

## *Supporting Information*

# **Transmission Spectroscopy and Kinetics in Crystalline Solids Using Aqueous Nanocrystalline Suspensions: The Spiropyran-Merocyanine Photochromic System**

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**General Information.** All commercially obtained reagents/solvents were used as received without further purification. Unless stated otherwise, reactions were conducted in oven-dried glassware under argon atmosphere.  $^1\text{H}$  and  $^{13}\text{C}$  NMR spectra were acquired on a Bruker Avance spectrometer at 500 MHz ( $^1\text{H}$ ) and 125 MHz ( $^{13}\text{C}$ ). All chemical shifts are reported in ppm on the  $\delta$ -scale relative to the residual solvent signal as reference ( $\text{CDCl}_3$   $\delta$  7.26 and  $\delta$  77.16 for proton and carbon, respectively,  $\text{CD}_3\text{CN}$   $\delta$  1.94 for proton and  $\delta$  1.32, 118.26 for carbon. Coupling constants ( $J$ ) are reported in hertz (Hz). Standard abbreviations indicating multiplicity were used as follows: s (singlet), b (broad), d (doublet), t (triplet), q (quartet), m (multiplet), dd (doublet of doublets), and td (triplet of doublets). High-resolution mass spectrum data was recorded on a DART spectrometer in positive (ESI+) ion mode. UV-Vis absorption and transmission spectra were recorded on Ocean Optics spectrometer (DT-MINI-2-GS UV-VIS-NIR LightSource and USB2000+ using SpectraSuite software package). Dynamic Light Scattering (DLS) data were recorded using a Beckman-Coulter N4 Plus particle analyzer with a 10 mW helium-neon laser at 632.8 nm. The particle size was

determined using the 62.6° detection angle and was calculated using the size distribution processor (SDP) analysis package provided by the manufacturer. Melting point values were recorded on a Melt-Point II® apparatus. Infrared spectra were recorded on a PerkinElmer® Spectrum 100 spectrometer equipped with a universal ATR sampling accessory. Powder X-ray diffraction measurements were performed on a flat stage using a PANalytical, Inc X'Pert Pro Cu K- $\alpha$  = 1.5406 Å radiation at power settings of 45 kV and 40 mA and a slit width of 0.5°. Data were collected at room temperature in the range of  $2\theta$  = 4–45°. All samples were irradiated using a Rayonet photochemical reactor with four 12 inch 8W (BLE-8T365) 365 nm bulbs. Fluorescence spectra were acquired using an Edinburgh Instruments FLSP920 spectrometer equipped with a 450 W xenon arc lamp (Xe900) as the light source, a single photon counting PMT as the detector, and the F900 software program.

#### **General preparation of solutions for UV-Vis absorption experiments (SP1–SP5)**

15  $\mu$ L of a stock solution (~10 mM) in acetonitrile was added to a quartz cuvette containing 2.5 mL of pure acetonitrile to give a final sample concentration of  $\sim 6 \times 10^{-5}$  M.

#### **General preparation of nanocrystalline suspensions for UV-Vis absorption experiments (SP1–SP5)**

10  $\mu$ L of a stock solution (~15 mM) in acetonitrile was injected via a micropipette into a culture tube containing 3 mL of vortexing Millipore water. Vortexing continued for 30 seconds after injection. Then the newly created suspension was transferred via pipette to a quartz cuvette.

#### **General method for UV-Vis absorption experiments in solution and nanocrystalline suspensions (SP1–SP5)**

An initial absorption spectrum was recorded, and then the sample was irradiated at 365 nm in a Rayonet photochemical reactor (4 bulbs) in 1 minute intervals until the photostationary state (PSS) was reached (as determined by monitoring the band growing in at ca. 550 nm). Absorption spectra were recorded after every irradiation interval. Once the PSS was reached, the sample was left to thermally decay under ambient light conditions, and the absorbance spectrum was recorded in regular time intervals until the absorbance of the visible band at ca. 550 nm remained constant.

#### **General preparation of solutions for fluorescence experiments (SP1)**

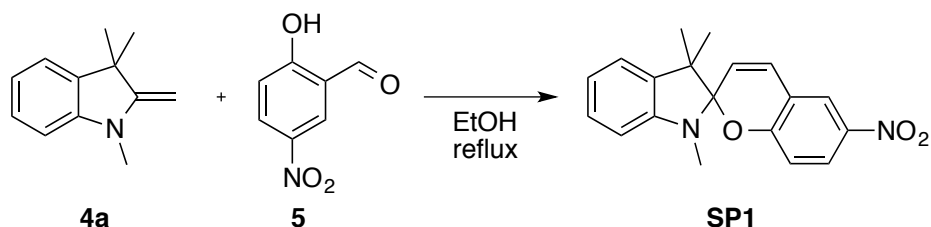
10  $\mu$ L of a stock solution (6.51 mM) in acetonitrile was added to a quartz cuvette containing 2.5 mL of pure acetonitrile to give a final sample concentration of  $2.6 \times 10^{-5}$  M.

#### **General preparation of nanocrystalline suspensions for fluorescence experiments (SP1)**

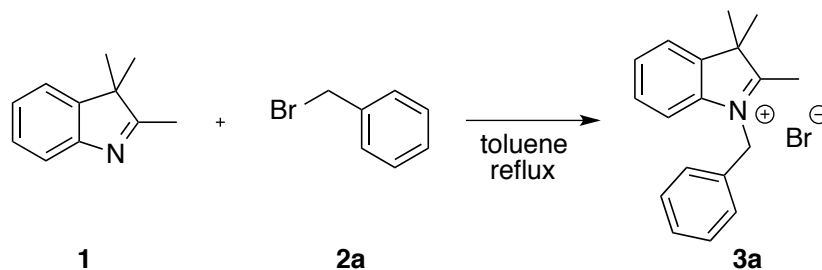
15  $\mu$ L of a stock solution (6.51 mM) in acetonitrile was injected via a micropipette into a culture tube containing 3 mL of vortexing Millipore water. Vortexing continued for 30 seconds after injection. Then the newly created suspension was transferred via pipette to a quartz cuvette.

#### **General method for fluorescence experiments in solution and nanocrystalline suspensions (SP1)**

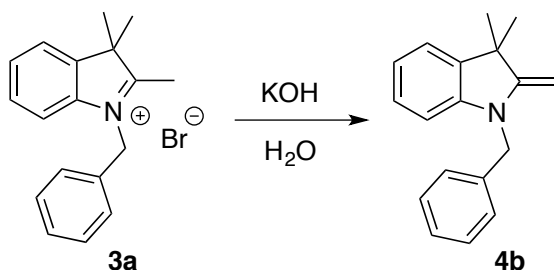
An initial emission spectrum was recorded with excitation at 530 nm and scanning from 540–720 nm, and then the sample was irradiated at 365 nm in a Rayonet photochemical reactor (4 bulbs) in 2 minute intervals for a total of 4–8 minutes irradiation, followed by a final irradiation interval of 5 minutes. Emission spectra were recorded after every irradiation interval. The samples were then allowed to thermally decay inside the fluorometer, and emission spectra were recorded in regular time intervals.



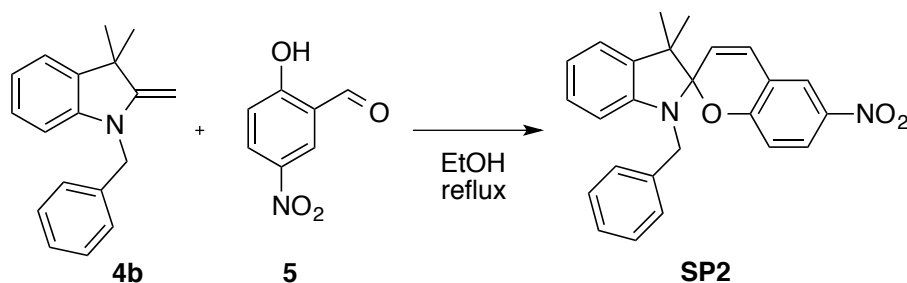
**1',3',3'-Trimethyl-6-nitrospiro[chromene-2,2'-indoline] (SP1):** SP1 was prepared with minor modifications to the experimental procedure in reference 1. 1,3,3-Trimethyl-2-methyleneindoline (**4a**) (0.275 mL, 1.55 mmol) was added dropwise to a solution containing 2-hydroxy-5-nitrobenzaldehyde (**5**) (0.2593 g, 1.55 mmol) in 8.5 mL of freshly distilled ethanol followed by refluxing for 16 h under argon. Once cooled to room temperature, the mixture was concentrated and filtered using ice cold ethanol to afford **SP1** as a yellow solid. Recrystallized from DCM/EtOH (1:1) to obtain yellow crystals (0.377 g, 75% yield). m.p. 181.7–182.7 °C (lit. 178–179 °C). IR (film): 3067, 2963, 2865, 1609, 1509, 1488, 1476, 1466, 1329, 1299, 1268, 1183, 1122, 1088, 1021, 1015, 948, 916, 806, 782, 750 cm<sup>-1</sup>. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz): δ 8.02 (dd, *J* = 8.71, 2.72 Hz, 1H), 8.00 (s, 1H), 7.21 (td, *J* = 7.62, 1.26 Hz, 1H), 7.09 (dd, *J* = 7.30, 0.81 Hz, 1H), 6.92 (d, *J* = 9.65 Hz, 1H), 6.89 (td, *J* = 7.39, 0.90 Hz, 1H), 6.77 (d, *J* = 9.38 Hz, 1H), 6.56 (d, *J* = 7.75 Hz, 1H), 5.86 (d, *J* = 10.37 Hz, 1H), 2.74 (s, 3H), 1.30 (s, 3H), 1.19 (s, 3H) ppm. <sup>13</sup>C NMR (CDCl<sub>3</sub>, 125 MHz): δ 160.0, 147.8, 141.1, 136.2, 128.4, 128.0, 126.0, 122.8, 121.8, 121.7, 119.9, 118.8, 115.6, 107.2, 106.5, 52.4, 29.0, 26.0, 20.1 ppm. HRMS (ESI-TOF) *m/z*: [M]<sup>++</sup> calcd. for C<sub>19</sub>H<sub>18</sub>N<sub>2</sub>O<sub>3</sub>: 322.13119, found: 322.13180.



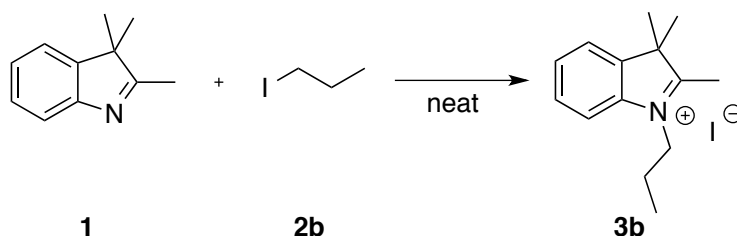
**1-Benzyl-2,3,3-trimethylindolenium bromide (3a):** Compound **3a** was prepared with minor modifications to the experimental procedure in reference 2. 2,3,3-Trimethylindolenine (**1**) (1.0 mL, 6.23 mmol) was added dropwise to a solution containing benzyl bromide (**2a**) (1.50 mL, 12.6 mmol) in 6 mL of dry toluene followed by refluxing for 22 h under argon. Let cool to room temperature, removed solvent under reduced pressure, and washed 3 x 5 mL with hexanes. Dried red solid **3a** under vacuum and stored in a desiccator (2.013 g, 97% yield).



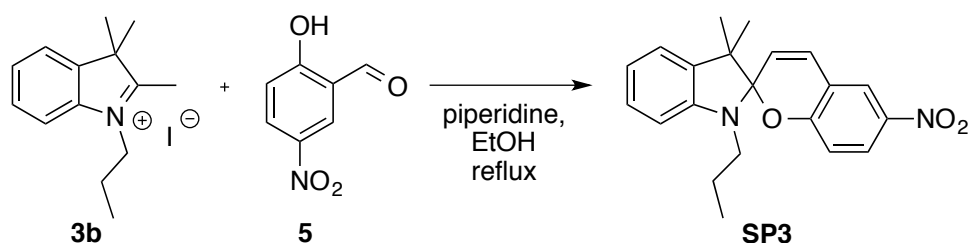
**1-Benzyl-3,3-dimethyl-2-methyleneindoline (4b):** Compound **4b** was prepared with minor modifications to the experimental procedure in reference 5. To a solution of 1-benzyl-2,3,3-trimethylindolenium bromide (**3a**) (2.0135 g, 6.1 mmol) dissolved in 23 mL of DI H<sub>2</sub>O was added a solution of KOH (1.061 g, 18.9 mmol) dissolved in 23 mL of DI H<sub>2</sub>O under argon. After 1 h at room temperature, extracted 3 x 40 mL with DCM, washed the combined organic layers 1 x 125 mL brine, dried over MgSO<sub>4</sub>, filtered and removed solvent under reduced pressure to obtain **4b** as a red-orange oil. Crude product used in next step without further purification.



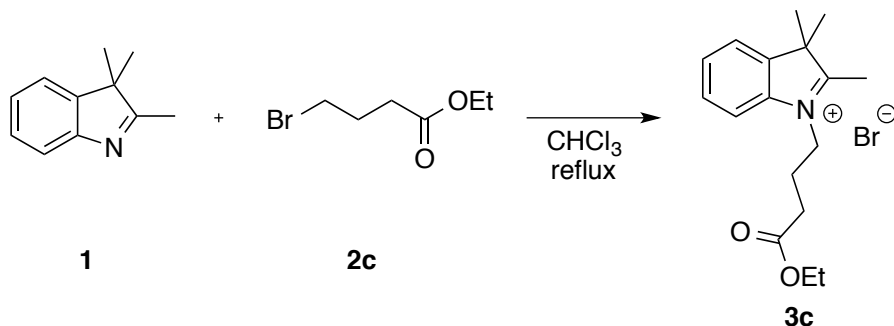
**1'-Benzyl-3',3'-dimethyl-6-nitrospiro[chromene-2,2'-indoline] (SP2):** **SP2** was prepared with minor modifications to the experimental procedure in reference 5. To a solution containing 2-hydroxy-5-nitrobenzaldehyde (**5**) (1.018 g, 6.1 mmol) in 20 mL of freshly distilled ethanol was added crude 1-benzyl-3,3-dimethyl-2-methylenindoline (**4b**) dissolved in 15 mL of ethanol followed by refluxing for 18 h under argon. After cooling to room temperature, removed solvent under reduced pressure and purified by flash chromatography on silica with hexanes/DCM as eluent (2:1→1:1) to afford **SP2** as a yellow solid. Recrystallized from ethanol to obtain yellow crystals (1.0231 g, 42% yield over 2 steps). m.p. 161.5–162.8 °C (lit. 162 °C)<sup>6</sup>. IR (film): 3030, 2966, 1611, 1518, 1481, 1458, 1336, 1273, 1090, 949, 810, 745 cm<sup>-1</sup>. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz): δ 8.03 (dd, *J* = 8.99, 2.78 Hz, 1H), 7.98 (d, *J* = 2.68 Hz, 1H), 7.33–7.23 (m, 5H), 7.12 (dd, *J* = 7.24, 0.82 Hz, 1H), 7.08 (td, *J* = 7.70, 1.30 Hz, 1H), 6.88 (dd, *J* = 14.76, 0.91 Hz, 1H), 6.87 (d, *J* = 10.19 Hz, 1H), 6.78 (d, *J* = 8.98 Hz, 1H), 6.35 (d, *J* = 7.72 Hz, 1H), 5.92 (d, *J* = 10.39 Hz, 1H), 4.51 (d, *J* = 16.54 Hz, 1H), 4.21 (d, *J* = 16.54 Hz, 1H), 1.34 (s, 3H), 1.31 (s, 3H) ppm. <sup>13</sup>C NMR (CDCl<sub>3</sub>, 125 MHz): δ 159.7, 147.2, 141.2, 138.7, 136.2, 128.8, 128.6, 127.9, 127.2, 126.7, 126.1, 122.9, 121.9, 121.8, 120.1, 118.7, 115.7, 108.1, 106.9, 52.9, 47.8, 26.3, 20.1 ppm. HRMS (ESI-TOF) *m/z*: [M]<sup>•+</sup> calcd. for C<sub>25</sub>H<sub>22</sub>N<sub>2</sub>O<sub>3</sub>: 398.16249, found 398.16092.



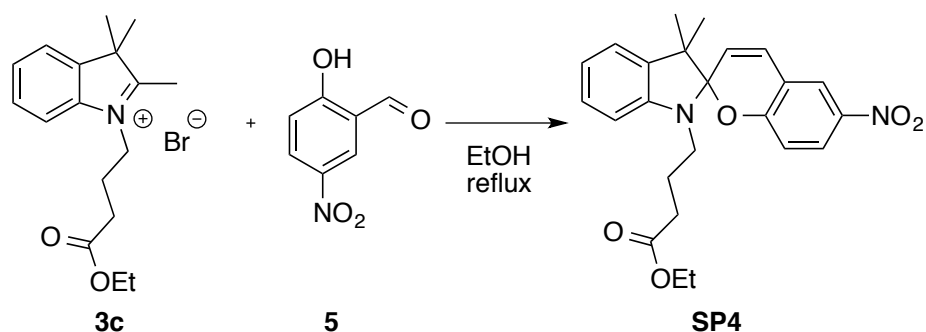
**2,3,3-Trimethyl-1-propylindolenium iodide (3b):** Compound **3b** was prepared with minor modifications to the experimental procedure in reference 3. 2,3,3-Trimethylindolenine (**1**) (0.24 mL, 1.5 mmol) was dissolved in 1-iodopropane (**2b**) (0.40 mL, 4.10 mmol) and heated to 95 °C for 7 h under argon. The resulting viscous oil (**3b**) was cooled to room temperature, rinsed with hexanes, and stored in a desiccator. Crude product used in next step without further purification.



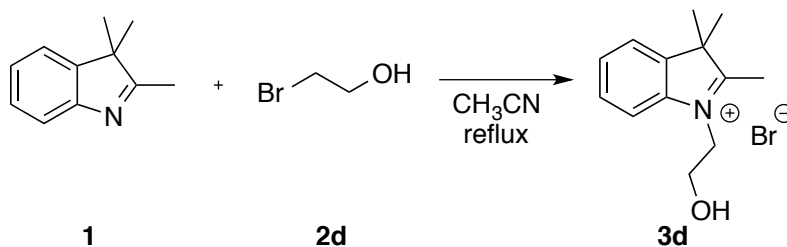
**3,3'-Dimethyl-6-nitro-1'-propylspiro[chromene-2,2'-indoline] (SP3):** SP3 was prepared with minor modifications to the experimental procedure in reference 3. To a solution containing crude 2,3,3-trimethyl-1-propylindolenium iodide (**3b**) in 5 mL of freshly distilled ethanol was added dropwise piperidine (0.18 mL, 1.82 mmol) under argon. After 25 min, a solution of 2-hydroxy-5-nitrobenzaldehyde (**5**) (0.2542 g, 1.52 mmol) in 7 mL of ethanol was added followed by refluxing for 18 h. The mixture was cooled to room temperature and purified by flash chromatography on silica with hexanes/DCM as eluent (1.5:1) to afford **SP3** as a yellow solid. Recrystallized from DCM/ethanol (1:1) to obtain yellow crystals (0.4321 g, 82% yield over 2 steps). m.p. 150.2–151.2 °C. IR (film): 3057, 2964, 1610, 1514, 1481, 1335, 1275, 1089, 957, 807, 748 cm<sup>-1</sup>. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz): δ 8.01 (dd, *J* = 10.3, 2.71 Hz, 1H), 7.99 (s, 1H), 7.18 (td, *J* = 7.65, 1.25 Hz, 1H), 7.08 (dd, *J* = 7.25, 0.94 Hz, 1H), 6.89 (d, *J* = 10.34 Hz, 1H), 6.86 (td, *J* = 7.43, 0.88 Hz, 1H), 6.74 (d, *J* = 8.33 Hz, 1H), 6.57 (d, *J* = 7.70 Hz, 1H), 5.87 (d, *J* = 10.41 Hz, 1H), 3.15–3.09 (m, 2H), 1.71–1.55 (m, 2H), 1.28 (s, 3H), 1.19 (s, 3H), 0.90 (t, *J* = 7.39, 3H) ppm. <sup>13</sup>C NMR (CDCl<sub>3</sub>, 125 MHz): δ 159.9, 147.3, 141.0, 136.0, 128.1, 127.8, 126.0, 122.8, 122.2, 121.8, 119.4, 118.6, 115.6, 106.9, 106.8, 52.8, 45.6, 26.1, 22.3, 20.0, 11.9 ppm. HRMS (ESI-TOF) *m/z*: [M]<sup>+</sup> calcd. for C<sub>21</sub>H<sub>22</sub>N<sub>2</sub>O<sub>3</sub>: 350.16249, found: 350.16220.



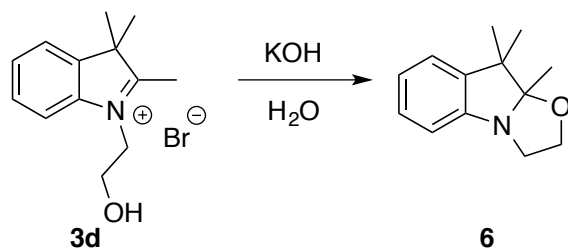
**1-(4-ethoxy-4-oxobutyl)-2,3,3-trimethylindolium bromide (3c):** Compound **3c** was prepared with minor modifications to the experimental procedure in reference 4. To a solution containing 2,3,3-trimethylindolenine (**1**) (0.32 mL, 1.99 mmol) in 5 mL of CHCl<sub>3</sub> was added ethyl-4-bromobutyrate (**2c**) (0.93 mL, 6.5 mmol) dropwise under argon followed by refluxing for 3 days. The crude mixture was cooled to room temperature and the solvent was removed under reduced pressure. The crude mixture was dissolved in methanol (2 mL) and then diethyl ether (10 mL) was added to precipitate out the product. Subsequent filtration afforded **3c** (0.360 g, 51% yield), which was used in the next step without further purification.



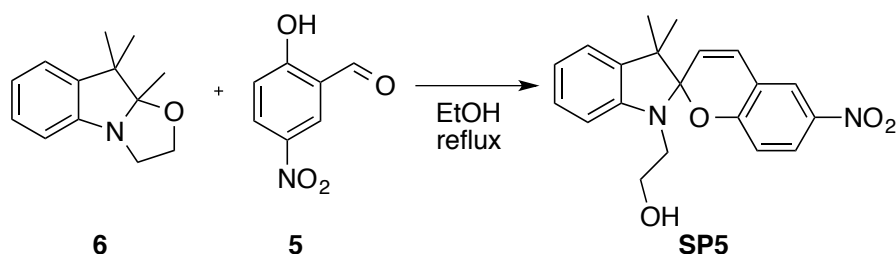
**Ethyl 4-(3',3'-dimethyl-6-nitrospiro[chromene-2,2'-indolin]-1'-yl)butanoate (SP4):** SP4 was prepared with minor modifications to the experimental procedure in reference 4. To a solution of **3c** (0.3412 g, 0.963 mmol) in 12 mL of freshly distilled ethanol was added 2-hydroxy-5-nitrobenzaldehyde (**5**) (0.1933 g, 1.16 mmol) under argon followed by refluxing for 18 h. After cooling to room temperature, removed solvent under reduced pressure. Dissolved the crude mixture in DCM, washed 3 x 10 mL with saturated Na<sub>2</sub>CO<sub>3</sub>, dried over Na<sub>2</sub>SO<sub>4</sub>, and concentrated. Purified by flash chromatography on silica with DCM/hexanes as eluent (2:1) to afford **SP4** as a golden yellow solid. Recrystallized from ethanol to obtain yellow needles (0.118 g, 29% yield). m.p. 122.0–123.2 °C (lit. 116 °C). IR (film): 3054, 2966, 1729, 1609, 1518, 1480, 1335, 1273, 1089, 954, 808, 747 cm<sup>-1</sup>. <sup>1</sup>H NMR (CDCl<sub>3</sub>, 500 MHz): δ 8.01 (dd, *J* = 11.46, 2.72 Hz, 1H), 7.99 (s, 1H), 7.19 (td, *J* = 7.66, 1.24 Hz, 1H), 7.08 (dd, *J* = 7.22, 1.03 Hz, 1H), 6.89 (d, *J* = 9.99 Hz, 1H), 6.88 (td, *J* = 7.81, 0.77 Hz, 1H), 6.74 (d, *J* = 8.58 Hz, 1H), 6.64 (d, *J* = 7.78 Hz, 1H), 5.87 (d, *J* = 10.36 Hz, 1H), 4.09 (qd, *J* = 7.13, 1.41 Hz, 2H), 3.24–3.17 (m, 2H), 2.34 (td, *J* = 7.20, 1.69 Hz, 2H), 2.01–1.80 (m, 2H), 1.28 (s, 3H), 1.23 (t, *J* = 7.15 Hz, 3H), 1.18 (s, 3H) ppm. <sup>13</sup>C NMR (CDCl<sub>3</sub>, 125 MHz): δ 173.2, 159.7, 147.2, 141.1, 136.1, 128.4, 128.0, 126.1, 122.9, 122.0, 121.9, 119.8, 118.6, 115.7, 107.0, 106.8, 60.6, 52.8, 43.2, 31.9, 26.1, 24.3, 20.0, 14.3 ppm. HRMS (ESI-TOF) *m/z*: [M]<sup>+</sup> calcd. for C<sub>24</sub>H<sub>26</sub>N<sub>2</sub>O<sub>5</sub>: 422.18362, found: 422.18276.



**1-(2-Hydroxyethyl)-2,3,3-trimethylindolium bromide (3d):** Compound **3d** was prepared with minor modifications to the experimental procedure in reference 5. To a solution of 2,3,3-trimethylindolenine (**1**) (1.0 mL, 6.23 mmol) in 8 mL of acetonitrile was added 2-bromoethanol (**2d**) (0.55 mL, 7.78 mmol) followed by heating to reflux for 24 h. After cooling to room temperature, concentrated the reaction mixture and then sonicated and filtered using hexanes to obtain **3d** as a purple solid, which was used in the next step without further purification.

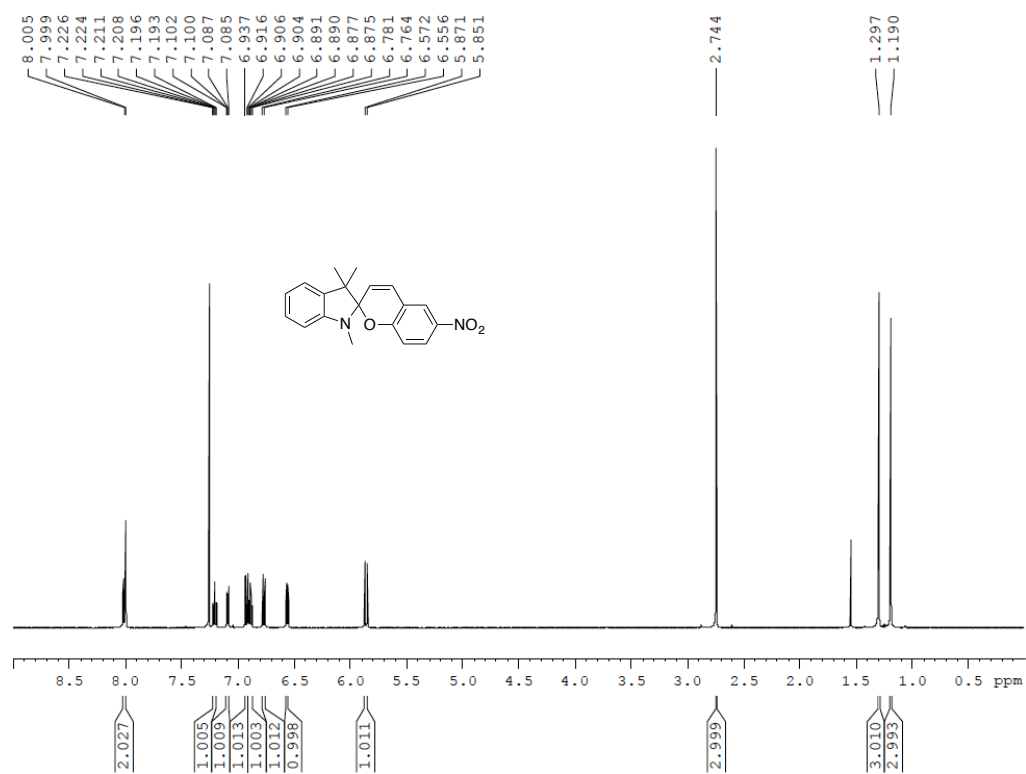


**9,9,9a-Trimethyl-2,3,9,9a-tetrahydrooxazolo[2,3-a]indole (6):** Compound **6** was prepared with minor modifications to the experimental procedure in reference 5. To a solution of **3d** in 10 mL of DI H<sub>2</sub>O was added a solution of KOH (0.46 g, 8.2 mmol) dissolved in 15 mL of DI H<sub>2</sub>O. Let stir for 10 min., then extracted the reaction mixture 3 x 40 mL of diethyl ether, dried over Na<sub>2</sub>SO<sub>4</sub>, filtered and concentrated under reduced pressure. Purified by flash chromatography on silica with hexanes/ethyl acetate as eluent (3:0.5) to obtain **6** (0.636 g, 50% yield over 2 steps) as a yellow-orange oil.

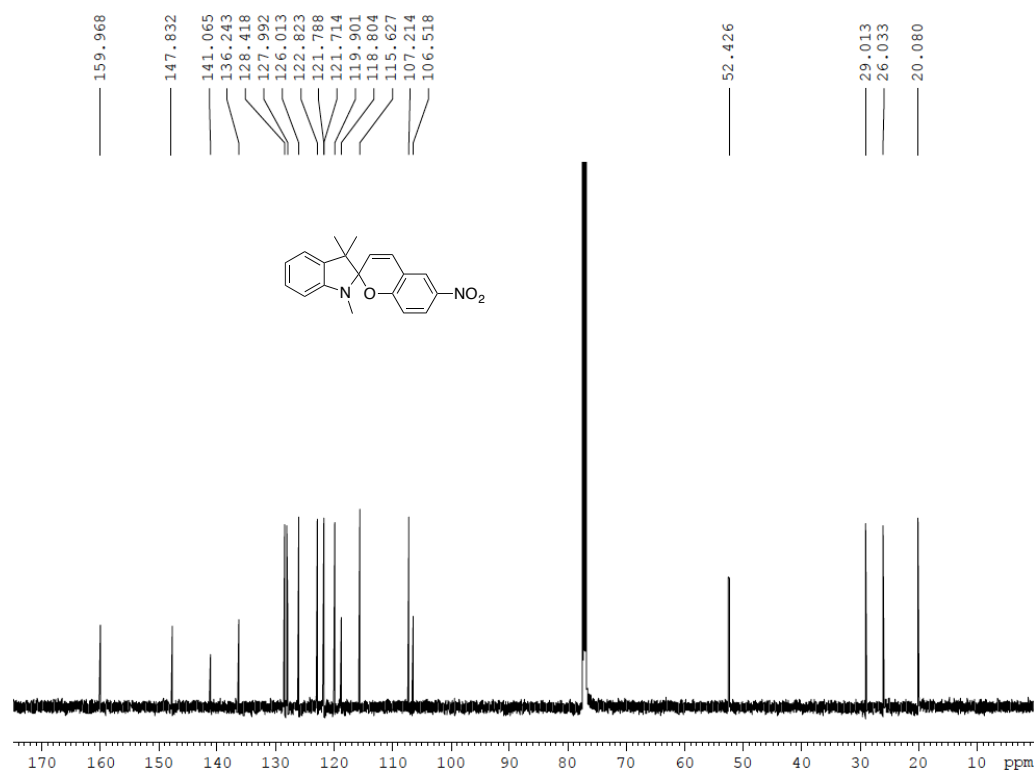


**2-(3',3'-Dimethyl-6-nitrospiro[chromene-2,2'-indolin]-1'-yl)ethanol (SP5):** SP5 was prepared with minor modifications to the experimental procedure in reference 5. To a solution of **6** (0.636 g, 3.13 mmol) in 10 mL of freshly distilled ethanol was added 2-hydroxy-5-nitrobenzaldehyde (0.654 g, 3.91 mmol) followed by refluxing for 6 h. After cooling to room temperature, removed solvent under reduced pressure. Recrystallized from DCM/ethanol (1:1) to obtain dark purple plates (0.90 g, 81% yield). m.p. 155.5–157.0 °C (lit. 171 °C). IR (film): 3411 (broad), 3069, 2964, 2924, 1610, 1517, 1481, 1459, 1336, 1273, 1089, 1026, 953, 807, 746 cm<sup>-1</sup>. <sup>1</sup>H NMR (CD<sub>3</sub>CN, 500 MHz): δ 8.07 (d, *J* = 2.75 Hz, 1H), 7.99 (dd, *J* = 9.00, 2.80 Hz, 1H), 7.14 (td, *J* = 7.69, 1.27 Hz, 1H), 7.11 (dd, *J* = 7.30, 0.85 Hz, 1H), 7.02 (d, *J* = 10.40 Hz, 1H), 6.83 (td, *J* = 7.41, 0.85 Hz, 1H), 6.71 (dd, *J* = 8.98, 0.23 Hz, 1H), 6.66 (d, *J* = 7.75 Hz, 1H), 5.99 (d, *J* = 10.40 Hz, 1H), 3.68–3.63 (m, 1H), 3.58–3.53 (m, 1H), 3.32 (dt, *J* = 14.75, 6.84 Hz, 1H), 3.24–3.19 (m, 1H), 1.25 (s, 3H), 1.15 (s, 3H) ppm. <sup>13</sup>C NMR (CD<sub>3</sub>CN, 125 MHz): δ 160.4, 148.3, 142.0, 136.9, 128.7, 128.6, 126.5, 123.6, 123.2, 122.7, 120.3, 120.0, 116.2, 108.0, 107.7, 60.8, 53.4, 46.8, 26.1, 20.0 ppm. HRMS (ESI-TOF) *m/z*: [M]<sup>•+</sup> calcd. for C<sub>20</sub>H<sub>20</sub>N<sub>2</sub>O<sub>4</sub>: 352.14176, found 352.14017.

**Figure S1.**  $^1\text{H}$  NMR of **SP1** in  $\text{CDCl}_3$  at 500 MHz

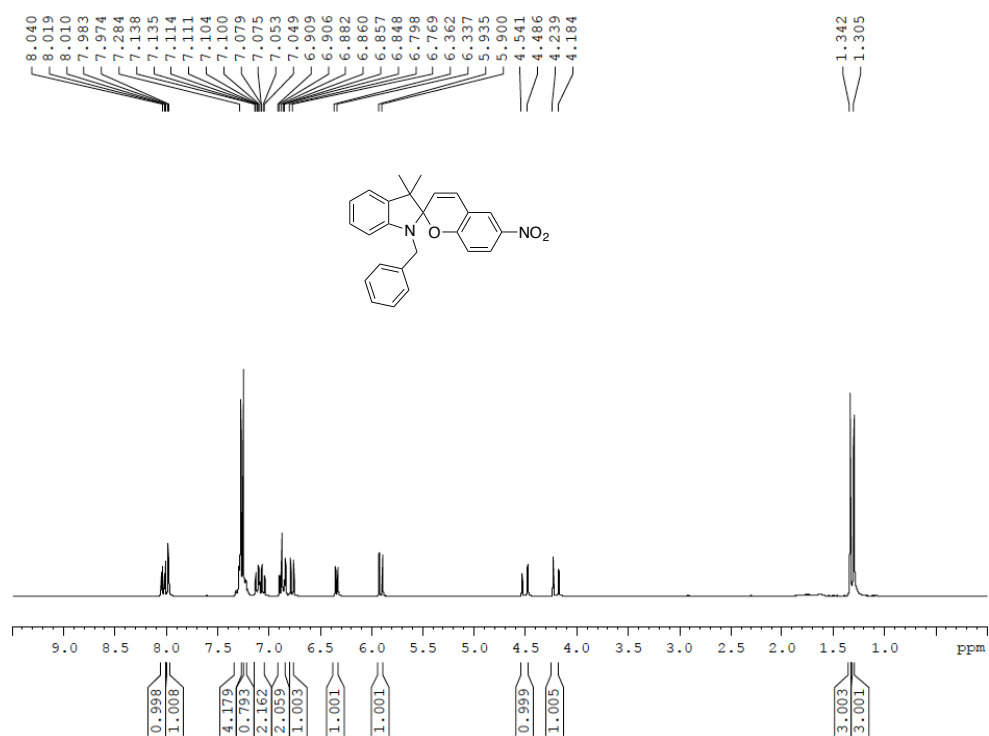


**Figure S2.**  $^{13}\text{C}$  NMR of **SP1** in  $\text{CDCl}_3$  at 125 MHz

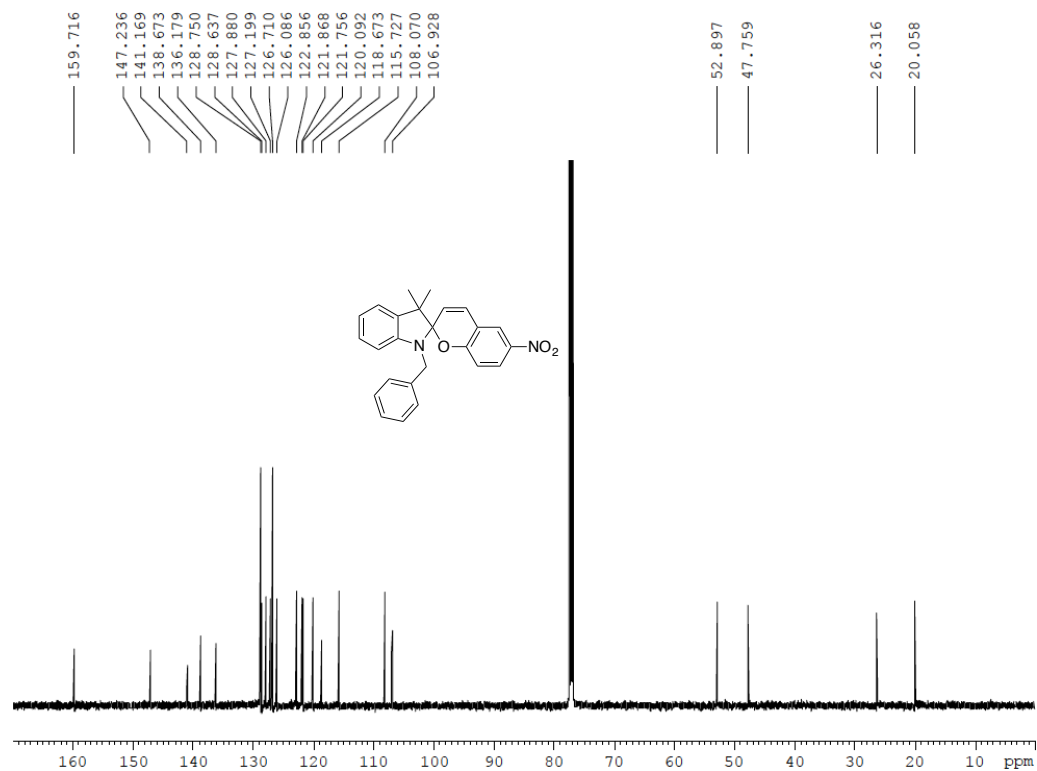




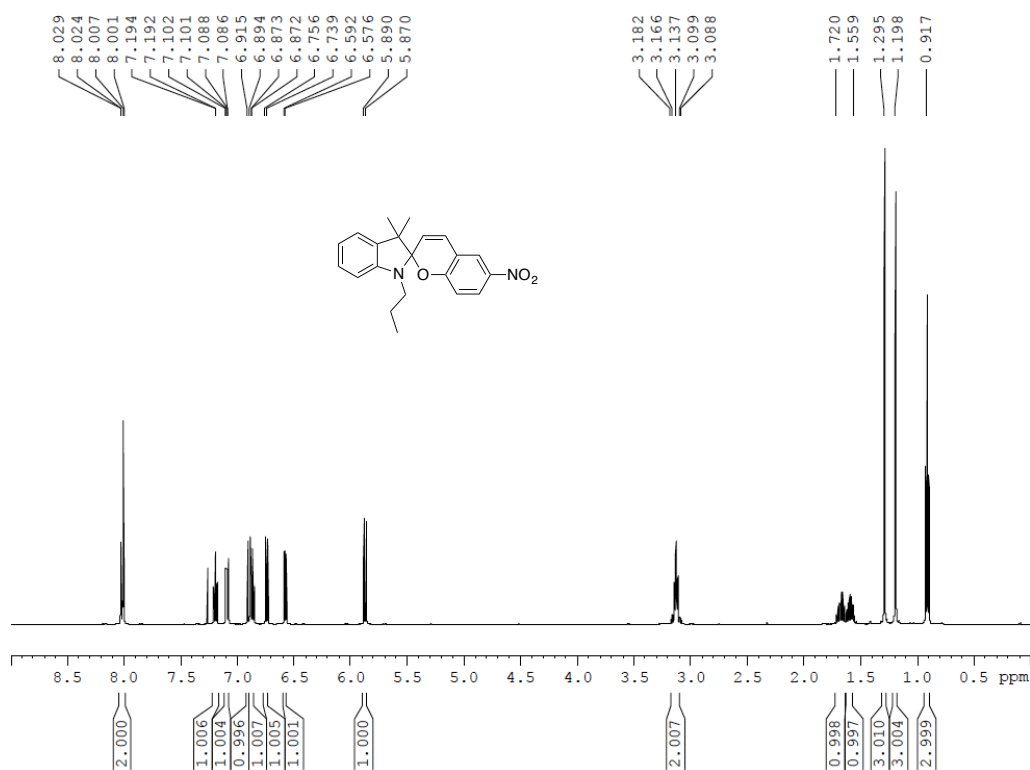
**Figure S3.**  $^1\text{H}$  NMR of **SP2** in  $\text{CDCl}_3$  at 500 MHz



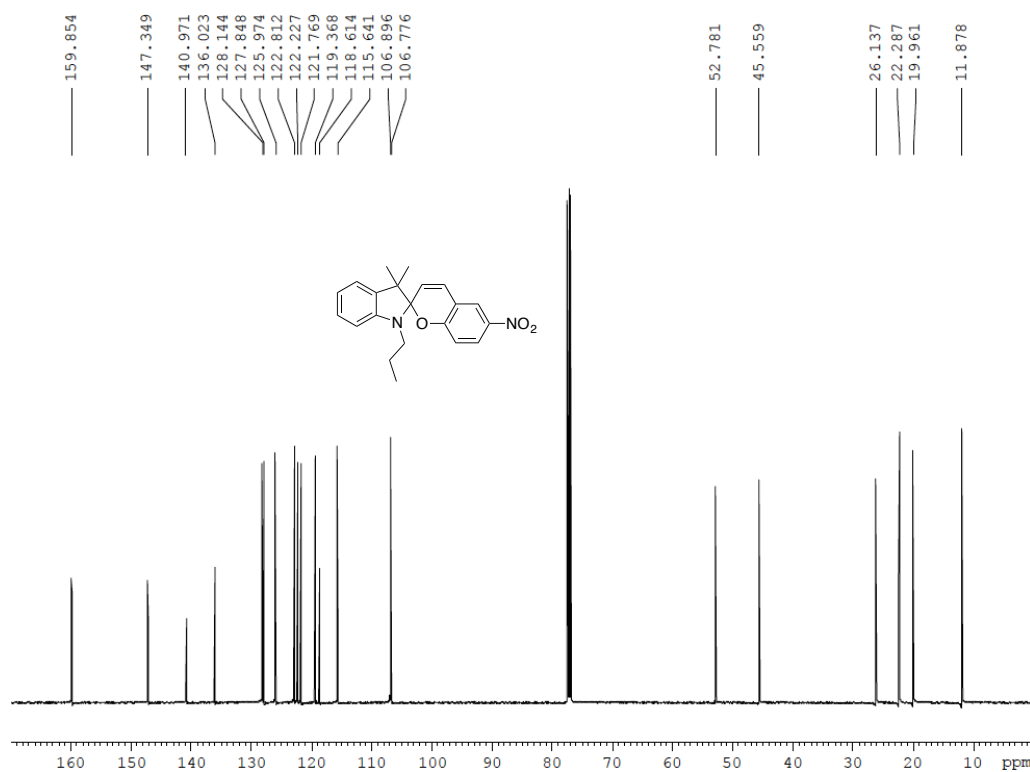
**Figure S4.**  $^{13}\text{C}$  NMR of **SP2** in  $\text{CDCl}_3$  at 125 MHz



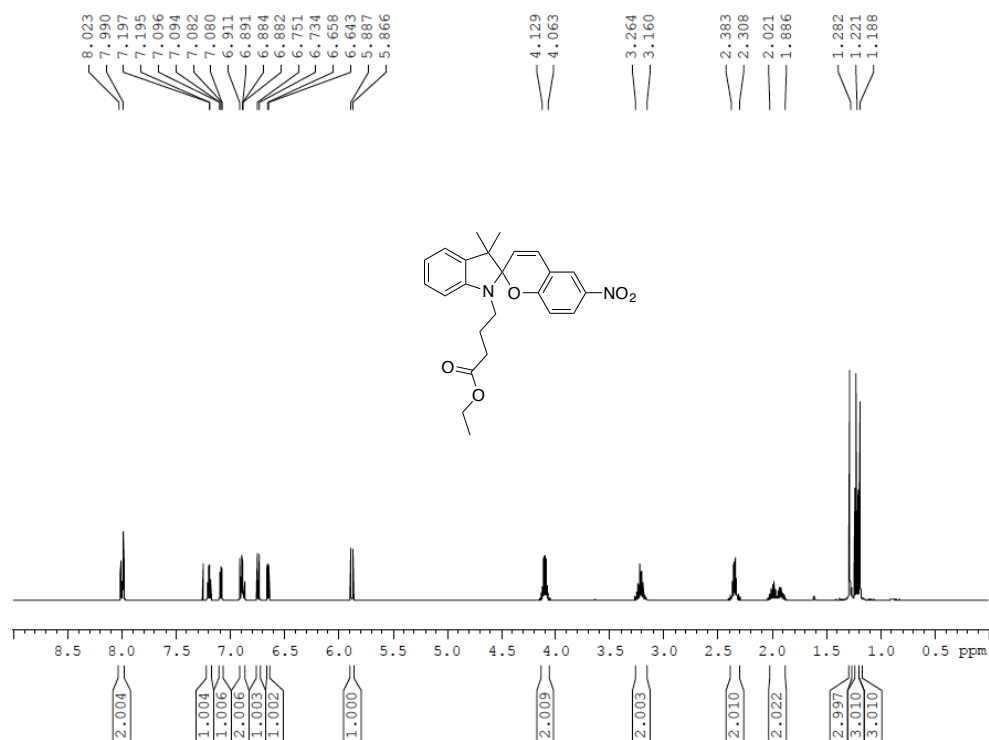
**Figure S5.**  $^1\text{H}$  NMR of **SP3** in  $\text{CDCl}_3$  at 500 MHz



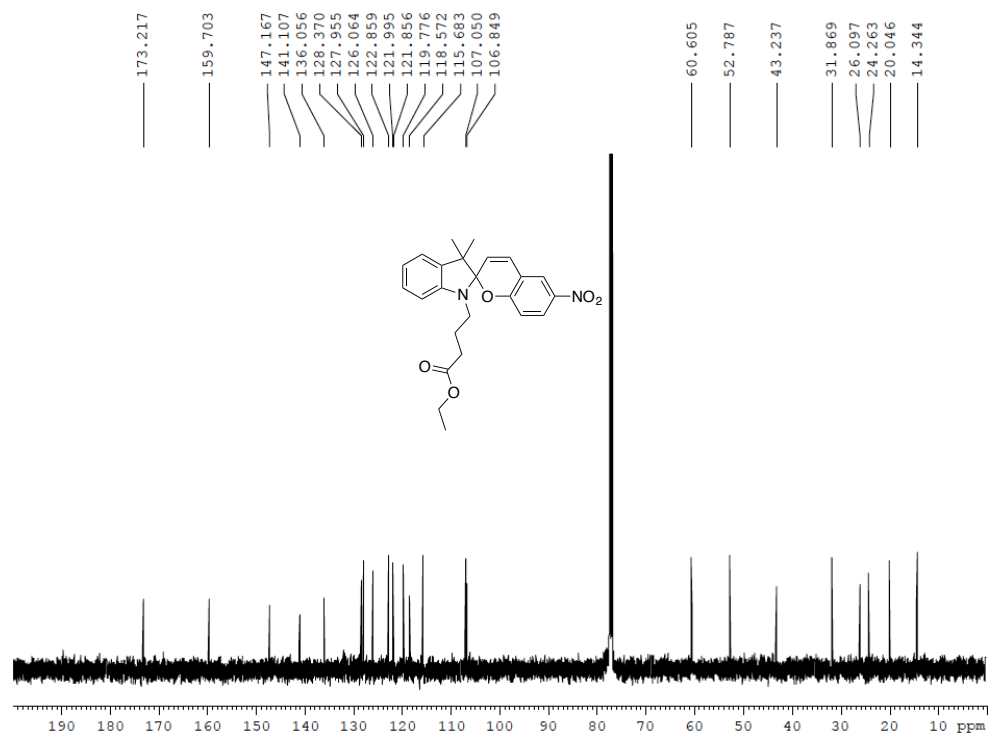
**Figure S6.**  $^{13}\text{C}$  NMR of **SP3** in  $\text{CDCl}_3$  at 125 MHz



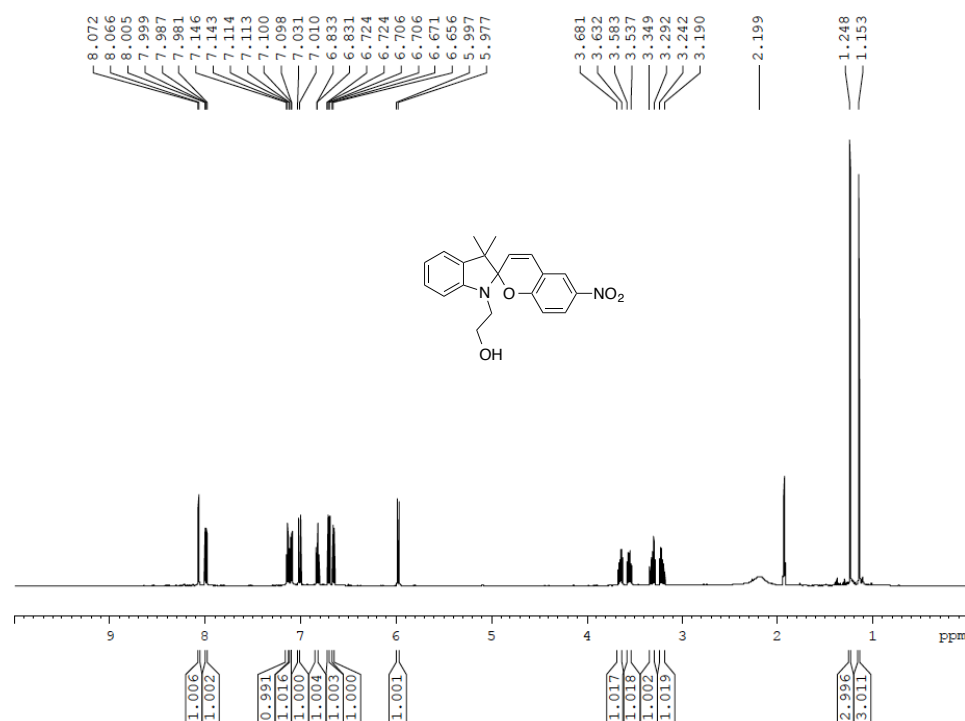
**Figure S7.**  $^1\text{H}$  NMR of **SP4** in  $\text{CDCl}_3$  at 500 MHz



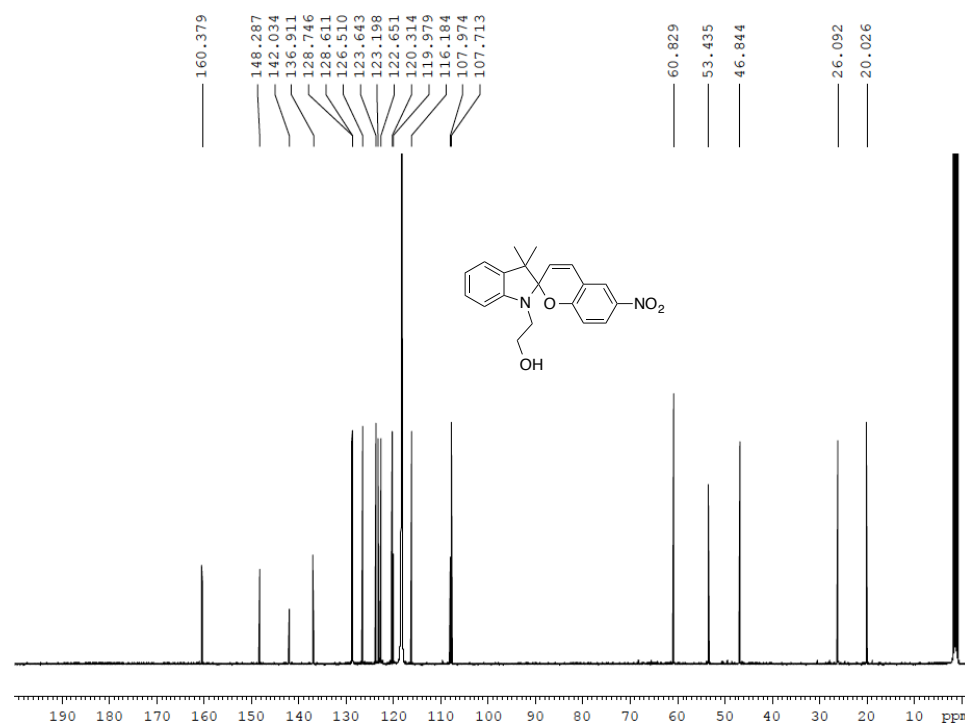
**Figure S8.**  $^{13}\text{C}$  NMR of **SP4** in  $\text{CDCl}_3$  at 125 MHz

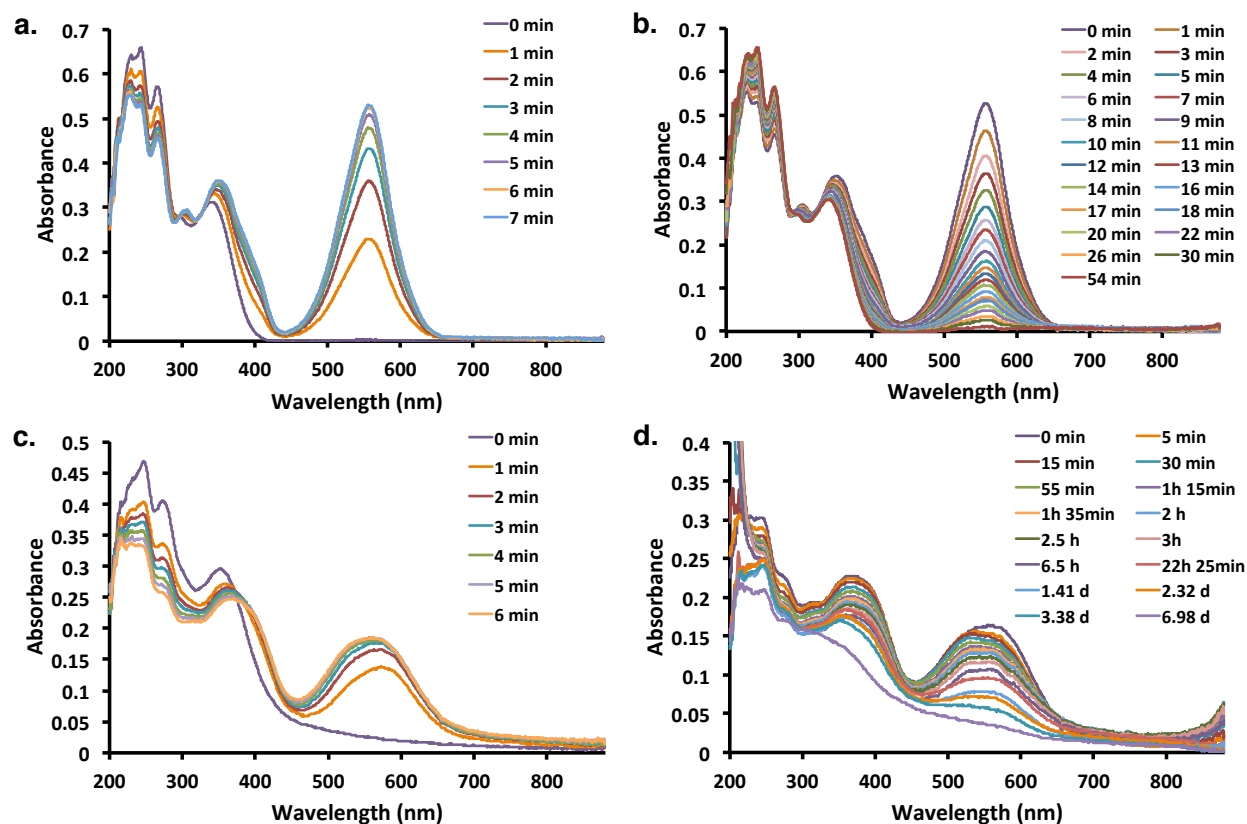


**Figure S9.**  $^1\text{H}$  NMR of **SP5** in  $\text{CD}_3\text{CN}$  at 500 MHz

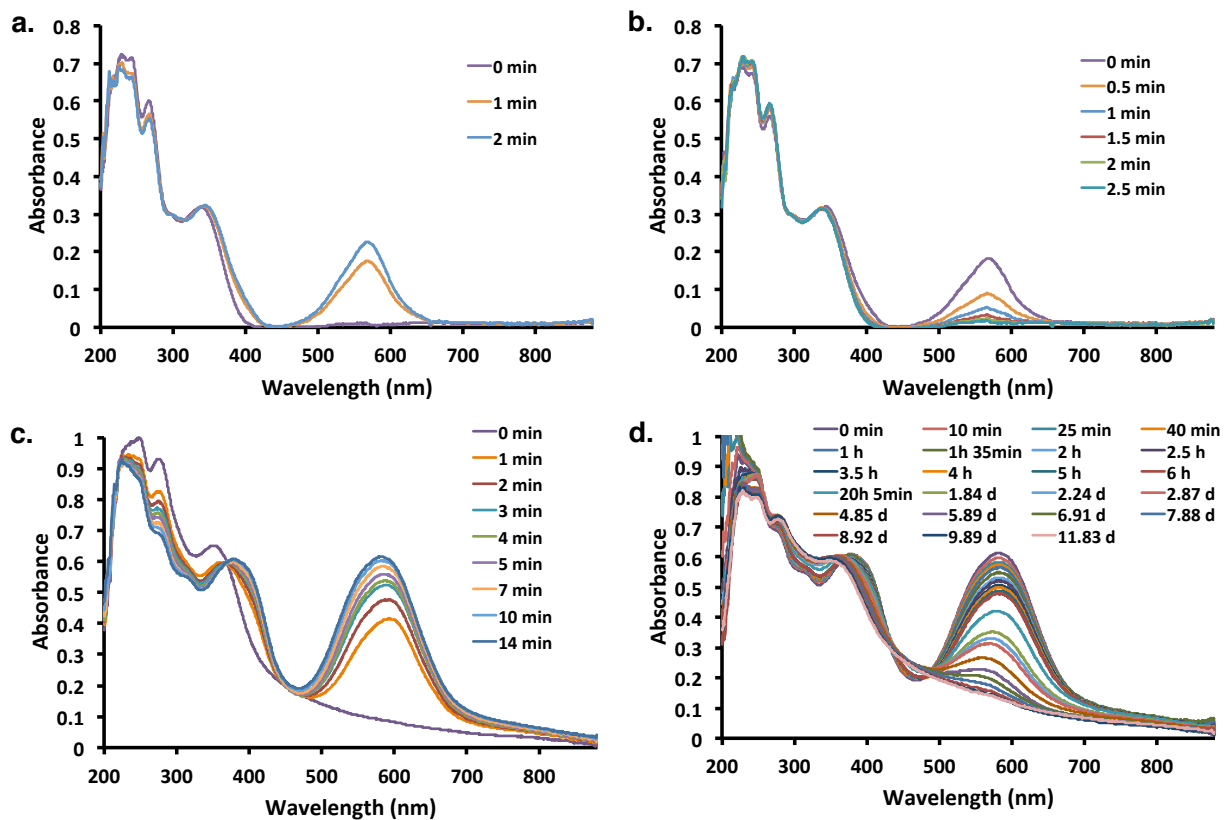


**Figure S10.**  $^{13}\text{C}$  NMR of **SP5** in  $\text{CD}_3\text{CN}$  at 125 MHz

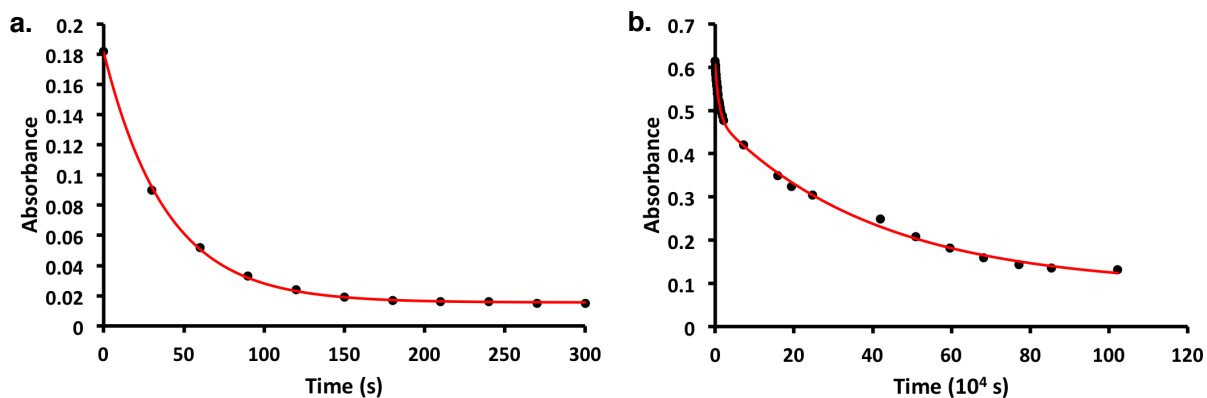




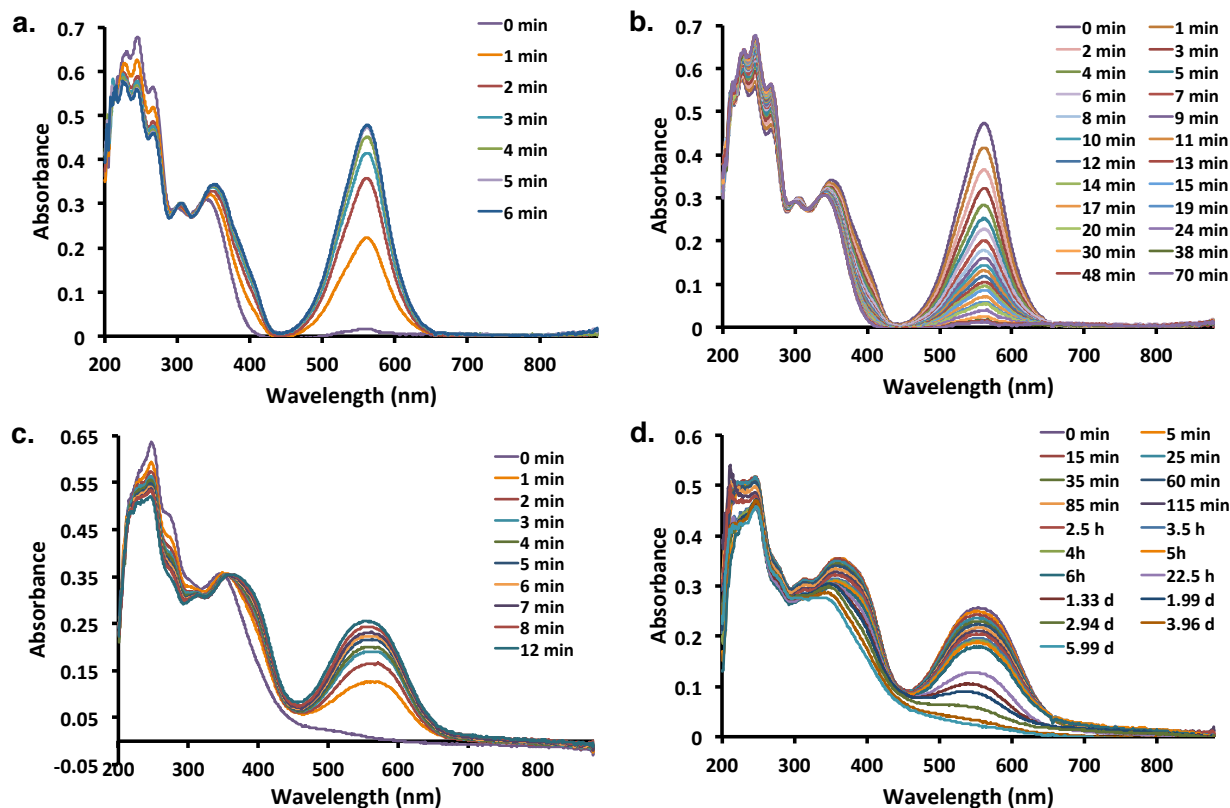
**Figure S11.** UV-Vis absorption spectra for **SP1** a) irradiation at 365 nm in MeCN, b) thermal decay at 557 nm in MeCN, c) irradiation at 365 nm in nanocrystalline suspension, and d) thermal decay at 557 nm in nanocrystalline suspension.



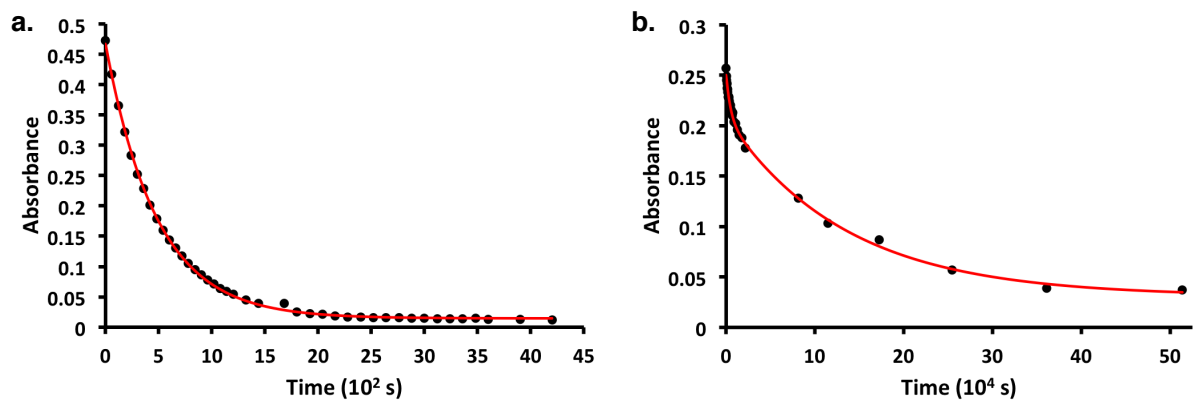
**Figure S12.** UV-Vis absorption spectra for **SP2** a) irradiation at 365 nm in MeCN, b) thermal decay at 566 nm in MeCN, c) irradiation at 365 nm in nanocrystalline suspension, and d) thermal decay at 582 nm in nanocrystalline suspension.



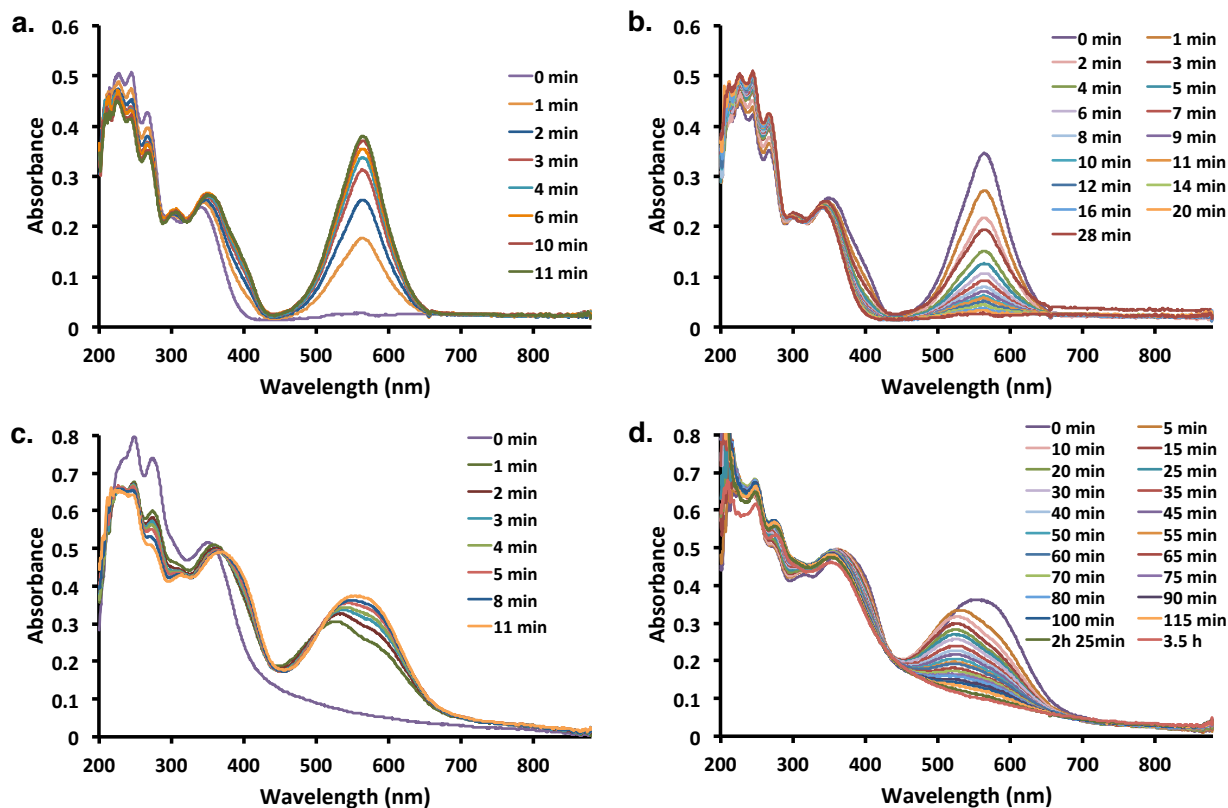
**Figure S13.** Thermal decay kinetic plots with trendline (red line) for **SP2** a) at 566 nm in MeCN and b) at 582 nm in NC suspension.



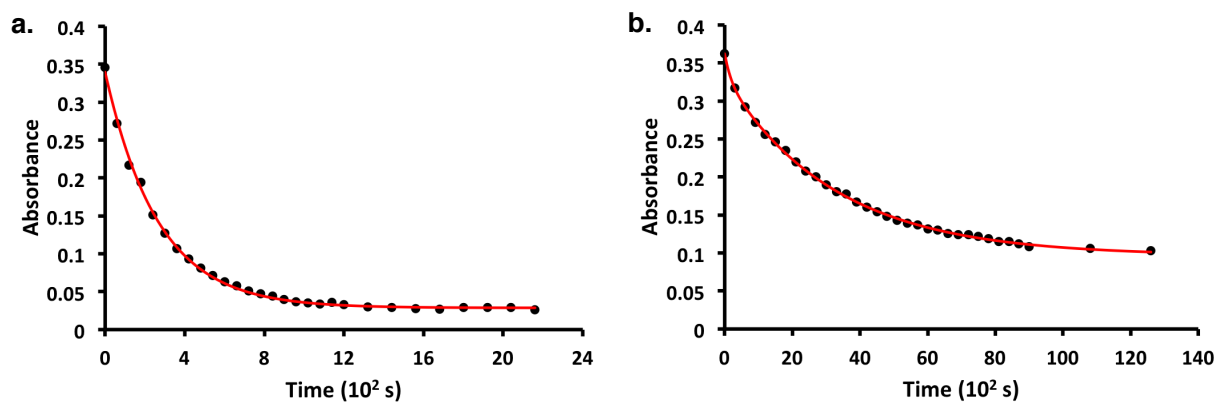
**Figure S14.** UV-Vis absorption spectra for SP3 a) irradiation at 365 nm in MeCN, b) thermal decay at 561 nm in MeCN, c) irradiation at 365 nm in nanocrystalline suspension, and d) thermal decay at 554 nm in nanocrystalline suspension.



**Figure S15.** Thermal decay kinetic plots with trendline (red line) for SP3 a) at 561 nm in MeCN and b) at 554 nm in NC suspension.

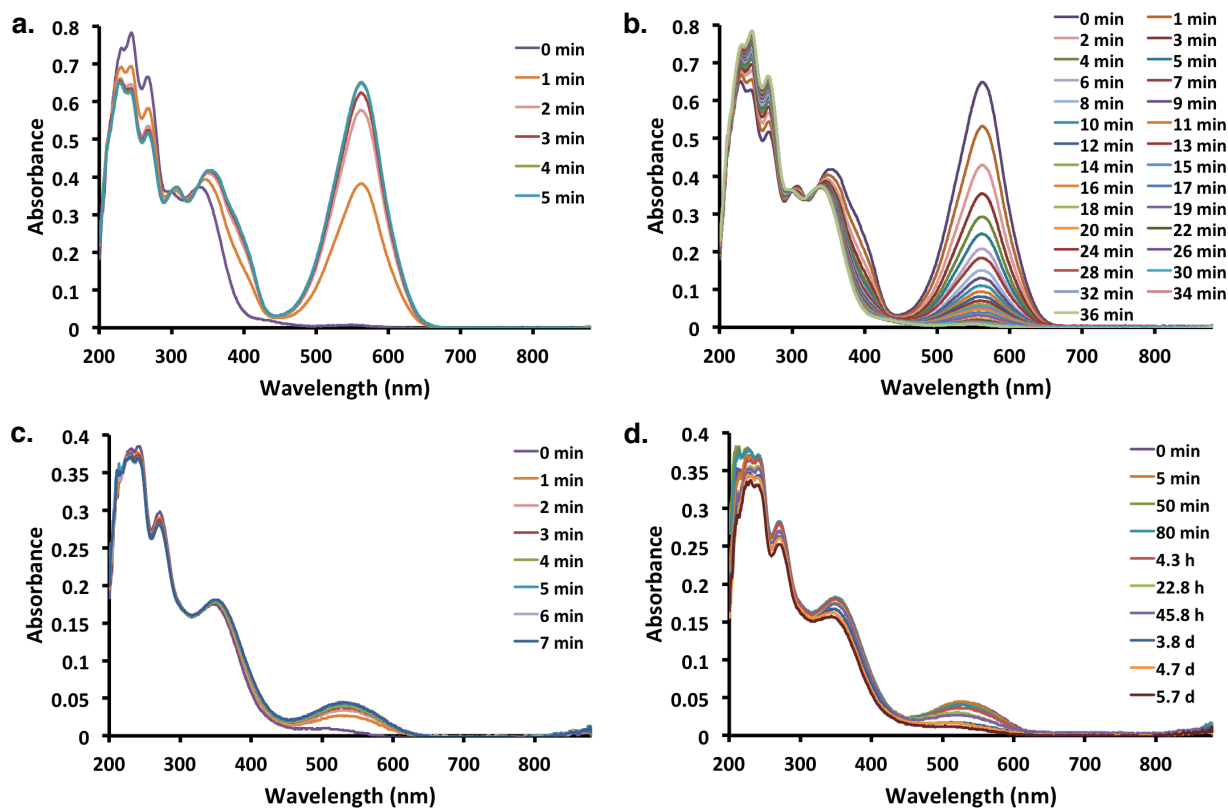


**Figure S16.** UV-Vis absorption spectra for SP4 a) irradiation at 365 nm in MeCN, b) thermal decay at 564 nm in MeCN, c) irradiation at 365 nm in nanocrystalline suspension, and d) thermal decay at 552 nm in nanocrystalline suspension.

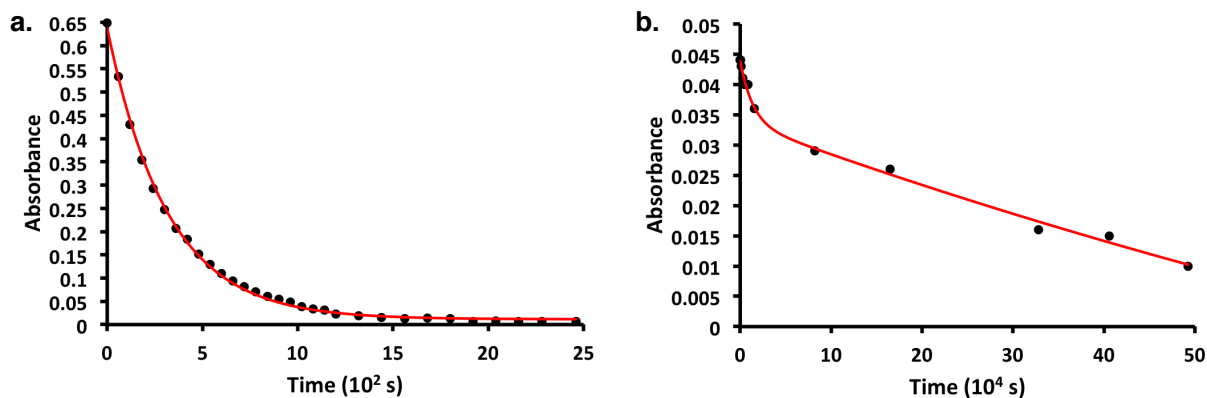


**Figure S17.** Thermal decay kinetic plots with trendline (red line) for SP4 a) at 564 nm in MeCN and b) at 552 nm in NC suspension.

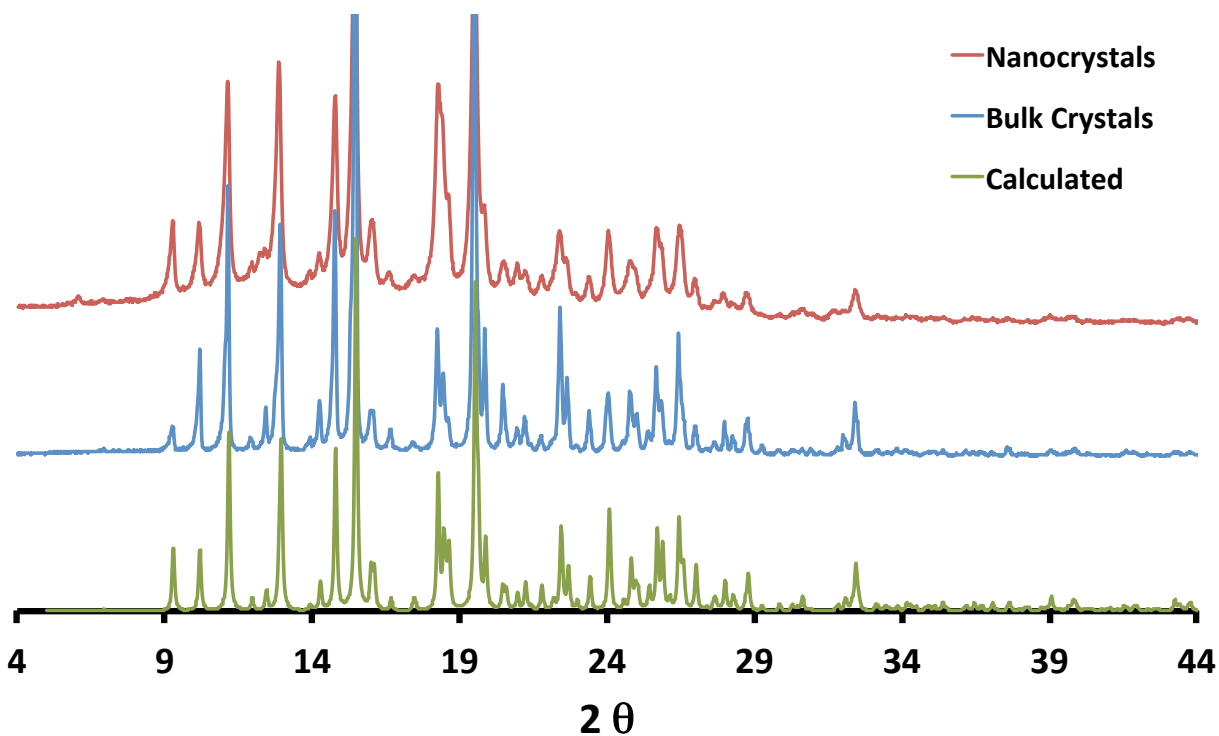




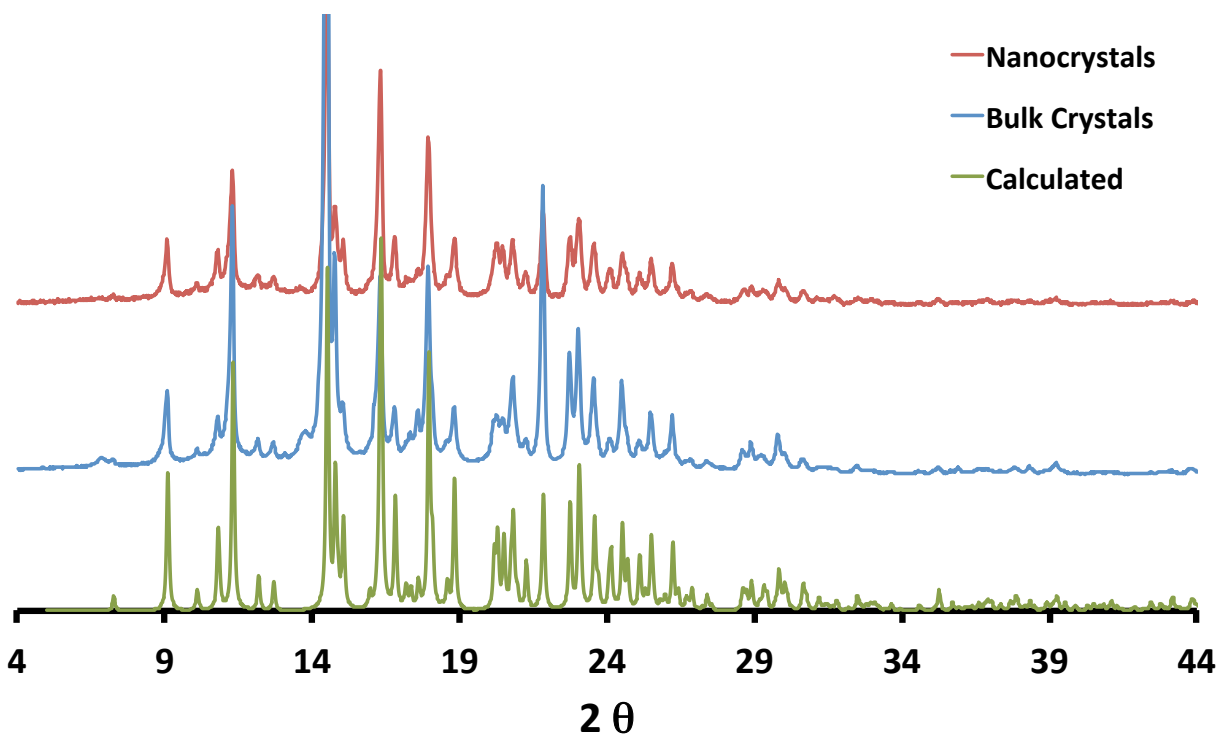
**Figure S18.** UV-Vis absorption spectra for SP5 a) irradiation at 365 nm in MeCN, b) thermal decay at 562 nm in MeCN, and c) irradiation at 365 nm in nanocrystalline suspension, and d) thermal decay at 531 nm in nanocrystalline suspension.



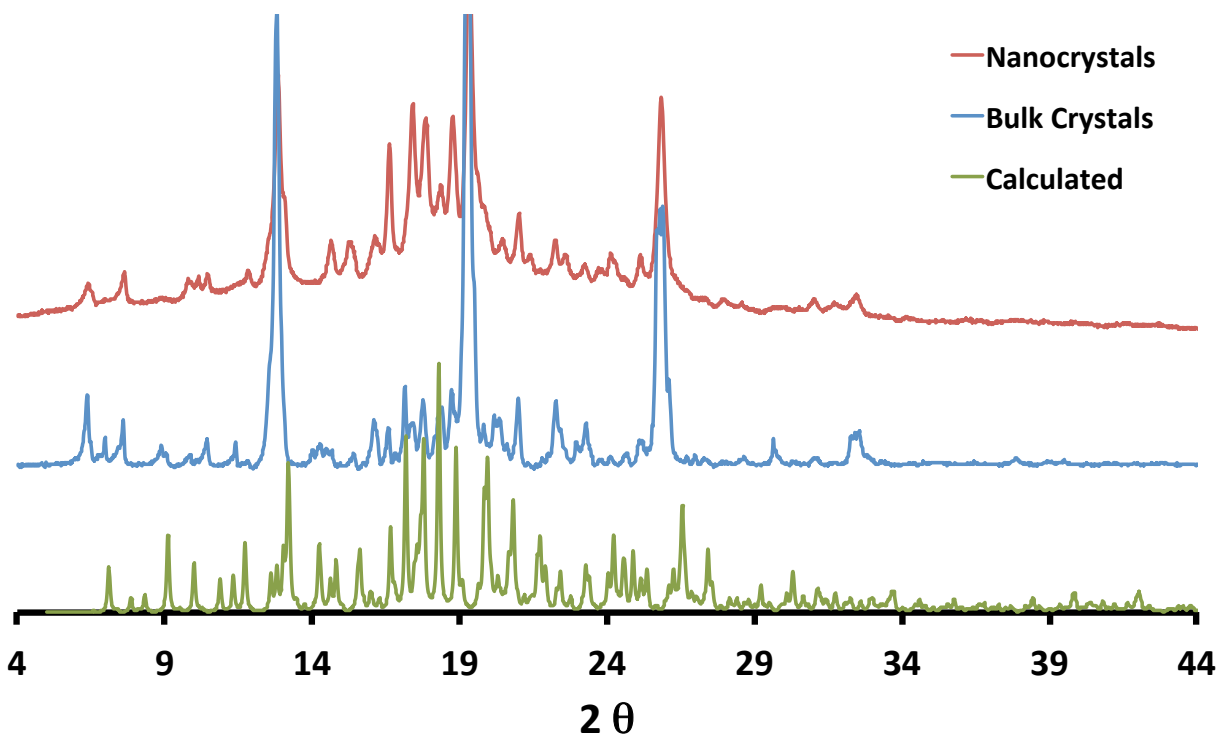
**Figure S19.** Thermal decay kinetic plots with trendline (red line) for SP5 a) at 562 nm in MeCN and b) at 531 nm in NC suspension.



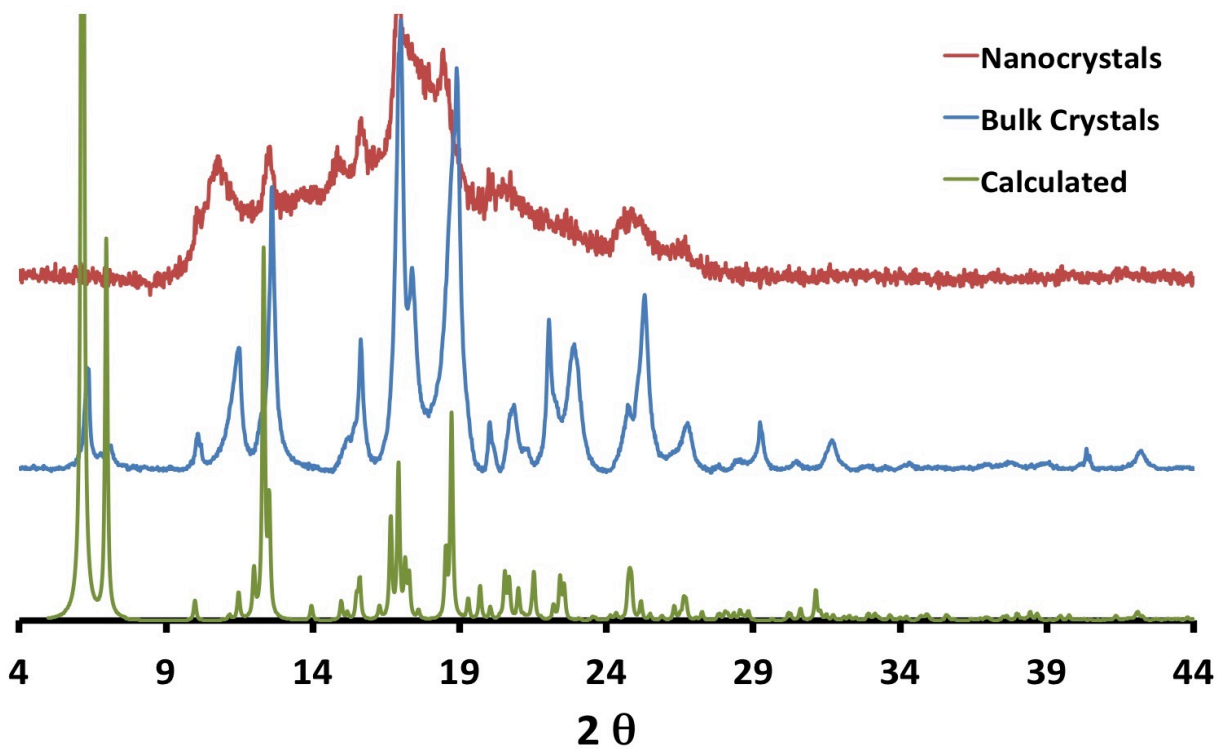
**Figure S20.** PXRD comparison of experimentally obtained nanocrystals (top) and bulk crystals (middle) to the calculated (bottom) diffractogram from the CSD for **SP2**.



**Figure S21.** PXRD comparison of experimentally obtained nanocrystals (top) and bulk crystals (middle) to the calculated (bottom) diffractogram from the CSD for **SP3**.



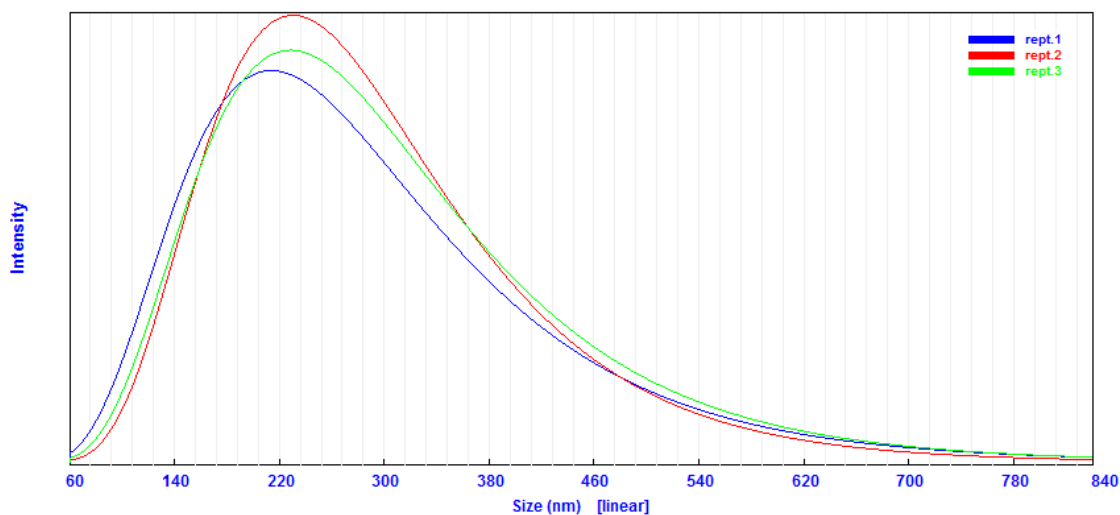
**Figure S22.** PXRD comparison of experimentally obtained nanocrystals (top) and bulk crystals (middle) to the calculated (bottom) diffractogram from the CSD for **SP4**.



**Figure S23.** PXRD comparison of experimentally obtained nanocrystals (top) and bulk crystals (middle) to the calculated (bottom) diffractogram from the CSD for **SP5**.

Unimodal Results Summary

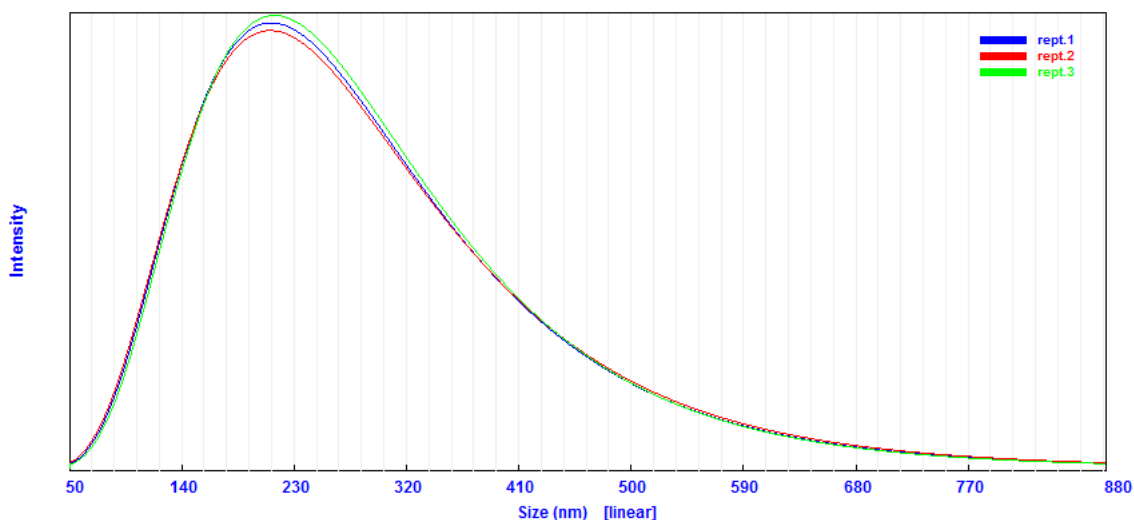
Rept#.	Mean (nm)	Std.Dev (nm)	Baseline Error	P.I.	Counts/s	Diff.Coeff (m <sup>2</sup> /s)	Overflow
Rept.1	224.5	87.0	0.02%	0.188	3.77e+05	2.02e-12	0
Rept.2	234.6	77.5	0.01%	0.097	3.69e+05	1.93e-12	0
Rept.3	236.8	86.2	0.04%	0.141	3.55e+05	1.91e-12	0
Average	232.0	83.54		0.142			



**Figure S24.** Dynamic light scattering results for the nanocrystals of **SP1** dispersed in water showing the average particle size to be 232 nm.

Unimodal Results Summary

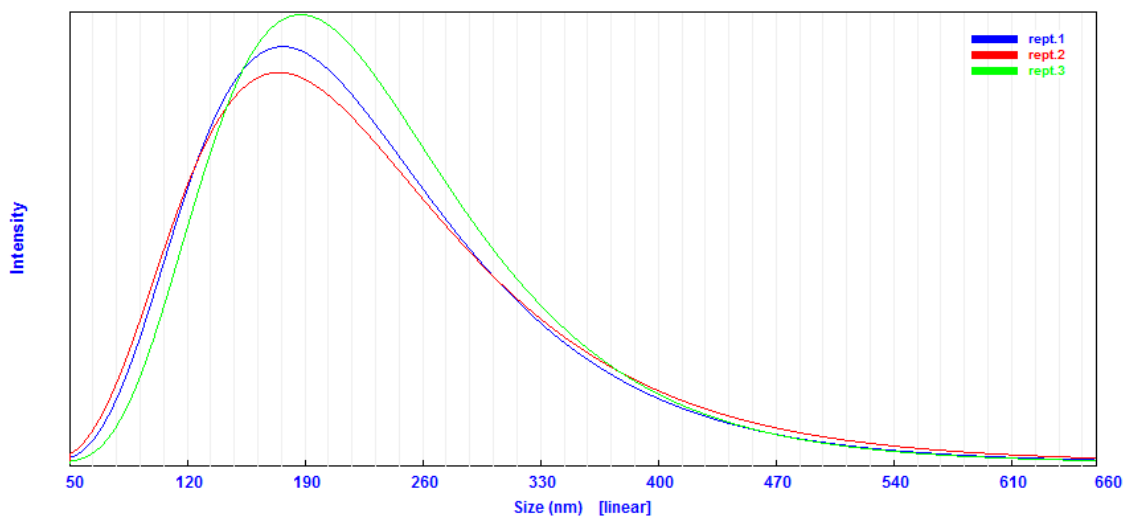
Rept#.	Mean (nm)	Std.Dev (nm)	Baseline Error	P.I.	Counts/s	Diff.Coeff (m <sup>2</sup> /s)	Overflow
Rept.1	223.4	88.6	-0.00%	0.212	1.59e+06	2.03e-12	0
Rept.2	223.6	90.6	0.00%	0.239	1.58e+06	2.02e-12	0
Rept.3	224.9	87.4	0.00%	0.190	1.58e+06	2.01e-12	0
Average	224.0	88.86		0.214			



**Figure S25.** Dynamic light scattering results for the nanocrystals of **SP2** dispersed in water showing the average particle size to be 224 nm.

Unimodal Results Summary

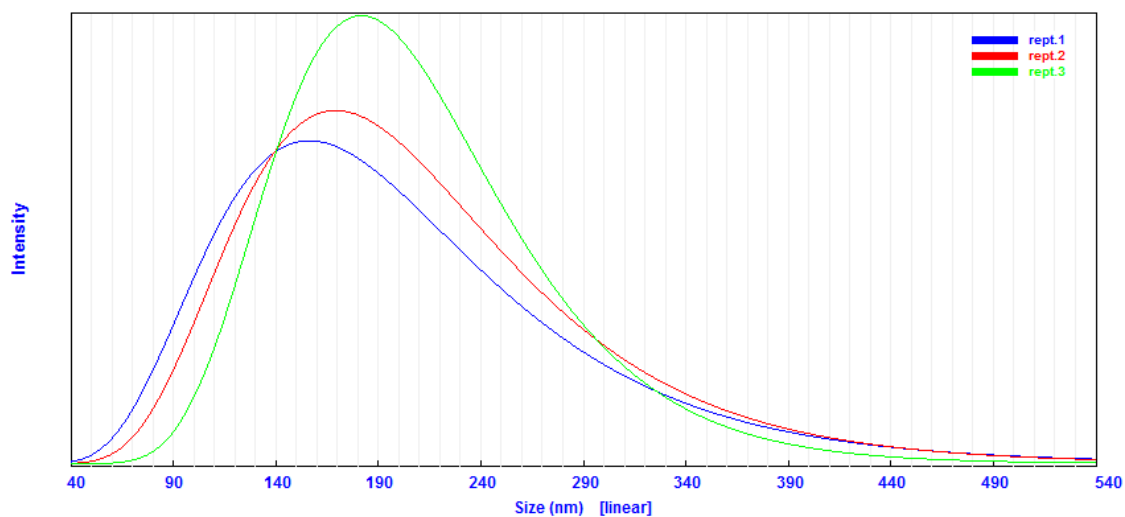
Rept#.	Mean (nm)	Std.Dev (nm)	Baseline Error	P.I.	Counts/s	Diff.Coeff (m <sup>2</sup> /s)	Overflow
Rept.1	181.7	63.5	0.00%	-0.119	6.59e+05	2.49e-12	0
Rept.2	182.0	68.7	0.01%	0.165	6.85e+05	2.49e-12	0
Rept.3	189.9	60.8	0.01%	0.087	7.17e+05	2.38e-12	0
Average	184.5	64.32		0.044			



**Figure S26.** Dynamic light scattering results for the nanocrystals of **SP3** dispersed in water showing the average particle size to be 184 nm.

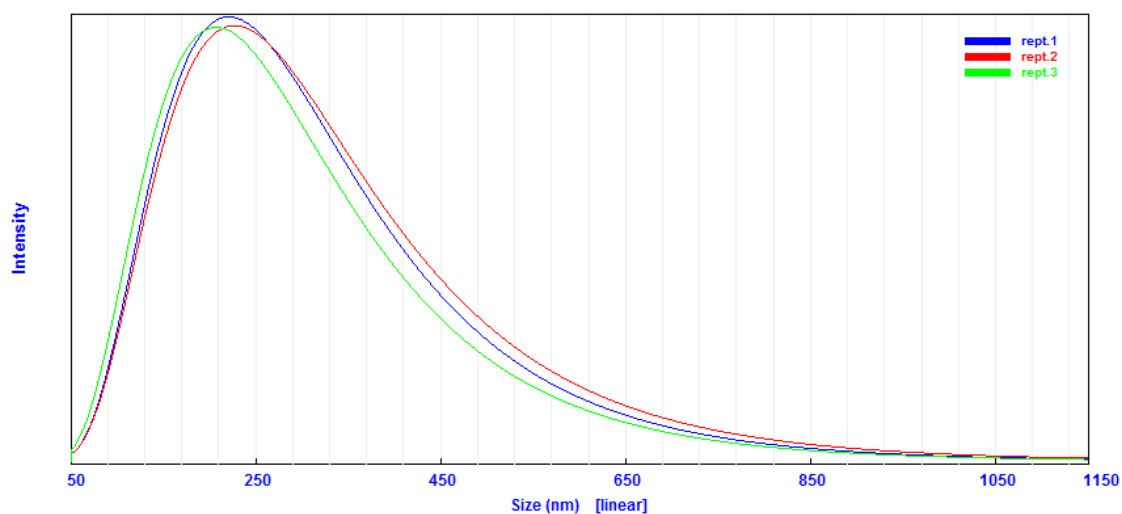
Unimodal Results Summary

Rept#.	Mean (nm)	Std.Dev (nm)	Baseline Error	P.I.	Counts/s	Diff.Coeff (m <sup>2</sup> /s)	Overflow
Rept.1	160.8	56.3	0.14%	0.121	4.46e+05	2.82e-12	0
Rept.2	171.2	54.0	0.06%	0.083	5.04e+05	2.64e-12	0
Rept.3	178.8	43.2	0.03%	0.038	5.46e+05	2.53e-12	0
Average	170.2	51.17		0.080			

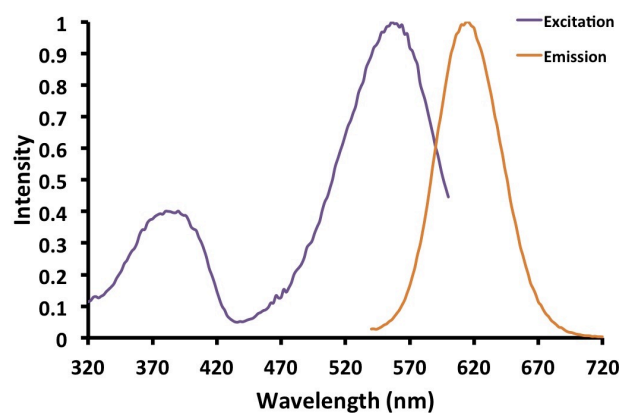


**Figure S27.** Dynamic light scattering results for the nanocrystals of **SP4** dispersed in water showing the average particle size to be 170 nm.

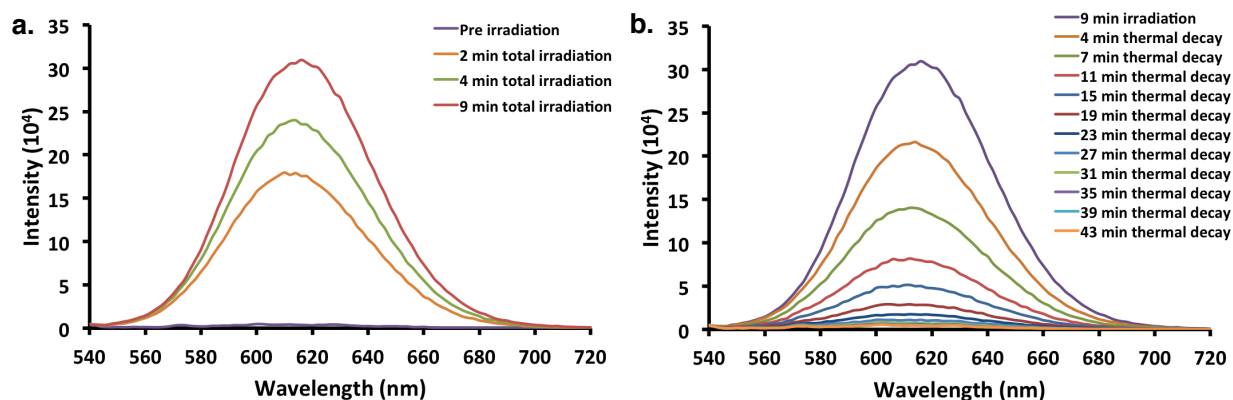
Unimodal Results Summary							
Rept.#	Mean (nm)	Std.Dev (nm)	Baseline Error	P.I.	Counts/s	Diff.Coeff (m <sup>2</sup> /s)	Overflow
Rept.1	238.3	103.0	4.13%	0.371	2.27e+05	1.90e-12	0
Rept.2	246.9	109.5	3.33%	0.463	2.22e+05	1.83e-12	0
Rept.3	225.4	100.5	0.27%	0.486	2.04e+05	2.01e-12	0
Average	236.9	104.37		0.440			



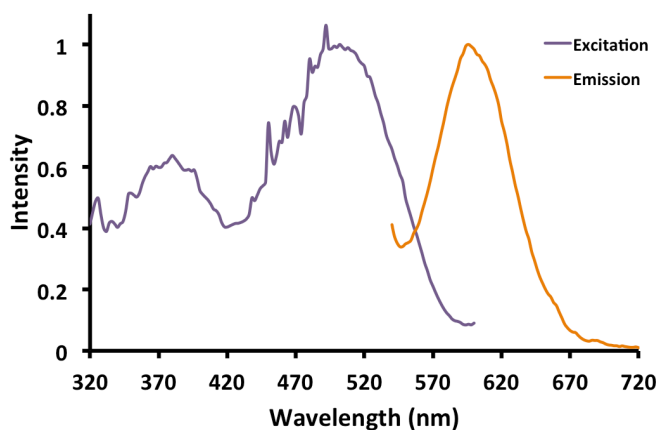
**Figure S28.** Dynamic light scattering results for the nanocrystals of **SP5** dispersed in water showing the average particle size to be 237 nm.



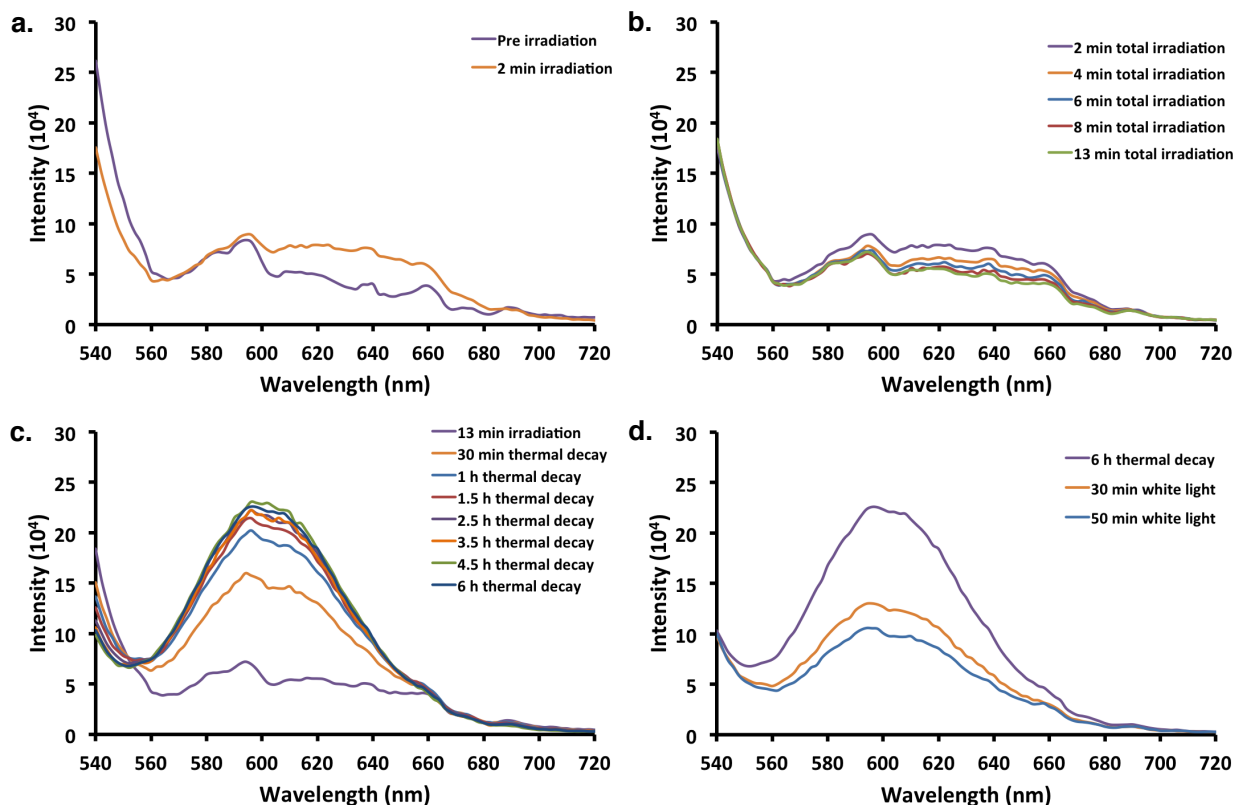
**Figure S29.** Normalized excitation and emission spectra for **SP1** in acetonitrile post 365 nm irradiation. Excitation and emission spectra were acquired using an emission wavelength of 612 nm and an excitation wavelength of 530 nm, respectively.



**Figure S30.** Emission spectra for **SP1** in acetonitrile using an excitation wavelength of 530 nm a) pre irradiation and post 2, 4, and 9 min irradiations at 365 nm and b) thermal decay of the merocyanine over time.



**Figure S31.** Normalized excitation and emission spectra for **SP1** in nanocrystalline suspension post 365 nm irradiation. Excitation and emission spectra were acquired using an emission wavelength of 612 nm and an excitation wavelength of 530 nm, respectively.



**Figure S32.** Emission spectra for **SP1** in nanocrystalline suspension using an excitation wavelength of 530 nm a) pre irradiation and post 2 min irradiation at 365 nm, b) repeated irradiations at 365 nm, c) fluorescence signal intensity increase during the thermal decay of the merocyanine over time, and d) white light induced decay of the merocyanine.

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