Room-temperature ferromagnetism in a β' -FeB monolayer with pentacoordinate boron

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The search for room-temperature ferromagnetic two-dimensional materials is crucial for the development of future functional spintronic devices. In this study, we identify β' -FeB as a two-dimensional pentacoordinate boron structure that energetically outperforms previously reported FeB structures. First-principles calculations reveal that β' -FeB is a ferromagnetic metal with a Curie temperature exceeding room temperature. Additionally, β' -FeB exhibits substantial resistance to external strain, maintaining its ferromagnetic ground state across a biaxial strain range of -5% to 5%. Furthermore, it exhibits superior thermodynamic, dynamic, and mechanical stability. These findings suggest β' -FeB holds promising application prospects in spintronic devices.

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I. INTRODUCTION

Spintronic devices, known for their rapid processing speeds and reduced energy consumption, are poised to become the next generation of high-performance devices [1–3]. Because of their atomically thin nature, two-dimensional (2D) ferromagnetic materials are of great interest in high-density data storage and spintronic applications [4,5]. Experimental discoveries of a series of 2D magnets, including CrI₃ [4], $Cr_2Ge_2Te_6$ [5], VSe₂ [6], and Fe₃GeTe₂ [7], have marked a significant advancement in the field of 2D magnetic materials.

It is well known that iron (Fe) with a body-centered cubic structure is ferromagnetic at room temperature. Studies have shown that combining Fe with 2D chalcogenide layers results in the formation of 2D magnets [8,9]. Consequently, iron-based 2D compounds are garnering significant interest. Predictions and designs of such materials, featuring high Curie temperatures, have been enabled by first-principles theoretical calculations. Notably, iron-phosphorus compounds such as FeP₃ and FeP₄, which have Curie temperatures of 420 K and 460 K, respectively, have been successfully designed [10,11]. Moreover, research by Niu et al. has demonstrated that two-dimensional iron-silicon nanosheets (FeSi_x) are ferromagnetic metals with Curie temperatures well above room temperature [12]. Furthermore, iron boride compounds like Fe_2B_6 and α -FeB₃ exhibit Curie temperatures of 420 K and 480 K, respectively [13,14].

In the exploration of 2D supercoordination, numerous binary phases featuring planar pentacoordinate with elemental ratio of 1:1 have been identified. In 2018, using the particleswarm optimization (PSO) method within the evolutionary scheme as implemented in the CALYPSO code, Li et al. discovered the lowest-energy configuration of two-dimensional CaSi, identifying it as a semiconductor with an indirect band gap of 0.5 eV [15]. The following year, Ma et al. predicted fully planar pentacoordinate carbon (ppC) in transition metal carbides (TMCs), including Co, Ni, and Cu, also used the PSO method in the CALYPSO code [16]. They identified α -TMC as the most stable phase and β -TMC as a metastable phase. In 2020, Hu et al. predicted an orthorhombic-FeC, which features a perfectly planar pentacoordinate carbon and a seven-coordinate iron, applying the same method [17]. Based on an adaptive generic algorithm structure search, our group discovered a NiB monolayer, which features pentacoordinate boron and exhibits topologically nontrivial electronic properties [18]. Recent research has revealed that the four-coordinate boron structure in a Fe:B = 1:1 ratio has a Curie temperature of 425 K [19], prompting inquiry into the magnetic properties of a similar 1:1 pentacoordinate FeB structure.

In this study, we employ first-principles calculations to investigate the structural, magnetic, and electronic properties of 2D Fe-B materials with a stoichiometric ratio of 1:1. We identify a stable two-dimensional material, β' -FeB, which features pentacoordinate boron and is energetically more favorable than the previously reported four-coordinate FeB. Notably, β' -FeB is a ferromagnetic metal with a Curie temperature exceeding room temperature. Additionally, β' -FeB shows substantial resistance to external strain, maintaining its ferromagnetic ground state across a biaxial strain range of-5% to 5%. This work opens new avenues for the design of two-dimensional magnetic materials.

II. COMPUTATIONAL DETAILS

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The Vienna Ab initio Simulation Package (VASP) software, based on the spin-polarized density functional theory

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FIG. 1. Top and side view of the optimized β' -FeB monolayer. The unit cell is marked with a dashed square.

was used for all calculations in this paper [20,21]. The exchange-correlation interaction was treated with the generalized gradient approximation (GGA) parameterized by the Perdew-Burke-Ernzerhof formula (PBE) [22]. The kinetic energy cutoff for the plane-wave expansion was set to 500 eV. The convergence criterion for energy and forces was set to 10^{-5} eV and 10^{-3} eV/Å, respectively. The thickness of the vacuum layer was set to 20 Å, and a k-point grid resolution of $2\pi \times 0.01 \text{ Å}^{-1}$ was employed for Brillouin zone integration. The phonon dispersion of the material was calculated using the PHONOPY software [23]. Ab initio molecular dynamics (AIMD) was conducted within the canonical ensemble (NVT) at 300 K for a total duration of 14 ps. Using the multidimensional Curie temperature simulation software developed by Zhang et al., Cuire temperature (Tc) of the material was calculated and 100×100×1 lattice was used in the Monte Carlo simulation [24]. The chemical bond strengths were calculated through the crystal orbital Hamiltonian population (COHP) implemented in the LOBSTER code [25–28].

III. RESULTS AND DISCUSSION

A. Atomic structure

Through structural searches, researchers have identified two pentacoordinate 2D carbides with different configurations, featuring a 1:1 ratio of metal to carbon [16,17]. Based on these discovered configurations, we derived two 2D ironboron compounds by element substitution, namely β' -FeB and α -FeB, as illustrated in Fig. 1 and Fig. S1(a) within the Supplemental Material (SM) [29]. Both structures consist of four Fe atoms and four B atoms, with each B atom coordinated to four Fe atoms and an adjacent B atom. The main difference between these two structures lies in the arrangement of the Fe six-membered rings. β' -FeB has optimized lattice constants of a = 6.234 Å and b = 4.847 Å with the Fe atoms arranged non-coplanarly: Fe1 and Fe3 atoms are about 0.062 Å above the plane, while Fe2 and Fe4 atoms are about 0.062 Å below the plane. The Fe-B bond lengths are 2.017 Å and 2.051 Å. Conversely, α -FeB presents a purely planar structure with optimized lattice constants a = 7.666 Å and b = 3.951 Å. Additionally, we also examined FeB structures in honeycomb and square configurations, termed honeycomb-FeB and square-FeB, respectively [illustrated in Figs. S1(b) and S1(c)]. The previously reported tetragonal-FeB [19], is shown in Fig. S1(d). As shown in Table S1 within the SM [29], the relative energies of the structures are ordered as follows: α -FeB < β' -FeB < tetragonal-FeB < square-FeB < honeycomb-FeB, with β' -FeB being only 3 meV/atom higher than α -FeB. It is worth noting that the reported relative energy of the tetragonal-FeB structure is 121 meV/atom higher than that of α -FeB.

B. Structural stability

To assess the stability of the structures, we calculated their cohesive energies E_{coh} , as defined by

$$E_{\rm coh} = \frac{nE_{\rm Fe} + mE_{\rm B} - E_{\rm tot}}{m+n},\tag{1}$$

where $E_{\rm Fe}$ and $E_{\rm B}$ represent the total energies of isolated Fe and B atoms, respectively, and $E_{\rm tot}$ is the total energy of the structure. Here, *m* and *n* denote the number of B and Fe atoms in the unit cell, respectively. The calculated cohesive energy for β' -FeB is 5.131 eV/atom, surpassing those of experimentally prepared FeB₂ (4.87 eV/atom) [30], silicene (3.71 eV/atom) [31,32], and Cu₂Si (3.46 eV/atom) [33]. The cohesive energy of α -FeB is 5.134 eV/atom. These results indicate a highly stable, strongly bonded network in α -FeB and β' -FeB.

To evaluate the thermal stability of the materials, we conducted AIMD simulations at 300 K using various supercell configurations: 3×3 for β' -FeB, 3×4 for α -FeB, 4×4 for square-FeB, and 5×5 for honeycomb-FeB. The results are depicted in Fig. 2(a) and Fig. S2 within the SM [29]. Each AIMD simulation ran for a total of 14 ps with a time step of 2 fs. The geometric snapshots captured at the end of the simulation reveal that α -FeB and β' -FeB maintain their structural integrity at room temperature. Conversely, square-FeB transformed into tetragonal-FeB, while honeycomb-FeB was destroyed. This indicates that tetragonal-FeB and honeycomb-FeB are unstable at room temperature. To determine the temperature limit of the α -FeB and β' -FeB, we performed AIMD simulations up to 2500 K. As shown in Fig. S3 within the SM [29], β' -FeB maintains its original configuration at 800 K, 1500 K, and 2000 K without significant structural distortion. However, at 2500 K, the structure was destroyed. Similarly, as shown in Fig. S4 within the SM [29], α -FeB remains stable at 2000 K but is destroyed at 2500 K. These results indicate that both α -FeB and β' -FeB can withstand temperatures in the range 2000-2500 K, which is comparable to the limits of Cu₂Si [33] (1500 K) and FeB₂ [30] (1500 K), but lower than TiB_4 [34] (3000 K).

Furthermore, to check the dynamic stability of α -FeB and β' -FeB, we calculated their phonon spectra, as depicted in Fig. 2(b) and Fig. S5 within the SM [29]. The



FIG. 2. (a) Evolution of total potential energy with simulation time at 300 K for 14 ps. The insets are the top and side views of the atomic structure at the end of the simulation, respectively. (b) The phonon dispersion of β' -FeB.

phonon spectrum of α -FeB exhibits significant imaginary frequencies, indicating instability. In contrast, the spectrum of β' -FeB shows no imaginary frequencies, confirming its dynamic stability. Additionally, the mechanical stability of the β' -FeB monolayer was evaluated by calculating its elastic constants, which must satisfy the criteria for two-dimensional materials: $C_{11}C_{22} > C_{12}^2$ and $C_{66} > 0$ [35]. The elastic constants for β' -FeB, $C_{11} = 140.88$ N/m, $C_{22} = 151.15$ N/m, $C_{12} = 24.43$ N/m, and $C_{66} = 61.60$ N/m, meet these stability criteria. From these elastic constants, the Young's moduli are evaluated as $E_a = \frac{C_{11}C_{22}-C_{12}^2}{C_{22}} = 136.93$ N/m and $E_b = \frac{C_{11}C_{22}-C_{12}^2}{C_{11}} = 146.91$ N/m. Although these values are lower than those of graphene ($E_a = E_b = 331.63$ N/m) [36], they indicate that β' -FeB possesses favorable mechanical properties, which could render it responsive to strain tuning.

To better understand the chemical bonding and electron distribution in β' -FeB, we computed the electron localization function (ELF). The ELF ranges from 0 to 1 [37], where lower values indicate areas with sparse electron density, while higher values denote regions with robust covalent bonding or lone electron pairs. An ELF value of 0.5 suggests a uniformly distributed electron gas. As seen from Fig. S6(a) within the SM [29], the B-B bonds show a covalent characteristic, whereas the Fe-B bonds exhibit a predominantly ionic nature. The blue regions surrounding the Fe atoms, characterized by an ELF value of 0, suggest electron depletion in the vicinity of the Fe atoms. Furthermore, electrons predominantly accumulate around the B dimer, as shown in Fig. S6(b). Bader charge analysis reveals that each B dimer receives approximately 0.85e from adjacent Fe atoms, suggesting significant ionization of Fe atoms. This electron transfer from Fe to B atoms contributes to the stabilization of the structural framework. To gain insight into the bonding strength, we performed a crystal orbital Hamiltonian population (COHP) analysis. The more negative the ICOHP value, the stronger the bonding strength. The results, shown in Fig. S7 within the SM [29], reveal that the B-B bond has the highest ICOHP value (-3.28/-3.31 for spin up/down), followed by the Fe-B bond (-0.98/-1.12), while the Fe-Fe bond exhibits the lowest ICOHP value (-0.14/-0.36). These findings indicate that the B-B bond exhibits the strongest bonding strength, whereas the Fe-Fe bond shows the weakest.

So far, β' -FeB has not been experimentally synthesized. Topochemical deinteraction has emerged as a powerful and promising technique for the development of 2D materials. It enables the synthesis of 2D materials under mild conditions and generally results in minimal structural alterations, allowing the resultant 2D materials to retain the key characteristics of their parent structure. By topochemical deintercalation of Li from lavered room-temperature RT-LiNiB, Zaikina et al. successfully synthesized monolayer NiB at room temperature [38]. It is also worth noting that the RT-LiNiB crystals can be synthesized using a facile route by heating the lithium hydride precursor to 1173 K for 12 hours, followed by slow cooling to room temperature [39]. The β' -FeB monolayer shows structural similarities to the NiB monolayer, suggesting that a similar preparation method could be employed to synthesize layered LiFeB and to obtain monolayer FeB through a topochemical deintercalation approach.

In addition, Zhao *et al.* have successfully prepared bulk β -FeB at high-temperature (1800 K) and high-pressure (5 GPa) conditions [40]. In our proposed β' -FeB, Fe atoms form hexagonal frameworks, with B dimers located at the centers of the Fe hexagonal rings, as illustrated in Fig. S8(e) within the SM [29]. Notably, similar structural characteristics can also be discerned in bulk β -FeB. As shown in Figs. S8(a)– S8(c), by extracting two layers of Fe and two layers of B from the bulk β -FeB, we obtain a FeB flake that features similar hexagonal iron framework, with boron dimers slightly inclined within the Fe hexagonal rings. However, it is important to note that this FeB flake retains a certain thickness, contrasting with the nearly planar configuration of our proposed FeB monolayer. For comparison, we also present the atomic structure of the two-dimensional α -FeB in Fig. S8(d). It is noteworthy that α -FeB and β' -FeB exhibit structural similarities to the NiB monolayers, which can be derived from high-temperature and room-temperature LiNiB, respectively [39]. In comparison with β' -FeB, the Fe hexagonal rings and B dimers in α -FeB and bulk β -FeB show distinct orientations. Given that bulk β -FeB is synthesized under high-temperature (1800 K) and



FIG. 3. (a) Electronic band structures and (b) partial density of states (PDOS) for the monolayer β' -FeB.

high-pressure (5 GPa) conditions, and considering that α -FeB is associated with high temperatures, we speculate that elevated temperatures may influence the orientations of the Fe hexagonal rings and B dimers, whereas high pressures could induce buckling between adjacent Fe atoms and lead to a transition of the B dimers from an in-plane to an out-ofplane configuration. This indicates that our proposed β' -FeB structure may be synthesized under milder conditions, such as ambient pressure and reduced temperatures. If the lowtemperature, ambient-pressure allotrope of bulk β -FeB can be successfully synthesized, it may be possible to subsequently employ a bond-breaking chemical method to obtain the β' -FeB monolayer. However, this hypothesis requires rigorous experimental validation to confirm its feasibility.

C. Magnetic and electronic properties

To determine the magnetic ground state of β' -FeB, we considered nonmagnetic (NM), ferromagnetic (FM), and four antiferromagnetic (AFM) configurations, as depicted in Fig. S9 within the SM [29]. The relative energies of these magnetic states are summarized in Table S2. Our calculations reveal that the FM state is energetically more favorable than the NM and AFM states, confirming that the ground state of β' -FeB is ferromagnetic. Subsequent band structure calculations show that the β' -FeB in FM state is metallic, as evident in Fig. 3(a). This metallicity was further verified by calculating the electronic band structure using the HSE06 functional, illustrated in Fig. S10 within the SM [29]. The partial density of states (PDOS), shown in Fig. 3(b), reveals that the magnetism of β' -FeB predominantly arises from the unpaired electrons in the 3d orbitals of Fe atoms. The magnetic moment of an Fe atom is calculated to be 2.16 μ_{B} . Additionally, the spin magnetic moments on each B atom is approximately 0.13 μ_B and antiparallel to the spin direction of the Fe atoms. This suggests that ferromagnetism in β' -FeB is driven by FM superexchange interactions between the partially occupied dorbitals of Fe atoms and the *p* orbitals of B atoms.

Magnetic anisotropy energy (MAE) critically influences the thermal stability of magnetic data storage devices. To understand the directional preference of β' -FeB's magnetization, we calculated the MAE for five different orientations based on spin-orbit coupling (SOC). As illustrated in Fig. 4(a), the easy axis (EA) of β' -FeB aligns along the (001) direction, demonstrating a preference for an out-of-plane orientation. Further analysis, shown in Fig. 4(b), reveals that the larger MAE predominantly arises from the SOC interactions between the dyz and dz^2 orbitals of Fe atoms.

The Curie temperature (T_c) is a crucial parameter that influences the viability of magnetic materials for applications in spintronic devices. We evaluated T_c of β' -FeB using Monte Carlo simulations based on the Heisenberg model, described by

$$E = E_0 - \sum_{i,j} J_{ij} \vec{S}_i \cdot \vec{S}_j.$$
⁽²⁾

In this model, E_0 represents the nonmagnetic energy, J_{ij} is the exchange integral between the *i*th and *j*th Fe atoms (as shown in Fig. S11 within the SM [29]), and \vec{S} denotes the spin quantum number of the Fe atoms. According to Eq. (2), the following equations can be established by combining the total energy of the five magnetic configurations:

$$E_{\rm FM} = E_0 - 4J_1S^2 - 8J_2S^2 - 4J_3S^2 - 16J_4S^2, \qquad (3)$$

$$E_{\text{AFM-1}} = E_0 - 4J_1S^2 + 8J_2S^2 - 4J_3S^2 + 16J_4S^2, \quad (4)$$

$$E_{\text{AFM-2}} = E_0 + 4J_1S^2 + 8J_2S^2 + 4J_3S^2 - 16J_4S^2, \quad (5)$$

$$E_{\text{AFM-3}} = E_0 + 4J_1S^2 - 8J_2S^2 + 4J_3S^2 + 16J_4S^2, \quad (6)$$

$$E_{\text{AFM-4}} = E_0 - 4J_1S^2 + 4J_3S^2.$$
⁽⁷⁾

The exchange constants J_1 , J_2 , J_3 , and J_4 for β' -FeB are calculated to be 55.8, 52.5, 12.7, and 8.2 meV/S², respectively. Generally speaking, the stronger the magnetic exchange, the higher the Curie temperature of the material [10]. Through Monte Carlo (MC) simulations, as shown in Fig. 4(c), the T_c of β' -FeB is determined to be 450 K, significantly surpassing room temperature (300 K).

The nonempirical, strongly constrained and appropriately normed (SCAN) meta-GGA functional, developed by Sun *et al.* [41], has been proven as a reliable functional to describe the structural, magnetic, and electronic properties of transition metal compounds with 3*d* electrons [42–44]. Here, we also employed the SCAN method to calculate the magnetic properties of β' -FeB. The magnetic moment of the Fe atom was



FIG. 4. (a) Magnetic anisotropy energies along different magnetization directions and (b) orbital-resolved MAE of *d* orbitals of Fe atom for β' -FeB. The variation of the magnetic moment (M) of Fe atom (blue) and susceptibility $\chi = (\langle \vec{M}^2 \rangle - \langle \vec{M} \rangle^2)/k_B T$ (red) with respect to temperature in the β' -FeB monolayer. The calculation constants of the Heisenberg model are derived from the (c) PBE functional and (d) SCAN functional.

calculated to be 2.41 μ_B , with the exchange coefficients J_1 , J_2 , J_3 , and J_4 measured at 68.3, 37.8, 0.17, and 13.1 meV/S², respectively. Additionally, the Curie temperature was determined to be 530 K [Fig. 4(d)].

D. Effects of strain

The properties of two-dimensional magnetic materials can be readily modulated by external strain [45,46]. We explored the effect of biaxial strain on the magnetic properties of β' -FeB. Biaxial strains ranging from -5% (compression) to 5% (tensile) were applied, with a focus on the relative stability between the FM and AFM states. The energy difference $\Delta E = E_{\text{AFM}} - E_{\text{FM}}$, where a positive ΔE signifies that the FM state is energetically favored. Our findings [Fig. 5(a)] indicate that the energy difference remains positive at all applied strains, confirming that β' -FeB maintains a FM ground state within this strain range.



FIG. 5. (a) Relative energies of ferromagnetic and antiferromagnetic states vary with biaxial strains ranging from -5% to 5%. (b) Phonon dispersion diagram of β' -FeB under 5% tensile strain.

Additionally, examining the structural stability of the β' -FeB structure under tensile and compressive strain is necessary. As shown in Fig. 5(b), the β' -FeB with 5% tensile strain has no imaginary frequencies in its phonon spectrum, indicating that it is dynamically stable. In contrast, the β' -FeB with 5% compressive strain exhibits significant imaginary frequencies (Fig. S12(a) within the SM [29]). Even a 1% compressive strain leads to imaginary frequencies [Fig. S12(b)], suggesting that the β' -FeB is dynamically unstable under compression. Furthermore, AIMD simulations at 300 K for the β' -FeB with 5% tensile strain [Fig. S13] show that it maintains its structural integrity throughout the simulation period, thereby confirming its thermodynamic stability. These results indicate that the β' -FeB remains stable under tensile strain but becomes unstable under compressive strain. The growth of two-dimensional materials requires the selection of appropriate substrates. Our results indicate that the lattice constant of the substrate should be greater than or equal to the lattice constant of the β' -FeB to avoid compressive strains during growth, which helps maintain the stability of the structure.

IV. CONCLUSIONS

In conclusion, our study has enhanced the understanding of β' -FeB as a promising two-dimensional magnetic material. Featuring pentacoordinate boron, β' -FeB is dynamically, thermodynamically, and mechanically stable. Moreover, it is energetically more favorable than previously reported FeB structures. We have discovered that β' -FeB exhibits a ferromagnetic ground state with a high Curie temperature, significantly surpassing room temperature, highlighting its promising potential for applications in spintronics. Under a range of biaxial strains from -5% to 5%, β' -FeB maintains its ferromagnetic ground state, making it a strong candidate for future research and applications in spintronics.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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