Supplementary Figures:

**Figure S1.** 1-1 NOESY regions (90% H2O/10% D2O, 500 MHz, 25 °C, t_{mix} = 200 ms) showing the Py6 H3 and H5 to amide NH6 cross peaks. (A) ImPyPy-γ-PyPyPy-Gly-Dp/TGTTT. (B) ImPyPy-γ-PyPyPy-Gly-Dp/GGTTT.

**Figure S2.** 1-D titrations of (A) 2:1 ImPyPy-γ-PyPyPy-Gly-Dp/TAACGT TA complex, and (B) 2:1 ImPyPy-γ-PyPyPy-Gly-Dp/TAACGT TA complex. Shown are the aromatic regions. Relative ratios of ligand to DNA are given. Note that in (A) the spectra broaden during the course of the titration and never sharpen where as in (B) the spectra broaden and then sharpen near the 2:1 ratio.