Electronic Supplementary Information (ESI)

S1 Experimental Section:

Materials and methods: All commercially available chemicals were used as supplied without further purification. The Q[5] was synthesized according to literature methods.[12] The C, H, and N microanalyses were carried out with a PE 240C elemental analyzer. The solid-state circular dichroism (CD) spectra were recorded on a Jasco J-810 spectropolarimeter with KBr pellets. The thermal gravimetric analyses (TGA) were carried out on a NETZSCH STA 449 C instrument under an air atmosphere with a heating rate of 20 °C /min. Powder X-ray diffraction measurements were recorded on a D/Max-2500 X-ray diffractometer using Cu_{Ka} radiation.

Preparation of compound 1: A mixture of Q[5]·$10H_2O$ (50 mg, 0.0495 mmol) and $Dy(NO_3)_3$ (103.5 mg, 0.297 mmol) in 1 mol·L^{-1} HCl (5 mL) was refluxed for 5 min and then filtered, the filtrate was allowed to stand to allow slow evaporation in air at room temperature, yielding colorless crystals of complex 1 within 5 days (39% yield on the basis of Q[5]·$10H_2O$). Anal. Calcd for $C_{30}H_{52}N_{22}O_{27}DyCl$: C, 26.67; H, 3.88; N, 22.81. Found: C, 26.59; H, 3.93; N, 22.73.

Preparation of compound 2: A mixture of Q[5]·$10H_2O$ (50 mg, 0.495 mmol), and a 4-fold excess of Hyq (217.8 mg, 1.98 mmol) were dissolved in 1 mol·L^{-1} HCl (5 mL) at 50°C while being stirred until the Q[5] and **Hyq** dissolved completely. A 6-fold excess of $Dy(NO_3)_3$ (103.5 mg, 2.97 mmol) was added with refluxing for 5 min and then filtered, the filtrate was allowed to stand to allow slow evaporation in air at room temperature. Red brown crystals were obtained from the solution over the course of 7 days. The yield in terms of Q[5]·$10H_2O$ was 51%. Anal. Calcd for $C_{36}H_{58}N_{23}O_{32}Dy$: C, 29.07; H, 3.93; N, 21.66. Found: C, 29.03; H, 3.97; N, 21.61.

S2 Crystallographic Information:

Diffraction data for compounds **1**, **2a** and **2b** were collected on a Bruker SMART Apex-II CCD diffractometer with graphite-monochromatic Mo-K_α radiation source (λ = 0.71073 Å) at 223 K. The data were corrected for absorption by SADABS program in BrukerAXS program package. All structures were solved by direct methods using SHELXS-97 and refined on F^2 by full-matrix least squares procedures with SHELXL-97.[22] All non-hydrogen atoms were treated anisotropically. Hydrogen atoms were placed in calculated positions and included as riding atoms with isotropic displacement parameters 1.2 – 1.5 times U_{eq} of the attached C or N atoms. It is should be mentioned that the program Squeeze in Platon[23] was applied to remove the anions because they could not be satisfactorily modeled for compound **2b**.

$C_{30}H_{52}N_{22}O_{27}DyCl$ (**1**) Monoclinic, space group $P21/n$, a = 14.473(2) Å, b = 22.512(3) Å, c = 16.595(3) Å, α = 90.00˚, β = 101.605(4) ˚, γ = 90.00˚, V = 5296.4(14) Å^3, Z = 4, Dc = 1.694 g·cm^{-3}, $F(000)$ = 2740, GOF = 1.060, R_1 = 0.0776 (I>2σ(I)), wR_2 = 0.2412 (all data).

$C_{36}H_{58}N_{23}O_{32}Dy$ (**2a**, right-handed helix): Hexagonal, space group $P6_1$, a = 14.4478(18) Å, b = 14.4478(18) Å, c = 46.109(12) Å, α = 90.00˚, β = 90.00˚, γ = 120.00˚, V = 8335(3) Å^3, Z = 6, Dc = 1.778 g·cm^{-3}, $F(000)$ = 4542, GOF = 1.005, R_1 = 0.0448 (I>2σ(I)), wR_2 = 0.1223 (all data).

$C_{36}H_{58}N_{23}O_{32}Dy$ (**2b**, left-handed helix): Hexagonal, space group $P6_5$, a = 14.5310(10) Å, b = 14.5310(10) Å, c = 46.070(7) Å, α = 90.00˚, β = 90.00˚, γ = 120.00˚, V = 8424.5(15) Å^3, Z = 6, Dc = 1.759 g·cm^{-3}, $F(000)$ = 4542, GOF = 1.069, R_1 = 0.0517 (I>2σ(I)), wR_2 = 0.1328 (all data).

CCDC 820006 for **1**, CCDC 820005 for **2a** and CCDC 844326 for **2b** 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.

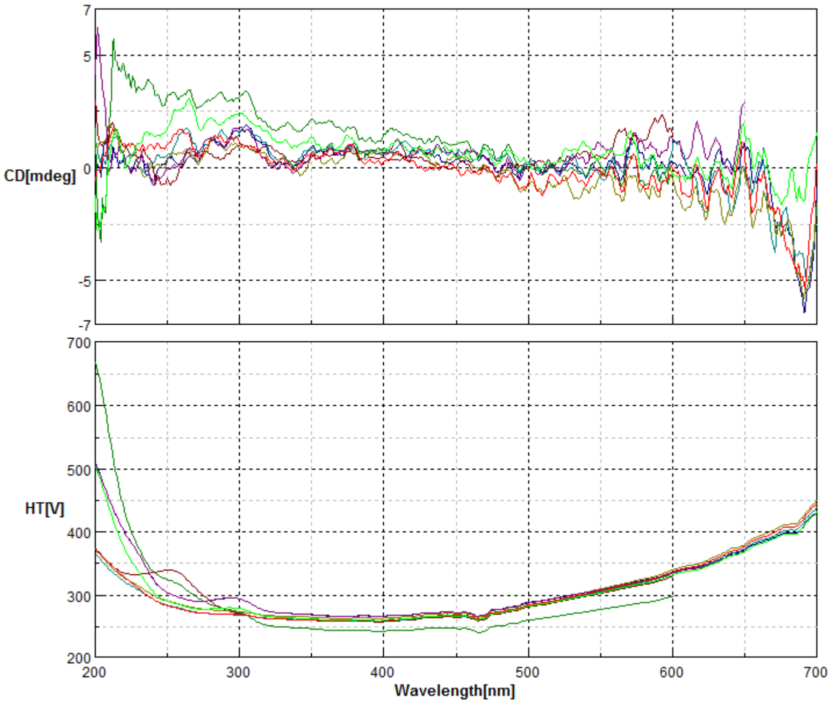
References:

22 (a) G. M. Sheldrick, SHELXS-97, *Program for X-ray Crystal Structure Determination*; University of Gottingen, Germany, **1997**. (b) G. M. Sheldrick, *SHELXL-97, Program for X-ray Crystal Structure Refinement*; University of Gottingen, Germany, **1997**. (c) G. M. Sheldrick, *Acta Crystallogr., Sect. A*, 2008, **64**, 112

23 A. L. Spek, *J. Appl. Crystallogr*. 2003, **36**, 7.

S3 Measurement of CD spectra:

Eight samples of individual single crystal of compound **2** were collected and dried. Each of these samples were mixed with KBr powder in a ratio of 1:200 and then pressed into a disk for use in the CD measurement. Following is the CD spectra.

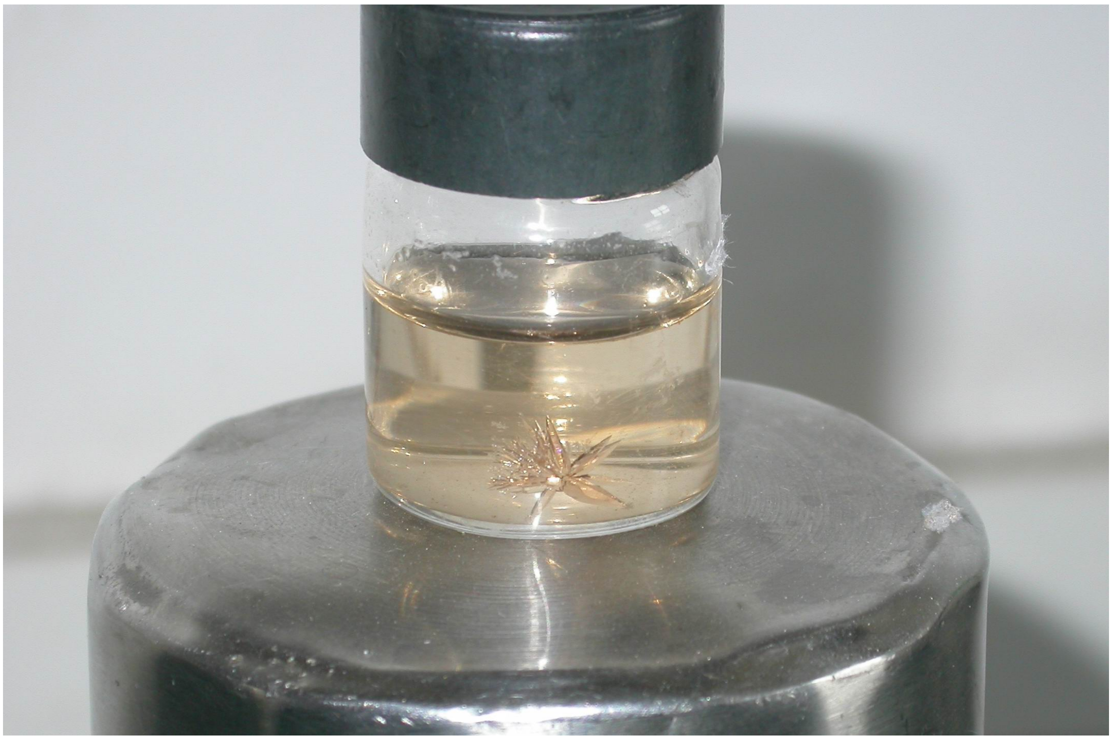


S4 Space group in a random beaker of compound 2.

For the first cluster, all the four single crystals were measured. For the other seven clusters, only one single crystal was measured.

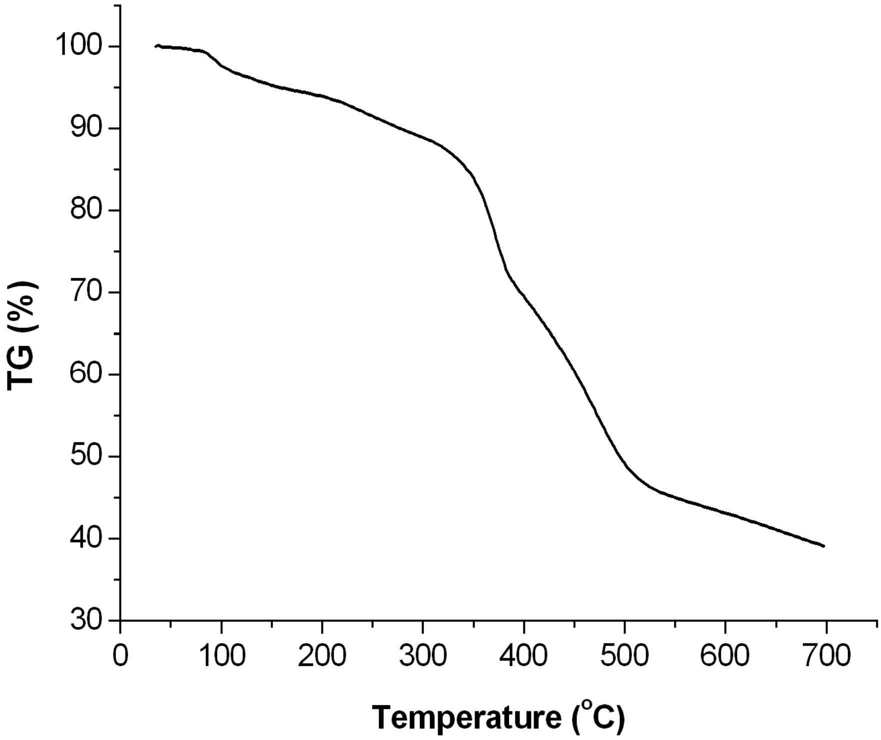
Crystal No.	Space Group	R Factor	Flack Parameter
1a	$P6_1$	0.0448	-0.001(10)
1b	$P6_1$	0.0372	0.000(11)
1c	$P6_1$	0.0413	0.001(10)
1d	$P6_1$	0.0462	0.004(10)
2	$P6_5$	0.0517	0.020(18)
3	$P6_1$	0.0581	-0.032(13)
4	$P6_5$	0.0511	-0.013(16)
5	$P6_1$	0.0432	0.024(16)
6	$P6_1$	0.0432	-0.02(2)
7	$P6_1$	0.0391	0.009(4)
8	$P6_1$	0.0378	0.001(4)

S5 The photo of a cluster of homochiral single crystals of compound 2.



S5 TG curve for compound 2

The TG study is performed under an air atmosphere. TG curve clearly shows the uncoordinated water molecules begin to eliminate when the temperature is increased to about 80 °C.



S6 The single-crystal photos of compounds 1 and 2.

S7 X-Ray powder diffractograms (XRPD) recorded for compound 2.

To verify the phase purity of samples, we carried out powder X-ray diffraction experiments.