

Above-Room-Temperature Half-Metallic Ferromagnetism in Fe-Doped MoS₂ Monolayers

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Abstract

This study investigates the electronic and magnetic properties of iron (Fe) doped molybdenum disulfide (MoS₂) monolayers through first-principles calculations based on density functional theory (DFT). Utilizing the GGA-PBE exchange-correlation functional and projector augmented-wave (PAW) pseudopotentials within the Quantum ESPRESSO framework, we simulated a $4 \times 4 \times 1$ supercell wherein two Mo atoms are substituted by Fe. While the pristine MoS₂ monolayer is confirmed to be a non-magnetic, direct-bandgap semiconductor with a gap of 1.76 eV, spin-polarized density of states (DOS) analyses demonstrate that Fe doping induces a highly spin-polarized, DOS-based half-metallic ferromagnetic ground state. Specifically, the majority-spin channel exhibits metallic behavior, whereas the minority-spin channel retains a semiconducting bandgap of 1.155 eV. The substituted system yields a total magnetic moment of 4.397 μ_B per supercell, confirming the establishment of stable diluted ferromagnetism. Furthermore, mean-field estimations predict a Curie temperature of 508 K. The realization of half-metallicity alongside above-room-temperature ferromagnetism underscores the significant potential of Fe-doped 2D MoS₂ for advanced spintronic applications and nanoscale magnetic memory devices.

Keywords

MoS₂ Monolayer, Density Functional Theory, Half-Metallicity, Ferromagnetism, Spintronics

1. Introduction

In recent years, two-dimensional (2D) materials have attracted tremendous re-

search interest due to their remarkable physical properties and substantial potential for next-generation technologies. Characterized by their atomic-scale thickness and the planar confinement of charge carriers, these materials exhibit unique electronic, optical, and magnetic phenomena that are fundamentally distinct from their bulk counterparts. Furthermore, their exceptionally large specific surface area and high sensitivity to external perturbations make them highly promising candidates for advanced applications in nanoelectronics, spintronics, and broader nanotechnologies [1] [2].

Among the most extensively studied 2D materials are transition metal dichalcogenides (TMDs), characterized by the general formula MX_2 (where M represents a transition metal and X a chalcogen). These compounds exhibit a layered structure stabilized by van der Waals interactions, enabling the isolation of stable monolayers with intrinsic properties that are fundamentally distinct from their bulk counterparts [3]. Molybdenum disulfide (MoS_2) occupies a prominent position within this family, primarily due to its indirect-to-direct bandgap crossover when scaling from the bulk to the monolayer limit. This dimensional quantum confinement makes it highly attractive for nanoelectronic and optoelectronic applications [4] [5].

However, despite its remarkable electronic characteristics, pristine 2D MoS_2 possesses several limitations. Most notably, its intrinsic non-magnetic ground state severely restricts its integration into spintronic devices [6]. Furthermore, its charge carrier mobility remains moderate compared to other 2D materials, and precise modulation of its band structure is difficult to achieve without external modification. These constraints necessitate advanced property engineering to broaden the material's application scope.

In this context, transition metal doping emerges as a robust strategy to tailor both the electronic and magnetic properties of 2D materials. Doping effectively modifies the density of states near the Fermi level and can induce local magnetic moments, thereby paving the way for spintronic applications, particularly in the development of diluted magnetic semiconductors [7]. Specifically, iron (Fe), owing to its partially filled $3d$ orbitals, serves as an exceptionally suitable dopant capable of significantly altering the electronic structure of MoS_2 while concurrently inducing robust magnetic polarization [8].

In this work, spin-polarized density functional theory (DFT) calculations are employed to systematically investigate the structural, electronic, and magnetic properties of substitutionally Fe-doped MoS_2 monolayers. Particular attention is devoted to elucidating the microscopic origin of the induced magnetism through spin-resolved electronic band structures, orbital-projected density of states, Löwdin population analyses, and real-space charge and spin-density distributions. Furthermore, the magnetic ground state, exchange interactions, and Curie temperature are evaluated to assess the thermal stability of the ferromagnetic phase. Our calculations predict that substitutional Fe doping transforms intrinsically non-magnetic MoS_2 into a robust half-metallic ferromagnet with a Curie temperature well above room temperature, highlighting its significant potential for next-

generation two-dimensional spintronic devices. By directly correlating orbital hybridization and charge redistribution with exchange interactions and magnetic stability, this study provides a comprehensive microscopic understanding of the fundamental mechanisms governing magnetism in Fe-doped MoS₂.

To ensure a clear logical progression, the remainder of this manuscript is structured as follows. Section 2 first details the computational methodology and the specific simulation parameters utilized in this study. Building upon this theoretical framework, Section 3 provides a comprehensive presentation and discussion of the structural, electronic, and magnetic properties of the Fe-doped MoS₂ monolayer. Finally, Section 4 synthesizes the principal conclusions.

2. Computational Methods

The present investigation was carried out using first-principles calculations based on Density Functional Theory (DFT) [9] [10], as implemented in the Quantum ESPRESSO package [11]. Exchange-correlation effects were treated using the Perdew-Burke-Ernzerhof (PBE) formulation of the Generalized Gradient Approximation (GGA) [12]. To accurately capture the long-range dispersion interactions inherent to 2D systems, non-local van der Waals corrections were explicitly included using the vdW-DF2 functional [13]-[16]. The interaction between valence electrons and ionic cores was modeled using Projector Augmented-Wave (PAW) pseudopotentials [17]. The specific pseudopotentials utilized for the Mo, S, and Fe atoms are detailed in **Table 1**. Following rigorous convergence tests, the electronic wave functions were expanded in a plane-wave basis set with a kinetic energy cutoff of 71 Ry, while the charge density cutoff was set to 400 Ry. This cutoff ratio (~ 5.6) is highly consistent with the requirements for PAW pseudopotentials, ensuring accurate augmentation charges and satisfactory convergence of both total energy and electronic properties. The out-of-plane lattice parameter 20.0 Å inherently incorporates a substantial vacuum region, ensuring the elimination of spurious interactions between adjacent periodic images and thus accurately simulating an isolated two-dimensional system.

Table 1. Summary of the Projector Augmented-Wave (PAW) pseudopotentials used for the Mo, S, and Fe atoms in the electronic structure calculations.

Atom	Pseudopotential File	Valence Electrons
Mo	Mo.pbe-spn-kjpaw_psl.1.0.0.UPF	14
S	S.pbe-n-kjpaw_psl.1.0.0.UPF	6
Fe	Fe.pbe-spn-kjpaw_psl.1.0.0.UPF	16

Brillouin-zone integrations for the self-consistent field (SCF) calculations were performed using a Γ -centered $8 \times 8 \times 1$ Monkhorst-Pack k -point mesh, which provides sufficient sampling given the dimensions of the $4 \times 4 \times 1$ supercell. A single k -point was sampled along the z -axis to properly reflect the two-dimensional nature of the system. The SCF convergence threshold was set to a rigorous

1.0×10^{-12} Ry to ensure high precision in determining the electronic ground state. To facilitate stable convergence within the large supercell framework, the charge-density mixing parameter was set to 0.2, with a maximum of 100 SCF iterations.

To properly account for the dopant, spin-polarized calculations were performed ($n_{\text{spin}} = 2$). Initial magnetic moments were set to zero for the non-magnetic Mo and S atoms, while an initial magnetization of 0.5 was assigned to the Fe atoms. This facilitated convergence toward a stable ferromagnetic state, consistent with the partially occupied $3d$ orbitals of iron.

Finally, for the non-self-consistent field (NSCF) and electronic band structure calculations, a denser Γ -centered $12 \times 12 \times 1$ k -point mesh was employed to achieve high accuracy in the density of states (DOS). The number of bands (nbnd) was set to 250. Given the total of 420 valence electrons in the supercell (contributed by 14 Mo, 32 S, and 2 Fe atoms), this configuration provides approximately 210 occupied bands and 20% additional empty bands. This is highly sufficient to accurately resolve the low-energy unoccupied electronic states near the Fermi level.

3. Results and Discussion

This section is organized around three complementary aspects: structural, electronic, and magnetic properties. The primary objective is to elucidate the effects of Fe doping on the local atomic geometry, the electronic density of states, orbital hybridization, and spin polarization, and to compare the resulting trends with those reported in the literature.

3.1. Structural Properties

The structural model employed in this study consists of a 2H-phase MoS_2 monolayer constructed as a $4 \times 4 \times 1$ supercell. This supercell contains a total of 48 atoms: 14 molybdenum (Mo), 32 sulfur (S), and 2 iron (Fe) atoms. Iron doping was achieved by substituting two Mo atoms with Fe atoms, yielding a substitutional doping concentration of 12.5% at the transition metal sites. The supercell lattice vectors are defined as $a_1 = (12.565, 0, 0)$ Å, $a_2 = (-6.283, 10.882, 0)$ Å, and $a_3 = (0, 0, 20.0)$ Å. This chosen supercell dimension is highly consistent with established first-principles methodologies for modeling Fe-doped MoS_2 and related transition-metal-doped frameworks. To accurately represent the 12.5% doping concentration while avoiding artificial clustering effects, the two substitutional Fe atoms were positioned with a spatial separation of 5.76 Å. The fully relaxed structural parameters for both the pristine and Fe-doped MoS_2 systems, calculated within the DFT framework, are summarized in **Table 2**.

Table 2. Optimized lattice parameters and structural properties for the pristine $4 \times 4 \times 1$ MoS_2 supercell and the corresponding Fe-doped system.

System	a (Å)	$d_{\text{Mo-S}}$ (Å)	$d_{\text{Mo-Fe}}$ (Å)	$d_{\text{Fe-Fe}}$ (Å)	Angle (S-Mo-S) (°)	Angle (Mo-Fe-Mo) (°)
Pristine MoS_2	3.119	2.392	-	-	81.386	-
Fe-doped MoS_2	3.149	2.392	3.237	5.76	82.087	60

The optimized atomic structure of the Fe-doped MoS₂ monolayer was visualized using the XCrySDen software package. **Figure 1** illustrates the top and side views of the fully relaxed $4 \times 4 \times 1$ supercell, explicitly elucidating the atomic arrangement and the local coordination environment surrounding the Fe dopants. These visual representations provide a critical framework for assessing the geometric distortions and structural modifications inherent to the transition-metal-substituted lattice.

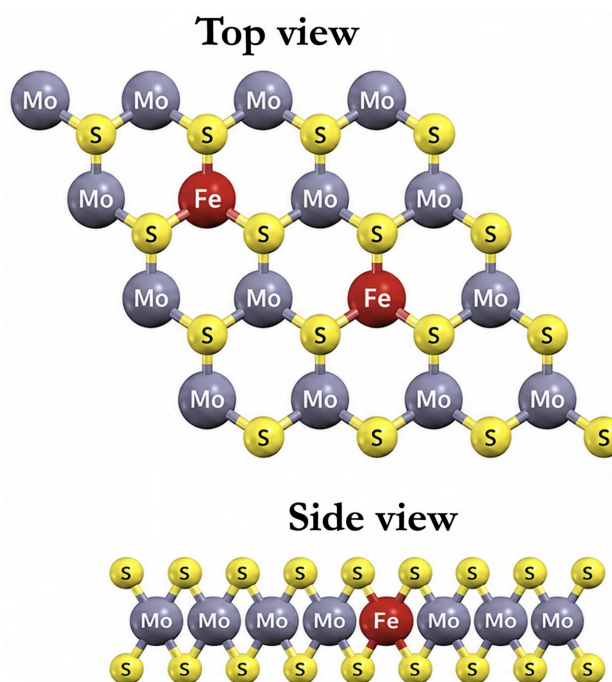


Figure 1. Optimized atomic structure of the Fe-doped 2D MoS₂ monolayer ($4 \times 4 \times 1$ supercell), illustrating both the top view and side view of the transition-metal dopant sites.

3.2. Charge Transfer

To quantitatively evaluate the charge redistribution induced by the substitution of Fe at the Mo sites, a Bader charge analysis was performed. This methodology partitions the continuous electron density into distinct atomic basins bounded by zero-flux surfaces of the charge-density gradient, thereby providing a rigorous and physically meaningful determination of localized atomic charges and interatomic charge transfer. The resulting Bader charge distribution for the doped system is illustrated in **Figure 2**.

Three principal conclusions emerge from the Bader charge analysis. First, the host Mo atoms exhibit an average Bader charge of $12.923 e^-$, which is lower than the 14 valence electrons explicitly treated by the PAW pseudopotential. This corresponds to an average charge donation of approximately $1.077 e^-$ from each Mo atom to its neighboring S atoms, underscoring the mixed covalent-ionic character of the Mo-S bonds. This behavior aligns with findings by Jia [18], who observed similar electron transfer dynamics in pristine MoS₂.

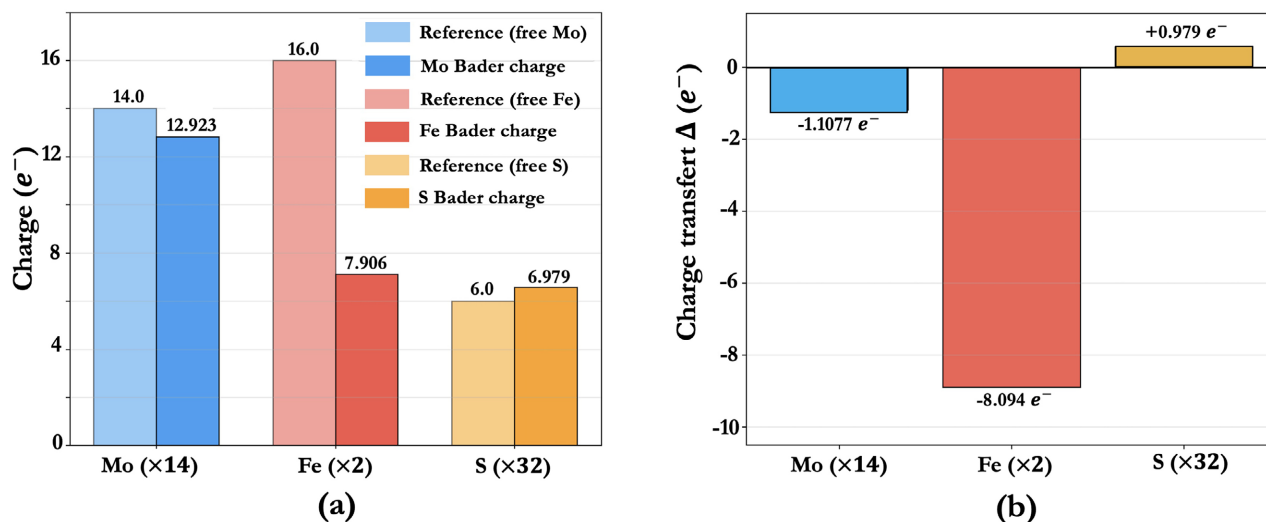


Figure 2. Bader charge analysis illustrating the quantitative charge redistribution and localized atomic charges within the Fe-doped MoS₂ monolayer.

Second, the substituted Fe dopants possess a calculated Bader charge of $7.906 e^-$. Given that the employed Fe pseudopotential includes 16 valence electrons (encompassing semi core states), this pronounced depletion reveals substantial charge redistribution following substitution at the Mo site. The reduced localized electron population around the Fe atoms indicates strong Fe-S chemical bonding and significant electron delocalization toward the adjacent sulfur atoms. The substantial disparity in retained charge between Mo ($12.923 e^-$) and Fe ($7.906 e^-$) originates from their distinct electronic configurations. Specifically, the partially filled Fe $3d$ orbitals interact intensely with the S $3p$ orbitals, driving enhanced orbital hybridization and charge sharing within the Fe-S bonds.

Third, the sulfur atoms gain an average of $0.979 e^-$, reaffirming their role as electron acceptors within the doped MoS₂ monolayer. This uptake is quantitatively comparable to that reported for pristine MoS₂ [18], indicating that while the global charge-transfer mechanism is preserved upon Fe substitution, the local redistribution is significantly amplified in the immediate vicinity of the dopant. This conclusion is strongly corroborated by the projected density of states (PDOS) presented in Figure 5, which reveals a robust overlap between the Fe $3d$ and S $3p$ states in the energy range from -5 to 0 eV. Together, the combined Bader charge and PDOS analyses conclusively demonstrate that the Fe-S interaction is characterized by a strongly polarized covalent-ionic bond, driven by deep orbital hybridization and partial electron transfer from the transition metal to the chalcogen.

To further elucidate the spatial distribution of valence electrons within the Fe-doped MoS₂ monolayer, the self-consistent charge density was calculated and visualized using the XCrySDen software package. The corresponding charge-density contour maps are presented in Figure 3. Panel (a) illustrates a two-dimensional cross-section passing directly through the transition-metal (Mo/Fe) basal plane, whereas panel (b) depicts the sulfur plane located approximately $\pm 1.55 \text{ \AA}$ away.

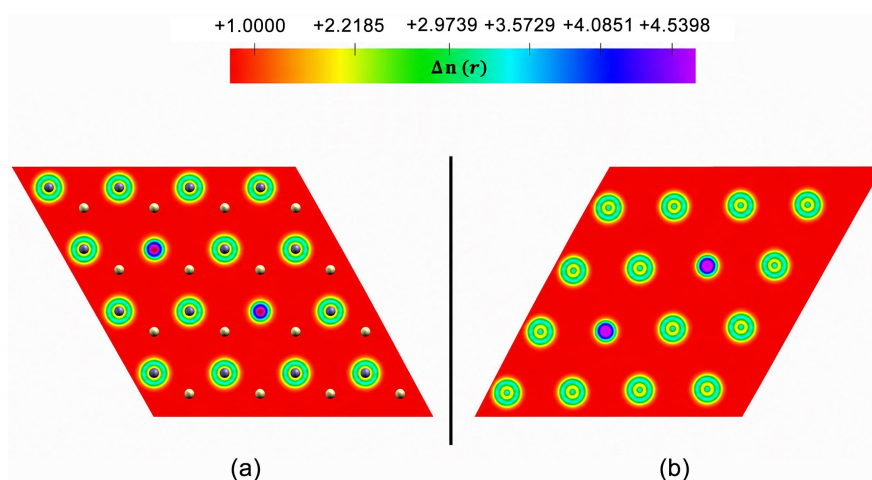


Figure 3. Contour maps of the self-consistent valence charge density for the Fe-doped MoS₂ monolayer. (a) Two-dimensional cross-section through the transition-metal (Mo/Fe) basal plane. (b) Cross-section through the chalcogen (sulfur) plane, located approximately $\pm 1.55 \text{ \AA}$ from the Mo/Fe layer. The color scale indicates the valence electron density in units of $e^{-1} \cdot \text{\AA}^{-3}$.

As depicted in **Figure 3(a)**, the valence electron density is strongly localized around both Mo and Fe atomic cores, evidenced by the high-density contours (indicated by the blue-to-violet color scale). The continuous charge-density contours bridging the transition-metal and sulfur atoms indicate substantial electron sharing, confirming the formation of robust chemical bonds throughout the lattice. Notably, the local charge-density topology around the Fe dopants closely mirrors that of the host Mo atoms, demonstrating that the substitutional Fe atoms integrate seamlessly into the MoS₂ framework without inducing severe geometric distortions.

In contrast, the charge density within the chalcogen plane (**Figure 3(b)**) exhibits a more homogeneous and diffuse distribution. This behavior stems from the spatially extended character of the S 3*p* orbitals compared to the highly localized Fe 3*d* and Mo 4*d* states. Consequently, the electron density surrounding the sulfur atoms is delocalized over a larger effective volume, resulting in a lower peak charge density relative to the transition-metal sites.

These charge-density maps are in excellent agreement with the preceding Bader analysis (**Figure 2**), visually corroborating the partial electron transfer from the transition metals to sulfur. Furthermore, the continuous charge bridging the Fe and neighboring S atoms is fully consistent with the PDOS (**Figure 5**), which highlighted a pronounced energetic overlap between the Fe 3*d* and S 3*p* orbitals. Collectively, these complementary analyses confirm that the Fe-S interaction is governed by a strongly polarized covalent-ionic bond, driven by deep orbital hybridization and concurrent charge donation.

Having established the fundamental electronic structure and bonding mechanisms of the Fe-doped MoS₂ system, the subsequent section focuses on its emergent magnetic properties, specifically detailing the distribution of local magnetic

moments, half-metallic spin polarization, and the estimated Curie temperature.

3.3. Magnetic Properties

Spin-polarized self-consistent field calculations converge to a stable magnetic ground state with a total energy of -7502.330 Ry. At convergence, the calculated total magnetic moment is $4.00 \mu_B$ per supercell, while the absolute magnetization reaches $4.75 \mu_B$ per supercell. These nonzero values definitively confirm that substitutional Fe doping induces a robust magnetic state within the MoS₂ monolayer, in stark contrast to the intrinsically non-magnetic ground state of pristine MoS₂.

Given that the supercell contains two Fe atoms, the total magnetic moment of $4.00 \mu_B$ corresponds to an effective average of $2.00 \mu_B$ per dopant. It is important to note that this value represents a macroscopic average, as a fraction of the spin polarization is delocalized onto the adjacent sulfur atoms and interstitial regions due to strong *p-d* hybridization. Nevertheless, this calculated magnetic moment is in excellent agreement with prior first-principles literature. For instance, Lan *et al.* [19] reported a magnetic moment of $1.849 \mu_B$ for a single substitutional Fe atom in a $3 \times 3 \times 1$ MoS₂ supercell. The marginally higher magnetic moment observed in the present study ($2.00 \mu_B$ per Fe) can be attributed to the elevated doping concentration (12.5% compared to approximately 11.1%), which facilitates enhanced ferromagnetic exchange coupling between adjacent Fe sites.

The theoretical prediction of a stable magnetic moment upon Fe substitution is strongly corroborated by recent experimental evidence. Utilizing aberration-corrected transmission electron microscopy (AC-TEM) and superconducting quantum interference device (SQUID) magnetometry, Fu *et al.* [20] demonstrated that the substitutional incorporation of Fe at Mo lattice sites in MoS₂ monolayers synthesized via chemical vapor deposition generates robust room-temperature magnetic ordering. While bulk macroscopic magnetic observables obtained experimentally cannot be compared exactly one-to-one with zero-Kelvin DFT magnetic moments, these empirical findings emphatically validate the transition to a magnetically ordered state predicted by our calculations.

3.3.1. Spin DOS

The spin-resolved total density of states (DOS) for the Fe-doped MoS₂ monolayer was computed across an energy window of -10.0 to $+10.0$ eV, utilizing a high energy resolution of 0.001 eV. For analytical clarity, the calculated Fermi energy ($E_F = -0.0169$ eV) was aligned to zero. The resulting spin-polarized total DOS, presented in Figure 4, exhibits two remarkable features.

First, a pronounced asymmetry between the spin-up and spin-down channels is clearly evident. At the Fermi level, the majority-spin (spin-up) channel demonstrates metallic behavior with a finite density of states of 27.14 states/eV, whereas the minority-spin (spin-down) channel displays a completely vanishing DOS. This strong spin asymmetry confirms that Fe substitution induces a highly spin-polarized electronic structure.

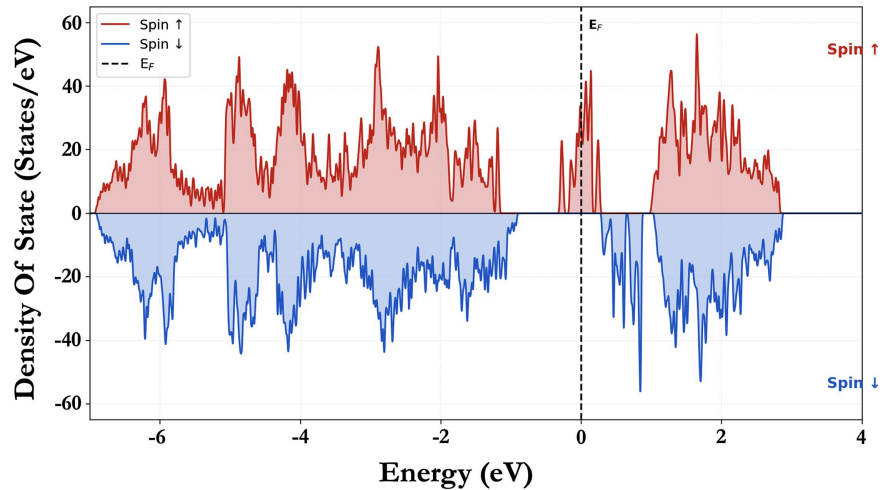


Figure 4. Spin-polarized total density of states (DOS) of the Fe-doped MoS₂ monolayer. The majority-spin (spin-up) and minority-spin (spin-down) channels are represented in red and blue, respectively.

Second, a detailed inspection of the electronic structure near the Fermi level reveals that the minority-spin channel retains a semiconducting bandgap of approximately 1.155 eV, with the valence band maximum (VBM) located at -0.890 eV and the conduction band minimum (CBM) at $+0.265$ eV. This energy gap is significantly narrower than the ~ 1.8 eV direct bandgap characteristic of pristine MoS₂ monolayers [4]. In stark contrast, the majority-spin channel exhibits continuous states crossing the Fermi level. This coexistence of a finite density of states in the spin-up channel and a vanished DOS in the spin-down channel serves as the defining DOS-based signature of half-metallicity, ultimately yielding 100% spin polarization at the Fermi level.

This predicted half-metallic phase is in excellent agreement with established theoretical literature. Lan *et al.* [19] similarly reported that the Fermi level exclusively intersects the majority-spin electronic states in substitutionally Fe-doped MoS₂, closely mirroring our DOS calculations. Furthermore, Xie *et al.* [21] demonstrated that Fe doping transforms monolayer MoS₂ into a half-metallic ferromagnet driven by strong p - d orbital hybridization between the Fe 3*d* and S 3*p* states. These corroborating studies provide robust validation for the half-metallic electronic structure and the fully spin-polarized ground state observed in the present work.

3.3.2. Spin PDOS

To determine the specific orbital contributions governing the half-metallic phase, the spin-polarized projected density of states (PDOS) was analyzed. By resolving the electronic structure into distinct atomic components, the PDOS elucidates the microscopic origin of the modifications induced by Fe substitution. Figure 5 presents the projected contributions of the Fe 3*d*, Mo 4*d*, and S 3*p* orbitals alongside the total density of states.

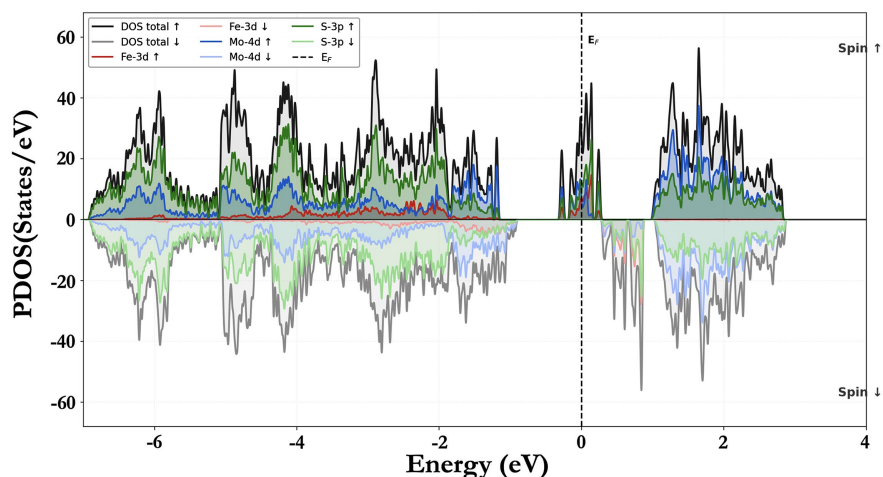


Figure 5. Calculated spin-polarized projected density of states (PDOS) for the Fe-doped MoS₂ monolayer. The total density of states is depicted in black/gray, while the projected orbital contributions are color-coded as follows: Fe 3*d* (red/pink), Mo 4*d* (blue/light blue), and S 3*p* (green/light green). The contrasting shades for each element distinguish between the majority-spin and minority-spin channels, with the Fermi level aligned to zero.

The PDOS analysis reveals two critical insights. First, in the immediate vicinity of the Fermi level ($E = 0$ eV), the S 3*p* and Mo 4*d* orbitals dominate the density of states in the majority-spin channel, while the direct contribution from the Fe 3*d* states is comparatively minor. This indicates that the metallic conductivity in the spin-up channel is primarily mediated by hybridized Mo--S host states that have been perturbed by the transition-metal dopant.

Second, a broader examination of the PDOS across the -7 to $+4$ eV energy range elucidates the fundamental origin of the macroscopic magnetic behavior. While the host Mo 4*d* and S 3*p* orbitals remain largely symmetric between the two spin channels over most of the energy spectrum, the Fe 3*d* states exhibit a pronounced and persistent exchange splitting that extends from the deep valence band up to the Fermi level. The Mo and S orbitals only develop noticeable spin asymmetry in the localized energy window adjacent to the Fermi level, as illustrated in **Figure 5**.

This persistent spin splitting of the Fe 3*d* states originates from the strong intra-atomic exchange interaction within the partially filled 3*d* shell of the dopant, serving as the primary microscopic driver of the induced magnetism. This interpretation is quantitatively validated by the local magnetic moment analysis, which demonstrates that the Fe atom carries a projected spin magnetic moment of $1.582 \mu_B$ substantially larger than the induced moments on the surrounding host atoms. Consequently, the substitutional Fe atom acts as a highly localized spin-polarizing center. Through robust orbital hybridization with the neighboring S 3*p* and Mo 4*d* states, it effectively induces a secondary, delocalized spin polarization throughout the MoS₂ host lattice.

3.3.3. Löwdin Charge

To quantify the atomic distribution of the magnetic moment, a Löwdin popula-

tion analysis was performed using the spin-polarized projected density of states, as calculated via the “projwfc.x” module of Quantum ESPRESSO.

Several critical observations emerge from this population analysis. First, the two substitutional Fe atoms exhibit nearly identical local magnetic moments of $1.588 \mu_B$ and $1.577 \mu_B$, respectively, confirming that the dopants occupy crystallographically equivalent environments within the relaxed supercell. Furthermore, almost the entirety of the magnetic moment on each Fe atom originates from its $3d$ orbitals (contributing $1.582 \mu_B$ and $1.569 \mu_B$, respectively). This demonstrates that the induced magnetism is overwhelmingly driven by the partially occupied $3d$ states, consistent with the localized nature of d electrons in transition-metal dopants.

Second, the nearest-neighbor Mo atoms exhibit induced magnetic moments of up to $0.182 \mu_B$, which are considerably larger than those of Mo atoms located further from the Fe dopants (approximately $0.049 \mu_B$). This spatial attenuation of the magnetic polarization indicates that the magnetic interaction is highly localized around the substitutional Fe centers and progressively weakens with increasing distance. The induced polarization of the surrounding Mo atoms suggests that the magnetic coupling is mediated through the Mo-S bonding network, rather than being confined exclusively to the Fe sites. Similar behavior has been documented in transition-metal-doped MoS_2 , where lattice-mediated interactions govern the coupling between magnetic dopants. In particular, Wu *et al.* [22] demonstrated that neighboring S atoms couple antiferromagnetically with Fe, whereas adjacent Mo atoms align ferromagnetically with the Fe dopants—a theoretical framework that is in excellent qualitative agreement with the positive spin polarization observed on the nearest-neighbor Mo atoms in the present work.

The total magnetic moment derived from the Löwdin population analysis is $4.397 \mu_B$ per supercell, which is in agreement with the self-consistent value of $4.00 \mu_B$ obtained directly from the SCF calculations (a discrepancy of only 9.9%). This minor deviation arises because the Löwdin projection scheme only accounts for the electronic population explicitly projected onto localized atomic orbitals, whereas a fraction of the spin density delocalizes into the interstitial regions and onto weakly polarized sulfur atoms, which is not entirely captured by the projection.

Finally, the orbital magnetic moment decomposition, summarized in **Table 3**, reveals that approximately 72% of the total magnetic moment is localized on the Fe atoms. This spin polarization resides almost entirely within the Fe $3d$ orbitals, with the d_{xy} and $d_{x^2-y^2}$ orbitals providing the dominant contributions. This specific orbital polarization reflects strong in-plane hybridization with the neighboring sulfur $3p$ orbitals within the basal plane. In contrast, the host Mo atoms contribute only 15.7% of the total magnetic moment, with their polarization decaying rapidly as a function of distance from the dopants. This confirms that the magnetic ordering in Fe-doped MoS_2 is fundamentally governed by the localized Fe moments, while the surrounding host lattice acquires only a secondary, induced spin polarization.

Table 3. Löwdin population analysis detailing the decomposition of the total magnetic moment in the Fe-doped MoS₂ monolayer. The table highlights the dominant spin-polarization contribution from the Fe dopants alongside the secondary induced polarization on the host Mo and S atoms.

Atomic Species (Count)	Magnetic Moment (μ_B)	Contribution (%)
Fe (×2)	3.165	72.0
Mo (×14)	0.689	15.7
S (×32)	0.543	12.3
Total	4.397	100.0

3.3.4. Spin Density

To elucidate the real-space distribution of the induced magnetic moments, the spatial spin density was computed and visualized using the XCrySDen software. The resulting spin-density isosurfaces are presented in **Figure 6**, where positive (majority-spin) and negative (minority-spin) charge densities are clearly delineated.

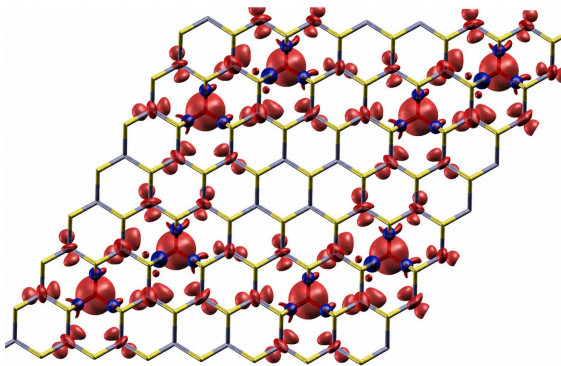


Figure 6. Calculated spatial spin-density distribution of the Fe-doped MoS₂ monolayer. Positive (majority-spin) and negative (minority-spin) spin densities are represented by contrasting isosurfaces.

This spin-density mapping provides a direct visual confirmation of the magnetic polarization within the lattice, fully corroborating the preceding SCF, PDOS, and Löwdin analyses. The substitutional Fe atoms are enveloped by nearly spherical majority-spin isosurfaces, underscoring the dominant contribution of their partially occupied 3*d* states. This quasi-isotropic spin-density distribution arises from the superimposed contributions of all five Fe 3*d* orbitals (*dz²*, *dxz*, *dyz*, *dx²-y²*, and *dxy*). This is in excellent accordance with Unsöld’s theorem, which postulates an approximately spherical continuous density distribution when all degenerate orbitals within a subshell are comparably populated.

In the immediate coordination sphere, the sulfur atoms directly bonded to the Fe dopants exhibit distinct minority-spin polarization. This provides direct visual evidence of a local antiferromagnetic alignment between the Fe centers and their nearest-neighbor chalcogens. This observation aligns perfectly with both our Lö-

wdin population analysis and the theoretical framework of Wu *et al.* [22], which predicted robust antiferromagnetic coupling between substitutional Fe and adjacent S atoms in MoS₂.

Conversely, the nearest-neighbor Mo atoms exhibit majority-spin polarization, visually corroborating the induced local magnetic moments of up to 0.182 μ_B derived from the earlier Löwdin projection. As the radial distance from the dopant increases, the spin density on the host Mo atoms decays markedly and adopts a characteristic dz^2 -like spatial profile, while the more distant sulfur atoms exhibit negligible spin polarization. These spatial features demonstrate that the magnetic perturbation remains highly localized around the transition-metal dopant and attenuates rapidly across the host lattice.

Overall, the real-space spin distribution visually confirms the transformation of the intrinsically non-magnetic MoS₂ monolayer into a robust 2D magnetic material upon Fe substitution. Furthermore, the coexistence of strongly localized primary moments on the Fe sites and secondary induced polarization on the surrounding Mo-S framework strongly implies that macroscopic magnetic ordering is governed by lattice-mediated exchange interactions. The precise thermodynamic strength and fundamental nature of these interactions are quantitatively assessed in the subsequent section by evaluating the total energies of the ferromagnetic and antiferromagnetic spin configurations.

3.3.5. Curie Temperature

To determine the magnetic ground state and quantitatively assess the relative thermodynamic stability of the ferromagnetic (FM) and antiferromagnetic (AFM) spin configurations, independent spin-polarized self-consistent field (SCF) calculations were executed. For the FM configuration, the initial magnetic moments of the two substitutional Fe atoms were aligned parallel to one another. Conversely, the AFM state was initialized by imposing a strictly antiparallel spin alignment between the two transition-metal centers. Because the energy differences associated with magnetic exchange coupling are inherently small, a highly stringent energy convergence threshold of 1.0×10^{-12} Ry was enforced to guarantee the numerical precision required to accurately evaluate the magnetic stability. The fully converged total energies and the corresponding net magnetic moments for both the FM and AFM states are summarized in **Table 4**.

Table 4. Calculated total energies, total magnetic moments, and absolute magnetizations for the ferromagnetic (FM) and antiferromagnetic (AFM) configurations of the Fe-doped MoS₂ monolayer.

Magnetic Configuration	Total Energy (Ry)	Total Magnetisation (μ_B)	Absolute Magnetisation (μ_B)
Ferromagnetic (FM)	-7502.33030000	4.00	4.75
Antiferromagnetic (AFM)	-7502.32547195	0.04	2.29
$\Delta E = E_{AFM} - E_{FM}$	+0.00482805	-	-

As detailed in **Table 4**, the calculated energy difference between the two magnetic configurations is $\Delta E = E_{AFM} - E_{FM} = +65.69$ meV per supercell. This positive value unambiguously establishes that the ferromagnetic (FM) configuration is energetically favored over the antiferromagnetic (AFM) configuration, thereby confirming the FM state as the magnetic ground state of the Fe-doped MoS₂ monolayer.

In the AFM configuration, the total magnetic moment is nearly quenched ($0.04 \mu_B$ per supercell), reflecting the near-perfect compensation of the antiparallel magnetic moments localized on the two Fe centers. Conversely, the absolute magnetization remains substantial at $2.29 \mu_B$ per supercell, confirming that the magnitude of the local magnetic moments is robustly preserved despite their antiparallel alignment. The negligible residual total magnetization in the AFM state is primarily a computational artifact, arising from minor numerical asymmetries associated with the finite convergence tolerance and slight structural deviations within the periodic supercell.

Crucially, the magnetic exchange energy (65.69 meV) significantly exceeds the thermal fluctuation energy at room temperature ($k_B \cdot T \approx 25.9$ meV at 300 K). While this direct energetic comparison does not rigorously define the magnetic phase transition, it strongly implies that the ferromagnetic ordering possesses substantial thermal stability against ambient fluctuations. To explicitly quantify this stability, a formal estimation of the Curie temperature (T_C), derived from the calculated magnetic exchange coupling, is detailed in the subsequent section.

To quantitatively evaluate the magnetic exchange interaction between the two substitutional Fe atoms, the first-principles total-energy difference between the ferromagnetic (FM) and antiferromagnetic (AFM) configurations was mapped onto an isotropic Heisenberg Hamiltonian, following the established approach of Liechtenstein *et al.* [23]. The corresponding spin Hamiltonian is expressed as:

$$\hat{H} = -J \cdot \mathbf{S}_1 \cdot \mathbf{S}_2, \quad (1)$$

where J is the exchange coupling constant ($J > 0$ for ferromagnetic coupling and $J < 0$ for antiferromagnetic coupling), while $\mathbf{S}_1$ and $\mathbf{S}_2$ denote the spin operators localized on the two Fe sites.

Within this localized spin framework, the total energies of the FM and AFM states can be expressed as:

$$E_{FM} = E_0 - JS^2, \quad (2)$$

$$E_{AFM} = E_0 + JS^2, \quad (3)$$

which yields an energy difference of

$$\Delta E = E_{AFM} - E_{FM} = 2JS^2, \quad (4)$$

and consequently

$$J = \frac{\Delta E}{2S^2}, \quad (5)$$

As demonstrated by the Löwdin population analysis (**Table 3**), the magnetic

moment is predominantly localized on the Fe centers, with projected local magnetic moments of $1.588 \mu_B$ and $1.577 \mu_B$. This high degree of localization rigorously justifies the description of the magnetic interaction via localized Fe spin vectors. Furthermore, because the total self-consistent magnetic moment averages approximately $2.00 \mu_B$ per dopant, an effective spin quantum number of $S = 1$ (corresponding to two unpaired d electrons) was adopted, adhering to conventional mapping procedures in first-principles studies of dilute magnetic semiconductors. Substituting the calculated energy difference $\Delta E = 65.69$ meV, the exchange constant is evaluated as:

$$J = \frac{65.69}{2 \times 1^2} = 32.84 \text{ meV}, \quad (6)$$

The positive value of the exchange constant unambiguously confirms that the coupling between the substitutional Fe atoms is ferromagnetic. Furthermore, this magnitude lies comfortably within the established range of exchange constants reported for transition-metal-doped MoS₂, which typically vary between 20 and 50 meV depending on the dopant configuration and concentration [18].

Building upon the determination of the exchange constant, the thermal stability of the ferromagnetic ground state was evaluated by estimating the Curie temperature (T_C) within the framework of the Mean-Field Approximation (MFA). Within the MFA, the Curie temperature of a ferromagnetic system composed of localized spins S coupled through an exchange interaction J is given by [24]:

$$T_C = \frac{2JS(S+1)}{3k_B}, \quad (7)$$

where $k_B = 1.381 \times 10^{-23} \text{ J}\cdot\text{K}^{-1}$ is the Boltzmann constant. By converting the exchange constant to Joules ($J = 5.262 \times 10^{-21} \text{ J}$) and substituting $S = 1$, the Curie temperature is obtained as:

$$T_C = \frac{2 \times 5.262 \times 10^{-21} \times 1 \times 2}{3 \times 1.381 \times 10^{-23}} = 508 \text{ K}, \quad (8)$$

The estimated Curie temperature is approximately 508 K (about 235°C). However, according to the Mermin-Wagner theorem, long-range magnetic order in an ideal two-dimensional isotropic Heisenberg system is suppressed at finite temperatures unless stabilized by magnetic anisotropy. Because spin-orbit coupling (SOC) was omitted from our present calculations, the magnetic anisotropy energy is not explicitly quantified here. Recent first-principles studies on transition-metal-doped MoS₂ confirm that the delicate interplay between localized exchange interactions and dopant-induced magnetic anisotropy energy (MAE) is fundamentally required to lift these restrictions and stabilize finite-temperature magnetic order [25]. Furthermore, the Mean-Field Approximation neglects thermal spin fluctuations and generally overestimates transition temperatures. Consequently, this estimated T_C does not definitively establish finite-temperature long-range ferromagnetism in an ideal free-standing monolayer. Rather, it serves as a robust metric for the strength of the localized ferromagnetic exchange interac-

tions, suggesting strong inherent magnetic stability that could be preserved in practical spintronic devices where substrate effects or extrinsic anisotropy break the strict 2D symmetry.

4. Conclusions

In this work, the structural, electronic, and magnetic properties of Fe-substituted MoS₂ monolayers were systematically investigated using spin-polarized density functional theory calculations. Structural optimization confirmed that substitutional incorporation of Fe into the Mo lattice is structurally stable and induces only moderate local distortions, thereby preserving the integrity of the two-dimensional crystal structure.

The electronic structure calculations revealed that Fe doping profoundly modifies the electronic properties of pristine MoS₂. The spin-resolved density of states demonstrated a pronounced asymmetry between the two spin channels, with a metallic majority-spin channel and a semiconducting minority-spin channel, establishing the DOS-based half-metallic character of the Fe-doped monolayer. The projected density of states further showed that the electronic and magnetic properties are primarily governed by the Fe 3*d* orbitals, whose strong hybridization with the neighboring S 3*p* and Mo 4*d* states gives rise to the observed spin polarization.

The microscopic origin of the magnetic behavior was further clarified through the Löwdin population analysis and spin-density visualization. The magnetic moments were found to be predominantly localized on the substitutional Fe atoms, while smaller induced magnetic moments develop on the neighboring Mo atoms through exchange interactions mediated by the Mo-S network. The spin-density distribution confirmed the localized nature of the magnetic polarization around the Fe dopants and provided direct real-space evidence of the magnetic ordering induced by Fe substitution.

A comparison of the total energies of the ferromagnetic and antiferromagnetic configurations demonstrated that the ferromagnetic state constitutes the magnetic ground state of the system, with an energy stabilization of 65.69~meV per supercell. Mapping the DFT energy difference onto the isotropic Heisenberg model yielded an exchange coupling constant of $J = 32.84$ meV. Within the Mean-Field Approximation (MFA), the corresponding Curie temperature was estimated to be approximately 508 K, significantly exceeding room temperature. Although the MFA generally overestimates the transition temperature because thermal spin fluctuations are neglected, the predicted value nevertheless indicates robust magnetic stability.

Overall, the present results demonstrate that substitutional Fe doping transforms intrinsically nonmagnetic monolayer MoS₂ into a half-metallic two-dimensional magnetic material exhibiting a stable ferromagnetic ground state and an estimated Curie temperature well above room temperature. These characteristics make Fe-doped MoS₂ a promising candidate for future spintronic devices, includ-

ing spin injectors, spin filters, and magnetic logic elements. The present work also provides a theoretical framework that may guide future experimental investigations of transition-metal-doped transition metal dichalcogenides for room-temperature spintronic applications.

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Conflicts of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability Statement

The data that support the findings of this study are available upon reasonable request from the authors.

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