

Fig. S1 Powder XRD patterns for compounds **1** (blue), **2** (red), **3** (black) and theoretical (light blue) for comparison

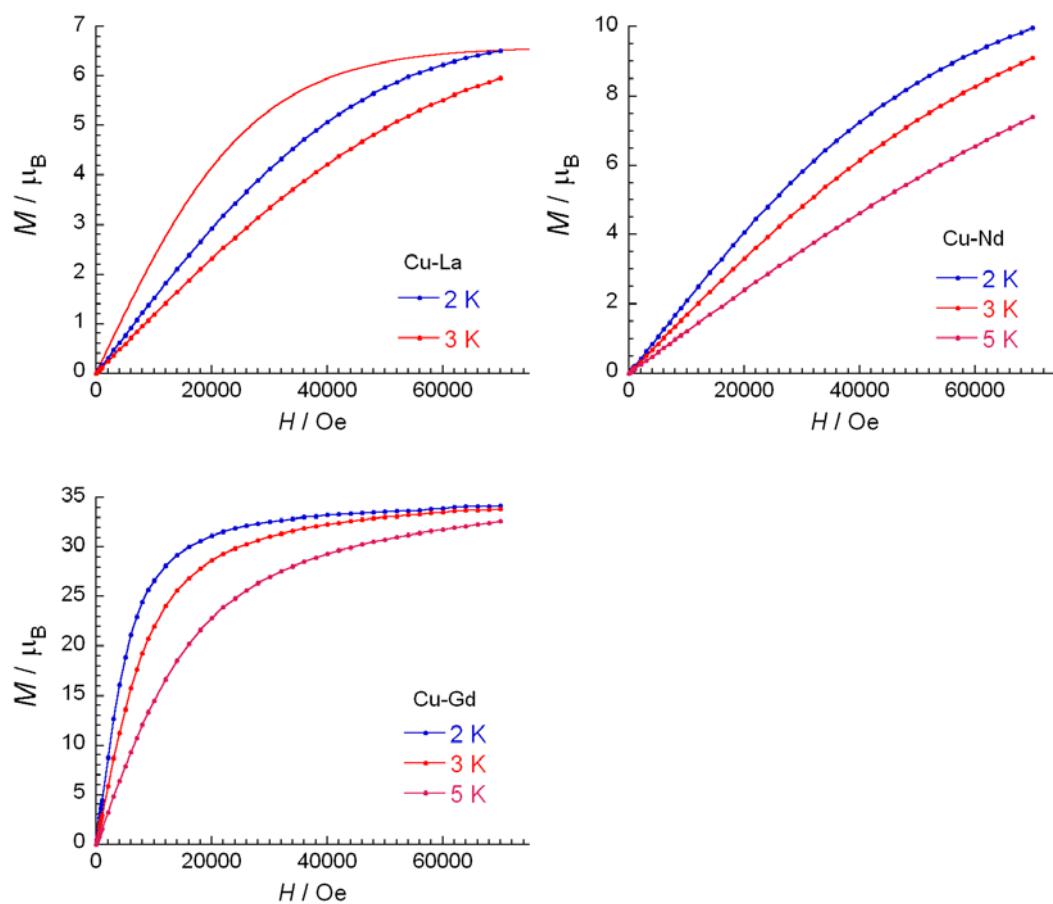


Figure S2 The magnetization of compounds **1-3**. The line without markers presented in **1** compound is the calculated curve corresponding to the sum of six Brillouin functions of $S = 1/2$ and $g = 2.2$ (see the main text for information).

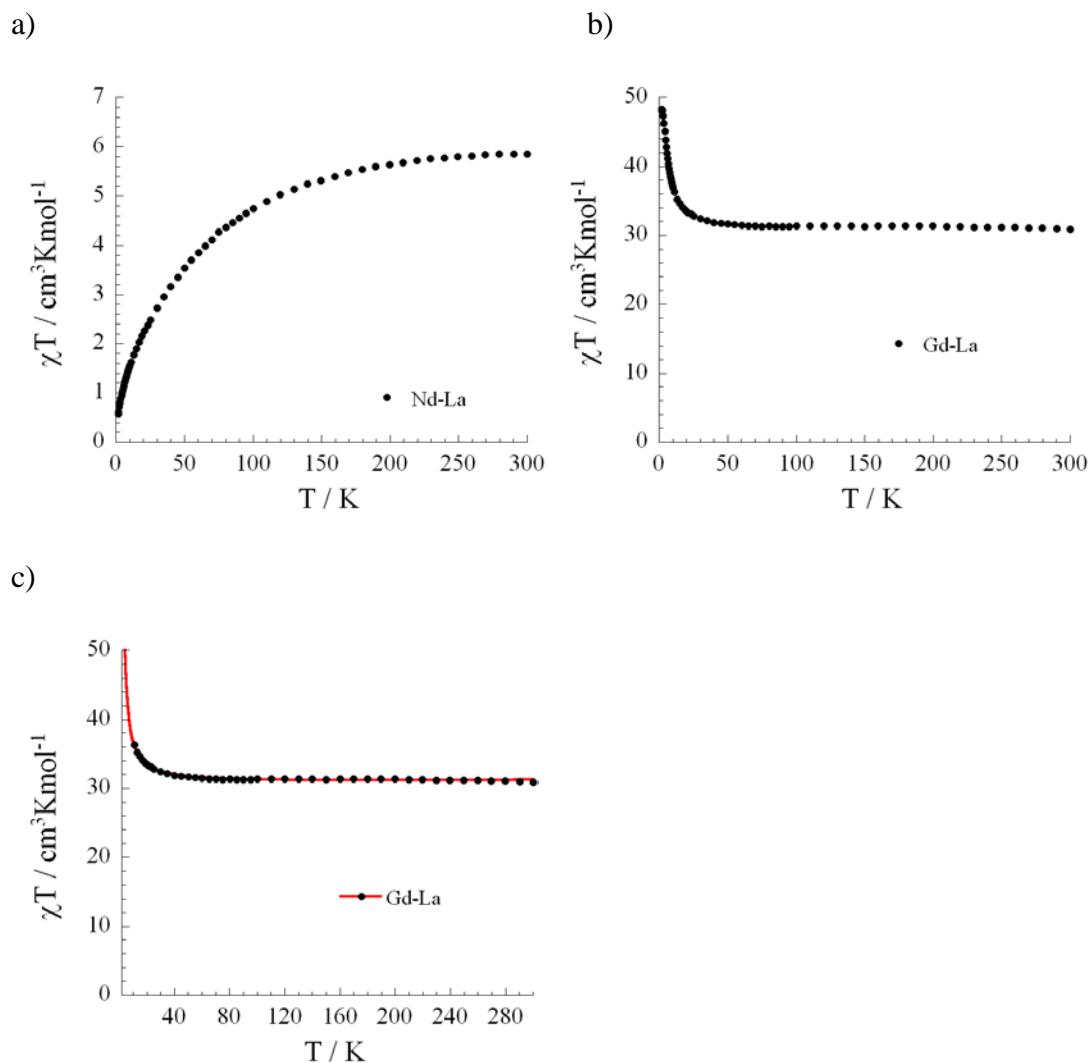


Figure S3 The χT vs T plots for **2**-Nd (a) and **3**-Gd (b) with the data for **1**-La subtracted, i.e. without the magnetic contribution of the six Cu ions. The Curie-Weiss fitting (above 20 K) of the χT vs T plot of **3**-Gd (b) with the data for **1**-La subtracted (c).

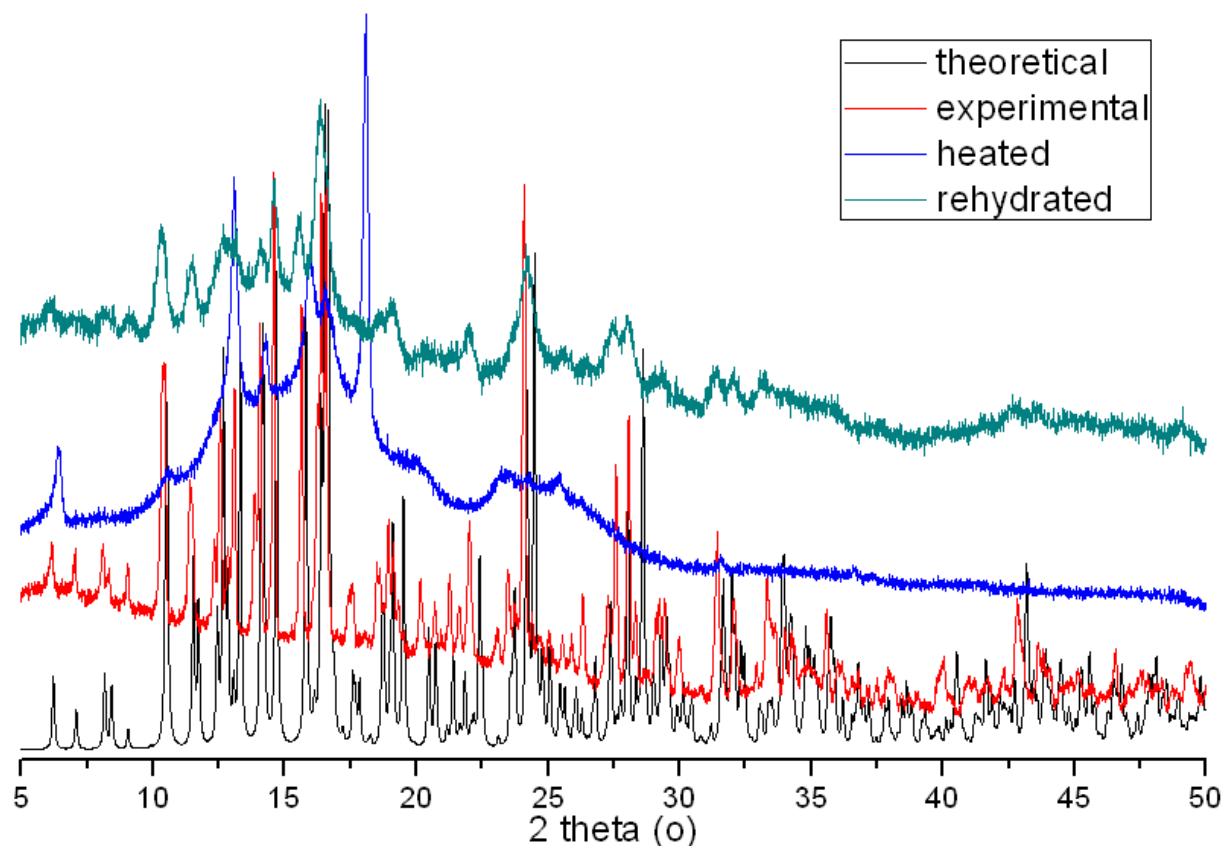


Fig S4 Powder XRD patterns for compound 2, theoretical (black), experimental (red), heated (blue) rehydrated (light blue)

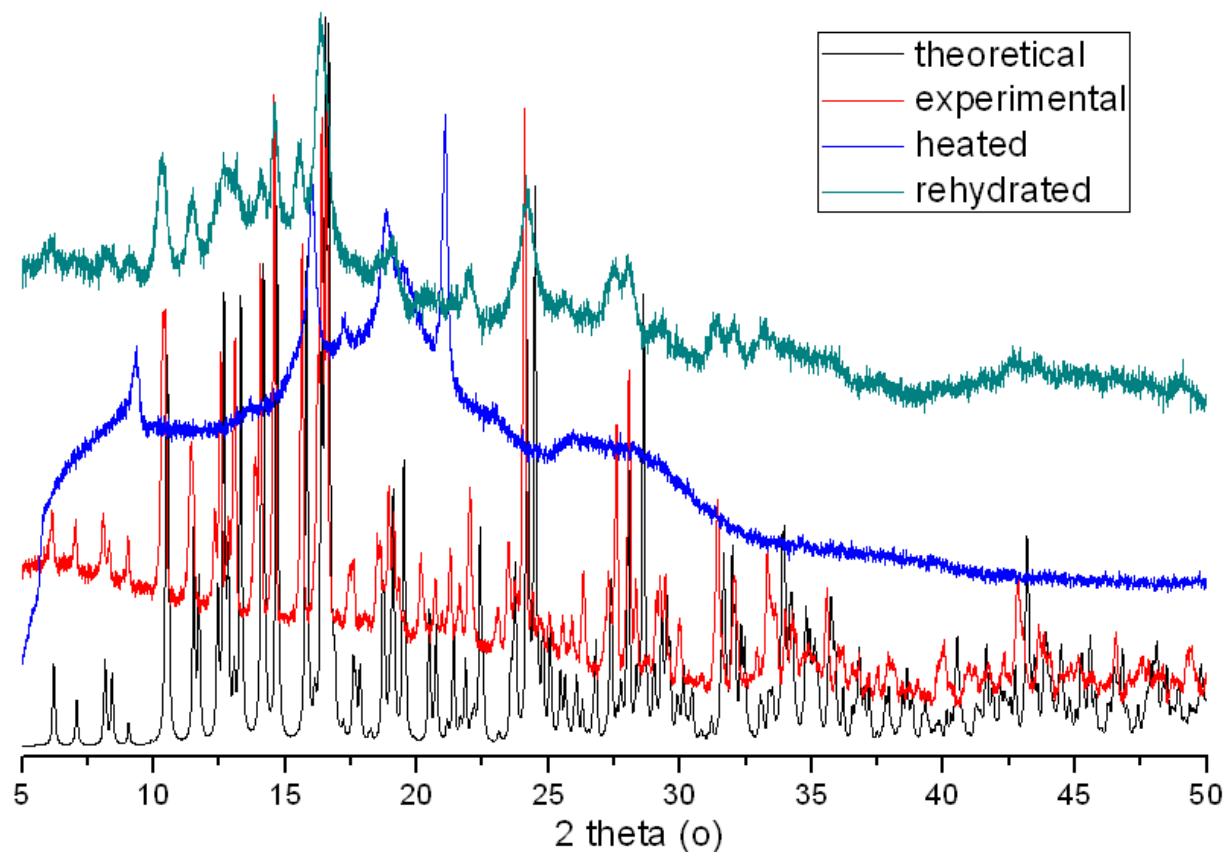


Fig S5 Powder XRD patterns for compound 3, theoretical (black), experimental (red), heated (blue) rehydrated (light blue)

Table S1. Bond lengths [\AA] for **1**

La(1)-O(3A)	2.406(7)	N(1A)-C(2A)	1.446(13)
La(1)-O(3B) ^{#1}	2.440(7)	N(2A)-C(5A)	1.292(13)
La(1)-O(4A) ^{#2}	2.442(7)	N(2A)-C(6A)	1.491(12)
La(1)-O(6B)	2.501(7)	C(1A)-C(2A)	1.515(15)
La(1)-O(4W)	2.541(7)	C(3A)-C(4A)	1.513(13)
La(1)-O(1W)	2.551(7)	C(4A)-C(5A)	1.522(13)
La(1)-O(2W)	2.555(8)	C(6A)-C(7A)	1.513(15)
La(1)-O(3W)	2.559(7)	Cu(2)-N(2B)	1.895(8)
O(1W)-H(11W)	0.8392	Cu(2)-N(1B)	1.907(8)
O(1W)-H(21W)	0.8964	Cu(2)-O(1B)	1.950(7)
O(2W)-H(12W)	0.8915	Cu(2)-O(5B)	1.955(7)
O(2W)-H(22W)	0.8269	O(1B)-C(1B)	1.276(12)
O(3W)-H(13W)	0.8333	O(2B)-C(1B)	1.257(12)
O(3W)-H(23W)	0.8217	O(3B)-C(3B)	1.259(12)
O(4W)-H(14W)	0.8798	O(3B)-La(1) ^{#1}	2.440(7)
O(4W)-H(24W)	0.8323	O(4B)-C(5B)	1.265(13)
La(2)-O(4B)	2.374(8)	O(5B)-C(7B)	1.289(12)
La(2)-O(4C) ^{#3}	2.433(7)	O(6B)-C(7B)	1.249(13)
La(2)-O(3C)	2.456(7)	N(1B)-C(3B)	1.325(13)
La(2)-O(8W)	2.519(7)	N(1B)-C(2B)	1.478(12)
La(2)-O(9W)	2.531(7)	N(2B)-C(5B)	1.300(13)
La(2)-O(7W)	2.540(8)	N(2B)-C(6B)	1.453(12)
La(2)-O(5W)	2.549(9)	C(1B)-C(2B)	1.510(14)
La(2)-O(6W)	2.594(7)	C(3B)-C(4B)	1.528(13)
O(5W)-H(15W)	0.9709	C(4B)-C(5B)	1.501(14)
O(5W)-H(25W)	0.8867	C(6B)-C(7B)	1.494(14)
O(6W)-H(16W)	0.8840	Cu(3)-N(1C)	1.898(9)
O(6W)-H(26W)	0.9221	Cu(3)-N(2C)	1.905(8)
O(7W)-H(17W)	0.8827	Cu(3)-O(5C)	1.932(8)
O(7W)-H(27W)	0.8611	Cu(3)-O(1C)	1.963(7)
O(8W)-H(18W)	0.8872	O(1C)-C(1C)	1.262(13)
O(8W)-H(28W)	0.9442	O(2C)-C(1C)	1.254(13)
O(9W)-H(19W)	0.8841	O(3C)-C(3C)	1.276(12)
O(9W)-H(29W)	0.8796	O(4C)-C(5C)	1.264(12)
Cu(1)-N(2A)	1.902(8)	O(4C)-La(2) ^{#3}	2.433(7)
Cu(1)-N(1A)	1.907(8)	O(5C)-C(7C)	1.307(14)
Cu(1)-O(2A)	1.922(7)	O(6C)-C(7C)	1.217(14)
Cu(1)-O(5A)	1.948(7)	N(1C)-C(3C)	1.325(13)
O(1A)-C(1A)	1.230(12)	N(1C)-C(2C)	1.454(13)
O(2A)-C(1A)	1.303(12)	N(2C)-C(5C)	1.317(13)
O(3A)-C(3A)	1.265(12)	N(2C)-C(6C)	1.459(13)
O(4A)-C(5A)	1.269(13)	C(1C)-C(2C)	1.528(14)
O(4A)-La(1) ^{#2}	2.442(7)	C(3C)-C(4C)	1.519(13)
O(5A)-C(7A)	1.277(12)	C(4C)-C(5C)	1.510(14)
O(6A)-C(7A)	1.247(12)	C(6C)-C(7C)	1.522(15)
N(1A)-C(3A)	1.312(13)		

Symmetry transformations used to generate equivalent atoms: #1 -x+1,-y+1,-z+2 #2 -x,-y,-z+2
 #3 -x+1,-y,-z+3

Table S2. Bond lengths [\AA] for 2

Nd(1)-O(3A)	2.347(3)	N(1A)-C(2A)	1.443(6)
Nd(1)-O(4A)#1	2.382(3)	N(2A)-C(5A)	1.280(6)
Nd(1)-O(3B)#2	2.385(3)	N(2A)-C(6A)	1.492(6)
Nd(1)-O(6B)	2.443(3)	C(1A)-C(2A)	1.521(7)
Nd(1)-O(4W)	2.474(3)	C(3A)-C(4A)	1.513(6)
Nd(1)-O(1W)	2.492(4)	C(4A)-C(5A)	1.512(6)
Nd(1)-O(3W)	2.499(3)	C(6A)-C(7A)	1.497(7)
Nd(1)-O(2W)	2.505(4)	Cu(2)-N(2B)	1.896(4)
O(1W)-H(11W)	0.8636	Cu(2)-N(1B)	1.912(4)
O(1W)-H(21W)	0.9250	Cu(2)-O(1B)	1.950(3)
O(2W)-H(12W)	0.9021	Cu(2)-O(5B)	1.963(3)
O(2W)-H(22W)	0.8425	O(1B)-C(1B)	1.281(6)
O(3W)-H(13W)	0.8507	O(2B)-C(1B)	1.239(6)
O(3W)-H(23W)	0.8514	O(3B)-C(3B)	1.258(5)
O(4W)-H(14W)	0.9035	O(3B)-Nd(1)#2	2.385(3)
O(4W)-H(24W)	0.8729	O(4B)-C(5B)	1.275(6)
Nd(2)-O(4B)	2.303(4)	O(5B)-C(7B)	1.281(6)
Nd(2)-O(4C)#3	2.369(3)	O(6B)-C(7B)	1.235(6)
Nd(2)-O(3C)	2.400(3)	N(1B)-C(3B)	1.302(6)
Nd(2)-O(8W)	2.454(3)	N(1B)-C(2B)	1.463(6)
Nd(2)-O(7W)	2.467(4)	N(2B)-C(5B)	1.302(6)
Nd(2)-O(9W)	2.470(3)	N(2B)-C(6B)	1.460(6)
Nd(2)-O(5W)	2.495(4)	C(1B)-C(2B)	1.513(7)
Nd(2)-O(6W)	2.553(3)	C(3B)-C(4B)	1.536(7)
O(5W)-H(15W)	1.0069	C(4B)-C(5B)	1.475(7)
O(5W)-H(25W)	0.8559	C(6B)-C(7B)	1.503(7)
O(6W)-H(16W)	0.8394	Cu(3)-N(2C)	1.905(4)
O(6W)-H(26W)	0.9626	Cu(3)-N(1C)	1.912(4)
O(7W)-H(17W)	0.9271	Cu(3)-O(5C)	1.942(3)
O(7W)-H(27W)	0.8872	Cu(3)-O(1C)	1.962(3)
O(8W)-H(18W)	0.8890	O(1C)-C(1C)	1.282(6)
O(8W)-H(28W)	0.9655	O(2C)-C(1C)	1.242(6)
O(9W)-H(19W)	0.8908	O(3C)-C(3C)	1.271(5)
O(9W)-H(29W)	0.8554	O(4C)-C(5C)	1.268(5)
Cu(1)-N(2A)	1.904(4)	O(4C)-Nd(2)#3	2.369(3)
Cu(1)-N(1A)	1.911(4)	O(5C)-C(7C)	1.302(6)
Cu(1)-O(2A)	1.936(3)	O(6C)-C(7C)	1.221(6)
Cu(1)-O(5A)	1.954(3)	N(1C)-C(3C)	1.311(6)
O(1A)-C(1A)	1.242(6)	N(1C)-C(2C)	1.449(6)
O(2A)-C(1A)	1.300(6)	N(2C)-C(5C)	1.311(6)
O(3A)-C(3A)	1.267(6)	N(2C)-C(6C)	1.456(6)
O(4A)-C(5A)	1.293(6)	C(1C)-C(2C)	1.508(6)
O(4A)-Nd(1)#1	2.382(3)	C(3C)-C(4C)	1.506(6)
O(5A)-C(7A)	1.300(5)	C(4C)-C(5C)	1.506(6)
O(6A)-C(7A)	1.248(5)	C(6C)-C(7C)	1.514(7)
N(1A)-C(3A)	1.297(6)		

Symmetry transformations used to generate equivalent atoms: #1 -x,-y,-z+2 #2 -x+1,-y+1,-z+2
 #3 -x+1,-y,-z+3

Table S3. Bond lengths [\AA] for **3**

Gd(1)-O(3A)	2.295(3)	N(1A)-C(2A)	1.451(7)
Gd(1)-O(4A)#1	2.327(3)	N(2A)-C(5A)	1.305(6)
Gd(1)-O(3B)#2	2.331(4)	N(2A)-C(6A)	1.470(6)
Gd(1)-O(6B)	2.394(4)	C(1A)-C(2A)	1.528(7)
Gd(1)-O(4W)	2.417(4)	C(3A)-C(4A)	1.517(7)
Gd(1)-O(1W)	2.431(4)	C(4A)-C(5A)	1.517(7)
Gd(1)-O(3W)	2.449(4)	C(6A)-C(7A)	1.513(7)
Gd(1)-O(2W)	2.457(4)	Cu(2)-N(2B)	1.894(4)
O(1W)-H(11W)	0.8645	Cu(2)-N(1B)	1.914(4)
O(1W)-H(21W)	0.9362	Cu(2)-O(1B)	1.954(3)
O(2W)-H(12W)	0.9274	Cu(2)-O(5B)	1.963(4)
O(2W)-H(22W)	0.8754	O(1B)-C(1B)	1.283(6)
O(3W)-H(13W)	0.8745	O(2B)-C(1B)	1.242(6)
O(3W)-H(23W)	0.8475	O(3B)-C(3B)	1.264(6)
O(4W)-H(14W)	0.9571	O(3B)-Gd(1)#2	2.331(4)
O(4W)-H(24W)	0.8916	O(4B)-C(5B)	1.274(6)
Gd(2)-O(4B)	2.259(4)	O(5B)-C(7B)	1.283(6)
Gd(2)-O(4C)#3	2.323(3)	O(6B)-C(7B)	1.240(6)
Gd(2)-O(3C)	2.351(3)	N(1B)-C(3B)	1.307(6)
Gd(2)-O(7W)	2.402(4)	N(1B)-C(2B)	1.454(6)
Gd(2)-O(8W)	2.404(4)	N(2B)-C(5B)	1.314(7)
Gd(2)-O(9W)	2.409(4)	N(2B)-C(6B)	1.441(6)
Gd(2)-O(5W)	2.430(4)	C(1B)-C(2B)	1.515(7)
Gd(2)-O(6W)	2.510(4)	C(3B)-C(4B)	1.521(7)
O(5W)-H(15W)	1.0502	C(4B)-C(5B)	1.482(8)
O(5W)-H(25W)	0.8665	C(6B)-C(7B)	1.505(7)
O(6W)-H(16W)	0.8166	Cu(3)-N(1C)	1.897(4)
O(6W)-H(26W)	0.9744	Cu(3)-N(2C)	1.906(4)
O(7W)-H(17W)	0.9920	Cu(3)-O(5C)	1.951(4)
O(7W)-H(27W)	0.7954	Cu(3)-O(1C)	1.961(4)
O(8W)-H(18W)	0.9007	O(1C)-C(1C)	1.278(7)
O(8W)-H(28W)	0.9956	O(2C)-C(1C)	1.249(6)
O(9W)-H(19W)	0.9151	O(3C)-C(3C)	1.267(6)
O(9W)-H(29W)	0.8814	O(4C)-C(5C)	1.272(6)
Cu(1)-N(2A)	1.905(4)	O(4C)-Gd(2)#3	2.323(3)
Cu(1)-N(1A)	1.907(4)	O(5C)-C(7C)	1.299(7)
Cu(1)-O(2A)	1.929(4)	O(6C)-C(7C)	1.223(7)
Cu(1)-O(5A)	1.952(3)	N(1C)-C(3C)	1.322(6)
O(1A)-C(1A)	1.239(6)	N(1C)-C(2C)	1.451(6)
O(2A)-C(1A)	1.285(6)	N(2C)-C(5C)	1.307(6)
O(3A)-C(3A)	1.277(6)	N(2C)-C(6C)	1.455(7)
O(4A)-C(5A)	1.269(6)	C(1C)-C(2C)	1.507(7)
O(4A)-Gd(1)#1	2.327(3)	C(3C)-C(4C)	1.506(7)
O(5A)-C(7A)	1.277(6)	C(4C)-C(5C)	1.511(7)
O(6A)-C(7A)	1.250(6)	C(6C)-C(7C)	1.510(7)
N(1A)-C(3A)	1.296(6)		

Symmetry transformations used to generate equivalent atoms: #1 -x,-y,-z+2 #2 -x+1,-y+1,-z+2
 #3 -x+1,-y,-z+3