Electronic properties of a two-dimensional van der Waals MoGe₂N₄/MoS₂N₄ heterobilayer: effect of the insertion of a graphene layer and interlayer coupling

D. K. Pham

van der Waals heterostructures (vdWHs) based on 2D layered materials with select properties are paving the way to integration at the atomic scale, and may give rise to new heterostructures exhibiting absolutely novel physics and versatility. Herein, we investigate the structural and contact types in a 2D vdW heterobilayer between MoGe₂N₄ and MoSi₂N₄ monolayers, and the monolayers in the presence of electrical graphene (GR) contact. In the ground state, the MoGe₂N₄/MoS₂N₄ heterobilayer forms type-II band alignment, which effectively promotes the separation of electrons and holes and provides opportunity for further electrons and holes. Thus, the MoGe₂N₄/MoS₂N₄ heterobilayer is promising for designing optoelectronic devices with significantly suppressed carrier-recombination. Interestingly, the insertion of the GR contact to a MoGe₂N₄/MoS₂N₄ heterobilayer gives rise to the formation of a metal/semiconductor contact. Depending on the GR position relative to the MoGe₂N₄/MoS₂N₄ heterobilayer, the GR-based heterostructure can form either an n-type or p-type Schottky contact. Intriguingly, the contact barriers in the GR contacted MoGe₂N₄/MoS₂N₄ heterobilayer are significantly smaller than those in the GR contacted with MoGe₂N₄ or MoSi₂N₄ monolayers, suggesting that the GR/MoGe₂N₄/MoS₂N₄ heterostructure offers an effective pathway to reduce the Schottky barrier, which is highly beneficial for improving the charge injection efficiency of the contact heterostructures. More interestingly, by controlling the interlayer coupling through stacking, both the Schottky barriers and contact types in the GR/MoGe₂N₄/MoS₂N₄ heterostructure can be manipulated. Our findings could provide theoretical insight into the design of nanodevices based on a GR and MoGe₂N₄/MoS₂N₄ heterobilayer.

1. Introduction

Two-dimensional (2D) materials have received extensive interest owing to their intriguing physical and chemical properties and broad device application potential, including in electronics, nanophotonics, optoelectronics and spintronics. A plethora of 2D materials, including graphene (GR), phosphorene, transition metal dichalcogenides (TMDs), and graphitic carbon nitrides, have been synthesized experimentally and predicted theoretically. These 2D materials exhibit outstanding properties and unique advantages over conventional bulk materials, and are particularly promising candidates for designing high-performance nanodevices, such as field-effect transistors and photodetectors.

Very recently, Hong et al. discovered a new family of 2D materials, namely MoSi₂N₄ monolayers, using chemical vapor deposition (CVD). The MoSi₂N₄ monolayer is a layered structure, which can be viewed as a MoN₂ layer sandwiched between two Si–N bilayers. The MoSi₂N₄ monolayer exhibits semiconducting characteristics with a band gap of about 1.94 eV, and possesses high carrier mobility up to 1200 cm² V⁻¹ s⁻¹. This monolayer is also stable and mechanically stronger than most other 2D semiconductors such as MoS₂ monolayers. The successful synthesis of MoSi₂N₄ monolayers has led to a new class of 2D materials with formula MA₂Z₄ where M represents an early transition metal (Mo, W, Cr), A = Si or Ge, and Z = N, P, or As. Motivated by this finding, many theoretical investigations have been performed to explore the electronic, optical and transport properties of MA₂Z₄ monolayers. For instance, Guo et al. studied the electronic properties and transport coefficient of MoSi₂N₄ monolayers alongside the strain effect, and showed that band gap and Seebeck coefficient of such a monolayer are strain-tunable. Wu and colleagues also predicted that the band gap of MoSi₂N₄ (M = Mo, W) monolayers can be altered by strain and electric field, which result in the transformation from indirect to direct band gap semiconductors. Furthermore, MoSi₂N₄, WSi₂N₄ and VSi₂N₄ monolayers are also predicted to be a potential valleytronic...
materials. These ever expanding first-principle calculations of MA₂Z₄ have continually revealed the enormous potential of MA₂Z₄ monolayers for future high-performance device applications.

Currently, constructing 2D van der Waals (vdW) heterostructures between two or more 2D materials is known to be an effective strategy to explore more new properties and extend the potential applications of the corresponding 2D materials. These 2D vdW heterostructures have been predicted theoretically using first-principles calculations, and synthesized experimentally via several methods, such as CVD and mechanical exfoliation. In addition, 2D vdW heterostructures have new desirable properties which could be used for fabricating high-efficiency nanodevices experimentally, such as for solar cells, field-effect transistors (FFTs), and photodetectors. Recently, the combination between MA₂Z₄ monolayers to form MA₂Z₄ bilayers, and between the GR layer and MA₂Z₄ monolayers has received much attention from the scientific community. For instance, Zhong et al. investigated the electronic features of MA₂Z₄ (M = Ti, Cr, Mo; A = Si and Z = N, P) bilayers under the strain effect. They predicted that the electronic properties of the MA₂Z₄ family are tunable with strain-induced transitions from semiconductor to metal. This finding makes the MA₂Z₄ family a potential candidate for fabricating electro-mechanical devices. Similarly, Wu and colleagues showed that applying an electric field can also tune the semiconductor-to-metal transition in both MoSi₂N₄ and WSi₂N₄ bilayers, they are therefore suitable for designing next generation nanoelectronics and optoelectronics. Nonetheless, the combination between MoSi₂N₄ and MoGe₂N₄ to form a heterobilayer, and between graphene and MoSi₂N₄/MoGe₂N₄ heterobilayer to form a GR/MoSi₂N₄/MoGe₂N₄ heterostructure, have not yet been explored thoroughly.

In this work, based on first-principles calculations, we investigate the electronic structures of the MoSi₂N₄/MoGe₂N₄ heterobilayer and explore its electronic properties and band alignment alongside the presence of the GR layer. Various combinations and stacking configurations of GR/MoSi₂N₄/MoGe₂N₄ heterostructures are investigated. Our results reveal that the MoSi₂N₄/MoGe₂N₄ heterobilayer exhibits an indirect band gap semiconductor and possesses type-II band alignment at the equilibrium state. With the presence of the GR layer in the MoSi₂N₄/MoGe₂N₄ heterobilayer, we see the formation of the Schottky contact with narrow barrier height of about 0.26 eV, which increases the carrier injection efficiency.

2. Computational details

In this work, all the calculations, including geometric optimization, electronic properties and band alignment of the MoSi₂N₄/MoGe₂N₄ heterobilayers and the heterostructures between GR and MoSi₂N₄/MoGe₂N₄ heterobilayer, were performed from first-principles calculations within density functional theory, which was implemented in Vienna Ab initio Simulation (VASP) and Quantum Espresso packages. The Perdew–Burke–Ernzerhof (PBE) in the framework of the generalized gradient approximation (GGA) was used to describe the exchange–correlation energy. The projector augmented wave (PAW) pseudopotentials were used to describe the electron-ion interaction. In addition, the DFT-D3 method of Grimme was also selected for describing the presence of the weak vdW forces that always occur in layered vdW systems, including heterobilayers and heterostructures. Furthermore, it should be noted that the traditional DFT method always underestimates the band gap values of materials, especially 2D materials, we therefore used the Heyd–Scuseria–Ernzerhof (HSE06) hybrid functional to avoid this issue and obtain an accurate band gap of the considered materials. The Brillouin zone (BZ) was sampled using the Monkhorst-Pack scheme. A 12 × 12 × 1 (6 × 6 × 1) k-point mesh was used for all the calculations within the DFT-PBE (HSE06) method with a cutoff energy of 510 eV. To avoid the interactions between periodical slabs, we set a large vacuum thickness of 25 Å along the z direction. The geometric optimization was fully obtained with the convergence threshold for energy of 10⁻⁶ eV and for a force of 10⁻² eV Å⁻¹. The spin–orbit coupling (SOC) was also taken into account for the electronic properties of the heterobilayer.

3. Results and discussion

3.1. Structural and electronic properties of MoGe₂N₄/MoSi₂N₄ heterobilayer

The relaxed atomic structures of the MoGe₂N₄/MoSi₂N₄ heterostructure for AA and AB stacking configurations are depicted in Fig. 1. In AA-stacking, both the Ge and N atoms in the MoGe₂N₄ layer are located directly above the Si and N atoms of the MoSi₂N₄ layer, as depicted in Fig. 1(a). Whereas, in AB-stacking, the Ge and N atoms in the MoGe₂N₄ layer are staggered with respect to the Si and N atoms of the MoSi₂N₄ layer, as depicted in Fig. 1(b). After geometric optimization, the interlayer spacings (D) between the MoGe₂N₄ and MoSi₂N₄ layers for the AA and AB stacking configurations are obtained as listed in Table 1. For the AA-stacking, the equilibrium interlayer spacing is 3.33 Å.
Table 1 Optimized lattice constant \(a\) (Å), equilibrium interlayer spacing \(D\) (Å), binding energy \(E_b\) (eV), band gap \(E_g\) (eV) obtained from PBE, HSE and PBE + SOC and band alignment of MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructures for different stacking configurations

<table>
<thead>
<tr>
<th>Stacking</th>
<th>(a)</th>
<th>(D)</th>
<th>(E_b)</th>
<th>Band alignment</th>
</tr>
</thead>
<tbody>
<tr>
<td>AA-stacking</td>
<td>2.96</td>
<td>3.33</td>
<td>-0.11</td>
<td>0.88 1.29 0.87 Type-II</td>
</tr>
<tr>
<td>AB-stacking</td>
<td>2.96</td>
<td>3.35</td>
<td>-0.16</td>
<td>0.90 1.33 0.89 Type-II</td>
</tr>
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Å, and it is 3.35 Å for the AB-stacking configuration. For checking the structural stability of such a heterostructure, we further calculate the binding energy as: \(E_b = E_{1\text{H}} - \Sigma E_{\text{Mi}}\), where \(E_{1\text{H}}\) is the total energy of the combined MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure. \(E_{\text{Mi}}\) is the total energy of the constituent MoGe\(_2\)N\(_4\) and MoSi\(_2\)N\(_4\) monolayers. The binding energy of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer for AA and AB stacking configurations is calculated to be \(-0.11\) and \(-0.16\) eV, respectively. The negative binding energy confirms that the heterobilayer is dynamically stable. In addition, the binding energy of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer for the AB stacking configuration is lower than that for the AA stacking configuration, suggesting that the AB stacking configuration can be considered as the most energetically favorable stacking configuration.

The band structures of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure for different stacking configurations are displayed in Fig. 2. We used different methods, including PBE, HSE and PBE + SOC to calculate the band gaps of the considered heterostructure. The obtained band gaps for the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure are listed in Table 1. For the AA-stacking, the band gaps of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure are calculated as 0.88 eV, 1.29 eV and 0.87 eV. It is clear that the HSE06 method gives rise to a larger band gap value than the PBE and PBE + SOC methods. However, all three methods predict the same characters of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure, which possesses the indirect band gap semiconductor. The valence band maximum (VBM) is located at the \(\Gamma\) point, while the conduction band minimum (CBM) is located at the \(M\) point. Interestingly, when the SOC effect is applied, it tends to split the valence bands at the \(M\) point of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure into two different parts, as depicted in Fig. 2(c). The energy of band splitting for the first and second valence bands of the AA-stacking is calculated to be 13.27 meV and 13.9 meV, respectively. However, the SOC effect splits only the valence bands at the \(M\) point, whereas the VBM of such heterostructure is located at the \(\Gamma\) point. Therefore, the SOC does not affect the band alignments of the heterostructures. Similar trends are also observed in the AB-stacking of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure. The PBE, HSE06 and PBE + SOC band gaps of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure for the AB-stacking are 0.90, 1.33 and 0.89 eV, respectively. The AB-stacking also predicts the indirect band gap semiconductor in the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure with the VBM at the \(\Gamma\) point and the CBM at the \(M\) point, as depicted in Fig. 2(d–f). Furthermore, the band offset is also critical for the optoelectronic application, we therefore calculate the band offsets for valence bands (\(\Delta V\)) and for conduction bands (\(\Delta C\)). The band offsets \(\Delta V\) and \(\Delta C\) for the AA-stacking are calculated to be 0.43 eV and 0.47 eV, respectively. Whereas, these values for AB-stacking are 0.40 eV and 0.43 eV, respectively. In addition, comparing AA and AB stacking, the band structure and alignment seems to be almost unchanged. This is a very convenient behaviour for experimental fabrication since the electronic properties are less affected by stacking configurations.

More interestingly, device performance based on vDW heterostructures depends crucially on the band alignment, forming between two different 2D materials. Depending on the band edge positions of the constituent monolayers, vDW heterostructure can be divided into three different types of band alignment, including type-I (straddling gap), type-II (staggered gap) and type-III (broken gap). To investigate the band alignment, we further calculate weighted band structures of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure, as depicted in Fig. 3. For both the AA-stacking and AB-stacking, one can find that the CBM of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure is mainly contributed by the Mo-orbital states of the MoGe\(_2\)N\(_4\) layer, as marked with green circles. Whereas, the VBM at the \(\Gamma\) point is mainly contributed by the Mo-orbital states of the MoSi\(_2\)N\(_4\) layer, as marked by the violet circles. The contributions by the different monolayers to the VBM and CBM demonstrate that the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure possesses type-II band alignment for both AA- and AB-stacking. The type-II band alignment promotes effective separation of electrons and holes and provides an opportunity for electrons and holes to separate in real space. Thus, the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterostructure is promising for designing optoelectronic devices with significantly suppressed carrier-recombination.

We further investigated the charge mechanism in the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer by calculating the charge
density difference as follows: \( \Delta \rho = \rho_{\text{HB}} - \Sigma \rho_{\text{Mi}} \), where \( \rho_{\text{HB}} \) and \( \rho_{\text{Mi}} \), respectively, are the charge density of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer and the constituent MoGe\(_2\)N\(_4\) and MoSi\(_2\)N\(_4\) monolayers. The charge density difference of the MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer for the AA- and AB stacking is illustrated in Fig. 4. The yellow and cyan regions represent the positive and negative charges, respectively. We find that the charges are mainly visualized at the interface. The yellow regions are located in the MoGe\(_2\)N\(_4\) side, while the cyan regions are in the MoSi\(_2\)N\(_4\) layer. This finding suggests that the charges are mainly accumulated in the MoGe\(_2\)N\(_4\) layer and depleted in the MoSi\(_2\)N\(_4\) layer.

3.2. Electrical graphene contact to a MoGe\(_2\)N\(_4\)/MoSi\(_2\)N\(_4\) heterobilayer

We now consider the formation of a triple-layered 2D-metal/2D-semiconductor heterostructure by including a graphene layer into the MoSi\(_2\)N\(_4\)/MoGe\(_2\)N\(_4\) heterobilayer. The atomic structures of all vDW heterostructures between the GR and MoSi\(_2\)N\(_4\)/MoGe\(_2\)N\(_4\) heterobilayer are depicted in Fig. 5(a–c). Three stacking sequences can be obtained: GR layer placed on top of the MoSi\(_2\)N\(_4\) layer in the MoSi\(_2\)N\(_4\)/MoGe\(_2\)N\(_4\) heterobilayer (Fig. 5(a)); GR layer between the MoSi\(_2\)N\(_4\) and MoGe\(_2\)N\(_4\) layers (Fig. 5(b)); and GR layer located below the MoGe\(_2\)N\(_4\) layer in the MoSi\(_2\)N\(_4\)/MoGe\(_2\)N\(_4\) heterobilayer (Fig. 5(c)). It is clear that the lattice constant of single-layer GR is calculated to be 2.46 Å, while the lattice constant of the MoSi\(_2\)N\(_4\)/MoGe\(_2\)N\(_4\) heterobilayer is calculated to be 2.98 Å. Therefore, in order to avoid the effect of lattice mismatch, we use a supercell, consisting of \( 2 \times \)
MoGe₂N₄ and MoSi₂N₄/MoGe₂N₄/GR heterostructures, we also suggest that the GR, MoSi₂N₄ and MoGe₂N₄ layers in their conducting feature of the MoSi₂N₄/MoGe₂N₄ heterobilayer is also maintained. The band gap of the MoSi₂N₄/MoGe₂N₄ heterobilayer is calculated to be 5.04 Å. Thus, the lattice mismatch between GR and the MoSi₂N₄/MoGe₂N₄ heterobilayer is 2.38%, which is reasonably small and does not affect the main results. The interlayer spacings between the GR, MoSi₂N₄ and MoGe₂N₄ are calculated and listed in Table 2. For the GR/MoSi₂N₄/MoGe₂N₄, the interlayer spacings D₁, D₂ and D₃ are calculated to be 3.58, 13.80 and 3.33 Å, respectively. These values indicate that the interactions between the GR, MoSi₂N₄ and MoGe₂N₄ are weak. In addition, we can see that the interlayer spacings of 3.33 Å and 3.58 Å are comparable with those in other 2D-layered heterostructures, which have typical vdW forces. This finding suggests that the GR, MoSi₂N₄ and MoGe₂N₄ layers in their corresponding heterostructures are bonded together via weak vdW interactions. It should be noted that the systems that are characterized by the vdW interactions are feasible and thus they can be easily fabricated in experiments using several common strategies, such as mechanical exfoliation and chemical vapor deposition (CVD). Furthermore, considering the MoSi₂N₄/GR/MoSi₂N₄ and MoSi₂N₄/MoGe₂N₄/GR heterostructures, we also find that they are characterized by weak vdW forces. Furthermore, in order to guarantee stability, we also perform the ab initio molecular dynamics simulations of such heterostructures and find that they are characterized by weak vdW forces. More interestingly, the contact between metallic GR and the semiconducting MoSi₂N₄/MoGe₂N₄ heterobilayer gives rise to the formation of a 2D metal/2D semiconductor interface. For the 2D metal/2D semiconductor interface, depending on the position of the Fermi level of metallic GR with respect to the band edges of the semiconductor, there is the formation of either Schottky contact (ShC) or Ohmic contact (OhC). One can find from the band structures in Fig. 5 that the Fermi level of the GR layer lies in the band gap region of the MoSi₂N₄/MoGe₂N₄ semiconductor, thus suggesting the formation of the ShC type in all three corresponding GR-based heterostructures. In addition, the Fermi level pinning is still weak in the heterostructures that are characterized by the weak vdW interactions. Therefore, we determine the contact barriers of such heterostructures using the Schottky-Mott rule. The n-type and p-type ShC, respectively, can be defined as \( \Phi_n = E_c - V \) and \( \Phi_p = E_V - E_f \), where \( E_c \), \( E_V \), and \( E_f \), respectively, are the CBM, VBM and Fermi level of the GR-based heterostructure. The calculated \( \Phi_n \) and \( \Phi_p \) of the GR/MoGe₂N₄/MoSi₂N₄, MoGe₂N₄/MoSi₂N₄/GR and MoSi₂N₄/MoGe₂N₄/GR heterostructures are listed in Table 2. We find that the \( \Phi_n \) in the GR/MoGe₂N₄/MoSi₂N₄ heterostructure is still smaller than the \( \Phi_p \), indicating that it forms an n-type ShC at the equilibrium state. Whereas, for the MoGe₂N₄/GR/MoSi₂N₄ and MoGe₂N₄/MoSi₂N₄/GR heterostructures, the \( \Phi_n \) is higher than the \( \Phi_p \), implying that they form the p-type ShC. The \( \Phi_n \) of the GR/MoGe₂N₄/MoSi₂N₄ heterostructure is calculated to be 0.33 eV, which is smaller than that of the GR/MoSi₂N₄/MoGe₂N₄/MoGe₂N₄/GR heterostructures, as illustrated in Fig. 7. The lower the contact barrier, the better the

<table>
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<th>Interlayer spacings</th>
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<td>Heterostructures</td>
<td>D₁, Å</td>
<td>D₂, Å</td>
<td>D₃, Å</td>
<td>E₀, meV per C atom</td>
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<tr>
<td>GR/MoSi₂N₄ (ref. 27)</td>
<td>3.16</td>
<td>—</td>
<td>—</td>
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<td>1.49</td>
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<tr>
<td>GR/MoGe₂N₄ (ref. 27)</td>
<td>—</td>
<td>3.38</td>
<td>—</td>
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<td>24.86</td>
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<td>3.58</td>
<td>13.80</td>
<td>3.33</td>
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<td>3.67</td>
<td>7.37</td>
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<tr>
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<td>3.31</td>
<td>3.45</td>
<td>—</td>
<td>0.47</td>
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</table>
device performance. Intriguingly, the contact barriers in the GR contacted MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer are significantly smaller than those in the GR contacted with MoGe$_2$N$_4$ and MoSi$_2$N$_4$ monolayers, as listed in Table 2. This suggests that the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure offers an effective pathway to reduce the Schottky barrier, which is highly beneficial for improving the charge injection efficiency of contact heterostructures.

The tuning of the contact barriers and contact types in GR-based vdW heterostructures is an important factor of nano-devices based on the GR heterostructures. Controlling the interlayer coupling through stacking is one of the most efficient strategies in tuning both the contact barriers and contact types of GR-based heterostructures. The interlayer coupling can be manipulated by changing the interlayer spacings between the GR and semiconductors in their corresponding heterostructures. One can observe from Table 2 that the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ has the lowest binding energy of $-67.66$ meV per C atom in comparison with that of other configurations, as it is the most stable configuration. Therefore, the effects of the interlayer couplings on the contact barriers and contact types of the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure are investigated as an illustrative example. The projected band structures of the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure as well as the variation in the contact barriers as functions of the interlayer couplings are displayed in Fig. 8. Red and black lines represent the contributions of the metallic GR and semiconducting MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer, respectively.

Interestingly, we find that changing interlayer couplings gives rise to a change not only in the Schottky barriers but also in the contact types of the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure. The changes in the Schottky barriers and contact types in the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure are illustrated in Fig. 8(a). The $\Phi_n$ decreases with increasing interlayer spacing, while the $\Phi_p$ increases accordingly. On the contrary, the $\Phi_n$ increases with decreasing the interlayer spacing, while the $\Phi_p$ decreases accordingly. When the $\Delta D$ is smaller than $-0.45$ Å, the $\Phi_n$ becomes larger than the $\Phi_p$, leading to the transition from n-type ShC to p-type. The physical mechanism of such transitions can be described by analyzing the change in the Fermi level relative to the band edges of the semiconducting MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer. With the applications of the compressive strain, i.e. $\Delta D < 0$, the Fermi level of graphene moves towards the VBM of the semiconducting MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer, thus resulting in an increase in the $\Phi_n/\Phi_p$. On the other hand, with the application of the tensile strain, i.e. $\Delta D > 0$, the Fermi level of graphene shifts upwards from the VBM to the CBM of the semiconducting MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer. Thus, the $\Phi_n$ is decreased and the $\Phi_p$ is increased accordingly. In this case, the heterostructure remains as n-type ShC. It can be concluded that with the application of different strains, both the Schottky barriers and contact types in the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure can be manipulated.
MoGe$_2$N$_4$ and MoSi$_2$N$_4$ monolayers, suggesting that the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure offers an effective pathway to reduce the Schottky barrier, which is highly beneficial for improving the charge injection efficiency of the contact heterostructures. More interesting, by controlling the interlayer coupling through stacking, both the Schottky barriers and contact types in the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure can be manipulated.

### Conflicts of interest

There are no conflicts to declare.

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### References


### 4. Conclusions

In summary, we have investigated the structural and contact types in a 2D vdW heterobilayer between MoGe$_2$N$_4$ and MoSi$_2$N$_4$ monolayers, as well as in the presence of electrical graphene. We found that the MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer forms type-II band alignment, which effectively promotes the separation of electrons and holes, and provides an opportunity for further electrons and holes. Interestingly, the insertion of the electrical graphene contact to a MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer gives rise to the formation of a metal/semiconductor contact, which is characterized by the contact type barriers. Depending on the graphene position relative to the MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer, the graphene-based heterostructure can form either an n-type or p-type Schottky contact. The graphene/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure exhibits an n-type Schottky contact, whereas the MoGe$_2$N$_4$/GR/MoSi$_2$N$_4$ and MoGe$_2$N$_4$/MoSi$_2$N$_4$/GR heterostructures form a p-type one. Intriguingly, the contact barriers in the GR contacted MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterobilayer are significantly smaller than those in the GR contacted MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure as a function of the interlayer couplings.

**Fig. 8** (a) The variation of the Schottky barriers of the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure as a function of the interlayer couplings. Projected band structures of the GR/MoGe$_2$N$_4$/MoSi$_2$N$_4$ heterostructure under (b) compressive and (c) tensile strains.