Predicted Hot Superconductivity in $LaSc_2H_{24}$ under Pressure

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The recent theory-driven discovery of a class of clathrate hydrides (e.g., CaH_6, YH_6, YH_9, and LaH_{10}) with superconducting critical temperatures (T_c) well above 200 K shed light on the findings of 'hot' superconductivity above room temperature at high pressure conditions. Recent efforts focus on the search of superconductors among ternary hydrides that accommodate more diverse material types and vast configurations compared to binary hydrides. Through extensively computational searches, we here report the prediction of a new class of thermodynamically stable clathrate superhydride structures consisting of two previously unreported H_{24} and H_{30} hydrogen clathrate cages at megabar pressures, among which $LaSc_2H_{24}$ shows potential hot superconductivity with a theoretical T_c of ∼330 K at 300 GPa. The superconductivity is attributed to an unusually large hydrogen-derived density of states at the Fermi level arising from the newly reported peculiar H_{30} as well as H_{24} cages. Our current theory on the hot superconductor via the introduction of Sc into La-H system sheds light on future design of other superconductors among stable ternary clathrate superhydrides.

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The quest for materials that superconduct at and even above room temperature remains one of the most important research topics in condensed matter and materials physics. The discovery of covalent H_3S (T_c = 203 K at 155 GPa) [1, 2] and a class of clathrate binary hydrides with high-T_c values in the range of 215-260 K at high pressures, including CaH_6 [3, 4], YH_6 [5, 6], YH_9 [6, 7], and LaH_{10} [8–10], all of which inspired by theoretical predictions assuming conventional superconductivity [11–15], has sparked great interest to search for room-temperature superconductivity among compressed hydrides. The first of these theoretically predicted superconducting clathrate hydrides CaH_6 (a predicted T_c of 220-235 K at 150 GPa) consisting of sodalite H_{24} cages [13], was recently confirmed experimentally in two independent studies [3, 4]. These exciting findings have encouraged the search for other superconducting hydrides with even higher critical temperatures among clathrate-like hydrogen structures.

Recently, attention has been focused on ternary hydrides, since they provide much wider range of candidate compositions and structures that may exhibit high-temperature superconductivity [16–21]. For example, our predicted metastable clathrate hydride Li_2MgH_{16} with the highest T_c calculated to date (∼473-351 K at 250-300 GPa) [22] was designed through a strategy of introducing additional electrons from guest metal Li into the host H_2-based MgH_{16} [23]. More recently, guided by theoretical predictions [24–27], the first stoichiometric high-T_c ternary $LaBeH_8$ hydride superconductor with a well-defined crystal structure was synthesized with a measured T_c of 110 K at 80 GPa [28]. Previous works also reported a series of nonstoichiometric high-T_c ternary alloy hydrides, where the random occupation of metal elements in the lattice sites can render the crystal structure effectively identical to the parent clathrate structure of the binary systems [29–39], as in theoretically predicted $CaYH_{12}$ (230-258 K at 180-200 GPa) [29, 30], and $(Y,Sr)H_{11}$ (240 K at 175 GPa) [35], as well as the experimentally synthesized $(La,Y)H_{10}$ (∼253 K at 183 GPa) [36], and $(La,Ce)H_9$ (148-178 K at 97-172 GPa) [37, 38]. These findings suggest the prospect of finding superconductors with the T_c exceeding room temperature in ternary hydrides. While ultimately such superconductors need to be stabilized and function at much lower pressures, it remains critical to establish whether any upper T_c limits exist for hydride superconductors. Therefore, searching for thermodynamically stable superconducting ternary hydrides with higher T_c than LaH_{10} (250-260 K at ∼180 GPa) is a pressing task.

LaH_{10} was the first metal superhydride (MH_n, $n >$ 6) to be predicted [14, 15], synthesized [40], and shown experimentally to be a near room-temperature superconductor, a result that has now been reproduced in experiments on the end-member and LaH_{10}-based alloys by at least three other groups [9, 10, 36, 39]. In addition, some

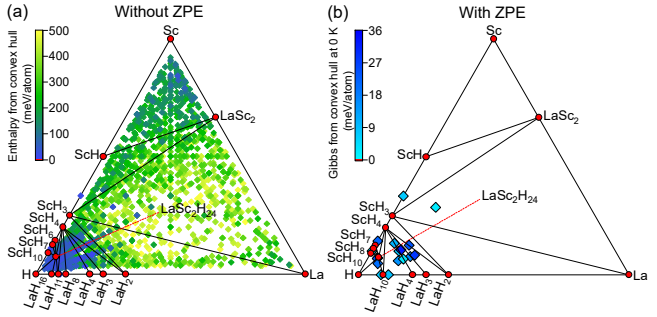

FIG. 1: (Color online) Calculated ternary phase diagram of the La-Sc-H system relative to elemental La [46–48], Sc [49], and H [50, 51], as well as binary La-H [14, 15, 52], Sc-H [53, 54], and La-Sc at 300 GPa (a) without or (b) with considering zero-point energy (ZPE). Red solid circles and colored squares indicate thermodynamically stable and unstable phases, respectively.

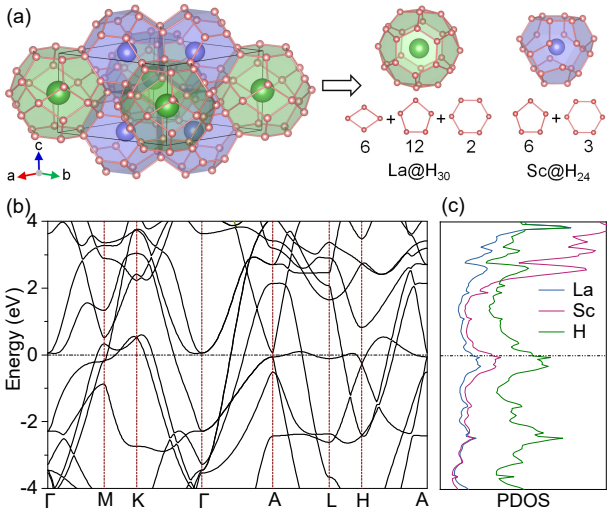

FIG. 2: (Color online) (a) The crystal structure of $LaSc_2H_{24}$ at 300 GPa, consisting of La-centered H_{30} cages and Sc-centered H_{24} cages. Each H_{30} cage contains six rhombuses, twelve pentagons, and two hexagons, whlie each H_{24} cage consists of six pentagons and three hexagons. (b) Electronic band structure and (c) projected electronic density of states (PDOS) of $LaSc_2H_{24}$ at 300 GPa.

studies indicated that the variable and higher T_c values found for LaH_{10} arise from different degrees of boron doping [41], and a decrease in T_c with the addition of magnetic impurities [39]. It is of great interest to examine the extent to which superconductivity could be further optimized by introducing other elements to form ternary hydrides with higher critical temperatures. An example is the substitutional alloy of (La,Ce)H_9 with a higher T_c than pure CeH_9 [37, 38], which is attributed to the element of La having a lower mass than Ce that could improve the Debye temperature of the system. Conversely, note that Sc has a similar valence electronic structure and a lower mass than La, allowing us to expect to tune the superconductivity in the La-H system by the introduction of Sc. To this end, using the swarm-intelligence-based CALYPSO structure prediction method [42–44] in combination with first-principles calculations, we systematically investigated the crystal structure and superconductivity of La-Sc-H system under high pressure. As a result, a clathrate structure of $LaSc_2H_{24}$ with two unusual H cages of H_{24} and H_{30} was found to be thermodynamically stable at 300 GPa. Remarkably, electron-phonon coupling (EPC) simulations with considering anharmonic effects revealed that this predicted structure is a promising hot superconductor with an estimated T_c of 330 K at 300 GPa.

Based on the results of variable-composition structure searches, we constructed the ternary phase diagram of La-Sc-H system at 300 GPa (see Supplemental Material [45] for computational details). A new hydrogen-rich compound with the formula $LaSc_2H_{24}$ and space group $P6/mmm$ is identified as thermodynamically stable at 300 GPa [Figs. 1(a) and S1(a) [45]]. As is well known, the contribution of zero-point energy (ZPE) can influence the stability of hydrogen-rich compounds due to the high vibrational frequency associated with the low mass of the H atoms. Including the effect of ZPE, we found that the new $LaSc_2H_{24}$ compound remains thermodynamically stable at 300 GPa [Figs. 1(b) and S1(b) [45]]. We also investigated the stability of $LaSc_2H_{24}$ at lower pressure by calculating the enthalpy differences in assuming several possible decomposition routes such as solid hydrogen [50, 51] and binary hydrides [14, 15, 52–54]. The results indicate that this new ternary hydride is stable at pressures as low above 158 GPa (ZPE included, see Fig. S2(b) [45]).

Moreover, the ternary $LaSc_2H_{24}$ compound crystallizes in a new superhydride clathrate structure that consists of La-centered H_{30} cages and Sc-centered H_{24} cages with the metal atoms arranged in the AlB_2 structure type [55] [Fig. 2(a)]. The H cages themselves have unusual structures, not found in known clathrate [13–15, 22, 56] or zeolite networks [57]: each H_{30} cage comprises six rhombuses, twelve pentagons, and two hexagons, while each H_{24} cage consists of six pentagons and three hexagons. It is also noteworthy that H-H bond lengths of 1.08, 1.12, 1.16, and 1.20 Å at 300 GPa are slightly longer than the H-H distance (0.98 Å) in monatomic solid hydrogen at 500 GPa [58, 59]. These H-H distances are similar to those found in CaH_6 (1.24 Å at 150 GPa) [13] and LaH_{10} (1.07 and 1.16 Å at 300 GPa) [14, 15], thereby forming an atomic hydrogen sublattice found in other clathrate hydride structures and associated with their very high-T_c superconductivity.

To examine the electronic properties, we calculated the electronic band structure and projected electronic

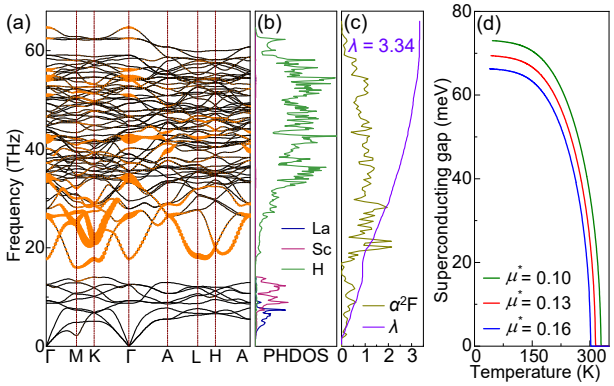

FIG. 3: (Color online) (a) Phonon spectra, where the orange solid circles show the phonon linewidth with a radius proportional to its strength, (b) projected phonon density of states (PHDOS), (c) Eliashberg spectral function $\alpha^2F(\omega)$, and the electron-phonon integral $\lambda(\omega)$ for $LaSc_2H_{24}$ at 300 GPa. (d) Superconducting gap of $LaSc_2H_{24}$ at 300 GPa.

density of states (PDOS) of $LaSc_2H_{24}$ at 300 GPa. As shown in Fig. 2(b), several electronic bands cross the Fermi level, indicating the metallic character of $LaSc_2H_{24}$. The PDOS of $LaSc_2H_{24}$ also shows a large H-derived DOS (0.11 states/spin/Ry/Å^3) at the Fermi level which is slightly higher than that in LaH_{10} (0.09 states/spin/Ry/Å^3) at 300 GPa [14, 15], indicating $LaSc_2H_{24}$ is expected to exhibit H-dominated high-temperature superconductivity. Full phonon and electron-phonon coupling (EPC) calculations were thus carried out. The phonon dispersion, with the absence of imaginary frequency modes, confirms the dynamical stability of the structure at 300 GPa [Fig. 3(a)]. The projected phonon density of states (PHDOS) shows that the phonon vibrations below 16 THz are associated with vibrations of heavy La and Sc atoms, and the frequency modes from 16 to 65 THz mainly originate from H atoms [Fig. 3(b)]. The calculated total EPC constant (λ) obtained by integrating over all available frequencies is 3.34, which is dominantly derived from the EPC of H modes (74%) [Fig. 3(c)], especially from the phonon softening between 16 and 29 THz, which shows a large phonon linewidth [Fig. 3(a)]. For strong EPC with λ larger than 1.5, the Eliashberg equations could give an accurate description through direct numerical solutions [60, 61]. The temperature-dependent superconducting energy gap [Fig. 3(d)] shows the predicted T_c of 303-329 K at 300 GPa, assuming the Coulomb pseudopotential μ^* of 0.16 to 0.1. In addition, our phonon simulations indicate that $LaSc_2H_{24}$ is dynamically stable down to 290 GPa (Fig. S3(a) [45]), before developing a harmonic instability at lower pressures (Fig. S4 [45]). We thus further explore the superconductivity in the pressure range 290 to 350 GPa within the harmonic approximation. As can be seen from Table I, the λ and T_c of $LaSc_2H_{24}$ decrease as pressure increases, whereas ω_{log} increases, which is dominantly attributed to less phonon softening with increasing pressure (Fig. S3 [45]).

TABLE I: The calculated EPC constant λ, phonon frequency logarithmic average ω_{log} (K), electronic density of states at the Fermi level $N(E_f)$ (states/spin/Ry/f.u.), and superconducting critical temperature T_c (K) with μ^* = 0.16-0.1 by numerically solving the Eliashberg equations for $LaSc_2H_{24}$ within the harmonic approximation and at the anharmonic level under given pressures (GPa).

Calculation	Pressure	λ	ω_{log}	$N(E_f)$	T_c
Harmonic	290	3.56	865	18.69	311-339
	300	3.34	968	17.27	303-329
	350	2.20	1294	15.64	266-294
Anharmonic	150	3.94	745	18.44	281-311
	200	3.21	965	19.31	291-325
	250	2.89	1075	19.63	296-331
	300	2.61	1165	19.61	293-330

It is known that anharmonic effects of quantum nuclear and/or thermal origin can affect both for the dynamic stability and superconductivity in many compressed hydrides [62–64]. We thus performed anharmonic phonon calculations of $LaSc_2H_{24}$ under high pressure by using the stochastic self-consistent harmonic approximation (SSCHA) approach [62, 65, 66]. The renormalized phonon spectra at the anharmonic level (Fig. S4 [45]) indicate that this hydride is dynamically stable down to 150 GPa, which is significantly lower than the harmonic approximation result of 290 GPa. The simulated results of superconductivity that consider anharmonic effects via renormalized phonon frequencies reveal that the calculated T_c could reach the highest value of 331 K at 250 GPa with μ^* of 0.1 (Table I). This calculated T_c value is larger than that (∼280 K at ∼210 GPa) originally calculated for LaH_{10} [14, 15], indicating the superconductivity in the La-H system could indeed be surpassed with the incorporation of Sc. Moreover, we found that there is essentially no change in calculated T_c (∼330 K at 300 GPa) after considering the self-consistent anharmonic and nuclear quantum effects.

In conclusion, the high-pressure phase diagram of the La-Sc-H system examined using the swarm-intelligence-based CALYPSO structure prediction method in combination with first-principles simulations reveals a novel hexagonal and thermodynamically stable $LaSc_2H_{24}$ phase at 300 GPa. The $LaSc_2H_{24}$ structure exhibits a unique three-dimensional H clathrate network consisting of unusual H_{24} and H_{30} cages. Most strikingly, self-consistent phonon calculations that take into account anharmonic and nuclear quantum effects predict a superconducting T_c up to 331 K at 250 GPa, which is well above room temperature. Our results further suggest that 'hot' superconducting hydrides with T_cs exceeding

300 K can exist in the experimentally accessible pressure range below 300 GPa and add to the diversity of clathrate hydride structures predicted by theory and confirmed in the laboratory.

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

[2] 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, Nat. Phys. **12**, 835 (2016).

[3] L. Ma, K. Wang, Y. Xie, X. Yang, Y. Wang, M. Zhou, H. Liu, X. Yu, Y. Zhao, H. Wang, G. Liu, and Y. Ma, High-temperature superconducting phase in clathrate calcium hydride CaH_6 up to 215 K at a pressure of 172 GPa, Phys. Rev. Lett. **128**, 167001 (2022).

[4] Z. Li, X. He, C. Zhang, X. Wang, S. Zhang, Y. Jia, S. Feng, K. Lu, J. Zhao, J. Zhang, B. Min, Y. Long, R. Yu, L. Wang, M. Ye, Z. Zhang, V. Prakapenka, S. Chariton, P. A. Ginsberg, J. Bass, S. Yuan, H. Liu, C. Jin, Superconductivity above 200 K discovered in superhydrides of calcium, Nat. Commun. **13**, 2863 (2022).

[5] I. A. Troyan, D. V. Semenok, A. G. Kvashnin, A. V. Sadakov, O. A. Sobolevskiy, V. M. Pudalov, A. G. Ivanova, V. B. Prakapenka, E. Greenberg, A. G. Gavriliuk, I. S. Lyubutin, V. V. Struzhkin, A. Bergara, I. Errea, R. Bianco, M. Calandra, F. Mauri, L. Monacelli, R. Akashi, and A. R. Oganov, Anomalous high-temperature superconductivity in YH_6, Adv. Mater. **33**, 2006832 (2021).

[6] P. Kong, V. S. Minkov, M. A. Kuzovnikov, A. P. Drozdov, S. P. Besedin, S. Mozaffari, 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, Nat. Commun. **12**, 5075 (2021).

[7] E. Snider, N. Dasenbrock-Gammon, R. McBride, X. Wang, N. Meyers, K. V. Lawler, E. Zurek, A. Salamat, and R. P. Dias, Synthesis of yttrium superhydride superconductor with a transition temperature up to 262 K by catalytic hydrogenation at high pressures, Phys. Rev. Lett. **126**, 117003 (2021).

[8] 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).

[9] A. P. Drozdov, P. P. Kong, V. S. Minkov, S. P. Besedin, M. A. Kuzovnikov, S. Mozaffari, L. Balicas, F. F. Balakirev, D. E. Graf, V. B. Prakapenka, E. Greenberg, D. A. Knyazev, M. Tkacz, and M. I. Eremets, Superconductivity at 250 K in lanthanum hydride under high pressures, Nature (London) **569**, 528 (2019).

[10] F. Hong, L. Yang, P. Shan, P. Yang, Z. Liu, J. Sun, Y. Yin, X.Yu, J. Cheng, and Z. Zhao, Superconductivity of lanthanum superhydride investigated using the standard four-probe configuration under high pressures, Chin. Phys. Lett. **37**, 107401 (2020).

[11] Y. Li, J. Hao, H. Liu, Y. Li, and Y. Ma, The metallization and superconductivity of dense hydrogen sulfide, J. Chem. Phys. **140**, 174712 (2014).

[12] D. Duan, Y. Liu, F. Tian, D. Li, X. Huang, Z. Zhao, H. Yu, B. Liu, W. Tian, and T. Cui, Pressure-induced metallization of dense $(H_2S)_2H_2$ with high-T_c superconductivity, Sci. Rep. **4**, 6968 (2014).

[13] H. Wang, J. S. Tse, K. Tanaka, T. Iitaka, and Y. Ma, Superconductive sodalite-like clathrate calcium hydride at high pressures, Proc. Natl. Acad. Sci. USA **109**, 6463 (2012).

[14] 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).

[15] H. Liu, I. I. Naumov, R. Hoffmann, N. W. Ashcroft, and R. J. Hemley, Potential high-T_c superconducting lanthanum and yttrium hydrides at high pressure, Proc. Natl. Acad. Sci. USA **114**, 6990 (2017).

[16] 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, Phys. Rep. **856**, 1 (2020).

[17] X. Zhang, Y. Zhao, and G. Yang, Superconducting ternary hydrides under high pressure, WIREs Comput. Mol. Sci. **12**, e1582 (2022).

[18] K. P. Hilleke and E. Zurek, Tuning chemical precompression: theoretical design and crystal chemistry of novel hydrides in the quest for warm and light superconductivity at ambient pressures, J. Appl. Phys. **131**, 070901 (2022).

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

[20] T. Ishikawa, T. Miyake, and K. Shimizu, Materials informatics based on evolutionary algorithms: application to search for superconducting hydrogen compounds, Phys. Rev. B **100**, 174506 (2019).

[21] X. Zhong, J. S. Tse, R. Hemley, and H. Liu, Theory-

directed discovery of high-temperature superconductivity in clathrate hydrides at high pressure, The Innovation **3**, 100226 (2022).

[22] 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).

[23] D. C. Lonie, J. Hooper, B. Altintas, and E. Zurek, Metallization of magnesium polyhydrides under pressure, Phys. Rev. B **87**, 054107 (2013).

[24] S. Di Cataldo, W. von der Linden, and L. Boeri, First-principles search of hot superconductivity in La-X-H ternary hydrides, npj Comput. Mater. **8**, 2 (2022).

[25] S. Di Cataldo, C. Heil, W. von der Linden, and L. Boeri, $LaBH_8$: towards high-T_c low-pressure superconductivity in ternary superhydrides, Phys. Rev. B **104**, L020511 (2021).

[26] X. Liang, A. Bergara, X. Wei, X. Song, L. Wang, R. Sun, H. Liu, R. J. Hemley, L. Wang, G. Gao, and Y. Tian, Prediction of high-T_c superconductivity in ternary lanthanum borohydrides, Phys. Rev. B **104**, 134501 (2021).

[27] Z. Zhang, T. Cui, M. J. Hutcheon, 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 a hydrogen-based alloy backbone at moderate pressure, Phys. Rev. Lett. **128**, 047001 (2022).

[28] Y. Song, J. Bi, Y. Nakamoto, K. Shimizu, H. Liu, B. Zou, G. Liu, H. Wang, and Y. Ma, Stoichiometric ternary superhydride $LaBeH_8$ as a new template for high-temperature superconductivity at 110 K under 80 GPa, Phys. Rev. Lett. **130**, 266001 (2023).

[29] X. Liang, A. Bergara, L. Wang, B. Wen, Z. Zhao, X.-F. Zhou, J. He, G. Gao, and Y. Tian, Potential high-T_c superconductivity in $CaYH_{12}$ under pressure, Phys. Rev. B **99**, 100505(R) (2019).

[30] H. Xie, D. Duan, Z. Shao, H. Song, Y. Wang, X. Xiao, D. Li, F. Tian, B. Liu, and T. Cui, High-temperature superconductivity in ternary clathrate $YCaH_{12}$ under high pressures, J. Phys.: Condens. Matter **31**, 245404 (2019).

[31] W. Sukmas, P. Tsuppayakorn-aek, U. Pinsook, and T. Bovornratanaraks, Near-room-temperature superconductivity of Mg/Ca substituted metal hexahydride under pressure, J. Alloys Compd. **849**, 156434 (2020).

[32] P. Song, Z. Hou, K. Nakano, K. Hongo and R. Maezono, Potential high-T_c superconductivity in $YCeH_x$ and $LaCeH_x$ under pressure, Mater. Today Phys. **28**, 100873 (2022).

[33] M. Du, H. Song, Z. Zhang, D. Duan, and T. Cui, Room-temperature superconductivity in Yb/Lu substituted clathrate hexahydrides under moderate pressure, Research **2022**, 9784309 (2022).

[34] L. Liu, F. Peng, P. Song, X. Liu, L. Zhang, X. Huang, C. Niu, C. Liu, W. Zhang, Y. Jia, and Z. Zhang, Generic rules for achieving room-temperature superconductivity in ternary hydrides with clathrate structures, Phys. Rev. B **107**, L020504 (2023).

[35] X.-L. He, P. Zhang, Y. Ma, H. Li, X. Zhong, Y. Wang, H. Liu, and Y. Ma, Potential high-temperature superconductivity in the substitutional alloy of (Y, Sr)H_{11} under high pressure, Phys. Rev. B **107**, 134509 (2023).

[36] D. V. Semenok, I. A. Troyan, A. G. Ivanova, A. G. Kvashnin, I. A. Kruglov, M. Hanfland, A. V. Sadakov, O. A. Sobolevskiy, K. S. Pervakov, I. S. Lyubutin, K. V. Glazyrin, N. Giordano, D. N. Karimov, A. L. Vasiliev, R. Akashi, V. M. Pudalov, and A. R. Oganov, Superconductivity at 253 K in lanthanum-yttrium ternary hydrides, Mater. Today **48**, 18 (2021).

[37] J. Bi, Y. Nakamoto, P. Zhang, K. Shimizu, B. Zou, H. Liu, M. Zhou, G. Liu, H. Wang, and Y. Ma, Giant enhancement of superconducting critical temperature in substitutional alloy (La,Ce)H_9, Nat. Commun. **13**, 5952 (2022).

[38] W. Chen, X. Huang, D. V. Semenok, S. Chen, D. Zhou, K. Zhang, A. R. Oganov, and T. Cui, Enhancement of superconducting properties in the La-Ce-H system at moderate pressures, Nat. Commun. **14**, 2660 (2023).

[39] D. V. Semenok, I. A. Troyan, A. V. Sadakov, D. Zhou, M. Galasso, A. G. Kvashnin, A. G. Ivanova, I. A. Kruglov, A. A. Bykov, K. Y. Terent'ev, A. V. Cherepahin, O. A. Sobolevskiy, K. S. Pervakov, A. Yu. Seregin, T. Helm, T. Förster, A. D. Grockowiak, S. W. Tozer, Y. Nakamoto, K. Shimizu, V. M. Pudalov, I. S. Lyubutin, and A. R. Oganov, Effect of magnetic impurities on superconductivity in LaH_{10}, Adv. Mater. **34**, 2204038 (2022).

[40] Z. M. Geballe, H. Liu, A. K. Mishra, M. Ahart, M. Somayazulu, Y. Meng, M. Baldini, and R. J. Hemley, Synthesis and stability of lanthanum superhydrides, Angew. Chem. Int. Ed. **57**, 688 (2018).

[41] Y. Ge, F. Zhang, and R. J. Hemley, Room-temperature superconductivity in boron- and nitrogen-doped lanthanum superhydride, Phys. Rev. B **104**, 214505 (2021).

[42] Y. Wang, J. Lv, L. Zhu, and Y. Ma, Crystal structure prediction via particle-swarm optimization, Phys. Rev. B **82**, 094116 (2010).

[43] Y. Wang, J. Lv, L. Zhu, and Y. Ma, CALYPSO: a method for crystal structure prediction, Comput. Phys. Commun. **183**, 2063 (2012).

[44] X. Shao, J. Lv, P. Liu, S. Shao, P. Gao, H. Liu, Y. Wang, and Y. Ma, A symmetry-orientated divide-and-conquer method for crystal structure prediction, J. Chem. Phys. **156**, 014105 (2022).

[45] See Supplemental Material at https://XXX for computational details, the ternary phase diagram of La-Sc-H system with LaH_2, ScH_3, and H as corners without or with considering zero-point energy (ZPE), relative enthalpies of $LaSc_2H_{24}$ as a function of pressure without or with considering ZPE, phonon dispersion curves at the harmonic and anharmonic levels, as well as structural parameters of $LaSc_2H_{24}$ under high pressure.

[46] F. Porsch and W. B. Holzapfel, Novel reentrant high pressure phase transtion in lanthanum, Phys. Rev. Lett. **70**, 4087 (1993).

[47] W. Chen, D. V. Semenok, I. A. Troyan, A. G. Ivanova, X. Huang, A. R. Oganov, and T. Cui, Superconductivity and equation of state of lanthanum at megabar pressures, Phys. Rev. B **102**, 134510 (2020).

[48] L. Chen, T. Liang, Z. Zhang, H. Song, Z. Liu, Q. Jiang, Y. Chen, and Defang Duan, Phase transitions and properties of lanthanum under high pressures, J. Phys.: Condens. Matter **34**, 204005 (2022).

[49] Y. Akahama, H. Fujihisa, and H. Kawamura, New helical chain structure for scandium at 240 GPa, Phys. Rev. Lett. **94**, 195503 (2005).

[50] C. J. Pickard and R. J. Needs, Structure of phase III of solid hydrogen, Nat. Phys. **3**, 473 (2007).

[51] H. Liu, H. Wang, and Y. Ma, Quasi-molecular and atomic phases of dense solid hydrogen, J. Phys. Chem. C **116**, 9221 (2012).

[52] I. A. Kruglov, D. V. Semenok, H. Song, R. Szcześniak, I. A. Wrona, R. Akashi, M. M. Davari Esfahani, D. Duan, T. Cui, A. G. Kvashnin, and A. R. Oganov, Superconductivity of LaH_{10} and LaH_{16} polyhydrides, Phys. Rev. B **101**, 024508 (2020).

[53] K. Abe, Hydrogen-rich scandium compounds at high pressures, Phys. Rev. B **96**, 144108 (2017).

[54] X. Ye, N. Zarifi, E. Zurek, R. Hoffmann, and N. W. Ashcroft, High hydrides of scandium under pressure: potential superconductors, J. Phys. Chem. C **122**, 6298 (2018).

[55] W. Hofmann and W. Jäniche, Die struktur von aluminiumborid AlB_2, Z. Phys. Chem. **31B**, 214 (1936).

[56] X. Zhong, Y. Sun, T. Iitaka, M. Xu, H. Liu, R. J. Hemley, C. Chen, and Y. Ma, Prediction of above-room-temperature superconductivity in lanthanide/actinide extreme superhydrides, J. Am. Chem. Soc. **144**, 13394 (2022).

[57] http://asia.iza-structure.org/IZA-SC/ftc_table.php.

[58] V. Labet, P. Gonzalez-Morelos, R. Hoffmann, and N. W. Ashcroft, A fresh look at dense hydrogen under pressure. I. An introduction to the problem, and an index probing equalization of H-H distances, J. Chem. Phys. **136**, 074501 (2012).

[59] J. M. McMahon and D. M. Ceperley, Ground-state structures of atomic metallic hydrogen, Phys. Rev. Lett. **106**, 165302 (2011).

[60] A. Sanna, J. A. Flores-Livas, A. Davydov, G. Profeta, K. Dewhurst, S. Sharma, and E. K. U. Gross, Ab initio eliashberg theory: making genuine predictions of superconducting features, J. Phys. Soc. Jpn. **87**, 041012 (2018).

[61] The elk code, url: http://elk.sourceforge.net/.

[62] I. Errea, M. Calandra, and F. Mauri, First-principles theory of anharmonicity and the inverse isotope effect in superconducting palladium-hydride compounds, Phys. Rev. Lett. **111**, 177002 (2013).

[63] I. Errea, M. Calandra, C. J. Pickard, J. Nelson, R. J. Needs, Y. Li, H. Liu, Y. Zhang, Y. Ma, and F. Mauri, High-pressure hydrogen sulfide from first principles: a strongly anharmonic phonon-mediated superconductor, Phys. Rev. Lett. **114**, 157004 (2015).

[64] I. Errea, F. Belli, L. Monacelli, A. Sanna, T. Koretsune, T. Tadano, R. Bianco, M. Calandra, R. Arita, F. Mauri, and J. A. Flores-Livas, Quantum crystal structure in the 250-kelvin superconducting lanthanum hydride, Nature (London) **578**, 66 (2020).

[65] I. Errea, M. Calandra, and F. Mauri, Anharmonic free energies and phonon dispersions from the stochastic self-consistent harmonic approximation: application to platinum and palladium hydrides, Phys. Rev. B **89**, 064302 (2014).

[66] 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, J. Phys: Condens. Matter **33**, 363001 (2021).