

### Supporting Information for:

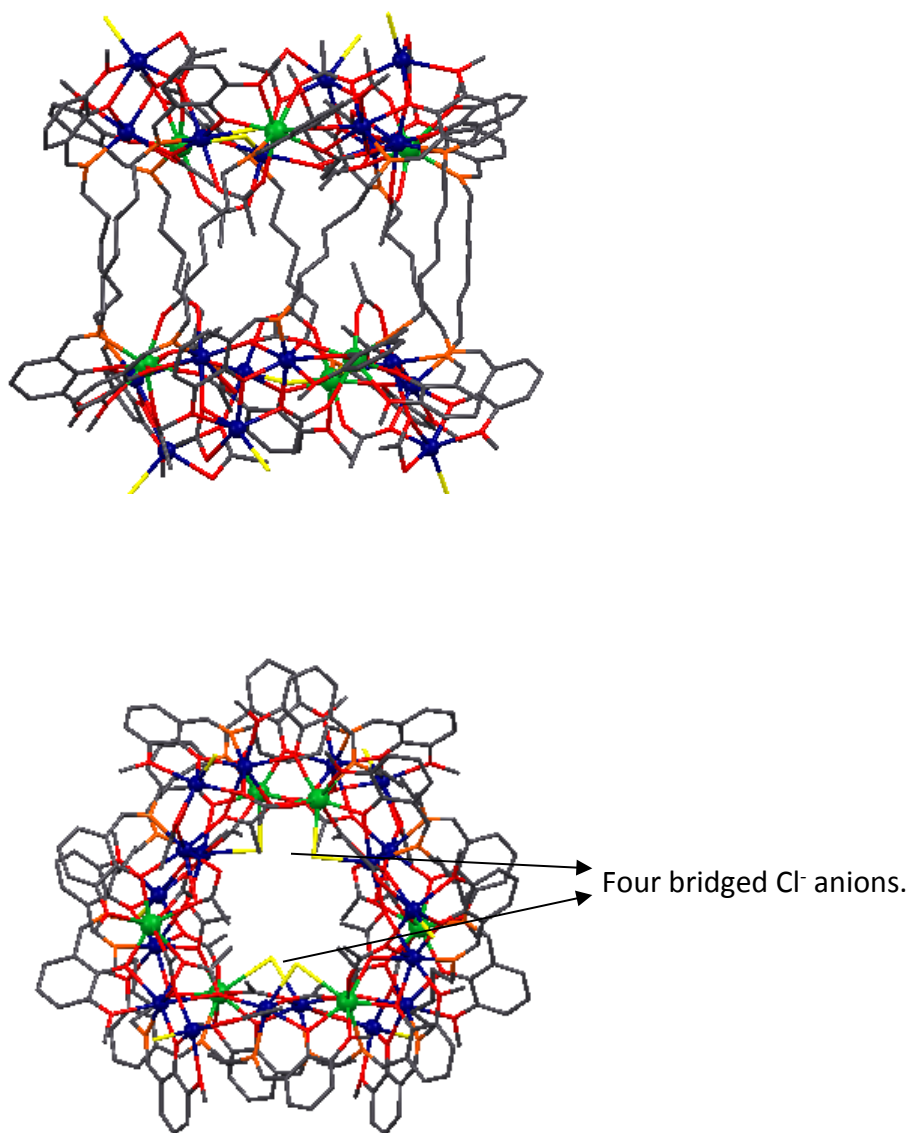
## Lanthanide Nano-drums: A New Class of Molecular Nanoparticles for Potential Biomedical Applications

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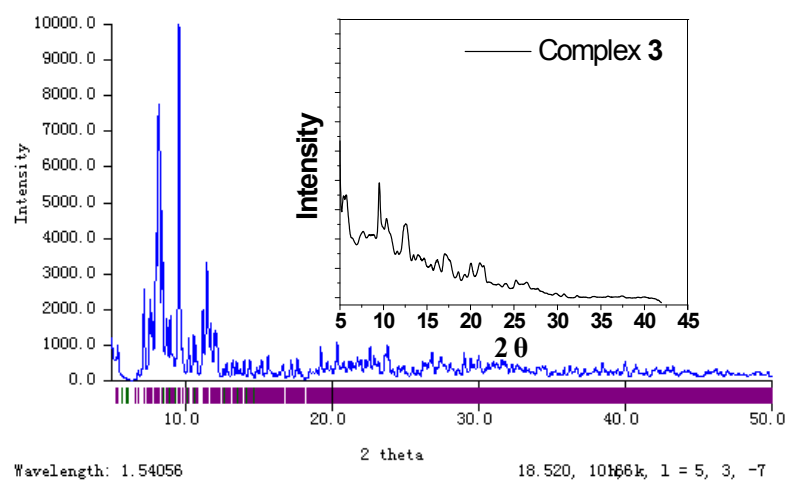
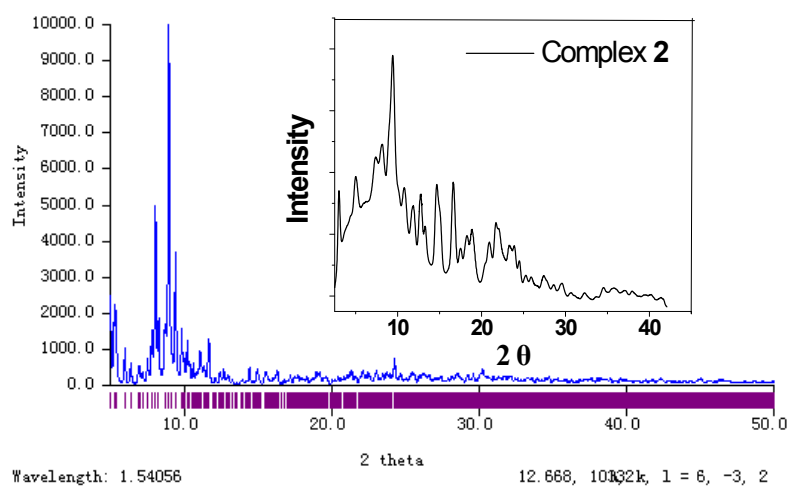
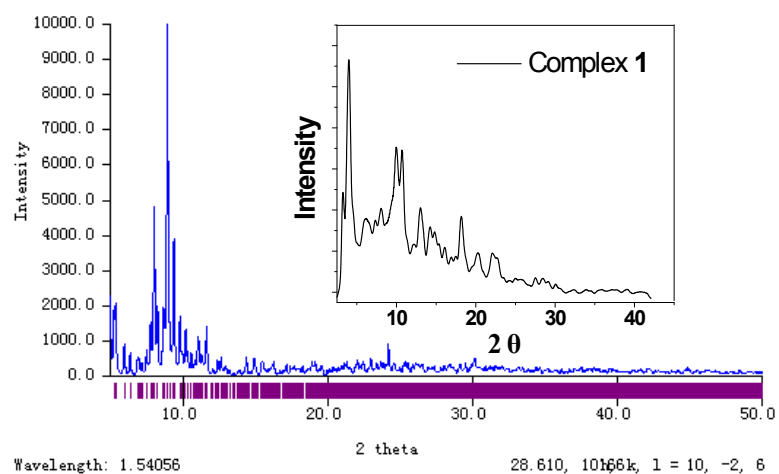
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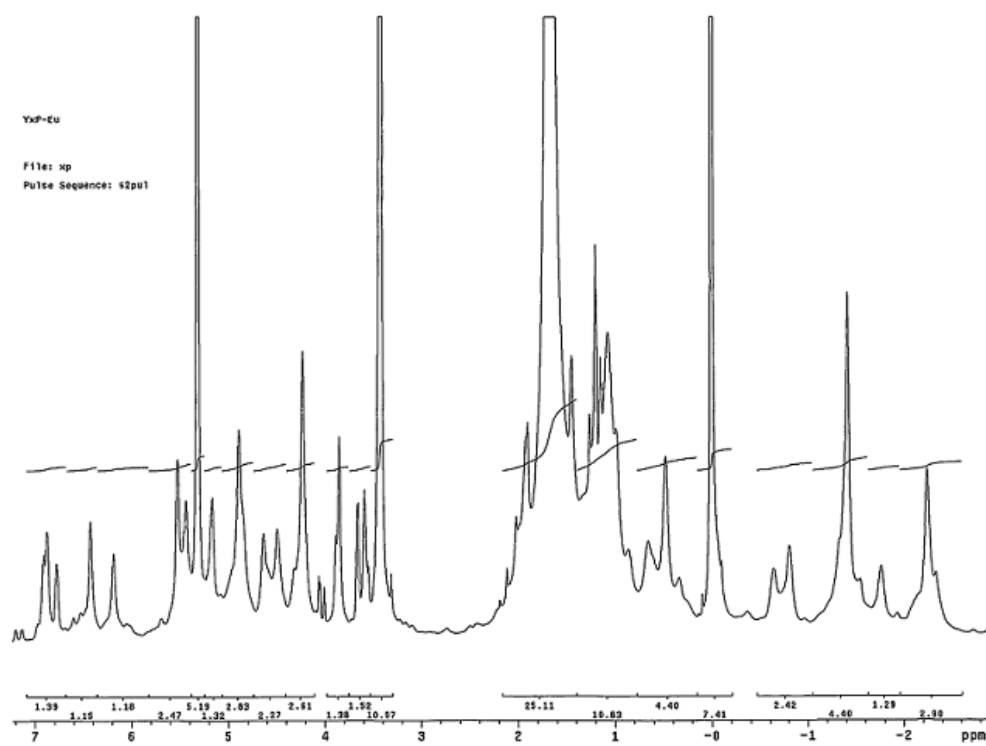
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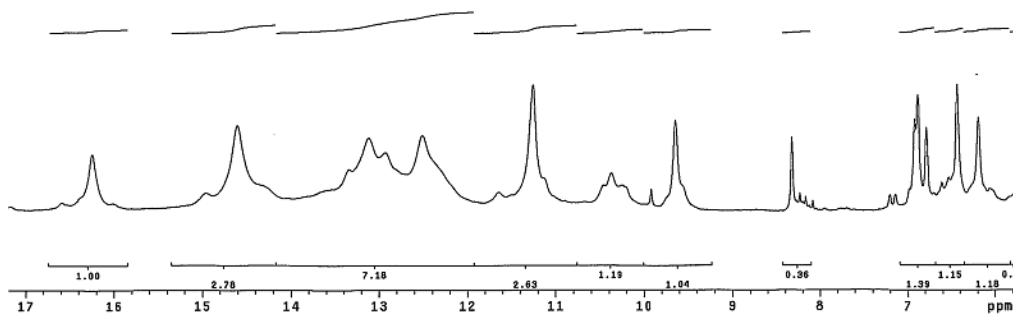
**Figure S1.** A view of the crystal structure of **3**. Viewed along the *b*-axis (up) and *a*-axis (down).  
(Eu<sup>3+</sup>: green; Cd<sup>3+</sup>: blue; Cl<sup>-</sup>: yellow)



**2. Figure S2. Powder XRD patterns of 1-3**

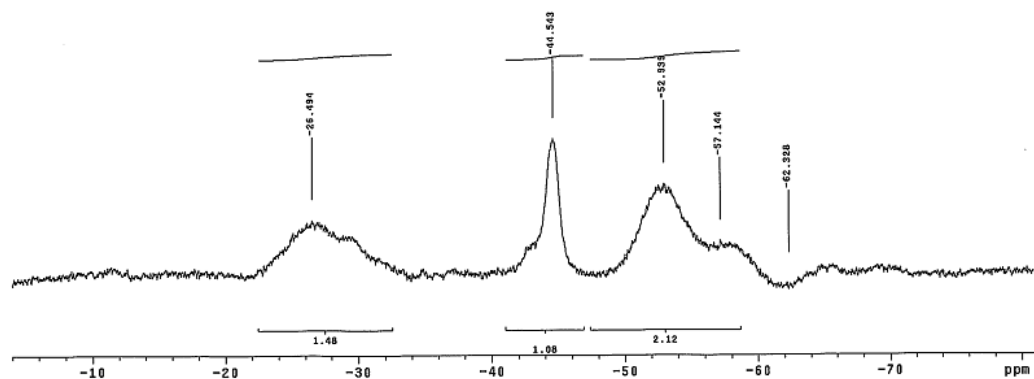


(a)

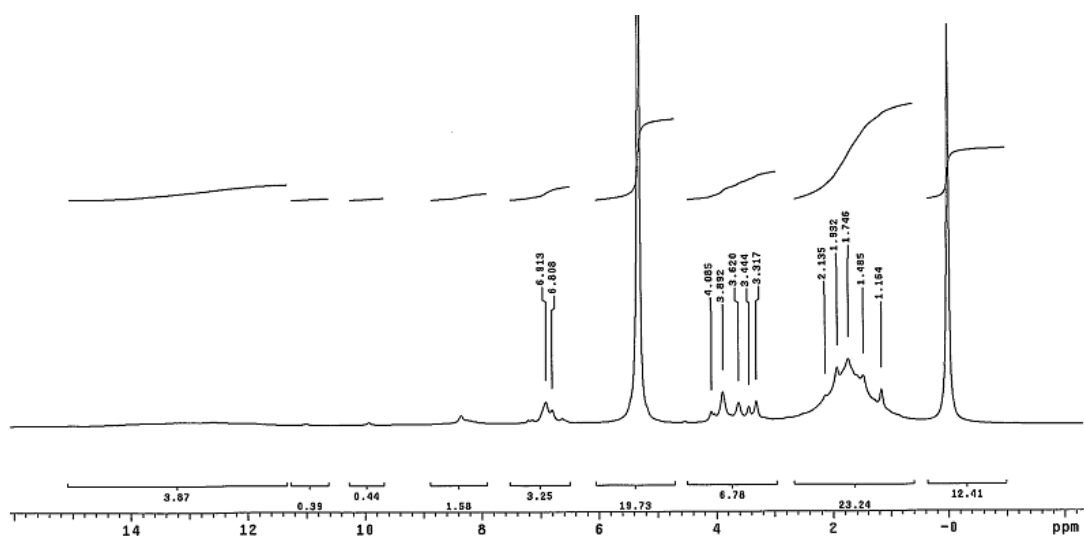


(b)

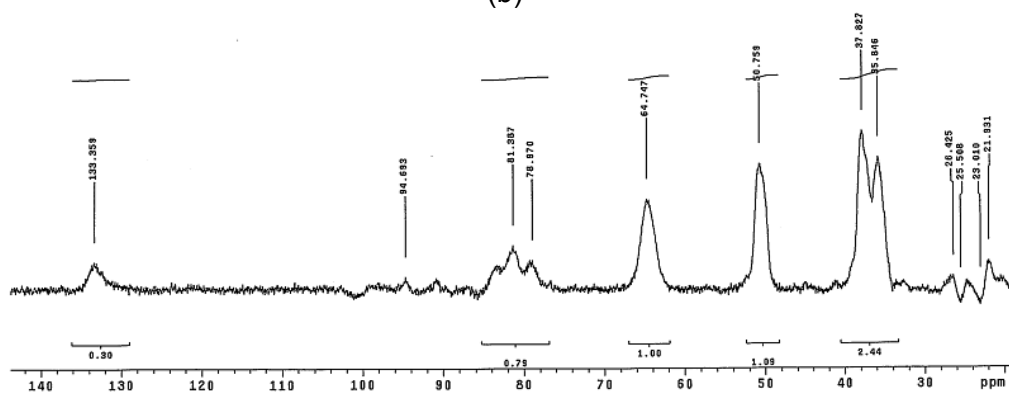
**Figure S3:**  $^1\text{H}$  NMR spectrum of **1** in  $\text{CD}_2\text{Cl}_2$ .



(a)

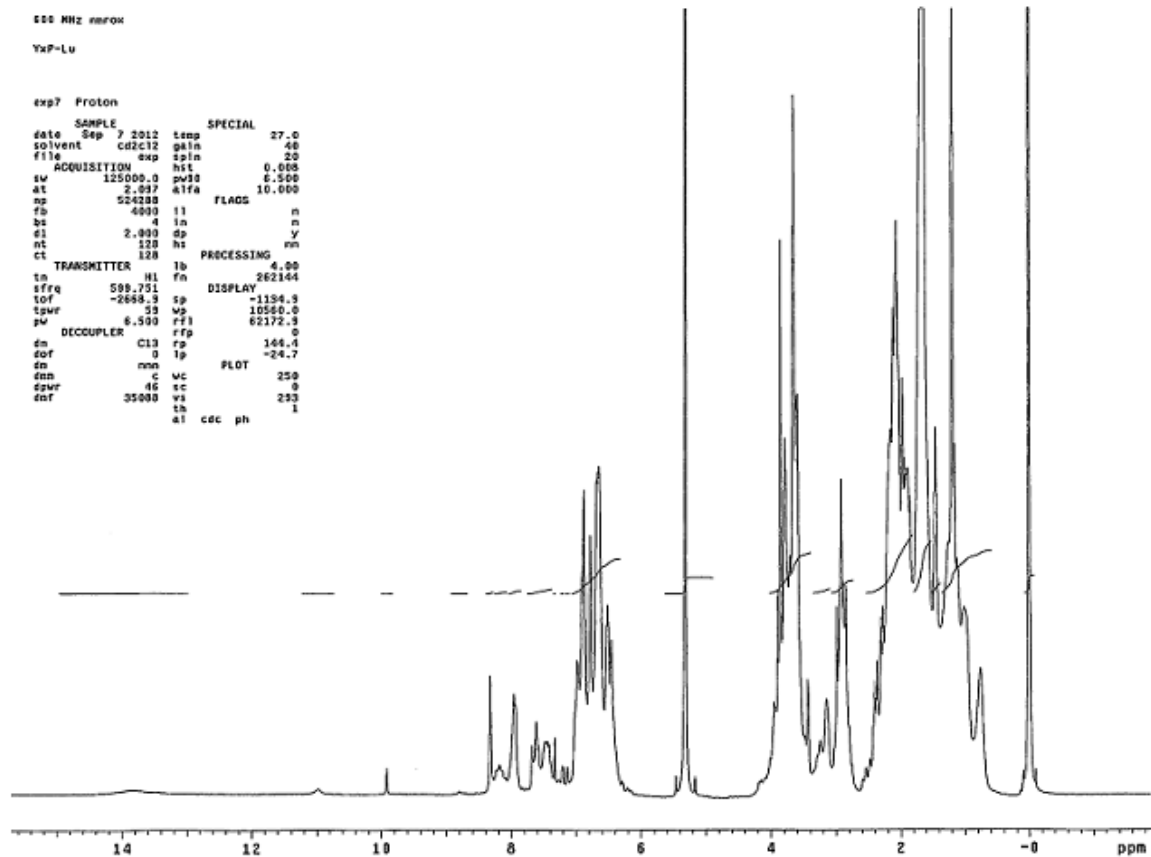


(b)

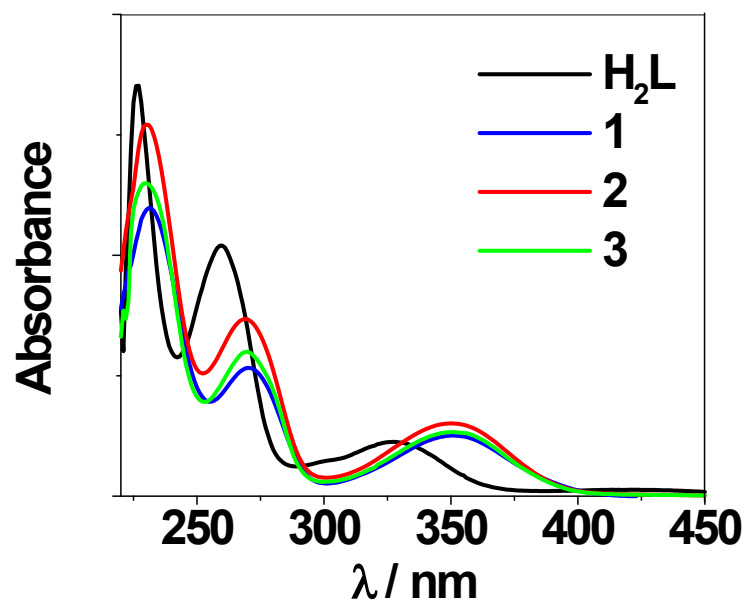


(c)

**Figure S4:**  $^1\text{H}$  NMR spectrum of **2** in  $\text{CD}_2\text{Cl}_2$ .



**Figure S5:**  $^1\text{H}$  NMR spectrum of **3** in  $\text{CD}_2\text{Cl}_2$ .



**Figure S6.** UV-Vis spectra of  $H_2L$  and complexes **1-3** in  $\text{CH}_2\text{Cl}_2$  ( $C = 10^{-6} - 10^{-5} \text{ M}$ ).

**Table S1.** Crystal data and structure refinement for complexes **1-3**.

|                                                                | <b>1</b>                                                                                                            | <b>2</b>                                                                                                            | <b>3</b>                                                                                                            |
|----------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------------------------|
| Formula                                                        | C <sub>274</sub> H <sub>385</sub> Cd <sub>18</sub> Cl <sub>8</sub> Eu <sub>6</sub> N <sub>18</sub> O <sub>110</sub> | C <sub>271</sub> H <sub>371</sub> Cd <sub>18</sub> Cl <sub>8</sub> Tb <sub>6</sub> N <sub>18</sub> O <sub>106</sub> | C <sub>269</sub> H <sub>370</sub> Cd <sub>18</sub> Cl <sub>8</sub> Lu <sub>6</sub> N <sub>18</sub> O <sub>102</sub> |
| Fw                                                             | 8900.72                                                                                                             | 8832.84                                                                                                             | 8900.84                                                                                                             |
| Crystal system                                                 | Hexagonal                                                                                                           | Hexagonal                                                                                                           | Monoclinic                                                                                                          |
| Space group                                                    | P-3                                                                                                                 | P-3                                                                                                                 | C2/c                                                                                                                |
| <i>a</i> [Å]                                                   | 51.304(7)                                                                                                           | 51.114(7)                                                                                                           | 41.871(8)                                                                                                           |
| <i>b</i> [Å]                                                   | 51.304(7)                                                                                                           | 51.114(7)                                                                                                           | 22.803(5)                                                                                                           |
| <i>c</i> [Å]                                                   | 24.418(5)                                                                                                           | 24.362(5)                                                                                                           | 47.322(10)                                                                                                          |
| $\alpha$ [deg]                                                 | 90.00                                                                                                               | 90.00                                                                                                               | 90.00                                                                                                               |
| $\beta$ [deg]                                                  | 90.00                                                                                                               | 90.00                                                                                                               | 112.91(3)                                                                                                           |
| $\gamma$ [deg]                                                 | 120.00                                                                                                              | 120.00                                                                                                              | 90.00                                                                                                               |
| <i>V</i> [Å <sup>3</sup> ]                                     | 55660(16)                                                                                                           | 55123(16)                                                                                                           | 41618(14)                                                                                                           |
| <i>d</i> / [g/cm <sup>3</sup> ]                                | 1.593                                                                                                               | 1.597                                                                                                               | 1.421                                                                                                               |
| <i>Z</i>                                                       | 6                                                                                                                   | 6                                                                                                                   | 4                                                                                                                   |
| <i>T</i> [K]                                                   | 153(1)                                                                                                              | 153(1)                                                                                                              | 153(1)                                                                                                              |
| <i>F</i> (000)                                                 | 26450                                                                                                               | 26153                                                                                                               | 17514                                                                                                               |
| $\mu$ , mm <sup>-1</sup>                                       | 2.138                                                                                                               | 2.288                                                                                                               | 2.435                                                                                                               |
| $\theta$ rang, deg                                             | 3.00-25.00                                                                                                          | 2.97-25.00                                                                                                          | 1.53-25.00                                                                                                          |
| reflns meads                                                   | 449154                                                                                                              | 368767                                                                                                              | 99822                                                                                                               |
| reflns used                                                    | 59227                                                                                                               | 60771                                                                                                               | 35936                                                                                                               |
| params                                                         | 4036                                                                                                                | 4024                                                                                                                | 2099                                                                                                                |
| R1 <sup>a</sup> , wR2 <sup>a</sup> / > 2 $\sigma$ ( <i>I</i> ) | 0.0754, 0.1806                                                                                                      | 0.0690, 0.1754                                                                                                      | 0.0727, 0.1800                                                                                                      |
| R1,                                                            | 0.1219, 0.2142                                                                                                      | 0.1155, 0.2225                                                                                                      | 0.0844, 0.1898                                                                                                      |

|                |       |       |       |
|----------------|-------|-------|-------|
| wR2 (all data) |       |       |       |
| Quality of fit | 1.021 | 1.062 | 1.108 |

<sup>a</sup> R1 =  $\Sigma|F_o| - |F_c|/\Sigma|F_o|$ . wR2 =  $[\Sigma w[(F_o^2 - F_c^2)^2]/\Sigma[w(F_o^2)^2]]^{1/2}$ .  $w=1/[\sigma^2(F_o^2)+(0.075P)^2]$ , where  $P = [\max(F_o^2, 0) + 2F_c^2]/3$ .

**Table S2.** Selected Bond Lengths (Å) and Angles (°) for **1**.

|             |           |              |           |
|-------------|-----------|--------------|-----------|
| Eu(1)-O(37) | 2.223(8)  | Eu(5)-O(7)   | 2.257(9)  |
| Eu(1)-O(2)  | 2.249(8)  | Eu(5)-O(3)   | 2.270(9)  |
| Eu(1)-O(6)  | 2.264(8)  | Eu(5)-O(81)  | 2.329(10) |
| Eu(1)-O(62) | 2.322(8)  | Eu(5)-O(74)  | 2.347(8)  |
| Eu(1)-O(39) | 2.346(10) | Eu(5)-O(78)  | 2.352(9)  |
| Eu(1)-O(58) | 2.351(9)  | Eu(5)-N(4)   | 2.505(11) |
| Eu(1)-N(1)  | 2.486(9)  | Eu(5)-O(4)   | 2.508(9)  |
| Eu(1)-O(5)  | 2.555(8)  | Eu(6)-O(19)  | 2.227(10) |
| Eu(2)-O(14) | 2.228(9)  | Eu(6)-O(69)  | 2.228(9)  |
| Eu(2)-O(47) | 2.230(8)  | Eu(6)-O(15)  | 2.258(10) |
| Eu(2)-O(18) | 2.264(9)  | Eu(6)-O(71)  | 2.287(11) |
| Eu(2)-O(46) | 2.312(9)  | Eu(6)-O(68)  | 2.290(11) |
| Eu(2)-O(44) | 2.358(8)  | Eu(6)-O(79)  | 2.346(10) |
| Eu(2)-N(7)  | 2.484(10) | Eu(6)-N(10)  | 2.483(12) |
| Eu(2)-O(17) | 2.549(8)  | Eu(6)-O(16)  | 2.537(9)  |
| Eu(2)-O(91) | 2.622(12) | Cd(16)-O(11) | 2.236(8)  |
| Eu(3)-O(55) | 2.209(9)  | Cd(16)-O(77) | 2.240(9)  |
| Eu(3)-O(26) | 2.228(9)  | Cd(16)-N(6)  | 2.254(11) |
| Eu(3)-O(30) | 2.251(9)  | Cd(16)-O(74) | 2.267(9)  |
| Eu(3)-O(54) | 2.322(10) | Cd(16)-O(7)  | 2.327(9)  |
| Eu(3)-O(50) | 2.345(9)  | Cd(16)-O(8)  | 2.602(9)  |
| Eu(3)-N(13) | 2.487(12) | Cd(7)-O(10)  | 2.248(8)  |
| Eu(3)-O(29) | 2.556(10) | Cd(7)-O(45)  | 2.273(9)  |
| Eu(3)-Cl(7) | 2.787(11) | Cd(7)-N(5)   | 2.274(11) |
| Eu(4)-O(63) | 2.201(9)  | Cd(7)-O(44)  | 2.281(9)  |
| Eu(4)-O(31) | 2.245(10) | Cd(7)-O(14)  | 2.336(8)  |
| Eu(4)-O(27) | 2.245(10) | Cd(7)-O(13)  | 2.535(9)  |
| Eu(4)-O(87) | 2.279(10) | Cd(3)-O(34)  | 2.257(9)  |
| Eu(4)-O(89) | 2.335(9)  | Cd(3)-N(17)  | 2.263(12) |
| Eu(4)-N(16) | 2.490(12) | Cd(3)-O(61)  | 2.268(9)  |
| Eu(4)-O(28) | 2.560(10) | Cd(3)-O(58)  | 2.283(9)  |
| Eu(4)-Cl(5) | 2.844(7)  | Cd(3)-O(2)   | 2.342(8)  |
| Eu(5)-O(83) | 2.241(8)  | Cd(3)-O(1)   | 2.531(9)  |

|               |           |                   |           |
|---------------|-----------|-------------------|-----------|
| Cd(2)-O(30)   | 2.293(9)  | Cd(14)-O(32)      | 2.535(10) |
| Cd(2)-N(15)   | 2.307(11) | Cd(8)-O(92)       | 2.046(16) |
| Cd(2)-O(56)   | 2.323(9)  | Cd(8)-N(9)        | 2.282(12) |
| Cd(2)-O(57)   | 2.342(9)  | Cd(8)-O(18)       | 2.292(9)  |
| Cd(2)-O(59)   | 2.404(11) | Cd(8)-O(48)       | 2.300(9)  |
| Cd(2)-Cl(7)   | 2.593(8)  | Cd(8)-O(49)       | 2.332(11) |
| Cd(6)-O(10)   | 2.269(9)  | Cd(8)-O(51)       | 2.499(11) |
| Cd(6)-O(45)   | 2.275(8)  | Cd(8)-O(91)       | 2.604(17) |
| Cd(6)-O(42)   | 2.318(10) | Cd(9)-O(22)       | 2.230(9)  |
| Cd(6)-O(41)   | 2.353(10) | Cd(9)-O(53)       | 2.277(10) |
| Cd(6)-O(9)    | 2.434(10) | Cd(9)-O(52)       | 2.295(11) |
| Cd(6)-Cl(2)   | 2.454(4)  | Cd(9)-O(21)       | 2.406(11) |
| Cd(5)-O(40)   | 2.277(13) | Cd(9)-O(51)       | 2.423(12) |
| Cd(5)-N(3)    | 2.284(11) | Cd(9)-Cl(4)       | 2.456(4)  |
| Cd(5)-O(38)   | 2.302(8)  | Cd(13)-N(14)      | 2.276(12) |
| Cd(5)-O(6)    | 2.316(8)  | Cd(13)-O(80)      | 2.294(10) |
| Cd(5)-O(43)   | 2.392(9)  | Cd(13)-O(27)      | 2.300(10) |
| Cd(5)-O(41)   | 2.496(10) | Cd(13)-O(64)      | 2.316(10) |
| Cd(5)-O(39)   | 2.598(9)  | Cd(13)-O(65)      | 2.412(12) |
| Cd(15)-O(82)  | 2.222(13) | Cd(13)-Cl(5)      | 2.543(5)  |
| Cd(15)-O(84)  | 2.281(9)  | Cd(12)-O(23)      | 2.209(11) |
| Cd(15)-N(2)   | 2.284(10) | Cd(12)-N(12)      | 2.265(14) |
| Cd(15)-O(3)   | 2.309(9)  | Cd(12)-O(67)      | 2.279(11) |
| Cd(15)-O(90)  | 2.386(10) | Cd(12)-O(79)      | 2.298(10) |
| Cd(15)-O(85)  | 2.576(12) | Cd(12)-O(19)      | 2.308(11) |
| Cd(4)-O(60)   | 2.236(10) | Cd(12)-O(20)      | 2.510(11) |
| Cd(4)-O(34)   | 2.237(9)  | Cd(1)-O(22)       | 2.232(9)  |
| Cd(4)-O(61)   | 2.303(9)  | Cd(1)-N(11)       | 2.258(13) |
| Cd(4)-O(33)   | 2.397(11) | Cd(1)-O(53)       | 2.268(10) |
| Cd(4)-Cl(3)   | 2.441(4)  | Cd(1)-O(50)       | 2.295(9)  |
| Cd(4)-O(59)   | 2.552(11) | Cd(1)-O(26)       | 2.298(9)  |
| Cd(18)-O(15)  | 2.316(9)  | Cd(1)-O(25)       | 2.509(10) |
| Cd(18)-N(8)   | 2.329(11) | Cd(11)-O(35)      | 2.234(11) |
| Cd(18)-O(70)  | 2.339(9)  | Cd(11)-O(86)      | 2.274(16) |
| Cd(18)-O(73)  | 2.356(9)  | Cd(11)-O(88)      | 2.299(10) |
| Cd(18)-O(75)  | 2.390(9)  | Cd(11)-O(85)      | 2.342(14) |
| Cd(18)-O(72)  | 2.438(11) | Cd(11)-Cl(6)      | 2.487(6)  |
| Cd(18)-O(71)  | 2.475(11) | Cd(11)-O(36)      | 2.498(12) |
| Cd(17)-O(76)  | 2.246(11) | Cd(10)-O(66)      | 2.197(15) |
| Cd(17)-O(11)  | 2.268(9)  | Cd(10)-O(23)      | 2.237(12) |
| Cd(17)-O(77)  | 2.312(9)  | Cd(10)-O(67)      | 2.273(12) |
| Cd(17)-O(75)  | 2.416(9)  | Cd(10)-Cl(8')     | 2.38(3)   |
| Cd(17)-Cl(1)  | 2.446(4)  | Cd(10)-O(24)      | 2.414(15) |
| Cd(17)-O(12)  | 2.474(10) | Cd(10)-Cl(8)      | 2.47(2)   |
| Cd(17)-C(248) | 2.707(16) | Cd(10)-O(65)      | 2.584(13) |
| Cd(14)-O(35)  | 2.227(10) | O(37)-Eu(1)-O(2)  | 114.6(3)  |
| Cd(14)-N(18)  | 2.230(13) | O(37)-Eu(1)-O(6)  | 86.2(3)   |
| Cd(14)-O(88)  | 2.254(11) | O(2)-Eu(1)-O(6)   | 137.0(3)  |
| Cd(14)-O(89)  | 2.288(9)  | O(37)-Eu(1)-O(62) | 148.4(3)  |
| Cd(14)-O(31)  | 2.329(10) | O(2)-Eu(1)-O(62)  | 80.6(3)   |

|                   |          |                   |          |
|-------------------|----------|-------------------|----------|
| O(6)-Eu(1)-O(62)  | 101.4(3) | O(18)-Eu(2)-O(17) | 64.2(3)  |
| O(37)-Eu(1)-O(39) | 77.2(3)  | O(46)-Eu(2)-O(17) | 69.6(3)  |
| O(2)-Eu(1)-O(39)  | 144.6(3) | O(44)-Eu(2)-O(17) | 136.6(3) |
| O(6)-Eu(1)-O(39)  | 74.0(3)  | N(7)-Eu(2)-O(17)  | 73.0(3)  |
| O(62)-Eu(1)-O(39) | 75.5(3)  | O(14)-Eu(2)-O(91) | 145.3(4) |
| O(37)-Eu(1)-O(58) | 78.3(3)  | O(47)-Eu(2)-O(91) | 78.2(4)  |
| O(2)-Eu(1)-O(58)  | 69.6(3)  | O(18)-Eu(2)-O(91) | 75.0(4)  |
| O(6)-Eu(1)-O(58)  | 153.3(3) | O(46)-Eu(2)-O(91) | 73.6(4)  |
| O(62)-Eu(1)-O(58) | 82.0(3)  | O(44)-Eu(2)-O(91) | 81.2(4)  |
| O(39)-Eu(1)-O(58) | 81.4(3)  | N(7)-Eu(2)-O(91)  | 139.9(4) |
| O(37)-Eu(1)-N(1)  | 74.5(3)  | O(17)-Eu(2)-O(91) | 115.5(4) |
| O(2)-Eu(1)-N(1)   | 74.2(3)  | O(55)-Eu(3)-O(26) | 114.5(3) |
| O(6)-Eu(1)-N(1)   | 76.3(3)  | O(55)-Eu(3)-O(30) | 86.9(3)  |
| O(62)-Eu(1)-N(1)  | 137.1(3) | O(26)-Eu(3)-O(30) | 134.8(3) |
| O(39)-Eu(1)-N(1)  | 139.9(3) | O(55)-Eu(3)-O(54) | 145.1(4) |
| O(58)-Eu(1)-N(1)  | 119.2(3) | O(26)-Eu(3)-O(54) | 82.7(4)  |
| O(37)-Eu(1)-O(5)  | 140.0(3) | O(30)-Eu(3)-O(54) | 102.2(4) |
| O(2)-Eu(1)-O(5)   | 77.7(3)  | O(55)-Eu(3)-O(50) | 78.7(3)  |
| O(6)-Eu(1)-O(5)   | 64.1(3)  | O(26)-Eu(3)-O(50) | 70.8(3)  |
| O(62)-Eu(1)-O(5)  | 68.2(3)  | O(30)-Eu(3)-O(50) | 154.3(3) |
| O(39)-Eu(1)-O(5)  | 115.8(3) | O(54)-Eu(3)-O(50) | 79.2(4)  |
| O(58)-Eu(1)-O(5)  | 138.8(3) | O(55)-Eu(3)-N(13) | 74.0(4)  |
| N(1)-Eu(1)-O(5)   | 72.8(3)  | O(26)-Eu(3)-N(13) | 73.5(4)  |
| O(14)-Eu(2)-O(47) | 114.2(3) | O(30)-Eu(3)-N(13) | 75.6(4)  |
| O(14)-Eu(2)-O(18) | 134.7(3) | O(54)-Eu(3)-N(13) | 140.9(4) |
| O(47)-Eu(2)-O(18) | 88.2(3)  | O(50)-Eu(3)-N(13) | 119.5(4) |
| O(14)-Eu(2)-O(46) | 81.1(3)  | O(55)-Eu(3)-O(29) | 142.6(3) |
| O(47)-Eu(2)-O(46) | 145.9(3) | O(26)-Eu(3)-O(29) | 76.4(3)  |
| O(18)-Eu(2)-O(46) | 102.5(3) | O(30)-Eu(3)-O(29) | 64.6(3)  |
| O(14)-Eu(2)-O(44) | 70.7(3)  | O(54)-Eu(3)-O(29) | 68.7(4)  |
| O(47)-Eu(2)-O(44) | 78.3(3)  | O(50)-Eu(3)-O(29) | 136.4(3) |
| O(18)-Eu(2)-O(44) | 154.6(3) | N(13)-Eu(3)-O(29) | 75.7(4)  |
| O(46)-Eu(2)-O(44) | 78.7(3)  | O(55)-Eu(3)-Cl(7) | 77.9(3)  |
| O(14)-Eu(2)-N(7)  | 74.0(3)  | O(26)-Eu(3)-Cl(7) | 146.3(3) |
| O(47)-Eu(2)-N(7)  | 75.0(3)  | O(30)-Eu(3)-Cl(7) | 74.1(3)  |
| O(18)-Eu(2)-N(7)  | 74.9(3)  | O(54)-Eu(3)-Cl(7) | 72.6(3)  |
| O(46)-Eu(2)-N(7)  | 139.0(3) | O(50)-Eu(3)-Cl(7) | 82.1(3)  |
| O(44)-Eu(2)-N(7)  | 121.0(3) | N(13)-Eu(3)-Cl(7) | 139.3(3) |
| O(14)-Eu(2)-O(17) | 75.6(3)  | O(29)-Eu(3)-Cl(7) | 113.6(3) |
| O(47)-Eu(2)-O(17) | 142.0(3) |                   |          |

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**Table S3.** Selected Bond Lengths (Å) and Angles (°) for **2**.

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|             |           |              |           |
|-------------|-----------|--------------|-----------|
| Tb(1)-O(37) | 2.224(8)  | Tb(6)-O(16)  | 2.543(9)  |
| Tb(1)-O(2)  | 2.250(8)  | Cd(1)-O(22)  | 2.240(9)  |
| Tb(1)-O(6)  | 2.258(8)  | Cd(1)-N(11)  | 2.253(13) |
| Tb(1)-O(62) | 2.315(8)  | Cd(1)-O(53)  | 2.265(10) |
| Tb(1)-O(58) | 2.344(8)  | Cd(1)-O(50)  | 2.284(9)  |
| Tb(1)-O(39) | 2.344(10) | Cd(1)-O(26)  | 2.286(9)  |
| Tb(1)-N(1)  | 2.460(9)  | Cd(1)-O(25)  | 2.507(10) |
| Tb(1)-O(5)  | 2.539(8)  | Cd(2)-O(30)  | 2.284(9)  |
| Tb(2)-O(47) | 2.225(8)  | Cd(2)-N(15)  | 2.297(10) |
| Tb(2)-O(14) | 2.228(8)  | Cd(2)-O(56)  | 2.316(8)  |
| Tb(2)-O(18) | 2.251(8)  | Cd(2)-O(57)  | 2.332(8)  |
| Tb(2)-O(46) | 2.307(8)  | Cd(2)-O(59)  | 2.403(10) |
| Tb(2)-O(44) | 2.347(7)  | Cd(2)-Cl(7)  | 2.581(7)  |
| Tb(2)-N(7)  | 2.489(9)  | Cd(3)-O(34)  | 2.259(9)  |
| Tb(2)-O(17) | 2.540(8)  | Cd(3)-O(61)  | 2.266(9)  |
| Tb(2)-O(91) | 2.610(10) | Cd(3)-N(17)  | 2.269(11) |
| Tb(3)-O(55) | 2.210(8)  | Cd(3)-O(58)  | 2.281(8)  |
| Tb(3)-O(26) | 2.226(9)  | Cd(3)-O(2)   | 2.350(8)  |
| Tb(3)-O(30) | 2.249(9)  | Cd(3)-O(1)   | 2.528(8)  |
| Tb(3)-O(54) | 2.302(10) | Cd(4)-O(60)  | 2.225(10) |
| Tb(3)-O(50) | 2.335(9)  | Cd(4)-O(34)  | 2.233(9)  |
| Tb(3)-N(13) | 2.478(11) | Cd(4)-O(61)  | 2.297(9)  |
| Tb(3)-O(29) | 2.554(9)  | Cd(4)-O(33)  | 2.396(10) |
| Tb(3)-Cl(7) | 2.764(10) | Cd(4)-Cl(3)  | 2.438(4)  |
| Tb(4)-O(63) | 2.207(8)  | Cd(4)-O(59)  | 2.545(10) |
| Tb(4)-O(27) | 2.229(9)  | Cd(4)-C(222) | 2.758(15) |
| Tb(4)-O(31) | 2.235(10) | Cd(5)-N(3)   | 2.284(10) |
| Tb(4)-O(87) | 2.284(10) | Cd(5)-O(40)  | 2.285(12) |
| Tb(4)-O(89) | 2.329(9)  | Cd(5)-O(38)  | 2.302(8)  |
| Tb(4)-N(16) | 2.485(11) | Cd(5)-O(6)   | 2.320(7)  |
| Tb(4)-O(28) | 2.548(9)  | Cd(5)-O(43)  | 2.401(8)  |
| Tb(4)-Cl(5) | 2.825(7)  | Cd(5)-O(41)  | 2.498(9)  |
| Tb(5)-O(7)  | 2.250(8)  | Cd(5)-O(39)  | 2.601(9)  |
| Tb(5)-O(83) | 2.251(8)  | Cd(6)-O(10)  | 2.257(8)  |
| Tb(5)-O(3)  | 2.271(8)  | Cd(6)-O(45)  | 2.281(8)  |
| Tb(5)-O(81) | 2.302(10) | Cd(6)-O(42)  | 2.292(9)  |
| Tb(5)-O(74) | 2.347(8)  | Cd(6)-O(41)  | 2.336(9)  |
| Tb(5)-O(78) | 2.349(9)  | Cd(6)-Cl(2)  | 2.442(4)  |
| Tb(5)-N(4)  | 2.493(11) | Cd(6)-O(9)   | 2.445(9)  |
| Tb(5)-O(4)  | 2.500(8)  | Cd(6)-C(204) | 2.664(14) |
| Tb(6)-O(19) | 2.221(10) | Cd(7)-O(10)  | 2.247(8)  |
| Tb(6)-O(69) | 2.223(9)  | Cd(7)-O(45)  | 2.267(8)  |
| Tb(6)-O(15) | 2.241(9)  | Cd(7)-N(5)   | 2.279(11) |
| Tb(6)-O(71) | 2.284(11) | Cd(7)-O(44)  | 2.281(8)  |
| Tb(6)-O(68) | 2.294(10) | Cd(7)-O(14)  | 2.332(7)  |
| Tb(6)-O(79) | 2.340(10) | Cd(7)-O(13)  | 2.524(8)  |
| Tb(6)-N(10) | 2.472(12) | Cd(8)-O(92)  | 2.060(13) |

|               |           |                   |           |
|---------------|-----------|-------------------|-----------|
| Cd(8)-N(9)    | 2.283(12) | Cd(16)-O(11)      | 2.237(8)  |
| Cd(8)-O(18)   | 2.288(8)  | Cd(16)-N(6)       | 2.241(10) |
| Cd(8)-O(48)   | 2.291(8)  | Cd(16)-O(77)      | 2.245(9)  |
| Cd(8)-O(49)   | 2.326(11) | Cd(16)-O(74)      | 2.261(8)  |
| Cd(8)-O(51)   | 2.494(11) | Cd(16)-O(7)       | 2.314(8)  |
| Cd(8)-O(91)   | 2.574(13) | Cd(16)-O(8)       | 2.595(9)  |
| Cd(9)-O(22)   | 2.222(10) | Cd(17)-O(76)      | 2.245(11) |
| Cd(9)-O(53)   | 2.272(9)  | Cd(17)-O(11)      | 2.254(8)  |
| Cd(9)-O(52)   | 2.305(11) | Cd(17)-O(77)      | 2.312(9)  |
| Cd(9)-O(51)   | 2.412(12) | Cd(17)-O(75)      | 2.414(10) |
| Cd(9)-O(21)   | 2.417(11) | Cd(17)-Cl(1)      | 2.435(4)  |
| Cd(9)-Cl(4)   | 2.445(4)  | Cd(17)-O(12)      | 2.478(10) |
| Cd(9)-C(214)  | 2.733(17) | Cd(17)-C(248)     | 2.702(16) |
| Cd(10)-O(66)  | 2.182(16) | Cd(18)-O(15)      | 2.315(9)  |
| Cd(10)-O(23)  | 2.231(12) | Cd(18)-N(8)       | 2.320(11) |
| Cd(10)-O(67)  | 2.269(12) | Cd(18)-O(70)      | 2.332(8)  |
| Cd(10)-Cl(8') | 2.32(3)   | Cd(18)-O(73)      | 2.339(9)  |
| Cd(10)-O(24)  | 2.405(15) | Cd(18)-O(75)      | 2.382(9)  |
| Cd(10)-Cl(8)  | 2.50(3)   | Cd(18)-O(72)      | 2.452(12) |
| Cd(10)-O(65)  | 2.571(13) | Cd(18)-O(71)      | 2.478(11) |
| Cd(11)-O(35)  | 2.227(10) | O(37)-Tb(1)-O(2)  | 114.5(3)  |
| Cd(11)-O(86)  | 2.268(17) | O(37)-Tb(1)-O(6)  | 86.1(3)   |
| Cd(11)-O(88)  | 2.287(10) | O(2)-Tb(1)-O(6)   | 136.4(3)  |
| Cd(11)-O(85)  | 2.309(14) | O(37)-Tb(1)-O(62) | 148.3(3)  |
| Cd(11)-Cl(6)  | 2.479(7)  | O(2)-Tb(1)-O(62)  | 81.0(3)   |
| Cd(11)-O(36)  | 2.505(12) | O(6)-Tb(1)-O(62)  | 101.6(3)  |
| Cd(12)-O(23)  | 2.209(12) | O(37)-Tb(1)-O(58) | 78.4(3)   |
| Cd(12)-N(12)  | 2.258(14) | O(2)-Tb(1)-O(58)  | 70.1(3)   |
| Cd(12)-O(67)  | 2.269(11) | O(6)-Tb(1)-O(58)  | 153.4(3)  |
| Cd(12)-O(79)  | 2.290(10) | O(62)-Tb(1)-O(58) | 81.9(3)   |
| Cd(12)-O(19)  | 2.298(11) | O(37)-Tb(1)-O(39) | 77.5(3)   |
| Cd(12)-O(20)  | 2.520(12) | O(2)-Tb(1)-O(39)  | 144.7(3)  |
| Cd(13)-N(14)  | 2.282(12) | O(6)-Tb(1)-O(39)  | 74.7(3)   |
| Cd(13)-O(80)  | 2.283(10) | O(62)-Tb(1)-O(39) | 75.1(3)   |
| Cd(13)-O(27)  | 2.299(10) | O(58)-Tb(1)-O(39) | 81.0(3)   |
| Cd(13)-O(64)  | 2.310(9)  | O(37)-Tb(1)-N(1)  | 74.7(3)   |
| Cd(13)-O(65)  | 2.414(12) | O(2)-Tb(1)-N(1)   | 73.8(3)   |
| Cd(13)-Cl(5)  | 2.530(6)  | O(6)-Tb(1)-N(1)   | 75.8(3)   |
| Cd(14)-N(18)  | 2.225(13) | O(62)-Tb(1)-N(1)  | 136.9(3)  |
| Cd(14)-O(35)  | 2.226(10) | O(58)-Tb(1)-N(1)  | 119.6(3)  |
| Cd(14)-O(88)  | 2.246(10) | O(39)-Tb(1)-N(1)  | 140.4(3)  |
| Cd(14)-O(89)  | 2.284(9)  | O(37)-Tb(1)-O(5)  | 139.9(3)  |
| Cd(14)-O(31)  | 2.317(10) | O(2)-Tb(1)-O(5)   | 77.5(3)   |
| Cd(14)-O(32)  | 2.527(10) | O(6)-Tb(1)-O(5)   | 63.9(3)   |
| Cd(15)-O(82)  | 2.224(12) | O(62)-Tb(1)-O(5)  | 68.3(3)   |
| Cd(15)-N(2)   | 2.254(10) | O(58)-Tb(1)-O(5)  | 138.9(3)  |
| Cd(15)-O(84)  | 2.274(8)  | O(39)-Tb(1)-O(5)  | 115.7(3)  |
| Cd(15)-O(3)   | 2.292(8)  | N(1)-Tb(1)-O(5)   | 72.5(3)   |
| Cd(15)-O(90)  | 2.379(9)  | O(47)-Tb(2)-O(14) | 114.1(3)  |
| Cd(15)-O(85)  | 2.573(12) | O(47)-Tb(2)-O(18) | 88.4(3)   |

|                   |          |                   |          |
|-------------------|----------|-------------------|----------|
| O(14)-Tb(2)-O(18) | 134.6(3) | O(55)-Tb(3)-O(30) | 87.3(3)  |
| O(47)-Tb(2)-O(46) | 146.2(3) | O(26)-Tb(3)-O(30) | 135.0(3) |
| O(14)-Tb(2)-O(46) | 81.2(3)  | O(55)-Tb(3)-O(54) | 144.5(4) |
| O(18)-Tb(2)-O(46) | 102.0(3) | O(26)-Tb(3)-O(54) | 82.7(4)  |
| O(47)-Tb(2)-O(44) | 78.1(3)  | O(30)-Tb(3)-O(54) | 102.2(4) |
| O(14)-Tb(2)-O(44) | 70.8(3)  | O(55)-Tb(3)-O(50) | 78.1(3)  |
| O(18)-Tb(2)-O(44) | 154.6(3) | O(26)-Tb(3)-O(50) | 70.4(3)  |
| O(46)-Tb(2)-O(44) | 79.2(3)  | O(30)-Tb(3)-O(50) | 154.6(3) |
| O(47)-Tb(2)-N(7)  | 75.0(3)  | O(54)-Tb(3)-O(50) | 79.3(4)  |
| O(14)-Tb(2)-N(7)  | 74.0(3)  | O(55)-Tb(3)-N(13) | 74.5(3)  |
| O(18)-Tb(2)-N(7)  | 75.0(3)  | O(26)-Tb(3)-N(13) | 73.5(3)  |
| O(46)-Tb(2)-N(7)  | 138.8(3) | O(30)-Tb(3)-N(13) | 75.7(4)  |
| O(44)-Tb(2)-N(7)  | 120.9(3) | O(54)-Tb(3)-N(13) | 140.9(4) |
| O(47)-Tb(2)-O(17) | 141.7(3) | O(50)-Tb(3)-N(13) | 119.1(3) |
| O(14)-Tb(2)-O(17) | 75.5(3)  | O(55)-Tb(3)-O(29) | 143.0(3) |
| O(18)-Tb(2)-O(17) | 64.0(3)  | O(26)-Tb(3)-O(29) | 76.8(3)  |
| O(46)-Tb(2)-O(17) | 69.6(3)  | O(30)-Tb(3)-O(29) | 64.3(3)  |
| O(44)-Tb(2)-O(17) | 136.9(3) | O(54)-Tb(3)-O(29) | 68.9(4)  |
| N(7)-Tb(2)-O(17)  | 72.6(3)  | O(50)-Tb(3)-O(29) | 136.6(3) |
| O(47)-Tb(2)-O(91) | 78.4(3)  | N(13)-Tb(3)-O(29) | 75.6(4)  |
| O(14)-Tb(2)-O(91) | 145.7(3) | O(55)-Tb(3)-Cl(7) | 77.8(3)  |
| O(18)-Tb(2)-O(91) | 74.6(3)  | O(26)-Tb(3)-Cl(7) | 146.1(3) |
| O(46)-Tb(2)-O(91) | 73.7(4)  | O(30)-Tb(3)-Cl(7) | 74.1(3)  |
| O(44)-Tb(2)-O(91) | 81.6(3)  | O(54)-Tb(3)-Cl(7) | 72.4(3)  |
| N(7)-Tb(2)-O(91)  | 139.7(4) | O(50)-Tb(3)-Cl(7) | 82.4(3)  |
| O(17)-Tb(2)-O(91) | 115.4(3) | N(13)-Tb(3)-Cl(7) | 139.5(3) |
| O(55)-Tb(3)-O(26) | 114.4(3) | O(29)-Tb(3)-Cl(7) | 113.4(3) |

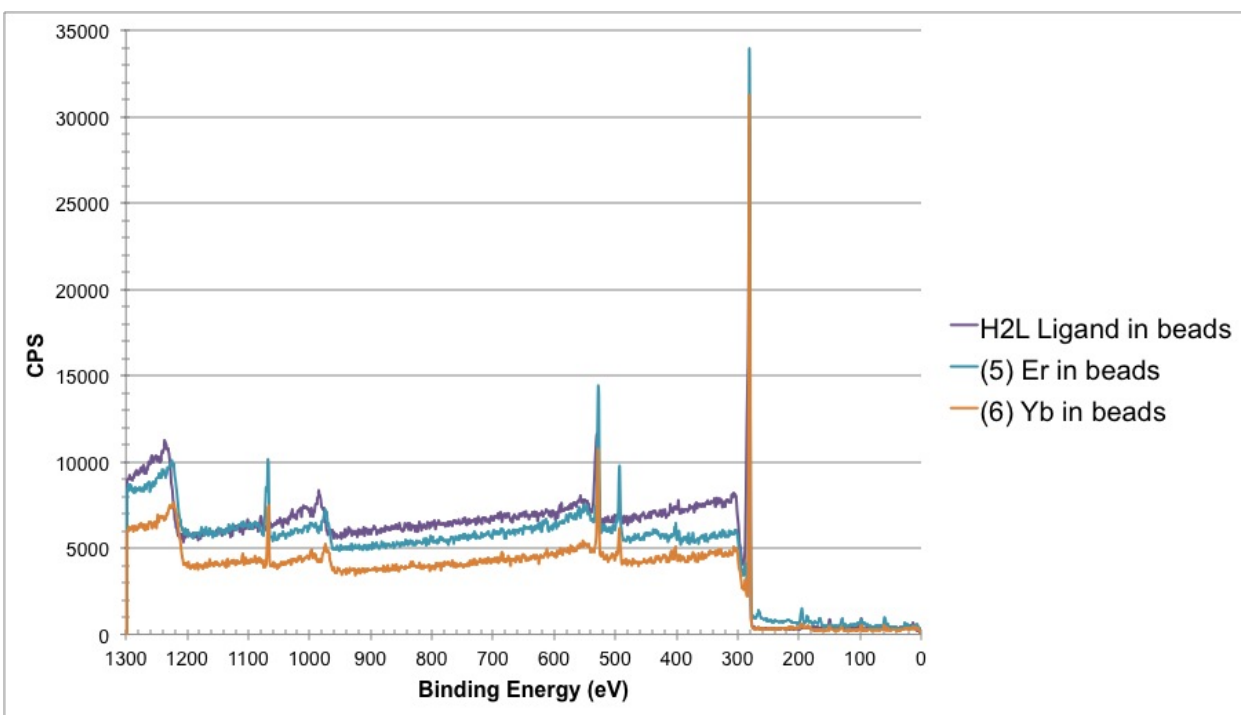
**Table S4.** Selected Bond Lengths (Å) and Angles (°) for **3**.

|             |          |             |           |
|-------------|----------|-------------|-----------|
| Lu(1)-O(27) | 2.211(6) | Lu(2)-N(1)  | 2.503(9)  |
| Lu(1)-O(10) | 2.239(6) | Lu(2)-O(13) | 2.533(7)  |
| Lu(1)-O(11) | 2.249(6) | Lu(2)-Cl(1) | 2.638(6)  |
| Lu(1)-O(26) | 2.282(7) | Lu(3)-O(35) | 2.221(7)  |
| Lu(1)-O(24) | 2.348(6) | Lu(3)-O(15) | 2.232(7)  |
| Lu(1)-N(6)  | 2.486(8) | Lu(3)-O(3)  | 2.246(7)  |
| Lu(1)-O(9)  | 2.550(6) | Lu(3)-O(34) | 2.307(7)  |
| Lu(1)-Cl(2) | 2.877(5) | Lu(3)-O(37) | 2.356(10) |
| Lu(2)-O(19) | 2.205(6) | Lu(3)-O(32) | 2.361(7)  |
| Lu(2)-O(2)  | 2.239(7) | Lu(3)-N(8)  | 2.488(8)  |
| Lu(2)-O(14) | 2.258(6) | Lu(3)-O(4)  | 2.569(7)  |
| Lu(2)-O(42) | 2.296(7) | Cd(1)-O(6)  | 2.251(6)  |
| Lu(2)-O(44) | 2.347(6) | Cd(1)-O(22) | 2.268(7)  |

|             |           |                   |            |
|-------------|-----------|-------------------|------------|
| Cd(1)-O(25) | 2.294(6)  | Cd(9)-O(24)       | 2.283(7)   |
| Cd(1)-O(5)  | 2.420(7)  | Cd(9)-O(11)       | 2.330(6)   |
| Cd(1)-O(21) | 2.440(8)  | Cd(9)-O(12)       | 2.493(7)   |
| Cd(1)-Cl(5) | 2.451(3)  | O(27)-Lu(1)-O(10) | 86.4(2)    |
| Cd(2)-N(7)  | 2.277(8)  | O(27)-Lu(1)-O(11) | 114.1(2)   |
| Cd(2)-O(14) | 2.304(6)  | O(10)-Lu(1)-O(11) | 135.7(2)   |
| Cd(2)-O(20) | 2.308(6)  | O(27)-Lu(1)-O(26) | 145.6(3)   |
| Cd(2)-O(23) | 2.382(7)  | O(10)-Lu(1)-O(26) | 101.5(2)   |
| Cd(2)-O(21) | 2.477(8)  | O(11)-Lu(1)-O(26) | 83.4(2)    |
| Cd(2)-Cl(1) | 2.607(4)  | O(27)-Lu(1)-O(24) | 79.1(2)    |
| Cd(3)-O(18) | 2.251(7)  | O(10)-Lu(1)-O(24) | 153.4(2)   |
| Cd(3)-O(41) | 2.268(7)  | O(11)-Lu(1)-O(24) | 70.9(2)    |
| Cd(3)-N(9)  | 2.275(10) | O(26)-Lu(1)-O(24) | 79.5(2)    |
| Cd(3)-O(44) | 2.295(6)  | O(27)-Lu(1)-N(6)  | 76.0(2)    |
| Cd(3)-O(2)  | 2.321(6)  | O(10)-Lu(1)-N(6)  | 75.4(3)    |
| Cd(3)-O(1)  | 2.506(7)  | O(11)-Lu(1)-N(6)  | 72.8(2)    |
| Cd(4)-O(18) | 2.247(8)  | O(26)-Lu(1)-N(6)  | 138.4(3)   |
| Cd(4)-O(40) | 2.269(8)  | O(24)-Lu(1)-N(6)  | 121.4(2)   |
| Cd(4)-O(41) | 2.305(7)  | O(27)-Lu(1)-O(9)  | 142.1(2)   |
| Cd(4)-Cl(4) | 2.443(3)  | O(10)-Lu(1)-O(9)  | 64.3(2)    |
| Cd(4)-O(17) | 2.447(8)  | O(11)-Lu(1)-O(9)  | 77.7(2)    |
| Cd(4)-O(39) | 2.456(8)  | O(26)-Lu(1)-O(9)  | 68.2(2)    |
| Cd(5)-N(2)  | 2.283(8)  | O(24)-Lu(1)-O(9)  | 137.0(2)   |
| Cd(5)-O(36) | 2.318(6)  | N(6)-Lu(1)-O(9)   | 73.7(2)    |
| Cd(5)-O(3)  | 2.324(6)  | O(27)-Lu(1)-Cl(2) | 78.21(19)  |
| Cd(5)-O(38) | 2.361(10) | O(10)-Lu(1)-Cl(2) | 73.04(18)  |
| Cd(5)-O(43) | 2.369(7)  | O(11)-Lu(1)-Cl(2) | 146.66(18) |
| Cd(5)-O(39) | 2.465(8)  | O(26)-Lu(1)-Cl(2) | 72.4(2)    |
| Cd(5)-O(37) | 2.522(8)  | O(24)-Lu(1)-Cl(2) | 82.21(18)  |
| Cd(6)-O(7)  | 2.231(6)  | N(6)-Lu(1)-Cl(2)  | 140.0(2)   |
| Cd(6)-N(4)  | 2.257(10) | O(9)-Lu(1)-Cl(2)  | 112.40(18) |
| Cd(6)-O(32) | 2.281(7)  | O(19)-Lu(2)-O(2)  | 112.7(2)   |
| Cd(6)-O(33) | 2.284(7)  | O(19)-Lu(2)-O(14) | 88.3(2)    |
| Cd(6)-O(15) | 2.315(7)  | O(2)-Lu(2)-O(14)  | 134.7(2)   |
| Cd(6)-O(16) | 2.498(7)  | O(19)-Lu(2)-O(42) | 145.7(2)   |
| Cd(7)-O(29) | 2.238(8)  | O(2)-Lu(2)-O(42)  | 82.9(3)    |
| Cd(7)-O(7)  | 2.247(7)  | O(14)-Lu(2)-O(42) | 102.1(2)   |
| Cd(7)-O(33) | 2.272(7)  | O(19)-Lu(2)-O(44) | 77.9(2)    |
| Cd(7)-O(8)  | 2.381(10) | O(2)-Lu(2)-O(44)  | 71.0(2)    |
| Cd(7)-Cl(3) | 2.470(3)  | O(14)-Lu(2)-O(44) | 154.3(2)   |
| Cd(7)-O(30) | 2.500(8)  | O(42)-Lu(2)-O(44) | 79.1(2)    |
| Cd(8)-N(5)  | 2.290(8)  | O(19)-Lu(2)-N(1)  | 74.1(3)    |
| Cd(8)-O(10) | 2.301(6)  | O(2)-Lu(2)-N(1)   | 73.5(3)    |
| Cd(8)-O(28) | 2.328(7)  | O(14)-Lu(2)-N(1)  | 74.8(3)    |
| Cd(8)-O(31) | 2.333(7)  | O(42)-Lu(2)-N(1)  | 140.0(3)   |
| Cd(8)-O(30) | 2.458(8)  | O(44)-Lu(2)-N(1)  | 120.7(3)   |
| Cd(8)-Cl(2) | 2.546(4)  | O(19)-Lu(2)-O(13) | 142.2(2)   |
| Cd(9)-O(6)  | 2.245(6)  | O(2)-Lu(2)-O(13)  | 76.1(2)    |
| Cd(9)-N(3)  | 2.254(9)  | O(14)-Lu(2)-O(13) | 64.5(2)    |
| Cd(9)-O(25) | 2.265(7)  | O(42)-Lu(2)-O(13) | 69.5(2)    |

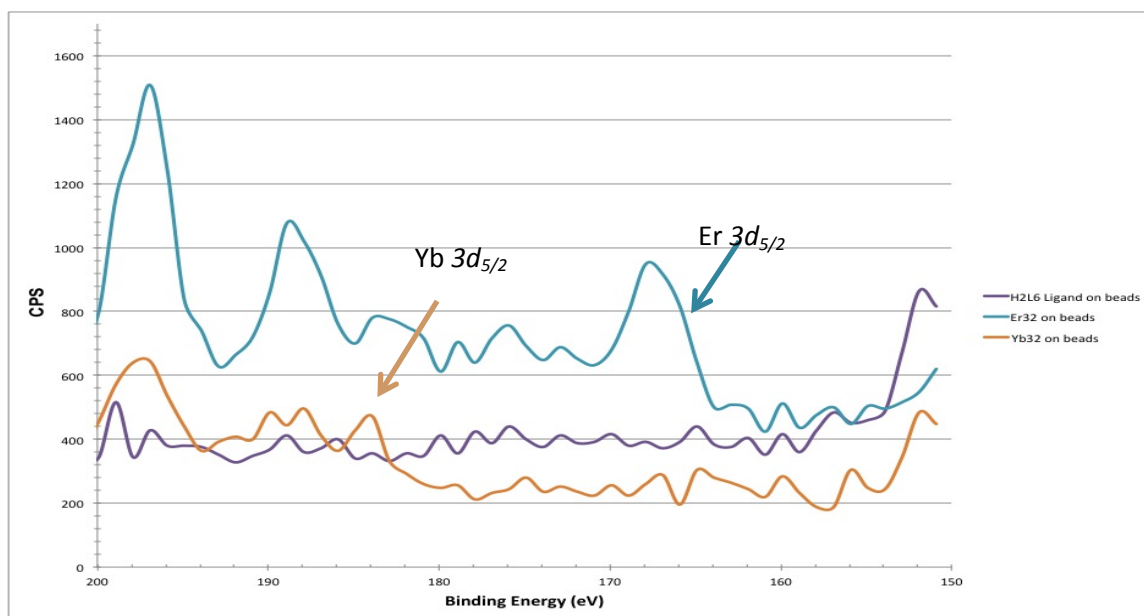
|                   |            |                   |          |
|-------------------|------------|-------------------|----------|
| O(44)-Lu(2)-O(13) | 136.7(2)   | O(35)-Lu(3)-O(32) | 77.6(2)  |
| N(1)-Lu(2)-O(13)  | 73.7(3)    | O(15)-Lu(3)-O(32) | 70.6(2)  |
| O(19)-Lu(2)-Cl(1) | 77.4(2)    | O(3)-Lu(3)-O(32)  | 150.6(2) |
| O(2)-Lu(2)-Cl(1)  | 146.84(19) | O(34)-Lu(3)-O(32) | 81.6(3)  |
| O(14)-Lu(2)-Cl(1) | 74.61(19)  | O(37)-Lu(3)-O(32) | 79.0(3)  |
| O(42)-Lu(2)-Cl(1) | 74.2(2)    | O(35)-Lu(3)-N(8)  | 75.1(3)  |
| O(44)-Lu(2)-Cl(1) | 81.19(19)  | O(15)-Lu(3)-N(8)  | 73.1(3)  |
| N(1)-Lu(2)-Cl(1)  | 138.3(2)   | O(3)-Lu(3)-N(8)   | 78.0(3)  |
| O(13)-Lu(2)-Cl(1) | 116.2(2)   | O(34)-Lu(3)-N(8)  | 135.8(3) |
| O(35)-Lu(3)-O(15) | 112.5(3)   | O(37)-Lu(3)-N(8)  | 141.9(3) |
| O(35)-Lu(3)-O(3)  | 87.0(2)    | O(32)-Lu(3)-N(8)  | 120.7(3) |
| O(15)-Lu(3)-O(3)  | 138.7(2)   | O(35)-Lu(3)-O(4)  | 139.3(3) |
| O(35)-Lu(3)-O(34) | 149.0(3)   | O(15)-Lu(3)-O(4)  | 79.4(2)  |
| O(15)-Lu(3)-O(34) | 81.3(3)    | O(3)-Lu(3)-O(4)   | 63.8(2)  |
| O(3)-Lu(3)-O(34)  | 100.6(3)   | O(34)-Lu(3)-O(4)  | 68.6(3)  |
| O(35)-Lu(3)-O(37) | 78.8(3)    | O(37)-Lu(3)-O(4)  | 115.3(3) |
| O(15)-Lu(3)-O(37) | 143.7(3)   | O(32)-Lu(3)-O(4)  | 140.5(2) |
| O(3)-Lu(3)-O(37)  | 73.5(3)    | N(8)-Lu(3)-O(4)   | 71.7(3)  |
| O(34)-Lu(3)-O(37) | 74.8(3)    |                   |          |

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XPS Surveys:

**Figure S7.** XPS survey scans of H<sub>2</sub>L ligand, (5), and (6) in beads



**Figure S8.** A closer look at the Er 4d<sub>5/2</sub> and Yb 4d<sub>5/2</sub> binding energies via XPS.

**Table S5.** Percent composition of Ln/Cd in beads

|                     | Er $4d_{5/2}$ | Yb $4d_{5/2}$ | Cd $3d_{3/2}$ |
|---------------------|---------------|---------------|---------------|
| Binding Energy (eV) | 168.1 eV      | 184.2 eV      | 405.6 eV      |
| 5 (Er) in beads     | 23.83 %       |               | 76.17 %       |
| 6 (Yb) in beads     |               | 26.78 %       | 73.22 %       |

**Figure S8.** A closer look at the Er  $4d_{5/2}$  and Yb  $4d_{5/2}$  binding energies via XPS.

