

Parallel folding topology-selective label-free detection and monitoring of conformational and topological changes of different G-quadruplex DNAs by emission spectral changes *via* FRET of *m*PPE-Ala–Pt(II) complex ensemble

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Electronic Supplementary Information

Experimental Section

Materials and Reagents. Polyethylene glycol (PEG) with M_w of 200 (PEG-200), human serum albumin (HSA), spermine, trypsin, lysozyme, poly(tyrosine) with M_w of 10000–40000 (PY), guanine, bovine serum albumin (BSA), poly(L-lysine hydrobromide) with M_w of 15000 (PK) and chondroitin 4-sulphate sodium salt from shark cartilage (CS) were purchased from Sigma-Aldrich Co. Ltd. *c-myc* (5'-TGAGGGTGGGGAGGGTGGGGAA-3'), *bcl-2* (5'-GGGCGCGGGAGGAA-GGGGGCGGG-3'), *c-kit1* (5'-AGGGAGGGCGCTGGGAGGAGGG-3'), human telomeric DNA (5'-TTAGGGTTAGGGTTAGGGTTAGGGTTA-3'), *c-myc-c* (5'-TTCCCCACCCTCCCCACCCTCA-3'), complementary sequence of human telomeric DNA (5'-TAACCCTAACCTAACCTAACCTAA-3') and other poly(nucleotides) were obtained from Tech Dragon Limited. *mPPE-Ala*¹ and **1**² were synthesized as described previously. The molecular weight of the precursor polymer, *mPPE-Ala-OBn*, was determined by organic-phase gel permeation chromatography (GPC). GPC (NMP, polystyrene standards): $M_n = 19901 \text{ g mol}^{-1}$, $M_w = 30143 \text{ g mol}^{-1}$, PDI = 1.51; the number of repeating units was estimated to be 74. All solvents were purified and distilled by standard procedures before use. All other reagents were of analytical grade and were used as received.

Physical Measurements and Instrumentation. UV-Vis absorption spectra were obtained by using a Cary 50 (Varian) spectrophotometer equipped with a Xenon flash lamp. Steady state emission spectra were recorded at room temperature using a Spex Fluorolog-3 Model FL3-211 fluorescence spectrofluorometer equipped with a R2658P photomultiplier tube (PMT) detector. Variable-temperature UV-Vis absorption spectra were obtained by using a Cary 50 (Varian) spectrophotometer equipped with a Xenon

flash lamp, with a single cell peltier thermostat to control the working temperatures in the range of 20 to 96 °C. Time-resolved emission decay profiles were recorded with a Horiba Jobin Yvon Fluorocube based on the time-correlated single photon counting method, using a nanoLED with peak wavelength and pulse duration of 371 nm and < 200 ps respectively as the excitation source. Resonance light scattering (RLS) experiments were performed on Spex Fluorolog-3 Model FL3-211 fluorescence spectrofluorometer with a Xenon flash lamp using a right-angle geometry to a R2658P PMT detector. The excitation and emission monochromator wavelengths were coupled and adjusted to scan simultaneously through the range of 280 to 650 nm. Circular dichroism (CD) measurements were performed on a Jasco (Tokyo, Japan) J-815 CD spectropolarimeter using a quartz CD cell with 10-mm path length. Microquartz cuvettes with 10-mm path length and 2-mm window width were used for UV-vis absorption, emission and CD measurements.

Preparation of G-quadruplex.³ In order to ensure the formation of G-quadruplex structure, *c-myc*, *bcl-2*, *c-kit1* or human telomeric DNA was dissolved in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with desired concentration. The solution was heated at 95 °C for 5 minutes, cooled slowly to room temperature and then incubated at room temperature overnight.

Preparation of Pre-formed Duplex DNA.³ In order to ensure the formation of duplex structure, the complementary DNA sequences (*c-myc* and *c-myc-c*; human telomeric DNA and complementary sequence of human telomeric DNA; poly(dA)₂₅ and poly(dT)₂₅; poly(dG)₂₅ and poly(dC)₂₅) were dissolved in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with desired concentration. The solution was

heated at 95 °C for 5 minutes, cooled slowly to room temperature and then incubated at room temperature overnight.

Preparation of G-quadruplex with Different Volume Percentages of PEG-200.⁴ In order to ensure the formation of G-quadruplex structure, *c-myc* or human telomeric DNA was dissolved in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with desired concentration. The solution was heated at 95 °C for 5 minutes, cooled slowly to room temperature and then incubated at room temperature overnight. PEG-200 of appropriate amount was added and the resultant mixture was further incubated at 37 °C for 4 hours before measurement.

Preparation of *mPPE-Ala-1* Ensemble. Mixtures of 500 μ L buffer solution (50 mM Tris-HCl, 50 mM KCl, pH 7.4), 664 μ L of 0.38 mM of *mPE-Ala* and water of appropriate amount were added to reach a final volume of 5 mL. The resultant mixture was sonicated at ambient temperature for 15 minutes. 80 μ L of 0.55 mM of **1** was added to 2 mL of *mPPE-Ala* solution and the resultant mixture was further incubated at ambient temperature for 15 minutes before measurement. All concentrations were calculated as the final concentration in the solution mixture, with buffer solution of 5 mM Tris-HCl, 50 mM KCl, pH 7.4, 50 μ M of *mPE-Ala* and 20 μ M of **1**.

UV Melting Study of **1 on *c-myc*, *bcl-2*, *c-kit1* or Human Telomeric DNA.**^{5,6} The electronic absorption spectra of *c-myc*, *bcl-2*, *c-kit1* or human telomeric DNA (3 μ M) in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the absence and in the presence of **1** (3 μ M) were monitored at different temperatures respectively. The

melting temperatures of the G-quadruplex structures, T_m , were determined graphically from the plots of absorbance against temperature.⁵

Assay Procedure for Measurement of G-quadruplex. Mixtures of 500 μL buffer solution (50 mM Tris-HCl, 500 mM KCl, pH 7.4), 1328 μL of 0.38 mM of *mPE-Ala* and water of appropriate amount were added to reach a final volume of 5 mL. The resultant mixture was sonicated at ambient temperature for 15 minutes. 80 μL of 1.10 mM of **1** was added to 2 mL of *mPPE-Ala* solution and the resultant mixture was incubated at ambient temperature for 15 minutes. G-quadruplex DNA, ensemble solution, buffer solution and water of appropriate amounts were added to reach the final volume of 400 μL and the resultant mixture was further incubated at ambient temperature for 15 minutes before measurement. All concentrations were calculated as the final concentration in the assay solution mixture, with buffer solution of 5 mM Tris-HCl, 50 mM KCl, pH 7.4, 50 μM of *mPE-Ala* and 20 μM of **1**. Emission spectra were recorded with an excitation wavelength of 299 nm at ambient temperature and corrected for PMT response.

Assay Procedure for Selectivity Analysis. The assay procedure was similar to that for the measurement of *c-myc* except that *c-myc* was replaced by same concentration (5 μM) of interfering substrate in aqueous buffer solution.

Assay Procedure for Competition Analysis. The assay procedure was similar to that for the measurement of selectivity analysis except that *c-myc* was added to the ensemble solution in aqueous buffer solution followed by incubation for another 15 minutes prior to the measurement.

Assay Procedure for Measurement of G-quadruplex in Molecular Crowding Conditions. The assay procedure was similar to that for the measurement of G-quadruplex DNA except that the G-quadruplex DNA solution with different volume

percentages of PEG-200 was mixed with the ensemble solution in aqueous buffer solution.

Determination of Binding Constant of **1 with *c-myc*.**⁷ **1** (50 μM) in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) was titrated with different concentrations of *c-myc*. The binding constants of **1** on *c-myc*, K_b , were determined from the Scatchard equation,⁷ which states that:

$$\frac{D}{\Delta\varepsilon_{\text{app}}} = \frac{D}{\Delta\varepsilon} + \frac{1}{(\Delta\varepsilon)K_b}$$

where D is the concentration of the base pairs of *c-myc*, $\Delta\varepsilon_{\text{app}} = [\varepsilon_A - \varepsilon_F]$ and $\Delta\varepsilon = [\varepsilon_B - \varepsilon_F]$. ε_A is calculated from the observed absorbance divided by the concentration of **1** (50 μM), while ε_B and ε_F correspond to the molar extinction coefficients of the *c-myc*-**1** adduct and the unbound **1**, respectively. By the plot of $D/\Delta\varepsilon_{\text{app}}$ against D , $\Delta\varepsilon$ and K_b can be found from the slope and the y-intercept of the graph respectively.

Determination of Detection Limit of *m*PPE-Ala-1** Ensemble on *c-myc*.**⁸ The detection limit was determined from the $3\sigma/m$ method based on the linear part of the calibration curve, where σ is the standard deviation of the blank emission, which was obtained from a series of five independent measurements, and m is the calibration sensitivity, which was determined from the slope of the linear plot.

References

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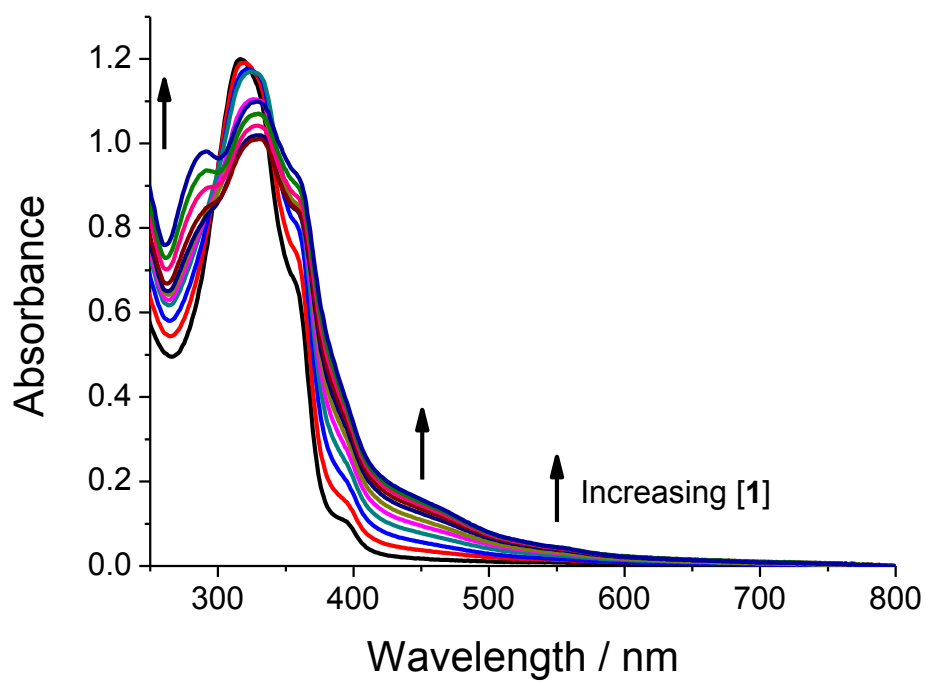


Fig. S1 Electronic absorption spectral changes of *mPPE-Ala* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with increasing concentration of **1** in the range of 0 to 50 μM . Concentration of *mPE-Ala* was 50 μM .

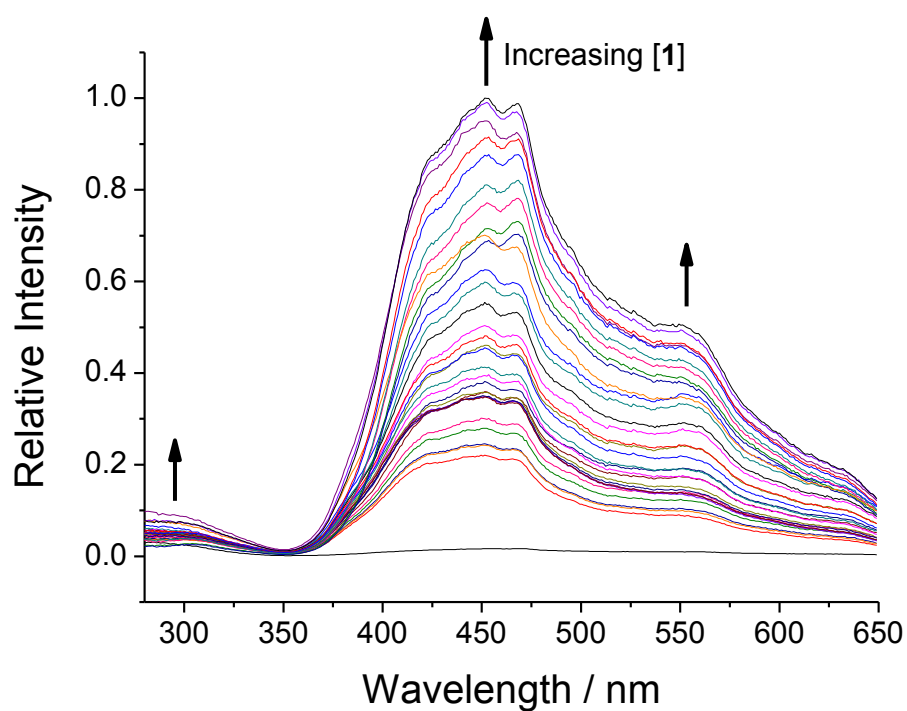


Fig. S2 Resonance light scattering spectra of *mPPE-Ala* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with increasing concentration of **1** in the range of 0 to 50 μM . Concentration of *mPE-Ala* was 50 μM . The black line represents the signal of aqueous buffer solution.

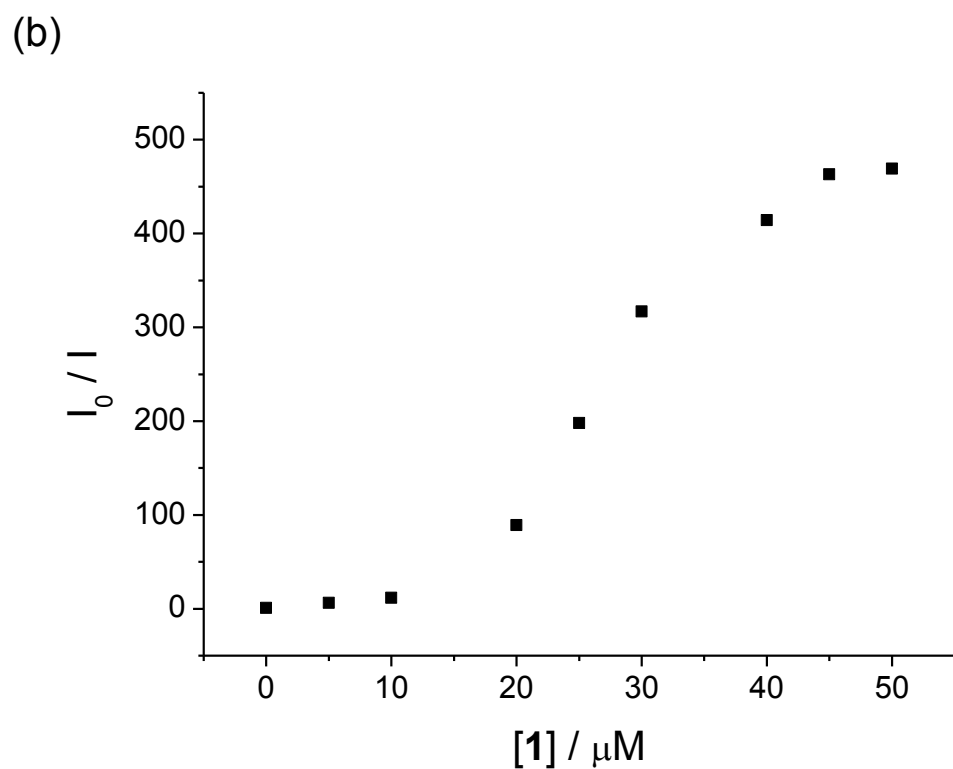
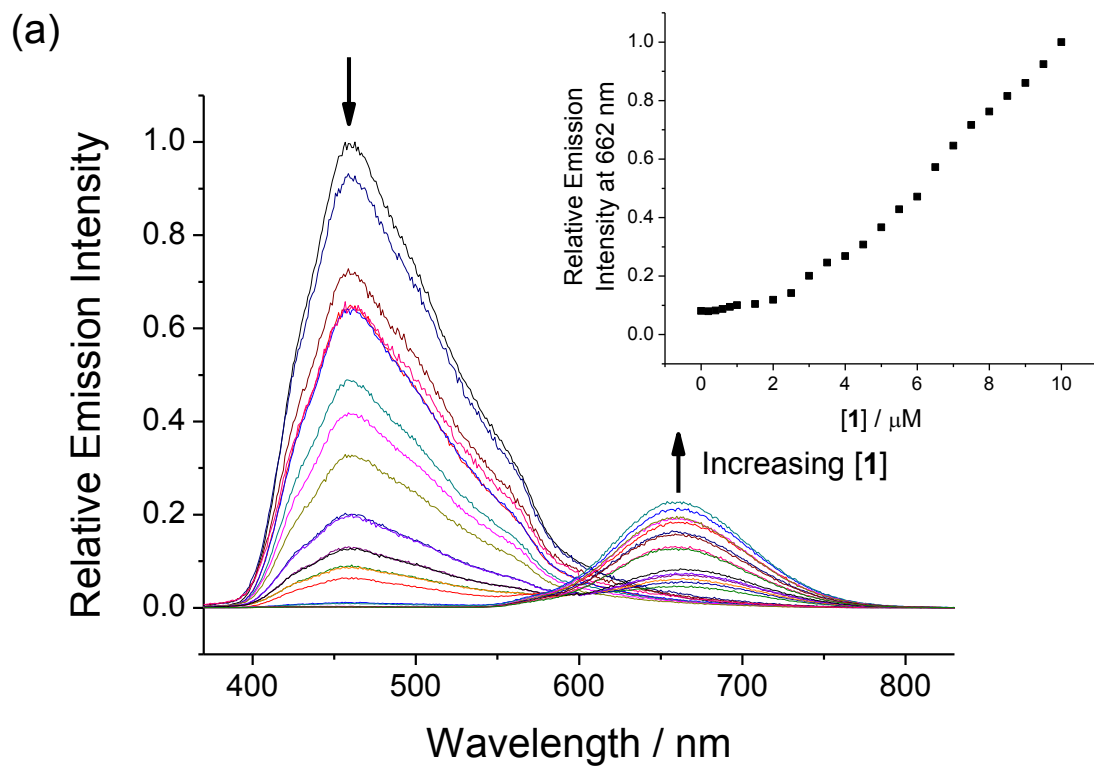


Fig. S3 (a) Emission spectral changes of *m*PPE-Ala in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with increasing concentration of **1**. Inset: a plot of relative emission intensity at 662 nm *versus* concentration of

1. (b) Stern–Volmer plot for the quenching of *m*PPE-Ala by **1** in aqueous buffer solution. Concentration of *m*PE-Ala was 50 μ M. Excitation was at 299 nm. I_0 and I are the emission intensity at 461 nm without **1** and with different concentrations of **1**, respectively.

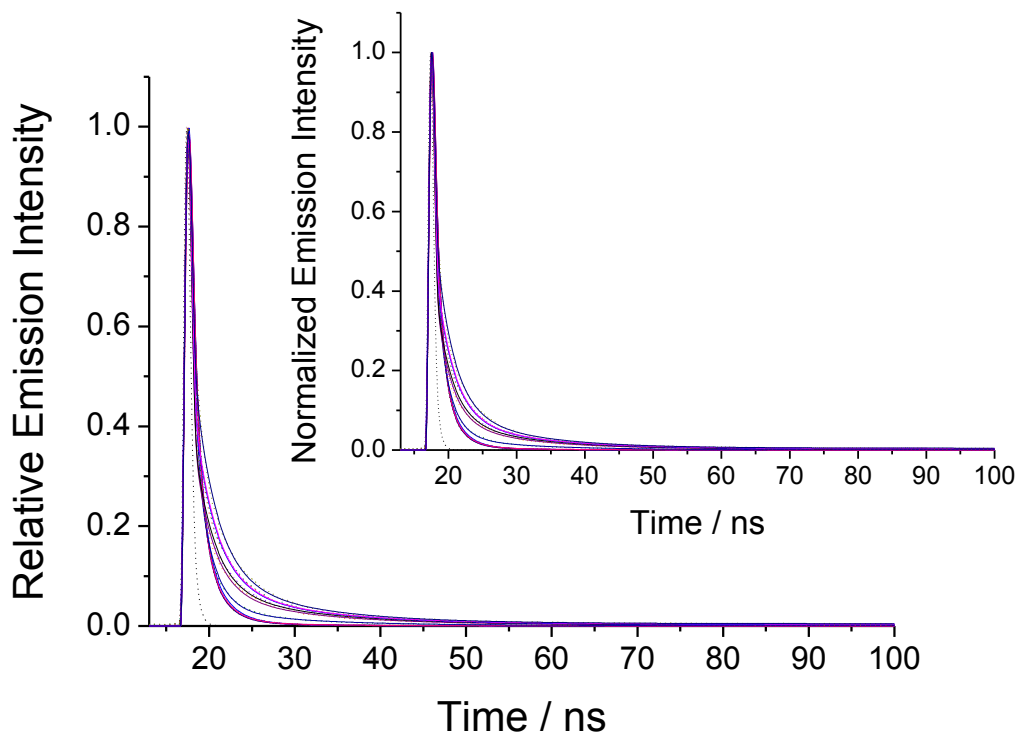


Fig. S4 Time-resolved emission decay profiles of *mPPE-Ala* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with different concentrations of **1** in the range of 0 to 50 μM . Inset: Normalized time-resolved emission decay profiles of *mPPE-Ala* with different concentrations of **1** in the range of 0 to 50 μM . Concentration of *mPE-Ala* was 50 μM . Excitation was at 371 nm and emission signals were monitored at 461 nm. The prompt signal is shown in black squares without any line fitting.

Table S1. Parameters obtained from steady-state emission spectra and time-resolved emission decay profiles of *m*PPE-Ala in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with different concentrations of **1**^a

[1] / μM	τ_1^b / ns		τ_2^b / ns		τ_3^b / ns		$K_{\text{SV}} \times 10^{-5c}$ / M^{-1}
0	9.07	5%	2.10	45%	0.71	50%	–
5	9.55	4%	2.03	43%	0.72	53%	6.42
10	9.27	4%	1.96	45%	0.69	51%	11.75
15	10.41	5%	1.98	46%	0.66	49%	– ^d
20	10.47	19%	1.82	46%	0.37	35%	89.09
25	10.86	32%	2.17	35%	0.16	33%	197.80
30	11.35	34%	2.31	34%	0.16	32%	317.00
35	11.64	34%	2.53	39%	0.18	27%	– ^d
40	11.37	34%	2.52	40%	0.17	26%	414.22
45	11.18	37%	2.57	42%	0.19	21%	462.93
50	– ^d	– ^d	– ^d	– ^d	– ^d	– ^d	469.07

^a Concentration of *m*PE-Ala was 50 μM .

^b τ_1 , τ_2 and τ_3 are the fluorescence lifetimes of the triexponential decay obtained from the time-resolved emission measurements.

^c K_{SV} is the Stern–Volmer quenching constant obtained from the steady-state emission measurements.

^d Not determined.

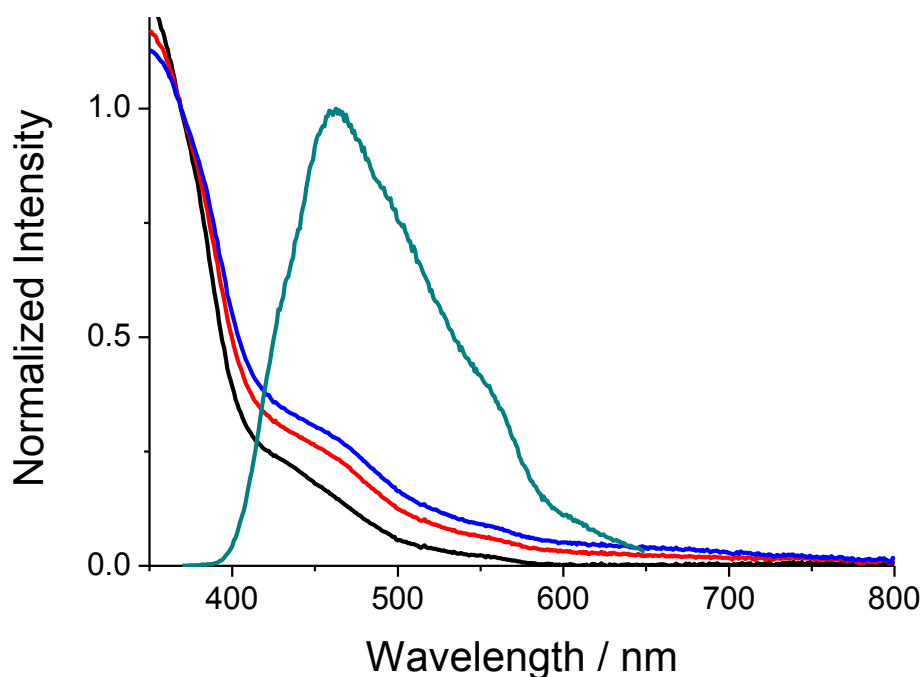


Fig. S5 Normalized electronic absorption spectra of **1** and emission spectrum of *mPPE-Ala* showing the spectral overlap between the electronic absorption spectra of **1** with the emission spectrum of *mPPE-Ala*. The electronic absorption spectra of **1** in the absence of polyelectrolyte (black), in the presence of poly(sodium *p*-styrenesulfonate) (red) with 25 μM sulfonate concentration and in the presence of poly(sodium *p*-styrenesulfonate) (blue) with 50 μM sulfonate concentration were normalized at 369 nm, while the emission spectrum of *mPPE-Ala* (dark cyan) with 50 μM of *mPPE-Ala* was normalized at 461 nm. All the spectra were recorded in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Electronic absorption spectra of aggregated **1** were recorded using poly(sodium *p*-styrenesulfonate) instead of *mPPE-Ala* in order to prevent the interference of the electronic absorption of **1** by the conjugated polymer.

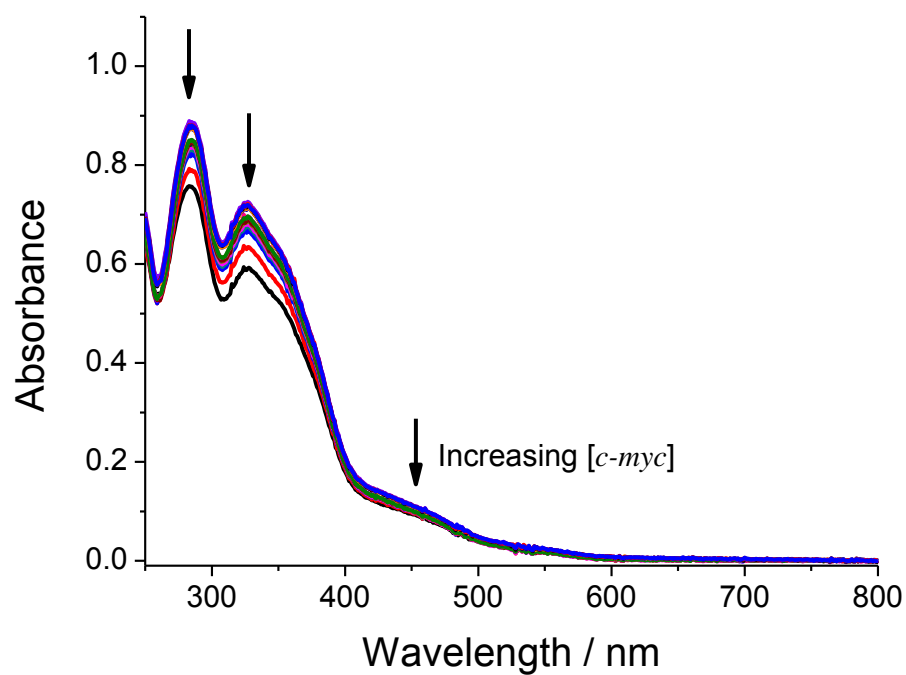


Fig. S6 Electronic absorption spectral changes of **1** with increasing concentration of *c-myc* in the range of 0 to 5 μM in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentration of **1** was 50 μM .

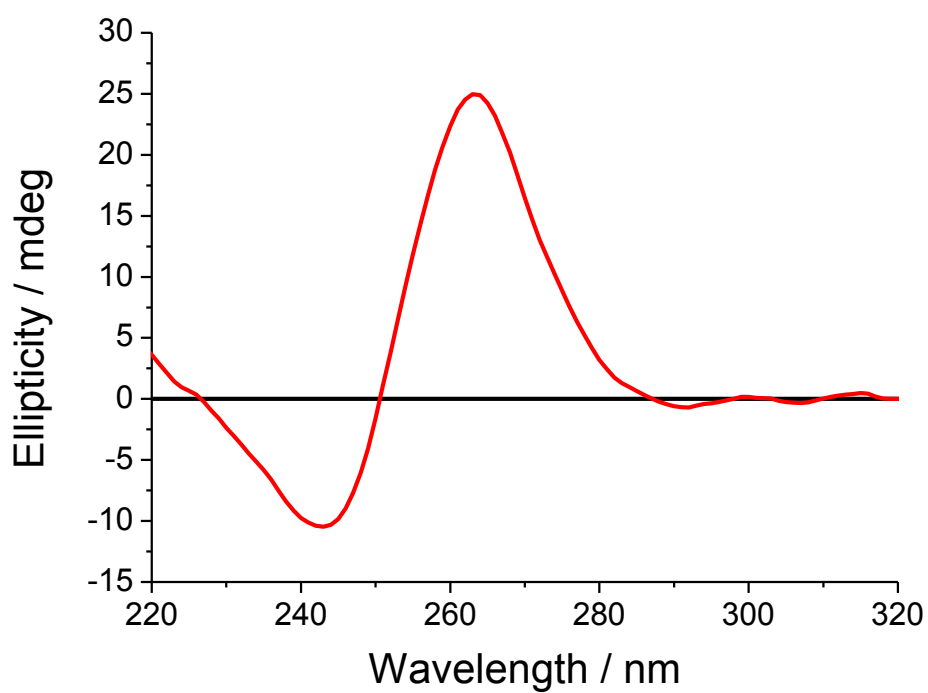


Fig. S7 CD spectrum of *c-myc* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentration of *c-myc* was 3 μM. The black line represents the signal of aqueous buffer solution.

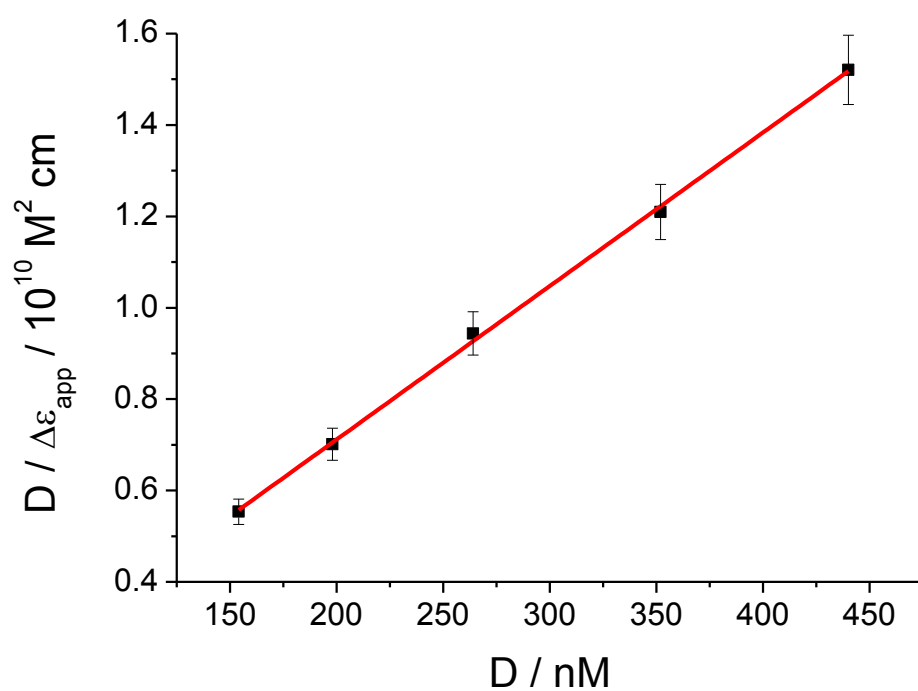


Fig. S8 A plot of $D/\Delta\epsilon_{\text{app}}$ against D for the determination of the binding constant of **1** with *c-myc*. D is the concentration of the base pairs of *c-myc* and $\Delta\epsilon_{\text{app}} = [\epsilon_{\text{A}} - \epsilon_{\text{F}}]$, where ϵ_{A} is calculated from the observed absorbance at 328 nm divided by the concentration of **1** (50 μM) and ϵ_{F} is the molar extinction coefficient of unbound **1** at 328 nm.

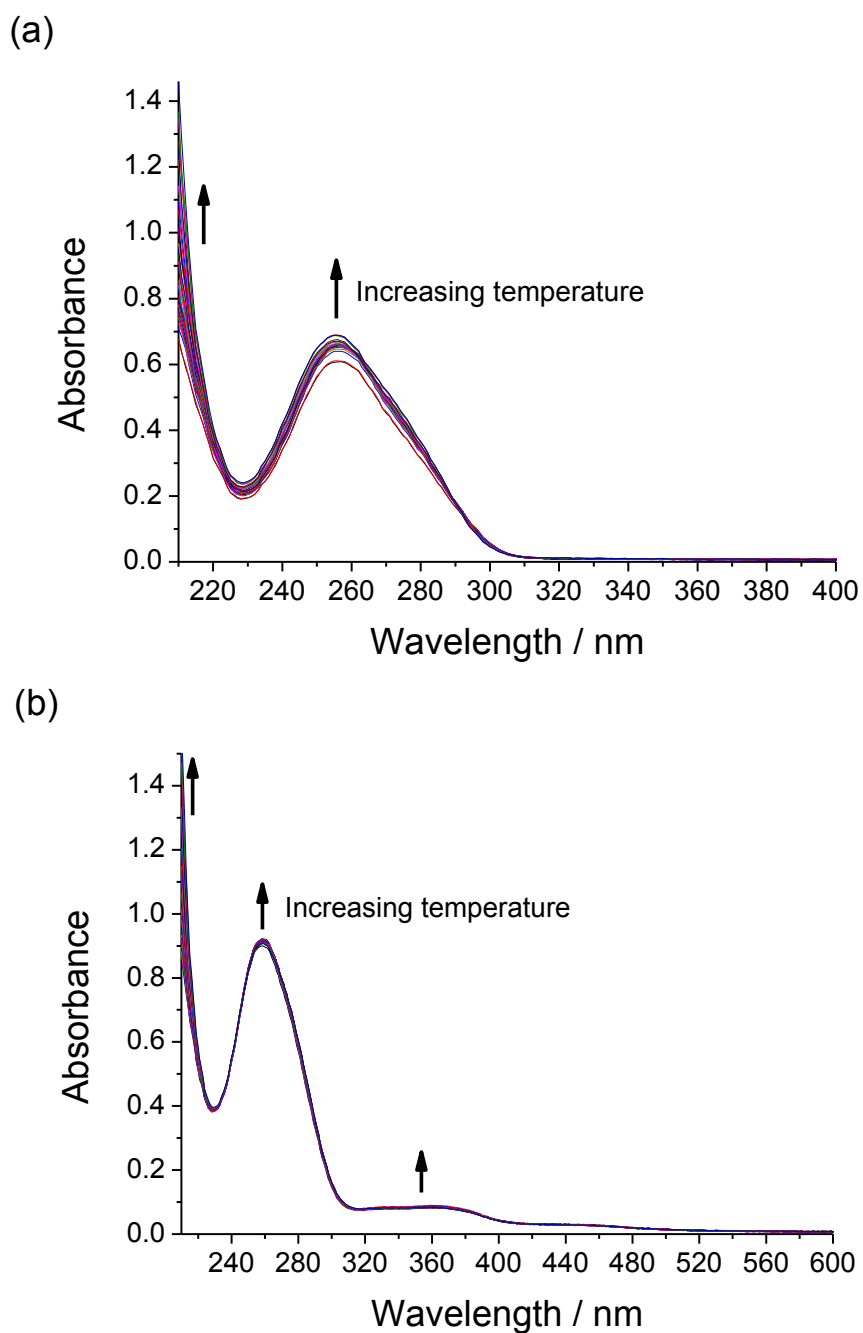


Fig. S9 Electronic absorption spectral changes of *c-myc* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with increasing temperature in the range of 20 to 96 °C (a) in the absence and (b) in the presence of **1**. Concentrations of *c-myc* and **1** were both 3 μ M.

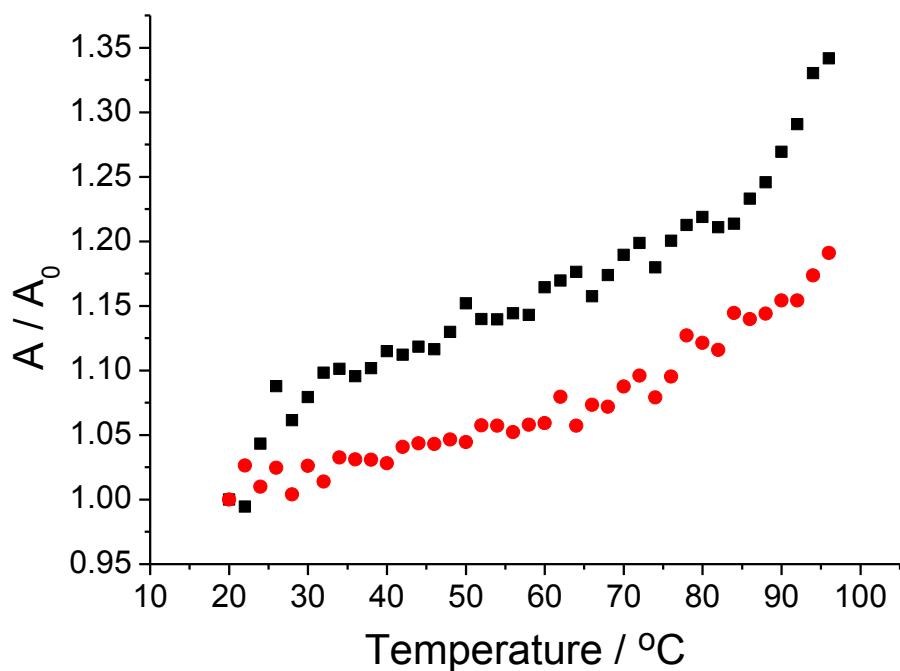


Fig. S10 UV melting profile of *c-myc* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the absence (■) and in the presence (●) of **1**. A_0 and A are the absorbance at 218 nm at 20 °C and at different temperatures respectively. The absorption change was monitored at 218 nm, which is the isosbestic point in the absorption spectra of **1** at different temperatures. Concentrations of *c-myc* and **1** were both 3 μ M.

Table S2. Folding topologies and UV melting temperatures of the G-quadruplex DNAs, T_m , in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the absence and in the presence of **1**^a

Guanine-rich G-quadruplex-forming DNA	G-quadruplex folding topology in dilute solution	T_m in the absence of 1 ^b / °C	T_m in the presence of 1 ^b / °C
<i>c-myc</i>	Intramolecular propeller parallel	91	> 96 ^c
<i>bcl-2</i>	Hybrid	75.5	77.5
<i>c-kit1</i>	Snap-back parallel	77	77
Human telomeric DNA	Mixed-hybrid	69.5	75.5

^a Concentrations of G-quadruplex DNAs and **1** were both 3 μ M.

^b UV-vis absorption changes were monitored at 218 nm.

^c The exact value cannot be determined under experimental conditions as T_m should be close to or even higher than the boiling point of water.

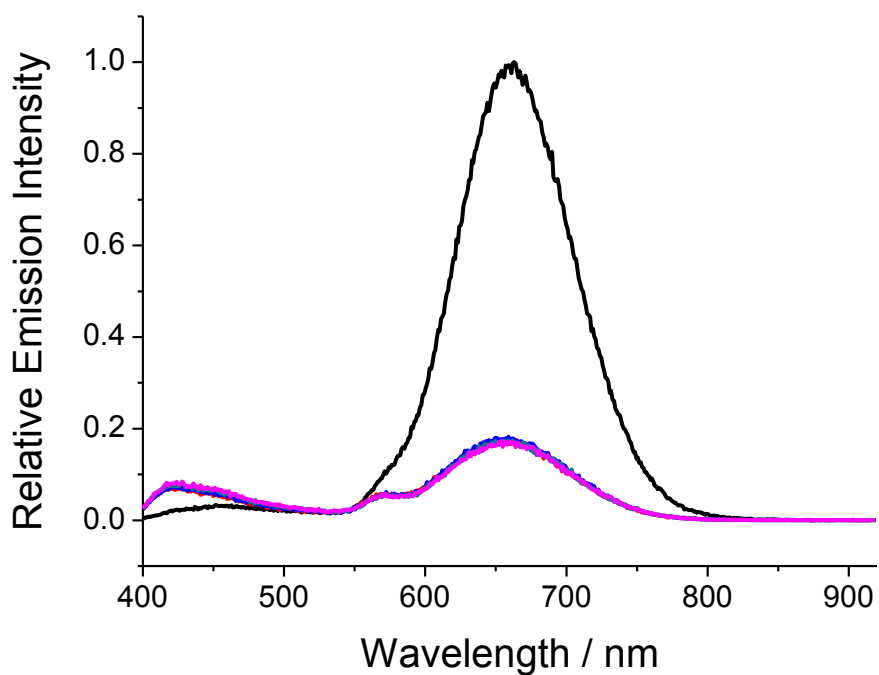
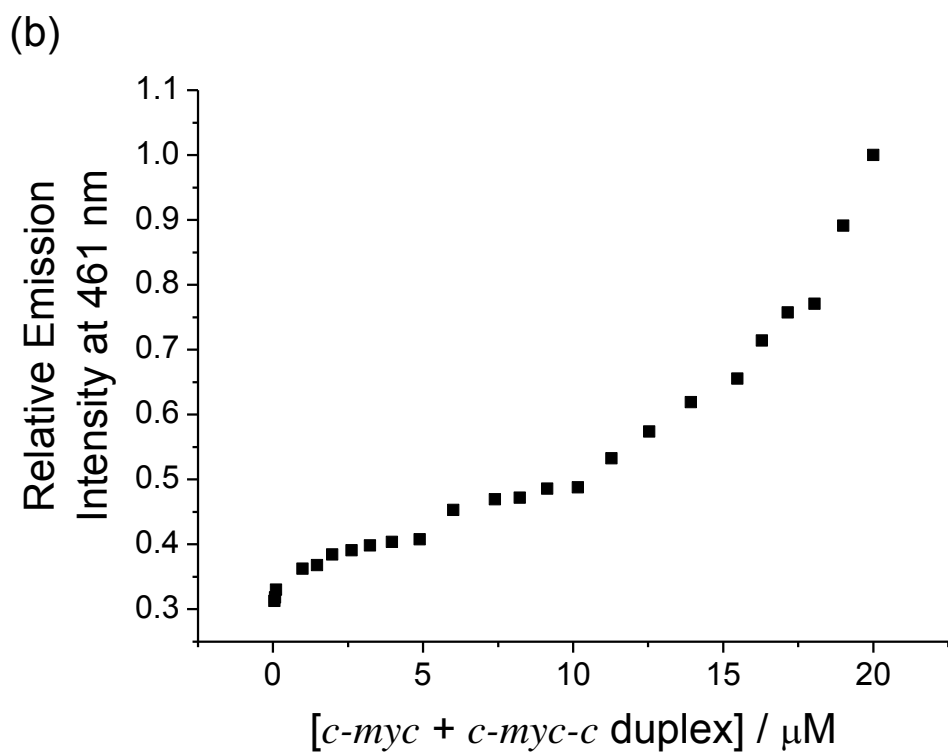
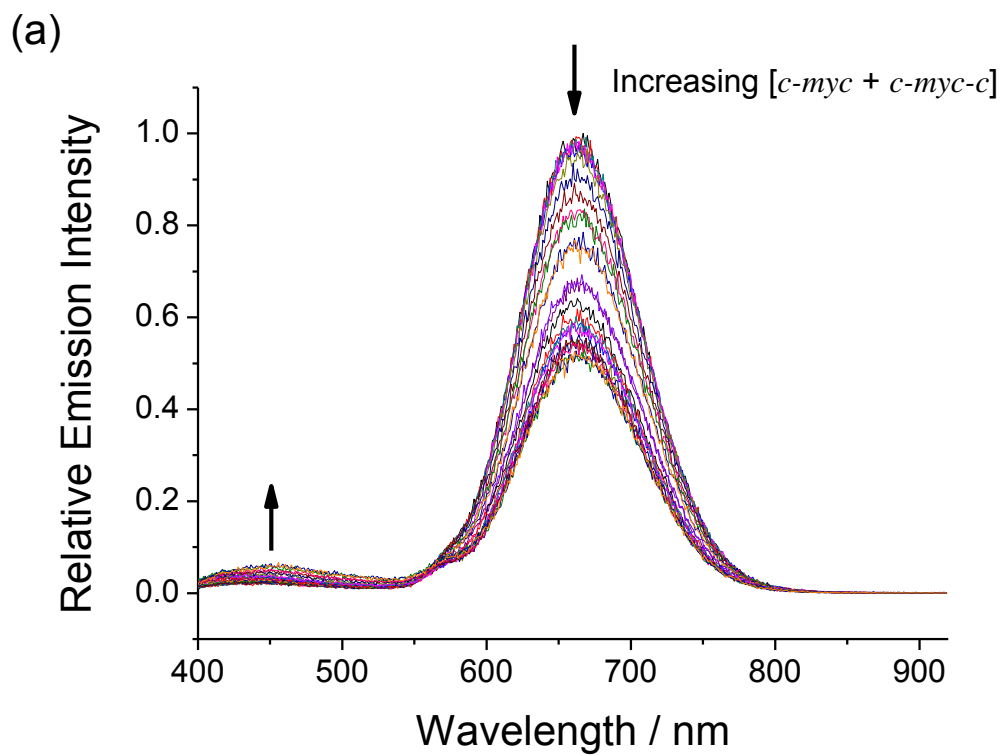


Fig. S11 Emission spectral changes of *mPPE-Ala* and **1** in the absence (black) and in the presence of 5 μM of *c-myc* (red), 5 μM of *c-myc* and 1.67 μM of *c-myc-c* (blue), 5 μM of *c-myc* and 3.33 μM of *c-myc-c* (dark cyan) and 5 μM of *c-myc* and 5 μM of *c-myc-c* (magenta) in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.



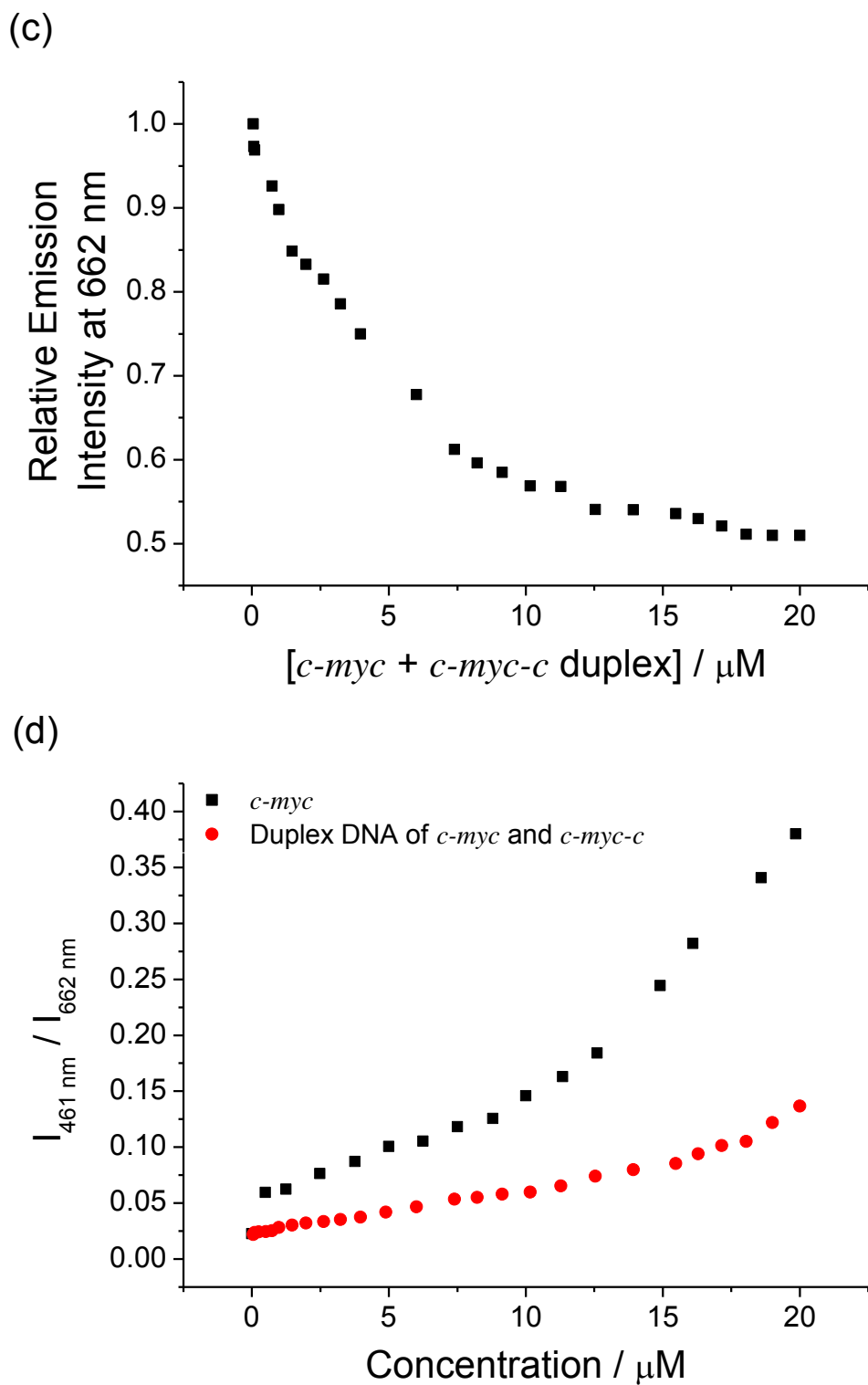


Fig. S12 (a) Emission spectral changes of *mPPE-Ala* and **1** with increasing concentration of pre-formed duplex of *c-myc* and *c-myc-c* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Relative emission intensity of *mPPE-Ala* and **1** at (b) 461 nm and (c) 662 nm with increasing

concentration of pre-formed duplex of *c-myc* and *c-myc-c*. (d) Ratiometric emission of *mPPE-Ala* and **1** at 461 and 662 nm with increasing concentrations of *c-myc* (■) and pre-formed duplex of *c-myc* and *c-myc-c* (●). Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

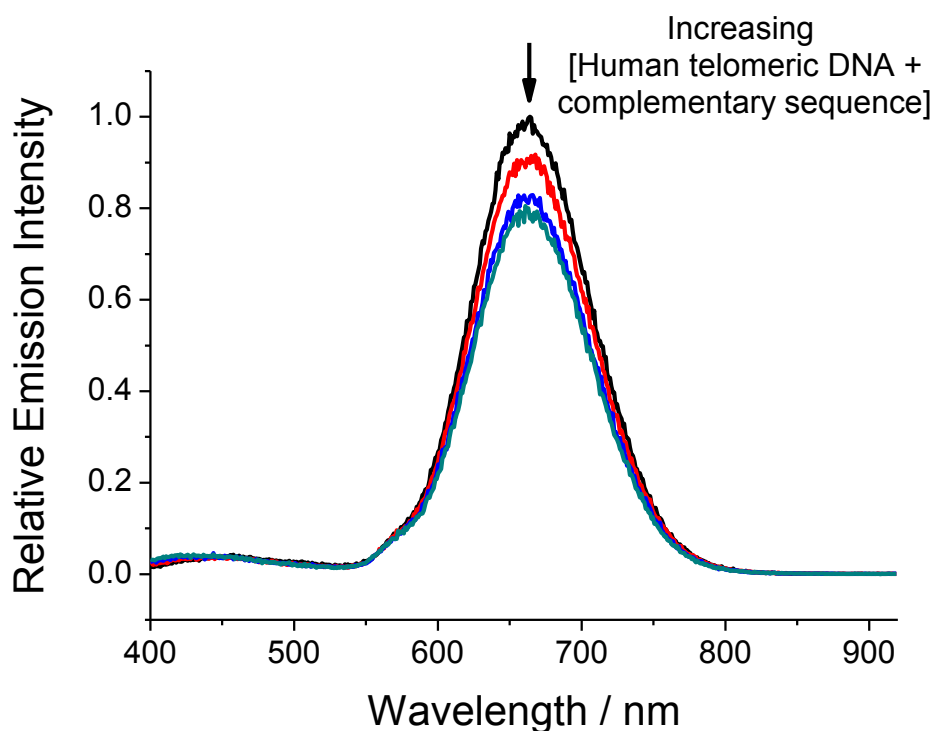


Fig. S13 Emission spectral changes of *mPPE-Ala* and **1** in the absence (black) and in the presence of pre-formed duplex DNA with 1.67 μM human telomeric DNA and 1.67 μM complementary sequence of human telomeric DNA (red), pre-formed duplex DNA with 3.33 μM human telomeric DNA and 3.33 μM complementary sequence of human telomeric DNA (blue) and pre-formed duplex DNA with 5 μM human telomeric DNA and 5 μM complementary sequence of human telomeric DNA (dark cyan) in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

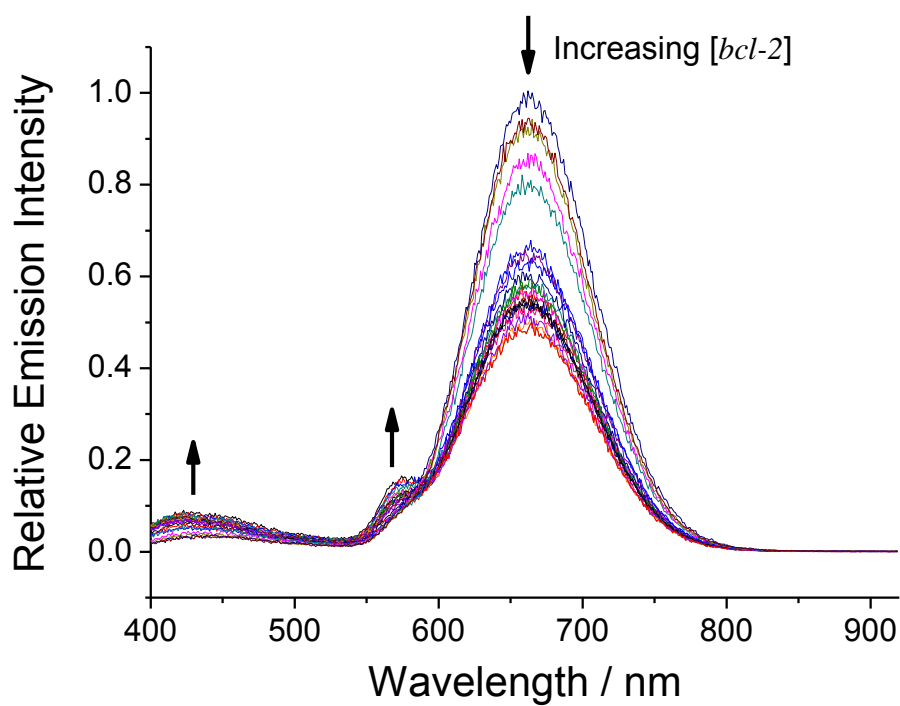


Fig. S14 Emission spectral changes of *mPPE-Ala* and **1** with increasing concentration of *bcl-2* in the range of 0 to 25 μM in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

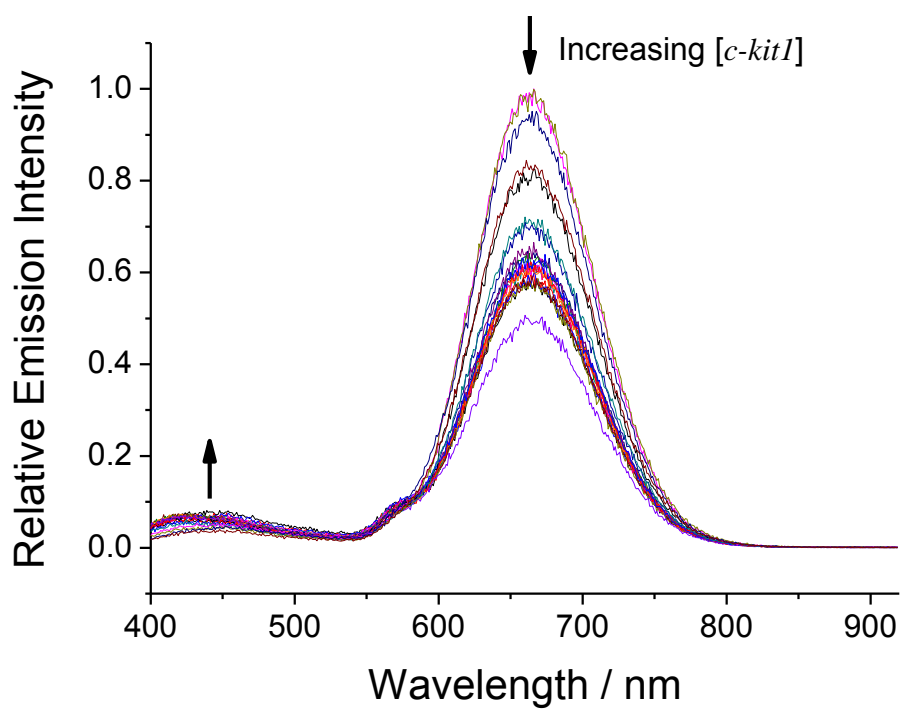


Fig. S15 Emission spectral changes of *mPPE-Ala* and **1** with increasing concentration of *c-kit1* in the range of 0 to 25 μM in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

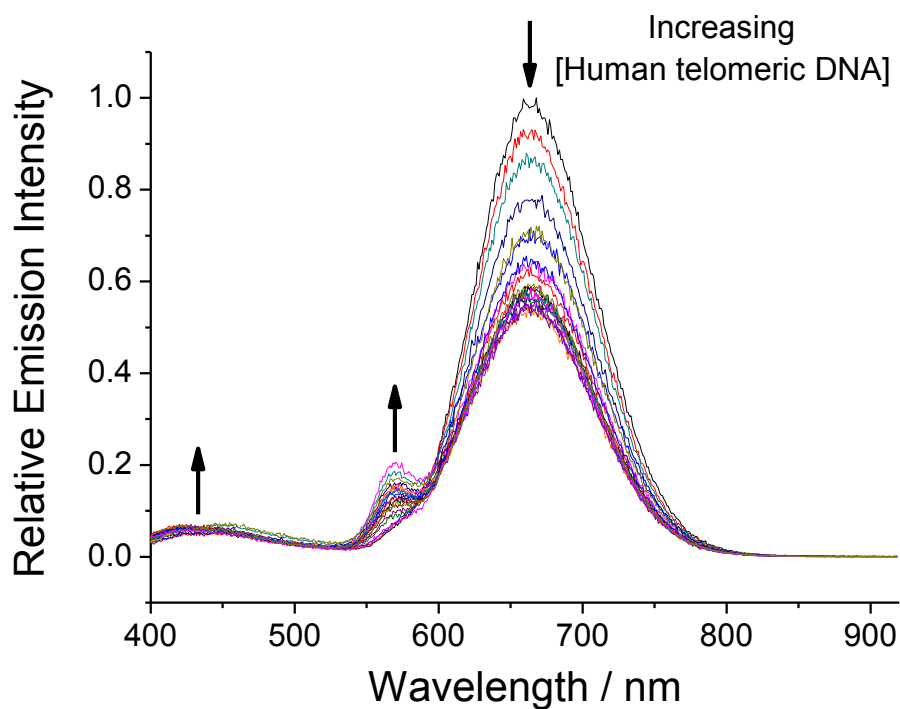


Fig. S16 Emission spectral changes of *mPPE-Ala* and **1** with increasing concentration of human telomeric DNA in the range of 0 to 25 μM in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentrations of *mPPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

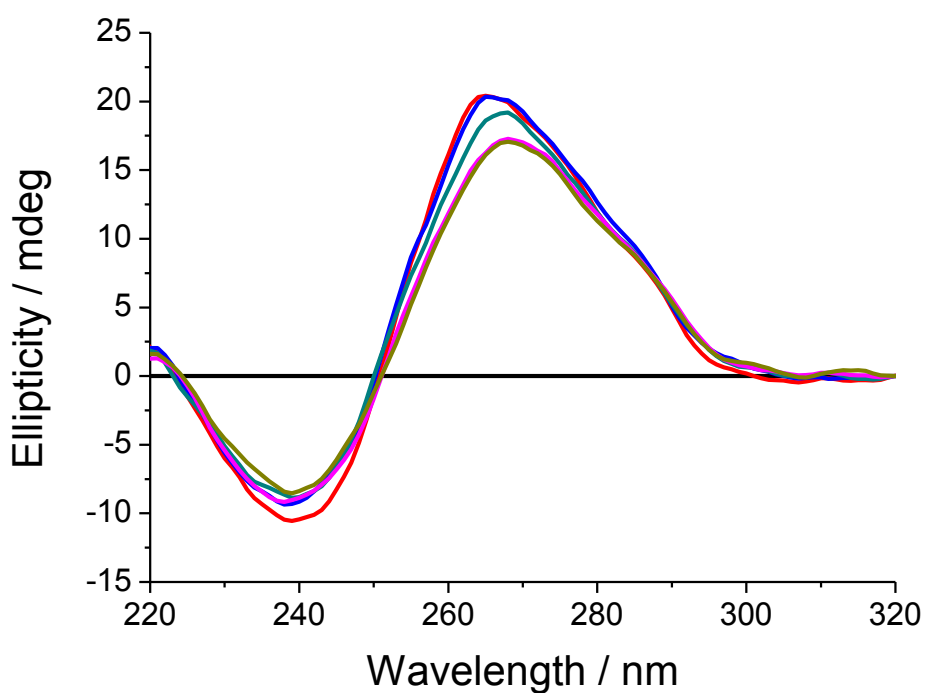
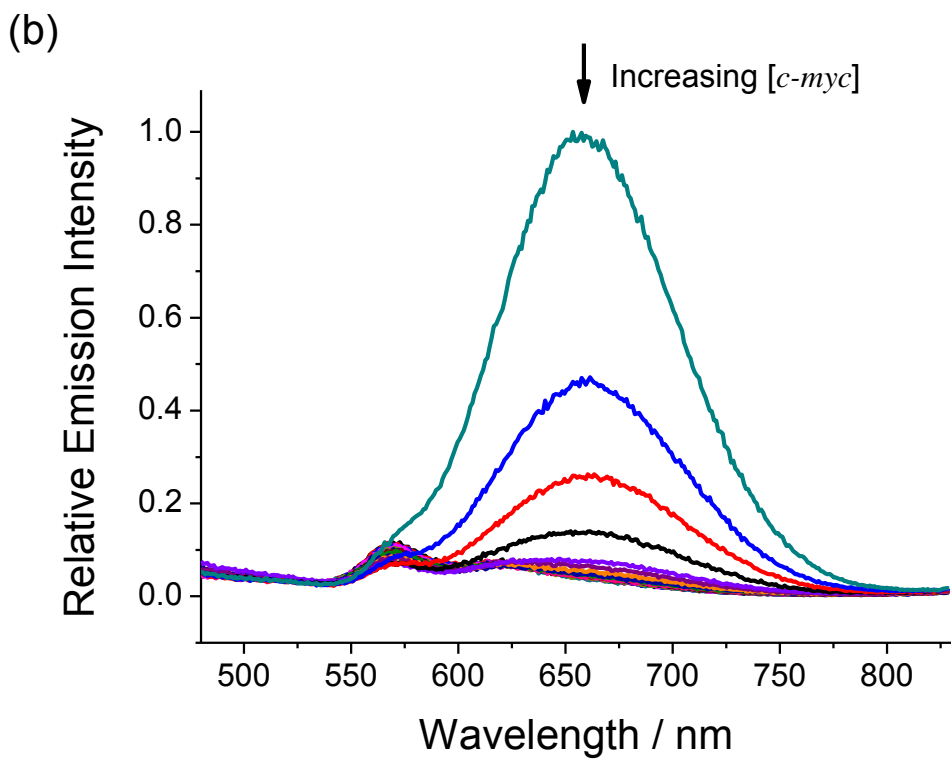
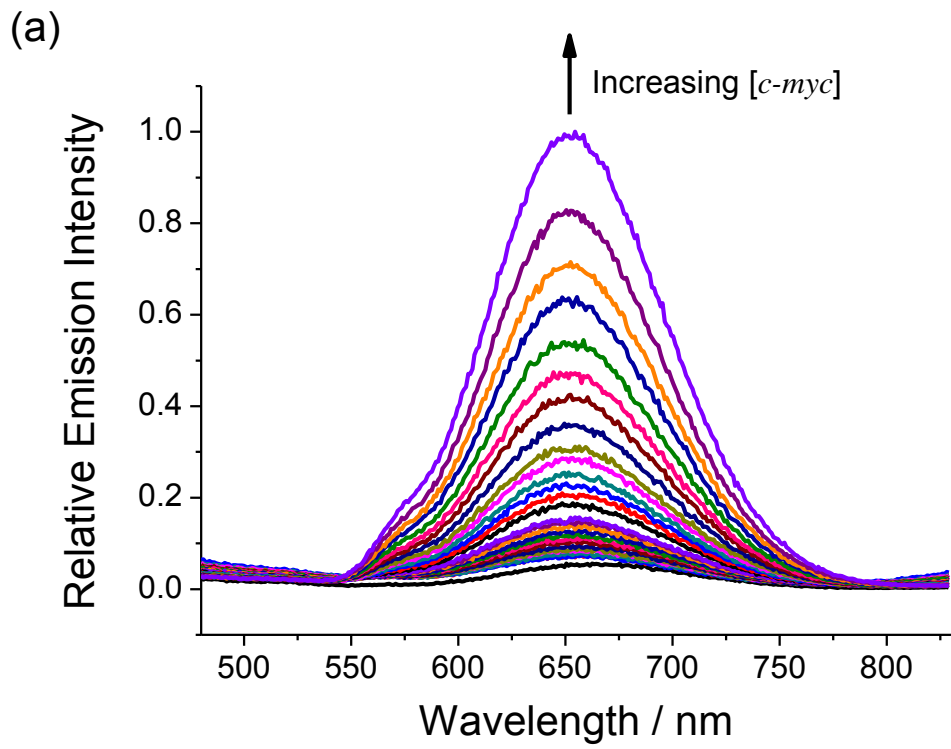
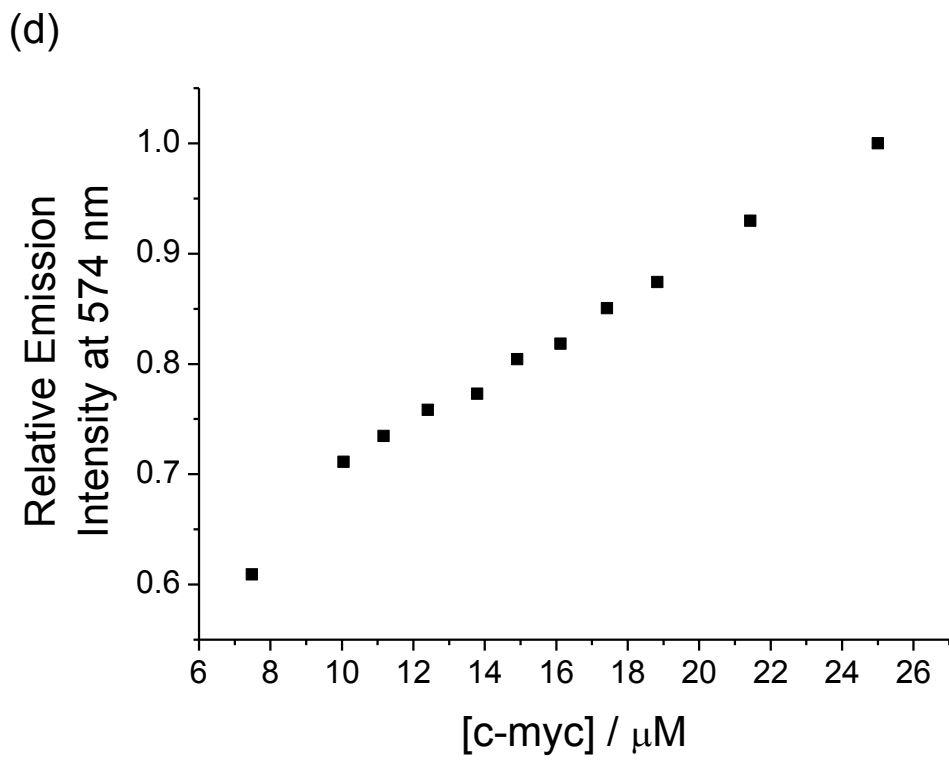
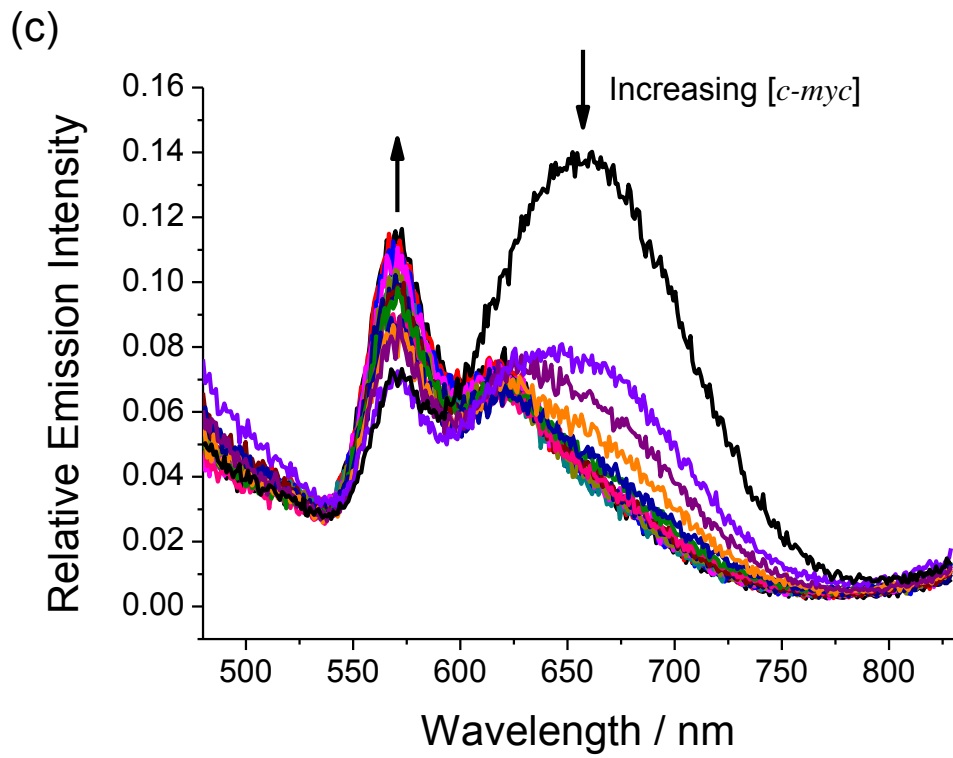


Fig. S17 CD spectra of pre-formed duplex of *c-myc* and *c-myc-c* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the absence (red) and in the presence of 1 μM (blue), 2 μM (dark cyan), 3 μM (magenta) and 4 μM (dark yellow) of **1**. Concentrations of *c-myc* and *c-myc-c* were both 3 μM . The black line represents the signal of aqueous buffer solution.





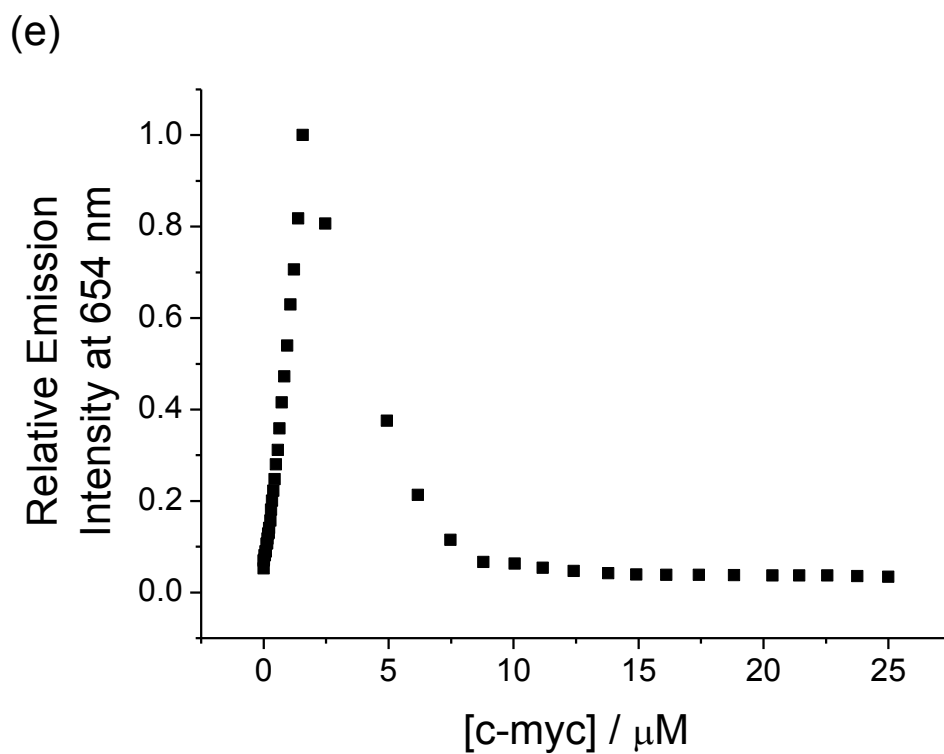


Fig. S18 Emission spectral changes of **1** in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) with (a) 0–1.57 μM , (b) 1.57–25 μM and (c) 7.48–25 μM of *c-myc*. Relative emission intensity of **1** at (d) 574 nm and (e) 654 nm with increasing concentration of *c-myc* in aqueous buffer solution. Concentration of **1** was 20 μM . Excitation was at 299 nm.

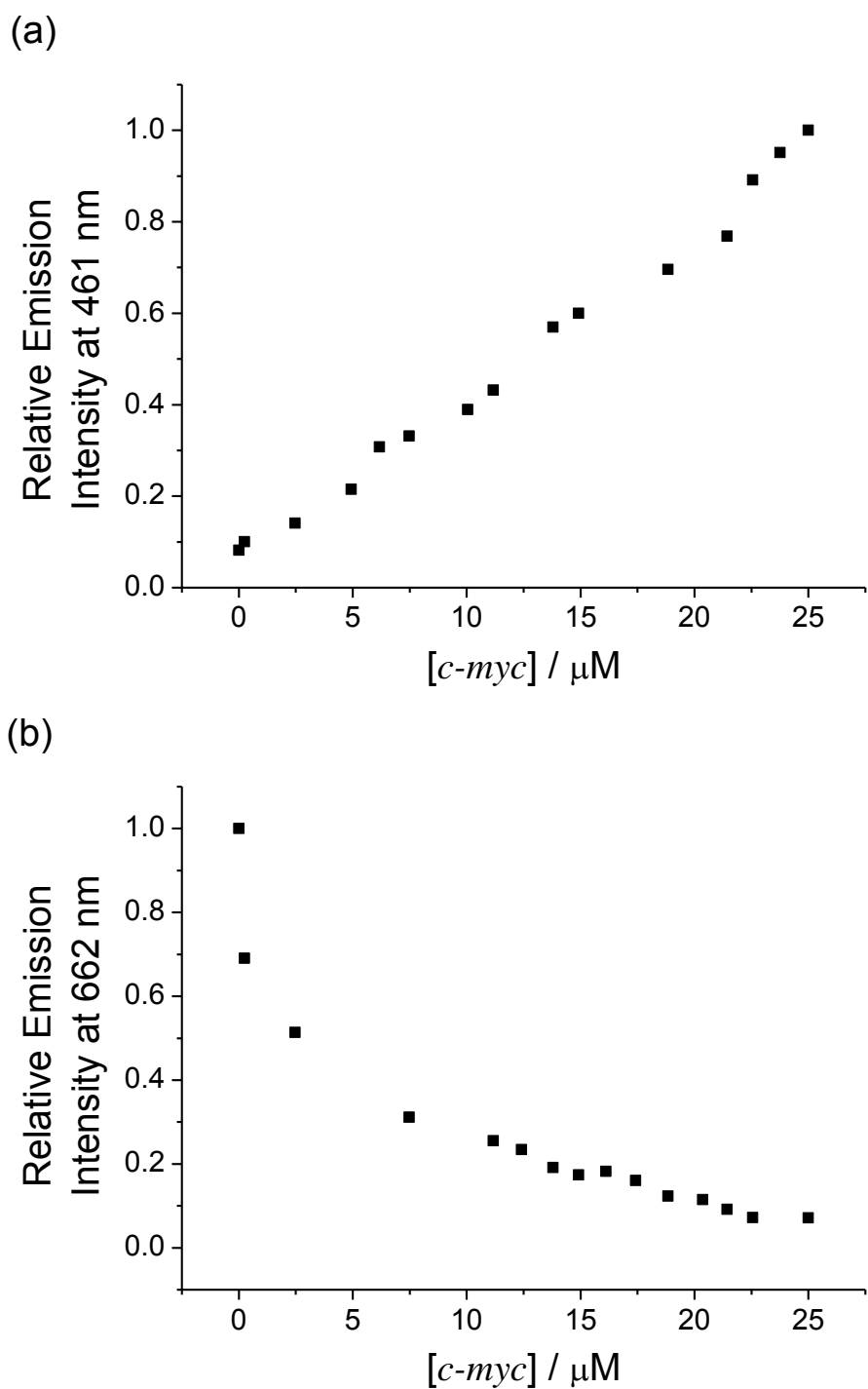
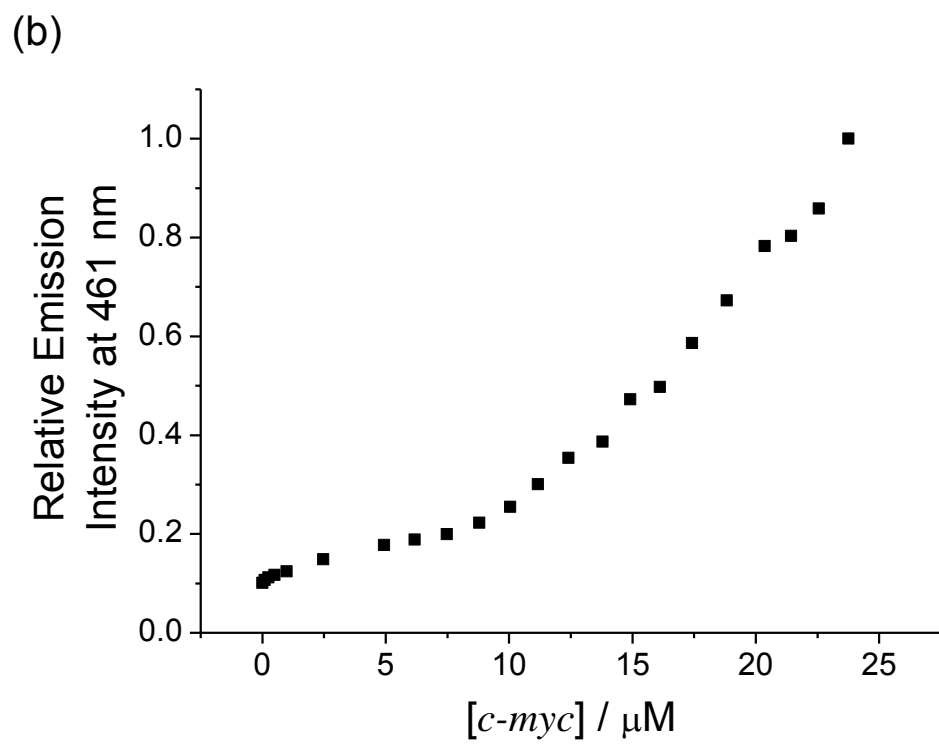
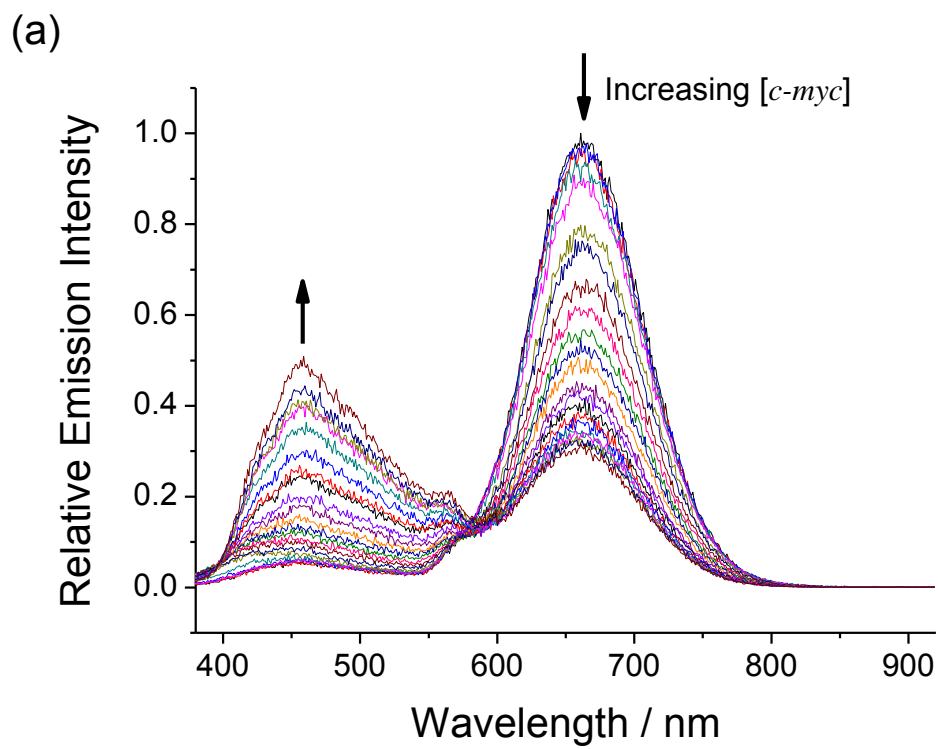


Fig. S19 Relative emission intensity of *m*PPe-Ala and **1** at (a) 461 nm and (b) 662 nm with increasing concentration of *c-myc* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the presence of 40 % (v/v) PEG-200. Concentrations of *m*PE-Ala and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.



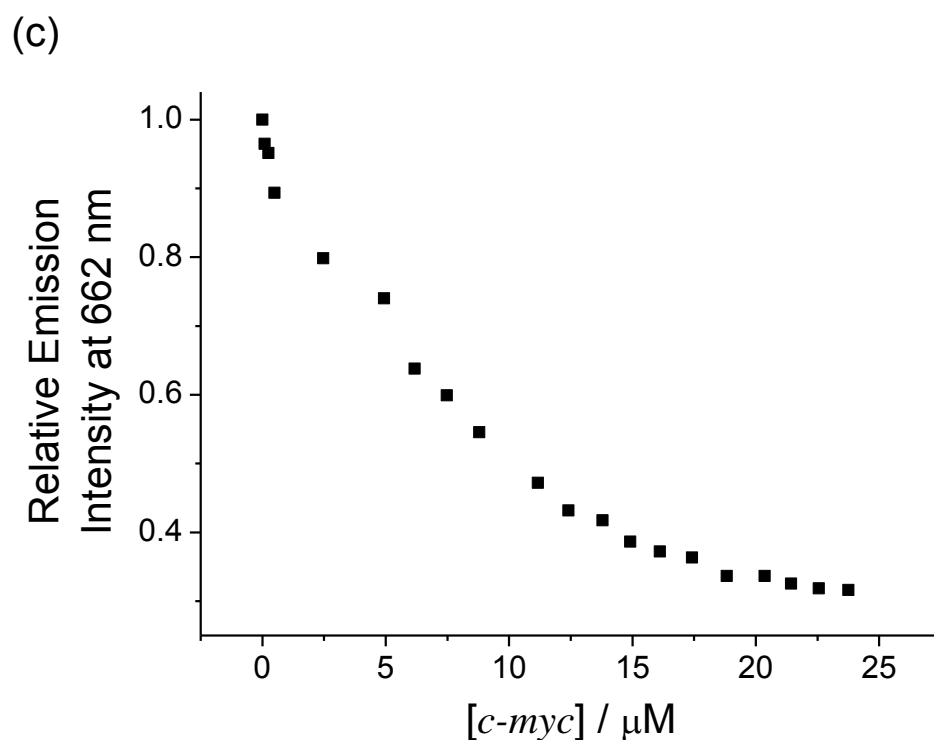
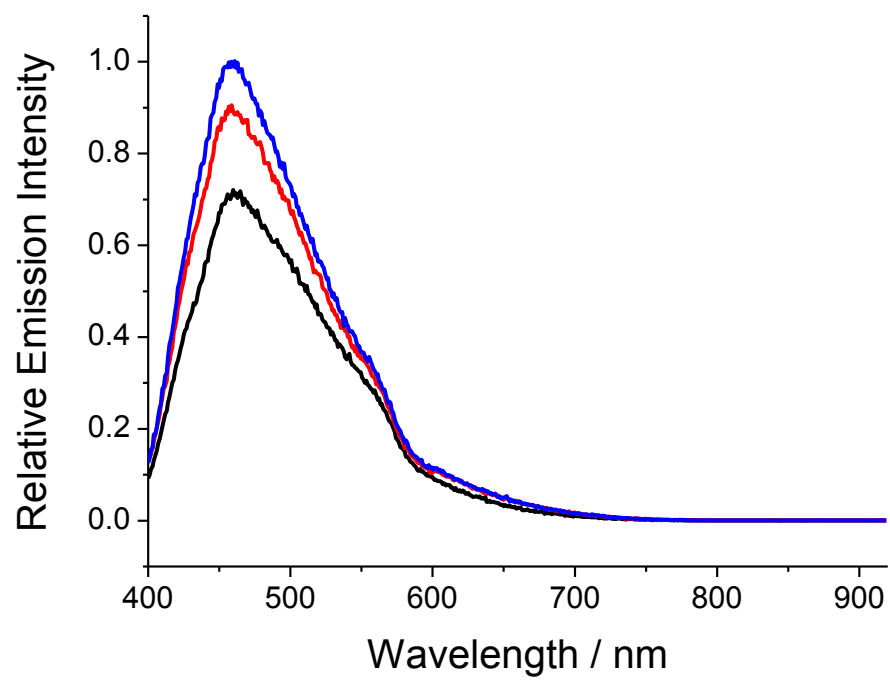
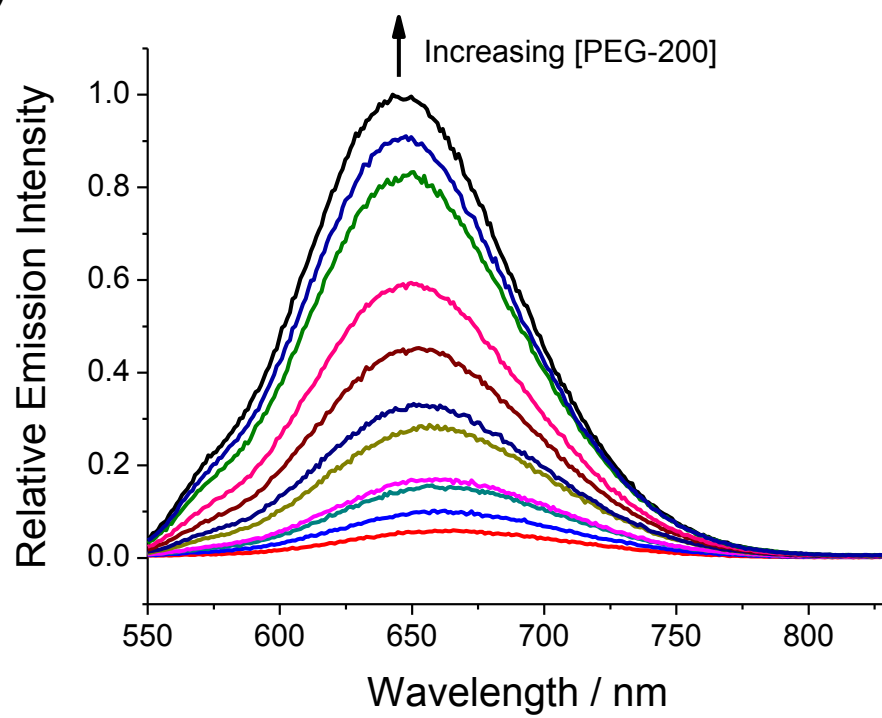


Fig. S20 (a) Emission spectral changes of *m*PPE-Ala and **1** with increasing concentration of *c-myc* in the range of 0 to 25 μM in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the presence of 36 % (v/v) PEG-200. Relative emission intensity of *m*PPE-Ala and **1** at (b) 461 nm and (c) 662 nm with increasing concentration of *c-myc* in aqueous buffer solution in the presence of 36 % (v/v) PEG-200. Concentrations of *m*PE-Ala and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

(a)



(b)



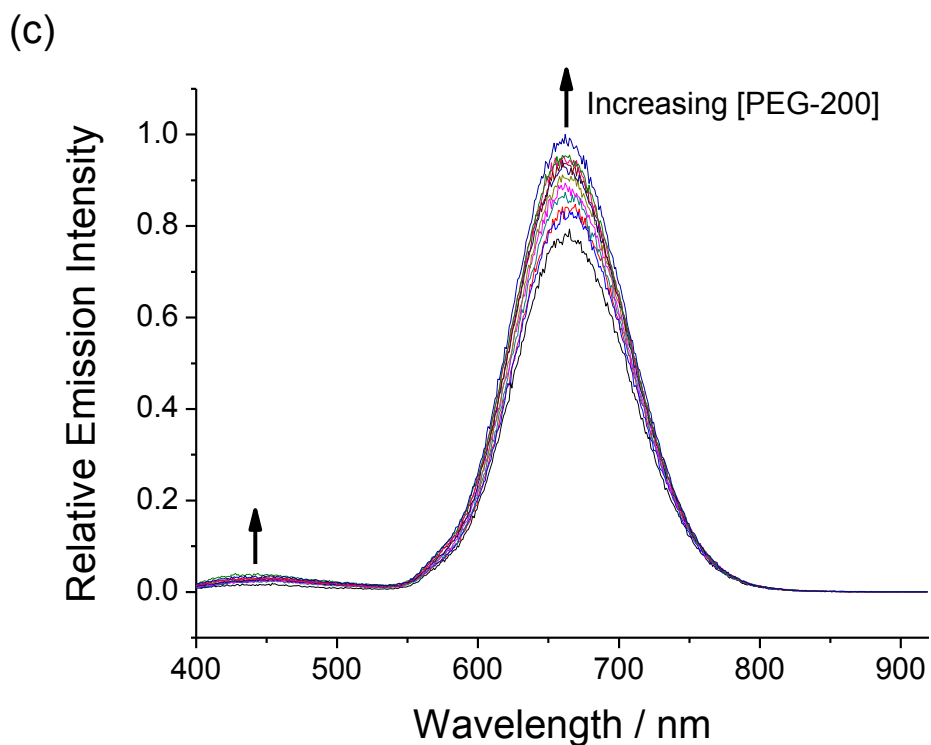


Fig. S21 (a) Emission spectra of *mPPE-Ala* in the absence (black) and in the presence of 20 % (v/v) (red) and 40 % (v/v) (blue) PEG-200 in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4). Concentration of *mPE-Ala* was 50 μ M. Excitation was at 299 nm. (b) Emission spectral changes of **1** with increasing concentration of PEG-200 in the range of 0 to 10 % (v/v) in aqueous buffer solution. Concentration of **1** was 20 μ M. Excitation was at 369 nm. (c) Emission spectral changes of *mPPE-Ala* and **1** with increasing concentration of PEG-200 in aqueous buffer solution. Concentrations of *mPE-Ala* and **1** were 50 μ M and 20 μ M respectively. Excitation was at 299 nm.

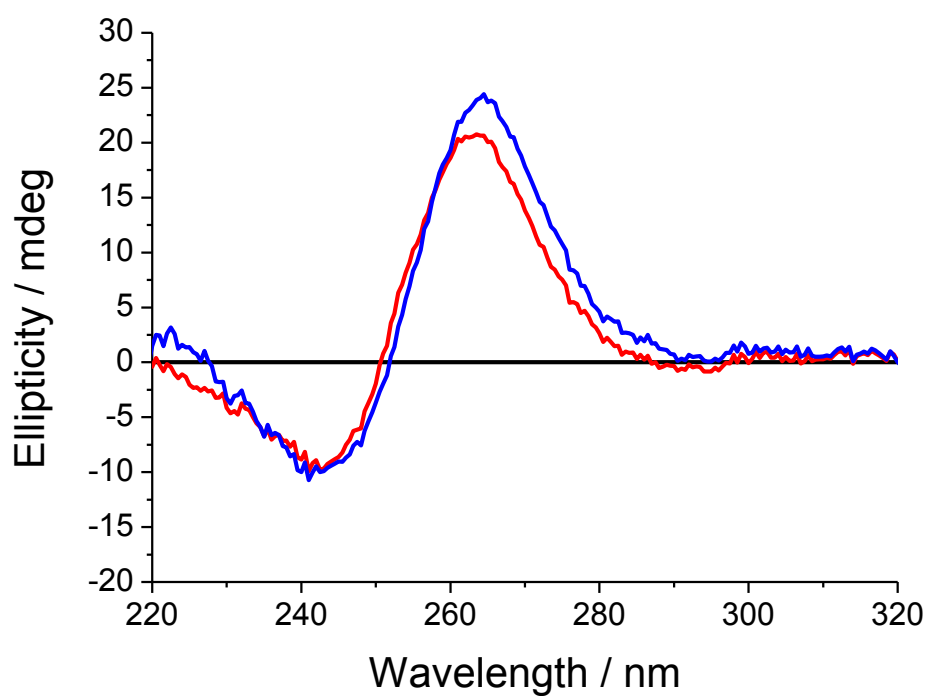


Fig. S22 CD spectra of *c-myc* in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the absence (red) and in the presence (blue) of 40 % (v/v) PEG-200. Concentration of *c-myc* was 3 μ M. The black line represents the signal of aqueous buffer solution.

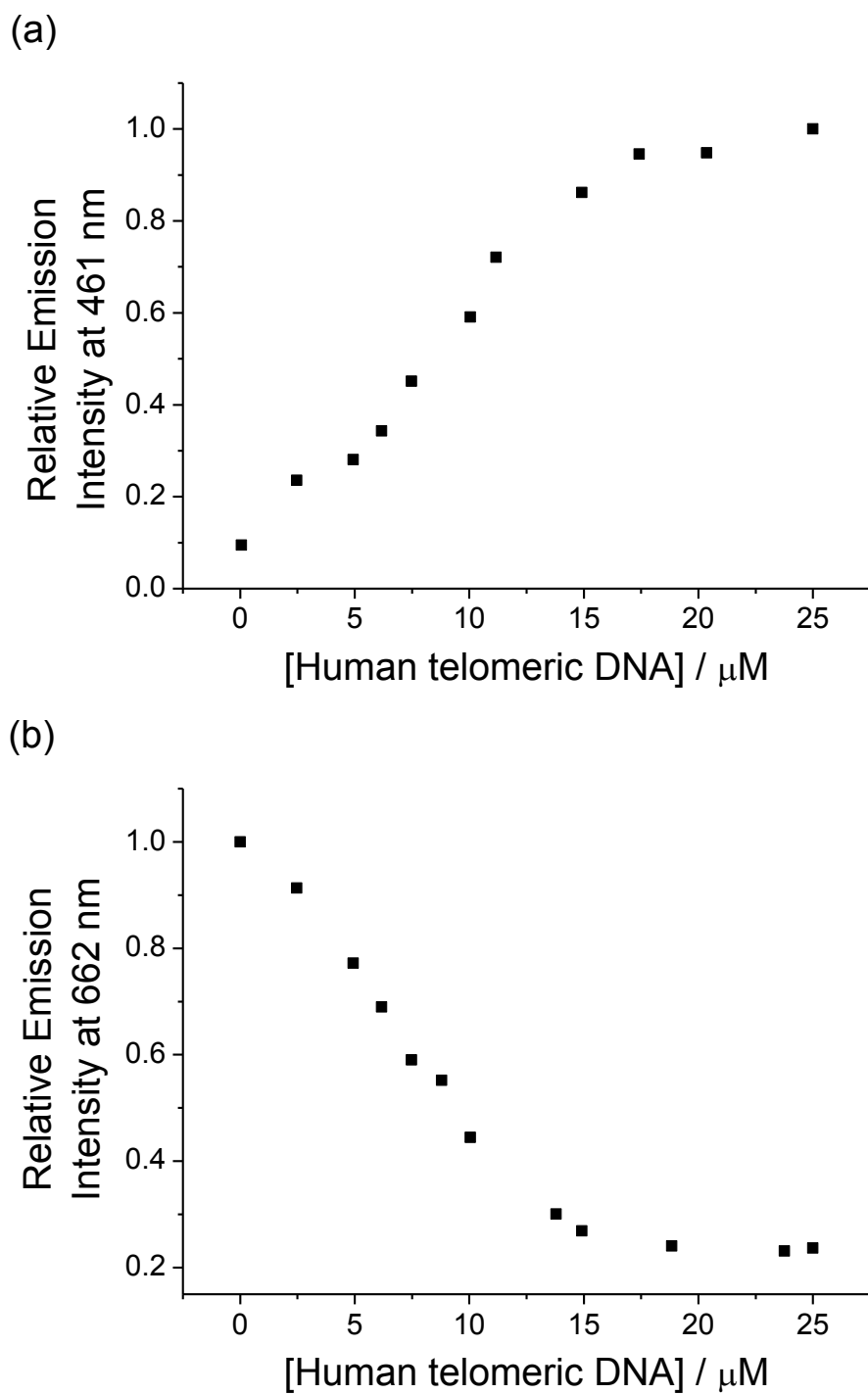


Fig. S23 Relative emission intensity of *m*PPe-Ala and **1** at (a) 461 nm and (b) 662 nm with increasing concentration of human telomeric DNA in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the presence of 40 % (v/v) PEG-200. Concentrations of *m*PE-Ala and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.

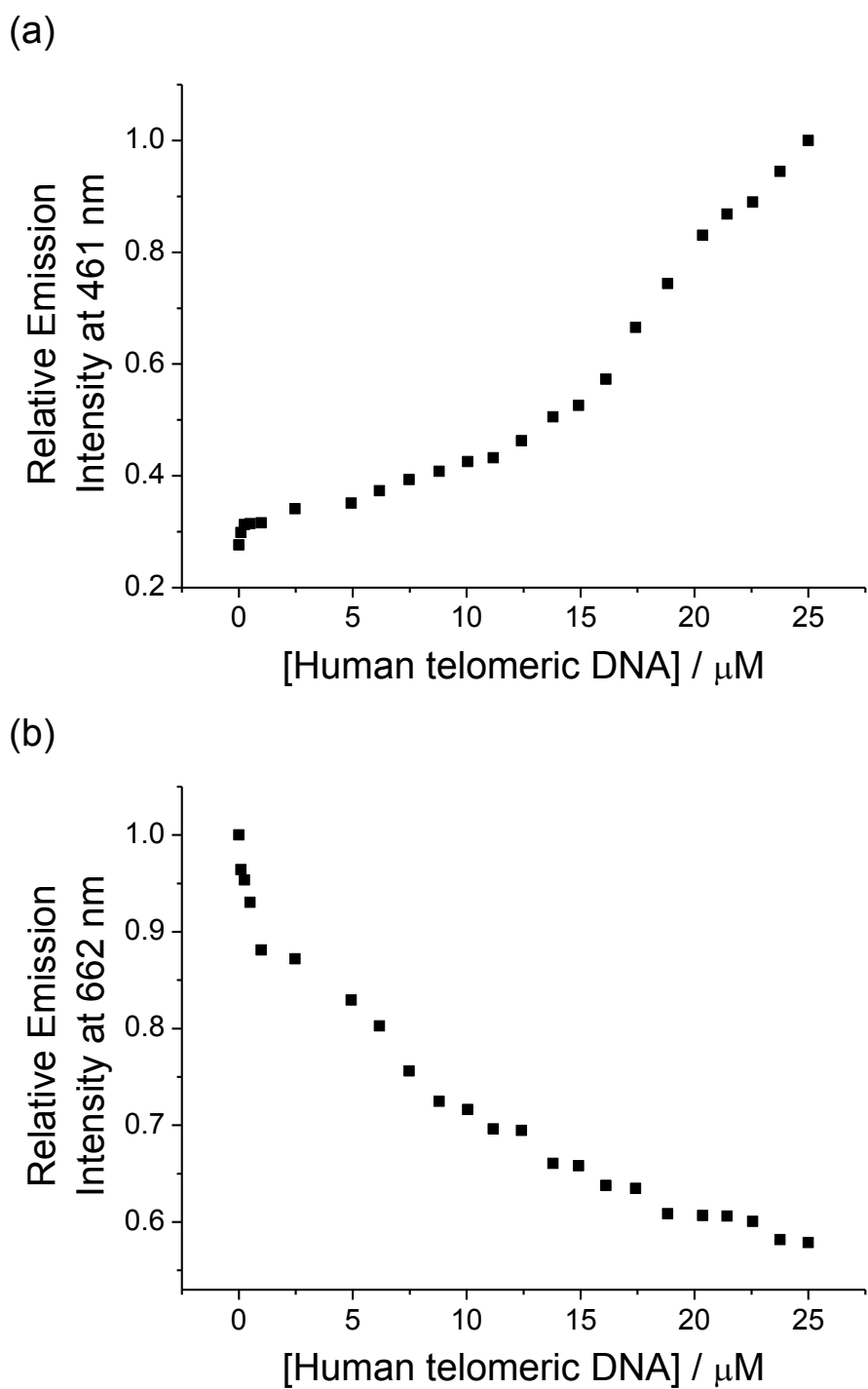


Fig. S24 Relative emission intensity of *mPPE-Ala* and **1** at (a) 461 nm and (b) 662 nm with increasing concentration of human telomeric DNA in aqueous buffer solution (5 mM Tris-HCl, 50 mM KCl, pH 7.4) in the presence of 36 % (v/v) PEG-200. Concentrations of *mPE-Ala* and **1** were 50 μM and 20 μM respectively. Excitation was at 299 nm.