## Enchaining EDTA-Chelated Lanthanide Molecular Magnets into Ordered 1D Networks

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under 1200 Oe applied dc field for 4. Temperature dependence of  $\chi_0$  and  $\chi_{inf}$  (top left),  $\chi_0$ - $\chi_{inf}$  (top right), and  $\alpha$  (bottom left). Relaxation times of the magnetization ln( $\tau$ ) vs. T<sup>-1</sup> for 2 (Arrhenius plot using  $\chi'$ (red) and  $\chi''$  (blue) ac data) under 1200 Oe applied dc field (bottom right). The solid red line corresponds to the fit of the data. 7

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## Supplementary Figures and Data



**Fig. S1** FTIR spectra of **1-4** for vacuum filtered crystals in the 4000-550 cm<sup>-1</sup> region.

Table S1. Sciected bond distances for complexes 1-	Table	<b>S1</b> .	Selected	bond	distances	for	comp	lexes	1-4
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Bond	Distance (Å)				
	1	2	3	4	
Ln1-01	2.333(8)	2.358(2)	2.392(3)	2.364(8)	
Ln1-03	2.395(7)	2.397(3)	2.435(3)	2.402(8)	
Ln1-05	2.351(7)	2.355(3)	2.388(3)	2.371(8)	
Ln1-07	2.397(8)	2.400(3)	2.422(4)	2.40(1)	
Ln1-09	2.422(7)	2.380(4)	2.430(4)	2.41(1)	
Ln1-010	2.513(7)	2.430(2)	2.471(3)	2.437(7)	
Ln1-011	2.371(8)	2.520(3)	2.550(3)	2.538(9)	
Ln1-N1	2.598(7)	2.655(3)	2.686(3)	2.64(1)	
Ln1-N2	2.650(9)	2.611(3)	2.642(4)	2.63(1)	
Ln2-012	-	2.402(3)	2.436(3)	2.409(8)	
Ln2-014	-	2.363(3)	2.394(4)	2.374(9)	
Ln2-016	-	2.392(2)	2.421(3)	2.394(8)	
Ln2-018	-	2.339(2)	2.386(3)	2.341(8)	
Ln2-020	-	2.390(2)	2.425(3)	2.382(7)	
Ln2-021	-	2.448(3)	2.470(4)	2.44(1)	
Ln2-022	-	2.532(3)	2.552(3)	2.513(9)	
Ln2-N3	-	2.667(4)	2.688(4)	2.70(1)	
Ln2-N4	-	2.614(3)	2.644(4)	2.61(1)	

Dihedral Angle	Gd-EDTA, Ref	Eu-EDTA, 2	Tb-EDTA, 3	Dy-EDTA, 4
	Gd1	Eu1	Tb1	Dy1
N2-N1-O3^O7-N2-O3	22.075(340)	21.445(157)	21.880(397)	20.902(135)
N2-N1-O3^O1-N2-N1	66.272(357)	66.697(125)	66.770(372)	67.411(123)
N2-N1-O3^O3-N1-O9	66.731(283)	67.090(119)	66.617(364)	67.570(108)
N2-07-03^07-03-010	53.199(261)	52.878(124)	53.346(312)	53.438(104)
N2-07-03^N2-07-05	61.510(253)	61.933(114)	62.031(299)	62.402(106)
O1-N2-N1^N1-O1-O9	58.226(302)	58.057(129)	57.454(342)	58.212(120)
01-N2-N1^01-N2-05	40.494(317)	40.215(127)	41.467(362)	41.02(11)
05-01-N2^011-01-05	38.221(253)	38.176(119)	36.871(290)	37.160(107)
05-01-N2^07-05-N2	59.007(261)	58.771(106)	59.322(314)	58.471(91)
011-05-01^09-01-011	54.455(269)	54.383(125)	55.187(323)	55.482(108)
011-05-01^011-010-05	75.165(241)	75.395(111)	75.10(28)	74.644(92)
010-09-03^07-03-010	54.385(249)	55.212(119)	54.349(311)	54.725(103)
010-05-011^010-09-011	69.787(287)	70.588(125)	69.717(329)	69.884(103)
09-010-011^011-01-09	56.485(288)	56.711(113)	56.709(333)	56.484(97)
09-010-011^09-03-010	50.199(271)	50.142(109)	49.625(307)	49.489(98)
09-011-01^09-N1-01	36.301(297)	35.759(131)	35.643(375)	35.183(114)
N1-09-01^09-03-N1	57.130(337)	56.806(144)	57.272(439)	57.298(111)
09-03-N1^09-03-010	35.427(257)	34.768(123)	35.352(346)	35.016(115)
05-07-010^011-05-010	8.541(273)	8.031(124)	9.261(300)	9.097(98)
05-07-010^N2-05-07	57.659(275)	57.926(129)	57.158(328)	57.364(109)
05-07-010^07-010-03	65.861(250)	66.589(110)	65.759(270)	66.057(82)
	Gd2	Eu2	Tb2	Dy2
022-020-021^020-012-021	<b>Gd2</b> 49.254(306)	<b>Eu2</b> 49.949(106)	<b>Tb2</b> 48.729(338)	<b>Dy2</b> 48.511(93)
022-020-021^020-012-021 022-020-021^020-014-022	<b>Gd2</b> 49.254(306) 56.674(315)	<b>Eu2</b> 49.949(106) 56.677(116)	<b>Tb2</b> 48.729(338) 56.159(357)	<b>Dy2</b> 48.511(93) 56.446(102)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021	<b>Gd2</b> 49.254(306) 56.674(315) 70.990(321)	<b>Eu2</b> 49.949(106) 56.677(116) 70.733(125)	Tb2   48.729(338)   56.159(357)   71.158(346)	<b>Dy2</b> 48.511(93) 56.446(102) 71.225(99)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)	<b>Dy2</b> 48.511(93) 56.446(102) 71.225(99) 55.594(92)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)	<b>Dy2</b> 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283)	Eu2   49.949(106)   56.677(116)   70.733(125)   55.291(111)   34.811(131)   66.610(103)   52.860(124)   61.984(113)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)	<b>Dy2</b> 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)	<b>Dy2</b> 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98) 37.603(106)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018 018-014-N4^014-N3-N4	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98) 37.603(106) 40.936(122)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018 018-014-N4^014-N3-N4 014-N3-N4^N4-N3-012	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345)	Eu2   49.949(106)   56.677(116)   70.733(125)   55.291(111)   34.811(131)   66.610(103)   52.860(124)   61.984(113)   58.730(121)   38.156(125)   40.227(136)   66.751(136)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98) 37.603(106) 40.936(122) 68.112(130)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^016-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018 018-014-N4^014-N3-N4 014-N3-N4^N3-020-014	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^021-016-021 020-021-012^021-016-021 016-012-021^018-021-016 016-012-021^018-N4-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018 018-014-N4^014-N3-N4 014-N3-N4^N3-020-014 020-N3-012^014-N3-020	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123) 56.778(140)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^018-04-012 018-016-N4^N4-016-012 018-016-N4^N4-014-018 018-014-N4^014-022-018 018-014-N4^014-N3-N4 014-N3-N4^N3-020-014 014-N3-N4^N3-020-014 020-N3-012^014-N3-020 020-N3-012^N3-N4-012	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349) 67.808(296)	Eu2   49.949(106)   56.677(116)   70.733(125)   55.291(111)   34.811(131)   66.610(103)   52.860(124)   61.984(113)   58.730(121)   38.156(125)   40.227(136)   66.751(136)   58.172(123)   56.778(140)   67.148(123)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 58.560(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104)
022-020-021^020-012-021 022-020-021^020-014-022 022-020-021^022-018-021 020-021-012^012-016-021 020-021-012^020-N3-012 016-012-021^018-021-016 016-012-021^018-042-012 018-016-N4^N4-016-012 018-016-N4^N4-014-012 018-016-N4^N4-014-018 018-014-N4^014-N3-N4 018-014-N4^014-N3-N4 014-N3-N4^N3-020-014 020-N3-012^014-N3-020 020-N3-012^N3-N4-012 022-018-021^014-022-018	Gd2   49.254(306)   56.674(315)   70.990(321)   55.559(252)   34.333(297)   66.683(229)   53.449(284)   62.705(283)   58.312(291)   37.998(257)   40.582(335)   68.027(345)   57.472(329)   56.858(349)   67.808(296)   74.594(266)	Eu2   49.949(106)   56.677(116)   70.733(125)   55.291(111)   34.811(131)   66.610(103)   52.860(124)   61.984(113)   58.730(121)   38.156(125)   40.227(136)   66.751(136)   58.172(123)   56.778(140)   67.148(123)   75.273(126)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)   74.272(275)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104) 74.286(92)
022-020-021^020-012-021   022-020-021^020-014-022   022-020-021^022-018-021   020-021-012^021-016-021   020-021-012^020-N3-012   016-012-021^018-021-016   016-012-021^016-N4-012   018-016-N4^N4-016-012   018-016-N4^N4-014-018   018-014-N4^014-N3-N4   014-N3-N4^N4-014-012   014-N3-N4^N3-020-014   020-N3-012^N3-N4-012   022-018-021^014-022-018   018-016-021^018-N4-016	Gd2   49.254(306)   56.674(315)   70.990(321)   55.559(252)   34.333(297)   66.683(229)   53.449(284)   62.705(283)   58.312(291)   37.998(257)   40.582(335)   68.027(345)   57.472(329)   56.858(349)   67.808(296)   74.594(266)   57.968(307)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123) 56.778(140) 67.148(123) 75.273(126) 57.779(127)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)   74.272(275)   57.703(326)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104) 74.286(92) 57.358(101)
022-020-021^020-012-021   022-020-021^020-014-022   022-020-021^022-018-021   020-021-012^021-016-021   020-021-012^020-N3-012   016-012-021^018-021-016   016-012-021^016-N4-012   018-016-N4^N4-016-012   018-016-N4^N4-014-018   018-014-N4^014-N3-N4   014-N3-N4^N3-020-014   020-N3-012^014-N3-020   020-N3-012^N3-N4-012   021-013-014-022-018   014-N3-N4^N3-020-014   020-N3-012^014-N3-020   020-N3-012^N3-N4-012   021-018-021^018-N4-016   012-N4-N3^016-012-N4	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349) 67.808(296) 74.594(266) 57.968(307) 19.798(414)	Eu2   49.949(106)   56.677(116)   70.733(125)   55.291(111)   34.811(131)   66.610(103)   52.860(124)   61.984(113)   58.730(121)   38.156(125)   40.227(136)   66.751(136)   58.172(123)   56.778(140)   67.148(123)   75.273(126)   57.779(127)   21.467(142)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)   74.272(275)   57.703(326)   19.463(393)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104) 74.286(92) 57.358(101) 20.293(137)
022-020-021^020-012-021   022-020-021^020-014-022   022-020-021^022-018-021   020-021-012^012-016-021   020-021-012^020-N3-012   016-012-021^018-021-016   016-012-021^016-N4-012   018-016-N4^N4-016-012   018-016-N4^N4-014-018   018-014-N4^014-N22-018   018-014-N4^014-N3-N4   014-N3-N4^N3-020-014   020-N3-012^N3-N4-012   022-018-021^014-N3-020   020-N3-012^N3-N4-012   022-018-021^018-N4-016   012-N4-N3^016-012-N4   014-020-N3^020-022-014	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349) 67.808(296) 74.594(266) 57.968(307) 19.798(414) 35.739(295)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123) 56.778(140) 67.148(123) 75.273(126) 57.779(127) 21.467(142) 35.607(133)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)   74.272(275)   57.703(326)   19.463(393)   35.257(333)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104) 74.286(92) 57.358(101) 20.293(137) 35.387(109)
022-020-021^020-012-021   022-020-021^020-014-022   022-020-021^022-018-021   020-021-012^012-016-021   020-021-012^020-N3-012   016-012-021^018-021-016   016-012-021^016-N4-012   018-016-N4^N4-016-012   018-016-N4^N4-014-018   018-014-N4^014-022-018   018-014-N4^014-N3-N4   014-N3-N4^N3-020-014   020-N3-012^N3-N4-012   020-N3-012^N3-N4-012   022-018-021^014-022-018   018-016-021^018-N4-016   012-N4-N3^016-012-N4   014-020-N3^020-022-014   014-020-N3^020-022-014	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349) 67.808(296) 74.594(266) 57.968(307) 19.798(414) 35.739(295) 8.578(279)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123) 56.778(140) 67.148(123) 75.273(126) 57.779(127) 21.467(142) 35.607(133) 7.997(116)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   58.440(305)   56.738(337)   68.629(345)   74.272(275)   57.703(326)   19.463(393)   35.257(333)   8.704(292)	Dy2 48.511(93) 56.446(102) 71.225(99) 55.594(92) 34.434(102) 66.049(80) 53.564(95) 62.736(98) 37.603(106) 40.936(122) 68.112(130) 57.501(113) 57.005(112) 67.637(104) 74.286(92) 57.358(101) 20.293(137) 35.387(109) 9.146(93)
022-020-021^020-012-021   022-020-021^020-014-022   022-020-021^022-018-021   020-021-012^012-016-021   020-021-012^020-N3-012   016-012-021^018-021-016   016-012-021^016-N4-012   018-016-N4^N4-016-012   018-016-N4^N4-014-018   018-014-N4^014-022-018   018-014-N4^014-N3-N4   014-N3-N4^N3-020-014   020-N3-012^N3-N4-012   022-018-021^014-N3-020   020-N3-012^N3-N4-012   022-018-021^018-N4-016   012-N4-N3^016-012-N4   014-020-N3^020-022-014   018-016-021^018-N4-016   012-N4-N3^016-012-N4   014-020-N3^020-022-014   018-021-016^021-022-018   020-022-014^018-014-022	Gd2 49.254(306) 56.674(315) 70.990(321) 55.559(252) 34.333(297) 66.683(229) 53.449(284) 62.705(283) 58.312(291) 37.998(257) 40.582(335) 68.027(345) 57.472(329) 56.858(349) 67.808(296) 74.594(266) 57.968(307) 19.798(414) 35.739(295) 8.578(279) 54.403(299)	Eu2 49.949(106) 56.677(116) 70.733(125) 55.291(111) 34.811(131) 66.610(103) 52.860(124) 61.984(113) 58.730(121) 38.156(125) 40.227(136) 66.751(136) 58.172(123) 56.778(140) 67.148(123) 75.273(126) 57.779(127) 21.467(142) 35.607(133) 7.997(116) 54.577(132)	Tb2   48.729(338)   56.159(357)   71.158(346)   55.972(326)   34.164(354)   66.580(285)   53.592(346)   62.655(294)   58.463(300)   38.125(319)   40.000(344)   68.477(340)   56.738(337)   68.629(345)   74.272(275)   57.703(326)   19.463(393)   35.257(333)   8.704(292)   54.706(315)	Dy2   48.511(93)   56.446(102)   71.225(99)   55.594(92)   34.434(102)   66.049(80)   53.564(95)   62.736(98)   58.560(98)   37.603(106)   40.936(122)   68.112(130)   57.501(113)   57.005(112)   67.637(104)   74.286(92)   57.358(101)   20.293(137)   35.387(109)   9.146(93)   55.053(109)

**Table S2.** Dihedral angles along the edges of coordination polyhedra, used to determine theanalogous measurement (AM) of complexes 2-4. Gd-EDTA was employed as reference.

**Table S3.** Dihedral angles (°) along the edges of the coordination polyhedra used to determine the analogous measurement (AM) of polyhedra Dy1 *vs*. Dy2 and Tb1 *vs*. Tb2, within complexes **3**, and **4**.

Dihedral Angle	Dy1	Tb1
05-07-010^011-05-010	9.097(98)	9.261(300)
N2-N1-O3^O7-N2-O3	20.902(135)	21.880(397)
09-03-N1^09-03-010	35.016(115)	35.352(346)
09-011-01^09-N1-01	35.183(114)	35.643(375)
05-01-N2^011-01-05	37.160(107)	36.871(290)
01-N2-N1^01-N2-05	41.02(11)	41.467(362)
09-010-011^09-03-010	49.489(98)	49.625(307)
N2-07-03^07-03-010	53.438(104)	53.346(312)
010-09-03^07-03-010	54.725(103)	54.349(311)
011-05-01^09-01-011	55.482(108)	55.187(323)
09-010-011^011-01-09	56.484(97)	56.709(333)
N1-09-01^09-03-N1	57.298(111)	57.272(439)
05-07-010^N2-05-07	57.364(109)	57.158(328)
01-N2-N1^N1-01-09	58.212(120)	57.454(342)
05-01-N2^07-05-N2	58.471(91)	59.322(314)
N2-07-03^N2-07-05	62.402(106)	62.031(299)
05-07-010^07-010-03	66.057(82)	65.759(270)
N2-N1-O3^O1-N2-N1	67.411(123)	66.770(372)
N2-N1-O3^O3-N1-O9	67.570(108)	66.617(364)
010-05-011^010-09-011	69.884(103)	69.717(329)
011-05-01^011-010-05	74.644(92)	75.10(28)
	Dy2	Tb2
018-021-016^021-022-018	9.146(93)	8.704(292)
012-N4-N3^016-012-N4	20.293(137)	19.463(393)
020-021-012^020-N3-012	34.434(102)	34.164(354)
014-020-N3^020-022-014	35.387(109)	35.257(333)
018-014-N4^014-022-018	37.603(106)	38.125(319)
018-014-N4^014-N3-N4	40.936(122)	40.000(344)
022-020-021^020-012-021	48.511(93)	48.729(338)
016-012-021^016-N4-012	53.564(95)	53.592(346)
020-022-014^018-014-022	55.053(109)	54.706(315)
020-021-012^012-016-021	55.594(92)	55.972(326)
022-020-021^020-014-022	56.446(102)	56.159(357)
020-N3-012^014-N3-020	57.005(112)	56.738(337)
018-016-021^018-N4-016	57.358(101)	57.703(326)
014-N3-N4^N3-O20-O14	57.501(113)	58.440(305)
018-016-N4^N4-014-018	58.560(98)	58.463(300)
018-016-N4^N4-016-012	62.736(98)	62.655(294)
016-012-021^018-021-016	66.049(80)	66.580(285)
020-N3-012^N3-N4-012	67.637(104)	68.629(345)
014-N3-N4^N4-N3-012	68.112(130)	68.477(340)
022-020-021^022-018-021	71.225(99)	/1.158(346)
022-018-021^014-022-018	74.286(92)	74.272(275)
AM	0.346301	0.78927



Fig. S2 Field dependence of the magnetisation of: (a) 2, (b) 3 and (c) 4 at indicated temperatures.



Fig. S3 *M* vs. *HT*<sup>-1</sup> plot for: (a) 2, (b) 3 and (c) 4 at indicated temperatures.



**Fig. S4** Data obtained from generalized Debye fits of  $\chi'$  and  $\chi''$  frequency dependent ac data under 3000 Oe applied dc field for **3**. Temperature dependence of  $\chi_0$  and  $\chi_{inf}$  (top left),  $\chi_0$ - $\chi_{inf}$  (top right), and  $\alpha$  (bottom left). Relaxation times of the magnetization  $\ln(\tau)$  vs. T<sup>-1</sup> for **3** (Arrhenius plot using  $\chi'$  (red) and  $\chi''$  (blue) ac data) under 3000 Oe applied dc field (bottom right). The solid red line corresponds to the fit of the data.



**Fig. S5** Data obtained from generalized Debye fits of  $\chi'$  and  $\chi''$  frequency dependent ac data under 1200 Oe applied dc field for **4**. Temperature dependence of  $\chi_0$  and  $\chi_{inf}$  (top left),  $\chi_0$ - $\chi_{inf}$  (top right), and  $\alpha$  (bottom left). Relaxation times of the magnetization ln( $\tau$ ) vs. T<sup>-1</sup> for **2** (Arrhenius plot using  $\chi'$  (red) and  $\chi''$  (blue) ac data) under 1200 Oe applied dc field (bottom right). The solid red line corresponds to the fit of the data.

<i>T</i> (K)	$\tau$ (s)	α	X <sub>inf</sub>	X <sub>0</sub>
6	0.000001739	0.72055	0	2.21193
5.5	0.000003238	0.67703	0	2.64156
5	0.000003631	0.69226	0	2.8015
4.5	0.000083010	0.65324	0.64556	3.60222
4	0.000503393	0.46878	1.4722	5.08173
3.75	0.001129997	0.41435	1.69645	5.60608
3.5	0.002739993	0.3676	1.8774	6.13707
3.25	0.007119981	0.3529	2.07863	6.66517
3	0.018079981	0.34831	2.29181	7.23623
2.75	0.037900158	0.36606	2.57326	7.85101
2.5	0.062090021	0.38118	2.87758	8.58645
2.25	0.089829666	0.39885	3.26144	9.50147
2	0.124259892	0.38299	3.51166	10.50988
1.8	0.144830672	0.38273	3.6697	11.04093

**Table S4.** Cole-Cole fitting values using a generalized Debye model for ac susceptibility data of4 under 1200 Oe applied dc field.