Supporting Information

Experimental Section

Materials and measurements:

DNA sequences used in the study.

G4：5′- CTCTTCGAGGGTTTTGGGTTTTGGGTTTTGGGAGCTA-3′

P：5′-AAAACCCAAAACCCAAAACCCGCGACGAGTCACAACAG-3′

T：5′-CTGTTGTGACTCGTCGCAATAAC-3′

T1：5′-CTGTT**C**TGACTCGTCGCAATAAC-3′

T2：5′-CTGTT**CA**GACTCGTCGCAATAAC-3′

T3：5′-CTGTT**CAC**ACTCGTCGCAATAAC-3′

* The G4 DNA was designed partially complementary to P DNA with two bases mismatched at 5′-terminus. When the G4/P probe was challenged with perfectly matched target DNA, ssDNA at 5′ end of the probe hybridized with the perfectly matched target DNA. Then the mismatched part of G4 would be displaced by the target DNA. Degraded by Exo III, the probe that hybridized with target DNA would release target DNA. Then the mismatched part of G4 DNA which was displaced by target DNA previously, would hybridize with P DNA again to make sure the digestion continue.

*Bold letters represent the mismatched bases.

DNAs were synthesized by Shanghai Sangon Biological Engineering Technology & Services (Shanghai, China). NMM was purchased from Porphyrin Products (Logan, UT), and its concentration was measured by using absorbance spectroscopy on a JASCO V-550 and found to be λ =379 nm, assuming an extinction coefficient of 1.45×10^5 $M^{-1}cm^{-1}$. Exonuclease III was purchased from Takara Biotechnology Co. Ltd. (Dalian, China) and used without further purification. Other chemicals were purchased from Sigma–Aldrich and used without further purification. All water used to prepare buffer solutions was obtained by using a Milli-Q water system. All measurements were performed in Mg-K buffer (10 mM Tris–HCl, 75 mM KCl, 10 mM $MgCl_2$, pH 8.0)， unless stated otherwise.

Fluorescence measurements：

Steady-state fluorescence measurements and time curves were carried out by using a JASCO FP-6500 spectrofluorometer. The fluorescence intensity of all samples was analyzed via a time base scan (λ_{ex}=399 nm, λ_{em}=609 nm). Slit widths for the excitation and emission were set at 10 and 10 nm, respectively. All measurements were performed in Mg-K buffer (10 mM Tris–HCl, 75 mM KCl, 10 mM $MgCl_2$, pH 8.0). The Mg buffer (10 mM Tris–HCl, 10 mM $MgCl_2$, pH 8.0) that was used for control analysis differ in their lack of corresponding salt components compared with

Mg-K buffer.

UV absorbance and melting studies：

UV melting experiments were carried out on a Cary 300 UV/Vis spectrophotometer equipped with a Peltier temperature control accessory. Absorbance changes at 295 nm versus temperature were collected at a heating rate of 1 °C min^{-1}.

CD spectroscopy：

Circular dichroism spectra were recorded with a JASCO J-810 spectropolarimeter equipped with a temperature controlled water bath. CD spectra were recorded from 350 to 210 nm in 1nm increments with an average time of 2 second and three scans were accumulated and automatically averaged.

Gel electrophoresis：

PAGE (20 %) was carried out in 1 × TBE buffer. The samples were incubated before initiating digestion at 25 °C for 5 min. Samples needed hydrolysis were initiated by addition of Exo III and maintained at 25 °C (reaction mixtures contained 50 U of Exo III). After 2h, digestions were stopped by adding stop buffer (57 mM EDTA). Samples were electrophoresed on 20% denaturing polyacrylamide gel. After Stains-All staining, gels were scanned. [DNA]= 3 μM.

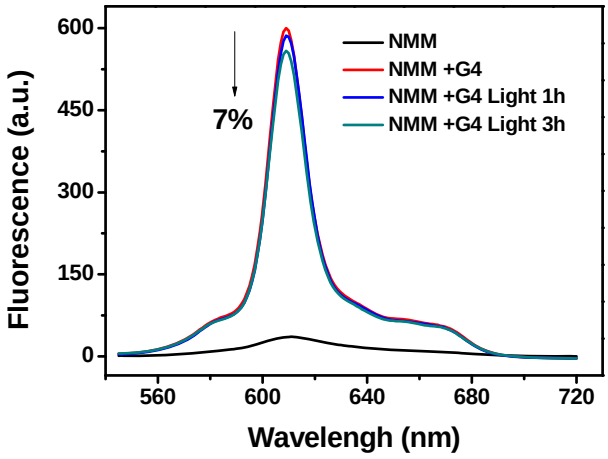

Fig. S1. Fluorescence spectra for NMM and fluorescence spectra recorded before and after 1h and 3h irradiation with visible light (350 nm <λ< 650nm) for 2μM NMM in the presence of 1 μM G4 in 10 mM Tris-HCl, 75 mM KCl, 10 mM $MgCl_2$, pH 8.0 buffer.λ_{ex} = 399 nm. Percent photobleaching calculated as the average loss in fluorescence at 609 nm for three separate trials.

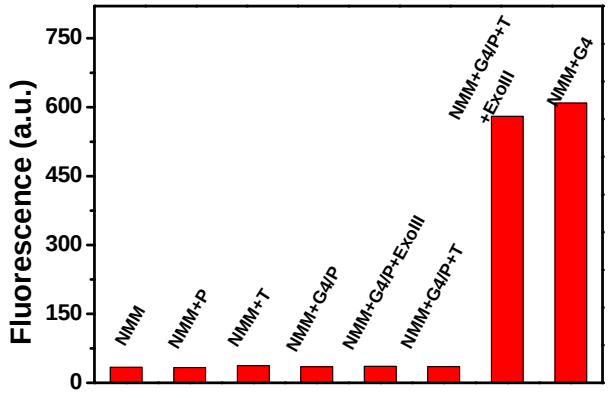

Fig. S2. Target triggered Exo III digesting the duplex G4/P probe and bringing about the fluorescence enhancement. Some control assays were carried out and control samples were indicated in the figure. [NMM]= 2μM, [DNA] = 1μM. All experiments were carried out in 10 mM Tris-HCl, 75 mM KCl, 10 mM $MgCl_2$, pH 8.0 buffer at RT.

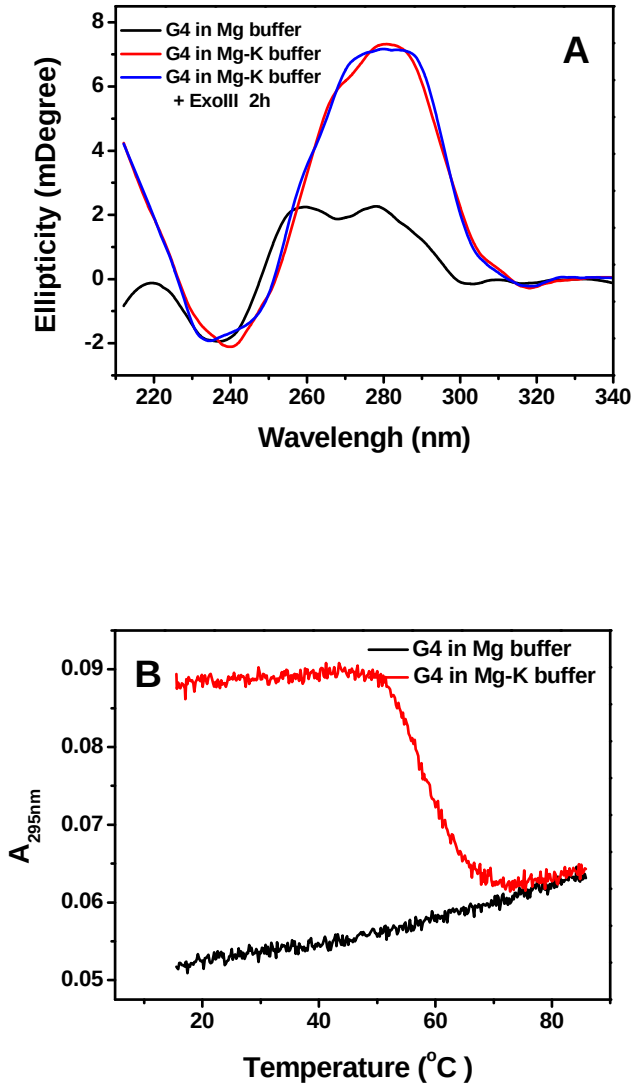

Fig. S3. (A) CD spectra of G4 (2 μM) in Mg buffer and Mg-K buffer in the absence and presence of Exo III. (B) UV melting curves of G4 (2 μM) in Mg buffer and Mg-K buffer. These results verified the quadruplex formation of G4 DNA in Mg-K buffer.

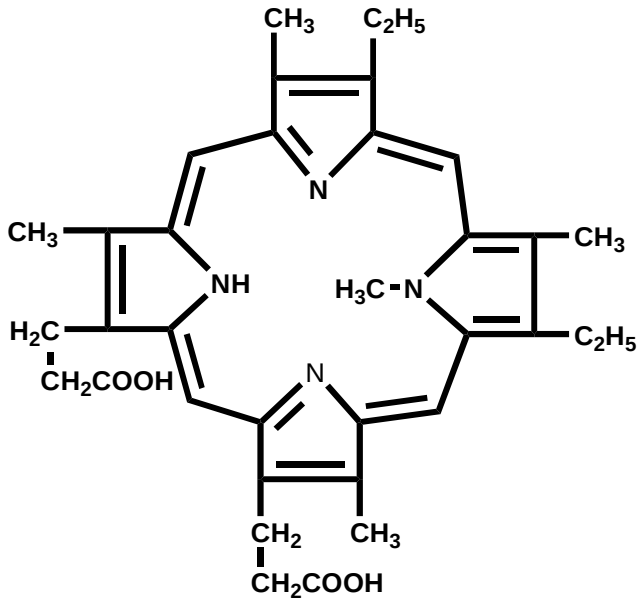

Fig. S4. Molecular structure of NMM.