One crystal of 1-Y was measured on an APEXII, Bruker-AXS diffractometer at Mo-Ka radiation ( $\lambda=0.71073 \AA$, graphite monochromator). The structure was solved by dualspace algorithm using the SHELXT program [1], and then refined with full-matrix leastsquares methods based on $F^{2}$ (SHELXL) [2]. The contribution of the disordered solvents to the calculated structure factors was estimated following the BYPASS algorithm [3], implemented as the SQUEEZE option in PLATON [4]. A new data set, free of solvent contribution, was then used in the final refinement. All non-hydrogen atoms were refined with anisotropic atomic displacement parameters. H atoms were finally included in their calculated positions and treated as riding on their parent atom with constrained thermal parameters.

One crystal of 1-Dy was measured on a D8 VENTURE Bruker AXS diffractometer at Mo$K \alpha$ radiation ( $\lambda=0.71073 \AA$ A, multilayer monochromator). The structure was solved by dual-space algorithm using the SHELXT program [1], and then refined with full-matrix least-squares methods based on $F^{2}$ (SHELXL) [2]. The contribution of the disordered solvents to the calculated structure factors was estimated following the BYPASS algorithm [3], implemented as the SQUEEZE option in PLATON [4]. A new data set, free of solvent contribution, was then used in the final refinement. All non-hydrogen atoms were refined with anisotropic atomic displacement parameters. Except water molecules hydrogen atoms that were introduced in the structural model through Fourier difference maps analysis, H atoms were finally included in their calculated positions and treated as riding on their parent atom with constrained thermal parameters.

| [1] | G. | Sheldrick, | Acta | Cryst. | A71 | (2015) | 3-8 |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| [2] |  | G.M., | Acta | Cryst. | C71 | (2015) | 3-8 |
| [3] | v.d | nd A.L. | Spek, | Cryst. | (1990) | A46, | 94-201 |

[4] A. L. Spek, J. Appl. Cryst. (2003), 36, 7-13

|  | 1-Dy | 1-Y |
| :---: | :---: | :---: |
| $\begin{gathered} \text { Formula } \\ - \\ \text { FW }\left(\text { g.mol }{ }^{-1}\right) \\ \text { Cryst. Syst. } \\ \text { Space group } \end{gathered}$ | $\begin{gathered} \mathrm{C}_{34} \mathrm{H}_{35} \mathrm{Dy} \mathrm{~F}_{6} \mathrm{~N}_{5} \mathrm{O}_{11} \mathrm{~S}_{2}, \mathrm{C} \\ \mathrm{~F}_{3} \mathrm{O}_{3} \mathrm{~S}, \mathrm{CH}_{2} \mathrm{Cl}_{2} \\ 1264.28 \\ \text { Trigonal } \\ \text { 'R -3' } \end{gathered}$ | $\begin{gathered} \mathrm{C}_{34} \mathrm{H}_{35} \mathrm{Y} \mathrm{~F}_{6} \mathrm{~N}_{5} \mathrm{O}_{11} \mathrm{~S}_{2}, \\ \mathrm{CH}_{2} \mathrm{Cl}_{2} \\ 1041.63 \\ \text { Trigonal } \\ \text { 'R -3' } \end{gathered}$ |
| $\begin{gathered} \mathbf{a}=\mathbf{b}(\AA) \\ \mathbf{c}(\AA) \\ \alpha=\beta\left({ }^{\circ}\right) \\ \gamma\left({ }^{\circ}\right) \\ \mathbf{V}\left(\AA^{3}\right) \\ \mathbf{Z} \\ D_{\mathrm{ca}}\left(\mathbf{g} . \mathrm{cm}^{-3}\right) \\ \mathbf{T}(\mathbf{K}) \\ \mathbf{R}[\mathbf{I}>\mathbf{2} \sigma] \\ \mathbf{R}_{\mathrm{w} 2}[\mathbf{I}>2 \sigma] \end{gathered}$ | $\begin{gathered} 47.4786(14) \\ 11.5392(4) \\ 90 \\ 120 \\ 22526.9(10) \\ 18 \\ 1.678 \\ 150(2) \\ 0.0439 \\ 0.1223 \end{gathered}$ | $\begin{gathered} 47.523(4) \\ 11.523(5) \\ 90 \\ 120 \\ 22482(3) \\ 18 \\ 1.385 \\ 150(2) \\ 0.0846 \\ 0.2048 \end{gathered}$ |

Table S1: X-Ray crystallographic details for complexes 1-Dy and 1-Y. Note that the squeeze procedure was used to treat disordered solvent in the case of $\mathbf{1 - D y}$ and disordered solvent and free triflate ion in the case of $\mathbf{1 - Y}$.


Figure S1: View of the packing of 1-Dy in the ab plane, obtained from the XRD analysis. Grey, blue, red, yellow, green, white and light blue spheres represent $C, N, O, S, F, H$ and Dy atoms, respectively.


Figure S2: Temperature dependence of $\chi_{M} T$ of 1-D $y_{\text {solid }}$ and simulation from ab-initio calculations. Inset: Field dependence of the magnetization 1-Dy solid at 2 K and simulation from ab-initio calculations.


Figure S3: Hysteresis curve of 1-Dy solid measured at 0.47 K with a field sweep rate of $16 \mathrm{Oe} . \mathrm{s}^{-1}$


Figure S4: Frequency dependence of $\chi_{M}{ }^{\prime \prime}$ of 1-D $y_{\text {solid, }}$ 1-D $y_{\text {solution }}$ and 1-D $y_{\text {photo }}$ measured with $H_{d c}=0$ Oe for temperatures ranging from 1.8 K (blue) to 5 K (red). Lines are guide to the eye.


Figure S5: Frequency dependence of $\chi_{\mathrm{M}}{ }^{\prime \prime}$ of $1-D y_{\text {solid, }} 1-D y_{\text {solution }}$ and $1-D y_{\text {photo }}$ at $4 K$ (top to bottom) with field from 0 Oe (red) to 2400 Oe (blue) with 300 Oe spacing. Lines are guide to the eye.


Figure S6: Frequency dependence of $\chi_{M}{ }^{\prime}$ of 1-D $y_{\text {solid, }}$ 1-Dy $y_{\text {solution }}$ and 1-D $y_{\text {photo }}$ measured with $H_{d c}=1200$ Oe for temperatures ranging from 2 K (blue) to 15 K (red). Lines are guide to the eye.


Figure S7: Temperature dependence of $\chi_{m}$ " of 1-D $y_{\text {solid, }}$ 1-D $y_{\text {solution }}$ and 1-D $y_{\text {photo }}$ measured with $H_{d c}=1200$ Oe for frequencies ranging from 1 Hz (red) to 1500 Hz (blue). Lines are guide to the eye.


Figure S8: Argand (Cole-Cole) plots and some selected fits for 1-Dy solid, $^{\text {1-Dy }}$ solution and 1-Dy photo measured with $\mathrm{H}_{\mathrm{dc}}=1200$ Oe for selected temperatures ranging from 2 K (blue) to 10 K (red).

Table S2: Extracted values from the fitting of $X^{\prime \prime}$ vs frequency curves under 1200 Oe field of 1-Dy ${ }_{\text {solid }}$

| $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ | $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ |
| :---: | :---: | :---: | :---: |
| 1.8 | 96084.56 | 7.5 | 1021.73 |
| 1.9 | 102399.6 | 8 | 799.17 |
| 2 | 100749.27 | 8.5 | 594.54 |
| 2.5 | 65555.84 | 9 | 473.52 |
| 3 | 36257.25 | 9.5 | 379.6 |
| 3.5 | 21709.96 | 10 | 329.27 |
| 4 | 13903.66 | 10.5 | 287.54 |
| 4.5 | 9001.14 | 11 | 254.31 |
| 5 | 5887.86 | 11.5 | 222.53 |
| 5.5 | 3960.92 | 12 | 184.9 |
| 6 | 2711.19 | 12.5 | 170.65 |
| 6.5 | 1907.88 | 13 | 164.44 |
| 7 | 1353.22 |  |  |

Table S3: Extracted values from the Argand plot under 1200 Oe field of 1-Dy solid

| T (K) | $\chi_{\text {s }}$ | $\chi^{\top}$ | $\alpha$ | $\mathrm{R}^{2}$ |
| :---: | :---: | :---: | :---: | :---: |
| 1.8 | 0.24104 | 6.74066 | 0.59547 | 0.99697 |
| 1.9 | 0.22679 | 6.61493 | 0.60664 | 0.99702 |
| 2 | 0.2258 | 6.39484 | 0.60017 | 0.99729 |
| 2.5 | 0.24422 | 5.27321 | 0.6126 | 0.99813 |
| 3 | 0.2895 | 4.2785 | 0.5896 | 0.99927 |
| 3.5 | 0.33186 | 3.59548 | 0.5615 | 0.99969 |
| 4 | 0.35132 | 3.12853 | 0.53968 | 0.99974 |
| 4.5 | 0.36933 | 2.76077 | 0.51603 | 0.99953 |
| 5 | 0.371 | 2.48 | 0.5029 | 0.99958 |
| 5.5 | 0.37736 | 2.24794 | 0.4884 | 0.99885 |
| 6 | 0.38143 | 2.0599 | 0.47694 | 0.99832 |
| 6.5 | 0.384 | 1.90268 | 0.46954 | 0.9973 |
| 7 | 0.38312 | 1.7713 | 0.46914 | 0.99319 |
| 7.5 | 0.399 | 1.65112 | 0.45342 | 0.99621 |
| 8 | 0.41581 | 1.54813 | 0.44111 | 0.99504 |
| 8.5 | 0.4211 | 1.4574 | 0.44035 | 0.99509 |
| 9 | 0.43326 | 1.37682 | 0.4321 | 0.99546 |
| 9.5 | 0.44717 | 1.30599 | 0.42633 | 0.99514 |
| 10 | 0.46948 | 1.24207 | 0.41361 | 0.99585 |
| 10.5 | 0.48938 | 1.18334 | 0.40483 | 0.99596 |
| 11 | 0.5079 | 1.1302 | 0.3942 | 0.99619 |
| 11.5 | 0.51832 | 1.08265 | 0.38992 | 0.99649 |
| 12 | 0.51214 | 1.03252 | 0.40479 | 0.99119 |
| 12.5 | 0.54784 | 0.99278 | 0.36998 | 0.99617 |
| 13 | 0.56059 | 0.95661 | 0.3597 | 0.99612 |
| 13.5 | 0.56146 | 0.92324 | 0.36467 | 0.9959 |
| 14 | 0.57109 | 0.89229 | 0.36147 | 0.99523 |
| 14.5 | 0.57884 | 0.8627 | 0.3546 | 0.99545 |
| 15 | 0.59261 | 0.83505 | 0.3444 | 0.99658 |

Table S4: Extracted values from the fitting of $X^{\prime \prime}$ vs frequency curves under 1200 Oe field of 1-Dy solution

| $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ | $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ |
| :---: | :---: | :---: | :---: |
| 1.8 | 143450.04376 | 6 | 1503.42543 |
| 1.9 | 131081.88343 | 6.5 | 1007.80015 |
| 2 | 118698.34227 | 7 | 719.47591 |
| 2.5 | 71002.95228 | 7.5 | 511.9609 |
| 3 | 40467.11818 | 8 | 384.37814 |
| 3.5 | 21854.94094 | 8.5 | 290.81952 |
| 4 | 12196.32838 | 9 | 233.64628 |
| 4.5 | 6868.09025 | 9.5 | 183.65093 |
| 5 | 3877.63302 | 10 | 160.39495 |
| 5.5 | 2369.11794 |  |  |

Table S5: Extracted values from the Argand plot under 1200 Oe field of 1-Dy solution

| $\mathrm{T}(\mathrm{K})$ | $\chi_{\mathrm{s}}$ | $\chi_{\mathrm{T}}$ | $\alpha$ | $\mathrm{R}^{2}$ |
| :---: | :---: | :---: | :---: | :---: |
| 1.8 | 0.04431 | 5.96841 | 0.37437 | 0.99964 |
| 1.9 | 0.03479 | 5.71858 | 0.38253 | 0.99939 |
| 2 | 0.03559 | 5.42408 | 0.38303 | 0.9997 |
| 2.5 | 0.02037 | 4.41179 | 0.40227 | 0.99985 |
| 3 | 0.01543 | 3.69211 | 0.40693 | 0.99965 |
| 4 | 0.00224 | 2.77965 | 0.41499 | 0.99756 |
| 4.5 | 0.03539 | 2.42977 | 0.39305 | 0.99682 |
| 5 | 0.02462 | 2.17785 | 0.39364 | 0.99283 |
| 5.5 | 0.04867 | 1.96565 | 0.37717 | 0.98823 |
| 6 | 0.06396 | 1.78769 | 0.37161 | 0.98415 |
| 6.5 | 0.09239 | 1.64132 | 0.35483 | 0.98651 |
| 7 | 0.12855 | 1.51655 | 0.33758 | 0.98793 |
| 7.5 | 0.15997 | 1.41119 | 0.32476 | 0.99043 |
| 8 | 0.19886 | 1.31954 | 0.31108 | 0.99125 |
| 8.5 | 0.24031 | 1.23594 | 0.28906 | 0.99154 |
| 9 | 0.27899 | 1.1635 | 0.27887 | 0.99119 |
| 9.5 | 0.31085 | 1.10116 | 0.26615 | 0.99061 |
| 10 | 0.34595 | 1.04271 | 0.2483 | 0.9947 |

Table S6: Extracted values from the fitting of $X^{\prime \prime}$ vs frequency curves under 1200 Oe field 1-Dy photo

| $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ | $\mathrm{T}(\mathrm{K})$ | $\tau(\mu \mathrm{s})$ |
| :---: | :---: | :---: | :---: |
| 1.8 | 23891.64684 | 5 | 2483.05791 |
| 1.9 | 22926.70661 | 5.5 | 1676.17778 |
| 2 | 22375.01916 | 6 | 1178.30995 |
| 2.25 | 20418.43954 | 6.5 | 838.62261 |
| 2.5 | 17682.37781 | 7 | 621.9 |
| 2.75 | 15026.57376 | 7.5 | 465.3299 |
| 3 | 12762.16544 | 8 | 357.88766 |
| 3.25 | 10681.60089 | 8.5 | 279.52919 |
| 3.5 | 8772.402 | 9 | 214.07418 |
| 3.75 | 7096.22103 | 9.5 | 190.4379 |
| 4 | 5714.96181 | 10 | 155.29641 |
| 4.5 | 3835.38525 |  |  |

Table S7: Extracted values from the Argand plot under 1200 Oe field of 1-Dy photo

| $\mathrm{T}(\mathrm{K})$ | $\chi_{\mathrm{s}}$ | $\chi_{\top}$ | $\alpha$ | $\mathrm{R}^{2}$ |
| :---: | :---: | :---: | :---: | :---: |
| 1.8 | 0.43191 | 9.36057 | 0.51867 | 0.99981 |
| 1.9 | 0.43872 | 8.96676 | 0.52102 | 0.99965 |
| 2 | 0.44582 | 8.63589 | 0.52492 | 0.99892 |
| 2.5 | 0.49941 | 7.11174 | 0.52628 | 0.99915 |
| 3 | 0.53038 | 5.95328 | 0.51645 | 0.99418 |
| 3.5 | 0.58371 | 5.07573 | 0.49391 | 0.99079 |
| 4 | 0.60278 | 4.40776 | 0.47152 | 0.99572 |
| 4.5 | 0.62667 | 3.91881 | 0.44965 | 0.99612 |
| 5 | 0.64001 | 3.49759 | 0.42886 | 0.99079 |
| 5.5 | 0.65017 | 3.17212 | 0.41155 | 0.99713 |
| 6 | 0.67306 | 2.90005 | 0.3918 | 0.99417 |
| 6.5 | 0.69023 | 2.67473 | 0.37949 | 0.99779 |
| 7 | 0.71638 | 2.47965 | 0.36161 | 0.99602 |
| 7.5 | 0.72926 | 2.31604 | 0.35418 | 0.99885 |
| 8 | 0.74631 | 2.17169 | 0.34437 | 0.99643 |
| 8.5 | 0.77511 | 2.03932 | 0.32621 | 0.99855 |
| 9 | 0.78060 | 1.92858 | 0.32844 | 0.99663 |
| 9.5 | 0.81975 | 1.82661 | 0.31666 | 0.99929 |
| 10 | 0.84427 | 1.73652 | 0.30292 | 0.997 |
| 10.5 | 0.85731 | 1.65346 | 0.30252 | 0.99821 |
| 11 | 0.88144 | 1.57906 | 0.2952 | 0.99588 |

Table S8: Computed energies levels (the ground state is set at zero), component values of the Lande factor $g$ and wavefunction composition for each $M_{J}$ state of the ground-state multiplet for 1-Dy.

|  | Energy ( $\mathrm{cm}^{-1}$ ) | $\mathrm{g}_{\mathrm{x}}$ | gy | $\mathrm{g}_{2}$ | Wavefunction composition ${ }^{\text {a }}$ |
| :---: | :---: | :---: | :---: | :---: | :---: |
| 1 | 0.0 | 0.00 | 0.00 | 19.69 | 0.98 \| $\pm 15 / 2>$ |
| 2 | 137.9 | 0.22 | 0.24 | 16.56 | 0.83\| $\pm 13 / 2>$ |
| 3 | 215.7 | 1.56 | 1.98 | 13.21 | $0.32\| \pm 11 / 2>+0.20\| \pm 9 / 2>+0.18 \mid \pm 1 / 2>$ |
| 4 | 258.5 | 1.77 | 4.75 | 11.14 | $\begin{gathered} 0.24\| \pm 7 / 2>+0.21\| \pm 11 / 2\rangle+0.18\| \pm 3 / 2\rangle \\ \quad+0.17\| \pm 5 / 2\rangle+0.11\| \pm 1 / 2\rangle \end{gathered}$ |
| 5 | 308.9 | 2.21 | 4.31 | 11.46 | $\begin{aligned} 0.30 \mid \pm 5 / 2>+ & 0.24\| \pm 3 / 2>+0.17\| \pm 11 / 2\rangle \\ & +0.13\| \pm 1 / 2\rangle \end{aligned}$ |
| 6 | 352.2 | 1.41 | 3.37 | 13.49 | $0.44\| \pm 1 / 2>+0.23\| \pm 3 / 2>+0.19 \mid \pm 9 / 2>$ |
| 7 | 394.8 | 0.42 | 1.20 | 18.17 | $\begin{gathered} 0.22\| \pm 3 / 2>+0.21\| \pm 5 / 2>+0.20\| \pm 7 / 2\rangle \\ +0.18\| \pm 9 / 2\rangle+0.13\| \pm 1 / 2\rangle \end{gathered}$ |
| 8 | 455.0 | 0.12 | 0.29 | 19.18 | $\begin{aligned} 0.33 \mid \pm 7 / 2> & +0.25\| \pm 9 / 2>+0.20\| \pm 5 / 2> \\ & +0.13\| \pm 11 / 2\rangle \end{aligned}$ |

${ }^{\text {a }}$ Only the contributions > 10\% are given.

