Supplementary Materials

A New \{\text{Dy}_3\} Single-Molecule Magnet Bearing the Schiff Base Ligand \text{N-Naphthalidene-2-amino-5-chlorophenol}

Table S1. Selected interatomic distances (Å) and angles (°) for complex \text{1:3MeOH-CHCl}_2.

<table>
<thead>
<tr>
<th></th>
<th>Dy1-O3</th>
<th>Dy3-O21</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Dy1-O4</td>
<td>2.551(4)</td>
<td>Dy3-O22</td>
<td>2.310(4)</td>
</tr>
<tr>
<td>Dy1-O7</td>
<td>2.270(4)</td>
<td>Dy3-O23</td>
<td>2.355(4)</td>
</tr>
<tr>
<td>Dy1-O8</td>
<td>2.378(4)</td>
<td>Dy3-N4</td>
<td>2.486(5)</td>
</tr>
<tr>
<td>Dy1-O24</td>
<td>2.327(4)</td>
<td>Dy4-O11</td>
<td>2.346(4)</td>
</tr>
<tr>
<td>Dy1-O25</td>
<td>2.389(5)</td>
<td>Dy4-O13</td>
<td>2.459(4)</td>
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<tr>
<td>Dy1-O26</td>
<td>2.313(4)</td>
<td>Dy4-O14</td>
<td>2.318(4)</td>
</tr>
<tr>
<td>Dy1-N1</td>
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<td>Dy4-O15</td>
<td>2.228(5)</td>
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<tr>
<td>Dy2-O2</td>
<td>2.447(5)</td>
<td>Dy4-O16</td>
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<td>Dy2-O4</td>
<td>2.434(4)</td>
<td>Dy4-O18</td>
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<td>Dy2-O6</td>
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<td>Dy5-O11</td>
<td>2.356(4)</td>
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<tr>
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<td>2.409(4)</td>
<td>Dy5-O12</td>
<td>2.415(4)</td>
</tr>
<tr>
<td>Dy3-O12</td>
<td>2.507(4)</td>
<td>Dy5-O13</td>
<td>2.300(4)</td>
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<td>Dy3-O13</td>
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<td>Dy2-O4-Dy1</td>
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<td>106.7(2)</td>
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<td>Dy5-O8-Dy1</td>
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<td>Dy1-O26-Dy2</td>
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<td>Dy4-O11-Dy5</td>
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<tr>
<td>Dy5-O12-Dy3</td>
<td>103.0(2)</td>
<td>Dy5-O26-Dy2</td>
<td>108.8(2)</td>
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<td>Dy3-O13-Dy4</td>
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<td>Dy3-O19-Dy4</td>
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<tr>
<td>Dy5-O13-Dy3</td>
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<td>Dy1-O24-Dy2</td>
<td>100.3(2)</td>
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<tr>
<td>Dy1...Dy2...Dy5</td>
<td>61.8(2)</td>
<td>Dy3...Dy4...Dy5</td>
<td>55.7(2)</td>
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<tr>
<td>Dy2...Dy5...Dy1</td>
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<td>Dy3...Dy5...Dy4</td>
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<tr>
<td>Dy5...Dy1...Dy2</td>
<td>62.4(2)</td>
<td>Dy5...Dy3...Dy4</td>
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Table S2. Continuous shape measures (CShM) of the 8-coordinate Dy(1-5) coordination polyhedra in complex 1.

<table>
<thead>
<tr>
<th>Polyhedronb</th>
<th>Dy1</th>
<th>Dy2</th>
<th>Dy3</th>
<th>Dy4</th>
<th>Dy5</th>
</tr>
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<tbody>
<tr>
<td>OP-8</td>
<td>29.89</td>
<td>31.53</td>
<td>29.46</td>
<td>30.61</td>
<td>34.32</td>
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<tr>
<td>HPY-8</td>
<td>24.36</td>
<td>21.09</td>
<td>21.77</td>
<td>24.03</td>
<td>22.68</td>
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<tr>
<td>HBPY-8</td>
<td>14.85</td>
<td>15.93</td>
<td>14.07</td>
<td>16.17</td>
<td>14.51</td>
</tr>
<tr>
<td>CU-8</td>
<td>10.59</td>
<td>13.00</td>
<td>8.06</td>
<td>12.33</td>
<td>7.79</td>
</tr>
<tr>
<td>SAPR-8</td>
<td>1.41</td>
<td>4.31</td>
<td>1.53</td>
<td>2.57</td>
<td>1.10</td>
</tr>
<tr>
<td>TDD-8</td>
<td>1.40</td>
<td>2.86</td>
<td>1.55</td>
<td>1.03</td>
<td>2.19</td>
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<tr>
<td>JGBF-8</td>
<td>13.15</td>
<td>13.48</td>
<td>14.30</td>
<td>12.30</td>
<td>15.47</td>
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<tr>
<td>JETBPY-8</td>
<td>29.10</td>
<td>26.83</td>
<td>27.13</td>
<td>28.50</td>
<td>29.11</td>
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<td>JBTBPR-8</td>
<td>1.92</td>
<td>2.22</td>
<td>3.22</td>
<td>2.47</td>
<td>3.54</td>
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<td>BTPR-8</td>
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<td>1.52</td>
<td>2.43</td>
<td>2.92</td>
<td>5.97</td>
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<td>TT-8</td>
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<td>13.70</td>
<td>8.65</td>
<td>13.09</td>
<td>8.48</td>
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<tr>
<td>ETBPY-8</td>
<td>25.19</td>
<td>22.69</td>
<td>22.14</td>
<td>24.69</td>
<td>24.27</td>
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</table>

a The values in boldface indicate the closest polyhedron according to the CShM.
b Abbreviations: OP-8, octagon; HPY-8, heptagonal pyramid; HBPY-8, hexagonal bipyramid; CU-8, cube; SAPR-8, square antiprism; TDD-8, triangular dodecahedron; JGBF-8, Johnson gyrobifastigium; JETBPY-8, Johnson elongated triangular bipyramid; JBTBPR-8, Johnson biaugmented trigonal prism; BTPR-8, biaugmented trigonal prism; JSD-8, Johnson snub diphenoid; TT-8, triakis tetrahedron; ETBPY-8, elongated trigonal bipyramid.

Table S3. Crystallographic data for complex 1-3MeOH-CH2Cl2.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>1-3MeOH-CH2Cl2</th>
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<tbody>
<tr>
<td>Empirical formula</td>
<td>C115H118N5O26Dy5Cl7</td>
</tr>
<tr>
<td>FW / g mol⁻¹</td>
<td>3046.79</td>
</tr>
<tr>
<td>Temperature / K</td>
<td>100(1)</td>
</tr>
<tr>
<td>Crystal system</td>
<td>triclinic</td>
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<tr>
<td>Space group</td>
<td>P-1</td>
</tr>
<tr>
<td>a / Å</td>
<td>14.4539(3)</td>
</tr>
<tr>
<td>b / Å</td>
<td>16.7348(3)</td>
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<tr>
<td>c / Å</td>
<td>27.4249(4)</td>
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<td>α / °</td>
<td>79.7590(10)</td>
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<td>β / °</td>
<td>79.6500(10)</td>
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<tr>
<td>γ / °</td>
<td>65.134(2)</td>
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<tr>
<td>Volume / Å³</td>
<td>5881.7(2)</td>
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<tr>
<td>Z</td>
<td>2</td>
</tr>
<tr>
<td>μ / mm⁻¹</td>
<td>3.369</td>
</tr>
<tr>
<td>F(000)</td>
<td>3000</td>
</tr>
<tr>
<td>Radiation</td>
<td>Mo Kα (λ = 0.71073)</td>
</tr>
<tr>
<td>Index ranges</td>
<td>-17 ≤ h ≤ 17</td>
</tr>
<tr>
<td></td>
<td>-19 ≤ k ≤ 19</td>
</tr>
<tr>
<td></td>
<td>-32 ≤ l ≤ 32</td>
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<tr>
<td>Reflections collected</td>
<td>66721</td>
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<tr>
<td>Independent reflections</td>
<td>17139 (Rint = 0.0610)</td>
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<tr>
<td>Goodness-of-fit on F²</td>
<td>1.043</td>
</tr>
<tr>
<td>Final R indexes [I ≥ 2σ(I)] ≫ b</td>
<td>R₁ = 0.0462</td>
</tr>
<tr>
<td></td>
<td>wR₂ = 0.1143</td>
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<tr>
<td>Final R indexes [all data]</td>
<td>R₁ = 0.0590</td>
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<tr>
<td></td>
<td>wR₂ = 0.1223</td>
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<tr>
<td>(Δρ)max/min / e Å⁻³</td>
<td>3.651 and -1.743</td>
</tr>
</tbody>
</table>

a R₁ = Σ(|Fo| - |Fc|)² / Σ|Fo|². b wR₂ = [Σ[w(Fo² - Fc²)]² / Σ[w(Fo²)]²]¹/², where p = [max(Fo², 0) + 2Fc²]/3.
Figure S1. Crystallographically established coordination modes of the ligands nacp$^2$ (top) and hpd$^-$ (bottom) present in 1.

Figure S2. Field ($H$) dependence of the relaxation time ($\tau$) of 1·3MeOH·CH$_2$Cl$_2$ measured at 2.0 K. The peak maximum corresponds to the optimum dc field.
**Figure S3.** $^1$H NMR spectrum of the ligand nacpH$_2$ in DMSO-$d_6$.

**Figure S4.** $^{13}$C NMR spectrum of the ligand nacpH$_2$ in DMSO-$d_6$. 

Chemical shift (ppm)
Figure S5. Positive ESI-MS spectra of the ligand nacpH₂ in MeCN. The inset spectra show a zoomed picture of the experimental data (top) and the theoretical simulation of the isotope model of the protonated ion of nacpH₂ (bottom).

Figure S6. Cole-Cole plots for 1:3MeOH-CH₂Cl₂ obtained using the ac susceptibility data at zero (top) and 200 Oe (bottom) applied dc fields. The solid lines are the best fit obtained from a generalized Debye model.