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Table S1: Levels of platinum per DNA strand for modified oligonucleotides^a

| Abbreviation | Oligonucleotide Sequence | Platinum/DNA |
|--------------|--|--------------|
| tsAG*G*A | 5' -CCTCTC AG *G* AT CTTC-3' | 1.13 |
| tsAG*G*T | 5' -CCTCTC AG *G* TT CTTC-3' | 1.11 |
| tsAG*G*C | 5' -CCTCTC AG *G* CT CTTC-3' | 1.12 |
| tsTG*G*A | 5' -CCTCTC TG *G* AT CTTC-3' | 1.13 |
| tsTG*G*T | 5' -CCTCTC TG *G* TT CTTC-3' | 1.03 |
| tsTG*G*C | 5' -CCTCTC TG *G* CT CTTC-3' | 1.09 |
| tsCG*G*A | 5' -CCTCTC CG *G* AT CTTC-3' | 0.94 |
| tsCG*G*T | 5' -CCTCTC CG *G* TT CTTC-3' | 1.13 |
| tsCG*G*C | 5' -CCTCTC CG *G* CT CTTC-3' | 1.17 |

^aPlatinum and DNA strand concentrations measured by atomic absorption and UV-Vis spectroscopy, respectively (see Experimental Procedures).

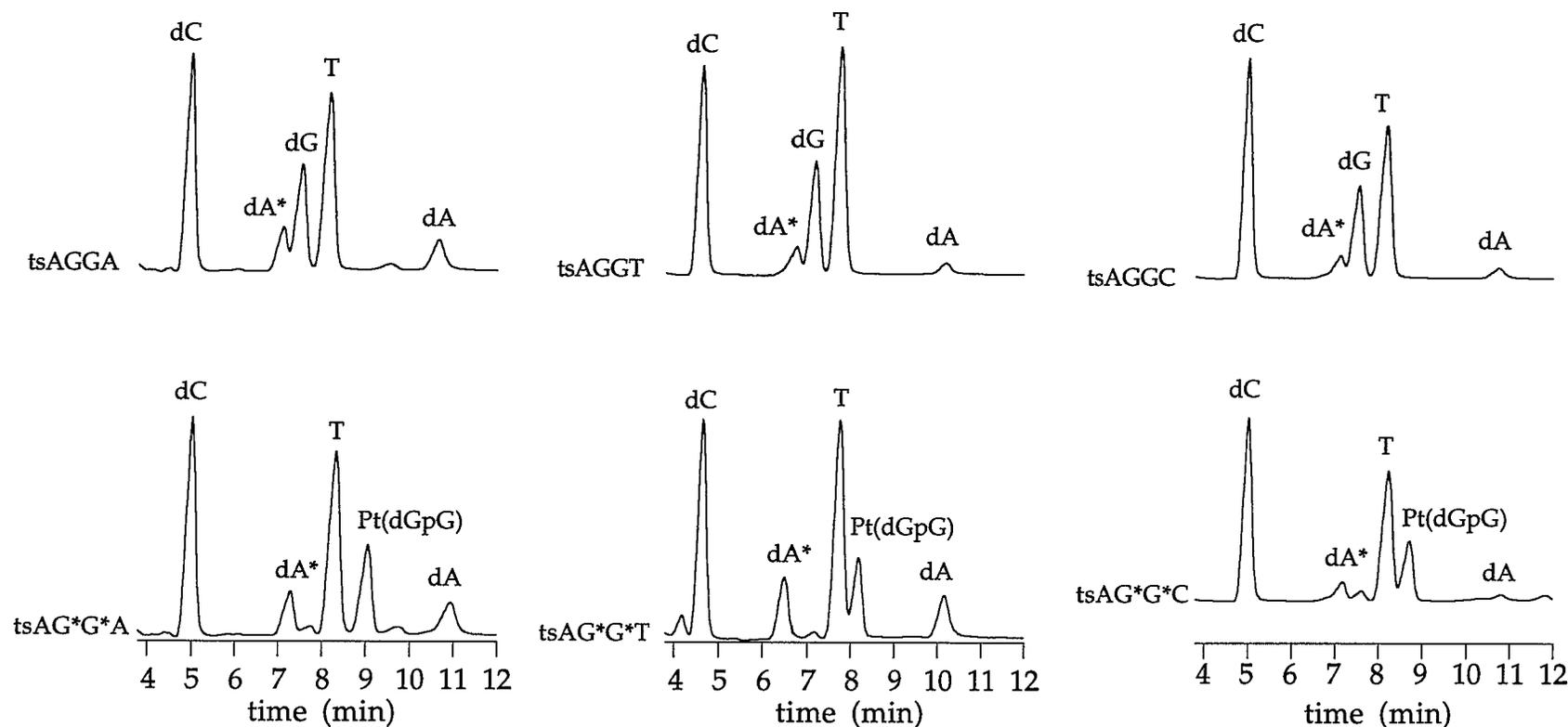
Table S2. Differences in free binding energies of isolated HMG-domain interactions with cisplatin-modified 15-bp DNA duplexes

| Sequence | K_{d2}/K_{d1}^a | ΔG_{Kd}^b (kcal/mol) |
|----------|-------------------|------------------------------|
| dsAG*G*A | 87 ± 27 | ~2.5 |
| dsCG*G*A | 170 ± 93 | ~2.8 |
| dsCG*G*C | 2.6 ± 0.7 | ~0.6 |

^a K_{d1} and K_{d2} are apparent dissociation constants for interaction of each oligonucleotide with HMG1domA and HMG1domB, respectively (See Table 4).

^bDifferences in free binding energies calculated at 277 K from

$$\Delta G_{Kd} = -RT \ln \left(\frac{K_{d2}}{K_{d1}} \right)$$



S4

Figure S1 (A). Reversed phase HPLC traces of enzymatic digestions of cisplatin-modified and unmodified 15mers (Table S1). Peak identities were determined by coinjections with original standards, where dC, dG, dA and T are the mononucleosides, dA* is a side-product from the digestion of dA with with excess P1 nuclease, and Pt(dGpG) is $[\text{Pt}(\text{NH}_3)_2\{\text{d}(\text{GpG})\}]^+$.

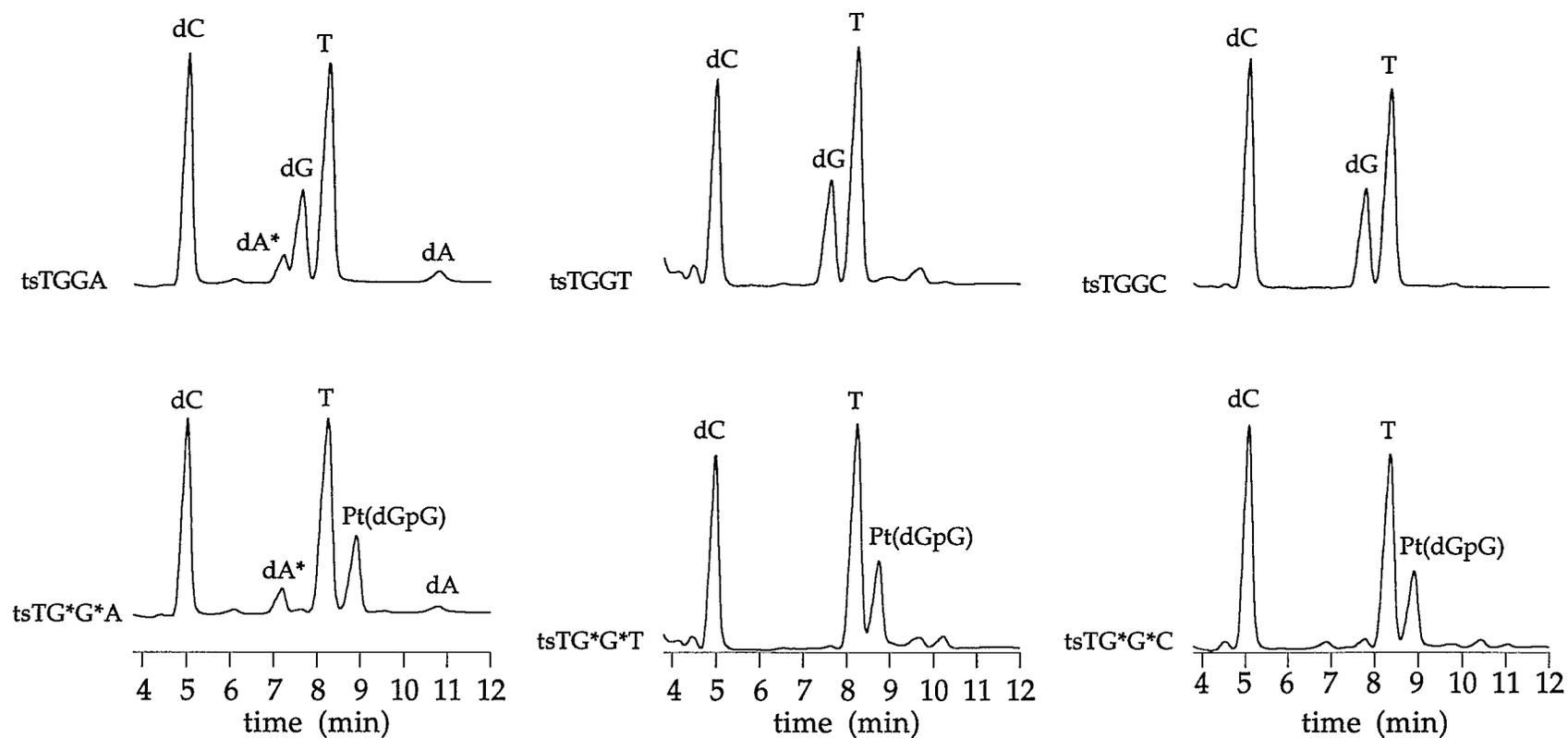


Figure S1 (B). Reversed phase HPLC traces of enzymatic digestions of cisplatin-modified and unmodified 15mers (Table S1). Peak identities were determined by coinjections with original standards, where dC, dG, dA and T are the mononucleosides, dA* is a side-product from the digestion of dA with with excess P1 nuclease, and Pt(dGpG) is $[\text{Pt}(\text{NH}_3)_2\{\text{d}(\text{GpG})\}]^+$.

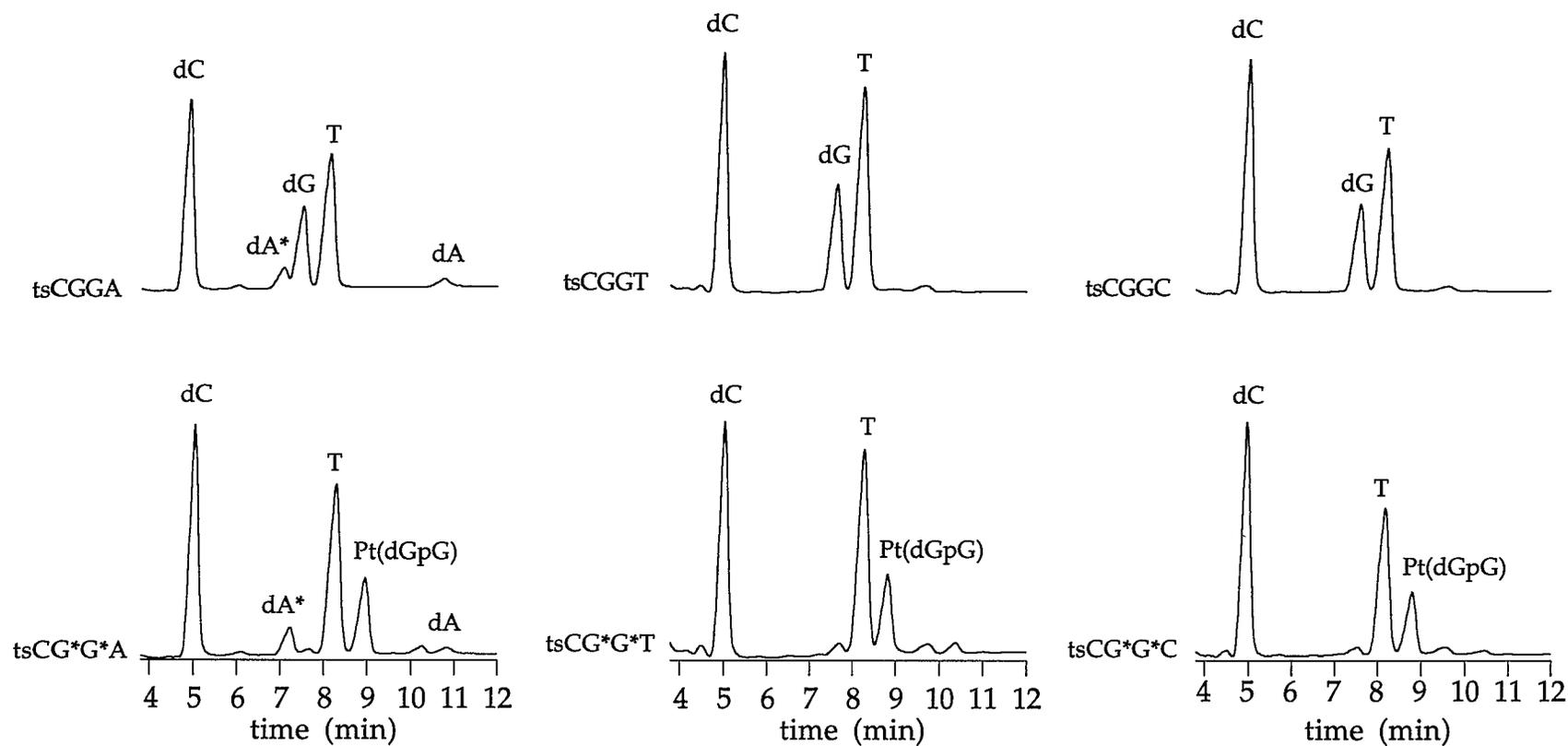


Figure S1 (C). Reversed phase HPLC traces of enzymatic digestions of cisplatin-modified and unmodified 15mers (Table S1). Peak identities were determined by coinjections with original standards, where dC, dG, dA and T are the mononucleosides, dA* is a side-product from the digestion of dA with with excess P1 nuclease, and Pt(dGpG) is $[\text{Pt}(\text{NH}_3)_2\{\text{d}(\text{GpG})\}]^+$.

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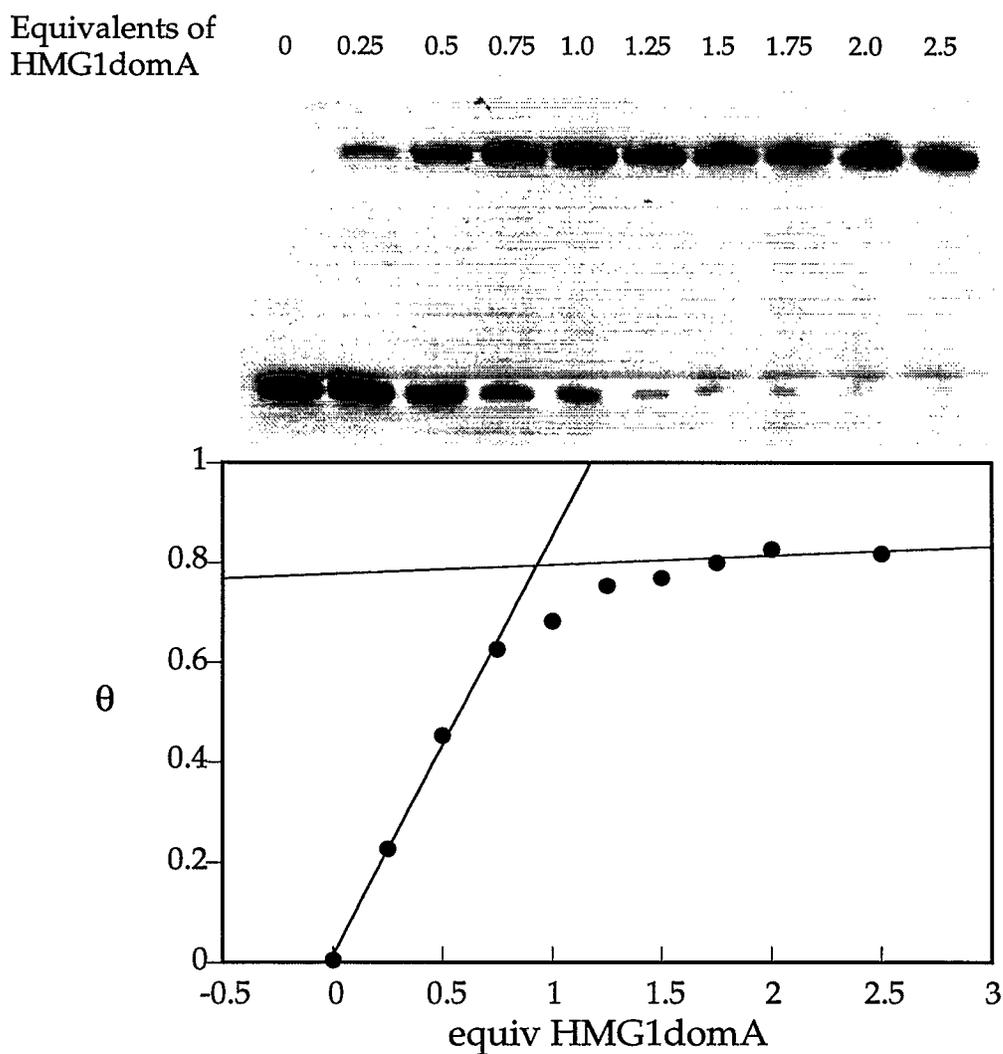


Figure S2. Gel mobility shift assay of 100 nM dsAG*G*A with increasing amounts of HMG1domA (top). Fit of the binding data to determine the stoichiometry of the interaction (bottom). The intersection of the two lines indicates that saturation of binding of the duplex occurs as 1.0 ± 0.2 equivalents of protein.