Supporting information for

Pair Distribution Function and ^{71}Ga NMR Study of Aqueous Ga^{3+} Complexes

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Data

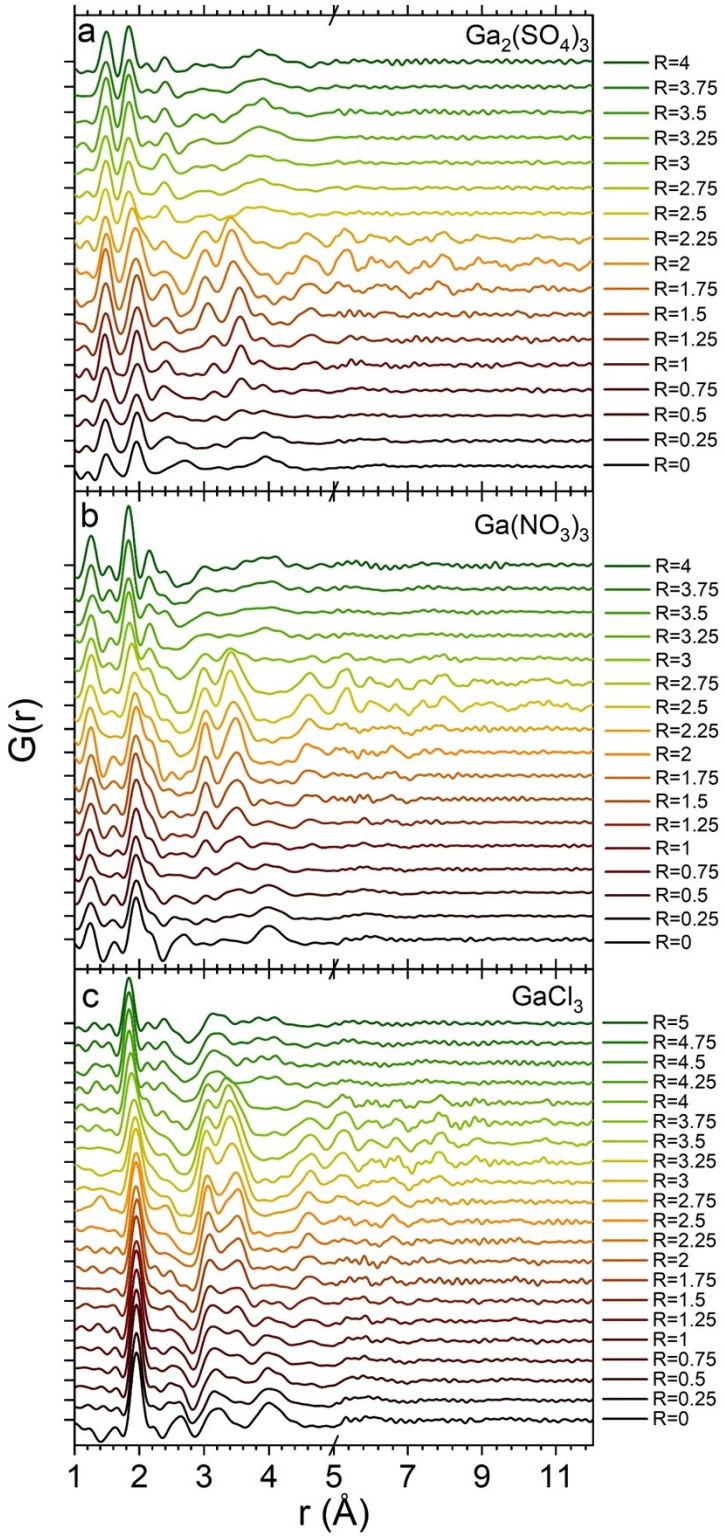

Figure S1. PDFs of the pH series for the three salts a) $Ga_2(SO_4)_3$, b) $Ga(NO_3)_3$ and c) $GaCl_3$. The hydrolysis ratio R denotes the calculated ratio of Ga^{3+} and OH^- ions in the solutions.

Anion signals and ligands

To identify the signals of the gallium structures, we first need to account for the signal of the anions, which are present in the solutions. It is clearly observed that some structural signals change with increasing hydrolysis ratio, R, while other signals are present independent of R (Figure S1). The latter originates form the anions: For the sulfate S-O and O-O distances are observed at 1.48 and 2.40 Å,[1] while for nitrate N-O and O-O distances are observed at 1.21 and 2.19 Å.[2] The chloride anion have previously been reported to induce a broad signal around 3.2 Å, which is ascribed to a Cl-O coordination.[3,4] A comparison showing similar features in the present data and simulated PDFs of the anion signals are presented in Figure S2.

The sulfate and nitrate correlations are narrow and well defined compared the Cl-O signal. This correspond well with the known nature of the anion nature: The bonds in nitrate and sulfate are covalent and thus strong and well defined, while the Cl-O bonding is a weaker solvent coordination, where the distances vary more thus giving a broader signal in the PDF.

Considering the coordination species for all the we must note that Raman spectroscopy has previously shown that anions coordinates to gallium in both first and second shell depending on the concentration and that sulfate bonds stronger than nitrate.[5,6] As first shell coordination of nitrate is at a level of less than 5% at concentrations similar to the ones used here, these effects are expected to give only minor changes in the PDF, they will not be treated further here. Thus, the modelled oxygen is here referred to as water or solvent as this is by far the main component of the coordination shells. Furthermore as the scattering power of hydrogen is very low, we will only use oxygen atoms as a descriptor for the ligated water.

The calculated anion contributions (Figure S2b) was included with refinement of a relative scale factor.

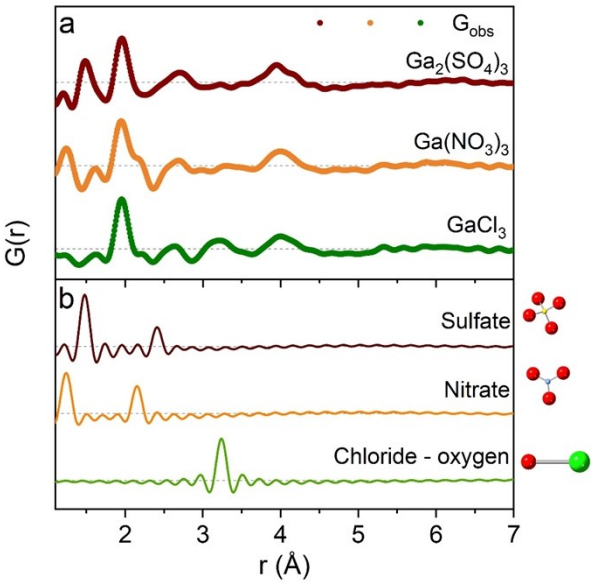

Figure S2. Comparison of a) the experimental PDFs of $Ga_2(SO_4)_3$, $Ga(NO_3)_3$ and $GaCl_3$ with R = 0 to b) the calculated PDFs of the anions SO_4^{2-}, NO_3^- and Cl^-.

Octahedral region

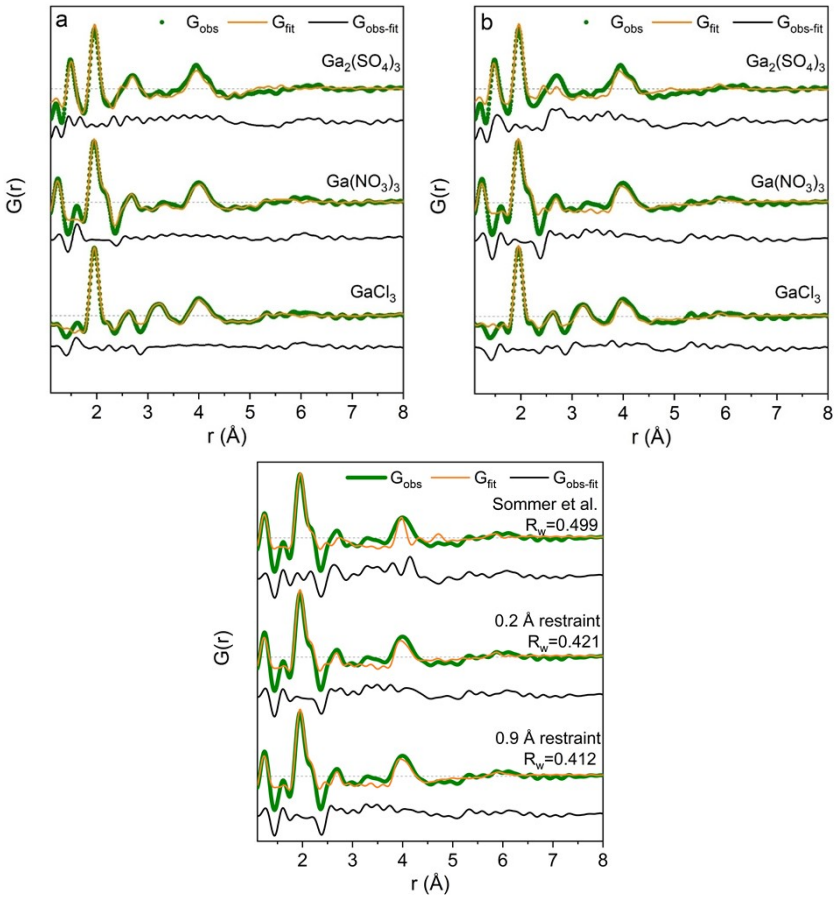

Figure S3. PDFs the three salts with R = 0 fitted with: a) Monomer model with 8 solvent oxygen with refined positions and a box restraint of 0.2 and b) Dimer with refined positions and box restraint of 0.9. c) shows the improvement obtained when relaxing the dimer model relative to the published structure by Sommer *et al.*

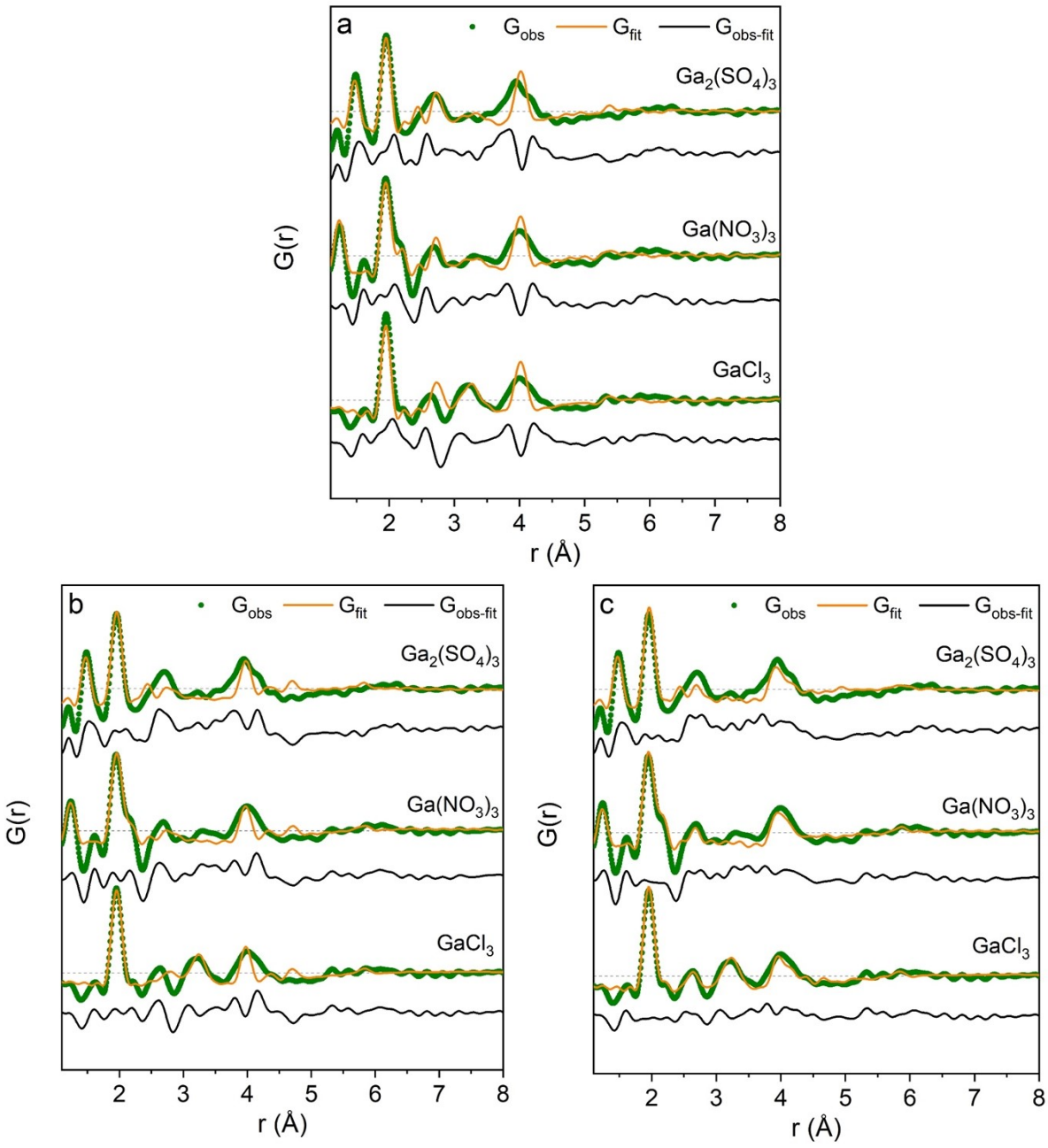

Figure S4. PDFs the three salts with R = 0 fitted with: a) Monomer model with 8 solvent directly from crystal structure[7] b) Dimer as in previous report[8], c) Dimer with refined positions and box restraint of 0.2.

Table S1. R_w and zoom values for the 5 models refined against the R = 0 PDFs of the three salts.

	$Ga(NO_3)_3$		$GaCl_3$		$Ga_2(SO_4)_3$	
	R_w	Zoom	R_w		R_w	
Monomer – zoom	0.460	1.001	0.495	1.003	0.543	1.003
Monomer – 0.2 restraint	0.284	-	0.208	-	0.322	-
Dimer – zoom	0.499	0.998	0.426	1.000	0.550	0.999
Dimer – 0.2 restraint	0.421	-	0.290	-	0.481	-
Dimer – 0.9 restraint	0.412	-	0.278	-	0.458	-

Table S2. Refined models against the three salts. Atomic positions for the original clusters and the refined positions for the three salts.

Original cluster from $Ga(NO_3)_3$ •9 H_2O [7]

Ga	6.980	0.000	0.000
O	6.149	-0.849	-1.529
O	8.803	-2.731	-2.272
O	3.961	-2.345	-1.420
O	6.357	-3.648	1.409
O	8.282	-2.094	3.190
O	5.846	-1.013	1.213
O	8.287	-1.438	0.066
O	7.603	3.648	-1.409
O	5.679	2.094	-3.190
O	8.115	1.013	-1.213
O	5.674	1.438	-0.066
O	7.812	0.849	1.529
O	5.158	2.731	2.272
O	10.000	2.345	1.420

Refined $GaCl_3$ R = 0 restraint 0.2

Ga	6.980000	0.000000	0.000000
O	6.209954	-1.050757	-1.452360
O	8.800810	-2.796772	-2.144862
O	4.025947	-2.491609	-1.229306
O	6.193931	-3.447107	1.381111
O	8.269910	-2.295188	3.391767
O	6.046921	-1.196219	1.228562
O	8.194466	-1.560837	-0.040743
O	7.804901	3.851368	-1.299531
O	5.478878	2.155416	-3.218800
O	7.912718	1.215080	-1.213113
O	5.644019	1.596245	-0.084558
O	7.611003	0.999746	1.465206
O	5.256305	2.931406	2.119614
O	9.875138	2.311488	1.219114

Refined $Ga(NO_3)_3$ R = 0 restraint 0.2

Ga	6.980000	0.000000	0.000000
O	6.084957	-1.050019	-1.516551
O	9.003598	-2.530215	-2.185884
O	3.980794	-2.322978	-1.218328
O	6.334670	-3.437772	1.202755
O	8.349012	-1.966551	3.379313
O	5.978980	-0.986473	1.134986
O	8.268634	-1.390398	-0.119191
O	7.733324	3.478653	-1.203604
O	5.879065	2.019369	-3.257977
O	8.080812	1.025998	-1.066177
O	5.666777	1.386968	-0.059847
O	7.734186	0.966007	1.388094
O	5.142330	2.576249	2.066845
O	9.797840	2.321711	1.214471

Refined $Ga_2(SO_4)_3$ R = 0 restraint 0.2

Ga	6.980000	0.000000	0.000000
O	6.103749	-0.901627	-1.512109
O	8.849986	-2.684628	-2.248716
O	3.885423	-2.313478	-1.482246
O	6.332507	-3.573180	1.209005
O	8.446494	-2.248218	3.283072
O	5.799251	-1.020233	1.249759
O	8.268396	-1.397679	-0.041388
O	7.402800	3.487235	-1.211105
O	5.878805	2.255116	-3.060234
O	8.260430	1.029715	-1.312777
O	5.630186	1.397840	0.017680
O	7.837193	0.846398	1.520427
O	5.090730	2.735466	2.187150
O	10.019824	2.324459	1.494240

Dimer from previous report[8]

Ga	4.470	-7.145	-3.099
Ga	4.022	-3.334	-4.100
O	3.887	-1.663	-4.973
O	3.006	-4.266	-5.470
O	6.292	-6.653	-3.486
O	2.522	-3.260	-2.912
O	4.508	-7.032	-1.176
O	4.874	-9.028	-3.096
O	4.355	-7.220	-5.074
O	5.627	-3.529	-5.265
O	2.571	-7.598	-2.765
O	4.002	-5.182	-3.270
O	5.171	-2.535	-2.697

Refined $Ga(NO_3)_3$ R = 0 restraint 0.2

Ga	4.470000	-7.145000	-3.099000
Ga	3.770564	-3.422481	-4.153727
O	3.758969	-1.733549	-5.099608
O	2.869799	-4.304055	-5.593109
O	6.440868	-6.458519	-3.508959
O	2.476610	-3.470450	-2.710498
O	4.307493	-6.845642	-1.017345
O	4.672023	-9.119847	-2.922847
O	4.377541	-7.371817	-5.014824
O	5.422995	-3.732183	-5.169929
O	2.569133	-7.385725	-2.969619
O	4.146575	-5.256542	-3.472989
O	4.970766	-2.745843	-2.810253

Refined $Ga(NO_3)_3$ R = 0 restraint 0.9

Ga	4.470000	-7.145000	-3.099000
Ga	4.001099	-3.385240	-4.099745
O	4.325116	-1.408365	-4.708365
O	2.938586	-4.370206	-5.648345
O	6.350631	-6.744695	-3.429840
O	2.538986	-3.482725	-2.852740
O	4.622221	-6.577633	-1.270138
O	5.050666	-8.997806	-2.999539
O	4.227352	-7.352531	-4.998583
O	5.516174	-3.806462	-5.221819
O	2.467505	-6.982115	-3.004395
O	4.120963	-5.244789	-3.472752
O	5.207346	-2.980647	-2.652112

Refined $GaCl_3$ R = 0 restraint 0.2

Ga	4.470000	-7.145000	-3.099000
Ga	3.864831	-3.308056	-3.827746
O	4.041315	-1.673639	-4.993074
O	2.874804	-4.062893	-5.319943
O	6.255545	-6.646659	-3.649613
O	2.321865	-3.474466	-2.708864
O	4.307943	-6.822615	-1.258286
O	5.076503	-9.027310	-3.296904
O	4.154691	-7.263633	-5.036578
O	5.487027	-3.732943	-5.185660
O	2.514813	-7.388291	-2.968847
O	4.071409	-5.261987	-3.426872
O	5.372455	-2.571735	-2.857417

Refined $GaCl_3$ R = 0 restraint 0.9

Ga	4.470000	-7.145000	-3.099000
Ga	3.986390	-3.329662	-3.977388
O	3.983127	-1.684615	-5.038929
O	3.023181	-4.123459	-5.480867
O	6.311253	-6.637163	-3.423430
O	2.367171	-3.361542	-3.006062
O	4.168803	-6.540971	-1.204609
O	5.363590	-9.091462	-3.330297
O	4.372463	-7.303968	-5.039922
O	5.544770	-3.744827	-5.160775
O	2.516266	-7.107568	-3.220449
O	4.145982	-5.227489	-3.512942
O	5.334033	-2.521367	-2.891243

Refined $Ga_2(SO_4)_3$ R = 0 restraint 0.2

Ga	4.470000	-7.145000	-3.099000
Ga	3.741583	-3.469782	-4.191678
O	3.855508	-1.482860	-4.833796
O	2.827030	-4.441589	-5.626131
O	6.303367	-6.597956	-3.285973
O	2.468997	-3.460670	-2.711946
O	4.700494	-7.232266	-1.130462
O	4.900906	-9.024661	-3.076732
O	4.394950	-7.341393	-5.060303
O	5.426457	-3.729501	-5.137306
O	2.593433	-7.396991	-2.965280
O	4.124843	-5.261045	-3.457397
O	4.995785	-2.736616	-2.740147

Refined $Ga_2(SO_4)_3$ R = 0 restraint 0.9

Ga	4.470000	-7.145000	-3.099000
Ga	4.019297	-3.401535	-4.052535
O	3.838158	-1.699896	-5.139726
O	2.874622	-4.207509	-5.389773
O	6.311314	-6.592271	-2.756102
O	2.541243	-3.466906	-2.781238
O	3.948497	-6.614080	-1.312174
O	5.250416	-9.071670	-2.948337
O	4.834727	-7.261012	-4.981244
O	5.558376	-3.838892	-5.151391
O	2.473715	-7.082997	-3.577512
O	4.258972	-5.242399	-3.455478
O	5.233675	-3.017075	-2.577154

Table S3. Extracted values of the EXAFS refinements. Structures were used as extracted for the PDF refinements. EXAFS data measured was on 1 M $Ga(NO_3)_3$ in water previously reported. For details on the data see Sommer et al.[8]

	R_w	Amplitude	Energy shift	Expansion	Ss1
Monomer – zoom	0.0290	1.206	9.289	0.00929	0.00595
Monomer – 0.2 restraint	0.0243	1.275	6.689	0.0559	0.00321
Dimer – zoom	0.0118	1.0002	3.151	-0.00059	0.00311 0.01878
Dimer – 0.2 restraint	0.0298	1.1654	5.185	0.007678	0.00373 0.01 (F)
Dimer – 0.9 restraint	0.0306	1.1352	5.074	0.006864	0.00361 0.01 (F)

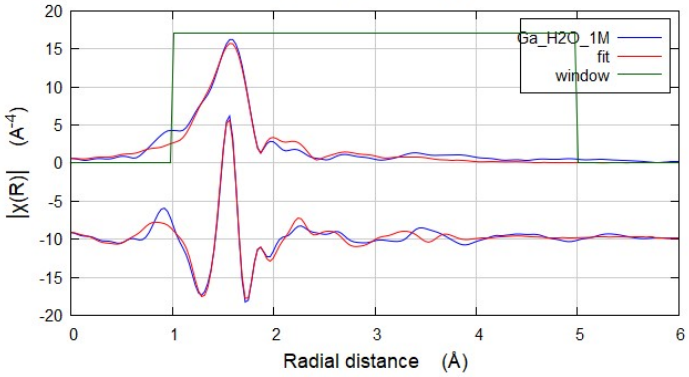

Figure S5. Refinement of EXAFS with the monomer model after refinement with zoomscale

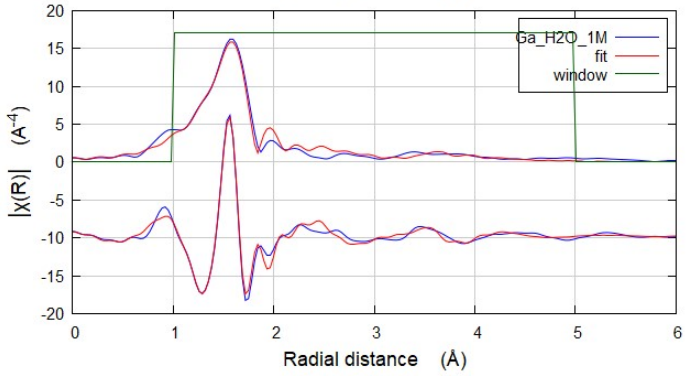

Figure S6. Refinement of EXAFS with of the monomer model after refinement with the 0.2 restraint

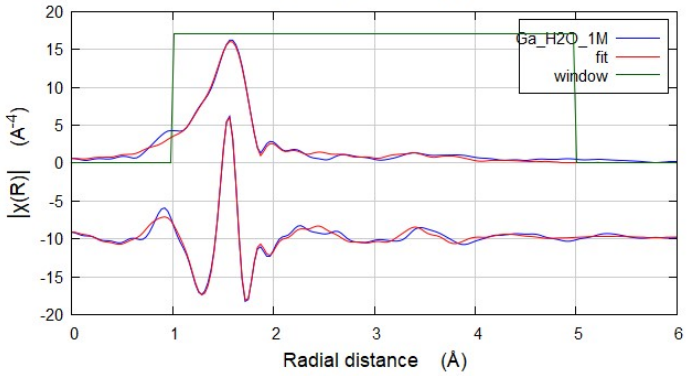

Figure S7. Refinement of EXAFS with the dimer model after refinement with zoomscale

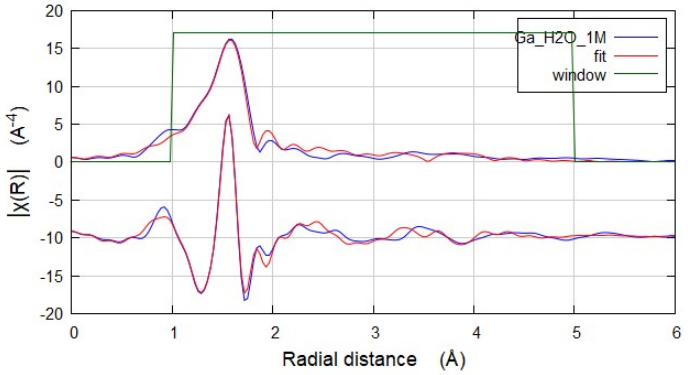

Figure S8. Refinement of EXAFS with the dimer model after refinement with the 0.2 restraint

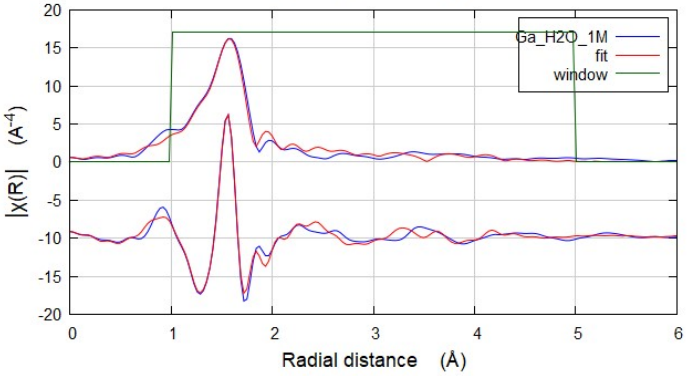

Figure S9. Refinement of EXAFS with the dimer model after refinement with the 0.9 restraint

Polyoxogallate region

Table S4. Nomenclature for reported polyoxogallate structures

Short name	Full structural nomenclautre	
Ga_8	$[Ga_8(heidi)_4(\mu_3\text{-}OH)_2(\mu_2\text{-}OH)_8(H_2O)_4(C_5H_5N)_2]^{2+}$ (H_3heidi = $N(CH_2COOH)_2(CH_2CH_2OH)$	9
Ga_9	$[(MesGaO)_9]$ (Mes = $Me_3C_6H_2$)	10
Ga_{12}	$[Ga_{12}TBu_{12}(\mu_3\text{-}O)_8(\mu\text{-}O)_2(\mu\text{-}OH)_4]$	11
planar Ga_{13}	$[Ga_{13}(\mu_3\text{-}OH)_6(\mu_2\text{-}OH)_{18}(H_2O)_{24}]$	9,12-13
Ga_{30}	$[Ga_{30}(\mu_4\text{-}O)_{12}(\mu_3\text{-}O)_4(\mu_3\text{-}OH)_4(\mu_2\text{-}OH)_{42}(H_2O)_{16}]^{12+}$	14
Ga_{32}	$[Ga_{32}(\mu_4\text{-}O)_{12}(\mu_3\text{-}O)_8(\mu_2\text{-}O)_7(\mu_2\text{-}OH)_{39}(H_2O)_{20}]^{3+}$	12

To identify the structure observed in this study, the atomic positions of all the previously reported gallium clusters and several known polyoxometalate clusters, such as the Keggin, Waugh, Wells-Dawson and Silverton clusters were extracted from single crystal data (ISCD database). For the latter, the metal atoms were substituted with gallium. For the relevant clusters, an additional motif was constructed where the central metal atom was removed. This have previously been reported to match data for Keggin ions for tungsten.[15] All these structures were refined against the PDF of the $Ga(NO_3)_3$ solution with R=2, as an example of the measured PDFs. The structures were allowed to expand or contract isotropically, to account for the elemental substitution as well as the motif being in a solvent and not in a crystal. A list of all the structures, modifications and parameters from the refinements are reported in Table S5.

Table S5. Extracted polyoxometalate clusters, their substitution and refinement parameters for a simple refinement with expansion/contraction.

#	Structure	Motif	Original	Rw	Zoomscale	Ref
1	Ga_8			0.5424	0.9890	[9]
2	Ga_9			0.6659	1.028	[10]
3	Ga_{12}			0.7736	1.053	[11]
4	Ga_{13}	Flat		0.6043 0.6577 0.6667	0.9806 0.9866 0.9852	[9] [12] [13]
5	Ga_{30}	Keggin-like		**0.3658**	1.002	[14]
6	Ga_{32}	Keggin-like		**0.3776**	1.001	[12]
7	Ga_{13}	α-Keggin	Al_{13} modified from #9	**0.5594**	1.034	
8	Ga_{13}	β-Keggin	Al_{13} modified from #9	**0.4566**	1.034	
9	Ga_{13}	γ-Keggin	Al_{13}	**0.3702**	1.034	[16]
10	Ga_{13}	δ-Keggin	Al_{13}	**0.3992**	1.032	[17]
11	Ga_{13}	ε-Keggin	Al_{13}	**0.6191**	1.036	[18]
12	Ga_{12}	α-Keggin, no center	Al_{12} modified from #7	0.7198	1.039	
13	Ga_{12}	β-Keggin, no center	Al_{12} modified from #8	0.6497	1.027	
14	Ga_{12}	γ-Keggin, no center	Al_{12} modified from #9	0.5793	1.028	
15	Ga_{12}	δ-Keggin, no center	Al_{12} modified from #10	0.6125	1.030	
16	Ga_{12}	ε-Keggin, no center	Al_{12} modified from #11	0.7659	1.039	
17	Ga_{20}	Wells-Dawson	P_2W_{18}	0.8685	0.9947	[19]
18	Ga_{18}	Wells-Dawson, no center	W_{18} modified from #17	0.8992	0.9423	
19	Ga_{13}	Silverton	$CeMo_{12}$	0.8876	0.9030	[20]
20	Ga_{12}	Silverton, no center	Mo_{12} modified from #19	0.8871	0.9097	
21	Ga_{10}	Waugh	$CoMo_9$	0.8872	0.9414	[21]
22	Ga_9	Waugh, no center	Mo_9 modified from #21	0.8983	0.9290	
23	Ga_7	Anderson	$GaMo_6$	0.8245	1.039	[22]
24	Ga_6	Anderson, no center	Mo_6 modified from #23	0.8863	1.043	
25	Ga_6	Lindqvist	Ta_6	0.8000	1.018	[23]

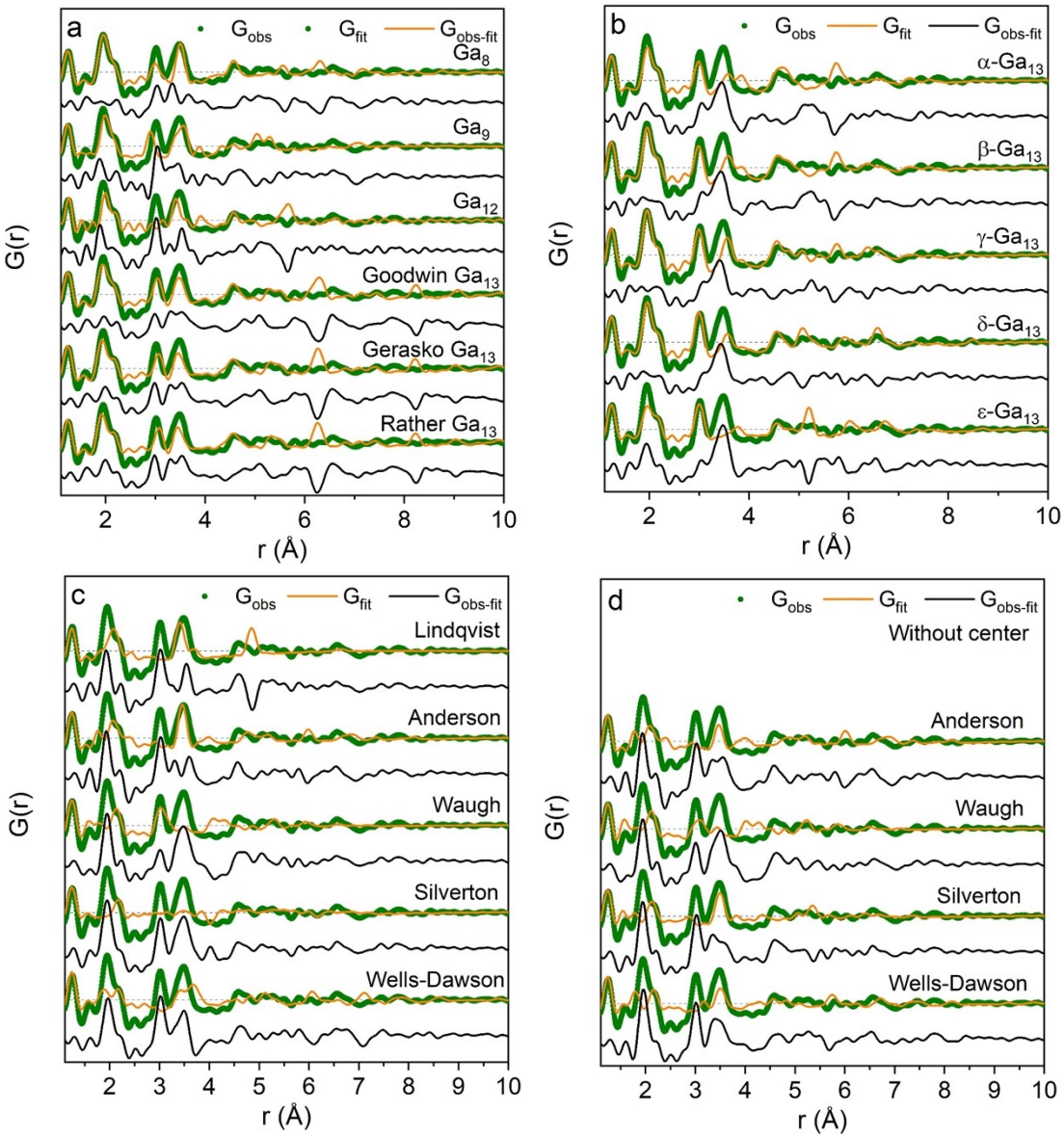

Figure S10. Fits of the most common polyoxometalate clusters to the $Ga(NO_3)_3$ R = 2 data. The refinements includes the nitrate ion to describe the peaks at 1.6 and 2.2 Å. a) Structures #1-4, b) Structures #12-16. c) Structures #17, 19, 21 23 and 25 and d) Structures #18, 20, 22 and 24.

Table S6. Atomic positions of the five Ga_{13} Keggin isomers, the Ga_{30} and Ga_{32} structures used for refinements.

α-Ga_{13}

Ga	-16.618	10.371	24.567
Ga	-19.612	10.683	23.302
Ga	-18.446	13.064	24.435
Ga	-17.450	12.074	21.905
Ga	-17.805	7.678	23.146
Ga	-17.717	11.254	27.447
Ga	-14.154	11.898	23.138
Ga	-15.605	9.045	21.751
Ga	-18.919	8.898	26.222
Ga	-16.277	8.695	27.532
Ga	-14.901	13.011	25.744
Ga	-13.577	10.411	25.620
Ga	-15.096	7.468	24.146
O	-17.808	11.378	23.707
O	-17.253	9.930	26.156
O	-16.335	8.918	23.574
O	-15.095	11.263	24.766
O	-20.150	12.421	23.872
O	-18.193	13.684	22.659
O	-19.167	11.401	21.623
O	-18.756	12.296	26.102
O	-19.839	10.126	25.063
O	-19.027	9.013	22.741
O	-16.708	10.491	21.232
O	-15.791	12.722	22.428
O	-16.646	13.607	24.773
O	-16.766	10.128	28.576
O	-19.304	10.203	27.548
O	-18.354	7.634	24.921
O	-16.999	7.810	21.405
O	-14.251	10.162	22.291
O	-12.707	11.125	24.077
O	-13.975	13.486	24.171
O	-17.967	7.845	27.532
O	-15.852	7.601	25.891
O	-16.546	6.365	23.572
O	-14.604	7.552	22.321
O	-13.800	8.800	24.548
O	-13.461	12.107	26.430
O	-14.660	9.971	27.250
O	-15.760	12.048	27.446
O	-19.151	14.712	25.016
O	-17.353	12.907	20.179
O	-21.382	10.181	22.830
O	-13.055	12.702	21.777
O	-14.943	8.932	19.948
O	-19.081	6.390	22.540
O	-18.276	12.341	28.930
O	-11.990	9.876	26.507
O	-14.075	6.043	24.634
O	-20.505	7.898	26.564
O	-15.688	7.631	28.950
O	-14.427	14.620	26.67

β-Ga_{13}

Ga	12.177	-3.639	28.735
Ga	10.240	-5.927	30.029
Ga	10.378	-3.418	31.442
Ga	8.917	-3.551	28.947
Ga	11.933	-6.391	26.990
Ga	13.961	-3.609	31.400
Ga	10.874	-0.732	29.233
Ga	10.633	-4.001	25.872
Ga	13.725	-6.142	29.977
Ga	15.565	-4.056	28.980
Ga	13.804	-0.727	29.165
Ga	12.311	-1.148	26.690
Ga	13.529	-4.293	25.792
O	10.693	-4.046	29.629
O	13.586	-4.217	29.631
O	12.073	-4.431	27.142
O	12.295	-1.880	28.519
O	9.791	-5.207	31.737
O	8.636	-3.048	30.787
O	8.581	-5.336	29.374
O	12.082	-3.935	31.981
O	11.947	-6.238	30.702
O	10.677	-6.567	28.342
O	9.255	-4.029	27.169
O	9.458	-1.798	28.662
O	10.843	-1.642	30.914
O	15.674	-3.430	30.706
O	14.158	-5.480	31.704
O	13.337	-6.636	28.184
O	10.579	-5.888	25.721
O	10.897	-2.194	26.071
O	11.006	-0.008	27.492
O	12.362	0.321	29.782
O	15.558	-5.895	29.421
O	14.776	-4.593	27.203
O	13.206	-6.169	25.640
O	12.051	-4.074	24.632
O	13.562	-2.416	26.090
O	13.663	-0.082	27.454
O	15.117	-2.044	28.709
O	14.058	-1.597	30.767
O	9.940	-2.882	33.195
O	7.082	-3.167	28.544
O	9.649	-7.669	30.498
O	9.702	0.645	29.895
O	9.305	-3.768	24.500
O	11.710	-8.250	26.604
O	14.589	-3.199	33.170
O	12.453	-0.172	25.071
O	14.850	-4.319	24.541
O	14.103	-7.953	30.436
O	17.410	-4.086	28.682
O	15.131	0.579	29.626

γ-Ga_{13}

Ga	1.644	-31.017	4.741
Ga	3.632	-31.985	2.339
Ga	1.011	-33.185	2.394
Ga	1.283	-30.431	1.547
Ga	4.531	-29.490	4.637
Ga	2.996	-33.928	5.115
Ga	-1.391	-30.953	3.654
Ga	2.173	-27.905	3.877
Ga	3.740	-31.839	7.007
Ga	1.141	-33.220	7.280
Ga	-1.161	-31.591	6.505
Ga	-0.609	-28.872	5.594
Ga	2.696	-28.547	6.669
O	1.842	-31.557	3.057
O	2.236	-32.269	5.839
O	2.583	-29.514	4.932
O	-0.070	-30.696	5.081
O	2.710	-33.472	1.579
O	0.576	-32.102	0.897
O	2.984	-30.905	0.945
O	1.602	-34.158	3.866
O	4.005	-33.072	3.803
O	4.472	-30.510	3.090
O	1.963	-28.830	2.240
O	-0.385	-30.145	2.311
O	-0.661	-32.644	3.143
O	1.899	-34.660	6.422
O	4.381	-33.504	6.354
O	4.762	-31.002	5.694
O	4.056	-27.896	3.672
O	0.381	-28.016	4.266
O	-1.922	-29.242	4.258
O	-2.375	-31.757	5.071
O	2.659	-32.765	8.313
O	2.947	-30.236	7.517
O	4.547	-28.473	6.205
O	2.481	-27.052	5.530
O	0.806	-28.694	6.818
O	-1.639	-29.851	6.830
O	0.293	-31.571	7.751
O	-0.441	-33.272	6.292
O	0.268	-34.732	1.616
O	0.800	-29.568	-0.097
O	5.267	-32.436	1.485
O	-2.914	-31.201	2.503
O	1.978	-26.239	2.934
O	6.397	-29.212	4.327
O	3.776	-35.646	4.747
O	-1.402	-27.262	6.205
O	2.951	-27.657	8.235
O	5.140	-31.657	8.288
O	0.419	-34.210	8.691
O	-2.401	-32.252	7.812

δ-Ga_{13}

Ga	9.446	-1.543	-20.830	O	8.329	0.541	-18.141	O	12.611	-3.961	-15.279
Ga	13.734	-3.865	-16.773	O	13.353	-0.951	-18.797	O	14.382	-2.174	-16.330
Ga	15.091	-1.585	-17.947	O	10.272	1.687	-16.839	O	9.436	2.764	-19.232
Ga	11.515	-5.154	-18.216	O	12.649	-0.176	-21.464	O	10.577	-5.136	-16.609
Ga	10.945	-3.389	-15.949	O	8.545	-1.878	-19.198	O	6.875	-1.474	-17.013
Ga	12.092	-1.883	-22.004	O	11.662	-3.671	-22.309	O	13.972	2.670	-17.535
Ga	14.232	-0.545	-20.560	O	11.881	1.486	-18.937	O	15.117	0.203	-17.425
Ga	10.082	1.054	-18.638	O	13.839	-2.163	-21.390	O	11.068	0.836	-14.299
Ga	13.503	0.886	-18.125	O	9.049	-0.345	-15.832	O	11.275	-7.087	-18.452
Ga	10.688	-4.202	-20.809	O	10.611	-0.655	-17.853	O	10.097	-5.799	-21.710
Ga	8.726	-1.200	-17.468	O	10.001	-4.669	-19.158	O	10.359	-1.313	-22.469
Ga	11.787	-1.820	-18.631	O	14.370	1.151	-19.780	O	7.778	-0.960	-21.689
Ga	10.880	0.074	-16.053	O	9.112	-3.346	-21.305	O	15.190	-4.737	-15.847
O	11.145	-2.271	-20.252	O	15.724	-1.206	-19.653	O	10.044	-3.799	-14.269
O	11.348	-1.618	-15.397	O	13.196	-5.466	-17.502	O	15.401	0.079	-21.969
O	9.298	-2.806	-16.708	O	12.625	0.562	-16.492	O	12.714	-1.683	-23.817
O	11.989	-3.276	-17.603	O	14.717	-3.368	-18.263	O	16.925	-1.881	-17.361
O	9.823	0.251	-20.335	O	12.162	-4.975	-19.960				

ε-Ga_{13}

Ga	-18.015	-2.072	-3.619	O	-17.312	3.117	-3.602	O	-16.738	-4.535	-0.547
Ga	-21.249	-1.621	-2.816	O	-16.683	-6.965	-4.903	O	-20.265	-3.460	-5.834
Ga	-20.700	-1.664	-5.702	O	-20.554	2.480	-4.200	O	-21.451	0.240	-2.987
Ga	-19.965	0.654	-4.076	O	-18.189	-5.189	-6.342	O	-16.208	-4.852	-3.189
Ga	-17.212	1.185	-3.513	O	-14.565	0.092	-0.174	O	-15.476	-2.252	-1.273
Ga	-15.730	-0.463	-1.650	O	-18.109	-3.074	-2.077	O	-14.834	-2.417	-4.535
Ga	-15.103	-0.556	-4.496	O	-15.722	1.188	-4.588	O	-20.220	-1.459	-1.240
Ga	-17.032	-2.714	-0.401	O	-16.208	-2.691	1.355	O	-14.268	-2.854	-7.261
Ga	-17.690	-5.034	-2.032	O	-21.263	-3.673	0.170	O	-16.281	1.312	-1.879
Ga	-19.796	-3.288	-1.008	O	-18.381	1.013	-4.930	O	-18.679	-2.950	0.426
Ga	-17.104	-5.076	-4.828	O	-22.929	-1.724	-1.887	O	-18.848	0.944	-2.618
Ga	-15.808	-2.835	-6.090	O	-18.479	-5.441	-3.649	O	-19.657	-1.338	-3.972
Ga	-18.621	-3.392	-6.702	O	-20.930	-3.385	-2.450	O	-19.294	-5.090	-1.080
O	-17.505	-3.133	-5.009	O	-15.888	-1.002	-6.101	O	-16.792	-0.759	-3.354
O	-13.429	-0.120	-5.367	O	-14.321	-0.318	-2.833	O	-19.196	-1.639	-6.773
O	-21.987	-1.692	-7.138	O	-15.619	-4.662	-5.829	O	-22.050	-1.953	-4.463
O	-19.402	-3.881	-8.383	O	-17.209	-0.848	-0.549	O	-17.294	-6.888	-1.720
O	-16.990	-3.075	-7.468	O	-20.864	0.179	-5.618				

Ga_{30}

Ga	13.550	-10.294	5.850	O	13.847	-13.383	7.127	O	15.825	-10.461	10.575
Ga	10.164	-11.487	6.094	O	11.940	-15.252	6.206	O	9.990	-16.272	8.034
Ga	12.032	-11.890	3.179	O	14.300	-14.209	4.442	O	14.798	-7.440	11.126
Ga	12.791	-13.682	5.513	O	12.171	-6.819	10.523	O	7.009	-15.209	4.277
Ga	13.646	-7.601	9.500	O	10.331	-8.804	9.660	Ga	9.182	-8.568	6.626
Ga	7.744	-13.647	5.220	O	8.593	-13.006	3.616	Ga	12.568	-7.375	6.382
Ga	15.850	-8.074	6.960	O	11.633	-13.551	4.012	Ga	10.701	-6.972	9.297
Ga	16.871	-10.825	6.342	O	9.277	-14.669	5.841	Ga	9.941	-5.180	6.963
Ga	12.563	-12.558	8.320	O	11.829	-14.280	8.816	Ga	9.086	-11.260	2.976
Ga	11.579	-9.753	8.561	O	16.514	-6.405	7.671	Ga	14.988	-5.215	7.256
Ga	14.937	-12.452	3.788	O	6.836	-14.002	6.910	Ga	6.882	-10.788	5.516
Ga	7.571	-12.604	8.010	O	13.623	-12.549	2.292	Ga	5.861	-8.036	6.134
Ga	16.151	-9.134	4.000	O	17.407	-9.181	7.251	Ga	10.170	-6.304	4.156
Ga	14.597	-10.440	9.005	O	16.184	-12.294	5.271	Ga	11.154	-9.108	3.915
Ga	10.505	-14.516	7.327	O	17.669	-10.066	4.738	Ga	7.795	-6.409	8.688
O	10.320	-9.941	7.104	O	15.870	-11.387	7.892	Ga	15.162	-6.258	4.466
O	15.284	-9.631	5.782	O	13.938	-12.185	9.600	Ga	6.581	-9.727	8.476
O	13.332	-11.797	4.808	O	11.237	-11.511	9.275	Ga	8.135	-8.422	3.471
O	13.093	-10.677	7.617	O	7.962	-10.983	8.979	Ga	12.227	-4.345	5.149
O	12.166	-7.900	8.112	O	15.392	-10.669	3.145	O	12.412	-8.921	5.372
O	11.330	-12.783	6.704	O	9.202	-13.522	8.404	O	7.448	-9.230	6.694
O	8.381	-11.984	6.227	O	6.636	-13.376	9.601	O	9.400	-7.065	7.668
O	13.082	-9.506	9.974	O	18.521	-11.788	6.836	O	9.639	-8.185	4.859
O	16.612	-7.505	5.112	O	16.320	-13.345	2.680	O	10.566	-10.962	4.364
O	14.932	-8.626	8.513	O	17.281	-8.646	2.448	O	11.402	-6.079	5.772

O	14.351	-6.878	6.249	O	13.455	-4.192	6.635	O	14.770	-7.878	3.497
O	9.650	-9.356	2.502	O	10.903	-4.581	3.660	O	7.340	-8.193	9.331
O	6.120	-11.357	7.364	O	6.218	-12.457	4.805	O	13.531	-5.339	4.072
O	7.801	-10.236	3.963	O	15.896	-4.860	5.566	O	16.097	-5.485	2.875
O	8.885	-5.478	5.349	O	9.109	-6.313	10.184	O	4.211	-7.073	5.640
O	10.792	-3.610	6.270	O	5.325	-9.680	5.225	O	6.412	-5.516	9.796
O	8.433	-4.652	8.034	O	6.549	-6.567	7.205	O	5.451	-10.216	10.028
O	10.561	-12.042	1.953	O	5.063	-8.796	7.738	O	6.907	-8.400	1.901
O	12.401	-10.058	2.816	O	6.862	-7.475	4.584	O	12.742	-2.590	4.442
O	14.139	-5.855	8.860	O	8.794	-6.677	2.876	O	7.934	-11.422	1.350
O	11.099	-5.311	8.464	O	11.495	-7.351	3.201	O	15.723	-3.652	8.199

Ga_{32}

Ga	18.640	9.625	-6.403	O	23.474	12.077	-6.027	Ga	18.316	13.598	-1.498
Ga	21.560	7.794	-5.859	Ga	19.735	10.942	-1.143	Ga	20.476	15.525	-5.751
Ga	20.578	9.557	-3.631	Ga	18.050	8.544	-2.043	Ga	23.535	14.025	-5.462
Ga	21.188	8.895	-8.659	O	21.323	10.186	-1.936	Ga	22.714	15.604	-3.033
Ga	19.027	6.968	-4.406	O	18.034	11.834	-0.720	Ga	21.962	17.406	-8.275
Ga	15.968	8.468	-4.694	O	19.082	9.238	-0.521	O	20.372	18.019	-9.329
Ga	16.789	6.889	-7.124	O	16.484	9.565	-1.483	O	23.590	16.956	-7.198
Ga	17.541	5.087	-1.882	O	14.702	8.339	-3.239	O	23.098	17.997	-9.782
O	19.131	4.474	-0.828	O	18.872	10.299	-2.898	O	22.231	19.314	-7.632
O	15.913	5.537	-2.958	Ga	24.065	12.983	-2.529	O	23.153	17.416	-2.431
O	16.405	4.495	-0.374	Ga	21.183	13.512	-0.516	O	16.184	15.237	-3.736
O	17.272	3.179	-2.524	O	21.968	14.187	1.156	O	17.327	13.845	-5.931
O	16.351	5.076	-7.725	O	25.679	12.547	-1.471	O	18.715	15.167	-2.581
O	23.319	7.256	-6.421	O	20.718	11.750	0.320	O	18.721	16.212	-5.186
O	22.176	8.648	-4.225	O	22.833	12.678	-1.092	O	21.165	16.163	-4.043
O	20.788	7.326	-7.576	O	24.274	14.853	-2.175	O	20.846	17.125	-6.769
O	20.782	6.280	-4.971	O	25.070	13.230	-4.284	O	21.892	15.629	-9.010
O	18.338	6.330	-6.113	O	22.527	13.700	-3.721	O	19.365	14.148	0.009
O	18.657	5.368	-3.388	O	20.122	12.732	-2.070	O	16.521	14.215	-1.208
O	17.611	6.864	-1.146	Ga	15.438	9.510	-7.628	O	21.582	15.173	-1.465
O	20.138	8.345	-10.165	Ga	18.320	8.981	-9.640	O	23.799	15.816	-4.702
O	22.982	8.278	-8.948	O	17.535	8.306	-11.312	O	22.002	14.564	-6.411
O	17.921	7.320	-8.691	O	13.825	9.946	-8.685	O	19.660	14.620	-7.378
O	15.704	6.677	-5.454	O	18.785	10.742	-10.477	O	19.724	13.785	-4.891
O	17.501	7.929	-3.746	O	16.670	9.815	-9.065	O	17.454	12.988	-3.261
O	19.843	7.873	-2.778	O	15.229	7.640	-7.982	Ga	17.665	11.283	-3.933
O	19.779	8.708	-5.266	O	14.433	9.263	-5.872	Ga	15.553	13.610	-2.811
O	22.049	9.505	-6.896	O	16.976	8.793	-6.435	Ga	14.490	11.216	-5.212
Ga	21.839	11.210	-6.224	O	19.381	9.761	-8.087	Ga	14.888	10.120	-2.446
Ga	23.950	8.883	-7.346	Ga	19.768	11.551	-9.014	O	13.731	9.667	-0.885
Ga	25.013	11.276	-4.945	Ga	21.454	13.949	-8.114	O	13.877	14.468	-2.248
Ga	24.615	12.373	-7.711	O	18.181	12.307	-8.220	O	12.902	11.773	-6.252
O	25.772	12.826	-9.272	O	21.470	10.659	-9.437	O	15.072	11.943	-1.917
O	25.626	8.025	-7.909	O	20.421	13.255	-9.636	O	15.524	11.310	-6.867
O	26.601	10.720	-3.904	O	23.019	12.928	-8.674	O	14.818	12.948	-4.510
O	24.432	10.550	-8.239	O	24.801	14.154	-6.918	O	13.424	10.703	-3.656
O	23.980	11.183	-3.289	O	20.631	12.194	-7.259	O	18.377	11.289	-5.641
O	24.685	9.545	-5.646	Ga	20.863	12.868	-3.754	O	16.029	10.416	-4.130
O	26.079	11.789	-6.500	Ga	17.943	14.699	-4.297				
O	21.126	11.204	-4.515	Ga	18.925	12.935	-6.526				

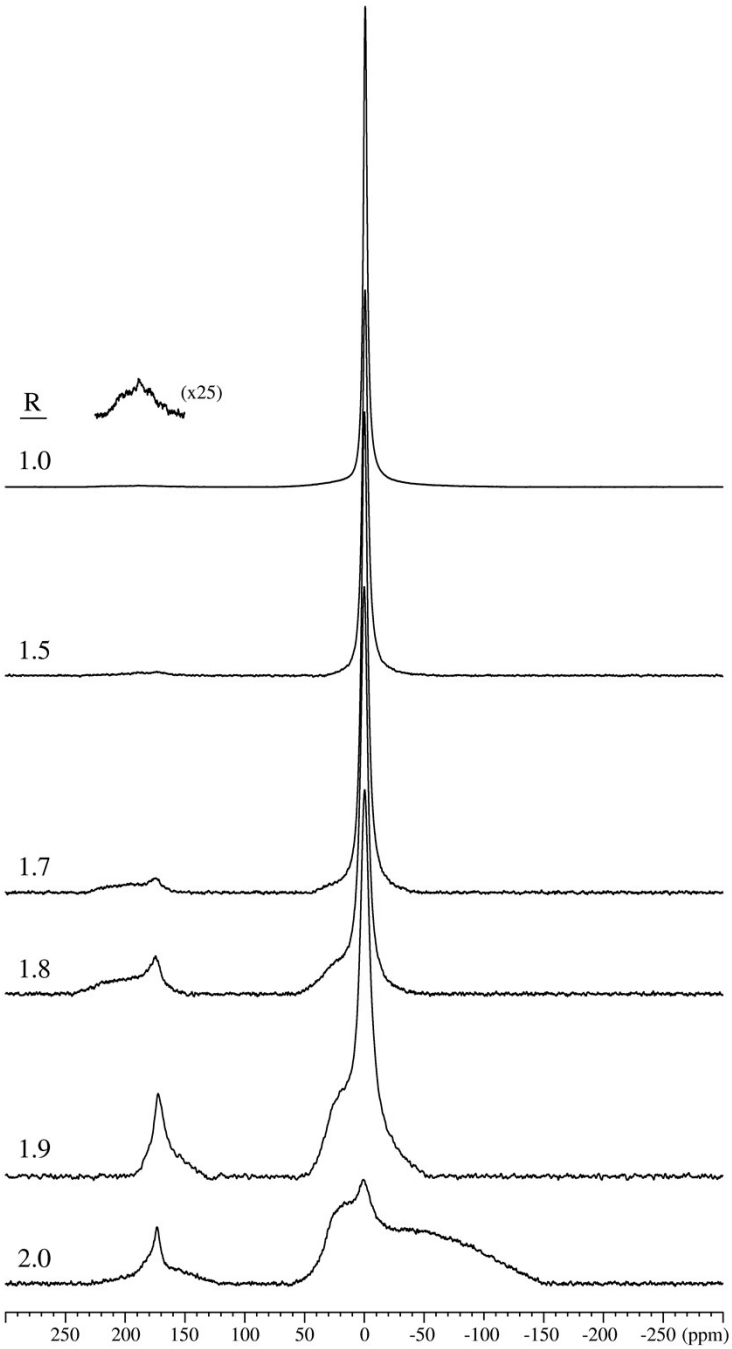

Figure S11. Single-pulse ^{71}Ga NMR spectra (14.09 T, $\nu_L(^{71}Ga)$ = 182.8 MHz) for the $Ga(NO_3)_3$ solutions in D_2O at different R-values (R = $[OH^-]/[Ga^{3+}]$). The spectra were acquired overnight in a 4 mm CP/MAS NMR probe using a 45° excitation pulse ($\gamma B_1/2\pi$ = 60 kHz), a relaxation delay of 1 s and typically 65.000 scans. The linebroadening and asymmetric lineshapes observed at increasing R values are tentatively ascribed to residual quadrupolar broadening caused by reduced motional averaging of the second-order quadrupolar interaction for the larger polyoxygallate complexes in solution.

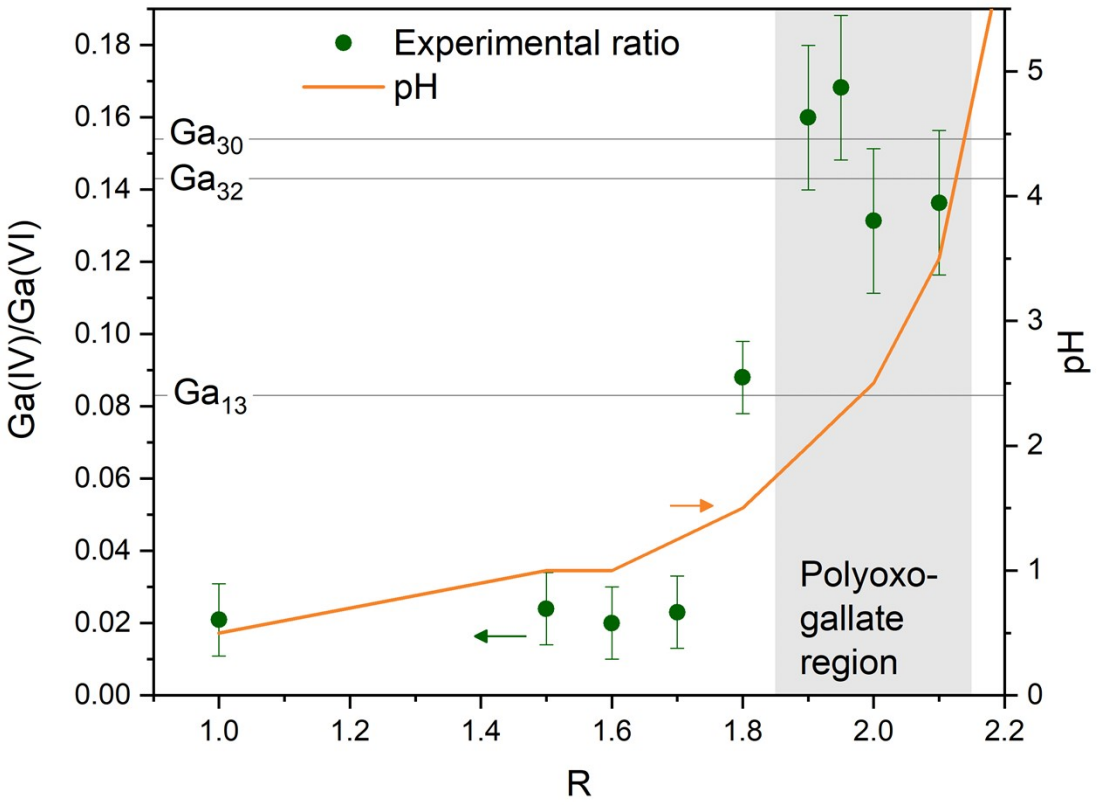

Figure S12. Ratio of tetrahedrally coordinated Ga^{3+} to octahedrally coordinated Ga^{3+} and pH as a function of R-value. ($R = [OH^-]/[Ga^{3+}]$).

$$13\ [Ga(OH)(H_2O)_5]^{2+} + 19\ OH^- \rightarrow [GaO_4Ga_{12}(OH)_{24}(H_2O)_{12}]^{7+} + 57\ H_2O$$

Reaction S1. Formation of Ga_{13} from octahedral coordinated Ga^{3+}

$$32\ [Ga(OH)(H_2O)_5]^{2+} + 61\ OH^- \rightarrow [Ga_{32}O_{27}(OH)_{39}(H_2O)_{20}]^{3+} + 167\ H_2O$$

Reaction S2. Formation of Ga_{32} from octahedral coordinated Ga^{3+}

GaOOH region

$[GaO_4Ga_{12}(OH)_{12}(H_2O)_{24}]^{7+} + 7\ OH^- \rightarrow 13\ GaOOH + 21\ H_2O$

Reaction S3. Formation of GaOOH from the Ga_{13}

$[Ga_{32}O_{27}(OH)_{39}(H_2O)_{20}]^{3+} + 3\ OH^- \rightarrow 32\ GaOOH + 25\ H_2O$

Reaction S4. Formation of GaOOH from the Ga_{32}

Table S7. Refinement values for the $Ga(NO_3)_3$ R = 2.5 data of single phase GaOOH (Model #1-4)

		#1	#2	#3	#4
	GaOOH[24]	Only unit cell	Unit cell and size	Unit cell and atomic positions	Unit cell, atomic positions and size
a (Å)	9.7907(8)	9.8384	10.2967	10.0096	9.5144
b (Å)	2.9732(2)	2.9732	2.9904	2.9762	2.9700
c (Å)	4.5171(4)	4.5073	4.1930	4.3215	4.6040
Size (Å)		24 (F)	9.31	24 (F)	10.14
Ga (x)	0.35544(9)	-	-	0.3569	0.3589
Ga (y)	0.25	-	-	-	-
Ga (z)	0.5517(2)	-	-	0.6337	0.5369
O1 (x)	0.1953(6)	-	-	0.1707	0.1587
O1 (y)	0.25	-	-	-	-
O1 (z)	0.3014(13)	-	-	0.5059	0.3812
O2 (x)	0.4447(7)	-	-	0.4560	0.4472
O2 (y)	0.75	-	-	-	-
O2 (z)	0.3030(15)	-	-	0.3985	0.3576
Rw		0.644	0.497	0.486	0.407

Table S8. Refinement values for the $Ga(NO_3)_3$ R = 2.5 data of two phase GaOOH (Model #5-7)

	GaOOH[24]	#5		#6		#7	
		Phase 1	Phase 2	Phase 1	Phase 2	Phase 1	Phase 2
Relative scale		0.1547		0.1547		0.2340	0.7660
a (Å)	9.7907(8)	-	-	9.8040	10.4009	9.7975	10.3356
b (Å)	2.9732(2)	-	-	2.9624	3.0053	2.9705	2.9532
c (Å)	4.5171(4)	-	-	4.5449	4.1320	4.6249	4.1267
Size (Å)		43.77	8.10	43.77	8.10	28.98	10.10
Ga (x)	0.35544(9)	-	-	-	-	0.3579	0.3615
Ga (y)	0.25	-	-	-	-	-	-
Ga (z)	0.5517(2)	-	-	-	-	0.5389	0.6281
O1 (x)	0.1953(6)	-	-	-	-	0.1911	0.1663
O1 (y)	0.25	-	-	-	-	-	-
O1 (z)	0.3014(13)	-	-	-	-	0.2397	0.4170
O2 (x)	0.4447(7)	-	-	-	-	0.4498	0.4294
O2 (y)	0.75	-	-	-	-	-	-
O2 (z)	0.3030(15)	-	-	-	-	0.3642	0.3409
Rw		0.431		0.431		0.307	

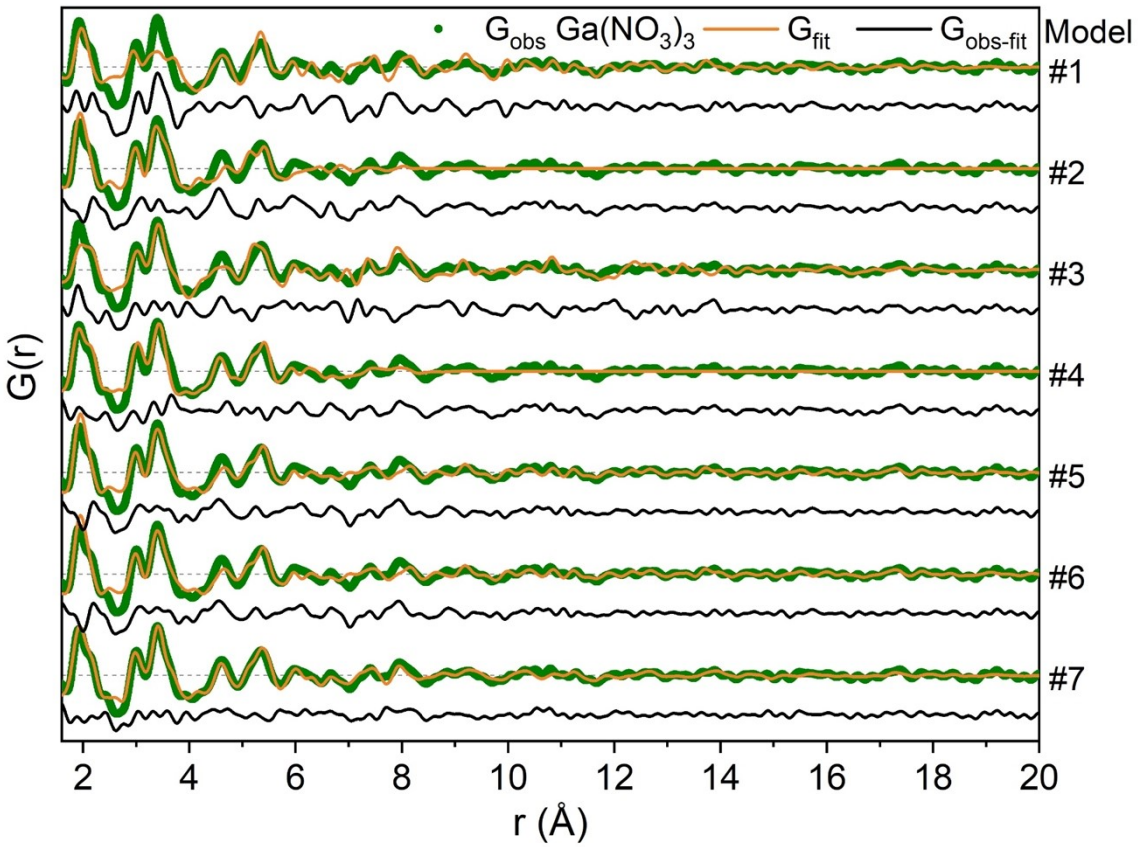

Figure S13. Fits for the $Ga(NO_3)_3$ R = 2.5 data of both single (Model #1-4) and two (Model #5-7) phase GaOOH

Table S9. Refinement values for the $Ga_2(SO_4)_3$ R = 2, $Ga(NO_3)_3$ R = 2.5 and $GaCl_3$ R = 3.5 data using one of the two phase GaOOH models (Model #7).

	GaOOH[24]	Phase 1			Phase 2		
		$Ga_2(SO_4)_3$	$Ga(NO_3)_3$	$GaCl_3$	$Ga_2(SO_4)_3$	$Ga(NO_3)_3$	$GaCl_3$
Relative scale		0.2076	0.2340	0.2689	0.7924	0.7660	0.7311
a (Å)	9.7907(8)	9.5467	9.7975	9.0606	10.3134	10.3356	10.2109
b (Å)	2.9732(2)	2.9785	2.9705	2.9660	2.9917	2.9532	3.0141
c (Å)	4.5171(4)	4.9157	4.6249	5.0321	4.0793	4.1267	4.0695
Size (Å)		24.77	28.98	15.00	10.48	10.10	8.63
Ga (x)	0.35544(9)	0.3610	0.3579	0.3550	0.3615	0.3615	0.3626
Ga (y)	0.25	-	-	-	-	-	-
Ga (z)	0.5517(2)	0.5340	0.5389	0.5887	0.6098	0.6281	0.6098
O1 (x)	0.1953(6)	0.2077	0.1911	0.2008	0.1418	0.1663	0.14218
O1 (y)	0.25	-	-	-	-	-	-
O1 (z)	0.3014(13)	0.2366	0.2397	0.2600	0.4227	0.4170	0.4228
O2 (x)	0.4447(7)	0.4451	0.4498	0.4792	0.4282	0.4294	0.4223
O2 (y)	0.75	-	-	-	-	-	-
O2 (z)	0.3030(15)	0.3905	0.3642	0.4880	0.3575	0.3409	0.3539
Rw		0.346	0.307	0.198			

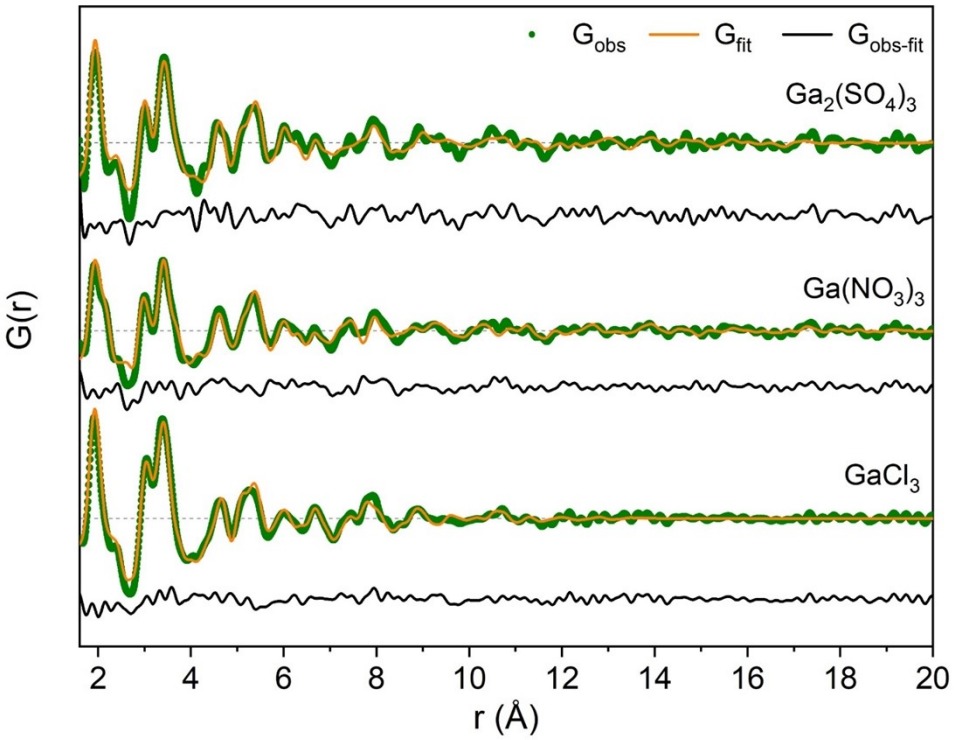

Figure S14. Fits for the $Ga_2(SO_4)_3$ R = 2, $Ga(NO_3)_3$ R = 2.5 and $GaCl_3$ R = 3.5 data using one of the two phase GaOOH models (Model #7).

Tetrahedral region

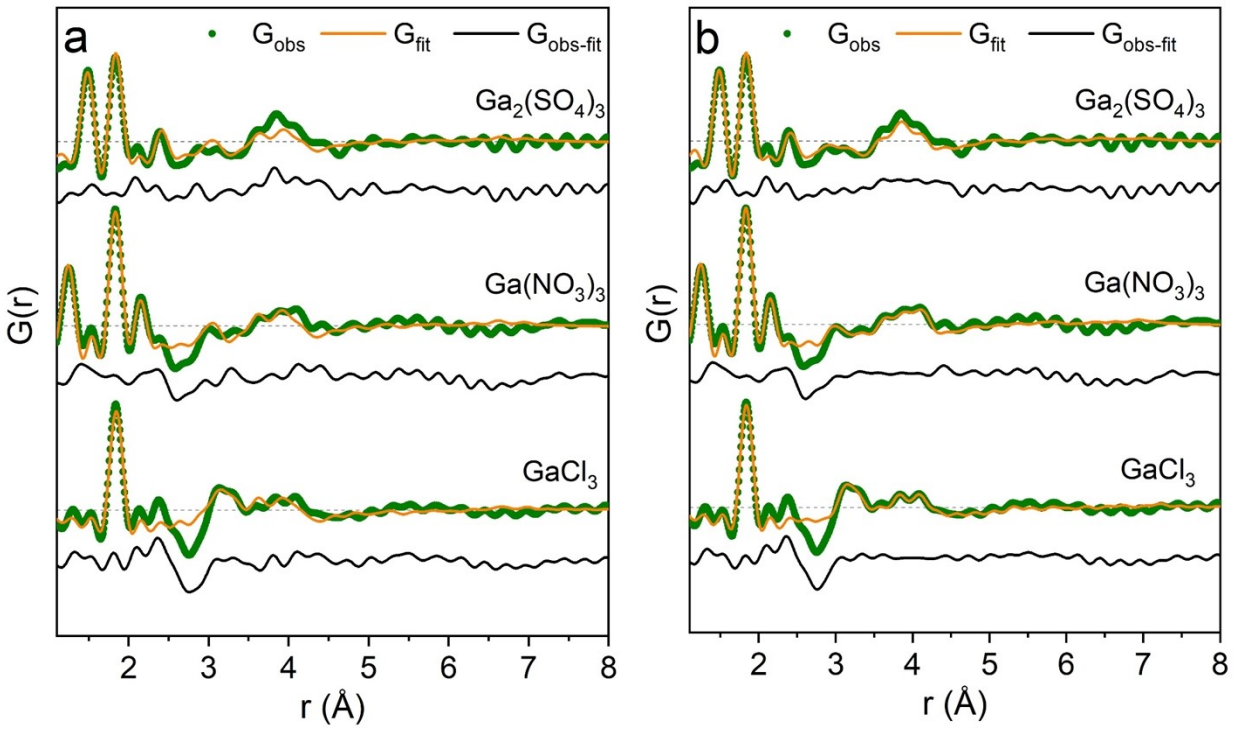

Figure S15. PDFs of a tetrahedral monomer with 5 solvent oxygen against the three salts for R = 5 $GaCl_3$ and R = 4 for $Ga(NO_3)_3$ and $Ga_2(SO_4)_3$. a) Only allowing for expansion (zoom) and b) with refinement of atomic positions with a 0.2 box restraint

Table S10. R_w and zoom values for the 5 models refined against the R = 5 PDF for $GaCl_3$ and R = 4 PDFs for $Ga_2(SO_4)_3$ and $Ga(NO_3)_3$.

	$Ga(NO_3)_3$		$GaCl_3$		$Ga_2(SO_4)_3$	
	R_w	Zoom	R_w		R_w	
Monomer with solvent – zoom	0.338	0.990	0.461	0.994	0.349	0.993
Monomer with solvent – 0.2 restraint	0.304	-	0.408	-	0.298	-

Table S11. Atomic positions of tetrahedral Ga^{3+} w. water before refinement and after for the three salts

Tetraeder w 5 water

Ga	-17.961	13.983	7.025
O	-18.833	12.438	7.554
O	-18.988	14.674	5.624
O	-18.006	15.066	8.539
O	-16.231	13.653	6.477
O	-17.204	13.024	3.029
O	-15.019	14.519	9.158
O	-21.827	14.351	7.707
O	-16.627	10.208	6.966
O	-16.587	17.042	5.238

Refined $Ga(NO_3)_3$ R = 4

Ga	-17.961	13.983	7.0250
O	-18.6320495	12.4321524	7.72124069
O	-19.0317877	14.4799483	5.60810299
O	-18.0960154	15.2661820	8.33641513
O	-16.2705853	13.7176293	6.36738061
O	-17.4044607	12.8234939	3.04927358
O	-15.1625696	14.6689081	9.32925885
O	-21.8632746	14.5511646	7.66860968
O	-16.4574265	10.1764707	6.76586685
O	-16.4984546	17.1966839	5.43960742

Refined $GaCl_3$ R = 5

Ga	-17.961	13.983	7.0250
O	-18.9860651	12.6414521	7.76374437
O	-18.8483033	14.8559552	5.63135611
O	-18.2113582	14.8558550	8.63808077
O	-16.2024945	13.5687103	6.59075646
O	-17.0038053	13.1157143	3.11259912
O	-14.9521846	14.4133131	9.29631819
O	-21.9652213	14.5520774	7.50614765
O	-16.4407038	10.0062085	6.76559037
O	-16.7880218	17.1523580	5.07256493

Refined $Ga_2(SO_4)_3$ R = 4

Ga	-17.961	13.983	7.0250
O	-18.7844008	12.4376520	7.70422553
O	-19.1236843	14.4734798	5.69950631
O	-18.2079226	15.2378032	8.33892556
O	-16.1828183	13.8530614	6.56007628
O	-17.0035452	13.2250166	3.06082036
O	-14.8179377	14.5458978	9.26717148
O	-21.8800558	14.3383660	7.90783095
O	-16.8281314	10.3830731	7.16700825
O	-16.7771345	17.1068126	5.12019507

Tetraeder w 4 water

Ga	-17.961000	13.983000	7.0250000
O	-18.772310	12.436301	7.6027502
O	-18.932119	14.588625	5.5787933
O	-18.163900	15.251935	8.3456078
O	-16.223992	13.817842	6.5121852
O	-17.001872	12.820389	3.1523900
O	-14.986104	14.708796	9.2857210
O	-21.747816	14.541609	7.8904572
O	-16.426752	10.239216	6.7617086

Tetraeder w 6 water

Ga	-17.961000	13.983000	7.0250000
O	-18.747141	12.362247	7.3531077
O	-18.903238	14.772678	5.6466696
O	-18.108355	15.056229	8.5064081
O	-16.234537	13.728053	6.4710983
O	-17.402262	12.902791	3.1114765
O	-14.859616	14.721096	9.2193889
O	-21.779137	14.552243	7.7915015
O	-16.422515	9.9957186	6.7640412
O	-16.687599	17.032405	5.4389059
O	-19.121371	12.357997	10.607568

Variations across salt

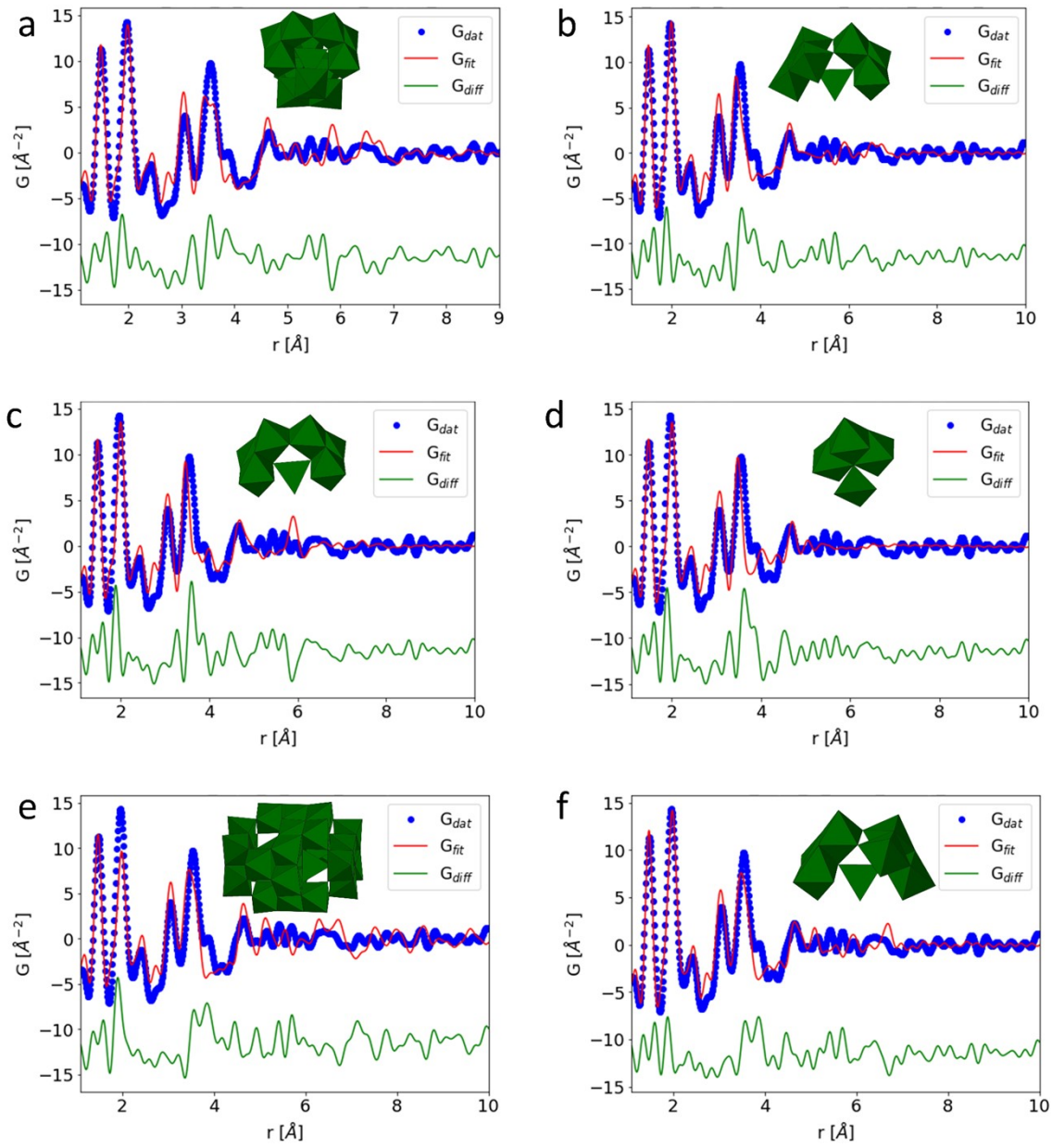

Figure S16. Fit of PDF of $Ga_2(SO_4)_3$ R = 1.5 with structures illustrated together with the fits. a) γ-Ga_{13} Keggin ion, b), c) and d) cut-outs of γ-Ga_{13} Keggin ion marked in red in Figure 7c, e) Ga_{30} ion and f) cut-out of Ga_{30} ion similar to the cut-out in c).

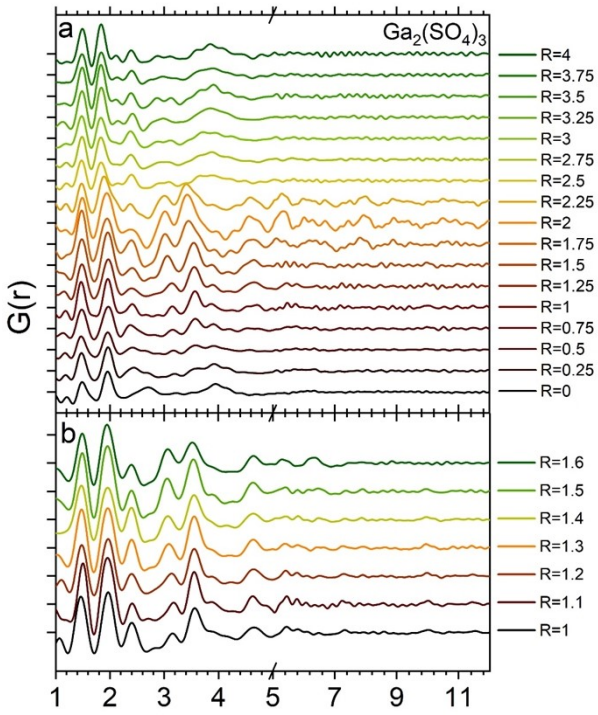

Figure S17. PDFs of $Ga_2(SO_4)_3$ solutions as function of R (R = $[OH^-]/[Ga^{3+}]$). a) Series with 0.25 steps in R and b) series with 0.1 steps in R around the polyoxogallate structure signal all showing a much stronger peak at 3.5 Å compared to the peak at 3.1 Å.

Concentration

Table S12. Measured pH for the concentration series of $Ga_2(SO_4)_3$, $Ga(NO_3)_3$ and $GaCl_3$ with R=0 and R=2 corresponding to the PDF data in Figure 8.

	R=0			**R=2**		
$c(Ga^{3+})$	$Ga_2(SO_4)_3$	$Ga(NO_3)_3$	$GaCl_3$	$Ga_2(SO_4)_3$	$Ga(NO_3)_3$	$GaCl_3$
0.5 M	2	1	1	10	8	2
1 M	2	0	0	11	9.5	1,5
2 M	1.5	0	0	8	10	1

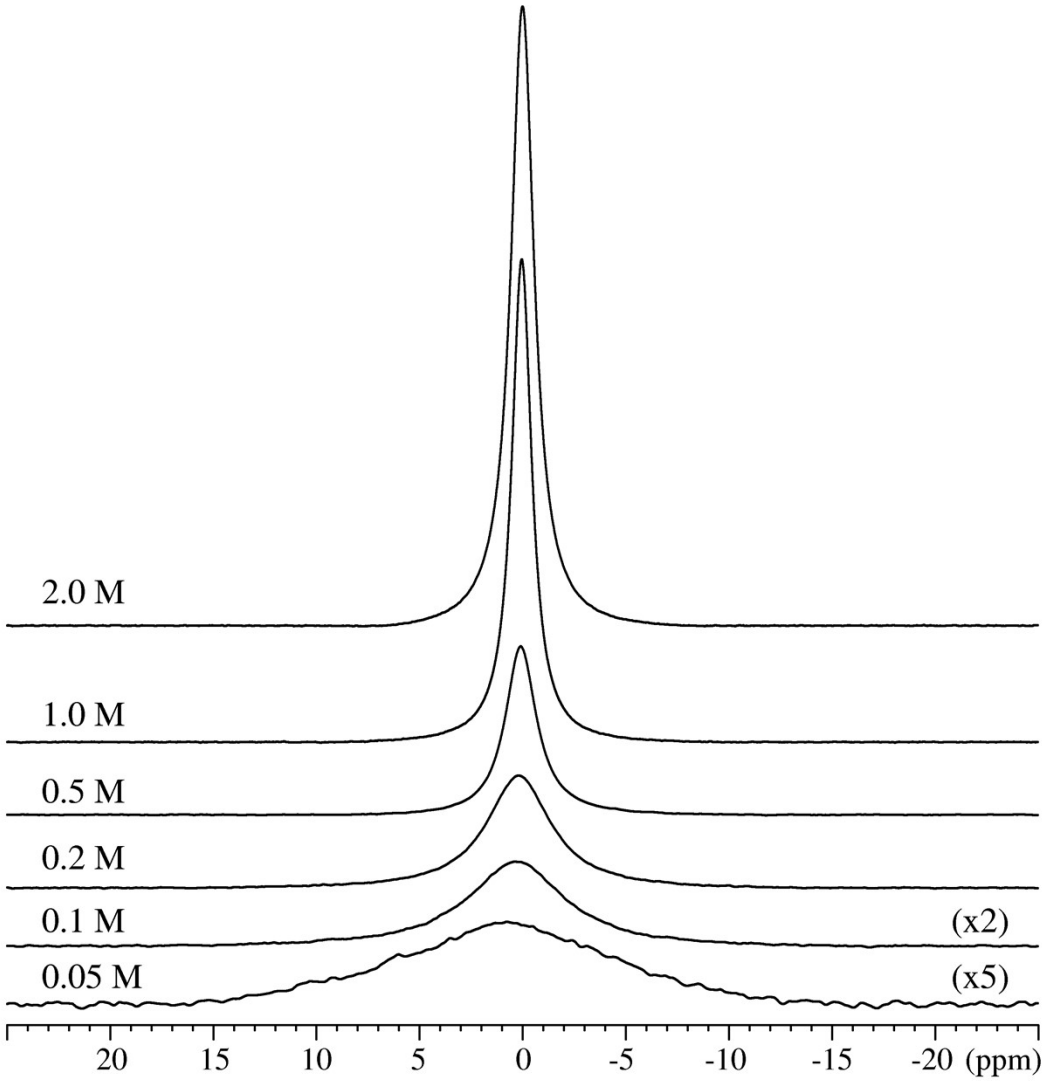

Figure S18. ^{71}Ga NMR spectra (9.39 T, $\nu_L(^{71}Ga)$ = 121.9 MHz) for $Ga(NO_3)_3$ dissolved in D_2O with concentrations of 0.05, 0.1, 0.2, 0.5, 1 and 2 M. The single-pulse spectra have be obtained at a solution-state spectrometer using a 90° excitation pulse ($\gamma B_1/2\pi$ = 20 kHz) , a 1 s relaxation delay and 64 – 256 scans.

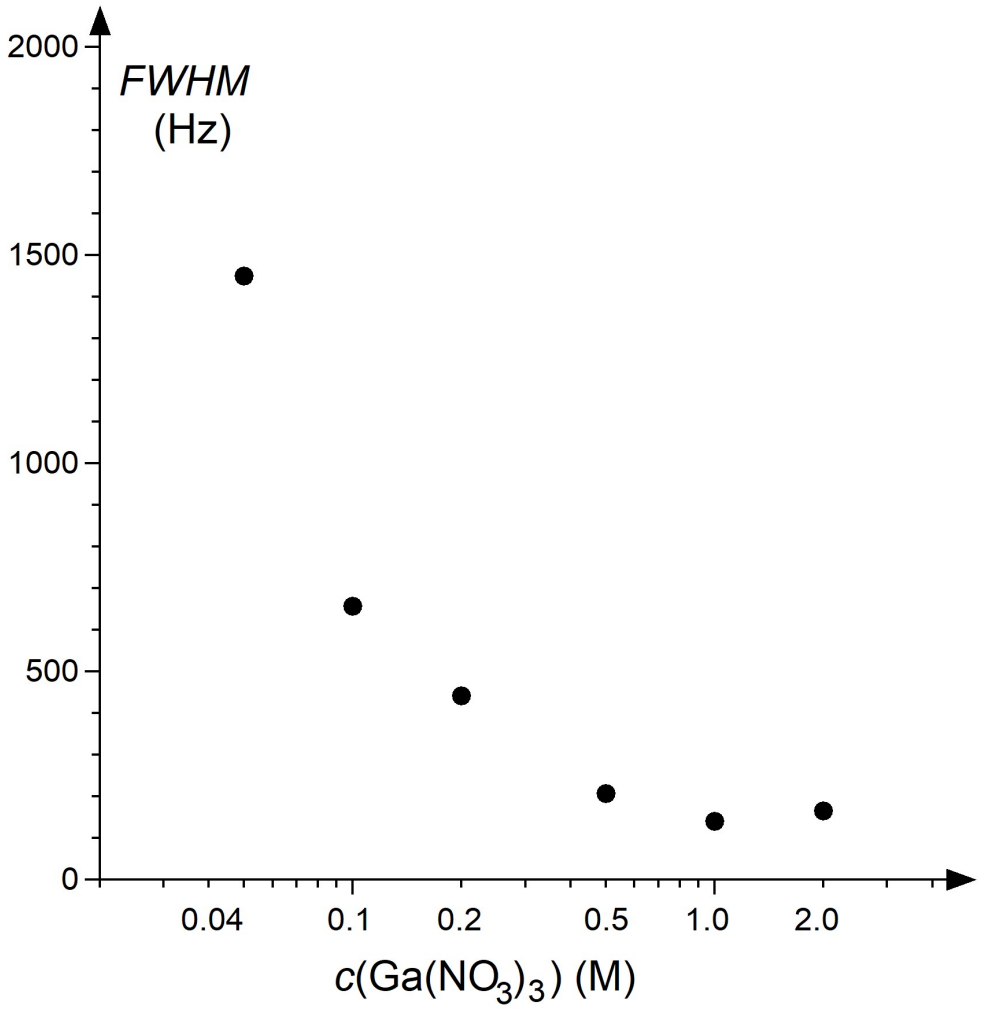

Figure S19. ^{71}Ga NMR linewidths *(FWHM*, full width at half maximum) of the resonances observed for the concentration series of $Ga(NO_3)_3$ in D_2O as a function of the $Ga(NO_3)_3$ concentration. The linewidths are determined from the spectra in Figure S19.

Table S13. Measured pH for the concentration series of $Ga(NO_3)_3$ in D_2O with NMR data in Figure S19-20.

c($Ga(NO_3)_3$)	**Measured pH**
2	0
1	0
0.5	0.5
0.2	1
0.1	1.5
0.05	2

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