Supporting Information

Prediction of pressure-induced superconductivity in the novel ternary system $ScCaH_{2n}$(n=1-6)

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Figure S1. The phonon spectrum of $ScCaH_{2n}$(n=1-3,5). There is absence imaginary frequency of each structure to show its dynamic stability.

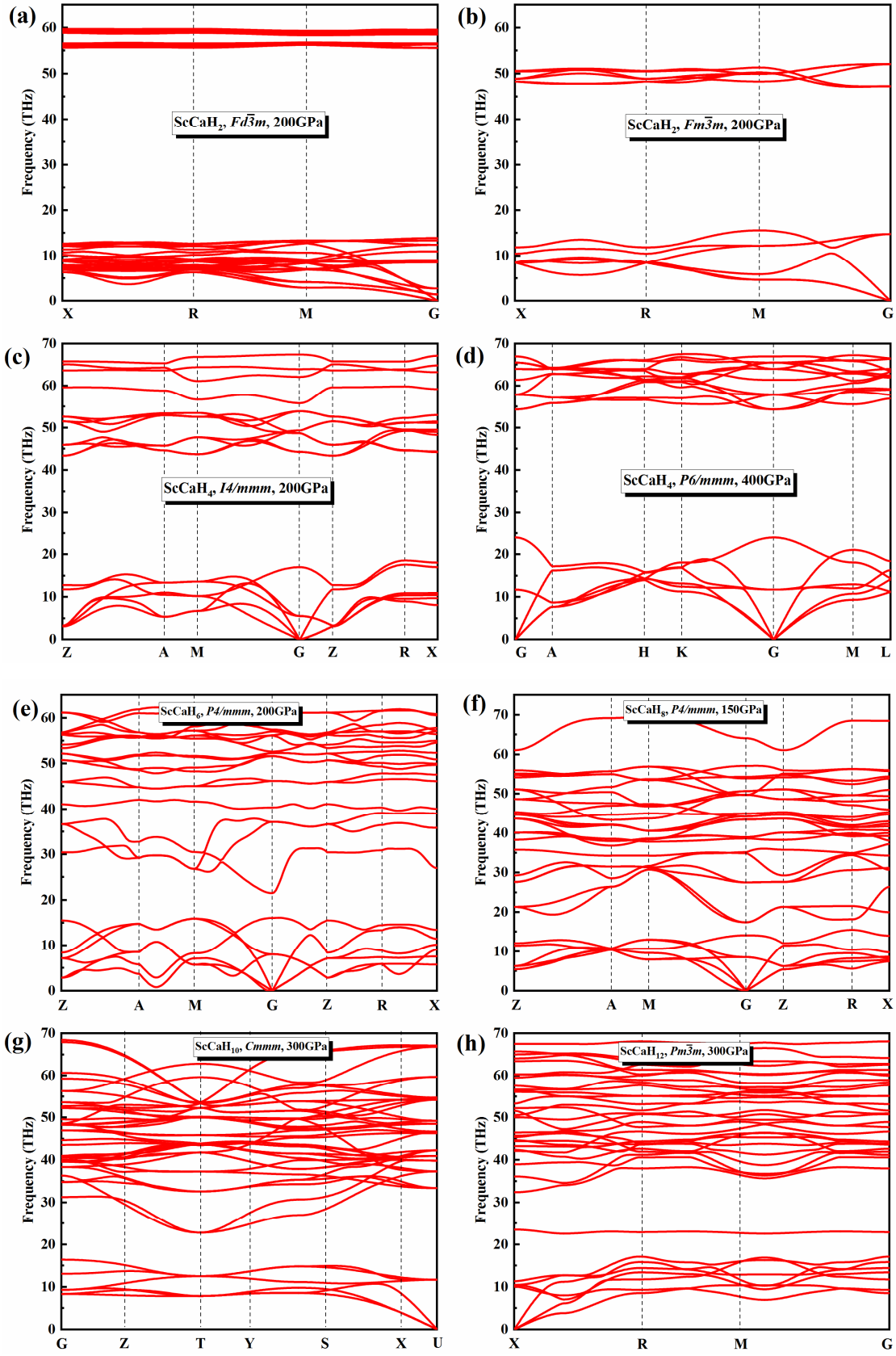


Figure S2. The enthalpies of formation with ZPE of $ScCaH_{2n}$(n=2-6) with respect to $ScCaH_2$ and H_2 at 100-300 GPa.

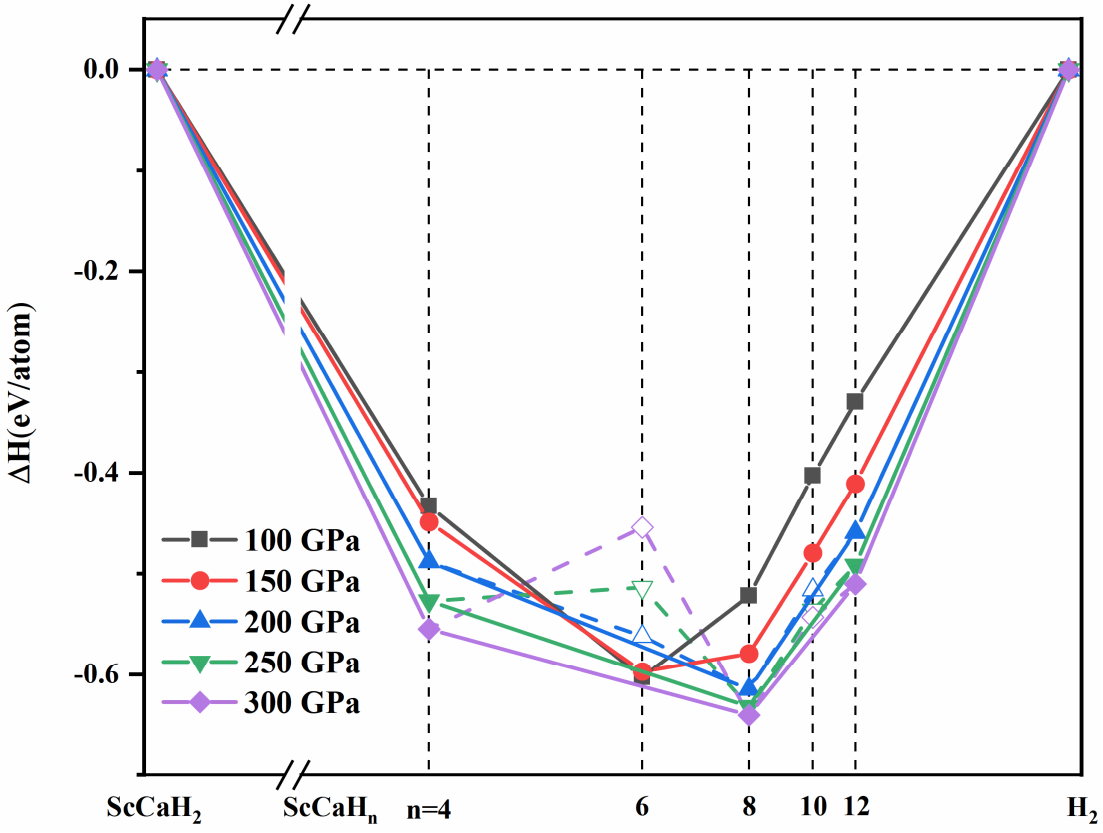
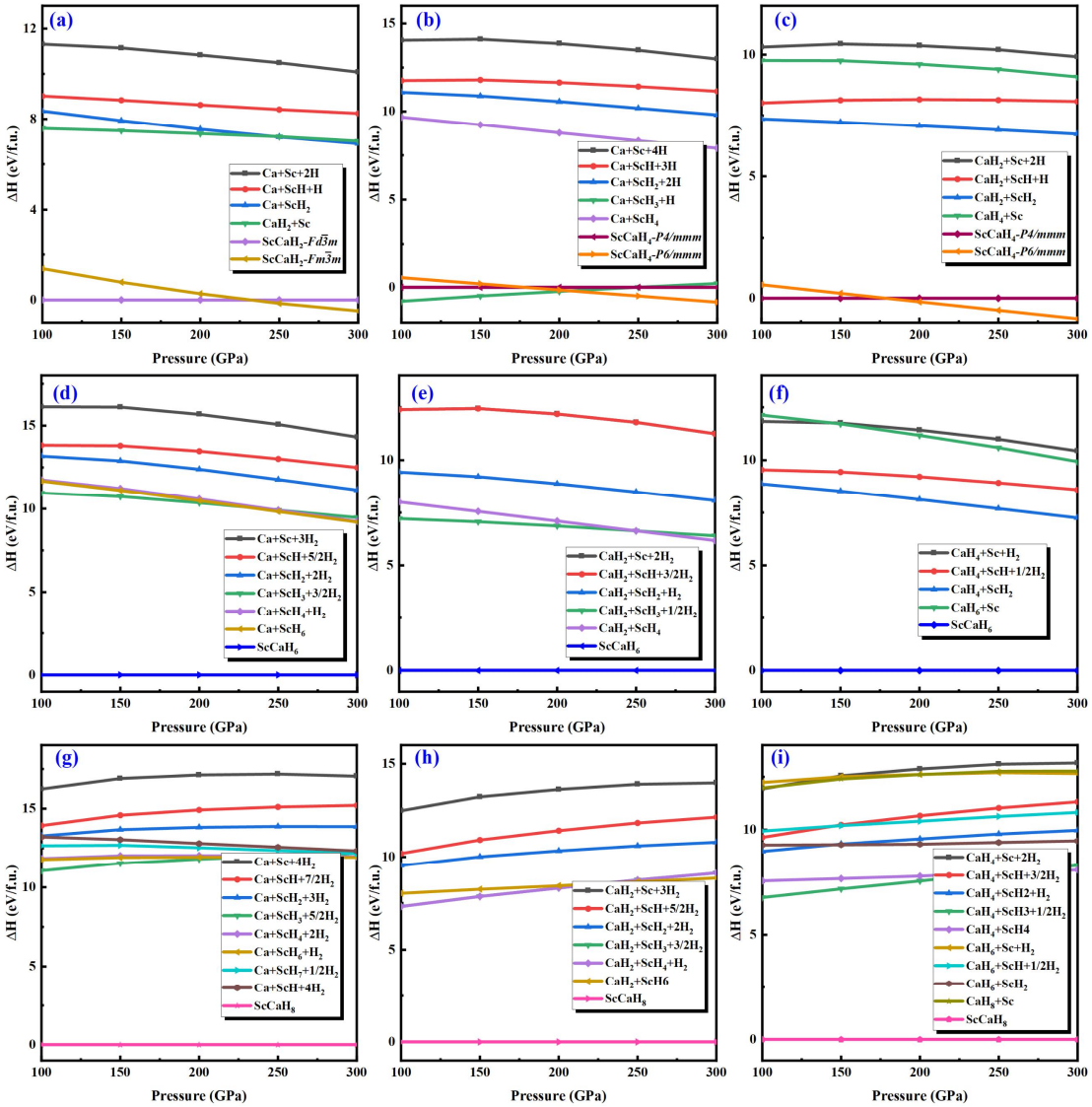


Figure S3. (a) The enthalpy difference curves with respect to $ScCaH_2$ as a function of pressure; (b)-(c) The enthalpy difference curves with respect to $ScCaH_4$ as a function of pressure; (d)-(f) The enthalpy difference curves with respect to $ScCaH_6$ as a function of pressure; (g)-(i) The enthalpy difference curves with respect to $ScCaH_8$ as a function of pressure; (j)-(m) The enthalpy difference curves with respect to $ScCaH_{10}$ as a function of pressure; (n)-(r) The enthalpy difference curves with respect to $ScCaH_{12}$ as a function of pressure.



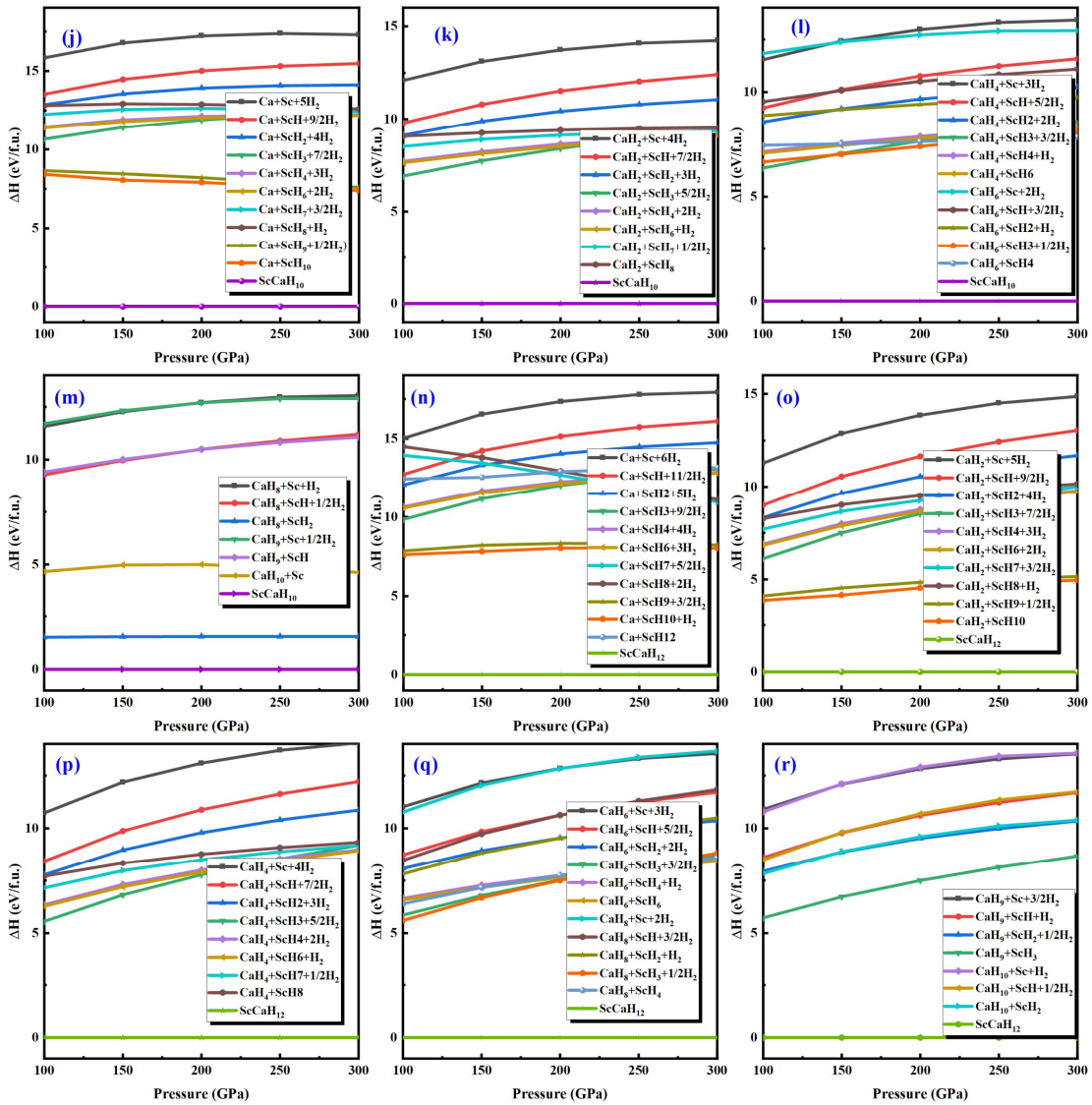

(j)
Ca+Sc+5H2
Ca+ScH+9/2H2
Ca+ScH2+4H2
Ca+ScH3+7/2H2
Ca+ScH4+3H2
Ca+ScH6+2H2
Ca+ScH7+3/2H2
Ca+ScH8+H2
Ca+ScH9+1/2H2)
Ca+ScH10
ScCaH10
(k)
CaH2+Sc+4H2
CaH2+ScH+7/2H2
CaH2+ScH2+3H2
CaH2+ScH3+5/2H2
CaH2+ScH4+2H2
CaH2+ScH6+H2
CaH2+ScH7+1/2H2
CaH2+ScH8
ScCaH10
(l)
CaH4+Sc+3H2
CaH4+ScH+5/2H2
CaH4+ScH2+2H2
CaH4+ScH3+3/2H2
CaH4+ScH4+H2
CaH4+ScH6
CaH6+Sc+2H2
CaH6+ScH+3/2H2
CaH6+ScH2+H2
CaH6+ScH3+1/2H2
CaH6+ScH4
ScCaH10
(m)
CaH8+Sc+H2
CaH8+ScH+1/2H2
CaH8+ScH2
CaH8+Sc+1/2H2
CaH9+ScH
CaH10+Sc
ScCaH10
(n)
Ca+Sc+6H2
Ca+ScH+11/2H2
Ca+ScH2+5H2
Ca+ScH3+9/2H2
Ca+ScH4+4H2
Ca+ScH6+3H2
Ca+ScH7+5/2H2
Ca+ScH8+2H2
Ca+ScH9+3/2H2
Ca+ScH10+H2
Ca+ScH12
ScCaH12
(o)
CaH2+Sc+5H2
CaH2+ScH+9/2H2
CaH2+ScH2+4H2
CaH2+ScH3+7/2H2
CaH2+ScH4+3H2
CaH2+ScH6+2H2
CaH2+ScH7+3/2H2
CaH2+ScH8+H2
CaH2+ScH9+1/2H2
CaH2+ScH10
ScCaH12
(p)
CaH4+Sc+4H2
CaH4+ScH+7/2H2
CaH4+ScH2+3H2
CaH4+ScH3+5/2H2
CaH4+ScH4+2H2
CaH4+ScH6+H2
CaH4+ScH7+1/2H2
CaH4+ScH8
ScCaH12
(q)
CaH6+Sc+3H2
CaH6+ScH+5/2H2
CaH6+ScH2+2H2
CaH6+ScH3+3/2H2
CaH6+ScH4+H2
CaH6+ScH6
CaH8+Sc+2H2
CaH8+ScH+3/2H2
CaH8+ScH2+H2
CaH8+ScH3+1/2H2
CaH8+ScH4
ScCaH12
(r)
CaH9+Sc+3/2H2
CaH9+ScH+H2
CaH9+ScH2+1/2H2
CaH9+ScH3
CaH10+Sc+H2
CaH10+ScH+1/2H2
CaH10+ScH2
ScCaH12
ΔH (eV/f.u.)
Pressure (GPa)

Figure S4. The predicted crystal structures for $ScCaH_{2n}$(n=1, 2). ELF in (-1471.49 -1 -1) sections for *Fd*$\bar{3}$*m*-$ScCaH_2$ at 150 GPa. ELF in (-1 -1 0) sections for *Fm*$\bar{3}$*m*-$ScCaH_2$ at 300 GPa. ELF in (1 1 0) sections for *P4/mmm*-$ScCaH_4$ at 150 GPa. ELF in (0 0 -1) sections for *P6/mmm*-$ScCaH_4$ at 300 GPa. It is obvious that in the two structures of $ScCaH_2$, there is no interaction between the hydrogen atoms. In *P4/mmm*-$ScCaH_4$, there are "H_2" structures and obviously interaction between these two hydrogen atoms. In *P6/mmm*-$ScCaH_4$, graphene layered structures are formed between the hydrogen atoms and the interaction between hydrogen atoms are strong.

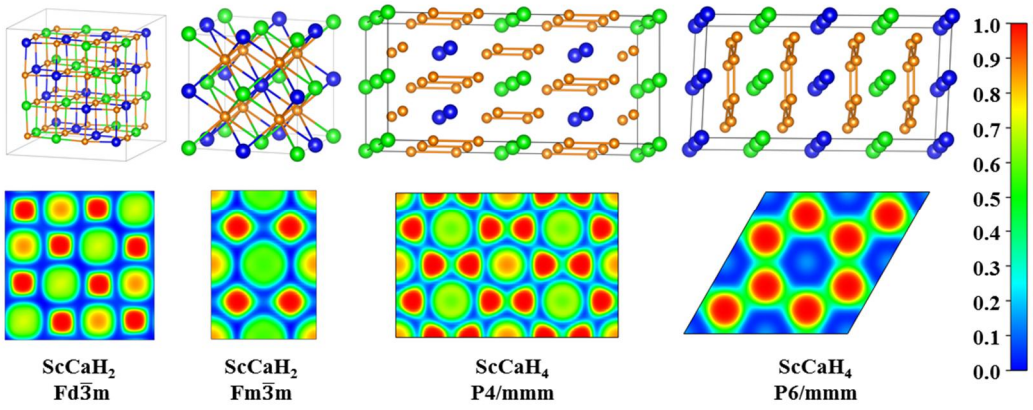
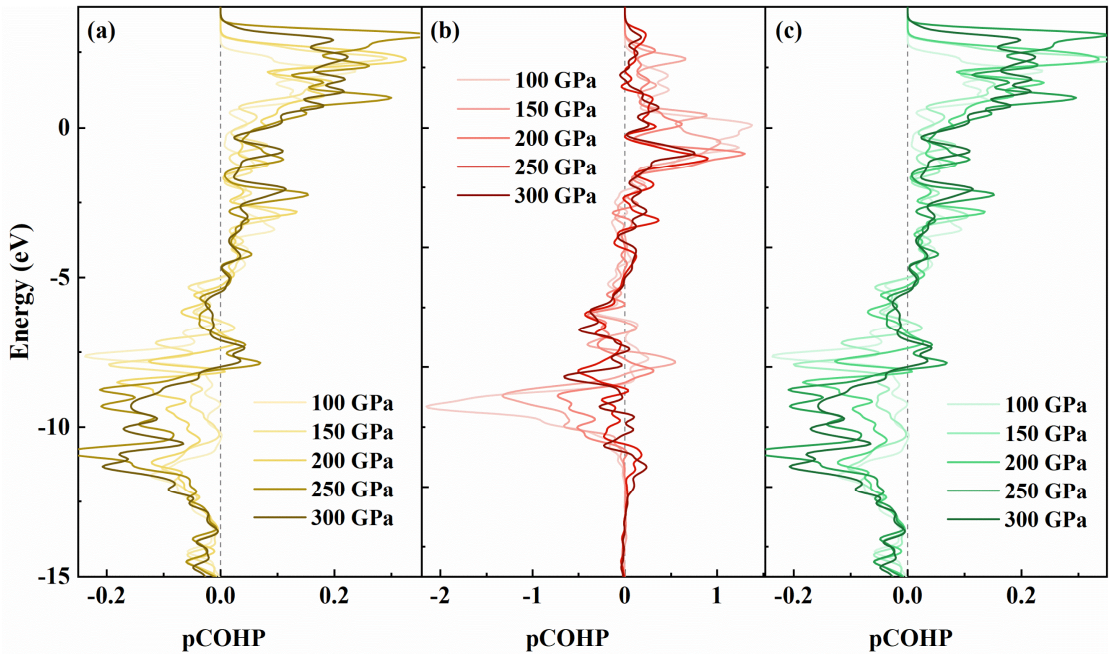


Figure S5. PCOHP for pairs of H…H in $ScCaH_8$ (a) Two hydrogen atoms connected by **a** bond (b) Two hydrogen atoms connected by **b** bond (c) Two hydrogen atoms connected by **c** bond at different pressures. Energies are normalized to the respective Fermi level. And the PCOHP for pairs of H…H in CaH6.



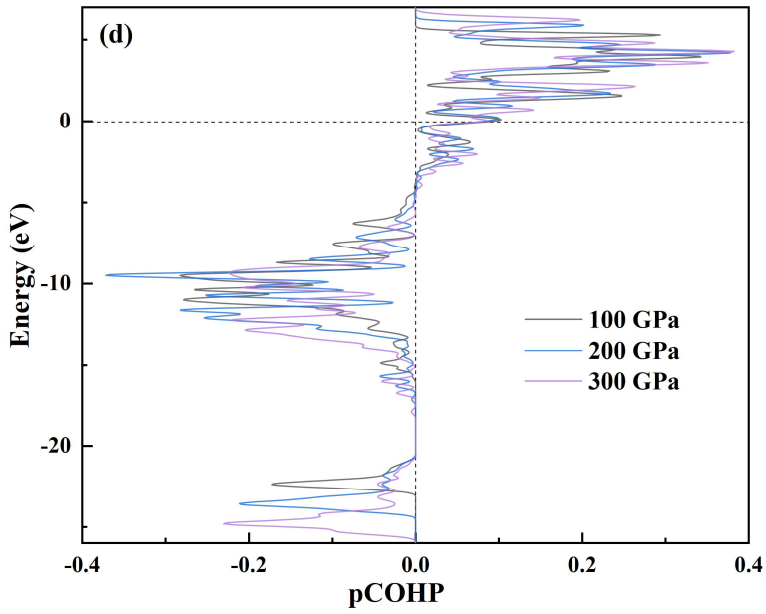


Figure S6. Electronic band structures and density of states of $ScCaH_{2n}$(n=1-3,5). The results show that these compounds are all metallic. In $ScCaH_2$ and $ScCaH_4$, the contribution of hydrogen atom at Fermi level in density of States are less than that of Ca and SC atoms. In $ScCaH_6$ and $ScCaH_{10}$, the contribution of hydrogen atom at Fermi level in the DOS are obviously increased.

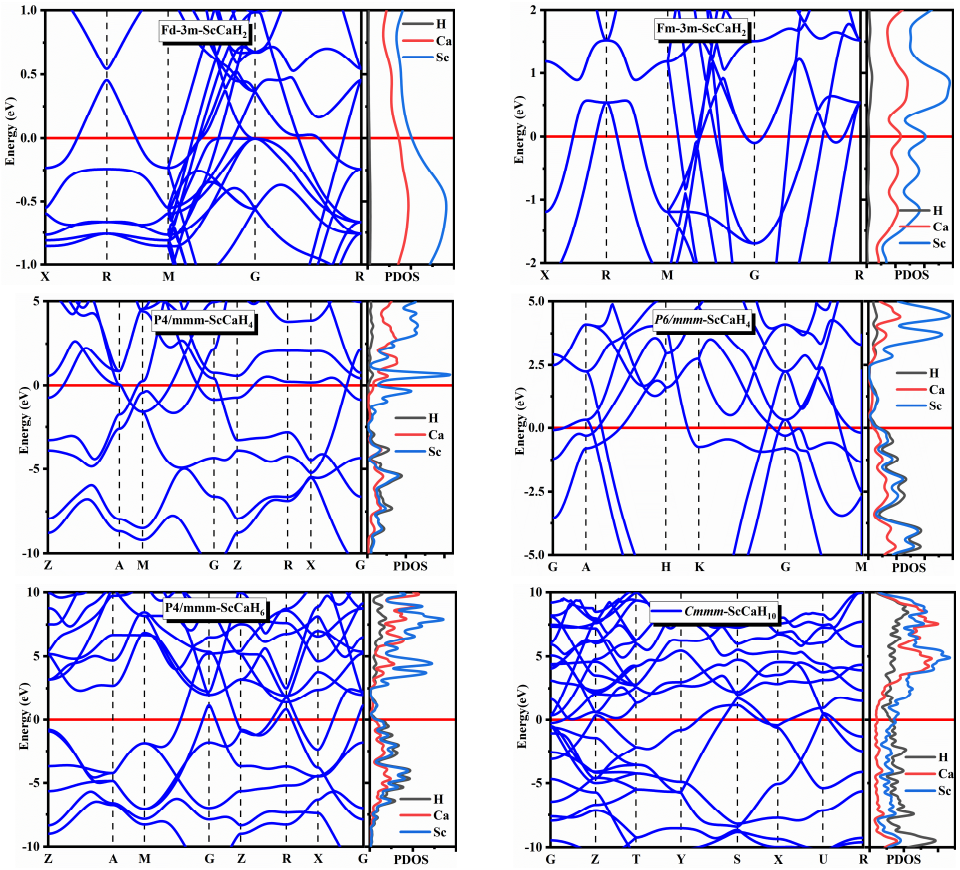


Figure S7. Critical temperature T_c as a function of the empirical parameter μ^*.

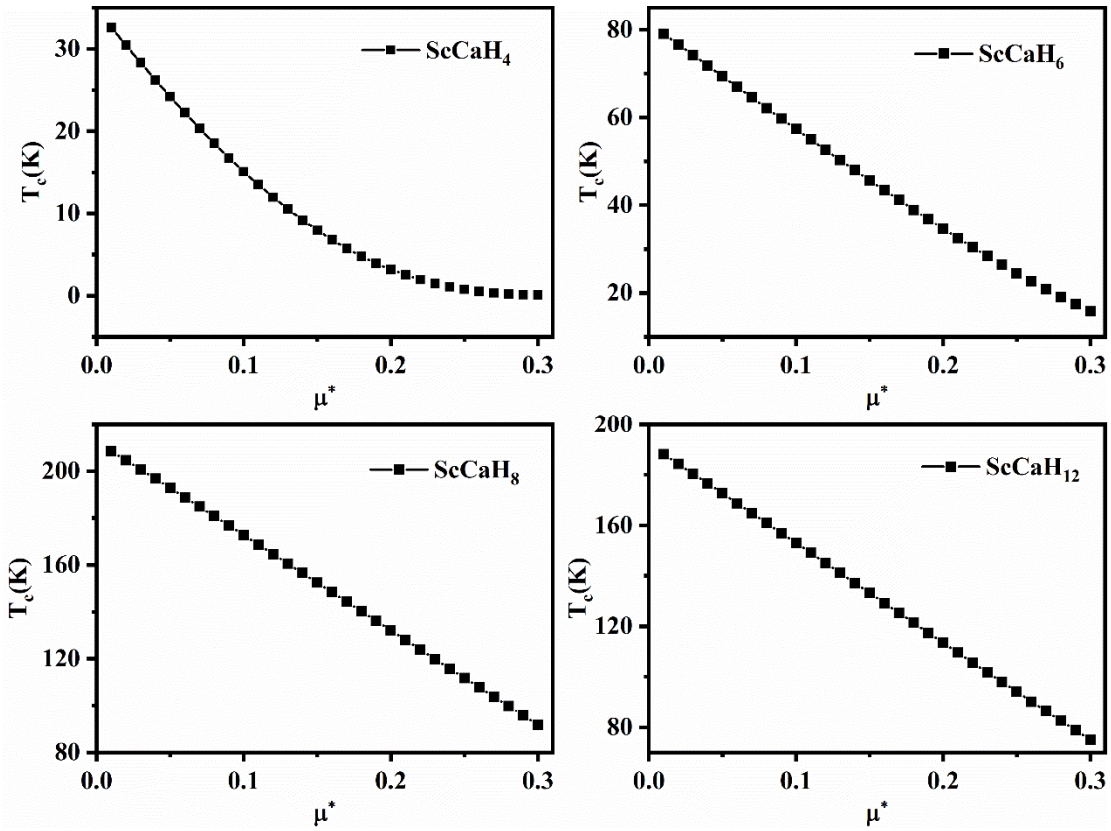


Table S1. Calculated structural parameters of the predicted phase for $ScCaH_2$

Compound	Space group	Lattice Parameters (Å)	Atomic coordinates (fractional)			
			Atoms	X	Y	Z
$ScCaH_2$	***$Fd\bar{3}m$***	a=b=c=7.4515 $\alpha=\beta=\gamma=90^\circ$	H(32e)	0.11746	0.88254	0.11746
			Ca(16d)	0.87500	0.87500	-0.37500
			Sc(16d)	0.62500	0.87500	0.37500
	$Fm\bar{3}m$	a=b=c4.2574 $\alpha=\beta=\gamma=90^\circ$	H(8c)	0.75000	0.25000	0.25000
			Ca(4b)	0.00000	0.00000	0.50000
			Sc(4a)	0.00000	0.00000	0.00000
$ScCaH_4$	***P4/mmm***	a=b=c=2.38750 $\alpha=\beta=\gamma=90^\circ$	H(2h)	0.50000	0.50000	0.15002
			H(2g)	0.00000	0.00000	0.33380
			Ca(1d)	0.50000	0.50000	0.50000
			Sc(1a)	0.00000	0.00000	0.00000
	P6/mmm	a=b=2.43200, c=4.41970 $\alpha=\beta=90^\circ$, $\gamma=120^\circ$	H(4h)	0.33333	0.66667	0.26548
			Ca(1a)	0.00000	0.00000	0.00000
			Sc(1b)	0.00000	0.00000	0.50000
$ScCaH_6$	***P4/mmm***	a=b=2.78960, c=4.02080 $\alpha=\beta=\gamma=90^\circ$	H(4i)	0.50000	0.0000	0.76734
			H(1c)	0.50000	0.50000	0.0000
			H(1b)	0.0000	0.0000	0.50000
			Ca(1d)	0.50000	0.50000	0.50000
			Sc(1a)	0.0000	0.0000	0.0000
$ScCaH_8$	***P4/mmm***	a=b=2.59780, c=4.86800 $\alpha=\beta=\gamma=90^\circ$	H(2g)	0.00000	0.00000	0.89919
			H(4i)	0.50000	0.00000	0.23478
			H(2h)	0.50000	0.50000	0.36819
			Ca(1b)	0.00000	0.00000	0.50000
			Sc(1c)	0.50000	0.50000	0.00000
$ScCaH_{10}$	***Cmmm***	a=c=4.71340, b=3.36210 $\alpha=\beta=\gamma=90^\circ$	H(8m)	0.75000	0.75000	0.77911
			H(8o)	0.86107	-0.00000	0.61550
			H(4g)	0.37705	-0.00000	0.00000
			Ca(2c)	0.50000	0.00000	0.50000
			Sc(2a)	0.0000	0.00000	0.00000
$ScCaH_{12}$	***$Pm\bar{3}m$***	a=b=c=4.00260 $\alpha=\beta=\gamma=90^\circ$	H(12h)	-0.00000	0.76123	0.50000
			Ca(1a)	0.00000	0.00000	0.00000
			Sc(1b)	0.50000	0.50000	0.50000

Table S2. Integrated COHP (ICOHP) up to the Fermi level for H…H in $ScCaH_8$.

Pressure	ICOHP (eV/atom pair)			
	a	**b**	**c**	**CaH_6**
100	-0.41772	-1.29912	-0.41699	-1.02540
150	-0.49917	-1.17591	-0.50345	-1.16565
200	-0.61116	-0.89808	-0.62243	-1.25351
250	-0.80201	-0.68686	-0.79061	-1.30817
300	-0.75937	-0.62798	-0.75859	-1.35751

Table S3. The calculated EPC parameter $\boldsymbol{\lambda}$, logarithmic average phonon frequency $\boldsymbol{\omega_{log}}$ (K), and superconducting transition temperatures $\boldsymbol{T_c}$ at high pressures.

Structure	Pressure (GPa)	$\boldsymbol{\lambda}$	$\boldsymbol{\omega_{log}}$(K)	$\boldsymbol{T_c}$(K) ($\boldsymbol{\mu^* = 0.1}$)
Cmcm-CaH_4	200	0.46	1398.0	11.6[a]
I4/mmm-ScH_4	250	0.81	1891.8	78.0[b]
P4/mmm-$ScCaH_8$	200	1.89	1252.4	212.1[c]
$Im\bar{3}m$-CaH_6	200	1.98	1237.7	211.3[d]
$Im\bar{3}m$-ScH_6	350	1.25	1433.4	169.0[b]
$Pm\bar{3}m$-$ScCaH_{12}$	200	1.70	1201.1	182.1[c]

[a] Inorg. Chem. 2019, 58, 2558

[b] J. Phys. Chem. C 2018, 122, 6298

[c] This work

[d] J. Phys.: Condens. Matter 2019, 31, 245404