Design Principles for High Temperature Superconductors with

Hydrogen-based Alloy Backbone at Moderate Pressure

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Abstract :

Hydrogen-based superconductors provide a route to the long-sought goal of room-temperature superconductivity, but the high pressures required to metallize these materials limit their immediate application. For example, carbonaceous sulfur hydride, the first room-temperature superconductor, can reach a critical temperature (T_c) of 288 K only at the extreme pressure of 267 GPa. The next recognized challenge is the realization of room-temperature superconductivity at significantly lower pressures. Here, we propose a strategy for the rational design of high-temperature superconductors at low pressures by alloying small-radius elements and hydrogen to form ternary hydride superconductors with alloy backbones. We identify a hitherto unknown 'fluorite-type' backbone in compositions of the form AXH₈, which exhibit high temperature superconductivity at moderate pressures. The $Fm\overline{3}m$ phase of LaBeH₈, with a 'fluorite-type' H-Be alloy backbone, is predicted to be metastable and superconducting with a $T_c \sim 191$ K at 50 GPa; a substantially lower pressure than that required by the geometrically similar clathrate hydride LaH₁₀ (170 GPa). Our approach paves the way for finding high- T_c ternary hydride superconductors at conditions close to ambient pressures.

Hydrogen, the lightest element, has been predicted to become a metallic solid and exhibit hightemperature superconductivity (with T_{cs} in the range 100-760 K) under extreme pressures [1]. However, metallization of solid hydrogen is still uncertain in high-pressure experiments up to 490 GPa [2]. It was predicted that comparable high-temperature superconductivity could be achieved in hydrogen dominant materials by "chemically pre-compressing" the hydrogen with other elements to produce the valence density sufficient for metallization at lower pressures [3]. Based on this principle, a series of H-based superconductors were theoretically predicted and some were successfully synthesized in the laboratory. Notably, H₃S was predicted to be a high-temperature superconductor with a T_c of 191-204 K [4], which was later confirmed by an experimentally measured T_c of 203 K at 155 GPa [5,6]. Following this success, several new hydrides in the clathrate hydride family, which consist of a pure hydrogen backbone pre-compressed by heavy metal atoms, were predicted and then synthesized, including LaH₁₀ with a T_c of 250-260 K at 170-180 GPa [7-11]. Several geometric classes of hydrides were found to facilitate high T_c ; in addition to the covalent six-fold cubic H₃S and the sodalite-type clathrate hydrides, a class of "penta-graphene-like" hydrides with high T_c were recently predicted at 250 GPa [12]. Although the pressures at which these H-based superconductors become stable are much lower than pure metallic hydrogen, the required pressure (> 150 GPa) is still difficult to obtain. The next challenge is therefore the realization of room-temperature superconductivity at significantly lower pressures (with a clear final goal of reaching ambient pressure).

Various routes have been explored to reduce the pressure at which hydride superconductors become stable. Doping known hydrogen-rich binary systems with extra elements or molecules is one way to achieve this. For example, the ionic bonds formed by doping a H₃S host with CH₄ molecules lead to stability of the compound at a much lower pressure of 100 GPa [13,14]. A careful choice of the elements used for pre-compression is also important. For example, low-pressure stability in lanthanide and actinide systems correlates strongly with the presence of *f* states at the Fermi level. As a result, metastable phases of YbH₆ and LuH₆ are predicted to exhibit high-*T*_c superconductivity at relatively low pressures (145 K at 70 GPa and 273 K at 100 GPa, respectively [15]). Turning to even lower pressures, *Fm*3*m* UH₈₊₈ [16], *F*43*m* EuH₉ [17] and *C2/c* NdH₇ [18] have been observed experimentally at 42 GPa, 86 GPa and 85 GPa, respectively, but with very low critical temperatures.

Whilst binary hydrides have been extensively explored [19-23], research into ternary hydrides is much more challenging. Because phase diagrams of ternary A-X-H systems that is much more complex than those of binary systems. Despite this difficulty, many ternary hydrides have been found to exhibit favorable properties when compared to current binary systems. For example, C doped H₃S

possesses a much higher T_c than that of H₃S in experiments [24], and Li₂MgH₁₆, a molecular Mg-H phase doped with Li, is predicted to have the highest T_c to date (473 K) [25]. The ability to methodically and efficiently explore ternary hydrides and identify those with desirable superconducting properties at low pressures is key to advancing research in superconductivity.

Here, we propose a strategy to design high- T_c ternary hydride superconductors at low pressures by engineering binary X-H backbones, which are subsequently "pre-compressed" by a large-radius element A. The resulting alloyed backbones are easier to metallize compared to pure H backbones, such as those found in the clathrate hydrides. These backbones can be designed by doping familiar structures with additional atoms (X), which break the local motif of the parent structure. This leads to a metallic H-rich phase with occupied overlapping bands, known as a *hydrogen alloy* phase [3]. The IIA and IIIB metals, with large radii, are known to be effective "pre-compressors" [7,8,26], so we investigate their combination with these designer backbones. We use a hard-sphere model to investigate the stability of these new materials in terms of the radius and bond lengths, leading to the prediction of a new class of high- T_c hydride superconductors with a 'fluorite-type' backbone.

We design the first of these alloy backbone materials from the pure H backbone of the hightemperature superconductor LaH₁₀ [10], which possesses the same symmetry as the low-pressure UH₈ superconductor [16]. Comparing the crystal structures of UH₈ and LaH₁₀ (as shown in Fig. 1(a)-(b)), the structure of LaH₁₀ can be regarded as a UH₈ parent structure doped with additional H atoms at vacant tetrahedral sites (as shown in Fig. 1(d)). The extra H atoms break the localization of cubic H₈ units and lead to the famous clathrate backbone in LaH₁₀. The extra dopant X atoms with smallradius can instead be located in vacancy sites at the centre of the cubic H₈ units, and the result is an *Fm*3*m* structure of LaXH₈. For example, atom Be may be suitable dopants at these cubic sites. This novel H-Be alloy backbone corresponds to a fluorite-type arrangement, in which Be atoms are located on the sites of a face-centered cubic lattice, and [H₄] tetrahedra are present in the tetrahedral vacancies found between the Be atoms (see Figs. 1(e)-(f)). This results in a novel class of ternary hydrides AXH₈, designed with X-H alloy backbones to potentially achieve high-temperature superconductivity at low pressure. We note that during the preparation of our manuscript, this same cubic structure was reported in the La-B-H ternary system [27,28].



FIG. 1 The parent structure and two types of doped framework. (a) The crystal structure of UH₈, with [H₈] cubic units shown. The U ions are shown in black and the H ions in pink. (b) The crystal structure of LaH₁₀, in which La ions are shown in green. The backbone in LaH₁₀ consists of cubic-unit H atoms (pink) and tetrahedron-center H atoms (gray). (c) The crystal structure of fluorite CaF₂, the Ca cations are shown in dark blue and the F ions in light blue. (d) The crystal structure of UH₈, with [H₄] tetrahedral units shown. (e) The crystal structure of LaBeH₈, in which La ions are shown in green. The backbone in LaBeH₈ consists of tetrahedral-unit H atoms (pink) and cubic-center Be atoms (blue). (f) The 'fluorite-type' cage of LaBeH₈, consisting of Be-H bonds and H-H bonds.

Having identified this fluorite-type ternary structure, we go on to investigate a wider class of fluorite-type backbone hydrides AXH_8 ($Fm\overline{3}m$) formed from "pre-compressor" elements A (A = Sc, Ca, Y, Sr, La, Ba) and small-radius elements X (X = Be, B, Al). The phonon dispersions of these AXH_8 structures were calculated in the pressure range 50-200 GPa, among which 7 compounds, CaBH₈, CaBeH₈, YBeH₈, SrBH₈, LaAlH₈, LaBH₈ and LaBeH₈, were found to be dynamically stable (as shown in Fig 2 and Fig. S26-S32). LaBeH₈ is dynamically stable at 50 GPa, a moderate pressure accessible to Kawai-Type Multi-Anvil presses (KMAPs) [29]. Four other compounds were found to be dynamically stable at or below 100 GPa; LaBH₈ (70 GPa), LaAlH₈ (100 GPa), CaBH₈ (100 GPa) and YBeH₈ (100 GPa). The other two compounds, SrBH₈ and CaBeH₈, become dynamically stable at 150 GPa and 210 GPa, respectively. We go on to determine the thermodynamic stability of these dynamically stable "fluorite-like" backbone hydrides using *ab initio* random structure searching

(AIRSS) [30], by constructing convex hulls (see Figs. S5-S11). Four compounds, LaBeH₈, CaBH₈, YBeH₈ and SrBH₈, sit on the convex hull. LaBeH₈ is thermodynamically stable at 98 GPa. CaBH₈, YBeH₈ and SrBH₈ become thermodynamically stable at, or above, 300 GPa as shown in the SM [31].



FIG. 2 Calculated enthalpy of alloyed ternary hydrides AXH₈ above the convex hull. (a) The radius of atom A is plotted on the x-axis and the radius of atom X on the y-axis. Dynamically unstable systems are shown as black crosses. Metastable phases are shown as circles, colored according to the calculated enthalpy above the convex hull. Thermodynamically stable phases are shown as carmine squares. (b) Calculated enthalpy as a function of pressure for La-Be-H structures relative to the $Fm\bar{3}m$ phase of LaBeH₈, where structures of LaH₆, BeH₂ and H are from Refs. [7-9,48,53], respectively.

Having investigated the stability of these fluorite-type backbone hydrides, we go on to investigate their superconducting properties. Based on Eliashberg equations, the values of T_c were determined using $\mu^* = 0.1$. As shown in Fig. 3, LaBeH₈ demonstrates high-temperature superconductivity with a T_c of 191 K at a remarkably low pressure of 50 GPa. This is possible because the local structure of the H₈ molecular units is broken by the Be dopants. These fluorite-type backbone structures can even exhibit room-temperature superconductivity, with the metastable hydride CaBeH₈ predicted to possess a T_c of 300 K at 210 GPa. Likewise, YBeH₈, CaBH₈, LaBH₈, LaAlH₈ and SrBH₈ also exhibit high-temperature superconductivity with 249 K at 100 GPa, 238 K at 100 GPa, 160 K at 70 GPa, 144 K at 100 GPa and 200 K at 150 GPa, respectively. As can be seen in Fig. 3, the threshold pressure at which fluorite-type backbone hydrides become dynamically stable is lower than that for typical high- T_c hydrides, whilst retaining a T_c that is much higher than the temperature of liquid nitrogen. LaBeH₈ is the first proposed H-based superconductor with a figure of merit [54] score around S = 3.



FIG. 3 Pressure dependence of T_c s calculated for alloyed clathrate hydrides shown alongside other high-Tc superconductors. The orange circles are T_c s of fluorite-type backbone hydrides at the lowest pressure where they become dynamically stable. The red circle is at the lowest pressure where LaBeH₈ becomes thermodynamically stable (98 GPa), and the suggested synthesis pressure range for cubic LaBeH₈ is highlighted in yellow. The blue squares are T_c s of clathrate binary hydrides at the lowest pressures reported in Refs. [4,7,8,26,54]. The purple stars are T_c s of well-known superconductors from experiment [5,10,24,54]. The background is shaded according to the figure of merit $S = \frac{T}{\sqrt{p^2 + T_{MgB_2}^2}}$

used to evaluate the significance of a particular superconductor [54].

The dynamic stability of fluorite-type hydrides depends on the radii of the pre-compressor element A as shown in Fig. 2(a), suggesting that a hard spheres model [55] derived from the geometry may allow us to draw general conclusions about this novel structure family. To construct this model, we make two simplifications: i) the large-radius atoms A are regarded as hard spheres; ii)

the backbone is characterized by H-H bonds and X-H bonds. The derivation of this model is presented in the SM [31] and the solution gives the lattice parameter of the cubic unit cell (*L*), the lengths of H-H bonds ($b_{\text{H-H}}$) and X-H bonds ($b_{\text{X-H}}$) as

follows: $\begin{cases} L = \frac{\sqrt{3}+1}{t+1} \cdot 2R_A \\ b_{H-H} = \frac{\sqrt{3}+1-2t}{t+1} \cdot \sqrt{6}R_A = A_{H-H}d_{H-H}, \\ b_{X-H} = \frac{3t-\sqrt{3}}{t+1} \cdot R_A = A_{H-X}d_{H-X} \end{cases}$

where R_A is the covalent radius [56] of atom A and t = 0.95-1.05 is a tolerance factor allowing slight overlap of the hard spheres. The bond lengths can also be represented as products of flexible factors (*A*) and bond lengths of binary hydrides (*d*) from the literatures.

Table. 1 The lattice parameter *L*, bond lengths b_{X-H} and b_{H-H} and flexible factors *A* calculated from the hard-spheres model (unprimed) and from DFT (primed) at 150 GPa. The amount of charge transferred to H is denoted as δ . Here we use *t*=1.03, *d*_{H-H} =1.1 Å [7,12,25], *d*_{Be-H} = 1.31 Å [48], *d*_{B-H} = 1.22 Å [50] and *d*_{Al-H} = 1.72 Å [51,57].

	L, Å	b _{X-н} , Å (А _{X-н})	<i>b</i> _{H-H} , Å (<i>А</i> _{H-H})	<i>L</i> ', Å	b' _{X-H} , Å (<i>A</i> ' _{X-H})	<i>b</i> ' _{н-н} , Å (<i>A</i> ' _{н-н})	δ , e-/atom
LaBeH ₈	5.03	1.25 (91%)	1.52 (137%)	5.17	1.36 (99%)	1.43 (130%)	0.36
LaBH ₈	5.03	1.25 (102%)	1.52 (137%)	5.13	1.33 (109%)	1.45 (131%)	0.32
LaAlH ₈	5.03	1.25 (73%)	1.52 (137%)	5.38	1.52 (88%)	1.32 (120%)	0.46
SrBH ₈	5.79	1.44 (118%)	1.74 (158%)	5.05	1.32 (108%)	1.41 (128%)	0.29
CaBH ₈	5.33	1.32 (109%)	1.60 (146%)	4.89	1.3 (107%)	1.33 (121%)	0.29
CaBeH ₈	5.33	1.32 (97%)	1.60 (146%)	4.93	1.32 (96%)	1.32 (120%)	0.32
YBeH ₈	4.79	1.19 (86%)	1.44 (131%)	5.02	1.34 (98%)	1.36 (124%)	0.40

The solutions for the 7 hydrides considered in this work are shown in Table 1, alongside with the values calculated from DFT at 150 GPa. The value of L obtained in the hard spheres model is similar to that calculated by DFT in the pressure range of 100-200 GPa. The geometry of the fluorite-type backbone has elongated H-H bond lengths by 30-60% or 20-30% (according to the hard-spheres model and DFT, respectively) compared to the H-H bond lengths in common hydrides [7,12,25], because of the large amount of charge transferred to the H-H bond (Table 1). The X-H bonds are also

affected by the geometry, but whether the bonds are elongated or shortened depends on the composition; the hard-spheres model and DFT calculations agree on the trend.

The hard-spheres model captures the general characteristics of AXH₈ and allows us to determine the criteria for elements that can be substituted for X in the crystal. In particular, candidate elements should form bonds to hydrogen with lengths in the range of 1.2-1.6 Å in binary systems. Therefore, we investigate the possibility of substituting Si, P and S into LaXH₈ since the X-H bond lengths are $d_{Si-H} \sim 1.6$ Å [58], $d_{P-H} \sim 1.4$ Å [59] and $d_{S-H} \sim 1.5$ Å [60]. We find that LaSiH₈, LaSH₈ and LaPH₈ are all dynamically stable high- T_c superconductors with 150 K at 100 GPa, 150 K at 200 GPa and 180 K at 200 GPa, respectively.



FIG. 4 The structures of $[H_3S]$ and $[BeH_8]$ frameworks with polarization vectors shown. (a) The polarization vectors of H-S bonds are aligned, in these bonds the positive charge is located at H atoms (pink) and negative charge is located at S atoms (yellow). (b) The polarization vectors of Be-H bonds converge, leading to a concentration of charge at the hydrogen atoms. In these bonds the positive charge is located at Be atoms (green) and the negative charge is located at H atoms (pink).

Unlike pure hydrogen frameworks, alloyed backbones in hydrides typically consist of polar bonds at high pressure. For example, the [SH₃] backbone of SCH₇ consists of H-S polar bonds as shown in Fig. 4(a). Similarly to hydrogen bonded structures at ambient pressure, end-to-end arrangements of polar bonds in these alloy backbones generally have low enthalpy. In contrast to the end-to-end arrangements found in common hydrides, in the [BeH₈] backbone, four H atoms with negative charge δ^- are found in close proximity and form a regular tetrahedron as shown in Fig. 4(b). This distribution of charge suggests that the fluorite-type backbone is formed in a different way to the common covalent hydrides. In the unique bonding environment of the fluorite-type backbone, a large amount of charge transfer to the H atoms is possible as shown in Table 1; these electrons occupy anti-bonding states along the H-H bonds (Fig. S19-S25). The electrons in the anti-bonding orbitals not only support the elongated H-H bonds, but also give rise to an increased H-derived density of states at the Fermi level. The unique chemistry in alloyed frameworks therefore provides a route to improved H-based superconductors.

As part of efforts to develop new H-based superconductors that are stable at low pressure, we design a new class of ternary hydrides AXH₈ with fluorite-type alloy backbones. The design of these fluorite-type hydrides is envisioned using a hard-spheres model, which allows the creation of new fluorite-type hydrides by geometry. In this work, we investigate a total of 10 different fluorite-type hydrides to demonstrate this design, which turns out to be highly successful. All predicted hydrides show both excellent superconductivity and low-pressure dynamic stability compared with other reported H-based superconductors. Although the lowest pressure at which any of these hydrides becomes dynamically stable (~ 50 GPa) is still high when compared to ambient pressure, it is decreased by a factor of 4 from the pressure needed to stabilize typical hydride superconductors. One of the systems we considered, LaBeH₈, is dynamically stable down to pressures obtainable in KMAPs. Synthesis of a high-temperature hydride superconductor in a KMAP would represent a breakthrough in the field. The hydrides in the fluorite-type backbone family would number about 20 if other elements in the lanthanide and actinide series were included. Future investigations of other ternary systems may identify high-temperature superconductors even closer to ambient pressure by similar methods.

In summary, designing hydrides with an alloyed backbone is shown to be an effective approach to obtaining low-pressure H-based superconductors. In these materials, small radius elements are alloyed with hydrogen in order to introduce new bonds into the backbone instead of pure H-H bonds. These new bonds are more stable than H-H bonds, and the resulting fluorite-type backbones can be stabilized at much lower pressures. Our results provide an effective method for the rational design of lower-pressure H-based superconductors and stimulate further experimental exploration of hydride superconductors at close-to-ambient pressures. The synthesis of hydridebased superconductors at closer to ambient pressures in experiment would represent an important breakthrough in the field of high-temperature superconductivity.

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Supplemental Materials

for

Design Principles for High Temperature Superconductors with Hydrogen-based Alloy Backbone at Moderate Pressure

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1. The hard-sphere model of "fluorite-like" backbone hydride

We built a hard-sphere model to describe "fluorite-like" backbone hydrides. The structures of our predicted fluorite-type hydrides are similar to perovskites and alloys, so we consult their method. In the view of perovskites, ionic radius is the main factor of the stability. For alloys, three main factors are atomic radii, electronegativity and valence of elements. In case of fluorite-type hydrides AXH₈, atomic radii of A, the length of X-H and H-H bonds, electronegativity are main factor in our model.

Here, the model of fluorite-type hydrides AXH₈ were based on two assumptions: (i) the geometric models of AXH₈ were carried out with atomic radius of atom A and bond lengths of X-H and H-H; (ii) the electronegativity of X elements were described by transferred charge from X atoms at pressure.

According to (i) and the structure of AXH₈, we can carry out the three equations:



FIG. S1. (100) surface of AXH₈

As shown in Fig. S1, length of a side *L* consists of two radii of hard spheres R_A and a side of cubic XH₈ $\frac{2\sqrt{3}}{3}b_{H-X}$ (where b_{H-X} is a half body diagonal of cubic XH₈). The equation is shown as following:

$$t \times L = 2R_A + \frac{2\sqrt{3}}{3}b_{H-X},\tag{S1}$$

where *L* is the length of side of cubic unit cell, R_A is the covalent radius of atom A from ref. [2], b_{X-H} and b_{H-H} are lengths of X-H bonds and H-H bonds. Here *t* is tolerance factor with value around 0.95~1.05. Because the cubic XH₈ is not hard, it could overlap a few with hard spheres.



FIG. S2. (110) surface of AXH₈

As shown in Fig. S2, diagonal of the face (100) $\sqrt{2}L$ consists of two diagonals of cubic XH₈ $2 \times \frac{2\sqrt{6}}{3} b_{H-X}$ and two H-H bonds $2b_{H-H}$. The equation is shown as following:

$$\frac{\sqrt{2}}{2}L = \frac{2\sqrt{6}}{3}b_{H-X} + b_{H-H}.$$
 (S2)



FIG. S3. ($\overline{2}11$) surface of AXH₈, here a is [111] and c is [01 $\overline{1}$].

As shown Fig. S3, body diagonal of unit cell $\sqrt{3}L$ consists of two bonds X-H $2b_{H-X}$, two heights of H tetrahedron $\frac{2\sqrt{6}}{3}b_{H-H}$ and two radii of hard spheres $2R_A$. The equation is shown as following:

$$\frac{\sqrt{3}}{2}L = R_A + \frac{\sqrt{6}}{3}b_{H-H} + b_{H-X},$$
 (S3)

Solutions of these equations are *L*, b_{X-H} , and b_{H-H} , they are side length of unit cell, lengths of H-X and H-H bonds, which are only depend on the radii of elements R_A in our model as shown in table S1 (these lengths are all Å).

Table S1| The side length of unit cell L(L'), bond lengths of H-X $b_{X-H}(b'_{X-H})$ and H-H $b_{X-H}(b'_{X-H})$ suggested by hard sphere model (calculated by DFT at 150 GPa). R_A is the covalent radius of atom A from ref.[2]

	R _A , Å	L, Å	<i>b</i> _{Х-н} , Å	$b_{ ext{H-H}}, ext{\AA}$	<i>L</i> ', Å	<i>b</i> ' _{Х-Н} , Å	<i>b</i> ' _{н-н} , Å
LaBeH ₈	1.87	5.03	1.25	1.52	5.17	1.36	1.43
LaBH ₈	1.87	5.03	1.25	1.52	5.13	1.33	1.45
LaAlH ₈	1.87	5.03	1.25	1.52	5.38	1.52	1.32
SrBH ₈	2.15	5.79	1.44	1.74	5.05	1.32	1.41
CaBH ₈	1.98	5.33	1.32	1.60	4.89	1.3	1.33
CaBeH ₈	1.98	5.33	1.32	1.60	4.93	1.32	1.32
YBeH ₈	1.78	4.79	1.19	1.44	5.02	1.34	1.36

L is the suggested length of unit cell by hard-sphere model without pressure, it is function of one variable of R_4 . From the table, we can see that the length of unit cell, bond lengths of H-X and H-H are similar to that at pressure of 100~200 GPa by DFT calculation. It is suggested that the AXH₈ is a pressure-stabilized hydride.

H-H bonds of H tetrahedron in AXH₈ were found to be longer than those in other hydrides at pressure such as those in clathrate hydrides (~ 1.1 Å), because of more transferred charge on hydrogen atoms as shown in Table S2. In order to compare bond length with other binary hydrides, the flexible factors A are used to describe the changes of lengths of H-H and X-H bonds. The bond

lengths (b) in AXH₈ represented by products of flexible factors A and bond lengths (d) in known binary hydrides and atomic phase of pure hydrogen, such that $b_{X-H} = A_{X-H}$. d_{X-H} and $b_{H-H} = A_{H-H}$.

Table S2| The length of H-H bonds b_{H-H} and its flexible factors A_{H-H} suggested by model and calculated by DFT. The positive charge means that charge transfers to H. Here $b_{H-H} = A_{H-H} \cdot d_{H-H}$, d_{H-H} is the nearest length of H-H bonds in common length of hydrides, here we use $d_{H-H} = 1.1$ Å.

	<i>b</i> н-н, Å	Ан-н	<i>b</i> ' _{Н-Н} , Å,	<i>А</i> 'н-н,	transferred charge to H, electrons
LaBeH ₈	1.52	137%	1.43	130%	0.36
LaBH ₈	1.52	137%	1.45	131%	0.32
$LaAlH_8$	1.52	137%	1.32	120%	0.46
$SrBH_8$	1.74	158%	1.41	128%	0.29
CaBH ₈	1.60	146%	1.33	121%	0.29
CaBeH ₈	1.60	146%	1.32	120%	0.32
YBeH ₈	1.44	131%	1.36	124%	0.40

The lengths of H-H bonds calculated by model and by DFT are about 30~60% and 20~30% longer than the common length of hydrides. Our result shows that it is the geometry that forces the H-H bonds to take longer lengths than other, and more charge makes the long bonds stable.

To find the replaceability of element X in AXH₈, it is suggested to study the electronegativity of X. Here, we studied the transferred charge from X atoms instead of electronegativity of element X.

Here we used common minimal X-H bond lengths d_{X-H} of BeH₂[3], BH[4] and AH₃[5] at about 150 GPa to calculate A_{X-H} with $b_{X-H} = A_{X-H}$. d_{X-H} as shown in Table S3.

Table S 3| The length of X-H bonds b_{X-H} and its flexible factors A_{X-H} suggested by model and calculated by DFT. The negative charge means that charge transfers from X. Here $b_{X-H} = A_{X-H} \cdot d_{X-H}$, d_{X-H} is length of common X-H bonds in known binary hydrides.

	<i>b</i> _{X-н} , Å	Ах-н	<i>b</i> ' _{Х-Н} , Å,	А'х-н	<i>d</i> _{X-н} , Å	transferred charge from X, electrons
LaBeH ₈	1.25	91%	1.36	99%	~1.37	-1.57
LaBH ₈	1.25	102%	1.33	109%	~1.22	-1.2
LaAlH ₈	1.25	75%	1.52	92%	~1.65	-2.28
SrBH ₈	1.44	118%	1.32	108%	~1.22	-1.28
CaBH ₈	1.32	109%	1.3	107%	~1.22	-1.29
CaBeH ₈	1.32	97%	1.32	96%	~1.37	-1.51
YBeH ₈	1.19	86%	1.34	98%	~1.37	-1.56

Compared with A_{X-H} and A'_{X-H} , they change consistently relative to the common minimal lengths of X-H bond d_{X-H} , because geometric factor controls the length of X-H bonds. And our results give an idea for the replaceability of element X in AXH₈: elements, of which length of bonds is about 1.2~1.6 Å and electrons transfer to H, is possibly replaceable. Following this idea, we had found LaSiH₈ and LaPH₈, and LaSH₈ which are also potential high-Tc superconductors. According to (ii), although electronegativity of B is similar to H, but at pressure transferred charge from B to H is large, therefore, pressure could increase the electronegativity of H in this structure.

In summary, the structure of fluorite-type hydrides AXH_8 was described by the hard-sphere model, from which structural parameters were similar to those calculated by DFT at pressure about 100~200 GPa. Therefore, the novel structure of fluorite-type hydrides AXH_8 was proven to be stabilized by pressure. And the lengths of bonds in the XH₈ backbone were forced by the geometry

of AXH_8 . The length of X-H bond is about $1.2\sim1.6$ Å. Because pressure increase the electronegativity in this structure, it is possible for H to get charge from elements with large electronegativity. Therefore, we had tried Si, P and S as candidates, which possess suitable bond lengths and electronegativity.

2. Computational details

High-pressure structural searches were performed from first principles using the AIRSS (ab initio Random Structure Searching) code[1], whose effectiveness has been confirmed by the successful applications to discovering the structures of solids, point defects, surfaces, and clusters. In pressure range of 50~200 GPa, we predict more than 15,000 structures in the La-Be-H system using variable composition structural searches, and the number of predicted structures at the LaBeH₈ stoichiometry is more than 1,000. And for systems Ca-Be-H, Ca-B-H, La-Al-H, La-B-H, Sr-B-H and Y-Be-H, each system we predicted around 5000 structures for their ternary convex hulls, and for each fluorite-type hydride we predicted around 1000 structures. Structural relaxations during the searches are performed using the CASTEP (Cambridge Sequential Total Energy Package) code with ultrasoft pseudopotentials[2]. Subsequent high-accuracy structural relaxations are performed using the projector augmented wave method (PAW) in the VASP code[3]. The cutoff energy was chosen to be 600 eV. The exchange-correlation functional was described using the Perdew-Burke-Ernzerhof (PBE) parametrization within the generalized gradient approximation (GGA)[4,5]. A Monkhorst-Pack k-point mesh of $2\pi \times 0.03$ Å⁻¹ was used to ensure that the enthalpy calculations were converged to within 1 meV/atom. We also perform Bader charge analysis to determine the extent of charge transfer[6,7]. Bonding in these hydrides was investigated by the crystal orbital Hamiltonian population (COHP)[8] analysis using LOBSTER code[9], which provides an atom-specific measure of the bonding character of states in a given energy region. We calculate electron-phonon matrix elements using density functional perturbation theory as implemented in the Quantum ESPRESSO package[10]. We use ultrasoft pseudopotentials[11] with an 80 Ry cutoff, an 18×18×18 k-point grid and a $6 \times 6 \times 6$ *q*-point grid.

3. Equations for calculating Tc and related parameters

(1) Self-consistent iteration solution of the Eliashberg equation

The Migdal-Eliashberg equation has a form[12-14]:

$$\Delta(i\omega_n)Z(i\omega_n) = \frac{\pi T}{N_F} \sum_{n'} \frac{\Delta(i\omega_n')}{\sqrt{\omega_n'^2 + \Delta^2(i\omega_n')}} \times [\lambda(\omega_n - \omega_{n'}) - N_F \mu^*]\delta(\epsilon)$$
(S4)

$$Z(i\omega_n) = 1 + \frac{\pi T}{N_F \omega_n} \sum_{n'} \frac{\omega_n'}{\sqrt{\omega_n'^2 + \Delta^2(i\omega_n')}} \lambda(\omega_n - \omega_{n'}) \delta(\epsilon)$$
(S5)

Here functions $\Delta(i\omega_n)$ and $Z(i\omega_n)$ represent pairing order parameter and the renormalization function, N_F is the density of electronic states at the Fermi level, and $\delta(\epsilon)$ is the Dirac delta function. $i\omega_n = i(2n+1)\pi T_c$ are the fermion Matsubara frequencies (we employ the themodynamic Green's functions formalism; μ^* is the Coulomb pseudopotential, for which we use the widely accepted range of 0.1-0.13. $\lambda(\omega_n - \omega_n)$ contains the electron-phonon coupling matrix, phonon propagator, and the phonon density of states, and is given by:

$$\lambda(\omega_n - \omega_n) = \int_0^\infty d\omega \frac{2\omega}{(\omega_n - \omega_n)^2 + \omega^2} \alpha^2 F(\omega)$$
(S6)

The equations for the order parameter and the renormalization function form a coupled nonlinear system and are solved self-consistently. We evaluated the renormalization function and the order parameter for each Matsubara frequency along the imaginary energy axis. After calculating $Z(i\omega_n)$ and $\Delta(i\omega_n)$, an analytic continuation is performed to the real axis using the Pade' functions. The calculation is performed for each T ($T_{\min} < T \le T_{\max}$) ($T_{\min} \approx 0$ and $T_{\max} \ge T_c$). The critical temperature Tc is obtained as an asymptotic value as $\Delta(i\omega_n)$ tends to zero.

(2) Gor'kov-Kresin's theory

Gor'kov and Kresin (G-K) introduced the coupling constants λ_{opt} and λ_{ac} describing the interaction of electrons with optical and acoustic phonons[15,16]. The generalized Eliashberg equation has the form (at $T=T_c$):

$$\Delta(\omega_n)Z = \pi T \sum_{\omega_n'} \left[\lambda_{\text{opt}} \frac{\tilde{\Omega}_{\text{opt}}^2}{\tilde{\Omega}_{\text{opt}}^2 + (\omega_n - \omega_{n'})^2} + \lambda_{\text{ac}} \frac{\tilde{\Omega}_{\text{ac}}^2}{\tilde{\Omega}_{\text{ac}}^2 + (\omega_n - \omega_{n'})^2} \right] \frac{\Delta(\omega_{n'})}{|\omega_{n'}|}_{|T=T_c}, \quad (S7)$$
$$\lambda_{\text{ac}} = 2 \int_0^{\omega_1} \frac{\alpha^2 F(\omega)}{\omega} d\omega, \ \lambda_{\text{opt}} = 2 \int_{\omega_1}^{\omega_m} \frac{\alpha^2 F(\omega)}{\omega} d\omega, \ \lambda_{\text{ac}} + \lambda_{\text{opt}} = \lambda, \quad (S8)$$

where ω_1 is the maximum frequency for the acoustic modes, ω_m is the maximum frequency value. The mean square average frequency values are defined as follows:

$$\widetilde{\omega}_{\rm ac} = \langle \omega_{\rm ac}^2 \rangle^{\frac{1}{2}}, \ \langle \omega_{\rm ac}^2 \rangle = \frac{2}{\lambda_{\rm ac}} \int_0^{\omega_1} d\omega \cdot \omega^2 \frac{\alpha^2 F(\omega)}{\omega} = \frac{2}{\lambda_{\rm ac}} \int_0^{\omega_1} \alpha^2 F(\omega) \omega d\omega, \quad (S9)$$
$$\widetilde{\omega}_{\rm opt} = \langle \omega_{\rm opt}^2 \rangle^{\frac{1}{2}}, \ \langle \omega_{\rm opt}^2 \rangle = \frac{2}{\lambda_{\rm opt}} \int_{\omega_1}^{\omega_{\rm m}} d\omega \cdot \omega^2 \frac{\alpha^2 F(\omega)}{\omega} = \frac{2}{\lambda_{\rm opt}} \int_{\omega_1}^{\omega_{\rm m}} \alpha^2 F(\omega) \omega d\omega, \quad (S10)$$

For our predicted hydrides the $\lambda_{ac} \ll \lambda_{opt}$, we assume that:

$$T_c = T_c^{opt} + \Delta T_c^{ac}$$
, and $T_c^{opt} \gg \Delta T_c^{ac}$ (S11)

As a result, the expression for Tc can be written in the form:

$$T_c = \left[1 + 2\frac{\lambda_{\rm ac}}{\lambda_{\rm opt} - \mu^*} \cdot \frac{1}{1 + \rho^{-2}}\right] T_c^0, \rho = \frac{\widetilde{\omega}_{\rm ac}}{\pi T_c^0}, T_c^0 \equiv T_c^{opt}.$$
 (S12)

Here the T_c^{0} is defined as the transition temperatures caused by the interaction of electrons with optical phonons only; for $\lambda_{opt} \le 1.5$:

$$T_{c}^{0} = \frac{\widetilde{\omega}_{\text{opt}}}{1.2} \exp\left[-\frac{1.04(1+\lambda_{\text{opt}})}{\lambda_{\text{opt}}-\mu^{*}(1+0.62\lambda_{\text{opt}})}\right].$$
 (S13)

For $\lambda_{opt} > 1.5$:

$$T_{c}^{0} = \frac{0.25\tilde{\omega}_{\text{opt}}}{\left[e^{\frac{2}{\lambda_{eff}}} - 1\right]^{1/2}}.$$
(S14)

Here the λ_{eff} is defined as follows:

$$\lambda_{\rm eff} = (\lambda_{\rm opt} - \mu^*) [1 + 2\mu^* + \lambda_{\rm opt} \mu^* t(\lambda_{\rm opt})]^{-1}, \qquad (S15)$$

$$t(x) = 1.5 \exp(-0.28) x.$$
(S16)

We used the following expression:

$$\alpha = \frac{1}{2} \left[1 - 4 \frac{\lambda_{ac}}{\lambda_{opt}} \frac{\rho^2}{(\rho^2 + 1)^2} \right]$$
(S17)

to calculate the isotope coefficient α .

(3) Allen-Dynes modified McMillan eqation

The Allen-Dynes modified McMillan equation which is the approximate analytic solution of T_c from Eliashberg equations at $\omega \approx 0$ or ∞ and $\lambda \ll 1.5[17]$:

$$T_c = \frac{\omega_{log}}{1.2} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right],$$
 (S18)

when $\lambda > 1.5$, we have the correction equation as follows:

$$T_{c} = \frac{f_{1}f_{2}\omega_{log}}{1.2} \exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^{*}(1+0.62\lambda)}\right],$$
 (S19)

two separate correction factors f_1 and f_2 are given by:

$$f_{1} = \sqrt[3]{\left[1 + \left(\frac{\lambda}{2.46(1+3.8\mu^{*})}\right)^{\frac{3}{2}}\right]}, f_{2} = 1 + \frac{\left(\frac{\overline{\omega}_{2}}{\omega_{log}} - 1\right)\lambda^{2}}{\lambda^{2} + \left[1.82(1+6.3\mu^{*})\frac{\overline{\omega}_{2}}{\omega_{log}}\right]^{2}},$$

$$\overline{\omega}_{2} = \sqrt{\frac{2}{\lambda}\int \alpha^{2}F(\omega)\,\omega d\omega},$$
(S20)

here $\overline{\omega}_2$ is mean square frequency, ω_{log} is the logarithmic average frequency and μ^* is the Coulomb pseudopotential, for which we use the widely accepted range of 0.1-0.13. The ω_{log} and EPC constant λ were calculated as:

$$\omega_{log} = \exp\left[\frac{2}{\lambda}\int\frac{d\omega}{\omega}\alpha^2 F(\omega)\ln\omega\right], \qquad (S22)$$

$$\lambda = 2 \int \frac{\alpha^{2} F(\omega)}{\omega} d\omega = \sum_{qj} \lambda_{qj} w(q), \qquad (S23)$$

$$\lambda_{qj} = \frac{\gamma_{qj}}{\pi \hbar N(\varepsilon_f) \omega_{qj}^2},\tag{S24}$$

where λ_{qj} is the mode EPC parameter and w(q) is the weight of phonon mode j at wave vector q in the first Brillouin zone (BZ).

Table S4. The calculated EPC parameter λ , logarithmic average phonon frequency ω_{log} (K), electronic density of states at Fermi level N(ϵ_f) (states/spin/Ry/f.u.) and superconducting transition

temperatures T_c (K) of flouride-type hydrides with $\mu^*=0.1$ at corresponding pressures (GPa). And the values of T_c are calculated from Elashberg equation with use of the approach with many iteration (IA), and with use of the Gorkov-Kresin theory (G-K). The table contains also the values obtained from the Allen- Dynes modified McMillan equation (Mc-A-D, See Eq. S19).

Hydrides	Pressure	λ	Wlog	Ν (ε _f)	T _c (Mc-A-D)	T _c (IA)	λ_{ac}	λ_{opt}	T _c (G-K)
LaBeH ₈	50	2.19	897.63	6.94	167	191	0.25	1.94	165
$LaBH_8$	70	1.98	843.63	4.63	144	160	0.30	1.67	155
LaAlH ₈	100	1.81	839.68	5.21	130	144	0.17	1.58	140
CaBeH ₈	210	3.91	798.06	5.02	254	302	1.12	2.77	291
$CaBH_8$	100	3.44	767.67	4.50	212	238	0.84	2.49	237
YBeH ₈	100	3.05	855.70	5.67	215	249	0.45	2.60	226
SrBH_8	150	1.79	1039.12	4.62	163	200	0.32	1.47	177
LaSH ₈	200	2.37	827.19	8.42	169	195	0.44	1.92	190
LaSiH ₈	100	2.41	659.62	5.17	137	150	0.49	1.73	159
LaPH ₈	200	1.41	1090.43	7.05	137	151	0.12	1.35	142



FIG. S4. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides CaBeH₈.



FIG. S5. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides CaBH₈.



FIG. S6. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides LaAlH₈.



FIG. S7. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides LaBeH₈.



FIG. S8. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides LaBH₈.



FIG. S9. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides SrBH₈.



FIG. S10. Calculated Eliashberg spectral function $\alpha^2 F(\omega)$ (black line), total electron-phonon integral λ_{tot} (red line), electron and acoustic phonon integral λ_{ac} (blue line) and electron and optical phonon integral λ_{opt} (green line) of alloyed ternary hydrides YBeH₈.

Space group	Lattice	Atomic coordinates	Sites
Pressure	parameters	(fractional)	
	(Å)		
	a = b = c = 5.643	Н 0.351 -0.351 -0.351	32f
Fm3m-LaBeH ₈		Be 0.500 0.500 0.500	4b
50 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.456	Н -0.647 0.352 -0.647	32f
Fm3m-LaBH ₈		B 0.500 0.500 0.500	4b
70 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.582	Н 0.338 -0.338 0.162	32f
Fm3m-LaAlH ₈		B 0.500 0.500 0.500	4b
100 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.540	Н -0.340 0.660 0.660	32f
Fm3m-LaSiH ₈		Si 0.500 0.500 0.500	4b
100 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.173	Н -0.334 0.666 0.666	32f
Fm3m-LaSH ₈		S 0.500 0.500 0.500	4b
200 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.186	Н -0.337 0.663 0.663	32f
Fm3m-LaPH ₈		P 0.500 0.500 0.500	4b
200 GPa		La 0.000 0.000 0.000	4a
	a = b = c = 5.074	Н -0.653 0.347 0.347	32f
Fm3m-CaBH ₈		B 0.500 0.500 0.500	4b
200 GPa		Ca 0.000 0.000 0.000	4a
Fm3m-CaBeH ₈	a = b = c = 4.752	Н 0.344 -0.344 0.344	32f

4. Structural information

210 GPa		Be 0.500 0.500 0.500	4b
		Ca 0.000 0.000 0.000	4a
	a = b = c = 5.051	Н -0.349 0.349 -0.151	32f
<i>Fm3m</i> -SrBH ₈		B 0.500 0.500 0.500	4b
150 GPa		Sr 0.000 0.000 0.000	4a
Emām VRaHa	a = b = c = 5.199	Н -0.846 0.654 -0.654	32f
100 GPa		Be 0.500 0.500 0.500	4b
		Y 0.000 0.000 0.000	4a
	a = b = c = 5.027	H1 0.606 -0.606 -0.394	16e
F43m-LaBeHa		H2 0.000 0.000 0.000	4a
50 GPa		Be 0.250 0.750 0.750	4c
50 61 4		La 0.750 0.250 0.250	4b
	a = 3.394	H1 0.157 0.157 0.487	1a
	b = 5.130	H2 0.071 0.874 0.131	la
	c = 5.219	H3 0.843 0.844 0.513	1a
	$\alpha = 73.0$	H4 0.929 0.126 0.869	1a
	$\beta = 71.2$	H5 0.341 0.934 0.338	1a
	$\gamma = 74.2$	H6 0.659 0.066 0.662	1a
$P\overline{1}$ -LaBeH ₈		H7 0.329 0.319 0.897	1a
100 GPa		H8 0.671 0.680 0.103	1a
		H9 0.496 0.214 0.692	1a
		H10 0.504 0.786 0.308	1a
		H11 0.811 0.515 0.845	la
		H12 0.188 0.485 0.155	la
		H13 0.711 0.490 0.510	la
		H14 0.289 0.509 0.490	1a

I	H15	0.414	0.077	0.008	1a
I	H16	0.586	0.923	0.992	1a
I	Be1	0.001	0.656	0.336	1a
I	Be2	0.999	0.344	0.663	1a
I	La1	0.255	0.759	0.769	1a
Ι	La2	0.745	0.241	0.231	1a

5. The ternary convex hull of "fluorite-like" backbone hydride



FIG. S11. Calculated enthalpy of alloyed ternary hydrides AXH₈ above the convex hull at (a) 200 GPa and (b) 300 GPa. The radius of atom A is plotted on the x-axis and the radius of atom X on the y-axis. Dynamically unstable systems are shown as black cross marks. Metastable phases are shown as circles, colored according to the calculated enthalpy above the convex hull. Thermodynamically stable phases are shown as carmine squares.



FIG. S12. The ternary phase diagram of Ca, Be, and H at 200 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big purple circle with edge indicates component CaBeH₈, which is a metastable phase. The structures of binary hydrides were from ref. [18-20].



FIG. S13. The ternary phase diagram of Ca, B, and H at 350 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big red square with edge indicates component CaBH₈, which is a stable phase. The structures of binary hydrides were from ref. [19-21].



FIG. S14. The ternary phase diagram of La, Al, and H at 200 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big purple circle with edge indicates component LaAlH₈, which is a metastable phase. The structures of binary hydrides were from ref. [22,23].



FIG. S15. The ternary phase diagram of La, Be, and H at 150 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big purple circle with edge indicates component LaBeH₈, which is a stable phase. The structures of binary hydrides were from ref. [18,23].



FIG. S16. The ternary phase diagram of La, B, and H at 200 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big purple circle with edge indicates component LaBH₈, which is a dynamically stable phase. The structures of binary hydrides were from ref. [21,23].



FIG. S17. The ternary phase diagram of Sr, B, and H at 300 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big purple circle with edge indicates component SrBH₈, which is a stable phase. The structures of binary hydrides were from ref. [21,24].



FIG. S18. The ternary phase diagram of Y, Be, and H at 300 GPa. The corresponding elements and boundary binary phases are chosen from the results of the previous works. Light blue circles indicate metastable phases. Red squares indicate stable phases. And the big red square with edge indicates component YBeH₈, which is a stable phase. The structures of binary hydrides were from ref. [18,23].

6. The Crystalline Orbital Hamiltonian Population (COHP) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP) of "fluorite-like" backbone hydride



FIG. S19. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-Be bonds **b** H-H bonds of CaBeH₈ at 200 GPa.



FIG. S20. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-B bonds **b** H-H bonds of CaBH₈ at 100 GPa.



FIG. S21. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-Al bonds **b** H-H bonds of LaAlH₈ at 100 GPa.



FIG. S22. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-Be bonds **b** H-H bonds of LaBeH₈ at 100 GPa.



FIG. S23. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-B bonds **b** H-H bonds of LaBH₈ at 100 GPa.



FIG. S24. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-B bonds **b** H-H bonds of SrBH₈ at 200 GPa.



FIG. S25. The calculated Crystalline Orbital Hamiltonian Population (COHP, top panel) and Integrated Crystalline Orbital Hamiltonian Population (ICOHP, bottom panel) of **a** H-Be bonds **b** H-H bonds of YBeH₈ at 100 GPa.



7. Electronic structure of "fluorite-like" backbone hydride

FIG. S26. Electronic structure of The of CaBeH₈ at 200 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** Be and **d** Ca.



FIG. S27. Electronic structure of The of CaBH₈ at 100 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** B and **d** Ca.



FIG. S28. Electronic structure of The of LaAlH₈ at 100 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** Al and **d** La.



FIG. S29. Electronic structure of The of LaBeH₈ at 100 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** Be and **d** La.



FIG. S30. Electronic structure of The of LaBH₈ at 100 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** B and **d** Ca.



FIG. S31. Electronic structure of The of SrBH₈ at 200 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** B and **d** Sr.



FIG. S32. Electronic structure of The of YBeH₈ at 100 GPa. **a** Electronic band structure (left panel) and projected density of states (right panel). And fatband of **b** H, **c** B and **d** Ca.



8. Phonon structure of "fluorite-like" hydrides

FIG. S33. Phonon structure of "fluorite-like" hydride CaBeH₈ at 210 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S34. Phonon structure of "fluorite-like" hydride CaBH₈ at 100 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S35. Phonon structure of "fluorite-like" hydride LaAlH₈ at 100 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S36. Phonon structure of "fluorite-like" hydride LaBeH₈ at 50 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S37. Phonon structure of "fluorite-like" hydride LaBH₈ at 100 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S38. Phonon structure of "fluorite-like" hydride $SrBH_8$ at 150 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).



FIG. S39. Phonon structure of "fluorite-like" hydride YBeH₈ at 100 GPa. Phonon structure with projective lambda (left panel) and phonon projected density of states (right panel).

9. Superconductivity of "fluorite-like" backbone hydride



FIG. S40. Gap calculated by Eliashberg equation of "fluorite-like" hydride CaBeH₈ at 210 GPa.



FIG. S41. Gap calculated by Eliashberg equation of "fluorite-like" hydride CaBH₈ at 100 GPa.



FIG. S42. Gap calculated by Eliashberg equation of "fluorite-like" hydride LaAlH₈ at 100 GPa.



FIG. S43. Gap calculated by Eliashberg equation of "fluorite-like" hydride LaBeH₈ at 50 GPa.



FIG. S44. Gap calculated by Eliashberg equation of "fluorite-like" hydride LaBH₈ at 100 GPa.



FIG. S45. Gap calculated by Eliashberg equation of "fluorite-like" hydride SrBH₈ at 150 GPa.



FIG. S46. Gap calculated by Eliashberg equation of "fluorite-like" hydride YBeH₈ at 100 GPa.

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