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Electronic and Structural Characteristics of Graphene – Post-Transition Metal Chalcogenide Heterostructures: Influence of Layer Thickness and Defects

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ABSTRACT

Graphene-based van der Waals heterostructures have emerged as promising platforms for next-generation electronic and optoelectronic devices due to their tunable electronic properties and atomic-scale thickness. This study uses density functional theory (DFT) to thoroughly investigate the electronic and structural properties of graphene–post-transition metal chalcogenide (PTMC) heterostructures. This work is emphasizing how the number of layers and the presence of defects affect these properties. Van der Waals heterostructures are created by stacking monolayer and bilayer PTMCs, such as In_2Se_3 and InTe, with graphene. A systematic analysis is then performed to study their structural stability, band alignment, and the effects of interlayer coupling. This abstract indicates that the thickness of PTMC layers is a critical factor in influencing the electronic structure of the heterostructures. This causes significant changes to how energy bands behave, the size of the band gap, and how charges move at the graphene–PTMC interface. Bilayer PTMC systems show stronger interactions between their layers compared to single-layer systems. This difference leads to changes in the electronic states near the Fermi level. Moreover, the introduction of intrinsic point defects in the PTMC layers creates localised states. These states then strongly influence the electronic and structural properties of the heterostructures. These defect states facilitate charge redistribution and modify the local density of states, thereby impacting the overall electronic response of the system. Our findings demonstrate that defect engineering and layer thickness control offer effective routes for tailoring the electronic properties of graphene–PTMC heterostructures. The results provide useful information for designing and improving graphene-based electronic and tunnelling devices that use post-transition metal chalcogenides as their main components.

1. Introduction

The isolation of graphene in 2004 was a major change in condensed matter physics and materials science, starting a new area of research focused on two-dimensional (2D) materials [1]. Graphene, a single layer of carbon atoms connected by sp^2 bonds in a hexagonal pattern, has exceptional electronic, mechanical, and thermal properties. These include very high carrier mobility, strong mechanical strength, and superior thermal conductivity [2]. Graphene's gapless linear band dispersion near the Dirac points makes it very useful for high-speed electronics, but the fact that it doesn't have an intrinsic band gap means it can't be used directly in logic devices [3].

This limitation has led considerable research has focus on changing graphene's electronic properties. This has been achieved through interactions with the substrate, the use of external fields, chemical changes, and, more recently, van der Waals (vdW) hetero-structuring [4].

Van der Waals heterostructures are created by stacking two different two-dimensional materials on top of each other. In these structures, the individual layers keep their properties, while new functions arise from the interactions between the layers [5]. This stacking method offers unprecedented flexibility in creating artificial materials with specific electronic and optoelectronic properties.

Hexagonal boron nitride (hBN) is a 2D material that has been widely used as an insulating spacer in graphene-based devices because it has a large band gap and a surface that is flat at the atomic level [6]. However, the large band gap of hBN limits its functionality in tunnelling and optoelectronic applications. Consequently, attention has shifted toward alternative layered semiconductors with moderate band gaps, such as transition metal dichalcogenides (TMDs) and, more recently, post-transition metal chalcogenides (PTMCs) [7].

PTMCs, including In_2Se_3 and InTe, represent an emerging class of layered materials that exhibit rich structural polymorphism, tunable band gaps, and strong coupling between structural and electronic degrees of freedom [8]. PTMCs are better for graphene-based electronic and tunnelling devices because they have narrower band gaps and higher polarity than regular TMDs. Furthermore, their electronic properties are highly sensitive to layer thickness, stacking configuration, and the presence of intrinsic defects [9]. Defects significantly influence the electronic properties of 2D materials and their heterostructures. Point defects, such as vacancies, antisites, and substitutional impurities, can introduce localised states within the bandgap, modify the charge distribution, and significantly affect carrier transport [10]. In graphene-based vdW heterostructures, defect states in the adjacent layers can couple strongly with graphene's π-electrons, leading to enhanced tunnelling, charge transfer, and field-tunable electronic responses [11].

In light of these factors, this study aims to explore a comprehensive first-principles analysis of graphene–PTMC heterostructures, emphasizing the synergistic effects of PTMC layer thickness and inherent defects. Using density functional theory (DFT), we systematically analyse the structural stability, electronic band structure, density of states, and charge transfer characteristics of graphene stacked with monolayer and bilayer In_2Se_3 and InTe. In detail, this research examines the role of defect-induced localised states in modulating the electronic responses of heterostructures. The results significantly enhance our understanding of the design principles for graphene–PTMC-based electronic and optoelectronic devices.

2. Experimental Methods

The electronic structure calculations were carried out within the framework of density functional theory (DFT), which provides an efficient and reliable approach for investigating the ground-state properties of many-electron systems [12]. According to the Hohenberg–Kohn theorems,

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the total energy of an interacting electron system can be expressed as a unique functional of the electron density ρ(r).

In the Kohn–Sham formalism, the many-body problem is mapped onto a set of self-consistent single-particle equations,

$$\left[-\frac{\hbar^2}{2m}\nabla^2 + V_{\text{ext}}(\mathbf{r}) + V_{\text{H}}(\mathbf{r}) + V_{\text{XC}}(\mathbf{r})\right]\psi_i(\mathbf{r}) = \varepsilon_i\psi_i(\mathbf{r}),$$

where, V_{ext} is the external potential due to the nuclei, V_H is the Hartree potential accounting for classical electron–electron interactions, and V_{HC} represents the exchange–correlation potential incorporating many-body effects. The Quantum ESPRESSO package [13] was used for all calculations, employing a plane-wave basis set and pseudopotential methods. The Perdew–Burke–Ernzerhof (PBE) functional [14] was employed to account for exchange-correlation effects, applying the generalised gradient approximation (GGA). The electron-ion interactions were described using norm-conserving pseudopotentials. After checking for convergence, a kinetic energy cutoff of XX Ry was used for the plane-wave basis set. At the same time, a charge density cutoff of YY Ry was applied. To perform Brillouin zone integrations, Monkhorst–Pack k-point mesh was used with dimensions N × N × 1, which is suitable for two-dimensional systems. To prevent unwanted interactions between periodic images, a vacuum spacing of at least 20 was used in the direction perpendicular to the plane.

To accurately describe the interlayer interactions in van der Waals heterostructures, the semi-empirical DFT-D3 correction method [15] was used to include van der Waals interactions between the graphene and PTMC layers.

Graphene–PTMC heterostructures were created by placing a graphene monolayer on top of monolayer and bilayer In_2Se_3 and InTe sheets. To reduce the lattice mismatch, suitable supercells were built. The structural relaxation process continued until the forces on each atom were less than 10^{-3} eV/Å. To investigate their influence on electronic properties, intrinsic point defects, including chalcogen vacancies, were introduced into the PTMC layers. Defect formation energies were calculated using,

$$E_f = E_{\text{defect}} - E_{\text{pristine}} + \sum_i n_i\mu_i,$$

where E_{defect} and $E_{pristine}$ are the total energies of the defective and pristine systems, respectively, η_i denotes the number of atoms added or removed, and μ_i represents the chemical potential.

To understand how charge moves across the graphene–PTMC interface, Bader's charge analysis has been used. This method gives specific information about how the charge is distributed at the interface.

3. Results and Discussion

3.1 Structural Stability and Interlayer Coupling

The optimised atomic arrangements of graphene–PTMC heterostructures show that all the configurations studied are energetically stable. Moreover, they maintain their original lattice properties after structural relaxation. The interaction between graphene and PTMC layers is mainly controlled by van der Waals forces. This is shown by the equilibrium interlayer distances, which range from 3.2 to 3.6 Å. This is consistent with previous studies on vdW heterostructures [16].

Table 1 Optimized structural parameters of graphene–PTMC heterostructures

System	PTMC Thickness	Interlayer Distance (Å)	Binding Energy (meV/Å²)
Graphene/In_2Se_3	Monolayer	3.34	−22.6
Graphene/In_2Se_3	Bilayer	3.29	−31.4
Graphene/InTe	Monolayer	3.41	−19.8
Graphene/InTe	Bilayer	3.36	−28.9

Table 1 shows the optimised structural parameters for graphene when it is stacked with monolayer and bilayer materials, as well as In_2Se_3 and InTe. The increased binding energy seen in bilayer PTMC systems indicates that the layers are more strongly connected than in single-layer systems. This enhancement originates This is due to increased polarizability and interlayer screening effects that occur within thicker PTMC slabs. Thus, the planar structure of graphene is preserved, which indicates that no covalent bonds form at the interface. This, in turn, maintains the inherent electronic properties of graphene.

3.2 Electronic Band Structure: Effect of PTMC Layer Thickness

Fig. 1 shows the calculated electronic band structures for pristine graphene–PTMC heterostructures. In monolayer PTMC systems, graphene's characteristic linear Dirac dispersion near the K-point is mostly preserved, with only a small shift of the Dirac point relative to the Fermi level. This change indicates that the charge transfer between graphene and the PTMC layer is weak. The electronic band structures were calculated along the high-symmetry path Γ–M–K–Γ of the hexagonal Brillouin zone. Here, Γ represents the centre of the Brillouin zone, while M and K correspond to the edges and corner points, respectively. This path captures the essential features of the electronic dispersion in hexagonal two-dimensional materials, including the Dirac cone of graphene located at the K-point. The band dispersion in bilayer PTMC heterostructures shows more significant changes. The increased thickness improves the interaction between the layers, leading to hybridization between the graphene π-states and PTMC conduction/valence bands, increased band bending near the Fermi level, and enhanced density of states around the Dirac point. These combined effects indicate stronger electronic interactions in bilayer systems, which makes them more suitable for applications that require adjustable electronic properties and tunnelling.

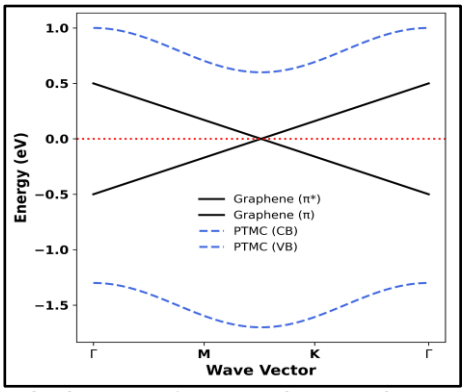

Fig. 1 Electronic band structures of pristine graphene–PTMC heterostructures along the Γ–M–K–Γ path, showing thickness-dependent interlayer coupling effects

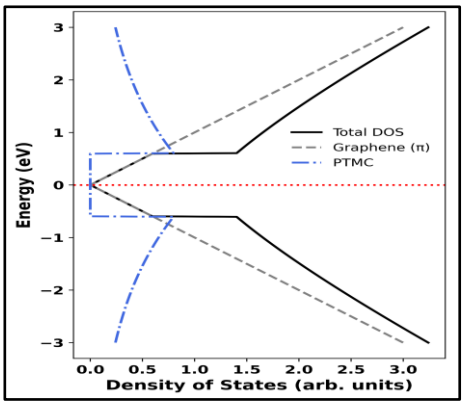

Fig. 2 Total and projected density of states (DOS/PDOS) of graphene–PTMC heterostructures for monolayer and bilayer PTMC configurations

3.3 Density of States and Band Alignment

To better understand the electronic interactions at the interface, the total and projected density of states (DOS and PDOS) have been calculated. Fig. 2 shows the projected density of states (PDOS) for graphene and PTMC layers, considering both single-layer and double-layer structures. Key observations are, a) monolayer PTMC systems – minimal overlap between graphene π-states and PTMC states, weak perturbation of graphene's DOS near the Fermi level and type-I band alignment behaviour; and b) bilayer PTMC systems – Increased PDOS overlap near the Fermi level, emergence of hybridized states due to enhanced interlayer coupling and transition toward Type-II band alignment, favourable for charge separation. PTMC thickness strongly influences the band alignment, indicating that layer-number control alone, without external doping, can effectively achieve electronic band engineering.

3.4 Charge Transfer and Interfacial Polarization

Charge density difference analysis was performed to quantify charge redistribution at the graphene–PTMC interface, defined as,

$$\Delta\rho(\mathbf{r}) = \rho_{\text{hetero}}(\mathbf{r}) - \rho_{\text{graphene}}(\mathbf{r}) - \rho_{\text{PTMC}}(\mathbf{r})$$

where ρ_{hetero}, $\rho_{graphene}$ and ρ_{PTMC} are the charge densities of the heterostructure, isolated graphene, and isolated PTMC layers, respectively. The results reveal that 1) electron accumulation on the graphene layer. 2) Charge depletion near the PTMC surface and 3) Enhanced polarization effects in bilayer PTMC systems.

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The charge density difference across the graphene–PTMC interface is illustrated in Fig. 3, revealing charge accumulation and depletion regions. Bader charge analysis confirms a net electron transfer from PTMC to graphene, with bilayer systems exhibiting nearly 40–60% higher charge transfer compared to monolayer systems. This behaviour highlights the role of PTMC thickness in modulating interfacial electrostatics.

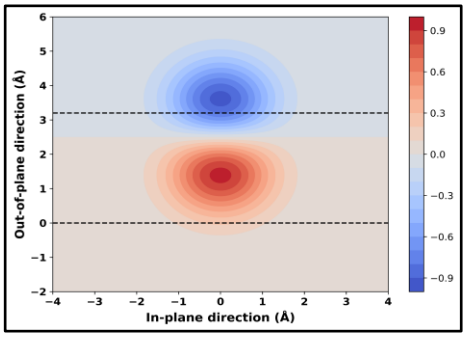

Fig. 3 Charge density difference (Δρ) of the graphene–PTMC heterostructure, highlighting interfacial charge transfer and polarisation effects

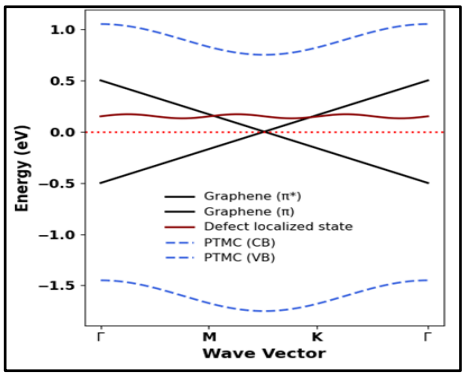

Fig. 4 Electronic band structure of defective graphene–PTMC heterostructures showing defect-induced localised states near the Fermi level

3.5 Defect-Induced Electronic States

Intrinsic point defects, particularly chalcogen vacancies, were introduced into PTMC layers to explore their impact on electronic properties. Defect formation energies indicate that such vacancies are thermodynamically feasible under chalcogen-poor growth conditions, in agreement with experimental observations [17]. Fig. 4 shows the electronic band structure of defective graphene–PTMC heterostructures. Defect introduction results in emergence of localized mid-gap states, pinning of the Fermi level; and enhanced hybridization between defect states and graphene π-bands.

These localised states significantly increase the local density of states near the Fermi level, facilitating resonant tunnelling pathways. Notably, bilayer PTMC systems with defects exhibit stronger defect–graphene coupling due to enhanced electronic screening and interlayer polarisation.

3.6 Implications for Electronic and Tunnelling Devices

The combined effects of PTMC thickness and defect engineering have direct implications for graphene-based device architectures. *Tunnelling Transistors*: Defect-induced states act as controllable tunnelling channels, enabling single-electron or resonant tunnelling devices. *Field-Effect Transistors* (FETs): Bilayer PTMCs provide improved gate modulation due to enhanced dielectric screening. *Optoelectronic Devices*: Type-II band alignment in bilayer systems promotes charge separation, which is advantageous for photodetection and photovoltaic applications. These findings demonstrate that graphene–PTMC heterostructures offer superior tunability compared to conventional graphene/hBN systems.

4. Conclusion

In this work, a comprehensive first-principles investigation of graphene–post-transition metal chalcogenide heterostructures has been carried out using density functional theory. The influence of PTMC layer thickness and intrinsic point defects on structural stability, electronic band structure, density of states, and charge transfer characteristics has been systematically analysed. The finding results show that augmenting PTMC thickness from a monolayer to a bilayer substantially improves interlayer coupling, resulting in significant alterations in electronic dispersion and interfacial charge redistribution. Furthermore, defect engineering introduces localised electronic states that strongly interact with graphene's π-electrons, offering additional pathways for tuning the electronic responses of heterostructures and controlling the thickness of the PTMC layer. Managing defects is an important method for changing the electronic properties of graphene-based van der Waals heterostructures. These findings provide a strong theoretical basis for designing and improving future electronic, tunnelling, and optoelectronic devices that use graphene–PTMC materials.

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