Red, blue, and green squares represent 120 mM, 60 mM, and 30 mM dye in PMMA, respectively.



**Figure S4: Dye loading effect.** Angle-dependent reflection measurements of (a) bPDI-2 at 120 mM dye loading, (b) bPDI-3 at 120 mM dye loading, and (c) bPDI-4 at 60mM dye loading. (d) We use the coupled oscillator model to reveal the Rabi splitting values for the samples.

## Table S1: Comparison

| Small molecules doped in a matrix |                           |                                    |                  |      |                        |
|-----------------------------------|---------------------------|------------------------------------|------------------|------|------------------------|
| Material                          | Threshold                 | Ω reported in                      | Ω                | PLQY | PLQY reported in       |
|                                   |                           |                                    | (meV)            |      |                        |
| bPDI-3                            | -                         | This work                          | 140 <sup>1</sup> | 45%  | Chem Mater. 2017,      |
| (120 mM)                          |                           |                                    |                  |      | <b>29</b> , 8395       |
| bPDI-3                            | -                         | This work                          | 105 1            | 62%  | Chem Mater. 2017,      |
| (60 mM)                           |                           |                                    |                  |      | <b>29</b> , 8395       |
| bPDI-3                            | -                         | This work                          | 65 <sup>1</sup>  | 89%  | Chem Mater. 2017,      |
| (30 mM)                           |                           |                                    |                  |      | <b>29</b> , 8395       |
| BODIPY                            | 527 μJ cm <sup>-2</sup>   | Adv. Opt. Mat.,                    | 91 <sup>2</sup>  | 11%  | Adv. Opt. Mat., 2016,  |
| (10% dye                          |                           | 2017, <b>5</b> 1700203             |                  |      | <b>4</b> , 1615        |
| loading in                        |                           |                                    |                  |      |                        |
| polystyrene)                      |                           |                                    |                  |      |                        |
| Protein                           |                           |                                    |                  |      |                        |
| eGFP                              | 2,400 μJ cm <sup>-2</sup> | <i>Sci Adv.</i> , 2016, <b>2</b> , | 97 <sup>2</sup>  | 60%  | Biophys. J., 1997, 73, |
|                                   |                           | E1600666                           |                  |      | 2782                   |
| Crystalline material              |                           |                                    |                  |      |                        |
| Crystalline                       | 320 µJ ст <sup>-2</sup>   | Nat. Phot. 2010, 4,                | 128 <sup>2</sup> | 94%  | Proc. Phys. Soc. B,    |
| anthracene                        |                           | 371                                |                  |      | 1995, <b>68</b> , 241  |
| Oligofluorene                     |                           |                                    |                  |      |                        |
| TDAF                              | 30 µJ cm <sup>-2</sup>    | Nat. Mat., 2014,                   | 293 <sup>2</sup> | 43%  | Adv. Opt. Mat., 2013,  |
|                                   | •                         | <b>13</b> , 271                    |                  |      | 1, 827                 |
| Polymer                           |                           |                                    |                  |      |                        |
| MELPPP                            | $500 \ \mu J \ cm^{-2}$   | Nat. Mat., 2014,                   | 116 <sup>2</sup> | 30%  | Appl. Phys. Lett.,     |
|                                   | -                         | <b>13</b> , 247                    |                  |      | 1996, <b>68</b> , 1090 |

<sup>1</sup> Reported in silver-silver cavity. <sup>2</sup> Reported in DBR-DBR cavity.

Note: Due to the thickness of our active material, the Rabi splitting should not be affected severely upon changing to DBR-DBR mirror.