

Article

Spin-Polarized Electronic Structure, Charge Analysis, and Magnetic Stability in Fe-Doped SiC Nanosheets: A DFT + U Study

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Abstract

In this work, the structural, electronic, charge-transfer, thermal, and magnetic properties of pristine and Fe-doped silicon carbide nanosheets (SiCNSHs) were systematically investigated using spin-polarized density functional theory (DFT) within the Local Spin Density Approximation including Hubbard correction (LSDA + U). A 4×4 SiCNSH supercell containing 80 atoms was considered, where Fe atoms were substitutionally introduced at carbon sites to evaluate dopant-induced modifications in the nanosheet. Structural optimization, energy convergence, force minimization, and stress evolution analyses confirm that Fe incorporation preserves the structural integrity of the SiCNSH and leads to energetically stable configurations. The calculated defect formation energy (-7.44 eV/atom) demonstrates the thermodynamic feasibility of Fe substitution, while ab initio molecular dynamics (AIMD) simulations at 300 K verify the thermal stability of the energetically favorable Fe-doped configuration. Electronic-structure calculations reveal that pristine SiCNSH exhibits a nonmagnetic semiconducting nature with a band gap of approximately 2.4 eV, whereas Fe incorporation significantly modifies the electronic structure through pronounced Fe-3d/C-2p/Si-3p orbital hybridization. The band gap is reduced to approximately 1.1 eV for the single-Fe-doped system and further decreases to 0.53/0.51 eV (spin-up/spin-down) in the double-Fe configuration, while preserving semiconducting behavior. Spin-polarized band structure and density of states analyses demonstrate clear spin asymmetry near the Fermi level, indicating strong dopant-induced spin polarization and exchange interactions. Charge-density difference and Bader charge analyses reveal substantial dopant-induced charge redistribution characterized by electron depletion around Fe atoms, enhanced electron accumulation on neighboring carbon atoms, and partial charge neutralization of nearby Si atoms, resulting in a more localized covalent Si-C-Fe bonding environment. Mulliken spin population analysis further demonstrates robust ferromagnetic ordering, where the Fe dopant acts as the dominant magnetic center with strong induced spin polarization extending into neighboring Si and C atoms. Comparison between ferromagnetic (FM) and antiferromagnetic (AFM) configurations confirms that the



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2Fe@C-doped SiCNSh stabilizes in a ferromagnetic ground state, exhibiting a favorable FM–AFM energy difference of 0.216 eV. Based on the mean-field approximation, the Curie temperature was estimated to be approximately 837 K, indicating strong magnetic stability significantly above room temperature. The present findings collectively demonstrate that Fe incorporation effectively tailors the electronic and magnetic properties of SiCNSh through band-gap engineering, spin-symmetry breaking, and stabilization of high-temperature ferromagnetism. These combined characteristics establish Fe-doped SiCNShs as promising candidates for spintronic devices, magnetic semiconductors, spin injectors, spin filters, and non-volatile magnetic memory applications.

Keywords: Fe-doped SiC nanosheet; density functional theory (DFT); spintronic materials; ferromagnetic semiconductors; curie temperature; charge redistribution

1. Introduction

Two-dimensional (2D) materials have attracted remarkable scientific interest due to their exceptional physical properties arising from reduced dimensionality, quantum confinement, and strong surface effects. Since the discovery of graphene, a wide range of atomically thin materials, including transition-metal dichalcogenides (TMDs), hexagonal boron nitride, silicene, phosphorene, and transition-metal carbides, have been extensively explored for next-generation nanoelectronic, optoelectronic, sensing, and spintronic applications. Their tunable electronic band structures, large surface-to-volume ratio, and controllable charge transport characteristics provide unique opportunities for designing multifunctional low-dimensional devices [1–3].

Among emerging 2D semiconductors, silicon carbide (SiC) nanosheets have attracted increasing attention owing to their outstanding thermal, mechanical, and chemical stability combined with semiconducting electronic behavior. In contrast to graphene, which exhibits a zero bandgap, SiC nanosheets possess an intrinsic wide bandgap, excellent thermal conductivity, superior radiation resistance, and high-temperature stability, making them attractive candidates for harsh-environment electronics, high-power devices, and spin-dependent transport technologies [4,5]. Furthermore, the mixed ionic–covalent nature of Si–C bonding provides additional flexibility for electronic and magnetic modulation through defect engineering and chemical doping [4,6].

Recent advances have demonstrated that the intrinsic properties of 2D SiC can be significantly modified through vacancies, surface functionalization, and transition-metal (TM) incorporation. In particular, substitutional doping with TM atoms has emerged as one of the most efficient strategies for inducing spin polarization, modifying charge redistribution, and tailoring band topology in low-dimensional semiconductors. Previous first-principles studies have revealed that TM dopants may introduce impurity states near the Fermi level, enhance exchange interactions, and even transform nonmagnetic semiconductors into half-metallic or spin-polarized systems suitable for spintronic applications [6–8]. Such dopant-induced modifications are strongly governed by orbital hybridization between transition-metal d states and host p orbitals, which critically determine the stability and magnetic ordering of doped systems [7,8].

Previous first-principles investigations have demonstrated that defect engineering, chemical functionalization, and transition-metal incorporation can significantly modify the electronic and magnetic characteristics of SiC-based low-dimensional systems. In particular, Fe-induced defects, fluorination, hydrogenation, and transition-metal substitution have been reported to induce spin polarization, ferromagnetic coupling, bandgap modulation,

and enhanced magnetic stability, highlighting the strong sensitivity of SiC nanostructures to local bonding environments and dopant-induced orbital hybridization [9–14]. For SiC-based nanostructures, several theoretical studies have investigated the influence of transition-metal doping on magnetic and electronic behavior. It has been shown that TM incorporation can induce localized magnetic moments, alter electronic conductivity, and stabilize ferromagnetic ordering in SiC monolayers and nanotube systems [6,8,13,14]. In particular, Fe doping has emerged as a highly promising route due to the partially filled Fe-3d orbitals, which strongly hybridize with neighboring atoms and promote spin asymmetry around the Fermi level. Previous reports on Fe-containing SiC nanostructures have suggested the possibility of half-metallicity, strong spin polarization, and Curie temperatures (T_C) exceeding room temperature, highlighting their technological relevance for magnetic semiconductors and spin-filtering devices [9,13,14]. In addition, chemical functionalization strategies such as fluorination and hydrogenation have also been reported to effectively tailor the electronic and magnetic characteristics of SiC monolayers, providing complementary routes for engineering spin-dependent properties [10,11].

Recent studies have further emphasized the remarkable tunability of SiC-based low-dimensional systems through transition-metal engineering and data-assisted computational approaches. For example, Co-doped SiC nanotubes were shown to exhibit significant electronic modulation under quantum confinement, while Ag incorporation in SiC nanotubes induced robust ferromagnetic behavior and enhanced magnetic stability [15,16]. Machine-learning-assisted DFT studies have additionally demonstrated improved prediction of density-of-states evolution in magnetically doped SiC nanotubes, highlighting the growing importance of hybrid computational frameworks for accelerated material design [17,18]. More recently, transition-metal-doped SiC nanotubes, including Fe-, V-, Al-, and Ga-containing systems, have been predicted to exhibit strong spin polarization, tunable magnetic moments, half-metallicity, and Curie temperatures exceeding room temperature, thereby reinforcing their potential for spintronic and magnetic semiconductor applications [19–23]. Furthermore, chirality-dependent stability and electronic behavior of SiC-based nanotubes have also been systematically explored, revealing the critical role of structural confinement and dopant configuration in governing magnetic exchange interactions and electronic transport characteristics [24]. These findings collectively highlight the exceptional versatility of SiC-based nanostructures and strongly motivate the investigation of Fe substitution in two-dimensional SiC nanosheets. Among the various transition-metal dopants, Fe has attracted particular attention because its partially occupied 3d orbitals can induce strong spin polarization, magnetic ordering, and significant modifications in the electronic structure of SiC-based nanomaterials. Previous theoretical studies have demonstrated that Fe incorporation can generate localized magnetic moments and spin-dependent electronic states, making Fe-doped SiC systems promising candidates for spintronic and magnetic semiconductor applications. Therefore, the present work focuses specifically on Fe substitution at carbon sites to provide a detailed understanding of the associated electronic, magnetic, thermal-stability, and charge-transfer mechanisms. Although other transition-metal dopants have also been investigated in SiC-based systems, Fe remains one of the most promising candidates for achieving robust magnetic behavior and spin-dependent electronic functionality.

The rapid advancement of next-generation spintronic technologies has created an increasing demand for low-dimensional materials capable of simultaneously exhibiting semiconducting behavior, robust magnetic ordering, and thermally stable spin polarization. Silicon carbide (SiC)-based nanostructures have emerged as promising candidates due to their exceptional thermal stability, chemical robustness, and tunable electronic properties. However, pristine SiC nanosheets are intrinsically nonmagnetic and exhibit limited spin

functionality, which significantly restricts their applicability in spin-dependent nanoelectronic devices. Although transition-metal doping has been proposed as an effective strategy for inducing magnetism in SiC-based systems, the underlying mechanisms governing magnetic stabilization, spin polarization, and thermal robustness remain insufficiently understood, particularly for substitutional Fe doping at carbon sites in two-dimensional SiC nanosheets. Moreover, previous studies have reported inconsistent magnetic behavior in Fe-doped SiC nanostructures, ranging from weak antiferromagnetism to ferromagnetic stabilization depending on dimensionality and substitutional configuration. Therefore, a systematic understanding of Fe-induced electronic reconstruction, charge redistribution, magnetic ordering, and Curie-temperature behavior in Fe-doped SiCNShs remains critically needed for their rational integration into future spintronic technologies.

Despite these promising developments, several important issues remain insufficiently understood. Most previous studies primarily focused on electronic band modulation or magnetic moment generation, whereas the combined influence of Fe substitution on structural stability, thermal robustness, charge redistribution, magnetic ordering, and Curie-temperature evolution in two-dimensional SiC nanosheets remains largely unexplored. In particular, a systematic understanding of how Fe substitution at carbon sites affects local bonding characteristics, electron transfer, and magnetic exchange interactions is still lacking. Moreover, few studies simultaneously combine *ab initio* molecular dynamics (AIMD), electron density difference (EED), Bader charge analysis, Mulliken population analysis, and magnetic exchange evaluation to comprehensively explain the microscopic origin of magnetism in Fe-doped SiC nanosheets [8,9,13,14].

The primary aim of this study is to systematically investigate the influence of substitutional Fe incorporation at carbon sites on the structural, electronic, charge-transfer, thermal, and magnetic properties of two-dimensional SiCNShs using spin-polarized DFT–LSDA + U calculations. Specifically, this work aims to (i) evaluate the thermodynamic and thermal stability of Fe-doped configurations through formation-energy analysis and AIMD simulations, (ii) elucidate the dopant-induced electronic reconstruction and band-gap engineering through spin-polarized band structure and DOS analyses, (iii) quantify charge redistribution and orbital hybridization mechanisms using electron density difference and Bader charge analyses, and (iv) determine the origin and stability of magnetic ordering through Mulliken spin populations, FM–AFM energy comparison, and Curie temperature estimation. Ultimately, this study seeks to assess the potential of Fe-doped SiCNShs as high-performance materials for spintronic, magnetic semiconductor, and nanoelectronic applications.

The novelty of the present work lies in the comprehensive first-principles investigation of carbon-site Fe substitution (Fe@C) in an 80-atom two-dimensional SiCNSh supercell, systematically combining structural optimization, defect energetics, thermal stability, electronic reconstruction, charge-transfer analysis, magnetic ordering, and Curie temperature prediction within the spin-polarized DFT–LSDA + U framework. Unlike previous studies that primarily focused on Fe substitution at silicon sites or reported weak magnetic stabilization in Fe(C)-doped SiC nanostructures, the present work demonstrates that 2Fe@C-doped SiCNSh stabilizes in a robust ferromagnetic semiconducting ground state, characterized by a favorable FM–AFM energy difference and a remarkably high predicted Curie temperature. Furthermore, the present study provides atomistic insight into the role of Fe-3d/C-2p/Si-3p hybridization, spin-density delocalization, and dopant-induced charge redistribution as the fundamental mechanisms governing magnetic stabilization and electronic modulation in Fe-doped SiCNShs. The simultaneous realization of high-temperature ferromagnetism, tunable semiconducting behavior, and structural stability distinguishes the present system from previously reported SiC-based magnetic nanostructures.

Motivated by these unresolved challenges, the present work employs spin-polarized density functional theory (DFT) within the LSDA + U framework to systematically investigate the structural, electronic, charge-transfer, and magnetic properties of pristine and Fe-substituted SiC nanosheets. Single- and double-Fe substitution at carbon sites are considered to evaluate the effect of dopant concentration and magnetic coupling on system behavior. Structural stability is assessed through total-energy analysis, formation-energy calculations, force/stress optimization, and finite-temperature AIMD simulations. The underlying electronic and bonding mechanisms are further clarified through spin-polarized band structures, total and partial density of states, electron density difference maps, Bader charge analysis, and Mulliken population calculations. In addition, magnetic moments, spin-density distributions, ferromagnetic–antiferromagnetic energy differences, and Curie temperatures are evaluated to assess the potential of Fe-substituted SiC nanosheets for future spintronic and magnetic semiconductor applications.

2. Computational Methodology

All first-principles calculations were carried out within the framework of spin-polarized density functional theory (DFT) [25] as implemented in the QuantumATK simulation package [26]. The exchange–correlation interaction was described using the Local Spin Density Approximation (LSDA) [27] with the Perdew–Zunger (PZ) parameterization [28].

To improve the accuracy of the electronic structure description and obtain more reliable band-gap estimation, Hubbard U corrections were incorporated into the DFT calculations. In the present work, an orbital-dependent LSDA + U formalism was employed within the simplified rotationally invariant approach proposed by S.L. Dudarev [29], corresponding to the $J = 0$ limit of the more general formulation introduced by A.I. Liechtenstein [30]. The Hubbard correction was applied to the Si d-projector states with $U = 5.0$ eV and $J = 0$ eV, and to the C p-states with $U = 4.8$ eV and $J = 0$ eV. These values were selected because they provide a semiconducting band structure and band-gap value for bulk three-dimensional SiC that are in reasonable agreement with previously reported experimental and theoretical data. The same U parameters were subsequently adopted for the pristine SiC nanosheet to ensure a consistent description of the Si–C electronic framework. To further assess the reliability of the adopted LSDA + U approach, additional HSE06 calculations were performed for the pristine SiC nanosheet, yielding a band gap of 2.91 eV and confirming the semiconducting character predicted by LSDA + U.

It is worth noting that, in our previous investigations of SiC-based nanostructures, including magnetically doped SiC nanotubes [17], various exchange–correlation approximations and computational approaches were systematically examined to assess their ability to reproduce the electronic properties of SiC systems. These benchmark studies demonstrated that the LSDA + U framework provides a reliable and physically meaningful description of the Si–C electronic structure, yielding band-gap values in reasonable agreement with available experimental and theoretical data. Based on these findings, the LSDA + U approach was adopted in the present work to ensure methodological consistency and to enable a comprehensive investigation of the electronic, magnetic, charge-transfer, and thermal properties of Fe-doped SiC nanosheets.

Although advanced many-body perturbation approaches such as the GW method generally provide highly accurate band-gap predictions, their application to large transition-metal-doped supercells remains computationally demanding, particularly when combined with extensive structural optimization, finite-temperature AIMD simulations, and magnetic-property calculations. Based on our previous investigations of bulk SiC and additional benchmark calculations using various exchange–correlation approximations, the LSDA + U approach was found to provide a reliable description of the electronic structure and to

yield band-gap values in reasonable agreement with available experimental and theoretical data. Therefore, LSDA + U was adopted in the present work as a computationally efficient framework capable of simultaneously describing localized Fe-3d electron correlations and enabling comprehensive investigations of structural, electronic, magnetic, charge-transfer, and thermal properties. For the Fe-doped configurations, an additional Hubbard correction was introduced for the Fe-3d states using $U = 4.6$ eV and $J = 0$ eV to properly account for the strong on-site Coulomb interactions associated with partially occupied transition-metal d orbitals. The selected U value lies within the commonly reported range (3.0–5.0 eV) for Fe-containing low-dimensional semiconductor systems and enables a more reliable description of electron localization, exchange splitting, and magnetic interactions induced by Fe incorporation [9,31–33]. The application of orbital-dependent Hubbard corrections was adopted to improve the treatment of localized electronic interactions and orbital hybridization effects governing the electronic states near the band edges. Since the electronic structure of SiC nanosheets is primarily influenced by hybridized Si-p/C-p states, the inclusion of Hubbard corrections enables a more realistic description of the electronic band structure compared with conventional LDA/GGA approaches, which are known to underestimate semiconducting band gaps. In Fe-doped systems, the Fe-3d orbitals play a dominant role in spin polarization, magnetic moment formation, and exchange interactions; therefore, the inclusion of the U parameter for Fe-3d orbitals is essential for accurately describing the magnetic and electronic behavior of the doped nanosheets. Accordingly, the selected Hubbard parameters were employed to achieve better agreement with previously reported theoretical and experimentally expected electronic characteristics of SiC-based nanostructures. The valence electronic configurations considered in the calculations were Si: [Ne] $3s^2 3p^2$, C: [He] $2s^2 2p^2$, and Fe: [Ar] $3d^6 4s^2$, corresponding to four valence electrons for Si and C atoms and eight valence electrons for Fe atoms.

A 4×4 hexagonal SiC nanosheet (SiCNSh) supercell, consisting of 80 atoms (40 Si + 40 C), was constructed to model the pristine system. To investigate the effect of Fe incorporation, one and two carbon atoms were substituted by Fe atoms, yielding single-Fe-doped ($\text{Si}_{40}\text{Fe}_1\text{C}_{39}$) and double-Fe-doped ($\text{Si}_{40}\text{Fe}_2\text{C}_{38}$) configurations, respectively. The relatively large supercell size minimizes artificial interactions between periodic dopant images and ensures a realistic representation of isolated impurity behavior. All atomic structures were fully optimized until the maximum residual force on each atom was below $0.01 \text{ eV } \text{\AA}^{-1}$, while the total-energy convergence threshold was set to 10^{-5} eV . Norm-conserving pseudopotentials and numerical atomic orbital (NAO) basis sets provided in QuantumATK were employed to describe electron-ion interactions. The Brillouin zone integration was performed using a $9 \times 9 \times 1$ Monkhorst-Pack k-point mesh [34], and an energy cutoff of 75 Ha was applied. To eliminate spurious interactions between periodic replicas along the out-of-plane direction, a vacuum spacing of 20 Å was introduced along the z-axis, while periodic boundary conditions were maintained in the in-plane directions. To evaluate the energetic and structural stability of the systems, total energies, residual forces, stress tensors, and formation energies were systematically analyzed. The thermodynamic feasibility of Fe substitution was assessed through formation-energy calculations according to:

$$E_{\text{form}} = E_{\text{Fe-SiCNSh}} - E_{\text{pristine}} + n\mu_{\text{C}} - n\mu_{\text{Fe}} \quad (1)$$

where $E_{\text{Fe-SiCNSh}}$ and E_{pristine} denote the total energies of doped and pristine SiC nanosheets, respectively, n represents the number of substituted carbon atoms, and μ_{C} and μ_{Fe} correspond to the chemical potentials of carbon and iron atoms.

The thermal stability of magnetic ordering was further assessed by estimating the Curie temperature (T_{C}), which represents the critical temperature separating the ferromagnetic and paramagnetic phases. In this work, T_{C} was evaluated within the mean-field

approximation (MFA), using the total-energy difference between the ferromagnetic (FM) and antiferromagnetic (AFM) configurations as a measure of the effective magnetic exchange interaction. The MFA expression is given as:

$$T_C = \frac{2}{3} \cdot \frac{\Delta E}{k_B} \quad (2)$$

where $\Delta E = E_{\text{AFM}} - E_{\text{FM}}$ is the energy difference between the AFM and FM states, and k_B is the Boltzmann constant (8.617×10^{-5} eV/K).

To further verify thermal stability under ambient conditions, ab initio molecular dynamics (AIMD) simulations were performed at 300 K using the canonical (NVT) ensemble with a Nosé–Hoover thermostat. The temporal evolution of total energy and structural integrity was monitored to evaluate the robustness of Fe-substituted SiC nanosheets against thermal fluctuations. The electronic properties of pristine and Fe-doped SiCNSh systems were investigated through spin-polarized band structure and total density of states (TDOS) calculations. Partial density of states (PDOS) analysis was additionally performed to identify the orbital contributions of Fe-3d, Si-3p, and C-2p states, particularly near the Fermi level, in order to elucidate the electronic modifications induced by Fe substitution.

To gain deeper insight into charge redistribution and chemical bonding characteristics, electron density difference (EED) analysis, Bader charge calculations, and Mulliken population analysis [35] were carried out. These analyses enable quantitative evaluation of charge transfer, bonding interactions, and local electronic rearrangements around Fe dopants and neighboring atoms. All calculations were performed for charge-neutral supercells without introducing additional vacancies or charge-compensation defects. In the present model, Fe atoms were incorporated through direct substitution of carbon atoms in the SiC nanosheet, thereby enabling the investigation of the intrinsic effect of substitutional Fe doping on the structural, electronic, and magnetic properties of the host lattice. After full geometry optimization, all investigated configurations remained energetically stable and exhibited no significant structural distortions or lattice instabilities. The favorable formation energies, successful convergence of the optimized structures, and structural robustness observed during the calculations indicate that explicit charge-compensation mechanisms are not required for the investigated Fe@C systems. Furthermore, the charge redistribution induced by Fe incorporation was naturally accommodated by the SiC host lattice and quantitatively analyzed using Bader charge analysis, Mulliken population analysis, and electron-density-difference calculations. Consequently, the intrinsic influence of Fe substitution could be systematically evaluated without the additional complexity associated with compensating defects. The magnetic behavior of the Fe-substituted SiC nanosheets was examined by calculating total and local magnetic moments together with spin density distributions. For double-Fe-doped systems, both ferromagnetic (FM) and antiferromagnetic (AFM) spin configurations were considered to determine the magnetic ground state. For the 2Fe@C-doped SiC nanosheet, two carbon atoms were replaced by Fe atoms at well-separated substitutional sites. The dopant positions were selected to maximize the Fe–Fe separation and to avoid direct Fe–Fe bonding, thereby enabling the investigation of magnetic interactions mediated by the SiC host lattice. Two different substitutional arrangements were considered during model construction, and both yielded qualitatively similar optimized structures and electronic characteristics. Consequently, one representative configuration was selected for the detailed analyses presented in this work.

Furthermore, the Curie temperature (T_C) was estimated using the mean-field approximation (MFA) based on the exchange-energy difference between FM and AFM states, providing insight into the thermal stability of long-range magnetic ordering. Thus, the adopted computational framework enables a comprehensive understanding of the rela-

relationship between Fe substitution, local bonding environment, charge redistribution, and magnetic exchange interactions in SiC nanosheets, thereby offering valuable insight into defect-engineered magnetism and spintronic functionality in two-dimensional semiconductor materials.

3. Results and Discussion

3.1. Structural Optimization of Fe@C-Doped SiCNSh

Structural stability is one of the most fundamental requirements for evaluating the physical viability and practical applicability of low-dimensional materials, particularly transition-metal-doped nanosystems intended for electronic and spintronic applications. Prior to investigating electronic and magnetic properties, it is essential to confirm that the proposed atomic configurations correspond to energetically favorable and mechanically stable states. In DFT-based materials modeling, structural optimization enables identification of the minimum-energy atomic arrangement through minimization of internal atomic forces and residual stress, thereby ensuring thermodynamic equilibrium and preventing artificial electronic or magnetic responses arising from metastable geometries.

The optimized atomic structures of pristine, single-Fe, and double-Fe substituted SiC nanosheets are presented in Figure 1. The pristine SiC nanosheet preserves its characteristic hexagonal arrangement, whereas Fe substitution at carbon sites induces localized structural distortions around the dopant region due to differences in atomic radius and bonding characteristics between Fe and C atoms. Compared to the single-Fe substituted system, the double-Fe configuration exhibits slightly more pronounced local lattice rearrangement. Nevertheless, the overall planar morphology and structural integrity of the SiC nanosheet remain well preserved after Fe incorporation, indicating that substitutional Fe doping does not induce severe lattice instability and suggesting favorable structural stability of the proposed nanosystems.

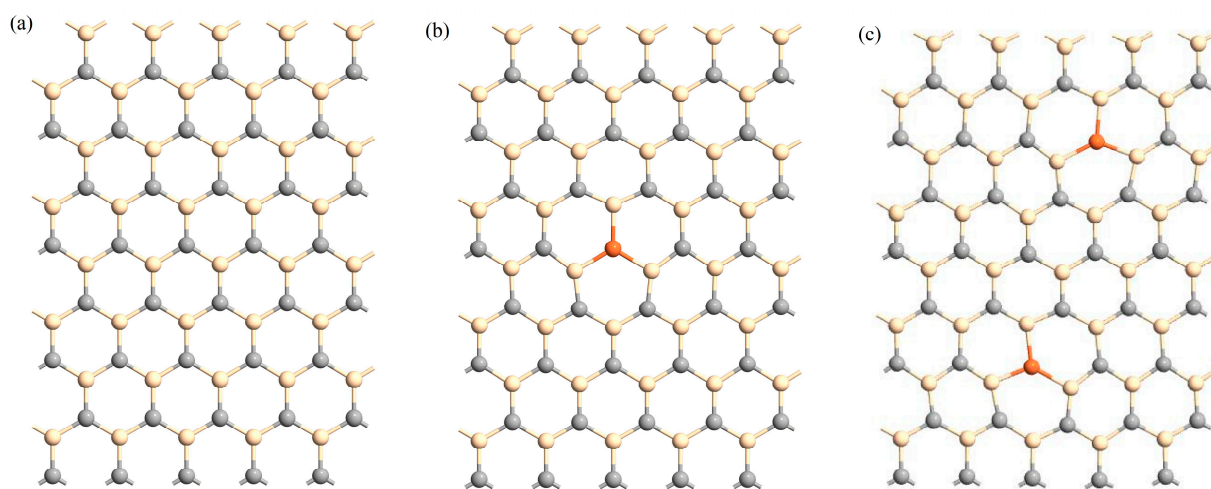


Figure 1. Optimized atomic structures of pristine (a), single-Fe (b), and double-Fe (c) substituted SiC nanosheets at C sites. Gray spheres represent C atoms, pale yellow (gold-colored) spheres represent Si atoms, and orange spheres represent Fe atoms.

Similar structural robustness upon transition-metal substitution has previously been reported for SiC-based low-dimensional systems [6,9,14,19,21]. Accordingly, full structural optimization of pristine and Fe-substituted SiC nanosheets was performed within the framework of spin-polarized density functional theory (DFT) to determine the most energetically favorable atomic configurations. During geometry relaxation, the evolution of total energy, atomic forces, and stress tensors was systematically monitored to ensure

complete convergence of both electronic and ionic subsystems. The gradual reduction of residual forces and stress values confirms the attainment of mechanical equilibrium, indicating that the optimized structures correspond to stable local minima on the potential-energy surface [4,9,14]. The convergence behavior of total energy during geometry optimization is shown in Figure S1 of the Supplementary Information. Initially, the total energy of the system was calculated as -614.33 eV and gradually decreased to approximately -618.50 eV within the first 20 optimization steps, after which the energy profile became nearly constant. This monotonic energy decrease indicates progressive structural relaxation toward a lower-energy and thermodynamically favorable configuration. From a microscopic perspective, the stabilization process is associated with local atomic rearrangement and optimization of the surrounding Si–C and Fe–C bonding environment. Furthermore, the flattening of the total-energy curve at later optimization stages confirms that the self-consistent convergence criteria were fully satisfied and that the optimized configuration reached structural equilibrium [6]. These findings provide strong evidence for the structural reliability and energetic stability of Fe-substituted SiC nanosheets, establishing a solid basis for subsequent electronic, charge-transfer, magnetic and thermal stability investigations.

Force convergence behavior during geometry optimization is illustrated in Figure S2 of the Supplementary Information. At the initial stage of structural relaxation, the atomic forces reach values as high as 9.60 eV/Å; however, they rapidly decrease and converge below 0.05 eV/Å after several optimization steps, indicating progressive structural stabilization and attainment of equilibrium atomic positions [9]. The near-zero residual force values demonstrate the absence of significant net mechanical stress acting on individual atoms, confirming the formation of a fully relaxed and energetically stable configuration. From a physical perspective, this behavior reflects the gradual minimization of the potential-energy gradient with respect to atomic displacements, thereby satisfying the Hellmann–Feynman convergence criterion for an optimized geometry. These results further support the structural reliability of the Fe-substituted SiC nanosheet and validate the suitability of the optimized geometry for subsequent electronic, magnetic, and thermal stability analyses.

Stress evolution and mechanical stability were evaluated through the variation of the stress tensor during structural optimization, as illustrated in Figure S3 of the Supplementary Information. The calculated stress values, expressed on the order of 10^{-5} eV/Å³, exhibit only minor fluctuations within the narrow range of approximately 1.60×10^{-5} to 1.63×10^{-5} eV/Å³ throughout the relaxation process. Such negligible variation indicates the absence of significant internal stress accumulation and confirms the preservation of mechanical equilibrium within the optimized system.

Furthermore, the nearly constant stress profile indicates that the SiC lattice effectively accommodates Fe-induced local distortions without generating significant residual strain, thereby preserving the mechanical equilibrium of the system. Combined with the monotonic decrease in total energy and the convergence of atomic forces toward near-zero values, these results confirm that the Fe-substituted SiC nanosheet reaches an energetically favorable, mechanically stable, and thermodynamically equilibrated configuration. Consequently, the optimized geometry provides a reliable foundation for subsequent investigations of electronic structure, projected density of states (PDOS), magnetic moment, Curie temperature, and thermal stability [4,14,21].

In general, the comparative analysis of pristine and Fe-doped SiCNShs demonstrates that Fe incorporation substantially alters both the electronic structure and energetic characteristics of the system. The energy contributions, including total, kinetic, electrostatic, exchange–correlation, and Hubbard energies, collectively highlight the superior energetic stability and strengthened electronic interactions of the 2Fe(2C)-doped SiCNSh compared to other investigated configurations. Notably, the enhanced exchange–correlation and Hub-

bard contributions indicate stronger magnetic interactions and improved spin-dependent behavior, emphasizing the magnetic robustness of the 2Fe(2C) system. The total energy and structural stability of pristine and Fe-doped (4×4) SiCNShs containing 80 atoms were systematically investigated using spin-polarized DFT within the LSDA + U framework. The pristine SiCNSh exhibits a total free energy of $-13,296.08$ eV, representing the energetically stable reference configuration of the undistorted Si–C network. The balanced electrostatic ($-12,981.93$ eV) and exchange–correlation (-6219.37 eV) energy contributions indicate stable bonding interactions and a homogeneous electronic distribution within the pristine lattice. Following substitution of a single Fe atom at a carbon site [1Fe(1C)], the total energy decreases significantly to $-13,890.91$ eV, indicating that Fe incorporation is thermodynamically favorable. The energetic stabilization can be attributed to the formation of strong Fe–C bonding interactions and enhanced orbital hybridization between Fe–3d and neighboring C–2p/Si–3p states. Simultaneously, the increase in kinetic energy from 5905.89 eV (pristine) to 6500.43 eV reflects enhanced electronic interaction and modification of charge-carrier dynamics following Fe incorporation.

The Hubbard correction term (-4.99 eV) further indicates moderate on-site electron correlation associated with localized Fe-derived electronic states and dopant-induced magnetic polarization. Upon substitution of two carbon atoms by Fe dopants [2Fe(2C)-FM], the system reaches the lowest total free energy ($-14,486.44$ eV), confirming this configuration as the most energetically favorable among all investigated models. The further reduction in electrostatic ($-14,721.55$ eV) and exchange–correlation (-6853.31 eV) energies suggests enhanced charge redistribution and strengthened Fe-mediated bonding interactions within the nanosheet.

In addition, the kinetic energy increases to 7094.03 eV, indicating stronger electronic interaction and more pronounced electronic reconstruction in the double-doped configuration. The larger magnitude of the Hubbard correction term (-5.61 eV) reflects enhanced electron correlation effects and stronger spin-dependent interactions between Fe centers, supporting stabilization of the ferromagnetic ground state. Overall, the progressive decrease in total energy from pristine to double-Fe-doped SiCNSh confirms that Fe incorporation substantially enhances energetic stability, with the 2Fe(2C)-FM configuration emerging as the most thermodynamically favorable system. These findings provide strong evidence that Fe doping effectively stabilizes the SiCNSh framework while simultaneously promoting electronic reconstruction and magnetic ordering, making the 2Fe(2C)-doped SiCNSh a promising candidate for future spintronic and nanoelectronic applications.

3.2. Defect Formation Energy

To evaluate the thermodynamic feasibility and energetic preference of Fe incorporation into the SiC nanosheet, the defect formation energy of Fe-substituted configurations was calculated. Defect formation energy is a key parameter for assessing the relative stability and experimental realizability of doped low-dimensional materials, since lower formation energies generally indicate a higher probability of defect formation under thermodynamic equilibrium conditions [4,14,21].

In the present study, Fe atoms were introduced substitutionally at carbon sites, and both single-Fe and double-Fe doped configurations were considered to identify the energetically most favorable defect arrangement. The corresponding formation energies were calculated according to Equation (1), presented in Section 2, to evaluate the thermodynamic feasibility and relative stability of Fe incorporation into the SiC nanosheet. According to Equation (1), $E_{\text{Fe-SiCNSh}}$ and E_{pristine} represent the total energies of the Fe-doped and pristine SiC nanosheets, respectively, while μ_{C} and μ_{Fe} denote the chemical potentials of carbon and Fe atoms. The parameter n corresponds to the number of substituted atoms.

Lower formation-energy values indicate energetically favorable defect incorporation and enhanced structural stability of the doped system.

The formation energy of the Fe-doped SiC nanosheet was calculated to evaluate the energetic feasibility of Fe incorporation into the host lattice. The negative value indicates that Fe substitution at the carbon site is energetically favorable and that the doped structure is thermodynamically stable after geometry optimization. Since Fe atoms were introduced substitutionally at carbon sites, the formation energy was determined by considering the removal of C atoms and the incorporation of Fe atoms.

The calculated total and formation energies for pristine SiCNSh, single-Fe-doped [1Fe(1C)], and double-Fe-doped [2Fe(2C)] configurations are summarized in Table 1. For the single-Fe-doped configuration, the formation energy was calculated as -7.44 eV/atom. The calculated formation energy of the double 2Fe(2C)-doped SiCNSh was found to be approximately -7.79 eV/atom, which is slightly lower than that of the single-Fe-doped configuration.

Table 1. Total and formation energy results of Fe-substituted SiC nanosheets.

System	Total Energy (eV)	Formation Energy (eV)
SiCNSh	$-13,296.07967$	—
Single Fe@C-doped SiCNSh	$-13,890.91176$	-7.44
Double Fe@C-doped SiCNSh	$-14,486.44170$	-7.79

This result confirms that double Fe incorporation is energetically more favorable and further enhances the stability of the SiCNSh framework, consistent with the total-energy analysis identifying the 2Fe(2C)-FM configuration as the thermodynamically most stable system among the investigated models. Moreover, the negative formation-energy values indicate that Fe atoms can be incorporated into the SiC nanosheet without inducing significant energetic instability, thereby supporting the structural feasibility and favorable thermodynamic stability of the proposed Fe-doped system.

3.3. Thermal Stability: AIMD Simulation

To further evaluate the thermal stability of the energetically favorable double Fe@C-doped SiCNSh, ab initio molecular dynamics (AIMD) simulations were performed at 300 K under the canonical (NVT) ensemble for 5 ps, as shown in Figure 2. A time step of 1 fs was employed, and the temperature was controlled using a Nosé–Hoover thermostat to mimic room-temperature conditions. The temporal evolution of the total energy and temperature during the AIMD simulation is presented in Figure 2. After an initial equilibration stage, both the total energy and temperature fluctuate around nearly constant average values without exhibiting any significant drift, confirming the thermal stability of the system at room temperature. Although small oscillations in these quantities are observed, such fluctuations are expected for low-dimensional nanosystems under finite-temperature conditions and remain within an acceptable range for thermodynamic stability. Importantly, no abrupt structural collapse, bond breaking, severe energy divergence, or irreversible atomic rearrangement was observed throughout the 5 ps AIMD simulation, indicating that the Fe@C-doped SiCNSh preserves its structural integrity and thermal stability. These results confirm the favorable thermal stability of the proposed configuration and support its potential applicability in nanoscale electronic and spintronic devices.

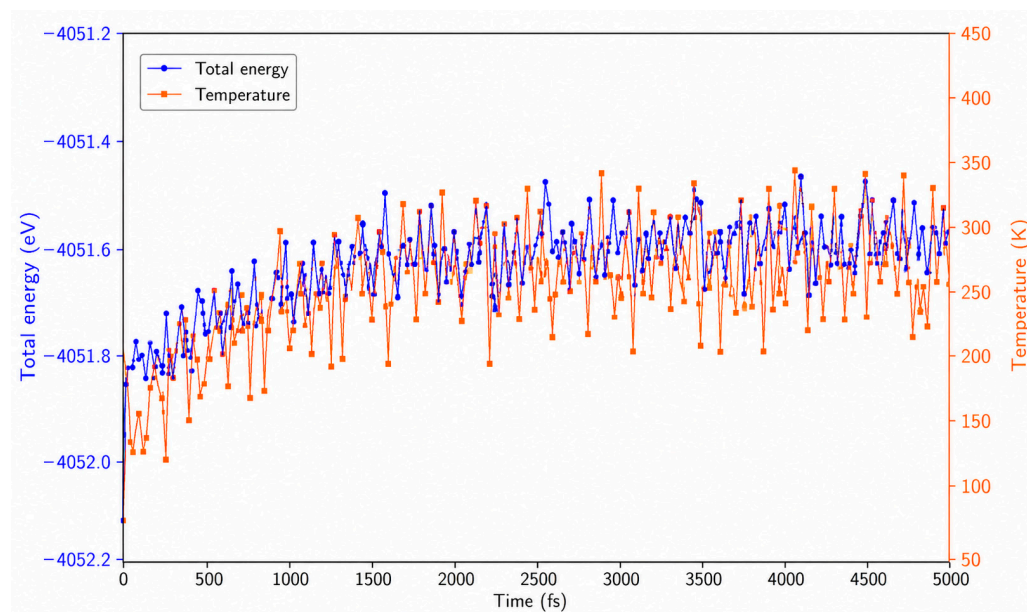


Figure 2. Variation of total energy and temperature during AIMD simulations at 300 K for Fe-doped SiC nanosheets.

The obtained AIMD results demonstrate that the double Fe@C-doped SiCNSh remains structurally stable under room-temperature conditions, exhibiting no significant structural degradation during the simulation. Such thermal robustness indicates the potential suitability of this nanosystem for future nanoscale electronic and spintronic applications.

3.4. Electron Density Redistribution and Charge Transfer Mechanism

To gain deeper insight into the electronic origin of magnetic and electronic modifications induced by Fe incorporation, the electron density redistribution in pristine and Fe-doped SiC nanosheets was investigated using electron difference density (EDD) analysis. The corresponding charge-density difference maps for single Fe-doped and double Fe-doped SiCNShs are presented in Figure 3. In these maps, positive values correspond to electron accumulation regions, whereas negative values indicate electron depletion. For the pristine SiC nanosheet, the charge redistribution remains relatively uniform throughout the lattice, reflecting the intrinsic covalent–ionic bonding nature of the Si–C framework.

As shown in Figure 3, electron density is predominantly localized around carbon atoms due to their higher electronegativity, while neighboring Si atoms exhibit relatively electron-deficient regions, confirming the polarized Si–C bonding network. This nearly periodic charge arrangement suggests an electronically balanced and nonmagnetic ground state of the pristine nanosheet. A substantial modification of the local electronic environment is observed after Fe substitution. In the single Fe-doped SiCNSh (Figure 3a), pronounced charge redistribution emerges around the dopant site, characterized by noticeable electron depletion near the Fe atom and enhanced electron accumulation around adjacent carbon atoms. This behavior indicates partial electron donation from Fe toward the surrounding lattice, resulting in strong local Fe–C hybridization. The redistribution pattern suggests significant overlap between Fe–3d and neighboring C–2p orbitals, which perturbs the local electronic symmetry and promotes spin polarization in the vicinity of the dopant atom. The induced charge asymmetry confirms that Fe incorporation strongly modifies the local bonding environment and electronic structure of the nanosheet. For the double Fe-doped configuration (Figure 3b), the electron redistribution becomes more spatially localized and pronounced around both Fe centers.

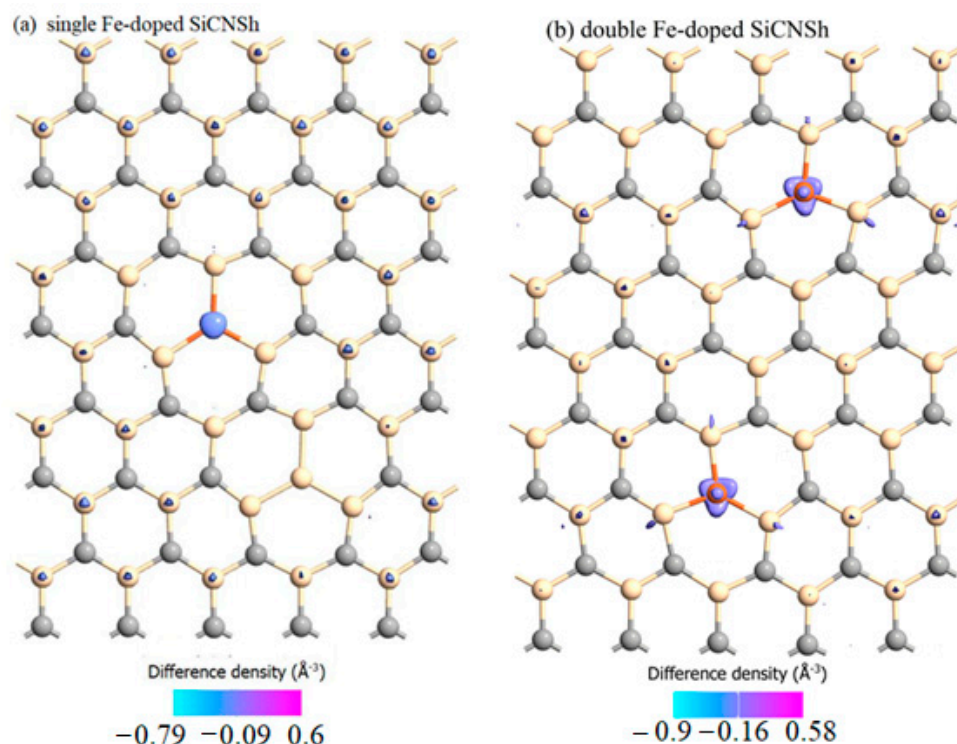


Figure 3. Electron density difference (EDD) maps of (a) single Fe-doped and (b) double Fe-doped SiC nanosheets. Gray spheres represent C atoms, pale yellow (gold-colored) spheres represent Si atoms, and orange spheres represent Fe atoms. Cyan and magenta regions indicate electron depletion and accumulation, respectively, revealing localized charge redistribution around Fe dopant atoms.

Compared with the single-doped system, stronger charge polarization is observed in the neighboring Si–C network, indicating enhanced electronic perturbation due to the cooperative interaction between Fe dopants. The overlapping redistribution regions around the Fe atoms imply increased Fe-mediated electronic coupling, which can facilitate magnetic exchange interactions through the surrounding lattice. Moreover, the preservation of electron accumulation around carbon atoms suggests that C atoms act as important mediators of charge transfer and magnetic communication between dopant sites.

Overall, the EDD analysis demonstrates that Fe doping induces highly non-uniform charge redistribution within the SiCNSh lattice, involving (i) electron donation from Fe atoms, (ii) electron accumulation around neighboring carbon atoms, (iii) local polarization of adjacent Si atoms, and (iv) enhanced Fe–C orbital hybridization. These electronic rearrangements play a crucial role in tailoring the magnetic and electronic characteristics of Fe-doped SiC nanosheets and support their potential applicability in spintronic and nanoscale electronic devices.

3.5. Bader Charge Analysis

To quantify the charge redistribution induced by Fe substitution within the 80-atom SiCNSh system, Bader charge analysis was performed. The calculated Bader electron populations indicate that carbon atoms possess average electron populations in the range of 7.9–8.1 e, whereas silicon atoms exhibit values of approximately 10.2–11.1 e. These populations correspond to effective charge differences of about -1.8 to -2.1 e for C atoms and $+3.0$ to $+3.5$ e for Si atoms, confirming the preservation of the intrinsic mixed ionic–covalent bonding character of the Si–C framework. The results indicate substantial electron transfer from Si atoms toward neighboring C atoms, which is consistent with the electronegativity

difference between Si and C and agrees with previously reported charge-transfer behavior in SiC-based low-dimensional systems.

Figure 4 illustrates the atom-resolved Bader charge difference distribution ($\Delta Q = N_{\text{Bader}} - Z$) for the Fe-doped 80-atom SiCNSh structure. A clear separation between the host atoms is observed: carbon atoms predominantly occupy the positive charge-difference region (approximately +1.7 to +2.1 e), while silicon atoms are distributed within the negative region (approximately -2.8 to -3.8 e). This well-defined charge partitioning further confirms the strong polarity of the Si–C network and reflects stable electronic charge transfer within the host lattice.

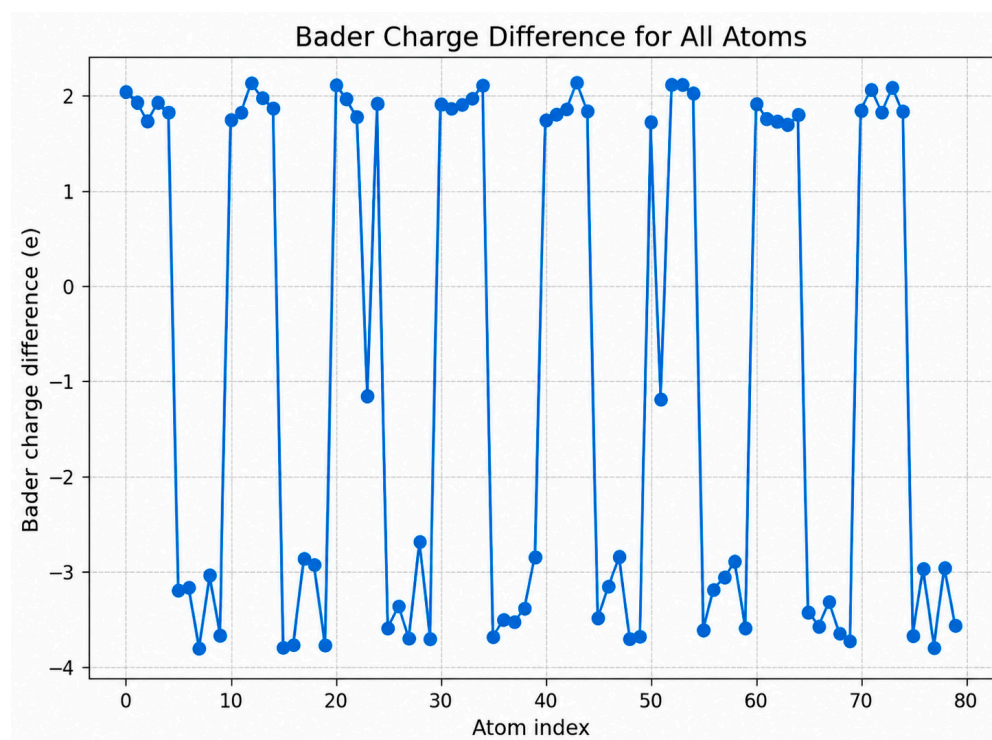


Figure 4. Bader charge difference as a function of atom index for the Fe-doped SiCNSh.

Importantly, the substitutional Fe dopant appears as a distinct localized state with a charge difference of approximately $\Delta Q \approx -1.20$ e, indicating partial electron depletion around the Fe center and suggesting that Fe behaves as a moderately cationic species inside the SiC host matrix. This behavior implies substantial electronic interaction between Fe-3d orbitals and neighboring C-2p/Si-3p states, promoting local charge redistribution and orbital hybridization near the defect region. Such dopant-induced electronic perturbation is expected to contribute significantly to the modified electronic and magnetic response of the Fe-doped SiCNSh, particularly through localized spin polarization and defect-mediated electronic states near the Fermi level.

The statistical distribution of the Bader charge differences for C, Si, and Fe atoms further clarifies the electronic environment within the Fe-doped SiCNSh system. Carbon atoms exhibit a relatively narrow and well-defined positive ΔQ distribution, indicating a consistent tendency toward electron accumulation across the SiC framework. In contrast, silicon atoms display a broader negative charge-difference range, reflecting stronger electron depletion and emphasizing their dominant role as electron donors in the intrinsic ionic-covalent Si–C bonding network. The substitutional Fe atom occupies an intermediate charge state between Si and C, exhibiting a significantly lower charge magnitude compared to the surrounding Si atoms. This behavior suggests reduced ionicity around the dopant site and points toward enhanced orbital overlap between Fe-3d, C-2p, and Si-3p states. Conse-

quently, Fe incorporation promotes a partial transition from the predominantly ionic Si–C bonding character toward a more localized covalent bonding environment, particularly in the vicinity of the substitutional defect.

To further examine the local electronic perturbation induced by Fe substitution, the atom-resolved Bader charge differences for the dopant and its nearest neighboring atoms are presented in Figure 5. The nearest carbon atoms exhibit slightly enhanced electron accumulation, with charge-difference values of approximately $\Delta Q \approx +2.03$ to $+2.12$ e, exceeding the average charge range observed for C atoms in the pristine lattice. This localized charge enrichment suggests stronger Fe–C electronic hybridization, reflecting increased orbital interaction near the defect region. Meanwhile, neighboring silicon atoms show noticeably reduced charge magnitudes compared with the average Si charge distribution. Such partial neutralization indicates localized redistribution of electron density caused by the Fe dopant and suggests weakening of the purely ionic Si–C interaction in favor of a more mixed covalent Si–C–Fe bonding configuration. These findings collectively demonstrate that Fe substitution not only perturbs the local charge landscape but also modifies the chemical bonding nature of the SiC nanosheet, potentially contributing to the observed changes in electronic and magnetic behavior.

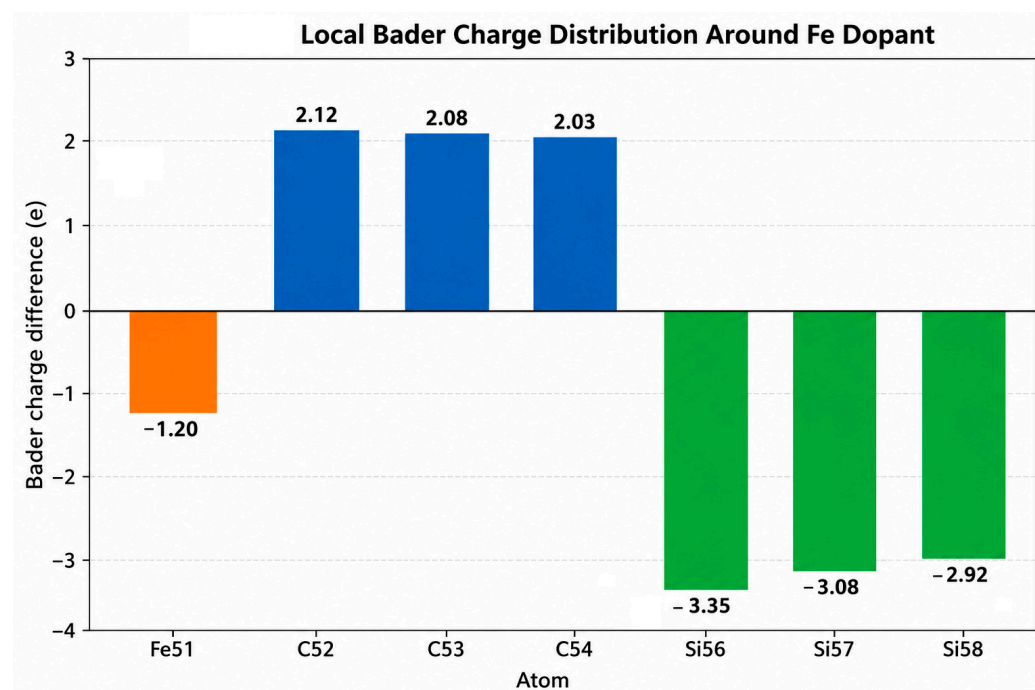


Figure 5. Local Bader charge distribution around Fe dopant in SiCNSh. Blue bars correspond to C atoms, green bars correspond to Si atoms, and the orange bar corresponds to the Fe dopant.

A spatial representation of the charge redistribution induced by Fe substitution is presented in Figure 6, which illustrates the two-dimensional Bader charge-difference map in the y – z plane. Each marker corresponds to an atomic site, while the color scale reflects the magnitude and direction of the charge difference relative to the neutral atomic state. A clear charge separation is observed throughout the SiC framework, where electron-enriched C atoms predominantly occupy the positive ΔQ region and electron-depleted Si atoms are distributed within the negative region, preserving the intrinsic ionic–covalent nature of the Si–C network. Importantly, the substitutional Fe dopant introduces localized charge perturbation, resulting in an asymmetric charge environment around the defect region. The neighboring atomic sites exhibit noticeable variations in charge magnitude compared with the bulk-like regions, indicating localized electronic polarization and redistribution.

of electron density induced by Fe incorporation. The spatial non-uniformity around the dopant site further suggests enhanced Fe–host orbital hybridization, particularly involving Fe–3d and neighboring C–2p/Si–3p states. These findings provide direct spatial evidence for the defect-mediated modification of the local electronic environment and are consistent with the Bader population trends discussed in Figures 4–6. The quantitative Bader charge-transfer characteristics of the substitutional Fe dopant and its nearest neighboring atoms are summarized in Table 2. The obtained values reveal enhanced charge accumulation around adjacent C atoms and partial charge neutralization of nearby Si atoms, confirming localized electronic redistribution induced by Fe incorporation within the SiCNSh framework.

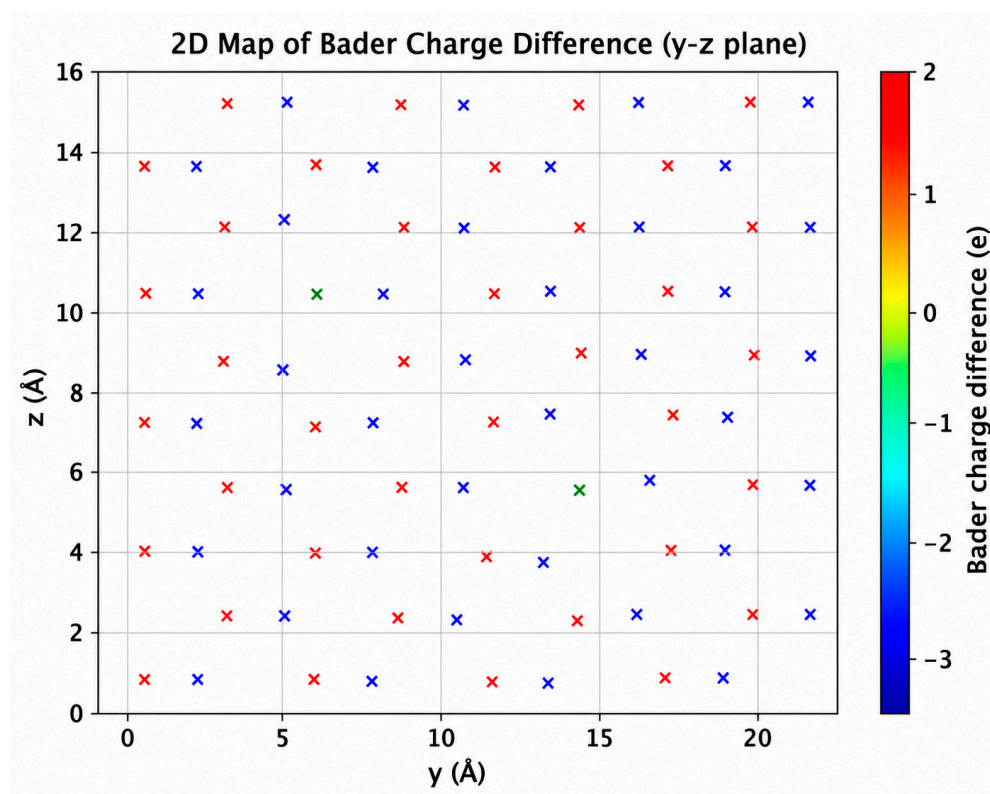


Figure 6. Two-dimensional Bader charge-difference map in the y–z plane for the Fe-doped 80-atom SiCNSh, illustrating spatial charge redistribution and localized electronic perturbation around the substitutional Fe dopant.

Table 2. Local Bader charge transfer characteristics of the substitutional Fe dopant and nearest neighboring atoms in Fe-doped SiCNSh.

Atom Type	Atom Number	Bader Charge Difference, ΔQ (e)	Electronic Behavior
Fe	51	−1.20	Moderately electron-depleted
C	52	+2.12	Electron-rich
C	53	+2.08	Electron-rich
C	54	+2.03	Electron-rich
Si	56	−3.35	Electron-depleted
Si	57	−3.08	Electron-depleted
Si	58	−2.92	Electron-depleted

Thus, the combined Bader charge analyses presented in Figures 4–6 reveal that Fe incorporation induces pronounced local charge redistribution within the SiCNSh framework. The substitutional Fe dopant promotes electron-density rearrangement around neighboring

atomic sites, characterized by enhanced charge accumulation on adjacent C atoms and partial neutralization of nearby Si atoms. These changes indicate increased covalent contributions within the Si–C–Fe bonding environment and highlight dopant-induced local electronic polarization. Consequently, charge redistribution emerges as a key mechanism governing the modification of the electronic structure, magnetic response, and potential spintronic functionality of Fe-doped SiCNSh systems.

3.6. Electronic Properties of Pristine SiCNSh

To establish a reliable reference framework for understanding dopant-induced modifications in the electronic and magnetic behavior of SiC nanosheets, the intrinsic electronic properties of pristine SiCNSh were first systematically investigated using density functional theory (DFT) calculations. All simulations were performed within the QuantumATK package employing spin-polarized calculations to accurately evaluate the magnetic ground state and electronic band characteristics of the system. A 4×4 hexagonal SiC supercell containing 80 atoms (40 Si and 40 C atoms) was considered to represent the undoped nanosheet configuration.

The calculated spin-polarized electronic band structures for both spin-up and spin-down channels are presented in Figure 7. As clearly observed, the spin-up and spin-down bands exhibit complete overlap throughout the entire Brillouin zone, indicating the absence of spin asymmetry in the electronic states. This perfect coincidence confirms that pristine SiCNSh stabilizes in a nonmagnetic ground state, with no spontaneous spin polarization or net magnetic moment.

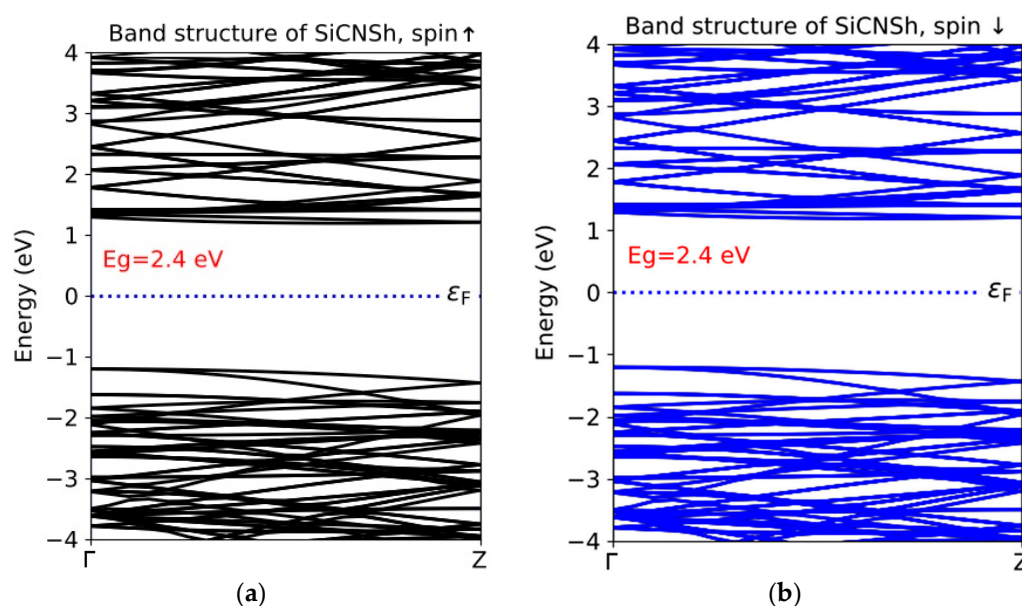


Figure 7. Spin-polarized electronic band structures of pristine SiCNSh for (a) spin-up and (b) spin-down channels. The complete overlap of both spin states and the finite band gap (~ 2.4 eV) confirm the nonmagnetic semiconducting nature of the pristine nanosheet.

Furthermore, the electronic band structure demonstrates a distinct separation between the valence band maximum (VBM) and conduction band minimum (CBM), confirming the semiconducting nature of the pristine SiCNSh. The calculated band gap is approximately 2.4 eV, as illustrated in Figure 7, with the Fermi level positioned at 0 eV. The absence of electronic states crossing the Fermi level further supports the semiconducting behavior of the nanosheet. The relatively moderate band gap suggests that pristine SiCNSh possesses favorable electronic characteristics for nanoscale semiconducting applications.

while maintaining electronic stability. For further validation of the adopted computational methodology, the calculated band-gap values of pristine SiC nanosheets were compared with previously reported theoretical results available in the literature. The corresponding comparison is presented in Table 3. As summarized in Table 3, the band-gap values reported for pristine SiC nanosheets vary considerably depending on the computational methodology employed. Previous GGA-PBE calculations yielded band gaps ranging from 2.50 to 3.76 eV, while HSE06 calculations predicted values of 3.38–3.43 eV. The LSDA + U band gap obtained in the present work (2.39–2.44 eV) is in good agreement with previously reported first-principles results, whereas the additional HSE06 calculation yielded a band gap of 2.91 eV, which lies within the range reported by previous hybrid-functional studies.

Table 3. Comparison of structure parameters and band gaps of pristine SiC nanosheets reported in previous studies and the present work. For entries containing two values, the indirect and direct band gaps are reported as E_g (indirect)/ E_g (direct), respectively.

System	Method	Si–C Bond Length (Å)	E_g (eV)	Reference
SiCNSh	FP-LAPW/GGA-PBE	—	2.51	[36]
SiCNSh	GGA-PBE	1.78	3.76	[37]
SiCNSh	GGA-PBE	1.78	2.50	[38]
SiCNSh	HSE06	—	3.38/3.43	[39]
SiCNSh	LSDA + U	1.84	2.39/2.44	Present work
SiCNSh	HybridGGA-HSE06	1.84	2.91	Present work

Overall, these comparisons support the reliability of the adopted computational methodology for describing the electronic structure of pristine SiC nanosheets. In addition to the electronic-structure comparison, the structural reliability of the pristine SiC nanosheet was also assessed. The initial model was constructed using a Si–C bond length of 1.82 Å, while full geometry optimization within the LSDA + U framework yielded an equilibrium Si–C bond length of 1.84 Å. This value is reasonably close to previously reported GGA-PBE results (~1.78 Å), indicating that the adopted computational approach preserves the essential structural characteristics of the SiC nanosheet while providing a reliable description of its electronic properties.

It should be noted that direct comparison with previously reported Fe-doped SiC nanosheets remains limited because most available studies considered Fe substitution at Si sites, different defect configurations, or did not report quantitative band-gap values for Fe@C systems. In contrast, the present work focuses specifically on substitutional Fe incorporation at carbon sites (Fe@C), for which electronic-structure data remain scarce in the literature. Therefore, the comparison presented in Table 3 is primarily intended to assess the consistency and reliability of the adopted computational methodology for the pristine SiC framework rather than to establish a direct correspondence between different Fe-doped defect models.

Importantly, the identical spin distributions observed in both spin channels demonstrate that the electronic structure remains completely spin-degenerate, implying that magnetic ordering is intrinsically absent in the pristine system. This result is fully consistent with the expected electronic nature of defect-free SiC-based two-dimensional nanostructures, where strong covalent Si–C bonding preserves charge balance and suppresses localized magnetic moments. Therefore, the pristine SiCNSh serves as an essential electronic reference system for evaluating the influence of Fe incorporation and defect engineering on spin polarization, band-gap modulation, and magnetic phase evolution. Any deviation from this spin-degenerate semiconducting state after Fe substitution can thus be directly

attributed to dopant-induced electronic hybridization, local charge redistribution, and exchange interactions within the Si–C lattice.

Further insight into the intrinsic electronic behavior of pristine SiCNSh was obtained from the spin-polarized total density of states (TDOS) and partial density of states (PDOS), as presented in Figure 8. The calculated TDOS exhibits a clear symmetry between the spin-up and spin-down channels over the entire investigated energy range, indicating complete spin degeneracy and confirming the absence of spontaneous magnetic ordering in the pristine nanosheet. No spin splitting is observed near the Fermi level, further validating the nonmagnetic ground-state nature of the undoped SiCNSh. The TDOS profile shows a pronounced depletion of electronic states around the Fermi level ($E_F = 0$ eV), where the density of states approaches nearly zero for both spin orientations. This feature confirms the existence of a finite electronic band gap and is fully consistent with the semiconducting character observed from the band-structure calculations. The absence of electronic states crossing the Fermi level indicates that pristine SiCNSh possesses an electronically stable semiconducting configuration without metallic conduction pathways.

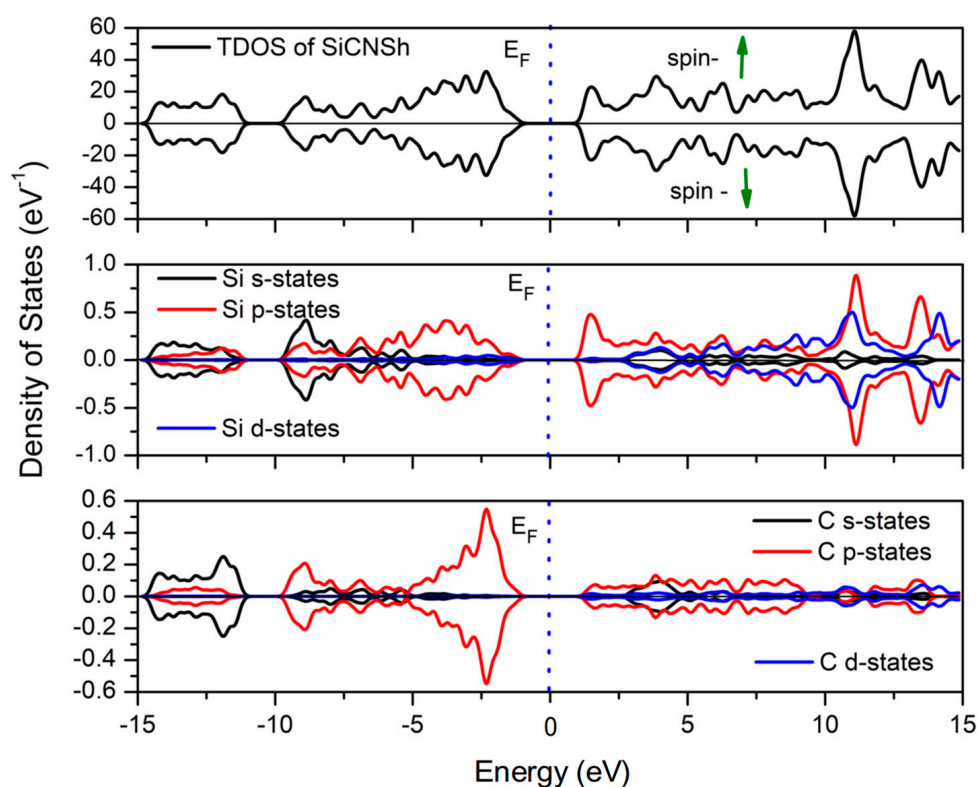


Figure 8. Spin-polarized TDOS and orbital-resolved PDOS of pristine SiCNSh. Contributions from Si and C s-, p-, and d-orbitals are presented for both spin channels. The symmetric spin distribution and near-zero DOS at the Fermi level ($E_F = 0$ eV) confirm the nonmagnetic semiconducting nature of pristine SiCNSh.

To clarify the orbital origin of the electronic states, orbital-resolved PDOS analysis was performed for both Si and C atoms. As illustrated in Figure 8, the electronic states near the valence- and conduction-band edges are predominantly governed by p-orbital contributions, particularly from Si-p and C-p states, highlighting strong orbital hybridization within the Si–C framework. In contrast, the contributions from s and d orbitals remain comparatively weak near the Fermi region, indicating their limited influence on the low-energy electronic transport characteristics of the pristine nanosheet.

Notably, the valence-band region below the Fermi level is mainly dominated by C-p states, whereas significant Si-p contributions become increasingly pronounced in the conduction-band region. This behavior suggests substantial Si-C covalent hybridization, which governs the electronic band topology and contributes to the intrinsic semiconducting stability of the nanosheet. The strong p-p orbital interaction between neighboring Si and C atoms plays a decisive role in maintaining charge balance and suppressing localized magnetic moments in the pristine structure. Overall, the TDOS and PDOS analyses consistently demonstrate that pristine SiCNSh is an intrinsically nonmagnetic semiconductor, characterized by spin symmetry, a finite band gap, and p-orbital-dominated electronic states. These results provide a fundamental electronic reference for understanding how Fe incorporation modifies orbital hybridization, induces spin asymmetry, and alters the electronic and magnetic properties of Fe-doped SiCNSh systems.

The calculated band gap of pristine SiCNSh was found to be approximately 2.4 eV, confirming its intrinsic semiconducting nature. This result is in good agreement with previous first-principles studies on pristine silicon carbide nanosheets. For instance, Houmad et al. reported a band gap of 2.51 eV for pristine SiC nanosheets using the full-potential linearized augmented plane wave (FP-LAPW) method implemented in the WIEN2k package within the GGA-PBE approximation [36]. To further assess the reliability of the electronic-structure calculations, an additional HSE06 hybrid-functional calculation was performed for the pristine SiCNSh, yielding a band gap of 2.91 eV. Although the HSE06 value is larger than the LSDA + U result, both approaches consistently predict a semiconducting ground state, supporting the robustness of the adopted computational methodology. The close agreement between the present result and the reported value supports the reliability of the adopted computational framework and demonstrates the capability of the employed LSDA + U methodology to accurately describe the electronic structure of SiC nanosheets. The minor deviation between the calculated band-gap values may reasonably originate from differences in the computational methodology, exchange–correlation treatment, basis formalism, and the inclusion of Hubbard U corrections in the present calculations. Overall, both studies consistently verify the intrinsic nonmagnetic semiconducting character of pristine SiC nanosheets.

3.7. Electronic Properties of Fe-Doped SiCNShs

This section provides an in-depth analysis of the electronic and magnetic properties of an Fe-doped silicon carbide nanosheet (Fe-SiCNSh), in which a carbon (C) atom is substitutionally replaced by an iron (Fe) atom. The computational model was constructed using an 80-atom hexagonal SiCNSh supercell, where one C atom was substituted by Fe to simulate transition-metal doping effects. Such substitutional doping strategies have been widely reported as an effective approach to tailor the electronic structure and induce magnetic ordering in SiC-based low-dimensional nanostructures [6,14,36]. The incorporation of Fe introduces localized perturbations in the electronic charge distribution and modifies the spin-dependent potential landscape of the SiC lattice.

The electronic structure of the Fe-doped system was investigated using spin-polarized density functional theory (DFT) within the LSDA + U formalism to accurately describe the strong on-site Coulomb interactions associated with partially occupied Fe-3d orbitals. The calculated spin-polarized band structures for the spin-up and spin-down channels are shown in Figure 9. Compared with pristine SiCNSh, Fe incorporation induces substantial modifications in the electronic states near the Fermi level, resulting in pronounced spin-dependent electronic behavior, in agreement with previous theoretical studies on transition-metal-doped SiC nanosystems [6,9,13,14].

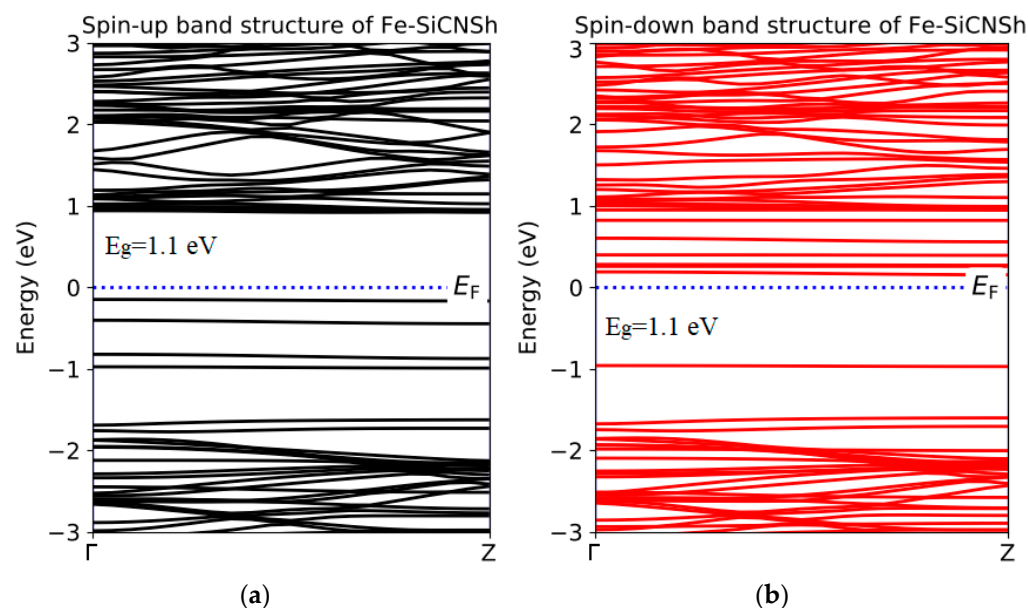


Figure 9. Spin-polarized electronic band structures of Fe-doped SiCNSH for (a) spin-up and (b) spin-down channels. Fe incorporation induces significant modification of the electronic structure, reducing the band gap to approximately 1.1 eV and introducing spin-dependent electronic behavior around the Fermi level.

As illustrated in Figure 9, both spin channels preserve a finite band gap of approximately 1.1 eV, which is considerably reduced compared with pristine SiCNSH (2.4 eV). This pronounced band-gap narrowing indicates that Fe incorporation strongly modifies the electronic band topology through orbital hybridization between Fe-3d and neighboring Si/C p-states. The reduced band gap suggests enhanced electronic conductivity and improved charge transport characteristics in the doped nanosheet. Importantly, although the system remains semiconducting in both spin channels, the spin-up and spin-down band structures are no longer perfectly overlapped, indicating the emergence of spin asymmetry induced by Fe substitution. The Fe dopant introduces localized electronic states close to the Fermi level, which alter the exchange interactions and contribute to magnetic moment formation in the system. Similar spin-dependent band modulation induced by transition-metal doping has previously been reported for SiC-based nanosheets and nanotube systems [6,13,19,21].

The electronic behavior of Fe-doped SiCNSH obtained in the present work was further compared with previous first-principles studies on transition-metal-doped SiC nanosheets. For example, Houmad et al. reported that Fe substitution in SiC nanosheets introduces localized Fe-3d states near the Fermi level, while preserving a nearly spin-symmetric electronic structure, indicating a predominantly nonmagnetic behavior [36]. In contrast, the present study adopts a fundamentally different substitutional model, where Fe is introduced at the carbon site (Fe@C) rather than substituting a silicon atom, as considered in Ref. [36]. Furthermore, Javan et al. demonstrated that Fe incorporation in monolayer SiC significantly modifies the electronic structure through the appearance of spin-polarized impurity states near the Fermi level [37]. Consistent with these findings, the present LSDA + U calculations reveal pronounced spin asymmetry and Fe-derived electronic states around the Fermi level arising from strong Fe-3d/C-2p/Si-3p orbital hybridization. These results confirm that the electronic response of Fe-doped SiC nanosheets is highly sensitive to the dopant configuration, substitutional site, and treatment of electron-correlation effects.

Such a difference in the dopant position significantly alters the local chemical environment and orbital interactions surrounding the Fe atom. As a consequence, Fe substitution at the carbon site induces pronounced modifications in the electronic structure of SiCNSH,

leading to a substantial band-gap reduction from 2.4 eV in pristine SiCNSh to approximately 1.1 eV in the Fe-doped configuration. This band-gap narrowing suggests that Fe incorporation introduces additional electronic states near the band edges and strongly modifies the intrinsic semiconducting character through Fe-3d and neighboring Si/C-p orbital hybridization.

More importantly, unlike the nearly symmetric spin behavior reported by Houmad et al. [36], the calculated spin-polarized band structures in the present work reveal a clear breaking of spin symmetry between the spin-up and spin-down channels. The absence of complete overlap between the two spin states indicates that Fe@C substitution induces spin-dependent electronic reconstruction, resulting in stronger exchange interactions and enhanced spin polarization within the SiC lattice. This behavior demonstrates that the electronic and magnetic response of SiCNSh is highly sensitive to the substitutional site of the Fe dopant, where carbon-site substitution produces significantly different spin characteristics compared with silicon-site substitution. Furthermore, the observed differences may also partly originate from the distinct computational methodologies employed, namely FP-LAPW/GGA-PBE in Ref. [36] and the LSDA + U approach adopted in the present work, where Hubbard corrections were included to better describe localized Fe-3d, Si-d, and C-p electronic interactions.

Further insight into the electronic reconstruction induced by Fe substitution was obtained from the spin-polarized total density of states (TDOS) and partial density of states (PDOS) analyses, as presented in Figure 10. In contrast to pristine SiCNSh, which exhibits complete spin symmetry and a nonmagnetic semiconducting ground state, the Fe-doped configuration shows a pronounced spin asymmetry between spin-up and spin-down channels, indicating the breaking of spin degeneracy induced by Fe incorporation. Similar transition-metal-induced modifications in SiC-based low-dimensional systems have been reported previously [6,13,14,36].

As shown in Figure 10a, the TDOS of Fe-doped SiCNSh exhibits a clear imbalance between spin-up and spin-down states throughout the investigated energy range, particularly in the vicinity of the Fermi level ($E_F = 0$ eV). Unlike pristine SiCNSh, where the DOS remains nearly zero around the Fermi region, Fe substitution introduces additional electronic states near the band edges, substantially modifying the intrinsic electronic structure. Nevertheless, a finite band gap of approximately 1.1 eV remains preserved in both spin channels, indicating that the system retains its semiconducting nature despite significant electronic reconstruction.

The pronounced reduction of the band gap from 2.4 eV in pristine SiCNSh to 1.1 eV after Fe doping demonstrates that Fe incorporation strongly perturbs the electronic band topology. This band-gap narrowing can be attributed to the emergence of dopant-induced impurity states arising from hybridization between Fe-3d orbitals and neighboring Si/C p-states, which shifts electronic states closer to the Fermi level and enhances spin-dependent interactions. The orbital-resolved PDOS presented in Figure 10b,c reveals that the electronic states near the Fermi level are predominantly governed by Fe-related contributions and hybridized Si-p/C-p orbitals. In particular, the chemically bonded silicon atoms surrounding the Fe dopant exhibit clear spin polarization, confirming that Fe substitution induces localized magnetic perturbations extending into the neighboring Si environment through Fe-Si/C orbital coupling. The observed spin imbalance in the Si-projected DOS indicates that magnetic interactions are not solely localized on the Fe atom but partially transferred to adjacent atoms through exchange-mediated hybridization.

The present findings were further compared with the previous first-principles study of Houmad et al. [36], who investigated Fe-doped SiC nanosheets using the FP-LAPW/GGA-PBE framework and reported nearly spin-symmetric electronic behavior with Fe-induced

states localized around the Fermi level. However, unlike Ref. [36], where Fe substitution was introduced at the silicon site, the present work considers Fe substitution at the carbon site (Fe@C). This difference in substitutional configuration substantially alters the local electronic environment surrounding the dopant atom and leads to stronger orbital hybridization and exchange interactions. Consequently, the present Fe@C-doped SiCNSh exhibits pronounced spin asymmetry and stronger spin-dependent electronic reconstruction, accompanied by a substantial reduction in band gap.

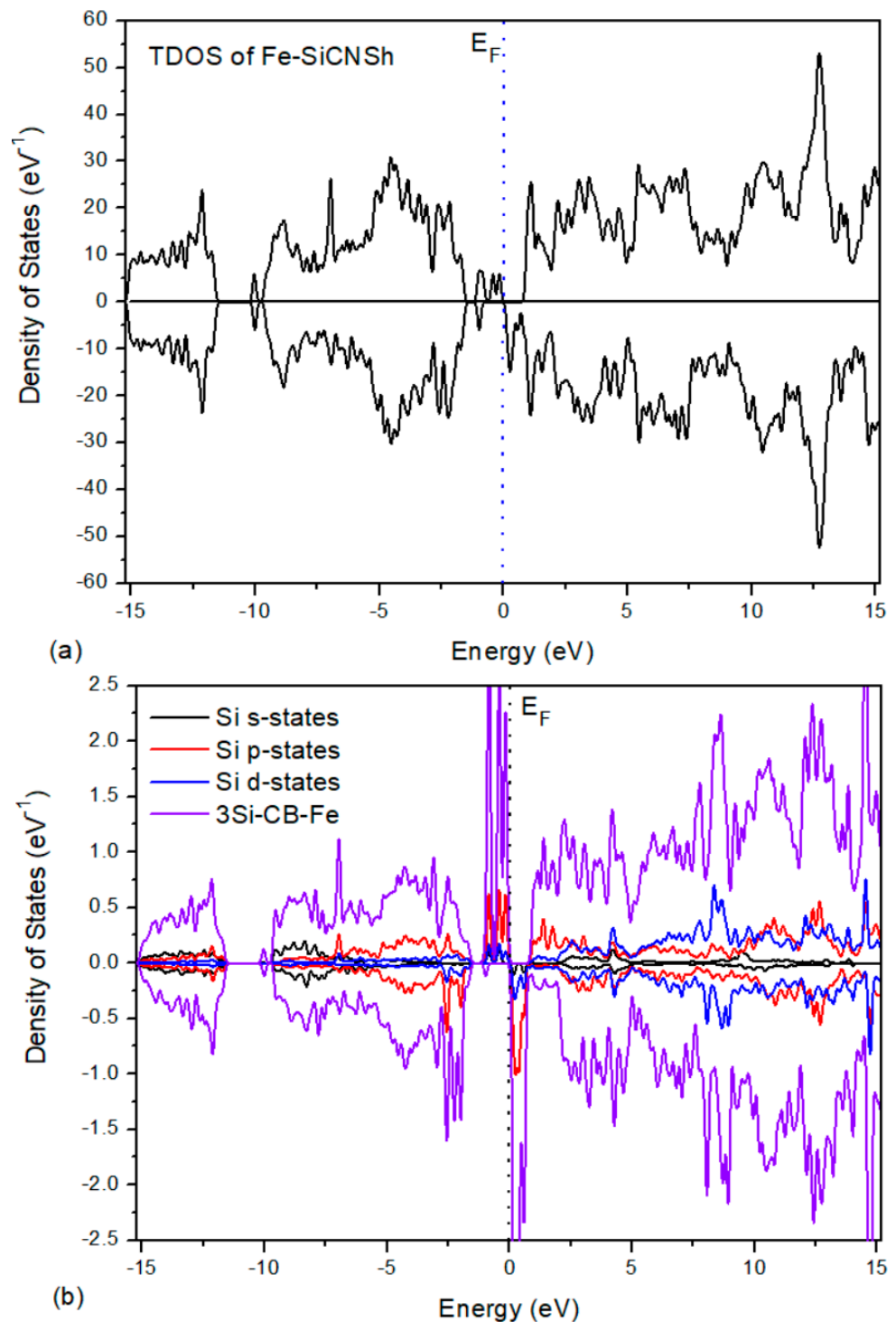


Figure 10. Cont.

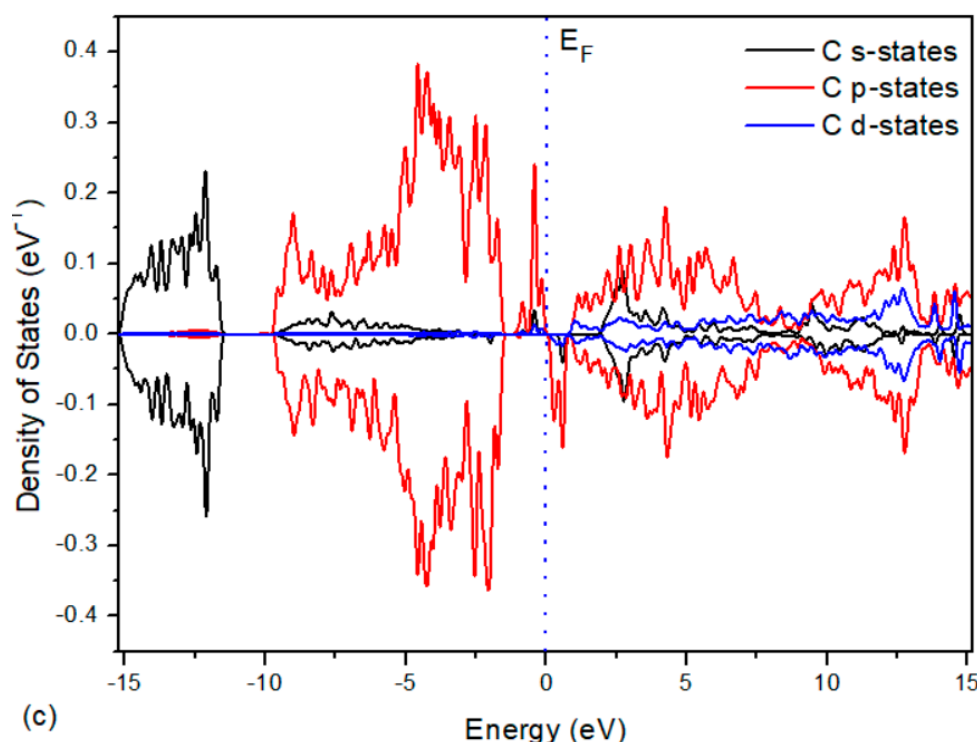


Figure 10. Total and partial density of states (TDOS/PDOS) of pristine and Fe-doped SiCNShs: (a) TDOS of the Fe-doped SiC nanosheet (Fe-SiCNSh), (b) PDOS of Si atoms and the Fe dopant, and (c) PDOS of C atoms. Spin-polarized partial density of states corresponding to the three Si atoms chemically bonded to Fe is presented to elucidate the Fe–Si hybridization mechanism and its contribution to the electronic structure modification.

The incorporation of a Fe dopant at the carbon substitutional site (Fe@C) induces pronounced modifications in the electronic and magnetic landscape of the two-dimensional SiCNSh. To elucidate the dopant-driven electronic reconstruction and spin-dependent behavior, the Fe@C configuration was systematically investigated using the spin-polarized DFT–LSDA + U framework, which enables an accurate treatment of localized electronic interactions associated with transition-metal impurities. Fe substitution fundamentally alters the intrinsic electronic character of pristine SiCNSh, resulting in a substantial band-gap reduction from 2.4 eV to approximately 1.1 eV. This narrowing of the electronic gap originates from the emergence of Fe-induced impurity states and enhanced orbital hybridization between Fe–3d and neighboring Si/C p-states, which collectively reshape the electronic states near the Fermi level. More importantly, the complete spin degeneracy observed in pristine SiCNSh is disrupted after Fe incorporation, as evidenced by the pronounced spin asymmetry between spin-up and spin-down electronic states. Such spin splitting reflects the formation of localized magnetic interactions and indicates the onset of dopant-mediated exchange coupling within the SiC framework. Collectively, the spin-polarized band structure, TDOS, and PDOS analyses demonstrate that Fe@C substitution serves as an effective route for engineering the electronic structure of SiCNSh, simultaneously enabling band-gap modulation, spin-symmetry breaking, and enhanced spin-dependent electronic interactions. These combined features position Fe-doped SiCNSh as a promising platform for next-generation spintronic and nanoelectronic technologies, where tunable electronic transport and spin manipulation are essential.

Table 4 summarizes the evolution of the electronic characteristics of pristine and Fe-substituted SiCNShs. The pristine Si₄₀C₄₀ nanosheet exhibits a semiconducting nature with an identical spin-up and spin-down band gap of 2.40 eV, confirming spin degeneracy in

the absence of magnetic dopants. Upon single Fe substitution ($\text{Si}_{40}\text{Fe}_1\text{C}_{39}$), the band gap decreases significantly to 1.10 eV in both spin channels, indicating a substantial modification of the intrinsic electronic structure while preserving semiconducting behavior.

Table 4. Comparative analysis of the electronic and magnetic properties of pristine and Fe-doped SiCNShs.

Compound	Band Gap (eV)		Electronic Nature
	Spin-Up	Spin-Down	
$\text{Si}_{40}\text{C}_{40}$	2.4	2.4	Semiconducting
$\text{Si}_{40}\text{Fe}_1\text{C}_{39}$	1.1	1.1	Semiconducting
$\text{Si}_{40}\text{Fe}_2\text{C}_{38}$	0.53	0.51	Semiconducting

Further Fe incorporation in the double-doped configuration ($\text{Si}_{40}\text{Fe}_2\text{C}_{38}$) results in an additional reduction of the band gap to approximately 0.53/0.51 eV for spin-up/spin-down states, respectively, revealing pronounced electronic reconstruction and the emergence of a narrow-gap semiconducting state. These findings demonstrate that Fe incorporation effectively tunes the electronic structure of SiCNShs through progressive band-gap narrowing, thereby enhancing their potential for spin-dependent nanoelectronic and optoelectronic applications.

To the best of our knowledge, direct experimental band-gap data for Fe-doped two-dimensional SiC nanosheets comparable to the present Fe@C model are not currently available in the literature. Most previous studies have focused on bulk SiC polytypes or Fe-doped SiC systems with different substitutional configurations, particularly Fe incorporation at Si sites. Therefore, the present results were primarily compared with available theoretical studies, while the magnetic properties were additionally evaluated against the limited Fe@C-doped SiC nanosheet data reported in the literature, showing good overall agreement.

3.8. Magnetic Properties and Curie Temperature

The magnetic behavior of Fe-doped SiCNShs was systematically investigated through spin-polarized DFT + U calculations combined with Mulliken spin population analysis. The obtained results reveal that substitutional Fe incorporation at the carbon site induces pronounced magnetization in the SiCNSh lattice, transforming the initially nonmagnetic pristine nanosheet into a magnetically active semiconducting system. For the single-Fe substituted configuration ($\text{Si}_{40}\text{Fe}_1\text{C}_{39}$), the magnetic response is predominantly governed by the Fe dopant, which exhibits a substantial localized magnetic moment of approximately $4.08 \mu_B$, originating mainly from partially occupied Fe-3d orbitals. This large local moment confirms that Fe acts as the principal magnetic center in the nanosheet, strongly perturbing the surrounding electronic environment. Simultaneously, neighboring lattice atoms develop induced magnetic polarization due to Fe-mediated orbital hybridization. In particular, the nearest-neighbor Si atoms chemically bonded to Fe display pronounced spin moments, reaching approximately $0.36\text{--}0.37 \mu_B$, indicating efficient spin-density transfer from the Fe center into the host lattice. Carbon atoms surrounding the dopant also exhibit smaller but non-negligible magnetic moments ($\sim 0.05\text{--}0.07 \mu_B$), confirming the participation of Fe-C and Fe-Si interactions in the magnetic exchange mechanism. Consequently, the overall supercell attains a total magnetic moment of approximately $6.00 \mu_B$, demonstrating robust dopant-induced ferromagnetic ordering.

The spin-polarized electronic structure further supports the emergence of magnetic semiconducting behavior in Fe-doped SiCNShs. Unlike the pristine nanosheet, which exhibits complete spin degeneracy, Fe substitution induces a clear asymmetry between spin-up and spin-down electronic states near the Fermi level. Nevertheless, both spin channels

preserve finite band gaps, indicating that the Fe-doped systems maintain a semiconducting nature rather than half-metallicity. Specifically, the band gap decreases from 2.40 eV in pristine SiCNSh to approximately 1.10 eV for the single-Fe substituted structure, while further Fe incorporation reduces the gap to nearly 0.53/0.51 eV for spin-up/spin-down channels, respectively. This progressive band-gap narrowing, together with enhanced spin polarization, highlights the strong influence of Fe-3d and neighboring Si/C orbital hybridization on the electronic reconstruction of the nanosheet.

The origin of magnetism in Fe-doped SiCNSh can be attributed to exchange interactions between localized Fe-3d states and surrounding Si/C p orbitals, which promote spin polarization through p-d hybridization.

As summarized in Table 5, the magnetic response of the 1Fe(1C)-doped SiCNSh is mainly dominated by the Fe dopant, which carries the largest local magnetic moment of 4.082 μ_B . The nearest Si atoms bonded to Fe, particularly Si(36), Si(37), and Si(47), also exhibit pronounced induced magnetic moments of approximately 0.36–0.37 μ_B , confirming efficient spin-density delocalization from Fe into the surrounding Si sublattice. In addition, several neighboring C atoms show moderate spin polarization, indicating that both Fe-Si and Fe-C interactions contribute to the total magnetic moment of 6.004 μ_B per supercell. The magnetic behavior obtained in the present work can be qualitatively compared with the Fe@C-doped SiC nanosheet reported by Javan et al., where a total magnetic moment of 2 μ_B and a local Fe magnetic moment of 2.031 μ_B were obtained. In both studies, Fe acts as the dominant magnetic center and induces spin polarization in the surrounding lattice. However, the present calculations predict substantially larger magnetic moments, with a local Fe moment of 4.0 μ_B and a total magnetic moment of 6.0 μ_B per supercell. This enhancement may be attributed to differences in supercell size, dopant concentration, and the inclusion of Hubbard-U corrections, which strengthen the localization and exchange splitting of Fe-3d states. The distribution of spin magnetic moments across the SiCNSh framework (Figure 11) reveals that magnetization is strongly concentrated around the Fe dopant, which acts as the dominant magnetic center with a substantial localized moment. Pronounced positive spin polarization is induced in the nearest-neighbor Si atoms, particularly those directly bonded to Fe, confirming efficient spin-density delocalization into the surrounding lattice. Neighboring carbon atoms also exhibit moderate magnetic moments, indicating that both Fe-Si and Fe-C orbital interactions contribute to the magnetic response of the nanosheet.

Table 5. Local and total magnetic moments for 1Fe(1C)-doped SiCNSh.

Atom Number	Element	Local Magnetic Moment (μ_B)	Magnetic Contribution
42	Fe	4.082	Main magnetic center
37	Si	0.372	Strong induced spin polarization
47	Si	0.361	Strong induced spin polarization
36	Si	0.360	Strong induced spin polarization
32	C	0.072	Moderate induced spin polarization
41	C	0.063	Moderate induced spin polarization
27	Si	0.063	Moderate induced spin polarization
51	C	0.061	Moderate induced spin polarization
52	C	0.059	Moderate induced spin polarization
43	C	0.052	Moderate induced spin polarization
31	C	0.051	Moderate induced spin polarization
46	Si	0.049	Moderate induced spin polarization
48	Si	0.045	Moderate induced spin polarization
—	Total supercell	6.004	Total magnetic moment

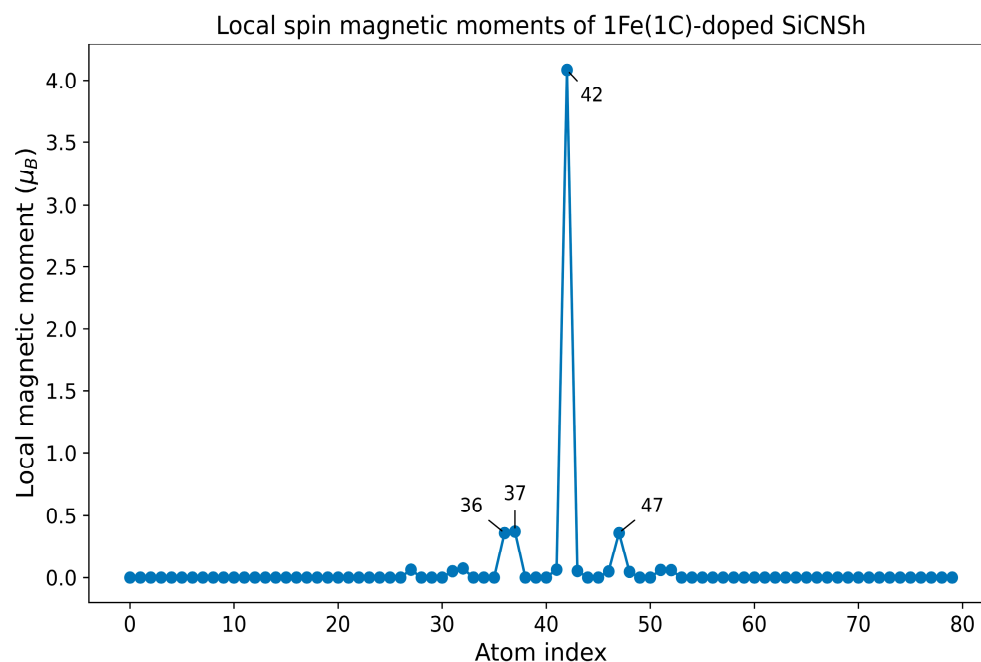


Figure 11. Local spin magnetic moments of 1Fe(1C)-doped SiCNSh as a function of atom index. The Fe dopant exhibits the dominant magnetic contribution, whereas neighboring Si and C atoms show induced spin polarization due to Fe-mediated orbital hybridization.

The spatial spin magnetic moment map (Figure 12) further illustrates the asymmetric localization of magnetic polarization around the Fe site. The intense magnetic region surrounding the dopant demonstrates that Fe substitution generates localized magnetic ordering rather than uniformly distributed magnetization throughout the nanosheet.

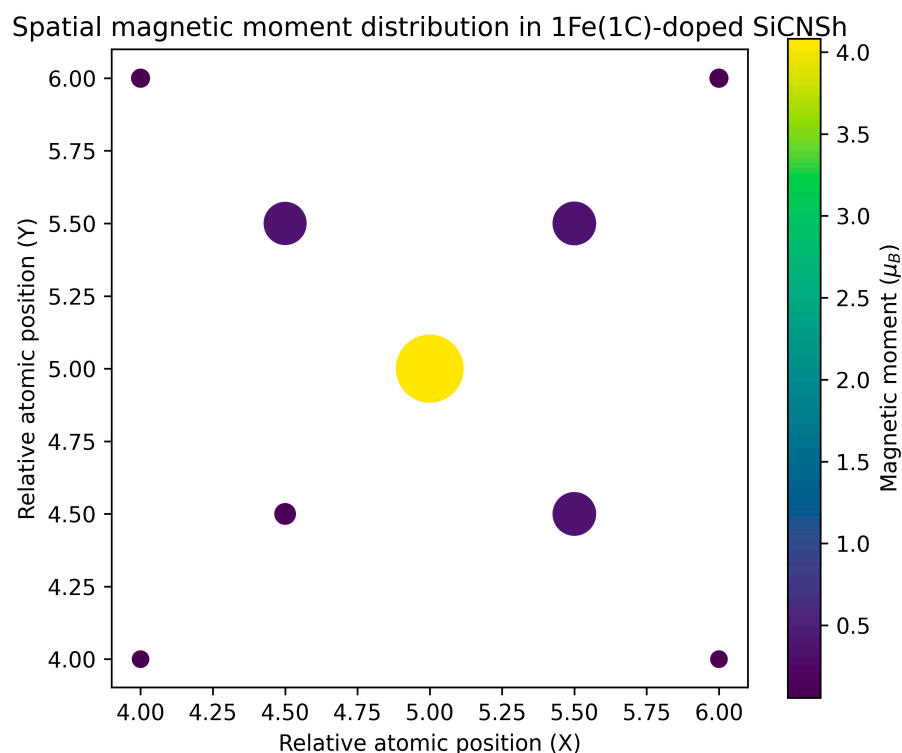


Figure 12. Spatial distribution of local magnetic moments in 1Fe(1C)-doped SiCNSh, illustrating the strong localization of magnetic polarization around the Fe dopant and induced spin density in neighboring Si and C atoms, indicative of cooperative spin alignment.

Importantly, the predominance of positive local magnetic moments suggests cooperative spin alignment instead of magnetic compensation, confirming the emergence of a stable ferromagnetic semiconducting state. The observed magnetization originates mainly from strong Fe–3d hybridization with neighboring Si/C orbitals, which enhances spin polarization and promotes dopant-induced magnetic ordering within the SiCNSh framework.

The partial delocalization of spin density toward neighboring Si atoms indicates that the magnetic coupling is not strictly confined to the Fe dopants but extends into the surrounding SiCNSh matrix, thereby enhancing magnetic stability. The pronounced spin polarization induced in adjacent Si atoms demonstrates the formation of extended magnetic exchange pathways mediated through Fe–Si/C interactions. Such behavior is particularly advantageous for spintronic and nanoscale magnetic semiconductor applications, where simultaneous control of charge transport and spin polarization is essential.

The energetic stability of magnetic ordering was further evaluated through comparison of the ferromagnetic (FM) and antiferromagnetic (AFM) spin configurations. The calculated total energies reveal that the FM state is energetically more favorable than the AFM configuration, with a total FM–AFM energy difference of 0.216 eV for the 2Fe@C-doped SiCNSh. Since two Fe atoms were incorporated into the supercell, the effective magnetic exchange energy per Fe pair was estimated as $\Delta E = 0.216$ eV, confirming robust ferromagnetic coupling between dopant centers. Furthermore, unlike previously reported Fe(C)-doped SiC nanotubes, where only weak antiferromagnetic stabilization ($\Delta E = -0.009$ eV) was reported by Zhang et al. [40], the present 2Fe@C configuration strongly favors a ferromagnetic ground state. This enhancement can be attributed to stronger Fe–3d/C–2p/Si–3p hybridization and more efficient spin-density delocalization toward neighboring Si atoms, which collectively strengthen magnetic exchange coupling and stabilize high-temperature ferromagnetism.

To evaluate the thermal stability of the magnetic ordering, the Curie temperature (T_C) was estimated within the framework of the mean-field approximation (MFA) using Equation (2) in Section 2. Using the calculated exchange energy, the Curie temperature was determined to be approximately 837 K, indicating exceptionally strong magnetic stability and robust ferromagnetic ordering well above room temperature. Such a high predicted T_C suggests that the magnetic phase of Fe-doped SiCNSh can remain thermally stable under conditions relevant for practical nanoelectronic and spintronic device operation. Although MFA-based approaches may somewhat overestimate Curie temperatures due to the neglect of spin fluctuations and many-body effects, particularly in low-dimensional materials, similar limitations have been reported in previous studies employing MFA for transition-metal-doped two-dimensional systems [41,42]. Therefore, the calculated T_C should be regarded as an upper-bound estimate of the magnetic transition temperature rather than an exact quantitative value. Nevertheless, the predicted value still confirms the strong intrinsic magnetic robustness of the Fe-doped nanosheet.

Previous studies have shown that Curie temperatures obtained within the MFA framework may differ from experimentally measured values because spin fluctuations are not explicitly taken into account. For example, Pajda et al. reported MFA-derived Curie temperatures of 1414 K and 1645 K for bcc Fe and fcc Co, respectively, compared with experimental values of 1044–1045 K and 1388–1398 K, indicating that MFA provides a reasonable estimate of magnetic ordering temperatures while tending to predict somewhat higher values in certain systems [43]. Therefore, the Curie temperature calculated in the present work should be regarded as an approximate upper-bound estimate of the magnetic transition temperature.

Furthermore, previous studies on transition-metal-doped two-dimensional materials have widely employed the MFA approach to estimate Curie temperatures and assess mag-

netic stability. For instance, Majid et al. successfully applied MFA to evaluate the magnetic ordering temperatures of transition-metal-doped SiC monolayers and demonstrated its effectiveness in describing magnetic trends in doped SiC-based systems [44]. Although MFA may somewhat overestimate the absolute values of T_C , it has consistently provided a reliable qualitative description of ferromagnetic ordering tendencies and relative magnetic stability in low-dimensional materials. Therefore, the predicted T_C of 837 K still provides strong evidence for robust ferromagnetic ordering and high thermal stability in the Fe-doped SiC nanosheet.

The enhanced magnetic stability of the 2Fe@C configuration originates primarily from strong hybridization between Fe-3d and neighboring C-2p/Si-3p orbitals, which promotes efficient spin polarization and stabilizes the ferromagnetic ground state. Because Fe atoms substitute carbon sites and establish relatively short and strongly covalent Fe-C bonds, the resulting exchange interactions become significantly strengthened, facilitating efficient magnetic coupling throughout the nanosheet. The present findings can be further compared with the first-principles study of Majid et al. [44], who investigated transition-metal-doped SiC monolayers for spintronic applications. In their work, the Fe-doped SiC monolayer exhibited a predicted Curie temperature of approximately 1150 K, indicating strong thermal magnetic stability mediated by transition-metal 3d-C 2p hybridization and exchange interactions within the SiC framework. Consequently, Fe incorporation not only tailors the electronic structure through band-gap narrowing but also induces stable high-temperature ferromagnetism, highlighting Fe-doped SiCNShs as promising candidates for next-generation magnetic semiconductors and spin-dependent nanoelectronic applications.

4. Conclusions

In this work, the structural, electronic, charge-transfer, thermal, and magnetic properties of pristine and Fe-doped silicon carbide nanosheets (SiCNShs) were systematically investigated using spin-polarized density functional theory within the LSDA + U framework. The obtained results demonstrate that substitutional Fe incorporation at carbon sites effectively modifies both the electronic and magnetic behavior of the SiCNSh while preserving its structural integrity and thermodynamic stability.

(i) Structural optimization, total-energy minimization, force convergence, and stress evolution analyses confirm that Fe substitution does not induce significant lattice instability, while the negative defect formation energy (-7.44 eV/atom) indicates thermodynamically favorable Fe incorporation into the SiCNSh lattice. Furthermore, *ab initio* molecular dynamics (AIMD) simulations performed at 300 K reveal that the energetically favorable Fe-doped configuration maintains structural integrity under finite-temperature conditions, confirming its thermal robustness and suitability for practical nanoscale applications.

(ii) Electronic-structure calculations reveal that pristine SiCNSh exhibits an intrinsically nonmagnetic semiconducting nature with a band gap of approximately 2.4 eV. Upon Fe substitution, substantial electronic reconstruction occurs through strong Fe-3d/C-2p/Si-3p hybridization, resulting in significant band-gap narrowing. The band gap decreases to approximately 1.1 eV for the single-Fe-doped configuration and further reduces to 0.53/0.51 eV for the double-Fe system while preserving semiconducting behavior. Importantly, Fe incorporation induces pronounced spin asymmetry between spin-up and spin-down electronic states, confirming the emergence of spin-dependent electronic reconstruction and enhanced exchange interactions.

(iii) Charge-density redistribution and Bader population analyses demonstrate substantial dopant-induced electronic perturbation, characterized by electron depletion around Fe atoms, enhanced electron accumulation on neighboring carbon atoms, and partial neutralization of nearby silicon atoms. These findings indicate the formation of a more localized

covalent Si–C–Fe bonding environment, where strong orbital hybridization plays a decisive role in governing the electronic and magnetic response of the nanosheet.

(iv) Magnetic analysis further reveals that Fe substitution transforms the initially nonmagnetic SiCNSh into a robust ferromagnetic semiconductor. The Fe dopant acts as the dominant magnetic center, carrying a local magnetic moment of approximately $4.08 \mu_B$, while neighboring Si and C atoms exhibit induced spin polarization due to efficient spin-density delocalization. The energetically favorable 2Fe@C configuration strongly stabilizes in the ferromagnetic ground state, exhibiting a positive FM–AFM energy difference of 0.216 eV and an estimated Curie temperature of approximately 837 K, indicating stable magnetic ordering far above room temperature.

Collectively, the present findings demonstrate that Fe incorporation provides an efficient strategy for tailoring the electronic structure, spin polarization, and magnetic stability of SiCNShs through controlled defect engineering. Compared with previous Fe@C-doped SiC nanosheet studies, the present work predicts enhanced magnetic stabilization and provides a more comprehensive description of the underlying mechanisms through AIMD simulations, charge-transfer analysis, FM–AFM energetics, and Curie-temperature estimation.

The coexistence of tunable semiconducting behavior, robust high-temperature ferromagnetism, and structural stability highlights Fe-doped SiCNShs as promising candidates for next-generation spintronic devices, magnetic semiconductors, spin injectors, spin-filter interfaces, magnetic tunnel junctions, and non-volatile magnetic memory technologies.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/micro6030047/s1>, Figure S1: Total-energy convergence during structural optimization of the Fe-substituted SiC nanosheet; Figure S2: Force convergence during structural optimization of the Fe-substituted SiC nanosheet; Figure S3: Stress convergence during structural optimization of the Fe-substituted SiC nanosheet (10^{-5} eV/Å³ scale).

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