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Second group of high-temperature lanthanide polyhydride superconductors under high pressure

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Rare-earth polyhydrides formed under pressure are promising conventional superconductors, with the critical temperature $T_c$ in compressed LaH$_{10}$ almost reaching room temperature. Here, we report a systematic computational investigation of the structural and superconducting properties of rare-earth (RE) polyhydrides formed under pressure across the whole lanthanide series. Analyses of the electronic and dynamical properties and electron-phonon coupling interaction for the most hydrogen-rich hydrides REH$_n$ ($n = 8, 9, 10$) that can be stabilized below 400 GPa show that enhanced $T_c$ correlates with a high density of H $s$-states and low number of RE $f$-states at the Fermi level. In addition to previously predicted and measured LaH$_{10}$ and CeH$_9$, we suggest YbH$_{10}$ and LuH$_8$ as additional potential high-$T_c$ superconductors. They form a ‘second island’ of high-$T_c$ superconductivity amongst the late lanthanide polyhydrides, with an estimated $T_c$ of 102 K for YbH$_{10}$ at 250 GPa.

Metallic hydrogen has been suggested as a potential conventional high-temperature superconductor [1]. Although hydrogen is predicted to become metallic and have high superconducting critical temperature $T_c$ at pressures beyond 450 GPa [2, 3], it is still a challenge to achieve such immense pressure in experiments, and the most recent studies report somewhat diverging properties of compressed hydrogen [3–5]. Meanwhile, Ashcroft suggested that ‘chemical precompression’ of hydrogen in hydride materials would lead to metallization at much lower pressures [6]. As a consequence, theoretical calculations have predicted a wide range of promising pressure induced metallic hydrides [7–14], such as sodalite-like clathrate CaH$_6$ and YH$_6$, where predicted $T_c$ values reach about 235 and 290 K at 150 and 300 GPa, respectively [7, 14]. Hydrogen sulfide was first theoretically reported as a good candidate for high-$T_c$ superconductivity [15–17], before experiments confirmed that H$_2$S has a $T_c$ of 203 K at pressures of 155 GPa [18]. The excellent agreement between theory and experiment greatly increased confidence in computational predictions. Encouraging results have spurred a flurry of interest in other compressed hydrides, that is, solid materials containing hydrogen atoms bonded to other elements for related hydrides with both higher $T_c$ and potentially broader ranges of stability [19–22].

Peng et al. have reported stable hydrogen-rich (H-rich) compounds with clathrate structures by using crystal structure prediction on rare-earth (RE) metallic hydrides under extreme conditions [23]. They found that clathrate H-rich hydrides allow high H content with purely atomic character, i.e. no H$_2$ molecules, and highlighted the cubic phase LaH$_{10}$ with a H$_{32}$ cage and surprisingly high $T_c$ of 288 K at 200 GPa. The calculated $T_c$ for sodalite-like fcc YH$_{10}$ is predicted to reach about 326 K, above room-temperature [20–23], elevating the class of H-rich hydrides to potential room-temperature superconductors [24, 25]. Most recently, experiments by Drozdov et al. [26] and Somayazulu et al. [27] have established LaH$_{10}$ as the highest measured $T_c$, reaching 250 K at 170 GPa, setting the current record of superconducting $T_c$ near room-temperature. While stability of the various RE polyhydrides had been studied by Peng et al. [23] across the entire lanthanide series of elements, superconductivity properties have only been reported for the early RE hydrides of La, Ce, and Pr. Calculations of electronic and $T_c$ properties have accompanied recent experimental reports on syntheses of individual RE polyhydrides, CeH$_9$ [28, 29], PrH$_6$ [30, 31], NdH$_9$ [32], EuH$_9$ [33] and ThH$_{10}$ [24]. However, a systematic investigation of electron-phonon coupling and superconducting $T_c$ across all RE hydrides is missing. A theoretical analysis of hydride superconductivity across the periodic table by Semenok et al. [34] used a neural network model to predict low $T_c$ across the lanthanide series, as increasing numbers of RE $f$-electrons at the Fermi level can weaken the electron-phonon coupling (EPC) interaction and suppress superconductivity.

In this article, we present a systematic study of potential hydride superconductivity across the entire lanthanide series, focusing on the most H-rich RE hydride species stabilized by each RE element under pressure. We mainly focus on the H-rich clathrate REH$_n$ ($n = 8, 9, 10$) hydride structures with atomic H$_{24}$, H$_{28}$, H$_{29}$, and H$_{32}$ cages and perform systematic studies of their energetic, dynamical, and electronic properties. Electronic structure properties reveal that large density of H-like electronic states at the Fermi level ($N_{cf}$) correlates with high superconducting $T_c$, while increasing the relative contribution of RE $f$-electrons is associated with...
weakening of EPC interaction. This leads to a strong suppression of $T_c$ in the middle of the lanthanide series but also a ‘second island’ of superconductivity in the late lanthanides, as we show that the late RE hydrides YbH_{10} and LuH_{8} (which become stable between 250 and 300 GPa) also have potential for high-temperature superconductivity ($T_c \sim 100$ K). We propose that H-rich hydrides of late lanthanides, with fully filled f-shells, should not be overlooked by experimental groups who attempt to synthesize the various RE polyhydrides.

We conducted crystal structure searches for the early lanthanide hydrides using the particle swarm optimization method as implemented in the CALYPSO package [35–37], focusing on the REH$_n$ (n = 3–10) stoichiometries at 100, 150, 200, 250, 300 and 400 GPa. Subsequently, all candidate structures from our own searches combined with previous reports [23, 34] were applied across the whole lanthanide series and the full 100–400 GPa pressure range. The validity of CALYPSO in structure prediction has been demonstrated for various systems which have been confirmed by experiment [15, 20, 23, 38], including high pressure lanthanide hydrides [20, 23]. The underlying total energy calculations and structure optimizations were performed at density functional theory (DFT) level of theory as implemented in the Vienna Ab Initio Simulation Package (VASP) [39]. Projector augmented wave (PAW) potentials with $5s^25p^66s^2(5d,4f)^n$ valence spaces were used to represent the electron-ion interactions [39, 40]. The Perdew-Burke-Ernzerhof (PBE) [41] functional was employed for exchange-correlation effects. Plane wave bases with cutoff energy of 800 eV and dense $k$-point meshes with $2\pi \times 0.03$ Å$^{-1}$ spacing gave excellent convergence of the structural relaxations and electronic calculations.

The electronic band structures, electron density of states (DOS) and electron-phonon coupling were determined using density functional perturbation theory (DFPT) as implemented in the QUANTUM-ESPRESSO code [42]. Ultrasoft pseudopotentials for lanthanide rare-earth RE and H atoms were chosen and plane wave cutoff energy was 80 Ry. For calculations of superconducting electron-phonon coupling (EPC) parameter $\lambda$, the Brillouin zone was sampled using a $4 \times 4 \times 4 \times 4 \times 4$ $q$-point mesh and much denser $24 \times 24 \times 24 \times 32 \times 32 \times 16$ $k$-point mesh for cubic (hexagonal) hydrides structures. The superconducting critical temperature ($T_c$) was estimated by solving the Allen-Dynes modified McMillan equation [43], which has been shown to give very good results in compressed metal hydrides [16, 23]. In our calculation, Coulomb pseudopotential $\mu^* = 0.1$ and 0.13 were used.

We explored the energetically stable phases of lanthanide hydrides (REH) at high pressures by constructing the relevant convex hulls. In Figs. 1, S1 and S2 in the Supplementary Information (SI), the convex hulls and resulting pressure-composition phase diagrams for all RE hydrides are shown relative to REH$_3$ and H$_2$ and across the pressure range from 100 to 400 GPa. For all lanthanides, REH$_4$ is a stable structure. For almost all lanthanides, REH$_{10}$ is stable at high pressures (exceptions are Ho, Er, Tm, and Lu), and usually REH$_9$, in a cubic or hexagonal structure, becomes stable at slightly lower pressures (the exception is La, where LaH$_{10}$ is very stable at relatively low pressures already). The sodalite-like REH$_6$ (Im3m) phase emerges as an additional stable structure from Pm onwards. The first stable REH$_{3}$ stoichiometry is predicted for NdH$_8$, which also occurs as preferred stable clathrate structure (> 250 GPa) in the late lanthanide hydrides of Ho, Er, Tm and Lu. With the exception of orthorhombic (Immm) LuH$_8$, all other REH$_8$ hydrides are stable in a cubic Fm3m phase. The most hydrogen-rich RE hydrides are therefore (at least up to 400 GPa) either REH$_8$, REH$_9$, or REH$_{10}$. For early RE hydrides La and Ce, we have reproduced the various structures (including LaH$_{10}$ and CeH$_8$), which have been experimentally synthesised under high pressure [26–29]. The predicted mid-lanthanide structures are in agreement with the results by Peng et al. [23]: the most H-rich clathrate hydride REH$_{10}$ is stabilised across RE = Nd, Pm, Sm, Eu, Gd, Tb, and Dy. For the late lanthanide systems of Er, Tm, Yb and Lu hydrides, the ErH$_8$, TmH$_8$, YbH$_{10}$ and LuH$_8$ phases emerged as stable hydrides at pressures above 250 GPa, which complements prior theoretical and experimental work [20, 23, 28]. As illustration, the thermodynamic convex hull construction for polyhydrides YbH$_n$ and LuH$_n$ at different pressures is depicted in Fig. 1(a, b), from which the clathrate hy-
drides $Fm\bar{3}m$ YbH$_{10}$, $F\bar{4}3m$ YbH$_{9}$ and $Immm$ LuH$_{8}$ are predicted as stable structures under high pressure. The corresponding pressure-composition diagrams of REH$_n$ (RE = La, Ce, Yb, Lu, $n = 8, 9, 10$) are shown in Fig. 1(c).

In Fig. 2, we have shown the four relevant, stable H-rich clathrate structures, which include $Fm\bar{3}m$ REH$_8$, $F\bar{4}3m$ REH$_9$, $P6_3/mmc$ REH$_8$ and $Fm\bar{3}m$ REH$_{10}$. In $Fm\bar{3}m$ REH$_8$ hydrides (Fig. 2(a)), $H_{24}$ cages surround the central metal atoms; this is the most H-rich phase found in the Nd, Ho, Er, and Tm hydride systems. The $Immm$ LuH$_8$ structure is a small distortion of this structure type. In the cubic phase ($F\bar{4}3m$) for REH$_9$ in Fig. 2(b), $H_{28}$ cages surround the central atoms; this structure appears at some conditions as the most H-rich hydride in the Pr, Nd, Pm, Sm, Ho and Yb systems. Meanwhile, $H_{29}$ cage structures in hexagonal phase REH$_9$ (Fig. 2(c)) appear stable in Ce, Eu, Gd, Tb and Dy hydrides. Lastly, in the cubic $Fm\bar{3}m$ REH$_{10}$ structure formed by most RE elements (see above), $H_{32}$ cages surround every metal atom (Fig. 2(d)). The absence of imaginary phonons in the dispersion curves (Fig. S3 and S4) confirms the dynamical stability of all predicted polyhydrides.

From here on, we focus on the properties of the predicted most H-rich REH$_n$ ($n = 8, 9, 10$) clathrate hydrides structures for each RE element. The electronic density of states (DOS) calculations of these phases (Figs. 3 and S5) confirm their metallic character across the series. As shown in Fig. S5, the striking narrow 4f-band of electronic states moves from above the Fermi level (LaH$_{10}$, CeH$_8$) to below the Fermi level (YbH$_{10}$, LuH$_8$) along the lanthanide series. The inset figures in Fig. S5 show the electronic DOS around the Fermi level, which are dominated by RE-f and H-s states. Fermi surfaces of all RE hydrides have been displayed in Figs. S6 and S7. The number of projected H-s and RE-f states at the Fermi level for the various H-rich RE hydrides is shown in Fig. 3(a). The projected f-DOS behaves as expected: essentially zero for the early RE elements (La, Ce and Pr), the RE f-states increase rapidly with atomic number. A maximum is reached for the Eu-H hydride system (EuH$_{10}$ structure), which corresponds to a half f-filled [Xe] 6s$^2$4f$^7$ one-electron configuration. From Eu-H towards the latter lanthanide hydrides, the RE f-DOS reduces as the 4f-states are mainly below the Fermi energy (shown in Fig. S5). For Yb and Lu hydrides, the f-DOS at the Fermi energy is basically zero again. For the H-s states (note different scale in Fig. 3(a)), their presence at the Fermi level decreases with atomic number across the early RE elements, from La to Pr. There is some structure amongst the mid-RE hydrides from Nd-H to Sm-H, but in general the H-s states reach a low plateau only to resurge again for the late RE hydrides of Tm, Yb and Lu. The general trend of electronic DOS of H-s and RE-f states at the Fermi level in the different polyhydrides is different structures notwithstanding) in agreement with the valence states of RE atoms in lanthanide compounds [44, 45].

The electronic structure in the late RE hydrides (Tm, Yb, Lu) thus mirrors the situation for the early RE hydrides (La, Ce, Pr): a high number of H-s states at
the Fermi level and low RE-$f$ character, which could be promising for strong electron-phonon coupling (EPC) and potential superconductivity [1, 46]. We therefore performed the EPC calculations for all H-rich lanthanide polyhydride systems, and collected superconducting properties including the EPC parameters $\lambda$, $\omega_{\log}$ and $T_c$ at different pressures in Table S1 in the SI. In Fig. 3(b), we show our calculated $T_c$ results across the entire lanthanide hydride series. Results for LaH$_{10}$ and CeH$_9$ hydrides ($T_c \sim 224$ and 87 K, $\mu^* = 0.1$) are in agreement with the established experimental results [26, 27] and calculations for LaH$_{10}$ ($T_c \sim 250$ K, $\mu^* = 0.1$) and CeH$_9$ ($T_c \sim 117$ K, $\mu^* = 0.1$) [28]. The trend for $T_c$ for lanthanide polyhydrides away from La and Ce is strongly downwards. When arriving at RE elements around the half-filled 4$f$ states (Eu, Gd, Dy and Ho), the DOS of H-$s$ states reaches the lowest values, while the RE-$f$ states have high values, resulting in weak EPC and low $T_c$. However, as the atomic number of the lanthanide increases further, a ‘second island’ of superconductivity appears in Yb and Lu polyhydrides, reaching 102 K at 250 GPa in YbH$_{10}$ (80 K in YbH$_9$) and 86 K at 300 GPa in LuH$_8$. The EPC parameters $\lambda$ for YbH$_9$ and YbH$_{10}$ (250 GPa) and LuH$_8$ (300 GPa) are 1.02, 1.10, and 2.18, respectively, which is larger than those predicted for CeH$_9$ [28], see Table 1. Our calculations focused on calculations of structurally similar H-rich clathrate hydrides across the entire lanthanide series, REH$_n$ ($n = 8, 9, 10$), and indicate the ‘secondary wave’ of superconductivity begins with Yb. There are hints that other structures hold promise as well: Semenik et al. [34] reported an upturn in $T_c$ in latter lanthanide atoms from Ho and Er onwards, with maximum $T_c$ for $I4/mmm$ HoH$_4$ and $P62m$ ErH$_{15}$ as 36.7 and 31.5 K ($\mu^* = 0.1$), respectively (see also Fig. 3(b)).

At low pressure, the divalent character of Yb coupled with propensity to valence fluctuations distinguish it from the other lanthanide elements [44, 47–49]. YbH$_2$ has been reported to undergo a structural and insulator-metal transition by neutron diffraction under high pressure, and suggested that superconductivity emerges at higher pressures with strong EPC driven by enhanced charge fluctuations [48]. As YbH$_{10}$ leads the ‘secondary wave’ of high-temperature superconductivity in our calculations, we show detailed electronic properties of YbH$_{10}$ at the single-particle level at 250 GPa in Fig. 4. The electronic band structure and DOS of YbH$_{10}$ shown in Fig. 4 (a) indicate the metallic character of YbH$_{10}$.

<table>
<thead>
<tr>
<th>Element</th>
<th>Polyhydrides</th>
<th>Pressure (GPa)</th>
<th>$\lambda$</th>
<th>$\omega_{\log}$, K</th>
<th>$T_c$ (K) $\mu^* = 0.10$</th>
<th>$\mu^* = 0.13$</th>
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<td>1653.9</td>
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<td>0.3</td>
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<td>953.6</td>
<td>1.7</td>
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<tr>
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<td>565.7</td>
<td>86.2</td>
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with several bands crossing the Fermi level. According to the projected DOS, the main contributions at the Fermi energy come from Yb-d, Yb-f and H-s orbitals (shown as inset in Fig. 4(a)). There is a sharp peak of Yb-f states just below the Fermi level. Detailed Fermi surfaces are displayed in Fig. 4(c-f) and agree qualitatively with the Fermi surface of LaH$_{10}$ (see Fig. S6): three electron pockets around Γ (two very small, one large and with multiple protrusions), and one sizeable hole pocket around X. The appearance of ‘flat band-steep band’ Fermi level crossings [46] and relatively large number of H-s states at the Fermi level again indicate YbH$_{10}$ could display high temperature superconductivity, which is consistent our EPC calculations above ($T_c \sim 102$ K). Phonon dispersion curves for YbH$_{10}$ structure are displayed in Fig. 4(b), together with projected phonon DOS, the Eliashberg EPC spectral function $\alpha^2 F(\omega)$ and the cumulative EPC integral $\lambda(\omega)$. There are two separate phonon frequency ranges: the acoustic phonon modes dominated by Yb atoms below 250 cm$^{-1}$, which contribute relatively little to the total EPC $\lambda$, and H-dominated optical modes above 675 cm$^{-1}$, vibrations in the clathrate H$_{32}$ cages, that contribute most to $\lambda$. For LuH$_8$ hydride, the detailed electronic properties and EPC parameters are displayed in a similar way in Fig. 5. The ‘flat band-steep band’ Fermi level crossings also emerge in Fig. 5(c), indicating that LuH$_8$ could support high temperature superconductivity. Detailed electron-phonon calculation results are shown in Fig. 5(b,d), which result in $T_c$ about 86.2 K, with large EPC $\lambda \sim 2.18$ (see also Table 1). In LuH$_8$, as broadly found in other high $T_c$ hydrides [19, 20, 28, 50], the high-frequency H-dominated vibrations make the largest contribution to EPC $\lambda$ and thus sizeable $T_c$.

Comparing the superconductivity trends across the whole lanthanide hydride series, our results show that, while LaH$_{10}$ remains the record holder for highest $T_c$ also in our calculations, YbH$_{10}$ and LuH$_8$ hydrides should also be considered as potential high temperature superconductors. The H-rich clathrate hydrides with atomic hydrogen cage structures are stable across the entire series at high pressure, and the high-frequency hydrogen sublattice vibrations contribute effectively to strong EPC and enhanced superconductivity. Recently, continuing synthesis efforts have made inroads into the lanthanide series of hydrides, including Pr [30, 31], Nd [32], Eu [33] and Dy [51]. Pushing towards the late lanthanides might add further insight into the limits of BCS superconductivity and the ultimate goal to achieve room-temperature $T_c$.

In summary, we have systematically explored superconductivity in H-rich REH$_n$ hydrides for the entire lanthanide hydride series, with combined structure searches and first-principles DFT calculations. We uncovered several new RE-H polyhydride structures with characteristic hydrogen clathrate cages; the most H-rich phases feature H$_{24}$, H$_{28}$, H$_{25}$ and H$_{32}$ cages in high-symmetry structures REH$_n$ ($n = 8, 9, 10$), with RE atoms located at the centers of the hydrogen cages. Detailed DOS and EPC calculations for the respective most H-rich phases across the series show that high number of H-s states and low number of RE-f states at the Fermi level lead to strong EPC interactions and high-$T_c$ superconductivity. To the known LaH$_{10}$ and CeH$_8$ hydrides we add YbH$_{10}$ and LuH$_8$ at the other end of the series with sizeable predicted $T_c$. We suggest experimental groups who attempt to synthesize RE polyhydrides with high-$T_c$ explore the ‘second island’ of superconductivity in the late RE polyhydrides.

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FIG. 5: (a) Crystal structure of Immm-LuH$_8$ at 300 GPa, (b) Fermi surface sheets of LuH$_8$, (c) Calculated electronic band structures and density of states (DOS) of LuH$_8$, (d) Phonon dispersion curves, phonon density of states (PHDOS) projected on Lu and H atoms and Eliashberg spectral function $\alpha^2 F(\omega)$ together with the electron-phonon integral $\lambda(\omega)$.


