

Toward ambient-pressure superconductivity from boron icosahedral superatoms

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We identify a family of boron-rich compounds consisting of interconnected B_{12} icosahedra and electropositive guest atoms (X) in interstitial sites. These structures were found through first-principles crystal structure prediction at 50 GPa, and are dynamically stable down to ambient pressure. When X is a mono- or trivalent element, the structures are metallic and superconducting. Predicted critical temperatures reach up to 42 K for CsB_{12} , rivaling MgB_2 , the highest- T_c ambient-pressure conventional superconductor. We interpret the XB_{12} phase as a superatomic crystal: the B_{12} units retain the icosahedral shape that they also exhibit in isolation, while forming an extended crystalline network. When X is a mono- or trivalent atom, the system is metallic, and the B–B covalent bonding promotes strong electron-phonon coupling. Unlike MgB_2 , where superconductivity is driven by a narrow subset of phonon modes, the XB_{12} compounds exhibit broad, mode- and momentum-distributed coupling through both intra- and intersuperatomic vibrations. Our results highlight the XB_{12} family as a promising platform for superconductivity and demonstrate the potential of superatoms as functional building blocks in solid-state materials design.

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I. INTRODUCTION

An emerging route to discover alternative ambient-pressure superconductors relies on stabilizing exotic phases through synthesis at moderate pressures, followed by recovery at ambient conditions in a metastable state. Structures rich in boron and/or carbon are particularly attractive for this purpose and have attracted interest in recent years [1–7]. In fact, both elements are characterized by light atomic masses, tend to form covalent bonds, and are known to form allotropes under pressure that are recoverable at ambient conditions (e.g., diamond). Concerning superconductivity, light masses lead to high phonon frequencies and covalent bonds to large electron-phonon matrix elements. These two are understood as they key driving factors of the 39 K critical temperature (T_c) of MgB_2 [8] and the record-high T_c of superhydrides [9,10]. In addition to light masses and covalent bonds, materials must also be metallic to host superconductivity. Most covalent materials, however, are insulating and require doping to achieve metallicity. While some exceptions exist, compounds become unstable even under modest doping levels [11–17]. The problem is that electron-phonon (ep) coupling, while being desirable for superconductivity, is also the main cause of structural instability if too large. This effect is particularly

strong if all the coupling is concentrated on a single phonon mode. The critical question is if we can identify structural motifs that tolerate doping, maintain strong covalent bonds, and distribute coupling. In this article, we present a family of boron-rich structures which fulfill these criteria.

These were found through first-principles crystal structure prediction at moderate pressures (50 GPa), with XB_{12} stoichiometry (X is an atom of groups I–III). For all elements but Sc, this phase is also dynamically stable both at 50 GPa and at ambient pressure. We interpret the XB_{12} crystal structure as consisting of boron icosahedral superatoms [18,19]—groups of atoms that coalesce in a single (meta)stable form, with properties that differ from those of the constituents. Once formed, superatoms can form stable crystals with other atoms, named superatomic crystals. A notable example of such a system are carbon fullerenes (C_{60}), which form a stable fcc lattice with metal atoms and become superconducting at ambient pressure [20].

We characterized the thermodynamic stability, electronic, and superconducting properties of XB_{12} superatomic structures by means of state-of-the-art first-principles calculations. At ambient pressure, XB_{12} phases with group I and III elements exhibit superconductivity with predicted T_c values of up to 42 K in CsB_{12} . Doping boron-icosahedral networks has already been presented as a possible route to strong covalent bonding and large electron-phonon coupling [21]. The key difference in the XB_{12} system is that here doping occurs at the interstitial sites, through the X atom, rather than on the covalent bonds themselves, which has a smaller impact on the structure and preserves the B–B bond network. At the same time, the electron-phonon coupling comes from covalent intra- and intersuperatom bonds whose states sit at the Fermi

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energy. In addition, the ep coupling distributes uniformly over modes and momenta, which makes the structure able to withstand even large coupling without becoming unstable.

This article is structured as follows. In Sec. II, we assess the thermodynamic stability of the XB_{12} family at 50 GPa, discussing their synthesizability and prospects for recovery at ambient conditions. In Sec. III, we present the concept of superatomic crystal and discuss how XB_{12} fulfills the criteria. In Sec. IV, we analyze the electronic structure XB_{12} structures in comparison with the α -boron structure, which also consists of icosahedral superatoms. This comparison confirms that B_{12} behaves consistently in different environments, as if it were a single atom, with corelike orbitals that do not interact with surrounding atoms, and valencelike ones that do. In Sec. V, we present superconductivity trends across the full XB_{12} family and discuss the role of electron-phonon coupling and phonon softening in determining their T_c . In Sec. VI, we validate these results for two limiting cases, CsB_{12} and LaB_{12} , using fully anisotropic SCDFT calculations, and analyze the Eliashberg function and the screened Coulomb interaction. Conclusions are drawn in Sec. VII.

II. THERMODYNAMICAL STABILITY

We begin our discussion by looking at the thermodynamics, to show how the predicted XB_{12} could form. The XB_{12} structure was found performing evolutionary crystal structure prediction at 50 GPa for the XB_{12} composition [22,23] ($X = K, Ca, Sc, Rb, Sr, Y, Cs, Ba, La$). This pressure was chosen to access structures with exotic bonding behavior, while remaining close enough to ambient pressure to enable their recovery at ambient conditions. The specific choice of B_{12} composition was motivated by the experimental report of a $Fm\bar{3}m$ phase with ZrB_{12} composition, in which boron forms a continuous network [24,25] that is superconducting at 6 K and ambient pressure [25,26]. Our working hypothesis was that similar structures would appear under pressure for elements other than Zr.

For Sc and Y, we found the $Fm\bar{3}m$ structure [24,25] to be energetically favored by more than 100 meV/atom. For all other X elements investigated, a structure with space group $Pm\bar{3}$ appeared spontaneously in all of our searches. This structure consists of B_{12} icosahedral units, with an enthalpy gain compared to the $Fm\bar{3}m$ phase ranging from 23 meV/atom (Ca) to 264 meV/atom (Ba). For alkaline earths, our results are consistent with a previous study, which also reported this phase for SrB_{12} , but did not study group I and group III elements [27].

To estimate the actual synthesizability of this phase, we computed the full X -B convex hulls at 0 and 50 GPa. An example is shown for SrB_{12} at 50 GPa in Fig. 1(a) (the results for other compositions are reported in Supplemental Material (SM) Figs. S1 and S2 [28]): Structures lying on the convex hull (blue circles) are thermodynamically stable, while those above it (red squares) are unstable or metastable. The SrB_{12} phase with $Pm\bar{3}$ space group lies on the convex hull, i.e., its enthalpy relative to the hull (ΔH_{hull}) is zero and should form spontaneously. In the following, we will refer to the enthalpy relative to the hull as ΔH_{hull} and relative to the pure elements as ΔH_f , respectively. The latter

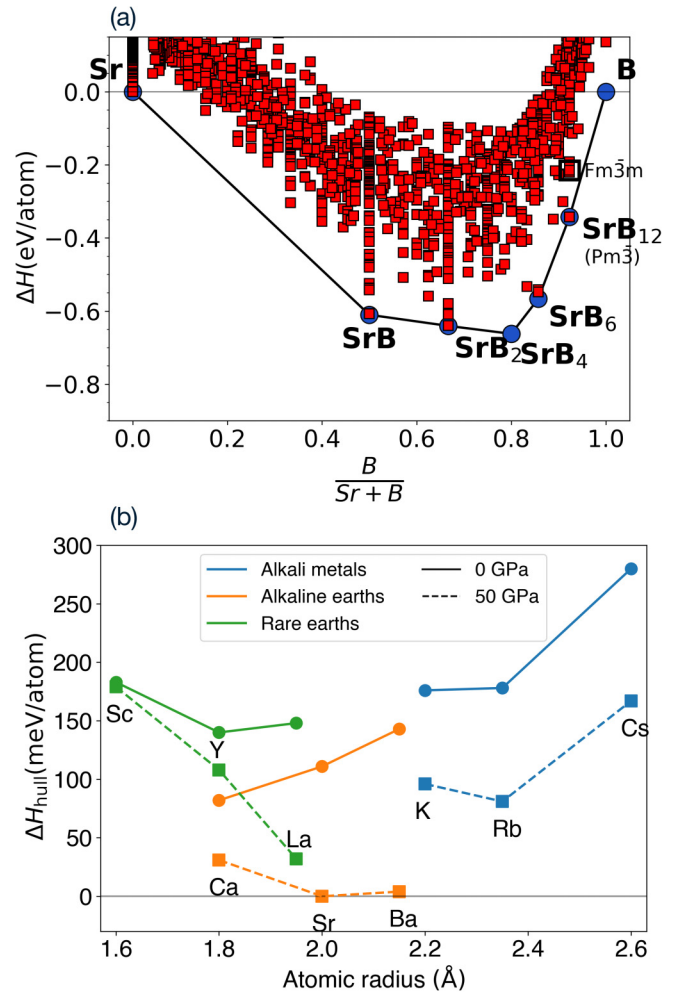


FIG. 1. Thermodynamical stability of X -B boride ($X = K, Ca, Sc, Rb, Sr, Y, Cs, Ba, La$) phases at 50 GPa. (a) Convex hull of Sr -B at 50 GPa. The superatomic ($Pm\bar{3}$) SrB_{12} phase is on the convex hull. The point corresponding to the known XB_{12} phase ($Fm\bar{3}m$) is indicated by a black square. (b) Distance from the convex hull of the superatomic XB_{12} phase at 0 (solid lines) and 50 GPa (dashed lines).

indicates if the structure would form from the pure elements, provided the pathway to other competing phases was inhibited.

Figure 1(b) shows ΔH_{hull} across all the X elements studied at both 0 and 50 GPa (for ΔH_f , we refer the reader to Fig. S3 of the SM [28] (see also Refs. [29,30] therein). At 50 GPa, the $Pm\bar{3}$ phase lies close to the hull for four elements: Sr and Ba are stable, and Ca and La are within 50 meV/atom. At ambient pressure, the $Pm\bar{3}$ - XB_{12} phase becomes less stable for all X , with ΔH_{hull} rising to 100–300 meV/atom (1030 kJ/mol). ΔH_f , on the other hand, at 50 GPa is negative for all elements but Rb (+10 meV/atom) and Cs (+167 meV/atom). At ambient pressure, it becomes larger across the board and becomes positive for La (+140 meV/atom), K (135 meV/atom), Rb (+198 meV/atom), and Cs (+297 meV/atom). The zero-point contribution to the free energy, evaluated for SrB_{12} , exhibits a very minor effect, stabilizing the $Pm\bar{3}$ phase by -3 meV/atom with respect to Sr and α -B (for details, we refer the reader to Table S1 of the SM).

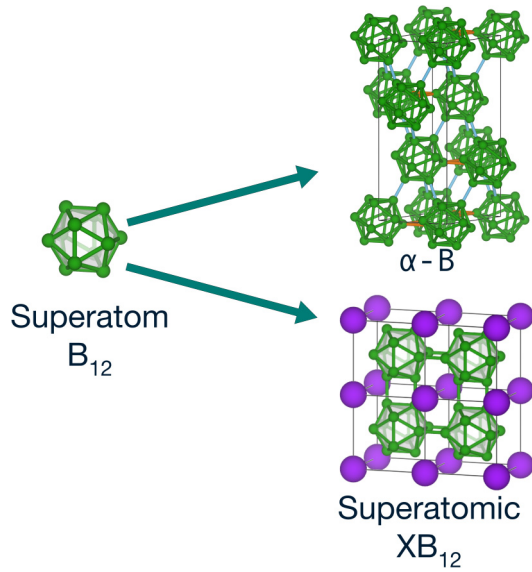


FIG. 2. Formation of the XB_{12} crystal from the B_{12} superatomic building block. We show an isolated B_{12} superatom, the crystal structure of α -B, with the B_{12} superatoms highlighted, and the $Pm\bar{3}$ - XB_{12} crystal structure. In the α -B structure, covalent and $2e3c$ bonds are highlighted in cyan and orange, respectively.

Precisely quantifying the lifetime of XB_{12} in a metastable state would require identifying the decomposition path, which is at the moment beyond the state of the art. Here, we argue based on qualitative arguments the plausibility that the $Pm\bar{3}$ - XB_{12} phase can be synthesized under pressure and recovered at ambient conditions. First, phonon calculations confirm that most compounds (excluding Sc) are dynamically stable at zero pressure (Fig. S12). Second, boron shares with carbon and nitrogen a propensity to form bonds with high cohesive energies. Statistically, this is associated with a window of metastability as wide as 200 meV/atom [31], which is comparable to our highest ΔH_{hull} values. These factors suggest that kinetic barriers, rather than thermodynamic driving forces, are the dominant constraint on decomposition. Third, the B_{12} icosahedron is a recurring motif that is present in pure boron [32], in boron-rich YB_{66} [33], and also as an isolated cluster [34,35]. In Fig. 2, we show the $Pm\bar{3}$ - XB_{12} structure, the B_{12} icosahedron, and the ground-state structure of boron (α -B). From a structural point of view, XB_{12} is like a molecular crystal, with interconnected B_{12} atoms, and X atoms in the interstitials. For as long as the B_{12} icosahedron remains intact, the decomposition is only possible towards a phase with intact icosahedra, i.e., α boron.

To further support the metastability argument, we performed three molecular dynamics simulations of CsB_{12} : at ambient pressure and 100, 600 K, and 1200 K, for a duration of 180 ps, representing, respectively, a temperature close to operation and two possible synthesis ones. In all cases, we find that boron atoms exhibit no diffusion, and the icosahedral shapes are preserved throughout the simulation (we refer the interested reader to Fig. S4 of the SM [28]).

We also compare our results for XB_{12} to those for SrB_3C_3 , since the two materials present several similarities: both consist of light elements (B/C) arranged in a covalent network

with large voids, where an electropositive element sits interstitially. SrB_3C_3 was experimentally synthesized under pressure and recovered at ambient conditions [36–39]. The synthesis was achieved by laser heating a mixture of SrB_6 , SrC_2 , and C at a pressure of 50 GPa, where it is thermodynamically stable. After synthesis, the compound was recovered at ambient conditions simply by releasing the pressure. Since the materials are quite similar, if one was synthesized and recovered, the other one should be too.

At 50 GPa, SrB_3C_3 was reported on the convex hull ($\Delta H_{\text{hull}} = 0$, $\Delta H_f = -0.5$ eV/atom) [40]. However, at ambient pressure it exhibits a positive ΔH_f (+30 meV/atom) [41]. $Pm\bar{3}$ - SrB_{12} is also on the convex hull at 50 GPa, and at ambient pressure exhibits a negative formation enthalpy ($\Delta H_f = -110$ meV/atom). In comparison, SrB_{12} is more stable. The synthesis pressure is about the same and, since the formation energy is negative, should be equally recoverable at ambient conditions.

We also propose a possible synthesis route of XB_{12} which does not require pressure. In fact, α -B already contains pre-formed B_{12} icosahedra, and moderate-temperature diffusion into this framework is known to be feasible: lightly Li-doped α - B_{12} ($Li_{0.3}B_{12}$) has been synthesized by diffusing Li into α -B without disrupting the icosahedral units [42]. This is a different case from the XB_{12} phases discussed here, since the geometry and connectivity of the B_{12} icosahedra differ from those of the α -B allotrope, but it demonstrates that X atoms can be incorporated by diffusion while leaving the B_{12} superatoms intact. Following the same principle, one could mix a finely ground powder of α -B and the selected X element and apply only moderate heat to promote diffusion of X atoms among B_{12} icosahedra. If the icosahedra do not break, the reaction toward phases with a lower boron content is inhibited, and XB_{12} can form as long as ΔH_f is negative, since no other phase we found contains B_{12} icosahedra.

III. XB_{12} AS SUPERATOMIC CRYSTALS

We now discuss the crystal structure of the $Pm\bar{3}$ - XB_{12} phase, show how the B_{12} icosahedron appears as a rigid building block, and discuss how it matches the definition of a superatom. This idea will be key to later understand the materials' electronic and vibrational properties.

A superatom is defined as an assembly of atoms that exhibits one or more atomlike properties [19]. This definition is intentionally broad and encompasses small molecules, radicals, and clusters. Assembling superatoms into periodically ordered superatomic crystals, where superatoms retain their identity while exhibiting collective behavior, is a major challenge in physical chemistry [18,43].

Superatomic crystals are characterized by two distinct length scales: an intrasuperatomic scale (the size of each superatom) and an intersuperatomic one (the distance between superatoms). This translates into two features that determine the electronic structure: discrete energy levels coming from orbitals localized on individual superatoms, and intersuperatomic hopping. The energy levels corresponding to localized states are low-lying and analogous to atomic semicore states. Those that arise from intersuperatomic hopping instead lead to dispersive bands, analogously to the interatom hopping of

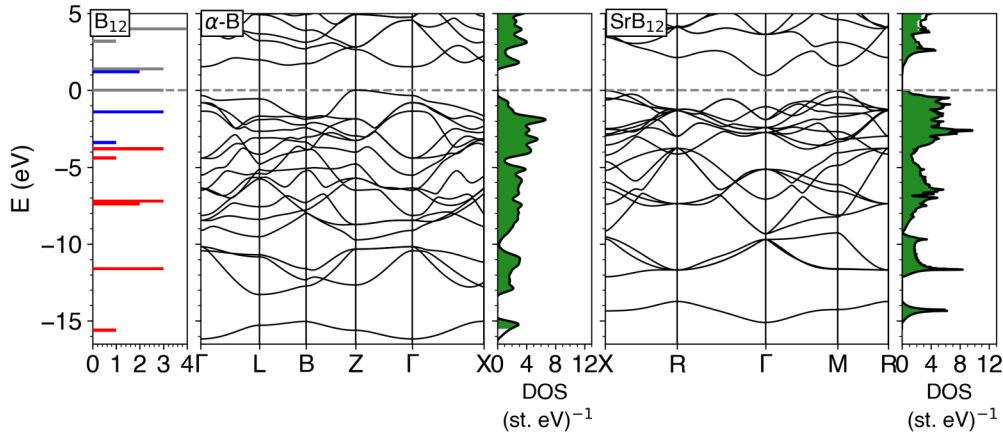


FIG. 3. Calculated electronic structure and total and atom-projected density of states (DOS) for an isolated B_{12} superatom, α -B, and SrB_{12} at ambient pressure. The energy zero is set at the top of the valence band. In the DOS panel for B_{12} , we indicate with red the B_{12} molecular states, and with blue and gray the nonbonding states that in the XB_{12} crystal become occupied and unoccupied, respectively. The total and B-projected DOS are shown as a black solid line and a green filled curve, respectively.

regular crystal. Depending on the intersuperatom distance, the hopping can be as large as for regular atoms, even though it involves molecular orbitals.

In the XB_{12} structure, B_{12} superatoms form 12 intersuperatom (B–B) bonds with their outward-facing orbitals. The inter- and intrasuperatom distances are both around 1.7 Å, hence the intersuperatom electronic hopping is comparable to the intrasuperatom one.

To support the idea that the XB_{12} structure does qualify as a superatomic crystal, we compare it with the ground-state structure of boron (α -B). There, B_{12} icosahedra are connected with their neighbors along the z axis through six B–B bonds, also 1.7 Å long, and to their neighbors in the xy plane via six two-electron, three-center bonds ($2e3c$) [44,45]. So, in the two structures, the B_{12} superatoms maintain well-defined geometries and bond lengths, while the periodic crystal is formed through intersuperatom bonds, which differ in coordination and geometry.

Concerning our specific goal of identifying conventional superconductors, the XB_{12} superatomic crystal possesses two important features. The first is that both inter- and intrasuperatom bonds involve the same atoms (B), and hence must be covalent [8]. The second feature is the ability of XB_{12} to withstand electron or hole doping over a broad range without becoming dynamically unstable. This aspect is discussed in more detail in Sec. V; the phonon dispersions at ambient pressure for all structures considered are shown in Fig. S12 of the SM [28].

We ascribe the stability of the XB_{12} crystal to two geometric factors. First, doping occurs through substitution at the interstitial X site, which in the system acts as a charge donor. This should be easier to realize experimentally, compared to doping the covalent bond (e.g., by C substitution) [13,21,46]. Second, the B_{12} superatoms present *internal* degrees of freedom. Therefore, the B_{12} can respond to doping with slight distortions, while the overall crystal structure remains unchanged, something that is not possible in regular crystals.

IV. ELECTRONIC STRUCTURE

We now describe the electronic structure of XB_{12} in comparison with α -B and the isolated B_{12} superatom. This comparison reveals that the low-lying electronic states of the B_{12} do not change when their arrangement changes, and supports the idea that the electronic behavior of the B_{12} icosahedron is atomlike.

To better understand the electronic structure of B_{12} , let us begin by considering it in isolation. Its molecular orbitals can be obtained from the $2s$ and $2p$ orbitals of the 12 boron atoms. These result in a total of 48 molecular orbitals, of which 13 are bonding, with A_g , T_{1u} , H_g symmetries (one, three, and nine molecular orbitals, respectively), 12 nonbonding, and 13 antibonding [47]. These orbitals must then be filled with 36 electrons (three per boron). The remaining ten electrons fill the nonbonding orbitals at higher energies. These orbitals face outward [47] and are those that form intersuperatomic bonds before they evolve into bands. These orbitals lead to different bonds in XB_{12} and α -B in a way that depends on the geometry.

In Fig. 3, we show the electronic structure for the isolated B_{12} , compared with SrB_{12} and α -B at ambient pressure, along with the total density of states (DOS). In the isolated B_{12} , one can count the 13 bonding states, going from -15.5 to -3.9 eV, followed by partially filled nonbonding states.

In α -B, there are 18 valence bands. It is immediately visible that the first, isolated band at -15 eV and the group of three bands around -10 eV match those of the isolated atom both in energy and electron count. According to Ref. [45], nine of the remaining bands derive from the other bonding states, and the last five from three covalent bonds and two two-electron-three-center bonds. The 36 electrons of boron completely fill these orbitals, hence the system is a semiconductor.

For the $Pm\bar{3}-XB_{12}$ compounds, the essential features of the electronic structure do not depend on X [48]. For all X elements, the band structure follows a rigid band behavior (for all the bands, we refer the reader to Fig. S8 of the SM [28]). When X is an alkali metal (group III element), the system is hole doped in the valence band (electron doped in the

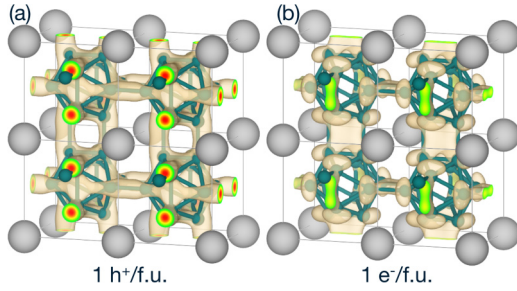


FIG. 4. (a) Local DOS for SrB_{12} with one extra hole per formula unit. (b) Local DOS for SrB_{12} with one extra electron per formula unit.

conduction band). In XB_{12} , the valence bands are 19. The single band at -15 eV and the group of three around -10 are immediately recognizable, like in α -B. Focusing on the Γ point, one can count a total of nine bands in the range from -9.5 to -3.5 eV, which correspond to the remaining B_{12} bonding orbitals (for the full evolution of the eigenvalues from isolated B_{12} to SrB_{12} , we refer the reader to Fig. S5 of the Supplemental Material [28]). The remaining six bands arise from the 12 intersuperatom covalent bonds. The 36 electrons from B fill 18 bands, leaving one unfilled. As a result, when X is a divalent atom, the system is insulating. It is also the valence of the X atom for which the system is thermodynamically most stable. The DFT band gap at ambient pressure is 1 eV for Ca, 1.5 eV for Sr, and 1.7 eV for Ba. A mono/trivalent X gives the metallic state needed for superconductivity.

In other words, we showed that the bonding molecular orbitals of B_{12} behave similarly to semi-core atomic states, which are only slightly perturbed by the environment, while the nonbonding ones behave analogously to valence ones, which change their behavior depending on the environment.

We finish this section by discussing how the bonds that are involved in superconductivity appear in real space, which revealed that both inter- and intrasuperatomic bonds participate.

The only electronic states that contribute to electron-phonon coupling are those near the Fermi level. In Figs. 4(a) and 4(b), we show the local DOS (the real-space localization of the DOS) at the Fermi level under hole and electron doping, respectively (for a detailed definition, we refer the reader to Sec. S1C of the SM [28]). In the hole-doped case, states localize over both the inter- and intra- B_{12} bonds. In the electron-doped case, states at the Fermi energy localize primarily on the single boron atoms and in the square formed by two inter- B_{12} bonds. In both cases, electron-phonon coupling involves contributions from both intra- and intersuperatom bonds, which indicates that all the degrees of freedom of superatoms contribute to superconductivity.

In both the electron- and hole-doped cases, the states near the Fermi level—just above and just below the gap, respectively—are of predominantly boron character (see Figs. S6 and S7 for the atom-projected DOS for all X atoms). These localized orbitals give rise to a strong electron-phonon coupling, comparable to that of MgB_2 . In fact, the concept of covalent-bonds driven metallic, originally proposed for MgB_2 [8], applies equally well to these materials. We will quantify this in the next section.

V. SUPERCONDUCTIVITY TRENDS

We now move on to discuss the superconducting properties of the $\text{Pm}\bar{3}-\text{XB}_{12}$ family. In this section, we focus on the T_c trends across the whole family to understand how much it is possible to optimize it. For this reason, we use the more transparent McMillan formula to estimate the T_c . In Sec. VI, we will focus on the two most promising compounds, LaB_{12} and CsB_{12} , using state-of-the-art Density Functional Theory for Superconductors.

The McMillan formula is [49,50]

$$T_c = \frac{\omega_{\log}}{1.2} \exp \left[-\frac{1.04(1 + \lambda)}{\lambda - \mu^*(1 + 0.62\lambda)} \right]; \quad (1)$$

here $\omega_{\log}$ is the logarithmic average phonon frequency, λ is the electron-phonon coupling constant, and μ^* is the Coulomb pseudopotential. According to this expression, T_c increases strongly with λ in the 0–2 range, increases linearly with $\omega_{\log}$, and decreases with μ^* . In general, λ is the dominating factor, but an increase in coupling is always associated with a suppression of the characteristic phonon energy $\omega_{\log}$. Beyond a certain threshold, the reduction in $\omega_{\log}$ offsets the gain from increased λ and can even lead to a structural instability—a behavior characteristic of the Cohen–Anderson limit [51].

An intuitive description of λ is given by Hopfield’s approximated expression [52]:

$$\lambda = \frac{N(\epsilon_F) \langle g^2 \rangle}{M \langle \omega^2 \rangle}, \quad (2)$$

where $N(\epsilon_F)$, $\langle g^2 \rangle$, M and $\langle \omega^2 \rangle$ are the DOS at the Fermi energy, electron-phonon matrix element, atomic mass, and average phonon frequency, respectively. M and $\langle \omega^2 \rangle$ depend on the material, while $\langle g^2 \rangle$ mainly depends on which bonds contribute to states at the Fermi energy, and $N(\epsilon_F)$ depends on the position of the Fermi energy.

Figure 5 summarizes the main superconducting properties for all metallic XB_{12} compounds at ambient pressure. Critical temperatures were predicted using Eq. (1) and a μ^* of 0.10 and 0.20, which is a conservative range of values assumed by most materials [53,54]. The calculated T_c ’s range from 10 K for trivalent X (Y, La) to over 70 K for CsB_{12} .

Using Eq. (1), it is clear that this variation is caused by the corresponding increase in λ [Fig. 5(b)], from ~ 0.7 in LaB_{12} to $\lambda \sim 1.5$ (strong coupling) in CsB_{12} . As shown in Fig. 5(e), the trend in λ closely follows the behavior of the electronic density of states (DOS) at the Fermi level $N(\epsilon_F)$, hence if we could increase it, λ should follow, and enhance the T_c .

We discussed in Sec. IV that the whole XB_{12} family follows a rigid band behavior (Fig. S8 of the SM [28]). Given the steep profile of the DOS, one could envision further boosting the DOS via hole doping. However, introducing Cs vacancies into CsB_{12} would leave a large void in the structure, which would lead to its collapse under pressure. Instead, to test the idea of hole doping in a more controlled way, we performed a theoretical study of hypothetical XB_{12} compounds with noble gas elements (Ar, Kr, Xe) at the X site.

Although these compounds exhibit prohibitively high formation enthalpies (>500 meV/atom) and are not synthesizable, they provide a useful test of the electronic mechanism to boost T_c . Indeed, noble gas substitutions significantly increase

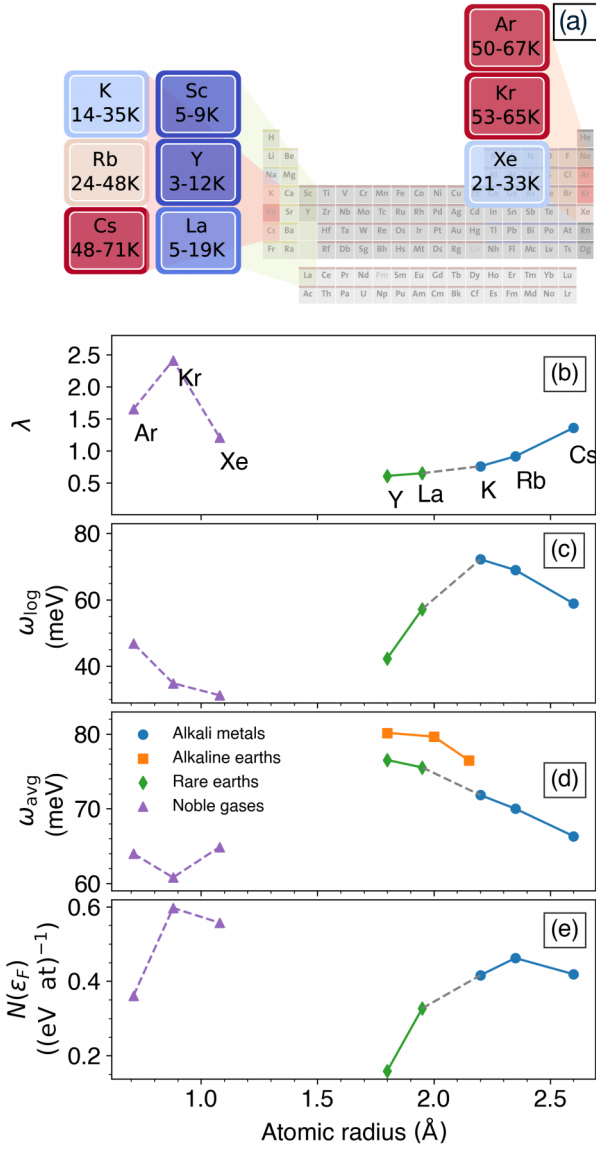


FIG. 5. Main superconducting observables for metallic XB_{12} as a function of the radius of the guest atom. (a) McMillan T_c (in a range of μ^* between 0.1 and 0.2) with respect to the X element in the periodic table. (b)–(e) The electron-phonon coupling λ , the logarithmic average of the phonon frequency $\omega_{\log}$, the plain average of the phonon frequency ω_{avg} , and the DOS at the Fermi energy $N(\epsilon_F)$, respectively. Alkali metals, alkaline earths, rare earths, and noble gases are shown as blue circles, orange squares, green diamonds, and purple triangles, respectively. Dashed grey lines between alkali metals and rare earths are added as a guide to the eye. The dashed lines between noble-gas compounds distinguish them from the others, indicating that their synthesis is unlikely.

both $N(0)$ and λ . Yet, despite these gains, the predicted T_c remains essentially unchanged. This is due to the concurrent phonon softening, which offsets the enhanced coupling strength, as is visible in the drop of $\omega_{\log}$ [Fig. 5(c)], that accompanies the increase in λ [Fig. 5(b)].

From Fig. 5(c), we note that $\omega_{\log}$ varies both with the doping type (alkali metals, rare earths, noble gases) and with the radius of the X atom; since these two vary together along the

series, their individual contributions cannot be disentangled from $\omega_{\log}$ alone. Moreover, $\omega_{\log}$ is only defined for metallic phases, since it requires finite electron-phonon coupling [50], so the insulating alkaline-earth compounds cannot be included in this comparison at all.

To separate the two effects, we turn to the average phonon frequency ω_{avg} . Since the alkaline-earth members (insulating, orange symbols) have radii similar to the rare earths (metallic, green symbols), comparing ω_{avg} between the two isolates the effect of coupling alone: with radius held fixed, coupling shifts the average frequency by 20 meV.

In principle, phonon softening could also arise from hole doping emptying bonding states and weakening the bond stiffness rather than from coupling itself. However, within a single isovalent group (Ar-Kr-Xe; K-Rb-Cs), where the charge is fixed, both λ and $\omega_{\log}$ still depend on the radius [Fig. 5(b)]. Since hole doping cannot produce a trend at constant charge, this radius dependence must instead reflect the coupling itself, confirming that coupling—rather than bond weakening from hole doping—is the dominant effect.

Because the decrease in $\omega_{\log}$ compensates the increase in λ , the T_c of CsB_{12} is already at the saturation limit for the XB_{12} family. As we will see in the following, the presence of a gap right above the Fermi energy also has consequences on μ^* , which is anomalously large due to reduced screening [55,56]. As a result, despite strong coupling and favorable DOS, the critical temperatures of these borides remain moderate. In this sense, CsB_{12} lies near the structural, dynamical, and electronic optimum of the XB_{12} family.

VI. SUPERCONDUCTING PROPERTIES OF CsB_{12} AND LaB_{12} FROM DENSITY FUNCTIONAL THEORY FOR SUPERCONDUCTORS

In this section, we focus on the two compounds that appeared as the most promising in the previous section: CsB_{12} (monovalent atom, highest T_c) and LaB_{12} (trivalent atom, most likely to be synthesized, lowest T_c). We examine their vibrational and superconducting properties in detail, identify the factors that enable XB_{12} to support superconductivity, and validate the results of the previous section.

To study CsB_{12} and LaB_{12} with a parameter-free theory, we solved the fully anisotropic equations of SCDFT (DFT for superconductors). SCDFT permits us to obtain the superconducting critical temperature and gap, accounting on equal footing for phonon-mediated pairing, Coulomb repulsion, and gap anisotropy effects [54].

Figure 6 displays the temperature-dependent superconducting gap [Fig. 6(a)] and its distribution over the Fermi surface [Fig. 6(b)]. In both compounds, the superconducting gap is nearly isotropic, suggesting that the pairing interaction is uniformly distributed in momentum space, without strong band- or mode-selective enhancements. The calculated critical temperatures are 42 K for CsB_{12} and 21 K for LaB_{12} —values that lie at the lower and upper limit of the range predicted with the McMillan formula for the two compounds, respectively. This result suggests that Coulomb interactions in both these systems are strong, while a ratio $\frac{2\Delta}{T_c}$ is 4.2 and 3.7 suggests that coupling is on the moderate/weak regime.

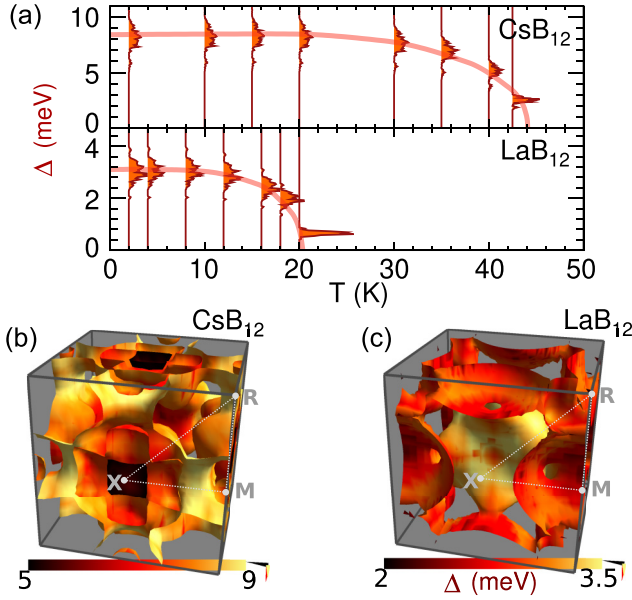


FIG. 6. Superconducting gap of CsB₁₂ and LaB₁₂. (a) Histograms of the anisotropic superconducting gaps as a function of temperature. The thick orange line is a guide to the eye. (b), (c) Fermi surfaces with, color coded, superconducting gap (at $T = 2$ K).

We move to analyze the two components of the pairing kernel: the screened electron–electron repulsion and the attractive electron–phonon interaction, described by the Eliashberg spectral function $\alpha^2F(\omega)$ and the static Coulomb interaction $W(\xi, \xi')$ [54,57,58], shown in Figs. 7(a) and 7(b), respectively.

The effect of Coulomb interaction is anomalously larger than most materials [54,59]. In fact, the partial gapping of states near the Fermi level reduces the efficiency of dynamical Coulomb renormalization [55,56]. This leads to an enhancement of the effective Coulomb interaction relative to constant-DOS models. In fact, the SCDFC critical temperatures fall at the lower end of the McMillan–Allen–Dynes predictions, indicating a high effective μ^* .

Nonetheless, the screened Coulomb interaction $W(\xi, \xi')$, shown in Fig. 7(b), displays the smooth, featureless profile characteristic of good metals [59,60]. The strength of Coulomb repulsion can be quantified as $\mu = W \times \text{DOS}$ at the Fermi level, yielding values of 0.55 for CsB₁₂ and 0.46 for LaB₁₂. While relatively large, these values are consistent with the high DOS.

The Eliashberg function $\alpha^2F(\omega)$ shown in Fig. 7(a) is broadly distributed over the phonon spectrum, both in terms of mode and momentum. As a result, the integrated coupling

$$\lambda(\omega) = 2 \int_0^\omega \frac{\alpha^2F(\omega')}{\omega'} d\omega'$$

(with $\lambda(\infty) = \lambda$) shown as a red line in Fig. 7 increases rather linearly with ω , up to 1.7 in CsB₁₂ and 0.9 in LaB₁₂. In LaB₁₂, the phonon modes are generally harder than in CsB₁₂, as visible from the larger ω_{avg} in Fig. 5(d). Even though the total coupling λ for LaB₁₂ is less, ω_{log} is smaller. This results from a different distribution of the Eliashberg function, as in LaB₁₂ about 25% of the total coupling comes from phonon

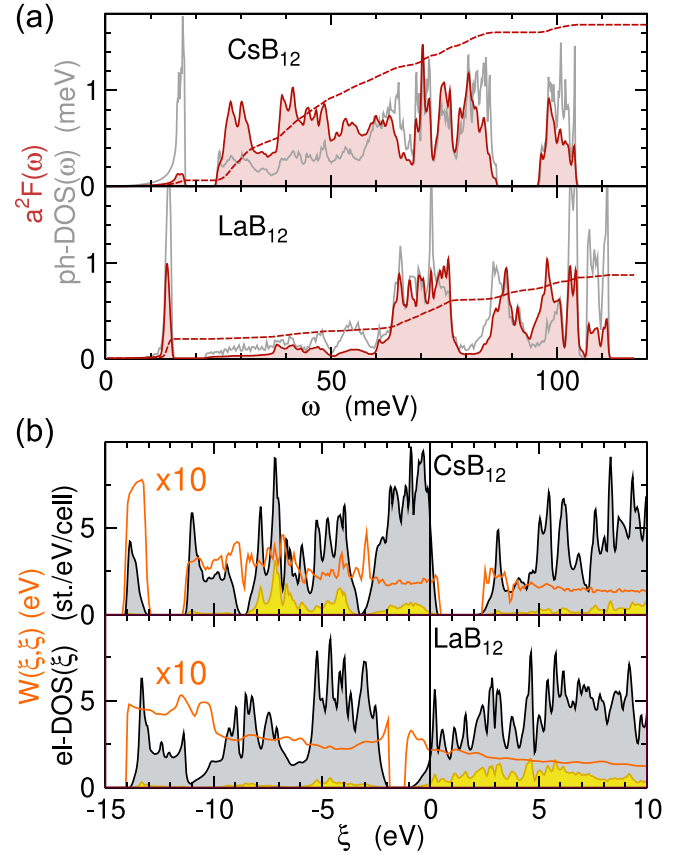


FIG. 7. Electron-phonon and electron-electron (Coulomb) interaction. (a) α^2F function of CsB₁₂ and LaB₁₂ at 0 GPa. Phonon density of states are shown in gray and electron-phonon spectral function (α^2F) is shown in red. The red dashed line is the integration curve of $2\alpha^2F/\omega$, leading to the coupling parameter λ . (b) Density of electronic states (black curve) and its projection on Cs and La (yellow). The orange curve is the diagonal part $W(\xi, \xi)$, the RPA screened electronic interaction entering the SCDFC gap equation [54].

modes at 15 meV (for the complete phonon dispersions with electron-phonon coupling, we refer the reader to Fig. S11 of the Supplemental Material).

In other superconductors, such as MgB₂, the electron-phonon coupling is often strongly mode- and momentum-selective, and concentrated in a narrow frequency range. Since a single mode carries all spectral weight, it is strongly renormalized. Even if one could increase coupling, the system would rapidly become unstable and undergo a structural transition. In contrast, in the XB₁₂ structure this effect is mitigated by the interconnected superatoms, whose phonons involve simultaneous deformations of both intra- and intersuperatom bonds along multiple directions. As a result, the electron-phonon interaction is spread more uniformly across phonon branches and throughout the Brillouin zone, reflecting the absence of a privileged direction and distance for the interaction.

VII. CONCLUSIONS

In conclusion, in this work we identify a class of promising boron-rich superconductors: $Pm\bar{3}-XB_{12}$ compounds

consisting of B_{12} icosahedral superatoms and interstitial electropositive elements, assembled in a superatomic crystal. These compounds can be synthesized under pressures of 50 GPa and, based on dynamical stability, could be recoverable at ambient conditions. XB_{12} crystals present unique advantages for superconductivity: First, boron atoms bond covalently both intra- and intersuperatom, leading to large electron-phonon matrix elements. Second, they are able to withstand extreme hole- and electron doping, which allows a fine tuning of the Fermi energy without compromising the structural stability. In fact, our DFT and SCDFD calculations predict superconducting critical temperatures up to 42 K at ambient pressure, rivaling MgB_2 , with electron-phonon coupling distributed broadly in mode and momentum space.

An alternative, nonequilibrium synthesis route may also be possible under ambient conditions. Since the B_{12} superatoms are already present in pure α -B, they could, in principle, be pulled apart from one another and intercalated with metal atoms. This is not without precedent: Nagatochi *et al.* induced superconductivity in α -B by diffusing lithium atoms into its lattice without disrupting the B_{12} icosahedra [42], though in that case the icosahedra retain the connectivity of the parent α -B allotrope rather than forming the distinct XB_{12} phases discussed here.

The potential to exploit the coupling of molecular states in a framework where the Fermi energy can be easily varied has been proposed as a promising route to find superconductors [61]. These findings highlight the potential of superatomic crystals to realize this idea.

VIII. METHODS

Structural searches were performed using the variable-composition evolutionary algorithm implemented in the USPEX code [22,23], using five steps for structural relaxations. The DFT calculations were performed using the Vienna Ab-initio Simulation Package [62–64], with the projector-augmented wave pseudopotentials provided with the code, and the Perdew-Burke-Ernzerhof approximation [65]. Further details are provided in the Supplemental Material [28].

Electronic structure, phonon and electron-phonon calculations were performed using Quantum ESPRESSO [66,67]. We employed optimized norm-conserving Vanderbilt pseudopotentials [68]. For the ground-state charge density, we employed a cutoff of 80 Ry on the plane wave expansion, and

a $8 \times 8 \times 8$ Monkhorst-Pack mesh and a smearing of 0.04 Ry for integrals over the Brillouin zone. Densities of states were computed non-self-consistently using the tetrahedron method over a $24 \times 24 \times 24$ grid for integrals over the Brillouin zone.

Phonon calculations for LaB_{12} and CsB_{12} were performed on a $6 \times 6 \times 6$ grid, and properties were also interpolated on a $18 \times 18 \times 18$ grid for both electrons and phonons. Convergence tests (see an example in Fig. S15 of the SM [28]) show that integration of phonon dispersions is already reasonably converged with $2 \times 2 \times 2$ grids, as a consequence of the uniformity of coupling in $\mathbf{q}$.

The Coulomb interaction for the SCDFD has been evaluated within the random phase approximation with an energy cutoff for the band summation of about 50 eV above the Fermi level and a maximum $|\mathbf{G}| = 3.0$ a.u. for the summation of crystal field factors [69].

Anisotropic simulations are done with a Monte Carlo algorithm using 50 K $\mathbf{k}$ -points accumulated with higher probability near the Fermi surface [70]. The same algorithm is used to compute the electron-phonon spectral functions shown in Fig. 7. Electronic and coupling parameters are linearly interpolated on the random mesh, which is used for the solution of the SCDFD Kohn-Sham gap equation. We adopt the functional from Ref. [71], also used for the calculation of the physical superconducting gaps [54]. Eliashberg simulations including Coulomb interactions are performed using the approach of Ref. [53].

Fermi surfaces are plotted with the Fermisurfer code [72].

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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