Electronic supplementary information for:

Exploring The Anion Chemical Space of $Ln_2OF_{2-x}Cl_xH_2$ (Ln = Y, La, Gd): An Electroelastic Material with High Mechanical Sensitivity and Energy Harvesting

Evgenii Strugovshchikov* and Aleksandr Pishtshev

Institute of Physics, University of Tartu, W.Ostwaldi 1, 50411 Tartu, Estonia

E-mail: evgenii.strugovshchikov@ut.ee, aleksandr.pishtshev@ut.ee

CONTENTS

1. Equilibrium atomic positions predicted for $Ln_2OF_{2-x}Cl_xH_2$ (Ln = Y, La, Gd)

Supplementary Table S1: $Y_2OF_2H_2$, P3m1

P3m1 (156); a = 3.712 Å, c = 6.941 Å					
Atom	Wyckoff position	Symm.	x	y	z
Y1	1a	3m.	0.0000	0.0000	0.6421
Y2	1c	3m.	0.6667	0.3333	0.2102
O1	1c	3m.	0.6667	0.3333	0.5306
F1	1a	3m.	0.0000	0.0000	0.0000
F2	1b	3m.	0.3333	0.6667	0.7721
H1	1a	3m.	0.0000	0.0000	0.3294
H2	1b	3m.	0.3333	0.6667	0.2326

Supplementary Table S2: $Y_2OF_2H_2$, R3m

R3m (160); a = 3.657 Å, c = 22.014 Å					
Atom	Wyckoff position	Symm.	x	y	z
Y1	3a	3m	0.0000	0.0000	0.0000
Y2	3a	3m	0.0000	0.0000	0.4710
O1	3a	3m	0.0000	0.0000	0.3659
F1	3a	3m	0.0000	0.0000	0.1964
F2	3a	3m	0.0000	0.0000	0.6168
H1	3a	3m	0.0000	0.0000	0.7927
H2	3a	3m	0.0000	0.0000	0.0949

Supplementary Table S3: $La_2OF_2H_2$, P3m1

P3m1 (156); a = 3.997 Å, c = 7.288 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	1a	3m.	0.0000	0.0000	0.6430
La2	1b	3m.	0.3333	0.6667	0.2028
O1	1b	3m.	0.3333	0.6667	0.5279
F1	1a	3m.	0.0000	0.0000	0.0000
F2	1c	3m.	0.6667	0.3333	0.7740
H1	1a	3m.	0.0000	0.0000	0.3250
H2	1c	3m.	0.6667	0.3333	0.2464

Supplementary Table S4: $La_2OF_2H_2$, R3m

R3m (160); a = 3.978 Å, c = 21.444 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	3a	3m	0.0000	0.0000	0.0000
La2	3a	3m	0.0000	0.0000	0.4847
O1	3a	3m	0.0000	0.0000	0.3705
F1	3a	3m	0.0000	0.0000	0.2130
F2	3a	3m	0.0000	0.0000	0.6163
H1	3a	3m	0.0000	0.0000	0.8035
H2	3a	3m	0.0000	0.0000	0.1046

Supplementary Table S5: $Gd_2OF_2H_2$, P3m1

P3m1 (156); a = 3.783 Å, c = 7.026 Å					
Atom	Wyckoff position	Symm.	x	y	z
Gd1	1a	3m.	0.0000	0.0000	0.6414
Gd2	1c	3m.	0.6667	0.3333	0.2079
O1	1c	3m.	0.6667	0.3333	0.5287
F1	1a	3m.	0.0000	0.0000	0.0000
F2	1b	3m.	0.3333	0.6667	0.7722
H1	1a	3m.	0.0000	0.0000	0.3289
H2	1b	3m.	0.3333	0.6667	0.2338

Supplementary Table S6: $Gd_2OF_2H_2$, R3m

R3m (160); a = 3.742 Å, c = 21.562 Å					
Atom	Wyckoff position	Symm.	x	y	z
Gd1	3a	3m	0.0000	0.0000	0.0000
Gd2	3a	3m	0.0000	0.0000	0.4760
O1	3a	3m	0.0000	0.0000	0.3676
F1	3a	3m	0.0000	0.0000	0.2024
F2	3a	3m	0.0000	0.0000	0.6159
H1	3a	3m	0.0000	0.0000	0.7972
H2	3a	3m	0.0000	0.0000	0.0981

Supplementary Table S7: Y_2OFClH_2, P3m1

P3m1 (156); a = 3.735 Å, c = 8.181 Å					
Atom	Wyckoff position	Symm.	x	y	z
Y1	1a	3m.	0.0000	0.0000	0.0000
Y2	1b	3m.	0.3333	0.6667	0.3637
O1	1b	3m.	0.3333	0.6667	0.0891
F1	1c	3m.	0.6667	0.3333	0.8849
Cl1	1a	3m.	0.0000	0.0000	0.5930
H1	1a	3m.	0.0000	0.0000	0.2660
H2	1c	3m.	0.6667	0.3333	0.3393

Supplementary Table S8: Y_2OFClH_2, R3m

R3m (160); a = 3.712 Å, c = 25.933 Å					
Atom	Wyckoff position	Symm.	x	y	z
Y1	3a	3m	0.0000	0.0000	0.0000
Y2	3a	3m	0.0000	0.0000	0.4484
O1	3a	3m	0.0000	0.0000	0.3604
F1	3a	3m	0.0000	0.0000	0.6270
Cl1	3a	3m	0.0000	0.0000	0.1863
H1	3a	3m	0.0000	0.0000	0.7724
H2	3a	3m	0.0000	0.0000	0.0829

Supplementary Table S9: La_2OFClH_2, P3m1

P3m1 (156); a = 4.048 Å, c = 8.374 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	1a	3m.	0.0000	0.0000	0.6153
La2	1c	3m.	0.6667	0.3333	0.2354
O1	1c	3m.	0.6667	0.3333	0.5166
F1	1b	3m.	0.3333	0.6667	0.7185
Cl1	1a	3m.	0.0000	0.0000	0.0000
H1	1a	3m.	0.0000	0.0000	0.3342
H2	1b	3m.	0.3333	0.6667	0.2672

Supplementary Table S10: La_2OFClH_2, R3m

R3m (160); a = 3.712 Å, c = 25.933 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	3a	3m	0.0000	0.0000	0.0000
La2	3a	3m	0.0000	0.0000	0.4603
O1	3a	3m	0.0000	0.0000	0.3660
F1	3a	3m	0.0000	0.0000	0.6315
Cl1	3a	3m	0.0000	0.0000	0.2049
H1	3a	3m	0.0000	0.0000	0.7823
H2	3a	3m	0.0000	0.0000	0.0928

Supplementary Table S11: Gd_2OFClH_2, P3m1

P3m1 (156); a = 3.812 Å, c = 8.181 Å					
Atom	Wyckoff position	Symm.	x	y	z
Gd1	1a	3m.	0.0000	0.0000	0.0000
Gd2	1b	3m.	0.3333	0.6667	0.3693
O1	1b	3m.	0.3333	0.6667	0.0923
F1	1c	3m.	0.6667	0.3333	0.8874
Cl1	1a	3m.	0.0000	0.0000	0.6009
H1	1a	3m.	0.0000	0.0000	0.2706
H2	1c	3m.	0.6667	0.3333	0.3434

Supplementary Table S12: Gd_2OFClH_2, R3m

R3m (160); a = 3.712 Å, c = 25.933 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	3a	3m	0.0000	0.0000	0.0000
La2	3a	3m	0.0000	0.0000	0.4502
O1	3a	3m	0.0000	0.0000	0.3613
F1	3a	3m	0.0000	0.0000	0.6274
Cl1	3a	3m	0.0000	0.0000	0.1887
H1	3a	3m	0.0000	0.0000	0.7735
H2	3a	3m	0.0000	0.0000	0.0843

Supplementary Table S13: $LaGdOFClH_2$, P3m1

P3m1 (156); a = 3.939 Å, c = 8.292 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	1a	3m.	0.0000	0.0000	0.6028
Gd1	1c	3m.	0.6667	0.3333	0.2288
O1	1c	3m.	0.6667	0.3333	0.4995
F1	1b	3m.	0.3333	0.6667	0.7163
Cl1	1a	3m.	0.0000	0.0000	0.0000
H1	1a	3m.	0.0000	0.0000	0.3207
H2	1b	3m.	0.3333	0.6667	0.2530

Supplementary Table S14: $LaGdOFClH_2$, R3m

R3m (160); a = 3.924 Å, c = 25.246 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	3a	3m	0.0000	0.0000	0.0000
Gd1	3a	3m	0.0000	0.0000	0.4563
O1	3a	3m	0.0000	0.0000	0.3667
F1	3a	3m	0.0000	0.0000	0.6277
Cl1	3a	3m	0.0000	0.0000	0.1971
H1	3a	3m	0.0000	0.0000	0.7806
H2	3a	3m	0.0000	0.0000	0.0916

Supplementary Table S15: $Y_2OCl_2H_2$, R3m

R3m (160); a = 3.756 Å, c = 30.481 Å					
Atom	Wyckoff position	Symm.	x	y	z
Y1	3a	3m	0.0000	0.0000	0.0000
Y2	3a	3m	0.0000	0.0000	0.4307
O1	3a	3m	0.0000	0.0000	0.3545
Cl1	3a	3m	0.0000	0.0000	0.1563
Cl2	3a	3m	0.0000	0.0000	0.6094
H1	3a	3m	0.0000	0.0000	0.7540
H2	3a	3m	0.0000	0.0000	0.0698

Supplementary Table S16: $La_2OCl_2H_2$, R3m

R3m (160); a = 4.056 Å, c = 30.999 Å					
Atom	Wyckoff position	Symm.	x	y	z
La1	3a	3m	0.0000	0.0000	0.0000
La2	3a	3m	0.0000	0.0000	0.4356
O1	3a	3m	0.0000	0.0000	0.3561
Cl1	3a	3m	0.0000	0.0000	0.1619
Cl2	3a	3m	0.0000	0.0000	0.6092
H1	3a	3m	0.0000	0.0000	0.7548
H2	3a	3m	0.0000	0.0000	0.0739

Supplementary Table S17: $Gd_2OCl_2H_2$, R3m

R3m (160); a = 3.824 Å, c = 30.722 Å					
Atom	Wyckoff position	Symm.	x	y	z
Gd1	3a	3m	0.0000	0.0000	0.0000
Gd2	3a	3m	0.0000	0.0000	0.4316
O1	3a	3m	0.0000	0.0000	0.3549
Cl1	3a	3m	0.0000	0.0000	0.1570
Cl2	3a	3m	0.0000	0.0000	0.6096
H1	3a	3m	0.0000	0.0000	0.7545
H2	3a	3m	0.0000	0.0000	0.0704

2. Bond distances and bond angles in $Ln_2OF_{2-x}Cl_xH_2$ (Ln = Y, La, Gd)

Supplementary Table S18: Bond distances (in Å) and bond angles (in degrees) in $Y_2OF_2H_2$

$Y_2OF_2H_2$, P3m1				$Y_2OF_2H_2$, R3m			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
Y1 – Y2	3.68			Y1 – Y2	3.69		
Y1 – O1	2.28	Y1 – O1 – Y2	109.9	Y1 – O1	2.23	Y1 – O1 – Y2	108.8
Y2 – O1	2.22			Y2 – O1	2.31		
Y1 – H2	2.17	Y2 – H1 – H2	166.8	Y1 – H2	2.09	Y2 – H1 – H2	169.0
Y2 – H1	2.15			Y2 – H1	2.13		
H1 – H2	2.25			H1 – H2	2.22		
H2 – F1	2.29	Y1 – H2 – F1	180.0	H2 – F1	2.23	Y1 – H2 – F1	180.0
Y1 – F2	2.33	Y2 – F1 – F2	177.8	Y1 – F2	2.38	Y1 – F2 – F1	165.2
Y1 – F1	2.48			Y2 – F2	3.21		
Y2 – F1	2.59			Y2 – F1	2.48		
F2 – F1	2.66			F2 – F1	2.85		

Supplementary Table S19: Bond distances (in Å) and bond angles (in degrees) in $La_2OF_2H_2$

$La_2OF_2H_2$, P3m1				$La_2OF_2H_2$, R3m			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
La1 – La2	3.95			La1 – La2	3.98		
La1 – O1	2.46	La1 – O1 – La2	110.0	La1 – O1	2.43	La1 – O1 – La2	109.1
La2 – O1	2.37			La2 – O1	2.45		
La1 – H2	2.32	La2 – H1 – H2	173.9	La1 – H2	2.24	La2 – H1 – H2	170.9
La2 – H1	2.33			La2 – H1	2.32		
H1 – H2	2.38			H1 – H2	2.40		
H2 – F1	2.37	La1 – H2 – F1	180.0	H2 – F1	2.33	La1 – H2 – F1	180.0
La1 – F2	2.50	La1 – F1 – F2	177.1	La1 – F2	2.54	La1 – F2 – F1	172.1
La1 – F1	2.60			La1 – F1	3.45		
La2 – F1	2.74			La2 – F1	2.65		
F2 – F1	2.84			F2 – F1	2.74		

Supplementary Table S20: Bond distances (in Å) and bond angles (in degrees) in $Gd_2OF_2H_2$

$Gd_2OF_2H_2$, P3m1				**$Gd_2OF_2H_2$, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
Gd1 – Gd2	3.75			Gd1 – Gd2	3.76		
Gd1 – O1	2.32	Gd1 – O1 – Gd2	109.9	Gd1 – O1	2.28	Gd1 – O1 – Gd2	108.9
Gd2 – O1	2.25			Gd2 – O1	2.34		
Gd1 – H2	2.20	Gd2 – H1 – H2	167.8	Gd1 – H2	2.11	Gd2 – H1 – H2	168.9
Gd2 – H1	2.19			Gd2 – H1	2.18		
H1 – H2	2.28			H1 – H2	2.27		
H2 – F1	2.31	Gd1 – H2 – F1	180.0	H2 – F1	2.25	Gd1 – H2 – F1	180.0
Gd1 – F2	2.37	Gd1 – F1 – F2	177.5	Gd1 – F2	2.42	Gd1 – F2 – F1	168.2
Gd1 – F1	2.52			Gd2 – F2	3.02		
Gd2 – F1	2.63			Gd2 – F1	2.51		
F2 – F1	2.71			F2 – F1	2.77		

Supplementary Table S21: Bond distances (in Å) and bond angles (in degrees) in Y_2OFClH_2

Y_2OFClH_2, P3m1				**Y_2OFClH_2, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
Y1 – Y2	3.67			Y1 – Y2	3.67		
Y1 – O1	2.28	Y1 – O1 – Y2	108.7	Y1 – O1	2.26	Y1 – O1 – Y2	108.2
Y2 – O1	2.25			Y2 – O1	2.28		
Y1 – H2	2.18	Y2 – H1 – H2	169.7	Y1 – H2	2.15	Y2 – H1 – H2	171.0
Y2 – H1	2.17			Y2 – H1	2.16		
H1 – H2	2.24			H1 – H2	2.22		
H2 – Cl1	2.68	Y1 – H2 – Cl1	180.0	H2 – Cl1	2.68	Y1 – H2 – Cl1	180.0
Y1 – F1	2.35	Y2 – Cl1 – F1	173.1	Y1 – F1	2.38	Y1 – F1 – Cl1	153.2
Y1 – Cl1	3.33			Y2 – F1	4.63		
Y2 – Cl1	2.86			Y2 – Cl1	2.83		
Cl1 – F1	3.22			Cl1 – F1	3.51		

Supplementary Table S22: Bond distances (in Å) and bond angles (in degrees) in La_2OFClH_2

La_2OFClH_2, P3m1				**La_2OFClH_2, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
La1 – La2	3.95			La1 – La2	3.94		
La1 – O1	2.48	La1 – O1 – La2	109.5	La1 – O1	2.47	La1 – O1 – La2	109.4
La2 – O1	2.36			La2 – O1	2.36		
La1 – H2	2.35	La2 – H1 – H2	173.0	La1 – H2	2.33	La2 – H1 – H2	173.1
La2 – H1	2.35			La2 – H1	2.35		
H1 – H2	2.40			H1 – H2	2.40		
H2 – Cl1	2.80	La1 – H2 – Cl1	180.0	H2 – Cl1	2.81	La1 – H2 – Cl1	180.0
La1 – F1	2.49	La2 – Cl1 – F1	174.9	La1 – F1	2.49	La1 – F1 – Cl1	155.7
La1 – Cl1	3.22			La2 – F1	4.29		
La2 – Cl1	3.06			La2 – Cl1	3.04		
Cl1 – F1	3.32			Cl1 – F1	3.30		

Supplementary Table S23: Bond distances (in Å) and bond angles (in degrees) in Gd_2OFClH_2

Gd_2OFClH_2, P3m1				**Gd_2OFClH_2, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
Gd1 – Gd2	3.74			Gd1 – Gd2	3.74		
Gd1 – O1	2.33	Gd1 – O1 – Gd2	108.9	Gd1 – O1	2.30	Gd1 – O1 – Gd2	108.4
Gd2 – O1	2.27			Gd2 – O1	2.30		
Gd1 – H2	2.21	Gd2 – H1 – H2	170.4	Gd1 – H2	2.18	Gd2 – H1 – H2	171.8
Gd2 – H1	2.21			Gd2 – H1	2.20		
H1 – H2	2.28			H1 – H2	2.26		
H2 – Cl1	2.70	Gd1 – H2 – Cl1	180.0	H2 – Cl1	2.71	Gd1 – H2 – Cl1	180.0
Gd1 – F1	2.39	Gd2 – F1 – Cl1	173.9	Gd1 – F1	2.41	Gd1 – F1 – Cl1	153.7
Gd1 – Cl1	3.26			Gd2 – F1	4.59		
Gd2 – Cl1	2.90			Gd2 – Cl1	2.87		
Cl1 – F1	3.21			Cl1 – F1	3.50		

Supplementary Table S24: Bond distances (in Å) and bond angles (in degrees) in $LaGdOFClH_2$

$LaGdOFClH_2$, P3m1				**$LaGdOFClH_2$, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
La1 – Gd1	3.85			La1 – Gd1	3.84		
Gd1 – O1	2.24	Gd1 – O1 – La1	110.6	Gd1 – O1	2.26	Gd1 – O1 – La1	110.4
La1 – O1	2.43			La1 – O1	2.42		
La1 – H2	2.34	Gd1 – H1 – H2	171.2	La1 – H2	2.31	Gd1 – H1 – H2	171.7
Gd1 – H1	2.28			Gd1 – H1	2.28		
H1 – H2	2.34			H1 – H2	2.33		
H2 – Cl1	2.66	La1 – H2 – Cl1	180.0	H2 – Cl1	2.66	La1 – H2 – Cl1	180.0
La1 – F1	2.46	Gd1 – Cl1 – F1	173.9	La1 – F1	2.47	La1 – F1 – Cl1	156.2
La1 – Cl1	3.29			Gd1 – F1	4.33		
Gd1 – Cl1	2.96			Gd1 – Cl1	2.93		
Cl1 – F1	3.27			Cl1 – F1	3.34		

Supplementary Table S25: Bond distances (in Å) and bond angles (in degrees) in $Y_2OCl_2H_2$ and $La_2OCl_2H_2$

$Y_2OCl_2H_2$, R3m				**$La_2OCl_2H_2$, R3m**			
Bond	Value	Angle	Value	Bond	Value	Angle	Value
Y1 – Y2	3.68			La1 – La2	3.94		
Y1 – O1	2.26	Y1 – O1 – Y2	106.6	La1 – O1	2.45	La1 – O1 – La2	
Y2 – O1	2.32			La2 – O1	2.47		
Y1 – H2	2.13	Y2 – H1 – H2	174.2	La1 – H2	2.29	La2 – H1 – H2	180.0
Y2 – H1	2.19			La2 – H1	2.38		
H1 – H2	2.23			H1 – H2	2.38		
H2 – Cl1	2.64	Y1 – H2 – Cl1	180.0	H2 – Cl1	2.73	La1 – H2 – Cl1	180.0
Y1 – Cl2	2.78	Y2 – Cl1 – Cl2	159.5	La1 – Cl2	2.94	La1 – Cl2 – Cl1	160.8
Y2 – Cl2	5.45			La2 – Cl2	5.38		
Y2 – Cl1	2.78			La1 – Cl2	2.94		
Cl2 – Cl1	4.24			Cl2 – Cl1	4.24		

Supplementary Table S26: Bond distances (in Å) and bond angles (in degrees) in $Gd_2OCl_2H_2$

$Gd_2OCl_2H_2$, R3m							
Bond	Value	Angle	Value				
Gd1 – Gd2	3.74						
Gd1 – O1	2.30	Gd1 – O1 – Gd2	106.7				
Gd2 – O1	2.36						
Gd1 – H2	2.16	Gd2 – H1 – H2	174.6				
Gd2 – H1	2.23						
H1 – H2	2.27						
H2 – Cl1	2.66	Gd1 – H2 – Cl1	180.0				
Gd1 – Cl2	2.82	Gd1 – Cl2 – Cl1	159.5				
Gd2 – Cl2	5.47						
Gd1 – Cl2	2.82						
Cl2 – Cl1	4.28						

3. Elastic properties of $Ln_2OF_{2-x}Cl_xH_2$ (Ln = Y, La, Gd)

Supplementary table S27: Components of the elasticity tensors C_{ij} (in GPa)

$Y_2OF_2H_2$ (P3m1)						$Y_2OF_2H_2$ (R3m)					
170.1	64.1	51.0	8.0	0	0	194.5	68.3	26.1	-12.1	0	0
64.1	170.1	51.0	-8.0	0	0	68.3	194.5	26.1	12.1	0	0
51.0	51.0	136.3	0	0	0	26.1	26.1	23.2	0	0	0
8.0	-8.0	0	25.6	0	0	-12.1	12.1	0	31.6	0	0
0	0	0	0	25.6	8.0	0	0	0	0	31.6	-12.1
0	0	0	0	8.0	53.0	0	0	0	0	-12.1	63.1
$La_2OF_2H_2$ (P3m1)						$La_2OF_2H_2$ (R3m)					
145.3	48.6	30.9	0.4	0	0	173.5	68.7	40.9	-17.7	0	0
48.6	145.3	30.9	-0.4	0	0	68.7	173.5	40.9	17.7	0	0
30.9	30.9	117.6	0	0	0	40.9	40.9	70.0	0	0	0
0.4	-0.4	0	20.3	0	0	-17.7	17.7	0	35.8	0	0
0	0	0	0	20.3	0.4	0	0	0	0	35.8	-17.7
0	0	0	0	0.4	48.3	0	0	0	0	-17.7	52.4
$Gd_2OF_2H_2$ (P3m1)						$Gd_2OF_2H_2$ (R3m)					
164.8	59.3	45.0	4.2	0	0	192.4	69.5	32.4	-15.0	0	0
59.3	164.8	45.0	-4.2	0	0	69.5	192.4	32.4	15.0	0	0
45.0	45.0	132.7	0	0	0	32.4	32.4	31.2	0	0	0
4.2	-4.2	0	25.5	0	0	-15.0	15.0	0	37.9	0	0
0	0	0	0	25.5	4.2	0	0	0	0	37.9	-15.0
0	0	0	0	4.2	52.7	0	0	0	0	-15.0	61.5

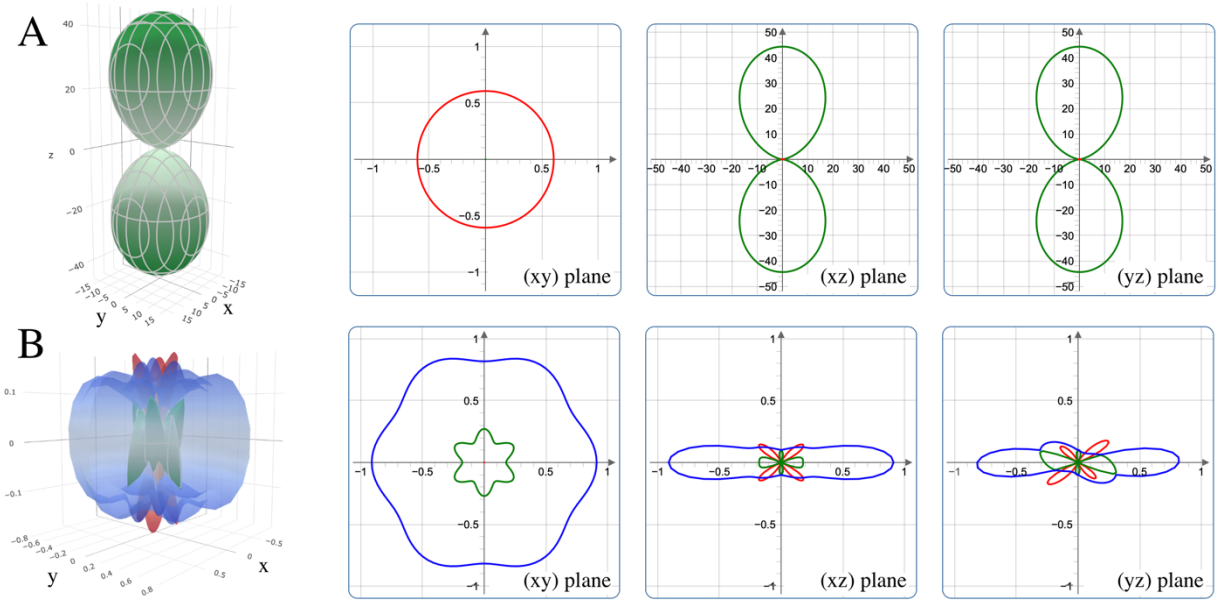
Y_2OFClH_2 (P3m1)						Y_2OFClH_2 (R3m)					
167.2	45.4	31.3	-5.2	0	0	158.9	43.2	11.2	-2.2	0	0
45.4	167.2	31.3	5.2	0	0	43.2	158.9	11.2	2.2	0	0
31.3	31.3	41.2	0	0	0	11.2	11.2	19.6	0	0	0
-5.2	5.2	0	24.4	0	0	-2.2	2.2	0	5.9	0	0
0	0	0	0	24.4	-5.2	0	0	0	0	5.9	-2.2
0	0	0	0	-5.2	60.9	0	0	0	0	-2.2	57.9
La_2OFClH_2 (P3m1)						La_2OFClH_2 (R3m)					
143.6	40.7	28.2	-4.0	0	0	139.4	44.5	21.7	-6.9	0	0
40.7	143.6	28.2	4.0	0	0	44.5	139.4	21.7	6.9	0	0

28.2	28.2	78.9	0	0	0	21.7	21.7	27.3	0	0	0
-4.0	4.0	0	18.8	0	0	-6.9	6.9	0	14.4	0	0
0	0	0	0	18.8	-4.0	0	0	0	0	14.4	-6.9
0	0	0	0	-4.0	47.0	0	0	0	0	-6.9	47.4
Gd_2OFClH_2 (P3m1)						Gd_2OFClH_2 (R3m)					
158.7	44.4	33.0	-4.9	0	0	151.5	41.2	12.0	-2.5	0	0
44.4	158.7	33.0	4.9	0	0	41.2	151.5	12.0	2.5	0	0
33.0	33.0	52.8	0	0	0	12.0	12.0	18.6	0	0	0
-4.9	4.9	0	24.7	0	0	-2.5	2.5	0	6.1	0	0
0	0	0	0	24.7	-4.9	0	0	0	0	6.1	-2.5
0	0	0	0	-4.9	57.1	0	0	0	0	-2.5	55.1
$LaGdOFClH_2$ (P3m1)						$LaGdOFClH_2$ (R3m)					
140.0	42.2	28.4	-2.5	0	0	143.6	42.7	20.4	-6.4	0	0
42.2	140.0	28.4	2.5	0	0	42.7	143.6	20.4	6.4	0	0
28.4	28.4	71.1	0	0	0	20.4	20.4	25.1	0	0	0
-2.5	2.5	0	20.0	0	0	-6.4	6.4	0	12.9	0	0
0	0	0	0	20.0	-2.5	0	0	0	0	12.9	-6.4
0	0	0	0	-2.5	48.9	0	0	0	0	-6.4	50.5
$Y_2OCl_2H_2$ (R3m)						$La_2OCl_2H_2$ (R3m)					
124.4	34.1	2.6	-0.2	0	0	98.8	30.0	2.9	-0.4	0	0
34.1	124.4	2.6	0.2	0	0	30.0	98.8	2.9	0.4	0	0
2.6	2.6	8.8	0	0	0	2.9	2.9	7.7	0	0	0
-0.2	0.2	0	1.5	0	0	-0.4	0.4	0	1.4	0	0
0	0	0	0	1.5	-0.2	0	0	0	0	1.4	-0.4
0	0	0	0	-0.2	45.1	0	0	0	0	-0.4	34.4
$Gd_2OCl_2H_2$ (R3m)											
116.7	32.3	2.3	-0.2	0	0						
32.3	116.7	2.3	0.2	0	0						
2.3	2.3	7.7	0	0	0						
-0.2	0.2	0	1.0	0	0						
0	0	0	0	1.0	-0.2						
0	0	0	0	-0.2	42.2						

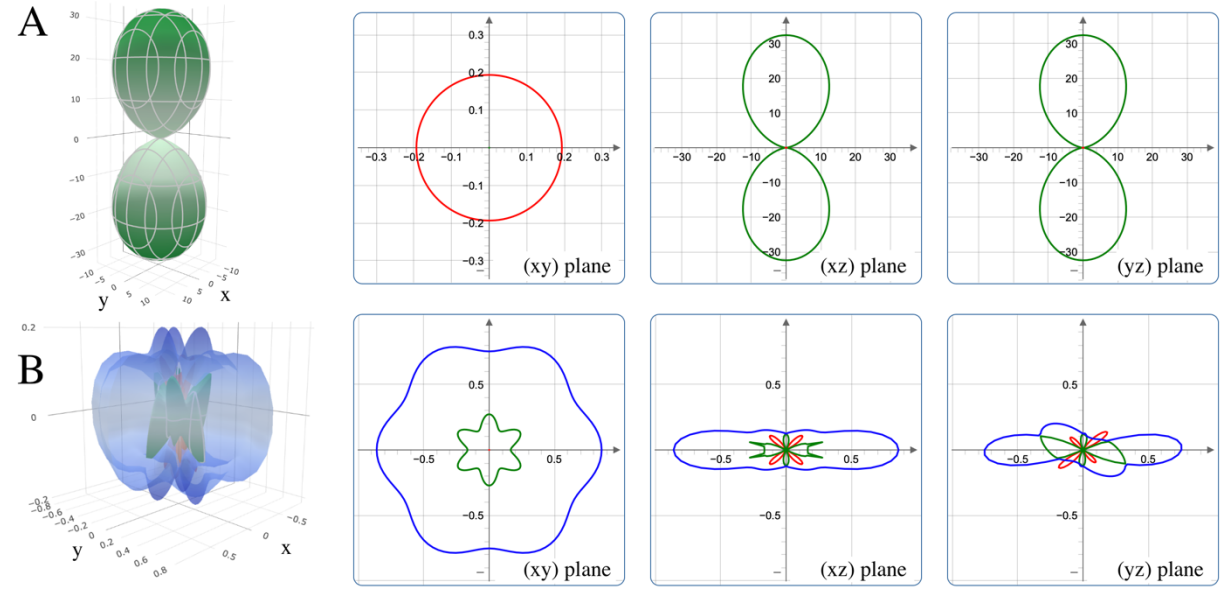
Supplementary table S28: Eigenvalues (λ) of the elasticity tensor (in Gpa)

	λ_1	λ_2	λ_3	λ_4	λ_5	λ_6
$Y_2OF_2H_2$ (P3m1)	23.4	24.0	55.1	98.1	107.5	272.4
$Y_2OF_2H_2$ (R3m)	17.7	27.4	28.6	67.3	129.3	268.3
$La_2OF_2H_2$ (P3m1)	20.3	20.3	48.3	96.7	97.7	213.8
$La_2OF_2H_2$ (R3m)	24.5	27.6	52.4	63.6	112.9	259.8
$Gd_2OF_2H_2$ (P3m1)	24.9	25.1	53.4	100.1	105.9	256.7
$Gd_2OF_2H_2$ (R3m)	22.5	30.6	32.9	68.7	127.9	270.7
Y_2OFClH_2 (P3m1)	23.7	23.8	30.5	61.6	122.4	223.4
Y_2OFClH_2 (R3m)	5.8	5.8	18.2	58.0	115.9	203.4
La_2OFClH_2 (P3m1)	18.3	18.4	47.5	64.6	94.3	189.7
La_2OFClH_2 (R3m)	13.0	13.2	21.5	48.8	96.1	189.7
Gd_2OFClH_2 (P3m1)	24.0	24.2	39.5	57.9	114.8	216.4
Gd_2OFClH_2 (R3m)	6.0	6.0	17.0	55.3	110.4	194.3
$LaGdOFClH_2$ (P3m1)	19.8	19.9	49.1	58.1	97.9	195.1
$LaGdOFClH_2$ (R3m)	11.8	12.0	20.0	51.1	101.9	191.3
$Y_2OCl_2H_2$ (R3m)	1.5	1.5	8.8	45.1	90.3	158.5
$La_2OCl_2H_2$ (R3m)	1.4	1.4	7.6	34.4	68.8	128.9
$Gd_2OCl_2H_2$ (R3m)	1.0	1.0	7.6	42.2	84.3	149.0

Spatial distributions of the shear modulus and Poisson's ratio were analyzed and visualized by using program tools implemented in ELATE open-source online application [1,2].



Supplementary Figure S1: Spatial distribution of the A) linear compressibility (in TPa^{-1}) and B) Poisson's ratio in $Y_2OF_2H_2$ (R3m). Green and blue colors represent the minimal and maximal values, respectively. Red color denotes the negative values



Supplementary Figure S2: Spatial distribution of the A) linear compressibility (in TPa^{-1}) and B) Poisson's ratio in $Gd_2OF_2H_2$ (R3m). Green and blue colors represent the minimal and maximal values, respectively. Red color denotes the negative values

4. Zone-centered optical vibrational modes calculated in the harmonic approximation

The frequencies were classified by using the program tools described in Ref. [3]

Supplementary Table S29: $Y_2OF_2H_2$ (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1317 cm^{-1}	A_1	H2 – H1
1311 cm^{-1}	E	H2 – H2
843 cm^{-1}	E	H1 – H2 – O1
673 cm^{-1}	A_1	H1 – Y2
495 cm^{-1}	A_1	O1 – F1 – Y1
408 cm^{-1}	E	F1 – O1 – Y1
347 cm^{-1}	A_1	F1 – F2 – Y1 – Y2
286 cm^{-1}	E	F2 – O1 – Y1
207 cm^{-1}	A_1	F1 – F2 – Y2
194 cm^{-1}	A_1	Y1 – Y2 – F2
168 cm^{-1}	E	F1 – Y1 – Y2
106 cm^{-1}	E	Y1 – Y2

Supplementary Table S30: $Y_2OF_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1434 cm^{-1}	A_1	H2 – H1
1352 cm^{-1}	E	H1 – H2
850 cm^{-1}	E	H2 – H1 – O1
693 cm^{-1}	A_1	H1 – Y2
465 cm^{-1}	A_1	O1 – Y1 – F1
425 cm^{-1}	E	O1 – F1 – Y2
359 cm^{-1}	A_1	F1 – Y1 – Y2
251 cm^{-1}	E	F1 – F2 – Y1
238 cm^{-1}	A_1	F2 – F1 – Y2
164 cm^{-1}	E	F1 – Y2
161 cm^{-1}	A_1	F2 – F1 – Y1 – Y2
102 cm^{-1}	E	Y1 – Y2 – F1

Supplementary Table S31: $La_2OF_2H_2$ (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1243 cm^{-1}	A_1	H2 – H1
1155 cm^{-1}	E	H1 – H2
723 cm^{-1}	E	H2 – H1
583 cm^{-1}	A_1	H1 – La2
429 cm^{-1}	A_1	O1 – La2
338 cm^{-1}	E	O1 – F1
283 cm^{-1}	A_1	O1 – F1 – La1
242 cm^{-1}	E	F2 – O1 – La1
203 cm^{-1}	A_1	F2 – F1 – La1 – La2
151 cm^{-1}	A_1	La1 – La2 – F1
145 cm^{-1}	E	F1 – La2
82 cm^{-1}	E	La1 – La2 – F2

Supplementary Table S32: $La_2OF_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1319 cm^{-1}	A_1	H2 – H1
1183 cm^{-1}	E	H1 – H2
721 cm^{-1}	E	H2 – H1 – O1
558 cm^{-1}	A_1	H1 – La2
406 cm^{-1}	A_1	O1 – La1 – F1
333 cm^{-1}	E	O1 – F2
291 cm^{-1}	A_1	F1 – F2 – La1 – La2
248 cm^{-1}	E	F2 – O1 – La1
178 cm^{-1}	A_1	F1 – F2 – La1
153 cm^{-1}	E	F1 – La2
133 cm^{-1}	A_1	La1 – La2 – F1
82 cm^{-1}	E	La1 – La2 – F1

Supplementary Table S33: $Gd_2OF_2H_2$ (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1303 cm^{-1}	A_1	H2 – H1
1269 cm^{-1}	E	H1 – H2
788 cm^{-1}	E	H2 – H1
652 cm^{-1}	A_1	H1 – Gd2
477 cm^{-1}	A_1	O1 – F1 – Gd1
389 cm^{-1}	E	O1 – F2
320 cm^{-1}	A_1	F1 – F2 – Gd1 – Gd2
269 cm^{-1}	E	F2 – O1 – Gd1
200 cm^{-1}	A_1	F2 – F1 – Gd2
159 cm^{-1}	E	F1 – Gd1
153 cm^{-1}	A_1	F2 – Gd1 – Gd2
81 cm^{-1}	E	Gd1 – Gd2 – F2

Supplementary Table S34: $Gd_2OF_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1418 cm^{-1}	A_1	H2 – H1
1298 cm^{-1}	E	H1 – H2
799 cm^{-1}	E	H2 – H1 – O1
661 cm^{-1}	A_1	H1 – Gd2
448 cm^{-1}	A_1	O1 – Gd1 – F2
395 cm^{-1}	E	O1 – F1
333 cm^{-1}	A_1	F1 – F2 – Gd1 – Gd2
249 cm^{-1}	E	F2 – O1 – Gd1
206 cm^{-1}	A_1	F1 – F2 – Gd2
163 cm^{-1}	E	F1 – Gd2
128 cm^{-1}	A_1	Gd1 – Gd2 – F1
80 cm^{-1}	E	Gd1 – Gd2

Supplementary Table S35: Y_2OFClH_2 (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1272 cm^{-1}	E	H1 – H2
1233 cm^{-1}	A_1	H2 – H1
847 cm^{-1}	E	H1 – H2 – O1
681 cm^{-1}	A_1	H1 – Y2
488 cm^{-1}	A_1	F1 – O1 – Y1
403 cm^{-1}	E	O1 – F1 – Y1
315 cm^{-1}	A_1	F1 – Cl1 – Y1 – Y2
246 cm^{-1}	E	F1 – O1 – Y1
221 cm^{-1}	A_1	F1 – Cl1 – Y2
156 cm^{-1}	A_1	Cl1 – Y1 – Y2
152 cm^{-1}	E	Cl1 – Y2
96 cm^{-1}	E	Y1 – Y2 – Cl1

Supplementary Table S36: Y_2OFClH_2 (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1285 cm^{-1}	E	H1 – H2
1274 cm^{-1}	A_1	H2 – H1
856 cm^{-1}	E	H2 – H1 – O1
686 cm^{-1}	A_1	H1 – Y2
479 cm^{-1}	A_1	O1 – Y1 – F1
411 cm^{-1}	E	O1 – F1 – Y1
314 cm^{-1}	A_1	F1 – Cl1 – Y1 – Y2
240 cm^{-1}	A_1	Cl1 – F1 – Y2
231 cm^{-1}	E	F1 – Y2 – O1
154 cm^{-1}	A_1	Cl1 – F1 – Y1 – Y2
154 cm^{-1}	E	Cl1 – Y2
90 cm^{-1}	E	Y1 – Y2 – Cl1

Supplementary Table S37: La_2OFClH_2 (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1121 cm^{-1}	E	H1 – H2
1119 cm^{-1}	A_1	H2 – H1
762 cm^{-1}	E	H2 – H1
562 cm^{-1}	A_1	H1 – La2
438 cm^{-1}	A_1	O1 – F1 – La1
332 cm^{-1}	E	O1 – F1
256 cm^{-1}	A_1	F1 – Cl1 – La1 – La2
222 cm^{-1}	E	F1 – O1 – La1
180 cm^{-1}	A_1	Cl1 – F1 – La2
140 cm^{-1}	A_1	La1 – La2 – Cl1
128 cm^{-1}	E	Cl1 – La2
75 cm^{-1}	E	La1 – La2 – Cl1

Supplementary Table S38: La_2OFClH_2 (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1144 cm^{-1}	A_1	H2 – H1
1127 cm^{-1}	E	H1 – H2
768 cm^{-1}	E	H2 – H1 – O1
565 cm^{-1}	A_1	H1 – La2
435 cm^{-1}	A_1	O1 – La1 – F1
333 cm^{-1}	E	O1 – F1
258 cm^{-1}	A_1	F1 – Cl1 – La1 – La2
225 cm^{-1}	E	F1 – O1 – La1
176 cm^{-1}	A_1	F1 – Cl1 – La2
130 cm^{-1}	A_1	La1 – La2 – Cl1
128 cm^{-1}	E	Cl1 – La2
73 cm^{-1}	E	La1 – La2 – Cl1 – F1

Supplementary Table S39: Gd_2OFClH_2 (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1228 cm^{-1}	E	H1 – H2
1208 cm^{-1}	A_1	H2 – H1
789 cm^{-1}	E	H2 – H1
656 cm^{-1}	A_1	H1 – Gd2
474 cm^{-1}	A_1	O1 – F1 – Gd1
382 cm^{-1}	E	O1 – F1
289 cm^{-1}	A_1	F1 – Cl1 – Gd1 – Gd2
235 cm^{-1}	E	F1 – O1 – Gd1
195 cm^{-1}	A_1	Cl1 – F1 – Gd2
139 cm^{-1}	E	Cl1 – Gd2
131 cm^{-1}	A_1	Cl1 – Gd1 – Gd2
77 cm^{-1}	E	Gd1 – Gd2 – Cl1 – F1

Supplementary Table S40: Gd_2OFClH_2 (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1254 cm^{-1}	A_1	H2 – H1
1242 cm^{-1}	E	H1 – H2
808 cm^{-1}	E	H2 – H1 – O1
661 cm^{-1}	A_1	H1 – Gd2
464 cm^{-1}	A_1	O1 – Gd1 – F1
389 cm^{-1}	E	O1 – F1
286 cm^{-1}	A_1	F1 – Cl1 – Gd1 – Gd2
221 cm^{-1}	E	F1 – O1 – Gd1
215 cm^{-1}	A_1	Cl1 – F1 – Gd2
138 cm^{-1}	E	Cl1 – Gd2
126 cm^{-1}	A_1	Gd1 – Gd2 – Cl1 – F1
72 cm^{-1}	E	Gd1 – Gd2 – Cl1 – F1

Supplementary Table S41: $LaGdOFClH_2$ (P3m1). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1171 cm^{-1}	A_1	H2 – H1
1111 cm^{-1}	E	H1 – H2
656 cm^{-1}	A_1	H1 – La1
644 cm^{-1}	E	H2 – H1
468 cm^{-1}	A_1	O1 – F1 – Gd1
350 cm^{-1}	E	O1 – F1
275 cm^{-1}	A_1	F1 – Cl1 – La1 – Gd1
253 cm^{-1}	E	F1 – O1 – La1
184 cm^{-1}	A_1	Cl1 – F1 – Gd1
139 cm^{-1}	A_1	Cl1 – La1 – Gd1
121 cm^{-1}	E	Cl1 – Gd1
75 cm^{-1}	E	Gd1 – La1 – Cl1 – F1

Supplementary Table S42: $LaGdOFClH_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1199 cm^{-1}	A_1	H2 – H1
1117 cm^{-1}	E	H1 – H2
661 cm^{-1}	E	H2 – H1 – O1
660 cm^{-1}	A_1	H1 – Gd1
462 cm^{-1}	A_1	O1 – La1 – F1
352 cm^{-1}	E	O1 – F1
275 cm^{-1}	A_1	F1 – Cl1 – La1 – Gd1
249 cm^{-1}	E	F1 – O1 – La1
188 cm^{-1}	A_1	Cl1 – F1 – Gd1
134 cm^{-1}	A_1	Gd1 – La1 – Cl1 – F1
119 cm^{-1}	E	Cl1 – Gd1
72 cm^{-1}	E	La1 – Gd1 – Cl1 – F1

Supplementary Table S43: $Y_2OCl_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

Frequency	Mode symmetry	Vibrational displacements
1325 cm^{-1}	A_1	H2 – H1
1226 cm^{-1}	E	H1 – H2
787 cm^{-1}	E	H2 – H1 – O1
688 cm^{-1}	A_1	H1 – Y2
456 cm^{-1}	A_1	O1 – Y1
383 cm^{-1}	E	O1 – Y1
280 cm^{-1}	A_1	Cl1 – Cl2 – Y1 – Y2
250 cm^{-1}	A_1	Cl1 – Cl2 – Y2 – Y1
166 cm^{-1}	E	Cl2 – Y2 – O1
154 cm^{-1}	E	Cl1 – Y2
140 cm^{-1}	A_1	Cl2 – Cl1 – Y2 – Y1
84 cm^{-1}	E	Y1 – Y2 – Cl1

Supplementary Table S44: $La_2OCl_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

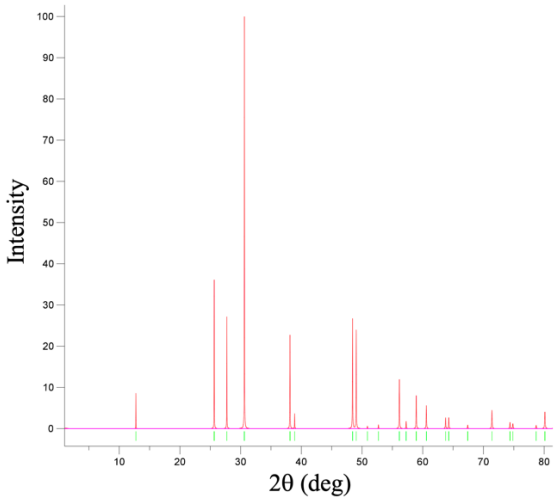
Frequency	Mode symmetry	Vibrational displacements
1228 cm^{-1}	A_1	H2 – H1
1073 cm^{-1}	E	H1 – H2
684 cm^{-1}	E	H2 – H1 – O1
584 cm^{-1}	A_1	H1 – La2
395 cm^{-1}	A_1	O1 – La1
305 cm^{-1}	E	O1 – Cl2
232 cm^{-1}	A_1	Cl1 – Cl2 – La1 – La2
211 cm^{-1}	A_1	Cl2 – Cl1 – La2
149 cm^{-1}	E	Cl2 – O1 – La1
134 cm^{-1}	E	Cl1 – La2
113 cm^{-1}	A_1	La1 – La2 – Cl1
66 cm^{-1}	E	La1 – La2 – Cl1 – Cl2

Supplementary Table S45: $Gd_2OCl_2H_2$ (R3m). $\Gamma_{optic} = 6A_1 + 6E$

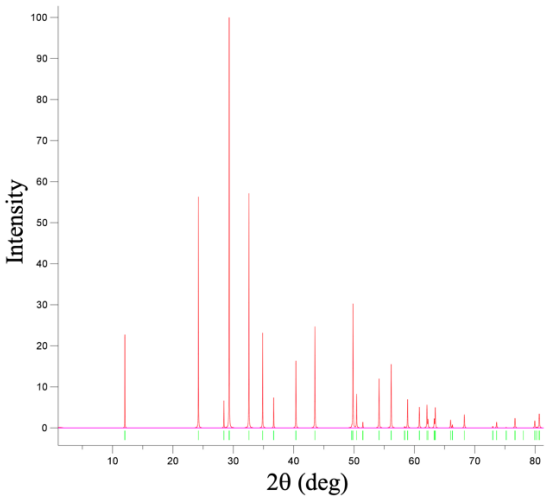
Frequency	Mode symmetry	Vibrational displacements
1304 cm^{-1}	A_1	H2 – H1
1189 cm^{-1}	E	H1 – H2
738 cm^{-1}	E	H2 – H1 – O1
661 cm^{-1}	A_1	H1 – Gd2
436 cm^{-1}	A_1	O1 – Gd1
361 cm^{-1}	E	O1 – Gd2
249 cm^{-1}	A_1	Cl1 – Cl2 – Gd1 – Gd2
230 cm^{-1}	A_1	Cl1 – Cl2 – Gd2
152 cm^{-1}	E	Cl2 – Gd1
140 cm^{-1}	E	Cl1 – Gd2
116 cm^{-1}	A_1	Gd1 – Gd2 – Cl1 – Cl2
68 cm^{-1}	E	Gd1 – Gd2 – Cl1 – Cl2

5. Simulation of X-ray diffraction patterns

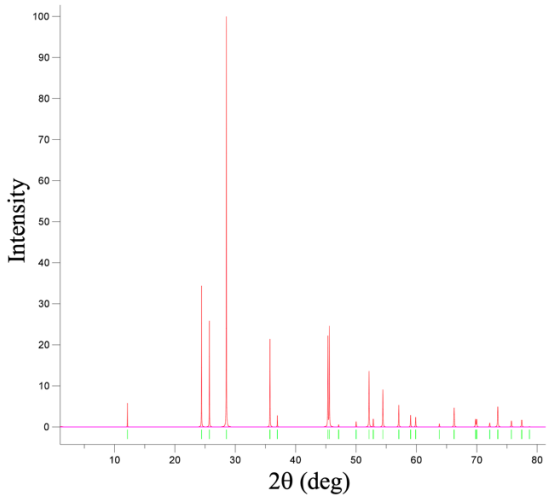
The Cu Kα monochromatic beam with wavelength λ=1.5406 Å was set as a parameter of calculation to determine the theoretical X-ray diffraction patterns. The illustrations have been made by using the VESTA program [4].



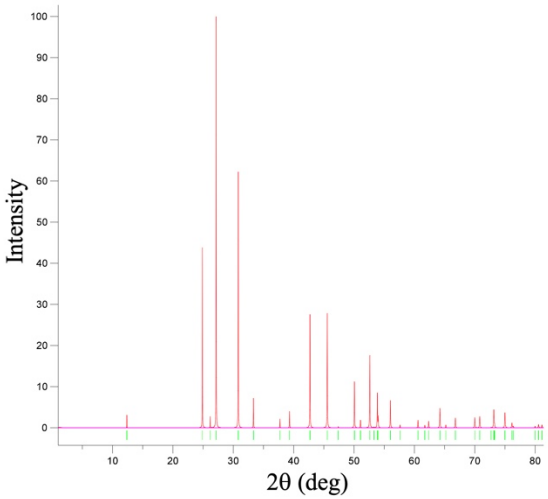
Supplementary Figure S3: $Y_2OF_2H_2$ (P3m1)



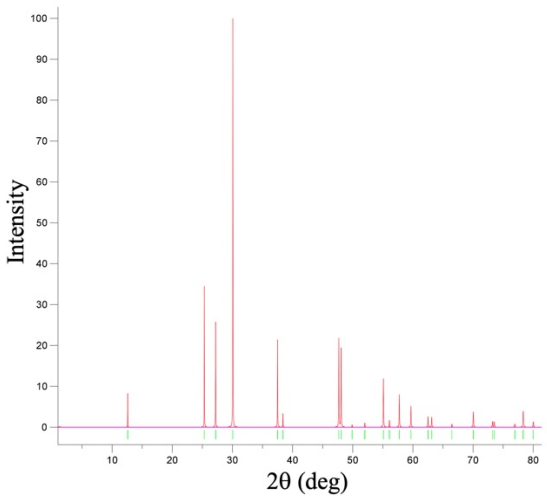
Supplementary Figure S4: $Y_2OF_2H_2$ (R3m)



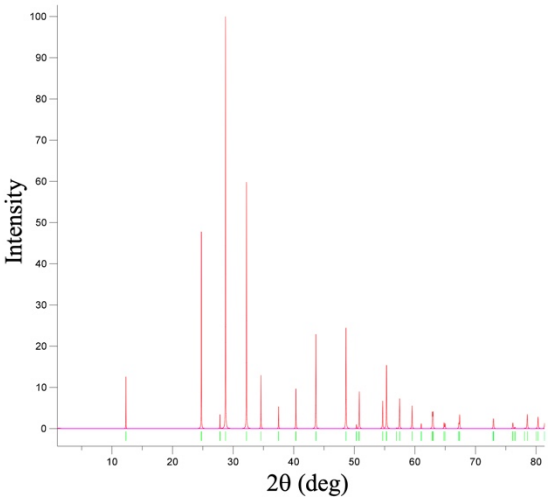
Supplementary Figure S5: $La_2OF_2H_2$ (P3m1)



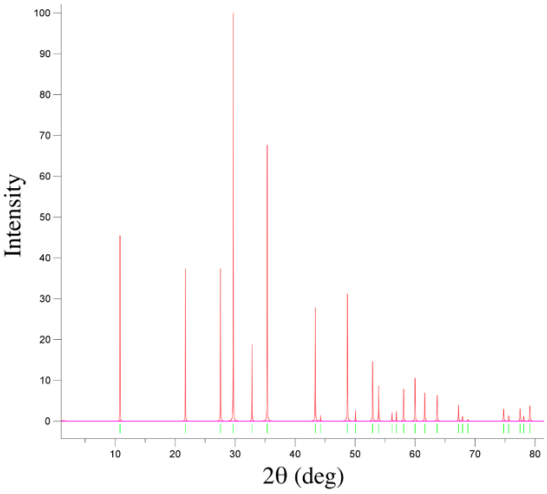
Supplementary Figure S6: $La_2OF_2H_2$ (R3m)



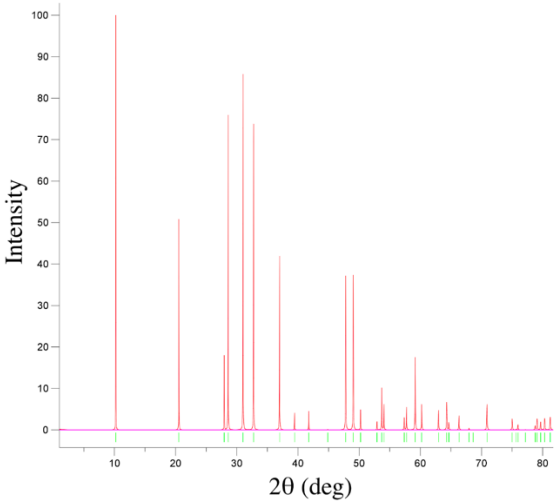
Supplementary Figure S7: $Gd_2OF_2H_2$ (P3m1)



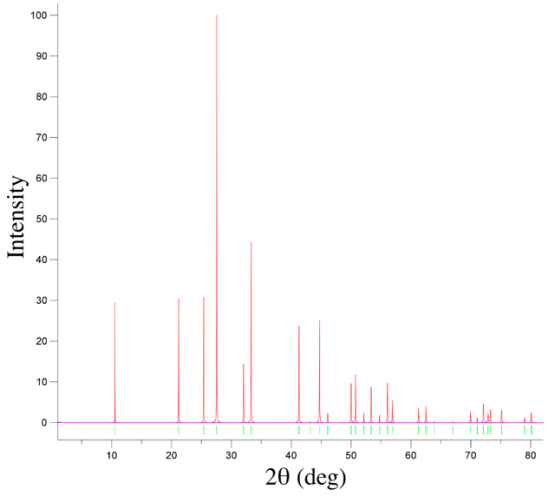
Supplementary Figure S8: $Gd_2OF_2H_2$ (R3m)



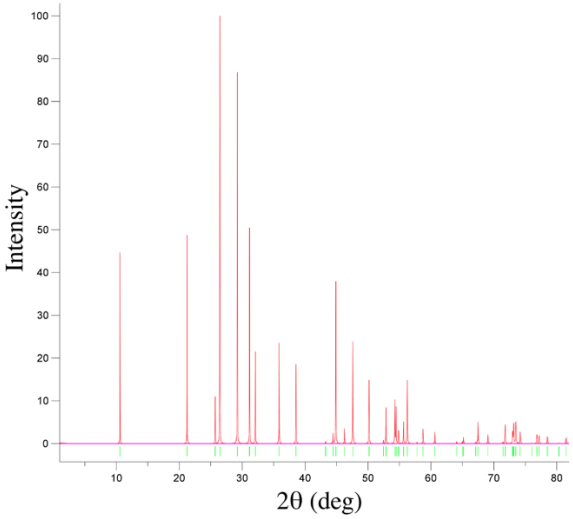
Supplementary Figure S9: Y_2OFClH_2 (P3m1)



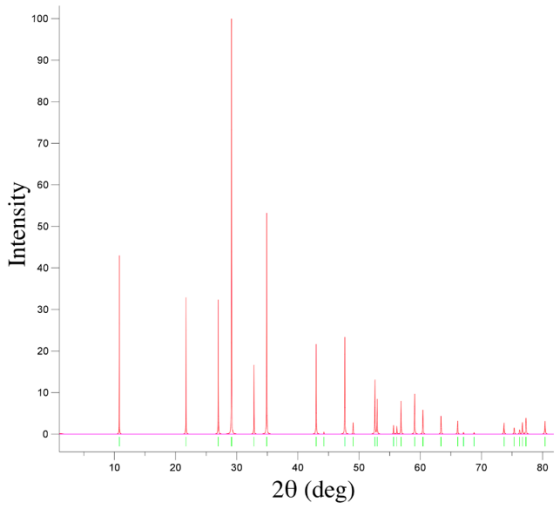
Supplementary Figure S10: Y_2OFClH_2 (R3m)



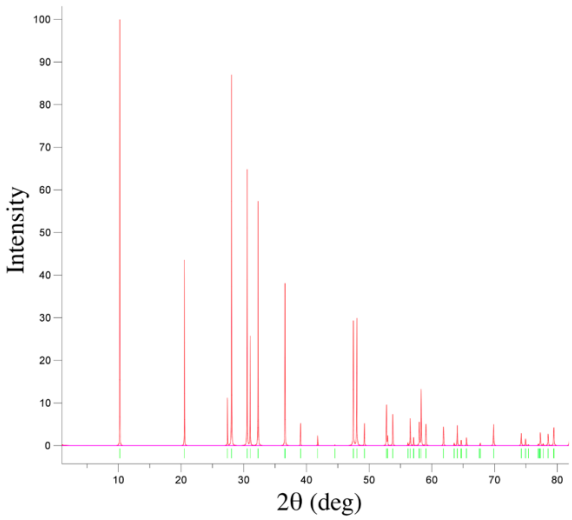
Supplementary Figure S11: La_2OFClH_2 (P3m1)



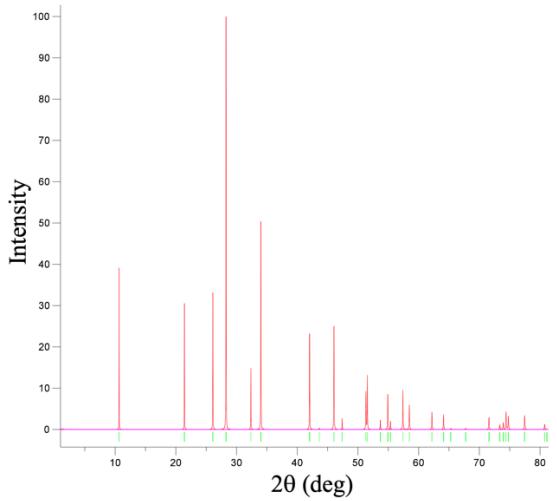
Supplementary Figure S12: La_2OFClH_2 (R3m)



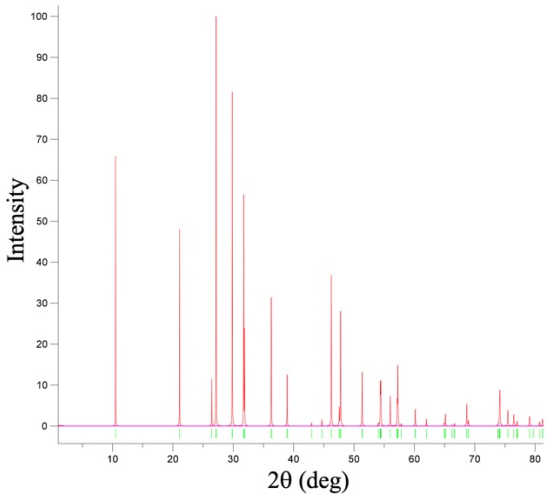
Supplementary Figure S13: Gd_2OFClH_2 (P3m1)



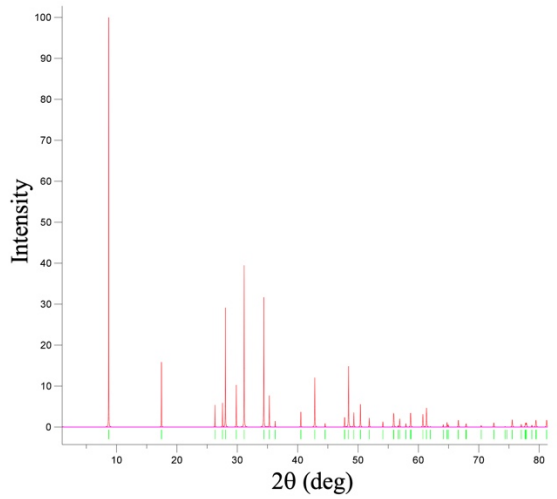
Supplementary Figure S14: Gd_2OFClH_2 (R3m)



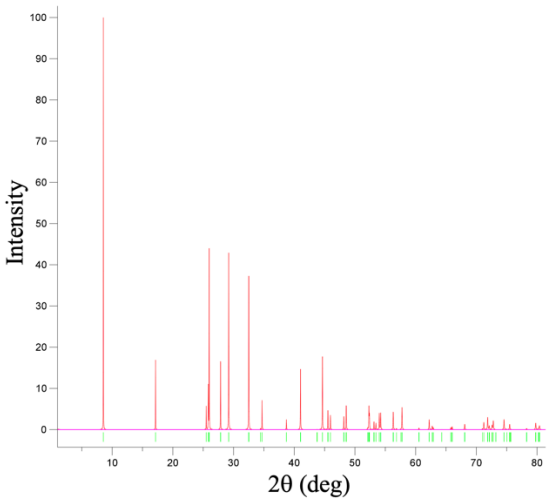
Supplementary Figure S15: $LaGdOFClH_2$ (P3m1)



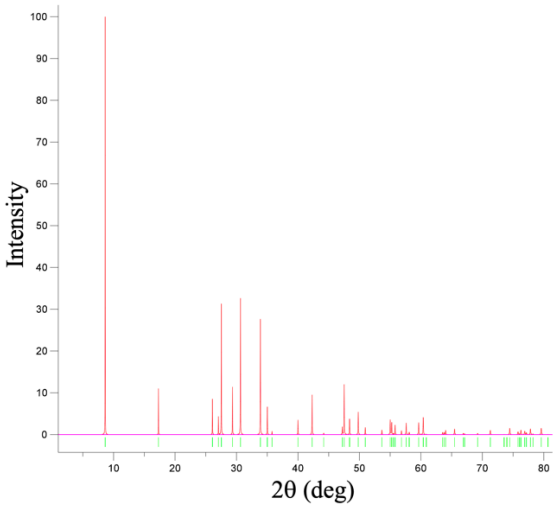
Supplementary Figure S16: $LaGdOFClH_2$ (R3m)



Supplementary Figure S17: $Y_2OCl_2H_2$ (R3m)



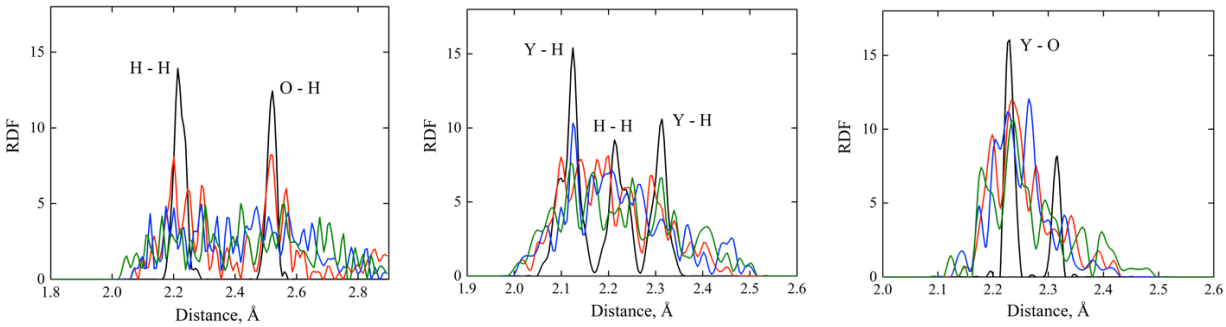
Supplementary Figure S18: $La_2OCl_2H_2$ (R3m)



Supplementary Figure S19: $Gd_2OCl_2H_2$ (R3m)

6. Radial distribution function evaluated for the shortest interatomic distances

Calculations have been performed for four temperatures: T= 12 K (results are shown in black), 100 K (shown in red), 200 K (shown in blue) and 300 K (shown in green).



Supplementary Figure S20: Radial distribution functions for the shortest interatomic distances between atoms in $Y_2OF_2H_2$ (R3m)

7. Piezoelectric properties of $Ln_2OF_{2-x}Cl_xH_2$ (Ln = Y, La, Gd)

Supplementary Table S46: Independent components of electronic and ionic contributions to direct piezoelectric tensor (eij) and dielectric tensor (εij) in $Ln_2OF_2H_2$

		e15	e22	e31	e33	ε11	ε33
$Y_2OF_2H_2$ (P3m1)	Elect.	-0.03	-0.03	0.08	-0.28	4.67	3.38
	Ion.	0.92	0.89	1.15	-0.06	13.21	10.09
$Y_2OF_2H_2$ (R3m)	Elect.	-0.04	0.10	0.002	0.04	4.47	3.13
	Ion.	-0.06	-0.82	0.22	-0.08	11.86	5.04
$La_2OF_2H_2$ (P3m1)	Elect.	-0.01	0.03	0.11	-0.34	4.48	3.44
	Ion.	0.60	-0.41	1.02	0.44	12.86	8.71
$La_2OF_2H_2$ (R3m)	Elect.	-0.02	0.07	-0.02	0.08	4.56	3.61
	Ion.	-0.09	-0.08	0.38	-0.62	13.91	10.96

Supplementary Table S47: Independent components of electronic and ionic contributions to direct piezoelectric tensor (eij) and dielectric tensor (εij) in Ln_2OFClH_2

		e15	e22	e31	e33	ε11	ε33
Y_2OFClH_2 (P3m1)	Elect.	0.01	0.002	0.07	-0.34	4.74	3.68
	Ion.	-0.03	0.25	0.23	-1.06	9.35	5.05
Y_2OFClH_2 (R3m)	Elect.	-0.01	0.03	0.02	-0.22	4.46	3.78
	Ion.	-0.14	-0.36	0.09	-0.43	9.26	2.44
La_2OFClH_2 (P3m1)	Elect.	0.04	0.01	0.10	-0.35	4.50	3.75
	Ion.	-0.05	-0.05	0.37	-0.54	10.13	5.74
La_2OFClH_2 (R3m)	Elect.	0.01	0.04	0.02	-0.21	4.55	3.65
	Ion.	-0.23	0.14	0.26	-0.89	10.69	5.39
Gd_2OFClH_2 (P3m1)	Elect.	0.02	-0.01	0.08	-0.37	4.70	3.80
	Ion.	-0.03	0.16	0.30	-0.99	10.10	5.49
Gd_2OFClH_2 (R3m)	Elect.	0.01	0.05	0.01	-0.20	4.62	3.34
	Ion.	-0.17	-0.24	0.12	-0.48	9.02	2.69
$LaGdOFClH_2$ (P3m1)	Elect.	0.06	0.003	0.05	-0.36	4.67	3.80
	Ion.	-0.05	0.20	0.36	-0.75	12.71	5.65
$LaGdOFClH_2$ (R3m)	Elect.	0.04	0.04	-0.02	-0.22	4.62	3.95
	Ion.	-0.22	-0.18	0.23	-0.82	12.51	4.31

Supplementary Table S48: Independent components of electronic and ionic contributions to direct piezoelectric tensor (eij) and dielectric tensor (εij) in $Ln_2OCl_2H_2$

		e15	e22	e31	e33	ε11	ε33
$Y_2OCl_2H_2$ (R3m)	Elect.	-0.02	0.004	0.02	-0.06	4.40	3.69
	Ion.	-0.01	-0.27	0.02	-0.04	8.05	0.96
$La_2OCl_2H_2$ (R3m)	Elect.	0.01	0.01	0.03	-0.04	4.25	3.40
	Ion.	-0.06	-0.10	0.06	-0.03	8.39	1.14
$Gd_2OCl_2H_2$ (R3m)	Elect.	-0.002	0.02	0.03	-0.04	4.42	3.65
	Ion.	-0.02	-0.22	0.02	-0.04	7.89	0.95

Supplementary Table S49: Comparison of the electromechanical properties of $Ln_2OF_{2-x}Cl_xH_2$ expressed in terms of piezoelectric voltage constant g_{ij} (10^{-3} ×[Vm/N]), piezoelectric coupling factors k_{ij} and k_t, and piezoelectric transduction coefficients $d_{ij}{\cdot}g_{ij}$ (10^{-15} ×[m^2/N])

Chem. Formula	Struc-ture	g_{33}	k_{33}	k_t	$d_{33}{\cdot}g_{33}$	g_{31}	k_{31}	$d_{31}{\cdot}g_{31}$	g_{15}	k_{15}	$d_{15}{\cdot}g_{15}$
$Y_2OF_2H_2$	P3m1	-64.8	23.9%	8.4%	500	58.3	23.2%	405	271	53.1%	11576
	R3m	-64.2	7.3%	3.1%	298	18.0	5.9%	23	9.3	1.9%	12
$La_2OF_2H_2$	P3m1	-22.7	7.7%	2.8%	55	57.8	21.2%	360	210.7	37.2%	6811
	R3m	-91.3	24.6%	17.6%	1074	27.1	10.7%	95	-22.1	4.9%	80
Y_2OFClH_2	P3m1	-600.0	94.4%	61.7%	27841	106.8	35.0%	882	-14.8	2.6%	27
	R3m	-723.3	69.3%	55.5%	26205	45.7	12.2%	105	-177	14.6%	3663
La_2OFClH_2	P3m1	-179.8	43.6%	33.3%	2719	60.4	18.8%	307	-0.9	0.1%	0.1
	R3m	-637.7	86.0%	58.9%	33367	93.6	28.2%	719	-143.6	19.4%	2809
Gd_2OFClH_2	P3m1	-427.5	79.5%	54.6%	15028	92.1	30.4%	697	-6.2	1.1%	5
	R3m	-770.9	73.7%	56.9%	31715	60.2	16.1%	193	-221.1	18.8%	5897
$LaGdOFClH_2$	P3m1	-238.0	54.4%	41.4%	4749	64.0	20.3%	342	-0.2	0.1%	0.1
	R3m	-724.3	88.9%	60.9%	38354	94.4	27.6%	651	-91.0	12.3%	1256
$Y_2OCl_2H_2$	R3m	-290.4	16.2%	15.7%	2999	10.4	2.1%	3.8	303.7	11.7%	9412
$La_2OCl_2H_2$	R3m	-249.6	13.8%	12.8%	2504	22.9	4.3%	21	-273.7	10.9%	8386
$Gd_2OCl_2H_2$	R3m	-260.6	14.5%	14.0%	2765	12.0	2.5%	6	-196.1	6.4%	4188

Supplementary Table S50: Comparison of the relaxed lattice geometry within the PBE+U approximation and the relevant experimental data for rare-earth oxides, fluorides, chlorides and hydrides

	La_2O_3, P-3m1		LaH_3, Fm-3m		LaF_3, P-3c1		$LaCl_3$, P63/m	
	PBE+U	exp.[5]	PBE+U	exp.[6]	PBE+U	exp.[7]	PBE+U	exp.[8]
a, Å	3.96	3.96	5.59	5.62	7.15	7.19	7.45	7.48
b, Å	3.96	3.96	5.59	5.62	7.15	7.19	7.45	7.48
c, Å	6.21	6.14	5.59	5.62	7.33	7.35	4.33	4.37
V, Å^3	84.17	83.24	174.7	177.5	324.3	328.6	208.4	211.2
	Gd_2O_3, Ia-3		GdH_3, P-3c1		GdF_3, Pnma		$GdCl_3$, P63/m	
	PBE+U	exp.[9]	PBE+U	exp.[10]	PBE+U	exp.[11]	PBE+U	exp.[8]
a, Å	10.85	10.79	6.44	6.47	6.55	6.57	7.32	7.37
b, Å	10.85	10.79	6.44	6.47	7.05	6.98	7.32	7.37
c, Å	10.85	10.79	6.70	6.72	4.48	4.39	4.05	4.11
V, Å^3	1276.7	1256.2	241.0	243.2	207.6	201.3	188.3	193.0

References:

1. R. Gaillac, P. Pullumbi, F.X. Coudert. J. Phys.: Condens. Matter. 2016, 28, 27.
2. R. Gaillac, F.X. Coudert, http://progs.coudert.name/elate
3. Kroumova, E.; Aroyo, M. I.; Perez-Mato, J. M.; Kirov, A.; Capillas, C.; Ivantchev, S.; Wondratschek, H. Bilbao Crystallographic Server I: Databases and crystallographic computing programs. *Phase Transitions* **2003**, *76*, 155–170.
4. Momma, K.; Izumi, F. VESTA 3 for three-dimensional visualization of crystal, volumetric and morphology data. *J. Appl. Crystallogr.* **2011**, *44*, 1272-1276
5. Cao Wang, Ming-qiu Tan, Chun-mu Feng, Zhi-feng Ma, Shuai Jiang, Zhu-an Xu, Guang-han Cao, Kazuyuki Matsubayashi, and Yoshiya Uwatoko. $La_2Co_2Se_2O_3$: A Quasi-Two-Dimensional Mott Insulator with Unusual Cobalt Spin State and Possible Orbital Ordering, *J. Am. Chem. Soc.*, **2010**, 132 (20), 7069-7073
6. H. Meng, M.A. Kuzovnikov, M. Tkacz, Phase stability of some rare earth trihydrides under high pressure, *Int. J. Hydrogen Energy*, **2017**, *42*, 29344-29349
7. Cheetham, A. K., Fender, B. E. F., Fuess, H. & Wright, A. F. A powder neutron diffraction study of lanthanum and cerium trifluorides, *Acta Cryst.*,**1976**, *B32*, 94-97
8. B. Morosin. Crystal Structures of Anhydrous Rare-Earth Chlorides, *J. Chem. Phys.*, **1968**, *49*, 3007-3012
9. Bartos, A., Lieb, K. P., Uhrmacher, M. & Wiarda, D. Refinement of atomic positions in bixbyite oxides using perturbed angular correlation spectroscopy, *Acta Cryst.* **1993**, *B49*, 165-169.
10. M. Ellner, H. Reule, E.J. Mittemeijer, The structure of the trihydride GdH_3, *J. Alloys Compd.*, **2000**, *309*, 1–2, 127-131
11. I.M. Ranieri, S.L. Baldochi, D. Klimm, The phase diagram GdF3–LuF3, *J. Solid State Chem.*, **2008**, *181*, 5, 1070-1074