Enhancement of superconducting properties in the La–Ce–H system at moderate pressures

Ternary hydrides are regarded as an important platform for exploring high-temperature superconductivity at relatively low pressures. Here, we successfully synthesized the hcp-(La,Ce)H$_{9-10}$ at 113 GPa with the initial La/Ce ratio close to 3:1. The high-temperature superconductivity was strikingly observed at 176 K and 100 GPa with the extrapolated upper critical field $H_{c2}(0)$ reaching 235 T. We also studied the binary La-H system for comparison, which exhibited a $T_c$ of 103 K at 78 GPa. The $T_c$ and $H_{c2}(0)$ of the La-Ce-H are respectively enhanced by over 80 K and 100 T with respect to the binary La-H and Ce-H components. The experimental results and theoretical calculations indicate that the formation of the solid solution contributes not only to enhanced stability but also to superior superconducting properties. These results show how better superconductors can be engineered in the new hydrides by large addition of alloy-forming elements.

Recent discoveries of hydrogen-based superconductors, such as H$_3$S$_{1-5}$, LaH$_{6-9}$, YH$_6$ and YH$_{9-10}$, CeH$_9$ and CeH$_{10}$$_{13-15}$, and CaH$_6$$_{16,17}$ have inspired enormous interest in multiple areas. Binary high-temperature superconducting (HTSC) hydrides can be basically divided into two categories. The first theoretically and experimentally discovered hydride H$_3$S belongs to the class of covalent polyhydrides$_2,3$, which require covalent bonding between hydrogen and light nonmetal elements located in the upper right corner of the periodic table. The second type is mainly alkaline earth and rare earth metal polyhydrides with sodalite-like clathrate structures$_{16-18}$, currently at the forefront of research. Besides, layered hydrides with graphene-like hydrogen nets are also predicted to have great potential as high-temperature superconductors$.^3$. Although the records of the superconducting $T_c$ keep getting updates, obtaining HTSC phases at moderate pressures is still an elusive goal. More attention has been turned to ternary systems, typically for La-based HTSC polyhydrides$.^{18-22}$ The complexity of ternary hydrides in theory invites an experimental approach$.^{20,21}$ However, this is thwarted by poorly controlled and complex synthesis.

Superconducting binary hydrides of lanthanum$^{6-9,25-29}$ and cerium$^{11,13,30-32}$ have been well studied before and have the similar crystal structures. Lanthanum polyhydride $Fm3m$-LaH$_{10}$ holds the record of $T_c$ (up to 250 K at 170 GPa), whereas cerium hydrides $P6_3/mmc$-CeH$_9$ and $Fm3m$-CeH$_{10}$ display HTSC properties at much lower pressures ($T_c$ up to 115 K at 95 GPa). Alloying these two hydrides may lead to ternary hydrides with enhanced properties that can be stable at moderate pressures. As neighbors in the periodic table, La and Ce have very close atomic radii and electronegativities and can form continuous and homogeneous solid solutions$.^{30,33,34}$ In this work, we have synthesized ternary HTSC-(La,Ce)H$_{9-10}$ with metal atoms forming the hexagonal close-packed ($P6_3/mmc$) sublattice. Superconductivity is strikingly preserved to $\sim$100 GPa with $T_c$ of 176 K and extrapolated $H_{c2}(0)$ of 235 T. This phase is stable under lower pressures with the giant enhancement of superconducting properties compared with the binary La–H and Ce–H system in the same pressure range. Present evidence suggests that the disordered state of (La,Ce)H$_{9-10}$ has a significant effect on the stability and superconducting properties.

Received: 26 August 2022
Accepted: 17 April 2023
Published online: 09 May 2023

Wuhao Chen$^1$, Xiaoli Huang$^1$, Dmitrii V. Semenok$^2$, Su Chen$^1$, Di Zhou$^2$, Kexin Zhang$^1$, Artem R. Oganov$^3$ & Tian Cui$^1,4$

1State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun 130012, China. 2Center for High Pressure Science and Technology Advanced Research (HPSTAR), Beijing, 100094, China. 3Skolkovo Institute of Science and Technology, Skolkovo Innovation Center, Bolshoy Boulevard 30, bldg. 1, Moscow 121205, Russia. 4School of Physical Science and Technology, Ningbo University, Ningbo 315211, China.  e-mail: huangxiaoli@jlu.edu.cn; cuitian@nbu.edu.cn
Results

Synthesis and characterization of the La–Ce alloys

We have chosen to synthesize ternary La–Ce–H hydride by a reaction of a La–Ce alloy with hydrogen, which is a relatively simple approach to synthesize multinary polyhydrides. Under pressure, the concentration of ~75% La is predicted to be the phase boundary in the La–Ce phase diagram that may be in favor of the lattice reconstruction. Hence, we used the La$_{0.75}$Ce$_{0.25}$ alloy (the ratio of La–Ce is 3:1) prepared by multitarget magnetron sputtering as the initial reactant for typical experimental runs (Supplementary Table S1 and Fig. S1). Before loading into the diamond anvil cell, we have characterized this solid solution with different methods. We got the exact concentration by scanning electron microscopy equipped with energy-dispersive X-ray spectroscopy (SEM + EDX), which also proved the homogeneous distribution of the elements (Supplementary Figs. S14–S16 and S21). The X-ray diffraction (XRD) measurement revealed that the La–Ce alloy belonged to $Fm\overline{3}m$ symmetry without any other impurity phase (Supplementary Fig. S2). We also discovered that the introduction of Ce atoms obviously suppressed the $T_c$ of pure La. The single superconducting transition judging from the $R$–$T$ curve partly indicated the homogeneity (Supplementary Figs. S19 and S20). It is noteworthy that La and Ce atoms retain random distribution over metal sites even after the formation of the polyhydride at high pressure.

Superconductivity in the La–Ce–H and La–H systems

To synthesize La–Ce–H or La–H compounds, we compressed the La–Ce alloy or pure La in the ammonia borane (NH$_3$BH$_3$) sample, which acted both as the pressure-transmitting medium and source of hydrogen. After that, the samples were laser-heated at the target pressure for a few seconds. In this process, hydrogen was released and reacted with La–Ce alloy or pure La at a temperature up to 1500 K, following which reaction products were quenched to room temperature. After synthesizing La–Ce–H compounds by laser heating at specific pressures, we conducted the electrical measurements and plotted the typical data in Fig. 1. To explore the possible high-temperature superconductivity at pressures lower than those of the known LaH$_{10}$, we laser-heated the samples in DACs #2 and #9 at 113 GPa and 120 GPa, respectively. Strikingly, the $T_c$ reaches 175 K (113 GPa, Fig. 1b) and 190 K (123 GPa, Fig. 1d), which is about 100 K higher than our previously discovered cerium polyhydrides. The obtained products are expected to remain metastable below the synthetic pressure. To explore this, we measured the pressure dependence of $T_c$ upon decompression and found $T_c$ of 155 K at 95 GPa (DAC #2) and 180 K at 104 GPa (DAC #9). To compare with LaH$_{10}$, we heated DAC #6 at 152 GPa and observed the main resistance drop starting from 188 K (Supplementary Fig. S17). We also noticed another slight drop at 206 K. This is because of the formation of two superconducting phases (see Supplementary Fig. S18).
$T_c$ slightly decreases with further compression to 156 GPa and 162 GPa, which is different from the behavior of C2/m-LaH$_{10}$\cite{29}. Noteworthy, the pressure scale (diamond Raman edge) used in this study, as well as from Somayazulu et al.\cite{8}, gives a higher value than using the hydrogen vibron by -18 GPa\cite{29}. The same diamond Raman edge scale has been used in both compression and decompression processes. DACs #3 and #5 were both laser-heated at about 130 GPa. Similar to sample #6, we also observed a slight resistance drop at 186 K for sample #3 (Supplementary Fig. S8). Sample #5 showed an obvious step-like transition which indicated the superconductivity of other phases at lower temperatures. The highest $T_c$, decreasing gradually along with the pressure, dropped to 132 K at 104 GPa. In contrast, the lower $T_c$ phase (~37 K, at the resistance close to zero) was robust during decompression from 127 GPa (Fig. 1c) to at least 80 GPa (Fig. 1d). Further decompression of sample #9 from 123 to 101 GPa showed the tendency similar to that of the high-$T_c$ phase in DACs #2 and #5 (Fig. 1b, c). The resistance of the La–Ce–H samples increased significantly when pressure decreased, and the width of the superconducting transition increased about 2.5 times from 104 GPa to 101 GPa (Fig. 1d), possibly due to disordering of the structure in the vicinity of a phase transition. Compared with the binary fcc-CeH$_{10}$ and hcp-CeH$_{10}$\cite{30}, we obtained higher $T_c$ in the ternary La–Ce–H system. However, there was no report on the superconductivity of binary La–H system at pressures lower than 120 GPa for our comparison. To fill this gap, we also explored the La–H system in the same pressure range (Fig. 1e). In DAC #L1, the La–H system showed the $T_c$ of 84 K after the first laser heating at 123 GPa. Further increasing the pressure and laser heating the sample led to another superconducting transition with a higher $T_c$ of 112 K at 129 GPa (Supplementary Fig. S30). During the decompression of DAC #L1 (Supplementary Fig. S31), superconductivity was preserved down to 78 GPa with a $T_c = 103$ K (Fig. 1e and Supplementary Fig. S28). These data indicate that La–Ce–H system has a greatly enhanced $T_c$ compared with La–H and Ce–H systems below 130 GPa.

The structural analysis of the La–Ce–H and La–H systems

To reveal the crystal structures of the superconducting polyhydrides, we performed synchrotron X-ray diffraction (XRD) measurements on the electrically characterized La–Ce–H samples in DACs #2 and #3 (Fig. 2a and Supplementary Figs. S4, S5, and S9), and newly prepared La–H samples in DAC #5 (Fig. 2a and Supplementary Fig. S35). The data were collected from three individual La–Ce–H electrical DACs at their pressures and from La–H DAC #5 during decompression. XRD reflections in phase mixtures were separated according to the phase distribution and the state of Debye rings. The Pb$_6$/mmce and Fm3m structures were discovered in DAC #3 at 131 GPa simultaneously, which can explain the high-$T_c$ phases. The Debye rings of the Pb$_6$/mmce structure were spot-like in DAC #2 while uniform in DAC #3. The lower pressure of synthesis and sufficient laser heating probably caused a better crystallization of sample #2. Noteworthy, the temperature (~1500 K) is not high enough to melt the La–Ce alloy at megabar pressures. Laser-heating may decompose the synthesized hydrides but cannot change the disordered distribution of La/Ce atoms. During the revision of our manuscript, we noticed another two La–Ce–H works with La/Ce of 1:1 that reported the Pb$_6$/mmce\cite{29} and Fm3m\cite{31} phases, respectively. The main differences between our and their data lie in the different La–Ce ratios and synthesized conditions, which contribute to the various results. By comparing with $P - V$ trend (Fig. 4c and Fig. S6), we propose that the Fm3m phase can possibly only be synthesized at pressures above 130 GPa (DACs #3 and #6) with lower $T_c$ than the Pb$_6$/mmce phase (Supplementary Figs. S8 and S18). Besides, the impurity phases with lower hydrogen content can explain the other lower $T_c$'s that we also observed. Current experimental techniques allow one just

![Image](https://doi.org/10.1038/s41467-023-38254-6)
to determine the metal sublattice and estimate the hydrogen content in metal polyhydrides. Hydrogen atoms cannot be directly determined due to their very low scattering factor. Neutron diffraction\textsuperscript{37} and nuclear magnetic resonance\textsuperscript{38} could be employed in the future to give more information on the structure of H-sublattice.

We plotted our data together with the calculated or experimentally reported equations of state (EoS) of binary Ce–H and La–H hydrides for comparison (Fig. 2b). The hydrogen content was determined by comparing the unit cell volume. We concluded that the main superconducting phase in La–Ce–H system below 130 GPa was deemed as $\text{Pm}\text{c}/\text{mm}c$-(La,Ce)H$\text{p}_{10}$. Considering that the electrical resistance and XRD measurements are not performed on the same DAC for the binary La–H system, $T_c$ values observed in DAC #L1 cannot be directly distinguished from those of the mixed phases $\text{Pm}\text{c}/\text{mm}c$-LaH$_x$ and $\text{C}2\text{m}$-LaH$_x$ and $\text{H}/\text{mm}c$-LaH$_x$, in DAC #S. However, this does not affect the conclusion that ternary hexagonal La–Ce–H system exhibits higher $T_c$ than the binary La–H system, both of which are synthesized at the same pressure-temperature conditions.

The upper critical magnetic field

To further confirm the superconductivity and study the upper critical field $H_{c2}(0)$ of the synthesized superconducting phases, we have applied an external magnetic field to different DACs from 150 GPa to 88 GPa (Fig. 3). We used the $T_{c\text{–med}}$ that can be easily recognized to fit with the Werthamer-Helfand-Hohenberg (WHH) model\textsuperscript{43}, simplified by Baumgartner\textsuperscript{44}. For all the DACs, the $T_c$ decreases with increasing magnetic field, as in all superconductors. The acceptable difference between the cooling and warming cycle is because of the temperature gradient between the temperature sensor and the target sample (Fig. 3c). We tried to apply the field parallel and perpendicular to the culet of DAC #9 and observed the trace of anisotropy effect (Fig. 3b). During the decompression of the La–Ce–H system, the upper critical field at 0 K (obtained by extrapolation using the simplified WHH model) increased from 135 T (150 GPa) to striking 235 T (100 GPa), accompanied by a broadening of the transition width. After the decomposition of (La,Ce)H$_{p_{10}}$, the $H_{c2}(0)$ for the residual phases dropped to ~25 T at 88–102 GPa and became steady in this pressure range. The La–H system shows a similar $H_{c2}(0)$ of 24.5 T at 123 GPa (Fig. S29). The strong enhancement of the upper critical magnetic field in the La–Ce–H system is probably related to the random distribution of La/Ce atoms and local distortion of the H-sublattice induced by it. This situation dramatically shortens the electronic mean free path and pushes the system into the dirty limit (Fig. S38). The H-sublattice then becomes
extremely unstable and distorted near the decomposition pressure of the polyhydride.

**Discussion**

We further analyzed the dependence of superconducting state parameters by fitting the experimental $R(T)$ data (see Supplementary Information for analysis). The fitted $\theta_D$ and $\omega_{ph}$ remain around 700–900 K at 90–150 GPa, suggesting the need of relatively high $\lambda = 2$–3 to ensure that the observed $T_c$ is above 160 K. Rather high $\lambda$ corresponds to the weakly ordered, soft, and highly defective structure of this superconducting La–Ce–H phase. The high-$T_c$ (La,Ce)H$_{9-10}$ can be preserved down to about 100 GPa and becomes dynamically unstable at lower pressures. However, we cannot rule out the possibility that (La,Ce)H$_{9-10}$ is metastable in our studied pressure range, considering that both LaH$_{9-10}$ and CeH$_{9-10}$ phases are not dynamically and thermodynamically stable at pressures below 150 GPa according to the predicted convex hull. The configurational entropy may contribute significantly to the stability at high temperatures because of the disordered state and laser heating to over a thousand kelvins. Since it is hard to provide direct evidence on the influence of configurational entropy in the experiment, we have resorted to theoretical calculations. We calculated the dynamical stability (Fig. 4a and Supplementary Fig. S42) and superconducting properties of periodic structure (Supplementary Fig. S40 and Table S4) with different Ce content at 120 GPa with the harmonic approximation. All hexagonal LaH$_{9-10}$, La$_3$CeH$_{36}$, and LaH$_8$ phases showed imaginary phonon frequencies, while adding Ce decreased the extent of this instability. This indicates that adding Ce is beneficial for stabilizing this phase. At the same time, we should also consider the effects of anharmonicity and configurational entropy on the stabilization (Fig. 4d). In order to calculate the harmonic Eliashberg functions, soft modes $<$5 THz were excluded. We obtained a relatively smooth Eliashberg function of La$_3$CeH$_{36}$ similar to what was reported in the disordered or amorphous superconductors before. However, the calculated $T_c$ (131 K, Supplementary Fig. S40c) is about 50 K lower than the experimental value (185 K) at 120 GPa. The random mixing may non-trivially affect the structure, chemical bonding, and electronic properties. Currently, theoretical calculations of La–Ce–H cannot provide an accurate prediction for this disordered system. The problem is in the computational complexity of calculations of ternary systems with variable element concentrations appearing due to the need to use unit cells with a large number of atoms (>40) and limited density of $k$ and q-meshes. The synthesized $P6_3/mmc$-(La,Ce)H$_{9-10}$ can be viewed as a high-entropy superconducting alloy composed of a distorted H-sublattice and randomly substituted metal sublattice analogous to the high-entropy ceramics. Based on this, the La–Ce alloy can be replaced by various related high-entropy reactants to further improve the stability and superconducting properties.

In conclusion, we successfully synthesized the HTSC ternary La–Ce–H (La: Ce = 2.5–3.5) and binary La–H compounds at pressures
lower than 130 GPa. \( T_c \) of hcp-(La,Ce)H\(_{10.76}\) was preserved to 176 K at about 100 GPa, which meant a high figure of merit (\( S = 1.62 \)) comparable to \( \text{Fm3m-LaH}_{16} \). Besides, the extrapolation of the upper critical magnetic field gives \( H_{c2}(0) = 235 \text{ T} \) at 100 GPa, the highest value for polyhydrides obtained so far by fitting with the simplified WHH formula. For comparison, \( T_c \) of the binary La–H phase was detected as 112 K at 129 GPa, decreased to 84 K at 118 GPa, and then increased to above 100 K at 78 GPa. Combining the analysis of experimental data and theoretical calculations, we think that the disordered state in the La–Ce–H system contributes to the giant enhancement of the superconducting \( T_c \) and \( H_{c2}(0) \) compared with binary La–H and Ce–H systems. Importantly, we propose a new strategy for searching for HTSC hydrides at moderate pressure that uses multiple appropriate elements to maximize the configurational entropy of clathrate structures.

**Methods**

Experimental details

The La–Ce alloys were prepared using the multigang magnetron sputtering (Supplementary Fig. S1). We sputtered the La (99.9%) and Ce (99.9%) metals simultaneously to the glass slide using DC and RF power supplies, respectively. The Ar pressure was 1.5 Pa, and both targets were pre-sputtered to remove the surface oxides. The La–Ce alloy was further characterized using the scanning electron microscope (SEM) Regulus 8100, equipped with the energy-dispersive X-ray spectroscopy (EDX). The La–Ce alloys for high-pressure experiment were located on the glass slide very close to the SEM-characterized area and kept strictly in the glove box (O\(_2\) < 0.01 ppm, H\(_2\)O < 0.01 ppm).

Normal type-1a diamonds with 60–150 \( \mu \text{m} \) culets single-beveled to 250–300 \( \mu \text{m} \) were used; the pressure was measured according to the Raman vibrational edge of diamond using Akahama’s calibration\(^{33,34}\). To protect the electrodes during decomposition, we tested the nanopolycrystalline diamonds (NPD) without bevels in some runs (Supplementary Fig. S31). Because of the high fluorescence background of the NPDs, their Raman edge cannot be distinguished with 532 nm laser. Therefore, the NPDs (200–300 \( \mu \text{m} \) culet) were combined with the normal diamonds, and the electrodes were set on the NPD side. The indention of a tungsten gasket was insulated using c-BN/epoxy and normal diamonds, and the electrodes were set on the NPD side. The and the bevel part was

Data availability

The authors declare that the main data supporting our findings of this study are contained within the paper and Supplementary Information. Because the current database contains many other miscellaneous items, we cannot sort them out in a short time. All other relevant data are available from the corresponding author upon request.

**Code availability**

USPEX code is free for academic use and available after registration at https://uspx-team.org/en. Quantum ESPRESSO code is free for academic use and available after registration at https://www.quantum-espresso.org/. VASP code is available for download on the developer page: https://www.vasp.at/. The processing of the Quantum ESPRESSO calculation was carried out using the code available on GitHub: https://github.com/GitGreg228/a2f.

**References**

1. Li, Y., Hao, J., Liu, H., Li, Y. & Ma, Y. The metallization and superconductivity of dense hydrogen sulfide. J. Chem. Phys. 140, 174712 (2014).
2. Duan, D. et al. Pressure-induced metallization of dense (H\(_2\)S)\(_2\)H\(_2\) with high-\( T_c \) superconductivity. Sci. Rep. 4, 6968 (2014).
3. Drozdov, A. P., Eremets, M. I., Troyan, I. A., Ksenofontov, V. & Shylin, S. I. Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system. Nature 525, 73 (2015).
4. Einaga, M. et al. Crystal structure of the superconducting phase of lanthanum hydride. Nat. Phys. 12, 835–838 (2016).
5. Huang, X. et al. High-temperature superconductivity in sulfur hydride evidenced by alternating-current magnetic susceptibility. Natl Sci. Rev. 6, 713–718 (2019).
6. Liu, H. Y., Naumov, I. I., Hoffmann, R., Ashcroft, N. W. & Hemley, R. J. Potential high-\( T_c \) superconducting lanthanum and yttrium hydrides at high pressure. Proc. Natl Acad. Sci. USA 114, 6980–6985 (2017).
7. Geballe, Z. M. et al. Synthesis and stability of lanthanum superhydrides. Angew. Chem. Int. Ed. 57, 688–692 (2018).
8. Somayazulu, M. et al. Evidence for superconductivity above 260 K in lanthanum superhydride at megabar pressures. Phys. Rev. Lett. 2020, 122, 027001 (2019).
9. Drozdov, A. P. et al. Superconductivity at 250 K in lanthanum hydride under high pressures. Nature 569, 528–531 (2019).
10. Snider, E. et al. Synthesis of Yttrium Superhydride Superconductor with a Transition Temperature up to 262 K by Catalytic Hydrogenation at High Pressures. Phys. Rev. Lett. 120, 117003 (2021).
11. Kong, P. et al. Superconductivity up to 243 K in the yttrium-hydrogen system under high pressure. Nat. Commun. 12, 5075 (2021).
12. Troyan, I. A. et al. Anomalous high-temperature superconductivity in YH\(_6\). Adv. Mater. 33, 2006832 (2021).
13. Li, X. et al. Polyhydride Ce\(_7\)H\(_{10}\) with an atomic-like hydrogen clathrate structure. Nat. Commun. 10, 3461 (2019).
24. Semenok, D. V. et al. Effect of Magnetic Impurities on Superconductivit

23. Semenok, D. V. et al. Superconductivity at 253 K in lanthanum hydride.

32. Jeon, H., Wang, C., Yi, S. & Cho, J.-H. Origin of enhanced chemical properties.

30. Peng, F. et al. Hydrogen clathrate structures in rare Earth hydrides.

34. Gschneidner, K. A. & Calderwood, F. W. in Handbook of the Physics and Chemistry of Rare Earths (Elsevier, 1986).

15. Chen, W. et al. High-temperature superconducting phases in cerium hydrides.

26. Errea, I. et al. Quantum crystal structure in the 250-kelvin superconducting lanthanum hydride.

16. Li, Z. et al. Superconductivity above 200 K discovered in superconducting lanthanum hydride.

19. Xie, H. et al. Hydrogen pentagraphenelike structure stabilized by hafnium: a high-temperature conventional superconductor.

20. Di Cataldo, S., Heil, C., Von Der Linden, W. & Boeri, L. LaBH8: a low-pressure superconducting lanthanum hydride. Preprint at https://arxiv.org/abs/2109.05199 (2022).

25. Liu, H. et al. Dynamics and superconductivity in compressed lanthanum superhydride.

27. Struzhkin, V. et al. Superconductivity in La and Y hydrides: remaining questions to experiment and theory. Matter. 5, 028201 (2020).

28. Sun, D. et al. High-temperature superconductivity on the verge of a structural instability in lanthanum superhydride. Nat. Commun. 12, 6863 (2021).

30. Peng, F. et al. Hydrogen clathrate structures in rare Earth hydrides at high pressures: possible route to room-temperature superconductivity. Phys. Rev. Lett. 119, 107001 (2017).

31. Li, B. et al. Predicted high-temperature superconductivity in cerium hydrides at high pressures. J. Appl. Phys. 126, 235901 (2019).

32. Jeon, H., Wang, C., Yi, S. & Cho, J.-H. Origin of enhanced chemical precompression in cerium hydride CeH3. Sci. Rep. 10, 16878 (2020).

33. King, E. & Harris, I. R. High pressure resistance measurements of some lanthanum-cerium alloys. J. Less Common Met. 27, 51–63 (1972).

34. Gschneidner, K. A. & Calderwood, F. W. in Handbook of the Physics and Chemistry of Rare Earths (Elsevier, 1986).

35. Schiwek, A., Porsch, F. & Holzapfel, W. B. Phase Diagrams for Cerium – Lanthanide Alloys under Pressure (2002).

36. Bi, J. et al. Giant enhancement of superconducting critical temperature in substitutional alloy (La0.5Ce0.5)H3. Nat. Commun. 13, 5952 (2022).

37. Ge Huang, T. L. et al. Synthesis of superconducting phase of La0.5Ce0.5H10 at high pressures. Preprint at https://arxiv.org/abs/2208.03519 (2022).

38. Haberl, B., Donnelly, M.-E., Molaison, J. J., Guthrie, M. & Boehler, R. Methods for neutron diffraction studies on hydride superconductors and other metal hydrides. J. Appl. Phys. 130, 215901 (2021).

39. Meier, T. et al. Pressure-induced hydrogen-hydrogen interaction in metallic FeH revealed by NMR. Phys. Rev. X 9, 031008 (2019).

40. Monserrat, B., Ashbrook, S. E. & Pickard, C. J. Nuclear magnetic resonance spectroscopy as a dynamical structural probe of hydrogen under high pressure. Phys. Rev. Lett. 122, 6 (2019).

41. Chen, D., Gao, W. & Jiang, Q. Distinguishing the structures of high-pressure hydrides with nuclear magnetic resonance spectroscopy. J. Phys. Chem. Lett. 11, 9439–9445 (2020).

42. Meier, T. et al. Proton mobility in metallic copper hydride from high-pressure nuclear magnetic resonance. Phys. Rev. B 102, 165109 (2020).

43. Werthamer, N. R., Helfand, E. & Hohenberg, P. C. Temperature and purity dependence of the superconducting critical field, Hc2. Ill. Electron spin and spin-orbit effects. Phys. Rev. 147, 295–302 (1966).

44. Baumgartner, T. et al. Effects of neutron irradiation on pinning force scaling in state-of-the-art Nb3Sn wires. Supercond. Sci. Technol. 27, 015005 (2014).

45. Manzoor, A., Pandey, S., Chakraborty, D., Philpot, S. R. & Aidhy, D. S. Entropy contributions to phase stability in binary random solid solutions. npj Comput. Mater. 4, 47 (2018).

46. Bergmann, G. Amorphous metals and their superconductivity. Phys. Rep. 27, 159–185 (1976).

47. Yao, Y. et al. High-entropy nanoparticles: Synthesis-structure-property relationships and data-driven discovery. Science 376, eabn3103 (2022).

48. Oses, C., Toher, C. & Curtarolo, S. High-entropy ceramics. Nat. Rev. Mater. 5, 295–309 (2020).

49. Baer, B. J., Chang, M. E. & Evans, W. J. Raman shift of stressed diamond anvils: pressure calibration and culet geometry dependence. J. Appl. Phys. 104, 034504 (2008).

50. Akahama, Y. & Kawamura, H. Pressure calibration of diamond anvil gauge to 310 GPa. J. Appl. Phys. 100, 043516 (2006).

51. Gao, C. et al. Accurate measurements of high pressure resistivity in a diamond anvil cell. Rev. Sci. Instrum. 76, 083912 (2005).

52. Prescher, C. & Prakashenka, V. B. DIOPTAS: a program for reduction and analysis of high pressure nuclear magnetic resonance data. High. Press. Res. 35, 223–230 (2015).

53. Petliček, V., Dušek, M. & Palatinus, L. Crystallographic computing system JANA2006: general features. Z. Kristallogr. Cryst. Mater. 229, 345–352 (2014).

54. Le Bail, A., Duroux, H. & Fourquet, J. L. Ab-initio structure determination of LiSbWO6 by X-ray powder diffraction. Mater. Res. Bull. 23, 447–452 (1988).

55. Giannozzi, P. et al. QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials. J. Phys. Condens. Matter 21, 395502 (2009).

56. Giannozzi, P. et al. Advanced capabilities for materials modelling with QUANTUM ESPRESSO. J. Phys. Condens. Matter 29, 465901 (2017).

57. Baroni, S., de Gironcoli, S., Dal Corso, A. & Giannozzi, P. Phonons and related crystal properties from density-functional perturbation theory. Rev. Mod. Phys. 73, 515–562 (2001).

58. Hartwigsen, C., Goedecker, S. & Hutter, J. Relativistic separable dual-space Gaussian pseudopotentials from H to Rn. Phys. Rev. B 58, 3641–3662 (1998).

59. Goedecker, S., Teter, M. & Hutter, J. Separable dual-space Gaussian pseudopotentials. Phys. Rev. B 54, 1703–1710 (1996).

60. Kawamura, M., Oghda, Y. & Tsumeyuki, S. Improved tetrahedron method for the Brillouin-zone integration applicable to response functions. Phys. Rev. B 89, 094515 (2014).

61. Pickard, C. J., Errea, I. & Eremets, M. I. Superconducting hydrides and hydrides under pressure. Annu. Rev. Condens. Matter Phys. 11, 57–76 (2020).
Acknowledgements
This work was supported by the National Key R&D Program of China (Grant No. 2022YFA1405500), the National Natural Science Foundation of China (Grants No. 52072188, and No. 11974133), the Program for Changjiang Scholars and Innovative Research Team in University (Grant No. IRT_15R23), Zhejiang Provincial Science and technology innovation Team (2021R01004), and Jilin Provincial Science and Technology Development Project (20210509038RQ). A.R.O. thanks the Russian Science Foundation (grant 19-72-30043). The authors thank the staff of the Shanghai Synchrotron Radiation Facility for their help during the synchrotron XRD measurements.

Author contributions
X.H. and T.C. conceived this project. W.C., S.C., K.Z., X.H., and T.C. performed the experiments. D.V.S. and D.Z. prepared the theoretical calculations. W.C., X.H., and D.V.S. analyzed the data. W.C., X.H. D.V.S., and A.R.O. wrote and revised the paper. All the authors discussed the results and offered useful inputs.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-023-38254-6.

Correspondence and requests for materials should be addressed to Xiaoli Huang or Tian Cui.

Peer review information Nature Communications thanks the anonymous reviewer(s) for their contribution to the peer review of this work.

Reprints and permissions information is available at http://www.nature.com/reprints

Publisher’s note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons license, and indicate if changes were made. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in a credit line to the material. If material is not included in the article’s Creative Commons license and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/.

© The Author(s) 2023