Supporting Information

Double-stranded metallo-triangles: from anion-templated nonanuclear to cation-templated tetraicosanuclear dysprosium clusters

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REFERENCES
Experimental Section

Synthesis

1-naphthylphosphonic acid (C_{10}H_{7}PO_{3}H_{2}) was prepared according to the methods reported in the literatures. All other reagents were purchased from commercial suppliers and as received without further purification. These two complexes were performed under aerobic conditions.

H_{2}L^{1}:

For (E)-N’-(3,5-di-tert-butyl-2-hydroxybenzylidene)pyrazine-2-carboxyhydrazide (H_{2}L^{1}): Pyrazine-2-carboxyhydrazide (10 mmol, 1.3812 g) was suspended together with 3,5-Di-tert-butyl-2-hydroxy-benzaldehyde (10 mmol, 2.3433 g) in methanol (40 ml), and the resulting mixture was stirred at the room temperature overnight. The pale yellow solid was collected by filtration (yield = 3.2031 g, 86%). Elemental analysis (%) calcd for C_{20}H_{17}N_{4}O_{2}: C, 69.55, H, 4.96, N, 16.22: found C, 68.06, H, 5.09, N, 15.65. IR (KBr, cm^{-1}): 3435(w), 3285(w), 2956(w), 1689(vs), 1651(w), 1585(m), 1524(s), 1467(w), 1437(m), 1398(w), 1357(w), 1309(m), 1269(w), 1251(m), 1236(m), 1209(m), 1157(w), 1107(m), 1052(m), 1022(s), 988(m), 963(w), 894(m), 860(m), 828(m), 770(m), 745(m), 672(m), 586(m), 446(m).

H_{4}L^{2}:

For (N^{1}E,N^{2}E)-N^{1},N^{2}-bis(2-hydroxy-3-methoxybenzylidene)oxalohydrazide (H_{4}L^{2}): Oxalyl dihydrazide (10 mmol, 1.1811 g) was suspended together with o-vanillin (20 mmol, 3.0436 g) in methanol (100 ml), and the resulting mixture was stirred at the room temperature overnight. The pale yellow solid was collected by filtration (yield = 3.7598 g, 89%). Elemental analysis (%) calcd for C_{18}H_{10}N_{4}O_{6}: C, 57.15, H, 2.66, N, 14.81: found C, 55.92, H, 2.49, N, 15.06. IR (KBr, cm^{-1}): 3333(w), 3218(w), 2998(m), 2939(m), 2840(m), 1682(vs), 1605(s), 1577(w), 1517(vs), 1468(vs), 1359(s), 1256(vs), 1186(w), 1172(w), 1081(s), 1004(m), 967(w), 831(s), 782(w), 728(s), 570(m), 551(m), 507(w), 450(m).

Dy_{9}:

For [Dy_{9}(L^{1})_{6}(O_{3}PC_{10}H_{7})_{6}((μ_{6}-Cl))(SCN)_{2}(DMF)_{6}]-4DMF-5MeCN-1H_{2}O (Dy_{9}): A
mixture of H$_2$L$^1$ (34.53 mg, 0.10 mmol), Dy(SCN)$_3$·6H$_2$O (200.72 mg, 0.45 mmol) and NaN$_3$ (6.53 mg, 0.10 mmol) in H$_2$O/MeOH/MeCN/DMF (1:1:4:2, 24 mL) was heated with triethylamine (0.20 mL, 1.50 mmol) at 100 °C. After 4 h, C$_{10}$H$_7$PO$_3$H$_2$ (41.63 mg, 0.20 mmol) and NaCl (5.84 mg, 0.10 mmol) were added slowly to the solution. The resulting clear yellow-orange solution was sealed and kept in a vacuum drying oven at 100 °C. Dark-red block-like crystals were isolated after two weeks in 29% yield. Elemental analysis (%) calcd for C$_{222}$H$_{273}$ClNa$_3$Dy$_9$O$_{44}$P$_6$S$_2$: C, 45.04; H, 4.65; N, 9.70; S, 1.08; Dy, 0.59; Cl, 0.00. Found: C, 43.59; H, 4.51; N, 9.48; S, 1.13; Dy, 25.94; Na, 0.00; Cl, 0.57. IR (KBr, cm$^{-1}$): 3423(w), 3051(m), 2954(w), 2064(w), 1660(vs), 1631(vs), 1614(vs), 1526(w), 1459(w), 1431(w), 1413(w), 1385(w), 1359(m), 1256(w), 1236(m), 1198(m), 1166(w), 1121(s), 1052(w), 1025(m), 1010(m), 963(m), 912(m), 912(m), 861(m), 840(w), 801(m), 790(w), 775(w), 751(m), 676(w), 637(m), 584(w), 520(w), 486(w), 470(m), 449(m).

**Dy$_{24}$:**

For [Dy$_{24}$Na$_3$(L$^2$)$_6$(O$_3$PC$_{10}$H$_7$)$_6$($\mu_4$-O)$_3$($\mu_3$-O)$_{18}$($\mu_2$-AcO)$_6$($\mu_2$-COO)$_3$(MeOH)$_6$ (H$_2$O)$_{18}$]-20MeOH·9H$_2$O (Dy$_{24}$): A mixture of H$_2$L$^2$ (37.83 mg, 0.10 mmol) and Dy(AcO)$_3$·6H$_2$O (71.07 mg, 0.20 mmol) in H$_2$O/MeOH/DMF (1:4:1, 24 mL) was heated with solid NaOH (4.81 mg, 0.12 mmol) at 100 °C. After 10 h, C$_{10}$H$_7$PO$_3$H$_2$ (41.63 mg, 0.20 mmol) and NaHCO$_3$ (8.41 mg, 0.10 mmol) were added slowly to the solution. The resulting clear yellow-orange solution was sealed and kept in a vacuum drying oven at 100 °C. Pale-yellow block-like crystals were isolated after 6 days in 36% yield. Elemental analysis (%) calcd for C$_{209}$H$_{473}$Dy$_{24}$N$_{24}$Na$_3$O$_{227}$P$_6$: C, 22.59; H, 4.29; N, 3.03; Dy, 35.10; Na, 0.62; Cl, 0.00. Found: C, 21.68; H, 4.43; N, 2.91; Dy, 35.64; Na, 0.59; Cl, 0.00. IR (KBr, cm$^{-1}$): 3422(w), 2934(m), 1663(vs), 1601(vs), 1469(vs), 1363(s), 1237(w), 1214(s), 1168(m), 1082(s), 1010(w), 967(w), 869(w), 829(m), 801(m), 780(w), 743(s), 664(w), 649(m), 636(m), 623(m), 564(w), 546(m), 518(m), 501(m).

**Analytical Procedures**

General methods: Elemental analyses were carried out in a PE$_{240}$C elemental
analyzer. The infrared spectra were recorded as KBr pellets on a Bruker Tensor 27 spectrometer in the range of 400-4000 cm\(^{-1}\). Thermal analyses were performed on a METTLER TOLEDO TGA/DSC 1 instrument in the range of 30-800 °C with Al\(_2\)O\(_3\) pan at a heating rate of 5 °C/min. Powder X-ray diffraction data were collected using a Bruker D8 ADVANCE PXRD equipped with a CuK\(_\alpha\) X-ray source over the 20 range of 5 to 50° at room temperature. Energy-dispersive X-ray spectra were recorded with the help of a HRTEM, Thermo Fischer, Talos F200x. The magnetic susceptibility data were obtained on a Quantum Design MPMS3 SQUID system. The direct current measurements were obtained with an external magnetic field of 1.0 kOe in the temperature range 1.8-300 K, and the alternating-current measurements were executed in a 3.0 Oe ac oscillating field at different frequencies from 1 to 1000 Hz. The diamagnetic contribution of the sample itself was estimated from Pascal’s constant.\(^{52}\)

Crystal structure determination and refinement: All crystals were taken from the mother liquid without further treatment, transferred to oil and mounted into a loop for single crystal X-ray data collection. Diffraction intensity data of Dy\(_9\) was collected on a ROD, XtaLAB Synergy Custom DW system, HyPix diffractometer using Cu K\(_\alpha\) (\(\lambda = 1.54184\ \text{Å}\)) at 100 K, whereas that of Dy\(_{24}\) on a multiwire proportional diffractometer using graphite-monochromated Mo K\(_\alpha\) (\(\lambda = 0.71073\ \text{Å}\)) at 100 K. The diffraction images were processed using the CrystAlis\(^{pro}\) software suite.\(^{53}\) These structures were solved using the charging-flipping algorithm, as implemented in the program SUPERFLIP\(^{54}\) and refined by full-matrix least-squares techniques against \(F_o^2\) using the SHELXL program\(^{55}\) through the OLEX2 interface.\(^{56}\) Hydrogen atoms were placed in calculated positions and refined isotropically by using a riding model. All structures were examined using the Addsym subroutine of PLATON\(^{57}\) to ensure that no additional symmetry could be applied to the models. Nevertheless, in this heavy-atom structure as it was not possible to see clear electron-density peaks in difference maps which would correspond with acceptable locations for the various H atoms bonded to water oxygen atoms, the refinement was completed with no
allowance for these H atoms in the model.

There are some solvent-accessible void volumes in the crystals of Dy$_9$ and Dy$_{24}$, which are occupied by highly disordered water and manthanol molecules. No satisfactory disorder model could be achieved despite numerous attempts, and therefore the SQUEEZE program$^{58}$ implemented in PLATON was further employed to remove these electron densities. For Dy$_9$, the SQUEEZE function of PLATON reveals a residual electron density of 288 electrons/cell in cell-remaining voids where the residual electron density was tentatively assigned to 4 dimethylformamide molecules, 5 acetonitrile molecules and 1 water molecules [288 = 4 (DMF) × 40e + 22 (MeCN) × 22e + 1 (H$_2$O) × 10e]. For Dy$_{24}$, the SQUEEZE function of PLATON reveals a residual electron density of 1260 electrons/cell in cell-remaining voids where the residual electron density was tentatively assigned to 20 methanol molecules and 90 water molecules [1260 = 20 (MeOH) × 18e + 90 (H$_2$O) × 10e]. The amount of lattice solvent molecules was proved by TG analysis (please see Figure S2). The reported sum formula has been corrected by taking into account the lattice methanol and water molecules. Pertinent crystallographic data collection and refinement parameters are collated in Table S1.

CCDC 2253521 (Dy$_9$) and CCDC 2253522 (Dy$_{24}$) contain the supplementary crystallographic data for this paper. These data can be obtained free of charge from the Cambridge Crystallographic Data Centre via www.ccdc.cam.ac.uk/data_request/cif.
Structure information

Figure S1. Infrared spectra of Dy$_9$ (a) and Dy$_{24}$ (b).

Figure S2. Thermal analysis of Dy$_9$ (a) and Dy$_{24}$ (b).

Figure S3. The powder XRD patterns for Dy$_9$ (a). The black circles are for the observed data (b). The red solid line is for the calculated data. The grey solid curve is for the difference. The vertical bars are the positions of Bragg peaks. Cell parameters: $R$-3c, $a = 23.84$ Å, $b = 23.84$ Å, $c = 78.24$ Å, $\alpha = 90.0^\circ$, $\beta = 90.0^\circ$, $\gamma = 120.0^\circ$, $V = 39016.1$ Å$^3$ ($w$R$\rho$ = 0.109).
Figure S4. The powder XRD patterns for Dy24 (a). The black circles are for the observed data (b). The red solid line is for the calculated data. The grey solid curve is for the difference. The vertical bars are the positions of Bragg peaks. Cell parameters: R-3c, a = 40.01 Å, b = 40.01 Å, c = 41.95 Å, α = 90.0°, β = 90.0°, γ = 120.0°, V = 59122.9 Å³ (wRp = 0.116).

Figure S5. Ball-and-stick views of Dy9. View along the crystallographic c axis (a). The [Dy6(μ6-Cl)(μ3-N3)2]^{15+} core (b). View along the crystallographic a axis (c). The hydrogen atoms are omitted for clarity.

Figure S6. Structure Dy9 highlighting the coordination modes of (L²)⁺ (a) and C_{10}H_{7}PO_{3}²⁻ (b). Coordination polyhedra observed for the metal centers (c).
Figure S7. Space-filling representation of Dy9 (a). The molecular packing viewed along the c axis (b).

Figure S8. Residual density map of Dy9.

Figure S9. Ball-and-stick views of Dy24. View along the crystallographic c axis (a). The [Dy4Na(O3PC10H7)2]9+ core (b). View along the crystallographic a axis (c). Hydrogen atoms are omitted for clarity.
**Figure S10.** Structure Dy$_{24}$ highlighting the coordination modes of (L$^2$)$_4^+$ (a) and C$_{10}$H$_7$PO$_5^-$ (b). Coordination polyhedra observed for the metal centers (c).

**Figure S11.** Space-filling representation of Dy$_{24}$ (a). The molecular packing viewed along the c axis (b).

**Figure S12.** Residual density map of Dy$_{24}$. 
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Symmetry codes: a: 1 - y, x - y, z; b: 1- x + y, 1- x, z; c: 1/3 + y, -1/3 + x, 7/6 - z; d: 4/3 - x, 2/3 -x + y, 7/6 - z; e: 1/3 + x - y, 2/3 - y, 7/6 - z.
Table S3. Selected bond lengths (Å) and angles (º) for Dy$_{24}$.

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<tr>
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<tr>
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<td>Dy2-O13</td>
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<td>Dy3-O5</td>
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<tr>
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<td>P1-O15</td>
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<td>P1-O16</td>
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<tr>
<td>P1-C19</td>
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<td></td>
<td>Na1-O5</td>
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<td>Na1-O6</td>
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<tr>
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<td></td>
<td>Na1-O5d</td>
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<td>Na1-O6d</td>
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<td></td>
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<td>Dy2-O12-Dy3</td>
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</tr>
</tbody>
</table>

Symmetry codes: a: 1 - y, x - y, z; b: 1 - x + y, 1 - x, z; c: 1/3 + y, -1/3 + x, 7/6 - z; d: 4/3 - x, 2/3 - x + y, 7/6 - z; e: 1/3 + x - y, 2/3 - y, 7/6 - z.
Table S4. Dy$^{III}$ geometry analysis of Dy$_9$ and Dy$_{24}$ by SHAPE 2.1 software.

| Geometry (CN = 8) | Dy$_1$ | Dy$_2$ | Dy$_3$ | Geometry (CN = 7) | Dy$_4$ | Geometry (CN = 6) | Na$_1$
|------------------|--------|--------|--------|------------------|--------|-------------------|--------
| OP-S             | 30.561 |        |        | EP-9             | 29.982 |                   |        |
| HPY-S            | 25.443 |        |        | OPY-9            | 21.513 |                   |        |
| HBPLY-S          | 14.585 |        |        | HBPLY-9          | 16.156 |                   |        |
| CU-S             | 12.425 |        |        | JTC-9            | 14.342 |                   |        |
| SAPR-S           | 4.273  |        |        | JCCU-9           | 9.380  |                   |        |
| TDD-S            | 2.224  |        |        | CCU-9            | 7.410  |                   |        |
| JGBF-S           | 9.582  |        |        | JCSAPR-9         | 7.180  |                   |        |
| JETBPY-S         | 24.551 |        |        | CSAPR-9          | 5.570  |                   |        |
| JBTPR-S          | 3.098  |        |        | JCTPR-9          | 7.750  |                   |        |
| BTPR-S           | 2.801  |        |        | TCTPR-9          | 6.020  |                   |        |
| JSD-S            | 1.788  |        |        | JTDIC-9          | 12.828 |                   |        |
| TT-S             | 13.240 |        |        | HH-9             | 6.001  |                   |        |
| ETBPY-S          | 21.118 |        |        | MFF-9            | 4.354  |                   |        |

| Geometry (CN = 8) | Dy$_1$ | Dy$_2$ | Dy$_3$ | Geometry (CN = 7) | Dy$_4$ | Geometry (CN = 6) | Na$_1$
|------------------|--------|--------|--------|------------------|--------|-------------------|--------
| OP-S             | 29.383 | 32.690 | 29.338 | HP-7             | 32.606 | HP-6              | 21.560 |
| HPY-S            | 23.995 | 20.301 | 23.707 | HPY-7            | 22.612 | PBY-6             | 10.093 |
| HBPLY-S          | 13.331 | 16.426 | 15.562 | PBPLY-7          | 1.216  | OC-6              | 13.627 |
| CU-S             | 10.370 | 14.492 | 12.295 | COC-7            | 5.498  | TPR-6             | 9.769  |
| SAPR-S           | 2.274  | 5.182  | 2.783  | CTRP-7           | 4.239  | JPR-6             | 12.552 |
| TDD-S            | 1.602  | 3.370  | 1.329  | JPBPY-7          | 4.073  |                   |        |
| JGBF-S           | 11.703 | 12.986 | 12.979 | JETPY-7          | 22.802 |                   |        |
| JETBPY-S         | 27.708 | 26.397 | 27.556 |                   |        |                   |        |
| JBTPR-S          | 2.618  | 3.666  | 2.558  |                   |        |                   |        |
| BTPR-S           | 1.812  | 2.128  | 1.920  |                   |        |                   |        |
| JSD-S            | 2.728  | 5.158  | 3.367  |                   |        |                   |        |
| TT-S             | 10.852 | 14.998 | 13.040 |                   |        |                   |        |
| ETBPY-S          | 24.757 | 21.008 | 23.780 |                   |        |                   |        |
Magnetic properties of Dy$_9$ and Dy$_{24}$

Figure S13. Temperature dependent of the $\chi_M T$ values at 1.0 kOe for compounds Dy$_9$ (a) and Dy$_{24}$ (b). Inset: the magnetization vs. field plots at different temperature.

Figure S14. Temperature dependence of $\chi_M'$ and $\chi_M''$ in zero dc field for Dy$_9$.

Figure S15. Frequency and field dependence of $\chi_M'$ (a) and $\chi_M''$ (b) at 2.0 K for Dy$_9$. The solid lines are the best fits to the experimental data, obtained with the generalized Debye model.
Figure S16. Cole-Cole plots at 2.0 K between 0 and 3.0 kOe for Dy$_9$ (a). The solid lines are the best fits to the experimental data, obtained with the generalized Debye model. Magnetization relaxation time, $\tau$ versus $H$ plots (b).

Figure S17. Temperature dependence of the $\chi'_M$ $T$ (a) and $\chi'_M$ (b) in a 800 Oe dc field for Dy$_9$.

Figure S18. Temperature dependence of $\chi''_M$ (a) and frequency dependence of $\chi'_M$ (b) in a 800 Oe dc field for Dy$_9$. 
**Figure S19.** Cole-Cole plots for compound Dy9 collected under a 800 Oe dc magnetic field. Data were collected at the temperature of 1.8 K and 8.0 K. The solid lines represent fits to the data using the generalized Debye model.

**Figure S20.** Temperature dependence of the $\chi_M T$ (a) and $\chi_M'$ (b) in zero dc field for Dy24.

**Figure S21.** Temperature dependence of $\chi_M''$ (a) and frequency dependence of $\chi_M'$ (b) in a 800 Oe dc field for Dy24.
Figure S22. Cole-Cole plots for compound Dy24 collected under zero applied dc magnetic field. Data were collected at the temperature of 1.8 K and 8.0 K. The solid lines represent fits to the data using the generalized Debye model.

Figure S23. Frequency and field dependence of $\chi_M'$ (a) and $\chi_M''$ (b) at 2.0 K for Dy24. The solid lines are the best fits to the experimental data, obtained with the generalized Debye model.

Figure S24. Cole-Cole plots at 2.0 K between 0 and 3.0 kOe for Dy24 (a). The solid lines are the best fits to the experiments with the generalized Debye model. Magnetization relaxation time, $\tau$ versus $H$ plots (b).
**Table S5.** Magnetization relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data for compound Dy$_9$ under the different applied dc field.

<table>
<thead>
<tr>
<th>$H$ (Oe)</th>
<th>$\chi_T$</th>
<th>$\chi_S$</th>
<th>$\alpha$</th>
<th>$\tau/s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>12.842(1)</td>
<td>12.693(1)</td>
<td>0.142(5)</td>
<td>3.306(5)$\times 10^{-4}$</td>
</tr>
<tr>
<td>400</td>
<td>12.504(6)</td>
<td>11.197(8)</td>
<td>0.189(7)</td>
<td>3.658(1)$\times 10^{-4}$</td>
</tr>
<tr>
<td>600</td>
<td>12.083(1)</td>
<td>9.203(1)</td>
<td>0.199(3)</td>
<td>4.088(1)$\times 10^{-4}$</td>
</tr>
<tr>
<td>800</td>
<td>11.543(1)</td>
<td>7.754(4)</td>
<td>0.236(7)</td>
<td>4.197(3)$\times 10^{-4}$</td>
</tr>
<tr>
<td>1000</td>
<td>11.006(2)</td>
<td>6.585(7)</td>
<td>0.284(2)</td>
<td>4.079(8)$\times 10^{-4}$</td>
</tr>
<tr>
<td>1200</td>
<td>10.358(5)</td>
<td>5.951(6)</td>
<td>0.285(1)</td>
<td>3.936(4)$\times 10^{-4}$</td>
</tr>
<tr>
<td>1400</td>
<td>9.757(1)</td>
<td>5.236(4)</td>
<td>0.295(9)</td>
<td>3.832(9)$\times 10^{-4}$</td>
</tr>
<tr>
<td>1600</td>
<td>9.129(5)</td>
<td>4.802(7)</td>
<td>0.308(3)</td>
<td>3.693(4)$\times 10^{-4}$</td>
</tr>
<tr>
<td>1800</td>
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<td>4.310(6)</td>
<td>0.327(6)</td>
<td>3.574(3)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2000</td>
<td>7.942(1)</td>
<td>4.200(8)</td>
<td>0.332(5)</td>
<td>3.469(6)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>7.333(6)</td>
<td>4.104(8)</td>
<td>0.347(7)</td>
<td>3.365(2)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2400</td>
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<td>3.988(8)</td>
<td>0.381(3)</td>
<td>3.291(6)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>3.270(2)</td>
<td>0.383(2)</td>
<td>3.204(1)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>3.261(8)</td>
<td>0.418(4)</td>
<td>3.135(9)$\times 10^{-4}$</td>
</tr>
<tr>
<td>3000</td>
<td>5.387(6)</td>
<td>3.752(8)</td>
<td>0.445(6)</td>
<td>3.032(8)$\times 10^{-4}$</td>
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</table>

**Table S6.** Magnetization relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data for compound Dy$_9$ under the optimal 800 Oe dc field.

<table>
<thead>
<tr>
<th>$T$ (K)</th>
<th>$\chi_T$</th>
<th>$\chi_S$</th>
<th>$\alpha$</th>
<th>$\tau/s$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.8</td>
<td>30.052(6)</td>
<td>9.955(3)</td>
<td>0.326(2)</td>
<td>1.828(7)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2.0</td>
<td>20.879(9)</td>
<td>10.988(5)</td>
<td>0.328(5)</td>
<td>1.735(4)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2.3</td>
<td>18.011(4)</td>
<td>11.120(1)</td>
<td>0.362(1)</td>
<td>1.632(6)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2.6</td>
<td>17.044(6)</td>
<td>12.130(3)</td>
<td>0.367(7)</td>
<td>1.564(4)$\times 10^{-4}$</td>
</tr>
<tr>
<td>2.9</td>
<td>15.964(7)</td>
<td>13.250(2)</td>
<td>0.371(3)</td>
<td>1.494(8)$\times 10^{-4}$</td>
</tr>
<tr>
<td>3.2</td>
<td>15.708(1)</td>
<td>13.241(4)</td>
<td>0.377(1)</td>
<td>1.429(9)$\times 10^{-4}$</td>
</tr>
<tr>
<td>3.5</td>
<td>15.236(4)</td>
<td>13.187(5)</td>
<td>0.382(6)</td>
<td>1.401(6)$\times 10^{-4}$</td>
</tr>
<tr>
<td>3.7</td>
<td>15.105(6)</td>
<td>13.082(2)</td>
<td>0.390(1)</td>
<td>1.400(7)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>12.979(3)</td>
<td>0.427(9)</td>
<td>1.367(3)$\times 10^{-4}$</td>
</tr>
<tr>
<td>4.5</td>
<td>14.487(4)</td>
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<td>0.443(5)</td>
<td>1.343(1)$\times 10^{-4}$</td>
</tr>
<tr>
<td>5.0</td>
<td>14.250(5)</td>
<td>12.749(4)</td>
<td>0.476(7)</td>
<td>1.303(4)$\times 10^{-4}$</td>
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<tr>
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<td>0.486(8)</td>
<td>1.276(1)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>14.020(6)</td>
<td>12.183(6)</td>
<td>0.495(2)</td>
<td>1.250(2)$\times 10^{-4}$</td>
</tr>
<tr>
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<td>13.917(7)</td>
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<td>0.543(9)</td>
<td>1.225(1)$\times 10^{-4}$</td>
</tr>
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<td>11.763(5)</td>
<td>0.559(6)</td>
<td>1.201(9)$\times 10^{-4}$</td>
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<td>11.291(8)</td>
<td>0.603(5)</td>
<td>1.183(1)$\times 10^{-4}$</td>
</tr>
<tr>
<td>8.0</td>
<td>13.749(7)</td>
<td>11.035(2)</td>
<td>0.622(4)</td>
<td>1.165(8)$\times 10^{-4}$</td>
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</table>
Table S7. Results obtained from the fitting of the frequency-dependent ac susceptibility for compound Dy\textsubscript{9} under the optimal 800 Oe dc field.

<table>
<thead>
<tr>
<th>Magnetic relaxation pathway</th>
<th>C (s\textsuperscript{-1}K\textsuperscript{-n})</th>
<th>n</th>
<th>A (s\textsuperscript{-1}K\textsuperscript{-1})</th>
</tr>
</thead>
<tbody>
<tr>
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<td>0.06</td>
<td>2.83</td>
<td>-</td>
</tr>
<tr>
<td>Direct process</td>
<td>-</td>
<td>-</td>
<td>6.48 \times 10\textsuperscript{-3}</td>
</tr>
<tr>
<td>Raman and direct processes</td>
<td>0.13</td>
<td>4.69</td>
<td>1.54 \times 10\textsuperscript{-4}</td>
</tr>
</tbody>
</table>

Table S8. Magnetization relaxation fitting parameters from least-squares fitting of \(\chi(\omega)\) data for compound Dy\textsubscript{24} under the different applied dc field.

<table>
<thead>
<tr>
<th>H (Oe)</th>
<th>(\chi_T)</th>
<th>(\chi_S)</th>
<th>(\alpha)</th>
<th>(\tau/\text{s})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>11.202(4)</td>
<td>6.754(1)</td>
<td>0.273(7)</td>
<td>4.909(1)E-5</td>
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<tr>
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<td>9.647(7)</td>
<td>5.833(1)</td>
<td>0.281(4)</td>
<td>4.083(9)E-5</td>
</tr>
<tr>
<td>400</td>
<td>8.479(6)</td>
<td>4.743(8)</td>
<td>0.293(2)</td>
<td>3.017(3)E-5</td>
</tr>
<tr>
<td>600</td>
<td>7.556(5)</td>
<td>3.300(1)</td>
<td>0.302(6)</td>
<td>2.437(3)E-5</td>
</tr>
<tr>
<td>800</td>
<td>6.812(6)</td>
<td>2.610(1)</td>
<td>0.303(5)</td>
<td>2.076(1)E-5</td>
</tr>
<tr>
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<td>0.310(1)</td>
<td>1.794(8)E-5</td>
</tr>
<tr>
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<td>0.337(1)</td>
<td>1.333(6)E-5</td>
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<tr>
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<td>1.727(1)</td>
<td>0.343(2)</td>
<td>1.232(8)E-5</td>
</tr>
<tr>
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<td>0.344(3)</td>
<td>1.141(1)E-5</td>
</tr>
<tr>
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<td>4.369(7)</td>
<td>1.390(1)</td>
<td>0.348(2)</td>
<td>1.006(9)E-5</td>
</tr>
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<td>2000</td>
<td>3.922(7)</td>
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<td>2200</td>
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<td>0.355(4)</td>
<td>9.886(2)E-6</td>
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<td>2400</td>
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<td>0.800(4)</td>
<td>0.367(3)</td>
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<td>0.771(7)</td>
<td>0.390(2)</td>
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<td>0.276(7)</td>
<td>0.406(1)</td>
<td>9.511(4)E-6</td>
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<td>3000</td>
<td>2.607(9)</td>
<td>0.267(8)</td>
<td>0.415(3)</td>
<td>9.437(1)E-6</td>
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### Table S9. Magnetization relaxation fitting parameters from least-squares fitting of $\chi(\omega)$ data for compound Dy$_{24}$ under zero dc field.

<table>
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<tr>
<th>$T$ (K)</th>
<th>$\chi_T$</th>
<th>$\chi_S$</th>
<th>$\alpha$</th>
<th>$\tau / s$</th>
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<tr>
<td>1.8</td>
<td>11.963(6)</td>
<td>6.532(1)</td>
<td>0.396(2)</td>
<td>2.672(1)E-5</td>
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<tr>
<td>2.0</td>
<td>10.310(1)</td>
<td>6.342(6)</td>
<td>0.419(1)</td>
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<tr>
<td>2.3</td>
<td>9.076(9)</td>
<td>5.466(5)</td>
<td>0.454(1)</td>
<td>1.505(2)E-5</td>
</tr>
<tr>
<td>2.6</td>
<td>8.092(5)</td>
<td>4.532(3)</td>
<td>0.443(6)</td>
<td>1.123(7)E-5</td>
</tr>
<tr>
<td>2.9</td>
<td>7.308(1)</td>
<td>2.966(8)</td>
<td>0.469(4)</td>
<td>8.718(5)E-6</td>
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<tr>
<td>3.2</td>
<td>6.670(1)</td>
<td>2.281(8)</td>
<td>0.534(6)</td>
<td>7.359(7)E-6</td>
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<td>3.5</td>
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<td>0.518(8)</td>
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<td>0.538(9)</td>
<td>5.209(4)E-6</td>
</tr>
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<tr>
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<td>1.314(8)</td>
<td>0.686(9)</td>
<td>2.400(4)E-6</td>
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<tr>
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<td>0.683(8)</td>
<td>1.968(6)E-6</td>
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### Table S10. Results obtained from the fitting of the frequency-dependent $ac$ susceptibility for compound Dy$_{24}$ under zero applied dc field.

<table>
<thead>
<tr>
<th>Magnetic relaxation pathway</th>
<th>$C$ (s$^4$K$^{-a}$)</th>
<th>$n$</th>
<th>$\tau_{tunnel}$ (s)</th>
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<tr>
<td>Quantum tunneling process</td>
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<td>-</td>
<td>0.006</td>
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<tr>
<td>Raman and quantum tunneling processes</td>
<td>1.45</td>
<td>6.18</td>
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REFERENCES


