High-temperature superconducting ternary Li-*R*-H superhydrides at high pressures (*R* = Sc, Y, La)

Ying Sun,[1] Yanchao Wang,[1,*] Xin Zhong,[2,†] Yu Xie,[1,3] and Hanyu Liu[1,4]

[1]*International Center for Computational Method & Software and State Key Laboratory of Superhard Materials, College of Physics, Jilin University, Changchun 130012, China*

[2]*Key Laboratory of Functional Materials Physics and Chemistry of the Ministry of Education, National Demonstration Center for Experimental Physics Education, Jilin Normal University, Changchun 130103, China*

[3]*Key Laboratory of Physics and Technology for Advanced Batteries (Ministry of Education), College of Physics, Jilin University, Changchun 130012, China*

[4]*International Center of Future Science, Jilin University, Changchun 130012, China*

(Received 15 February 2022; accepted 30 June 2022; published 26 July 2022)

Compressed clathrate superhydrides have been the most promising candidates for room-temperature superconductors since the theory-oriented findings of CaH_6, YH_9, LaH_{10} *et. al.*, where the hydrogen clathrate framework was believed to play a critical role in improving superconductivity. Recently, a ternary superhydride of Li_2MgH_{16} was predicted to be a "hot" superconductor with a theoretical T_c value up to 473 K at 250 GPa, although it exhibits the metastable feature under high pressure. With the aim of seeking thermodynamically stable ternary clathrate superhydrides, by exploring the high-pressure phase diagram of the Li–*R*–H (*R* = Sc, Y, and La) systems at 300 GPa, we identified several thermodynamically ternary superhydrides with high-temperature superconductivity. Among these predicted stable structures, as a result of extensive simulations, clathrate structured Li_2YH_{17} and Li_2LaH_{17} are predicted to be high-temperature superconductors with a superconducting critical temperature (T_c) up to 108 and 156 K, at 200 and 160 GPa, respectively. Interestingly, a superhydride, *Immm*–Li_2ScH_{20}, with mixed molecular and atomic hydrogen, is predicted to possess a high T_c of 242 K at 300 GPa. The present results may stimulate the future experiment for the investigation of structural, electronic, and superconducting properties of metal-doped rare-earth superhydrides, which thus help the further design and discovery of superconducting clathrate superhydrides.

DOI: 10.1103/PhysRevB.106.024519

I. INTRODUCTION

Clathrate superhydride is a new class of hydrides in which metal atoms act as guests in hydrogen (H) cages, while H atoms are weakly covalently bonded with each other [1–11] and the H–H distance of approximately 1.0 Å [12–14] is close to that (0.98 Å) in atomic hydrogen at 500 GPa [15]. The first-ever sodalite-like clathrate superhydride CaH_6 is predicted to have high T_c values of 220–235 K at 150 GPa [12], which has been recently successfully synthesized by two independent experiments [16,17]. After the seminal prediction of CaH_6 [12], compressed clathrate superhydrides, such as LaH_{10} and YH_{10}, are predicted to have high T_c at high pressure, approaching room-temperature superconductivity [13,14]. Encouraged by these remarkable predictions, subsequent experiments synthesized a variety of rare-earth (*R*) clathrate superhydrides [1–3], of which the measured T_cs of YH_6, YH_9, and LaH_{10} reach high values of 220 K [18,19] at 180 GPa, 243 K [18] at 200 GPa, and 250–260 K at 180 GPa [20,21], respectively. The analysis of these high-temperature superconducting superhydrides suggests that high superconductivity arises from atomic-H cages due to the fact that the H electrons contribute substantially to the electron density of states at the Fermi level ($N(E_f)$) [13,14]. These concrete theoretical and experimental efforts indicate that the hydrogen clathrate framework plays an important role in improving superconductivity, thereby implying clathrate superhydrides are the most promising candidates for room-temperature superconductivity.

Recently, ternary hydrides are found to provide a unique opportunity to investigate high superconductivity, since they have more abundant and complex structures resulting from diverse chemical compositions and synergistic charge transfer as well as strong electron-phonon coupling constant, which make them an appealing contender for superconductors [6,11,22]. There have been many theoretical and experimental studies on ternary superconducting superhydride at high pressure [23–40], including several predicted and synthesized clathrate structured materials, such as $LaBH_8$ with a T_c of 126 K at 50 GPa [28,29], $CaYH_{12}$ with T_cs of 230–258 K at 180–200 GPa [23,41], and cubic hexahydride $(La,Y)H_6$ with a T_c of ∼237 K and decahydrides $(La,Y)H_{10}$ with a T_c of ∼253 K at 183 GPa [26]. In our previous work, through the strategy of introducing extra electrons via metal doping into known H_2-rich binary systems, we have already identified a metastable hot superconductor of Li_2MgH_{16} with a theoretical T_c value well above room temperature (T_c = 473 K at 250 GPa) [27]. However, this interesting Li_2MgH_{16} phase exhibits a metastable feature, thus the search for

*wyc@calypso.cn
†zhongxin@calypso.cn

thermodynamically stable ternary clathrate superhydrides with high T_c values remains a key issue in the research field of high-temperature superconductivity.

In an attempt to seek ternary stable clathrate superhydrides that have even higher H content than Li_2MgH_{16}, the H lattices of ternary hydrides should involve more electrons from the metal elements than that in Li_2MgH_{16}. It is noteworthy that R elements have more valence electrons than Mg, where these R elements such as Y could donate their valence electrons into hydrogen lattice due to the low electronegativities. This indicates that the Li–R–H compound may be an ideal system to offer a platform for the search for high-temperature superconductivity. In this work, therefore, we performed extensive simulations on the exploration of the stability for various Li–R–H (R = Sc, Y, La) compounds under high pressure. By focusing on H-rich species by performing structure searches, we identified several thermodynamically stable and high-temperature superconducting ternary compounds. Among these predicted stable structures, $Fd\bar{3}m$ structured Li_2YH_{17} and Li_2LaH_{17} are clathrate superhydrides with predicted T_c values up to 112 and 156 K, respectively. Moreover, we have also predicted several metastable superhydrides with high T_c approaching room temperature.

II. COMPUTATIONAL DETAILS

We employed our developed swarm-intelligence-based CALYPSO structure prediction method [42–45] for the investigation of phase stability of Li–R–H compounds at 300 GPa. Firstly, universal variable-composition structure searches for $Li_xR_yH_z$ (R = Sc, Y, and La. x = 1–3, y = 1–2, z = 10–48) systems were performed at 300 GPa, where more than 300 structures for each stoichiometry have been studied. The energetic stability of different ternary stoichiometries is evaluated by their formation enthalpy of dissociated into the most competing element and binary compounds [13,14,46–49]. For compositions with formation enthalpies lower than 100 meV/atom, separate structural predictions with fixed stoichiometries were conducted for confirmation of their results. 1000 ∼ 1200 structures were investigated for each fixed stoichiometry to ensure the convergence of our structure searches. The underlying energetic calculations are performed with the plane-wave pseudopotential method as implemented in the VASP code [50]. The Perdew-Burke-Ernzerhof generalized gradient approximation [51,52] is chosen for the exchange-correlation functional. The electron-ion interaction is described by projector-augmented-wave [53] potentials with 1, 3, 11, 11, and 11 valence electrons for H, Li, Sc, Y, and La atoms, respectively. A kinetic cutoff energy of 700 eV and Monkhorst-Pack $\boldsymbol{k}$ meshes with grid spacing of 0.25 $Å^{-1}$ are adopted to ensure the enthalpy converges to better than 1 meV/atom. The phonon spectrum and electron-phonon coupling (EPC) of the stable compounds are calculated within the framework of linear response theory through the Quantum-ESPRESSO code [54], where ultrasoft pseudopotentials [55] were used with a kinetic energy cutoff for wave functions of 60 Ry and a kinetic energy cutoff for charge density and potential of 750 Ry. $3 \times 3 \times 3$ $\boldsymbol{q}$ meshes and $12 \times 12 \times 12$ $\boldsymbol{k}$ meshes were used for stable Li–R–H ternaries to compute the EPC matrix elements. EPC constant λ, $\omega_{\log}$, and T_c are solved using the elk code [56], as derived by the direct solution of the isotropic Migdal-Eliashberg equation [57–59].

III. RESULTS AND DISCUSSION

The previous prediction suggests that YH_{24} is the most H-rich binary rare-earth hydride, containing 12 H_2 molecules per formula unit [13]. To mimic the behavior of introducing extra electrons via Li doping into 1–2 $f.u.$ binary rare-earth superhydrides, we examine the phase diagram of $Li_xR_yH_z$ (R = Sc, Y, and La. x = 1–3, y = 1–2, z = 10–48) at 300 GPa, as shown in Fig. 1. It is noteworthy that our structure searches are more focused on H-rich species in this work. In previous high-throughput material discovery studies, the convention structure searches for the threshold between metastable (likely to be synthesized) or unstable (unlikely to be synthesized) compounds are 50 meV/atom [60]. The same criterion was adopted in this work: stable, metastable, and unstable compounds with formation enthalpy values lower than 100 meV/atom are represented by green circles, blue squares, and red squares, respectively (Fig. 1). As a result, $LiScH_{10}$, Li_2ScH_{20}, Li_2YH_{17}, $Li_3Y_2H_{36}$, and Li_2LaH_{17} are predicted to become stable at 300 GPa Figs. 1(a)–1(c)]. The corresponding parameters of these predicted structures at 300 GPa are listed in the Appendix Table II.

It is well known that the zero-point energy (ZPE) is critical in determining the phase stabilities of H-rich compounds because of the high vibrational frequency arising from the light element of hydrogen. Compared with the atomic–H, H_2 molecular units often give higher vibrational frequency in H-rich compounds. The energy difference between molecular and atomic hydrogen at 500 GPa was calculated to be 30 meV/atom [27]. The stable and metastable Li–R–H phases with formation enthalpy values lower than 30 meV/atom were further explored by considering ZPE, as shown in Figs. 1(d)–1(f). As a result, Li_2ScH_{20}, Li_2YH_{17}, $Li_3Y_2H_{36}$, and Li_2LaH_{17} compounds remain thermally stable at 300 GPa with considering ZPE.

Four newly predicted stable ternary Li–R–H compounds at 300 GPa are shown in Fig. 2. Space group of Li_2ScH_{20}, Li_2YH_{17}, $Li_3Y_2H_{36}$, and Li_2LaH_{17} is $Immm$, $Fd\bar{3}m$, $C2/m$, and $Fd\bar{3}m$, respectively. Li_2ScH_{20} can be referred as $Li_2Sc(H_2)_4H_{12}$. In this predicted structure, 40% of H atoms form covalently bonded H_2 molecule units with H–H distances of 0.78 and 0.90 Å, while the other 60% of H atoms form weakly covalently bonded atomic-H layer with H–H distances of 1.03 and 1.15 Å. Clathrate-structured Li_2YH_{17}, $Li_3Y_2H_{36}$ and Li_2LaH_{17} are composed of H_{20} and H_{28} cages, with Li or H_2 molecules and Y or La in the center of atomic-H cages. Interestingly, Li_2YH_{17} and Li_2LaH_{17} share the same structure, while $Li_3Y_2H_{36}$ can be regarded as Li_2YH_{17} after replacing a quarter of Li atoms with H_2 molecules, which thus can be referred as $[(H_2)Li_3]Y_2H_{34}$. The symmetry of $Li_3Y_2H_{36}$ is much lower than that of Li_2YH_{17}, which is attributed to the symmetry breaking introduced by the H_2 molecule units. It should be noted that all of these four Li–R–H compounds contain large proportions of atomic-H with less or almost no proportions of H_2 molecule units. This suggests that these predicted superhydrides with

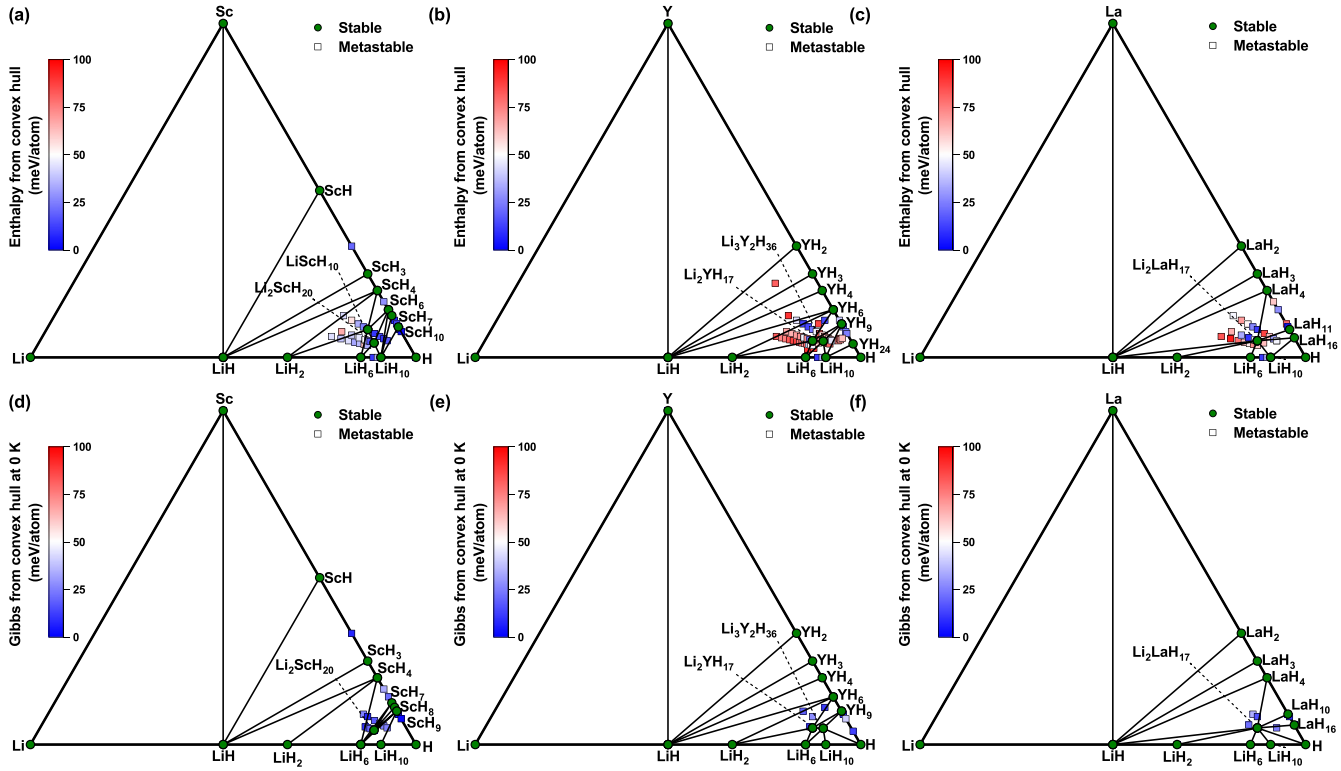

FIG. 1. Calculated enthalpy phase diagrams of Li–*R*–H (R = Sc, Y, La) system at 300 GPa (a)–(c) without or (d)–(f) with considering zero-point energy (ZPE). Blue and red squares denote metastable or unstable phases with different formation enthalpies. Green circles indicate thermally stable phases. Black lines between green circles connect stable phases.

a large number of atomic-H may exhibit high-temperature superconductivity.

All predicted stable Li–*R*–H ternaries are metallic at 300 GPa, as confirmed by the electronic density of states calculations (see Fig. 3). Similar to other clathrate superhydrides (such as CaH_6 [12], YH_9 [13], LaH_{10} [13,14], and Li_2MgH_{16}) [27], clathrate structured Li–*R*–H compounds possess high H-dominated $N(E_f)$. Li_2ScH_{20} with H_2 molecular unit and atomic hydrogen layers, also proposes large $N(E_f)$ values, where $N(E_f)$ is also H-dominated and there is a large proportion of H atoms by forming atomic-H

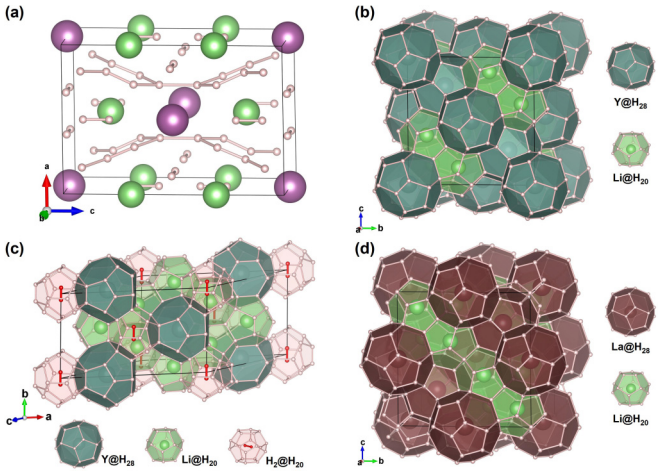

FIG. 2. Conventional cells of (a) $Immm$–Li_2ScH_{20}, (b) $Fd\bar{3}m$–Li_2YH_{17}, (c) $C2/m$–$Li_3Y_2H_{36}$, (d) $Fd\bar{3}m$–Li_2LaH_{17} at 300 GPa. The light green, purple, dark green, and brown spheres represent Li, Sc, Y, and La metal atoms, respectively. Both red and pink spheres are H atoms, and different colors are used to characterize bonding characteristics between H atoms. $Fd\bar{3}m$–Li_2YH_{17}, $C2/m$–$Li_3Y_2H_{36}$, and $Fd\bar{3}m$–Li_2LaH_{17} are all clathrate structures, consisting of Li-centered H_{20}, H_2-centered H_{20}, Y-centered H_{28}, and La-centered H_{28} cage units. All the hydrogen cage units are presented using translucent polyhedrons.

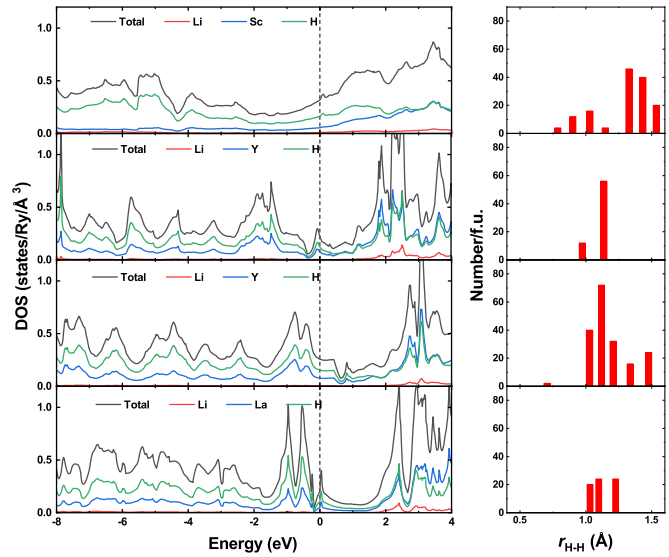

FIG. 3. Projected density of states (left panel) and histogram of H–H distances (r_{H-H}, right panel) for thermally stable Li–*R*–H compounds at 300 GPa.

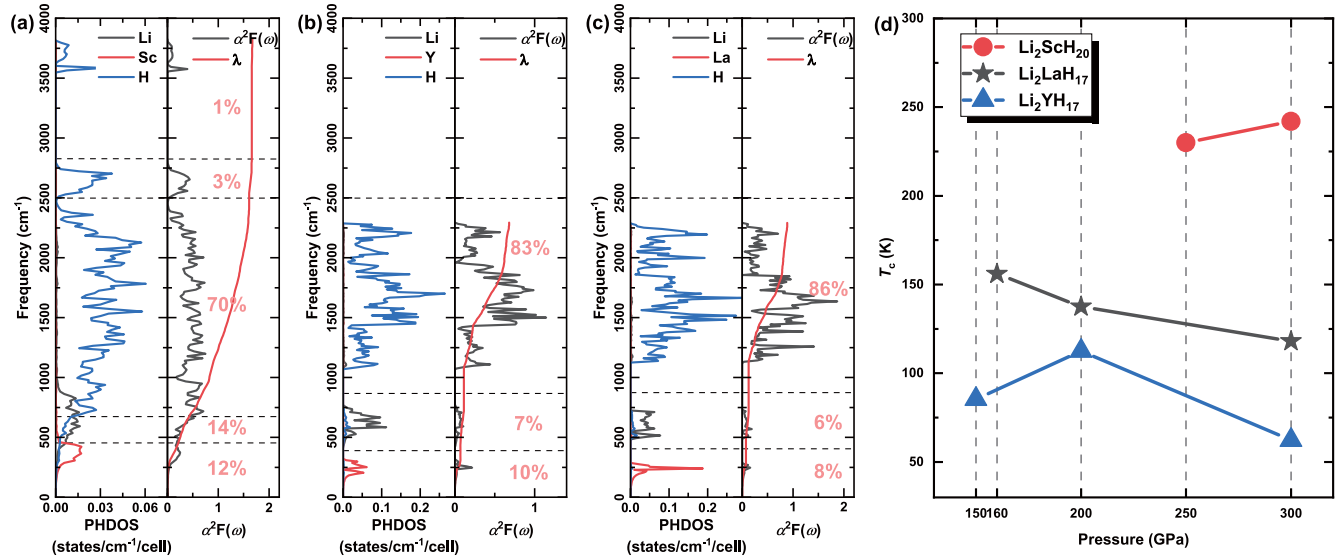

FIG. 4. Projected phonon densities of states (left panel) and isotropic Eliashberg spectral function $\alpha^2F(\omega)$ and EPC parameter $\lambda(\omega)$ (right panel) of (a) $Immm$–Li_2ScH_{20}, (b) $Fd\bar{3}m$–Li_2YH_{17}, and (c) $Fd\bar{3}m$–Li_2LaH_{17} at 300 GPa. Vibration frequencies dominated by different elements are separated by black dashed lines. The contribution proportion of different vibration frequency areas to λ is represented in pink characters. (d) Predicted T_c of Li_2ScH_{20}, Li_2YH_{17}, and Li_2LaH_{17} at different pressures.

layers. A high H-dominated $N(E_f)$ is often associated with high-temperature superconductivity [2,13,61]. Our further lattice dynamics analysis of Li–*R*–H compounds indicated that only $Immm$–Li_2ScH_{20}, $Fd\bar{3}m$–Li_2YH_{17}, and $Fd\bar{3}m$–Li_2LaH_{17} can be dynamically stable at 300 GPa. These results show that Li_2ScH_{20}, Li_2YH_{17}, and Li_2LaH_{17} are dynamically stable at pressures down to 250, 150, and 160 GPa, respectively.

EPC constants (λ) of $Immm$–Li_2ScH_{20}, $Fd\bar{3}m$–Li_2YH_{17}, and $Fd\bar{3}m$–Li_2LaH_{17} at 300 GPa are estimated to be 1.81, 0.69, and 0.86, respectively, mainly from the contribution of atomic–H's bending modes (800–2500 cm^{-1}), as shown in Figs. 4(a)–4(c). For $Immm$–Li_2ScH_{20}, as shown in Fig. 4(a), middle-frequency vibrations of 700–2800 cm^{-1} derived from bending modes of atomic-H layers and contribute 70% to the total λ, while high-frequency vibrations of 2500–2800 cm^{-1} and 3500–3800 cm^{-1} correspond with H_2 molecule units with H–H distance of 0.90 and 0.78 Å, contribute 3% and 1% to the total λ, respectively. For clathrate-structured Li_2YH_{17} [Fig. 4(b)] and Li_2LaH_{17} [Fig. 4(c)], no high-frequency vibration is consistent with the absence of H_2 molecule unit in the structure, and the highest phonon frequency of 2300 cm^{-1} can also be comparable with other systems containing atomic-H, such as 2600 cm^{-1} in the $I4_1/amd$ phase of metallic hydrogen at 500 GPa [62], and 2300 cm^{-1} in the $Fm\bar{3}m$–LaH_{10} clathrate superhydride at 300 GPa [13,14].

In terms of large EPC λ in these predicted superhydrides, the superconducting energy gap values versus temperature were calculated by numerically solving the isotropic Migdal-Eliashberg equation [57–59], then the maximum value of temperature corresponding to the superconducting energy gap is not zero defines the T_c value. When the Coulomb shielding pseudopotential μ^* is set to be 0.10, T_c of Li_2ScH_{20}, Li_2YH_{17}, and Li_2LaH_{17} is estimated to be up to 242 K at 300 GPa, 108 K at 200 GPa, and 156 K at 160 GPa, respectively, and the corresponding EPC λ is 1.81, 0.99 and 1.80, respectively. Although neither Li_2YH_{17} nor Li_2LaH_{17} are potential room-temperature superconductors, they are thermodynamically stable clathrate structural prototypes. We have also identified several metastable clathrate superhydrides in Li–*R*–H system at 300 GPa, including $Fm\bar{3}m$–Li_2REH_{16} (R = Sc, Y) and $Fm\bar{3}m$–Li_2ScH_{17}, which could be dynamically stable and share the same structure with $Fm\bar{3}m$–Li_2MgH_{16} [27] and $Fm\bar{3}m$–Li_2YH_{17}, respectively. Of these metastable superhydrides, Li_2ScH_{16} and Li_2YH_{16} are predicted to have estimated high T_c of 281 K at 230 GPa and 285 K at 170 GPa, respectively, as listed in Table I. These results suggest that partially replacing Li/Y/La atoms with other metal atoms or small molecules may help to further design high-temperature superconductivity in clathrate superhydrides.

IV. CONCLUSION

In summary, by searching for the high-pressure phase diagram of the Li–*R*–H system using the CALYPSO crystal structure prediction method and software, several thermodynamically stable superhydrides were proposed to be high-temperature superconductors. T_c values of clathrate structured $Fd\bar{3}m$–Li_2YH_{17} and $Fd\bar{3}m$–Li_2LaH_{17} are estimated to be up to 108 and 156 K at 200 and 160 GPa, respectively. Besides, a superhydride, $Immm$–Li_2ScH_{20}, with mixed H_2 molecular units and the atomic hydrogen layers, is predicted to be a high-temperature superconductor with the highest T_c value of 242 K at 300 GPa. Clathrate structured Li_2YH_{17} and Li_2LaH_{17} could be used as a prototype structure, and superconductivity might be improved by replacing Li and Y/La via other metal elements. Ternary compounds may be promising candidates for exploring high-temperature superconductivity and intriguing physical phenomena [63,64] under high pressure.

TABLE I. $N(E_f)$ (states/Ry/f.u.), λ, $\omega_{\log}$ (K), and T_c (K) values estimated using $\mu^\star = 0.10(0.13)$ of Li–*R*–H compounds at different pressures (GPa). Superhydrides charactered with (M) indicate their metastable properties at 300 GPa.

Compound	Pressure	$N(E_f)$	λ	$\omega_{\log}$	T_c
Immm–Li_2ScH_{20}	300	7.55	1.81	1404	242(223)
	250	7.46	1.86	1294	230(211)
Fd-3*m*–Li_2YH_{17}	300	2.99	0.69	1758	64(50)
	200	3.03	0.99	1332	108(93)
	150	3.19	0.85	1390	83(69)
Fd-3*m*–Li_2LaH_{17}	300	4.87	0.86	1854	118(99)
	200	4.58	1.14	1324	138(123)
	160	4.56	1.80	1274	156(142)
$R\bar{3}m$–$LiScH_{10}$	300	4.96	0.67	1538	52(40)
(M)	200	5.50	0.78	1351	65(53)
	150	5.90	0.90	1161	75(65)
Fd-3*m*–Li_2ScH_{16}	300	5.64	2.04	1153	262(244)
(M)	250	5.89	2.74	815	276(260)
	230	6.00	3.47	515	281(265)
Fd-3*m*–Li_2ScH_{17}	300	3.94	1.11	941	94(80)
(M)	250	3.57	1.25	660	83(69)
	220	2.66	1.27	312	36(30)
Fd-3*m*–Li_2YH_{16}	300	5.19	1.66	1502	251(233)
(M)	250	5.37	1.99	1308	263(246)
	200	5.57	2.78	924	276(260)
	170	5.71	4.57	546	285(271)

This paper is dedicated to the 70th anniversary of the physics of Jilin University.

ACKNOWLEDGMENTS

We thank Dr. J. Yang for helping collect data from the supercomputer. We also thank Prof. Yanming Ma for his valuable discussions and suggestions. This work was supported by the National Key Research and Development Program of China (Grant No. 2021YFA1400203), the Major Program of the National Natural Science Foundation of China (Grant No. 52090024), the National Natural Science Foundation of China (Grants No. 12074138, 11874175, 11874176), the China Postdoctoral Science Foundation (Grants No. 2020M681032, No. 2021T140264), and the Strategic Priority Research Program of Chinese Academy of Sciences (Grant No. XDB33000000).

APPENDIX: STRUCTURAL PARAMETERS

TABLE II. Structural parameters of Li–*R*–H compounds at 300 GPa, where M indicates the metastable phase.

Compound	Lattice	Atomic coordinates (fractional)			
(S.G.)	(Å,°)	Atoms	*X*	*Y*	*Z*
Li_2ScH_{20}	$a = 6.621$	Li(4f)	0.192	0.500	0.000
(*Immm*)	$b = 4.514$	Sc(2a)	0.000	0.000	0.000
	$c = 2.889$	H(8l)	0.000	0.641	0.846
	$\alpha = 90.0$	H(16o)	0.812	0.209	0.768
	$\beta = 90.0$	H(4e)	0.287	0.000	0.000
	$\gamma = 90.0$	H(8n)	0.587	0.678	0.000
		H(4e)	0.406	0.000	0.000
Li_2YH_{17}	$a = 6.886$	Li(16c)	0.000	0.000	0.000
($Fd\bar{3}m$)	$b = 6.886$	Y(8b)	0.375	0.375	0.375
	$c = 6.886$	H(96g)	0.675	0.675	0.368
	$\alpha = 90.0$	H(32e)	0.216	0.216	0.216
	$\beta = 90.0$	H(8a)	0.125	0.125	0.125
	$\gamma = 90.0$				
$Li_3Y_2H_{36}$	$a = 8.427$	Li(4e)	0.250	0.250	0.000
($C2/m$)	$b = 4.862$	Li(2a)	0.000	0.000	0.000
	$c = 6.878$	Y(4i)	0.251	0.000	0.625
	$\alpha = 90.0$	H(8j)	0.937	0.708	0.011
	$\beta = 145.0$	H(4i)	0.067	0.000	0.853
	$\gamma = 90.0$	H(8j)	0.541	0.814	0.365
		H(8j)	0.563	0.298	0.640
		H(4i)	0.436	0.000	0.226
		H(8j)	0.255	0.100	0.369
		H(8j)	0.252	0.821	0.219
		H(4i)	0.258	0.000	0.138
		H(4h)	0.000	0.428	0.500
		H(4i)	0.355	0.000	0.989
		H(4i)	0.146	0.000	0.778
		H(8j)	0.957	0.812	0.779
Li_2LaH_{17}	$a = 7.011$	Li(16c)	0.000	0.000	0.000
($Fd\bar{3}m$)	$b = 7.011$	La(8b)	0.375	0.375	0.375
	$c = 7.011$	H(96g)	0.075	0.075	0.361
	$\alpha = 90.0$	H(32e)	0.215	0.215	0.215
	$\beta = 90.0$	H(8a)	0.125	0.125	0.125
	$\gamma = 90.0$				
$LiScH_{10}$(M)	$a = 4.178$	Li(1b)	0.500	0.500	0.500
($R\bar{3}m$)	$b = 4.178$	Sc(1a)	0.000	0.000	0.000
	$c = 4.178$	H(2c)	0.737	0.737	0.737
	$\alpha = 38.9$	H(6h)	0.972	0.972	0.448
	$\beta = 38.9$	H(2c)	0.377	0.377	0.377
	$\gamma = 38.9$				
Li_2ScH_{16}(M)	$a = 6.652$	Li(16c)	0.000	0.000	0.000
($Fd\bar{3}m$)	$b = 6.652$	Sc(8b)	0.375	0.375	0.375
	$c = 6.652$	H(96g)	0.819	0.819	0.631
	$\alpha = 90.0$	H(32e)	0.210	0.210	0.210
	$\beta = 90.0$				
	$\gamma = 90.0$				
Li_2ScH_{17}(M)	$a = 6.740$	Li(16c)	0.000	0.000	0.000
($Fd\bar{3}m$)	$b = 6.740$	Sc(8b)	0.375	0.375	0.375
	$c = 6.740$	H(96g)	0.075	0.075	0.376
	$\alpha = 90.0$	H(32e)	0.219	0.219	0.219
	$\beta = 90.0$	H(8a)	0.125	0.125	0.125
	$\gamma = 90.0$				
Li_2YH_{16}(M)	$a = 6.808$	Li(16d)	0.500	0.500	0.500
($Fd\bar{3}m$)	$b = 6.808$	Y(8a)	0.125	0.125	0.125
	$c = 6.808$	H(96g)	0.071	0.071	0.863
	$\alpha = 90.0$	H(32e)	0.709	0.709	0.709
	$\beta = 90.0$				
	$\gamma = 90.0$				

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