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Stoichiometric Ternary Superhydride $LaBeH_8$ as a New Template for High-Temperature Superconductivity at 110 K under 80 GPa

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The search for high-temperature superconducting superhydrides has recently moved into a new phase by going beyond extensively probed binary compounds and focusing on ternary ones with vastly expanded material types and configurations for property optimization. Theoretical and experimental works have revealed promising ternary compounds that superconduct at or above room temperature, but it remains a pressing challenge to synthesize stoichiometric ternary compounds with a well-resolved crystal structure that can host high-temperature superconductivity at submegabar pressures. Here, we report on the successful synthesis of ternary $LaBeH_8$ obtained via compression in a diamond anvil cell under 110–130 GPa. X-ray diffraction unveils a rocksalt-like structure composing La and BeH_8 units in the lattice. Transport measurements determined superconductivity with critical temperature T_c up to 110 K at 80 GPa, as evidenced by a sharp drop of resistivity to zero and a characteristic shift of T_c driven by a magnetic field. Our experiment establishes the first superconductive ternary compound with a resolved crystal structure. These findings raise the prospects of rational development of the class of high-T_c superhydrides among ternary compounds, opening greatly expanded and more diverse structural space for exploration and discovery of superhydrides with enhanced high-T_c superconductivity.

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In his seminal work published in 2004, Ashcroft proposed to find high-temperature superconductivity in hydrogen dominant metallic alloys (superhydrides) [1], opening a research area that has since seen explosive growth across many scientific disciplines in concerted efforts to search for such intriguing superconductors at much lower pressures compared to those required for producing metallic hydrogen [2,3]. Among the early milestone developments are the theoretical prediction of ionic clathrate CaH_6 [4] and the experimental realization of covalent SH_3 with a critical temperature (T_c) of 203 K [5]. Inspired by the CaH_6 work, extensive efforts have focused on compounds with similar clathrate structures, culminating in recent computational discoveries of rare-earth (RE) superhydrides REH_6, REH_9, and REH_{10} consisting of H_{24}, H_{29}, and H_{32} cages, respectively, which were predicted to possess higher T_c approaching the room temperature [6–8]. These remarkable results were confirmed by ensuing experimental synthesis and characterization, as seen in a plethora of binary superhydrides, including YH_6 [9,10], YH_9 [10,11], CeH_9, CeH_{10} [12], ThH_9, ThH_{10} [13], and LaH_{10} [14–17] with measured T_c values ranging from 57 to 262 K. Most recently, experimental efforts successfully synthesized the long missing first predicted clathrate CaH_6 compound that has a T_c of 210–215 K [18,19], representing the highest T_c among non-RE ionic superhydrides.

Despite the impressive accomplishments in the study of high-T_c superhydrides to date, further development of this field toward establishing more diverse and robust superconducting superhydrides that can operate at or above room temperature as desired for most practical applications has been hindered by two major factors. First, most studies on superhydrides have so far concentrated on binary compounds, which are relatively easy for computational prediction and experimental characterization of the related crystal structure, but impose severe limits on the structural configurational space, reducing the variety of metal elements that can facilitate the formation of atomiclike hydrogen in superhydrides [20–24]. Second, while ternary systems offer more structural prototypes [21] that expand the material basis for exploring potentially superior superconducting properties, the required structure search and characterization are much more challenging, impeding an understanding of the structure-property relation that is the foundation to elucidating superconducting phenomena and the underlying mechanism.

The quest for ternary superconducting superhydrides has been led by some notable recent theoretical and experimental efforts (please refer to the review paper on ternary superhydrides [25]). A computational study proposed to introduce Li to donate extra electrons into the molecularlike hydrogen in MgH_{16} for generating atomiclike H_{18} and H_{28} cages and forming ternary Li_2MgH_{16} compound, which was predicted to host hot superconductivity well above room temperature albeit under multimegabar pressures [26]. Recent experiments found nonstoichiometric alloyed ternary superhydrides ($(La, Y)H_6$, $(La, Y)H_{10}$ [27], and $(La, Ce)H_9$ [28,29]), where the random occupation of the lattice sites renders the crystal structure effectively identical to the clathrate structures of the previously examined binary systems. While the superconductivity has been experimentally reported in some ternary hydrides like Li_5MoH_{11} [30] and $BaReH_9$ [31], these hydrides do not carry good prototype structures for high T_c superconductivity since their T_c values are rather low at or below 7 K even under high pressure. To date, there remains a formidable challenge on the synthesis of stoichiometric ternary superhydrides with high-T_c superconductivity, presenting a major bottleneck for the study of ternary superconducting superhydrides that have the needed data on definitive structural and composition for theoretical evaluation and moderate (e.g., submegabar) pressures for access by experimental characterization.

Recent theoretical studies have predicted an appealing ternary XYH_8-type structure, where atomiclike hydrogens are stabilized by chemical compression originating from the X-Y scaffolding [32–34]. The combination of heavy X and light Y elements with disparate sizes makes this ternary structure exhibit a more efficient stacking compared to previously explored binary clathrate structures [33,34]. More interestingly, when the X element is replaced with another light element Be, the resulting isomorphic $LaBeH_8$ is predicted to be a high-temperature superconductor and thermodynamically stable at an experimentally accessible pressure of about 100 GPa [35], thus providing a desirable material platform for experimental verification.

In this Letter, we report on the synthesis of stoichiometric $LaBeH_8$ with a well-resolved crystal structure in $Fm\text{-}3m$ symmetry at 110–130 GPa with an X-Y skeleton and novel BeH_8 unit that is clearly distinguished from the previously studied binary clathrate REH_n compounds [4,6,7]. The T_c up to 110 K obtained below megabar pressure is confirmed by our electrical transport experiments. The present work produces the first successful experimental synthesis of a distinct stoichiometric ternary compound in XYH_8 structure prototype [32–34], opening a promising avenue for exploring an expanded class of superconducting superhydrides.

We used a diamond anvil cell for the high-pressure synthesis. Equimolar La-Be alloy and NH_3BH_3, which was used as a main hydrogen source [9,13,17,18,36,37], are used as the precursors in this work. The samples were pressurized to target pressures at room temperature. Laser heating was performed using a pulsed yttrium-aluminum-garnet laser (1064 nm) or SPI fiber laser (1070 nm). A total of 12 cells were prepared to synthesize the target ternary hydride and characterize its superconductivity. The samples were pressurized to 110–130 GPa at room temperature and then heated to about 800–2300 K for 1 min using infrared laser. The color of the sample becomes black, along with a pressure reduction after irradiation, indicating the expected synthetic reaction accompanied by a volume collapse. The experimental details [e.g., the x-ray diffraction (XRD) and electronic transport measurements, etc.] are summarized in the Supplemental Material [38].

In situ XRD measurements were carried out to determine the crystal structure of the synthesized product. As shown in Fig. 1(a), the main peaks at 7.66°, 8.84°, 12.52°, and 14.69° can be indexed to be contributed by the $(1\,1\,1)$, $(2\,0\,0)$, $(2\,2\,0)$, and $(3\,1\,1)$ planes of a high-symmetric face-centered cubic structure (space group $Fm\text{-}3m$) with the cell parameter of $a = 5.36$ Å, which is consistent with the simulated value (5.28 Å) of the predicted $Fm\text{-}3m$ phase of $LaBeH_8$ [35]. Although the observed diffraction peaks are well refined with $LaBeH_8$, the exact positions of H and Be cannot be directly determined by the current beam flux of x-ray sources, owing to the weak x-ray scattering cross sections of the light elements. This is a well-known problem in the XRD-directed structure determination of superhydrides and the same difficulty inevitably appears in structure solution of other known superhydrides (e.g., H_3S [59], LaH_{10} [14,15], CaH_6 [18], YH_6, and YH_9 [9,10]). Nevertheless, the experimental hydrogen content was indeed determined to be ∼8 via the method of the lattice volume expansion derived from hydrogen [38], similar to the strategy adopted in previous other experiments on syntheses of polyhydrides [10,12,14,15,18,28]. Also, the experimental equation of state measured on the synthetic sample is highly consistent with the theoretical data of $Fm\text{-}3m$ $LaBeH_8$ [Fig. 1(b)]. These results and analysis support that we have successfully synthesized a ternary hydride $LaBeH_8$, which can be considered as a rocksalt (B_2) structure composing La and BeH_8 units [Fig. 1(b)] that have not been previously observed in any binary or nonstoichiometric ternary alloy superhydrides. We checked the structure stability at other pressures by carrying out the XRD measurements up to 145 GPa (Fig. S2 [38]), at which pressure the diamond anvil cracked. From the data, it is seen that the $Fm\text{-}3m$ structured $LaBeH_8$ appears at 121 GPa and is stable at least up to 145 GPa.

We performed a series of electrical transport measurements on $LaBeH_8$ to explore its superconductivity under high pressure. Representative electrical resistance data as a function of temperature at different pressures are shown in Fig. 2(a), and the results indicate obvious superconducting transitions as evidenced by the sharp resistance drop at

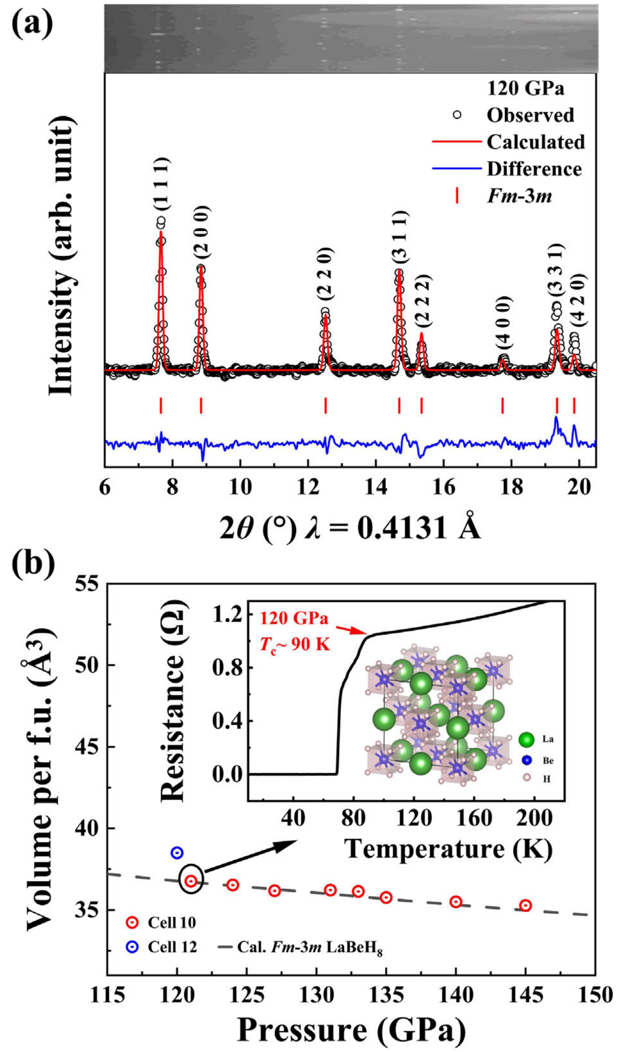

FIG. 1. (a) Synchrotron x-ray diffraction pattern of the heated sample (cell 12) at 120 GPa and the Rietveld refinement of the Fm-3m $LaBeH_8$ structure. The high-intensity points caused by single-crystal-like diffraction are masked, and the continuous background is removed before performing the integration and Rietveld analysis. The black circles and red and blue curves correspond to the experimental data, Rietveld refinement fit, and residue, respectively. The red ticks indicate the calculated peak positions for Fm-3m $LaBeH_8$. The crystal surface indexes are marked next to the peaks. The cake plot shows some textures that can be fitted by spherical harmonics, where the harmonic order is 10 and the texture index is 1.386. (b) Experimental equation-of-state data (circle symbols) for the synthetic samples are compared with the theoretical data (dashed line) derived from the Fm-3m structured $LaBeH_8$. The experimental data from cell 10 and cell 12 are marked with red and blue circles, respectively. Inset figure depicts the superconducting transition with $T_c \sim 90$ K in the electrical measurement for cell 10 at 120 GPa and the crystal structure of $LaBeH_8$.

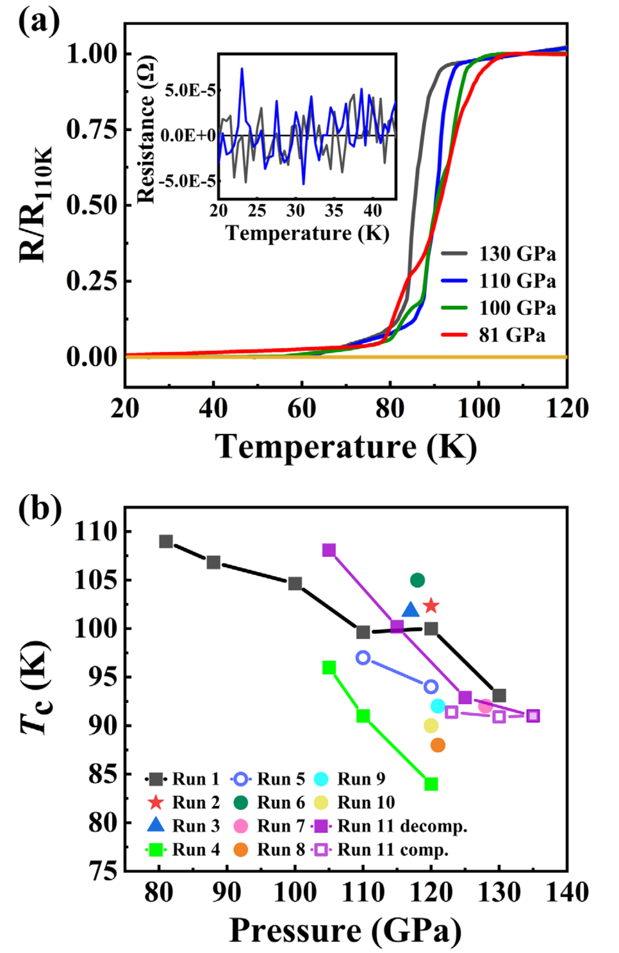

FIG. 2. (a) The observation of superconductivity in $LaBeH_8$ (cell 1). The vertical axis is the resistance divided by the resistance at 110 K. The resistance data with near zero values are shown on a smaller scale in the left inset. (b) The dependence of the onset of superconducting temperature T_c data on pressure for 11 different experimental runs from cells 1–11 (Figs. S4–S15 [38]). The experimental data are marked in different colors. The open and solid symbols represent the data obtained from compression and decompression, respectively.

93 K, 100 K, and 104 K around 130 GPa, 120 GPa, and 100 GPa, respectively. In these measurements, observation of zero resistance [insets of Figs. 2(a), S4, S5, S10, S11, S12, and S14 [38]] excludes the possibility of other temperature-induced transitions where an abrupt drop of resistance may appear. Notably, we observed a sharp drop of resistance around 90 K at 120 GPa in cell 10, and the XRD measurement (Fig. S2 [38]) was then performed for the same cell to determine the Fm-3m structure of the superconducting sample. The experimentally observed onset of T_c rises with decreasing pressure as summarized in Fig. 2(b). The highest measured T_c is about 110 K at 80 GPa, at which pressure the cracking of diamond anvil leads to the breaking of electrodes, preventing further measurements at lower pressure. Experimentally observed superconductivity is highly sensitive to the symmetry and stoichiometry of hydride [12]. In high-pressure experiments, uncontrolled anisotropic stress often leads to slight lattice distortion that is difficult to be distinguished by XRD

measurements, as typically occurred in the La-H system [14,15,60]. In addition, polyhydrides with slightly different hydrogen concentrations are often seen in the syntheses, such as $LaH_{10\pm x}$ [14,15,17,60] and $CeH_{9\pm x}$ [12,61,62]. Here, in our work, both lattice distortion and slight deviation from ideal stoichiometry (e.g., possible defects of hydrogen and beryllium) of synthetic samples for different runs may result in T_c variation, as also occurred in other superconducting polyhydrides [12,14,18]. It should be pointed out that our experimentally observed T_c value of 104 K at 100 GPa is significantly lower than the theoretical T_c result of 166–192 K calculated at the same pressure by Zhang *et al.* [35], but is in good consistence with our calculated T_c result at 106–117 K [38] where much denser k and q points sets are used in our calculation.

Most superconductive superhydrides exhibit T_c values first increasing then decreasing with reducing pressure [10,14,18], displaying domelike shaped T_c-pressure curves that stem from their structural destabilization as pressure is released. In LaH_{10}, for example, the high-symmetric cubic structure tends to be distorted, forming a monoclinic phase below 160 GPa, which is attributed to the pressure-induced phonon softening that causes the decrease of T_c at lower pressures [15,60]. In contrast, the onset (T_c^{onset}) of the superconducting transitions of $LaBeH_8$ increases linearly with decreasing pressure within the range probed in this work, which can be approximately fitted by a function of $T_c^{\text{onset}} = kP + T_0$ with $k = -0.37 \pm 0.08$ K/GPa and $T_0 = 139.20 \pm 9.02$ K. The robust dynamic stability of $LaBeH_8$ [35] plays a crucial role in maintaining its high symmetry over a wide range of pressure, thus allowing the steady enhancement of its superconductivity in the lower pressure range. Besides, we also track the offset temperature (T_c^{offset}) of the superconducting transitions as shown in Fig. S3(b) [38], as well as the transition width (ΔT) that is determined by $\Delta T = T_c^{\text{onset}}\text{-}T_c^{\text{offset}}$. We note that the evolution of offset shows an opposite trend to the onset's one above 88 GPa. The broadening of the superconducting transitions becomes wider during decompression, which might be caused by inhomogeneities or grain or surface effects [63,64] in the high-temperature synthesis.

Limited by currently available experimental techniques, it is difficult to collect the weak signals of magnetic flux expulsion (i.e., Meissner) effect at extremely high pressures [11,14]. But it is feasible to examine T_c as a function of external magnetic field, as shown in Fig. 3(a), where the measured results reveal that the resistance drop gradually shifts to lower temperatures as the magnetic field is increased in the range 0–9 T at 120 GPa, thus verifying the nature of the superconducting transition in $LaBeH_8$ as expected by the conventional superconductivity theory. The upper critical field as a function of temperature is shown in the inset of Fig. 3(b). The application of a magnetic field reduces T_c by about 15 K at $\mu_0 H = 9$ T. The extrapolation values of the upper critical field $\mu_0 H_{c2}(T)$ and the coherence length toward T = 0 K are 33.5 T and 31.4 Å and 44.5 T and 27.2 Å fitted by the Ginzburg-Landau (GL) [65,66] and Werthamer-Helfand-Hohenberg (WHH) [67] models, respectively. The magnitude of the estimated coherence length (27.2–31.4 Å) is well below the typical value (hundreds of angstroms) for type-I superconductors, indicating that $LaBeH_8$ is a type-II superconductor.

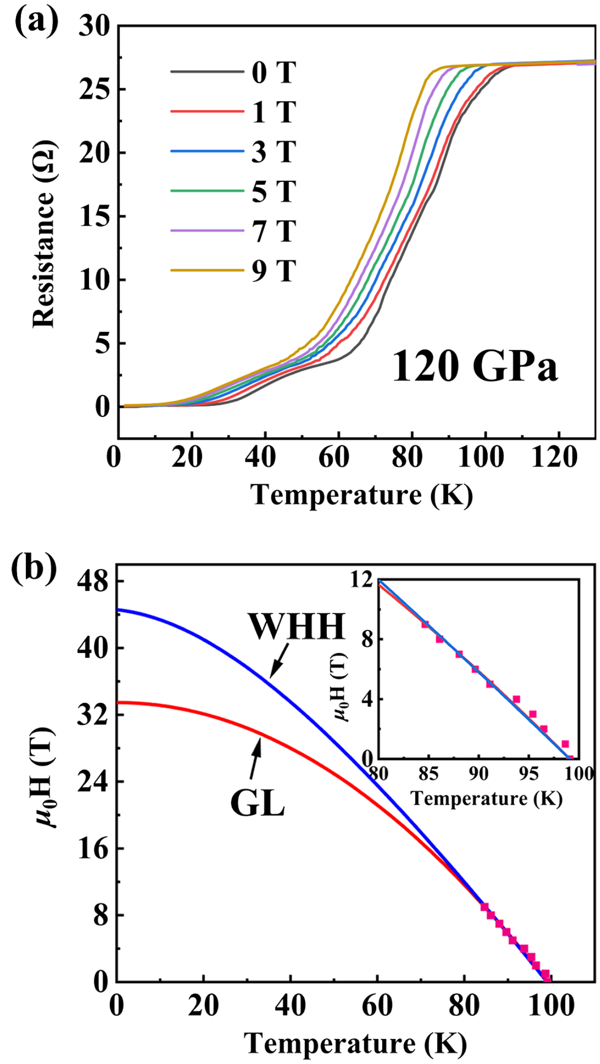

FIG. 3. (a) Temperature dependence of the electrical resistance in cell 3 under applied magnetic fields of H = 0, 1, 3, 5, 7, and 9 T at 120 GPa. (b) Upper critical field H_{c2} versus temperature following the criteria of 90% of the resistance in the metallic state at 120 GPa, fitted with the GL and WHH models. Inset: the dependence of the T_c under the applied magnetic field.

In our experiments, NH_3BH_3 was used as a hydrogen source for the $LaBeH_8$ synthesis. It is noteworthy that a previous theory [33] predicted that $LaBH_8$ is a thermodynamically stable phase at the experimental pressure of 120 GPa. This might raise a possibility on the synthesis of $LaBH_8$ in the experimental product. Below, we discussed from three aspects to exclude this possibility and give further evidence for the assignment of our superconducting sample as $LaBeH_8$, other than $LaBH_8$. First, under high temperature, NH_3BH_3 is thermally decomposed into H_2 and cubic BN (c-BN) [68–70], of which the latter accommodating strong covalent B-N bonds is chemically rather stable to against its chemical interaction with elements or compounds [71]. This is the underlying origin on why there is no any report on the formation of B-bearing hydrides through the use of NH_3BH_3 as hydrogen source in the syntheses of a large number of superconducting superhydrides (e.g., YH_6, YH_9, LaH_{10}, CeH_9, CeH_{10}, CaH_6, EuH_6, EuH_9, ThH_9, ThH_{10}, etc. [9,10,12,13,17,18,72]). Second, at the synthetic pressure of 120 GPa, a recent theory [34] revealed that $LaBH_8$ is not a thermodynamically stable phase anymore once $LaBH_6$ and $LaBH_7$ are considered in the convex hull calculations, and it becomes stable only above 160 GPa. As further evidence, we also performed enthalpy simulations at 120 GPa for the designed synthetic routes of $LaBeH_8 + c\text{-BN}$ over $LaBH_8 + 1/3\ Be_3N_2 + 1/6\ N_2$ and found that formation of $LaBeH_8$ is ($\sim$160 meV/atom) energetically much more favorable than that of $LaBH_8$. Third, in order to avoid introducing B in the experiment, we used an alternative precursor of paraffin (C_nH_{2n+2}, n = 18–39) as the hydrogen source, which has been earlier used in the synthesis of other superhydrides [73–76]. The La-Be alloy was sandwiched between paraffin in a diamond anvil cell, and the sample was heated above 2000 K at a loaded pressure of 123 GPa. The resultant electrical resistance data for the newly synthesized La-Be-H compound are shown in Fig. S15(a) [38], which clearly show high-temperature superconducting behaviors (e.g., $T_c = 108$ K at 105 GPa). The experimental T_c values with the variation of pressure for the synthesized product with the uses of different hydrogen sources of paraffin and NH_3BH_3 are consistent with each other as shown in Fig. S15(b) [38]. This experiment by using paraffin without containing any B elements gives us a further support on the assignment of the synthesized superconducting superhydride to $LaBeH_8$.

Unlike nonstoichiometric ternary alloy superhydrides that have similar metal elements and share the identical structure framework as binary systems [27–29], the stoichiometric $LaBeH_8$ structural prototype contains two types of nonhydrogen elements with different atomic sizes [32–35]. In this stoichiometric ternary XYH_8 structure, small Be atoms have weak interaction with hydrogen atoms and form BeH_8 units, which fill the interspace of La sublattice, thereby inducing more efficient packing and producing more pronounced chemical precompression compared to binary hydrides. By means of proper selection and combination of metal elements in $LaBeH_8$, large amounts of atomiclike hydrogen can form near or even below megabar pressure, whereas it is impossible to form the hydrogen cage structure in a binary La-H system at such reduced pressures. The present successful experimental synthesis and characterization of $LaBeH_8$ are encouraging for the exploration of wide-ranging ternary superhydrides that may exhibit further enhanced T_c values under similarly moderate or lower pressures.

In summary, using diamond anvil cell compression and laser heating techniques, we have successfully synthesized a stoichiometric ternary superhydride $LaBeH_8$ in a distinct structural framework containing X-Y skeleton and BeH_8 units. The measured superconducting critical temperature T_c of up to 110 K was obtained at 80 GPa. To our knowledge, this is the first experimental realization of an archetype ternary prototype with exact stoichiometry, well-resolved structure, and T_c beyond 100 K. Our results establish an important paradigm in search of stoichiometric ternary or higher multinary high-T_c superhydrides via a suitable combination of metal elements, potentially leading to further enhanced T_c under reduced pressures toward more accessible scientific exploration and ultimate practical applications.

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