Supporting Information

for

Theoretical Study on the Structural and Thermodynamic Properties of U-He compounds under High Pressure

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Table S1. Crystal structure information of *Fmmm*-U_4He and $P\bar{1}$-U_6He at 150 GPa.

Phase	Space group	Lattice parameters	Atom	Fractional coordinates			Wyckoff Position
				x	y	z	
		a=10.320 Å	U1	0.664	0.500	-0.165	16n
U_4He	*Fmmm*	b=3.434 Å	U5	0.341	0.500	-0.500	8g
		c=13.017 Å	U7	0.500	0.500	-0.346	8i
		α=β=γ=90°	He1	0.936	0.500	-0.500	8g
		a=2.746 Å					
		b=4.481 Å	U1	0.533	0.676	0.716	2i
U_6He	$P\bar{1}$	c=7.023 Å	U2	0.801	0.853	0.353	2i
		α=100.0°	U4	0.819	0.241	0.964	2i
		β=92.3°	He1	0.000	0.500	0.500	1g
		γ=90.7°					

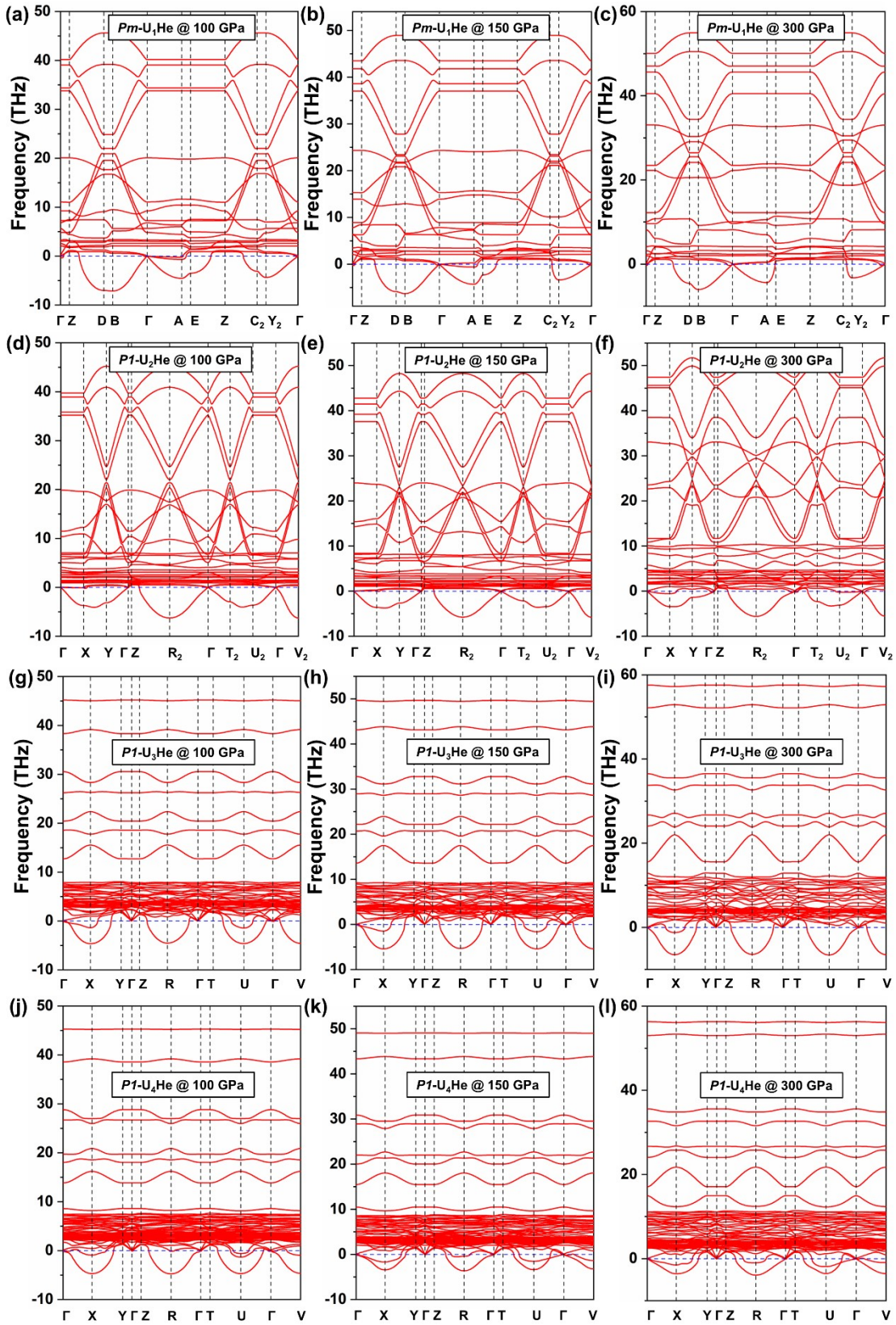

Fig. S1. The phonon dispersion spectra at 100, 150, and 300 GPa of (a)-(c) *Pm*-UHe, (d)-(f) *P1*-U_2He, (g)-(i) *P1*-U_3He and (j)-(l) *P1*-U_4He, respectively.

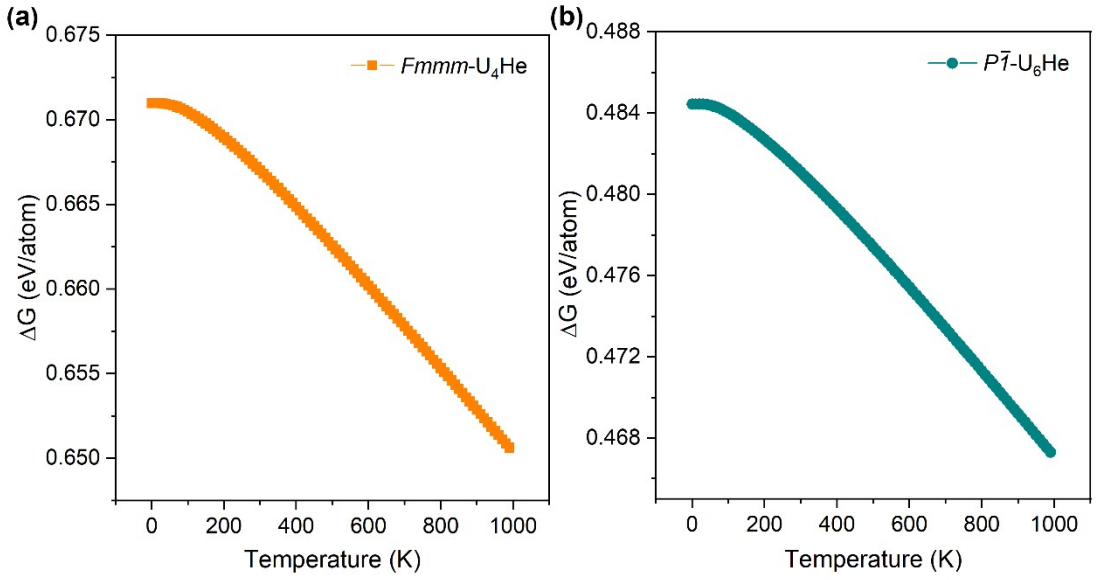

Fig. S2. Gibbs free energy of (a) *Fmmm*-U_4He and (b) $P\bar{1}$-U_6He within quasi-harmonic approximation (QHA) at 150 GPa.

Under the quasi-harmonic approximation (QHA), [1] the Gibbs free energy of *Fmmm*-U_4He can be expressed as:

$$\Delta G = \frac{G(U_nHe) - nG(U) - G(He)}{n+1} \tag{1}$$

where *G(U)* and *G (He)* represented the Gibbs free energy of the most stable structure of element uranium (α-U) and helium (*hcp*-He), respectively.

Table S2. Bader charge (in e) of *Fmmm*-U_4He and $P\bar{1}$-U_6He at 150 GPa.

Fmmm-U_4He					
	U1	U2	U3	U4	U5
Charge	+0.023	+0.023	+0.029	+0.029	-0.253
	U6	U7	U8	He1	He2
Charge	-0.253	+0.083	+0.083	+0.118	+0.118
$P\bar{1}$-U_6He					
	U1	U2	U3	U4	U5
Charge	-0.127	+0.062	+0.062	-0.008	-0.008
	U6	He1			
Charge	-0.127	+0.147			

Table S3. The calculated independent elastic constants C_{ij}, bulk modulus B, shear modulus G, Young's modulus E (in GPa), Poisson's ratio v, and longitudinal v_l, transverse v_t, average sound velocities v_m (in m/s), Debye temperature θ_D (in K), and B/G of α-U under 0 and 100 GPa, as well as experimental data and other theoretical calculations.

Pressure (GPa)	C_{11}	C_{22}	C_{33}	C_{44}	C_{55}	C_{66}	C_{12}	C_{13}	C_{23}
0 (Expt.[2])	215	199	267	124	73	74	46	22	108
0 (Calc.[3])	296	216	367	153	129	99	60	29	141
0 (Calc.[4])	295	215	347	143	130	102	68	25	149
0 (Calc.[5])	287	220	352	151	117	101	66	28	152
0	300.0	226.2	361.5	155.0	126.2	99.2	60.1	27.3	138.9
100 (Calc.[5])	1165	770	1020	355	293	310	224	167	453
100	1153.1	758.5	1108.7	367.7	312.3	309.4	209.9	200.5	428.8
Pressure (GPa)	B	G	E	v	v_l	v_t	v_m	θ_D	B/G
0 (Expt. [2])	115	87		0.20				251	1.322
0 (Calc.[3])	149	108	261	0.21				287	1.380
0 (Calc.[4])	147	108	261	0.204				284	1.357
0 (Calc.[5])	148	107	259	0.207	3846	2338	2583	283	1.383
0	146.6	114.1	271.8	0.191	3893.1	2406.0	2653.6	290.8	1.285
100 (Calc.[5])	513	320	795	0.242	5895	3439	3815	464	1.603
100	515.6	333.2	822.4	0.234	5959.5	3511.3	3897.1	473.7	1.547

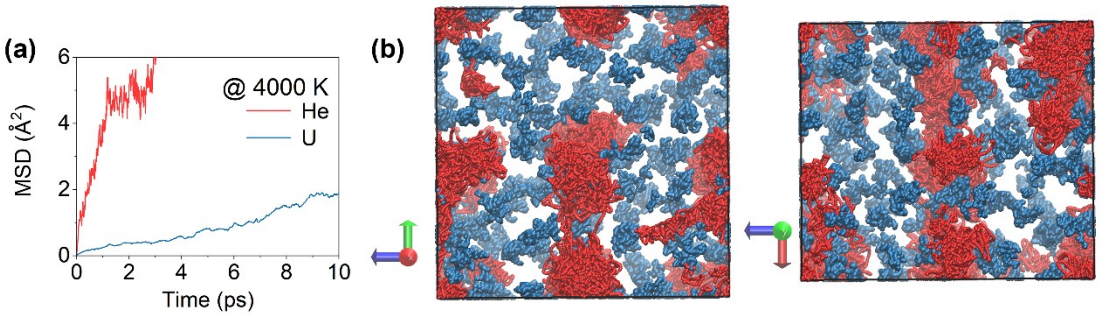

Fig. S3. (a) The calculated mean squared displacement (MSD) and (b) the trajectories of U (red) and He (blue) atoms in *Fmmm*-U_4He at 4000 K.

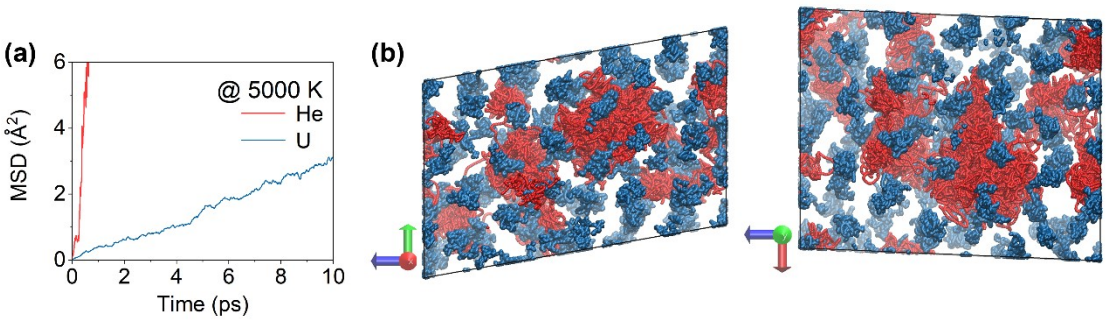

Fig. S4. (a) The calculated mean squared displacement (MSD) and (b) the trajectories of U (red) and He (blue) atoms in $P\bar{1}$-U_6He at 5000 K.

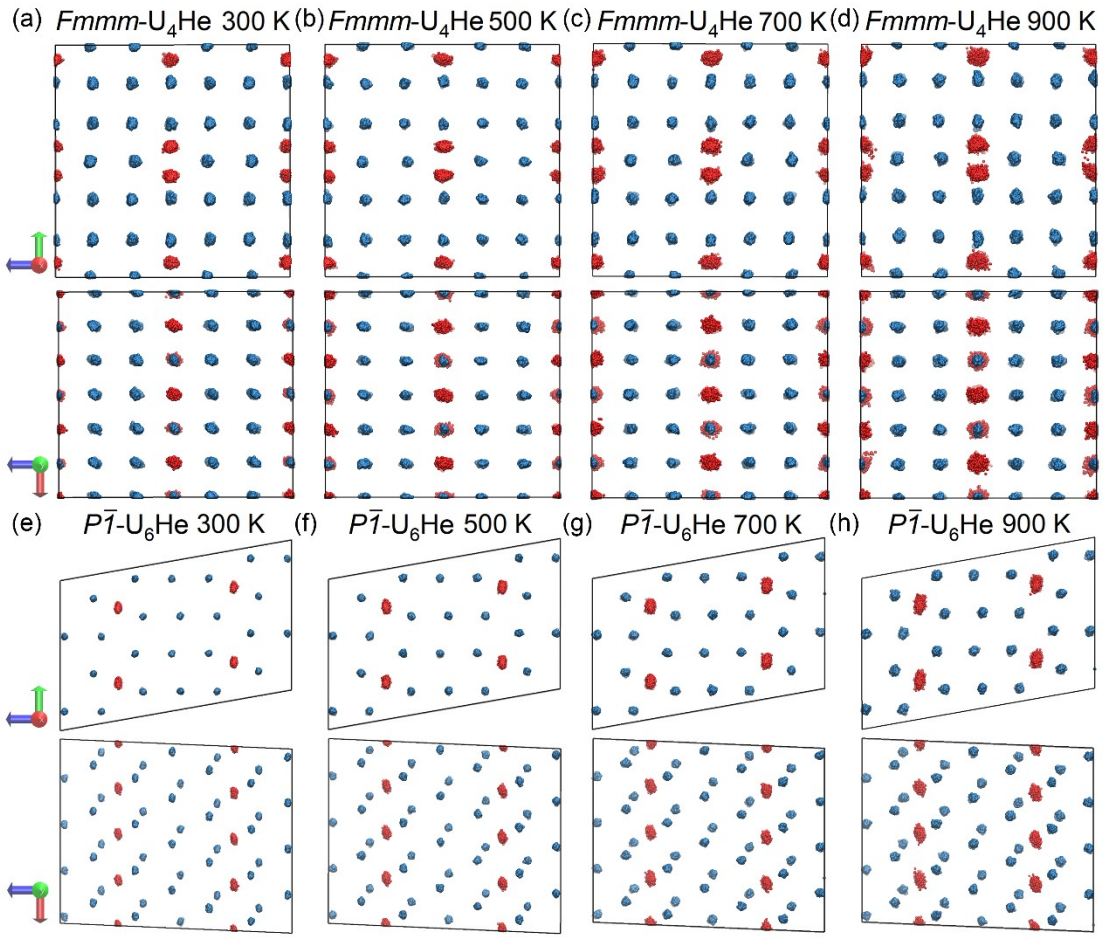

Fig. S5. The trajectories of He (red) and U (blue) atoms of (a)-(d) *Fmmm*-U_4He and (e)-(h) $P\bar{1}$-U_6He at 300, 500, 700 and 900 K.

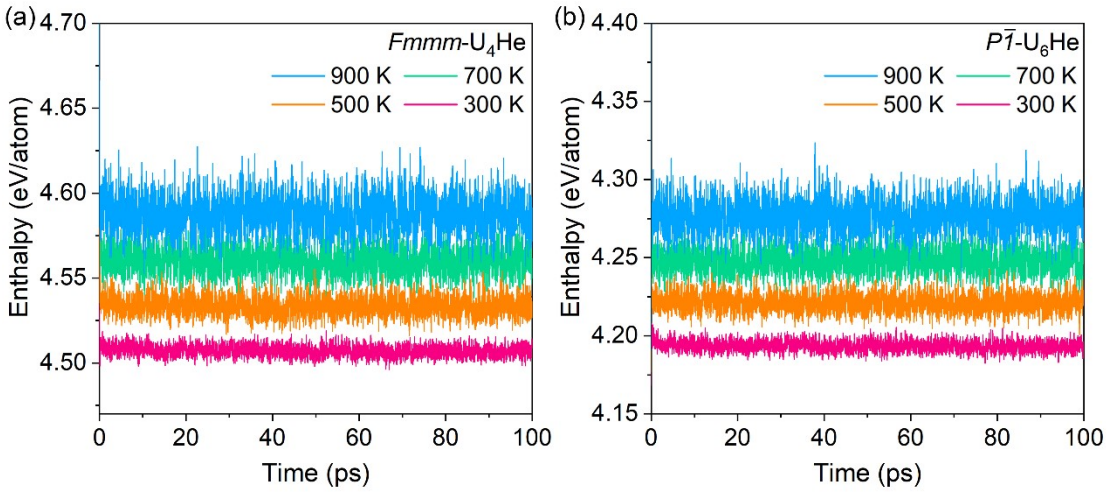

Fig. S6. The evolution of enthalpy with time of (a) *Fmmm*-U_4He and (b) $P\bar{1}$-U_6He at 300, 500, 700 and 900 K.

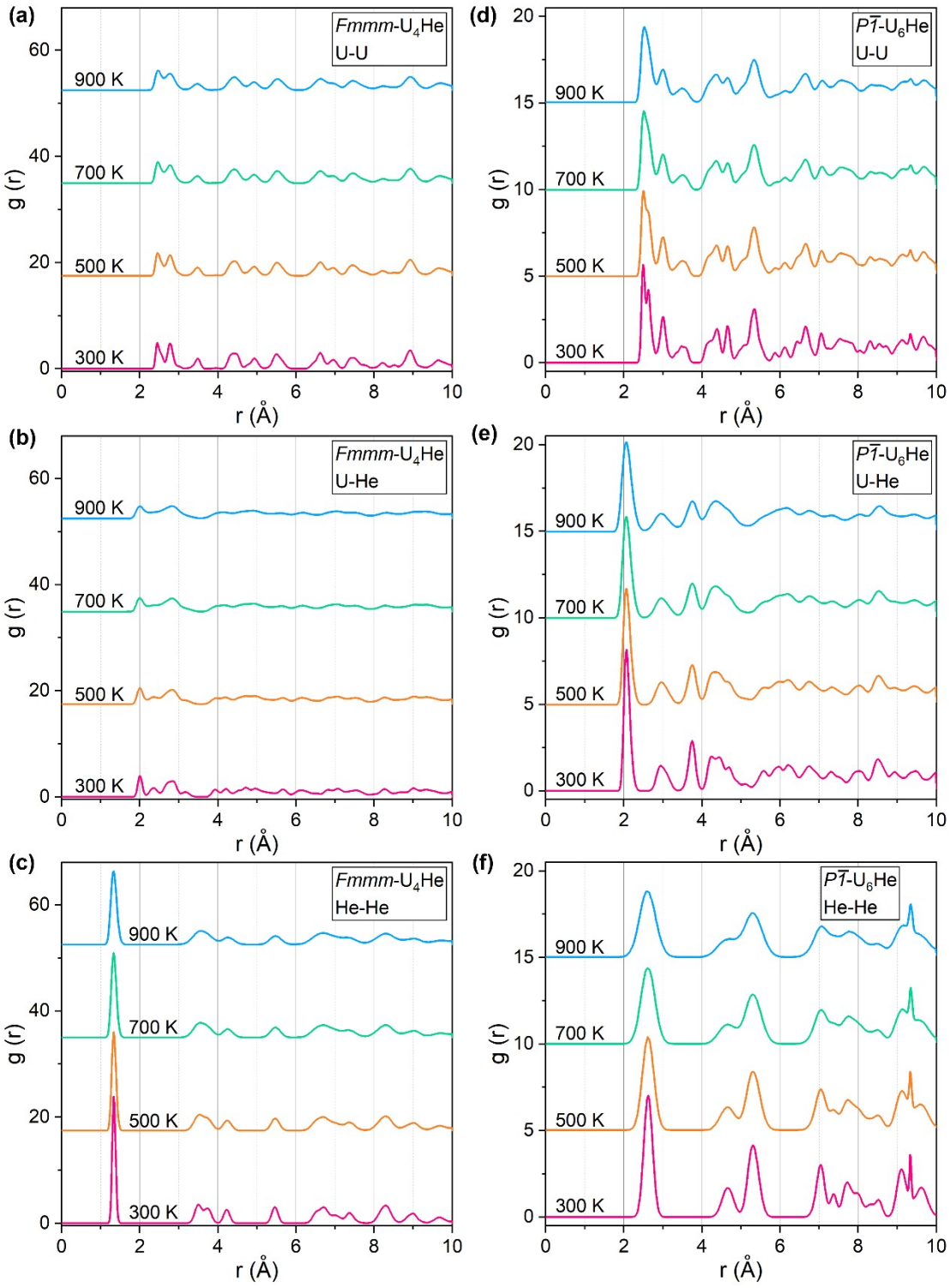

Fig. S7. The radial distribution function of (a)-(c) *Fmmm*-U_4He and (d)-(f) $P\bar{1}$-U_6He at select temperature.

References

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