Here we report the high-pressure synthesis of a series of lanthanum–yttrium ternary hydrides obtained at pressures of 170–196 GPa via the laser heating of P63/mmc La–Y alloys with ammonia borane. As a result, we discovered several novel compounds: cubic hexahydride (La,Y)H6 and decahydrides (La,Y)H10 with a maximum critical temperature $T_C$ of 253 K and an extrapolated upper critical magnetic field $B_{C2}(0)$ of up to 135 T at 183 GPa. The current–voltage measurements show that the critical current density $J_C$ in (La,Y)H10 is 12–27.7 kA/mm² at 4.2 K, which is comparable with that of commercial superconducting wires such as NbTi and Nb₃Sn. (La,Y)H₆ and (La,Y)H₁₀ are among the first examples of ternary high-$T_C$ superconducting hydrides. Our experiments show that part of metal atoms in the structures of recently discovered Im₃m-YH₆ and Fm₃m-LaH₁₀ can be replaced with lanthanum (~70%) and yttrium (~25%), respectively, with the formation of unique ternary superhydrides containing metal-encapsulated cages La@H₂₄ and Y@H₃₂, which are specific for Im₃m-LaH₆ and Fm₃m-YH₁₀. This work demonstrates that hydrides, unstable in pure form such as LaH₆ and YH₁₀, may nevertheless be stabilized at relatively low pressures in solid solutions with superhydrides having the desired structure.

Keywords: Superconductivity; Hydrides; USPEX; SCFDT; High pressure
Introduction
Metallic hydrogen is expected to display remarkable superconducting properties stemming from its high Debye temperature and strong electron–phonon coupling [1–4]. Theoretical calculations [5–7] show that metallization of hydrogen takes place only at extremely high pressures above 450 GPa, which have not been technically achievable for a long time. The renaissance of hydride superconductivity began in 2004 when Ashcroft [1] suggested that hydrogen–dominant metallic alloys may have high critical superconducting temperatures at much lower pressures. This scenario was realized in sulfur hydride [8] and in lanthanum hydride [9,10] at pressures below 200 GPa. Moreover, near-room-temperature superconducting transition (at 288 K), which is believed to be described within the conventional phonon-mediated mechanism of coupling, has been reported lately in carbonaceous sulfur hydride [11] and strong electron–phonon coupling [1–4]. Theoretical calculations [5–7] show that metallization of hydrogen takes place only at extremely high pressures above 450 GPa, which have not been technically achievable for a long time. The renaissance of hydride superconductivity began in 2004 when Ashcroft [1] suggested that hydrogen–dominant metallic alloys may have high critical superconducting temperatures at much lower pressures. This scenario was realized in sulfur hydride [8] and in lanthanum hydride [9,10] at pressures below 200 GPa. Moreover, near-room-temperature superconducting transition (at 288 K), which is believed to be described within the conventional phonon-mediated mechanism of coupling, has been reported lately in carbonaceous sulfur hydride [11] and strong electron–phonon coupling [1–4].

We wondered whether these unstable compounds could be stabilized in ternary La–Y alloys with a similar structure. Theoretical studies of several ternary hydride systems such as Ca–Y–H [19], Ca–Mg–H [20], and Li–Mg–H [21] carried out in recent years indicated a possibility of increasing the superconducting critical temperature in comparison with binary hydrides. However, ternary La–Y hydrides have not been systematically examined yet. Some hints of their promising superconducting properties have been made: using the neural network [22], we have found that La–Y–H is one of the most encouraging systems to achieve room-temperature superconductivity. At the same time, Kostrzewa et al. [23], analyzing the dependence of the logarithmically averaged frequency \( \nu_{\text{log}} \) on the mass of atoms, have predicted that the La–Y–H ternary hydrides may have \( T_c \) up to 274 K at 190 GPa.

In this work, we theoretically and experimentally investigated the superconducting properties of cubic hexahydrides \( \text{La}_x\text{Y}_{1-x}\text{H}_6 \), and decahydrides \( \text{La}_x\text{Y}_{1-x}\text{H}_{10} \), obtained via laser heating of yttrium–lanthanum alloys with ammonia borane (AB, \( \text{NH}_3\text{BH}_3 \)) used as a hydrogen source. Samples were compressed to 170–196 GPa in diamond anvil cells and heated by a series of laser pulses of 100–400 milliseconds duration. The superconducting properties of yttrium–lanthanum polyhydrides were investigated using the electrical transport measurements in different current modes and external magnetic fields. Ab initio calculations were used to unravel the composition and structure of the obtained compounds, as well as details of the mechanism of their superconductivity.

Results and discussion
Structure prediction
The study of yttrium hexahydride, synthesized at high pressures [13], has demonstrated that its critical temperature of superconductivity is significantly lower than theoretically predicted [24]. Is it possible to improve the parameters of the superconducting state by introducing another metal into the Y–H system? We noticed that lanthanum could form \( \text{Im} \bar{3} \text{m-LaH}_{10} \), which lies only slightly above thermodynamic stability (i.e. the convex hull, Fig. 1) at 150–200 GPa [18]. This cubic \( \text{LaH}_6 \) can be, at least kinetically, stabilized by an isostructural environment of \( \text{YH}_6 \), which is of great interest in terms of high-temperature superconductivity because of the structural analogy with yttrium hexahydride.

Using evolutionary algorithm USPEX [25–27], we searched for thermodynamically stable phases in the La–Y–H system (Fig. 1) and found that at 200 GPa and 0 K none of the ternary hydrides are on the 3D convex hull except \( \text{LaYH}_{20} \). Below in this paragraph we denote the symmetry of compounds without distinguishing Y and La. Surprisingly, \( P6_3/mmc-\text{LaYH}_{20} \) is stable at 0 K (Fig. 1a), whereas the cubic modification \( Fm \bar{3} m-\text{LaYH}_{20} \) is 50 meV/atom above the convex hull. An increase in temperature to 1000 K leads to the stabilization of tetragonal \( I4/mmm-\text{La}_2\text{YH}_{12} \) and cubic hexahydrides \( \text{La}_3\text{YH}_{10} \) and \( \text{LaYH}_{12} \). These phases are metastable at 0 K, with the distances from the convex hull equal to 82, 38, and 53 meV/atom, respectively. Moreover, \( \text{La}_3\text{YH}_{10} \), which belongs to the \( \text{La}_1_{-x}\text{Y}_{1+x}\text{H}_{10} \) structure type \( (x = 0.2) \), becomes stable at 1000 K. These structures can be considered as solid solutions of \( \text{YH}_{10} \) in \( Fm \bar{3} m-\text{LaH}_{10} \). Other phases that belong to the \( \text{La}_1_{-x}\text{Y}_{1+x}\text{H}_{10} \) group \( (\text{La}_2\text{YH}_{10}, \text{LaYH}_{12}, \text{La}_3\text{YH}_{10}, \text{LaYH}_{12}) \) are 5, 8, 13, and 17 meV/atom above the convex hull at 1000 K (Fig. 1b). Further increase in temperature to 1500 K leads to the stabilization of \( \text{La}_6\text{YH}_{30} \) and \( \text{La}_7\text{YH}_{18} \) (Fig. 1c), whereas higher temperature of 2000 K stabilizes the \( \text{La}_9\text{YH}_{50} \) phase (Fig. 1d). Above 1800 K, \( Fm \bar{3} m-\text{LaH}_{20} \) also becomes stable (at 2000 K, \( P6_3/mmc-\text{LaH}_{20} \) is 5 meV/atom above the convex hull).

Recently, Song et al. [28] conducted a more detailed analysis of the thermodynamic stability of \( \text{LaH}_{12} \) clathrate structures, proposing several new structures in the \( \text{La–Y–H} \) system: semimetallic nonsuperconducting \( R3\text{c-LaH}_{12} \), stable at 100–130 GPa, and \( Cm \bar{m} m-\text{LaH}_{12} \), stable at 140–300 GPa, with relatively low \( T_c \) = 140 K at 200 GPa. At 0 K and 200 GPa, phase \( R3\text{c} \) (with 78 atoms per unit cell) has the enthalpy of formation which is 24 meV/atom lower than that of \( Fm \bar{3} m-\text{LaH}_{12} \) studied in this work. The enthalpy of formation of another polymorphic
modification proposed by Song et al. [29], \textit{Cmmm}-LaYH$_{12}$ (28 atoms per unit cell), is 6 meV/atom lower than that of phase \textit{Pm}$\bar{3}$\textit{m}. Calculations at temperatures of 1500–2000 K in the harmonic approximation show the same order of the structures by the energy. It is important that all mentioned polymorphic modifications of LaYH$_{12}$ have very similar X-ray diffraction patterns. Moreover, as we show below, in all the experimentally studied compounds, the critical temperature is significantly higher than 200 K and may not be attributed to the \textit{R}$\bar{3}$\textit{c} or \textit{Cmmm} modifications of LaYH$_{12}$.

Phonon calculations show that only La$_2$YH$_{18}$ is dynamically stable within the harmonic approximation (Supporting Information Figs. S38–S40). However, an anharmonicity induced all investigated La–Y hydrides become dynamically stable, although in a number of cases (LaH$_x$, LaYH$_{20}$) their crystal structure is distorted (Supporting Information Figs. S41–S43). Under the entire range of studied pressure–temperature conditions, pure \textit{Im}$\bar{3}$\textit{m}-LaH$_6$ and \textit{Fm}$\bar{3}$\textit{m}-YH$_{10}$ were thermodynamically unstable.

**High-pressure synthesis**

To synthesize promising superconducting ternary yttrium–lanthanum hydrides, we prepared a series of La–Y alloys: La$_4$Y, La$_3$Y, La$_2$Y, LaY, and LaY$_4$. The yttrium (>$99.99\%$) and lanthanum (>99.99\%) metal pieces were mixed with the selected molar ratio and a total weight of 1 g, pressurized, and melted several times in an argon arc while the sample was rotated to homogenize the melt. Subsequent X-ray and electron diffraction, X-ray fluorescence (XRF), and energy-dispersive X-ray (EDX) analyses showed that the hexagonal phase \textit{P6}_3\textit{mmc}-La$_{x}$Y$_{1-x}$ is the main product of the alloying (Supporting Information Figs. S3–S5, S10–S11, S14–S15).

For loading high-pressure diamond anvil cells (DACs), we took the material from a homogeneous region of the alloy surface with the desired La:Y ratio determined by the EDX and XRF. To measure the synchrotron X-ray diffraction (XRD) and the critical temperature of superconductivity of synthesized lanthanum–yttrium hydrides, we used seven DACs (M1-2, M2_S, SL1, SL1_S, SL3, and SL3_S) with a 50 \textmu m culet beveled to 300 \textmu m at 8.5°, equipped with four $\sim$200 nm thick Ta electrodes with $\sim$80 nm gold plating (Fig. 3c) that were sputtered onto the piston diamond. Ammonia borane was used as a hydrogen source, following the technique that has shown good results in previous studies [13,29–33]. The detailed description of the DACs is presented in Supporting Information.

In five out of seven diamond anvil cells prepared for the XRD studies, the main observed phase was \textit{Fm}$\bar{3}$\textit{m}-(La,Y)H$_{10}$ (Fig. 2a,b,}

---

**FIGURE 1**

Ternary convex hulls of the La–Y–H system at a pressure of 200 GPa and temperatures of (a) 0 K, (b) 1000 K, (c) 1500 K, and (d) 2000 K, calculated with a contribution of the configurational entropy $T$S$_{conf}$ and the zero-point energy (ZPE). The invisible Z axis, perpendicular to the plane of the figure, corresponds to the enthalpy of formation. Stable and metastable phases are shown in red and blue, respectively.
Supporting Information Figs. S18–S21) with a unit cell volume of 31.8 Å³ at 180 GPa, which is about 0.5–0.83 Å³/f.u. lower than the volume of $Fm\bar{3}m$-LaH₁₀ found by Geballe et al. [12] (Fig. 2d, Table 1 and Supporting Information Table S5). The decrease in the unit cell volume is due to the replacement of large La atoms with Y, which have a smaller size. In the discovered ternary cubic La–Y polyhydride, the substitution of the La atoms with Y resulted in the formation of the Y@H₃₂ fragments with a locally distorted H-cage specific to $Fm\bar{3}m$-YH₁₀ [18].

There are large differences in the parameters of the unit cell of LaH₁₀ reported by different research groups [9,10,12,18,34]. In the first experiment by Geballe et al. [12], the unit cell volume of LaH₁₀ at about 170 GPa was over 33 Å³, which is significantly larger than 31.2 Å³ calculated theoretically using VASP (Fig. 1d, red circles). This prompted the authors of Ref. [12] to attribute the undefined stoichiometry LaH₉₋₃ to the discovered compound [12].

In the subsequent studies by Drozdov et al. [10] and Sun et al. [35], the experimental cell volume values of the synthesized LaH₁₀ ($V = 33.2$ Å³ at 150 GPa) are smaller by about 1.2 Å³/f.u. than those observed by Geballe et al. [12] (Fig. 2d). However, they are larger than those predicted using VASP [36–39], and are in close agreement with the equations of state (EoS) calculated using the Quantum ESPRESSO [40,41] pseudopotentials (Fig. 2d, “QE”, blue line). In both cases the Perdew–Burke–Ernzerhof functional [42] (generalized gradient approximation) was used. The experimental data of Sun et al. [35] were obtained in the low pressure range of 120–150 GPa, where distortion of the ideal cubic structure of $Fm\bar{3}m$-LaH₁₀ ($\tilde{C}2/m$) occurs [35] and, at the same time, there is a possibility of losing some hydrogen from the crystal structure.

Given the substantial uncertainty in both theoretical (Supporting Information Table S4) and experimental equations of states, and taking into account that the substitution of La with Y in LaH₁₀ should lead to a decrease in the unit cell volume, we performed an additional experiment with pure La compressed with AB in DAC SL3_S at 171 GPa. After the laser heating of the sample, we detected the formation of $Fm\bar{3}m$-LaH₁₀ with the unit cell volume of 32.82 Å³ (Supporting Information Fig. S22) and an impurity hexagonal (P6₃mc) LaH₁₀ phase with the unit cell volume of 33.05 Å³ (Supporting Information Fig. S23), which was also detected in previous experiments [10]. Thus, taking into
account that our experimental result for \( V(\text{LaH}_{10}) \) is close to the data of Ref. [12], we chose the 3rd order Birch–Murnaghan interpolation [43,44] of the experimental EoS found by Geballe et al. [12] as a reference (Fig. 2d, red dotted line). Using this EoS, we determined the composition of synthesized (La,Y)H\(_{10}\) as La\(_3\)YH\(_4\) or La\(_{0.75}\)Y\(_{0.25}\)H\(_{10}\) (Fig. 2e), in which the La:Y ratio is close to that

<table>
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<tr>
<th>DAC</th>
<th>Pressure, GPa</th>
<th>( a ), Å</th>
<th>( V ), Å(^3)</th>
</tr>
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<tbody>
<tr>
<td>( Fm\overline{3}m )-(La,Y)H(_{10})</td>
<td>M1</td>
<td>180</td>
<td>5.038(1)</td>
</tr>
<tr>
<td></td>
<td>M2</td>
<td>180</td>
<td>5.026(1)</td>
</tr>
<tr>
<td></td>
<td>SL1</td>
<td>180</td>
<td>5.031(1)</td>
</tr>
<tr>
<td></td>
<td>SL3</td>
<td>170</td>
<td>5.071(1)</td>
</tr>
<tr>
<td>( Im\overline{3}m )-(La,Y)H(_{6})</td>
<td>M2</td>
<td>175</td>
<td>3.65(1)</td>
</tr>
<tr>
<td></td>
<td>M2_S</td>
<td>180</td>
<td>3.64(1)</td>
</tr>
<tr>
<td>( Fm\overline{3}m )-LaH(_{10})</td>
<td>SL3</td>
<td>171</td>
<td>5.08(1)</td>
</tr>
<tr>
<td></td>
<td>171</td>
<td>5.07(1)</td>
<td>130.8(1)</td>
</tr>
<tr>
<td>( P\overline{6}<em>3\text{mc} )-LaH(</em>{10})</td>
<td>SL3</td>
<td>171</td>
<td>3.60(1)</td>
</tr>
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of the loaded sample in DACs M2, SL1, and SL1_S. Indeed, the results of the X-ray structure analysis show that the cleanest samples of (La,Y)H10 were obtained in DACs SL1, SL1_S, and M2, whereas in DACs M1 (loaded with LaY) and SL3 (loaded with LaY₄) we detected a notable amount of previously described [13] impurities: pseudocubic Imm2-YH7 and, probably, Cmcm-LaH₃ (Supporting Information Figs. S19–S20), formed as decomposition products: (La,Y)H10 $\rightarrow$ LaH₃ + YH₇.

For DAC M2_S, loaded with La₂Y/AB, the X-ray diffraction patterns (Fig. 3a,b) were identical to the one previously observed for Im3m-YH₆ [13]. However, the obtained unit cell volumes of 48.5–48.3 Å³ ($Z = 2$, 175–180 GPa) are in sharp contrast with that of YH₆ (<46 Å³, Fig. 3e) in the same pressure interval. Given that the volume of hypothetical Im3m-LaH₆ is above 50 Å³ ($Z = 2$), the resulting product (La,Y)H₆ can be described as La₂YH₁₈ (more accurately, La₀.7Y₀.3H₆, Figs. 3d and 2e) with a structure of Im3m-Y₃H₁₈ where approximately every two out of three yttrium atoms are replaced by lanthanum. At the center of DAC M2_S, at 180 GPa we also detected a tetragonal phase impurity similar to previously detected I4/mmm-YH₄ [13], but with a unit cell volume larger by 1.4 Å³/f.u. at this pressure. This enables us to assign this compound the composition La₂YH₁₂ (Fig. 3e). The significant difference between the results of syntheses in DACs M1–2, SL1, SL3, and M2_S is probably due to the lack of hydrogen in DAC M2_S, which is indicated by the presence of impurities of lower tetrahydrides (Fig. 3).

**Superconductivity**

The superconducting properties of the obtained lanthanum–yttrium hydrides were studied by measuring the temperature dependence of the electrical resistance of the samples in various current modes using the four-probe method, with and without an external magnetic field (Fig. 4). The Fm3m-(La,Y)H₁₀ polyhydrides, synthesized from La₂Y and La₄Y, have similar properties and exhibit relatively wide (15–17 K) superconducting transition with $T_C = 245–253$ K at 183–199 GPa, with the resistance drop to 0.1 mΩ (Fig. 4a,b). In several DACs we detected an additional shelf in the $R(T)$ dependence at 237 ± 5 K, possibly due to the presence of Im3m-(La,Y)H₆ formed with less hydrogen. The admixture of YH₆, which forms sometimes, corresponds to the superconducting transition at 224–226 K (Supporting Information Figs. S27 and S29).

We investigated the pressure dependence of $T_C$ for the (La,Y)H₁₀ samples obtained from La₂Y, La₃Y, and La₄Y alloys (Supporting Information Figs. S28 and S30). For the first sample (La₂Y), when pressure in the DAC decreased from 196 to 183 GPa, the critical temperature of the corresponding superhydride increased from 244.5 K to 253 K with a gradient $dT_C/dP = 0.65$ K/GPa.

![FIGURE 4](image)

**Superconducting transitions in Fm3m-(La,Y)H₁₀:** (a) Temperature dependence of the electrical resistance for the sample obtained from La₂Y. Inset: residual resistance after cooling below $T_C$. (b) Temperature dependence of the resistance for the sample obtained from La₃Y. Insets: residual resistance after cooling below $T_C$ and a photo of the DAC culet with electrodes. (c) Dependence of the electrical resistance of (La,Y)H₁₀ on the external magnetic field (0–16 T) at 186 GPa and 0.1 mA current. The critical temperatures were determined at the onset of the resistance drop. (d) Dependence of the critical current on the temperature and external magnetic field (0–16 T). The critical currents were measured near $T_C$. Inset: current–voltage characteristic near a superconducting transition. (e) Extrapolation of the upper critical magnetic field using the Werthamer–Helfand–Hohenberg theory [45] (WHH) and Ginzburg–Landau theory [46] (GL). (f) Dependence of the critical temperature $T_C$ on the applied magnetic field.
whereas for La-Y, the measured gradient was −0.13 K/GPa (Supporting Information Fig. S28a). The observed increase in the critical temperature is probably due to the near loss of dynamical stability of the $\text{Fm\overline{3}m-}\La\text{H}_{10}$ structure and increase of the electron–phonon coupling ($EPC$) coefficient. The opposite situation was observed for La$_3$Y: with an increase in pressure from 157 to 181 GPa, the critical temperature grew very slowly from 243 to $\sim$ 245 K with a slope $dT_c/dP = +0.071$ K/GPa (Supporting Information Fig. S30). Thus, the use of La$_3$Y alloy yielded the most efficient (La$_3$Y)$_{H_{10}}$ superconductor.

Measurements in external magnetic fields of 0–16 T (Fig. 4c, d, Supporting Information Figs. S27–S28) showed an almost linear $T_c(B)$ dependence with the slope $dB_{c2}/dT = -0.76$ T/K near 230–250 K (Fig. 4f). The extrapolated upper critical magnetic field $\mu_0H_{c2}(0)$ is 90–135 T, which is lower than that of YH$_{10}$ even though $T_c$ of (La,Y)$_{H_{10}}$ is notably higher (see Supporting Information) than those of YH$_{10}$. This additionally indicates a possible anomaly in the mechanism of superconductivity in $\text{Im\overline{3}m-}\La\text{H}_{10}$.

In all the studied cases the upper critical magnetic field demonstrated a linear dependence $B_{c2}(T) \sim |T-T_c|$. This allows us to extrapolate the $I_c(T)$ data to low temperatures within the single vortex model $I_c = I_{c0}(1 - |T/T_c|)^{1/2}$. The extrapolation shows that at 4.2 K, the critical current $I_c$ in the sample may reach 6 A and the critical current density $J_c$ may exceed 12 kA/mm$^2$. The extrapolation using Ginzburg–Landau model [46] ($I_c = I_{c0}(1 - |T/T_c|)^{1/2}$) gives lower values at 4.2 K: the extrapolated critical current $I_c$ in the sample can reach 1.25 A and the extrapolated critical current density $J_{c0}$ at 0 K exceeding 2500 A/mm$^2$. These values are comparable with the parameters of NbTi and YBCO [48], and slightly higher than those in recently studied $\text{Im\overline{3}m-}\La\text{H}_{10}$.[13]. Taking into account the more accurate SEM and optical measurements of the sample sizes, we can refine the estimate for $J_c$ as minimum 27.7 kA/mm$^2$ at 4.2 K (GL-model).

The first-principles analysis of the superconducting properties of the La–Y–H phases was performed within the harmonic approximation for phonons. We calculated parameters of the electron–phonon coupling and the superconducting state for a series of hexahydrides ($\text{Im\overline{3}m-}\La\text{H}_{10}$, cubic La$_2$YH$_{18}$, $\text{Pm\overline{3}m-}\La\text{H}_{12}$, and for a decahydride pseudocubic $\text{Rm\overline{3}m-}\La\text{H}_{20}$ (obtained from $\text{Im\overline{3}m-}\La\text{H}_{20}$ by the La–Y replacement) at 180 GPa using the tetrahedron method of integration over the Brillouin zone [49] (Table 2, Supporting Information Tables S7–S8). To simplify the analysis, here we discuss in detail only the results of the calculations for high-symmetry decahydride $\text{Rm\overline{3}m-}\La\text{H}_{20}$ with a regular arrangement of the lanthanum and yttrium atoms. At 180 GPa, the EPC coefficient $\lambda$ of $\text{Pm\overline{3}m-}\La\text{H}_{12}$ reaches 2.82, $\theta_{\text{Eliash}}$ is 847 K, and the critical temperature $T_c$ calculated using the Migdal–Eliashberg approach [50,52] is 223–241 K within the common range of the Coulomb pseudopotential $\mu^* = 0.15$–0.1. A decrease in the concentration of yttrium in (La,Y)H$_{10}$ leads to an increase in $T_c$ of La$_2$YH$_{18}$ and La$_2$YH$_{20}$ to 265–270 K ($\mu^* = 0.1$). Keeping in mind that the H sublattice in (La,Y)H$_{10}$ has almost the same structure as in YH$_{10}$, we can expect a negative anharmonic contribution $\Delta T_c \sim 25–30$ K, as in pure yttrium hexahydride [13]. These estimates are in good agreement with the experimentally observed critical temperature of $237 \pm 5$ K. Calculations within the Migdal–Eliashberg (ME) theory show that the expected upper critical magnetic field $\mu_0H_{c2}(0)$

```
<table>
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<tr>
<th>Parameter</th>
<th>$\text{Rm\overline{3}m-}\La\text{H}_{20}$</th>
<th>$\text{Pm\overline{3}m-}\La\text{H}_{12}$</th>
<th>$\text{Im\overline{3}m-}\La\text{H}_{20}$</th>
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<td>$\lambda$</td>
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<td>2.82</td>
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<td>$\omega_{\text{Eliash}}$</td>
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<td>847</td>
<td>1011</td>
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<td>$T_c$ (AD), K</td>
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<td>176–203</td>
<td>180–207</td>
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<tr>
<td>$T_c$ (E), K</td>
<td>281–300*</td>
<td>223–241</td>
<td>217–235</td>
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<td>$T_c$ (SCDFT), K</td>
<td>252</td>
<td>191</td>
<td>176</td>
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<td>0.8</td>
<td>1.62</td>
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<td>$\Delta(0)$, meV</td>
<td>67–71</td>
<td>53–57.5</td>
<td>49–54</td>
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<tr>
<td>$\mu_0H_{c2}(0), T$</td>
<td>99–101</td>
<td>73–77</td>
<td>98–106</td>
</tr>
<tr>
<td>$R_0 = 2\Delta(0)/\mu_0T_c$</td>
<td>5.48–5.54</td>
<td>5.5</td>
<td>5.23–5.35</td>
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* $T_c$ (E) = 266 K at $\mu^* = 0.2$. 

TABLE 2

Parameters of the superconducting state of $\text{Rm\overline{3}m-}\La\text{H}_{20}$, $\text{Pm\overline{3}m-}\La\text{H}_{12}$, and $\text{Im\overline{3}m-}\La\text{H}_{20}$ at 180 GPa calculated using the isotropic Migdal–Eliashberg equations (E) [50] and the Allen–Dynes formula (AD) [51] with $\mu^* = 0.15$–0.1.
of (La,Y)H_{6} is ~ 70–80 T and the superconducting gap is around 60 meV (Table 2, Supporting Information Table S8). We believe that the enhanced superconducting properties of (La,Y)H_{6} compared to pure YH_{6} are related to the presence of distorted \( \text{Im}^{3}m\)-LaH_{6} in the crystal structure, which results in an increase in the electron–phonon interaction coefficient from 2.24 in YH_{6} (harmonic approach [13]) to 2.41–2.82 in (La,Y)H_{6}.

Our calculations within Migdal–Eliashberg (ME) [50,52] theory show that decahydride \( \text{K}^{3}m\)-LaH_{20} has the EPC coefficient of 3.87 and \( \omega_{0/8} \approx 868 \, \text{K} \), which are comparable to the parameters of LaH_{10} [53], and result in theoretical \( T_{C}^{\text{ME}} = 281–300 \, \text{K} \) within the common range of the Coulomb pseudopotential \( \mu^{*} = 0.15–0.1 \). Because the structure of \( \text{K}^{3}m\)-LaH_{20} is similar to that of LaH_{10}, we can expect the negative anharmonic contribution \( \Delta T_{C} \) of ~ 25–35 K [34] to reduce \( T_{C} \) to about 265 K. Calculations within Migdal–Eliashberg theory show that for (La,Y)H_{10} the expected upper critical magnetic field \( \mu_{0}H_{c2}(0) = 100 \, \text{T} \), the superconducting gap is around 70 meV, and the coherence length \( \xi_{\text{cS}} = 0.5 \sqrt{\hbar / m_{e}c_{2}} \) is close to 16 Å (Supporting Information Table S8). In contrast to the \( \text{Im}^{3}m\)-XH_{6} (X = La, Y) structures, the calculations within density functional theory for superconductors (SCDFT) yield \( T_{C} = 252 \, \text{K} \) for \( \text{K}^{3}m\)-LaH_{20} at 180 GPa, which is close to both the ME theory and experimental results. Experimentally found \( T_{C} = 253 \, \text{K} \) and extrapolated \( \mu_{0}H_{c2}(0) = 100–135 \, \text{T} \) of (La,Y)H_{10} are in close agreement with the theoretically calculated values, which means that superconductivity in the La–Y–H ternary decahydrides is described well in the framework of the classical Bardeen–Cooper–Schrieffer [54,55] and Migdal–Eliashberg theories.

Conclusions

Ternary hydrides represent the most promising direction for studies of superconductivity under pressure. The primary objective in this field is to achieve room-temperature transition at the lowest possible pressure. Such superconductors, synthesized in miniature (\( \text{d}_{\text{out}} \approx 10–15 \, \text{mm} \)) high-pressure anvil cells, may find multiple applications in microelectronics. In fact, diamond anvil cell provides enough space (20x20x20 \( \mu \text{m} \)) for creating relatively complex optical and magnetic sensors using lithographic methods.

In this work, novel high-\( T_{C} \) ternary superconducting hydrides \( \text{Im}^{3}m\)-(La,Y)H_{6} and \( \text{Fm}^{3}m\)-(La,Y)H_{10} \) were experimentally discovered together with \( \text{Im}^{4}mmm\)-(La,Y)H_{4} \) at pressures of 170–196 GPa. Using the La–Y alloys (with 1:1, 2:1, 3:1, 4:1, and 1:4 ratios) as a precursor for high-pressure synthesis, we replaced about 25% of lanthanum atoms in the structure of LaH_{10} with yttrium. Moreover, we found that about 70% of the yttrium atoms in YH_{6} can be replaced by La without decomposition of the \( \text{Im}^{3}m \) sodalite-like structure of the hexahydride. In other words, superhydrides that do not exist at 180 GPa can nevertheless be stabilized as a solid solution: \( \text{YH}_{12} \) cages with the local H environment specific for \( \text{Fm}^{3}m\)-YH_{10} can exist in the \( \text{Fm}^{3}m\)-LaH_{10} environment, whereas \( \text{Im}^{3}m\)-LaH_{6} can be stabilized by introducing only 30% of yttrium.

At 183 GPa, the obtained \( \text{Fm}^{3}m\)-(La,Y)H_{10} decahydrides demonstrate outstanding \( T_{C} \) of up to 253 K, the superconducting gap of ~ 70 meV, and the extrapolated upper critical magnetic field of 100–135 T in close agreement with the calculations within ME theory. The estimated critical current density (12–27.7 kA/mm^{2}) at 4.2 K in lanthanum–yttrium decahydride may exceed that of YBCO, NbTi, and YH_{6}.

We believe the performed experiments will have a strong impact on subsequent studies of ternary metal–hydrogen systems, opening promising ways to stabilize such exotic compounds as MgH_{6} [56,57], ZrH_{10} [58,59], CaH_{6} [60], PdH_{2} [61], PdH_{6} [62], and others which cannot be obtained by direct synthesis but may exist in the form of a solid solution with known superhydrides at relatively low pressures.

Methods

Experiment

To perform the X-ray diffraction study, seven diamond anvil cells (DACs M1, M2, M2_S, SL1, SL1_S, SL3, and SL3_S) were loaded. The diameter of the working surface of the diamond anvil was 280 \( \mu \text{m} \) beveled at an angle of 8.5° to a culet of 50 \( \mu \text{m} \). Composite gaskets consisting of a tungsten ring and a CaF2/epoxy mixture were used to isolate the electrical leads. Lanthanum–yttrium pieces with a thickness of ~ 1–2 \( \mu \text{m} \) were sandwiched between the electrodes and ammonia borane NH_{3}BH_{3} (AB) in the gasket hole with a diameter of 20 \( \mu \text{m} \) and a thickness of 10–12 \( \mu \text{m} \). Laser heating of samples above 2000 K at pressures of 170–196 GPa by a series of laser pulses with a duration of 100–400 milliseconds led to the formation of ternary lanthanum–yttrium hydrides the structure of which was analyzed using X-ray diffraction. Because each laser heating and cooling cycle resulted in some pressure change in the DACs, the pressure in each experiment was determined separately. Raman and X-ray diffraction studies showed the absence of byproducts of the AB thermal decomposition in the obtained hydrides.

X-ray diffraction patterns of the samples in DACs M1-2, SL1, and SL3 were recorded at the ID15B synchrotron beamline at the European Synchrotron Radiation Facility (Grenoble, France) using a focused (5 \times 5 \( \mu \text{m} \)) monochromatic X-ray beam with a wavelength of 0.4111 Å and Mar355 detector. The exposure time was 60 s. CeO_{2} standard was used for the distance calibration. The X-ray diffraction data were analyzed and integrated using Dioptas software package (version 0.5) [63]. The full profile analysis of the diffraction patterns and the calculation of the unit cell parameters were performed in JANA2006 program [64] using the Le Bail method [65]. The pressure in the DACs was determined via the Raman signal of diamond [66].

X-ray diffraction patterns of the La\_Y\_H\_6 sample (DAC M2_S) were recorded at the BL10XU beamline (Spring-8, Japan) using monochromatic synchrotron radiation and an imaging plate detector at room temperature [67,68]. The X-ray beam with a wavelength of 0.413 Å was focused in a 3 \times 8 \( \mu \text{m} \) spot with a polymer refractive lens (SU-8, produced by ANKA).

Powder XRD experiments with DACs SL1_S and SL3_S were carried out at beamline P02.2 of PETRA III, DESY (Hamburg) [69]. The X-ray wavelength was \( \lambda = 0.2906 \, \text{Å} \), and the beam size was 2 \times 2 \( \mu \text{m} \) at full width at half maximum (FWHM). The calibration was performed using CeO_{2} standard. For data collection, a fast flat panel detector XRD1621 from Perkin Elmer (2048 pix-
els × 2048 pixels with 200 × 200 μm pixel size) was used. All (La, Y)H₅ samples were mapped using a 10 × 10 or 12 × 12 grid with a step of 2 μm, and XRD was examined for each point of this map.

The initial La-Y alloys used as the precursors for the high-pressure synthesis of ternary hydrides were fused and then characterized using XRD, energy-dispersive X-ray fluorescence (EDXRF), and scanning/transmission electron microscopy (SEM/TEM). To prepare La₂Y alloy, pure metals La and Y (99.9%, CHEMCRAFT Ltd.) were crushed, washed in dilute HCl and acetone to remove impurities, and dried in a glove box. The components were weighed and mixed in a specified ratio. Heating was carried out resistively. The melt was kept in a nitrogen atmosphere (helium) for one hour and quenched at an initial rate of 200 K/min.

XRD patterns of the La-Y alloys were recorded using the powder X-ray diffractometer Rigaku MiniFlex 600 (Rigaku, Japan) with CuKα radiation (40 kV, 15 mA, Ni-Kα-filter) in the 2θ range from 20° to 115° with a scanning step of 0.02° and a rate of 0.7°/min. The phases were identified in the PXDL program (Rigaku, Japan) using ICDD PDF-2 datasets (release 2017). The unit cell parameters were refined in JANA2006 program [64] using Le Bail method [65]. The elemental analysis of the initial La-Y alloys was performed using the energy-dispersive EDXRF in µ-XRF system ORBIS PC (EDAX, USA). The EDXRF spectra were collected under vacuum from 2 mm areas of the samples irradiated using Rh X-ray tube at 30 kV and 100 μA during 30 s.

The morphology and composition of the initial LaY alloy used as a precursor for La₂YH₅ hydride (DAC M2.S) were studied using the scanning electron microscope (SEM) Scios (ThermoFisher Scientific, USA) equipped with an energy-dispersive X-ray (EDX) spectrometer and an Octane Elite silicon drift detector (SDD) with Si₃N₄ window (EDAX, USA). The acceleration voltage was 30 kV. For the SEM and EDX studies, the chunk of the alloy was polished on one side using diamond powder with sequentially reduced particle sizes. The specimen for scanning/transmission electron microscopy (S/TEM) was prepared in the focused ion beam (FIB) scanning electron microscope Scios (ThermoFisher Scientific, USA) by a standard lift-out FIB technique at an accelerating voltage of Ga⁺ ions of 30 kV at the initial and 5 kV at the final step. Microstructural and elemental analyses were performed in an Osiris TEM/STEM (ThermoFisher Scientific, USA) equipped with the EDX Super-X SDD spectrometer (Bruker, USA), the high-angle annular dark field (HAADF) electron detector (Fischione, USA) and a 2048 × 2018 Gatan CCD (Gatan, USA). Image processing was performed using Digital Micrograph (Gatan, USA) and TIA (ThermoFisher Scientific, USA) software. Simulations of the ED patterns and images were produced using the Stadelmann’s EMS software package [70].

Magnetotransport measurements were performed on samples with at least two hydride phases, therefore the voltage contacts in the Van der Pauw method might have been connected to a low-Tc phase. As a result, the superconducting transition in the main phase (La,Y)H₁₀ can be observed as an upward feature of the resistance–temperature curves because of the shunting effect in fine-grained samples.

In Fig. S29, the plot of the normalized volume pinning force \( F_p / F_p^{\text{max}} \) versus the reduced field \( h = H / H_c^2 \) is drawn on the basis of interpolation of the \( I_c(T, B) \) data. The experimental data were fitted using the Dew-Hughes (DH) model [47] for surface type normal pinning centers \( f \sim H^0(1 – h)^q \). For this model, the parameters are: \( p = 0.5 \), \( q = 2 \), \( n_{\text{max}} = 0.2 \), which is close to our fit. Depinning critical current for this type of pinning can be described within the model of single vortices, where vortices are pinned on randomly distributed weak pinning centers via spatial fluctuations of the charge carrier mean free path, or, in other words, “dl-pinning”.

**Theory**

Computational predictions of thermodynamic stability of the La–Y–H phases at 200 GPa were carried out using the variable-composition evolutionary algorithm USPEX [27,71,72]. The first generation consisting of 120 structures was produced using the random symmetric [72] and random topological [73] generators, whereas all subsequent generations contained 20% of random structures and 80% of those created using heredity, softmutation, and transmutation operators. The evolutionary searches were combined with structure relaxations using density functional theory (DFT) [74,75] within the Perdew–Burke–Ernzerhof functional (generalized gradient approximation) [42] and the projector augmented wave method [39,76] as implemented in the VASP code [36,38,77]. The kinetic energy cutoff for plane waves was 600 eV. The Brillouin zone was sampled using \( \Gamma \)-centered K-points meshes with a resolution of \( 2\pi \times 0.05 \text{ Å}^{-1} \). This methodology is similar the one used in our previous works [78,79].

The same techniques and parameters as those used for structure relaxations were implemented to calculate the equations of state. We also calculated the phonon densities of states of the studied materials using the finite displacements method (VASP and PHONOPY [80,81]).

The calculations of the critical temperature of superconductivity \( T_c \) were carried out using Quantum ESPRESSO (QE) package [40,41]. The phonon frequencies and electron–phonon coupling (EPC) coefficients were computed using density functional perturbation theory [82], employing the plane-wave pseudopotential method and Perdew–Burke–Ernzerhof exchange-correlation functional [42]. In our ab initio calculations of the electron–phonon coupling (EPC) coefficient \( \lambda \), the first Brillouin zone was sampled using a 3 × 3 × 3 or 4 × 4 × 4 q-points mesh and a denser 16 × 16 × 16 k-points mesh for the La–Y–H phases. \( T_c \) was calculated by solving Eliashberg equations [50] using the iterative self-consistent method for the imaginary part of the order parameter \( \Delta(T, \omega) \) (superconducting gap) and the renormalization wave function \( Z(T, \omega) \) [83]. More approximate estimates of \( T_c \) were made using the Allen–Dynes formula [51].

Finally, we also solved the gap equation in density functional theory for superconductors (SCDFIT) [84,85] for evaluating \( T_c \) of Im\( \overline{3} \)-La₅Ho, LaYH₁₂, and R\( \overline{3} \)-LaYH₂₀ nonempirically:

\[
\Delta_{nk}(T) = -Z_{nk}(T)\Delta_{nk}(0) \frac{1}{2} \sum_{k'k} K_{nk'k'}(T) \frac{\tanh \frac{E_{nk}}{2T}}{E_{nk}} \Delta_{nk'}(T),
\]

where \( E_{nk} = \sqrt{\frac{\pi k_n^2}{m_n} + \Delta_{nk}^2} \). The temperature (as \( \beta = 1/T \)) dependence of the order parameter \( \Delta_{nk} \) indicates \( T_c \). Labels \( n, n', k, \) and \( k' \) denote the Kohn–Sham band and momentum indices,
**Author contributions**


**Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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**Data availability**

The raw and processed data required to reproduce these findings are available to download as Supporting Information for the manuscript.

**Appendix A. Supplementary data**

Supplementary data to this article can be found online at https://doi.org/10.1016/j.mattod.2021.03.025.

**References**
