2. A solution of Rhodamine B (14.2 g, 30.0 mmol) in CH_2Cl_2 (40 mL) was treated with EDCI (5.73 g, 30.0 mmol) for 10 min. Bromoethylamine hydrobromide (6.15 g, 30.0 mmol) and TEA (4.21 mL, 3.03 g, 30.0 mmol) were added. The reaction was allowed to proceed for 24 h. Water was added to the solution, the phases were separated, and the aqueous layer was extracted with CH_2Cl_2. The combined organic phases were washed with water and dilute acid (0.1 M HCl). The organic layer was dried ($MgSO_4$) and concentrated. Chromatography [silica, CH_2Cl_2/MeOH (0→1%)] afforded a pale pink solid (7.08 g, 41%): ESI-MS 550 $(M + H)^+$; ^1H NMR δ 1.10 (t, J = 7.0 Hz, 12H), 2.91 (t, J = 7.3 Hz, 2H), 3.26 (q, J = 7.0 Hz, 8H), 3.45 (t, J = 7.3 Hz, 2H), 6.18 (d, J = 2.8 Hz, 1H), 6.21 (d, J = 2.6 Hz, 1H), 6.31 (d, J = 2.6 Hz, 1H), 6.34 (s, 1H), 6.37 (s, 1H), 6.96-7.02 (m, 1H), 7.33-7.39 (m, 2H), 7.80-7.86 (m, 1H); ^{13}C NMR 12.54, 41.76, 44.31, 64.65, 97.72, 105.07, 108.12, 122.89, 123.77, 128.04, 128.64, 130.50, 132.64, 148.84, 153.15, 153.54.

3. Cyclen (3.21 g, 18.66 mmol) was dissolved in 100% EtOH (25 mL). TEA (5.24 mL, 3.77 g, 37.33 mmol) and **2** (7.08 g, 12.44 mmol) were added. The solution was refluxed under argon for 24 h. The ethanol and the triethylamine were evaporated, and the residue was dissolved in CH_2Cl_2. The solution was washed with water, and the aqueous phase was extracted with CH_2Cl_2. The combined organic phases were washed with water and dried ($MgSO_4$). The sample was purified by column chromatography [basic alumina (pH 9.5), CH_2Cl_2/MeOH (0.2→4%)]. A pale pink solid was isolated after evaporation of the solvents (3.12 g, 39%): ESI-MS 641 $(M + H)^+$; ^1H NMR δ 1.10 (t, J = 7.0 Hz, 12H), 2.17-3.37 (m, 28H), 6.17-6.21 (m, 2H), 6.31-6.41 (m, 5H), 6.97-7.01 (m, 1H), 7.32-7.36 (m, 1H), 7.44-7.47 (m, 1H), 7.80-7.86 (m, 1H); ^{13}C NMR 12.56, 37.83, 41.76, 44.32, 45.36, 46.32, 47.18, 51.13, 52.05, 64.78, 97.83, 105.68, 108.00, 122.57, 123.68, 127.87, 128.84, 131.41, 132.11, 148.67, 153.34, 153.45, 167.61.

4. A sample of **3** (3.12 g, 4.73 mmol) in anhydrous $CHCl_3$ (55 mL) was treated with TEA (6.6 mL, 47.3 mmol) and the solution was flushed with Ar for 10 min. N-Boc-bromoethylamine (1.06 g, 4.73 mmol) in $CHCl_3$ (15 mL) was added drop-wise (1 h) to the reaction mixture. The stirring was continued for 36 h at room temperature. Water was added to the solution. The aqueous layer was extracted with CH_2Cl_2. The organic phase was washed with water and dried ($MgSO_4$). Silica column chromatography [CH_2Cl_2/^{i}PrNH_2 (2.5→10%)] yielded an off-white solid (0.75 g, 20%): ESI-MS 784 $(M + H)^+$; ^{1}H NMR δ 1.10 (t, *J* = 7.0 Hz, 12H), 1.29 (s, 9H), 2.08-2.64 (m, 20H), 2.92-3.30 (m, 12H) 6.18 (d, *J* = 2.7 Hz, 1H), 6.21 (d, *J* = 2.7 Hz , 1H), 6.30-6.35 (m, 5H), 6.44 (br s, 1H), 6.99-7.04 (m, 1H), 7.34-7.40 (m, 2H), 7.80-7.84 (m, 1H); ^{13}C NMR: 12.62, 28.50, 34.79, 38.76, 44.31, 45.28, 45.36, 46.75, 47.46, 48.11, 50.73, 51.38, 51.63, 52.39, 53.26, 64.79, 97.54, 105.61, 108.03, 122.53, 123.79, 128.05, 129.11, 131.81, 132.17, 148.75, 153.11, 153.49, 167.36.

5. To a sample of **4** (1.08 g, 1.56 mmol) in CH_3CN (8 mL) were added K_2CO_3 (3.50 g, 25.0 mmol), NaI (3.74 g, 25.0 mmol) and ethyl bromoacetate (1.70 mL, 15.6 mmol). The mixture was refluxed under Ar for 24 h. The solvent was evaporated and the residue was dissolved in a mixture of water and CH_2Cl_2. The phases were separated and the aqueous phase was extracted with CH_2Cl_2. The combined organic phases were washed with water and dried ($MgSO_4$). After column chromatography [silica, CH_2Cl_2/MeOH (0→2.5%)] an off-white solid was obtained (1.24 g, 83%): ESI-MS 957 $(M + H)^+$; ^1H NMR δ 1.07-1.25 (m, 18H), 1.34 (s, 9H), 2.18-3.58 (m, 36H), 4.00-4.53 (m, 4H), 6.18 (d, *J* = 2.6 Hz, 1H), 6.21 (d, *J* = 2.6 Hz, 1H), 6.26-6.37 (m, 4H), 7.01-7.04 (m, 1H), 7.36-7.39 (m, 2H), 7.80-7.83 (m, 1H); ^{13}C NMR: 12.61, 14.24, 14.27, 28.48, 44.32, 50.75, 51.27, 51.91, 52.41, 53.40, 60.18, 97.62, 105.57, 108.02, 122.58, 123.79, 128.04, 129.00, 148.76, 153.46, 156.23, 167.47, 171.39.

6. A sample of **5** (1.24 g, 1.30 mmol) in CH_2Cl_2 (12 mL) was treated with TFA (6 mL). The reaction mixture was stirred at room temperature for 30 min. The volatile components were evaporated. The residue was dissolved in CH_2Cl_2. Concentrated aqueous $KHCO_3$ was added in small portions until pH 8 was reached. The phases were separated and the aqueous layer was extracted with CH_2Cl_2. The combined organic phases were washed with water and dried ($MgSO_4$). Evaporation of the solvent afforded a pale purple solid (0.93 g, 84%): ES-MS 856 $(M + H)^+$, 428 $(M + 2H)^{2+}$; ^1H NMR δ 1.07-1.23 (m, 18H), 2.10-3.39 (m, 36H), 3.98 (q, *J* = 7.2 Hz, 2H), 4.07-4.14 (m, 2H), 6.20 (d, *J* = 2.6 Hz, 1H), 6.23 (d, *J* = 2.6 Hz, 1H), 6.30-6.37 (m, 4H), 7.01-7.04 (m, 1H), 7.35-7.40 (m, 2H), 7.81-7.84 (m, 1H), 8.07 (br s, 2H); ^{13}C NMR δ 12.58, 14.01, 34.87, 37.00, 44.31, 49.13, 49.59, 49.92, 50.79, 51.33, 52.35, 55.62, 60.35, 60.91, 64.93, 97.51, 105.50, 108.10, 122.69, 123.77, 128.12, 129.06, 131.45, 132.33, 148.80, 152.95, 153.46, 167.61, 171.35, 171.89.

7a and 7b. To a solution of **6** (0.86 g, 1.0 mmol) in DMF (7 mL) was added a solution of 5’-carboxylic nucleotide (1.1 mmol) and EDCI (0.23 g, 1.2 mmol). The reaction mixture was stirred at room temperature for 24 hours. The DMF was removed at reduced pressure. The oily residue was dissolved in a mixture of CH_2Cl_2 and water. The phases were separated and the aqueous layer was extracted with CH_2Cl_2. The combined organic phases were washed with water and dried ($MgSO_4$). Purification by silica column chromatography (CH_2Cl_2/^{i}PrNH$_2$, 0-20%) afforded off-white solids:

7a (B = U): Yield: 0.54 g, 48%; ESI-MS 1136 $(M + H)^+$; HRESI-MS ($C_{60}H_{83}N_{10}O_{12}$) Calc 1136.6245 $(M + H)^+$, Found 1136.6251; Anal Calc for $C_{60}H_{83}N_{10}O_{12}$ C, 63.42; H, 7.36; N, 12.33; Found C, 63.62; H, 7.41; N, 12.28. ^{1}H NMR δ 1.07-1.21 (m, 18H), 1.25 (s, 3H), 1.48 (s, 3H), 1.87-3.31 (m, 39H), 4.01-4.10 (m, 5H), 4.55 (br s, 1H), 5.63 (d, *J* = 8.1 Hz, 2H), 6.18-6.23 (m, 2H), 6.30-6.35 (m, 3H), 7.02-7.05 (m, 1H), 7.38-7.41 (m, 2H), 78.2-7.85 (m, 1H); ^{13}C NMR δ 12.60, 14.20, 24.52, 26.96, 33.52, 44.35, 45.25, 50.66, 50.74, 51.91, 52.48, 52.17, 60.48, 77.93, 84.21, 89.70, 97.61, 102.67, 108.09, 109.86, 122.79, 123.86, 125.49, 128.94, 129.05, 132.48, 132.92, 148.85, 153.48, 171.31, 173.66.

7b (B = A): Yield: 0.46 g, 40%; ESI-MS 1163 $(M + H)^+$; HRESI-MS ($C_{61}H_{84}N_{13}O_{10}$) Calc 1159.6540 $(M + H)^+$, Found 1159.6551; Anal. Calc. for $C_{61}H_{84}N_{13}O_{10}$ C, 63.19; H, 7.30; N, 15.71; Found C, 63.13; H, 7.27; N, 15.78. ^{1}H NMR δ 1.06-1.22 (m, 18H), 1.30 (s, 3H), 1.52 (s, 3H), 1.87-3.29 (m, 39H), 4.01-4.10 (m, 5H), 5.89 (br s, 2H), 6.17-6.22 (m, 2H), 6.29-6.37 (m, 3H), 7.01-7.04 (m, 1H), 7.36-7.39 (m, 2H), 7.80-7.83 (m, 1H), 8.06 (s, 1H), 8.22 (s, 1H); ^{13}C NMR δ 12.59, 14.24, 14.30, 25.11, 26.83, 44.33, 45.24, 51.54, 52.09, 55.23, 55.34, 60.03, 60.11, 64.87, 83.70, 84.33, 87.07, 91.51, 97.62, 97.71, 105.61, 108.01, 113.72, 119.75, 122.60, 123.80, 128.04, 128.97, 129.14, 132.21, 140.34, 148.74, 149.49, 153.08, 153.43, 155.50, 167.43, 168.60, 171.48, 171.59.

Nd7a and Nd7b. Samples of **7a** and **7b** (75 mg and 80 mg, 0.066 mmol and 0.069 mmol, respectively) in HPLC-grade methanol (0.5 mL) were treated with anhydrous $NdCl_3$ (15.7 mg and 16.4 mg, 0.95 equiv). The solution was refluxed under Ar for 12 h. The reaction mixture was allowed to cool back to room temperature. The mixture was poured into cold diethyl ether (20 mL). The mixture was centrifuged and the ether was decanted from the white precipitate. The solid was suspended in ether (5 mL), and centrifuged again. The ether was poured off again, the residue dissolved in distilled water (3 mL), filtered through a plug of cotton wool and the sample was freeze-dried. Pale pink solids were obtained:

Nd7a (B = Uridine): Yield: 62.5 mg, 82%; ESI-MS 1183 (Ligand – Both ethyl esters and acetal + H)$^+$; HRESI-MS $C_{60}H_{83}N_{10}O_{12}NdCl_4$ Calc 1423.4182, Found 1216.8581 corresponds to ($C_{60}H_{83}N_{10}O_{12}NdCl$ – 2 ethyl groups and acetal)$^+$; Anal. Calc for $C_{60}H_{83}N_{10}O_{12}NdCl_4$ C, 50.64; H, 5.89; N, 9.84; Found C, 50.82; H, 6.02; N, 9.98.

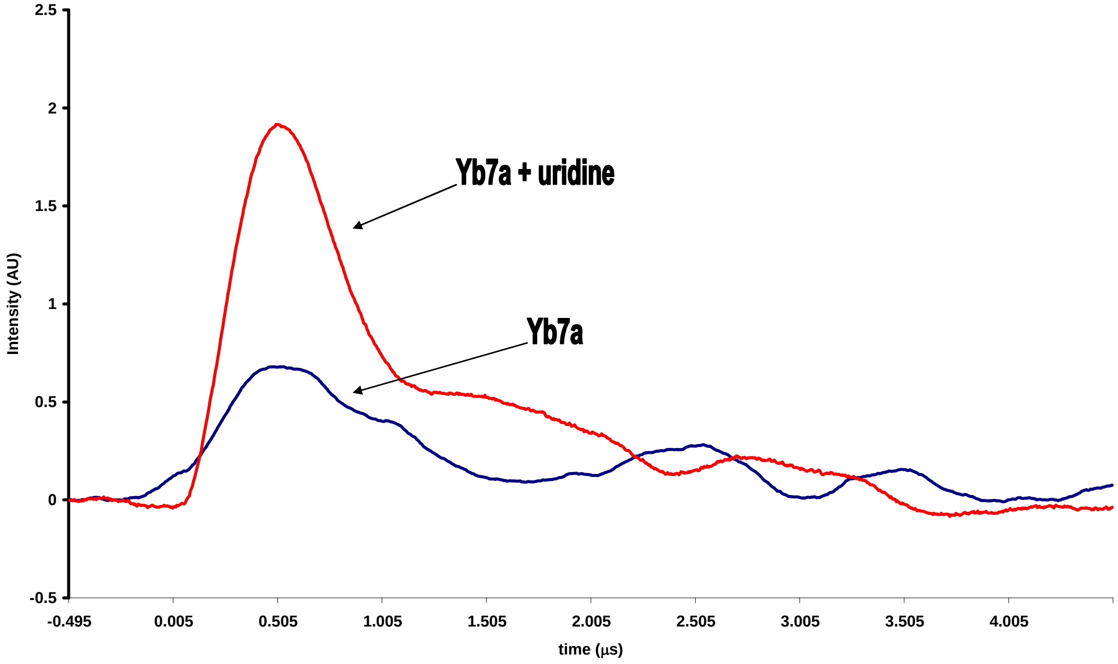
Nd7b (B = Adenosine): Yield: 49.5 mg, 58%; ESI-MS 1204 (Ligand – Both ethyl esters and acetal + H)$^+$ HRESI-MS $C_{61}H_{84}N_{13}O_{10}NdCl_4$ Calc 1444.7927 Found 1238.2333 corresponds to ($C_{61}H_{84}N_{13}O_{10}NdCl$ – 2 ethyl groups and acetal)$^+$; Anal Calc for $C_{61}H_{84}N_{13}O_{10}NdCl_4$ C, 50.74; H, 5.86; N, 12.61; Found C, 50.86; H, 5.98; N, 12.96.

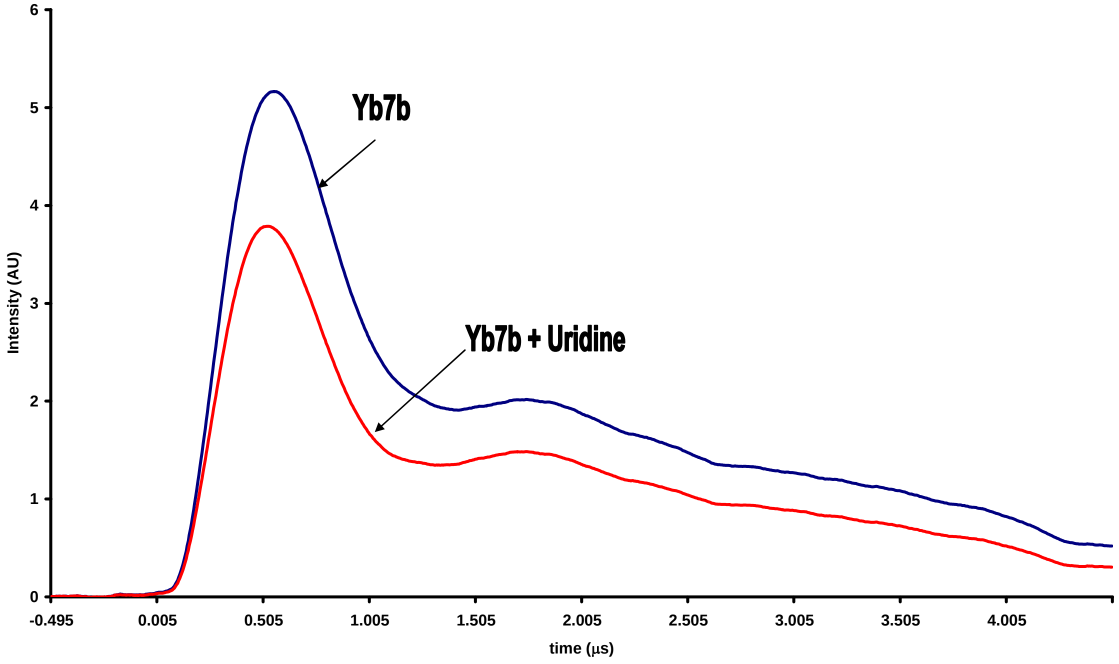
Yb7a and Yb7b. Samples of **7a** and **7b** (75 mg and 80 mg, 0.066 mmol and 0.069 mmol, respectively) in HPLC-grade methanol (0.5 mL) were treated with anhydrous $YbCl_3$ (17.5 mg and 18.3 mg, 0.95 equiv). The solution was refluxed under Ar for 12 h. The reaction mixture was allowed to cool back to room temperature. The mixture was poured into cold diethyl ether (20 mL). The mixture was centrifuged and the ether was poured off the white precipitate. The solid was suspended in ether (5 mL), and centrifuged again. The ether was poured off again. The residue was dissolved in distilled water (3 mL) and was filtered through a plug of cotton wool. The samples were freeze-dried. White solids were obtained:

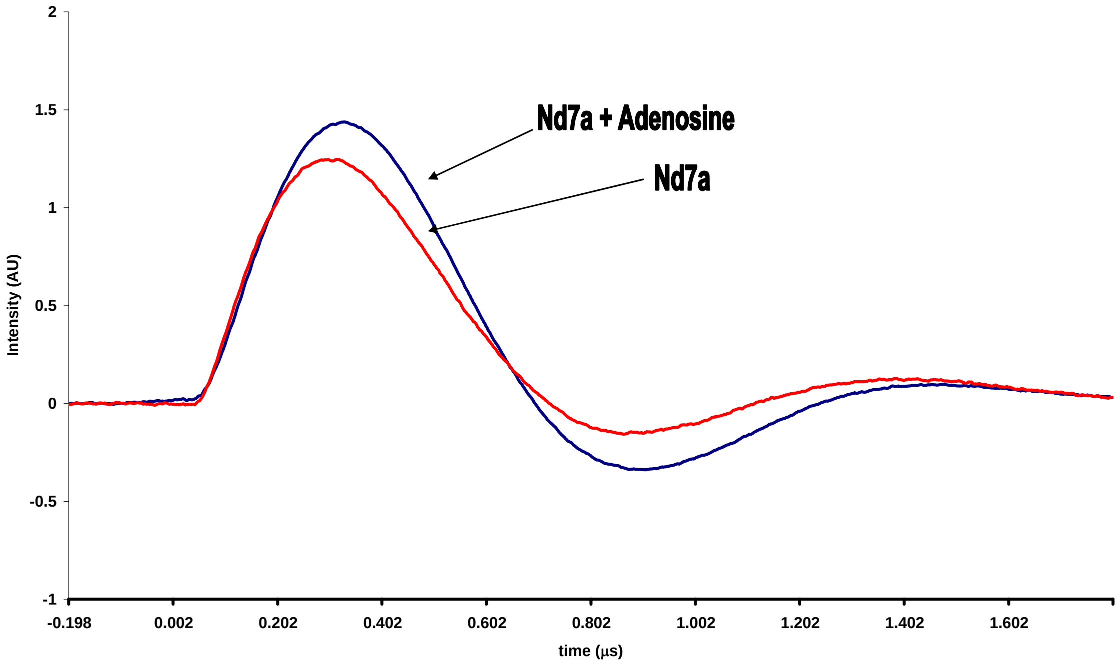
Yb7a (B = U): Yield: 26 mg, 30%; ESI-MS 1183 (Ligand – Both ethyl esters and acetal + H)$^+$ HRESI-MS ($C_{60}H_{83}N_{10}O_{12}YbCl_4$) Calc 1450.4707, Found 1243.7542 corresponds to ($C_{60}H_{83}N_{10}O_{12}YbCl$ – 2 ethyl groups and acetal); Anal Calc for $C_{60}H_{83}N_{10}O_{12}YbCl_4$ C, 49.70; H, 5.77; N, 9.66; Found C, 49.54; H, 5.99; N, 10.01.

Yb7b (B = A): Yield: 44 mg, 50%; ESI-MS 1204 (Ligand – Both ethyl esters and acetal + H)$^+$; HRESI-MS ($C_{61}H_{84}N_{13}O_{10}YbCl_4$) Calc 1473.5007, Found 1266.9392 corresponds to ($C_{61}H_{84}N_{13}O_{10}NdCl$ – 2 ethyl groups and acetal); Anal Calc for $C_{61}H_{84}N_{13}O_{10}YbCl_3$ C, 49.74; H, 5.75; N, 12.36; Found C, 49.65; H, 5.93; N, 12.72.

Time Resolved Emission spectra for the complexes **Yb7a**, **Yb7b** and **Nd7a**. The spectra were recorded in methanol monitoring the emission at 980 nm for the Yb complexes and 890 nm for the Nd complexes. The effect of added nucleoside on the emission intensity was also recorded.







Steady State emmission spectra of Nd7b recorded in methanol (3mM) excitation = 355nm