Dear Editor,

Please find enclosed our revised manuscript "Superconductivity in RbH_{12} at low pressures: an *ab initio* study", which we would like to resubmit to SciPost Physics.

We would like to thank the Referees for their valuable comments on our manuscript. We have fully addressed the issues raised by the Referees and modified the paper accordingly. We believe that the revised manuscript is now suitable for publication in SciPost Physics. Below we have included a detailed response to the Referees and a description of the changes to the manuscript.

Response to the Referee 1.

(*)*I think this paper should be published. The main complaint will be that the more accurate calculations that account for multiple real physical effects missed in previous work do not, in fact, change the overall pressure-dependent phase hierarchy of this material. However, in general, those effects cannot be neglected when they are of the same order as the relevant energy differences calculated in the simplest manner and the accidental reproduction of previous results doesn't change that reality. The value of this work is in its careful explication of this fact, explanation of the correct methodologies and clear presentation of the right way to examine potential meta-stability in this important materials.*

We thank the Referee for taking the time to carefully read our manuscript and for his/her positive assessment and encouraging remarks about our methodology and results.

1.*One extremely trivial matter:*

In the last paragraph of the introduction the phrase "leaving *to* Immm and Cmcm phases to emerge" occurs. The "to" is odd. Is it supposed to be "two"? Or maybe "the"?

We thank the referee for noticing this. The phrasing was a literal translation from our native language and sounds unnatural in English. We have corrected it to "leaving the Immm and Cmcm phases to emerge" in the revised version.

Response to Referee 2.

(*)*In this paper the authors study partially the phase diagram, and the superconducting properties of rubidium hydrides under pressure. For the calculation of the phonon properties, they employ an advanced method, that is the stochastic self-consistent harmonic approximation, which allows them to include anharmonic and quantum effects in the phonon spectrum. The importance of these effects*

in hydrides was demonstrated in the previous literature by these and others developers of SSCHA, and it is often to promote a stabilization of hydride structure towards lower pressures.

From the side of the importance, it is hard to evaluate this paper. Rubidium hydrides have been studied already from the computational and experimental side. This paper proposes an improved study, in the sense that it employs SSCHA to compute the phonon properties. On the other hand, the authors do not study the experimentally published phases, but rather study a phase predicted in a previous high-throughput study. The main effect to SSCHA is to lower the stabilization pressure of this known phase, and may motivate further experimental studies.

Overall, I think this article satisfies the general acceptance criteria, but I believe it does not meet the more stringent expectations of SciPost physics. There is no breakthrough, novel link, or ground-breaking discovery.

We thank the referee for taking the time to review our work and for the thoughtful assessment. We understand the referee's point that our results are not of a "breakthrough" nature and that the study does not identify a particularly promising low-pressure hydride superconductor. Indeed, much of our work was carried out before experimental data on rubidium hydrides at lower pressures became available.

Nevertheless, we believe that our results make a meaningful contribution to the collective understanding of hydride superconductors. In particular, our application of the SSCHA method provides an advanced anharmonic and quantum-level characterization of the RbH_{12} phase, which may help guide and benchmark future theoretical and experimental investigations. Additionally, in our calculation of superconducting critical temperatures we have explicitly included the Coulomb repulsion providing a fully first principles estimation of superconducting T_C in these materials. We therefore view this study as a valuable addition to the broader effort to map and understand the complex phase diagram of hydrides under pressure.

We respond to the referee's specific comments and suggestions point-by-point below.

1.*In the abstract, and later in the results, the authors write that the Immm and P63/mmc phases will be dynamically stable down to a certain pressure even though their phonon dispersion exhibit imaginary phonons. The reason, verbatim, is "these are probably interpolation issues". They refer to the fact that a point for which the SSCHA matrices were computed has become stable, while another for which they were interpolated remained unstable. It is very hard to consider this argument as reliable evidence to support the claim that the structure is metastable at this pressure. I understand that the a larger supercell may be too expensive to compute, but this is not a valid argument to claim*
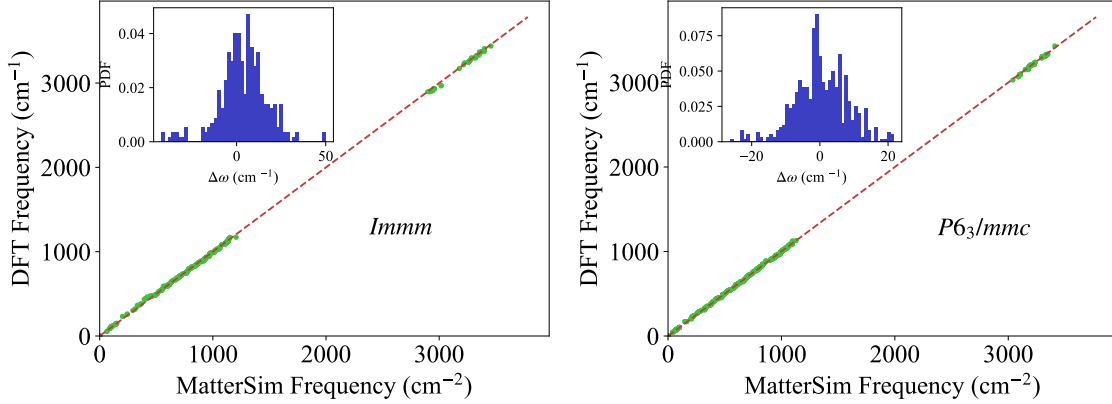
FIG. 1. Comparison between DFT and Mattersim calculated SSCHA auxiliary phonon frequencies in $2 \times 2 \times 2$ supercells of $Immm$ and $P6_3/mmc$ phase. The inset shows the histogram of calculated errors.

stability at that pressure.

To further substantiate our interpretation, we have performed additional calculations beyond those presented in the original submission.

Given the high computational cost associated with evaluating the required number of atomic forces, energies, and stresses, we employed the recently developed MatterSim machine learning potential [1]. Specifically, we fine-tuned the foundation model ("mattersim-v1.0.0-1M") using our previously computed DFT data for smaller supercells to accurately reproduce the relevant portions of the potential energy surface. TO justify the use of this interatomic potential we show the comparison between DFT and Mattersim calculated SSCHA auxiliary phonon frequencies in $2 \times 2 \times 2$ supercell in Fig. 1.

We then carried out SSCHA relaxations for the $Immm$ phase of RbH$_{12}$ at 25 GPa using a $3 \times 3 \times 3$ supercell (see Fig. 2). In this case, all Hessian phonon frequencies—both directly computed and interpolated—are positive. This confirms our earlier interpretation that the negative frequencies reported previously originated from interpolation artifacts rather than true dynamical instabilities.

We also repeated the analysis for the $P6_3/mmc$ phase. Since this structure contains two formula units per primitive cell, the largest feasible supercell corresponds to $3 \times 3 \times 2$ (468 atoms). Larger supercells are computationally prohibitive because the calculation of third-order force constants scales as N^3. In this system, we observe a clear renormalization and hardening of the Hessian phonon frequencies. The residual small imaginary modes are confined near the Γ point, consistent with remaining interpolation inaccuracies rather than genuine instabilities.

These additional results strengthen our conclusion that the previously reported imaginary frequencies were numerical artifacts and that both $Immm$ and $P6_3/mmc$ phases are dynamically stable at
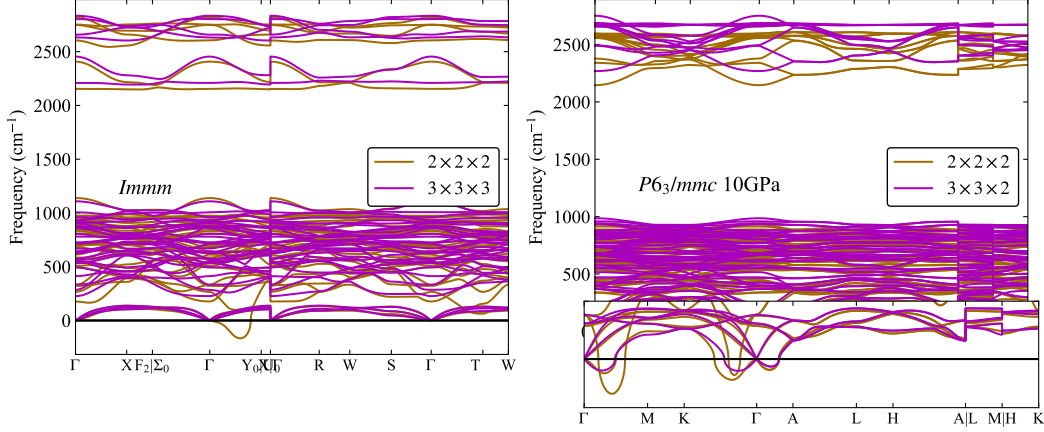
FIG. 2. Phonon band structures of $Immm$ (10 GPa) and $P6_3/mmc$ phases calculated from the Hessian of the total free energy.

the corresponding pressures. We have added this discussion to the Supplementary Material. The superconductivity calculations were still done with smaller supercell DFT results.

2. *The authors estimate the Raman activity of the phonon modes in a rather unconventional way. Since in metals the polarizability is not defined, they take a random tensor, symmetrize it, and compute some intensities. This is a clever trick, but I do not think that showing a simulated spectrum that is wrong (by the authors' own admission) is a good idea. I suggest they just show the peak centers as vertical lines with different colors to mark which one is active or not (by symmetry).*

We thank the referee for this comment and for recognizing our approach to identifying Raman-active phonon modes. We understand that the referee's main concern lies not with our method of determining mode activity, but with the way we present the results — namely, by showing the phonon spectral functions of Raman-active modes instead of displaying only the auxiliary phonon frequencies.

We respectfully note that, in our view, plotting only the auxiliary (harmonic) phonon frequencies would be less representative of the actual vibrational behavior, as it would neglect the anharmonic renormalization captured by the phonon spectral functions. Although the absolute Raman intensities obtained from our symmetrized random tensor approach are not physically meaningful, the spectral functions still convey valuable information on line broadening, peak shifts, and relative mode visibility. These are relevant physical features that are lost when showing only vertical lines at auxiliary frequencies.

For this reason, we prefer to retain the current representation, which we believe provides a more informative visualization of the Raman-active modes while clearly stating in the text that the
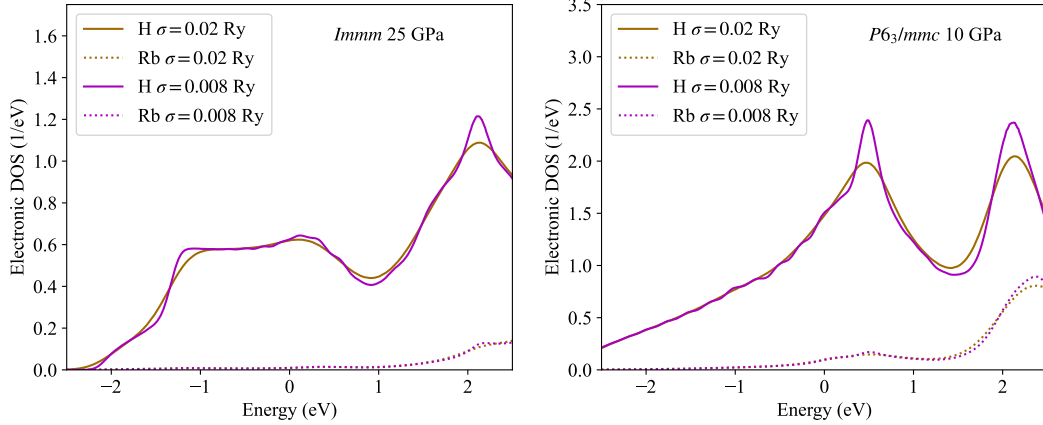
FIG. 3. Electronic density of states calculated for $Immm$ and $P6_3/mmc$ phases of RbH_{12} for two different smearing parameters (0.02 Ry (brown) and 0.008 Ry (magenta)).

intensities are not to be interpreted quantitatively.

3. *The smearing on the DOS in Fig. 5 appears to be the 0.02 Ry used to compute the charge density. The result unfortunately is a DOS which is all smeared out. I suggest they employ the 0.008 Ry smearing which they used for the matrix elements, which also gives an indication if the chosen k-grid is reasonable.*

We have repeated the DOS calculation using the smaller smearing of 0.008 Ry, consistent with the value employed for the matrix elements. The comparison between the original and updated results is presented in Fig. 3. The new calculation with reduced smearing is now included in the revised manuscript.

4. *There is an inconsistency between the y axis label of Fig. 1 and what the authors write ("Fig. 1 shows the Gibbs free energy"). I think it's because they take it at T = 0, but distinguish it from enthalpy because there is the zero point energy. However this is all very misleading. At T = 0 it makes no sense to call this a free energy. The zero-point energy is not part of the free energy, even though one may include it in the expression for Fh.*

The Gibbs free energy in the manuscript is defined at 0 K, where it effectively reduces to the enthalpy including the zero-point energy contribution. We have added a clarifying sentence stating that at 0 K the Gibbs free energy equals the enthalpy, and the figure caption and text have been revised for consistency.

5. *Concerning Fig. 4, it is also rather worrying that the lines are so irregular, and that they include so few points, especially since these are done with plain DFT calculations which should be very*

cheap. Moreover, there should also be a "RbH+11H" line, or something similar, i.e. a line showing how much higher are the proposed structure from the most important points in the convex hull. Showing only the enthalpy (or free energy) compared between RbH12 phases may mislead the reader into thinking that there is one RbH12 phase on the hull even at ambient pressure, which is not the case.

The apparent irregularity of the lines originates from the inclusion of the 0 GPa point. The large volume change between 0 GPa and 10 GPa leads to significant differences in relative enthalpy at these pressures, giving the curves their uneven appearance. For clarity, the 0 GPa point has been removed from the revised figure.

Regarding the reference to the convex hull, the figure was intended only to compare the relative enthalpies of the different RbH_{12} phases, all referenced to the *Immm* structure. The manuscript text already states this explicitly and clarifies that these values do not represent distances from the convex hull. We have reviewed this section to ensure that this distinction is now unambiguous and cannot mislead the reader.

5. *A minor point is that the authors write that "a first ab initio prediction of high-temperature superconductivity was done for H3S, followed immediately by the experimental confirmation". The real story is slightly more complicated, as the group of Eremets had been working on hydrogen sulfide independently, and tried multiple times to double-check the validity of the experiment. In fact, in the time between Duan's and Eremets' arXiv papers there are only a few days, which would not have been enough to do all those experiments. This was written by Eremets himself in Physics Reports 856, 1-78 (2020). I think it would be right to give the late Eremets credit for that.*

The sentence has been revised to state that the theoretical prediction and the experimental discovery of superconductivity in H_3S were published almost simultaneously, thereby properly reflecting the independent contributions of both groups.

6. *The authors used a mu* of 0.1 to solve the isotropic Eliashberg equations. In Ref. Nature Reviews Physics 6 509–523 (2024) it is clearly argued that this is inappropriate if the goal is to compare the results with the McMillan formula, as that formula assumes a mu* with a different cutoff on the Matsubara frequencies. Please take it into account.*

In response to this comment, we have performed a first-principles calculation of the Coulomb interaction for the *Immm* phase of RbH_{12}. From this, we obtained a value of $\mu = 0.159$. Using this value in the isotropic Eliashberg equations does not affect critical temperature at all, with the estimate from the full bandwidth calculation (including Coulomb interaction explicitly and allowing

for the Fermi level to change) remaining 98 K. The details of the calculation and the estimation procedure for μ are now included in the Supplementary Material.

With kind regards,

Đorđe Dangić, Yue-Wen Fang, Ion Errea

List of changes:

1. The sentence in the last paragraph of the introduction has been revised following the first referee's suggestion.

2. A reference to the first-principles Coulomb-interaction calculation has been added in the Methods section.

3. The first paragraph on pg. 3 now explicitly states that at 0 K the Gibbs free energy equals the enthalpy.

4. The second-to-last paragraph on pg. 3 now clarifies that the values shown in Fig. 1 are relative enthalpies and not distances from the convex hull.

5. The first paragraph on pg. 5 has been updated to clarify that the negative phonon frequencies in Fig. 3 arise from interpolation artifacts.

6. Additional information regarding the ab-initio treatment of Coulomb repulsion in the Migdal–Eliashberg calculations has been included in the second paragraph on pg. 6.

7. Figure 5 now presents the electronic density of states calculated using the same Gaussian smearing employed in the electron-phonon calculations.

8. The caption of Fig. 6 has been revised to better explain how the superconducting critical temperatures were obtained.

9. All Migdal–Eliashberg calculations involving μ^* have been repeated using $\mu^* = 0.118$, consistent with the value obtained for the *Immm* phase.

10. A section describing additional calculations using larger SSCHA supercells has been added to the Supplementary Material.

11. A section explaining our approach to solving the Migdal–Eliashberg equations with an ab-initio Coulomb interaction has been added to the Supplementary Material.

12. Nine additional references have been included to cite the relevant work that enabled the additional calculations.

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[1] H. Yang, C. Hu, Y. Zhou, X. Liu, Y. Shi, J. Li, G. Li, Z. Chen, S. Chen, C. Zeni, M. Horton, R. Pinsler, A. Fowler, D. Zügner, T. Xie, J. Smith, L. Sun, Q. Wang, L. Kong, C. Liu, H. Hao, and Z. Lu, Mattersim: A deep learning atomistic model across elements, temperatures and pressures, arXiv preprint arXiv:2405.04967 (2024), arXiv:2405.04967 [cond-mat.mtrl-sci].