Supporting Figure 1: Quantitative DNase I footprinting titration with ImPyβImPy-γ-PyPyβImPy-β-Dp (**1**) on the 292-bp restriction fragment from the plasmid pSES 19-1: lane 1, A reaction; lane 2, DNase I control; lanes 3-12, 10 pM, 20 pM, 50 pM, 100 pM, 200 pM, 500 pM, 1 nM, 2 nM, 5 nM, and 10 nM **1**; lane 13, intact DNA. The four putative binding sites are shown on the right of the autoradiograms. All reactions contain a 15 kcpm restriction fragment, 10 mM Tris•HCl (pH 7.0), 10 mM KCl, 10 mM $MgCl_2$, and 5 mM $CaCl_2$.

Supporting Figure 2: Quantitative DNase I footprinting titration with ImPyβImPy-γ-PyPy^{S}IsImPy-β-Dp (**4**) on the 292-bp restriction fragment from the plasmid pSES 19-1: lane 1, A reaction; lane 2, DNase I control; lanes 3-12, 10 pM, 20 pM, 50 pM, 100 pM, 200 pM, 500 pM, 1 nM, 2 nM, 5 nM, and 10 nM **4**; lane 13, intact DNA. The four putative binding sites are shown on the right of the autoradiograms. All reactions contain a 15 kcpm restriction fragment, 10 mM Tris•HCl (pH 7.0), 10 mM KCl, 10 mM $MgCl_2$, and 5 mM $CaCl_2$.

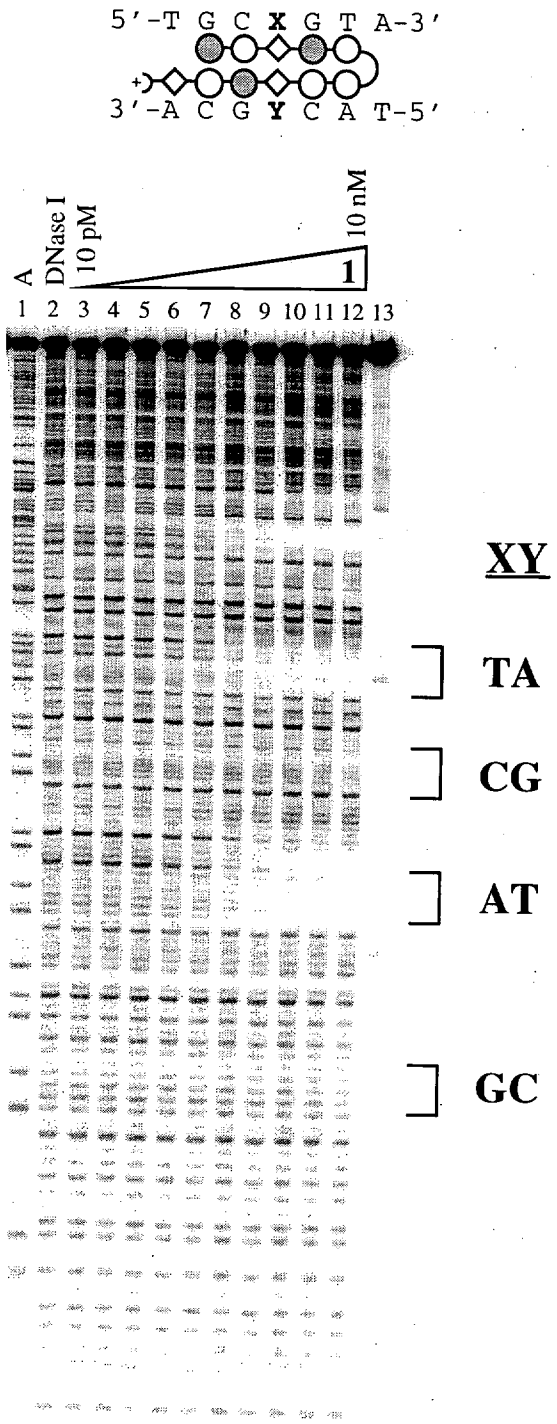
Supporting Figure 3: Quantitative DNase I footprinting titration with ImPyAaImPy-γ-PyPyβImPy-β-Dp (**5**) on the 292-bp restriction fragment from the plasmid pSES 19-1: lane 1, A reaction; lane 2, DNase I control; lanes 3-11, 100 pM, 200 pM, 500 pM, 1 nM,

2 nM, 5 nM, 10 nM, 20 nM, and 50 nM nM **5**. The four putative binding sites are shown on the right of the autoradiograms. All reactions contain a 15 kcpm restriction fragment, 10 mM Tris•HCl (pH 7.0), 10 mM KCl, 10 mM $MgCl_2$, and 5 mM $CaCl_2$.

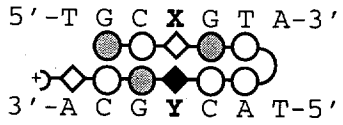
Supporting Figure 4: Quantitative DNase I footprinting titration with ImPyFbImPy-γ-PyPyβImPy-β-Dp (**6**) on the 292-bp restriction fragment from the plasmid pSES 19-1: lane 1, A reaction; lane 2, DNase I control; lanes 3-12, 100 pM, 200 pM, 500 pM, 1 nM, 2 nM, 5 nM, 10 nM, 20 nM, 50 nM, and 100 nM **6**; lane 13, intact DNA. The four putative binding sites are shown on the right of the autoradiograms. All reactions contain a 15 kcpm restriction fragment, 10 mM Tris•HCl (pH 7.0), 10 mM KCl, 10 mM $MgCl_2$, and 5 mM $CaCl_2$.

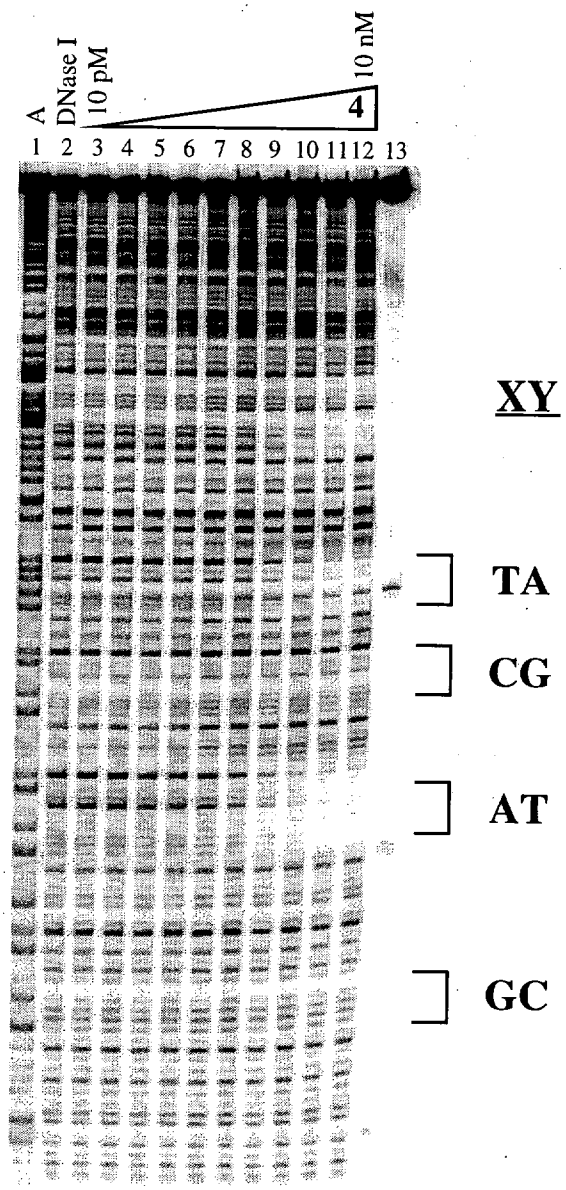
Supporting Figure 5: (a) MPE•Fe(II) footprinting titration of **2** and affinity cleaving study of **2E** on the 292-bp 5'-^{32}P-labeled restriction fragment from the plasmid pSES 19-1: lane 1, intact DNA; lane 2, A reaction; lane 3, MPE control; lanes 4-6, 100 pM, 1 nM, and 10 nM **2**; lanes 7-9, 100 pM, 1 nM, and 10 nM **2E**. The sequences 5'-TGCTGTA-3' and 5'-TGCAGTA-3' are shown at the right of the autoradiogram. All reactions contain a 15 kcpm restriction fragment, 20 mM HEPES buffer (pH 7.0), and 10 mM NaCl. (b) Results from MPE•Fe(II) footprinting titration of **2**. Bold sequences represent binding

sites determined by the published model. Bar heights are proportional to the relative protection from cleavage at each band. (c) Results from the affinity cleaving study of **2E**. Bold sequences represent binding sites determined by the published model. Line heights are proportional to the relative cleavage at each band.

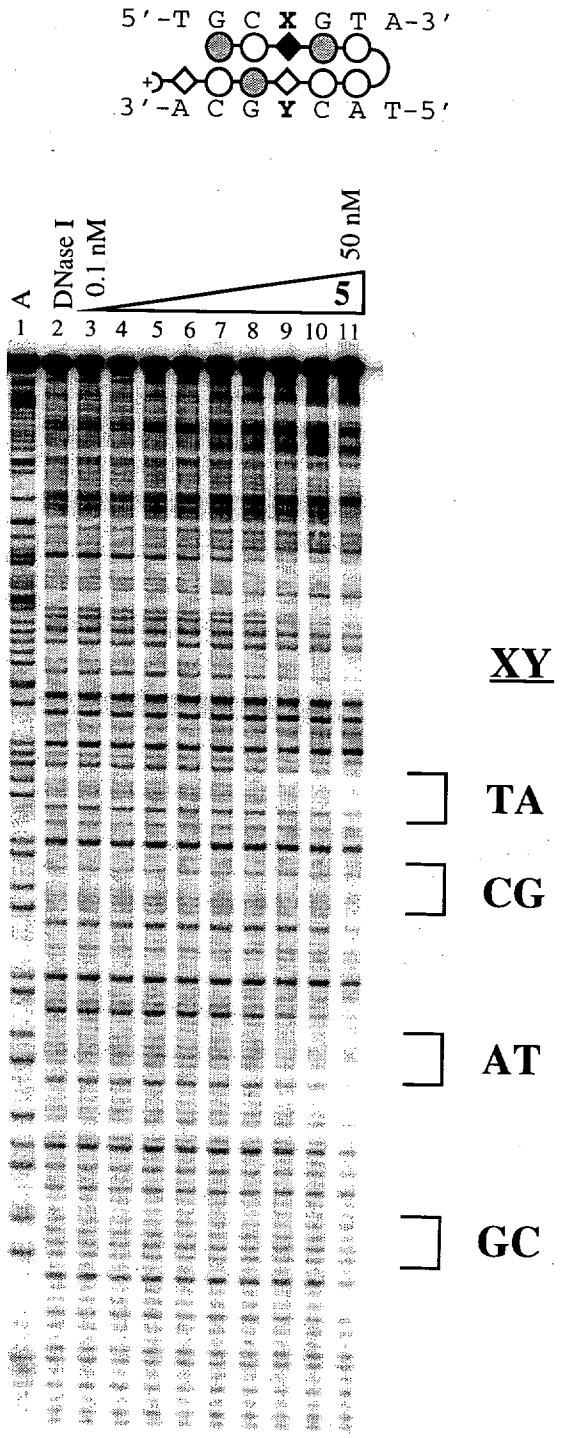

5′-T G C X G T A-3′
3′-A C G Y C A T-5′
A
DNase I
10 pM
10 nM
1
1 2 3 4 5 6 7 8 9 10 11 12 13
XY
TA
CG
AT
GC

SI Figure 1

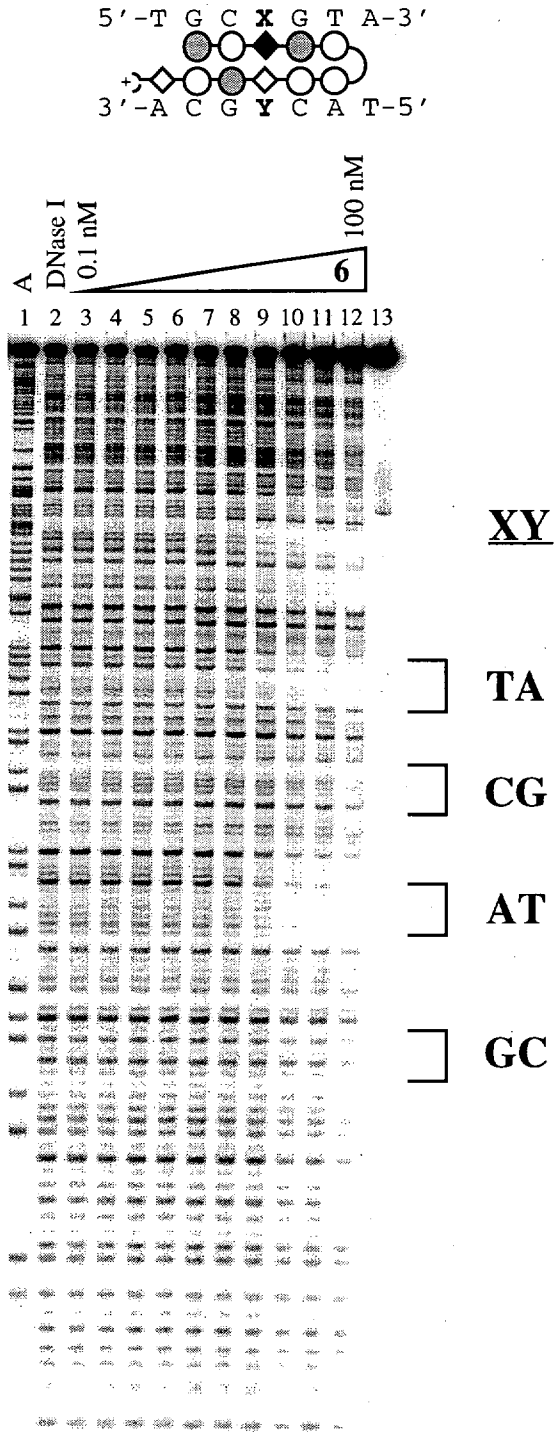

5′-T G C X G T A-3′
3′-A C G Y C A T-5′


A
DNase I
10 pM
10 nM
4
1 2 3 4 5 6 7 8 9 10 11 12 13
XY
TA
CG
AT
GC

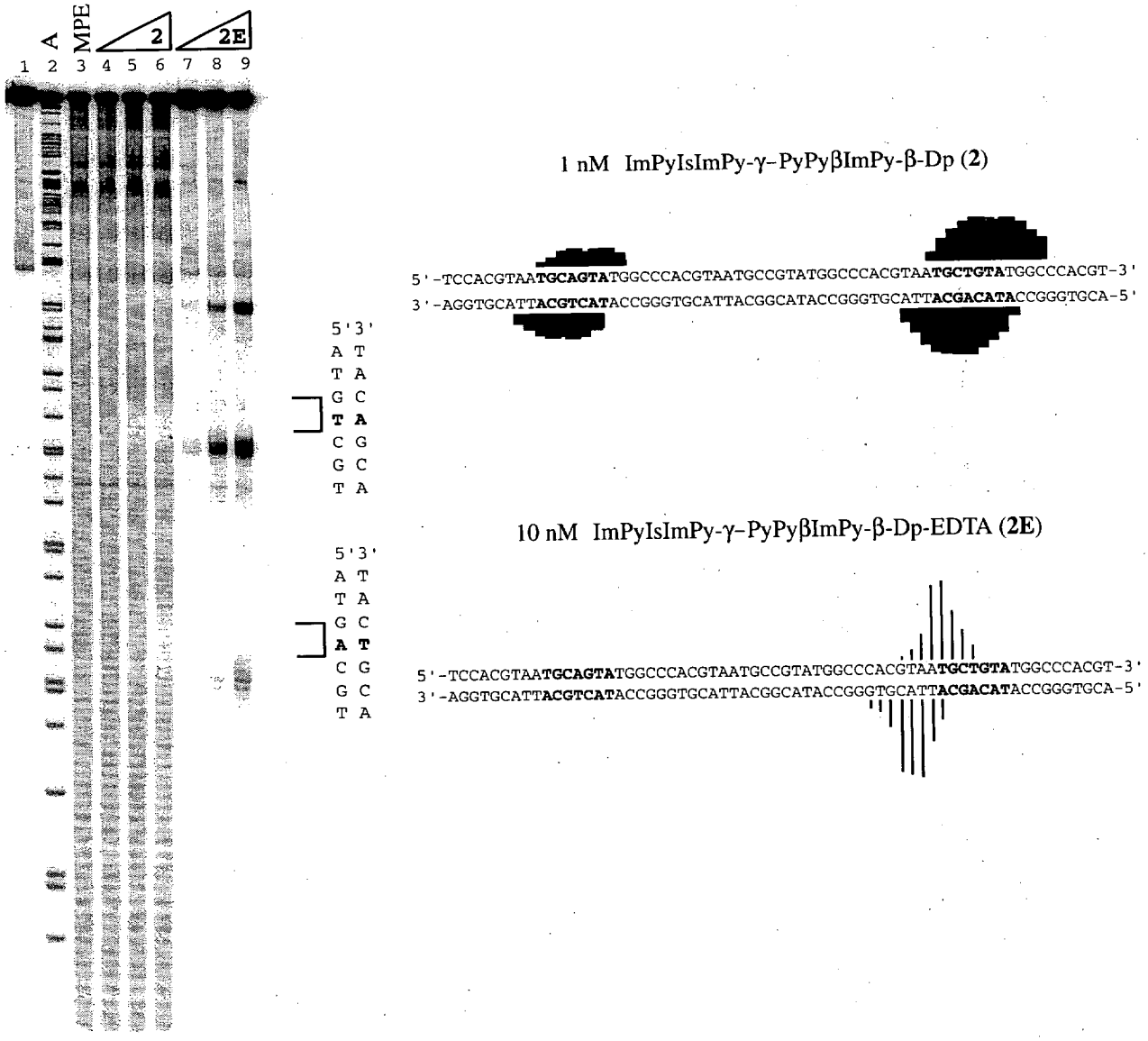
SI Figure 2


5′-T G C X G T A-3′
3′-A C G Y C A T-5′
A
DNase I
0.1 nM
50 nM
5
1 2 3 4 5 6 7 8 9 10 11
XY
TA
CG
AT
GC

SI Figure 3


5′-T G C X G T A-3′
3′-A C G Y C A T-5′
A
DNase I
0.1 nM
100 nM
6
1 2 3 4 5 6 7 8 9 10 11 12 13
XY
TA
CG
AT
GC

SI Figure 4


A
MPE
2
2E
1 2 3 4 5 6 7 8 9
5'3'
A T
T A
G C
T A
C G
G C
T A
5'3'
A T
T A
G C
A T
C G
G C
T A
1 nM ImPyIsImPy-γ-PyPyβImPy-β-Dp (2)
5'-TCCACGTAATGCAGTATGGCCCACGTAATGCCGTATGGCCCACGTAATGCTGTATGGCCCACGT-3'
3'-AGGTGCATTACGTCATACCGGGTGCATTACGGCATACCGGGTGCATTACGACATACCGGGTGCA-5'
10 nM ImPyIsImPy-γ-PyPyβImPy-β-Dp-EDTA (2E)
5'-TCCACGTAATGCAGTATGGCCCACGTAATGCCGTATGGCCCACGTAATGCTGTATGGCCCACGT-3'
3'-AGGTGCATTACGTCATACCGGGTGCATTACGGCATACCGGGTGCATTACGACATACCGGGTGCA-5'

SI FIgure 5