Supporting information for:

Fast Photo-driven Electron Spin Coherence Transfer: The Effect of Electron-Nuclear Hyperfine Coupling on Coherence Dephasing

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General. Naphthalene-1,8:4,5-tetracarboxylic dianhydride was purchased from TCI America and used as obtained. All other commercially available reagents were ordered from Aldrich and used as obtained. Toluene and tetrahydrofuran (ACS reagent grade, Aldrich) were dried using a Glass Contour solvent system. All column chromatography was performed on standard silica gel, 60 Å porosity, 40-63 μm diameter (Sorbent Technologies). Proton and carbon-13 nuclear magnetic resonance spectra were obtained on a Varian Inova 500 MHz or a Bruker Avance III 500 MHz spectrometer with TMS as an internal standard. Mass spectrometry was performed on a Bruker Daltonics Autoflex III Smartbeam MALDI spectrometer with a matrix of 2-hydroxy-1-naphthoic acid.

1. Synthesis

The synthesis of PER-d_{11}-NDI_1-NDI_2 proceeded similarly to the route previously published for PER-h_{11}-NDI_1-NDI_2, starting with perylene-d_{12} (98%) purchased from Cambridge Isotopes. Characterization of the final product (two rotamers) was performed using a Bruker Avance III 500 MHz NMR and a Bruker Autoflex III mass spectrometer. ^{1}H-NMR (500 MHz, $CDCl_3$) δ 8.95 – 8.88 (m, 8H), 7.63 (dd, *J* = 8.7, 1.0 Hz, 1H), 7.54 – 7.53 (m, 4H), 7.36 – 7.27 (m, 2H), 7.06-7.00 (m, 1H), 2.28 – 2.20 (m, 6H) 2.11 (s, 6H), 1.96 (s, 6H), 1.37 – 1.32 (m, 9H), 1.32 – 1.28 (m, 9H). ^{13}C NMR (126 MHz, $CDCl_3$) δ 163.95, 163.21, 162.55, 162.50, 150.56, 144.02, 143.85, 140.62, 140.38, 139.24, 135.22, 135.13, 134.83, 134.77, 133.65, 132.96, 132.94, 132.15, 132.13, 131.76, 131.68, 131.63, 131.57, 131.53, 131.47, 131.31, 131.27, 130.88, 130.84, 130.24, 129.19, 129.12, 128.81, 128.75, 128.59, 127.68, 127.65, 127.58, 127.51, 127.48, 127.42, 127.39, 127.34, 127.26, 127.13, 127.09, 126.87, 126.20 (br), 119.90 (br), 35.75, 34.45, 31.92, 31.36, 21.21, 18.51, 17.99, 17.60. MS (MALDI-TOF) *m/z* for $C_{86}H_{53}D_{11}N_4O_8$ (M^+) calculated: 1291.54, found: 1291.60.

2. Femtosecond Transient Absorption Spectroscopy

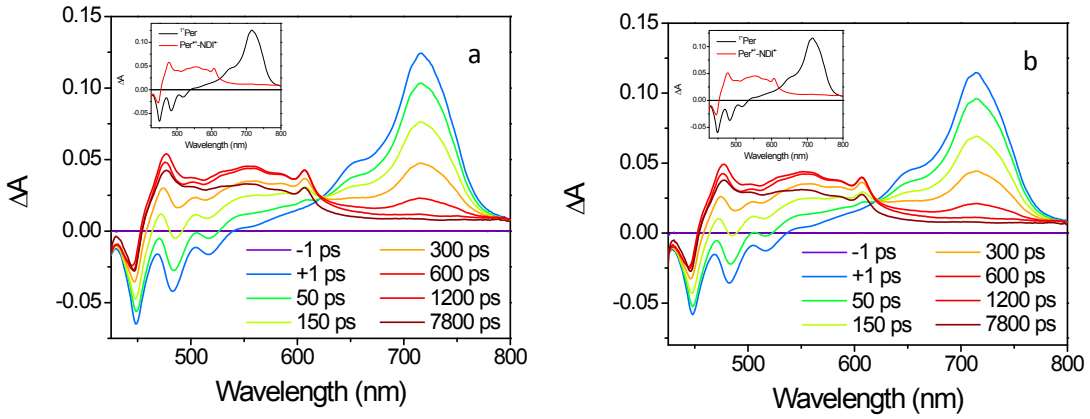

Figure S1: Transient absorption of (a) HPer-DTol and (b) DPer-HTol following a 416 nm, 120 fs laser pulse. Inset: Spectra of ^{1*}PER and PER$^{+\bullet}$-NDI$^{-\bullet}$ obtained by SVD and global analysis of transient absorption dataset.

3. Nanosecond Transient Absorption Spectroscopy

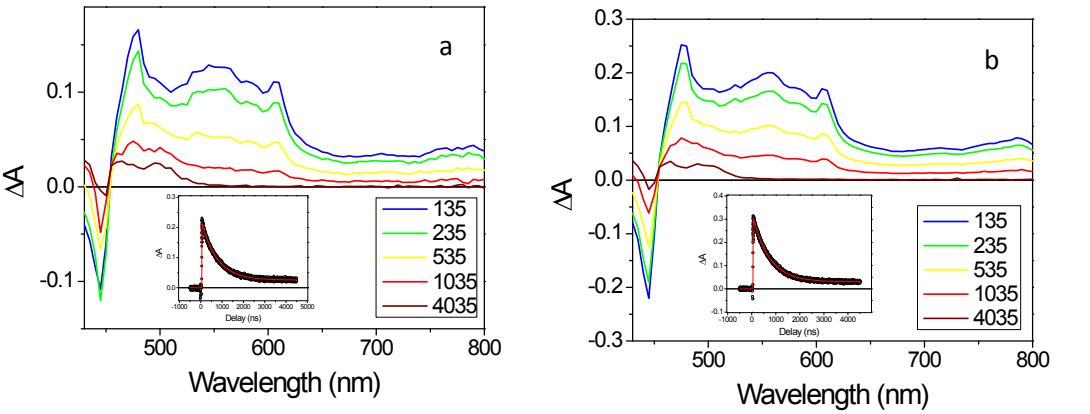

Figure S2: Transient absorption of (a) HPer-DTol and (b) DPer-HTol following a 416 nm, 7 ns laser pulse. Inset: Transient kinetics were monitored at 480 nm