Supporting Information

High-temperature Superconductivities and Crucial Factors Influencing Stability of $LaThH_{12}$ under Moderate Pressures

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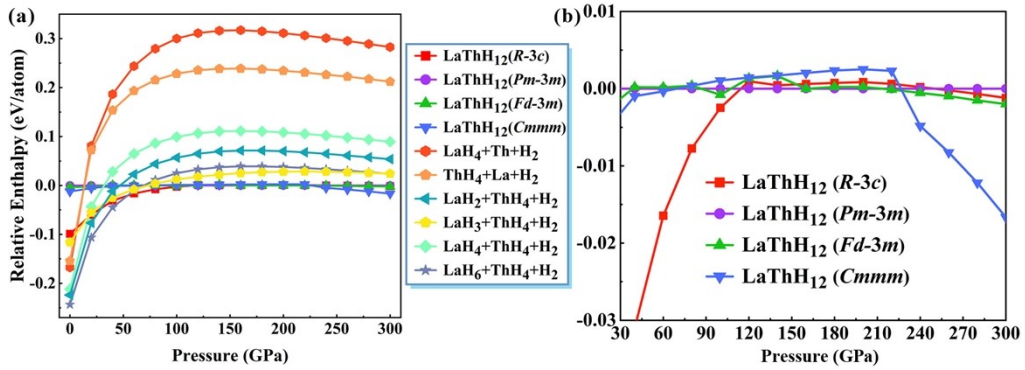

Fig. S1. (a) The calculated enthalpies as a function of pressure for predicted $LaThH_{12}$ relative to the *Pm*-3*m* phase. (b) Enthalpies of *R*-3*c*, *Fd*-3*m* and *Cmmm* structures relative to the *Pm*-3*m* structure.

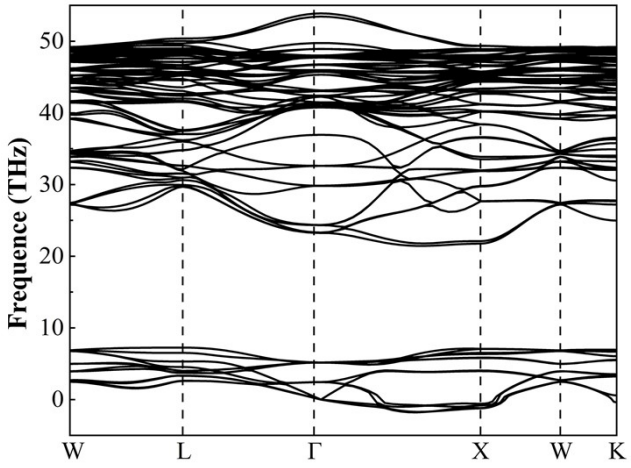

Fig. S2. The phonon dispersion curve for *Fd*-3*m*-$LaThH_{12}$ at 225GPa.

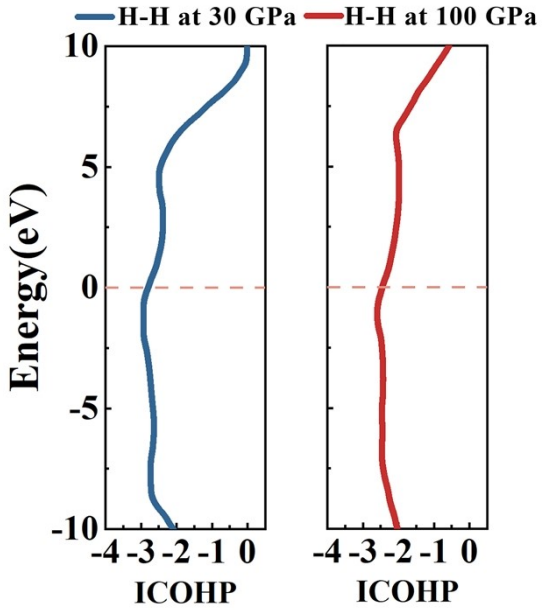

Fig. S3. The calculated iCOHP of H–H bonds of *R*-3*c*-$LaThH_{12}$ at 30 and 100 GPa.

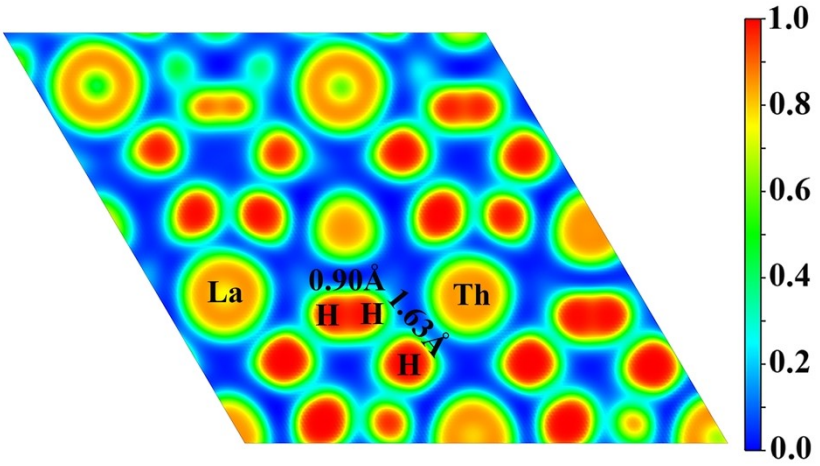

Fig. S4. Calculated the 2D ELF of 1D profile locating at the H-H distance of 0.9 Å and 1.63 Å in the *R*-3*c*-$LaThH_{12}$ structure at 30 GPa.

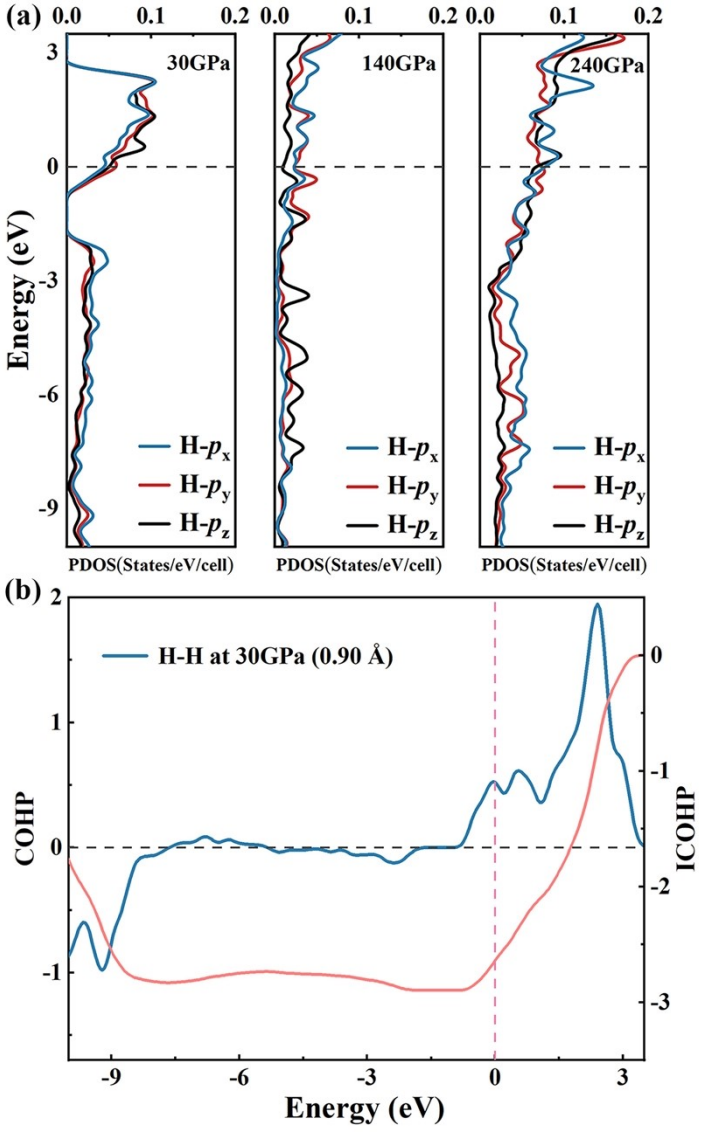

Fig. S5. (a) Partial density of states (DOS) projected onto hydrogen atoms for *R*-3*c*-

$LaThH_{12}$ at 30 GPa, *Pm*-3*m*-$LaThH_{12}$ at 140 GPa and *Cmmm*-$LaThH_{12}$ at 240 GPa, where all the electron states originated from H p_x, p_y, and p_z orbitals near the Fermi level contribute to the DOS. (b) COHP (blue line) and ICOHP (red line) of the pair of H-H bond in *R*-3*c*-$LaThH_{12}$ at 30 GPa.

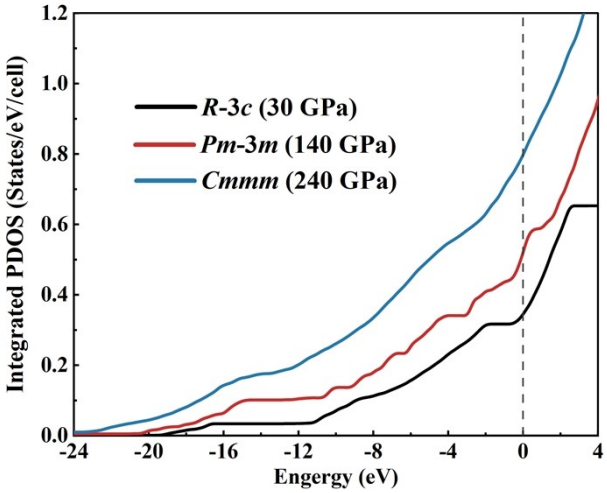

Fig. S6. Integrated PDOS of H *p* orbitals of $LaThH_{12}$ at pressures up to 240 GPa. The calculated integrated projected DOS clearly shows a gradual increase and widening of the H *p*-valence band of $LaThH_{12}$ with increasing pressure. The number of *p* electrons has more than doubled (from 0.34*e*/state to 0.8*e*/state) as the pressure is increased from 30 to 240 GPa.

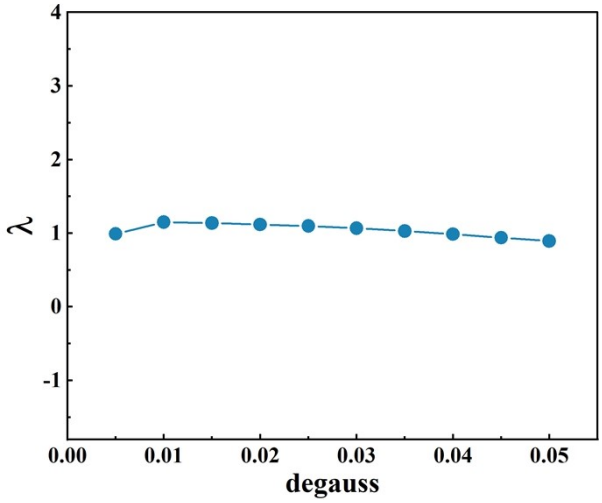

Fig. S7. The calculated electron-phonon coupling λ with different double-delta smearing values for *R*-3*c*-$LaThH_{12}$ at 30 GPa, the value of λ becomes convergent more perfectly at the smearing width value of 0.03.

Supplementary structural information of our predicted other hydrides in La-Th-H system:

In order to facilitate a more detailed understanding of the La-Th-H system, we have provided a supplementary description of the predicted new structures of the system. As shown in Fig. S8, we successfully reproduced the previously predicted structures such as *Pm*3-$LaThH_6$, *P*-1-La_3ThH_{16}, *P*2/*m*-$LaTh_3H_{16}$, *Cmmm*-$LaThH_8$, *C*2/*m*-La_2ThH_{12}, *C*2/*m*-$LaTh_2H_{12}$, *I*4/*mmm*-$LaTh_3H_{40}$ and *R*-3*m*-$LaThH_{20}$, confirming the reliability of our work, their corresponding T_c's and atomic coordinates are summarized in Table S1 and Table S4 [1]. Our predicted clathrate hydrides, such as *P*-3*m*1-$LaTh_2H_{18}$, *Fm*-3*m*-$LaTh_3H_{24}$, and *C*2/*m*-La_2ThH_{18}, were found to be dynamically stable within the range of 50 – 300 GPa, with formation energies slightly higher than the convex hull diagram by 20 – 50 meV/atom. $LaTh_2H_{18}$ and La_2ThH_{18} maintain dynamic stability at 200 GPa, while $LaTh_3H_{24}$ can be stabilized at 150 GPa (see Fig. S9). Furthermore, the metastable hydrides $LaTh_2H_{18}$, La_2ThH_{18}, and $LaTh_3H_{24}$ can also be viewed as stacked configurations of La-H_{24} cages and Th-H_{24} cages, and their corresponding geometries have been listed in Fig. S10. The lattice parameters and superconductivities of corresponding structures of *P*-3*m*1-$LaTh_2H_{18}$, *Fm*-3*m*-$LaTh_3H_{24}$, and *C*2/*m*-La_2ThH_{18} are summarized in Table S2 and Table S5.

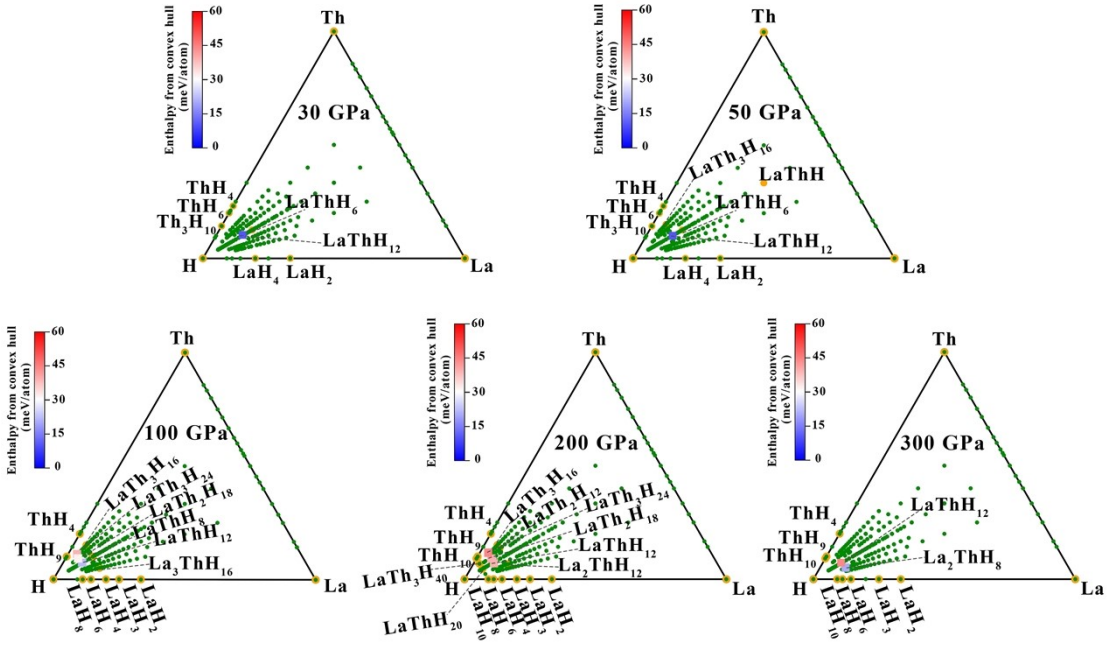

Fig. S8. Trigonal formation enthalpies convex hull of La-Th-H system at 30 GPa, 50 GPa, 100 GPa, 200 GPa, and 300 GPa, respectively. Orange circles denoted thermodynamically stable stoichiometries. Green circles denoted thermodynamically unstable stoichiometries. Differently colored squares denoted metastable stoichiometries. The range of distance from the convex hull was given in the legend.

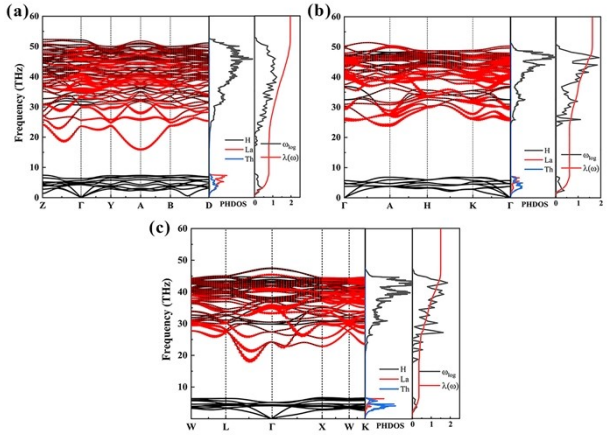

Fig. S9. The phonon dispersion curve, projected phonon density of states (PHDOS), Eliashberg spectral function $\alpha^2F(\omega)$, together with the electron-phonon parameter $\lambda(\omega)$ for (a) *C*2/*m*-La_2ThH_{18} at 200 GPa, (b) *P*-3*m*1-$LaTh_2H_{18}$ at 200 GPa, (c) *Fm*-3*m*-$LaTh_3H_{24}$ at 150 GPa.

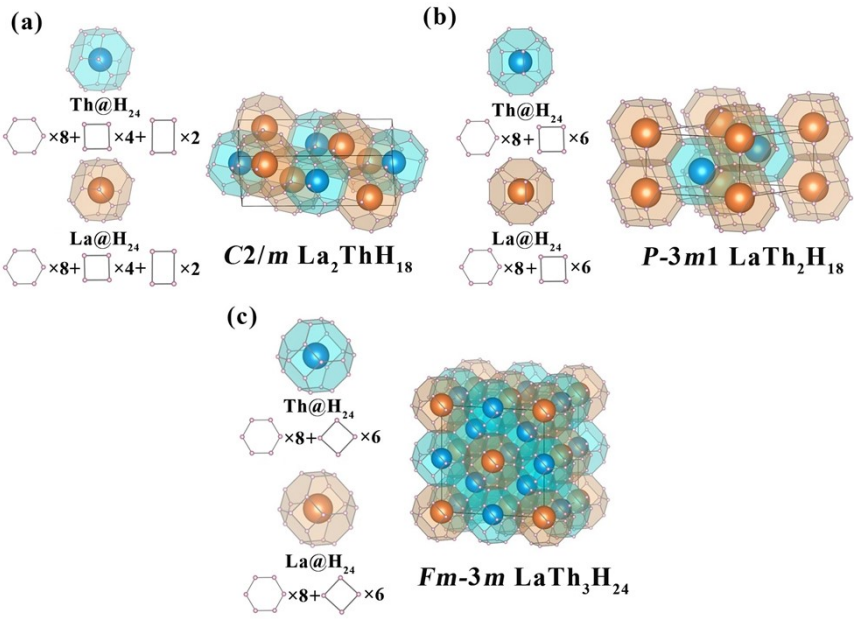

Fig. S10. The predicted crystal structure of (a) *C*2/*m*-La_2ThH_{18}, (b) *P*-3*m*1-$LaTh_2H_{18}$ and (c) *Fm*-3*m*-$LaTh_3H_{24}$, consisting of Th-centered H_{24} cages (light blue) and La-centered H_{24} cages (light orange).

Table S1. Superconducting transition temperatures of stable structures at different pressure.

Phase	Space group	P (GPa)	T_c (K)
$LaThH_6$	*Pm*3	50	0.009
$LaThH_8$ [1]	*Cmmm*	100	10.1
La_3ThH_{16} [1]	*P*-1	100	34.7
$LaTh_2H_{12}$ [1]	*C*2/*m*	200	1.0
La_2ThH_{12} [1]	*C*2/*m*	200	10.7
$LaTh_3H_{40}$ [1]	*I*4/*mmm*	200	163.8
$LaThH_{20}$ [1]	*R*-3*m*	200	179.2

Table S2. Structural information of predicted metastable hydrides in this work.

Space group	P (GPa)	Lattice parameters	Atomic coordinates (fractional)			
			Atoms	x	y	z
R-3*c*-$LaThH_{12}$	30	a = 5.95772 b = 5.95772 c = 15.2468 α = 90.0° β = 90.0° γ = 120.0°	H(36f) H(36f) La(6b) Th(6a)	0.58025 0.61571 0.33333 0.33333	0.67731 0.67729 0.66667 0.66667	0.79949 0.55951 0.16667 0.91667
Fm-3*m*-$LaTh_3H_{24}$	100	a = 7.3709 b = 7.3709 c = 7.3709 α = 90.0° β = 90.0° γ = 90.0°	H(96j) La(4a) Th(8c) Th(4b)	-0.00000 0.50000 0.75000 0.00000	0.87566 -0.00000 0.75000 0.00000	0.75054 0.50000 0.75000 0.50000
P-3*m*1-$LaTh_2H_{18}$	100	a = 5.1994 b = 5.1994 c = 3.1926 α = 90.0° β = 90.0° γ = 120.0°	H(12j) H(6h) La(1a) Th(2d)	0.66926 0.00000 0.00000 0.66667	0.58474 0.75043 0.00000 0.333333	0.83098 0.50000 0.00000 0.33012
Pm-3*m*-$LaThH_{12}$	200	a = 3.8020 b = 3.8020 c = 3.8020 α = 90.0° β = 90.0° γ = 90.0°	H(12h) La(1a) Th(1b)	0.50000 0.00000 0.50000	0.75113 0.00000 0.50000	0.00000 0.00000 0.50000
Cmmm-$LaThH_{12}$	300	a = 3.5888 b = 4.7125 c = 5.5053 α = 90.0° β = 90.0° γ = 90.0°	La(2c) Th(2a) H(8n) H(8n) H(8m)	0.5000 0.00000 0.5000 0.5000 0.2500	-0.0000 0.00000 0.38785 0.1139 -0.2500	-0.5000 0.00000 -0.36094 -0.14285 -0.25295

*C*2/*m*-La_2ThH_{18}	300	a = 9.1758	H(8j)	0.62067	0.11388	0.01893
		b = 4.8356	H(8j)	0.83244	0.25179	0.66413
		c = 3.3156	H(8j)	0.71297	0.11248	0.64787
		α = 90.0°	H(8j)	0.95409	0.11084	0.68297
		β = 93.5924°	H(4g)	0.00000	0.74751	0.00000
		γ = 90.0°	La(4i)	0.666667	0.50000	0.83642
			Th(2d)	0.00000	0.50000	0.50000

Table S3. The average charge value (e) and number of electrons transferred $\delta(e)$ for different atoms in *R*-3*c*-$LaThH_{12}$ and *R*-3*c*-$LaYH_{12}$ at 50 GPa.

Structure	Atom	Pressure	Charge Value	$\delta(e)$
R-3*c*-$LaThH_{12}$	La (2)	50GPa	9.54	1.46
	Th (2)		10.16	1.84
	H1 (12)		1.41	-0.41
	H2 (6)		1.13	-0.13
	H3 (6)		1.15	-0.15
R-3*c*-$LaYH_{12}$	La (2)	50GPa	9.26	1.74
	Y (2)		9.14	1.86
	H1 (12)		1.52	-0.52
	H2 (6)		1.09	-0.09
	H3 (6)		1.07	-0.07

Table S4. Structural information of our predicted another stable hydrides in this work.

Space group	P (GPa)	Lattice parameters	Atomic coordinates (fractional) Atoms x y z
*Pm*3-$LaThH_6$	30	a = 4.0752 b = 4.0752 c = 4.0752 α = 90.0° β = 90.0° γ = 90.0°	H(6g) 0.50000 0.00000 0.238510 La(1b) 0.50000 0.50000 0.50000 Th(1a) 0.00000 0.00000 0.00000
*P*4/*nmm*-LaThH	50	a = 3.22219 b = 3.22219 c = 8.86673 α = 90.0° β = 90.0° γ = 90.0°	H(8j) 0.50000 0.00000 0.62949 La(8j) 0.50000 1.00000 0.36625 Th(8j) 0.50000 1.00000 -0.11626
*Pm*3-$LaThH_6$	50	a = 3.9545 b = 3.9545 c = 3.9545 α = 90.0° β = 90.0° γ = 90.0°	H(6f) 1.23866 -1.00000 -0.50000 La(1b) 0.50000 -0.50000 -0.50000 Th(1a) -0.00000 -1.00000 -1.00000

Pm-3*n*- $LaTh_3H_{16}$	50	a = 6.3358 b = 6.3358 c = 6.3358 α = 90.0° β = 90.0° γ = 90.0°	H(24k) H(8e) La(2a) Th(6c)	1.00000 0.25000 0.00000 0.50000	0.15804 0.67838 0.25000 0.25000 0.00000 0.00000 0.75000 0.00000
Cmmm- $LaThH_8$	100	a = 4.2311 b = 5.9785 c = 4.2353 α = 90.0° β = 90.0° γ = 90.0°	H(8m) H(4j) H(4i) La(2c) Th(2a)	0.25000 -0.00000 -0.00000 0.00000 0.00000	0.25000 0.23415 0.13664 0.50000 0.36448 0.00000 0.50000 0.50000 0.00000 0.00000
P-1- La_3ThH_{16}	100	a = 3.6724 b = 5.6233 c = 5.6129 α = 73.2990° β = 77.4613° γ = 90.0846°	H(2i) H(2i) H(2i) H(2i) H(2i) H(2i) H(2i) H(2i) La(2i) La(1g) Th(1e)	0.04329 0.05814 0.18614 0.20547 0.29875 0.31066 0.43091 0.44564 0.25096 0.00000 0.50000	0.33003 0.91770 0.62062 0.82209 0.87292 0.44013 0.19378 0.32648 0.81025 0.15145 0.13446 0.05950 0.37764 0.69609 0.67577 0.60517 0.00194 0.74804 0.50000 0.50000 0.50000 0.00000
*C*2/*m*- La_2ThH_{12}	200	a = 9.7457 b = 3.9161 c = 3.4991 α = 90.0° β = 92.8528° γ = 90.0°	H(8j) H(4i) H(4i) H(4i) La(4i) H(4g) Th(2d)	0.66296 0.04695 0.61135 0.71022 0.16727 0.50000 0.50000	-0.24223 0.32912 -0.00000 0.30195 -0.00000 0.02770 -0.00000 0.64208 -0.00000 0.82951 -0.23804 0.00000 0.00000 0.50000
*C*2/*m*- $LaTh_2H_{12}$	200	a = 9.7356 b = 3.8833 c = 3.4958 α = 90.0° β = 92.6646° γ = 90.0°	H(8j) H(4i) H(4i) H(4i) Th(4i) H(4h) La(2a)	0.66973 0.54933 0.12239 0.21642 0.666667 0.50000 0.00000	-0.24672 0.83760 -0.00000 0.81184 -0.00000 0.52624 -0.00000 0.14056 -0.00000 0.33814 -0.24052 0.50000 0.00000 0.00000
R-3*m*- $LaThH_{20}$	200	a = 3.5289 b = 3.5289 c = 17.4089 α = 90.0° β = 90.0° γ = 120.0°	H(18h) H(18h) H(6c) H(6c) H(6c) H(6c) La(3b) Th(3a)	-0.32598 0.34490 -0.00000 -0.00000 -0.00000 -0.00000 -0.00000 0.00000	-0.16299 0.76785 0.17245 0.60553 0.00000 0.12130 0.00000 0.18958 0.00000 0.31076 0.00000 0.37382 -0.00000 0.50000 0.00000 0.00000
*I*4/*mmm*- $LaTh_3H_{40}$	200	a = 4.9929 b = 4.9929 c = 9.9937 α = 90.0° β = 90.0° γ = 90.0°	H(32o) H(16m) H(16m) H(16m) La(2b) Th(2a) Th(4d)	-0.38155 -0.12259 -0.12410 -0.24954 -0.00000 0.00000 -0.50000	0.12287 0.43785 0.12259 0.81194 0.12410 0.69165 0.24954 0.62456 -0.00000 0.50000 0.00000 0.00000 0.00000 0.25000
*P*2/*m*-		a = 3.4919 b = 3.8823 c = 6.5240	H(4o) H(2i) H(2m) H(2m) H(2n)	0.25269 0.00000 0.03611 0.46584 0.21681	0.25676 0.74414 0.25941 0.00000 0.00000 0.18471 0.00000 0.32696 0.50000 0.56942

$LaTh_3H_{16}$	200	α = 90.0°	H(2n)	0.28661	0.50000 0.92673
		β = 97.2805°	Th(2n)	0.24338	0.50000 0.24860
		γ = 90.0°	H(2l)	0.50000	0.25829 0.50000
			La(1d)	0.50000	0.00000 -0.00000
			Th(1c)	0.00000	0.00000 0.50000

Table S5. Summary of the main superconducting properties of other metastable hydrides. λ, ω_{log} and T_c expressed in Kelvin (K) in the table. The DOS at E_f $N(E_F)$ is in units of (states/spin/Ry/cell).

	P(GPa)	λ	ω_{log}	$N(E_F)$	T_c (μ^*=0.1)
***C*2/*m*-La_2ThH_{18}**	200	1.96	646.55	16.33	112.85
***P*-3*m*1-$LaTh_2H_{18}$**	200	1.63	712.09	16.22	102.50
***Fm*-3*m*-$LaTh_3H_{24}$**	150	1.50	965.05	22.62	125.29
***Fm*-3*m*-$LaTh_3H_{24}$**	200	1.55	756.81	22.30	103.04

References

1. P. Song, Z. Hou, K. Hongo and R. Maezono, arXiv:2210.06371, 2022.