Author's response to the comments

The reviewers commend the paper for clearly framing the challenge of discovering thermodynamically stable metal superhydrides, executing a polished workflow that melds chemical templates with machine-learning–guided screening, and expanding the catalogue of structural prototypes, demonstrating promising in-silico discoveries. Efficiency claims lack support because no computational-cost benchmarks are given and the authors could instead have run high-throughput DFT on all 8,000 lattices.

The conditional acceptance is based upon addressing the following:

Comments: *Explain why a stack-ensembling AutoGluon approach was selected and compare its performance to graph-based ML architectures cited in the literature.*

Authors' reply: Graph based ML architectures were not cited. This work uses a supervised learning approach to screen templates, it is not feasible to benchmark the model against graph-based ML architectures. The prediction of the high-accuracy thermodynamic stability of metal superhydrides was performed using ensembling multiple ML algorithms integrated into the AutoGluon framework, which has demonstrated high reliability in previous works [1–3].

Comments: *Provide complete, reproducible details of the ensemble setup (base models, meta-learner, hyper-parameters, training protocol, code).*

Authors' reply: The details of the ensemble setup are uploaded into Github: https://github.com/hison001/Metal-superhydrides/tree/main.

Comments: *List and justify the 46 input features shown in Figure 1B.*

Authors' reply: The input features and their importance in Figure 1B are shown below.

Feature name	**Feature importance**
max packing efficiency	1.019936
ELFmax	0.326033
maximum neighbor distance variation	0.171131
minimum neighbor distance variation	0.136029
mean neighbor distance variation	0.110488
number_H	0.09614
dimensionality	0.087374
ELFavg	0.066839
mean Average bond length	0.051677
packing fraction	0.044652
vpa	0.028975
MagpieData avg_dev MeltingT formula	0.02519
ratio_H2M	0.020042
MagpieData avg_dev GSvolume_pa formula	0.019598

MagpieData mean MendeleevNumber formula	0.019044
MagpieData avg_dev MendeleevNumber formula	0.017875
density	0.016197
MagpieData mean GSvolume_pa formula	0.015639
PymatgenData mean atomic_radius formula	0.014709
number_M	0.013134
MagpieData mean MeltingT formula	0.01156
MagpieData mean Column formula	0.011531
MagpieData range Electronegativity formula	0.010626
MagpieData avg_dev Column formula	0.01
MagpieData avg_dev CovalentRadius formula	0.009912
M_MagpieData mean CovalentRadius	0.008839
M_MagpieData mean MendeleevNumber	0.008722
MagpieData mean NdValence formula	0.008402
MagpieData avg_dev NdUnfilled formula	0.008157
PymatgenData std_dev atomic_radius formula	0.007511
MagpieData mean CovalentRadius formula	0.00741
MagpieData mean Electronegativity formula	0.005855
MagpieData avg_dev Electronegativity formula	0.005351
MagpieData maximum AtomicWeight formula	0.005034
MagpieData avg_dev NValence formula	0.004871
MagpieData mean NValence formula	0.004327
MagpieData minimum Electronegativity formula	0.003902
MagpieData avg_dev AtomicWeight formula	0.003231
MagpieData range Number formula	0.002902
PymatgenData mean thermal_conductivity formula	0.001599
MagpieData avg_dev Number formula	0.000866
MagpieData avg_dev NUnfilled formula	0.000562
MagpieData mean Number formula	0.000093
MagpieData mean Row formula	-0.001119
MagpieData mean AtomicWeight formula	-0.001633

Comments: *Describe the training data in full: name the 57 starting prototypes, outline the metal-substitution augmentation, and show that none re-appear in the evaluation split.*

Authors' reply: The 57 starting prototypes are shown below.

No.	Protptype	No.	Protptype
1	AcH_{10}_Cm	30	PaH_4_Fmmm
2	AcH_{16}_P6/mmm	31	PrH_6_C2/m
3	AcH_4_Cmcm	32	PrH_8_$P6_3mc$
4	AcH_5_C2/m	33	ScH_{10}_Cmcm
5	AcH_7_$Cmc2_1$	34	ScH_{12}_C2/c

6	AcH_8_C2/m	35	ScH_{12}_Cmcm
7	BaH_{10}_Cmmm	36	ScH_6_Cmcm
8	BaH_{12}_ I4/mmm	37	ScH_7_Cmcm
9	BaH_{12}_ $P6_3/mmc$	38	ScH_8_Immm
10	BaH_{12}_$P2_1$	39	ScH_9_$I4_1md$
11	BaH_6_Imm2	40	SrH_{10}_$P2_1/c$
12	BaH_8_Imma	41	SrH_{12}_C2/m
13	CaH_{12}_C2/c	42	SrH_8_$P2_1/c$
14	CaH_{12}_$R\bar{3}$	43	ThH_{10}_$Fm\bar{3}m$
15	CaH_6_$Im\bar{3}m$	44	ThH_7_$P2_1/c$
16	CaH_9_C2/m	45	Ti_2H_{13}_Immm
17	CeH_9_$P6_3/mmc$	46	TiH_{14}_C2/m
18	HfH_{14}_C2/m	47	TiH_{16}_Pbam
19	HfH_4_Fddd	48	TiH_{22}_C2/m
20	HfH_6_$Cmc2_1$	49	UH_7_$P6_3/mmc$
21	LaH_{11}_P4/nmm	50	UH_8_$Fm\bar{3}m$
22	LaH_{16}_P6/mmm	51	YH_{12}_C_2/m
23	LaH_4_I4/mmm	52	YH_{13}_$R\bar{3}m$
24	LaH_5_C2/m	53	YH_9_$P6_3/m$
25	LaH_6_$R\bar{3}c$	54	ZrH_{10}_$P6_3/mmc$
26	LaH_7_$Cmc2_1$	55	ZrH_6_$Cmc2_1$
27	LaH_8_C2/m	56	ZrH_6_ $P2_1/c$
28	LaH_9_Cc	57	ZrH_8_ I4/mmm
29	NdH_9_$F\bar{4}3m$		

Comments: *Specify how the test set was constructed and report model metrics on this held-out data.*

Authors' reply: The efficient identification of suitable metal lattices is crucial for advancing the discovery of metal superhydrides. Besides the body-centered cubic (BCC), face-centered cubic (FCC), and hexagonal close-packed (HCP) frameworks, which typically host only a few metal atoms per primitive cell and rarely yield stable superhydrides with non-integer $\frac{x_H}{x_M}$ (e.g., Ti_2H_{13} [4]), we broadened the search to include 52 complex metal lattice prototypes selected from the Crystal Lattice Structure sites (http://cst-www.nrl.navy.mil/lattice/) and from variant stacking sequences of close-packed atomic layers [5].

Comments: *Define the "rapid evaluation method" that served as a proxy for full electron-phonon calculations.*

Authors' reply: The estimated T_c by applying $T_c = (750\Phi_{DOS} - 85)K$ proposed by Belli et al [6]. Where, $\Phi_{DOS} = \varphi H_f \sqrt[3]{H_{DOS}}$, φ is the maximum electron localization function (ELF)

value that the 3D ELF network spanning through the entire cell, H_f is the fraction of number of hydrogen atoms with respect to number of whole atoms in the primitive cell, H_{DOS} is the hydrogen fraction of the total density of states (DOS) at the Fermi level, respectively.[more detailed explanations are shown in *Nat. Commun. 12, 5381 (2021)*]

Comments: *Unambiguously state that the 30 predicted compounds that did not receive full electron-phonon calculations are preliminary results for which this calculation will need to be performed as the next step.*

Authors' reply: Following the reviewer's suggestion, we add the clear statement in the revised manuscript.

References

[1] C. Shen et al., Accelerated Screening of Ternary Chalcogenides for Potential Photovoltaic Applications, J. Am. Chem. Soc. **145**, 21925 (2023).

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[6] F. Belli, T. Novoa, J. Contreras-García, and I. Errea, Strong correlation between electronic bonding network and critical temperature in hydrogen-based superconductors, Nat. Commun. **12**, 5381 (2021).