frontiers | Frontiers in Electronic Materials

MINI REVIEW
published: 25 May 2022
doi: 10.3389/femat.2022.906213

# Advances in the Synthesis and Superconductivity of Lanthanide Polyhydrides Under High Pressure

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Room-temperature superconductors have long been the ultimate goal of scientists. Pressure-stabilized hydrides are a new rapidly growing class of high-temperature superconductors and are believed to be a new superconducting system, undoubtedly leading to a surge in the discovery of new hydrogen-rich materials. They are the forefront of physics and material science. Lanthanide polyhydrides formed under pressure are promising conventional superconductors. Especially, both the theoretical and experimental reports on lanthanum superhydrides under pressure, exhibiting superconductivity at temperatures as high as 250 K, have further stimulated an intense search for room-temperature superconductors in hydrides. This review focuses on the recent advances of crystal structures, stabilities, and superconductivity of lanthanide polyhydrides at high pressures, including the experimental results from our group. By using *in situ* four-probe electrical measurements and the synchrotron X-ray diffraction technique, we have identified several high-temperature superconducting phases: a lanthanum superhydride and two cerium superhydrides. The present work indicates that superconductivity declines along the La–Ce–Pr–Nd series, while magnetism becomes more and more pronounced. These discoveries have enriched the binary system of clathrate superhydrides and provided more hints for studying the role of rare earth metal elements having high-temperature superconductivity.

**Keywords: high pressure, lanthanide hydrides, superconductivity, crystal structure, magnetic properties**

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**Specialty section:**
This article was submitted to
Superconducting Materials,
a section of the journal
Frontiers in Electronic Materials

**Received:** 28 March 2022
**Accepted:** 19 April 2022
**Published:** 25 May 2022

**Citation:**
Guo J, Chen S, Chen W, Huang X and Cui T (2022) Advances in the Synthesis and Superconductivity of Lanthanide Polyhydrides Under High Pressure. Front. Electron. Mater. 2:906213. doi: 10.3389/femat.2022.906213

## INTRODUCTION

Since Kamerlingh Onnes first discovered superconductivity with $T_c$~4.2 K in mercury (Onnes, 1911), room-temperature superconductivity has been one of the most challenging projects in multiple fields such as physics and material science. In 1957, Bardeen, Cooper, and Schrieffer established the microscopic BCS theory to provide an explanation for conventional superconductors (Bardeen et al., 1957), which was also considered a road map to high-temperature superconductors. For these years, the search for superconducting materials with higher $T_c$ has been ongoing. The famous and representative conventional superconductor $MgB_2$ had a $T_c$ of 39 K at ambient pressure (Nagamatsu et al., 2001), which was far lower than the $T_c$ for cuprates with 133 K (Iqbal et al., 1994). By the application of hydrostatic pressure, $T_c$ of the HgBaCaCuO compound was increased to ~164 K (Gao et al., 1994). However, cuprate-based superconductors are beyond the conventional BCS theory, and there is shortage of a generally accepted theory to explain the microscopic mechanism of them.

Back in 1935, Winger and Huntington proposed that hydrogen could be metallized under high pressure (Winger et al., 1935). According to the BCS theory, high phonon frequencies, large electronic density of states (DOS) near the Fermi level, and strong electron phonon coupling (EPC) can result in high $T_c$. In 1968, Ashcroft predicted that the realization of solid metallic hydrogen would open the pathway to high $T_c$ superconductors (Ashcroft., 1968). Achieving a metallic state of hydrogen, referred to as metallic hydrogen, is dubbed as the Holy Grail in the high-pressure community. Experimental evidences on the transformation of hydrogen to the atomic state were reported close to 500 GPa (Loubeyre et al., 2020; Dias et al., 2017; Eremets et al., 2019), but such higher pressure limited its properties and application research. In 2004, Ashcroft suggested an alternative approach that metallization and superconductivity of hydrogen-rich compounds existed at much lower pressures (Ashcroft., 2004). Only 10 years later, Ashcroft's idea found its experimental proofs, and extraordinarily, high-temperature superconductivities were demonstrated in compressed $H_3S$ and $LaH_{10}$ (Drozdov et al., 2015; Einaga et al., 2016; Troyan et al., 2016; Huang et al., 2019; Somayazulu et al., 2019; Drozdov et al., 2019). It has been found that binary hydride $LaH_{10}$ can achieve high temperature superconductivity of 250–260 K, representing a new breakthrough in the binary system until now. These encouraging results have spurred a flurry of interests in other compressed hydrides, particularly for the ones from the same group. In this review, we are going to summarize the recent progresses on the lanthanide polyhydrides under high pressure. Through the discussions, we hope that some hints could be concluded for future studies in new hydrides with high-temperature superconductivity.

## DICUSSIONS

### Lanthanum Superhydride

Lanthanum superhydride $LaH_{10}$ was first theoretically reported as a good candidate for high-$T_c$ superconductivity (Peng et al., 2017; Liu et al., 2017), before experiments confirmed its high-temperature superconductivity (Somayazulu et al., 2019). Peng et al. and Liu et al. have independently performed systematic density functional theory (DFT) studies and proposed that the high-temperature superconducting phase *fcc*-$LaH_{10}$ with $T_c$ reaching 280 K. *fcc*-$LaH_{10}$ showed a clathrate-like structure constituted by H cages, with La atoms occupying the *fcc* lattice positions. The high $T_c$ has been explained as the strong electron -phonon coupling, which is associated with the strongly hybridized La *f* and H *s* orbitals (Liu et al., 2019; Wang et al., 2020). These theoretical predictions that lanthanum superhydride has great potential to be a near room-temperature superconductor have motivated experimental verifications. However, such experiments are very challenging, limited by hydrogen permeability and sample size.

Several experimental groups have successfully synthesized the predicted *fcc*-$LaH_{10}$ phase by laser heating La metal with pure hydrogen or ammonia borane ($NH_3BH_3$) at high pressure in a diamond anvil cell. The initial experimental observation of $LaH_x$ with $T_c$ at 215 K and 150 GPa was claimed by Drozdov et al. (Drozdov et al., 2018). They have obtained the target sample by heating the sample of La and pure hydrogen below 1,000 K at 170 GPa. The sample had a $T_c$ of 209 K at 170 GPa, which increased to 215 K when the pressure reduced to 150 GPa. Then, Somayazulu et al. reported the superconducting lanthanum superhydride with $T_c$ ~ 260 K at 180–200 GPa (Somayazulu et al., 2019). Different from Drozdov et al., they chose ammonia borane as the hydrogen source. In addition, they also found that $T_c$ decreased with increasing measuring current. Subsequently, Drozdov et al. changed the synthesized conditions and reported that $LaH_{10}$ had a highest $T_c$ of 250 K at 170 GPa and decreased linearly as pressure increased (Drozdov et al., 2019). In their work, they also confirmed the crystal structure of $LaH_{10}$ as $Fm\overline{3}m$ symmetry *via* synchrotron X-ray diffraction and observed the isotope effects in the experiment. Besides those two groups, Hong et al. observed that the highest $T_c$ for the La–H system was 246 K at 136 GPa (Hong et al., 2020), but the real phase was not known. Recently, Sun et al. found that $T_c$ decreased to 191 K as the pressure dropped from 138 to 120 GPa, which was associated with the phase transition changing from $Fm\overline{3}m$ to $C2/m$ (Sun et al., 2021).

During this time, we have also explored the superconductivity in the lanthanum superhydride. We used $NH_3BH_3$ as the hydrogen source considering the heating decompression reaction: $NH_3BH_3 \rightarrow 3H_2 + c\text{-}BN$ (Chen et al., 2021; Shao et al., 2021b; Kondrat'ev et al., 2015). The La and $NH_3BH_3$ samples were loaded inside the hole of the gasket. We have applied the four-probe method to study the electrical resistance, and four Mo electrodes were sputtered onto the diamonds with a top flat of 60 μm beveled 300 μm, which have been successfully used in our previous work (Chen et al., 2021). The Raman shift of diamond was measured to calibrate the pressure (Akahama et al., 2006). By laser heating a mixture of La and $NH_3BH_3$ to about 1,400 K at 171 GPa, a resistance drop was triggered at about 238.4 K during the cooling cycle (**Figure 1A**). To further confirm this superconductivity, we have also measured the temperature-dependent electrical resistance under a series of applied magnetic fields from 0–6 T. $T_c$ decreased along with the increasing magnetic fields, and the superconducting state was suppressed by the external magnetic fields, further proving the superconductivity. The upper critical magnetic fields were extrapolated based on the single-band Werthamer–Helfand–Hohenberg (WHH) (Werthamer et al., 1966) and Ginzburg–Landau (GL) (Ginzburg et al., 1950) models. The two models yielded the $H_{c2}(0)$ as 158.1 and 114.9 T, respectively. These values are consistent with the reported experimental data (Drozdov et al., 2019). The coherence length can be calculated based on $\mu_0 H_{c2} = \Phi_0/(2\pi\xi^2)$. The $\xi_{\text{WHH}}(0)$ and $\xi_{\text{GL}}(0)$ are equal to 1.44 and 1.69 nm, respectively. In Shukor's work (Shukor, 2021), the value of coherence length for $LaH_{10}$ is 15.6–18.6 Å calculated by using $\xi_{\text{GL}} = 0.74\,\xi_0\,(1-T/T_c)^{1/2}$, which is consistent with our work.

### Cerium Polyhydrides

From 2018, our group has carried out a series of work on cerium polyhydrides, particularly the discovery of two high-temperature

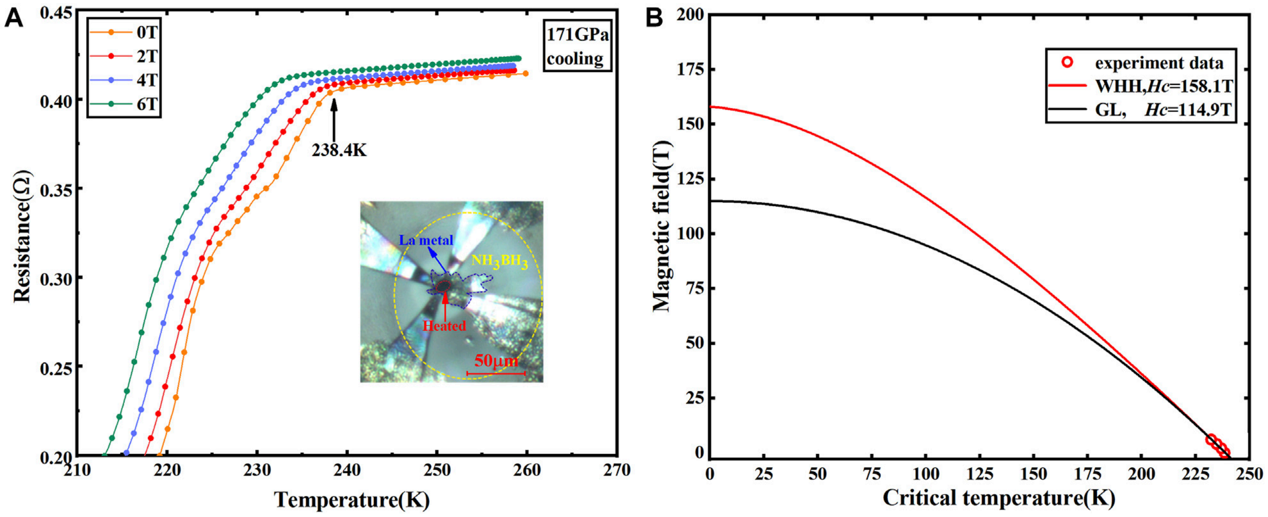

**FIGURE 1 |** Our work measured the superconductivity of lanthanum superhydride at 171 GPa. **(A)** Temperature-dependent electrical resistance at 171 GPa during the cooling process and upon the external magnetic fields (0–6 T), measured by the four-probe technique. The inset shows that the sample chamber consists of La and $NH_3BH_3$ after laser heating. **(B)** Upper critical magnetic fields estimated by the single-band Werthamer–Helfand–Hohenberg (WHH) (Werthamer et al., 1966) and Ginzburg–Landau (GL) (Ginzburg et al., 1950) models.

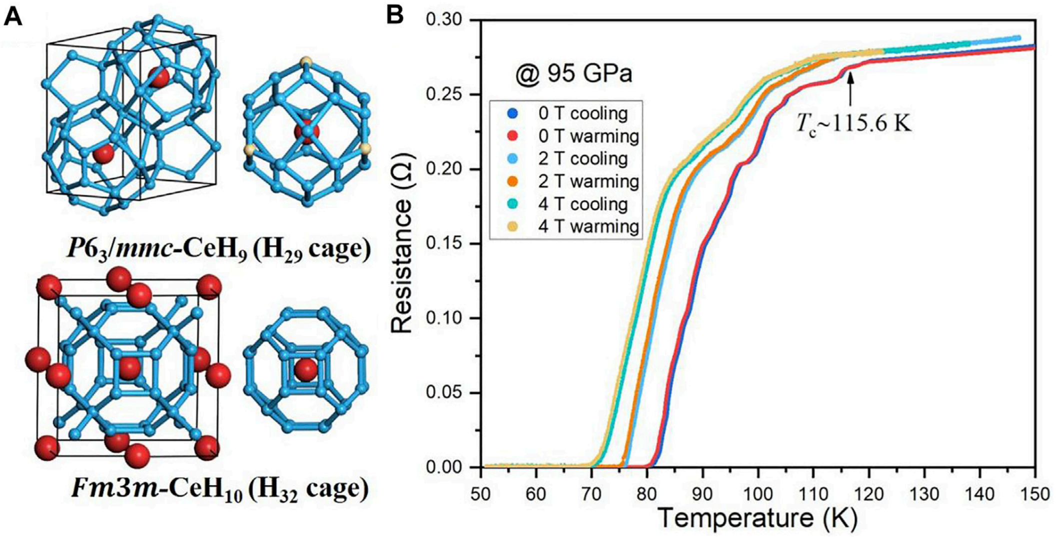

**FIGURE 2 |** Crystal structures and superconducting transitions in the Ce–H system (Chen et al., 2021). **(A)** Crystal structures and hydrogen sub-lattice of $P6_3/mmc$-$CeH_9$ and $Fm\overline{3}m$-$CeH_{10}$ phases. **(B)** Superconducting transitions for the $Fm\overline{3}m$-$CeH_{10}$ phase determined by the electrical resistance measurements, with the external magnetic fields of 0–4 T at 95 GPa.

superconducting cerium polyhydrides. Several new hydrides are studied by the treatment of different compression pathways (Li X. et al., 2019; Li et al., 2021). We have successfully synthesized a series of cerium polyhydrides by the direct reaction of Ce and $H_2$ upon cold compression up to 159 GPa (Li X. et al., 2019). The Ce polyhydrides ($CeH_3$, $CeH_{3+x}$, $CeH_4$, $CeH_{9-\delta}$, and $CeH_9$) presented an increase of hydrogen content as pressure increased. The formed $CeH_9$ had a unique clathrate-like structure consisting of H29 cages surrounding Ce atoms occupying the hexagonal $P6_3/mmc$ symmetry (see **Figure 2A**). $CeH_9$ was also with the nearest-neighbor H–H distances closest to predictions for solid atomic metallic hydrogen in all synthesized hydrides. The electron localization function of $CeH_9$ indicated an ionic bonding between Ce and H atoms, and the band structure confirmed its metallic character. The density of electronic-state calculations indicated the significant contribution of H at the Fermi level. Our cold-compression experiment provides a facile route to potential superconductors in superhydrides. The discovery of $CeH_9$ with atomic-like hydrogen sub-lattice suggests a low-pressure route for bulk dense atomic hydrogen stabilized by other element atoms. Independently, Salke et al. synthesized the clathrate hydride $CeH_9$ at 80–100 GPa by laser heating the mixture of the Ce sample and $H_2$ gas up to 2,000 K (Salke et al., 2019). They decompressed the sample and observed that the hexagonal $CeH_9$ became unstable below 93 GPa.

Recently, we have also further conducted deep experiments to explore the possible superconducting phases, and two phases were acquired: $P6_3/mmc$-$CeH_9$ and $Fm\overline{3}m$-$CeH_{10}$ (Chen et al., 2021). The synchrotron X-ray diffraction verified that the superconductivity arose from $CeH_9$ and $CeH_{10}$. Both $P6_3/mmc$-$CeH_9$ and $Fm\overline{3}m$-$CeH_{10}$ had high symmetric sodalite-like clathrate structures. For these two phases, zero resistance was evidenced by four-probe measurement in our experiment, which showed an obvious decrease of sample resistance during the cooling process. We first synthesized the $Fm\overline{3}m$-$CeH_{10}$ phase, isostructural to $Fm\overline{3}m$-$LaH_{10}$, which showed an almost linear increase of $T_c$ during the release of pressure. The measured $T_c$ reached 115 K at a pressure below 1 Mbar, as shown in (**Figure 2B**). In addition, the measured $T_c$ also decreased with the enhancement of applied external magnetic fields.

The isotope effects were also experimentally observed by substituting the hydrogen in $CeH_9$ with heavier deuterium and the decreased $T_c$ was measured, which substantiated the conventional electron–phonon coupling mechanism of the BCS theory. According to the reported theoretical calculations (Li B. et al., 2019; Peng et al., 2017), the much heavier and larger cerium atoms provided sufficient electrons to hydrogen and stabilized the H29 and H32 fascinating cages. The predicted $E_f$ and electron–phonon coupling coefficient $\lambda$ were both at high level, which contributed to an optimistic superconducting $T_c$. Presently, our result enriches the binary system of clathrate superhydrides and provides more evidence for studying the role of rare earth metal elements. This work also verifies that Ce is a potential choice for designing ternary or even more complex high-temperature superconducting hydrides.

## Praseodymium and Neodymium Polyhydrides

We also performed a similar experimental method on the praseodymium and neodymium polyhydrides (Zhou et al., 2020a; Zhou et al., 2020b). Under high-pressure and high-temperature conditions, the introduction of lanthanide heavy metal atoms stabilized the hydrogen structure, and new superhydrides with high hydrogen content were obtained. We have adopted $NH_3BH_3$ as the hydrogen source, which greatly improved the success rate of experimental studies on superhydrides. At 130 GPa, we synthesized two novel superhydrides $F\overline{4}3m$-$PrH_9$ and $P6_3/mmc$-$PrH_9$, both of which had similar hydrogen-cage configurations as *fcc*-$LaH_{10}$ and $P6_3/mmc$-$CeH_9$. Resistance measurements showed that the synthesized mixture of cubic and hexagonal $PrH_9$ demonstrated a resistance drop around 9 K in high pressure, indicating a possible superconducting transition below 9 K (Zhou et al., 2020). Similar experimental methods were used in the Nd–H system, and three compounds, $I4/mmm$-$NdH_4$, $C2/c$-$NdH_7$, and $P6_3/mmc$-$NdH_9$, were obtained (Zhou et al., 2020b). The resistance measurements of synthesized $NdH_9$ demonstrated that there was no superconducting transition at 5–300 K over 100 GPa. Further calculation results showed that a magnetic order and electron–phonon interaction co-existed in a very close pressure range in praseodymium hydrides, which might have affected the low $T_c$. Subsequently, another group successfully synthesized several praseodymium polyhydrides at around 100 GPa (Pena-Alvarez et al., 2019). They heated the sample between 1,000 and 1,400 K at 85 GPa for 10 s, and both hexagonal $PrH_7$ and $PrH_9$ diffraction peaks could be observed in X-ray diffraction patterns. During the decompressing cycle, $PrH_9$ remained stable down to around 80 GPa, and the $PrH_7$ could decompose to $PrH_4$ below 54 GPa.

Our group has obtained new hydrides $PrH_9$ and $NdH_9$ in experiment. The theoretical calculations found that $PrH_9$ possessed weak superconductivity, while $NdH_9$ displayed strong magnetism (Zhou et al., 2020a; Zhou et al., 2020b). With the increase of *f* electrons, the outer *f* electrons had a significant effect on the superconductivity of these new superhydrides. The present results on lanthanide superhydrides show that superconductivity declines along the La–Ce–Pr–Nd series, while magnetism becomes more and more pronounced (see **Figure 3**). Generally, based on the BCS theory, the electron–phonon coupling directly affected the superconductivity of the conventional superconductors. The existence of magnetism prevented the formations of cooper pairs and then was not favorable to the superconductivity. Therefore, compared with lanthanum hydride, the increased number of *f* electrons in neodymium displayed pronounced magnetic properties, at the same time suppressing the conventional superconductivity based on electron–phonon coupling. It was further clarified that magnetism is an important factor to affect the conventional superconductivity in superhydrides.

## Europium Polyhydrides

Europium as an active metal is a divalent element in general and reacts with hydrogen under pressure. Our previous work has obtained a series of Eu hydrides ($F\overline{4}3m$-$EuH_9$, $P6_3/mmc$-$EuH_9$ and $Pm\overline{3}n$-$Eu_8H_{46}$) by laser heating the Eu metal and $NH_3BH_3$ at high pressure (Semenok et al., 2021). $EuH_9$ adopted $F\overline{4}3m$ structure from 86 to 130 GPa, and the $P6_3/mmc$ phase required higher stable pressure. We also discovered the $Pm\overline{3}n$-$Eu_8H_{46}$ phase when heated to 1,600 K at 74 GPa, and the content of $Eu_8H_{46}$ increased as pressure increased. These three polyhydrides were all clathrate-like structures and magnetic. The Monte Carlo simulation suggested the anti-ferromagnetic ordering critical temperature for $F\overline{4}3m$-$EuH_9$ with $T_N = 24$ K. The $P6_3/mmc$-$EuH_9$ and $Pm\overline{3}n$-$Eu_8H_{46}$ were calculated to be with ferromagnetic ordering critical temperatures $T_c = 137$ and 336 K, respectively. Ma et al. also obtained the same hexagonal clathrate-structure $EuH_9$ by laser heating samples to 2,800 K at 170 GPa (Ma et al., 2021). Moreover, they also obtained clathrate-structure $EuH_6$. Their electrical resistance measurements indicated the possible magnetic order

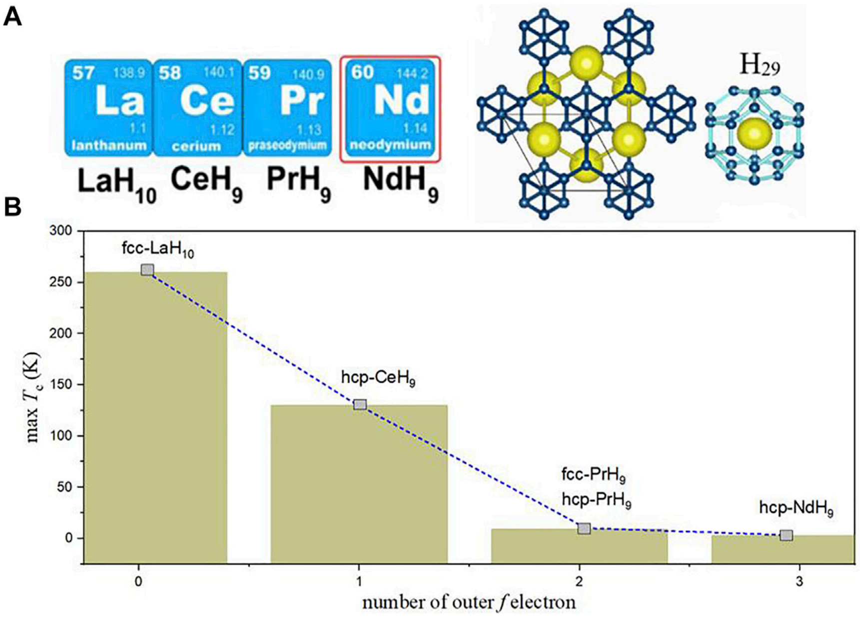

**FIGURE 3 |** Crystal structure and superconducting transition temperature $T_c$ of novel lanthanide metal hydrides synthesized experimentally. **(A)** Crystal structures and hydrogen cage configurations of $CeH_9$, $PrH_9$, and $NdH_9$ found in this work; **(B)** Relationship of maximum $T_c$ and outer *f* electrons of these new superhydrides.

transition temperature at 225 and 258 K for $Pm\bar{3}n$-$EuH_5$ and $Im\bar{3}m$-$EuH_6$, respectively. The weak superconductivity of Eu hydrides was attributed to the strong magnetism similar to Nd hydrides.

## Other Heavy Lanthanide Polyhydrides

The heavy lanthanide hydrides are predicted as promising superconductors under pressure. Several groups have carried on the theoretical predictions of heavy lanthanide hydrides with high hydrogen content (Peng et al., 2017; Sun et al., 2020; Hai et al., 2021; Song et al., 2021). Peng et al. reported a serious of stable rare earth (RE) hydrides $REH_6$, $REH_9$, and $REH_{10}$ with unusual H clathrate structures at high pressure. The H-rich RE hydrides usually exhibited high-temperature superconductivity related to the H clathrate structures with strong EPC and large H-derived DOS near the Fermi surface, but the heavy lanthanide polyhydrides had much lower $T_c$ due to heavier atoms which reduced the superconductivity (Peng et al., 2017). Subsequently, Sun et al. performed the EPC calculations for lanthanide hydrides with high hydrogen content and summarized the $T_c$ for them. For the RE elements with half-filled 4*f* states (Eu, Gd, Dy, and Ho), they had the lowest DOS H-*s* values, resulting in a weak EPC and low $T_c$ below 10 K. As the number of outer electrons increased, the $T_c$s for $YbH_{10}$ and $LuH_8$ reached as high as 102 K at 250 GPa and 86 K at 300 GPa, respectively (Sun et al., 2020). A series of stable superconducting Tb hydrides under pressure have been studied by Hai et al. $TbH_n$ ($n$ = 1, 2, 3) were predicted to be stable under low pressure, while the terbium hydrides with high hydrogen content were stable at high pressure. The terbium polyhydrides exhibited high-temperature superconductivity especially for $Fm\bar{3}m$-$TbH_{10}$, with a $T_c$ beyond 270 K above 230 GPa (Hai et al., 2021). The other theoretical work indicated that $YbH_6$ and $LuH_6$ showed the $T_c$ as high as 145 K at 70 GPa and 273 K at 100 GPa, respectively (Song et al., 2021). In terms of the experiment, our work has

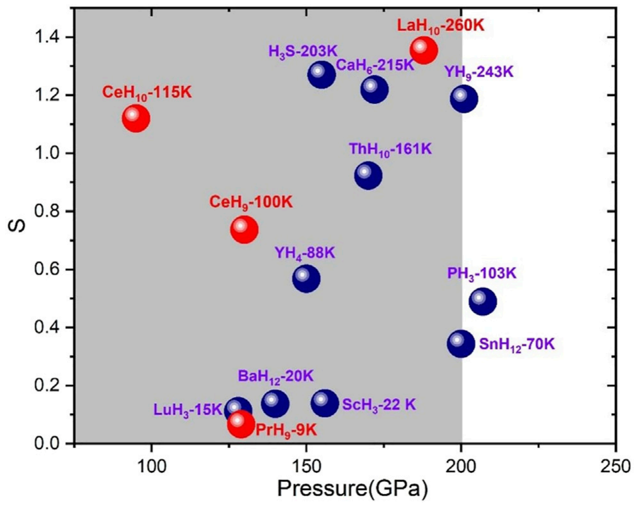

**FIGURE 4 |** S for different superconducting hydrides reported from the experiment. Merit index *S*, can be explained as follows: $S=T_c/(T^2_{c,\mathrm{MgB2}}+P^2)^{1/2}$ (Pickard et al., 2020; Shipley et al., 2021; Wang et al., 2021), where the temperature is expressed in Kelvin and the pressure is expressed in GPa. $T_{c,\mathrm{MgB2}}$ is the superconductivity critical temperature of $MgB_2$ at ambient pressures, which equals to 39 K.

successfully synthesized $Fm\overline{3}m$-$LuH_3$ under high pressure (Shao et al., 2021a). The $T_c$ for $LuH_3$ increased, first with compression to 15 K at 128 GPa, then decreased sharply, and increased to 14 K at 156 GPa at last. The upper critical magnetic fields $H_{c2}(0)$ were estimated to be 8.47 and 7.17 T by WHH and GL models, respectively. The failure of synthesizing high $T_c$ lutetium superhydrides above a pressure of 1 Mbar indicated that it was much tougher for Lu to form high hydrogen content hydrides than light lanthanide elements.

## PERSPECTIVE

It is important to find the high-temperature superconductors; at the same time, it is essential that the pressures required are reduced. Merit index $S$ reflects the relationship between $T_c$ and pressure (Pickard et al., 2020). We set $S(MgB_2)$ as 1. A superconductor with a higher value of $S$ means it has a higher critical temperature at mild pressure. As **Figure 4** shows, $LaH_{10}$ and $CeH_{10}$ have high $S$ values 1.35 and 1.12, which indicates that these hydrides have a stronger superconductivity under similar pressure. Thus, lanthanide elements have played a key role in forming high-temperature superconductors.

In conclusion, we summarize the experimental results of lanthanide hydrides at high pressure. The lanthanide hydrides with high hydrogen content have clathrate structures with hydrogen cages surrounding metal atoms and show cubic or hexagonal symmetries in general. Although several high-temperature superconducting lanthanum hydrides have been discovered, the extremely high synthetic pressures limit their practical applications. Therefore, there is still a long way to obtain a high-temperature superconductor at ambient pressure.

## AUTHOR CONTRIBUTIONS

XH conceived this project. SC and WC performed the experiment. JG analyzed the experimental data. XH and JG wrote and revised the article. All authors discussed the results and offered the useful discussions.

## FUNDING

This work was supported by the National Key R&D Program of China (Grant No. 2018YFA0305900), the National Natural Science Foundation of China (Grant Nos. 11974133, 52072188, 51632002, and 51720105007), and the Program for Changjiang Scholars and Innovative Research Team in University (Grant No. IRT_15R23).

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**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.