Supporting Information

For **Crystallography, DFT study and nature of the bonding in the series of AnO_2^{2+} acetates (An = U, Np, Pu)**

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Table S1. Hydrogen bonds for **U-RT** structure.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
O1W	H1A	O2W	0.848(19)	1.82(3)	2.591(6)	151(6)
O1W	H1B	O4[1]	0.828(19)	2.29(3)	2.990(4)	143(3)
O1W	H1B	O4[2]	0.828(19)	2.29(3)	2.990(4)	143(3)
O2W	H2	O3[3]	0.852(16)	1.942(14)	2.789(3)	173(3)

[1]1-X,-1/2+Y,1-Z; [2]1-X,1-Y,1-Z; [3]3/2-X,-Y,1/2+Z

Table S2. Hydrogen bonds for **U-LT** structure.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
O1W	H1A	O6[1]	0.84(3)	1.96(3)	2.780(8)	166(8)
O1W	H1B	O2W	0.84(3)	1.79(4)	2.584(8)	157(9)
O2W	H2D	O4[2]	0.84(3)	1.96(4)	2.762(8)	159(9)
O2W	H2E	O3[3]	0.86(3)	1.92(3)	2.774(8)	173(10)

[1]1-X,1-Y,1-Z; [2]-1/2+X,1/2-Y,-1/2+Z; [3]1/2+X,1/2-Y,-1/2+Z

Table S3. Hydrogen bonds for **Np-LT** structure.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
O1W	H1A	O6[1]	0.85(3)	1.96(5)	2.775(8)	160(10)
O1W	H1B	O2W	0.85(3)	1.83(6)	2.600(8)	150(10)
O2W	H2E	O3[2]	0.84(3)	1.93(3)	2.772(8)	173(10)
O2W	H2D	O4[3]	0.84(3)	1.93(3)	2.761(8)	168(9)

[1]1-X,1-Y,1-Z; [2]1/2+X,1/2-Y,-1/2+Z; [3]-1/2+X,1/2-Y,-1/2+Z

Table S4. Hydrogen bonds for **Pu-LT** structure.

D	H	A	d(D-H)/Å	d(H-A)/Å	d(D-A)/Å	D-H-A/°
O1W	H1A	O6[1]	0.85(2)	1.94(3)	2.772(11)	167(11)
O1W	H1B	O2W	0.85(2)	1.76(2)	2.608(11)	176(14)
O2W	H2D	O4[2]	0.85(2)	1.91(3)	2.751(11)	172(14)
O2W	H2E	O3[3]	0.85(2)	1.96(4)	2.773(11)	160(12)

[1]1-X,1-Y,1-Z; [2]-1/2+X,1/2-Y,-1/2+Z; [3]1/2+X,1/2-Y,-1/2+Z

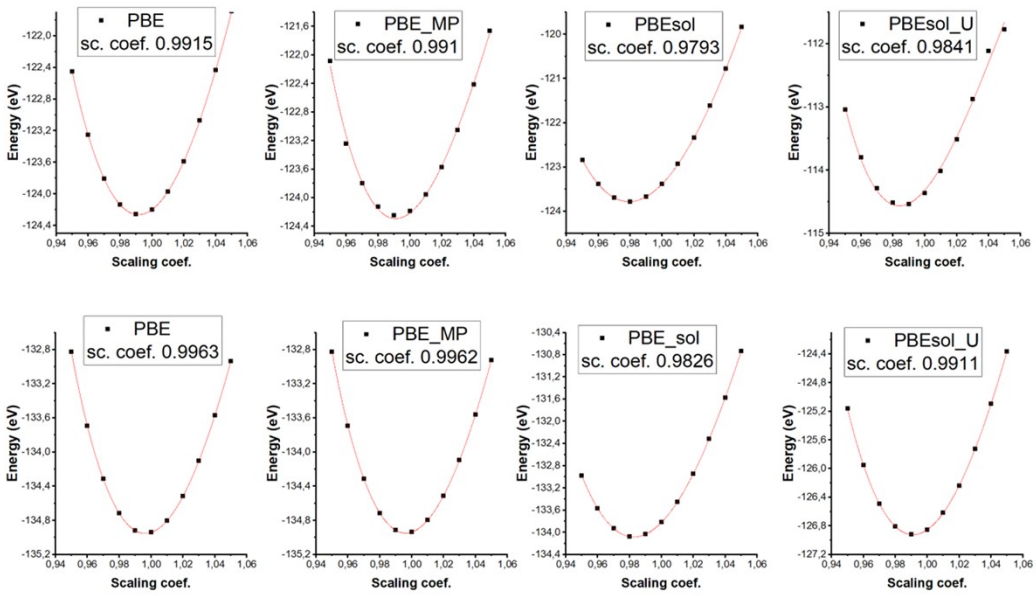

Figure S1. Energy vs. lattice scaling coefficient curves for **UO_2_fcc** (top) and **PuO_2_fcc** (bottom) obtained within different exchange-correlation functionals; black dots – data points, red solid line – bets fit of Birch–Murnaghan (EOS). Tolerance level for Birch–Murnaghan (EOS) fit > 0.99 for all considered levels of theory.

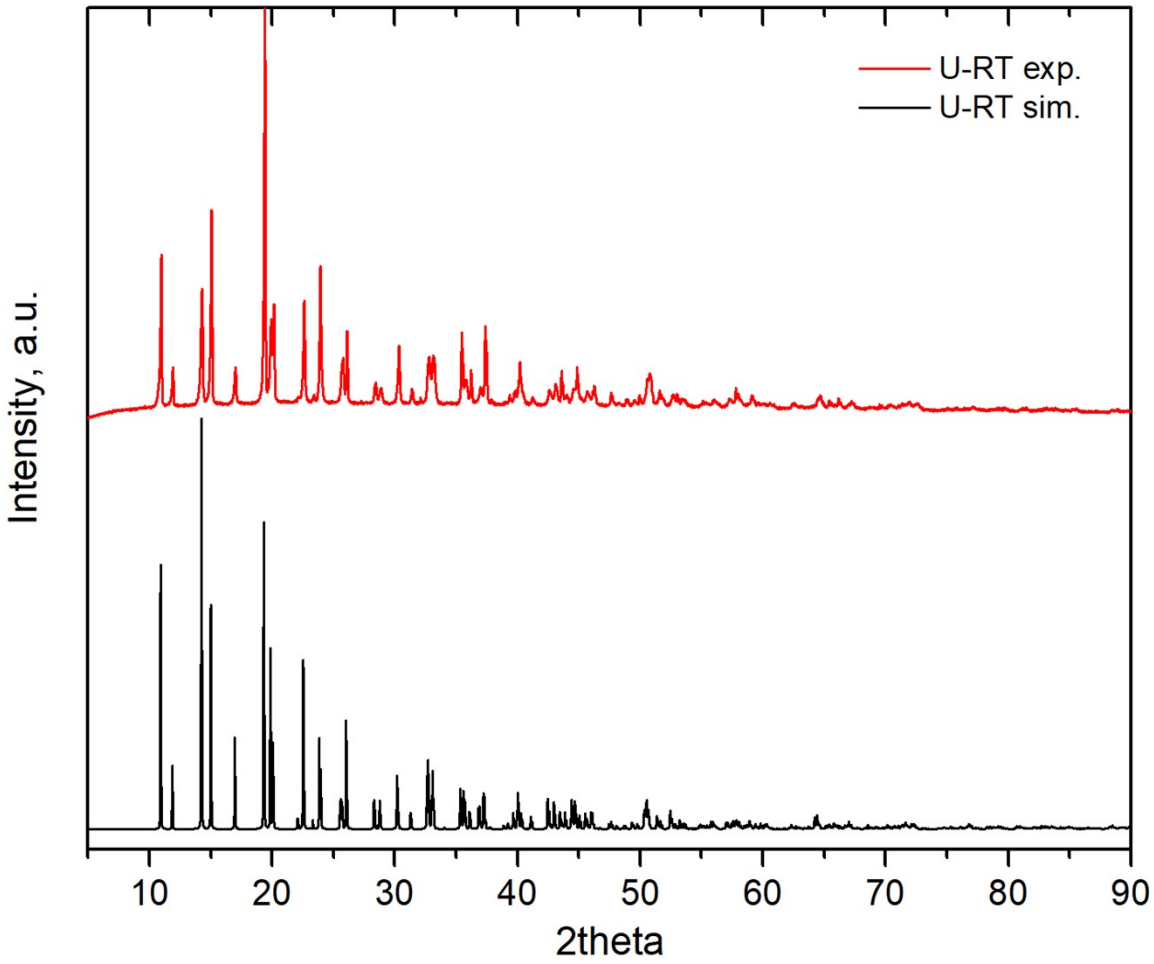

Figure S2. Experimental and simulated powder X-ray diffraction of **U-RT**.

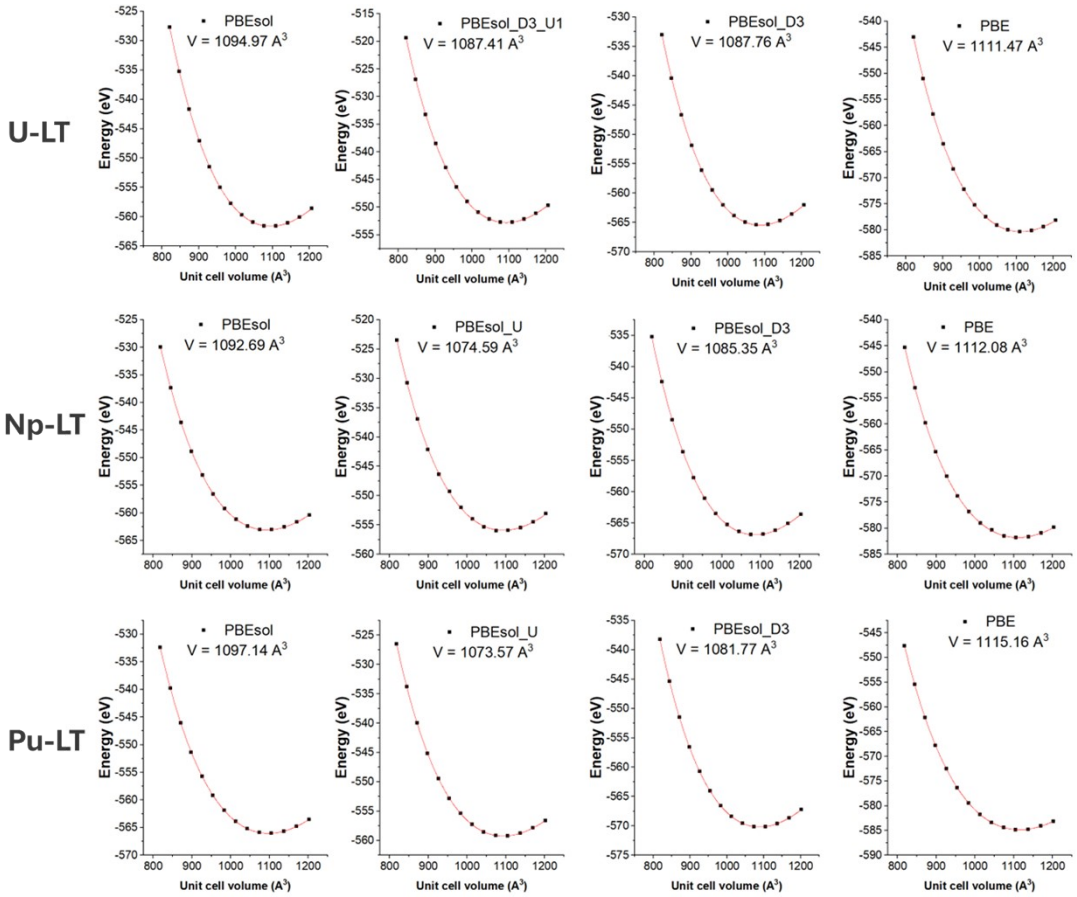

Figure S3. Energy vs. unit cell volume plots for An-LT structures along with polymorph obtained within different approximations. Tolerance level for Birch–Murnaghan (EOS) fit > 0.99 for all considered levels of theory.

Table S5. XYZ coordinates

U-LT	U	1.392580	8.529243	13.465688
	O	1.407637	6.747258	13.669539
	O	2.555199	8.310721	11.288173
	O	0.360019	8.336557	11.244625
	O	6.151894	8.611104	13.748248
	O	3.923419	8.721926	13.842066
	H	1.329627	9.442599	8.832251
	H	0.680729	7.775392	8.695177
	H	2.462948	8.049966	8.722220
	H	3.999934	6.610245	12.337228
	H	4.838376	5.992975	13.772204
	H	5.809028	6.480020	12.360166
	C	1.472560	8.321260	10.601171
	C	1.493335	8.385932	9.116185
	C	5.006577	8.095611	13.533482
	C	4.909755	6.722081	12.944578
	O	-0.984001	8.611104	13.748248
	O	1.430234	8.720440	15.773752
	O	1.441426	10.312594	13.308285

	C -2.129317 8.095611 13.533482 H 1.500113 9.618173 16.253253 H 1.319034 7.972668 16.483499 O -3.212476 8.721926 13.842066 C -2.226140 6.722081 12.944578 H -3.135961 6.610245 12.337228 H -1.326867 6.480020 12.360166 H -2.297519 5.992975 13.772204
Np-LT	Np 1.355090 8.625499 13.285503 O 1.356092 6.862636 13.400864 O 2.515228 8.477619 11.093237 O 0.312905 8.502604 11.065224 O 6.059789 8.718692 13.589544 O 3.835486 8.804219 13.684540 H 1.396869 9.541075 8.604207 H 0.550804 7.967518 8.536165 H 2.356358 8.026120 8.542623 H 3.951060 6.759162 12.069926 H 4.732538 6.086189 13.515361 H 5.754818 6.617304 12.153208 C 1.423837 8.473861 10.421805 C 1.436333 8.486154 8.931965 C 4.923699 8.200194 13.351126 C 4.838036 6.843450 12.716059 O -1.032387 8.718692 13.589544 O 1.388751 8.667526 15.643781 O 1.381445 10.387133 13.189942 C -2.168476 8.200194 13.351126 H 1.267066 7.884334 16.298288 H 1.455368 9.523757 16.184792 O -3.256690 8.804219 13.684540 C -2.254140 6.843450 12.716059 H -3.141115 6.759162 12.069926 H -1.337357 6.617304 12.153208 H -2.359637 6.086189 13.515361
Pu-LT	Pu 4.923809 1.268520 2.037065 O 4.960320 3.005577 1.931013 O 3.782362 1.392275 4.238962 O 5.980061 1.444269 4.263404 O 0.225674 1.134406 1.723382 O 2.450792 1.063103 1.626551 H 5.036424 0.376673 6.729632 H 5.671052 2.047036 6.799749 H 3.893108 1.753862 6.791958 H 2.331345 3.110649 3.217999 H 1.529619 3.772426 1.775234 H 0.531326 3.251060 3.163291 C 4.872761 1.432529 4.909141 C 4.862630 1.416819 6.400344 C 1.355847 1.663966 1.955803

	C	1.437309	3.022299	2.582861
	O	4.871431	-0.469123	2.136776
	O	4.927703	1.217327	-0.287063
	O	7.300794	1.134406	1.723382
	H	4.893593	0.349661	-0.815963
	H	5.044883	1.997274	-0.946517
	C	8.430966	1.663966	1.955803
	C	8.512428	3.022299	2.582861
	O	9.525912	1.063103	1.626551
	H	8.604738	3.772426	1.775234
	H	7.606446	3.251060	3.163291
	H	9.406465	3.110649	3.217999