Structure and Properties of Cubic PuH_2 and PuH_3: A Density Functional Theory Study

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Supporting Information

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Table S1. ***k***-point and cut off convergence testing for cubic PuH_2 and PuH_3 using PBEsol+U+SOC.

k-point Convergence Testing					
	Differences in Energy per PuH_2 unit (eV)				
Magnetic Configuration	(7×7×7) - (5×5×5)	(9×9×9) - (7×7×7)	(11×11×11) - (9×9×9)	(13×13×13) - (11×11×11)	(15×15×15) - (13×13×13)
FM <111>	-9.45×10^{-4}	6.82×10^{-4}	1.28×10^{-4}	9.70×10^{-4}	-1.37×10^{-4}
L3kAFM	1.00×10^{-3}	-2.65×10^{-4}	1.02×10^{-4}	2.65×10^{-4}	-1.18×10^{-4}
T3kAFM	9.20×10^{-4}	-2.85×10^{-4}	9.5×10^{-5}	2.68×10^{-4}	-1.18×10^{-4}
	Differences in Energy per PuH_3 unit (eV)				
FM <111>	-1.42×10^{-3}	5.45×10^{-4}	-1.92×10^{-4}	2.70×10^{-5}	1.84×10^{-3}
L3kAFM	-8.10×10^{-4}	1.05×10^{-4}	7.00×10^{-6}	-5.50×10^{-5}	2.17×10^{-4}
T3kAFM	-9.12×10^{-4}	1.15×10^{-4}	8.00×10^{-6}	-5.30×10^{-5}	2.15×10^{-4}
Cut off Convergence Testing					
	Differences in Energy per PuH_2 unit (eV)				
	500 – 400 (eV)	600 – 500 (eV)	700 – 600 (eV)	800 – 700 (eV)	900 – 800 (eV)	1000 – 900 (eV)
FM <111>	-7.69×10^{-3}	-2.15×10^{-3}	-3.48×10^{-4}	-1.01×10^{-3}	-1.60×10^{-4}	-6.30×10^{-5}
L3kAFM	-7.67×10^{-3}	-2.12×10^{-3}	-7.57×10^{-4}	-5.92×10^{-4}	-1.63×10^{-4}	-5.70×10^{-5}
T3kAFM	-7.66×10^{-3}	-2.11×10^{-3}	-7.60×10^{-4}	-5.92×10^{-4}	-1.60×10^{-4}	-6.00×10^{-5}
	Differences in Energy per PuH_3 unit (eV)				
FM <111>	-8.06×10^{-3}	-2.69×10^{-3}	-8.70×10^{-4}	-6.37×10^{-4}	-1.75×10^{-4}	-7.70×10^{-5}
L3kAFM	-8.03×10^{-3}	-2.68×10^{-3}	-8.72×10^{-4}	-6.30×10^{-4}	-1.77×10^{-4}	-7.02×10^{-5}
T3kAFM	-8.00×10^{-3}	-2.68×10^{-3}	-8.72×10^{-4}	-6.33×10^{-4}	-1.75×10^{-4}	-7.30×10^{-5}

Section S2: Energetics and structural properties of cubic PuH_2 and PuH_3 calculated with PBEsol+*U*+SOC+D3

Table S2. Relative energies per formula unit, calculated when considering van der Waal's dispersion corrections (D3) of Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV), for cubic PuH_2 and PuH_3 with: FM <100>, <110> and <111>; longitudinal AFM 1k, 2k and 3k; and transverse AFM 1k, 2k and 3k magnetic orders.

Magnetic Order	PuH_x	PBEsol+*U*+SOC+D3 - *U* (eV)							
		0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
FM <100>	PuH_2	0.065	0.044	0.047	0.048	0.050	0.053	0.057	0.062
	PuH_3	0.000	0.010	0.014	0.022	0.028	0.033	0.039	0.048
FM <110>	PuH_2	0.054	0.042	0.019	0.009	0.004	0.001	0.000	0.000
	PuH_3	0.003	0.000	0.063	0.048	0.039	0.031	0.025	0.020
FM <111>	PuH_2	0.051	0.050	0.028	0.020	0.017	0.016	0.018	0.020
	PuH_3	0.002	0.005	0.000	0.000	0.000	0.000	0.087	0.090
L1kAFM	PuH_2	0.031	0.030	0.029	0.034	0.039	0.045	0.052	0.059
	PuH_3	0.053	0.039	0.046	0.045	0.045	0.046	0.049	0.054
L2kAFM	PuH_2	0.030	0.023	0.007	0.003	0.001	0.001	0.002	0.003
	PuH_3	0.074	0.061	0.041	0.025	0.015	0.070	0.056	0.044
L3kAFM	PuH_2	0.029	0.029	0.018	0.016	0.016	0.017	0.088	0.070
	PuH_3	0.083	0.098	0.067	0.046	0.035	0.028	0.025	0.026
T1kAFM	PuH_2	0.005	0.004	0.023	0.034	0.042	0.048	0.055	0.062
	PuH_3	0.052	0.061	0.054	0.049	0.044	0.036	0.031	0.029
T2kAFM	PuH_2	0.000	0.000	0.000	0.000	0.000	0.000	0.001	0.003
	PuH_3	0.052	0.063	0.037	0.020	0.011	0.004	0.000	0.000
T3kAFM	PuH_2	0.000	0.008	0.008	0.009	0.011	0.013	0.017	0.094
	PuH_3	0.076	0.075	0.051	0.034	0.026	0.020	0.018	0.020

Table S3. Spin, orbital and total magnetic moment, calculated when considering van der Waal's dispersion corrections (D3) of Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV), for cubic PuH_2 and PuH_3 with: FM <100>, <110> and <111>; longitudinal AFM 1k, 2k and 3k; and transverse AFM 1k, 2k and 3k magnetic orders.

Magnetic Order	PuH_x	PBEsol+*U*+SOC+D3 - *U* (eV)							
		0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
		μ_{spin} (μ_B/ Pu ion)							
FM <100>	PuH_2	4.56	4.67	4.66	4.70	4.75	4.79	4.83	4.87
	PuH_3	4.56	4.60	4.62	4.65	4.68	4.72	4.76	4.80
FM <110>	PuH_2	4.55	4.66	4.66	4.70	4.74	4.78	4.82	4.86
	PuH_3	4.57	4.60	4.64	4.68	4.72	4.76	4.80	4.84
FM <111>	PuH_2	4.54	4.69	4.70	4.74	4.78	4.82	4.86	4.90
	PuH_3	4.57	4.63	4.66	4.70	4.73	4.76	4.76	4.80
L1kAFM	PuH_2	4.47	4.58	4.64	4.69	4.73	4.78	4.82	4.86
	PuH_3	4.48	4.57	4.61	4.64	4.68	4.73	4.77	4.81
L2kAFM	PuH_2	4.45	4.55	4.62	4.67	4.72	4.76	4.80	4.84
	PuH_3	4.44	4.52	4.57	4.61	4.66	4.73	4.79	4.84
L3kAFM	PuH_2	4.44	4.57	4.65	4.71	4.76	4.80	4.77	4.82
	PuH_3	4.44	4.57	4.62	4.67	4.71	4.76	4.80	4.85
T1kAFM	PuH_2	4.42	4.54	4.62	4.68	4.73	4.77	4.81	4.85
	PuH_3	4.46	4.55	4.59	4.63	4.68	4.73	4.78	4.83
T2kAFM	PuH_2	4.42	4.56	4.62	4.68	4.72	4.76	4.80	4.84
	PuH_3	4.46	4.52	4.56	4.62	4.67	4.71	4.75	4.80
T3kAFM	PuH_2	4.42	4.58	4.66	4.72	4.76	4.81	4.85	4.82
	PuH_3	4.47	4.56	4.61	4.67	4.71	4.76	4.80	4.85
		$\mu_{orbital}$ (μ_B / Pu ion)							
FM <100>	PuH_2	2.50	2.94	3.24	3.35	3.44	3.53	3.61	3.70
	PuH_3	2.38	2.78	2.99	3.13	3.26	3.37	3.48	3.59
FM <110>	PuH_2	2.55	2.91	3.22	3.36	3.48	3.58	3.67	3.75
	PuH_3	2.42	2.74	3.33	3.49	3.60	3.69	3.76	3.82
FM <111>	PuH_2	2.51	2.95	3.25	3.38	3.47	3.56	3.64	3.71
	PuH_3	2.43	2.82	3.04	3.21	3.35	3.46	4.34	4.44
L1kAFM	PuH_2	2.37	2.94	3.18	3.30	3.40	3.49	3.58	3.67
	PuH_3	2.24	2.65	2.89	3.04	3.17	3.28	3.40	3.52
L2kAFM	PuH_2	2.32	2.89	3.14	3.29	3.41	3.52	3.62	3.71
	PuH_3	2.22	2.68	2.94	3.13	3.28	3.64	3.71	3.78
L3kAFM	PuH_2	2.33	2.94	3.16	3.30	3.40	3.50	2.70	3.72
	PuH_3	2.21	2.74	2.98	3.14	3.26	3.38	3.48	3.58
T1kAFM	PuH_2	2.38	2.87	3.13	3.26	3.37	3.46	3.56	3.66
	PuH_3	2.15	2.66	2.90	3.09	3.26	3.39	3.49	3.58
T2kAFM	PuH_2	2.44	2.83	3.10	3.27	3.40	3.51	3.61	3.71
	PuH_3	2.15	2.73	2.98	3.15	3.29	3.42	3.54	3.65
T3kAFM	PuH_2	2.41	2.88	3.15	3.30	3.41	3.51	3.59	2.75
	PuH_3	2.27	2.83	3.05	3.20	3.32	3.43	3.52	3.62
		μ_{total} (μ_B / Pu ion)							
FM <100>	PuH_2	2.07	1.73	1.42	1.36	1.31	1.26	1.22	1.17
	PuH_3	2.18	1.82	1.63	1.52	1.43	1.35	1.28	1.21
FM <110>	PuH_2	2.00	1.75	1.44	1.34	1.26	1.20	1.15	1.11
	PuH_3	2.15	1.86	1.31	1.19	1.12	1.07	1.04	1.03
FM <111>	PuH_2	2.02	1.74	1.44	1.36	1.30	1.26	1.23	1.19
	PuH_3	2.15	1.81	1.62	1.48	1.38	1.30	0.42	0.35
L1kAFM	PuH_2	2.10	1.63	1.46	1.39	1.34	1.29	1.24	1.18
	PuH_3	2.24	1.92	1.72	1.61	1.52	1.44	1.37	1.29
L2kAFM	PuH_2	2.13	1.66	1.48	1.38	1.31	1.24	1.18	1.13
	PuH_3	2.22	1.84	1.63	1.48	1.39	1.09	1.08	1.06
L3kAFM	PuH_2	2.11	1.63	1.49	1.41	1.36	1.31	2.08	1.09
	PuH_3	2.22	1.83	1.64	1.52	1.45	1.38	1.32	1.27
T1kAFM	PuH_2	2.03	1.67	1.49	1.42	1.36	1.31	1.25	1.20
	PuH_3	2.31	1.89	1.68	1.55	1.42	1.34	1.29	1.25
T2kAFM	PuH_2	1.98	1.73	1.52	1.40	1.32	1.25	1.19	1.13
	PuH_3	2.31	1.79	1.58	1.47	1.38	1.29	1.22	1.15
T3kAFM	PuH_2	2.01	1.71	1.52	1.42	1.35	1.30	1.25	2.07
	PuH_3	2.19	1.72	1.56	1.46	1.39	1.33	1.28	1.23

Table S4. Structural symmetries, obtaining from the calculations when considering van der Waal's dispersion corrections (D3) of Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV) with a tolerance of 10^{-5} Å, for cubic PuH_2 and PuH_3 with: FM <100>, <110> and <111>; longitudinal AFM 1k, 2k and 3k; and transverse AFM 1k, 2k and 3k orders. For PuH_3, the space groups are the same as PuH_2 apart from the labels in brackets, which refers to the space group for PuH_3 when it differs from that of PuH_2. Space group numbers are attributed to the respective structural symmetries as: *I*4/*mmm* (139), $P\bar{1}$ (2), *C*2/*m* (12), *Immm* (71), $R\bar{3}m$ (166), $Fm\bar{3}m$ (225), *Fmmm* (69), *Cmca* (64), *Pbca* (61) and $Pa\bar{3}$ (205).

Magnetic order	**PBEsol+*U*+SOC+D3 – *U* (eV)**							
	0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
FM <100>	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*
FM <110>	*C*2/*m*	*C*2/*m*	*C*2/*m*	*C*2/*m*	*C*2/*m*	*C*2/*m*	*C*2/*m*	*C*2/*m*
FM <111>	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$	$R\bar{3}m$
L1kAFM	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*
L2kAFM	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*
L3kAFM	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$	$Fm\bar{3}m$
T1kAFM	*Fmmm* (*Cmca*)	*Fmmm*	*Fmmm*	*Fmmm*	*Fmmm*	*Fmmm*	*Fmmm*	*Fmmm*
T2kAFM	*Cmca*	*Cmca*	*Cmca*	*Cmca*	*Cmca*	*Cmca*	*Cmca*	*Cmca*
T3kAFM	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$	$Pa\bar{3}$

Table S5. Cell lengths and cell angles, calculated when considering van der Waal's dispersion corrections (D3) of Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV), for cubic PuH_2. a, b and c in Å. Cell volume in $Å^3$. α, β, γ in degree.

Magnetic order	Parameter	PBEsol+*U*+SOC+D3 - *U* (eV)							
		0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
FM <100>	a	5.11	5.19	5.22	5.25	5.27	5.29	5.31	5.33
	b	5.11	5.1	5.22	5.25	5.27	5.29	5.31	5.33
	c	5.11	5.19	5.22	5.25	5.27	5.29	5.30	5.31
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	133.51	138.28	142.17	144.65	146.65	148.23	149.58	150.67
FM <110>	a	5.10	5.18	5.23	5.26	5.28	5.30	5.31	5.32
	b	5.10	5.18	5.23	5.26	5.28	5.30	5.31	5.32
	c	5.14	5.18	5.21	5.24	5.27	5.30	5.31	5.32
	α	90.00	90.00	90.00	90.00	90.00	89.99	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.01	90.00	90.00
	γ	89.49	89.66	89.76	89.84	89.90	89.98	90.01	90.04
	Lattice Volume	133.38	139.07	142.66	145.15	147.05	148.66	149.89	150.79
FM <111>	a	5.11	5.18	5.22	5.25	5.28	5.30	5.31	5.32
	b	5.11	5.18	5.22	5.25	5.28	5.30	5.31	5.32
	c	5.11	5.18	5.22	5.25	5.28	5.30	5.31	5.32
	α	89.60	89.76	89.80	89.87	89.89	89.93	89.97	89.98
	β	89.60	89.76	89.80	89.87	89.89	89.93	89.97	89.98
	γ	89.60	89.76	89.80	89.87	89.89	89.93	89.97	89.98
	Lattice Volume	133.24	139.16	142.60	145.01	146.98	148.54	149.77	150.72
L1kAFM	a	5.01	5.09	5.17	5.21	5.24	5.27	5.29	5.31
	b	5.01	5.09	5.17	5.21	5.24	5.27	5.29	5.31
	c	5.26	5.29	5.30	5.30	5.32	5.33	5.34	5.34
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.94	137.17	141.38	144.20	146.36	147.99	149.37	150.40
L2kAFM	a	5.11	5.18	5.23	5.26	5.29	5.31	5.32	5.33
	b	5.11	5.18	5.23	5.26	5.29	5.31	5.32	5.33
	c	5.0582	5.14	5.18	5.22	5.25	5.28	5.29	5.30
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	132.15	138.00	141.89	144.69	146.82	148.51	149.78	150.87
L3kAFM	a	5.09	5.16	5.22	5.25	5.27	5.30	5.31	5.34
	b	5.09	5.16	5.22	5.25	5.27	5.30	5.31	5.34
	c	5.09	5.16	5.22	5.25	5.27	5.30	5.31	5.34
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	132.19	137.77	141.85	144.65	146.59	148.67	149.80	151.87
T1kAFM	a	5.13	5.16	5.20	5.23	5.26	5.28	5.30	5.31
	b	5.14	5.17	5.20	5.23	5.26	5.28	5.30	5.31
	c	4.99	5.12	5.21	5.26	5.29	5.30	5.32	5.33
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.82	136.57	141.06	144.07	146.34	148.06	149.39	150.39
T2kAFM	a	5.09	5.17	5.19	5.23	5.26	5.28	5.30	5.31
	b	5.10	5.15	5.20	5.25	5.27	5.31	5.31	5.32
	c	5.08	5.07	5.25	5.27	5.29	5.29	5.33	5.33
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.84	137.32	141.68	144.66	146.80	148.48	149.76	150.78
T3kAFM	a	5.09	5.16	5.21	5.25	5.27	5.29	5.31	5.32
	b	5.09	5.16	5.21	5.25	5.27	5.29	5.31	5.32
	c	5.09	5.16	5.21	5.25	5.27	5.29	5.31	5.32
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.51	137.27	141.67	144.58	146.78	148.28	149.69	150.64

Table S6. Cell lengths and cell angles, calculated when considering van der Waal's dispersion corrections (D3) of Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV) for cubic PuH_3. A, b and c in Å. Cell volume in $Å^3$. α, β, γ in degree.

Magnetic order	Parameter	PBEsol+*U*+SOC+D3 - *U* (eV)							
		0	**1**	**2**	**3**	**4**	**5**	**6**	**7**
FM <100>	a	4.93	5.06	5.10	5.16	5.21	5.24	5.28	5.30
	b	5.15	5.15	5.18	5.20	5.21	5.22	5.22	5.24
	c	5.15	5.15	5.18	5.20	5.21	5.22	5.22	5.24
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	130.59	134.13	137.03	139.51	141.4	142.95	144.22	145.17
FM <110>	a	5.06	5.10	5.14	5.16	5.19	5.21	5.23	5.25
	b	5.06	5.10	5.14	5.16	5.19	5.21	5.23	5.25
	c	5.11	5.16	5.20	5.24	5.25	5.27	5.26	5.27
	α	90.00	90.00	90.00	90.01	90.00	90.01	90.00	90.00
	β	90.00	90.00	90.00	89.99	90.00	89.99	90.00	90.00
	γ	89.48	90.03	89.71	89.83	89.89	89.92	89.95	89.96
	Lattice Volume	130.79	134.44	137.63	139.82	141.62	143.08	144.22	145.06
FM <111>	a	5.08	5.12	5.16	5.19	5.21	5.23	5.24	5.25
	b	5.08	5.12	5.16	5.19	5.21	5.23	5.24	5.25
	c	5.08	5.12	5.16	5.18	5.21	5.23	5.24	5.25
	α	89.54	89.82	89.85	89.85	89.87	89.88	89.83	89.88
	β	89.54	89.82	89.85	89.85	89.87	89.88	89.83	89.88
	γ	89.54	89.82	89.85	89.85	89.87	89.88	89.83	89.88
	Lattice Volume	130.69	134.37	137.36	139.78	141.73	143.21	144.07	144.96
L1kAFM	a	5.07	5.13	5.16	5.15	5.17	5.18	5.19	5.20
	b	5.07	5.13	5.16	5.15	5.17	5.18	5.19	5.20
	c	5.10	5.12	5.16	5.27	5.30	5.34	5.36	5.38
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.23	134.57	137.69	139.99	141.87	143.42	144.59	145.51
L2kAFM	a	5.07	5.12	5.17	5.21	5.24	5.24	5.26	5.27
	b	5.07	5.12	5.17	5.21	5.24	5.24	5.26	5.27
	c	5.10	5.16	5.17	5.18	5.18	5.24	5.23	5.24
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.2	135.19	138.18	140.44	142.24	143.92	144.98	145.63
L3kAFM	a	5.08	5.14	5.17	5.20	5.22	5.24	5.25	5.26
	b	5.08	5.14	5.17	5.20	5.22	5.24	5.25	5.26
	c	5.08	5.14	5.17	5.20	5.22	5.24	5.25	5.26
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	130.98	135.60	138.33	140.53	142.34	143.82	144.98	145.83
T1kAFM	a	4.99	5.08	5.10	5.12	5.09	4.96	4.97	4.99
	b	4.89	5.03	5.10	5.15	5.13	5.01	5.01	5.02
	c	5.34	5.26	5.29	5.32	5.45	5.81	5.84	5.84
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	130.16	134.42	137.56	140.13	142.17	144.18	145.39	146.26
T2kAFM	a	4.99	5.17	5.22	5.21	5.23	5.23	5.23	5.22
	b	4.89	5.07	5.09	5.15	5.18	5.20	5.20	5.22
	c	5.34	5.15	5.19	5.23	5.26	5.28	5.32	5.36
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	130.17	135.11	137.92	140.34	142.19	143.68	144.83	145.72
T3kAFM	a	5.08	5.13	5.17	5.20	5.22	5.24	5.25	5.26
	b	5.08	5.13	5.17	5.20	5.22	5.24	5.25	5.26
	c	5.08	5.13	5.17	5.20	5.22	5.24	5.25	5.26
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Lattice Volume	131.29	135.14	138.01	140.37	142.19	143.64	144.75	145.63

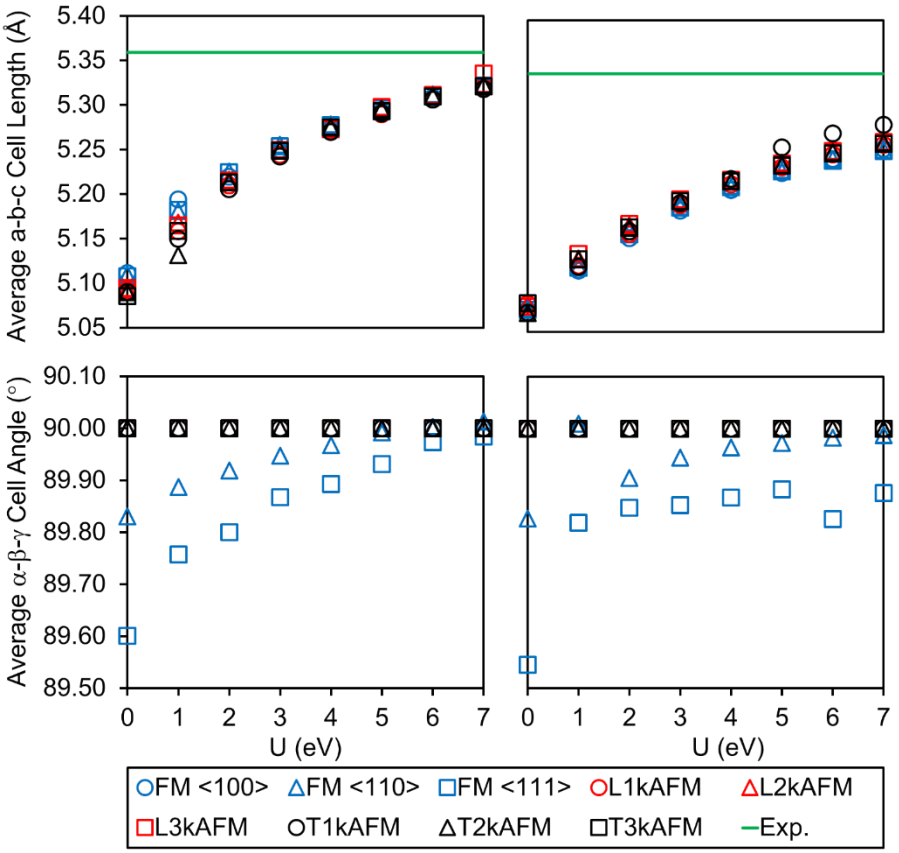

Figure S1. Average cell lengths a-b-c (Å) for cubic (a) PuH_2 and (b) PuH_3 and average cell angles α-β-γ (degree) for (c) PuH_2 and (d) PuH_3 calculated the inclusion of van der Waals correction Grimme *et al.* [1], *i.e.* PBEsol+*U*+SOC+D3 (*U* = 0 - 7 eV). Circles, triangles and squares symbols represent the <100>, <110> and <111> alignments respectively for FM orders in blue, longitudinal AFM in red, and transverse AFM in black. Experimental cell lengths represented in green for PuH_2 (Mulford et al. [2]) and PuH_3 (Muromura et al. [3]).

Section S3: Magnetic, structural and electronic properties of cubic PuH_2 and PuH_3

Table S7. Magnetic wavevectors in PuH_2 and PuH_3 with ferromagnetic <100>, <110> and <111>, longitudinal antiferromagnetic 1k, 2k and 3k and transverse antiferromagnetic 1k, 2k and 3k orders after optimisation using HSE06sol+SOC.

PuH_2			
Ferromagnetic			
Pu ionic position	**FM <100>**	**FM <110>**	**FM <111>**
(0, 0, 0)	(0, 0, 4.7)	(0, 3.3, 3.3)	(2.7, 2.7, 2.7)
(0.5, 0.5, 0)	(0, 0, 4.7)	(0, 3.3, 3.3)	(2.7, 2.8, 2.7)
(0.5, 0, 0.5)	(0, 0, 4.7)	(0, 3.3, 3.3)	(2.8, 2.7, 2.7)
(0, 0.5, 0.5)	(0, 0, 4.7)	(0, 3.3, 3.3)	(2.7, 2.7, 2.7)
Longitudinal antiferromagnetic			
Pu ionic position	**L1kAFM**	**L2kAFM**	**L3kAFM**
(0, 0, 0)	(0, 0, 4.6)	(0, 3.3, 3.3)	(2.7, 2.7, 2.7)
(0.5, 0.5, 0)	(0, 0, 4.6)	(0, -3.2, 3.3)	(-2.7, -2.7, 2.7)
(0.5, 0, 0.5)	(0, 0, -4.6)	(0, 3.2, -3.3)	(-2.7, 2.7, -2.7)
(0, 0.5, 0.5)	(0, 0, -4.6)	(0, -3.3, -3.3)	(2.7, -2.7, 2.7)
Transverse antiferromagnetic			
Pu ionic position	**T1kAFM**	**T2kAFM**	**T3kAFM**
(0, 0, 0)	(0, 0, 4.6)	(0, 3.1, 3.4)	(2.7, 2.7, 2.7)
(0.5, 0.5, 0)	(0, 0, -4.6)	(0, -3.1, -3.4)	(2.7, -2.7, -2.7)
(0.5, 0, 0.5)	(0, 0, 4.6)	(0, -3.1, 3.4)	(-2.7, -2.7, 2.7)
(0, 0.5, 0.5)	(0, 0, -4.6)	(0, 3.1, -3.4)	(-2.7, 2.7, -2.7)
PuH_3			
Ferromagnetic			
Pu ionic position	**FM <100>**	**FM <110>**	**FM <111>**
(0, 0, 0)	(0, 0, 4.6)	(0, 3.2, 3.2)	(2.7, 2.7, 2.7)
(0.5, 0.5, 0)	(0, 0, 4.6)	(0, 3.3, 3.2)	(2.7, 2.7, 2.6)
(0.5, 0, 0.5)	(0, 0, 4.6)	(0, 3.3, 3.3)	(2.7, 2.6, 2.7)
(0, 0.5, 0.5)	(0, 0, 4.6)	(0, 3.2, 3.2)	(2.6, 2.7, 2.7)
Longitudinal antiferromagnetic			
Pu ionic position	**L1kAFM**	**L2kAFM**	**L3kAFM**
(0, 0, 0)	(0, 0, 4.5)	(0, 3.2, 3.2)	(2.6, 2.6, 2.6)
(0.5, 0.5, 0)	(0, 0, 4.5)	(0, -3.2, 3.2)	(-2.6, -2.6, 2.6)
(0.5, 0, 0.5)	(0, 0, -4.5)	(0, 3.2, -3.2)	(-2.6, 2.6, -2.6)
(0, 0.5, 0.5)	(0, 0, -4.5)	(0, -3.2, -3.2)	(2.6, -2.6, 2.6)
Transverse antiferromagnetic			
Pu ionic position	**T1kAFM**	**T2kAFM**	**T3kAFM**
(0, 0, 0)	(0, 0, 4.6)	(0, 3.2, 3.2)	(2.6, 2.6, 2.6)
(0.5, 0.5, 0)	(0, 0, -4.6)	(0, -3.2, -3.2)	(2.6, -2.6, -2.6)
(0.5, 0, 0.5)	(0, 0, 4.6)	(0, -3.2, 3.2)	(-2.6, -2.6, 2.6)
(0, 0.5, 0.5)	(0, 0, -4.6)	(0, 3.2, -3.2)	(-2.6, 2.6, -2.6)

Table S8. Cell lengths and cell angles for cubic PuH_2 using PBEsol+*U*+SOC (*U* = 0 - 7 eV) and HSE06sol+SOC methods. a, b and c in Å. Cell volume in $Å^3$. α, β, γ in degree.

Magnetic order	Parameter	PBEsol+*U*+SOC - *U* (eV								HSE06sol +SOC
		0	1	2	3	4	5	6	7	
FM <100>	a	5.20	5.28	5.29	5.32	5.34	5.36	5.37	5.39	5.29
	b	5.20	5.28	5.29	5.32	5.34	5.36	5.37	5.39	5.29
	c	5.20	5.21	5.29	5.32	5.34	5.35	5.36	5.36	5.33
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	140.46	144.89	148.06	150.32	152.20	153.66	154.80	155.79	149.36
FM <110>	a	5.19	5.26	5.30	5.33	5.35	5.36	5.37	5.38	5.33
	b	5.19	5.26	5.30	5.33	5.35	5.36	5.37	5.38	5.33
	c	5.21	5.27	5.28	5.31	5.34	5.36	5.37	5.38	5.28
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	89.99	90.00
	β	90.00	90.01	90.00	90.00	90.00	90.00	90.00	90.01	90.00
	γ	89.72	89.82	89.82	89.89	89.92	89.97	89.98	90.01	89.85
	Cell Volume	140.26	145.79	148.55	150.76	152.58	153.98	155.15	156.03	149.80
FM <111>	a	5.19	5.26	5.29	5.32	5.34	5.36	5.37	5.38	5.31
	b	5.19	5.26	5.29	5.32	5.34	5.36	5.37	5.38	5.31
	c	5.19	5.26	5.29	5.32	5.34	5.36	5.37	5.38	5.31
	α	89.77	89.85	89.84	89.87	89.91	89.94	89.97	89.98	89.89
	β	89.77	89.85	89.84	89.87	89.91	89.94	89.97	89.98	89.89
	γ	89.77	89.85	89.84	89.87	89.91	89.94	89.97	89.98	89.89
	Cell Volume	140.15	145.73	148.42	150.65	152.45	153.93	155.03	155.93	149.66
L1kAFM	a	5.15	5.20	5.24	5.28	5.31	5.34	5.35	5.37	5.22
	b	5.15	5.20	5.24	5.28	5.31	5.34	5.35	5.37	5.22
	c	5.23	5.33	5.35	5.37	5.38	5.39	5.40	5.40	5.43
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.80	143.87	147.24	149.80	151.78	153.39	154.72	155.58	147.77
L2kAFM	a	5.19	5.26	5.30	5.33	5.35	5.37	5.38	5.39	5.29
	b	5.19	5.26	5.30	5.33	5.35	5.37	5.38	5.39	5.29
	c	5.16	5.22	5.26	5.29	5.32	5.34	5.36	5.37	5.31
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.86	144.39	147.79	150.32	152.30	153.84	155.15	156.02	148.55
L3kAFM	a	5.18	5.24	5.29	5.32	5.34	5.36	5.37	5.38	5.29
	b	5.18	5.24	5.29	5.32	5.34	5.36	5.37	5.38	5.29
	c	5.18	5.24	5.29	5.32	5.34	5.36	5.37	5.38	5.29
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.84	144.05	147.82	150.25	152.36	153.89	155.17	156.01	148.44
T1kAFM	a	5.20	5.23	5.27	5.30	5.33	5.35	5.37	5.38	5.26
	b	5.21	5.25	5.27	5.30	5.33	5.34	5.36	5.37	5.25
	c	5.11	5.21	5.29	5.33	5.35	5.37	5.38	5.39	5.34
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.50	143.14	147.03	149.72	151.80	153.42	154.72	155.70	147.54
T2kAFM	a	5.17	5.20	5.26	5.30	5.33	5.35	5.36	5.37	5.31
	b	5.17	5.23	5.28	5.31	5.34	5.35	5.37	5.38	5.35
	c	5.18	5.28	5.32	5.34	5.36	5.37	5.39	5.40	5.23
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.47	143.77	147.56	150.27	152.28	153.88	155.11	156.01	148.35
T3kAFM	a	5.17	5.24	5.28	5.32	5.34	5.36	5.37	5.38	5.29
	b	5.17	5.24	5.28	5.32	5.34	5.36	5.37	5.38	5.29
	c	5.17	5.24	5.28	5.32	5.34	5.36	5.37	5.38	5.29
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	138.31	143.83	147.56	150.33	152.19	153.69	154.98	155.84	148.28

Table S9. Cell lengths and cell angles for PuH_3 using PBEsol+*U*+SOC (*U* = 0 - 7 eV) and HSE06sol+SOC methods. Cell volume in $Å^3$. α, β, γ in degree.

Magnetic order	Parameter	PBEsol+*U*+SOC - *U* (eV)								HSE06sol +SOC
		0	1	2	3	4	5	6	7	
FM <100>	a	5.21	5.20	5.23	5.25	5.26	5.27	5.27	5.28	5.24
	b	5.21	5.20	5.23	5.25	5.26	5.27	5.27	5.28	5.24
	c	5.02	5.16	5.20	5.24	5.29	5.33	5.37	5.39	5.20
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	136.07	139.58	142.29	144.53	146.41	147.92	149.15	150.07	143.12
FM <110>	a	5.13	5.17	5.21	5.23	5.26	5.28	5.29	5.30	5.19
	b	5.13	5.17	5.21	5.23	5.26	5.28	5.29	5.30	5.19
	c	5.18	5.23	5.27	5.30	5.30	5.31	5.32	5.33	5.33
	α	90.00	89.99	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.01	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	89.61	90.03	89.78	89.87	89.92	89.96	89.94	89.96	89.80
	Cell Volume	136.23	139.83	142.83	144.90	146.65	148.07	149.14	149.96	144.20
FM <111>	a	5.14	5.19	5.22	5.25	5.27	5.29	5.30	5.31	5.24
	b	5.14	5.19	5.22	5.25	5.27	5.29	5.30	5.31	5.24
	c	5.14	5.19	5.22	5.25	5.27	5.29	5.30	5.31	5.24
	α	89.69	89.84	89.84	89.85	89.87	89.89	89.79	89.85	89.76
	β	89.69	89.84	89.84	89.85	89.87	89.89	89.79	89.85	89.76
	γ	89.69	89.84	89.84	89.85	89.87	89.89	89.79	89.85	89.76
	Cell Volume	136.14	139.73	142.51	144.88	146.68	148.17	149.14	149.85	143.46
L1kAFM	a	5.15	5.19	5.22	5.21	5.23	5.24	5.24	5.25	5.17
	b	5.15	5.19	5.22	5.21	5.23	5.24	5.24	5.25	5.17
	c	5.15	5.19	5.25	5.35	5.37	5.41	5.24	5.45	5.38
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	136.65	139.86	142.87	145.08	146.82	148.30	149.47	150.39	143.63
L2kAFM	a	5.14	5.19	5.24	5.28	5.30	5.31	5.33	5.34	5.28
	b	5.14	5.19	5.24	5.28	5.30	5.31	5.33	5.34	5.28
	c	5.19	5.22	5.23	5.22	5.24	5.28	5.28	5.28	5.18
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	136.76	140.58	143.39	145.45	147.26	148.85	149.85	150.51	144.32
L3kAFM	a	5.15	5.20	5.24	5.26	5.28	5.30	5.31	5.32	5.24
	b	5.15	5.20	5.24	5.26	5.28	5.30	5.31	5.32	5.24
	c	5.15	5.20	5.24	5.26	5.28	5.30	5.31	5.32	5.24
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	136.80	140.88	143.56	145.76	147.25	148.53	149.81	150.70	144.20
T1kAFM	a	5.09	5.16	5.15	5.16	5.14	5.13	5.13	5.24	4.97
	b	4.99	5.12	5.18	5.21	5.20	5.18	5.19	5.31	5.02
	c	5.35	5.30	5.35	5.40	5.50	5.60	5.63	5.40	5.77
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	135.96	139.92	142.81	145.23	147.15	148.69	149.81	150.34	144.01
T2kAFM	a	5.09	5.23	5.26	5.26	5.28	5.28	5.28	5.29	5.24
	b	5.00	5.22	5.26	5.30	5.31	5.25	5.27	5.27	5.21
	c	5.34	5.15	5.17	5.21	5.24	5.36	5.39	5.40	5.28
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume	135.99	140.52	143.17	145.39	147.17	148.60	149.75	150.53	144.09
T3kAFM	a	5.15	5.20	5.23	5.26	5.28	5.30	5.31	5.32	5.24
	b	5.15	5.20	5.23	5.26	5.28	5.30	5.31	5.32	5.24
	c	5.15	5.20	5.23	5.26	5.28	5.30	5.31	5.32	5.24
	α	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	β	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	γ	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00	90.00
	Cell Volume ($Å^3$)	136.95	140.55	143.28	145.34	147.18	148.61	149.63	150.54	144.11

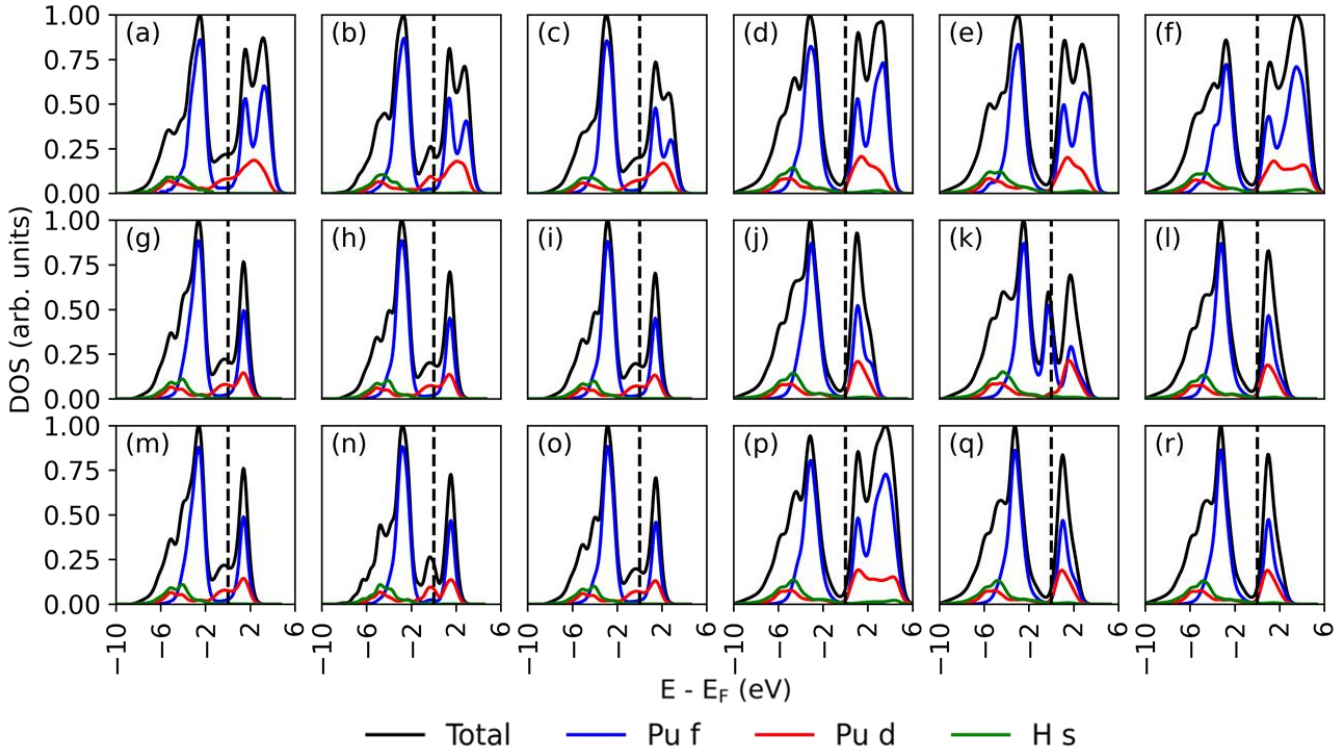

Figure S2. eDOS plots calculated using PBEsol+*U*+SOC (*U* = 4 eV) for cubic PuH_2/PuH_3 with: FM (a/d) <100>, (b/e) <110> and (c/f) <111>; longitudinal AFM (g/j) 1k, (h/k) 2k, and (i/l) 3k; and transverse AFM (m/p) 1k, (n/q) 2k, and (o/r) 3k. The Fermi level is at 0 eV denoted by a vertical dashed line.

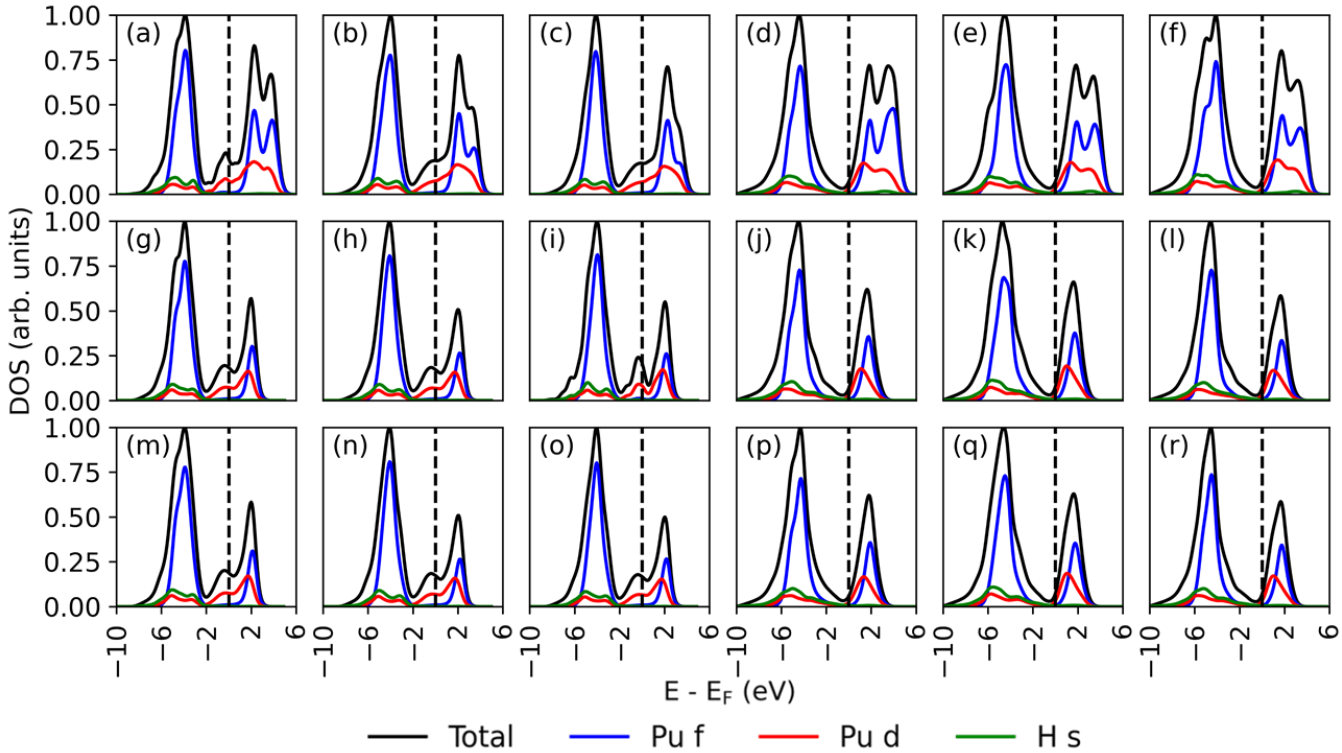

Figure S3. eDOS plots calculated using PBEsol+*U*+SOC (*U* = 6 eV) for cubic PuH_2/PuH_3 with: FM (a/d) <100>, (b/e) <110> and (c/f) <111>; longitudinal AFM (g/j) 1k, (h/k) 2k, and (i/l) 3k; and transverse AFM (m/p) 1k, (n/q) 2k, and (o/r) 3k. The Fermi level is at 0 eV denoted by a vertical dashed line.

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