In-silico synthesis of novel lowest-pressure high-$T_c$ ternary superhydrides

Roman Lucrezi, Simone Di Cataldo, Wolfgang von der Linden, Lilia Boeri, and Christoph Heil

Institute of Theoretical and Computational Physics, Graz University of Technology, NAWI Graz, 8010 Graz, Austria
 Dipartimento di Fisica, Sapienza Università di Roma, 00185 Roma, Italy

We report the theoretical prediction of two novel high-performing hydride superconductors BaSiH$_8$ and SrSiH$_8$. They are thermodynamically stable above pressures of 130 and 174 GPa, respectively, and remain dynamically stable down to much lower pressures ($p_c = 3$ and 27 GPa), with superconducting $T_c$’s of 71 and 126 K, respectively.

INTRODUCTION

The unexpected discovery of high-temperature superconductivity in high-pressure hydrides has revamped the hopes of ending the century-long quest for superconductivity at ambient conditions [1–12]. In less than five years, superhydrides have established higher and higher records for critical temperatures, starting with SH$_3$ (203 K) [3], LaH$_{16}$ (265 K) [4, 5], and C-S-H (288 K) [9].

While it is very exciting to strive for new superconductors with even higher $T_c$’s attaining hot superconductivity [13–15], lowering the required stabilization pressures while retaining $T_c$ above the boiling point of nitrogen (77K) is of even greater importance [16–21]. In fact, the discovery of a conventional (s-wave) superconductor with these properties would open up a wide array of technological applications in key strategical sectors such as energy conservation, climate change, and medicine.

After exhausting the search space of binary hydrides, the focus of superhydride research is rapidly shifting to ternary hydrides, where the parameter space is much larger [16]. We have recently predicted, for example, that a ternary hydride with LaBH$_8$ composition could form an $Fm\bar{3}m$ ternary sodalite-like clathrate (SLC) structure and remain stable down to a critical pressure $p_c \sim 35$ GPa, with a $T_c = 126$ K [15, 18]. Our findings were later confirmed by independent studies on the La-B-H system [20, 22]. The predicted $p_c$ of LaBH$_8$ is a factor of four lower than in binary hydrides, where $p_c$’s are in the Megabar range, but still too high to envision any large-scale applications for this particular compound. However, through the identification of the $Fm\bar{3}m$ XYH$_8$ structural template, the discovery of LaBH$_8$ paved the road to the study of a whole new family of potential high-$T_c$ ternary hydrides, where $p_c$ and $T_c$ may be improved even further.

In this work, using first-principles methods for crystal structure prediction and superconductivity, we identify two novel high-$T_c$ rare-earth/silicon superhydrides, BaSiH$_8$ and SrSiH$_8$. We predict that both compounds will spontaneously form in the $Fm\bar{3}m$ $XYH_8$ structure at high pressures (130 and 174 GPa, respectively), and remain dynamically stable down to much lower pressures ($p_c = 3$ and 27 GPa), with superconducting $T_c$’s of 71 and 126 K, respectively.

We also succeed in deriving a rigorous limit for the stability of BaSiH$_8$, calculating explicitly the energy barrier protecting the metastable $Fm\bar{3}m$ structure from decomposition as a function of pressure, using the Variable-Cell Nudged Elastic Band (VCNEB) method [23]. We find that, indeed, $Fm\bar{3}m$ BaSiH$_8$ should remain metastable at pressures well below 130 GPa. However, the kinetic critical pressure $p_{kin}$ determined by the energy barrier is significantly higher (30 GPa) than the value estimated from anharmonic lattice dynamics. Similar discrepancies between dynamical and kinetic pressure of stability may explain the systematic underestimation of the predicted pressures of (meta)stability found in other high-pressure hydrides compared to experiments [11, 24, 26].

RESULTS AND DISCUSSION

Initial Screening: BaSiH$_8$ and SrSiH$_8$ were first identified through a high-throughput screening of possible substitutions of La and B by neighbouring elements into the $Fm\bar{3}m$ $XYH_8$ structure, as shown in Fig. 1(a). In this structure – Fig. 1(b) – hydrogen atoms form rhombicuboctahedral cages around lanthanum; boron, being much smaller, fills the six cubic voids surrounding the cages. This structure permits a very efficient realisation of the chemical precompression mechanism proposed by Neil Ashcroft and observed in SLC binary hydrides [2, 24, 27]. To elaborate: In addition to the large central atom (La), also the smaller atom in the interstitials (B) exerts an additional chemical pressure on the hydrogen sublattice, lowering the minimum stabilization pressure $p_c$ [18].
We assumed that, considering different combinations of large \((X)\) and small \((Y)\) atoms, the superconducting properties of \(\text{LaBH}_8\) could be improved even further. Aiming at identifying compounds with low critical pressures, we performed structural relaxations at 50 GPa for all combinations of \(X = [\text{K}, \text{Rb}, \text{Cs}, \text{Ca}, \text{Sr}, \text{Ba}, \text{Sc}, \text{Y}, \text{and La}]\) and \(Y = [\text{B}, \text{Al}, \text{Ga}, \text{C}, \text{Si}, \text{and Ge}]\) elements in the \(\text{Fm}\overline{3}m\) \(XYH_8\) template, and evaluated the dynamical stability of the resulting compounds by calculating the harmonic phonon dispersions on a \(4\times4\times4\) grid in reciprocal space.

As shown in Fig. S2 of the Supplementary Material (SM) \[28\], the lattice constants of the relaxed structures exhibit an almost perfect linear dependence on the sum of the \(X\) and \(Y\) atomic radii \(R = R_X + R_Y\), with slopes determined by the total number of valence electrons \(N_e = N_X + N_Y\). Out of the 54 compounds investigated, only three compounds with \(N_e = 6\) – green ticks in Fig. 1(a) – are dynamically stable at 50 GPa: Apart from the already reported \(\text{LaBH}_8\) \[13\], we also found two silicon hydrides, \(\text{BaSiH}_8\) and \(\text{SrSiH}_8\). Both compounds exhibit a larger \(R\) than \(\text{LaBH}_8\), suggesting that chemical precompression in both compounds will be more efficient, and hence their \(p_c\) may be lower. In particular, \(\text{BaSiH}_8\), where \(R\) is considerably larger than in the other two compounds, looks very promising in this respect. Indeed, our harmonic phonon calculations yield \(p_c = 5\) and 30 GPa for \(\text{BaSiH}_8\) and \(\text{SrSiH}_8\), respectively, lower than the \(p_c = 40\) GPa in \(\text{LaBH}_8\).

**Convex Hulls and Phase Diagrams:** Having established that \(\text{BaSiH}_8\) and \(\text{SrSiH}_8\) are dynamically stable in the \(\text{Fm}\overline{3}m\) structure close to ambient pressure, we determined the pressures at which the two compounds may spontaneously crystallize in the \(\text{Fm}\overline{3}m\) structure starting from appropriate precursors. To this end, we computed the full ternary convex hulls for the two ternary \(X\text{-Si-H}\) systems at ambient pressure and at 100 GPa, employing \(\text{ab-initio}\) variable-composition evolutionary crystal structure prediction methods as implemented in \text{Us-}

In the \(\text{Ba-Si-H}\) system, the \(\text{BaSiH}_8\) composition is thermodynamically stable, i.e., it lies on the hull at 100 GPa in a distorted \(P1\) phase. The \(\text{Fm}\overline{3}m\) phase is slightly higher in enthalpy (32 meV/atom), but becomes enthalpically favourable above 130 GPa. In the \(\text{Sr-Si-H}\) system at 100 GPa, according to our calculations, \(\text{SrSiH}_8\) should decompose into \(\text{SrSi}_6\) and \(\text{SrSi}_{12}\). However, the \(\text{Fm}\overline{3}m\) phase lies only 54 meV/atom above the convex hull. From a comparison of the formation enthalpies, we predict that it should become stable at 174 GPa.

**Anharmonic lattice dynamics:** Preliminary phonon calculations in the harmonic approximation indicated that both \(\text{BaSiH}_8\) and \(\text{SrSiH}_8\) experience remarkable phonon softening at lower pressures. More specifically, we find soft phonon modes, mainly at \(\Gamma\) \((T_{2g}, E_g, \text{and } A_{1g})\) and \(X\) \((E_g, \text{and } A_{2g})\) in \(\text{SrSiH}_8\), as shown in Fig. 2. Eventually, imaginary frequencies appear for the \(E_g\) mode at \(X\) below 5 GPa for \(\text{BaSiH}_8\) and
Soft-mode behaviour, associated with strong anharmonicity, has been reported for many hydrogen-rich materials [24, 25, 31–33]. Anharmonic lattice effects have been shown to crucially affect the range of dynamical stability, phonon frequencies and eigenvectors of superhydrides, and their inclusion is essential to obtain accurate estimates of these quantities.

In order to account for anharmonic effects on the phonon dispersions of BaSiH$_8$ and SrSiH$_8$, we evaluated the adiabatic potential energy surface (APES) for every soft mode of a $2\times2\times2$ wave-vector grid, which includes explicitly the special points $\Gamma$, $L$, and $X$, and solved the resulting Schrödinger equations. (More information can be found in the method section, Sec. S2C of the SM [28], and Ref. [34].)

The harmonic and the anharmonic phonon dispersions for BaSiH$_8$ and SrSiH$_8$ close to their critical pressures of stability are shown as dashed and full lines in Fig. 2. Anharmonicity causes a considerable hardening of the $T_{2g}$ (at $\Gamma$) and $E_g$ (at $\Gamma$ and $X$) modes, leading to a decrease of the critical pressure of dynamical stability $p_c$: Taking this hardening into account, SrSiH$_8$ is stable down to a pressure of 27 GPa and, even more excitingly, BaSiH$_8$ down to 3 GPa.

Electronic Structure and Superconductivity: Having determined that both BaSiH$_8$ and SrSiH$_8$ remain dynamically stable close to ambient pressure, we are left with the question whether at these relatively low pressures they can still be considered superhydrides. A first positive indication comes from the analysis of their electronic structure.

In Fig. 3 we show the electronic dispersions and DOS for BaSiH$_8$ (left) and SrSiH$_8$ (right), at 5 and 30 GPa, respectively. Despite a scaling of the total bandwidth due to the different pressures, the band structures are qualitatively very similar. The Fermi level is located just above a large DOS shoulder; in this region and up to $-5.5$ eV for BaSiH$_8$ ($-3.5$ meV for SrSiH$_8$), the bands
are of purely hydrogen character. This is an important prerequisite for high-$T_c$ conventional superconductivity in hydrides, since it can imply a strong electron-phonon ($ep$) coupling between electronic states to the light hydrogen sublattice [35].

The resulting Fermi surface topology is the same in the two compounds: a large, spheric-like electron pocket is centered around the Brillouin zone center, while a more complicated hole-like network extends around the faces of the Brillouin zone, enclosing the $X$ and $W$ points (see the insets of Fig. 4).

These qualitative electronic structure arguments are confirmed by actual calculations of the superconducting properties. Fig. 4 shows the energy distribution of the superconducting gap $\Delta$ as a function of temperature $T$ for BaSiH$_8$ at 5 GPa pressure and for SrSiH$_8$ at 30 GPa, obtained by solving the anisotropic Migdal-Eliashberg (ME) equations on a $30 \times 30 \times 30$ $k$- and $q$-grid using the anharmonically corrected phonon dispersions with the Epw code [36, 37]. (The superconducting properties at all other considered pressures and further computational details are provided in the SM [28].) We observe two distinct superconducting gaps: The inset of the figure shows that large $\Delta$ values correspond to the $\Gamma$-centered, spherical electron pocket, while lower values occur on the hole-like tubular network around the $X$ and $W$ points. The superconducting critical temperatures are predicted to be 71 and 126 K for BaSiH$_8$ and SrSiH$_8$, respectively.

Further details on the origin of the remarkable $T_c$’s of BaSiH$_8$ and SrSiH$_8$ can be obtained from an analysis of the distribution of their $ep$ coupling over phonon branches. In panel (b) of Fig. 2, the mode and wavevector resolved $ep$ coupling $\lambda_{q,\nu}$ are overlayed onto the phonon dispersions as fat bands; panel (d) of the same figure shows the Eliashberg spectral function $\alpha^2 F(\omega)$, and the total frequency-dependent $ep$ coupling parameter $\lambda(\omega)$.

The figure clearly shows that the largest contributions to the total coupling come from low-energy modes; a substantial fraction is associated to soft phonon modes around $\Gamma$ and $X$. In the case of BaSiH$_8$, for example, we estimate that the phonons related to $\Gamma - T^2_g$ contribute roughly 40% to the total $\lambda$ and around $X - E^g$ about 35%. These modes are of purely hydrogen character and their patterns can be more easily visualized in terms of the H cubes around Si: The $T^2_g$ mode at $\Gamma$ is a centrosymmetric stretching and squeezing along one of the space diagonals of the H cube, while the $E^g$ mode at $X$ is a non-deforming rotation around a face normal of the H cube, with alternating phase in two neighbouring cells.

Fig. 5 shows the evolution of the superconducting properties of BaSiH$_8$ and SrSiH$_8$ as a function of pressure from the lowest dynamical stability pressure up to 100 GPa; open circles and solid lines correspond to anharmonic re-
FIG. 6. Minimum energy path (MEP) for BaSiH$_8$ at various pressures, calculated using the VCNEB method. Each point represents an individual crystal structure (image) along the path. Particularly relevant intermediate states are shown.

sults, while dashed lines refer to harmonic values. Interestingly, $T_c$ is approximately constant with pressure – panel (a); this is true even close to $p_c$, where one would expect a strong deviation due to anharmonicity. An inspection of panels (b) and (c) shows that this happens because of the compensating effect of phonon hardening on $\omega_{\log}$ and $\lambda$. The effect is more marked in SrSiH$_8$ and in BaSiH$_8$ close to the instability point, where harmonic and anharmonic dispersions diverge the most. For SrSiH$_8$ at 30 GPa, we find an anharmonic (harmonic) $\lambda$ of 3.21 (3.96) and an $\omega_{\log}$ of 33.1 meV (22.3 meV), and for BaSiH$_8$ at 5 GPa, we find a $\lambda$ of 1.61 (2.55) and an $\omega_{\log}$ of 33.3 meV (23.7 meV).

Metastability: In all theoretical studies of high-pressure hydrides performed so far, the range of metastability of high-pressure phases has been assumed to coincide with the range of dynamical stability, eventually including quantum corrections to lattice dynamics. However, a comparison of these predictions and available experimental data suggests that theoretical estimates of the critical pressure are systematically lower than experimental values [11, 24–26].

Indeed, dynamical stability is only a prerequisite for thermodynamic metastability. The latter is determined by the existence of an energy (enthalpy) barrier protecting a metastable phase from decomposition into other phases [38]. Attempts to quantitatively estimate the barrier height are extremely rare. For $Fm\bar{3}m$ BaSiH$_8$, where the dynamical instability pressure $p_c$ is extremely close to ambient pressure, this issue is obviously crucial, since the presence of a sufficiently high barrier could be considered the definitive proof of synthesizability.

In this work, we estimated the barrier height using the VCNEB as implemented in the USPEX code [28]. In this method, a number of intermediate structures (images) are created between the metastable structure of interest and the ground-state structure at a given pressure; elastic forces between each image are added to the "physical" forces, and the whole chain of images thus created is finally relaxed to obtain the energy/enthalpy profile of the transition.

We ran VCNEB simulations for BaSiH$_8$ at six different pressures: 10, 25, 35, 50, 100, and 200 GPa. As end-members for the VCNEB path, we chose the $Fm\bar{3}m$ phase and a $P1$ phase, identified as the ground-state structure at 10 GPa through a fixed-composition structural search. Due to its large unit cell and low symmetry, it can be assumed that the $P1$ phase, relaxed at the different pressures, approximates quite well the true ground-state of the system; indeed, at all pressures the $P1$ BaSiH$_8$ phase lies only a few meV above the hull. In practice, analyzing the VCNEB images in Fig. 6, we observe that the transition from the $Fm\bar{3}m$ to the $P1$ phase corresponds to the decomposition of BaSiH$_8$ into BaSiH$_6$ + H$_2$, with the expulsion of hydrogen in molecular form.

The potential barrier, shown in Fig. S5 [28], decreases with pressure from 153 meV/atom at 100 GPa to 57 meV/atom at 35 GPa; a sharp transition is visible at 25 GPa, where the kinetic barrier abruptly drops to 9 meV/atom. Although a small barrier survives down to 10 GPa, this sharp decrease is the signature of an impending kinetic instability, i.e., the metastable state will be short-lived and most likely will not be observed in experiments.

Combining the convex hull results in Fig. 1 with the VCNEB analysis, we can argue with confidence that an $Fm\bar{3}m$ BaSiH$_8$ phase could be synthesized above 100 GPa, and retained down to ~ 30 GPa, where a clearly visible enthalpy barrier exists. At lower pressures, metastable $Fm\bar{3}m$ BaSiH$_8$ will decompose, even though (anharmonic) lattice dynamics calculations predict it to be stable. Hence, kinetic stability poses a stricter bound for synthesizability than dynamical stability.
**Discussion:** In summary, our study shows that indeed the superconducting properties of the \( XYH_8 \) template can be optimized by a suitable choice of the \( X \) and \( Y \) elements. The two silicides identified in this work represent an improvement compared to \( \text{LaBH}_8 \); for \( \text{SrSiH}_8 \) we predict a dynamical stability pressure \( p_c \) of 27 GPa, with a \( T_c \) of 127 K. Using the figure of merit \( S \) introduced by the authors of Ref. [16], this means passing from 1.3 in \( \text{H}_3\text{S} \) and \( \text{LaA}_10 \) to 2.2 in \( \text{LaBH}_8 \) to 2.7 in \( \text{SrSiH}_8 \). Even more remarkably, \( \text{BaSiH}_8 \) is predicted to be dynamically stable down to pressures of 3 GPa, with a critical temperature of 71 K, which is substantially higher than all established [39] and claimed [40] experimental \( T_c \) records for conventional superconductors at ambient pressure.

VCNEB calculations also demonstrate that the shape of the potential energy landscape of \( \text{BaSiH}_8 \) is favorable for its synthesis in a wide range of pressures. A possible synthesis path involves synthesis at high pressure (\( p > 130 \) GPa) and/or laser heating, and rapid quenching of the resulting phase to lower pressure, i.e., down to \( \sim 30 \) GPa, where the metastable \( \text{Fm}3\text{m} \) crystal structure is protected by a sizable potential barrier. This defines a kinetic threshold pressure, \( p_{\text{kin}} \), which is substantially higher than \( p_c \).

By defining a concrete method to estimate the synthesizability of a proposed structure, our study sets a new standard for the \textit{ab-initio} design of new superconductors at high pressures, based on the more rigorous concept of kinetic stability, rather than dynamical stability. The existence of a distinct kinetic stability criterion may also be invoked to explain why many long-standing predictions of high-\( T_c \) superconductors have not been realized experimentally [41]–[43].

We strongly believe that the proposed method represents a major step forward towards achieving high-\( T_c \) conventional superconductivity at room pressure.

**METHODS**

Crystal structure prediction runs were carried out using evolutionary algorithms as implemented in the \textit{Uspex} package [29]–[31], [44]; the underlying total energy and structural relaxation calculations were performed using plane-waves and pseudopotentials as implemented in the Vienna \textit{ab-initio} Simulation Package \textit{VASP} [45]. Further computational details are provided in Sec. S2A of the SM [28].

All density-functional theory (DFT) and density-functional perturbation theory (DFPT) calculations of electronic and vibrational properties were carried out using the plane-wave pseudopotential code \textit{QUANTUM ESPRESSO} [46], scalar-relativistic optimized norm-conserving Vanderbilt pseudopotentials (ONCV) [47], and the PBE-GGA exchange and correlation functional [48]. Computational details are provided in Sec. S1A of the SM [28].

The phase transition path between the \( P1 \) and \( \text{Fm}3\text{m} \) \( \text{BaSiH}_8 \) was evaluated using the VCNEB method as implemented in \textit{Uspex} [23]–[29], using variable elastic constants and a variable number of images between the endpoints. The energy and forces were calculated using \textit{VASP}. Further computational details are provided in Sec. S2B of the SM [28].

The interpolation of the \( ep \) matrix elements onto dense wave-vector grids and the subsequent self-consistent solution of the fully anisotropic Migdal-Eliashberg equations were done with \textit{EPW} [37]. Based on our previous work on \( \text{LaBH}_8 \), where we calculated the Morel-Anderson pseudopotential \( \mu^* \) from first principles using \textit{GW} and found consistent values for \( \mu^* \) of about 0.1 [18]–[24], [35], we chose the same value in the current work. Further computational details are provided in Sec. S2D of the SM [28].

Anharmonic corrections to phonon frequencies were obtained by explicitly solving the Schrödinger equation for the APES of every soft mode on a \( 2 \times 2 \times 2 \) wave-vector grid. By calculating and solving 2D APES, we also checked that phonon-phonon interactions for various modes and wave vectors can be neglected in a good first approximation. The interpolation of the real-space force constants obtained on the \( 2 \times 2 \times 2 \) \( q \)-grid was performed using the corresponding harmonic support DFPT solution as implemented in the \textit{CELLCONSTRUCTOR} package [49]. Further computational details are provided in Sec. S2C of the SM [28].

**ACKNOWLEDGMENTS**

This work was supported by the Austrian Science Fund (FWF) Projects No. P 30269-N36 and No. P 32144-N36, the dCluster of the Graz University of Technology and the VSC-4 of the Vienna University of Technology. L.B. acknowledges support from Fondo Ateneo-Sapienza 2017-2019. S.D.C. acknowledges computational Resources from CINECA, proj. IsC90-HTS-TECH_C.

**AUTHOR CONTRIBUTIONS**

R. Lucrezi and S. Di Cataldo performed the calculations, and L. Boeri and C. Heil conceived and supervised the project. All authors contributed to the discussion of the results and participated in preparing the manuscript.

[1] lilia.boeri@uniroma1.it

† Corresponding author; christoph.heil@tugraz.at
I. A. Troyan, D. V. Semenok, A. G. Kvashnin, A. V. W. Chen, D. V. Semenok, X. Huang, H. Shu, X. Li, A. Grockowiak, M. Ahart, T. Helm, W. Coniglio, R. Kumar, M. Somayazulu, Y. Meng, M. Oliff, V. Williams, N. Ashcroft, et al., Hot hydride superconductivity above 550 K, arXiv preprint arXiv:2006.03004 (2020).

[1] N. W. Ashcroft, Metallic hydrogen: A high-temperature superconductor?, Phys. Rev. Lett., 21, 1748 (1968).

[2] N. W. Ashcroft, Hydrogen dominant metallic alloys: High temperature superconductors?, Phys. Rev. Lett. 92, 187002 (2004).

[3] A. Drozdov, M. Eremets, I. Troyan, V. Ksenofontov, and S. I. Shylin, Conventional superconductivity at 203 kelvin at high pressures in the sulfur hydride system, Nature 525, 73 (2015).

[4] A. Drozdov, P. Kong, V. Minkov, S. Besedin, M. Kuzovnikov, S. Mozaafari, L. Balicas, F. Balakirev, D. Graf, V. Prakapenka, et al., Superconductivity at 250 K in lanthanum hydride under high pressures, Nature 569, 528 (2019).

[5] M. Somayazulu, M. Ahart, A. K. Mishra, Z. M. Geballe, M. Baldini, Y. Meng, V. V. Struzhkin, and R. J. Hemley, Evidence for superconductivity above 260 K in lanthanum superhydride at megabar pressures, Phys. Rev. Lett. 122, 027001 (2019).

[6] J. A. Flores-Livas, L. Boeri, A. Sanna, G. Profeta, R. Arita, and M. Eremets, A perspective on conventional high-temperature superconductors at high pressure: Methods and materials, Physics Reports 856, 1 (2020).

[7] Y. Sun, J. Lv, Y. Xie, H. Liu, and Y. Ma, Route to a superconducting phase above room temperature in electron-doped hydride compounds under high pressure, Phys. Rev. Lett. 123, 097001 (2019).

[8] D. V. Semenok, I. A. Troyan, A. G. Ivanova, A. G. Kvasnin, I. A. Kruglov, M. Hanfland, A. V. Sadakov, O. A. Sobolevskiy, K. S. Pervakov, I. S. Lyubutin, et al., Superconductivity at 253 K in lanthanum–yttrium ternary hydrides, Materials Today (2021).

[9] E. Snider, N. Darenbrock-Gammon, R. McBride, M. Debessai, H. Vindana, K. Vencatasamy, K. V. Lawler, A. Salamat, and R. P. Dias, Room-temperature superconductivity in a carbonaceous sulfur hydride, Nature 586, 373 (2020).

[10] I. A. Troyan, D. V. Semenok, A. G. Kvasnin, A. V. Sadakov, O. A. Sobolevskiy, V. M. Pudalov, A. G. Ivanova, V. B. Prakapenka, E. Greenberg, A. G. Gavriliuk, et al., Anomalous high-temperature superconductivity in YHe, Advanced Materials 33, 2006832 (2021).

[11] P. Kong, V. S. Minkov, M. A. Kuzovnikov, A. P. Drozdov, S. P. Besedin, S. Mozaafari, L. Balicas, F. F. Balakirev, V. B. Prakapenka, S. Chariton, D. A. Knyazev, E. Greenberg, and M. I. Eremets, Superconductivity up to 243 K in the yttrium-hydrogen system under high pressure, Nature Comm. 12, 5075 (2021).

[12] W. Chen, D. V. Semenok, X. Huang, H. Shu, X. Li, D. Duan, T. Cui, and A. R. Oganov, High-temperature superconducting phases in cerium superhydride with a Tc up to 115 K below a pressure of 1 megabar, Phys. Rev. Lett. 127, 117001 (2021).

[13] F. Peng, Y. Sun, C. J. Pickard, R. J. Needs, Q. Wu, and Y. Ma, Hydrogen clathrate structures in rare earth hydrides at high pressures: possible route to room-temperature superconductivity, Phys. Rev. Lett. 119, 107001 (2017).

[14] A. Grockowiak, M. Ahart, T. Helm, W. Coniglio, R. Kumar, M. Somayazulu, Y. Meng, M. Oliff, V. Williams, N. Ashcroft, et al., Hot hydride superconductivity above 550 K, arXiv preprint arXiv:2006.03004 (2020).

[15] S. Di Cataldo, W. von der Linden, and L. Boeri, La-X-H hydrides: is hot superconductivity possible?, arXiv preprint arXiv:2106.07266 (2021).

[16] C. J. Pickard, I. Errea, and M. I. Eremets, Superconducting hydrides under pressure, Annual Review of Condensed Matter Physics 11, 57 (2020).

[17] J. Lv, Y. Sun, H. Liu, and Y. Ma, Theory-orientated discovery of high-temperature superconductors in superhydrides stabilized under high pressure, Matter and Radiation at Extremes 5, 068101 (2020).

[18] S. Di Cataldo, C. Heil, W. von der Linden, and L. Boeri, LaBH5: Towards high-Tc, low-pressure superconductivity in ternary superhydrides, Phys. Rev. B 104, L020511 (2021).

[19] A. M. Shipley, M. J. Hutcheson, R. J. Needs, and C. J. Pickard, High-throughput discovery of high-temperature conventional superconductors, Phys. Rev. B 104, 054501 (2021).

[20] Z. Zhang, T. Cui, M. J. Hutcheson, A. M. Shipley, H. Song, M. Du, V. Z. Kresin, D. Duan, C. J. Pickard, and Y. Yao, Design principles for high temperature superconductors with hydrogen-based alloy backbone at moderate pressure, arXiv preprint arXiv:2106.09879 (2021).

[21] S. Di Cataldo, W. Von Der Linden, and L. Boeri, Phase diagram and superconductivity of calcium borohydrides at extreme pressures, Phys. Rev. B 102, 014516 (2020).

[22] X. Liang, A. Bergara, X. Wei, X. Song, L. Wang, R. Sun, H. Liu, R. J. Hemley, L. Wang, G. Gao, et al., Prediction of high-Tc superconductivity in ternary lanthanum borohydrides, Phys. Rev. B 104, 134501 (2021).

[23] G.-R. Qian, X. Dong, X.-F. Zhou, Y. Tian, A. R. Oganov, and H.-T. Wang, Variable cell nudged elastic band method for studying solid–solid structural phase transitions, Comput. Phys. Commun. 184, 2111 (2013).

[24] C. Heil, S. Di Cataldo, G. B. Bachelet, and L. Boeri, Superconductivity in sodalite-like yttrium hydride clathrates, Phys. Rev. B 99, 220502 (2019).

[25] I. Errea, M. Calandra, C. J. Pickard, R. J. Needs, Y. Li, H. Liu, Y. Zhang, Y. Ma, and F. Mauri, Quantum hydrogen-bond symmetrization in the superconducting hydrogen sulfide system, Nature 532, 81 (2016).

[26] M. Einaga, M. Sakata, T. Ishikawa, K. Shimizu, M. I. Eremets, A. P. Drozdov, I. A. Troyan, N. Hirao, and Y. Ohishi, Crystal structure of the superconducting phase of sulfur hydride, Nature Physics 12, 835–838 (2016).

[27] H. Liu, I. I. Naumov, R. Hoffmann, N. Ashcroft, and R. J. Hemley, Potential high-Tc superconducting lanthanum and yttrium hydrides at high pressure, Proceedings of the National Academy of Sciences 114, 6990 (2017).

[28] The Supplementary Material is available at [...]..

[29] C. W. Glass, A. R. Oganov, and N. Hansen, USPEx - evolutionary crystal structure prediction, Comput. Phys. Commun. 175, 713 (2006).

[30] A. R. Oganov, A. O. Lyakhov, and M. Valle, How evolutionary crystal structure prediction works—and why, Acc. Chem. Res. 44, 227 (2011), pMID: 21361336, https://doi.org/10.1021/ar1001318.

[31] I. Errea, F. Belli, L. Monacelli, A. Sanna, T. Koretune, T. Tadano, R. Bianco, M. Calandra, R. Arita, F. Mauri, et al., Quantum crystal structure in the 250-kelvin superconducting lanthanum hydride, Nature 578, 66 (2020).
M. Borinaga, I. Errea, M. Calandra, F. Mauri, and A. Bergara, Anharmonic effects in atomic hydrogen: Superconductivity and lattice dynamical stability, Phys. Rev. B 93, 174308 (2016).

C. Heil and L. Boeri, Influence of bonding on superconductivity in high-pressure hydrides, Phys. Rev. B 92, 060508 (2015).

C. Heil, S. Poncé, H. Lambert, M. Schlipf, E. R. Margine, and F. Giustino, Origin of superconductivity and latent charge density wave in NbS$_2$, Phys. Rev. Lett. 119, 087003 (2017).

C. Heil, G. B. Bachelet, and L. Boeri, Absence of superconductivity in iron polyhydrides at high pressures, Phys. Rev. B 97, 214510 (2018).

E. R. Margine and F. Giustino, Anisotropic Migdal-Eliashberg theory using Wannier functions, Phys. Rev. B 87, 024505 (2013).

S. Poncé, E. Margine, C. Verdi, and F. Giustino, Epw: Electron-phonon coupling, transport and superconducting properties using maximally localized Wannier functions, Comput. Phys. Commun. 209, 116 (2016).

W. Sun, S. T. Dacek, S. P. Ong, G. Hautier, A. Jain, W. D. Richards, A. C. Gamst, K. A. Persson, and G. Ceder, The thermodynamic scale of inorganic crystalline metastability, Sci. Adv. 2, 1 (2016).

J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, Superconductivity at 39K in magnesium diboride, Nature 410, 63 (2001).

A. Bhaumik, R. Sachan, and J. Narayan, High-temperature superconductivity in boron-doped q-carbon, ACS Nano 11, 5351–5357 (2017).

H. Rosner, A. Kitaigorodsky, and W. E. Pickett, Prediction of high $T_c$ superconductivity in hole-doped LiBC, Phys. Rev. Lett. 88, 127001 (2002).

G. Savini, A. C. Ferrari, and F. Giustino, First-principles prediction of doped graphene as a high-temperature electron-phonon superconductor, Phys. Rev. Lett. 105, 037002 (2010).

S. Saha, S. Di Cataldo, M. Amsler, W. von der Linden, and L. Boeri, High-temperature conventional superconductivity in the boron-carbon system: Material trends, Phys. Rev. B 102, 024519 (2020).

A. O. Lyakhov, A. R. Oganov, H. T. Stokes, and Q. Zhu, New developments in evolutionary structure prediction algorithm USPEX, Comput. Phys. Commun. 184, 1172 (2013).

G. Kresse and J. Furthmüller, Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set, Comput. Mater. Sci. 6, 15 (1996).

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

D. R. Hamann, Optimized norm-conserving Vanderbilt pseudopotentials, Phys. Rev. B 88, 085117 (2013).

J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized gradient approximation made simple, Phys. Rev. Lett. 77, 3865 (1996).

L. Monacelli, R. Bianco, M. Cherubini, M. Calandra, I. Errea, and F. Mauri, The stochastic self-consistent harmonic approximation: Calculating vibrational properties of materials with full quantum and anharmonic effects, Journal of Physics: Condensed Matter (2021).