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Prediction of high-temperature superconductivity in LaH₄ at low pressures

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Superconducting hydrides have received significant attention in the last decade due to their potential for room-temperature superconductivity. However, achieving high critical temperatures (T_c s) typically requires extreme pressures exceeding 150 GPa. Recently, a new, low-pressure $R\bar{3}m$ -LaH₄ phase was observed to form above approximately 20 GPa. Here, we perform first-principles calculations to investigate the electron–phonon interactions and superconducting properties of the new phase across a range of pressures. At the harmonic level, the system is found to be dynamically unstable, but this is remedied through the inclusion of anharmonic effects. We estimate that T_c reaches up to 115 K at 25 GPa, driven by a high density of states at the Fermi level (N_F) and soft phonon modes. However, superconductivity is suppressed with increasing pressure, as T_c rapidly decreases to 34 K at 60 GPa and 11 K at 100 GPa, due to a reduction in N_F and phonon hardening.

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1 Introduction

Room-temperature superconductivity has long been a highly sought after goal in materials science. The discovery of superconductivity in H₃S at 203 K and 155 GPa¹ marked a pivotal breakthrough, demonstrating that high- T_c superconductivity could be realized in hydrogen-rich systems under pressure. This was soon followed by the realization of near-room-temperature superconductivity in LaH₁₀ at 250 K and 170 GPa,^{2,3} firmly establishing binary hydrides as front-runners in the search for high- T_c materials. Yttrium hydrides, such as YH₆ and YH₉, have since been both predicted and experimentally confirmed to reach T_c values above 240 K at megabar pressures.^{4,5} Other binary hydrides have been experimentally reported to exhibit superconductivity, including CaH₆ (215 K at 172 GPa),^{6,7} CeH₉ (100 K at 100 GPa),^{8–10} and ThH₁₀ (161 K at 175 GPa).¹¹ These findings demonstrate that binary hydrides have a strong potential to achieve high- T_c , particularly when the hydrogen fraction in a compound exceeds 0.6.¹² The high phonon frequencies of hydrogen and the lower frequency modes from the heavier ions both couple strongly to result in significant electron–phonon (e–ph) coupling, and, therefore, T_c . Although these

superhydrides significantly reduce the pressure needed to dissociate molecular H₂ by providing a lattice structure that “chemically pre-compresses” the hydrogen,¹³ the required pressures remain prohibitively high for practical applications.

Among binary hydrides, significant research has focused on the lanthanum hydrides following the report of superconductivity in LaH₁₀.^{2,3,14–27} In this case, studies have demonstrated strong e–ph coupling,²⁰ stability at lower pressures,¹⁷ and the importance of anharmonic corrections for achieving dynamical stability.¹⁸ Recently, several lanthanum hydrides with different La : H ratios have been successfully synthesized in the pressure range of 50 to 180 GPa,²⁸ with the cubic La₄H₂₃ phase shown to exhibit superconductivity with a maximum T_c of 105 K at 118 GPa.^{29,30}

Building on these results and motivated by the need to identify hydrogen-rich compounds that can achieve high- T_c superconductivity at lower pressures, some of the present authors investigated the formation of La hydrides in cryomilled elemental lanthanum exposed to ammonia borane³¹. Structural evolution under pressures up to 60 GPa revealed a distortion from the $Fm\bar{3}m$ phase to an $R\bar{3}m$ phase, with the new rhombohedral structure becoming kinetically stable at pressures above approximately 20 GPa. Using the observed excess cell volume and comparison with the equation of state calculated *via* density functional theory (DFT) for various lanthanum hydrides, the stoichiometry of the $R\bar{3}m$ phase was determined to correspond to LaH_{*x*} with $x \sim 4$.³¹

Based on these experimental observations, we perform first-principles calculations to investigate the superconducting properties of the rhombohedral LaH₄ structure shown in Fig. 1.

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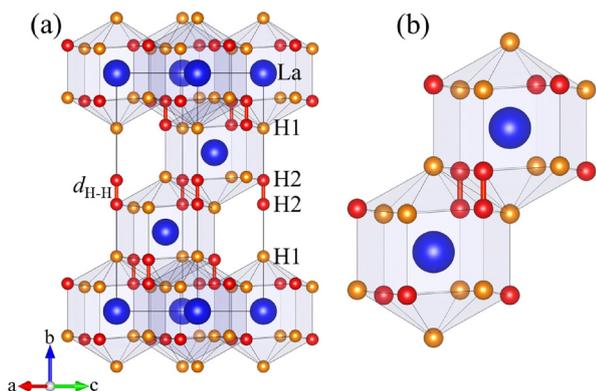


Fig. 1 Crystal structure of $R\bar{3}m$ -LaH₄ at 25 GPa. The two nonequivalent hydrogen atoms are labeled as H1 and H2 and the shortest H–H distance is indicated as $d_{\text{H-H}}$. The figures were generated with VESTA.⁴⁷

Our calculations predict a T_c reaching 115 K at 25 GPa, nearly half that of LaH₁₀^{15,19} and comparable to La₄H₂₃,²⁹ but at significantly lower pressures. For instance, a tetragonal LaH₄ phase was previously predicted to exhibit a T_c of only 10 K at 300 GPa.¹⁵ Our findings reinforce the potential of the lanthanum hydride family for the development of high-temperature superconducting materials at low to ambient pressures.

2 Methods

First principle calculations were carried out with the Quantum ESPRESSO (QE) package.^{32,33} We employed optimized norm-converging Vanderbilt pseudopotentials (ONCVSP)³⁴ from the Pseudo Dojo library³⁵ generated with the scalar-relativistic revised Perdew–Burke–Ernzerhof (PBEsol) parametrization.³⁶ A plane wave kinetic-energy cutoff of 120 Ry for the wavefunctions, and 480 Ry for the charge density and potential, were used. For the Brillouin-zone integration of the 5-atom unit cell, we used a Γ -centered $12 \times 12 \times 12$ \mathbf{k} -mesh³⁷ with a Methfessel–Paxton³⁸ smearing of 0.02 Ry. The atomic positions and lattice parameters were optimized until the total energy was converged within 10^{-6} Ry and the force on each atom was less than 10^{-4} Ry \AA^{-1} . The dynamical matrices and the linear variation of the self-consistent potential were first computed using density-functional perturbation theory (DFPT)³⁹ on the irreducible set of a regular $6 \times 6 \times 6$ \mathbf{q} -mesh.

The force constants computed by DFPT were used as a starting point to calculate phonon anharmonicity *via* the anharmonic special displacement method (ASDM)^{40–42} within the framework of the self-consistent phonon theory.⁴³ The ASDM is used as implemented in the Zacharias–Giustino (ZG) code of the EPW package.⁴⁴ For our ASDM calculations, we employed finite differences of amplitude 0.2 \AA to iteratively compute the interatomic force constants of ZG configurations at 300 K in $2 \times 2 \times 2$ supercells. We find that such large displacements in low-symmetry systems involving light-mass atoms improve numerical stability and provide a better representation of the anharmonic behavior, particularly for capturing higher-order contributions to the effective force constants. In contrast to high-symmetry

hydrides, such as the $Im\bar{3}m$ phase of H₃S, where small 0.01 \AA displacements work effectively,⁴⁵ such small displacement values lead to phonon instabilities in the soft-mode regions near Γ , as shown in Fig. S1.⁴⁶ With the aid of iterative mixing,⁴² the convergence of the interatomic force constants was achieved at the 10th iteration, where three successive iterations showed little, to no, change in the anharmonic phonon dispersion. The converged anharmonic force constants were then used to construct the dynamical matrix *via* standard Fourier interpolation on a $6 \times 6 \times 6$ \mathbf{q} -mesh.

The EPW^{44,48–50} code was employed to investigate the e–ph interactions and superconducting properties. The electronic wavefunctions required for the Wannier–Fourier interpolation^{51,52} were calculated on a Γ -centered $12 \times 12 \times 12$ \mathbf{k} -mesh. Nine maximally localized Wannier functions (five d orbitals for La and one s orbital for each H) were used to describe the electronic structure. The anisotropic full-bandwidth Migdal–Eliashberg equations^{44,53} were solved with a sparse intermediate representation of the Matsubara frequencies⁵⁴ on fine uniform $80 \times 80 \times 80$ \mathbf{k} - and $40 \times 40 \times 40$ \mathbf{q} -grids with an energy window of ± 0.2 eV around the Fermi level. The semiempirical Coulomb parameter, μ^* , was varied from 0.1–0.2.

3 Results and discussion

Guided by experiments, we consider the $R\bar{3}m$ rhombohedral phase (space group no. 166) for the LaH₄ structure. Fig. 2(a) shows our X-ray Diffraction (XRD) results for cryomilled La mixed with ammonia borane, measured in a diamond anvil cell at a pressure of 29.1 GPa. The XRD data reveal the co-existence of $R\bar{3}m$ and FCC La-hydride phases. Fig. 2(b) shows the cell volume of La-hydride inferred from XRD data as a function of pressure. The experimental cell volume is systemically larger than that of the FCC LaH₃ phase over the investigated pressure range.

Utilizing the experimentally determined unit cell and positions of the La atoms, the positions of the H atoms were derived in ref. 31 based on symmetry considerations and atomic relaxation using DFT. The enthalpy of the predicted rhombohedral $R\bar{3}m$ LaH₄ structure was found to be lower by about 0.2 eV f.u.^{−1} than that of several alternative, symmetry-broken LaH₄ configurations obtained *via ab initio* molecular dynamics followed by atomic relaxation. On the other hand, the $I4/mmm$ proposed in ref. 15 remains thermodynamically more favorable than the $R\bar{3}m$ phase in the considered pressure range, as shown in Fig. S2.⁴⁶ While the $R\bar{3}m$ phase is not the thermodynamic ground state, our theoretical calculations (including anharmonic effects) indicate that it is dynamically stable, meaning it is kinetically stable in the limit of low temperature. Our experimental data corroborate this, suggesting that the $R\bar{3}m$ phase is observable (thermodynamically metastable) at room temperature. This indicates that it is even more likely to be observable (kinetically stabilized) at low temperatures, particularly near the predicted transition temperature T_c . A similar kinetics-based stabilization mechanism has been shown to overcome the highly unfavorable thermodynamics of Alane at room temperature.⁵⁵



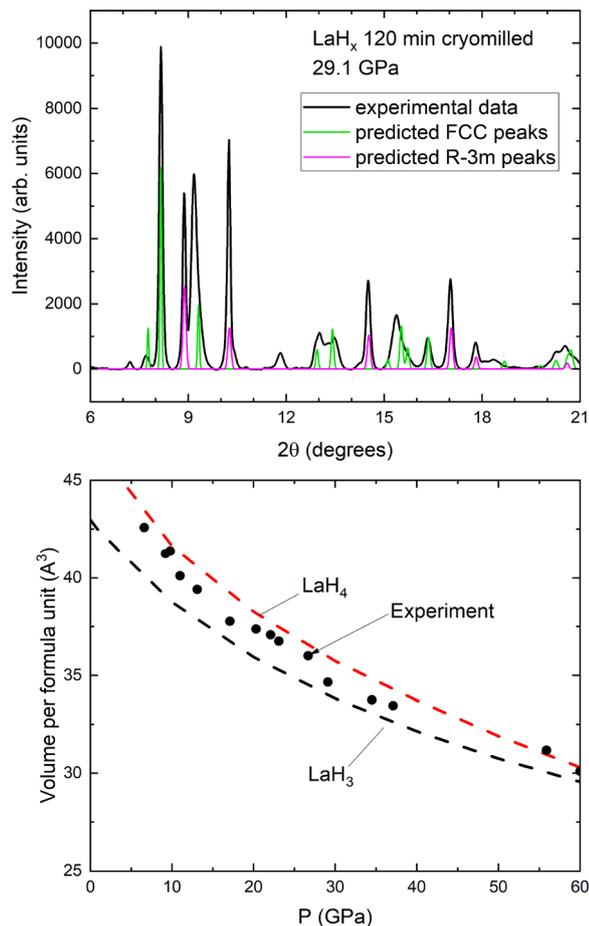


Fig. 2 (a) Powder X-ray diffraction data for cryomilled La mixed with ammonia borane, showing the co-existence of $R\bar{3}m$ and FCC La-hydride phases under high pressure (29.1 GPa). (b) The cell volume of the La-hydride phase inferred from X-ray diffraction data as a function of pressure. DFT simulations of the equation of state are shown for the proposed $R\bar{3}m$ LaH_4 phase and the FCC LaH_3 phase, as indicated. The data presented in this figure are from ref. 31.

The $R\bar{3}m$ - LaH_4 phase is predicted to adopt a clathrate structure that is characteristic of other high-temperature superconducting hydrides.^{7,18,56–63} The structure consists of a La atom occupying the 3a (0, 0, 0) Wyckoff site and two nonequivalent H atoms, labeled as H1 and H2, located at 6c (0, 0, z_1) and 6c ($1/3$, $2/3$, z_2) Wyckoff sites. The calculated structural parameters at 25, 60, and 100 GPa are provided in Table 1. Each La atom is enclosed in a cage formed by 14 H atoms, as seen in Fig. 1. This matches the crystal structure of $R\bar{3}m$ - TeH_4 predicted to stabilize above 230 GPa and reach a T_c of about 70 K.⁶⁴ The H-hexagonal rings are slightly buckled at 25 GPa (~ 0.1 Å) but, as in $R\bar{3}m$ - TeH_4 , become nearly flat at higher pressures. The H1 atoms form a H_2 unit as marked by the red bond in Fig. 1. This bond marks the shortest H–H distance in the structure, with a value $d_{\text{H-H}}$ of 0.96, 0.92, and 0.88 Å for 25, 60, and 100 GPa, respectively. These bond length values are close to the H–H distance of 1 Å predicted for metallic hydrogen near 500 GPa⁶⁵ and 0.86 Å for TeH_4 at 300 GPa,⁶⁴ but slightly smaller than 1.1 Å for LaH_{10} in the 150–300 GPa pressure range^{15,66} and 1.3 Å for La_4H_{23} at 118 GPa.²⁹

Table 1 Calculated lattice parameters, shortest H–H distance, and atomic coordinates for the conventional cell of LaH_4 at 25, 60, and 100 GPa. The structure adopts the $R\bar{3}m$ space group with $\gamma = 120^\circ$ at all pressures

Pressure (GPa)	$a = b$ (Å)	c (Å)	$d_{\text{H-H}}$ (Å)	Atomic coordinates (fractional)			
25	3.54	9.87	0.96	La (3a)	0.000	0.000	0.000
				H1 (6c)	0.000	0.000	0.231
				H2 (6c)	0.333	0.666	0.217
60	3.20	9.89	0.92	La (3a)	0.000	0.000	0.000
				H1 (6c)	0.000	0.000	0.215
				H2 (6c)	0.333	0.666	0.213
100	2.99	9.78	0.88	La (3a)	0.000	0.000	0.000
				H1 (6c)	0.000	0.000	0.209
				H2 (6c)	0.333	0.666	0.212

The electronic band structure and density of state (DOS) plots in Fig. 3 indicate that the $R\bar{3}m$ - LaH_4 phase is metallic across all investigated pressures. The DOS at the Fermi level (N_{F}) comes from a combination of La and H states with La boasting about 50%, 68%, and 81% increase over H states at 25, 60, and 100 GPa. For hydrogen, the unit formed by the vertically adjacent H2 atoms, as seen in Fig. 1, account for the bulk of the hydrogen states at the Fermi level, contributing 86%, 82%, and 81% of the total hydrogen states at the Fermi level, as depicted in the second panel of Fig. 3. At 25 GPa, N_{F} reaches its highest value of 0.81 states per eV f.u.⁻¹, comparable to values reported

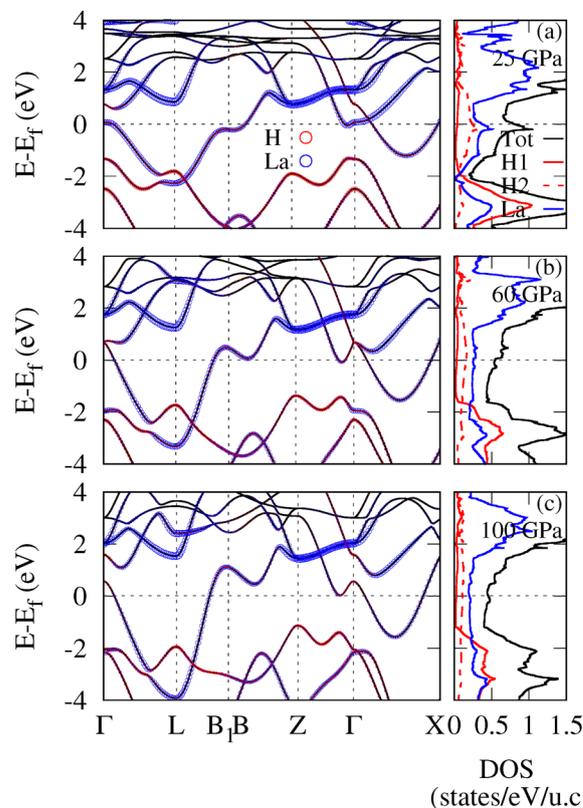


Fig. 3 Electronic band structure with orbital characters, and total and projected density of states (DOS) of LaH_4 at (a)–(c) 25, 60, and 100 GPa. The orbital character is shown in red for H and blue for La states. The total DOS is shown in black, while the projected DOS corresponding to H1, H2, and La atoms are shown with red, dashed red, and blue lines, respectively.



for other high- T_c La-H phases (e.g., ~ 0.93 states per eV f.u. $^{-1}$ for LaH₁₀ at 150 GPa¹⁸ and 0.79 states per eV per La for La₄H₂₃ at 120 GPa²⁹). This high DOS is largely attributed to a flat electronic band around the Γ point. Furthermore, approximately 0.2 eV below the Fermi level, a Van-Hove-like singularity emerges due to another flat band along the L-B₁ and B-Z high-symmetry directions. The presence of these dispersionless bands at 25 GPa induces a significant variation in the DOS in the vicinity of E_F that is captured by the full-bandwidth approach used to solve the superconducting Migdal-Eliashberg equations.⁵³ At higher pressures, the flat bands evolve into parabolic dispersions, resulting in a marked decrease in N_F by approximately 33% at 60 GPa and 46% at 100 GPa, respectively. To note, the effect of spin-orbit coupling on the electronic structure was considered and found to be negligible, as shown in Fig. S3.⁴⁶

To further analyze the electronic structure, we calculated the charge density difference between the LaH₄ crystal structure and the sum of its constituent atoms at 25 and 100 GPa. As shown in Fig. 4(a) and (b), there is a buildup of charge on the hydrogen

atoms, but, surprisingly, no charge depletion around the La atoms. This suggests minimal to no charge transfer between La and H atoms. Meanwhile, charge depletion is observed between two vertically adjacent H2 atoms, becoming significantly more pronounced at 100 GPa. This trend indicates the formation of H₂ units at higher pressure, resembling molecular hydrogen. This picture is further supported by the electron localization function (ELF) plots in Fig. 4(c) and (d), taken along the (1 1 0) and (0 0 1) planes, respectively. Panel (c) reveals the formation of H₂ units between adjacent H2 atoms, as well as the repulsion between La and H1 atoms, which gives rise to the teardrop-shaped features around the H1 atoms. Panel (d) illustrates how the H atoms arrange into a hexagonal ring surrounding the La atom, highlighting the structural characteristics of $R\bar{3}m$ -LaH₄.

Fig. 5 presents the vibrational properties of LaH₄. At the harmonic level, depicted by the solid gray lines, the structure exhibits dynamic instabilities across all investigated pressures, with the most pronounced negative phonon modes observed at 25 GPa. This behavior is common among many superconducting hydrides and is typically resolved by accounting for quantum anharmonic effects.^{18,68-71} When applying the ASDM, these instabilities are eliminated, as shown by the solid black lines. Notably, the low-frequency La modes at 25 GPa undergo significant renormalization under the ASDM, which has important implications for the superconducting properties of the material.

The phonon dispersion displays a distinct separation between modes associated with lanthanum and hydrogen.

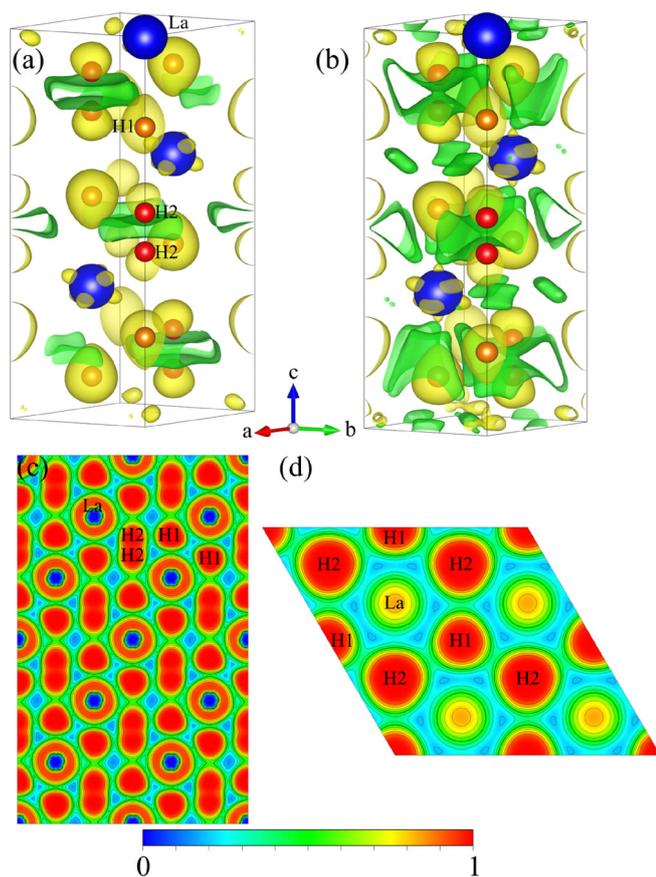


Fig. 4 Charge density difference between the LaH₄ crystal structure and the sum of its constituent atoms at (a) 25 GPa and (b) 100 GPa. The yellow and green colors represent charge accumulation and depletion regions, respectively, with an isosurface value set to $7 \times 10^{-2} \text{ e } \text{\AA}^{-3}$. Electron localization function (ELF) for LaH₄ at 25 GPa taken along the (c) (1 1 0) and (d) (0 0 1) Miller planes, respectively, with a contour spacing set to 0.1. All plots were generated with VESTA.⁴⁷

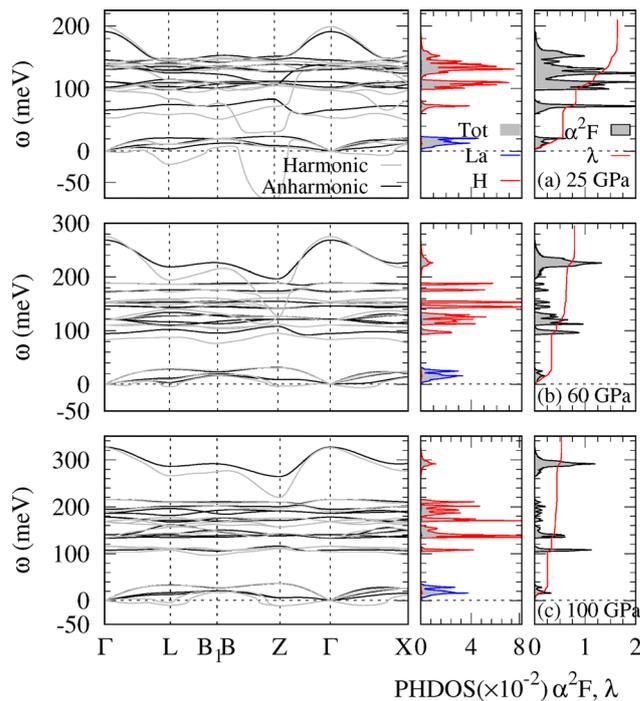


Fig. 5 (a)–(c) Harmonic and anharmonic phonon dispersion, phonon density of states (PHDOS), isotropic Eliashberg spectral function (α^2F), and electron-phonon coupling strength (λ) of LaH₄ at 25, 60, and 100 GPa, respectively. The total PHDOS is decomposed with respect to the vibrations of H (red) and La (blue) atoms. The anharmonic phonon dispersions were computed for a temperature of 300 K.



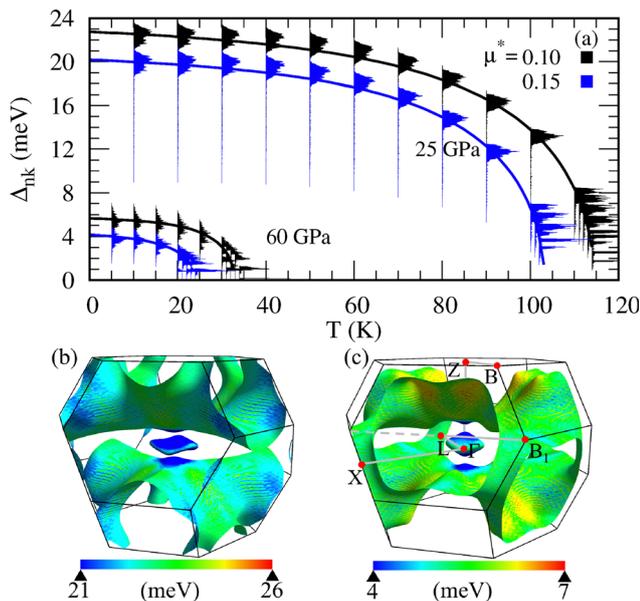


Fig. 6 (a) Histograms of the energy-dependent distribution of the superconducting gap Δ_{nk} as a function of temperature in LaH₄ at 25 and 60 GPa with $\mu^* = 0.10$ (black) and $\mu^* = 0.15$ (blue). The solid lines are guides to the eye. (b) and (c) Momentum-resolved superconducting gap on the Fermi surface in LaH₄ at 25 and 60 GPa with $\mu^* = 0.10$, generated with FermiSurfer.⁶⁷

The acoustic modes arise solely from the vibration of the La atoms, while the intermediate- and high-frequency modes originate exclusively from the motion of the H atoms. From the comparison of the Eliashberg spectral function (α^2F) and the phonon density of states (PHDOS) in the middle and right panels of Fig. 5, we can see that most of the e–ph coupling comes from the hydrogen phonon modes. At 25 GPa, for example, 66% of the total e–ph coupling constant λ of 1.64 comes from the H modes. This λ value is comparable to those computed for LaH₁₀ (1.78 and 1.86 at 300 GPa^{15,19}) and La₄H₂₃ (1.49 at 100 GPa⁷²), and almost four times larger than that of a predicted tetragonal LaH₄ phase (0.43 at 300 GPa¹⁵). As pressure increases, the phonon frequencies harden across all regions, and the DOS at the Fermi level decreases. These combined effects lead to a reduction in λ , with values of 0.79 and 0.53 at 60 GPa and 100 GPa, respectively. A similar pressure dependence has been observed in LaH₁₀ and La₄H₂₃ superconductors.^{3,15,29}

To understand the superconducting properties, we solved the anisotropic full-bandwidth Migdal–Eliashberg equations for a Coulomb parameter μ^* in the 0.10–0.20 range, consistent with values used in other studies on superconductivity in lanthanum hydrides.^{15,19,29} Fig. 6 presents the energy distribution of the superconducting gap, Δ_{nk} , as a function of temperature along with its momentum-resolved k -dependence on the Fermi surface at select pressure and μ^* values. Our calculations reveal that LaH₄ exhibits a single anisotropic gap, with an estimated T_c^{aME} of 115 K for $\mu^* = 0.10$ at 25 GPa. In comparison, the McMillan (McM) equation,⁷³ the Allen–Dynes (AD) formula,⁷⁴ the machine-learned (ML) SISSO model,⁷⁵ and the isotropic Migdal–Eliashberg (iME) formalism yield $T_c^{\text{McM}} = 72.5$ K, $T_c^{\text{AD}} = 85.4$ K, $T_c^{\text{ML}} = 102.7$ K, and $T_c^{\text{iME}} = 117$ K, respectively. To put this into context, T_c^{iME} was predicted to be 254 K at 300 GPa in LaH₁₀,¹⁵ 95 K at 100 GPa in La₄H₂₃,²⁹ and only 10 K at 300 GPa in a tetragonal LaH₄ phase¹⁵ for $\mu^* = 0.10$. The estimated T_c^{aME} in $R\bar{3}m$ -LaH₄ is less than half that of LaH₁₀ but slightly higher than that in La₄H₂₃, and more importantly, it could be achieved at significantly lower pressures than in these cases. The critical temperature decreases significantly with pressure, with T_c^{aME} dropping to 33 K at 60 GPa and 14 K at 100 GPa. Results for different μ^* values and various approaches for calculating the critical temperature are summarized in Table 2.

As mentioned above, the rapid decrease in T_c with increasing pressure can be directly attributed to a reduction in the DOS at the Fermi level and the hardening of the phonon frequencies. Delving deeper, superconducting hydrides are designed to provide a lattice that promotes the dissociation of molecular H₂, facilitating the formation of metallic hydrogen under pressure.¹³ Consequently, a reduction in the H–H distance within the H₂ units may suppress superconductivity, aligning with our observations. As summarized in Table 1, the highest T_c of 115 K is achieved at 25 GPa, where the shortest H–H distance is $d_{\text{H–H}} = 0.96$ Å. However, T_c drops sharply as pressure increases, reaching just 11 K at 100 GPa where $d_{\text{H–H}} = 0.88$ Å. For reference, the bond length of a H₂ molecule is approximately 0.74 Å,⁷⁶ while the H–H distance in metallic hydrogen is predicted to be around 1 Å at 500 GPa.⁶⁵ This trend, along with the increased charge depletion between the H₂ units at higher pressures, suggests that hydrogen may transition toward a

Table 2 Properties of LaH₄ at 25, 60, and 100 GPa: density of states at the Fermi level (N_F), logarithmic average phonon frequency (ω_{log}), total electron–phonon coupling strength (λ), semiempirical Coulomb parameter (μ^*), and superconducting critical temperature (T_c). The T_c values are calculated using different methodologies: McMillan equation (T_c^{McM}), Allen–Dynes formula (T_c^{AD}), machine-learned SISSO mode (T_c^{ML}), isotropic Migdal–Eliashberg formalism (T_c^{iME}), and anisotropic Migdal–Eliashberg formalism (T_c^{aME})

Pressure (GPa)	N_F (states per eV f.u. ^{−1})	ω_{log} (meV)	λ	μ^*	T_c^{McM} (K)	T_c^{AD} (K)	T_c^{ML} (K)	T_c^{iME} (K)	T_c^{aME} (K)
25	0.81	50.6	1.64	0.10	72.5	85.4	102.7	117	115
				0.15	62.8	71.5	85.1	103	102
				0.20	53.1	59.1	68.7	93	92
				0.10	26.0	27.4	25.3	29	34
60	0.54	48.7	0.79	0.15	17.8	18.6	16.2	21	24
				0.20	10.9	11.2	10.3	16	15
				0.10	9.8	10.1	7.3	9	11
				0.15	4.4	4.5	3.0	9	9
100	0.44	56.1	0.53	0.20	1.3	1.3	0.9	—	6



molecular H₂-like state rather than a metallic phase under compression.

4 Conclusion

We carried out an *ab initio* study to explore the superconducting potential of a new low-pressure rhombohedral LaH₄ phase. Our results show that the inclusion of anharmonic corrections is crucial for stabilizing the crystal structure, highlighting the importance of these effects when investigating lanthanum hydrides under pressure. Furthermore, we find that the superconducting temperature can reach up to 115 K at 25 GPa but decreases rapidly with increasing pressure, a common trend in hydride superconductors. Besides experimentally confirming superconductivity of the rhombohedral phase in the 25–100 GPa range, it would be interesting to explore whether this phase could be stabilized down to ambient pressure. These results are exciting as they contribute to the ongoing search for hydrogen-rich compounds capable of achieving high-temperature superconductivity at lower pressures.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article is included in the manuscript.

Supplementary information is available. See DOI: <https://doi.org/10.1039/d5tc02600h>.

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