Supplementary Data

Table S1. Fluorescence decay parameters for the APout-M.HhaI-AdoHcy single crystal. Fluorescence was excited at 320nm and decays obtained at the 3 emission wavelengths shown were fitted globally to 4 common lifetimes. The global fluorescence lifetimes and global χ^2 are given, followed by the A factors and local χ^2 for each decay.

Global Lifetimes: τ1=0.07ns, τ2=0.53ns, τ3=2.07ns, τ4=7.36ns
Global χ^2=1.052

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.62	0.21	0.14	0.03	1.038
390	0.64	0.19	0.14	0.03	1.073
410	0.66	0.16	0.15	0.03	1.045

Table S2. Fluorescence decay parameters for the APtarget-M.HhaI-AdoHcy single crystal.

Global Lifetimes: τ1=1.10ns, τ2=6.30ns, τ3=10.85ns
Global χ^2=1.023

Emission Wavelength/ nm	A1	A2	A3	Local χ^2
370	0.12	0.21	0.67	1.042
390	0.07	0.16	0.77	1.004
410	0.04	0.12	0.84	1.023

Table S3. Fluorescence decay parameters for the APout duplex in aqueous solution.

Global Lifetimes: τ1=0.04ns, τ2=0.50ns, τ3=2.95ns, τ4=11.01ns
Global χ^2=1.121

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.78	0.08	0.07	0.07	1.132
390	0.70	0.12	0.10	0.08	1.049
410	0.65	0.19	0.10	0.06	1.181

Table S4. Fluorescence decay parameters for the APout duplex with M.HhaI (wild type) and AdoHcy in aqueous solution.

Global Lifetimes: τ1=0.03ns, τ2=0.47ns, τ3=2.82ns, τ4=10.25ns
Global χ^2=1.171

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.80	0.07	0.06	0.07	1.197
390	0.70	0.12	0.08	0.10	1.121
410	0.70	0.14	0.08	0.08	1.196

Table S5. Fluorescence decay parameters for the APtarget duplex in aqueous solution.

Global Lifetimes: τ1=0.08ns, τ2=0.58ns, τ3=2.94ns, τ4=9.60ns
Global χ^2=1.137

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.60	0.18	0.13	0.09	1.170
390	0.53	0.27	0.14	0.06	1.092
410	0.51	0.30	0.14	0.05	1.149

Table S6. Fluorescence decay parameters for the APtarget duplex with M.HhaI (T250G) in aqueous solution.

Global Lifetimes: τ1=0.14ns, τ2=1.01ns, τ3=5.26ns, τ4=12.58ns
Globaļ χ^2=1.045

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
70	0.19	0.14	0.22	0.45	1.065
390	0.19	0.16	0.19	0.46	1.008
410	0.26	0.20	0.16	0.38	1.062

Table S7. Fluorescence decay parameters for the APtarget duplex with M.HhaI (T250G) and AdoMet in aqueous solution.

Global Lifetimes: τ1=0.17ns, τ2=1.14ns, τ3=6.01ns, τ4=12.63ns
Globaļ χ^2=1.004

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.16	0.12	0.25	0.47	0.992
390	0.16	0.14	0.20	0.50	0.996
410	0.24	0.17	0.16	0.43	1.024

Table S8. Fluorescence decay parameters for the APadj-M.HhaI-AdoHcy single crystal.

Global Lifetimes: τ1=0.19ns τ2=0.91ns, τ3=3.54s, τ4=10.13ns
Global χ^2=1.085

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.61	0.30	0.07	0.02	1.117
390	0.54	0.32	0.10	0.04	1.052
410	0.49	0.32	0.14	0.05	1.087

Table S9. Fluorescence decay parameters for the APopp-M.HhaI-AdoHcy single crystal.

Global Lifetimes: τ1=0.15ns τ2=0.94ns, τ3=3.41s, τ4=9.39ns
Global χ^2=1.084

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.58	0.27	0.11	0.04	1.145
390	0.50	0.30	0.15	0.05	1.045
410	0.44	0.31	0.17	0.08	1.062

Table S10. Fluorescence decay parameters for the APadj duplex in aqueous solution.

Global Lifetimes: τ1=0.04ns, τ2=0.45ns, τ3=2.63ns, τ4=10.33ns
Globaļ χ^2=1.128

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ^2
370	0.89	0.06	0.03	0.02	1.103
390	0.84	0.09	0.05	0.02	1.121
410	0.77	0.14	0.07	0.02	1.159

Table S11. Fluorescence decay parameters for the APadj duplex with M.HhaI (wild-type) and AdoHcy in aqueous solution.

Global Lifetimes: τ1=0.08ns, τ2=0.32ns, τ3=2.35ns, τ4=9.91ns
Globaļχ^2= 1.133

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ2
370	0.83	0.12	0.03	0.02	1.152
390	0.79	0.15	0.04	0.02	1.096
410	0.74	0.18	0.06	0.02	1.152

Table S12. Fluorescence decay parameters for the APopp duplex in aqueous solution.

Global Lifetimes: τ1=0.05ns, τ2=0.50ns, τ3=2.98ns, τ4=9.81ns
Global χ^2=1.089

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ2
370	0.69	0.12	0.09	0.10	1.100
390	0.64	0.16	0.10	0.10	1.086
410	0.59	0.20	0.12	0.09	1.082

Table S13. Fluorescence decay parameters for the APopp duplex with M.HhaI (wild-type) and AdoHcy in aqueous solution.

Global Lifetimes: τ1=0.06ns, τ2=0.44ns, τ3=2.68ns, τ4=9.37ns
Global χ^2= 1.132

Emission Wavelength/ nm	A1	A2	A3	A4	Local χ2
370	0.69	0.16	0.07	0.08	1.149
390	0.65	0.18	0.09	0.08	1.137
410	0.65	0.19	0.09	0.07	1.111