Quantum crystal structure in the 250-kelvin superconducting lanthanum hydride

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The discovery of superconductivity at 200 kelvin in the hydrogen sulfide system at high pressures¹ demonstrated the potential of hydrogen-rich materials as hightemperature superconductors. Recent theoretical predictions of rare-earth hydrides with hydrogen cages^{2,3} and the subsequent synthesis of LaH₁₀ with a superconducting critical temperature (T_c) of 250 kelvin^{4,5} have placed these materials on the verge of achieving the long-standing goal of room-temperature superconductivity. Electrical and X-ray diffraction measurements have revealed a weakly pressure-dependent T_c for LaH₁₀ between 137 and 218 gigapascals in a structure that has a face-centred cubic arrangement of lanthanum atoms⁵. Here we show that quantum atomic fluctuations stabilize a highly symmetrical $Fm\overline{3}m$ crystal structure over this pressure range. The structure is consistent with experimental findings and has a very large electronphonon coupling constant of 3.5. Although ab initio classical calculations predict that this $Fm\overline{3}m$ structure undergoes distortion at pressures below 230 gigapascals^{2,3}, yielding a complex energy landscape, the inclusion of quantum effects suggests that it is the true ground-state structure. The agreement between the calculated and experimental T_c values further indicates that this phase is responsible for the superconductivity observed at 250 kelvin. The relevance of quantum fluctuations calls into question many of the crystal structure predictions that have been made for hydrides within a classical approach and that currently guide the experimental quest for room-temperature superconductivity⁶⁻⁸. Furthermore, we find that quantum effects are crucial for the stabilization of solids with high electron-phonon coupling constants that could otherwise be destabilized by the large electron-phonon interaction⁹, thus reducing the pressures required for their synthesis.

The potential of metallic hydrogen as a high-T_c superconductor¹⁰ was identified shortly after the development of Bardeen-Cooper-Schrieffer theory, which explains superconductivity using the electron-phonon coupling mechanism. The main argument in favour of metallic hydrogen was that T_c can be maximized for light compounds owing to their high vibrational frequencies. Because high pressures are required to metallize hydrogen¹¹, chemical precompression with heavier atoms^{12,13} was suggested as a pathway by which to decrease the pressure needed to reach metallicity and, therefore, superconductivity. These ideas have been realised using modern ab initio crystal structure prediction methods based on density functional theory (DFT)^{7,14,15}. Hundreds of hydrogen-rich compounds have been predicted to be thermodynamically stable at high pressures, and their T_c values have been estimated by calculating the electron-phonon interaction^{6,7}. The success of this co-operation between DFT crystal-structure predictions and T_c calculations was exemplified by the discovery of superconductivity in H₃S at 200 K^{1,16,17}. The prospects for discovering hydrogen-based high- T_c superconductors in the near future are therefore high, with rare-earth hydrides with sodalite-like clathrate structures showing particular promise^{2,3}. This is in clear contrast to other high- T_c superconducting families such as cuprates or pnictides, in which the lack of a clear understanding of the superconducting mechanism hinders an in silico-guided approach.

DFT predictions of the La–H system proposed LaH₁₀ to be thermodynamically stable against decomposition at high pressures². A sodalite-type structure with space group $Fm\bar{3}m$ and $T_c \approx 280$ K at pressures greater than around 220 GPa was suggested as a candidate for high- T_c superconductivity^{2,3} (Fig. 1). Distorted versions of the $Fm\bar{3}m$ structure with space group C2/m and a rhombohedral lanthanum sublattice were also discussed¹⁸, and shortly after the first predictions^{2,3}, a lanthanum superhydride was synthesized by heating a lanthanum sample with a laser in a hydrogen-rich atmosphere inside a diamond anvil cell (DAC)¹⁹.

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Fig. 1 | Quantum effects stabilize the symmetric $Fm\overline{3}m$ phase of LaH₁₀. a, Enthalpy as a function of pressure for different structures of LaH₁₀, neglecting zero-point energy in the calculations. Here, pressure is calculated classically from $V(\mathbf{R})$, neglecting quantum effects on $V(\mathbf{R})$. **b**, Left, a Born-Oppenheimer energy surface V(R), exemplifying the presence of many local minima belonging to distorted structures. R represents the positions of atoms treated classically as simple points. Right, the configurational energy surface $E(\mathcal{R})$, including quantum effects. R represents the quantum centroid positions, which determine the centre of the ionic wave functions-that is, the average atomic positions. By including quantum effects, all phases collapse to a single phase: the highly symmetric $Fm\overline{3}m$

On the basis of the unit cell volume obtained by X-ray diffraction, the hydrogen-to-lanthanum ratio was estimated to be between 9 and 12. The lanthanum atoms adopted a face-centred cubic (fcc) arrangement at pressures greater than about 160 GPa, whereas at lower pressures the lattice was rhombohedral with a lanthanum sublattice of the $R\overline{3}m$ space group. Owing to the small X-ray cross-section of hydrogen, it is not experimentally possible to resolve the hydrogen sublattice directly. More recently, evidence of a superconducting transition at 260 K and 188 GPa was reported in a lanthanum superhydride^{4,20}. These findings were subsequently confirmed by the measurement of a T_c of 250 K from 137 to 218 GPa in a structure with an fcc arrangement of the lanthanum atoms, suggesting a LaH₁₀ stoichiometry⁵.

Although it is tempting to assign the superconductivity at 250 K to the previously predicted $Fm\overline{3}m$ phase²⁻⁵, there is a clear problem: the $Fm\overline{3}m$ structure is predicted to be dynamically unstable over the whole pressure range in which a 250 K T_c has been observed. This would imply that the $Fm\overline{3}m$ phase is not a minimum of the Born-Oppenheimer energy surface, and consequently a T_c has not been estimated for this phase in the experimental pressure range. The contradiction between the observation of superconductivity and the predicted instability of the $Fm\overline{3}m$ phase may indicate a problem with the classical treatment of the atomic vibrations in the calculations. Considering that quantum proton fluctuations symmetrize hydrogen bonds in the high-pressure X phase of ice²¹ and in $H_3S^{22,23}$, a similar situation is expected in La H_{10} . Here we show how quantum atomic fluctuations completely reshape the energy landscape by removing classical local minima, rendering the $Fm\overline{3}m$ phase the true ground state and the state responsible for the observed superconducting critical temperature.

We start by using DFT to calculate the lowest-enthalpy structures of LaH₁₀ as a function of pressure, using state-of-the-art methods for the prediction of crystal structure²⁴. The contribution associated with atomic fluctuations is not included, so that the energy corresponds solely to the Born–Oppenheimer energy $V(\mathbf{R})$, where \mathbf{R} represents the position of atoms treated classically as simple points. As shown in Fig. 1, different distorted phases of LaH₁₀ are thermodynamically more stable

than the $Fm\bar{3}m$ phase. At pressures greater than about 250 GPa, all phases merge to the $Fm\bar{3}m$ symmetric phase. These results are in agreement with previous calculations², even though we identify other possible distorted structures with lower enthalpy—such as the $R\bar{3}m$, C2 and P1 (not shown) phases. These phases feature distortion not only in the position of the hydrogen atoms but also in the lanthanum sublattice, leading to a non-fcc arrangement that should be detectable by X-ray analysis (see Extended Data Fig. 1). The fact that many structures are predicted emphasizes that the classical V(**R**) energy surface has a multifunnel structure that is tractable to many different saddle and local minima, as shown in Fig. 1.

This picture completely changes when we include the energy of quantum atomic fluctuations-the zero-point energy. We calculate the zero-point energy within the stochastic self-consistent harmonic approximation (SSCHA)^{25,26}. The SSCHA is a variational method that calculates the energy of the system $(E(\mathcal{R}))$ including atomic quantum fluctuations as a function of the centroid positions \mathcal{R} , which determine the centre of the ionic wave functions. The calculations are performed without approximating $V(\mathbf{R})$, keeping all of its anharmonic terms. We perform a minimization of $E(\mathcal{R})$ and determine the centroid positions at its minimum. By calculating the stress tensor from $E(\mathcal{R})$ (ref.²⁶), we relax the lattice parameters in order to find structures with isotropic stress conditions considering quantum effects. We start the quantum relaxation for both $R\overline{3}m$ and C2 phases with the lattice that yields a classical isotropic pressure of 150 GPa and vanishing classical forcesthat is, calculated from $V(\mathbf{R})$. All quantum relaxations quickly evolve into the $Fm\overline{3}m$ phase (Extended Data Fig. 4). This suggests that the quantum energy $(E(\mathcal{R}))$ landscape is much simpler than the classical $V(\mathbf{R})$ landscape, as shown in Fig. 1, and that the ground state of LaH₁₀ over the pressure range of interest is the $Fm\overline{3}m$ phase with sodalite-type symmetry. The quantum effects are substantial, reshaping the energy landscape and stabilizing structures by more than 60 meV per LaH₁₀.

Our results further confirm that the $Fm\bar{3}m$ phase of LaH₁₀ is responsible for the superconductivity at 250 K. This is consistent with the fcc



Fig. 2| **The phonon band structure of** $Fm\overline{3}m$ LaH₁₀ at different pressures. The harmonic phonons show large instabilities in several regions of the Brillouin zone. Only at the high-pressure limit–for example, at pressures greater than 220–250 GPa– is dynamic (harmonic) stabilization reached. The anharmonic phonons obtained from the Hessian of the quantum energy $E(\mathcal{R})$ within the SSCHA are dynamically stable over the experimentally relevant pressure range.

arrangement of lanthanum atoms that is found experimentally⁵. We verified that the experimental resolution reported⁵ is sufficient to discard the classically obtained distorted structures (Extended Data Fig. 1). However, another study¹⁹ observed a rhombohedral distortion at pressures lower than about 160 GPa, in which the lanthanum sublattice occupied the $R\overline{3}m$ space group and the rhombohedral angle was approximately 61.3° ($c/a \approx 2.38$ in the hexagonal representation). Our calculations show that this distortion is compatible with the hypothesis of slight anisotropic stress, which could be present in some experiments inside the DAC. Indeed, by performing an SSCHA minimization for the $R\overline{3}m$ phase but keeping the rhombohedral angle fixed at 62.3° (the value that yields an isotropic pressure of 150 GPa at the classical level), the quantum stress tensor shows a 6% anisotropy between the diagonal direction and the perpendicular plane. This suggests that anisotropic conditions inside the DAC can produce the $R\overline{3}m$ phase, although we cannot rule out the possibility that other experimental stress conditions could favour other crystalline phases.

The phonon spectra of the $Fm\bar{3}m$ phase, calculated in the harmonic approximation from the Hessian of $V(\mathbf{R})$, show clear phonon instabilities in a broad region of the Brillouin zone (Fig. 2). These instabilities appear at pressures lower than about 230 GPa, which is consistent with the finding that many possible atomic distortions lower the enthalpy of this composition at these pressures. Conversely, when the calculation is performed using the Hessian of $E(\mathcal{R})$ (ref.²⁵)–which effectively

In the case of deuterium, an instability develops at low pressures (126 GPa), which is consistent with experimental evidence. The pressure stated corresponds to that calculated from $E(\mathcal{R})$, which considers quantum effects. The grey shading marks the regions with imaginary phonon frequencies, which are depicted as negative frequencies.

captures the full anharmonicity of $V(\mathbf{R})$ —no phonon instability is observed (Fig. 2). This again confirms that the $Fm\overline{3}m$ phase is a minimum in the quantum-energy landscape over the whole pressure range in which superconductivity at 250 K was observed. Whereas the $Fm\overline{3}m$ phase of LaH₁₀ remains a minimum of $E(\mathcal{R})$ at pressures as low as 129 GPa, instabilities are seen at 126 GPa in the case of LaD₁₀. This implies that—at this pressure—the $Fm\overline{3}m$ phase of LaD₁₀ distorts to a new phase, as has been suggested previously⁵.

The breakdown of the classical harmonic approximation for phonons hinders the estimation of T_c at pressures lower than around 230 GPa in the $Fm\overline{3}m$ phase. It also calls into question the certainty of harmonic calculations at higher pressures^{2,27}, considering that large anharmonic effects are persistent well above 260 GPa (Fig. 2). However, with anharmonic phonons derived from the Hessian of $E(\mathcal{R})$, we can readily calculate the electron-phonon interaction and the superconducting T_c over the experimental pressure range (137–218 GPa). T_c is estimated fully ab initio-without any empirical parameter-by solving Migdal-Eliashberg equations and applying superconducting DFT (SCDFT). As shown in Fig. 3, the numerical solutions of Migdal-Eliashberg equations with an anisotropic energy gap match well with the experimental values. The values obtained from SCDFT calculations systematically show a slightly lower T_c . Our reported values of T_c provide evidence for the phonon-driven mechanism of superconductivity, and confirm that the $Fm\overline{3}m$ structure of LaH₁₀ is responsible for what is, to our knowledge,



Fig. 3 | **Summary of experimental and theoretical** T_c **values.** Superconducting critical temperatures calculated using anisotropic Migdal–Eliashberg equations and SCDFT. In both cases the anharmonic phonons obtained with the SSCHA were used. The results are compared with the experimental values measured in refs. ^{4,5}. The error bars in the data from ref. ⁵ correspond to the experimental uncertainty in the determination of T_c .

the highest T_c reported to date. Our calculations for LaD₁₀ in the $Fm\overline{3}m$ phase are also in agreement with the experimental data point reported. Despite the presence of large anharmonic effects, the isotope coefficient $\alpha = -[\ln T_c(\text{LaD}_{10}) - \ln T_c(\text{LaH}_{10})]/\ln 2$ is close to 0.5 (0.43 at around 160 GPa)-as expected by Bardeen-Cooper-Schrieffer theory-and is in agreement with the experimentally reported value of $\alpha = 0.46$.

Finally, we also calculated T_c in the presence of the subtle rhombohedral distortion that can be induced in experiments by anisotropic pressure conditions. When the rhombohedral angle is fixed at 62.3°, the T_c obtained for the $R\bar{3}m$ phase at 160 GPa is 9% lower than for the $Fm\bar{3}m$ phase. The observed weak pressure dependence of T_c is therefore consistent with the absence of a rhombohedral distortion, as suggested by the X-ray data⁵. However, as argued above, undesired anisotropic stress conditions in the DAC can induce phase transitions. For cases in which measurements of T_c have yielded lower values (around 200 K), it is highly probable that the corresponding structures are distorted as a result of anisotropic pressure conditions. We can also safely rule out the possibility that compositions such as LaH₁₁– which are proposed to have a high superconducting critical temperature⁵–are responsible for T_c values in the range observed here (Extended Data Fig. 8).

In summary, this work demonstrates the importance of quantum effects in determining the ground-state structures of superconducting hydrides-challenging current predictions and evidencing flaws in standard theoretical methods. Similar effects are expected in other high-T_c compounds with hydrogen clathrate structures, for which syntheses have recently been reported²⁸⁻³⁰. We also illustrated that quantum fluctuations are essential in order to sustain crystals with large electron-phonon coupling constants (the value of 3.6, found at 129 GPa for LaH₁₀, is to our knowledge the highest reported); such structures would otherwise be destabilized by the substantial electron-phonon interaction, resulting in distorted (low-symmetry) structures in which the electronic density of states at the Fermi level is reduced⁹ (Extended Data Fig. 7). Our results may therefore help to increase the prospect of attaining high- T_c superconductivity in hydrogen-based structures at much lower pressures than would be expected classically.

Online content

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41586-020-1955-z.

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Methods

Calculation details

First-principles calculations were performed within DFT and the generalized gradient approximation (GGA) as parametrized by Perdew, Burke and Ernzerhof (PBE)³¹. Harmonic phonon frequencies were calculated within density functional perturbation theory (DFPT)³² making use of the Quantum ESPRESSO code^{33,34}. The SSCHA^{25,26,35,36} minimization requires the calculation of energies, forces and stress tensors in supercells. These were also calculated within DFT at the PBE level with Quantum ESPRESSO. For the final SSCHA populations, 1,000 configurations were used to reduce the stochastic noise. In all calculations we used ultrasoft pseudopotentials including 11 electrons for the La atoms, a plane-wave cut-off energy of 50 Ry for the kinetic energy and 500 Ry for the charge density.

In the harmonic phonon calculations for the $Fm\bar{3}m$ and the $R\bar{3}m$ phases, we used the primitive and rhombohedral lattices, respectively, with one LaH₁₀ formula unit in the unit cell. A 20 × 20 × 20 Monkhorst–Pack shifted electron-momentum grid was used for these calculations with a Methfessel–Paxton smearing of 0.02 Ry. The DFT calculations performed for the SSCHA on supercells were performed on a coarser electron-momentum grid, which would correspond to a 12 × 12 × 12 grid in the unit cell. We explicitly verified that this coarser mesh yields a fully converged SSCHA gradient with respect to the electron-momentum grid, thus not affecting the SSCHA minimization. The DFT supercell calculations for the SSCHA minimization on the *C*2 phase were performed keeping the same **k**-point density.

All phonon frequencies for q-points that were not commensurate with the supercell used in the SSCHA minimization were obtained by directly Fourier-interpolating the real space force constants obtained in this supercell, which are calculated form the Hessian of $E(\mathcal{R})$. For the $Fm\overline{3}m$ phase, the SSCHA calculation was performed both on a 2×2×2 and on a 3×3×3 supercell containing, respectively, 88 and 297 atoms. The phonon spectra shown in Fig. 2 for the $Fm\overline{3}m$ phase were obtained by directly Fourier-interpolating the SSCHA energy Hessian force constants obtained in a 3 × 3 × 3 supercell. In Extended Data Fig. 2 we show that the phonon spectrum obtained by directly interpolating the force constants in a 2 × 2 × 2 supercell yields similar results, indicating that the energy Hessian force constants are short-range and can be Fourier-interpolated. Indeed, the T_c calculated with the $2 \times 2 \times 2$ and $3 \times 3 \times 3$ force constants for interpolating phonons differs by only around 3 K. Because the value of estimated T_c only negligibly depends on the cell size for the $Fm\overline{3}m$ phase, the SSCHA quantum structural relaxations in the $R\overline{3}m$ and C2 phases were performed in $2 \times 2 \times 2$ supercells with 88 atoms.

As shown in ref.²⁵, the Hessian of $E(\mathcal{R})$ is

$$\frac{\partial^2 E(\mathcal{R})}{\partial \mathcal{R} \partial \mathcal{R}} = \boldsymbol{\phi} + \boldsymbol{\phi} A \left(1 - \boldsymbol{\phi} A \right)^{-1} \boldsymbol{\phi}^{(3)} \boldsymbol{\phi}$$
(1)

Bold notation represents matrices and tensors in compact notation. In equation (1), ϕ are the variational force constants of the SSCHA minimization, ϕ the quantum statistical averages taken with the SSCHA density matrix of the *n*th order derivatives of $V(\mathbf{R})$, and Λ a tensor that depends on the temperature and ϕ . 1 is the identity matrix. As we show in Extended Data Fig. 2, setting $\phi = 0$ has a negligible effect on the pho-(4) nons obtained from the Hessian defined in equation (1). Therefore, ϕ is neglected throughout, and all superconductivity calculations in the $Fm\overline{3}m$ and $R\overline{3}m$ phases are performed making use of the phonon frequencies and polarization vectors obtained from the Hessian of $E(\mathcal{R})$ with $\phi = 0$. We also estimated T_c with the phonon frequencies and polarization vectors obtained instead from ϕ , resulting in a critical temperature 12 K lower within the Allen–Dynes-modified McMillan equation. This difference is small and within the uncertainty of the T_c calculation between SCDFT and anisotropic Migdal–Eliashberg calculations (see Fig. 3 and below).

The Eliashberg spectral function, which we used for the T_c calculations, is defined as

$$\alpha^{2}F(\omega) = \frac{1}{N_{E_{\rm F}}} \sum_{n\mathbf{k},m\mathbf{q},\nu} \left| g_{n\mathbf{k},m\mathbf{k}+\mathbf{q}}^{\nu} \right|^{2} \delta(\varepsilon_{n\mathbf{k}} - E_{\rm F}) \\ \times \delta(\varepsilon_{m\mathbf{k}+\mathbf{q}} - E_{\rm F}) \delta(\omega - \omega_{\mathbf{q}\nu})$$
(2)

where $N_{F_{e}}$ is the electronic density of states (DOS) at the Fermi energy $(E_{\rm F})$, n and m are band indices, **k** is a crystal momentum, ε_{nk} is a band energy, ω_{av} is the phonon frequency of mode v at wavevector **q**, and $g_{n\mathbf{k},m\mathbf{k}+\mathbf{q}}^{\nu}$ is the electron-phonon matrix element between a state $n\mathbf{k}$ and $m\mathbf{k} + \mathbf{q}$. We calculated $\alpha^2 F(\omega)$ combining the SSCHA phonon frequencies and polarization vectors obtained from the Hessian of $E(\mathcal{R})$ with the electron-phonon matrix elements calculated with DFPT. For the $Fm\overline{3}m$ and $R\overline{3}m$ phases, the electron-phonon matrix elements were calculated in a $6 \times 6 \times 6$ **q**-point grid and a $40 \times 40 \times 40$ **k**-point grid. These were combined with the SSCHA phonons and polarization vectors obtained by Fourier interpolation to the $6 \times 6 \times 6$ q-point grid from the real space force constants coming from the Hessian of $E(\mathcal{R})$ in a $3 \times 3 \times 3$ supercell for the $Fm\overline{3}m$ phase and in a $2 \times 2 \times 2$ supercell for the $R\overline{3}m$ phase. The Dirac deltas on the band energies are estimated by substituting them with a Gaussian of width 0.004 Ry. The calculated $\alpha^2 F(\omega)$ functions for the *Fm*3*m* phase are shown in Extended Data Fig. 3, and in Extended Data Fig. 5 we show the results for the $R\overline{3}m$ phase.

Crystal phase diagram exploration

To sample the enthalpy landscape of LaH₁₀ we used the minima-hopping method^{24,37}, which has been successfully employed for global geometry optimization in a large variety of applications-including superconducting materials such as H₃S, PH₃, and elemental solids at high pressure³⁸⁻⁴⁰. This composition was thoroughly explored with 1, 2, 3 and 4 formula unit simulation cells. Variable composition simulations were also performed for other La-H compositions. Energy, atomic forces and stresses were evaluated at the DFT level with the GGA-PBE parametrization to the exchange-correlation functional. A plane wave basis-set with a high cut-off energy of 900 eV was used to expand the wave function together with the projector-augmented wave method as implemented in the Vienna Ab initio Simulation Package (VASP)⁴¹. Geometry relaxations were performed with tight convergence criteria such that the forces on the atoms were less than 2 meV $Å^{-1}$ and the stresses were less than 0.1 eV Å⁻³. Extended Data Fig. 1 shows our calculated convex hull of enthalpy formation without considering the zeropoint energy at 100, 150 and 200 GPa. Notably, there are many stable compositions in the convex hull. LaH₁₀ becomes enthalpically stable (classically) at around 175 GPa and remains in the convex well above 300 GPa. We have verified that LaH₁₀ ($Fm\bar{3}m$) does not decompose into LaH₃ (*Cmcm*) and LaH₁₁ (P4/nmm) by around 0.3 eV per LaH₁₀ once quantum effects are included in the calculation of the enthalpy formation at 150 GPa, contrary to the conclusion drawn by classical calculations in ref.³. Below 150 GPa, $R\overline{3}m$ and C2 phases (LaH₁₀) show unstable harmonic phonons at Γ , becoming saddle points of $V(\mathbf{R})$. However, harmonically one can find P1 stable structures (decreasing symmetry) by following the instability pattern (softening direction-that is, along eigenvector polarization). P1 structures are degenerate in enthalpy within less than 3 meV per LaH₁₀ with respect to C2. We therefore used the C2 as a representative of highly distorted structures for our study.

Superconductivity calculations in the $Fm\overline{3}m$ phase

Superconductivity calculations were performed within two different approaches that represent the state-of-the-art of ab initio superconductivity: SCDFT and the Eliashberg equations with full Coulomb interaction. SCDFT is an extension to DFT for a superconducting ground state^{42,43}. By assuming that the *n***k** anisotropy in the electron–phonon coupling is negligible (see ref. ⁴³ for further details), the critical temperature is computed by solving an (isotropic) equation for the Kohn–Sham gap:

$$\Delta_{s}(\varepsilon) = \mathcal{Z}(\varepsilon)\Delta_{s}(\varepsilon) - \int d\varepsilon' \mathcal{K}(\varepsilon, \varepsilon') \frac{\tanh\left\lfloor\frac{\beta \mathcal{E}(\varepsilon')}{2}\right\rfloor}{2\mathcal{E}(\varepsilon')} \Delta_{s}(\varepsilon')$$
(3)

where ε is the electron energy and β the inverse temperature. The kernels \mathcal{K} and \mathcal{Z} come from the exchange correlation functional of the theory⁴³⁻⁴⁸ and depend on the properties of the pairing interactions: electron–phonon coupling and screened electron–electron repulsion. Equation (3) enables us to calculate T_c completely ab initio, without introducing an empirical μ^* parameter (Coulomb pseudopotential). Dynamic effects on the Coulomb interaction (plasmon) were also tested and did not show any substantial effect. In its isotropic form, the screened Coulomb interaction in SCDFT is accounted for by a function $\mu(\varepsilon, \varepsilon')$, which is given by the average⁴⁹ random phase approximation (RPA) Coulomb matrix element on the iso-energy surfaces ε and ε' times the DOS at $\varepsilon'(N(\varepsilon'))$:

$$\mu(\varepsilon,\varepsilon') = \sum_{n,m} \iint d^{3}(kk') V_{n\mathbf{k},m\mathbf{k}'}^{\text{RPA}} \frac{\delta(\varepsilon - \varepsilon_{n\mathbf{k}})}{N(\varepsilon_{n\mathbf{k}})} \delta(\varepsilon' - \varepsilon_{m\mathbf{k}'})$$
(4)

The full energy dependence of the DOS is accounted for in the calculations, whereas the electron–phonon coupling is described by the $\alpha^2 F(\omega)$ of equation (2).

The second approach we use to simulate the superconducting state is the anisotropic Eliashberg approach⁵⁰. Here we include, together with the energy dependence of the electron DOS, the anisotropy of the electron–phonon coupling. The Green's function form of the Eliashberg equation we solve is given as

$$\Sigma_{n\mathbf{k}}(\mathbf{i}\omega_i) = -\frac{1}{N\beta} \sum_{\mu,\mathbf{q},m} V_{mn}^{\mathrm{ph}}(\mathbf{q},\mathbf{i}\omega_\mu) G_{m\mathbf{k}+\mathbf{q}}(\mathbf{i}\omega_\mu + \mathbf{i}\omega_i)$$
(5)

$$\Delta_{n\mathbf{k}}(\mathbf{i}\omega_{i}) = -\frac{1}{N\beta} \sum_{\mu,\mathbf{q},m} \{V_{mn}^{\text{ph}}(\mathbf{q},\mathbf{i}\omega_{\mu}) + V_{mn}^{\text{C}}(\mathbf{q},\mathbf{i}\omega_{\mu})\} \\ \times |G_{m\mathbf{k}+\mathbf{q}}(\mathbf{i}\omega_{\mu}+\mathbf{i}\omega_{i})|^{2} \Delta_{m\mathbf{k}+\mathbf{q}}(\mathbf{i}\omega_{\mu}+\mathbf{i}\omega_{i})$$
(6)

Here, $\Sigma_{n\mathbf{k}}(i\omega_i)$ and $\Delta_{n\mathbf{k}}(i\omega_i)$ are the normal and anomalous self energy, and $V_{mn}^{\text{ph}}(\mathbf{q}, i\omega_{\mu})$ and $V_{mn}^{\text{C}}(\mathbf{q}, i\omega_{\mu})$ are the **k**-averaged phonon-mediated interaction and Coulomb interaction, respectively. The explicit form of $V_{mn}^{\text{ph}}(\mathbf{q}, i\omega_{\mu})$ is given as

$$V_{mn}^{\text{ph}}(\mathbf{q}, \mathbf{i}\omega_{\mu}) = \sum_{\nu} |g_{nm}^{\nu}(\mathbf{q})|^2 D_{\nu}(\mathbf{q}, \mathbf{i}\omega_{\mu})$$
(7)

where $|g_{nm}^{\nu}(\mathbf{q})|^2$ is a **k**-averaged electron-phonon matrix element

$$\left|g_{nm}^{\nu}(\mathbf{q})\right|^{2} = \frac{\sum_{\mathbf{k}} \left|g_{n\mathbf{k},m\mathbf{k}+\mathbf{q}}^{\nu}\right|^{2} \delta(\varepsilon_{n\mathbf{k}}-E_{\mathrm{F}}) \delta(\varepsilon_{m\mathbf{k}+\mathbf{q}}-E_{\mathrm{F}})}{\sum_{\mathbf{k}} \delta(\varepsilon_{n\mathbf{k}}-E_{\mathrm{F}}) \delta(\varepsilon_{m\mathbf{k}+\mathbf{q}}-E_{\mathrm{F}})}$$
(8)

and $D_v(\mathbf{q}, i\omega_\mu)$ is a free-phonon Green's function, $D_v(\mathbf{q}, i\omega_\mu) = -2\omega_{\mathbf{q}v}/(\omega_\mu^2 + \omega_{\mathbf{q}v}^2)$. The electron–phonon matrix elements are calculated through a DFPT calculation with $6 \times 6 \times 6 \mathbf{q}$ -point grid and are combined with the phonon frequencies and polarization vectors obtained by directly Fourier-interpolating to this grid the force constants arising from the $E(\mathcal{R})$ Hessian in the $3 \times 3 \times 3$ supercell. For the Coulomb interaction, $V_{mn}^C(\mathbf{q}, i\omega_\mu)$ is approximated by **k**-averaged static Coulomb interaction within the random phase approximation, $\frac{1}{N_k} \sum_{\mathbf{k}} V_{mk,nk+\mathbf{q}}^{\text{RPA}}(i\omega_\mu = 0)$. Using equation (5), the Dyson equation was solved self-consistently and then equation (6) was solved to estimate T_c with $36 \times 36 \times 36$ **k**-point grid and 512 Matsubara frequencies. In Extended Data Table 1 we summarize all calculated T_c values within anisotropic ME and isotropic SCDFT. We also include the values obtained with the McMillan equation and the Allen–Dynes-modified McMillan equation ($\mu^* = 0.1$). The calculated electron–phonon coupling constant, $\lambda = 2 \int_0^{\infty} d\omega \alpha^2 F(\omega)/\omega$ and the logarithmic frequency average, $\omega_{\log} = \exp\left(\frac{2}{\lambda} \int_0^{\infty} d\omega \frac{\alpha^2 F(\omega)}{\omega} \log\omega\right)$ are also included in the table.

Quantum structural relaxations in the $R\overline{3}m$ and C2 phases

In Extended Data Fig. 5 we show the evolution of the pressure calculated along the different Cartesian directions for the $R\overline{3}m$ throughout the SSCHA minimization but keeping the rhombohedral angle fixed at 62.3° . Thus, the centroid positions \mathcal{R} are optimized only considering the internal degrees of freedom of the $R\overline{3}m$ phase. Even if, at the classical level, the stress is isotropic (within a 0.5%), after the SSCHA quantum relaxation an anisotropic stress of a 6% is created between the z and x-y directions. The phonons obtained at the end of the minimization are shown in Extended Data Fig. 5. Second, in Extended Data Fig. 4, we show that starting from the result of this minimization but now also relaxing the lattice, the $R\overline{3}m$ phase evolves into the $Fm\overline{3}m$ phase. It is clear how the pressure calculated with quantum effects becomes isotropic when the rhombohedral angle becomes 60°, the angle corresponding to an fcc lattice in a rhombohedral description. It is also evident that the Wyckoff positions of the $R\overline{3}m$ phase evolve clearly into the $Fm\overline{3}m$ Wyckoff positions, which are summarized in Extended Data Table 2.

In Extended Data Fig. 6 we show the evolution of the diagonal components of the pressure along the three different Cartesian directions for the monoclininc C2 when the lattice structure is relaxed with the SSCHA. The starting point is obtained by first performing a SSCHA relaxation of only internal atomic coordinates, keeping the lattice parameters that yield an isotropic stress of 150 GPa. It is clear that quantum effects create an anisotropic stress if the lattice parameters are not modified. When the quantum relaxation of the lattice is performed, the lattice parameters are modified and an isotropic stress is recovered.

Extended Data Fig. 7 shows the structures of the $R\bar{3}m$ and C2 phases obtained classically and after the quantum SSCHA relaxation. After the quantum relaxation, the symmetry of both structures is recognized as $Fm\bar{3}m$ with a tolerance of 0.001 Å for lattice vectors and 0.005 Å for ionic positions, consistent with the stochastic accuracy of the SSCHA. In the same figure, the electronic DOS as a function of pressure is plotted. A highly symmetric motif ($Fm\bar{3}m$) maximizes $N_{E_{F'}}$, whereas in distorted structures ($R\bar{3}m$ and C2) the occupation at the Fermi level is reduced by more than 20%. This underlines that the classical distortions would lower $N_{E_{F'}}$ reducing λ , as expected in a system that is destabilized by the electron–phonon interaction.

Transition temperatures from other La-H compositions

Different compositions on the La–H phase diagram have been reported to be thermodynamically stable. Presumably, the stabilization of these compositions and the measurement of different T_c values (see ref. ⁵) demonstrate that other stoichiometries are responsible for these measured T_c values. Notably, these T_c values appear substantially lower–for instance, the values decrease from 250 K, to 215 K, 110 K and to 70 K. Experimentally there is not a clear correlation between sample preparation, T_c and pressure. In the sample preparation in ref. ⁵, pressures can vary from 100 to 200 GPa (gradient inside the DAC) and it was proposed that other stoichiometries (low-hydrogen content) are responsible for systematically lower values of T_c .

Conversely, in a later publication, the same authors suggested that another hydrogen-rich system that is enthalpically competitive (LaH_{II}) could possibly be responsible for other high- T_c values. In order to verify this hypothesis, we considered structure-prediction runs with this stoichiometry, and found crystalline structures that were previously reported in ref.³. Extended Data Fig. 8 shows the structural motif and

the corresponding phonons and $\alpha^2 F(\omega)$ spectral function. We can rule out the possibility that high T_c values, as measured in different samples, arise from LaH₁₁ in its P4/nmm (129) structure (lowest enthalpy structure for this composition at relevant experimental pressures). As seen in Extended Data Fig. 8, this phase has a strong molecular-crystal character, composed of H₂ units weakly interacting with the La lattice. This phase is indeed a poor metal—with low occupation of electrons at the Fermi level—owing to its molecular character, and it cannot explain T_c values of 70 K or higher. Our estimated T_c with the Allen–Dynes formula, harmonic phonons and using a $\mu^* = 0.1$ is 7 K at 100 GPa, reaching around 24 K at 200 GPa. More importantly, this phase does not show marked anharmonicity.

Data availability

All the data generated in this work is available upon request from I.E. and J.A.F.-L.

Code availability

Quantum ESPRESSO is an open-source suite of computational tools available at https://www.quantum-espresso.org. VASP is a proprietary program. The SSCHA and the SCDFT codes are private codes developed by some of the authors, and are being prepared for distribution as an open-source code.

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Author contributions The project was conceived by I.E. and J.A.F.-L. The SSCHA was developed by I.E., L.M., R.B., M.C. and F.M. In particular, R.B. developed the method to compute the quantum energy Hessian and the anharmonic phonon dispersions, and L.M. developed the method to perform a quantum relaxation of the lattice parameters. I.E. and F.B. performed the SSCHA calculations. A.S., T.K., T.T., R.A. and J.A.F.-L. conducted studies on structure prediction and superconductivity. All authors contributed to the editing of the manuscript.

Competing interests The authors declare no competing interests.

Additional information

Correspondence and requests for materials should be addressed to J.A.F.-L. **Peer review information** *Nature* thanks Yanming Ma and the other, anonymous, reviewer(s) for their contribution to the peer review of this work.

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Extended Data Fig. 1 | Convex hull of enthalpy formation and diffraction pattern of candidate LaH₁₀ phases. Top, classical calculations of enthalpy (without zero-point energy) at different hydrogen contents at 100, 150 and 200 GPa. At low pressure (100 GPa), LaH₁₀ is not stable and only develops as stable point in the convex hull of enthalpy formation at pressures above about 175 GPa. Bottom, diffraction patterns of different structures at 150 GPa (classical pressure), compared to the experimental data reported in ref.⁵ for LaH₁₀ in the $Fm\bar{3}m$ phase at 150 GPa. The pattern is shown in the vicinity of the (111) peak of the $Fm\bar{3}m$ phase. This peak is clearly split in the distorted C2 and $R\bar{3}m$ phases predicted classically. The figure provides confirmation that the experimental resolution in ref.⁵ would have been sufficient to distinguish between these phases.



Extended Data Fig. 2 | **Convergence of SSCHA-phonon supercells and** different anharmonic phonon calculations for LaH₁₀ at 163 GPa. Left, the phonon spectra shown are calculated by directly Fourier-interpolating the force constants obtained from the Hessian of $E(\mathcal{R})$ in a real space $2 \times 2 \times 2$ and a $3 \times 3 \times 3$ supercell. The similarity of both phonon spectra obtained by Fourier interpolation indicates that these SSCHA force constants are short-ranged and can be Fourier-interpolated. Right, phonon spectra obtained from the SSCHA energy Hessian of equation (1), making different level of approximations. The purple solid line is the phonon spectrum calculated with the full-energy Hessian without any approximation. In the blue dotted spectrum we set $\phi = 0$ in the equation. For the orange dash-dotted line we set $\Phi = \Phi = 0$, so that the phonon spectra correspond to that arising directly from the SSCHA variational force constants Φ . These results clearly show that whereas the effect of ϕ is

important, setting $\phi = 0$ has minimal effect. All phonon spectra are obtained by directly Fourier-interpolating the real space anharmonic force constants in a $2 \times 2 \times 2$ supercell.



Extended Data Fig. 3 | $\alpha^2 F(\omega)$ values for the $Fm\overline{3}m$ phase of LaH₁₀ and LaD₁₀. Calculated $\alpha^2 F(\omega)$ values for different pressures together with the integrated electron–phonon coupling constant, which is defined as $\lambda(\omega) = 2 \int_0^{\omega} d\Omega \alpha^2 F(\Omega) / \Omega$. The results show that high frequency, optical modes of hydrogen are

responsible for the large value of the electron–phonon coupling constant λ . It is worth noting that acoustic modes with La character contribute between 0.2 and 0.5 to λ and cannot be neglected when aiming to estimate an accurate value of T_c .





Extended Data Fig. 4 | **Details of the** $R\bar{3}mLaH_{10}$ **cell relaxation, including quantum effects.** The initial point for the relaxation is the output from the previous internal relaxation with fixed angle presented in Extended Data Fig. 5. The $R\bar{3}m$ phase in the rhombohedral description is described by three vectors of the same length ($\mathbf{a} = \mathbf{b} = \mathbf{c}$) and by the angles between them ($\alpha = \beta = \gamma$). The top left panel shows the evolution of the rhombohedral angle and the top right panel shows the evolution of the rhombohedral lattice parameter (a = b = c). The progression of the stress tensor in the quantum SSCHA minimization is shown in the bottom left panel. It is clear that at the end of the minimization the structure has an angle of 60°, which matches the angle of an fcc lattice and, in

this case, the stress is isotropic. In the bottom right panel, we show the evolution of the Wyckoff positions in the minimization and we compare it with that of the $Fm\bar{3}m$ phase. The occupied Wyckoff positions for both $R\bar{3}m$ LaH₁₀ and $Fm\bar{3}m$ LaH₁₀ are summarized in Extended Data Table 2. Here, the evolution of ε_a , ε_b , ε_x and ε_y parameters in the minimization can be seen. The atoms in the first set of 6c positions approach the 8c Wyckoff site of the $Fm\bar{3}m$ phase, whereas the atoms in the second set of 6c positions and those in 18h sites approach the atoms in the 32fWyckoff site of the $Fm\bar{3}m$ phase, where $\varepsilon = 0.12053$.



Extended Data Fig. 5 | Phonon dispersion in $R\overline{3}m$ -phase LaH₁₀ and the anisotropic pressure created in a fixed-cell quantum relaxation. Left, harmonic and anharmonic phonon spectrum, maintaining a 62.3° rhombohedral angle. The harmonic calculation is performed with the internal atomic positions that yield classical vanishing forces. The anharmonic calculation is performed after relaxing (with the SSCHA) the internal degrees of freedom but maintaining the 62.3° rhombohedral angle. At the harmonic level there are unstable phonon modes even at Γ . Symmetry prevents the relaxation of this structure according to the unstable phonon mode at Γ . The harmonic phonons are calculated at a classic pressure of 150 GPa. Quantum effects add around an extra 10 GPa to the pressure. To the right of the graph is shown the behaviour of $\lambda(\omega)$ and $\alpha^2 F(\omega)$ for the anharmonic calculation. Right, pressure along the different Cartesian directions during the SSCHA relaxation of the internal parameters, keeping the rhombohedral angle fixed at 62.3°. At step 0 the pressure reported is obtained directly from $V(\mathbf{R})$, neglecting quantum effects. It is isotropic within 1 GPa of difference between the *x*-*y* and *z* directions. At each of the other steps it is calculated from the quantum $E(\mathcal{R})$ and along the minimization it becomes anisotropic. When the minimization stops at step 12–that is, the internal coordinates are at the minimum of the $E(\mathcal{R})$ for this lattice—the stress anisotropy between the *z* and the *x*-*y* directions is about 6%. This clearly indicates that quantum effects act to relax the crystal lattice—in particular, because P_z is larger—by reducing the rhombohedral angle. It is worth noting that quantum effects increase the total pressure by approximately 10 GPa, which is calculated as $P = (P_x + P_y + P_z)/3$. The initial cell parameters before the minimization are a = 3.5473398 Å and $a = 62.34158^\circ$. The initial values of the free Wyckoff parameters, which yield classical vanishing forces and a 150-GPa isotropic stress, are $\varepsilon_a = 0.26043$, $\varepsilon_b = 0.09950$, $\varepsilon_x = 0.10746$ and $\varepsilon_y = 0.12810$. See Extended Data Table 2 for more details.



Extended Data Fig. 6 | **Anisotropic pressure of the C2 phase of LaH**₁₀ **in a cell quantum relaxation.** Pressure along the different Cartesian directions is plotted during the SSCHA cell minimization. The target pressure for this minimization is 160 GPa. At the end of the minimization the isotropy of the stress tensor is recovered. A symmetry analysis performed on the structure at the end of the minimization confirms that the $C2LaH_{10}$ evolves in the $Fm\overline{3}m$ -phase LaH₁₀. The initial values $P_x = 163.2$ GPa, $P_y = 159.7$ GPa, $P_z = 155.0$ GPa are obtained by an atomic internal relaxation performed using the SSCHA with a fixed cell.



Extended Data Fig.7 | **SSCHA minimization on LaH**₁₀ and **DOS.** Top left and top right, two initial structures (C2 and $R\overline{3}m$) of low enthalpy that were considered in our SSCHA simulations. When considering quantum effects, both structures evolve towards the $Fm\overline{3}m$ structure. The corresponding total electronic DOS at different pressures is plotted for each structure (for comparison, at the same energy scale). The highly symmetric motif ($Fm\overline{3}m$)

maximizes N_{E_r} , whereas in distorted structures ($R\bar{3}m$ and C2) the occupation at the Fermi level is reduced by more than 23% for C2 and by 11% for $R\bar{3}m$ (with respect to $Fm\bar{3}m$ at 150 GPa). Values at classical pressures are shown for comparison. Note that the shape of the DOS plot is also strongly modified at different pressures.



Extended Data Fig. 8 | **Details of LaH**₁₁. Left, crystal structure of the *P*4/*nmm* phase of LaH₁₁ at 100 GPa, which is thermodynamically stable in the convex hull. Top right, dispersion of harmonic phonons along the momentum space for LaH₁₁: it is dynamically stable. Bottom right, superconducting Eliashberg

spectrum function ($\alpha^2 F(\omega)$) calculated for LaH₁₁ at the pressure indicated with harmonic phonons. The T_c estimated using the Allen–Dynes formula ($\mu^* = 0.1$) is around 7 K at 100 GPa (harmonic phonons).

Extended Data Table 1 | Summary of calculated T_c values

| System | Pressure (GPa) | λ | ω_{\log} (meV) | $ T_{c}{}^{Mc}_{\mu^*=0.1}$ (K) | $ T_{c\mu^*=0.1}^{AD}(K) $ | $ T_{c_{ani}}^{ME}(K) $ | $ T_{c}^{SCDFT}(K) $ |
|---------------------------|----------------|-----------|-----------------------|---------------------------------|----------------------------|-------------------------|----------------------|
| LaH_{10} | 129 | 3.62 | 76.4 | 171.8 | 252.6 | 255.3 | 230 |
| LaH_{10} | 163 | 2.67 | 96.4 | 190.4 | 247.0 | 242.8 | 225 |
| LaH_{10} | 214 | 2.06 | 115.5 | 196.3 | 235.9 | 237.9 | 210 |
| LaH_{10} | 264 | 1.73 | 126.6 | 189.5 | 219.2 | 216.9 | 201 |
| LaD_{10} | 159 | 3.14 | 63.5 | 135.0 | 184.2 | 180.4 | 171 |
| LaD_{10} | 210 | 2.21 | 81.7 | 145.5 | 176.5 | 172.9 | 158 |
| LaD_{10} | 260 | 1.80 | 92.2 | 142.2 | 164.6 | 157.9 | 151 |

Values are calculated using different approaches ranging from empirical to fully ab initio: McMillan equation $(T_{c_{\mu=0,l}}^{Mc})$, Allen–Dynes-modified McMillan equation $(T_{c_{\mu=0,l}}^{AD})$, anisotropic treatment of Migdal–Eliashberg (T_{cani}^{Mc}) and SCDFT (T_{c}^{SCDFT}) . Values of λ and ω_{log} are also given.

Extended Data Table 2 | Details of the crystal structures

| $Fm\overline{3}m$ (C) | | $Fm\overline{3}r$ | m (R) | $R\overline{3}m$ (R) | | |
|-----------------------|--|-------------------|---|----------------------|--|--|
| 1 La 4b | $\left[\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\right]$ | 1 La 4b | $\left[\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\right]$ | 1 La 3b | $\left[\tfrac{1}{2}, \tfrac{1}{2}, \tfrac{1}{2}\right]$ | |
| 2 H 8c | $\begin{bmatrix} \frac{1}{4}, \frac{1}{4}, \frac{1}{4} \\ \begin{bmatrix} \frac{3}{4}, \frac{3}{4}, \frac{3}{4} \end{bmatrix}$ | 2 H 8c | $\begin{bmatrix} \frac{1}{4}, \frac{1}{4}, \frac{1}{4} \end{bmatrix} \begin{bmatrix} \frac{3}{4}, \frac{3}{4}, \frac{3}{4} \end{bmatrix}$ | 2 H 6c | $\begin{bmatrix} \epsilon_a, \epsilon_a, \epsilon_a \end{bmatrix} \\ \begin{bmatrix} -\epsilon_a, -\epsilon_a, -\epsilon_a \end{bmatrix}$ | |
| 8 H 32f | $[\epsilon, \epsilon, \epsilon]$ | 8 H 32f | $[\epsilon, \epsilon, \epsilon]$ | 2 H 6c | $\begin{bmatrix} \epsilon_b, \epsilon_b, \epsilon_b \end{bmatrix}$ | |
| | $[\epsilon, \epsilon, -\epsilon]$ | | $[-\epsilon, -\epsilon, 3\epsilon]$ | 6 H 18h | $[-\epsilon_x, -\epsilon_x, 3\epsilon_y]$ | |
| | $[\epsilon, -\epsilon, \epsilon]$ $[-\epsilon, \epsilon, \epsilon]$ | | $[-\epsilon, 3\epsilon, -\epsilon]$ $[3\epsilon, -\epsilon, -\epsilon]$ | | $\begin{bmatrix} -\epsilon_y, 5\epsilon_x, -\epsilon_x \end{bmatrix} \\ \begin{bmatrix} 3\epsilon_y, -\epsilon_x, -\epsilon_x \end{bmatrix}$ | |
| | $[-\epsilon, -\epsilon, \epsilon]$ $[-\epsilon, \epsilon, -\epsilon]$ | | $[\epsilon, \epsilon, -3\epsilon]$ $[\epsilon, -3\epsilon, \epsilon]$ | | $\begin{bmatrix} \epsilon_x, \epsilon_x, -3\epsilon_y \end{bmatrix}$ $\begin{bmatrix} \epsilon_x, -3\epsilon_y, \epsilon_x \end{bmatrix}$ | |
| | $[\epsilon, -\epsilon, -\epsilon]$ | | $[-3\epsilon,\epsilon,\epsilon]$ | | $[-3\epsilon_y,\epsilon_x,\epsilon_x]$ | |

Composition (Space group) Lattice parameters

Wyckoff positions

| LaH_{10} ($Immm$) | a = 3.58303 Å b = 3.61834 Å c = 5.08749 Å | La H H | 2c 8m 8l 4i | [0.50000, 0.50000, 0.00000] [0.75841, 0.00000, 0.11649] [0.00000, 0.75742, 0.87548] [0.50000, 0.00000, 0.74573] |
|--|--|------------------------|--|--|
| LaH_{10} (C2) | a = 6.15468 Å b = 3.60628 Å c = 7.23776 Å $\beta = 55.71434^{\circ}$ | La H H H | 4c 4c 4c 4c 4c | [0.49244, 0.00070, 0.25292] [0.13978, 0.24567, -0.05243] [0.09798, 0.24122, 0.45027] [0.36015, 0.25590, 0.05238] |
| | | H H H H H | 4c 4c 4c 4c 4c 4c 4c 4c | [0.40204, 0.26021, 0.54971] [-0.09751, 0.00051, -0.05100] [0.86810, 0.00071, 0.43706] [0.88713, 0.00076, 0.69398] [0.87083, 0.00068, 0.19089] [0.73058, 0.00043, 0.88088] |
| LaH ₁₁ (<i>P</i> 4/ <i>nmm</i>) | a = 3.87435 Å b = 3.87435 Å c = 5.27636 Å | H La H H H | 4c 2c 4e 8i 8i 2a | [0.76156, 0.00071, 0.36763] [0.25000, 0.25000, 0.78577] [0.00000, 0.00000, 0.50000] [0.25000, -0.02052, 0.17824] [0.25000, 0.55418, 0.35160] [0.75000, 0.25000, 0.00000] |
| | | | | |

Top, the table summarizes the occupied Wyckoff positions for different structures found with the minima hopping method and used in SSCHA for minimization. We describe the Wyckoff positions using crystal coordinates, so that the [x, y, z] coordinate should be understood as an xa + yb + zc atomic position with a, b, c the lattice vectors. For the $R\overline{3}m$ phase we use the rhombohedral lattice (R), where the three lattice vectors have the same length (a = b = c) and the angle between them is the same ($a = \beta = \gamma$). The $Fm\overline{3}m$ phase is described both in this rhombohedral description (R) and, for comparison, in the standard cubic conventional lattice (C). In the $Fm\overline{3}m$ phase the lanthanum atom is described by the 4b sites, two hydrogen atoms occupy the 8c sites, and the remaining eight hydrogen atoms occupy the 32f sites. Most of the atomic positions are fixed by symmetry, and overall the $Fm\overline{3}m$ structure can be described by one single free parameter (ε). In the $R\overline{3}m$ phase the lanthanum atom is locked in the 3b sites, two pairs of hydrogen atoms occupy the 6c sites and the remaining six hydrogen atoms occupy the 18h sites. In this case symmetry allows for more freedom and overall the structure of the $R\overline{3}m$ phase can be described by four free parameters ($\varepsilon_w, \varepsilon_w, \varepsilon_x$ and ε_y). The bottom table shows lattice parameters and atomic coordinates for LaH₁₀ (*Immm*) and LaH₁₀ (C2) at 150 GPa and LaH₁₁ P4/nmm at 100 GPa. These pressures are estimated classically. The positions below give vanishing forces at classical level.