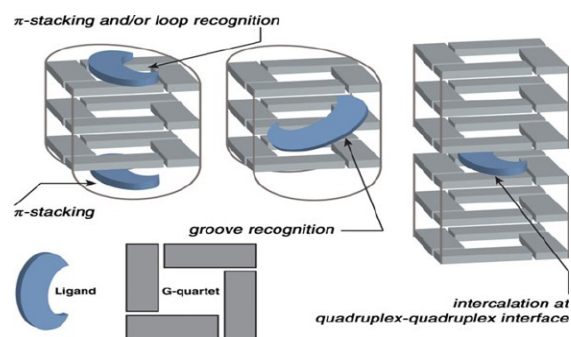
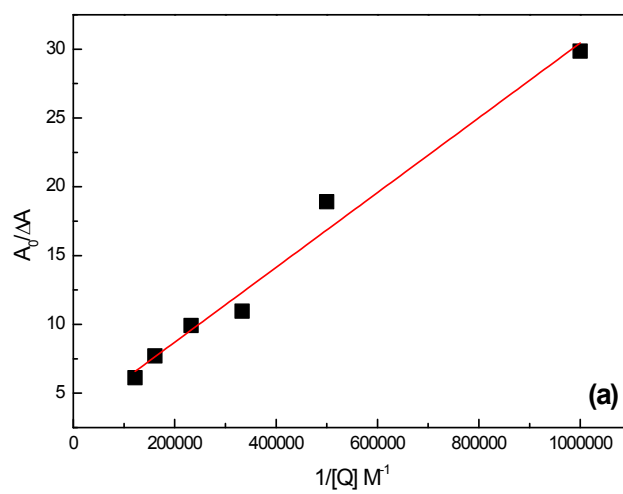
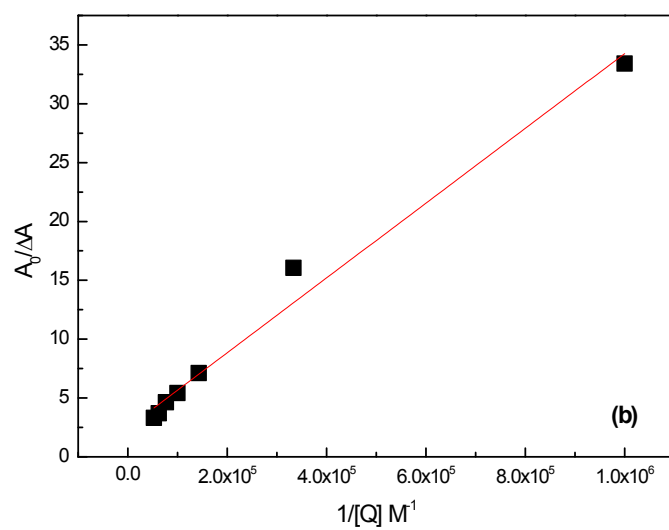


### Electronic Supplementary Information

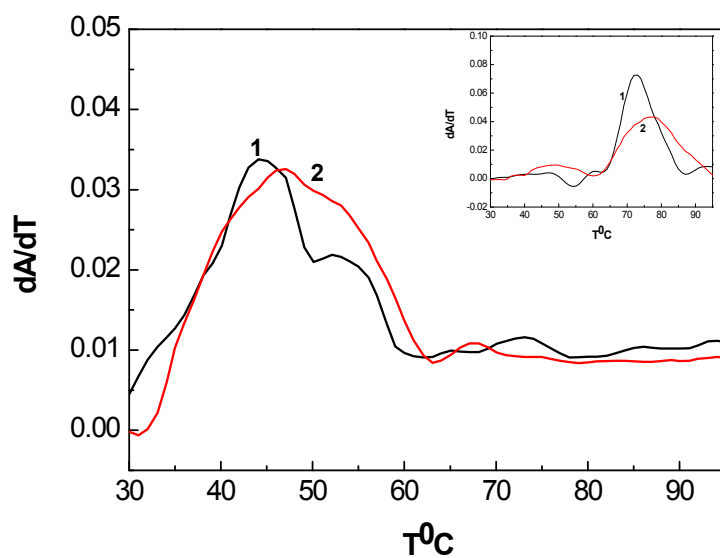


**Fig. S1** Different binding modes of ligand-GQ-DNA interaction.

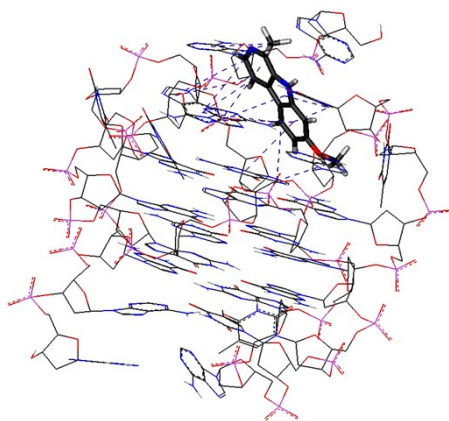




**Fig. S2.** Modified Benesi-Hildebrand plot for (a) Harmine-GQ-DNA absorption, (b) Harmine-B-DNA absorption.



**Figure S3.** Derivative plots of melting profile of G-quadruplex (inset: B-DNA) in the (1) absence and (2) presence of HR as indicated on the graph.



**Fig. S4.** Docked structure of HR-GQ-DNA complex (another representative snapshot in different view of loop binding).

**Table S1.** Comparative FID results for HR.

DNA	DC <sub>50</sub>	Estimated Selectivity(2.5/°C)
B-DNA	> 2.5μM	1.7
GQ-DNA	> 2.5μM	

**Table S2.** Fitted parameters of anisotropy decays of HR in the presence of GQ-DNA and B-DNA.

Sample	Fluorescence Lifetime (ns)	Rotational relaxation time (ps)	Pre-exponential Factor	Rotational relaxation time (ns)	Pre-Exponential Factor(β)	χ <sup>2</sup>
HR	6.50	520	1	-	-	1.04
HR+12 μM GQ-DNA	6.50	539	0.88	5.89	0.12	1.01
HR+ 12 μM B-DNA	6.50	443	0.83	3.36	0.17	1.00