

Hydrogen Vacancy Induced Superconductivity Collapse in A15 Lanthanum Hydride

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Hydrogen-rich lanthanum compounds show the highest known superconducting transition temperatures at high pressure. Despite the pivotal role of hydrogen within these systems, there has been no systematic exploration of how the composition and superconducting properties are intertwined. Our experimental and computational studies demonstrate that A15-type $\text{LaH}_{5.75-x}$ hosts high- T_c superconductivity ($T_c = 98$ K at 94 GPa) when available interstitial sites are fully occupied with hydrogen. Upon decompression, a site-selective hydrogen depopulation drives a superconductor-to-insulator transition with a threshold composition of LaH_5 . Strikingly, the A15 framework is experimentally retained from 120 to 4 GPa, with reversible pressure-dependent hydrogen content changes from $\text{LaH}_{5.75}$ down to $\text{LaH}_{3.25}$. Our results demonstrate the exceptional stability and tunability of the A15 framework, offering a unique platform to probe the interplay between composition, structure, and superconductivity in hydrogen-rich materials.

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Lanthanum hydrides exhibit a rich structural and stoichiometry diversity [1–5], with a wide range of observed electronic properties. These span from electrically insulating LaH_3 to LaH_{10} with the highest confirmed critical temperature (T_c) to date (250 K at 170 GPa) [1,2,5–7]. While LaH_{10} has been extensively investigated, there exist several intermediate phases (LaH_6 , LaH_{3-4} , $\text{LaH}_{5.75}$, and LaH_9) with limited experimental characterization [2,7,8]. Of particular interest is A15 $\text{LaH}_{5.75}$, which is superconducting with T_c above liquid nitrogen temperatures but at substantially lower pressures than LaH_{10} [3,4]. Despite this, the stability and superconducting behavior at low pressure remain unknown.

Understanding the direct correlation between hydrogen content and superconducting properties within the same crystallographic framework is essential to guide the rational design of next-generation hydride-based superconductors [9]. In rare-earth hydrides, the A15 structure has emerged as a recurring motif [2,10–16], providing an efficient framework for densely packing atomic hydrogen within a metallic lattice. Across different metal-hydride systems,

hydrogen occupancy in the tetrahedral interstitial sites varies widely [2,10–17], giving rise to a broad range of possible stoichiometries and hydrogen-ordering patterns. This diversity makes A15-type hydrides an ideal model system for investigating the interplay between pressure, stoichiometry, and superconductivity. Computational studies [18,19] suggest that introducing hydrogen vacancies could influence the pressure required to stabilize high- T_c phases, yet this possibility remains experimentally unexplored. Recent measurements on lanthanum hydrides have suggested a gradual decrease in T_c over time [20]. While this is suggested to be linked to hydrogen loss [20], studies that provide a clear connection between crystal structure, hydrogen content, and superconducting behavior are of paramount importance [21].

Through synchrotron x-ray diffraction, Raman spectroscopy, and electrical transport measurements supported by density functional theory (DFT) and molecular dynamics (MD) simulations, we reveal a hydrogen stoichiometry-controlled superconductor–insulator transition that occurs entirely within the same crystallographic A15 framework. We experimentally demonstrate that A15 $\text{LaH}_{5.75-x}$ ($x \approx 0$ –2.5) is stable upon decompression down to 4 GPa but the hydrogen content H/La continuously decreases from close to maximal $\text{H}/\text{La} = 5.75$ down to $\text{H}/\text{La} = 3.25$, as supported by DFT calculations. We show that pressure-dependent hydrogen content changes are responsible for a rapid loss of superconductivity below 90 ± 5 GPa. These findings establish a direct correlation

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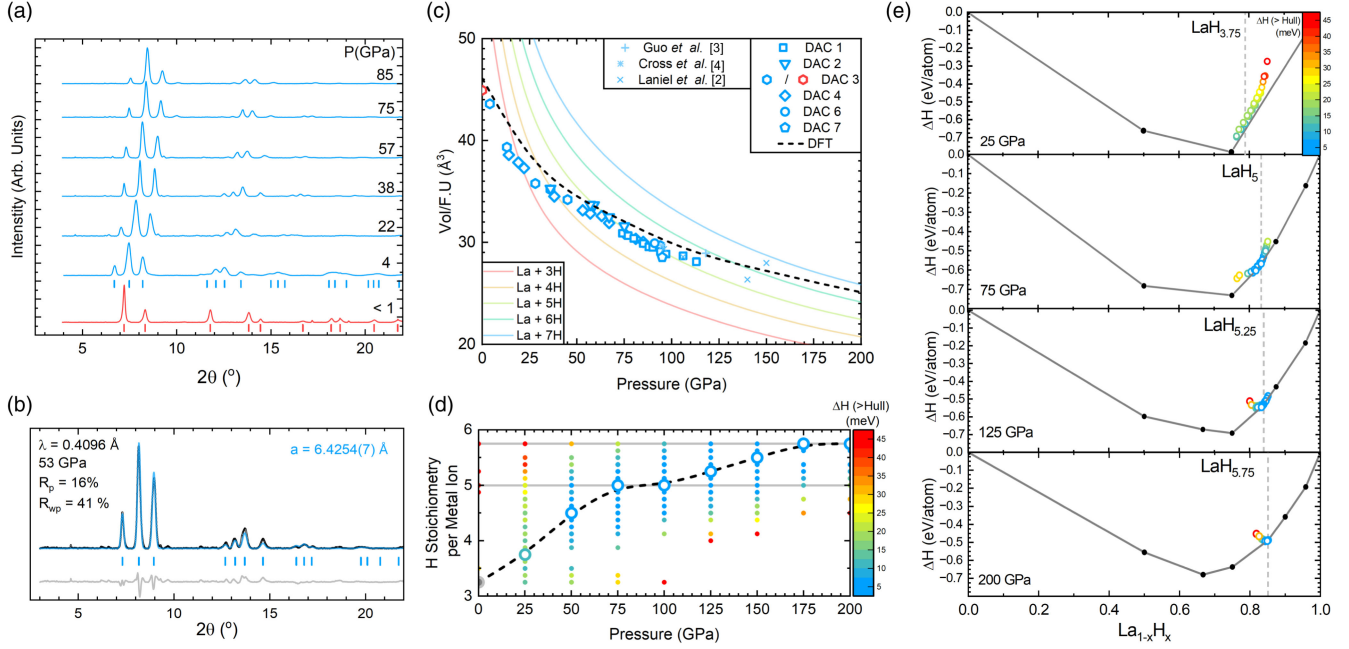


FIG. 1. (a) X-ray diffraction patterns of A15 $\text{LaH}_{5.75-x}$ collected upon decompression. Tick marks of A15 $\text{LaH}_{5.75-x}$ are shown in blue. Data and tick marks shown in red depict the decomposition to fcc- LaH_3 . Data taken with incident x-rays of wavelength 0.4096 $\text{\AA}$. (b) Rietveld refinement to A15 $\text{LaH}_{5.75-x}$. Black data points show experimental data, with blue line showing the Rietveld refinement. The gray line is the difference between the experimental and calculated patterns. (c) DFT calculated and experimentally observed volumes per formula unit (F.U.) for the A15 lanthanum hydride. Colored curves show additions of the volumes of $\text{La} + \text{H}_x$, taken from Refs. [24] and [25], respectively. Blue points are experimentally measured volumes of A15 $\text{LaH}_{5.75-x}$ from x-ray experiments in this Letter, in which different symbols correspond to different experimental runs. The red data point shows the measured volume of fcc- LaH_3 . Runs labeled DAC 1 was measured upon compression, while the other measurements are taken upon decompression. The dashed black line shows the calculated volumes from DFT. (d) AIRSS calculated hydrogen stoichiometries. The dashed black line links A15 structures with the highest hydrogen content, calculated to be closest to, or on the convex hull of enthalpies. (e) Convex hulls of the $\text{La}_{1-x}\text{H}_x$ system. Black points show the convex hull, whilst hollow points are enthalpies from structures with an A15 lanthanum sublattice. Color gradient (blue to red) indicates distance from the convex hull.

between hydrogen loss and the suppression of superconductivity, and provide a benchmark for the low-pressure stability of extended hydrogen networks in metal hydrides.

A detailed description of both experimental and computational methods is provided in the Supplemental Material [22]. At pressures between 70 and 100 GPa, laser heating lanthanum in a hydrogen medium (or ammonia borane as the hydrogen source) readily forms A15 $\text{LaH}_{5.75-x}$ [see Fig. 1(a)]. Upon decompression, the A15-type sublattice remains stable from 115 to 4 GPa. In Fig. 1(b) we present the Rietveld refinement for the A15-type lanthanum sublattice at 53 GPa, in which lanthanum atoms occupy the $2a$ (0, 0, 0) and $6d$ (0, 0.5, 0.25) Wyckoff positions, detailed in Table S1 [22]. Below 1 GPa, only fcc- LaH_3 was observed [7,23], indicating the transformation of the A15 hydride to prior known fcc- LaH_3 between 1 and 4 GPa.

Previously, A15 lanthanum hydride was only studied between 95 to 150 GPa, with this Letter significantly extending the known pressure range of this phase [2–4]. Above 90 GPa, our data are in good agreement with the experimental volumes previously reported for A15 lanthanum hydride [2–4]. Comparing our results with calculated

constant-stoichiometry equation of state (EOS) curves formed from $V(\text{La}) + xV(\text{H})$ [24,25] (colored lines), we observe that between 100 and 115 GPa the volume per formula unit (F.U.) lies between $\text{La} + 5\text{H}$ and $\text{La} + 6\text{H}$. The experimental volume per formula unit follows a unique trend that does not match any of the constant-stoichiometry EOS curves, but instead intersects through each ($\text{La} + 5\text{H}$ to $\text{La} + 3\text{H}$) upon decreasing pressure. This atypical EOS behavior indicates a significant change of hydrogen content in A15 $\text{LaH}_{5.75-x}$, occurring on both compression and decompression (inverted triangles and diamonds or hexagons, respectively). This reproducible pressure dependence suggests that the hydrogen stoichiometry variability is intrinsic to the A15 sublattice, rather than due to time-dependent kinetics or sample decomposition.

To explore the effect of stoichiometry change in A15-type lanthanum hydride, we have performed *ab initio* random structure searching (AIRSS) calculations. The corresponding formation enthalpies of the computed structures are shown in Fig. 1(e). Colored points are from structures with an A15 lanthanum sublattice, in which the color gradient depicts the distance from the convex hull.

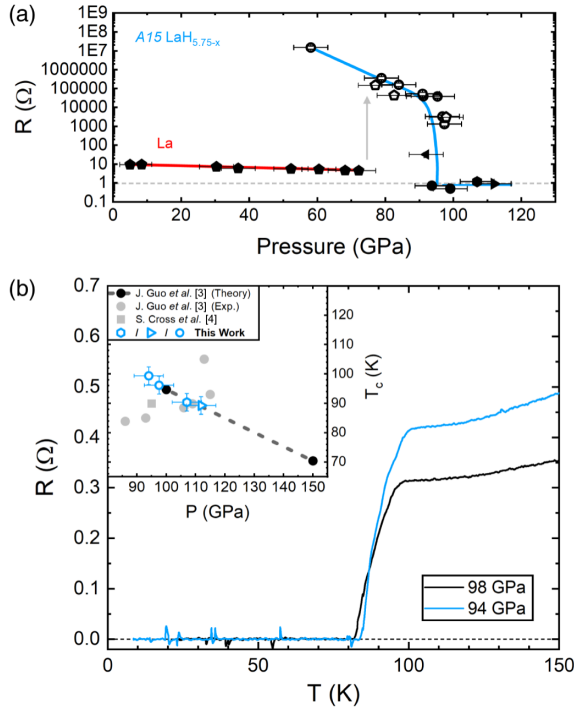


FIG. 2. (a) Resistance for A15 $\text{LaH}_{5.75-x}$ at room temperature. Empty symbols are two-point resistance values, whilst solid symbols are four-probe electrical resistance values. Pentagons connected by the red line indicate resistances measured upon compression for elemental La in ammonia-borane (DAC7) before laser heating. The increase in resistance upon laser heating is indicated by the grey arrow. Circular points (DAC6) correspond to an experiment with decompression from 98 to 91 GPa, compression from 91 to 97 GPa and decompression to 58 GPa. Further details on different experimental runs are provided in Table S2 [22]. The blue line is a guide to the eye. (b) Electrical resistance measured for A15 $\text{LaH}_{5.75-x}$ upon warming. The onset of T_c was determined to be at the intersection between the normal state resistance and a linear fit to the resistance drop. The inset shows extracted T_c values for this Letter and those from previous publications [3,4]. A dashed line is used to join our experimental results with theoretical values from [3].

Black points are from additional La-H structures not of A15 type. While A15 $\text{LaH}_{5.75}$ is only thermodynamically stable above 175 GPa, we observe that hydrogen deficient variants A15 $\text{LaH}_{5.75-x}$ remain stable down to 75 GPa, with decreasing hydrogen content as pressure is reduced. Below this pressure, all searched A15 structures lie above the convex hull by 5–45 meV, indicating metastability, in good agreement with our experimental synthesis pathways.

The corresponding evolution with pressure in stable hydrogen stoichiometry is presented in Fig. 1(d). Computational results evidence a slight hydrogen content loss in A15 $\text{LaH}_{5.75-x}$ from $\text{H/La} = 5.75$ at 175 GPa to $\text{H/La} = 5$ at 100 GPa, before dropping more sharply below 75 GPa, reaching $\text{H/La} = 3.25$ at ambient pressure. These trends are qualitatively supported by experimental

estimations of hydrogen content (Fig. S1 [22]), which indicate a similar drop in hydrogen content, although the large variance in the hydrogen equation of state [25,26] limits their reliability at low pressure. Nevertheless, as shown by the dashed black curve in Fig. 1(c), the measured volumes across 4 to 115 GPa align closely with those predicted for a A15 lanthanum hydride with a hydrogen content that varies with pressure.

Figure 2(a) presents the evolution of the room temperature electrical resistance in A15 $\text{LaH}_{5.75-x}$ below 120 GPa. Upon laser heating La in NH_3BH_3 , there is a sharp increase in the measured electrical resistance from that of metallic elemental lanthanum at 77 GPa, up to 150 k Ω , signaling the formation of a nonmetallic phase. Further compression then drives a rapid drop in resistance [see Fig. 2(a)]. Above 94 GPa A15 $\text{LaH}_{5.75-x}$ exhibits high-temperature superconductivity with an onset of T_c of 96 K at 98 GPa, as shown from drops to zero resistivity presented in Fig. 2(b). Decompression marginally increases this T_c value up to 98 K at 94 GPa; this behavior differs from the previous nonmonotonic experimentally derived pressure dependence of critical temperature [3], possibly arising from phase inhomogeneities, impurities, or uncertainty in pressure determination [27]. Our negative $T_c(P)$ trend however agrees with recently reported calculations on A15- La_4H_{23} [3] and additional experiments with partial resistance drops (Fig. S2 [22]). The nonzero residual resistance in these samples is a result of an incomplete synthesis of A15- $\text{La}_{5.75-x}$, with residual metallic but nonsuperconducting phases, such as *Cmcm* LaH_3 [2,7] (see Figs. S3 and S4 [22]).

The superconducting state of A15-type $\text{LaH}_{5.75-x}$ disappears below 94 GPa, showing a drastic loss of metallicity upon decompression. At pressures below 94 GPa, the resistance of A15-type $\text{LaH}_{5.75-x}$ dramatically increases [Fig. 2(a)] indicating a nonmetallic state. The experimental data shown in Fig. 2 were obtained on compression and decompression, supporting the reversibility of this transition. This metal-insulator transition is additionally demonstrated by intermediate semiconducting behavior at 92 GPa presented in Fig. S5 [22]. The electrical resistance of A15 $\text{LaH}_{5.75-x}$ were measured down to 58 GPa, when it increased to 15 M Ω . The observed consistency between both compression and decompression is an excellent indicator that this resistance response to pressure is intrinsic to A15 $\text{LaH}_{5.75-x}$. XRD data (Figs. S3 and S4 [22]) and Raman spectroscopy data (Figs. S6 and S7 [22]) confirm the synthesis of A15- $\text{LaH}_{5.75-x}$ in all experimental runs, while XRD data presented in Fig. 1 show that the A15-type La sublattice does not undergo structural changes or decomposition down to 4 GPa.

To understand the mechanism behind the abrupt loss of metallicity on decompression, we use DFT calculations to examine the role of hydrogen occupancy across specific tetrahedral interstitial sites and the impact on the electronic

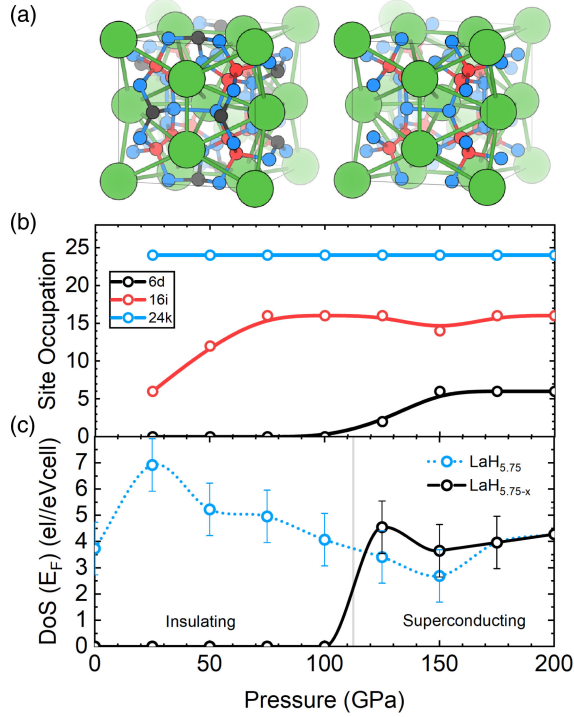


FIG. 3. (a) Crystal structure of A15 LaH_{5.75-x}. Lanthanum atoms are shown in green, while the black, red, and blue points represent the calculated hydrogen positions at sites 6c, 16i, and 16i + 24k, respectively, for full occupancy (left) and for only 24k occupancy (right). (b) Hydrogen occupation at sites 6c, 16i, and 24k calculated with static DFT for the structures on the convex hull (above 75 GPa), or those calculated to be closest to the convex hull (below 75 GPa). See main text for further details. Hydrogen site occupation is determined by mapping the relaxed hydrogen positions from AIRSS structures onto the nearest equivalent Wyckoff site in $Pm-3n$ space group. (c) eDOS calculated for the corresponding stable A15 LaH_{5.75-x} stoichiometries presented in Fig. 1(d), black line. Blue points depict the calculated eDOS for fully occupied A15 LaH_{5.75} at pressures outside its stability region (below 175 GPa). eDOS as a function of hydrogen stoichiometry is plotted in Fig. S8 [22].

density of states (eDOS). In Fig. 3(b), we present the computed hydrogen site occupations for the stable A15 LaH_{5.75-x} structures between 0 and 200 GPa. Two distinct regimes of dehydrogenation are observed.

Upon decompression, between 150 and 100 GPa, the 6c sites (black points) begin to depopulate, while 16i (red points) and 24k (blue points) remain fully occupied. A more substantial loss of hydrogen then occurs below 75 GPa, where the 16i sites also depopulate. Notably, the 24k sites retain full occupancy across the entire pressure range in which A15 LaH_{5.75-x} remains stable. This is consistent with previous reports on A15-type β -UH₃, where only the 24k sites host hydrogen [28].

We investigate the impact of this hydrogen content on the calculated electronic density of states shown in Fig. 3(c). Individual eDOS plots from which the density of states at the Fermi level (E_F) is derived are presented in Fig. S9 [22].

We find insulating materials if only the 16i + 24k sites are filled ($< \text{LaH}_5$), and metallic ones when the 6c sites are also occupied ($> \text{LaH}_{5.25}$). For LaH₅, band structures indicate semimetallic behaviour at 100 GPa, with a nonzero eDOS arising from small hole pockets shown in Fig. S10 [22]. We find that the tuning of electronic behavior between these regimes is driven primarily by hydrogen content as opposed to pressure, with hydrogenation above A15 LaH_{5.0} raising the Fermi level into the conduction band. Calculated electronic band gaps for structures with fixed hydrogen stoichiometry within and beyond their pressure range of stability are presented in Table S3 [22]. We find that while the $\text{DOS}(E_F)$ collapses for A15 LaH_{5.75-x} at 100 GPa, DFT calculations on fixed full hydrogen occupation (Fig. S11 [22]) show a consistently high $\text{DOS}(E_F)$ for all calculated pressures. These calculations evidence that the insulator-metal transition is not directly caused by pressure-dependent band gap closure, but instead is driven by hydrogen content and site occupancy in A15 LaH_{5.75-x}.

Within the uncertainties in pressure determination within both DFT calculations and high-pressure experiments, the predicted metal-insulator transition between 100 and 125 GPa is in excellent agreement with our experimental data presented in Fig. 2(a), in which high values of the measured resistance are observed at pressures below 90–100 GPa. However, the influence of further changes in the electronic DOS above 150 GPa on the superconducting properties remains unresolved and warrants detailed electron-phonon calculations that consider the evolving stoichiometry across this pressure range.

Raman spectroscopy experiments evidence a broad and distinct Raman continuum observed below 1500 cm⁻¹ in A15 LaH_{5.75-x}, as shown in Fig. S7 [22]. *Ab initio* molecular dynamics (MD) provides insight into the highly dynamic behavior of the hydrogens atoms in A15 LaH_{5.75} observing transient H₂ molecules of various lengths (Fig. S12 [22]). While each hydrogen remains in its tetrahedral site (Fig. S13 [22]), hydrogen pairs continuously form and break (Fig. S14 [22]) with separation between 0.85 and 1 Å (Fig. S12 [22]) through a triangle of La [22]. The short-lived nature of these modes broadens and suppresses any calculated spectral features [29,30] related to quasi-molecular H₂ above 2000 cm⁻¹) [31–33], well reproducing the experimentally observed Raman continuum [29,30]. Thus, this allows us to further identify the Raman spectra of A15 LaH_{5.75-x} as a signature for its formation.

Extended hydrogen networks have been recognized as a recurring structural motif in many high- T_c hydrides, but destabilize on decompression due to symmetry-breaking distortions [7,34–37]. Our results reveal a unique pathway in A15-LaH_{5.75-x}, where a reversible superconductor-insulator transition is driven purely by hydrogen content. Combined experimental and computational evidence shows that selective vacancy formation, first at 6c sites, then 16i,

progressively depletes the hydrogen network. This results in a collapse in the eDOS at the Fermi level and a drop in electrical conductivity. Given that in Bardeen-Cooper-Schrieffer (BCS) theory the eDOS is explicitly tied to the T_c [38], this sudden drop in $\text{DOS}(E_F)$ rapidly suppresses superconductivity and metallicity, which we experimentally observe below 90 ± 5 GPa. This contrasts with fcc-LaH₁₀ (and also bcc-H₃S), where the collapse of superconductivity is attributed to a cubic-to-rhombohedral lattice distortion.

While other A15-type compounds such as Lu₄H₂₃ and (Lu, Y)₄H₂₃ exhibit moderate T_c values (60–110K) [15,39], these have thus far been experimentally studied only at pressures close to maximal hydrogen occupancy. Recent predictions of A15 Ca₈H_{44–46} [19] and A15 NaY₃H₂₀ [18] find that small deviations from full hydrogen occupancy improve phase stability at low pressures, but no pressure-dependent hydrogen content changes were predicted. At lower hydrogen stoichiometries, A15-type hydrides such as CeH₃ and β -UH₃ [28,40] show no stoichiometry changes, where hydrogen site occupancy remains confined to the 24k sites only [28]. Our experimental and computational work shows a substantial evolution of hydrogen stoichiometry in A15 lanthanum hydride from LaH_{5.75}-LaH_{3.25}. The A15 structure of La atoms is therefore favorable to superconductivity when close to full hydrogen saturation, but is unable to retain full hydrogen content at lower pressures. Given that ternary substitution has proven a viable strategy for stabilizing high-temperature superconducting phases to lower pressures in La [41] and Y-based hydrides [42,43], the subsequent introduction of ternary dopant elements into A15 LaH_{5.75–x} may therefore offer a viable strategy for maintaining the superconducting properties down to lower pressure.

We establish A15-type hydrides as an unprecedented system for isolating the role of hydrogen occupancy in superconducting behavior. More broadly, we identify that the hydrogen content can undergo substantial changes within the A15 structure, which stabilizes A15 LaH_{5.75–x} at low pressure but at the cost of the metallicity and therefore the superconductivity. Our results on binary A15 LaH_{5.75–x} therefore provide a key benchmark for the design of next-generation hydrogen-rich superconductors and tunable quantum materials.

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The authors declare no competing interests.

Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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