Supporting Information:

The systematic study on the phase diagram and superconductivity of ternary clathrate Ca−Sc−H at high pressures

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Table S1. Crystallographic data of the newly predicted structures of ternary Ca-Sc-H compounds under high pressure (P, in GPa). The unit of lattice parameters (*a*, *b*, and *c*) is given in Å.

Compound	Space group	*P* (GPa)	Lattice parameters (Å, °)	Atomic coordinates (fractional)			
				Atoms	x	y	Z
$CaSc_2H_3$	*Immm*	200	*a* = 7.7776	Ca(2c)	0.00000	0.00000	0.50000
			b = 3.6064	Sc(4f)	-0.16565	0.50000	0.00000
			c = 2.4821	H(4e)	-0.17429	0.00000	0.00000
			α =β =γ =90	H(2b)	-0.50000	0.00000	0.00000
$CaSc_2H_6$	$P6_3/mmc$	200	*a* = *c* = 2.9638	Ca(2c)	0.66667	0.33333	0.75000
			b =10.6052	Sc(4f)	0.66667	0.33333	0.41596
				H(4f)	0.66667	0.33333	0.57831
			α =γ = 90	H(4e)	1.00000	0.00000	0.16075
			β = 120	H(2a)	1.00000	0.00000	0.00000
				H(2d)	0.66667	0.33333	0.25000
$CaSc_2H_9$	*Immm*	200	*a* = 8.4205	Ca(2d)	0.00000	0.50000	0.00000
			b=3.9777	Sc(4f)	-0.33183	0.50000	0.00000
			c=2.7802	H(8n)	-0.16856	0.24599	0.00000
				H(4e)	-0.33056	0.00000	0.00000
			α =β =γ =90	H(4h)	-0.50000	0.26894	0.00000
				H(2a)	0.00000	0.00000	0.00000
$CaSc_3H_{12}$	$Pm\bar{3}m$	200	*a* = *b* = c=3.9532	Ca(1b)	0.50000	0.50000	0.50000

				Sc(3d)	0.00000	0.00000	0.50000
				H(8g)	0.24424	0.24424	0.24424
			α = β = γ = 90	H(3c)	0.00000	0.50000	0.50000
				H(1a)	0.00000	0.00000	0.00000
$CaScH_4$	$P\bar{6}m2$	200	*a* = b=2.9681	Ca(1a)	2.00000	1.00000	0.00000
			c=3.5625	Sc(1d)	1.33333	0.66667	0.50000
				H(2i)	0.66667	0.33333	0.26483
			α =β = 90	H(1b)	2.00000	1.00000	0.50000
			γ =120	H(1c)	1.33333	0.66667	0.00000
$CaScH_6$	$R\bar{3}m$	200	*a*=*b*=2.7310	Ca(3b)	0.33333	0.66667	0.66667
			c=14.5510	Sc(3a)	0.33333	0.66667	0.16667
				H(6c)	0.33333	0.66667	0.28040
			α = β =90	H(6c)	0.33333	0.66667	0.42319
			γ =120	H(6c)	0.33333	0.66667	0.54641
$CaScH_2$	$I4_1/amd$	200	*a* = *b* = 3.6074	Ca(4a)	0.00000	-0.50000	0.25000
			c=7.2067	Sc(4b)	0.00000	-0.50000	-0.25000
			α = β =γ=90	H(8e)	0.00000	-0.50000	-0.00674
$CaScH_3$	$P6_3/mmc$	200	*a* = *b* =2.8823	Ca(2c)	1.33333	0.66667	-0.75000
			c=7.1247	Sc(2a)	2.00000	1.00000	0.00000
			α = β =90	H(4f)	1.33333	0.66667	-0.39801
			γ =120	H(2b)	0.00000	0.00000	-0.25000
$CaScH_7$	$P4/mmm$	200	*a*=*b*=2.6547	Ca(1b)	0.00000	0.00000	0.50000
			c=4.5960	Sc(1c)	0.50000	0.50000	0.00000
				H(4i)	0.00000	0.50000	0.23784
			α = β = γ = 90	H(2h)	0.50000	0.50000	0.40427
				H(1a)	0.00000	0.00000	0.00000
Ca_2ScH_{10}	$C2/m$	200	*a*=4.8583	Ca(4i)	0.13504	0.50000	0.67529
			b=2.7538	Sc(2a)	0.00000	0.00000	0.00000
			c=7.2546	H_1(4i)	0.16251	0.50000	0.91684
				H_2(4i)	0.18179	0.50000	0.15147
				H_3(4i)	0.20351	0.50000	0.43464
			α =γ=90	H_4(4i)	0.02745	-0.00000	0.23080
			β =93.52	H_5(4i)	0.09487	-0.00000	0.49793
$CaScH_{12}$	$Pm\bar{3}m$	200	*a*=*b*=*c*=3.37410	Ca(1a)	0.00000	0.00000	0.00000
				Sc(1b)	0.50000	0.50000	0.50000
			α = β = γ = 90	H(12h)	0.00000	0.75294	0.50000

Table S2. Calculated elastic constants C_{ij} and bulk (B), shear (G), Young's (Y) moduli and Poisson's ratio (σ) of the stable phase of Ca-Sc-H systems selected pressures. All moduli are in GPa.

	$CaScH_6$ $R\bar{3}m$	$CaSc_2H_9$ $Immm$	Ca_2ScH_{10} $C2/m$	$CaScH_{12}$ $Pm\bar{3}m$
C_{11}	534.91	520.02	412.87	882.55
C_{12}	347.22	246.09	200.89	258.99
C_{13}	441.77	206.93	210.09	258.99
C_{22}	534.91	472.53	420.67	882.55
C_{23}	441.77	253.63	208.1	258.99
C_{33}	722.16	519.86	464.49	882.55
C_{44}	169.09	141.95	119.34	254.99
C_{55}	169.09	95.57	135.12	254.99
C_{66}	93.84	165.44	111.98	254.99
B	472.62	325.08	281.8	466.85
G	123.82	134.31	118.55	277.71
B/G	3.82	2.42	2.38	1.68
Y	341.63	354.16	311.92	695.27
σ	0.38	0.32	0.32	0.25

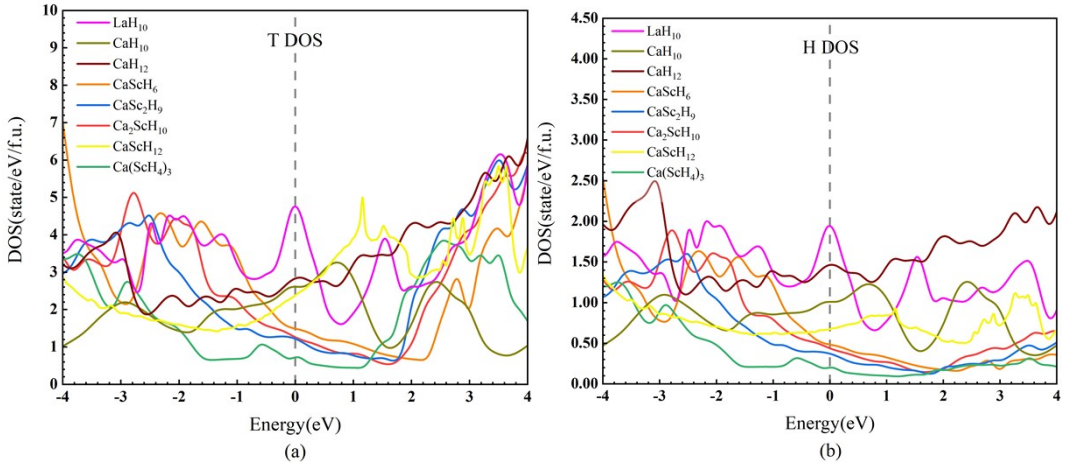

Figure S1. (a) Total electronic density of states (DOS) of and (b) partial DOS of H atoms in LaH_{10},[1] CaH_{10}, CaH_{12},[2] $CaScH_6$, Ca_2ScH_{10}, $CaSc_2H_9$, $CaScH_{12}$, and $Ca(ScH_4)_3$ at 200 GPa.

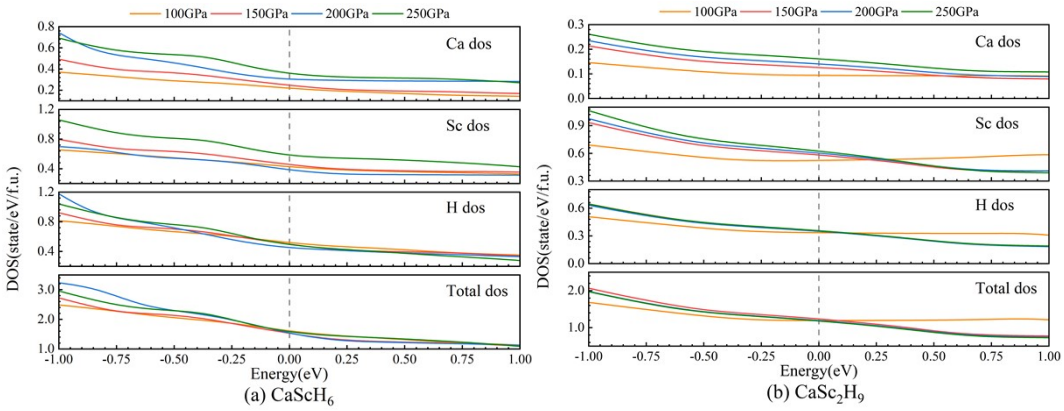

Figure S2. Electronic density of states for (a) $CaScH_6$ and (b) $CaSc_2H_9$ under the pressure from 100 to 250 Gpa.

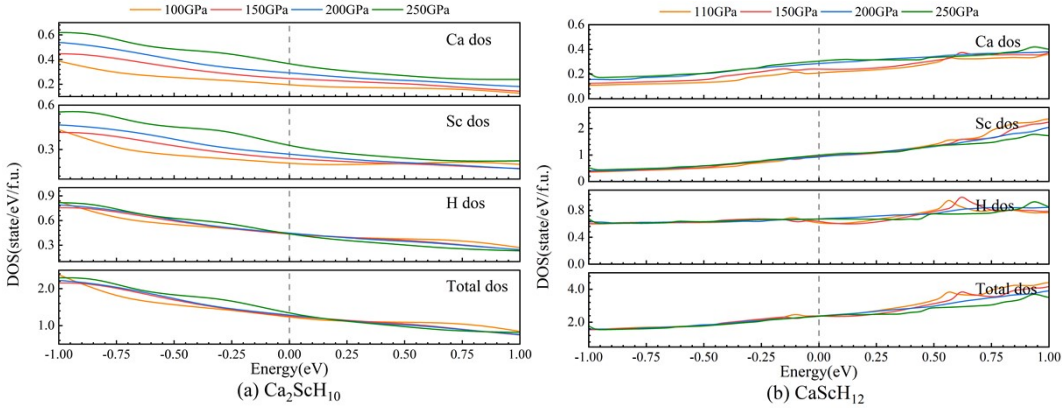

Figure S3. Electronic density of states for (a) Ca_2ScH_{10} and (b) $CaScH_{12}$ under the pressure from 100 to 250 GPa.

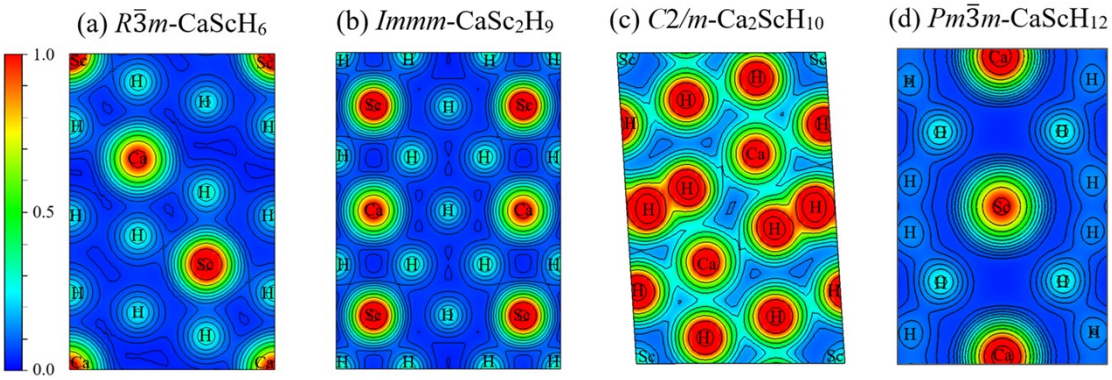

Figure S4. Charge state densities of $CaScH_6$, $CaSc_2H_9$, Ca_2ScH_{10}, and $CaScH_{12}$ at 200GPa. $CaScH_6$ along the (1 1 0) plane, (b) *Immm*- $CaSc_2H_9$ along the (1 0 0) plane, (c) *C2/m*-Ca_2ScH_{10} along the (1 0 0) plane, (d) $Pm\bar{3}m$-$CaScH_{12}$ along the (-2.02378 1 1.02378) plane at 200 GPa.

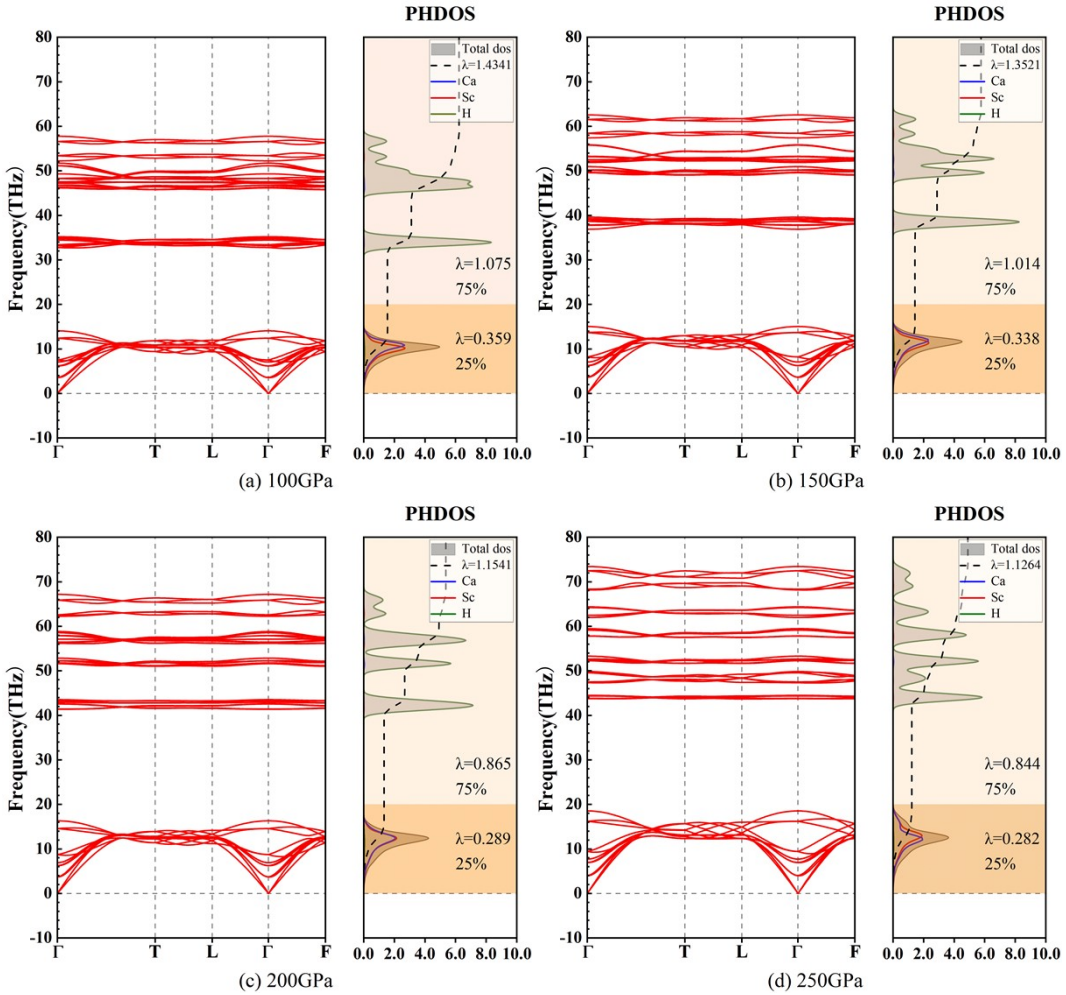

Figure S5. Pressure-dependent phonons and electron-phonon coupling spectra for $CaScH_6$.

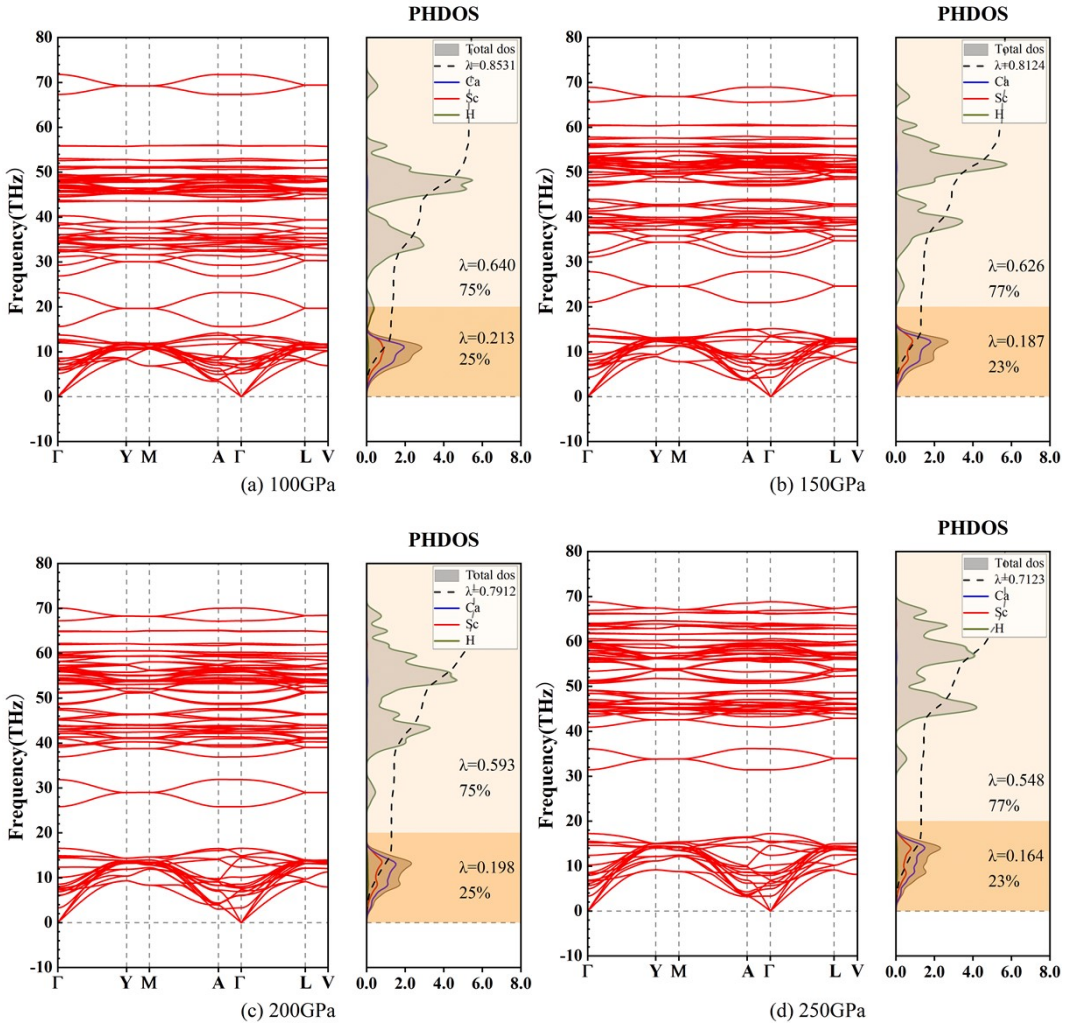

Figure S6. Pressure-dependent phonons and electron-phonon coupling spectra for Ca_2ScH_{10}.

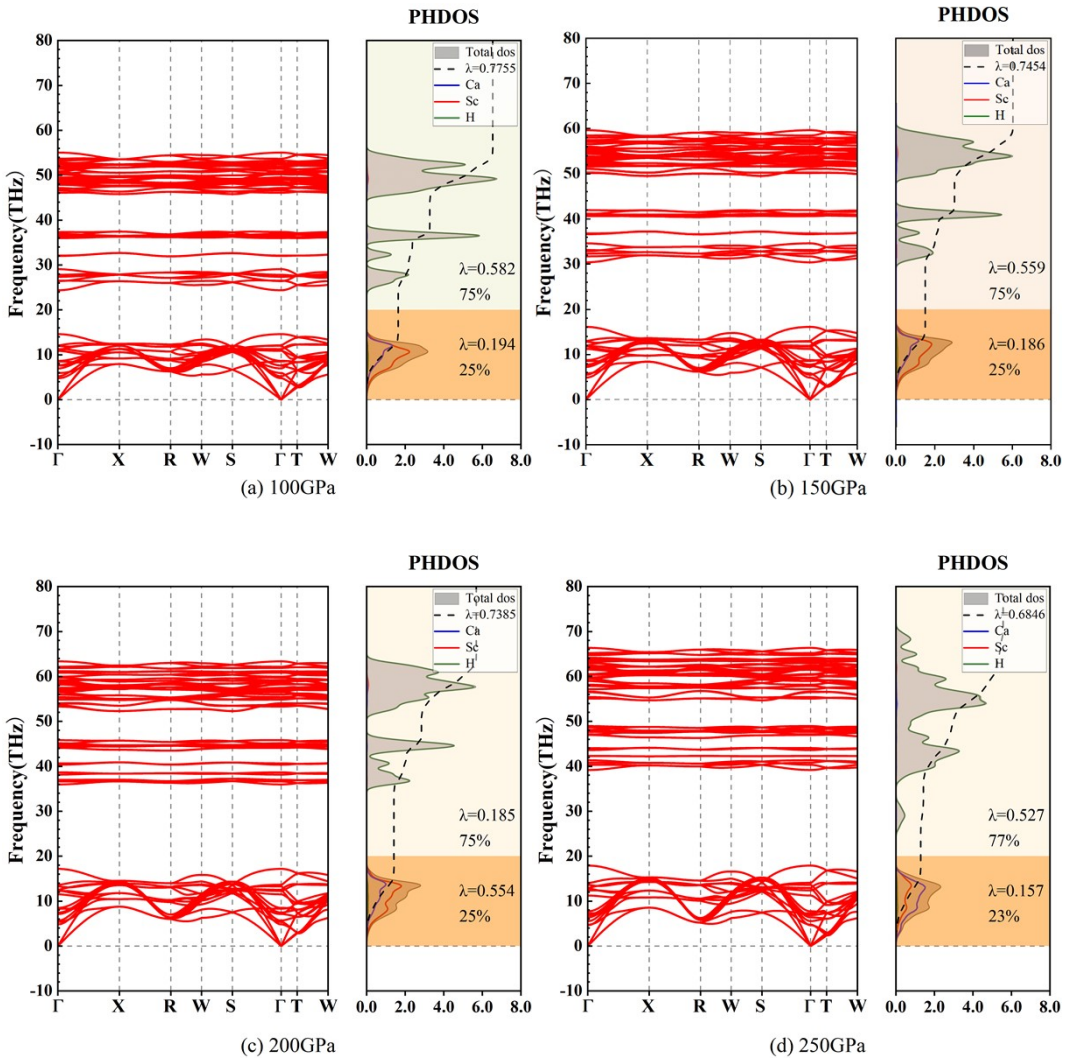

Figure S7. Pressure-dependent phonons and electron-phonon coupling spectra for $CaSc_2H_9$.

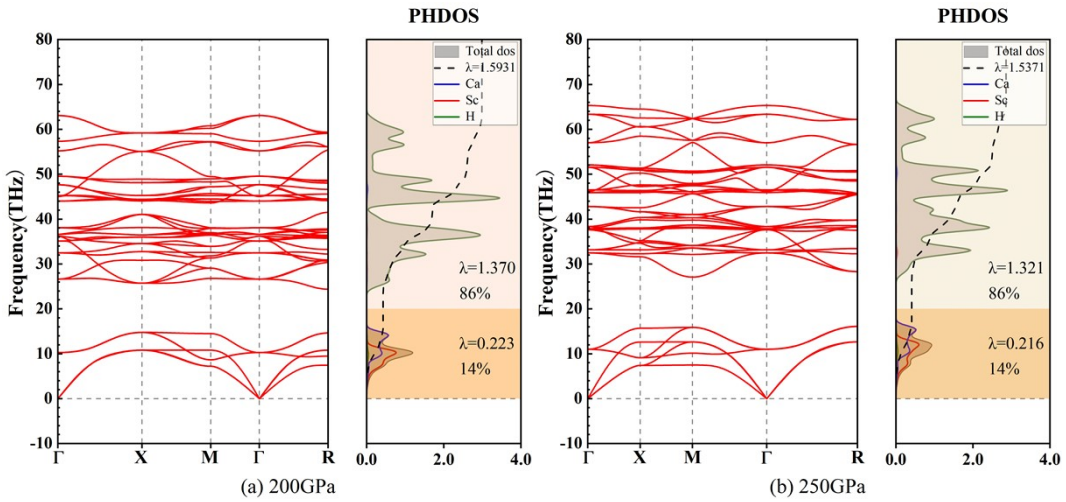

Figure S8. Pressure-dependent phonons and electron-phonon coupling spectra for $CaScH_{12}$.

POSCAR: $R\bar{3}m$-$CaScH_6$

1.00000000000000

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-1.3655303564336718 2.3651679682557902 0.0000000000000000

0.0000000000000000 0.0000000000000000 14.5510734252590499

Ca Sc H

3 3 18

Direct

0.0000000000000000 -0.0000000000000000 0.5000000000000000

0.6666666870000029 0.3333333429999996 0.8333333129999971

0.3333333429999996 0.6666666870000029 0.1666666719999981

-0.0000000000000000 -0.0000000000000000 0.0000000000000000

0.6666666870000029 0.3333333429999996 0.3333333429999996

0.3333333429999996 0.6666666870000029 0.6666666870000029

-0.0000000000000000 0.0000000000000000 0.1139987074155152

-0.0000000000000000 -0.0000000000000000 0.8860012775844800

0.6666666870000029 0.3333333429999996 0.4473320364155137

0.6666666870000029 0.3333333429999996 0.2193346205844867

0.3333333429999996 0.6666666870000029 0.7806653494155179

0.3333333429999996 0.6666666870000029 0.5526679635844863

-0.0000000000000000 0.0000000000000000 0.2560792338191430

0.0000000000000000 -0.0000000000000000 0.7439207371808580

0.6666666870000029 0.3333333429999996 0.5894125768191427

0.6666666870000029 0.3333333429999996 0.0772541021808524

0.3333333429999996 0.6666666870000029 0.9227458898191470

0.3333333429999996 0.6666666870000029 0.4105874231808573

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0.0000000000000000 -0.0000000000000000 0.6205891900781521

0.6666666870000029 0.3333333429999996 0.7127441229218451

0.6666666870000029 0.3333333429999996 0.9539225630781542

0.3333333429999996 0.6666666870000029 0.0460774479218432

0.3333333429999996 0.6666666870000029 0.2872558770781550

POSCAR:*Immm*-$CaSc_2H_9$

1.00000000000000

2.9568861110424893	0.0000000000000000	0.0000000000000000
0.0000000000000000	4.2242466708092792	0.0000000000000000
0.0000000000000000	0.0000000000000000	8.9283821013229794

Ca	Sc	H
2	4	8

Direct

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0.5000000000000000	-0.0000000000000000	0.5000000000000000
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0.0000000000000000	0.5000000000000000	0.6680458254090924
0.5000000000000000	-0.0000000000000000	0.8319541745909076
0.5000000000000000	-0.0000000000000000	0.1680458254090926
0.0000000000000000	0.2424798849811872	0.1700110720092442
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0.5000000000000000	0.7424798849811873	0.3299889429907535
-0.0000000000000000	-0.0000000000000000	0.3299590604263207
0.0000000000000000	-0.0000000000000000	0.6700409105736735
0.5000000000000000	0.5000000000000000	0.8299590894263265
0.5000000000000000	0.5000000000000000	0.1700409395736793
-0.0000000000000000	0.2706487328768900	0.5000000000000000
-0.0000000000000000	0.7293512671231100	0.5000000000000000
0.5000000000000000	0.7706487328768900	-0.0000000000000000
0.5000000000000000	0.2293512671231101	-0.0000000000000000
-0.0000000000000000	-0.0000000000000000	-0.0000000000000000
0.5000000000000000	0.5000000000000000	0.5000000000000000

POSCAR:*C*2/*m*-Ca_2ScH_{10}

1.00000000000000

5.1612985613551778 0.0000000000000000 -0.0360512758029978

0.0000000000000000 2.9986583081895786 0.0000000000000000

-0.2886784461768619 0.0000000000000000 7.6333558977245541

Ca Sc H

4 2 20

Direct

0.1444041610647362 0.5000000000000000 0.6769673770373635

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0.5000000000000000 0.5000000000000000 0.0000000000000000

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0.6758684837595180 0.0000000000000000 0.1471187808730320

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0.8028439683650961 0.5000000000000000 0.5704978987881320

0.6971560316349039 -0.0000000000000000 0.4295020712118658

0.3028439683650961 -0.0000000000000000 0.5704978987881320

POSCAR:$Pm\bar{3}m$-$CaScH_{12}$

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0.0000000000000000	3.3740634318908138	0.0000000000000000
0.0000000000000000	0.0000000000000000	3.3740634318908138

Ca	Sc	H
12	1	1

Direct

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0.5000000000000000	0.0000000000000000	0.7529382654494629
0.5000000000000000	0.0000000000000000	0.2470617345505372
0.7529382654494629	0.5000000000000000	0.0000000000000000
0.2470617345505372	0.5000000000000000	-0.0000000000000000
0.7529382654494629	0.0000000000000000	0.5000000000000000
0.2470617345505372	0.0000000000000000	0.5000000000000000
-0.0000000000000000	0.5000000000000000	0.2470617345505372
0.0000000000000000	0.5000000000000000	0.7529382654494629
0.5000000000000000	0.7529382654494629	-0.0000000000000000
0.5000000000000000	0.2470617345505372	0.0000000000000000
0.0000000000000000	0.0000000000000000	-0.0000000000000000
0.5000000000000000	0.5000000000000000	0.5000000000000000

References

1. Liu, H.; Naumov, I. I.; Hoffmann, R.; Ashcroft, N. W.; Hemley, R. J., Potential High-Tc Superconducting Lanthanum and Yttrium Hydrides at High Pressure. *Proceedings of the National Academy of Sciences* **2017**, *114*, 6990-6995.

2. Wang, H.; Tse, J. S.; Tanaka, K.; Iitaka, T.; Ma, Y., Superconductive Sodalite-Like Clathrate Calcium Hydride at High Pressures. *Proceedings of the National Academy of Sciences* **2012**, *109*, 6463-6466.