

Design of high-temperature superconductors at moderate pressures by alloying AlH_3 or GaH_3

Xiaowei Liang,^{1,2} Xudong Wei,¹ Eva Zurek,³ Aitor Bergara,^{4,5,6} Peifang Li,⁷ Guoying Gao,^{1, a)} Yongjun Tian¹

¹*Center for High Pressure Science (CHiPS), State Key Laboratory of Metastable Materials Science and Technology, Yanshan University, Qinhuangdao, Hebei, 066004, China*

²*School of Materials Science and Engineering, Henan University of Technology, Zhengzhou, 450001, China*

³*Department of Chemistry, State University of New York at Buffalo, Buffalo, New York 14260-3000, United States*

⁴*Physics Department and EHU Quantum Center, University of the Basque Country, UPV/EHU, 48080 Bilbao, Spain*

⁵*Donostia International Physics Center (DIPC), 20018 Donostia, Spain*

⁶*Centro de Física de Materiales CFM, Centro Mixto CSIC-UPV/EHU, 20018 Donostia, Spain*

⁷*Extreme Conditions Physics Research Team, College of Mathematics and Physics, Inner Mongolia Minzu University, Tongliao 028043, China*

^{a)} **Authors to whom correspondence should be addressed:** gaoguoying@ysu.edu.cn

ABSTRACT

Since the discovery of hydride superconductors, a significant challenge has been to reduce the pressure required for their stabilization. In this context, we propose that alloying could be an effective strategy to get this. We focused on a series of alloyed hydrides with the AMH_6 composition, which could be made via alloying $\text{A}_{15} \text{AH}_3$ ($\text{A}=\text{Al}$ and Ga) with M ($\text{M}=\text{IIIB}$ and IVB group metals), and studied their behavior under pressure. According to the research, seven of them were predicted to maintain the A_{15} -type structure, similar to AH_3 under pressure, providing a platform for studying the effects of alloying on the stability and superconductivity of AH_3 . Among them, the A_{15} -type phases of AlZrH_6 and AlHfH_6 were found to be thermodynamically stable in the pressure range of 40-150 and 30-181 GPa, respectively. Furthermore, they remain dynamically stable at even lower pressures, as low as 13 GPa for AlZrH_6 and 6 GPa for AlHfH_6 . These pressures are significantly lower than what is required for stabilizing A_{15}

AlH₃. Additionally, the introduction of Zr and Hf into the alloys increases the electronic density of states at the Fermi level compared to AlH₃. This enhancement leads to higher critical temperatures (T_c) of 75 and 76 K for AlZrH₆ and AlHfH₆ at 20 and 10 GPa, respectively. In the case of GaMH₆ alloys, where M represents Sc, Ti, Zr or Hf, these metals reinforce the stability of the A15-type structure and reduce the lowest thermodynamically stable pressure for GaH₃, from 160 GPa to 116, 95, 80 and 85 GPa, respectively. Particularly noteworthy is the A15-type GaMH₆ alloys, which remain dynamically stable at low pressures of 97, 28, 5 and 6 GPa, simultaneously exhibiting high T_c s of 88, 39, 70 and 49 K at 100, 35, 10 and 10 GPa, respectively. Overall, these findings enrich the family of A15-type superconductors and provide insights for the future exploration of high-temperature hydride superconductors that can be stabilized at lower pressures.

I. INTRODUCTION

In recent years, hydrides have emerged as potential materials in the search of room-temperature superconductivity, not only from theoretical predictions¹⁻⁵ but also from experimental verifications.⁶⁻²² In 2015, the predicted cubic H₃S was synthesized for the first time and was experimentally confirmed to have a T_c of 203 K at 155 GPa.^{7,8} Four years later, the T_c record passed to the LaH₁₀ metal hydride with an H-clathrate structure, which was observed to have T_c values of 250-260 K at 170-190 GPa.^{9,10} Metallic clathrate hydrides were predicted to be a large class²³⁻²⁸ and some of the predicted superconductors in this family have been successively verified experimentally, such as ThH_{9/10} (146/161 K at 170-175 GPa),¹¹ YH₆ (224 K at 166 GPa; 220 K at 183 GPa),^{12,13} YH₉ (243 K at 201 GPa; 262 K at 182 GPa)^{13,14} and CaH₆ (215 K at 172 GPa; 210 K at 160 GPa).^{15,16} These achievements have greatly encouraged the exploration of room-temperature superconductivity in hydrides.

However, the possible application of superconducting hydrides is not only influenced by the superconducting transition temperature, as in the case of other superconductors, but also by the stabilization pressure. Although the hydrides mentioned above have high T_c values, the pressures required to stabilize them are also extremely high (> 150 GPa). Therefore, it is essential to obtain superconducting hydrides that are stable at low pressures. Our previous study proposed that the introduction of light B into the La-H system leads to a stable $Fm\bar{3}m$ LaBH₈ phase with BH₈ units,²⁹⁻³¹ which remains dynamically stable at 55 GPa and exhibits a T_c of 155 K. Subsequently, some other B, C and Si-based ternary hydrides were also estimated to show a good superconductivity at moderate pressures.³²⁻³⁵ In these hydrides, the covalent units formed by including light elements with H contribute significantly to their low-pressure stability. Furthermore, the La-Y alloy tetrahydride was synthesized at 110 GPa, exhibiting a T_c of 92 K, and can be recovered at 80 GPa, both of which are lower pressure thresholds than those required for the synthesis of YH₄.¹⁸ Bi *et al.* synthesized the (La,Ce)H₉ alloy at megabar pressures of 97–172 GPa, which exhibited a T_c of 148–178 K.¹⁹ More recently, the metastable compound $P6_3/mmc$ -LaH₁₀ was

stabilized at 146 GPa by introducing Al atoms to form $P6_3/mmc$ -(La,Al)H₁₀, with a T_c of 178 K.²⁰ An increase in the configurational entropy of a mixed alloy hydride will decrease its Gibbs free energy and enhance its stability. Therefore, alloying binary metal hydrides may be an alternative approach to optimize the stabilization pressure of superconducting hydrides.

Trihydrides are commonly found in binary metal hydrides and their superconductivity under pressure has been extensively studied.³⁶⁻⁴⁵ Based on their structures, common metal trihydrides can be divided into two categories, one with the $Pm\bar{3}n$ structure and the other with the $Fm\bar{3}m$ structure. This provides a platform to tune the stability and superconductivity of hydrides by alloying. The $Pm\bar{3}n$ structure, also called the A15 structure, is well known for its excellent superconducting performance. To date, about 50 alloys with this structure are superconductors, and some of them remain the most promising materials for practical applications.⁴⁶ Among binary hydrides, AlH₃, GaH₃, ZrH₃ and HfH₃ were predicted to become stable in the A15 structure at 73, 160, 8 and 27 GPa, respectively, in which metal atoms form a body-centered cubic (bcc) lattice with the six H atoms occupying half of the tetrahedral interstices of the bcc lattice.³⁶⁻⁴⁰ Interestingly, the A15 phase of AlH₃ and ZrH₃ were confirmed in experiments at 100 and 30 GPa, respectively, however, they do not present high values of T_c .^{37,39,41} On the other hand, the A15 phase of GaH₃ was estimated to have the highest T_c of 102 K at 120 GPa.³⁸ However, A15 GaH₃ presents relatively high minimum pressures of ~160 and ~84 GPa for thermodynamic and dynamic stability respectively,^{38,47} making its synthesis difficult. Below 160 GPa, A15 GaH₃ becomes thermodynamically unstable and transforms into a structure containing H₂ units, with an insulating character.²⁶ Comparing to Al and Ga, IIIB and IVB group metal atoms are less electronegative and could transfer more electrons to H₂ units, allowing structures with atomic hydrogens to remain stable at lower pressures. In addition, IIIB and IVB group metals can also exist in the trivalent state and form trihydrides. Therefore, alloying AlH₃/GaH₃ with IIIB /IVB group metals could give stable alloy hydrides with the A15-type structure and the expected superconductivity at low pressures. For ternary

hydrides, A15-type GaAsH₆ and YZrH₆ were predicted to have T_c values of 98 and 16 K at 180 GPa and ambient pressure, respectively.^{28, 48} Recently, our group designed a series of A15 ternary hydrides, with the CaSnH₆ alloy demonstrating the lowest thermodynamically stable pressure of 110 GPa,⁴⁷ which also facilitated our exploration of ternary alloy hydrides. To some extent, alloying AlH₃/GaH₃ with IIIB /IVB group metals also provides a way to control the physical properties of ternary hydrides through the choice of their constituent elements.

In this work, we investigate theoretically the structure, stability and superconductivity of alloy hydrides with the AMH₆ composition (A=Al and Ga; M=IIIB and IVB group metals). **We chose the AMH₆ composition because the substitution of an M atom for an A atom in the A₂H₆ unit cell is one of the most intuitive ways to form alloy hydrides. Furthermore, considering the promoting effect of configurational entropy in material formation, mixing metal elements in equal proportions may yield a highly disordered alloy hydride, making it more favorable for experimental synthesis.** The results show that seven ternary hydrides, AlMH₆ (M=Ti, Zr and Hf) and GaMH₆ (M=Sc, Ti, Zr and Hf) are identified to be stable in the A15 structure. A15 AlZrH₆, AlHfH₆ and all GaMH₆ are thermodynamically stable at pressures much lower pressures than those required to stabilize A15 AlH₃ and GaH₃. Except for GaScH₆, all of these hydrides also have significant advantages over AlH₃ and GaH₃ in terms of the lowest dynamically stable pressures, with AlZrH₆, AlHfH₆, GaZrH₆ and GaHfH₆ able to maintain their dynamical stability at ~13, 6, 5 and 6 GPa, respectively. Electron-phonon coupling (EPC) calculations show that all of these ternary hydrides are superconducting. Among them, the estimated T_c values for AlZrH₆, AlHfH₆, GaScH₆, GaZrH₆ and GaHfH₆ are 75, 76, 88, 70 and 49 K at 20, 10, 100, 10 and 10 GPa, respectively. In addition, AlScH₆ and AlYH₆ are predicted to be stable in structures similar to $Fm\bar{3}m$ Sc(Y)H₃ and they are also potential superconductors under pressure. Our results indicate that they have great potential to explore low-pressure stable high- T_c superconductors in alloy hydrides.

II. COMPUTATIONAL DETAILS

Structure searches of AMH_6 with simulation cells containing up to 4 formula units were performed at 50-300 GPa by using the particle swarm optimization technique implemented in the CALYPSO code.^{49,50} Structural relaxations and calculations of electronic properties were performed using the VASP code based on the density functional theory with the Perdew-Burke-Ernzerhof generalized gradient approximation.^{51,52} The ion-electron interaction was described by projector-augmented-wave potentials, where $1s^1$, $3s^23p^1$, $3d^{10}4s^24p^1$, $3s^23p^63d^14s^2$, $4s^24p^64d^15s^2$, $5s^25p^65d^16s^2$, $3s^23p^63d^24s^2$, $4s^24p^64d^25s^2$, $5p^65d^26s^2$ configurations are treated as valence electrons for H, Al, Ga, Sc, Y, La, Ti, Zr and Hf atoms, respectively.⁵³ The plane wave kinetic energy cutoff was set to 700 eV and corresponding Monkhorst-Pack (MP) k -point meshes were adopted for different structures to ensure that enthalpy converges to 1 meV/atom. Phonon calculations were performed by using the PHONOPY⁵⁴ or Quantum-ESPRESSO codes.⁵⁵ EPC calculations of $Pm\bar{3}$ ternary hydrides were carried out with the Quantum-ESPRESSO code using ultrasoft pseudopotentials for all the atoms. A $9\times9\times9$ q -point mesh in the first Brillouin zone (BZ) was used in the EPC calculation and a MP grid of $36\times36\times36$ was considered to ensure k -point sampling convergence.

III. RESULTS AND DISCUSSION

We performed structure prediction searches for twelve ternary hydrides with the AMH_6 composition in the pressure range of 50-300 GPa. Seven hydrides (AlTiH_6 , AlZrH_6 , AlHfH_6 , GaScH_6 , GaTiH_6 , GaZrH_6 and GaHfH_6) were identified as having an A15-type structure with $Pm\bar{3}$ symmetry, as shown in Fig. 1(a). In this structure, two metal atoms occupy the vertex and the center positions to form a bcc lattice, with H atoms occupying near their tetrahedral interstices. The A-H, M-H and H-H bond distances with $Pm\bar{3}$ AMH_6 at 100 GPa are shown in Table S1. The A-H distance is shorter than the M-H distance in AlZrH_6 and AlHfH_6 , while the reverse happens in AlTiH_6 and GaMH_6 . Different metal atoms in the structure lead to two different lengths

between adjacent H atoms. The calculated H-H distances of 1.45-1.75 Å are much longer than those of 0.74 and 1.2 Å in H₂ and LaH₁₀ at 100 GPa, respectively, indicating that H atoms are not bonded to each other. Except for GaTiH₆ and GaScH₆, the other five hydrides have no phase transition in the entire pressures range from 50 to 300 GPa. For GaTiH₆ and GaScH₆, *C2/m* and *Pmma* structures were predicted to be stable at 50 GPa, respectively, and they do not present H₂ molecules in their structures (Fig. S1). Furthermore, AlScH₆ and AlYH₆ were predicted to have structures similar to *Fm* $\bar{3}$ *m* Sc(Y)H₃, where metal atoms form a face-centered cubic (fcc) lattice and H atoms are located at the tetrahedral and octahedral interstices of the fcc lattice. As shown in Fig. S2, *Pmmn* AlScH₆ can be seen as a 2×2×1 supercell of *Fm* $\bar{3}$ *m* Sc(Y)H₃ with two metal atoms arranged alternately in two directions. *P4/mmm* AlScH₆ is formed by replacing the two Sc atoms in *Fm* $\bar{3}$ *m* ScH₃ with Al atoms. *I4/mmm* AlYH₆ is similar to the *P4/mmm* AlScH₆, but with slightly shifted H positions. The predicted structures of AlLaH₆, GaYH₆ and GaLaH₆ are also shown in Figure S3.

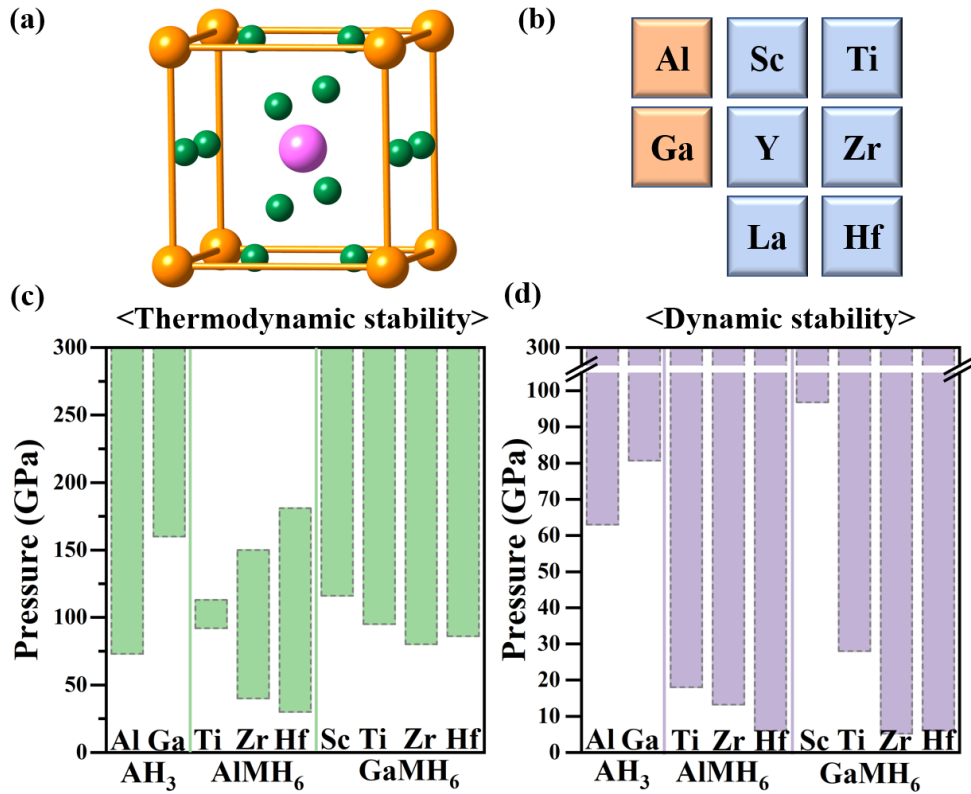


FIG. 1. (a) The crystal structure of the A15-type ternary hydrides. Orange, magenta and green balls represent A (Al and Ga), M (III B and IV B group metal) and H atom, respectively. (b) The considered

elements for the ternary hydrides. (c) Thermodynamic and (d) dynamic stability phase diagram of the $Pm\bar{3}$ hydrides with pressure.

Bader charge analyses⁵⁶ were performed on A15-type ternary hydrides at 100 GPa, as shown in Table S2. The results demonstrate the transfer of electronic charges from metal to H atoms, suggesting an ionic bonding nature between them. Each H atom in $AlMH_6$ and $GaMH_6$ accepts approximately 0.60-0.64 and 0.39-0.43 e, respectively. In $AlMH_6$, Al atoms transfer around 2.31-2.71 e to H atoms, surpassing the ~ 1.31 -1.54 e transferred from the M atoms. In $GaMH_6$, the M atom loses a greater charge (~ 1.34 -1.61 e) compared to the Ga atom (~ 0.94 -1.00 e), indicating a stronger ionic bonding in the M-H bond. The acquired electrons by the H_2 molecule occupy its antibonding orbitals, leading to an elongation of the H-H bond length and potentially even dissociation of the H_2 unit. In $AlMH_6$, the significant electron transfer from Al to H atoms compensates for the relatively smaller electrons transfer from the M atom. Contrasted with GaH_3 , the doping M atoms enable H atoms to gain more charge in $GaMH_6$ at the same pressure, thereby allowing these ternary hydrides to exhibit structures with atomic H at lower pressures. Additionally, the electron localization functions (ELFs) were calculated for these A15 ternary hydrides at 100 GPa with an isosurface value of 0.5, as depicted in Fig. S4. The ELFs indicate that electrons are predominantly localized around the H atoms, confirming their role as electron acceptors. The ELF values below 0.5 between adjacent H atoms indicate the absence of H-H covalent bonds. The ELF values between the metal and H atoms approach zero, validating their ionic bonding character, which aligns with the aforementioned Bader charge analysis.

The thermodynamical stability of these ternary hydrides has been assessed by calculating their formation enthalpies relative to elemental solids and binary compounds.^{36,38-40,42-45} The relative enthalpy curves for AMH_6 are presented in Fig. 2 and S5. The calculations consider the most stable configuration for each component and the total energy of AH_3 and MH_3 is used as the reference energy. Within certain pressure ranges, AMH_6 exhibits lower formation enthalpy compared to possible

decomposition pathways, indicating that AMH_6 is thermodynamically stable. For AlMH_6 (M=Ti, Zr and Hf), the predicted A15-type phases remain stable within the pressure ranges of 78-165, 69-123 and 43-157 GPa, respectively. Upon accounting for zero-point energy (ZPE), the stable pressure range for AlTiH_6 shrinks to 92-113 GPa and the relative formation enthalpy reduces to only -10 meV/f.u. Conversely, with ZPE corrections, the stable pressure ranges for AlZrH_6 and AlHfH_6 expand to 40-150 and 30-181 GPa, respectively. Furthermore, the stability pressure thresholds for both hydrides are lower than the 73 GPa threshold for AlH_3 ³⁶ (Fig. 1c). Regarding GaMH_6 (M=Sc, Ti, Zr and Hf), the predicted A15-type phases are stable above 126, 96, 85 and 95 GPa, respectively, and remain stable with increasing pressure. After considering ZPE, the minimal stable pressures change to 116, 95, 80 and 86 GPa, respectively, which are well below the stability threshold of 160 GPa for GaH_3 (Fig. 1c). Therefore, it is anticipated that the experimentally synthesis of A15-type GaMH_6 might be easier compared to GaH_3 . Inspired by the successful synthesis of the equal-atomic (La,Y) H_4 and (La,Ce) H_9 alloy, the high temperature and high pressure reaction of AM alloys with NH_3BH_3 might be a promising route for the synthesis of alloy AMH_6 .¹⁸⁻¹⁹ Fig. S5 demonstrates that the $Fm\bar{3}m$ -like AlScH_6 is stable above 150 GPa and undergoes a transformation from the $Pmmn$ phase to the $P4/mmm$ phase at 318 GPa. The $C2/m$ phase of AlYH_6 is stable above 45 GPa and transforms to the $Fm\bar{3}m$ -like $I4/mmm$ phase at 115 GPa. AlLaH_6 is predicted to exhibit the $P6_3/mmc$ and $Cmcm$ structures below approximately 200GPa. For GaYH_6 and GaLaH_6 , the low-symmetry $P2_12_12$ and $C2/m$ structures are stable near 100 GPa and below 150 GPa, respectively. Additionally, we also performed structure predictions and First-principles calculations for hydrides with higher H content in the Al-Zr-H, Ga-Sc-H, Ga-Zr-H, and Ga-Hf-H systems at 200 and 300 GPa. As shown in the Fig. S6, these H-rich hydrides have higher formation enthalpies relative to $\text{AH}_3+\text{BH}_3+\text{H}_2$ or AMH_6+H_2 , indicating that they are unstable at the corresponding pressures.

To better understand the reason why the A15-type AMH_6 structure is stable, we conducted an analysis of the influence of the relative internal energy (ΔU) and the

product of pressure and volume (ΔPV) to the relative enthalpy (ΔH) under pressure. The findings for AlZrH_6 and AlHfH_6 are illustrated in Fig. S7. As pressure increases, the ΔPV term also rises and eventually exhibits positive values. Conversely, the ΔU term displays an opposite trend, indicating that the bonding in both ternary hydrides significantly contributes to their stabilization. For the A15-type GaMH_6 (Fig. S8), the ΔPV contribution in all four hydrides is negative in comparison the benchmark, indicating that the ΔPV term plays a dominant role in their thermodynamic stability. Meanwhile, the magnitude of ΔU decreases with increasing pressure, leading to an overall enhancement in the stability of these ternary hydrides. Furthermore, in GaScH_6 the ΔPV and ΔU terms exhibit contrasting trends when compared to $\text{ScH}_3+\text{GaH}_3$. The negative ΔPV contribution counteracts the negative effect of the ΔU term, ultimately resulting in the stabilization of GaScH_6 . Both the ΔPV and ΔU contribution in GaTiH_6 , GaZrH_6 and GaHfH_6 have favorable effects on the thermodynamic stability relative to their respective binary hydrides.

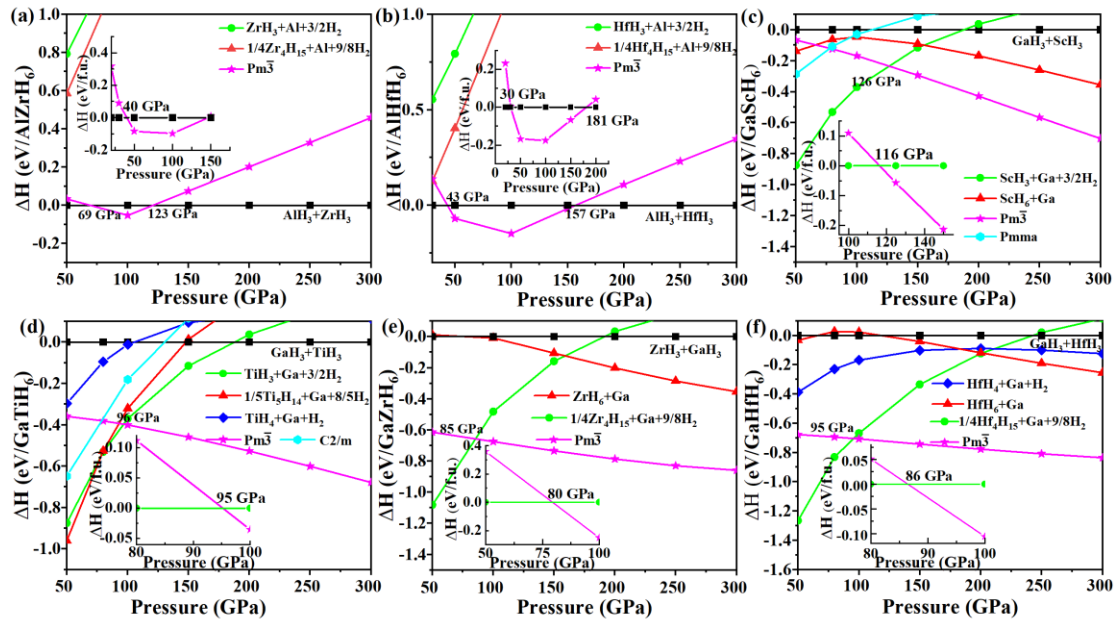


FIG. 2. The relative enthalpy curves of predicted structures for (a) AlZrH_6 , (b) AlHfH_6 , (c) GaScH_6 , (d) GaTiH_6 , (e) GaZrH_6 and (f) GaHfH_6 with respect to AH_3 ($A=\text{Al}$ and Ga) and MH_3 ($M=\text{Sc}$, Ti , Zr)

and Hf) under pressure. The insets show the relative enthalpies considering ZPE. The following structures for the elemental solids and binary hydrides were used for the ΔH calculations: $P6_3/m$, $C2/c$ and $Cmca$ for H_2 ; $Fm\bar{3}m$ and $P6_3/mmc$ for Al; $Fm\bar{3}m$ for Ga; $R\bar{3}c$, $Pnma$ and $Pm\bar{3}n$ for AlH_3 ; $P2_1/m$ and $Pm\bar{3}n$ for GaH_3 ; $Pm\bar{3}n$ and $R\bar{3}c$ for ZrH_3 ; $I\bar{4}3d$ for Zr_4H_{15} , $Cmc2_1$ and $I4/mmm$ for ZrH_6 ; $Pm\bar{3}n$ for HfH_3 ; $I\bar{4}3d$ for Hf_4H_{15} ; $Cmc2_1$ for HfH_6 ; $Fm\bar{3}m$ for ScH_3 ; $Cmcm$, $P6_3/mmc$ and $Im\bar{3}m$ for ScH_6 ; $Fm\bar{3}m$ for TiH_3 ; $I\bar{4}$ for Ti_5H_{14} ; $Fddd$ for TiH_4 .

We also investigated the dynamical stability of the predicted A15-type AMH_6 compounds by calculating the phonon spectra using the supercell approach implemented in the PHONOPY code⁵⁴ (Figs. S14 and S15). Within their respective thermodynamically stable pressure ranges, no imaginary frequencies were observed in the phonon spectra of these compounds, indicating their dynamical stability. Furthermore, we systematically explored the minimum pressure required for dynamical stability of these A15-type AMH_6 . As pressure decreases, phonon softening begins to occur, eventually leading to instability with the appearance of imaginary frequencies at certain q -wave vectors. We plotted the evolution of the frequency with pressure at the q -wave vector where the largest imaginary frequency occurs. As depicted in Figs. S9 and S10, A15-type $GaScH_6$ exhibited the highest critical pressure for dynamical stability, estimated to be around 97 GPa. $GaTiH_6$ followed with a critical pressure of approximately 28 GPa. $AlTiH_6$ and $AlZrH_6$ were found to be dynamically stable at lower pressures of about 18 and 13 GPa, respectively. Interestingly, the critical pressures of dynamical stability of $AlHfH_6$, $GaZrH_6$ and $GaHfH_6$ were considerably lower, around 6, 5 and 6 GPa, respectively. Moreover, the threshold pressures for dynamic stability of the five ternary hydrides ($GaTiH_6$, $AlTiH_6$, $AlHfH_6$, $GaZrH_6$ and $GaHfH_6$) were much lower compared to those of AH_3 and MH_3 (Figs. S11, S13 and 1d). This suggests that these ternary hydrides could potentially be recovered at lower pressures, provided that the barriers preventing them from decomposing are sufficiently high. For $Fm\bar{3}m$ -like ternary hydrides, $Pmmn$ $AlScH_6$, $P4/mmm$ $AlScH_6$ and $I4/mmm$ $AlYH_6$ could maintain dynamically stable to 65, 139 and 43 GPa, respectively (Figs. S12 and S16).

The electronic properties of the predicted A15-type AMH₆ compounds were investigated at different pressures, and the results are displayed in Fig. 3, as well as Figs. S17 and S18. The calculated density of states (DOS) reveals the presence of electronic states at the Fermi energy level (E_f), indicating that these hydrides exhibit metallic behavior within the studied pressure range. The contributions of the metal and hydrogen atoms to the total DOS are also depicted in the figures. Fig. 3d highlights a distinct difference between the DOS of GaScH₆ and the other six hydrides composed of Al/Ga and group IVB metals. In GaScH₆, the hydrogen atoms significantly contribute to the DOS at the E_f , suggesting the potential for excellent superconducting properties. In the remaining six hydrides, the group IVB metals have one additional valence electron compared to Sc atoms, causing a shift of the E_f to higher energies relative to GaScH₆. These six hydrides exhibit higher or comparable total DOS at the E_f compared to GaScH₆. However, the DOS contributions from Al and Ga atoms are relatively lower due to significant charge transfer to the H atoms. In the case of the group IVB metals, the DOS at E_f is mainly contributed by d orbitals. Furthermore, the electronic band structures of AlHfH₆, GaScH₆ and GaZrH₆ were examined as examples at pressures of 10, 100 and 10 GPa, respectively. The band projections onto different elements are also displayed in the band structures. In these three hydrides, a band associated with Al/Ga atoms is observed to cross the E_f steeply along the M - R - Γ direction. Additionally, in AlHfH₆ and GaZrH₆ there is an electron pocket at the M point and a flat band along the Γ - M direction, dominated by Hf and Zr atoms, located near the E_f . In the case of GaScH₆, electron and hole pockets are observed along the X - M direction, along with a flat band along the Γ - M direction, which are attributed to H atoms. These localized electronic states contribute to a high DOS at the Fermi energy and play a role in electron-phonon interactions. Moreover, a comparison was made between the DOS of A15-type AlH₃, AlTiH₆, AlZrH₆ and AlHfH₆ at 100 GPa. As shown in Fig. S17, the DOS at E_f for AlTiH₆, AlZrH₆ and AlHfH₆ are 0.030, 0.018 and 0.018 states/eV/Å³ respectively, which are significantly higher than that of AlH₃ (0.008 states/eV/Å³). The doping of AlH₃ with Ti, Zr and Hf atoms elevated the E_f , resulting in an increased DOS at the E_f .

Ternary hydrides exhibit a more pronounced metallic character compared to AlH_3 , potentially enhancing their superconducting properties.

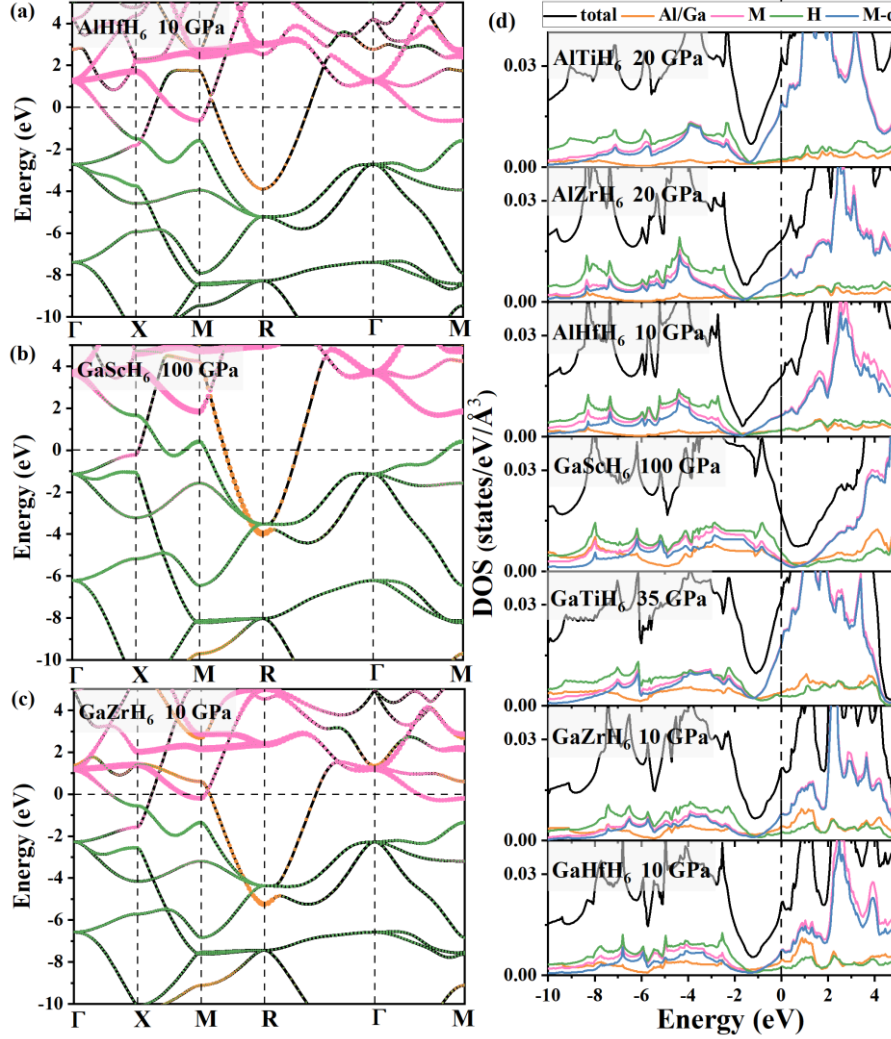


FIG. 3. The calculated band structures and electronic density of states of $Pm\bar{3}$ ternary hydrides at different pressures.

After determining the stability and metallicity of A15-type ternary hydrides, we conducted EPC calculations to investigate their superconductivity. The calculated phonon dispersion curves, projected phonon density of states (PDOS), Eliashberg phonon spectral function $\alpha^2F(\omega)/\omega$, and its integral $\lambda(\omega)$ for $Pm\bar{3}$ AlHfH_6 , GaScH_6 and GaZrH_6 at pressures of 10, 100 and 10 GPa are displayed in Fig. 4. From the PDOS, it is evident that the high-frequency and low-frequency phonon modes are associated with the vibration of H and metal atoms, respectively. The right panels of Fig. 4 illustrate that the peaks of $\alpha^2F(\omega)/\omega$ for AlHfH_6 , GaScH_6 and GaZrH_6 are predominantly

distributed below 15, 20 and 15 THz, respectively, with the corresponding $\lambda(\omega)$ increasing rapidly. The EPC strength on the different phonon modes is also depicted on the phonon dispersions. As a result, the significant contribution to the EPC comes from the soft modes associated with H-atom vibrations and the modes dominated by metal atoms. This leads to a total EPC parameter λ of 2.18, 1.56 and 1.80 for AlHfH₆, GaScH₆ and GaZrH₆ at pressures of 10, 100 and 10 GPa, respectively.

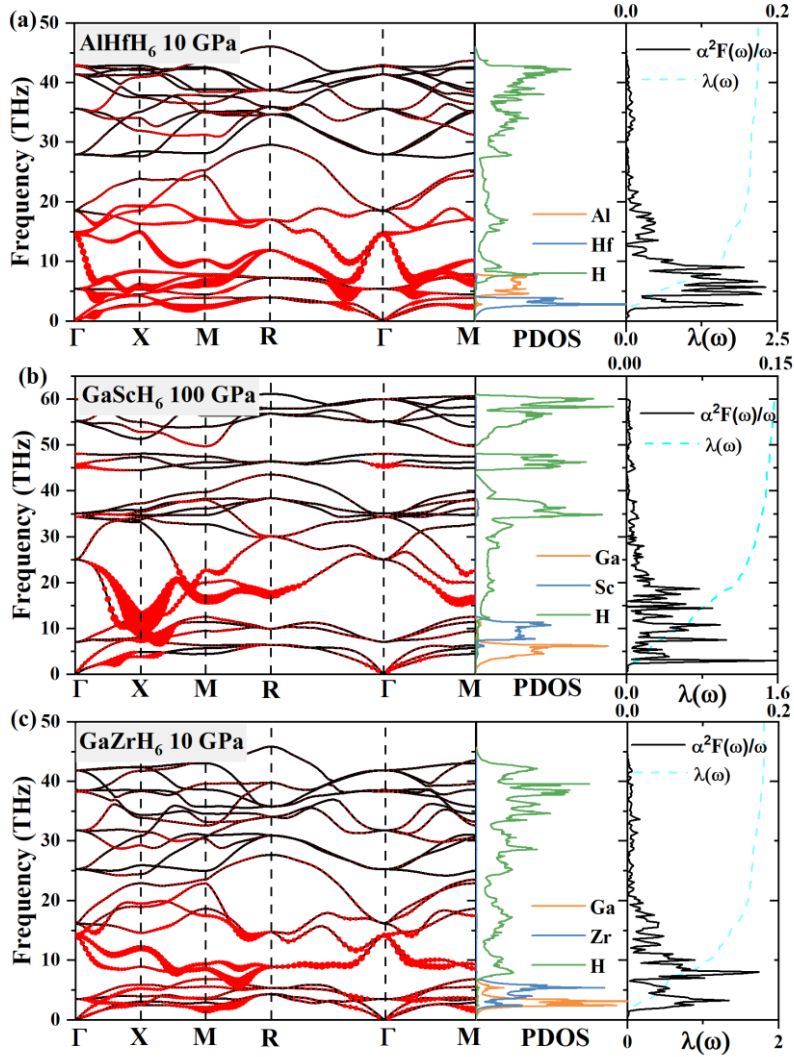


FIG. 4. Calculated phonon dispersion curves (the area of the red circles is proportional to the EPC strength), projected phonon density of states (PDOS), the Eliashberg phonon spectral function $\alpha^2F(\omega)/\omega$ and its integral $\lambda(\omega)$ for (a) $Pm\bar{3}$ AlHfH₆ at 10 GPa, (b) $Pm\bar{3}$ GaScH₆ at 100 GPa and (c) $Pm\bar{3}$ GaZrH₆ at 10 GPa.

The superconducting critical temperature for the predicted A15-type ternary

hydrides were calculated using the Allen-Dynes modified McMillan equation with a Coulomb pseudopotential parameter (μ^*) ranging from 0.1 to 0.13.⁵⁷ The calculated T_c values, along with the EPC parameter λ and the phonon frequency logarithmic average $\omega_{\log}$ are presented in Fig. 5 and Table 1. For AlTiH₆, the calculated EPC parameter λ is 1.15 and the phonon frequency logarithmic average $\omega_{\log}$ is 423 K. This results in a T_c of 32-36 K at 20 GPa. AlZrH₆ and AlHfH₆ present stronger EPC interactions, with λ of 1.72 and 2.18, and the T_c values are estimated to be 59-64 and 57-60 K at 20 and 10 GPa, respectively. Considering that the λ values are higher than 1.5, the calculated T_c values are further rectified with the strong coupling (f_1) and shape correction (f_2) and improved to be 68-75 and 70-76 K, respectively. The T_c values obtained for AlMH₆ are indeed higher than those of AlH₃ and the corresponding MH₃ compounds.^{37,39,40,45} Similarly, GaScH₆ exhibits the highest T_c among the studied hydrides, reaching a range of 79-88 K at a higher pressure of 100 GPa, However, its T_c is slightly lower than that of GaH₃, which has a T_c of 102 K of at 120 GPa. In case of GaTiH₆, it shows a lower T_c range of 34-39 K at 35 GPa compared to the T_c of 63-70 K for GaZrH₆ and 45-49 K for GaHfH₆ at 10 GPa.

The evolution of T_c with pressure was also investigated for $Pm\bar{3}$ AMH₆. For AlZrH₆, the calculated EPC parameter λ decreased from 1.72 at 20 GPa to 1.18 at 30 GPa, while the $\omega_{\log}$ increased from 494 to 667 K, resulting in a slight decrease in the T_c with μ^* of 0.1 from 64 to 59 K (Allen-Dynes modified McMillan equation). Similarly, as the pressure increased from 10 to 20 GPa, the λ for AlHfH₆ decreased from 2.18 to 1.28 and the $\omega_{\log}$ increased from 397 to 596 K. As a result, the calculated T_c with μ^* of 0.1 decreased slightly from 60 to 58 K. The increase in pressure leads to a stiffening of the phonon modes, which results in a decrease in EPC parameter and an increase in $\omega_{\log}$. This compensating effect prevents a significant reduction in T_c . In addition, the T_c values of GaMH₆ compounds decrease with increasing pressure, due to the competition between the descending EPC parameter (λ) and the increasing phonon frequency logarithmic average ($\omega_{\log}$). For $Fm\bar{3}m$ -like ternary hydrides, $Pm\bar{3}n$ AlScH₆, $P4/mmm$ AlScH₆ and $I4/mmm$ AlYH₆ found to be metallic (Fig. S19) and subsequent EPC

calculations suggest that they exhibit superconductivity. The T_c values for these hydrides are estimated to be 42, 32 and 52 K at pressures of 80, 200 and 60 GPa, respectively (Table S3).

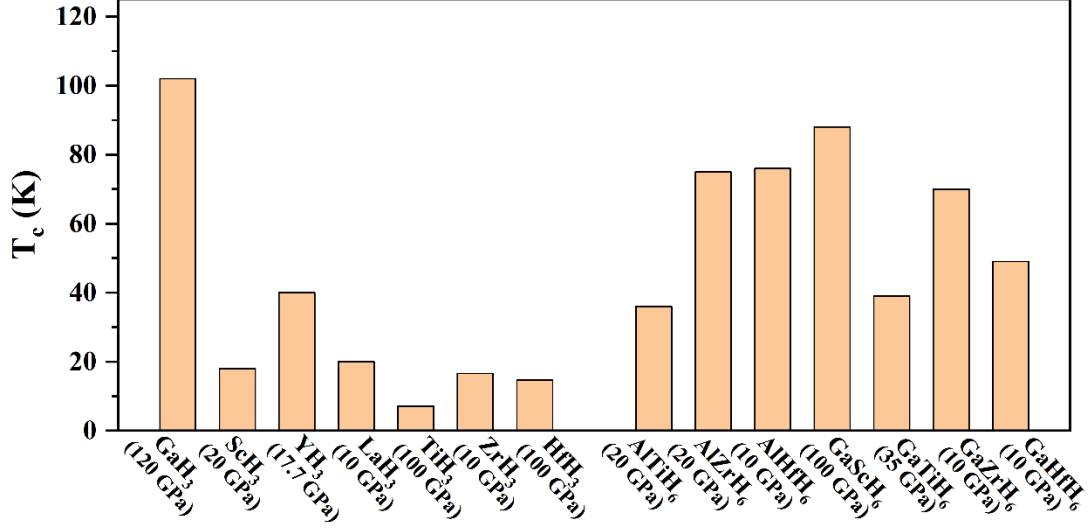


FIG. 5. The estimated T_c for the predicted A15 AMH₆ using the Allen-Dynes modified McMillan equation and for already known A(M)H₃ compounds extracted from literature.^{38-40,43-45}

Table 1. The calculated values for electron-phonon coupling parameter λ , phonon frequency logarithmic average $\omega_{\log}$, critical temperature T_c ($\mu^*=0.1-0.13$) using the Allen-Dynes modified McMillan equation without and with strong-coupling and shape corrections for $Pm\bar{3}$ ternary hydrides.

Phase	Pressure (GPa)	λ	$\omega_{\log}$	T_c (K)	T_c (K) with f_l and f_2
				$\mu^*=0.1-0.13$	$\mu^*=0.1-0.13$
AlTiH ₆	20	1.15	423	32-36	
AlZrH ₆	20	1.72	494	59-64	68-75
	30	1.18	667	52-59	
AlHfH ₆	10	2.18	397	57-60	70-76
	20	1.28	596	52-58	
GaScH ₆	100	1.56	641	69-76	79-88

	120	1.09	890	71-62	
GaTiH ₆	35	1.17	448	34-39	
	100	0.68	887	29-22	
GaZrH ₆	10	1.80	438	54-58	63-70
	80	0.81	898	43-35	
GaHfH ₆	10	1.42	454	45-49	
	86	0.74	802	32-25	

IV. CONCLUSIONS

In summary, we have carried out crystal structural prediction and first-principles calculations on alloy hydrides with the AMH₆ composition under pressure. Seven ternary hydrides, AlMH₆ (M=Ti, Zr and Hf) and GaMH₆ (M=Sc, Ti, Zr and Hf) were predicted to adopt the A15-type structure, and the calculated enthalpy curves indicated that the A15-type AlMH₆ are stable within specific pressure ranges of 92-113, 40-150 and 30-181, respectively. The onset stable pressures for AlZrH₆ and AlHfH₆ are lower than the stability threshold of 73 GPa for AlH₃. Phonon calculations demonstrated that A15-type AlTiH₆, AlZrH₆, AlHfH₆ can be dynamically stable at relatively low pressures, such as, 18, 13 and 6 GPa, respectively. Additionally, the addition of Ti, Zr and Hf into AlH₃ influenced the position of the Fermi level, resulting in improved metallicity compared to AlH₃. Consequently, the ternary hydrides exhibited higher T_{cs} of 36, 75 and 76 K at 20, 20 and 10 GPa, respectively. The A15-type GaMH₆ hydrides (GaScH₆, GaTiH₆, GaZrH₆, and GaHfH₆) were found to have minimum thresholds of thermodynamically stable pressure at 116, 95, 80 and 86 GPa respectively, which are significantly lower than the stability threshold of 160 GPa for GaH₃. Additionally, dynamical stability calculations indicated their potential retention down to lower pressures of 97, 28, 5 and 6 GPa, respectively. EPC calculations revealed that A15-type GaMH₆ hydrides are superconducting, with T_{cs} of 88, 39, 70 and 49 K at 100, 35, 10 and 10 GPa, respectively. AlScH₆ and AlYH₆ were predicted to be stable in $Fm\bar{3}m$ Sc(Y)H₃-like structures under pressure. The estimated T_c values for $Pmmn$, $P4/mmm$

AlScH₆ and *I4/mmm* AlYH₆ were 42, 32 and 52 K at 80, 200 and 60 GPa, respectively. These findings suggest that alloying holds promise as a route to lower the stabilization pressure of hydrides, enabling the exploration of high-*T_c* hydride superconductors that can be stable at lower or ambient pressures.

SUPPLEMENTARY MATERIAL

See supplementary material for the predicted structures of GaTiH₆, GaScH₆, AlScH₆, AlYH₆, AlLaH₆, GaYH₆ and GaLaH₆; ELF's; the relative enthalpy curves of AlTiH₆, AlScH₆, AlYH₆, AlLaH₆, GaYH₆ and GaLaH₆; The formation enthalpies of hydrides with higher H content in the Al-Zr-H, Ga-Sc-H, Ga-Zr-H and Ga-Hf-H system with respect to decomposition into AH₃+BH₃+H₂ or AMH₆+H₂ at 200 and 300 GPa; the relative internal energies ΔU and ΔPV components of the enthalpy for the A15 ternary hydrides; the evolution of the frequency with pressure for the studied hydrides; phonon spectra, electronic density of states, structural information; Bader charge analyses; superconductivity of the *Pmmn*, *P4/mmm* AlScH₆ and *I4/mmm* AlYH₆.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

DATA AVAILABILITY

The data supporting the findings of this study are publicly available from the corresponding author upon request.

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