Supplementary Information

Using fluorescence changes of ^{Fl}U units at terminal and midloop positions to probe i-motif structures

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Sequence	MALDI-TOF signal $[M^+]$	
	Calc. m/z	Found <i>m</i> / <i>z</i>
ODN tF	6678.2	6679.3
ODN mF	6678.2	6678.3
ODN G	6653.4	6654.2
ODN N	6504.3	6506.0

 Table S1 MALDI-TOF mass spectral data.



Figure S1 MALDI-TOF mass spectra of (a) ODN tF, (b) ODN mF (c) ODN G, (d) ODN N.

Experimental method for picosecond Time-Resolved Fluorescence (TRF)

The laser system consists of home-built femtosecond Ti:Sapphire oscillator, multi-pass Ti:Sapphire amplifier operating at 10 kHz, and a noncollinear optical parametric amplifier (NOPA). The NOPA was pumped by the second harmonic of the amplified fundamental at 800 nm to produce 50 fs pulses tunable across the visible. The output of the NOPA around 620 nm was frequency doubled to 310 nm for excitation source.

Picosecond TRF was measured by the time correlated single photon counting (TCSPC) method. In the TCSPC experiment, fluorescence was collected by a parabolic mirror, dispersed by a monochromator, and detected with a thermoelectrically cooled microchannel plate photomultiplier tube (Hamamatsu R3809U-51). Outputs of the photomultiplier tube (start pulse) and a fast photodiode (stop pulse) were processed by a pair of 1 GHz amplifier/discriminator (EG&G Ortec, 9327) and analyzed by a picosecond time analyzer (EG&G Ortec, 9308). The width of the instrument response function (IRF) was 50 ps.





Figure S2 All residuals of fitted values for multiple exponentials and their reduced χ^2 values. All fitting parameters are achieved with reduced χ^2 values of around 1.