Supplementary information for

"Phase diagrams and superconductivity of ternary Ca-Al-H compounds under high pressure"

Ming Xu[1], Defang Duan[2], Wendi Zhao[1], Decheng An[2] Hao Song[1,*] and Tian Cui[1,2]†

[1]*Institute of High Pressure Physics, School of Physical Science and Technology, Ningbo University, Ningbo 315211, China*

[2]*State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun 130012, China*

*Corresponding author: songhao@nbu.edu.cn
†Corresponding author: cuitian@nbu.edu.cn

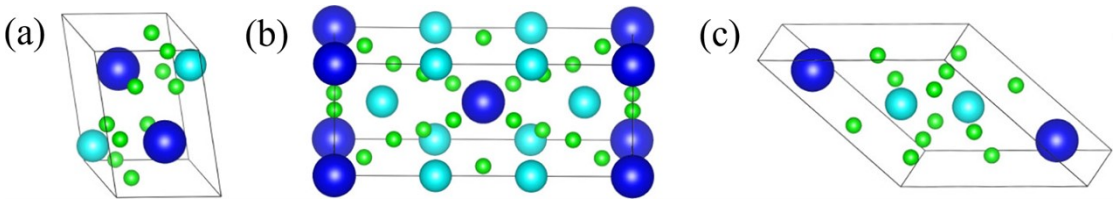

Fig. S1. The crystal structures of (a) *Cmcm*-$CaAlH_5$, (b) *Pnnm*-$CaAl_2H_8$, (c) $P2_1/m$-$CaAlH_5$. Among them, calcium atoms are dark blue, aluminum atoms are light blue, and hydrogen atoms are green.

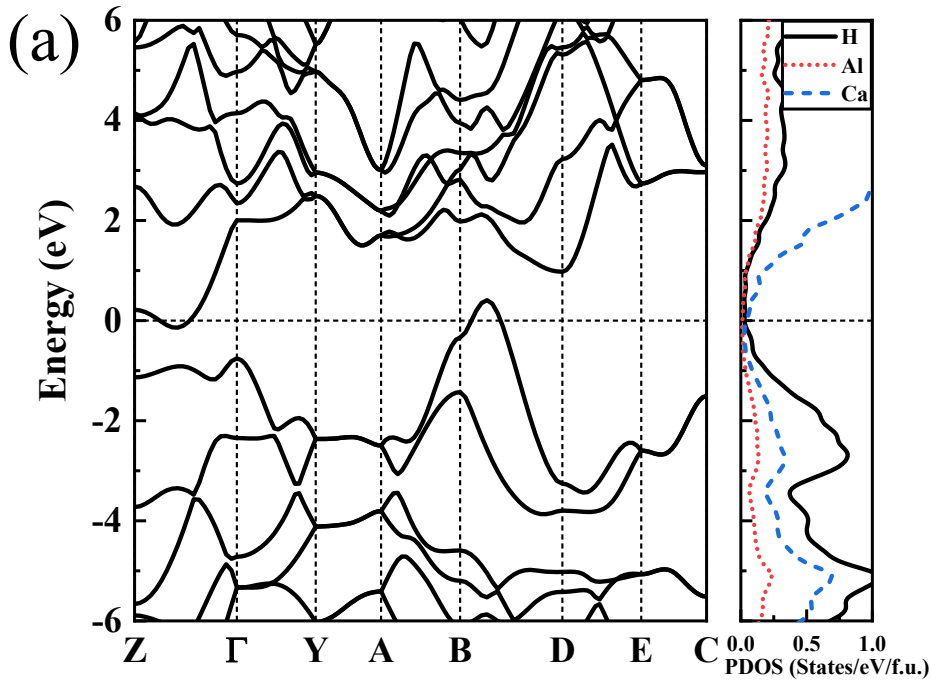

Fig.S2. The band structures and partial electronic density of states (PDOS) of (a) $P2_1/m$-$CaAlH_5$ at 200 GPa using the GGA-PBE functionals.

With the pressure increases, the T_c of $CaAlH_7$ decreases significantly as shown in the Fig. S3. (a) At the same time, we also give the superconducting transition temperature T_c of $CaAlH_7$ for different Coulomb potential μ* at 50 GPa. (b) The empiric values of the two Coulomb potential μ* we commonly use are 0.1 and 0.13, and their corresponding superconducting transition temperatures T_c are 71 K and 62 K, respectively.

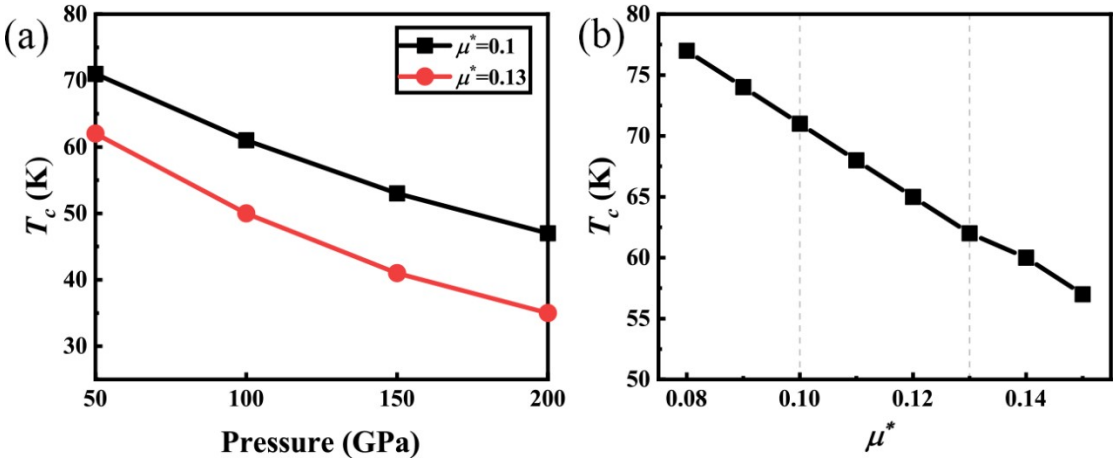

Fig. S3 (a) Calculated the superconducting transition temperature T_c of $CaAlH_7$ at different pressures. (b) Calculated the superconducting transition temperatures T_c of $CaAlH_7$ at 50 GPa as a function of Coulomb potential μ*.

TABLE I. Structural information of predicted hydrides.

Structure	Lattice parameters(Å)	Atomic	coordinates			Sites
		H1	0.000000	1.000000	-0.889920	2g
*P*4/*mmm*	a=b=2.6015	H2	0.000000	0.500000	-0.288020	4i
$CaAlH_7$	c=4.2674	H3	0.500000	0.500000	-0.500000	1d
200 GPa	α=β=γ=90	Al1	0.000000	0.000000	-0.500000	1b
		Ca1	-0.500000	0.500000	0.000000	1c
	a= 5.0112	H1	0.495540	0.496650	0.206350	4f
$P2_1/m$	b= 3.7364	H2	-0.115550	0.454200	0.244450	4f
$CaAlH_5$	c= 3.2078	H3	0.194180	0.250000	1.210440	2e
200 GPa	α=γ=90	Al1	0.662960	0.750000	0.377780	2e
	β= 63.9052	Ca1	0.186540	0.750000	0.209030	2e
		H1	-0.114290	0.211810	0.800710	8h
Cmcm	a= 3.8830	H2	0.748210	-0.505780	0.500000	4g
$CaAlH_5$	b= 3.4174	H3	1.000000	0.000000	0.601300	4e
50 GPa	c= 8.8034	Al1	0.500000	0.000000	0.839420	4f
	α=β=γ=90	Ca1	0.000000	-0.500000	1.000000	4c
		H1	0.000000	-0.398440	-1.500000	4h
Ca_2AlH_{12}	a= 8.0124	H2	0.343450	-0.716860	-1.000000	8n
Immm	b= 4.4908	H3	0.186400	-0.908070	-1.000000	8n
200 GPa	c= 2.6524	H4	0.000000	-0.805120	-2.000000	4g
	α=β=γ=90	Al1	0.000000	0.000000	-0.500000	2c
		Ca1	0.342920	1.000000	-1.500000	4f
		H1	-0.114290	0.211810	0.800710	8h
$CaAl_2H_8$	a= 3.8830	H2	0.748210	-0.505780	0.500000	4g
Pnnm	b= 3.4174	H3	1.000000	0.000000	0.601300	4e
50 GPa	c= 8.8034	Al1	0.500000	0.000000	0.839420	4f
	α=β=γ=90	Ca1	0.000000	-0.500000	1.000000	4c