

Supplementary Information for

## Trinuclear Nickel-Lanthanide Compounds

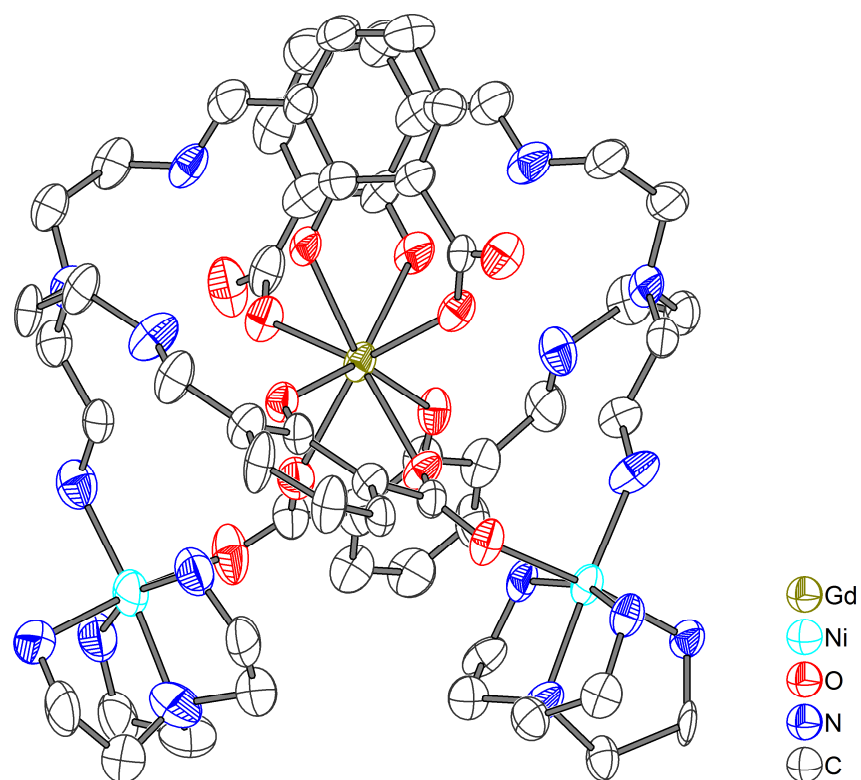
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**Figure S1.** Thermal ellipsoid plot of the molecular structure of the cation of **1**. Thermal ellipsoids are drawn at the 50% probability level. Hydrogen atoms, water molecules, and  $\text{NO}_3^-$  ions are omitted for clarity.

Table S1. Crystal data and structure refinement for **1**.

|                                    |  |                  |
|------------------------------------|--|------------------|
| Crystal data                       |  |                  |
| Identification code                | 484  |                  |
| Habitus, colour                    | needle, colourless   |                  |
| Crystal size                       | 0.41 x 0.08 x 0.07 mm <sup>3</sup>   |                  |
| Crystal system                     | Triclinic  |                  |
| Space group                        | P -1   | Z = 2            |
| Unit cell dimensions               | a = 15.9709(9) Å   | α = 109.136(4)°. |
|                                    | b = 16.0363(7) Å   | β = 112.815(4)°. |
|                                    | c = 20.1336(11) Å  | γ = 88.400(4)°.  |
| Volume                             | 4461.2(4) Å <sup>3</sup>   |                  |
| Cell determination                 | 24560 peaks with Theta 1.4 to 24.2°.   |                  |
| Empirical formula                  | C <sub>56</sub> H <sub>124</sub> GdN <sub>19</sub> Ni <sub>2</sub> O <sub>41</sub><br>C <sub>56</sub> H <sub>84</sub> GdN <sub>16</sub> Ni <sub>2</sub> O <sub>12</sub> , 3(N O <sub>3</sub> ), 20(H <sub>2</sub> O) |                  |
| Formula weight                     | 1994.41  |                  |
| Density (calculated)               | 1.485 Mg/m <sup>3</sup>  |                  |
| Absorption coefficient             | 1.249 mm <sup>-1</sup>   |                  |
| F(000)                             | 2082   |                  |
| Data collection:                   |  |                  |
| Diffractometer type                | STOE IPDS 2T   |                  |
| Wavelength                         | 0.71073 Å  |                  |
| Temperature                        | 100(2) K   |                  |
| Theta range for data collection    | 1.35 to 25.00°.  |                  |
| Index ranges                       | -18<=h<=17, -19<=k<=18, 0<=l<=23   |                  |
| Data collection software           | STOE X-AREA (STOE & Cie, 2006)   |                  |
| Cell refinement software           | STOE X-AREA  |                  |
| Data reduction software            | STOE X-AREA  |                  |
| Solution and refinement:           |  |                  |
| Reflections collected              | 38926  |                  |
| Independent reflections            | 15625 [R(int) = 0.097]   |                  |
| Completeness to theta = 25.00°     | 99.6 %   |                  |
| Observed reflections               | 8108[I>2sigma(I)]  |                  |
| Reflections used for refinement    | 15625  |                  |
| Absorption correction              | Semi-empirical from equivalents  |                  |
| Max. and min. transmission         | 0.8773 and 0.5314  |                  |
| Largest diff. peak and hole        | 4.354 and -1.510 e.Å <sup>-3</sup>   |                  |
| Solution                           | Lu compound as start model   |                  |
| Refinement                         | Full-matrix least-squares on F <sup>2</sup>  |                  |
| Treatment of hydrogen atoms        | Calculated positions, riding model   |                  |
| Programs used                      | SHELXL-97 (Sheldrick, 1997)  |                  |
|                                    | DIAMOND (Crystal Impact)   |                  |
|                                    | STOE IPDS2 software  |                  |
| Data / restraints / parameters     | 15625 / 126 / 892  |                  |
| Goodness-of-fit on F <sup>2</sup>  | 0.957  |                  |
| R index (all data)                 | wR2 = 0.1929   |                  |
| R index conventional [I>2sigma(I)] | R1 = 0.0819  |                  |

Table S2. Bond lengths [Å] and angles [°] for **1**.

|          |           |          |           |
|----------|-----------|----------|-----------|
| C1-O1    | 1.265(12) | C29-O9   | 1.314(11) |
| C1-O2    | 1.300(13) | C29-C30  | 1.372(13) |
| C1-C2    | 1.430(16) | C30-C31  | 1.417(14) |
| C3-O3    | 1.272(11) | C30-C34  | 1.469(14) |
| C3-C2    | 1.451(15) | C31-C32  | 1.388(14) |
| C3-C4    | 1.465(16) | C31-H31  | 0.9500    |
| C4-C8    | 1.378(16) | C32-C33  | 1.371(13) |
| C4-C5    | 1.428(15) | C32-H32  | 0.9500    |
| C5-C6    | 1.399(17) | C33-H33  | 0.9500    |
| C5-H5    | 0.9500    | C34-N35  | 1.246(13) |
| C6-C7    | 1.386(17) | C34-H34  | 0.9500    |
| C6-H6    | 0.9500    | C36-N35  | 1.458(13) |
| C7-C2    | 1.383(15) | C36-C37  | 1.544(14) |
| C7-H7    | 0.9500    | C36-H36A | 0.9900    |
| C8-N9    | 1.335(14) | C36-H36B | 0.9900    |
| C8-H8    | 0.9500    | C37-N38  | 1.492(12) |
| C10-N9   | 1.471(14) | C37-H37A | 0.9900    |
| C10-C11  | 1.486(15) | C37-H37B | 0.9900    |
| C10-H10A | 0.9900    | C39-N38  | 1.424(12) |
| C10-H10B | 0.9900    | C39-C40  | 1.527(14) |
| C11-N12  | 1.483(14) | C39-H39A | 0.9900    |
| C11-H11A | 0.9900    | C39-H39B | 0.9900    |
| C11-H11B | 0.9900    | C40-N41  | 1.468(13) |
| C13-C14  | 1.463(15) | C40-H40A | 0.9900    |
| C13-N12  | 1.492(13) | C40-H40B | 0.9900    |
| C13-H13A | 0.9900    | C42-N41  | 1.347(14) |
| C13-H13B | 0.9900    | C42-C43  | 1.380(15) |
| C14-N15  | 1.487(13) | C42-H42  | 0.9500    |
| C14-H14A | 0.9900    | C43-C48  | 1.403(14) |
| C14-H14B | 0.9900    | C43-C44  | 1.462(13) |
| C16-N15  | 1.243(14) | C44-O10  | 1.291(11) |
| C16-C17  | 1.453(15) | C44-C45  | 1.468(14) |
| C16-H16  | 0.9500    | C45-C46  | 1.354(14) |
| C17-C22  | 1.413(15) | C45-C49  | 1.518(13) |
| C17-C18  | 1.465(13) | C46-C47  | 1.408(15) |
| C18-O4   | 1.269(11) | C46-H46  | 0.9500    |
| C18-C19  | 1.427(14) | C47-C48  | 1.367(15) |
| C19-C20  | 1.451(14) | C47-H47  | 0.9500    |
| C19-C23  | 1.472(13) | C48-H48  | 0.9500    |
| C20-C21  | 1.412(14) | C49-O12  | 1.232(12) |
| C20-H20  | 0.9500    | C49-O11  | 1.296(12) |
| C21-C22  | 1.342(15) | C50-N38  | 1.433(13) |
| C21-H21  | 0.9500    | C50-C51  | 1.519(14) |
| C22-H22  | 0.9500    | C50-H50A | 0.9900    |
| C23-O5   | 1.232(12) | C50-H50B | 0.9900    |
| C23-O6   | 1.246(12) | C51-N52  | 1.502(14) |
| C24-N12  | 1.449(12) | C51-H51A | 0.9900    |
| C24-C25  | 1.522(14) | C51-H51B | 0.9900    |
| C24-H24A | 0.9900    | C54-N53  | 1.496(12) |
| C24-H24B | 0.9900    | C54-C55  | 1.557(13) |
| C25-N26  | 1.428(11) | C54-H54A | 0.9900    |
| C25-H25A | 0.9900    | C54-H54B | 0.9900    |
| C25-H25B | 0.9900    | C55-N56  | 1.447(13) |
| C27-O8   | 1.225(11) | C55-H55A | 0.9900    |
| C27-O7   | 1.273(11) | C55-H55B | 0.9900    |
| C27-C28  | 1.517(12) | C57-N53  | 1.489(12) |
| C28-C33  | 1.384(13) | C57-C58  | 1.535(12) |
| C28-C29  | 1.472(12) | C57-H57A | 0.9900    |

|          |           |               |           |
|----------|-----------|---------------|-----------|
| C57-H57B | 0.9900    | N56-H56B      | 0.9200    |
| C58-N59  | 1.479(12) | N59-Ni1       | 2.128(7)  |
| C58-H58A | 0.9900    | N59-H59A      | 0.9200    |
| C58-H58B | 0.9900    | N59-H59B      | 0.9200    |
| C60-N53  | 1.402(13) | N62-Ni1       | 2.109(7)  |
| C60-C61  | 1.516(14) | N62-H62A      | 0.9200    |
| C60-H60A | 0.9900    | N62-H62B      | 0.9200    |
| C60-H60B | 0.9900    | N63-Ni2       | 2.046(11) |
| C61-N62  | 1.469(12) | N66-Ni2       | 2.091(9)  |
| C61-H61A | 0.9900    | N66-H66A      | 0.9200    |
| C61-H61B | 0.9900    | N66-H66B      | 0.9200    |
| C64-N63  | 1.454(17) | N69-Ni2       | 2.129(10) |
| C64-C65  | 1.55(2)   | N69-H69A      | 0.9200    |
| C64-H64A | 0.9900    | N69-H69B      | 0.9200    |
| C64-H64B | 0.9900    | N72-Ni2       | 2.154(9)  |
| C65-N66  | 1.427(15) | N72-H72A      | 0.9200    |
| C65-H65A | 0.9900    | N72-H72B      | 0.9200    |
| C65-H65B | 0.9900    | O1-Gd1        | 2.345(6)  |
| C67-C68  | 1.45(2)   | O3-Gd1        | 2.393(6)  |
| C67-N63  | 1.55(2)   | O3-H3         | 0.9500    |
| C67-H67A | 0.9900    | O4-Gd1        | 2.369(6)  |
| C67-H67B | 0.9900    | O4-H4         | 0.9500    |
| C68-N69  | 1.416(15) | O5-Gd1        | 2.392(7)  |
| C68-H68A | 0.9900    | O6-Ni2        | 2.143(7)  |
| C68-H68B | 0.9900    | O7-Gd1        | 2.342(6)  |
| C70-N63  | 1.522(15) | O8-Ni1        | 2.138(6)  |
| C70-C71  | 1.526(19) | O9-Gd1        | 2.370(6)  |
| C70-H70A | 0.9900    | O9-H9         | 0.9500    |
| C70-H70B | 0.9900    | O10-Gd1       | 2.389(6)  |
| C71-N72  | 1.472(15) | O10-H10       | 0.9500    |
| C71-H71A | 0.9900    | O11-Gd1       | 2.376(7)  |
| C71-H71B | 0.9900    | N200-O201     | 1.234(8)  |
| N26-Ni1  | 2.050(9)  | N200-O202     | 1.251(8)  |
| N26-H26A | 0.9200    | N200-O200     | 1.257(8)  |
| N26-H26B | 0.9200    | O102-N100     | 1.245(11) |
| N52-Ni2  | 2.056(10) | O101-N100     | 1.241(11) |
| N52-H52A | 0.9200    | N100-O100     | 1.199(11) |
| N52-H52B | 0.9200    | O300-N300     | 1.262(11) |
| N53-Ni1  | 2.104(8)  | N300-O302     | 1.199(11) |
| N56-Ni1  | 2.134(8)  | N300-O301     | 1.254(11) |
| N56-H56A | 0.9200    |               |           |
| O1-C1-O2 | 119.6(11) | N9-C8-H8      | 119.0     |
| O1-C1-C2 | 122.8(10) | C4-C8-H8      | 119.0     |
| O2-C1-C2 | 117.5(10) | N9-C10-C11    | 109.2(10) |
| O3-C3-C2 | 121.9(10) | N9-C10-H10A   | 109.9     |
| O3-C3-C4 | 121.0(10) | C11-C10-H10A  | 109.9     |
| C2-C3-C4 | 117.0(9)  | N9-C10-H10B   | 109.9     |
| C8-C4-C5 | 116.9(11) | C11-C10-H10B  | 109.9     |
| C8-C4-C3 | 121.7(10) | H10A-C10-H10B | 108.3     |
| C5-C4-C3 | 121.3(11) | N12-C11-C10   | 112.8(8)  |
| C6-C5-C4 | 118.7(12) | N12-C11-H11A  | 109.0     |
| C6-C5-H5 | 120.7     | C10-C11-H11A  | 109.0     |
| C4-C5-H5 | 120.7     | N12-C11-H11B  | 109.0     |
| C7-C6-C5 | 119.9(12) | C10-C11-H11B  | 109.0     |
| C7-C6-H6 | 120.1     | H11A-C11-H11B | 107.8     |
| C5-C6-H6 | 120.1     | C14-C13-N12   | 113.3(8)  |
| C2-C7-C6 | 124.6(12) | C14-C13-H13A  | 108.9     |
| C2-C7-H7 | 117.7     | N12-C13-H13A  | 108.9     |
| C6-C7-H7 | 117.7     | C14-C13-H13B  | 108.9     |
| N9-C8-C4 | 121.9(11) | N12-C13-H13B  | 108.9     |

|               |           |               |           |
|---------------|-----------|---------------|-----------|
| H13A-C13-H13B | 107.7     | C32-C33-C28   | 123.9(9)  |
| C13-C14-N15   | 110.1(8)  | C32-C33-H33   | 118.0     |
| C13-C14-H14A  | 109.6     | C28-C33-H33   | 118.0     |
| N15-C14-H14A  | 109.6     | N35-C34-C30   | 126.3(9)  |
| C13-C14-H14B  | 109.6     | N35-C34-H34   | 116.8     |
| N15-C14-H14B  | 109.6     | C30-C34-H34   | 116.8     |
| H14A-C14-H14B | 108.2     | N35-C36-C37   | 110.9(8)  |
| N15-C16-C17   | 127.3(10) | N35-C36-H36A  | 109.5     |
| N15-C16-H16   | 116.4     | C37-C36-H36A  | 109.5     |
| C17-C16-H16   | 116.4     | N35-C36-H36B  | 109.5     |
| C22-C17-C16   | 120.9(9)  | C37-C36-H36B  | 109.5     |
| C22-C17-C18   | 120.2(10) | H36A-C36-H36B | 108.0     |
| C16-C17-C18   | 118.9(10) | N38-C37-C36   | 109.4(7)  |
| O4-C18-C19    | 124.2(8)  | N38-C37-H37A  | 109.8     |
| O4-C18-C17    | 119.3(9)  | C36-C37-H37A  | 109.8     |
| C19-C18-C17   | 116.5(9)  | N38-C37-H37B  | 109.8     |
| C18-C19-C20   | 119.7(9)  | C36-C37-H37B  | 109.8     |
| C18-C19-C23   | 125.0(9)  | H37A-C37-H37B | 108.2     |
| C20-C19-C23   | 115.3(9)  | N38-C39-C40   | 114.8(8)  |
| C21-C20-C19   | 121.3(10) | N38-C39-H39A  | 108.6     |
| C21-C20-H20   | 119.3     | C40-C39-H39A  | 108.6     |
| C19-C20-H20   | 119.3     | N38-C39-H39B  | 108.6     |
| C22-C21-C20   | 118.8(11) | C40-C39-H39B  | 108.6     |
| C22-C21-H21   | 120.6     | H39A-C39-H39B | 107.6     |
| C20-C21-H21   | 120.6     | N41-C40-C39   | 108.6(8)  |
| C21-C22-C17   | 123.4(10) | N41-C40-H40A  | 110.0     |
| C21-C22-H22   | 118.3     | C39-C40-H40A  | 110.0     |
| C17-C22-H22   | 118.3     | N41-C40-H40B  | 110.0     |
| O5-C23-O6     | 122.6(9)  | C39-C40-H40B  | 110.0     |
| O5-C23-C19    | 119.3(9)  | H40A-C40-H40B | 108.4     |
| O6-C23-C19    | 118.1(9)  | N41-C42-C43   | 125.3(10) |
| N12-C24-C25   | 113.3(9)  | N41-C42-H42   | 117.4     |
| N12-C24-H24A  | 108.9     | C43-C42-H42   | 117.4     |
| C25-C24-H24A  | 108.9     | C42-C43-C48   | 121.2(10) |
| N12-C24-H24B  | 108.9     | C42-C43-C44   | 118.9(10) |
| C25-C24-H24B  | 108.9     | C48-C43-C44   | 119.9(9)  |
| H24A-C24-H24B | 107.7     | O10-C44-C43   | 121.6(9)  |
| N26-C25-C24   | 116.9(9)  | O10-C44-C45   | 122.7(9)  |
| N26-C25-H25A  | 108.1     | C43-C44-C45   | 115.8(9)  |
| C24-C25-H25A  | 108.1     | C46-C45-C44   | 119.8(9)  |
| N26-C25-H25B  | 108.1     | C46-C45-C49   | 119.6(10) |
| C24-C25-H25B  | 108.1     | C44-C45-C49   | 120.6(9)  |
| H25A-C25-H25B | 107.3     | C45-C46-C47   | 124.1(11) |
| O8-C27-O7     | 125.1(8)  | C45-C46-H46   | 117.9     |
| O8-C27-C28    | 115.2(8)  | C47-C46-H46   | 117.9     |
| O7-C27-C28    | 119.7(8)  | C48-C47-C46   | 117.6(11) |
| C33-C28-C29   | 117.7(8)  | C48-C47-H47   | 121.2     |
| C33-C28-C27   | 118.4(8)  | C46-C47-H47   | 121.2     |
| C29-C28-C27   | 123.8(8)  | C47-C48-C43   | 122.8(10) |
| O9-C29-C30    | 120.6(8)  | C47-C48-H48   | 118.6     |
| O9-C29-C28    | 122.2(8)  | C43-C48-H48   | 118.6     |
| C30-C29-C28   | 117.1(8)  | O12-C49-O11   | 124.7(9)  |
| C29-C30-C31   | 123.0(9)  | O12-C49-C45   | 116.9(10) |
| C29-C30-C34   | 121.8(9)  | O11-C49-C45   | 118.4(9)  |
| C31-C30-C34   | 115.2(8)  | N38-C50-C51   | 111.9(9)  |
| C32-C31-C30   | 119.1(9)  | N38-C50-H50A  | 109.2     |
| C32-C31-H31   | 120.5     | C51-C50-H50A  | 109.2     |
| C30-C31-H31   | 120.5     | N38-C50-H50B  | 109.2     |
| C33-C32-C31   | 118.8(9)  | C51-C50-H50B  | 109.2     |
| C33-C32-H32   | 120.6     | H50A-C50-H50B | 107.9     |
| C31-C32-H32   | 120.6     | N52-C51-C50   | 113.6(9)  |

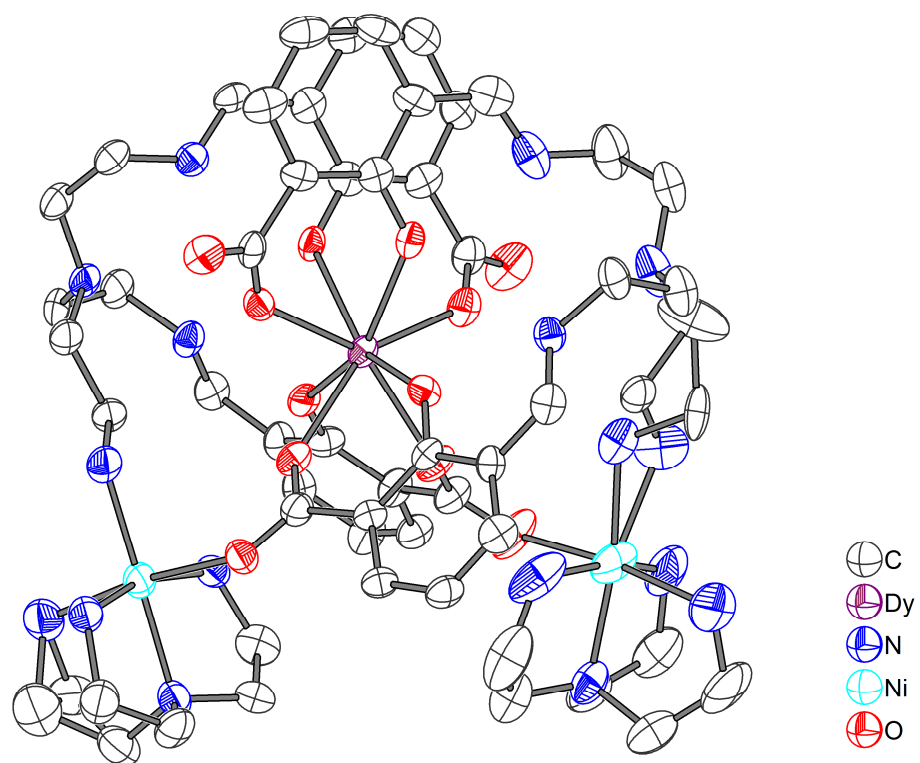
|               |           |               |           |
|---------------|-----------|---------------|-----------|
| N52-C51-H51A  | 108.8     | C67-C68-H68A  | 109.6     |
| C50-C51-H51A  | 108.8     | N69-C68-H68B  | 109.6     |
| N52-C51-H51B  | 108.8     | C67-C68-H68B  | 109.6     |
| C50-C51-H51B  | 108.8     | H68A-C68-H68B | 108.1     |
| H51A-C51-H51B | 107.7     | N63-C70-C71   | 108.1(10) |
| N53-C54-C55   | 110.3(8)  | N63-C70-H70A  | 110.1     |
| N53-C54-H54A  | 109.6     | C71-C70-H70A  | 110.1     |
| C55-C54-H54A  | 109.6     | N63-C70-H70B  | 110.1     |
| N53-C54-H54B  | 109.6     | C71-C70-H70B  | 110.1     |
| C55-C54-H54B  | 109.6     | H70A-C70-H70B | 108.4     |
| H54A-C54-H54B | 108.1     | N72-C71-C70   | 109.7(11) |
| N56-C55-C54   | 109.7(9)  | N72-C71-H71A  | 109.7     |
| N56-C55-H55A  | 109.7     | C70-C71-H71A  | 109.7     |
| C54-C55-H55A  | 109.7     | N72-C71-H71B  | 109.7     |
| N56-C55-H55B  | 109.7     | C70-C71-H71B  | 109.7     |
| C54-C55-H55B  | 109.7     | H71A-C71-H71B | 108.2     |
| H55A-C55-H55B | 108.2     | C7-C2-C1      | 123.1(11) |
| N53-C57-C58   | 112.8(8)  | C7-C2-C3      | 118.4(11) |
| N53-C57-H57A  | 109.0     | C1-C2-C3      | 118.3(9)  |
| C58-C57-H57A  | 109.0     | C8-N9-C10     | 126.6(10) |
| N53-C57-H57B  | 109.0     | C24-N12-C11   | 113.3(9)  |
| C58-C57-H57B  | 109.0     | C24-N12-C13   | 114.3(8)  |
| H57A-C57-H57B | 107.8     | C11-N12-C13   | 110.0(7)  |
| N59-C58-C57   | 112.4(7)  | C16-N15-C14   | 121.6(9)  |
| N59-C58-H58A  | 109.1     | C25-N26-Ni1   | 124.8(7)  |
| C57-C58-H58A  | 109.1     | C25-N26-H26A  | 106.1     |
| N59-C58-H58B  | 109.1     | Ni1-N26-H26A  | 106.1     |
| C57-C58-H58B  | 109.1     | C25-N26-H26B  | 106.1     |
| H58A-C58-H58B | 107.9     | Ni1-N26-H26B  | 106.1     |
| N53-C60-C61   | 110.9(8)  | H26A-N26-H26B | 106.3     |
| N53-C60-H60A  | 109.5     | C34-N35-C36   | 123.3(9)  |
| C61-C60-H60A  | 109.5     | C39-N38-C50   | 113.6(8)  |
| N53-C60-H60B  | 109.5     | C39-N38-C37   | 109.7(8)  |
| C61-C60-H60B  | 109.5     | C50-N38-C37   | 110.3(8)  |
| H60A-C60-H60B | 108.0     | C42-N41-C40   | 125.2(9)  |
| N62-C61-C60   | 108.7(8)  | C51-N52-Ni2   | 115.6(7)  |
| N62-C61-H61A  | 109.9     | C51-N52-H52A  | 108.4     |
| C60-C61-H61A  | 109.9     | Ni2-N52-H52A  | 108.4     |
| N62-C61-H61B  | 109.9     | C51-N52-H52B  | 108.4     |
| C60-C61-H61B  | 109.9     | Ni2-N52-H52B  | 108.4     |
| H61A-C61-H61B | 108.3     | H52A-N52-H52B | 107.4     |
| N63-C64-C65   | 108.8(11) | C60-N53-C57   | 110.9(8)  |
| N63-C64-H64A  | 109.9     | C60-N53-C54   | 113.1(7)  |
| C65-C64-H64A  | 109.9     | C57-N53-C54   | 112.3(8)  |
| N63-C64-H64B  | 109.9     | C60-N53-Ni1   | 107.7(7)  |
| C65-C64-H64B  | 109.9     | C57-N53-Ni1   | 110.0(6)  |
| H64A-C64-H64B | 108.3     | C54-N53-Ni1   | 102.5(6)  |
| N66-C65-C64   | 111.4(12) | C55-N56-Ni1   | 108.6(6)  |
| N66-C65-H65A  | 109.3     | C55-N56-H56A  | 110.0     |
| C64-C65-H65A  | 109.3     | Ni1-N56-H56A  | 110.0     |
| N66-C65-H65B  | 109.3     | C55-N56-H56B  | 110.0     |
| C64-C65-H65B  | 109.3     | Ni1-N56-H56B  | 110.0     |
| H65A-C65-H65B | 108.0     | H56A-N56-H56B | 108.4     |
| C68-C67-N63   | 111.0(12) | C58-N59-Ni1   | 106.7(5)  |
| C68-C67-H67A  | 109.4     | C58-N59-H59A  | 110.4     |
| N63-C67-H67A  | 109.4     | Ni1-N59-H59A  | 110.4     |
| C68-C67-H67B  | 109.4     | C58-N59-H59B  | 110.4     |
| N63-C67-H67B  | 109.4     | Ni1-N59-H59B  | 110.4     |
| H67A-C67-H67B | 108.0     | H59A-N59-H59B | 108.6     |
| N69-C68-C67   | 110.4(10) | C61-N62-Ni1   | 112.0(5)  |
| N69-C68-H68A  | 109.6     | C61-N62-H62A  | 109.2     |

|                |           |                |           |
|----------------|-----------|----------------|-----------|
| Ni1-N62-H62A   | 109.2     | N62-Ni1-N56    | 163.3(3)  |
| C61-N62-H62B   | 109.2     | N59-Ni1-N56    | 92.4(3)   |
| Ni1-N62-H62B   | 109.2     | N26-Ni1-O8     | 91.2(3)   |
| H62A-N62-H62B  | 107.9     | N53-Ni1-O8     | 90.6(3)   |
| C64-N63-C70    | 114.1(11) | N62-Ni1-O8     | 82.0(3)   |
| C64-N63-C67    | 110.2(11) | N59-Ni1-O8     | 173.1(3)  |
| C70-N63-C67    | 109.8(11) | N56-Ni1-O8     | 91.7(3)   |
| C64-N63-Ni2    | 114.3(9)  | N63-Ni2-N52    | 175.4(4)  |
| C70-N63-Ni2    | 104.5(7)  | N63-Ni2-N66    | 81.9(4)   |
| C67-N63-Ni2    | 103.3(9)  | N52-Ni2-N66    | 94.5(4)   |
| C65-N66-Ni2    | 108.6(7)  | N63-Ni2-N69    | 83.6(4)   |
| C65-N66-H66A   | 110.0     | N52-Ni2-N69    | 94.0(4)   |
| Ni2-N66-H66A   | 110.0     | N66-Ni2-N69    | 96.2(4)   |
| C65-N66-H66B   | 110.0     | N63-Ni2-O6     | 93.3(4)   |
| Ni2-N66-H66B   | 110.0     | N52-Ni2-O6     | 90.1(3)   |
| H66A-N66-H66B  | 108.3     | N66-Ni2-O6     | 173.9(4)  |
| C68-N69-Ni2    | 109.1(8)  | N69-Ni2-O6     | 79.5(3)   |
| C68-N69-H69A   | 109.9     | N63-Ni2-N72    | 82.1(4)   |
| Ni2-N69-H69A   | 109.9     | N52-Ni2-N72    | 100.8(4)  |
| C68-N69-H69B   | 109.9     | N66-Ni2-N72    | 91.4(4)   |
| Ni2-N69-H69B   | 109.9     | N69-Ni2-N72    | 162.7(4)  |
| H69A-N69-H69B  | 108.3     | O6-Ni2-N72     | 91.6(3)   |
| C71-N72-Ni2    | 109.8(8)  | O7-Gd1-O1      | 146.5(2)  |
| C71-N72-H72A   | 109.7     | O7-Gd1-O4      | 79.6(2)   |
| Ni2-N72-H72A   | 109.7     | O1-Gd1-O4      | 87.9(2)   |
| C71-N72-H72B   | 109.7     | O7-Gd1-O9      | 71.9(2)   |
| Ni2-N72-H72B   | 109.7     | O1-Gd1-O9      | 102.9(2)  |
| H72A-N72-H72B  | 108.2     | O4-Gd1-O9      | 142.8(2)  |
| C1-O1-Gd1      | 132.8(6)  | O7-Gd1-O11     | 72.7(2)   |
| C3-O3-Gd1      | 130.8(6)  | O1-Gd1-O11     | 140.8(2)  |
| C3-O3-H3       | 114.6     | O4-Gd1-O11     | 104.8(2)  |
| Gd1-O3-H3      | 114.6     | O9-Gd1-O11     | 89.2(2)   |
| C18-O4-Gd1     | 136.5(6)  | O7-Gd1-O10     | 128.4(2)  |
| C18-O4-H4      | 111.8     | O1-Gd1-O10     | 76.9(2)   |
| Gd1-O4-H4      | 111.8     | O4-Gd1-O10     | 145.7(2)  |
| C23-O5-Gd1     | 138.9(7)  | O9-Gd1-O10     | 71.2(2)   |
| C23-O6-Ni2     | 131.1(7)  | O11-Gd1-O10    | 72.0(2)   |
| C27-O7-Gd1     | 138.0(6)  | O7-Gd1-O5      | 73.4(2)   |
| C27-O8-Ni1     | 130.2(6)  | O1-Gd1-O5      | 73.2(2)   |
| C29-O9-Gd1     | 134.0(5)  | O4-Gd1-O5      | 71.1(2)   |
| C29-O9-H9      | 113.0     | O9-Gd1-O5      | 78.1(2)   |
| Gd1-O9-H9      | 113.0     | O11-Gd1-O5     | 146.0(2)  |
| C44-O10-Gd1    | 130.8(5)  | O10-Gd1-O5     | 130.5(2)  |
| C44-O10-H10    | 114.6     | O7-Gd1-O3      | 130.4(2)  |
| Gd1-O10-H10    | 114.6     | O1-Gd1-O3      | 71.7(2)   |
| C49-O11-Gd1    | 137.5(6)  | O4-Gd1-O3      | 70.5(2)   |
| O201-N200-O202 | 116.0(8)  | O9-Gd1-O3      | 146.6(2)  |
| O201-N200-O200 | 122.8(8)  | O11-Gd1-O3     | 77.8(2)   |
| O202-N200-O200 | 121.0(7)  | O10-Gd1-O3     | 75.6(2)   |
| N26-Ni1-N53    | 177.4(3)  | O5-Gd1-O3      | 128.1(3)  |
| N26-Ni1-N62    | 98.3(3)   | O100-N100-O101 | 123.2(16) |
| N53-Ni1-N62    | 80.0(3)   | O100-N100-O102 | 121.2(16) |
| N26-Ni1-N59    | 93.8(3)   | O101-N100-O102 | 115.6(12) |
| N53-Ni1-N59    | 84.2(3)   | O302-N300-O301 | 123.8(14) |
| N62-Ni1-N59    | 92.5(3)   | O302-N300-O300 | 116.7(14) |
| N26-Ni1-N56    | 97.3(3)   | O301-N300-O300 | 116.7(13) |
| N53-Ni1-N56    | 84.6(3)   |                |           |

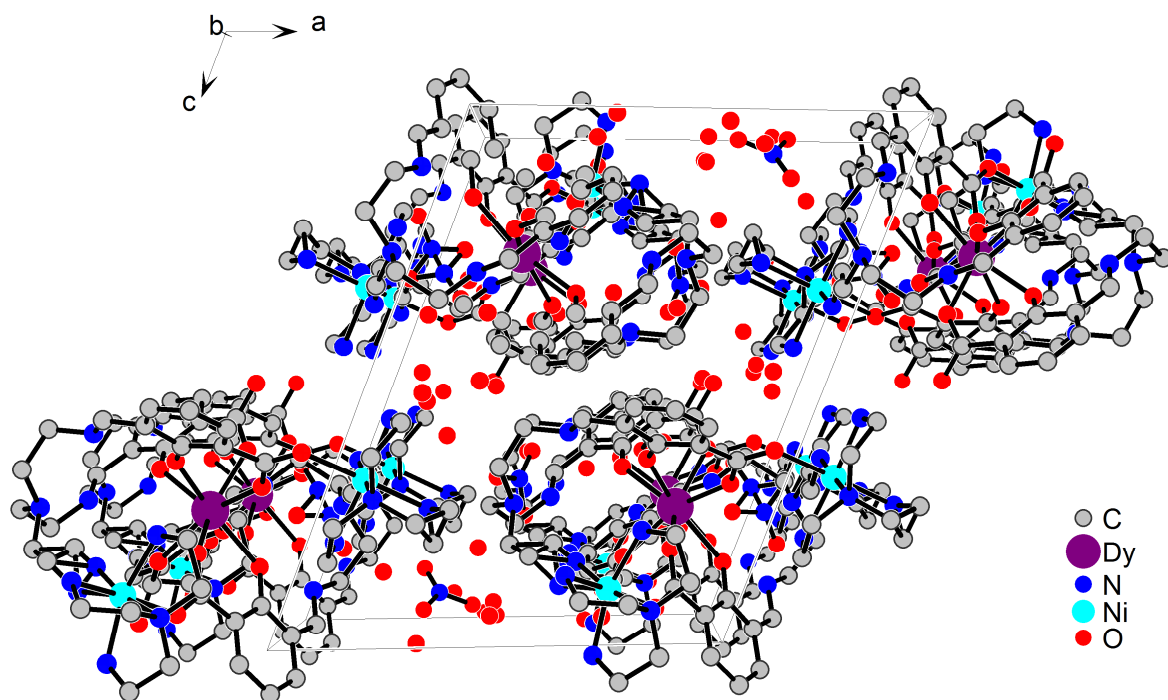
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Symmetry transformations used to generate equivalent atoms:





**Figure S2.** Thermal ellipsoid plot of the molecular structure of the cation of **2**. Thermal ellipsoids are drawn at the 50% probability level. Hydrogen atoms, water molecules, and  $\text{NO}_3^-$  ions are omitted for clarity.



**Figure S3:** Cell Plot of compound **2** omitting the hydrogen atoms. View along the b-axis.

Selected closest intermolecular distances [Å]: Dy-Dy' 9.8271(8), Dy-Ni1' 8.8027(13), Dy-N2' 10.7852(15), Ni1-Ni1' 7.642(2), Ni1-N2' 8.190(2), Ni2-Ni2' 8.873(2).

Table S3. Crystal data and structure refinement for **2**.

|                                    |   |
|------------------------------------|---|
| Crystal data                       |   |
| Identification code                | 1045a   |
| Habitus, colour                    | plate, colourless   |
| Crystal size                       | 0.20 x 0.12 x 0.06 mm <sup>3</sup>  |
| Crystal system                     | Triclinic   |
| Space group                        | P -1  |
| Unit cell dimensions               | Z = 2   |
|                                    | a = 14.8795(9) Å  |
|                                    | α = 102.704(4)°   |
|                                    | b = 16.0919(8) Å  |
|                                    | β = 106.785(5)°   |
|                                    | c = 18.6121(10) Å   |
|                                    | γ = 103.584(4)°   |
| Volume                             | 3944.0(4) Å <sup>3</sup>  |
| Cell determination                 | 16527 peaks with Theta 4.6 to 26.8°.  |
| Empirical formula                  | C <sub>56</sub> H <sub>14</sub> Dy N <sub>19</sub> Ni <sub>2</sub> O <sub>36</sub>                                      |
| Formula weight                     | 1909.58   |
| Density (calculated)               | 1.608 Mg/m <sup>3</sup>   |
| Absorption coefficient             | 1.511 mm <sup>-1</sup>  |
| F(000)                             | 1986  |
| Data collection:                   |   |
| Diffractometer type                | STOE IPDS 2T  |
| Wavelength                         | 0.71073 Å   |
| Temperature                        | 100(2) K  |
| Theta range for data collection    | 4.68 to 25.00°.   |
| Index ranges                       | -17<=h<=17, -19<=k<=19, -22<=l<=21  |
| Data collection software           | STOE X-AREA (STOE & Cie, 2006)  |
| Cell refinement software           | STOE X-AREA   |
| Data reduction software            | STOE X-AREA   |
| Solution and refinement:           |   |
| Reflections collected              | 25555   |
| Independent reflections            | 13546 [R(int) = 0.0858]   |
| Completeness to theta = 25.00°     | 97.6 %  |
| Observed reflections               | 7334[I>2sigma(I)]   |
| Reflections used for refinement    | 13546   |
| Absorption correction              | Integration   |
| Max. and min. transmission         | 0.9320 and 0.7325   |
| Largest diff. peak and hole        | 0.947 and -0.680 e.Å <sup>-3</sup>  |
| Solution                           | Charge flipping   |
| Refinement                         | Full-matrix least-squares on F <sup>2</sup>   |
| Treatment of hydrogen atoms        | Caqlculated positions, riding model   |
| Programs used                      | Superflip (Palatinus & Chapuis, 2007)<br>SHELXL-97 (Sheldrick, 2008)<br>DIAMOND (Crystal Impact)<br>STOE IPDS2 software |
| Data / restraints / parameters     | 13546 / 264 / 1093  |
| Goodness-of-fit on F <sup>2</sup>  | 0.855   |
| R index (all data)                 | wR2 = 0.1218  |
| R index conventional [I>2sigma(I)] | R1 = 0.0594   |

Table S4. Bond lengths [Å] and angles [°] for **2**

|          |           |           |           |
|----------|-----------|-----------|-----------|
| C1-O2    | 1.265(10) | C28-C29   | 1.420(11) |
| C1-O1    | 1.281(9)  | C29-O9    | 1.279(9)  |
| C1-C2    | 1.489(12) | C29-C30   | 1.430(10) |
| C2-C7    | 1.387(12) | C30-C31   | 1.412(11) |
| C2-C3    | 1.425(12) | C30-C34   | 1.425(12) |
| C3-O3    | 1.308(9)  | C31-C32   | 1.341(12) |
| C3-C4    | 1.413(11) | C31-H31   | 0.9500    |
| C4-C8    | 1.412(11) | C32-C33   | 1.399(11) |
| C4-C5    | 1.421(12) | C32-H32   | 0.9500    |
| C5-C6    | 1.351(12) | C33-H33   | 0.9500    |
| C5-H5    | 0.9500    | C34-N35   | 1.321(10) |
| C6-C7    | 1.396(12) | C34-H34   | 0.9500    |
| C6-H6    | 0.9500    | C36-N35   | 1.473(10) |
| C7-H7    | 0.9500    | C36-C37   | 1.508(12) |
| C8-N9    | 1.301(10) | C36-H36A  | 0.9900    |
| C8-H8    | 0.9500    | C36-H36B  | 0.9900    |
| C10-N9   | 1.452(11) | C37-N38   | 1.480(10) |
| C10-C11  | 1.531(12) | C37-H37A  | 0.9900    |
| C10-H10A | 0.9900    | C37-H37B  | 0.9900    |
| C10-H10B | 0.9900    | C39-N38   | 1.476(12) |
| C11-N12  | 1.453(10) | C39-C40   | 1.498(12) |
| C11-H11A | 0.9900    | C39-H39A  | 0.9900    |
| C11-H11B | 0.9900    | C39-H39B  | 0.9900    |
| C13-N12  | 1.467(10) | C40-N41   | 1.457(12) |
| C13-C14  | 1.498(12) | C40-H40A  | 0.9900    |
| C13-H13A | 0.9900    | C40-H40B  | 0.9900    |
| C13-H13B | 0.9900    | C42-N41   | 1.295(11) |
| C14-N15  | 1.494(10) | C42-C43   | 1.413(13) |
| C14-H14A | 0.9900    | C42-H42   | 0.9500    |
| C14-H14B | 0.9900    | C43-C48   | 1.401(12) |
| C16-N15  | 1.279(10) | C43-C44   | 1.427(12) |
| C16-C17  | 1.425(11) | C44-O10   | 1.303(9)  |
| C16-H16  | 0.9500    | C44-C45   | 1.417(11) |
| C17-C22  | 1.403(11) | C45-C46   | 1.391(11) |
| C17-C18  | 1.433(10) | C45-C49   | 1.492(12) |
| C18-O4   | 1.294(9)  | C46-C47   | 1.400(13) |
| C18-C19  | 1.407(11) | C46-H46   | 0.9500    |
| C19-C20  | 1.390(11) | C47-C48   | 1.343(13) |
| C19-C23  | 1.515(10) | C47-H47   | 0.9500    |
| C20-C21  | 1.407(10) | C48-H48   | 0.9500    |
| C20-H20  | 0.9500    | C49-O11   | 1.265(9)  |
| C21-C22  | 1.375(11) | C49-O12   | 1.268(9)  |
| C21-H21  | 0.9500    | C50-N38   | 1.445(14) |
| C22-H22  | 0.9500    | C50-C51A  | 1.60(2)   |
| C23-O6   | 1.249(9)  | C50-C51   | 1.600(19) |
| C23-O5   | 1.277(9)  | C50-H50A  | 0.9900    |
| C24-N12  | 1.495(10) | C50-H50B  | 0.9900    |
| C24-C25  | 1.521(11) | C50-H50C  | 0.9900    |
| C24-H24A | 0.9900    | C50-H50D  | 0.9900    |
| C24-H24B | 0.9900    | C51-N52   | 1.474(17) |
| C25-N26  | 1.482(10) | C51-H51A  | 0.9900    |
| C25-H25A | 0.9900    | C51-H51B  | 0.9900    |
| C25-H25B | 0.9900    | C51A-N52A | 1.457(18) |
| C27-O8   | 1.255(10) | C51A-H51C | 0.9900    |
| C27-O7   | 1.273(10) | C51A-H51D | 0.9900    |
| C27-C28  | 1.508(11) | C54-N53   | 1.477(11) |
| C28-C33  | 1.393(11) | C54-C55   | 1.525(12) |

|           |           |           |           |
|-----------|-----------|-----------|-----------|
| C54-H54A  | 0.9900    | Dy1-O3    | 2.365(5)  |
| C54-H54B  | 0.9900    | Dy1-O11   | 2.368(5)  |
| C55-N56   | 1.473(11) | N26-Ni1   | 2.052(7)  |
| C55-H55A  | 0.9900    | N26-H26A  | 0.9200    |
| C55-H55B  | 0.9900    | N26-H26B  | 0.9200    |
| C57-N53   | 1.493(11) | N52-Ni2   | 2.112(16) |
| C57-C58   | 1.511(13) | N52-H52A  | 0.9200    |
| C57-H57A  | 0.9900    | N52-H52B  | 0.9200    |
| C57-H57B  | 0.9900    | N52A-Ni2  | 2.124(15) |
| C58-N59   | 1.505(12) | N52A-H52C | 0.9200    |
| C58-H58A  | 0.9900    | N52A-H52D | 0.9200    |
| C58-H58B  | 0.9900    | N53-Ni1   | 2.100(7)  |
| C60-N53   | 1.484(11) | N56-Ni1   | 2.105(7)  |
| C60-C61   | 1.51(2)   | N56-H56A  | 0.9200    |
| C60-C61A  | 1.518(16) | N56-H56B  | 0.9200    |
| C60-H60A  | 0.9900    | N59-Ni1   | 2.123(7)  |
| C60-H60B  | 0.9900    | N59-H59A  | 0.9200    |
| C60-H60C  | 0.9900    | N59-H59B  | 0.9200    |
| C60-H60D  | 0.9900    | N62-Ni1   | 2.111(7)  |
| C61-N62   | 1.54(2)   | N62-H62A  | 0.9200    |
| C61-H61A  | 0.9900    | N62-H62B  | 0.9200    |
| C61-H61B  | 0.9900    | N62-H62C  | 0.9200    |
| C61A-N62  | 1.541(16) | N62-H62D  | 0.9200    |
| C61A-H61C | 0.9900    | N63-Ni2   | 2.098(8)  |
| C61A-H61D | 0.9900    | N66-Ni2   | 2.084(9)  |
| C64-C65   | 1.478(15) | N66-H66A  | 0.9200    |
| C64-N63   | 1.485(11) | N66-H66B  | 0.9200    |
| C64-H64A  | 0.9900    | N69-Ni2   | 2.104(7)  |
| C64-H64B  | 0.9900    | N69-H69A  | 0.9200    |
| C65-N66   | 1.506(14) | N69-H69B  | 0.9200    |
| C65-H65A  | 0.9900    | N72-Ni2   | 2.093(10) |
| C65-H65B  | 0.9900    | N72-H72A  | 0.9200    |
| C67-C68   | 1.484(17) | N72-H72B  | 0.9200    |
| C67-N63   | 1.501(11) | Ni1-O8    | 2.160(5)  |
| C67-H67A  | 0.9900    | Ni2-O6    | 2.095(5)  |
| C67-H67B  | 0.9900    | O3-H3     | 0.8507    |
| C68-N69   | 1.458(14) | O4-H4     | 0.8501    |
| C68-H68A  | 0.9900    | O9-H9     | 0.8504    |
| C68-H68B  | 0.9900    | O10-H10   | 0.8501    |
| C70-N63   | 1.477(12) | N100-O101 | 1.184(12) |
| C70-C71   | 1.522(14) | N100-O102 | 1.260(13) |
| C70-H70A  | 0.9900    | N100-O103 | 1.278(13) |
| C70-H70B  | 0.9900    | N200-O203 | 1.231(13) |
| C71-N72   | 1.448(16) | N200-O202 | 1.264(12) |
| C71-H71A  | 0.9900    | N200-O201 | 1.339(13) |
| C71-H71B  | 0.9900    | N300-O302 | 1.158(12) |
| Dy1-O4    | 2.311(5)  | N300-O300 | 1.175(13) |
| Dy1-O7    | 2.316(6)  | N300-O301 | 1.194(12) |
| Dy1-O1    | 2.321(5)  | N30A-O301 | 1.123(13) |
| Dy1-O9    | 2.332(5)  | N30A-O30A | 1.170(14) |
| Dy1-O10   | 2.357(5)  | N30A-O302 | 1.219(13) |
| Dy1-O5    | 2.362(5)  |           |           |
| O2-C1-O1  | 122.6(8)  | C4-C3-C2  | 118.6(8)  |
| O2-C1-C2  | 118.3(7)  | C8-C4-C3  | 122.7(8)  |
| O1-C1-C2  | 119.1(7)  | C8-C4-C5  | 118.4(8)  |
| C7-C2-C3  | 119.1(8)  | C3-C4-C5  | 118.8(8)  |
| C7-C2-C1  | 119.6(8)  | C6-C5-C4  | 122.5(8)  |
| C3-C2-C1  | 121.2(8)  | C6-C5-H5  | 118.8     |
| O3-C3-C4  | 120.1(8)  | C4-C5-H5  | 118.8     |
| O3-C3-C2  | 121.2(8)  | C5-C6-C7  | 118.3(9)  |

|               |          |               |           |
|---------------|----------|---------------|-----------|
| C5-C6-H6      | 120.8    | H24A-C24-H24B | 107.9     |
| C7-C6-H6      | 120.8    | N26-C25-C24   | 111.8(7)  |
| C2-C7-C6      | 122.4(9) | N26-C25-H25A  | 109.3     |
| C2-C7-H7      | 118.8    | C24-C25-H25A  | 109.3     |
| C6-C7-H7      | 118.8    | N26-C25-H25B  | 109.3     |
| N9-C8-C4      | 121.2(8) | C24-C25-H25B  | 109.3     |
| N9-C8-H8      | 119.4    | H25A-C25-H25B | 107.9     |
| C4-C8-H8      | 119.4    | O8-C27-O7     | 123.4(7)  |
| N9-C10-C11    | 108.9(7) | O8-C27-C28    | 117.1(7)  |
| N9-C10-H10A   | 109.9    | O7-C27-C28    | 119.5(8)  |
| C11-C10-H10A  | 109.9    | C33-C28-C29   | 119.6(7)  |
| N9-C10-H10B   | 109.9    | C33-C28-C27   | 118.0(7)  |
| C11-C10-H10B  | 109.9    | C29-C28-C27   | 122.4(7)  |
| H10A-C10-H10B | 108.3    | O9-C29-C28    | 124.1(7)  |
| N12-C11-C10   | 112.2(6) | O9-C29-C30    | 119.1(7)  |
| N12-C11-H11A  | 109.2    | C28-C29-C30   | 116.8(7)  |
| C10-C11-H11A  | 109.2    | C31-C30-C34   | 117.4(7)  |
| N12-C11-H11B  | 109.2    | C31-C30-C29   | 121.3(7)  |
| C10-C11-H11B  | 109.2    | C34-C30-C29   | 121.3(7)  |
| H11A-C11-H11B | 107.9    | C32-C31-C30   | 120.4(7)  |
| N12-C13-C14   | 110.3(7) | C32-C31-H31   | 119.8     |
| N12-C13-H13A  | 109.6    | C30-C31-H31   | 119.8     |
| C14-C13-H13A  | 109.6    | C31-C32-C33   | 120.0(8)  |
| N12-C13-H13B  | 109.6    | C31-C32-H32   | 120.0     |
| C14-C13-H13B  | 109.6    | C33-C32-H32   | 120.0     |
| H13A-C13-H13B | 108.1    | C28-C33-C32   | 121.9(8)  |
| N15-C14-C13   | 109.7(7) | C28-C33-H33   | 119.1     |
| N15-C14-H14A  | 109.7    | C32-C33-H33   | 119.1     |
| C13-C14-H14A  | 109.7    | N35-C34-C30   | 125.5(7)  |
| N15-C14-H14B  | 109.7    | N35-C34-H34   | 117.2     |
| C13-C14-H14B  | 109.7    | C30-C34-H34   | 117.2     |
| H14A-C14-H14B | 108.2    | N35-C36-C37   | 110.7(7)  |
| N15-C16-C17   | 126.2(7) | N35-C36-H36A  | 109.5     |
| N15-C16-H16   | 116.9    | C37-C36-H36A  | 109.5     |
| C17-C16-H16   | 116.9    | N35-C36-H36B  | 109.5     |
| C22-C17-C16   | 117.0(7) | C37-C36-H36B  | 109.5     |
| C22-C17-C18   | 121.1(7) | H36A-C36-H36B | 108.1     |
| C16-C17-C18   | 121.9(7) | N38-C37-C36   | 111.1(7)  |
| O4-C18-C19    | 123.9(7) | N38-C37-H37A  | 109.4     |
| O4-C18-C17    | 119.4(7) | C36-C37-H37A  | 109.4     |
| C19-C18-C17   | 116.7(7) | N38-C37-H37B  | 109.4     |
| C20-C19-C18   | 120.6(7) | C36-C37-H37B  | 109.4     |
| C20-C19-C23   | 115.1(7) | H37A-C37-H37B | 108.0     |
| C18-C19-C23   | 124.3(7) | N38-C39-C40   | 112.8(8)  |
| C19-C20-C21   | 122.4(7) | N38-C39-H39A  | 109.0     |
| C19-C20-H20   | 118.8    | C40-C39-H39A  | 109.0     |
| C21-C20-H20   | 118.8    | N38-C39-H39B  | 109.0     |
| C22-C21-C20   | 117.7(7) | C40-C39-H39B  | 109.0     |
| C22-C21-H21   | 121.2    | H39A-C39-H39B | 107.8     |
| C20-C21-H21   | 121.2    | N41-C40-C39   | 110.4(8)  |
| C21-C22-C17   | 121.4(7) | N41-C40-H40A  | 109.6     |
| C21-C22-H22   | 119.3    | C39-C40-H40A  | 109.6     |
| C17-C22-H22   | 119.3    | N41-C40-H40B  | 109.6     |
| O6-C23-O5     | 123.9(7) | C39-C40-H40B  | 109.6     |
| O6-C23-C19    | 116.5(7) | H40A-C40-H40B | 108.1     |
| O5-C23-C19    | 119.6(7) | N41-C42-C43   | 125.3(10) |
| N12-C24-C25   | 112.1(7) | N41-C42-H42   | 117.3     |
| N12-C24-H24A  | 109.2    | C43-C42-H42   | 117.3     |
| C25-C24-H24A  | 109.2    | C48-C43-C42   | 119.2(9)  |
| N12-C24-H24B  | 109.2    | C48-C43-C44   | 120.9(9)  |
| C25-C24-H24B  | 109.2    | C42-C43-C44   | 119.9(9)  |

|                |           |                |           |
|----------------|-----------|----------------|-----------|
| O10-C44-C45    | 120.6(8)  | C54-C55-H55B   | 109.3     |
| O10-C44-C43    | 121.7(8)  | H55A-C55-H55B  | 108.0     |
| C45-C44-C43    | 117.6(8)  | N53-C57-C58    | 109.5(7)  |
| C46-C45-C44    | 119.3(8)  | N53-C57-H57A   | 109.8     |
| C46-C45-C49    | 118.2(8)  | C58-C57-H57A   | 109.8     |
| C44-C45-C49    | 122.5(7)  | N53-C57-H57B   | 109.8     |
| C45-C46-C47    | 121.5(9)  | C58-C57-H57B   | 109.8     |
| C45-C46-H46    | 119.3     | H57A-C57-H57B  | 108.2     |
| C47-C46-H46    | 119.3     | N59-C58-C57    | 111.1(8)  |
| C48-C47-C46    | 120.3(9)  | N59-C58-H58A   | 109.4     |
| C48-C47-H47    | 119.9     | C57-C58-H58A   | 109.4     |
| C46-C47-H47    | 119.9     | N59-C58-H58B   | 109.4     |
| C47-C48-C43    | 120.5(9)  | C57-C58-H58B   | 109.4     |
| C47-C48-H48    | 119.8     | H58A-C58-H58B  | 108.0     |
| C43-C48-H48    | 119.8     | N53-C60-C61    | 112.4(15) |
| O11-C49-O12    | 122.1(8)  | N53-C60-C61A   | 113.3(9)  |
| O11-C49-C45    | 120.0(7)  | C61-C60-C61A   | 44.3(13)  |
| O12-C49-C45    | 117.9(8)  | N53-C60-H60A   | 109.1     |
| N38-C50-C51A   | 98.3(11)  | C61-C60-H60A   | 109.1     |
| N38-C50-C51    | 125.2(10) | C61A-C60-H60A  | 136.4     |
| C51A-C50-C51   | 38.3(9)   | N53-C60-H60B   | 109.1     |
| N38-C50-H50A   | 106.0     | C61-C60-H60B   | 109.1     |
| C51A-C50-H50A  | 144.2     | C61A-C60-H60B  | 67.3      |
| C51-C50-H50A   | 106.0     | H60A-C60-H60B  | 107.9     |
| N38-C50-H50B   | 106.0     | N53-C60-H60C   | 108.9     |
| C51A-C50-H50B  | 91.4      | C61-C60-H60C   | 67.7      |
| C51-C50-H50B   | 106.0     | C61A-C60-H60C  | 108.9     |
| H50A-C50-H50B  | 106.3     | H60A-C60-H60C  | 45.4      |
| N38-C50-H50C   | 112.1     | H60B-C60-H60C  | 139.5     |
| C51A-C50-H50C  | 112.1     | N53-C60-H60D   | 108.9     |
| C51-C50-H50C   | 76.1      | C61-C60-H60D   | 137.5     |
| H50A-C50-H50C  | 33.9      | C61A-C60-H60D  | 108.9     |
| H50B-C50-H50C  | 130.7     | H60A-C60-H60D  | 64.8      |
| N38-C50-H50D   | 112.1     | H60B-C60-H60D  | 45.7      |
| C51A-C50-H50D  | 112.1     | H60C-C60-H60D  | 107.7     |
| C51-C50-H50D   | 115.1     | C60-C61-N62    | 109.6(14) |
| H50A-C50-H50D  | 82.7      | C60-C61-H61A   | 109.7     |
| H50B-C50-H50D  | 23.6      | N62-C61-H61A   | 109.7     |
| H50C-C50-H50D  | 109.7     | C60-C61-H61B   | 109.7     |
| N52-C51-C50    | 107.6(12) | N62-C61-H61B   | 109.7     |
| N52-C51-H51A   | 110.2     | H61A-C61-H61B  | 108.2     |
| C50-C51-H51A   | 110.2     | C60-C61A-N62   | 109.1(10) |
| N52-C51-H51B   | 110.2     | C60-C61A-H61C  | 109.9     |
| C50-C51-H51B   | 110.2     | N62-C61A-H61C  | 109.9     |
| H51A-C51-H51B  | 108.5     | C60-C61A-H61D  | 109.9     |
| N52A-C51A-C50  | 107.8(12) | N62-C61A-H61D  | 109.9     |
| N52A-C51A-H51C | 110.1     | H61C-C61A-H61D | 108.3     |
| C50-C51A-H51C  | 110.1     | C65-C64-N63    | 112.5(9)  |
| N52A-C51A-H51D | 110.1     | C65-C64-H64A   | 109.1     |
| C50-C51A-H51D  | 110.1     | N63-C64-H64A   | 109.1     |
| H51C-C51A-H51D | 108.5     | C65-C64-H64B   | 109.1     |
| N53-C54-C55    | 110.7(7)  | N63-C64-H64B   | 109.1     |
| N53-C54-H54A   | 109.5     | H64A-C64-H64B  | 107.8     |
| C55-C54-H54A   | 109.5     | C64-C65-N66    | 108.9(9)  |
| N53-C54-H54B   | 109.5     | C64-C65-H65A   | 109.9     |
| C55-C54-H54B   | 109.5     | N66-C65-H65A   | 109.9     |
| H54A-C54-H54B  | 108.1     | C64-C65-H65B   | 109.9     |
| N56-C55-C54    | 111.4(7)  | N66-C65-H65B   | 109.9     |
| N56-C55-H55A   | 109.3     | H65A-C65-H65B  | 108.3     |
| C54-C55-H55A   | 109.3     | C68-C67-N63    | 112.8(9)  |
| N56-C55-H55B   | 109.3     | C68-C67-H67A   | 109.0     |

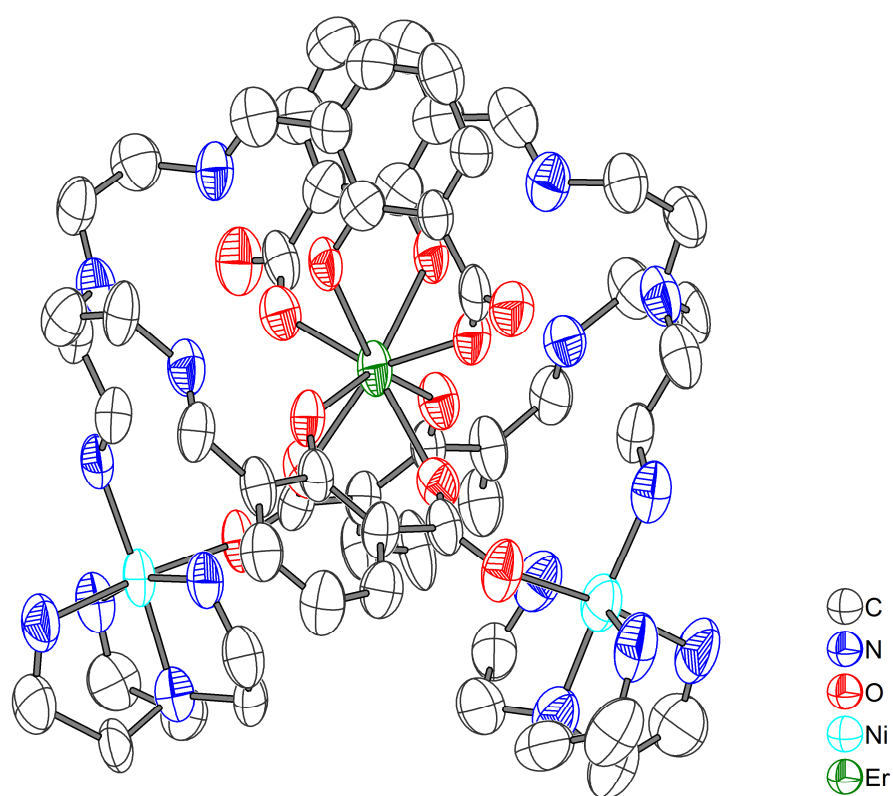
|               |            |                |           |
|---------------|------------|----------------|-----------|
| N63-C67-H67A  | 109.0      | C34-N35-C36    | 121.2(7)  |
| C68-C67-H67B  | 109.0      | C50-N38-C39    | 108.7(7)  |
| N63-C67-H67B  | 109.0      | C50-N38-C37    | 112.6(8)  |
| H67A-C67-H67B | 107.8      | C39-N38-C37    | 110.3(7)  |
| N69-C68-C67   | 113.4(10)  | C42-N41-C40    | 126.1(9)  |
| N69-C68-H68A  | 108.9      | C51-N52-Ni2    | 115.1(11) |
| C67-C68-H68A  | 108.9      | C51-N52-H52A   | 108.5     |
| N69-C68-H68B  | 108.9      | Ni2-N52-H52A   | 108.5     |
| C67-C68-H68B  | 108.9      | C51-N52-H52B   | 108.5     |
| H68A-C68-H68B | 107.7      | Ni2-N52-H52B   | 108.5     |
| N63-C70-C71   | 108.5(9)   | H52A-N52-H52B  | 107.5     |
| N63-C70-H70A  | 110.0      | C51A-N52A-Ni2  | 115.5(14) |
| C71-C70-H70A  | 110.0      | C51A-N52A-H52C | 108.4     |
| N63-C70-H70B  | 110.0      | Ni2-N52A-H52C  | 108.4     |
| C71-C70-H70B  | 110.0      | C51A-N52A-H52D | 108.4     |
| H70A-C70-H70B | 108.4      | Ni2-N52A-H52D  | 108.4     |
| N72-C71-C70   | 113.4(10)  | H52C-N52A-H52D | 107.5     |
| N72-C71-H71A  | 108.9      | C54-N53-C60    | 112.7(7)  |
| C70-C71-H71A  | 108.9      | C54-N53-C57    | 111.5(6)  |
| N72-C71-H71B  | 108.9      | C60-N53-C57    | 111.0(7)  |
| C70-C71-H71B  | 108.9      | C54-N53-Ni1    | 104.5(5)  |
| H71A-C71-H71B | 107.7      | C60-N53-Ni1    | 110.4(5)  |
| O4-Dy1-O7     | 78.31(18)  | C57-N53-Ni1    | 106.3(5)  |
| O4-Dy1-O1     | 86.67(18)  | C55-N56-Ni1    | 110.4(5)  |
| O7-Dy1-O1     | 147.00(19) | C55-N56-H56A   | 109.6     |
| O4-Dy1-O9     | 142.40(18) | Ni1-N56-H56A   | 109.6     |
| O7-Dy1-O9     | 72.28(17)  | C55-N56-H56B   | 109.6     |
| O1-Dy1-O9     | 105.63(17) | Ni1-N56-H56B   | 109.6     |
| O4-Dy1-O10    | 147.43(18) | H56A-N56-H56B  | 108.1     |
| O7-Dy1-O10    | 128.49(17) | C58-N59-Ni1    | 108.3(5)  |
| O1-Dy1-O10    | 77.59(19)  | C58-N59-H59A   | 110.0     |
| O9-Dy1-O10    | 69.98(17)  | Ni1-N59-H59A   | 110.0     |
| O4-Dy1-O5     | 72.51(17)  | C58-N59-H59B   | 110.0     |
| O7-Dy1-O5     | 74.55(19)  | Ni1-N59-H59B   | 110.0     |
| O1-Dy1-O5     | 72.9(2)    | H59A-N59-H59B  | 108.4     |
| O9-Dy1-O5     | 77.39(17)  | C61-N62-C61A   | 43.5(12)  |
| O10-Dy1-O5    | 127.47(18) | C61-N62-Ni1    | 106.6(12) |
| O4-Dy1-O3     | 71.77(18)  | C61A-N62-Ni1   | 106.4(7)  |
| O7-Dy1-O3     | 127.28(17) | C61-N62-H62A   | 110.4     |
| O1-Dy1-O3     | 73.35(18)  | C61A-N62-H62A  | 140.5     |
| O9-Dy1-O3     | 145.61(18) | Ni1-N62-H62A   | 110.4     |
| O10-Dy1-O3    | 76.43(17)  | C61-N62-H62B   | 110.4     |
| O5-Dy1-O3     | 131.55(17) | C61A-N62-H62B  | 70.2      |
| O4-Dy1-O11    | 106.08(18) | Ni1-N62-H62B   | 110.4     |
| O7-Dy1-O11    | 71.65(18)  | H62A-N62-H62B  | 108.6     |
| O1-Dy1-O11    | 141.25(19) | C61-N62-H62C   | 70.1      |
| O9-Dy1-O11    | 86.52(17)  | C61A-N62-H62C  | 110.5     |
| O10-Dy1-O11   | 72.31(18)  | Ni1-N62-H62C   | 110.5     |
| O5-Dy1-O11    | 145.62(19) | H62A-N62-H62C  | 42.5      |
| O3-Dy1-O11    | 76.28(17)  | H62B-N62-H62C  | 136.8     |
| C8-N9-C10     | 124.9(7)   | C61-N62-H62D   | 140.3     |
| C11-N12-C13   | 111.5(6)   | C61A-N62-H62D  | 110.5     |
| C11-N12-C24   | 111.5(7)   | Ni1-N62-H62D   | 110.5     |
| C13-N12-C24   | 113.3(7)   | H62A-N62-H62D  | 69.3      |
| C16-N15-C14   | 123.6(7)   | H62B-N62-H62D  | 42.4      |
| C25-N26-Ni1   | 118.3(5)   | H62C-N62-H62D  | 108.6     |
| C25-N26-H26A  | 107.7      | C70-N63-C64    | 112.5(7)  |
| Ni1-N26-H26A  | 107.7      | C70-N63-C67    | 111.7(8)  |
| C25-N26-H26B  | 107.7      | C64-N63-C67    | 111.6(7)  |
| Ni1-N26-H26B  | 107.7      | C70-N63-Ni2    | 105.9(6)  |
| H26A-N26-H26B | 107.1      | C64-N63-Ni2    | 104.9(6)  |



|               |          |                |           |
|---------------|----------|----------------|-----------|
| C67-N63-Ni2   | 109.7(6) | N66-Ni2-N52    | 104.6(7)  |
| C65-N66-Ni2   | 111.1(7) | N72-Ni2-N52    | 88.0(7)   |
| C65-N66-H66A  | 109.4    | O6-Ni2-N52     | 86.2(4)   |
| Ni2-N66-H66A  | 109.4    | N63-Ni2-N52    | 170.2(6)  |
| C65-N66-H66B  | 109.4    | N69-Ni2-N52    | 101.9(5)  |
| Ni2-N66-H66B  | 109.4    | N66-Ni2-N52A   | 81.8(7)   |
| H66A-N66-H66B | 108.0    | N72-Ni2-N52A   | 112.3(7)  |
| C68-N69-Ni2   | 107.9(6) | O6-Ni2-N52A    | 93.8(6)   |
| C68-N69-H69A  | 110.1    | N63-Ni2-N52A   | 164.1(7)  |
| Ni2-N69-H69A  | 110.1    | N69-Ni2-N52A   | 93.3(7)   |
| C68-N69-H69B  | 110.1    | N52-Ni2-N52A   | 25.2(6)   |
| Ni2-N69-H69B  | 110.1    | C1-O1-Dy1      | 134.3(5)  |
| H69A-N69-H69B | 108.4    | C3-O3-Dy1      | 129.2(4)  |
| C71-N72-Ni2   | 109.0(6) | C3-O3-H3       | 108.8     |
| C71-N72-H72A  | 109.9    | Dy1-O3-H3      | 112.1     |
| Ni2-N72-H72A  | 109.9    | C18-O4-Dy1     | 136.2(5)  |
| C71-N72-H72B  | 109.9    | C18-O4-H4      | 109.1     |
| Ni2-N72-H72B  | 109.9    | Dy1-O4-H4      | 112.6     |
| H72A-N72-H72B | 108.3    | C23-O5-Dy1     | 135.0(5)  |
| N26-Ni1-N53   | 178.7(3) | C23-O6-Ni2     | 134.3(5)  |
| N26-Ni1-N56   | 98.6(3)  | C27-O7-Dy1     | 140.7(5)  |
| N53-Ni1-N56   | 82.7(3)  | C27-O8-Ni1     | 126.7(5)  |
| N26-Ni1-N62   | 96.4(3)  | C29-O9-Dy1     | 136.2(5)  |
| N53-Ni1-N62   | 83.8(3)  | C29-O9-H9      | 109.1     |
| N56-Ni1-N62   | 91.6(3)  | Dy1-O9-H9      | 114.7     |
| N26-Ni1-N59   | 95.1(3)  | C44-O10-Dy1    | 131.0(5)  |
| N53-Ni1-N59   | 83.6(3)  | C44-O10-H10    | 109.5     |
| N56-Ni1-N59   | 164.7(3) | Dy1-O10-H10    | 107.8     |
| N62-Ni1-N59   | 93.8(3)  | C49-O11-Dy1    | 135.1(5)  |
| N26-Ni1-O8    | 91.3(2)  | O101-N100-O102 | 124.8(13) |
| N53-Ni1-O8    | 88.4(2)  | O101-N100-O103 | 118.3(12) |
| N56-Ni1-O8    | 88.4(2)  | O102-N100-O103 | 116.7(12) |
| N62-Ni1-O8    | 172.2(3) | O203-N200-O202 | 126.9(13) |
| N59-Ni1-O8    | 84.4(2)  | O203-N200-O201 | 116.3(12) |
| N66-Ni2-N72   | 163.1(4) | O202-N200-O201 | 116.1(12) |
| N66-Ni2-O6    | 82.3(3)  | O302-N300-O300 | 116.0(13) |
| N72-Ni2-O6    | 87.5(3)  | O302-N300-O301 | 125.3(14) |
| N66-Ni2-N63   | 82.9(3)  | O300-N300-O301 | 117.4(13) |
| N72-Ni2-N63   | 83.4(4)  | O301-N30A-O30A | 113.3(14) |
| O6-Ni2-N63    | 88.7(3)  | O301-N30A-O302 | 126.2(14) |
| N66-Ni2-N69   | 94.7(4)  | O30A-N30A-O302 | 120.5(14) |
| N72-Ni2-N69   | 93.5(4)  | N30A-O301-N300 | 49.1(13)  |
| O6-Ni2-N69    | 171.9(3) | N300-O302-N30A | 47.8(13)  |
| N63-Ni2-N69   | 83.4(3)  |                |           |

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Symmetry transformations used to generate equivalent atoms:



**Figure S4.** Thermal ellipsoid plot of the molecular structure of the cation of **3**. Thermal ellipsoids are drawn at the 50% probability level. Hydrogen atoms, water molecules, and  $\text{NO}_3^-$  ions are omitted for clarity.

Table S5. Crystal data and structure refinement for **3**.

|                                    |   |
|------------------------------------|---|
| Crystal data                       |   |
| Identification code                | ab1341  |
| Habitus, colour                    | needle, colourless  |
| Crystal size                       | 0.36 x 0.08 x 0.05 mm <sup>3</sup>  |
| Crystal system                     | Triclinic   |
| Space group                        | P -1  |
| Unit cell dimensions               | Z = 2   |
|                                    | a = 16.2644(15) Å   |
|                                    | b = 16.3118(12) Å   |
|                                    | c = 19.4070(14) Å   |
|                                    | $\alpha = 110.568(5)^\circ$ .   |
|                                    | $\beta = 111.377(6)^\circ$ .  |
|                                    | $\gamma = 88.898(7)^\circ$ .  |
| Volume                             | 4453.8(6) Å <sup>3</sup>  |
| Cell determination                 | 11535 peaks with Theta 1.2 to 23.8°.  |
| Empirical formula                  | C <sub>56</sub> H <sub>124</sub> Er N <sub>19</sub> Ni <sub>2</sub> O <sub>41</sub> |
| Formula weight                     | 2004.42   |
| Density (calculated)               | 1.495 Mg/m <sup>3</sup>   |
| Absorption coefficient             | 1.449 mm <sup>-1</sup>  |
| F(000)                             | 2090  |
| Data collection:                   |   |
| Diffractometer type                | STOE IPDS 2   |
| Wavelength                         | 0.71073 Å   |
| Temperature                        | 100(2) K  |
| Theta range for data collection    | 1.36 to 25.12°.   |
| Index ranges                       | -19<=h<=19, -19<=k<=18, -22<=l<=23  |
| Data collection software           | STOE X-AREA (Stoe & Cie., 2006)   |
| Cell refinement software           | STOE X-AREA   |
| Data reduction software            | STOE X-AREA   |
| Solution and refinement:           |   |
| Reflections collected              | 30546   |
| Independent reflections            | 15737 [R(int) = 0.1237]   |
| Completeness to theta = 25.12°     | 98.8 %  |
| Observed reflections               | 5500[I>2sigma(I)]   |
| Reflections used for refinement    | 15737   |
| Absorption correction              | Semi-empirical from equivalents   |
| Max. and min. transmission         | 0.9397 and 0.7459   |
| Largest diff. peak and hole        | 0.882 and -1.919 e.Å <sup>-3</sup>  |
| Solution                           | Direct methods  |
| Refinement                         | Full-matrix least-squares on F <sup>2</sup>   |
| Treatment of hydrogen atoms        | Calculated positions, riding model  |
| Programs used                      | SIR2008 (Burla et al., 2007)  |
|                                    | SHELXL-97 (Sheldrick, 2008)   |
|                                    | DIAMOND (Crystal Impact)  |
|                                    | STOE IPDS2 software   |
| Data / restraints / parameters     | 15737 / 160 / 1062  |
| Goodness-of-fit on F <sup>2</sup>  | 0.747   |
| R index (all data)                 | wR2 = 0.1908  |
| R index conventional [I>2sigma(I)] | R1 = 0.0707   |

Table S6. Bond lengths [Å] and angles [°] for **3**.

|          |           |          |           |
|----------|-----------|----------|-----------|
| C1-O2    | 1.228(13) | C28-C29  | 1.420(15) |
| C1-O1    | 1.275(15) | C29-O9   | 1.280(12) |
| C1-C2    | 1.490(17) | C29-C30  | 1.459(12) |
| C2-C7    | 1.370(15) | C30-C34  | 1.399(16) |
| C2-C3    | 1.422(15) | C30-C31  | 1.407(17) |
| C3-O3    | 1.298(13) | C31-C32  | 1.379(17) |
| C3-C4    | 1.447(16) | C31-H31  | 0.9500    |
| C4-C5    | 1.387(16) | C32-C33  | 1.412(14) |
| C4-C8    | 1.452(16) | C32-H32  | 0.9500    |
| C5-C6    | 1.349(16) | C33-H33  | 0.9500    |
| C5-H5    | 0.9500    | C34-N35  | 1.304(15) |
| C6-C7    | 1.427(17) | C34-H34  | 0.9500    |
| C6-H6    | 0.9500    | C36-N35  | 1.459(15) |
| C7-H7    | 0.9500    | C36-C37  | 1.516(17) |
| C8-N9    | 1.271(15) | C36-H36A | 0.9900    |
| C8-H8    | 0.9500    | C36-H36B | 0.9900    |
| C10-N9   | 1.476(15) | C37-N38  | 1.472(14) |
| C10-C11  | 1.551(15) | C37-H37A | 0.9900    |
| C10-H10A | 0.9900    | C37-H37B | 0.9900    |
| C10-H10B | 0.9900    | C39-N38  | 1.480(16) |
| C11-N12  | 1.438(14) | C39-C40  | 1.540(15) |
| C11-H11A | 0.9900    | C39-H39A | 0.9900    |
| C11-H11B | 0.9900    | C39-H39B | 0.9900    |
| C13-N12  | 1.470(13) | C40-N41  | 1.470(15) |
| C13-C14  | 1.475(16) | C40-H40A | 0.9900    |
| C13-H13A | 0.9900    | C40-H40B | 0.9900    |
| C13-H13B | 0.9900    | C42-N41  | 1.310(15) |
| C14-N15  | 1.467(13) | C42-C43  | 1.451(17) |
| C14-H14A | 0.9900    | C42-H42  | 0.9500    |
| C14-H14B | 0.9900    | C43-C48  | 1.378(17) |
| C16-N15  | 1.284(15) | C43-C44  | 1.404(17) |
| C16-C17  | 1.425(16) | C44-O10  | 1.303(14) |
| C16-H16  | 0.9500    | C44-C45  | 1.461(16) |
| C17-C22  | 1.410(16) | C45-C46  | 1.343(17) |
| C17-C18  | 1.459(13) | C45-C49  | 1.468(17) |
| C18-O4   | 1.295(12) | C46-C47  | 1.397(18) |
| C18-C19  | 1.445(14) | C46-H46  | 0.9500    |
| C19-C20  | 1.348(15) | C47-C48  | 1.434(17) |
| C19-C23  | 1.541(13) | C47-H47  | 0.9500    |
| C20-C21  | 1.420(14) | C48-H48  | 0.9500    |
| C20-H20  | 0.9500    | C49-O11  | 1.216(15) |
| C21-C22  | 1.353(15) | C49-O12  | 1.289(14) |
| C21-H21  | 0.9500    | C50-N38  | 1.470(16) |
| C22-H22  | 0.9500    | C50-C51  | 1.535(18) |
| C23-O5   | 1.226(13) | C50-H50A | 0.9900    |
| C23-O6   | 1.255(13) | C50-H50B | 0.9900    |
| C24-N12  | 1.491(15) | C51-N52  | 1.415(14) |
| C24-C25  | 1.518(15) | C51-H51A | 0.9900    |
| C24-H24A | 0.9900    | C51-H51B | 0.9900    |
| C24-H24B | 0.9900    | C54-N53  | 1.495(12) |
| C25-N26  | 1.481(13) | C54-C55  | 1.537(15) |
| C25-H25A | 0.9900    | C54-H54A | 0.9900    |
| C25-H25B | 0.9900    | C54-H54B | 0.9900    |
| C27-O8   | 1.232(13) | C55-N56  | 1.474(14) |
| C27-O7   | 1.281(13) | C55-H55A | 0.9900    |
| C27-C28  | 1.508(13) | C55-H55B | 0.9900    |
| C28-C33  | 1.390(15) | C57-N53  | 1.481(13) |

|          |           |               |           |
|----------|-----------|---------------|-----------|
| C57-C58  | 1.561(15) | N56-H56B      | 0.9200    |
| C57-H57A | 0.9900    | N59-Ni1       | 2.070(8)  |
| C57-H57B | 0.9900    | N59-H59A      | 0.9200    |
| C58-N59  | 1.429(16) | N59-H59B      | 0.9200    |
| C58-H58A | 0.9900    | N62-Ni1       | 2.134(10) |
| C58-H58B | 0.9900    | N62-H62A      | 0.9200    |
| C60-N53  | 1.474(14) | N62-H62B      | 0.9200    |
| C60-C61  | 1.542(16) | N63-Ni2       | 2.097(12) |
| C60-H60A | 0.9900    | N66-Ni2       | 2.110(10) |
| C60-H60B | 0.9900    | N66-H66A      | 0.9200    |
| C61-N62  | 1.445(15) | N66-H66B      | 0.9200    |
| C61-H61A | 0.9900    | N69-Ni2       | 2.097(11) |
| C61-H61B | 0.9900    | N69-H69A      | 0.9200    |
| C64-C65  | 1.38(2)   | N69-H69B      | 0.9200    |
| C64-N63  | 1.510(15) | N72-Ni2       | 2.109(11) |
| C64-H64A | 0.9900    | N72-H72A      | 0.9200    |
| C64-H64B | 0.9900    | N72-H72B      | 0.9200    |
| C65-N66  | 1.384(19) | O1-Er1        | 2.318(8)  |
| C65-H65A | 0.9900    | O3-Er1        | 2.355(7)  |
| C65-H65B | 0.9900    | O3-H3         | 0.9500    |
| C67-N63  | 1.468(15) | O4-Er1        | 2.300(7)  |
| C67-C68  | 1.51(2)   | O4-H4         | 0.9500    |
| C67-H67A | 0.9900    | O5-Er1        | 2.323(8)  |
| C67-H67B | 0.9900    | O6-Ni2        | 2.124(7)  |
| C68-N69  | 1.496(19) | O7-Er1        | 2.299(9)  |
| C68-H68A | 0.9900    | O8-Ni1        | 2.116(7)  |
| C68-H68B | 0.9900    | O9-Er1        | 2.294(7)  |
| C70-C71  | 1.488(17) | O9-H9         | 0.9500    |
| C70-N63  | 1.499(16) | O10-Er1       | 2.349(8)  |
| C70-H70A | 0.9900    | O10-H10       | 0.9500    |
| C70-H70B | 0.9900    | O11-Er1       | 2.310(9)  |
| C71-N72  | 1.468(16) | N300-O302     | 1.297(15) |
| C71-H71A | 0.9900    | N300-O300     | 1.316(15) |
| C71-H71B | 0.9900    | N300-O301     | 1.335(15) |
| N26-Ni1  | 2.103(9)  | N301-O303     | 1.304(14) |
| N26-H26A | 0.9200    | N301-O302     | 1.329(14) |
| N26-H26B | 0.9200    | N301-O301     | 1.356(14) |
| N52-Ni2  | 2.091(11) | N200-O202     | 1.348(14) |
| N52-H52A | 0.9200    | N200-O200     | 1.389(15) |
| N52-H52B | 0.9200    | N200-O201     | 1.399(14) |
| N53-Ni1  | 2.091(10) | N100-O101     | 1.304(14) |
| N56-Ni1  | 2.098(9)  | N100-O102     | 1.319(17) |
| N56-H56A | 0.9200    | N100-O100     | 1.340(15) |
| O2-C1-O1 | 123.2(13) | C7-C6-H6      | 121.0     |
| O2-C1-C2 | 117.6(13) | C2-C7-C6      | 122.5(12) |
| O1-C1-C2 | 119.1(12) | C2-C7-H7      | 118.8     |
| C7-C2-C3 | 119.9(12) | C6-C7-H7      | 118.8     |
| C7-C2-C1 | 119.2(11) | N9-C8-C4      | 125.0(13) |
| C3-C2-C1 | 120.9(12) | N9-C8-H8      | 117.5     |
| O3-C3-C2 | 122.6(11) | C4-C8-H8      | 117.5     |
| O3-C3-C4 | 120.8(11) | N9-C10-C11    | 108.1(10) |
| C2-C3-C4 | 116.6(12) | N9-C10-H10A   | 110.1     |
| C5-C4-C3 | 120.9(11) | C11-C10-H10A  | 110.1     |
| C5-C4-C8 | 121.5(13) | N9-C10-H10B   | 110.1     |
| C3-C4-C8 | 117.5(12) | C11-C10-H10B  | 110.1     |
| C6-C5-C4 | 121.9(13) | H10A-C10-H10B | 108.4     |
| C6-C5-H5 | 119.0     | N12-C11-C10   | 112.1(9)  |
| C4-C5-H5 | 119.0     | N12-C11-H11A  | 109.2     |
| C5-C6-C7 | 118.0(13) | C10-C11-H11A  | 109.2     |
| C5-C6-H6 | 121.0     | N12-C11-H11B  | 109.2     |

|               |           |               |           |
|---------------|-----------|---------------|-----------|
| C10-C11-H11B  | 109.2     | C31-C30-C29   | 118.5(11) |
| H11A-C11-H11B | 107.9     | C32-C31-C30   | 123.5(10) |
| N12-C13-C14   | 112.3(10) | C32-C31-H31   | 118.2     |
| N12-C13-H13A  | 109.1     | C30-C31-H31   | 118.2     |
| C14-C13-H13A  | 109.1     | C31-C32-C33   | 116.9(12) |
| N12-C13-H13B  | 109.1     | C31-C32-H32   | 121.6     |
| C14-C13-H13B  | 109.1     | C33-C32-H32   | 121.6     |
| H13A-C13-H13B | 107.9     | C28-C33-C32   | 123.1(11) |
| N15-C14-C13   | 110.3(10) | C28-C33-H33   | 118.5     |
| N15-C14-H14A  | 109.6     | C32-C33-H33   | 118.5     |
| C13-C14-H14A  | 109.6     | N35-C34-C30   | 126.3(10) |
| N15-C14-H14B  | 109.6     | N35-C34-H34   | 116.8     |
| C13-C14-H14B  | 109.6     | C30-C34-H34   | 116.8     |
| H14A-C14-H14B | 108.1     | N35-C36-C37   | 109.3(10) |
| N15-C16-C17   | 127.4(11) | N35-C36-H36A  | 109.8     |
| N15-C16-H16   | 116.3     | C37-C36-H36A  | 109.8     |
| C17-C16-H16   | 116.3     | N35-C36-H36B  | 109.8     |
| C22-C17-C16   | 119.1(10) | C37-C36-H36B  | 109.8     |
| C22-C17-C18   | 121.3(11) | H36A-C36-H36B | 108.3     |
| C16-C17-C18   | 119.6(12) | N38-C37-C36   | 109.0(9)  |
| O4-C18-C19    | 125.2(9)  | N38-C37-H37A  | 109.9     |
| O4-C18-C17    | 119.7(10) | C36-C37-H37A  | 109.9     |
| C19-C18-C17   | 115.1(11) | N38-C37-H37B  | 109.9     |
| C20-C19-C18   | 120.2(10) | C36-C37-H37B  | 109.9     |
| C20-C19-C23   | 118.3(10) | H37A-C37-H37B | 108.3     |
| C18-C19-C23   | 121.2(10) | N38-C39-C40   | 110.9(10) |
| C19-C20-C21   | 124.2(11) | N38-C39-H39A  | 109.5     |
| C19-C20-H20   | 117.9     | C40-C39-H39A  | 109.5     |
| C21-C20-H20   | 117.9     | N38-C39-H39B  | 109.5     |
| C22-C21-C20   | 117.8(12) | C40-C39-H39B  | 109.5     |
| C22-C21-H21   | 121.1     | H39A-C39-H39B | 108.0     |
| C20-C21-H21   | 121.1     | N41-C40-C39   | 107.9(10) |
| C21-C22-C17   | 121.2(10) | N41-C40-H40A  | 110.1     |
| C21-C22-H22   | 119.4     | C39-C40-H40A  | 110.1     |
| C17-C22-H22   | 119.4     | N41-C40-H40B  | 110.1     |
| O5-C23-O6     | 126.3(9)  | C39-C40-H40B  | 110.1     |
| O5-C23-C19    | 120.5(11) | H40A-C40-H40B | 108.4     |
| O6-C23-C19    | 113.1(11) | N41-C42-C43   | 122.2(13) |
| N12-C24-C25   | 111.5(10) | N41-C42-H42   | 118.9     |
| N12-C24-H24A  | 109.3     | C43-C42-H42   | 118.9     |
| C25-C24-H24A  | 109.3     | C48-C43-C44   | 122.5(14) |
| N12-C24-H24B  | 109.3     | C48-C43-C42   | 117.1(14) |
| C25-C24-H24B  | 109.3     | C44-C43-C42   | 120.2(13) |
| H24A-C24-H24B | 108.0     | O10-C44-C43   | 120.0(12) |
| N26-C25-C24   | 111.8(9)  | O10-C44-C45   | 121.7(13) |
| N26-C25-H25A  | 109.3     | C43-C44-C45   | 118.2(13) |
| C24-C25-H25A  | 109.3     | C46-C45-C44   | 116.3(14) |
| N26-C25-H25B  | 109.3     | C46-C45-C49   | 125.0(13) |
| C24-C25-H25B  | 109.3     | C44-C45-C49   | 118.6(12) |
| H25A-C25-H25B | 107.9     | C45-C46-C47   | 127.6(15) |
| O8-C27-O7     | 126.2(9)  | C45-C46-H46   | 116.2     |
| O8-C27-C28    | 116.6(11) | C47-C46-H46   | 116.2     |
| O7-C27-C28    | 117.2(11) | C46-C47-C48   | 115.3(13) |
| C33-C28-C29   | 119.9(9)  | C46-C47-H47   | 122.4     |
| C33-C28-C27   | 116.0(10) | C48-C47-H47   | 122.4     |
| C29-C28-C27   | 124.1(11) | C43-C48-C47   | 120.0(14) |
| O9-C29-C28    | 124.6(9)  | C43-C48-H48   | 120.0     |
| O9-C29-C30    | 117.4(10) | C47-C48-H48   | 120.0     |
| C28-C29-C30   | 118.0(11) | O11-C49-O12   | 123.0(13) |
| C34-C30-C31   | 119.2(10) | O11-C49-C45   | 123.1(13) |
| C34-C30-C29   | 122.2(11) | O12-C49-C45   | 113.6(14) |

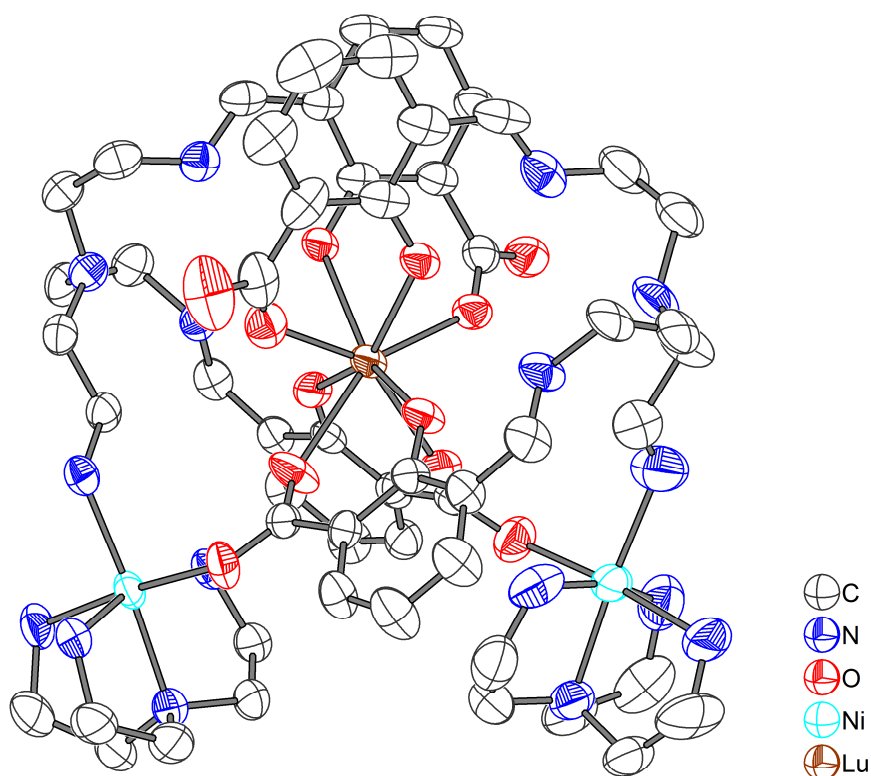
|               |           |               |           |
|---------------|-----------|---------------|-----------|
| N38-C50-C51   | 112.1(11) | N63-C67-H67A  | 109.0     |
| N38-C50-H50A  | 109.2     | C68-C67-H67A  | 109.0     |
| C51-C50-H50A  | 109.2     | N63-C67-H67B  | 109.0     |
| N38-C50-H50B  | 109.2     | C68-C67-H67B  | 109.0     |
| C51-C50-H50B  | 109.2     | H67A-C67-H67B | 107.8     |
| H50A-C50-H50B | 107.9     | N69-C68-C67   | 107.5(12) |
| N52-C51-C50   | 114.7(11) | N69-C68-H68A  | 110.2     |
| N52-C51-H51A  | 108.6     | C67-C68-H68A  | 110.2     |
| C50-C51-H51A  | 108.6     | N69-C68-H68B  | 110.2     |
| N52-C51-H51B  | 108.6     | C67-C68-H68B  | 110.2     |
| C50-C51-H51B  | 108.6     | H68A-C68-H68B | 108.5     |
| H51A-C51-H51B | 107.6     | C71-C70-N63   | 109.1(10) |
| N53-C54-C55   | 109.6(8)  | C71-C70-H70A  | 109.9     |
| N53-C54-H54A  | 109.8     | N63-C70-H70A  | 109.9     |
| C55-C54-H54A  | 109.8     | C71-C70-H70B  | 109.9     |
| N53-C54-H54B  | 109.8     | N63-C70-H70B  | 109.9     |
| C55-C54-H54B  | 109.8     | H70A-C70-H70B | 108.3     |
| H54A-C54-H54B | 108.2     | N72-C71-C70   | 112.7(12) |
| N56-C55-C54   | 110.6(10) | N72-C71-H71A  | 109.0     |
| N56-C55-H55A  | 109.5     | C70-C71-H71A  | 109.0     |
| C54-C55-H55A  | 109.5     | N72-C71-H71B  | 109.0     |
| N56-C55-H55B  | 109.5     | C70-C71-H71B  | 109.0     |
| C54-C55-H55B  | 109.5     | H71A-C71-H71B | 107.8     |
| H55A-C55-H55B | 108.1     | C8-N9-C10     | 125.8(12) |
| N53-C57-C58   | 111.1(10) | C11-N12-C13   | 110.7(9)  |
| N53-C57-H57A  | 109.4     | C11-N12-C24   | 112.7(10) |
| C58-C57-H57A  | 109.4     | C13-N12-C24   | 113.7(10) |
| N53-C57-H57B  | 109.4     | C16-N15-C14   | 123.6(10) |
| C58-C57-H57B  | 109.4     | C25-N26-Ni1   | 118.6(7)  |
| H57A-C57-H57B | 108.0     | C25-N26-H26A  | 107.7     |
| N59-C58-C57   | 112.6(11) | Ni1-N26-H26A  | 107.7     |
| N59-C58-H58A  | 109.1     | C25-N26-H26B  | 107.7     |
| C57-C58-H58A  | 109.1     | Ni1-N26-H26B  | 107.7     |
| N59-C58-H58B  | 109.1     | H26A-N26-H26B | 107.1     |
| C57-C58-H58B  | 109.1     | C34-N35-C36   | 123.3(9)  |
| H58A-C58-H58B | 107.8     | C50-N38-C37   | 113.7(11) |
| N53-C60-C61   | 109.2(10) | C50-N38-C39   | 111.6(11) |
| N53-C60-H60A  | 109.8     | C37-N38-C39   | 110.2(10) |
| C61-C60-H60A  | 109.8     | C42-N41-C40   | 123.5(12) |
| N53-C60-H60B  | 109.8     | C51-N52-Ni2   | 119.1(8)  |
| C61-C60-H60B  | 109.8     | C51-N52-H52A  | 107.5     |
| H60A-C60-H60B | 108.3     | Ni2-N52-H52A  | 107.5     |
| N62-C61-C60   | 111.4(11) | C51-N52-H52B  | 107.5     |
| N62-C61-H61A  | 109.4     | Ni2-N52-H52B  | 107.5     |
| C60-C61-H61A  | 109.4     | H52A-N52-H52B | 107.0     |
| N62-C61-H61B  | 109.4     | C60-N53-C57   | 111.3(10) |
| C60-C61-H61B  | 109.4     | C60-N53-C54   | 111.8(8)  |
| H61A-C61-H61B | 108.0     | C57-N53-C54   | 109.5(8)  |
| C65-C64-N63   | 114.9(14) | C60-N53-Ni1   | 108.4(7)  |
| C65-C64-H64A  | 108.5     | C57-N53-Ni1   | 109.7(7)  |
| N63-C64-H64A  | 108.5     | C54-N53-Ni1   | 106.0(7)  |
| C65-C64-H64B  | 108.5     | C55-N56-Ni1   | 111.0(7)  |
| N63-C64-H64B  | 108.5     | C55-N56-H56A  | 109.4     |
| H64A-C64-H64B | 107.5     | Ni1-N56-H56A  | 109.4     |
| C64-C65-N66   | 122.3(13) | C55-N56-H56B  | 109.4     |
| C64-C65-H65A  | 106.8     | Ni1-N56-H56B  | 109.4     |
| N66-C65-H65A  | 106.8     | H56A-N56-H56B | 108.0     |
| C64-C65-H65B  | 106.8     | C58-N59-Ni1   | 108.8(7)  |
| N66-C65-H65B  | 106.8     | C58-N59-H59A  | 109.9     |
| H65A-C65-H65B | 106.6     | Ni1-N59-H59A  | 109.9     |
| N63-C67-C68   | 113.1(12) | C58-N59-H59B  | 109.9     |

|               |           |                |           |
|---------------|-----------|----------------|-----------|
| Ni1-N59-H59B  | 109.9     | N53-Ni1-N62    | 81.4(4)   |
| H59A-N59-H59B | 108.3     | N56-Ni1-N62    | 162.9(4)  |
| C61-N62-Ni1   | 111.4(7)  | N26-Ni1-N62    | 96.4(4)   |
| C61-N62-H62A  | 109.3     | O8-Ni1-N62     | 82.2(3)   |
| Ni1-N62-H62A  | 109.3     | N52-Ni2-N63    | 177.8(4)  |
| C61-N62-H62B  | 109.3     | N52-Ni2-N69    | 95.7(5)   |
| Ni1-N62-H62B  | 109.3     | N63-Ni2-N69    | 83.3(5)   |
| H62A-N62-H62B | 108.0     | N52-Ni2-N72    | 99.1(4)   |
| C67-N63-C70   | 113.6(10) | N63-Ni2-N72    | 82.3(5)   |
| C67-N63-C64   | 111.8(10) | N69-Ni2-N72    | 162.6(5)  |
| C70-N63-C64   | 110.4(11) | N52-Ni2-N66    | 95.7(5)   |
| C67-N63-Ni2   | 104.9(9)  | N63-Ni2-N66    | 82.5(5)   |
| C70-N63-Ni2   | 106.3(8)  | N69-Ni2-N66    | 95.0(5)   |
| C64-N63-Ni2   | 109.4(8)  | N72-Ni2-N66    | 92.7(4)   |
| C65-N66-Ni2   | 109.5(9)  | N52-Ni2-O6     | 91.0(4)   |
| C65-N66-H66A  | 109.8     | N63-Ni2-O6     | 90.7(3)   |
| Ni2-N66-H66A  | 109.8     | N69-Ni2-O6     | 80.4(3)   |
| C65-N66-H66B  | 109.8     | N72-Ni2-O6     | 90.2(3)   |
| Ni2-N66-H66B  | 109.8     | N66-Ni2-O6     | 172.2(4)  |
| H66A-N66-H66B | 108.2     | O9-Er1-O7      | 73.0(3)   |
| C68-N69-Ni2   | 111.0(10) | O9-Er1-O4      | 144.1(3)  |
| C68-N69-H69A  | 109.4     | O7-Er1-O4      | 78.2(3)   |
| Ni2-N69-H69A  | 109.4     | O9-Er1-O11     | 86.4(3)   |
| C68-N69-H69B  | 109.4     | O7-Er1-O11     | 72.7(3)   |
| Ni2-N69-H69B  | 109.4     | O4-Er1-O11     | 105.3(3)  |
| H69A-N69-H69B | 108.0     | O9-Er1-O1      | 105.7(3)  |
| C71-N72-Ni2   | 110.0(8)  | O7-Er1-O1      | 144.5(3)  |
| C71-N72-H72A  | 109.7     | O4-Er1-O1      | 85.7(2)   |
| Ni2-N72-H72A  | 109.7     | O11-Er1-O1     | 142.6(3)  |
| C71-N72-H72B  | 109.7     | O9-Er1-O5      | 77.8(3)   |
| Ni2-N72-H72B  | 109.7     | O7-Er1-O5      | 73.2(3)   |
| H72A-N72-H72B | 108.2     | O4-Er1-O5      | 73.5(2)   |
| C1-O1-Er1     | 133.7(7)  | O11-Er1-O5     | 145.3(3)  |
| C3-O3-Er1     | 129.3(6)  | O1-Er1-O5      | 71.9(3)   |
| C3-O3-H3      | 115.4     | O9-Er1-O10     | 71.1(3)   |
| Er1-O3-H3     | 115.4     | O7-Er1-O10     | 131.7(3)  |
| C18-O4-Er1    | 137.0(6)  | O4-Er1-O10     | 144.6(3)  |
| C18-O4-H4     | 111.5     | O11-Er1-O10    | 73.8(3)   |
| Er1-O4-H4     | 111.5     | O1-Er1-O10     | 76.9(3)   |
| C23-O5-Er1    | 139.8(7)  | O5-Er1-O10     | 127.4(2)  |
| C23-O6-Ni2    | 131.6(7)  | O9-Er1-O3      | 144.7(3)  |
| C27-O7-Er1    | 140.7(7)  | O7-Er1-O3      | 127.8(2)  |
| C27-O8-Ni1    | 130.3(7)  | O4-Er1-O3      | 71.0(3)   |
| C29-O9-Er1    | 137.6(6)  | O11-Er1-O3     | 76.1(3)   |
| C29-O9-H9     | 111.2     | O1-Er1-O3      | 74.0(3)   |
| Er1-O9-H9     | 111.2     | O5-Er1-O3      | 131.9(3)  |
| C44-O10-Er1   | 130.1(7)  | O10-Er1-O3     | 74.6(3)   |
| C44-O10-H10   | 115.0     | O302-N300-O300 | 110(2)    |
| Er1-O10-H10   | 115.0     | O302-N300-O301 | 105.7(18) |
| C49-O11-Er1   | 133.5(8)  | O300-N300-O301 | 145(2)    |
| N59-Ni1-N53   | 84.6(4)   | O303-N301-O302 | 123.7(16) |
| N59-Ni1-N56   | 93.0(4)   | O303-N301-O301 | 127.0(17) |
| N53-Ni1-N56   | 83.1(4)   | O302-N301-O301 | 102.8(16) |
| N59-Ni1-N26   | 93.2(4)   | N300-O301-N301 | 67.0(17)  |
| N53-Ni1-N26   | 176.8(3)  | N300-O302-N301 | 68.9(17)  |
| N56-Ni1-N26   | 99.4(4)   | O202-N200-O200 | 116.3(13) |
| N59-Ni1-O8    | 173.5(4)  | O202-N200-O201 | 125.2(14) |
| N53-Ni1-O8    | 90.7(3)   | O200-N200-O201 | 115.9(12) |
| N56-Ni1-O8    | 90.8(3)   | O101-N100-O102 | 119.6(15) |
| N26-Ni1-O8    | 91.4(3)   | O101-N100-O100 | 116.5(13) |
| N59-Ni1-N62   | 92.7(4)   | O102-N100-O100 | 110.9(14) |



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Symmetry transformations used to generate equivalent atoms:



**Figure S5.** Thermal ellipsoid plot of the molecular structure of the cation of **4**. Thermal ellipsoids are drawn at the 50% probability level. Hydrogen atoms, water molecules, and  $\text{NO}_3^-$  ions are omitted for clarity.

Table S7. Crystal data and structure refinement for **4**.

Crystal data

|                        |                                      |                 |
|------------------------|--------------------------------------|-----------------|
| Identification code    | ab1342                               |                 |
| Habitus, colour        | prism, colourless                    |                 |
| Crystal size           | 0.49 x 0.23 x 0.10 mm <sup>3</sup>   |                 |
| Crystal system         | Triclinic                            |                 |
| Space group            | P -1                                 | Z = 2           |
| Unit cell dimensions   | a = 16.3087(6) Å                     | α = 110.903(3)° |
|                        | b = 16.3388(5) Å                     | β = 110.911(3)° |
|                        | c = 19.2433(7) Å                     | γ = 90.224(3)°  |
| Volume                 | 4423.8(3) Å <sup>3</sup>             |                 |
| Cell determination     | 36150 peaks with Theta 1.2 to 27.1°. |                 |
| Empirical formula      | C56 H122 Lu N19 Ni2 O40              |                 |
| Formula weight         | 1994.12                              |                 |
| Density (calculated)   | 1.497 Mg/m <sup>3</sup>              |                 |
| Absorption coefficient | 1.625 mm <sup>-1</sup>               |                 |
| F(000)                 | 2076                                 |                 |

Data collection:

|                                 |  |
|---------------------------------|--|
| Diffractometer type             | STOE IPDS 2                              |
| Wavelength                      | 0.71073 Å                                |
| Temperature                     | 100(2) K                                 |
| Theta range for data collection | 1.23 to 25.12°                           |
| Index ranges                    | -19 ≤ h ≤ 19, -19 ≤ k ≤ 19, -22 ≤ l ≤ 21 |
| Data collection software        | STOE X-AREA                              |
| Cell refinement software        | STOE X-AREA                              |
| Data reduction software         | STOE X-AREA                              |

Solution and refinement:

|                                   |  |
|-----------------------------------|--|
| Reflections collected             | 42995  |
| Independent reflections           | 15710 [R(int) = 0.0578]  |
| Completeness to theta = 25.12°    | 99.4 %   |
| Observed reflections              | 10841 [I > 2σ(I)]  |
| Reflections used for refinement   | 15710  |
| Absorption correction             | Semi-empirical from equivalents  |
| Max. and min. transmission        | 0.7758 and 0.5340  |
| Largest diff. peak and hole       | 1.446 and -1.023 e.Å <sup>-3</sup>   |
| Solution                          | Direct methods   |
| Refinement                        | Full-matrix least-squares on F <sup>2</sup>  |
| Treatment of hydrogen atoms       | Calculated positions, riding model   |
| Programs used                     | SIR-92<br>SHELXL-97 (Sheldrick, 1997)<br>DIAMOND (Crystal Impact)<br>STOE IPDS2 software |
| Data / restraints / parameters    | 15710 / 321 / 1225   |
| Goodness-of-fit on F <sup>2</sup> | 0.917  |
| R index (all data)                | wR2 = 0.1335   |
| R index conventional [I > 2σ(I)]  | R1 = 0.0512  |

Table S8. Bond lengths [Å] and angles [°] for **4**.

|          |           |          |           |
|----------|-----------|----------|-----------|
| C1-O2    | 1.246(8)  | C28-C29  | 1.435(8)  |
| C1-O1    | 1.264(8)  | C29-O9   | 1.277(7)  |
| C1-C222  | 1.493(9)  | C29-C30  | 1.439(9)  |
| C3-O3    | 1.306(7)  | C30-C34  | 1.400(9)  |
| C3-C222  | 1.429(10) | C30-C31  | 1.421(10) |
| C3-C4    | 1.430(9)  | C31-C32  | 1.366(10) |
| C4-C5    | 1.415(9)  | C31-H31  | 0.9500    |
| C4-C8    | 1.420(10) | C32-C33  | 1.399(10) |
| C5-C6    | 1.365(10) | C32-H32  | 0.9500    |
| C5-H5    | 0.9500    | C33-H33  | 0.9500    |
| C6-C7    | 1.408(9)  | C34-N35  | 1.306(9)  |
| C6-H6    | 0.9500    | C34-H34  | 0.9500    |
| C7-C222  | 1.401(9)  | C36-N35  | 1.476(8)  |
| C7-H7    | 0.9500    | C36-C37  | 1.479(13) |
| C8-N9    | 1.309(8)  | C36-H36A | 0.9900    |
| C8-H8    | 0.9500    | C36-H36B | 0.9900    |
| C10-N9   | 1.438(9)  | C37-N38  | 1.480(11) |
| C10-C11  | 1.502(10) | C37-H37A | 0.9900    |
| C10-H10A | 0.9900    | C37-H37B | 0.9900    |
| C10-H10B | 0.9900    | C39-N38  | 1.475(9)  |
| C11-N12  | 1.482(8)  | C39-C40  | 1.532(13) |
| C11-H11A | 0.9900    | C39-H39A | 0.9900    |
| C11-H11B | 0.9900    | C39-H39B | 0.9900    |
| C13-N12  | 1.487(8)  | C40-N41  | 1.443(11) |
| C13-C14  | 1.520(10) | C40-H40A | 0.9900    |
| C13-H13A | 0.9900    | C40-H40B | 0.9900    |
| C13-H13B | 0.9900    | C42-N41  | 1.315(10) |
| C14-N15  | 1.462(9)  | C42-C43  | 1.400(12) |
| C14-H14A | 0.9900    | C42-H42  | 0.9500    |
| C14-H14B | 0.9900    | C43-C44  | 1.421(10) |
| C16-N15  | 1.305(8)  | C43-C48  | 1.429(11) |
| C16-C17  | 1.417(9)  | C44-O10  | 1.308(8)  |
| C16-H16  | 0.9500    | C44-C45  | 1.430(11) |
| C17-C22  | 1.402(9)  | C45-C46  | 1.376(11) |
| C17-C18  | 1.438(9)  | C45-C49  | 1.503(12) |
| C18-O4   | 1.295(7)  | C46-C47  | 1.369(14) |
| C18-C19  | 1.425(9)  | C46-H46  | 0.9500    |
| C19-C20  | 1.374(9)  | C47-C48  | 1.397(14) |
| C19-C23  | 1.507(9)  | C47-H47  | 0.9500    |
| C20-C21  | 1.399(10) | C48-H48  | 0.9500    |
| C20-H20  | 0.9500    | C49-O11  | 1.206(10) |
| C21-C22  | 1.382(10) | C49-O12  | 1.290(10) |
| C21-H21  | 0.9500    | C50-N38  | 1.454(12) |
| C22-H22  | 0.9500    | C50-C51  | 1.580(12) |
| C23-O5   | 1.252(8)  | C50-H50A | 0.9900    |
| C23-O6   | 1.256(8)  | C50-H50B | 0.9900    |
| C24-N12  | 1.453(9)  | C51-N52  | 1.350(12) |
| C24-C25  | 1.509(9)  | C51-H51A | 0.9900    |
| C24-H24A | 0.9900    | C51-H51B | 0.9900    |
| C24-H24B | 0.9900    | C54-N53  | 1.477(8)  |
| C25-N26  | 1.490(8)  | C54-C55  | 1.509(10) |
| C25-H25A | 0.9900    | C54-H54A | 0.9900    |
| C25-H25B | 0.9900    | C54-H54B | 0.9900    |
| C27-O8   | 1.257(7)  | C55-N56  | 1.473(8)  |
| C27-O7   | 1.259(8)  | C55-H55A | 0.9900    |
| C27-C28  | 1.489(8)  | C55-H55B | 0.9900    |
| C28-C33  | 1.381(9)  | C57-N53  | 1.479(10) |

|            |           |               |           |
|------------|-----------|---------------|-----------|
| C57-C58    | 1.523(12) | N56-H56B      | 0.9200    |
| C57-H57A   | 0.9900    | N59-Ni1       | 2.095(5)  |
| C57-H57B   | 0.9900    | N59-H59A      | 0.9200    |
| C58-N59    | 1.473(10) | N59-H59B      | 0.9200    |
| C58-H58A   | 0.9900    | N62-Ni1       | 2.130(6)  |
| C58-H58B   | 0.9900    | N62-H62A      | 0.9200    |
| C60-N53    | 1.490(10) | N62-H62B      | 0.9200    |
| C60-C61    | 1.519(10) | N63-Ni2       | 2.089(6)  |
| C60-H60A   | 0.9900    | N66-Ni2       | 2.089(7)  |
| C60-H60B   | 0.9900    | N66-H66A      | 0.9200    |
| C61-N62    | 1.476(9)  | N66-H66B      | 0.9200    |
| C61-H61A   | 0.9900    | N69-Ni2       | 2.122(9)  |
| C61-H61B   | 0.9900    | N69-H69A      | 0.9200    |
| C64-C65    | 1.451(13) | N69-H69B      | 0.9200    |
| C64-N63    | 1.497(10) | N72-Ni2       | 2.086(7)  |
| C64-H64A   | 0.9900    | N72-H72A      | 0.9200    |
| C64-H64B   | 0.9900    | N72-H72B      | 0.9200    |
| C65-N66    | 1.338(12) | O1-Lu1        | 2.286(4)  |
| C65-H65A   | 0.9900    | O3-Lu1        | 2.319(4)  |
| C65-H65B   | 0.9900    | O3-H3         | 0.9500    |
| C67-N63    | 1.469(11) | O4-Lu1        | 2.280(4)  |
| C67-C68    | 1.470(14) | O4-H4         | 0.9500    |
| C67-H67A   | 0.9900    | O5-Lu1        | 2.311(5)  |
| C67-H67B   | 0.9900    | O6-Ni2        | 2.113(5)  |
| C68-N69    | 1.454(13) | O7-Lu1        | 2.291(4)  |
| C68-H68A   | 0.9900    | O8-Ni1        | 2.121(4)  |
| C68-H68B   | 0.9900    | O9-Lu1        | 2.262(4)  |
| C70-N63    | 1.474(11) | O9-H9         | 0.9500    |
| C70-C71    | 1.531(13) | O10-Lu1       | 2.309(4)  |
| C70-H70A   | 0.9900    | O10-H10       | 0.9500    |
| C70-H70B   | 0.9900    | O11-Lu1       | 2.288(6)  |
| C71-N72    | 1.463(11) | N100-O102     | 1.262(11) |
| C71-H71A   | 0.9900    | N100-O101     | 1.290(10) |
| C71-H71B   | 0.9900    | N100-O100     | 1.303(10) |
| N26-Ni1    | 2.091(5)  | N200-O201     | 1.330(12) |
| N26-H26A   | 0.9200    | N200-O202     | 1.383(12) |
| N26-H26B   | 0.9200    | N200-O200     | 1.482(14) |
| N52-Ni2    | 2.081(7)  | N300-O302     | 1.170(12) |
| N52-H52A   | 0.9200    | N300-O300     | 1.264(12) |
| N52-H52B   | 0.9200    | N300-O301     | 1.300(11) |
| N53-Ni1    | 2.096(5)  | O301-N301     | 1.256(13) |
| N56-Ni1    | 2.106(5)  | O302-N301     | 1.269(13) |
| N56-H56A   | 0.9200    | N301-O303     | 1.171(13) |
| O2-C1-O1   | 123.1(6)  | C6-C7-H7      | 119.3     |
| O2-C1-C222 | 118.2(6)  | N9-C8-C4      | 122.9(6)  |
| O1-C1-C222 | 118.6(7)  | N9-C8-H8      | 118.6     |
| O3-C3-C222 | 122.3(5)  | C4-C8-H8      | 118.6     |
| O3-C3-C4   | 120.3(6)  | N9-C10-C11    | 110.8(6)  |
| C222-C3-C4 | 117.5(5)  | N9-C10-H10A   | 109.5     |
| C5-C4-C8   | 118.9(6)  | C11-C10-H10A  | 109.5     |
| C5-C4-C3   | 120.4(6)  | N9-C10-H10B   | 109.5     |
| C8-C4-C3   | 120.7(6)  | C11-C10-H10B  | 109.5     |
| C6-C5-C4   | 121.2(6)  | H10A-C10-H10B | 108.1     |
| C6-C5-H5   | 119.4     | N12-C11-C10   | 111.7(6)  |
| C4-C5-H5   | 119.4     | N12-C11-H11A  | 109.3     |
| C5-C6-C7   | 119.4(6)  | C10-C11-H11A  | 109.3     |
| C5-C6-H6   | 120.3     | N12-C11-H11B  | 109.3     |
| C7-C6-H6   | 120.3     | C10-C11-H11B  | 109.3     |
| C222-C7-C6 | 121.3(7)  | H11A-C11-H11B | 107.9     |
| C222-C7-H7 | 119.3     | N12-C13-C14   | 110.5(5)  |

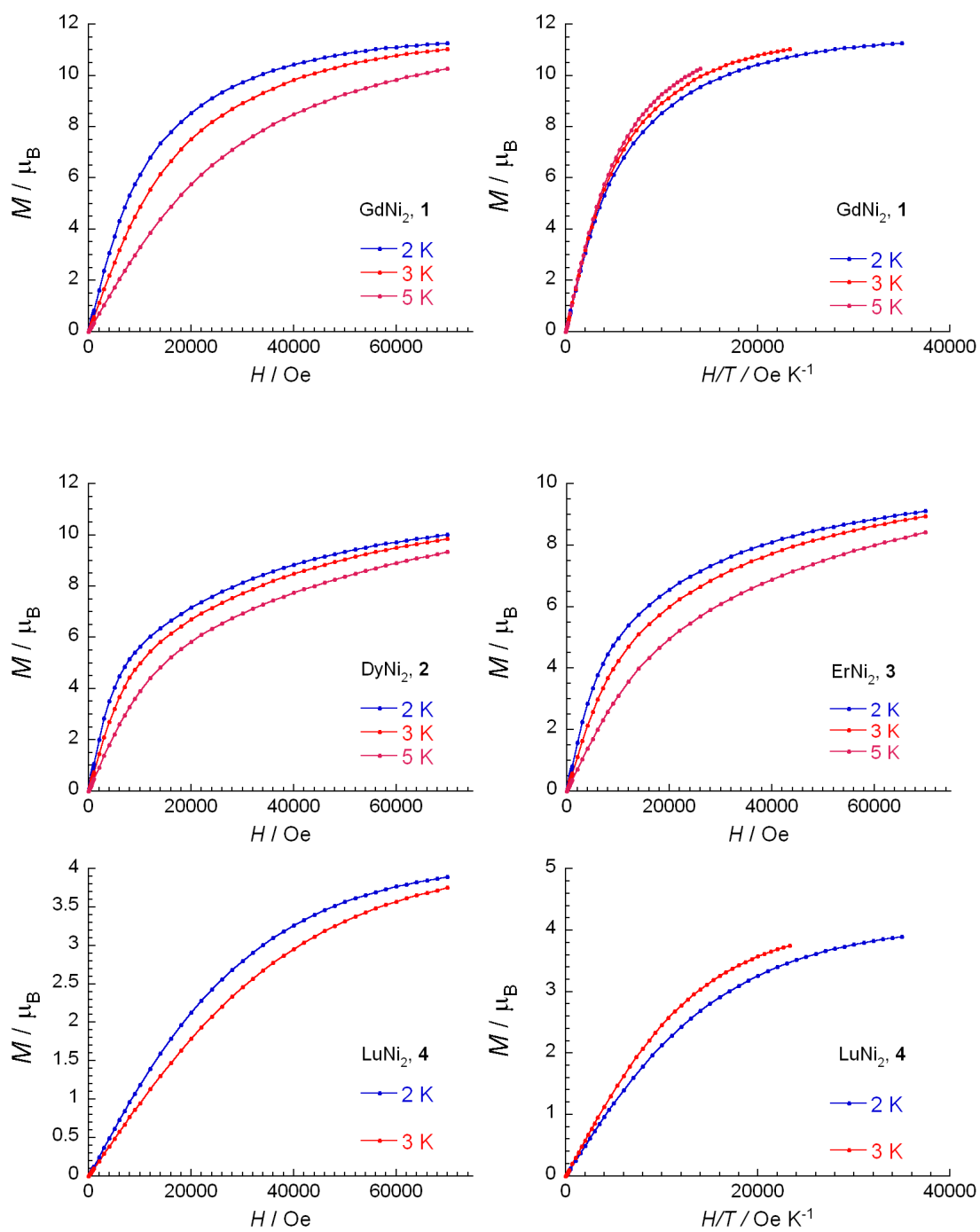
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|---------------|----------|---------------|-----------|
| N12-C13-H13A  | 109.5    | C32-C33-H33   | 118.5     |
| C14-C13-H13A  | 109.5    | N35-C34-C30   | 125.3(7)  |
| N12-C13-H13B  | 109.5    | N35-C34-H34   | 117.3     |
| C14-C13-H13B  | 109.5    | C30-C34-H34   | 117.3     |
| H13A-C13-H13B | 108.1    | N35-C36-C37   | 110.9(6)  |
| N15-C14-C13   | 110.3(6) | N35-C36-H36A  | 109.4     |
| N15-C14-H14A  | 109.6    | C37-C36-H36A  | 109.4     |
| C13-C14-H14A  | 109.6    | N35-C36-H36B  | 109.4     |
| N15-C14-H14B  | 109.6    | C37-C36-H36B  | 109.4     |
| C13-C14-H14B  | 109.6    | H36A-C36-H36B | 108.0     |
| H14A-C14-H14B | 108.1    | C36-C37-N38   | 110.1(7)  |
| N15-C16-C17   | 125.5(6) | C36-C37-H37A  | 109.6     |
| N15-C16-H16   | 117.3    | N38-C37-H37A  | 109.6     |
| C17-C16-H16   | 117.3    | C36-C37-H37B  | 109.6     |
| C22-C17-C16   | 116.4(6) | N38-C37-H37B  | 109.6     |
| C22-C17-C18   | 121.2(6) | H37A-C37-H37B | 108.2     |
| C16-C17-C18   | 122.3(6) | N38-C39-C40   | 111.4(6)  |
| O4-C18-C19    | 124.9(6) | N38-C39-H39A  | 109.3     |
| O4-C18-C17    | 118.1(6) | C40-C39-H39A  | 109.3     |
| C19-C18-C17   | 117.0(5) | N38-C39-H39B  | 109.3     |
| C20-C19-C18   | 119.6(6) | C40-C39-H39B  | 109.3     |
| C20-C19-C23   | 117.6(6) | H39A-C39-H39B | 108.0     |
| C18-C19-C23   | 122.8(6) | N41-C40-C39   | 109.0(8)  |
| C19-C20-C21   | 123.1(7) | N41-C40-H40A  | 109.9     |
| C19-C20-H20   | 118.4    | C39-C40-H40A  | 109.9     |
| C21-C20-H20   | 118.4    | N41-C40-H40B  | 109.9     |
| C22-C21-C20   | 118.7(7) | C39-C40-H40B  | 109.9     |
| C22-C21-H21   | 120.7    | H40A-C40-H40B | 108.3     |
| C20-C21-H21   | 120.7    | N41-C42-C43   | 125.0(7)  |
| C21-C22-C17   | 120.2(7) | N41-C42-H42   | 117.5     |
| C21-C22-H22   | 119.9    | C43-C42-H42   | 117.5     |
| C17-C22-H22   | 119.9    | C42-C43-C44   | 120.3(7)  |
| O5-C23-O6     | 123.3(6) | C42-C43-C48   | 118.0(7)  |
| O5-C23-C19    | 120.9(6) | C44-C43-C48   | 121.7(8)  |
| O6-C23-C19    | 115.8(6) | O10-C44-C43   | 119.8(7)  |
| N12-C24-C25   | 112.0(5) | O10-C44-C45   | 123.1(6)  |
| N12-C24-H24A  | 109.2    | C43-C44-C45   | 117.0(7)  |
| C25-C24-H24A  | 109.2    | C46-C45-C44   | 121.0(8)  |
| N12-C24-H24B  | 109.2    | C46-C45-C49   | 119.2(8)  |
| C25-C24-H24B  | 109.2    | C44-C45-C49   | 119.8(6)  |
| H24A-C24-H24B | 107.9    | C47-C46-C45   | 120.7(10) |
| N26-C25-C24   | 113.3(5) | C47-C46-H46   | 119.6     |
| N26-C25-H25A  | 108.9    | C45-C46-H46   | 119.6     |
| C24-C25-H25A  | 108.9    | C46-C47-C48   | 122.5(9)  |
| N26-C25-H25B  | 108.9    | C46-C47-H47   | 118.8     |
| C24-C25-H25B  | 108.9    | C48-C47-H47   | 118.8     |
| H25A-C25-H25B | 107.7    | C47-C48-C43   | 117.1(8)  |
| O8-C27-O7     | 122.8(6) | C47-C48-H48   | 121.5     |
| O8-C27-C28    | 116.5(6) | C43-C48-H48   | 121.5     |
| O7-C27-C28    | 120.6(5) | O11-C49-O12   | 122.6(8)  |
| C33-C28-C29   | 119.7(6) | O11-C49-C45   | 121.4(7)  |
| C33-C28-C27   | 117.4(6) | O12-C49-C45   | 115.9(8)  |
| C29-C28-C27   | 122.8(5) | N38-C50-C51   | 110.7(8)  |
| O9-C29-C28    | 124.2(6) | N38-C50-H50A  | 109.5     |
| O9-C29-C30    | 119.0(5) | C51-C50-H50A  | 109.5     |
| C28-C29-C30   | 116.8(5) | N38-C50-H50B  | 109.5     |
| C34-C30-C31   | 117.7(6) | C51-C50-H50B  | 109.5     |
| C34-C30-C29   | 121.7(6) | H50A-C50-H50B | 108.1     |
| C31-C30-C29   | 120.6(6) | N52-C51-C50   | 114.6(9)  |
| C32-C31-C30   | 120.7(7) | N52-C51-H51A  | 108.6     |
| C32-C31-H31   | 119.6    | C50-C51-H51A  | 108.6     |
| C30-C31-H31   | 119.6    | N52-C51-H51B  | 108.6     |
| C31-C32-C33   | 118.9(7) | C50-C51-H51B  | 108.6     |
| C31-C32-H32   | 120.5    | H51A-C51-H51B | 107.6     |
| C33-C32-H32   | 120.5    | N53-C54-C55   | 111.3(6)  |
| C28-C33-C32   | 122.9(6) | N53-C54-H54A  | 109.4     |
| C28-C33-H33   | 118.5    | C55-C54-H54A  | 109.4     |

|               |          |               |          |
|---------------|----------|---------------|----------|
| N53-C54-H54B  | 109.4    | C70-C71-H71B  | 109.6    |
| C55-C54-H54B  | 109.4    | H71A-C71-H71B | 108.1    |
| H54A-C54-H54B | 108.0    | C7-C222-C3    | 120.0(6) |
| N56-C55-C54   | 110.1(6) | C7-C222-C1    | 118.3(6) |
| N56-C55-H55A  | 109.6    | C3-C222-C1    | 121.6(5) |
| C54-C55-H55A  | 109.6    | C8-N9-C10     | 125.1(6) |
| N56-C55-H55B  | 109.6    | C24-N12-C11   | 111.7(5) |
| C54-C55-H55B  | 109.6    | C24-N12-C13   | 112.9(5) |
| H55A-C55-H55B | 108.2    | C11-N12-C13   | 110.5(5) |
| N53-C57-C58   | 113.1(6) | C16-N15-C14   | 122.6(6) |
| N53-C57-H57A  | 109.0    | C25-N26-Ni1   | 119.0(4) |
| C58-C57-H57A  | 109.0    | C25-N26-H26A  | 107.6    |
| N53-C57-H57B  | 109.0    | Ni1-N26-H26A  | 107.6    |
| C58-C57-H57B  | 109.0    | C25-N26-H26B  | 107.6    |
| H57A-C57-H57B | 107.8    | Ni1-N26-H26B  | 107.6    |
| N59-C58-C57   | 110.5(6) | H26A-N26-H26B | 107.0    |
| N59-C58-H58A  | 109.6    | C34-N35-C36   | 122.6(6) |
| C57-C58-H58A  | 109.6    | C50-N38-C39   | 109.8(7) |
| N59-C58-H58B  | 109.6    | C50-N38-C37   | 114.1(8) |
| C57-C58-H58B  | 109.6    | C39-N38-C37   | 111.7(6) |
| H58A-C58-H58B | 108.1    | C42-N41-C40   | 127.7(7) |
| N53-C60-C61   | 110.4(7) | C51-N52-Ni2   | 117.2(7) |
| N53-C60-H60A  | 109.6    | C51-N52-H52A  | 108.0    |
| C61-C60-H60A  | 109.6    | Ni2-N52-H52A  | 108.0    |
| N53-C60-H60B  | 109.6    | C51-N52-H52B  | 108.0    |
| C61-C60-H60B  | 109.6    | Ni2-N52-H52B  | 108.0    |
| H60A-C60-H60B | 108.1    | H52A-N52-H52B | 107.2    |
| N62-C61-C60   | 110.4(6) | C54-N53-C57   | 112.1(6) |
| N62-C61-H61A  | 109.6    | C54-N53-C60   | 111.5(6) |
| C60-C61-H61A  | 109.6    | C57-N53-C60   | 113.3(6) |
| N62-C61-H61B  | 109.6    | C54-N53-Ni1   | 104.3(4) |
| C60-C61-H61B  | 109.6    | C57-N53-Ni1   | 108.6(5) |
| H61A-C61-H61B | 108.1    | C60-N53-Ni1   | 106.4(4) |
| C65-C64-N63   | 113.4(7) | C55-N56-Ni1   | 110.3(4) |
| C65-C64-H64A  | 108.9    | C55-N56-H56A  | 109.6    |
| N63-C64-H64A  | 108.9    | Ni1-N56-H56A  | 109.6    |
| C65-C64-H64B  | 108.9    | C55-N56-H56B  | 109.6    |
| N63-C64-H64B  | 108.9    | Ni1-N56-H56B  | 109.6    |
| H64A-C64-H64B | 107.7    | H56A-N56-H56B | 108.1    |
| N66-C65-C64   | 119.8(8) | C58-N59-Ni1   | 107.1(5) |
| N66-C65-H65A  | 107.4    | C58-N59-H59A  | 110.3    |
| C64-C65-H65A  | 107.4    | Ni1-N59-H59A  | 110.3    |
| N66-C65-H65B  | 107.4    | C58-N59-H59B  | 110.3    |
| C64-C65-H65B  | 107.4    | Ni1-N59-H59B  | 110.3    |
| H65A-C65-H65B | 106.9    | H59A-N59-H59B | 108.5    |
| N63-C67-C68   | 110.7(8) | C61-N62-Ni1   | 110.8(4) |
| N63-C67-H67A  | 109.5    | C61-N62-H62A  | 109.5    |
| C68-C67-H67A  | 109.5    | Ni1-N62-H62A  | 109.5    |
| N63-C67-H67B  | 109.5    | C61-N62-H62B  | 109.5    |
| C68-C67-H67B  | 109.5    | Ni1-N62-H62B  | 109.5    |
| H67A-C67-H67B | 108.1    | H62A-N62-H62B | 108.1    |
| N69-C68-C67   | 110.2(8) | C67-N63-C70   | 112.5(7) |
| N69-C68-H68A  | 109.6    | C67-N63-C64   | 111.9(6) |
| C67-C68-H68A  | 109.6    | C70-N63-C64   | 112.3(7) |
| N69-C68-H68B  | 109.6    | C67-N63-Ni2   | 105.0(5) |
| C67-C68-H68B  | 109.6    | C70-N63-Ni2   | 105.2(4) |
| H68A-C68-H68B | 108.1    | C64-N63-Ni2   | 109.6(5) |
| N63-C70-C71   | 109.4(7) | C65-N66-Ni2   | 110.8(6) |
| N63-C70-H70A  | 109.8    | C65-N66-H66A  | 109.5    |
| C71-C70-H70A  | 109.8    | Ni2-N66-H66A  | 109.5    |
| N63-C70-H70B  | 109.8    | C65-N66-H66B  | 109.5    |
| C71-C70-H70B  | 109.8    | Ni2-N66-H66B  | 109.5    |
| H70A-C70-H70B | 108.2    | H66A-N66-H66B | 108.1    |
| N72-C71-C70   | 110.3(7) | C68-N69-Ni2   | 108.9(6) |
| N72-C71-H71A  | 109.6    | C68-N69-H69A  | 109.9    |
| C70-C71-H71A  | 109.6    | Ni2-N69-H69A  | 109.9    |
| N72-C71-H71B  | 109.6    | C68-N69-H69B  | 109.9    |

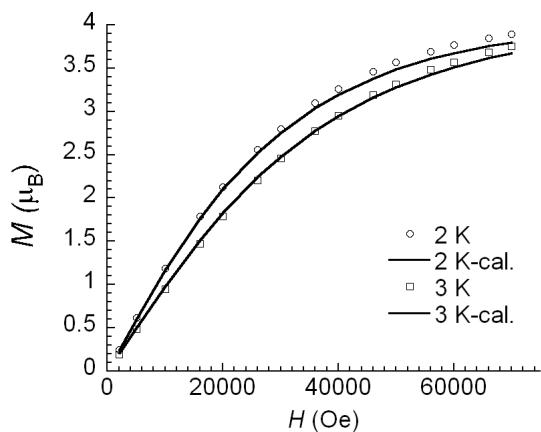
|                |           |             |            |
|----------------|-----------|-------------|------------|
| Ni2-N69-H69B   | 109.9     | N59-Ni1-N62 | 93.8(2)    |
| H69A-N69-H69B  | 108.3     | N53-Ni1-N62 | 82.2(2)    |
| C71-N72-Ni2    | 110.7(5)  | N56-Ni1-N62 | 163.1(2)   |
| C71-N72-H72A   | 109.5     | O8-Ni1-N62  | 82.7(2)    |
| Ni2-N72-H72A   | 109.5     | N52-Ni2-N72 | 101.5(3)   |
| C71-N72-H72B   | 109.5     | N52-Ni2-N63 | 175.1(3)   |
| Ni2-N72-H72B   | 109.5     | N72-Ni2-N63 | 83.0(3)    |
| H72A-N72-H72B  | 108.1     | N52-Ni2-N66 | 95.1(3)    |
| C1-O1-Lu1      | 135.1(4)  | N72-Ni2-N66 | 94.8(4)    |
| C3-O3-Lu1      | 128.5(4)  | N63-Ni2-N66 | 82.6(3)    |
| C3-O3-H3       | 115.7     | N52-Ni2-O6  | 91.2(3)    |
| Lu1-O3-H3      | 115.7     | N72-Ni2-O6  | 88.8(3)    |
| C18-O4-Lu1     | 136.3(4)  | N63-Ni2-O6  | 90.8(2)    |
| C18-O4-H4      | 111.8     | N66-Ni2-O6  | 171.9(3)   |
| Lu1-O4-H4      | 111.8     | N52-Ni2-N69 | 93.5(4)    |
| C23-O5-Lu1     | 137.0(4)  | N72-Ni2-N69 | 162.6(3)   |
| C23-O6-Ni2     | 134.5(5)  | N63-Ni2-N69 | 82.3(3)    |
| C27-O7-Lu1     | 138.1(4)  | N66-Ni2-N69 | 92.6(4)    |
| C27-O8-Ni1     | 131.7(4)  | O6-Ni2-N69  | 82.0(3)    |
| C29-O9-Lu1     | 137.5(4)  | O9-Lu1-O4   | 143.94(15) |
| C29-O9-H9      | 111.2     | O9-Lu1-O1   | 106.20(16) |
| Lu1-O9-H9      | 111.2     | O4-Lu1-O1   | 84.83(16)  |
| C44-O10-Lu1    | 129.5(4)  | O9-Lu1-O11  | 85.39(18)  |
| C44-O10-H10    | 115.3     | O4-Lu1-O11  | 106.44(18) |
| Lu1-O10-H10    | 115.3     | O1-Lu1-O11  | 143.14(17) |
| C49-O11-Lu1    | 135.4(5)  | O9-Lu1-O7   | 74.48(15)  |
| O102-N100-O101 | 126.5(11) | O4-Lu1-O7   | 77.11(16)  |
| O102-N100-O100 | 115.9(11) | O1-Lu1-O7   | 144.52(19) |
| O101-N100-O100 | 117.3(10) | O11-Lu1-O7  | 72.0(2)    |
| O201-N200-O202 | 142.5(14) | O9-Lu1-O10  | 70.90(14)  |
| O201-N200-O200 | 108.9(12) | O4-Lu1-O10  | 144.70(15) |
| O202-N200-O200 | 102.0(11) | O1-Lu1-O10  | 76.24(15)  |
| O302-N300-O300 | 117.3(11) | O11-Lu1-O10 | 74.90(18)  |
| O302-N300-O301 | 113.4(10) | O7-Lu1-O10  | 133.33(18) |
| O300-N300-O301 | 126.0(11) | O9-Lu1-O5   | 76.44(17)  |
| N301-O301-N300 | 54.4(10)  | O4-Lu1-O5   | 74.62(16)  |
| N300-O302-N301 | 57.0(10)  | O1-Lu1-O5   | 72.09(16)  |
| O303-N301-O301 | 132.6(14) | O11-Lu1-O5  | 144.44(18) |
| O303-N301-O302 | 117.5(12) | O7-Lu1-O5   | 73.8(2)    |
| O301-N301-O302 | 109.9(11) | O10-Lu1-O5  | 125.11(16) |
| N26-Ni1-N59    | 93.1(2)   | O9-Lu1-O3   | 144.95(14) |
| N26-Ni1-N53    | 176.1(2)  | O4-Lu1-O3   | 70.76(14)  |
| N59-Ni1-N53    | 84.4(2)   | O1-Lu1-O3   | 74.98(15)  |
| N26-Ni1-N56    | 100.1(2)  | O11-Lu1-O3  | 76.03(17)  |
| N59-Ni1-N56    | 92.8(2)   | O7-Lu1-O3   | 125.15(16) |
| N53-Ni1-N56    | 83.0(2)   | O10-Lu1-O3  | 75.63(14)  |
| N26-Ni1-O8     | 91.65(19) | O5-Lu1-O3   | 133.67(17) |
| N59-Ni1-O8     | 174.3(2)  |             |            |
| N53-Ni1-O8     | 90.7(2)   |             |            |
| N56-Ni1-O8     | 89.44(19) |             |            |
| N26-Ni1-N62    | 95.0(2)   |             |            |



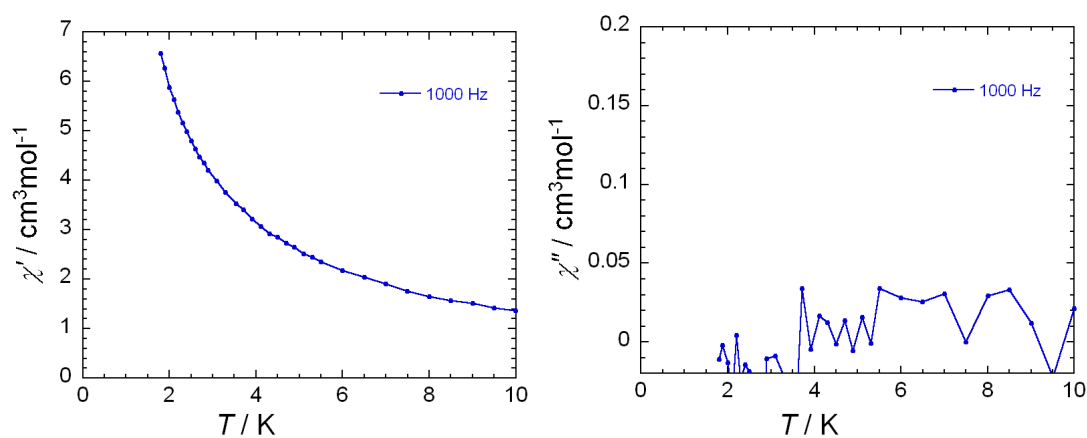
## Magnetic data



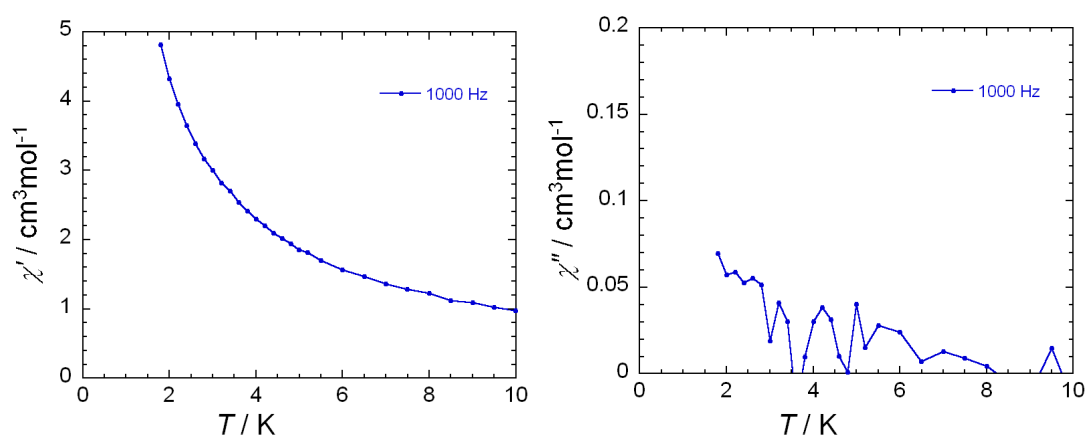
**Figure S6** Field dependence of the magnetization at low temperatures. Sample codes are indicated in the Figure.



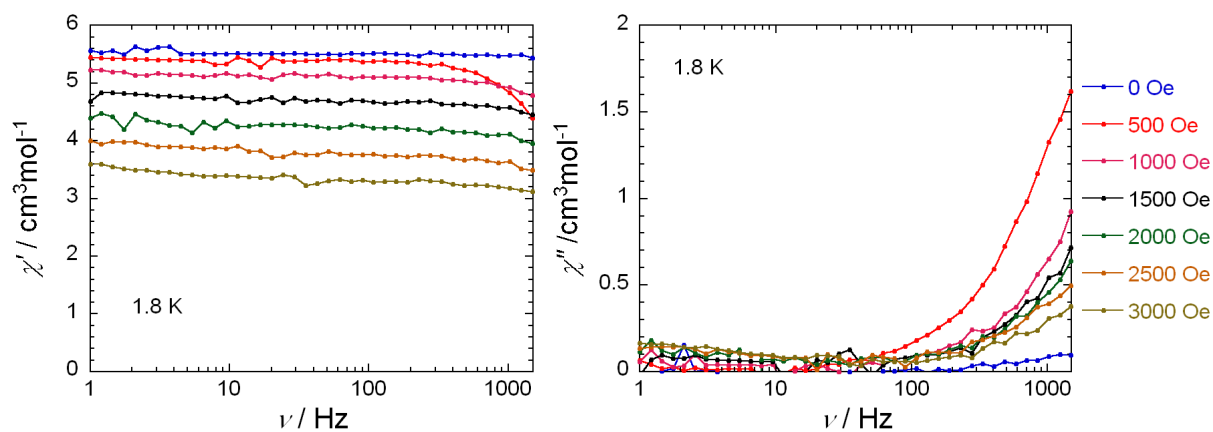
**Figure S7** The magnetization curve of compound **4** at 2 and 3 K. The solid lines correspond to the best fittings using Anisofit program. See the main text for information on the parameters.



**Figure S8** Temperature dependence of *ac* susceptibilities under zero *dc* field for compound **2**.



**Figure S9** Temperature dependence of *ac* susceptibilities under zero *dc* field for compound **3**.



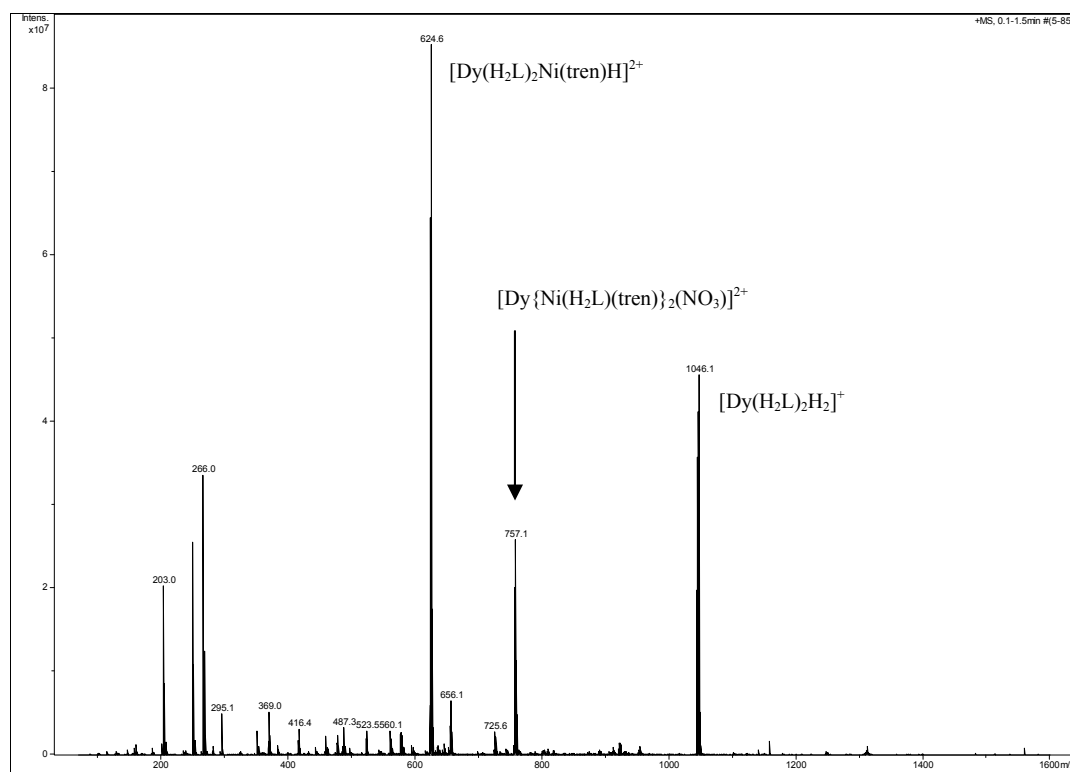
**Figure S10** Frequency dependence of *ac* susceptibilities at 1.8 K under a dc field for compound **3**.

## Electrospray Ionization Mass Spectrometry

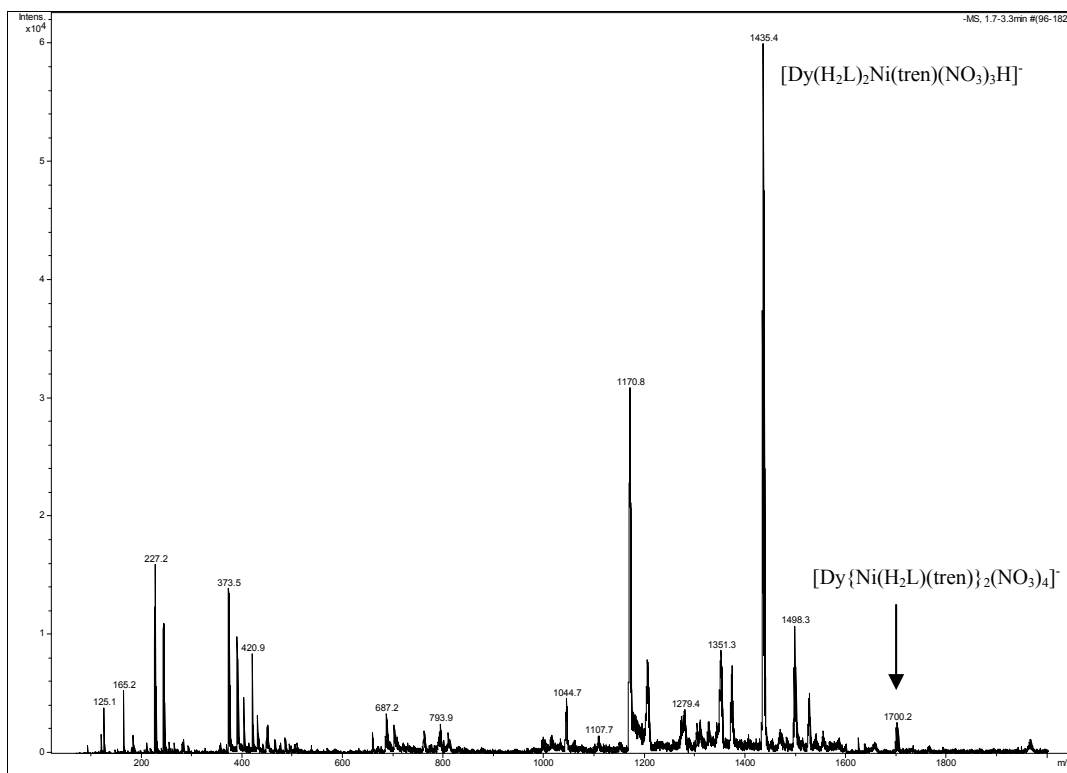
Abbreviations:

tren = C<sub>6</sub>H<sub>18</sub>N<sub>4</sub>

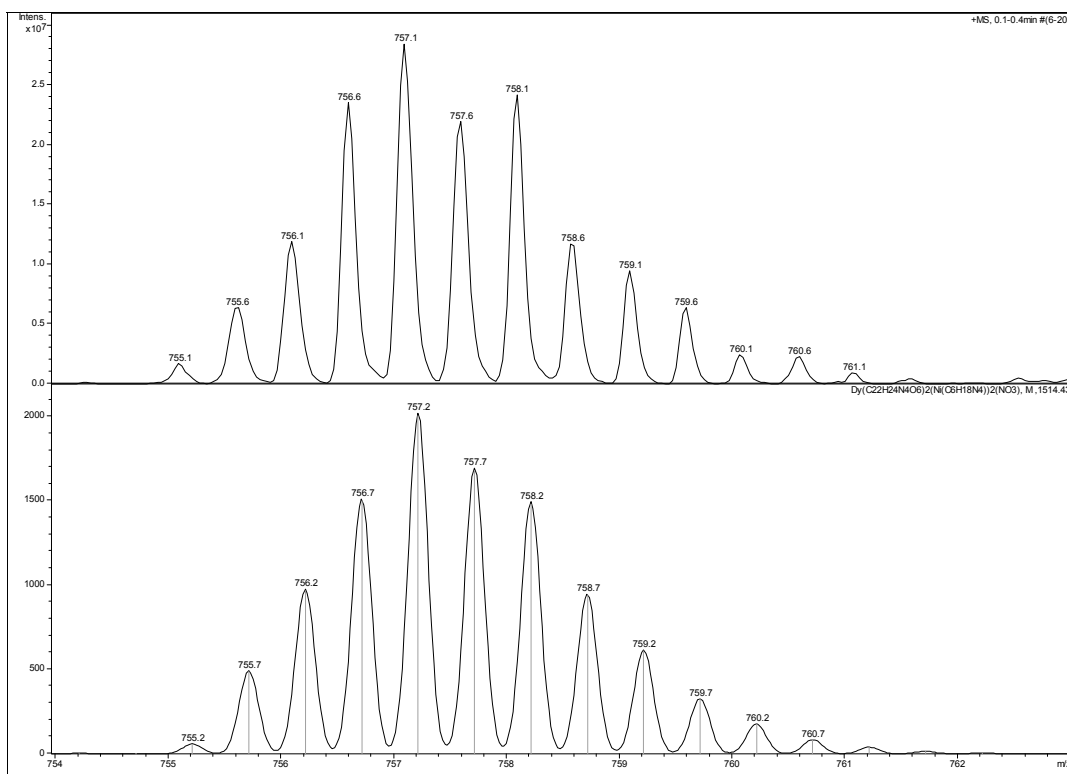
H<sub>2</sub>L = C<sub>22</sub>H<sub>24</sub>N<sub>4</sub>O<sub>6</sub>; (H<sub>2</sub>L)<sub>2</sub> = C<sub>44</sub>H<sub>48</sub>N<sub>8</sub>O<sub>12</sub>



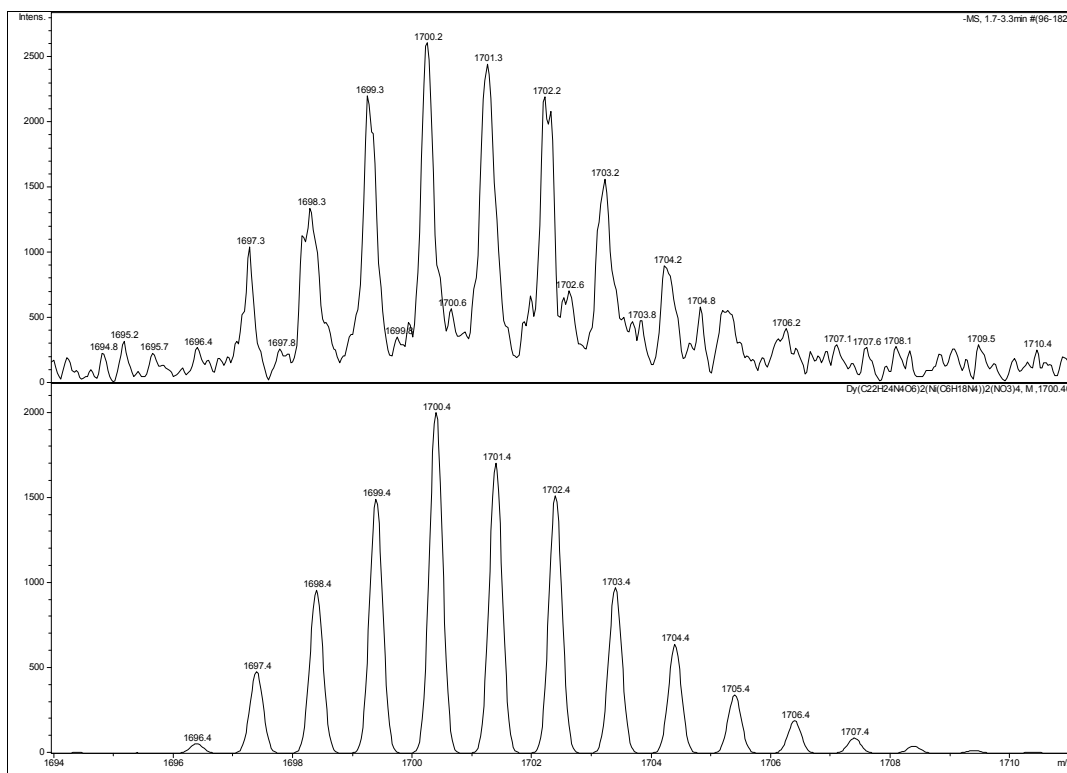
**Figure S11a** ESI-MS spectrum of a solution of compound **2** in acetonitrile detected in positive ion mode.



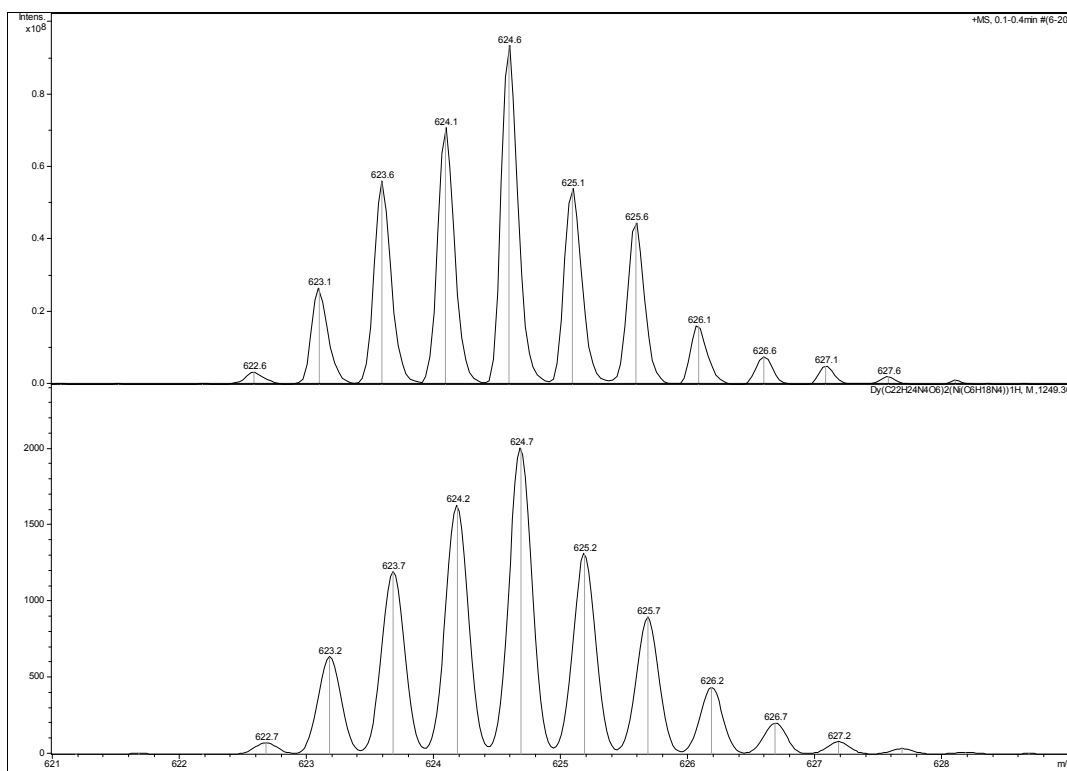
**Figure S11b** ESI-MS spectrum of a solution of compound **2** in acetonitrile detected in negative ion mode.



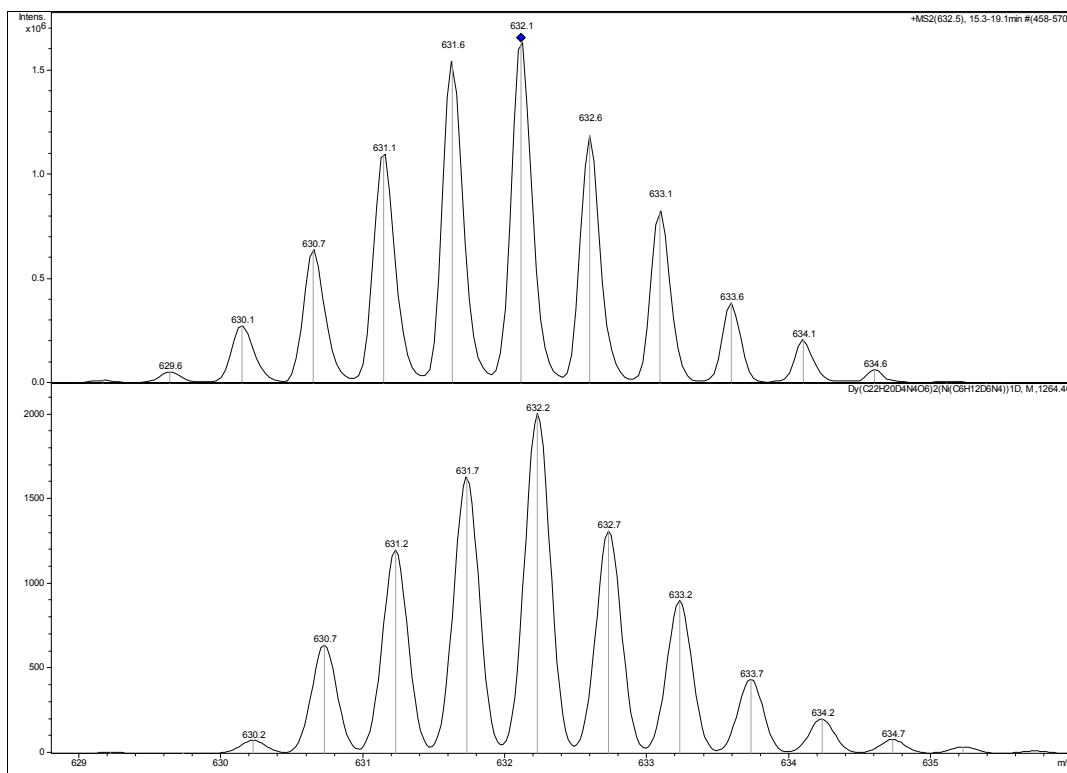
**Figure S12a** Top: experimental isotope pattern of  $[\text{Dy}\{\text{Ni}(\text{H}_2\text{L})(\text{tren})\}_2(\text{NO}_3)_2]^{2+}$ ; bottom: simulation



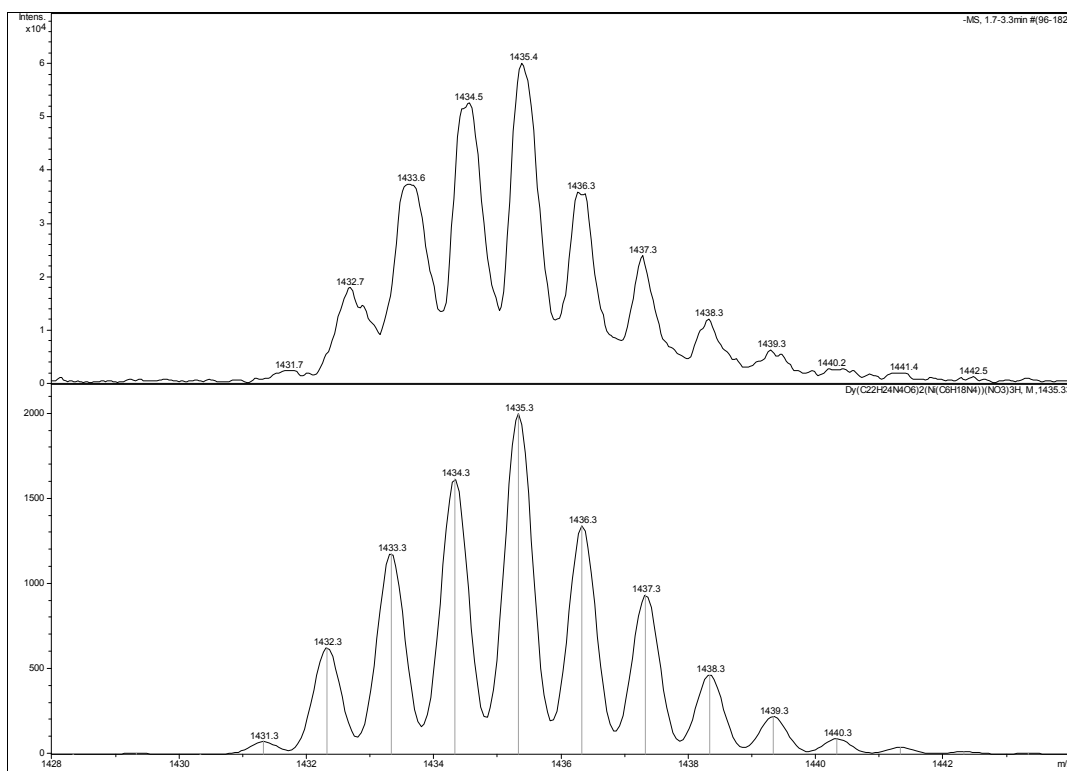
**Figure S12b** Top: experimental isotope pattern of  $[\text{Dy}\{\text{Ni}(\text{H}_2\text{L})(\text{tren})\}_2(\text{NO}_3)_4]$ ; bottom: simulation



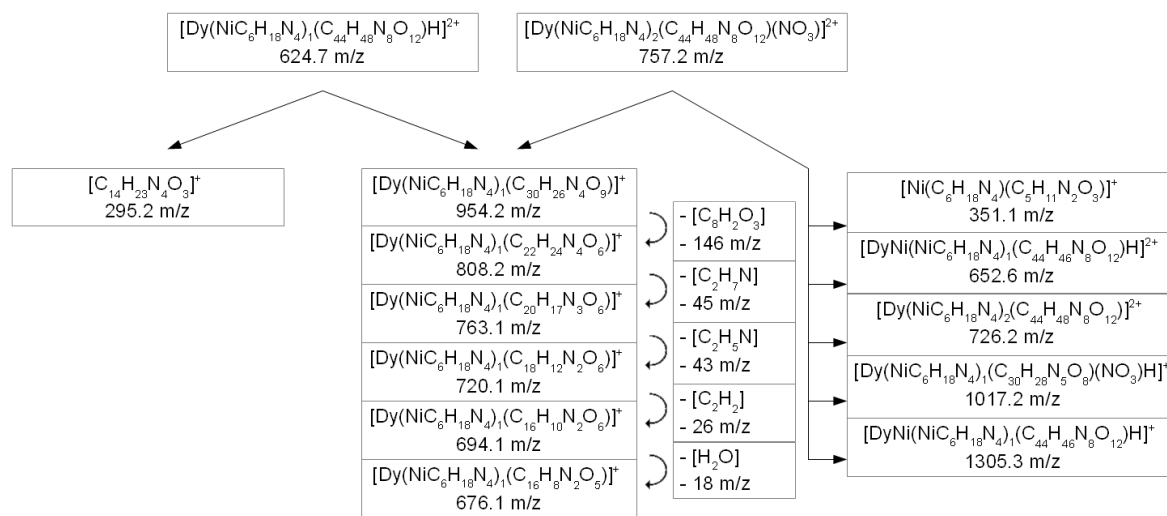
**Figure S12c** Top: experimental isotope pattern of  $[\text{Dy}(\text{H}_2\text{L})_2\text{Ni}(\text{tren})\text{H}]^{2+}$ ; bottom: simulation



**Figure S12d** Top: experimental isotope pattern of  $[\text{Dy}(\text{H}_2\text{L})_2\text{Ni}(\text{tren})\text{H}]^{2+}\text{-d}_{15}$ ; bottom: simulation



**Figure S12e** Top: experimental isotope pattern of  $[\text{Dy}(\text{H}_2\text{L})_2\text{Ni}(\text{tren})(\text{NO}_3)_3\text{H}]^-$ ; bottom: simulation



**Scheme S1** Fragmentation scheme (MS<sup>n</sup>) for the two major, doubly charged cations of compound 2:  $[\text{Dy}(\text{H}_2\text{L})_2\text{Ni}(\text{tren})\text{H}]^{2+}$  and  $[\text{Dy}\{\text{Ni}(\text{H}_2\text{L})(\text{tren})\}_2(\text{NO}_3)]^{2+}$ . Some of the neutral losses could be assigned to alternative compositions.