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Van der Waals graphene/MoS$_2$ heterostructure: Tuning the electronic properties and Schottky barrier by biaxial strain

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ABSTRACT: First principles calculations are performed to study the effects of the interlayer distance and biaxial strain on electronic properties and contact properties of graphene/MoS$_2$ heterostructures. The interlayer interaction is weakened and the charge transfer from the graphene layer to MoS$_2$ layer is reduced with increasing interlayer distance in graphene/MoS$_2$ heterostructures, resulting in shift of Fermi level to a high energy state. The n-type Schottky barrier is formed with $\Phi_{SB,N}$ are 0.647 eV, 0.568 eV, 0.509 eV, and 0.418 eV when the interlayer distances are 3.209 Å, 3.346 Å, 3.482 Å, and 3.755 Å, respectively. The interlayer distance and charge density difference change slightly, but the electronic structure of the graphene/MoS$_2$ heterostructure changes obviously by applying the biaxial strain. For the biaxial strain from $-4\%$ to $+6\%$, the $\Phi_{SB,P}$ gradually increases for the graphene/MoS$_2$ heterostructure, while the $\Phi_{SB,N}$ increases initially and then decreases. Moreover, the $\Phi_{SB,N}$ is only 0.080 eV under the biaxial strain of $+6\%$, indicating the Ohmic contact nearly forms. The results demonstrate the significant effects of biaxial strain on the physical properties of 2D heterostructures.

Keywords: Graphene; MoS$_2$; biaxial strain; electronic properties; Schottky barrier
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1. Introduction

Since the discovery of graphene in 2004, its unique physics and novel applications have triggered extensive research.\textsuperscript{1-3} Although the extremely high electrical conductivity makes graphene a potential candidate material to replace silicon–based electronic, Klein tunneling causes the electrical transport of Dirac fermions insensitive to the electrostatic potentials, which leads to a low current on/off ratio of graphene–based field effect transistors.\textsuperscript{4-6} In order to realize graphene electronics, it is very important to manipulate the electronic properties without impairing its high mobility.

An important milestone is the creation of heterostructures based on graphene and other two dimensional (2D) materials, which can be assembled into three dimensional stacks with atomic layer precision.\textsuperscript{7-9} Such layered structures have already demonstrated a range of fascinating physical phenomena. Wang et al. found that for MoS\textsubscript{2}/ZnO heterostructure the strong optical absorption in the visible region, indicating that it has potential for application in photovoltaic and photocatalytic devices.\textsuperscript{10} The type-II band alignment occurs at MX\textsubscript{2}/graphene-like zinc oxide interfaces, together with the large built-in electric filed across the interface.\textsuperscript{11,12} Due to the inherent weak absorption characteristics and small built-in potential of 2D materials photodetectors, their external quantum efficiency is severely limited to the range of \(-0.1-1\%\).\textsuperscript{13,14} Duan et al had modulated the amplitude and polarity of photocurrent in the gated vertical graphene/MoS\textsubscript{2} heterostructures by the electric field of an external gate. The maximum external quantum efficiency and internal quantum efficiency are estimated to be 55\% and 85\%, respectively.\textsuperscript{15} Moreover, graphene/MoS\textsubscript{2} heterostructures display the highly sensitive photodetection and gate tunable persistent photoconductivity.\textsuperscript{16} The responsivity of the heterostructures is nearly \(1\times10^{10}\) AW\textsuperscript{-1} at 130 K and \(5\times10^{8}\) AW\textsuperscript{-1} at room temperature. The heterostructures could also function as a rewritable optoelectronic switch or memory when irradiated by the time-dependent photoillumination, where the persistent state shows almost no relaxation or decay within experiment
timescales, indicating near-perfect charge retention.

Compared with metal/2D material heterostructures, the use of 2D materials to construct van der Waals vertically stacked heterostructures is one of the effective and feasible ways to reduce the effect of Fermi level pinning. Graphene has potential applications in 2D transistors due to its excellent electrical conductivity. For example, the high-performance devices and circuits based on graphene/MoS$_2$ heterostructures, where MoS$_2$ is used as the transistor channel and graphene as contact electrodes.$^{17}$ As well all know, there are two mechanisms by which charge can be injected into a semiconductor: thermionic emission over the semiconductor and field emission across the semiconductor. The thermionic emission-diffusion theory describes the current-voltage characteristics of a metal/semiconductor heterostructure as a function of Schottky barrier height. Carrier recombination can also be a current-limiting process if an inversion layer is present near the contact. Contrary to the bulk case, where the diffusion region extends both laterally and vertically into the semiconductor, in a metal/2D semiconductor heterostructure with no hybridization, the position of the bands vary only laterally, so that charge carriers injected far from the contact edge first encounter the flat-band region before the diffusion region. In this case, the relative contributions from thermionic emission and tunnelling become difficult to predict. On the basis of a thermionic field emission model, the barrier height at the graphene/MoS$_2$ heterostructure was determined to be 0.23 eV and the tunability of graphene work function with electrostatic doping significantly improves the Ohmic contact.$^{18}$ Wei-Qing Huang et al.$^{19}$ found that the transformation from n-type Schottky barrier to p-type Schottky barrier can be realized in MoS$_x$Se$_{2-x}$/graphene heterostructures when the Se concentration is greater than 25%. Interestingly, the Schottky type, Schottky barrier height, and contact types at interface can be tuned by external electric field.$^{19,20}$ On the other hand, because of their excellent Young’s modulus, graphene and MoS$_2$ holds promise for applications in flexible electronic devices. A highly flexible transistor was developed based on an exfoliated MoS$_2$ channel and CVD–grown graphene source/drain electrodes, and a low Schottky barrier (~22 meV) forms.$^{21–24}$ The graphene/MoS$_2$
heterostructures exhibit a Young’s modulus that is about three times that of monolayer MoS₂, while correspondingly exhibiting a yield strain that is about 30%–40% smaller than that of monolayer MoS₂ due to lateral buckling of the outer graphene layers. Zhou et al. found that the tensile strain can enhance the optical absorption in the visible range and increase the solar energy conversion efficiency for 2D heterostructures. Moreover, with appropriate compressive strain of 2% and 3%, the WTe₂–As heterostructures show transition from type-I to type-II band alignment, which could slow down electron-hole pair recombination. However, up to date the effects of the strain on the structural and electronic properties of the graphene/MoS₂ heterostructures have not yet been studied systematically.

Herein, first principles calculations are done to systematically study the effects of the interlayer distance and biaxial strain on the electronic structure and contact properties of the graphene/MoS₂ heterostructures. Although the charge transfer from the graphene layer to MoS₂ layer, the electronic band structure seems to be a simple sum of those of each constituent when graphene stacks on top of MoS₂ layer. And the contact properties are insensitive to the interlayer distance. While the biaxial strain engineering is a valuable method to modulate electronic structure and contact properties of the graphene/MoS₂ heterostructures.

2. Computational method

The all calculations are performed by VASP based on the density function theory. The electron–ion core interaction and the exchange–correlation interaction are described by the projector augmented wave potentials and the Perdew–Burke–Ernzerhof formulation of the generalized gradient approximation, respectively. The DFT–D2 approach in the Grimme scheme is adopted to include the contribution from the vdW interaction between layers. The cutoff energy for the plane–waves is chosen to be 400 eV. According to the Monkhorst–Pack scheme, the Brillouin–zone integration is performed by using an 11×11×1 k–mesh and Gaussian
smeared broadening of 0.2 eV is adopted. In order to relax the ions to the ground states with an
energy convergence of $1.0 \times 10^{-5}$ eV and a force convergence of 0.02 eV/Å on each ion, a
conjugate-gradient algorithm is employed. Visualizations of supercells and structures are done
with the software VESTA.$^{35}$

The thermodynamic stability of graphene/MoS$_2$ heterostructures is evaluated by calculating the
binding energy ($E_b$) as shown in the following equation:

$$E_b = \frac{E_{\text{graphene/MoS}_2} - (E_{\text{graphene}} + E_{\text{MoS}_2})}{N_C},$$

(1)

where $E_b$, and $E_{\text{graphene/MoS}_2}$, $E_{\text{graphene}}$, and $E_{\text{MoS}_2}$ are the binding energy, and the total energy of
the graphene/MoS$_2$ heterostructure, graphene and MoS$_2$ monolayers, respectively. $N_C$ is the
number of carbon atoms in the supercell. The negative $E_b$ is, the system is stable. And the
larger absolute value of the $E_b$ value is, the stronger the heterostructure binding is. To gain
further insight into the bonding nature and interlayer interaction, the plane-averaged charge density
difference ($\Delta \rho$) was calculated as shown in the following equation:

$$\Delta \rho = \rho_{\text{graphene/MoS}_2} - \rho_{\text{graphene}} - \rho_{\text{MoS}_2},$$

(2)

in which $\rho_{\text{graphene/MoS}_2}$, $\rho_{\text{graphene}}$, and $\rho_{\text{MoS}_2}$ are the plane-averaged charge densities of the
graphene/MoS$_2$ heterostructure, graphene and MoS$_2$ monolayers, respectively.

3. Results and discussion

3.1 Graphene/MoS$_2$ heterostructure

The optimized lattice constants of monolayer graphene and MoS$_2$ are 2.460 Å and 3.160 Å [as
shown in Fig. 1 (a,b)]. To limit the lattice mismatch between graphene and MoS$_2$ in our
calculations, the simulation cell is built from a 5×5 graphene supercell and a 4×4 MoS$_2$ supercell
[Fig. 1 (c)]. The lattice constant of the graphene/MoS$_2$ heterostructure fixed as that of MoS$_2$ and
change the lattice constant of graphene to adjust to those of MoS$_2$, where graphene is under a
biauxial tensile strain of 2.69% and no significant changes on the electronic properties in the heterostructure. Additional, the previous work indicated that the structural and electronic properties of the graphene/MoS$_2$ heterostructure are independent of the atomic configuration.$^{36}$ Thus, the other atomic configuration of graphene adsorbs on MoS$_2$ is not taken into account.

The equilibrium structure is yielded by applying the two-step optimization on the designed lattice. For step one, the interlayer distance is changed and the binding energy was calculated, as well as the results were fitted into the well-known Buckingham potential equation:

$$E_b = A e^{-Bd} - \frac{C}{d^6},$$

in which $A$, $B$, and $C$ are fitting parameters of $-7.492$ eV, $1.158$ Å, and $-136.063$ eV·Å, respectively.$^6$ $d$ and $E_b$ are the interlayer distance and the binding energy, respectively. The binding energy as a function of the interlayer distance and fitting curve is shown in Fig. 1 (d). The predicted equilibrium interlayer distance read from the fitting curve is about $3.410$ Å. For step two, a structure with an interlayer distance of $3.410$ Å is constructed and placed into a full relaxation to yield the final equilibrium structure. After optimization, the interlayer distance between the monolayer MoS$_2$ and graphene is $3.414$ Å, with a binding energy of $-39$ meV per C atom, which is of the same order of magnitude as that of other heterostructures such as MoS$_2$/graphene,$^{37}$ black phosphorene/graphene,$^{38-41}$ and arsenene/graphene.$^{42,43}$

Next, the calculated electronic band structure of graphene/MoS$_2$ heterostructure is displayed in Fig. 2 (c). For comparison, the electronic band structures of pristine graphene and isolated MoS$_2$ monolayers are also shown in Fig. 2 (a,b). For graphene/MoS$_2$ heterostructure, the electronic band structure seems to be a simple sum of those of each constituent. The linear dispersion bands of graphene appear in the large energy gap of MoS$_2$, and the character of the electronic band structure of pristine graphene seems to be preserved, indicating the weak interaction between graphene and MoS$_2$. In Fig. 3 (a), the band alignment is schematically demonstrated to show the formation of interlayer excitons. In particular, excited electrons from graphene migrate to MoS$_2$, while holes go
oppositely owing to the difference between their the Fermi levels, and the electrons and holes can be held together by strong Coulomb interactions. The charge difference between the graphene/MoS\(_2\) heterostructure and the constituents. As expected, the charge redistribution mainly occurs at the interface between the layers, with an accumulation of electrons on the MoS\(_2\) layer and a depletion of charges on the graphene layer [Fig. 3 (b,c)]. Furthermore, it can be found that the potential of graphene (8.881 eV) in the graphene/MoS\(_2\) heterostructure is deepened as shown in Fig. 3 (d). The large potential drop of the graphene/MoS\(_2\) heterostructure indicates a strong electronic field across the interface, which may affect the kinetics of photo-generated carriers.\(^{44}\)**

### 3.2 Effects of interlayer distance on Graphene/MoS\(_2\) heterostructure

Charging the interlayer distance is an effective approach to tune the electronic properties of the van der Waals heterostructure. Moreover, the interlayer distance in the van der Waals heterostructure can be easily controlled in experiments by vacuum thermal annealing, nanomechanical pressure, or diamond anvil cell.\(^{45-47}\) Thus, it is necessary to consider the influence of interlayer distance on the electronic properties of the graphene/MoS\(_2\) heterostructure.

Fig. 4 displays the charge density difference of the graphene/MoS\(_2\) heterostructure as the interlayer distance changes. From Fig. 4 (a), one can observe that the fluctuation of differential charge density curve increases when the interlayer distance is reduced from 3.755 Å to 3.209 Å, indicating the more charge transfers from graphene layer to MoS\(_2\) layer. Moreover, according to Mulliken charge analysis, 0.120 e, 0.107 e, 0.088 e, and 0.062 e transfer from graphene layer to MoS\(_2\) layer when the interlayer distances are 3.209 Å, 3.346 Å, 3.482 Å, and 3.755 Å, respectively. The Fermi level of the graphene shifts downwards due to its charge transfers to MoS\(_2\) layer. While the electronic band structures of pristine graphene and MoS\(_2\) monolayers are preserved. The band gaps of the MoS\(_2\) layer in heterostructure are 1.747 eV, 1.737 eV, 1.747 eV, and 1.757 eV when the interlayer distances are 3.209 Å, 3.346 Å, 3.482 Å, and 3.755 Å, respectively (Fig. 5). All these results indicate that the interlayer distance is effective to control the charge transferring, while limited to regulate the electronic structure.
3.3 Effects of biaxial strain on Graphene/MoS$_2$ heterostructure

It is worth noting that the electronic properties of MoS$_2$ monolayer are very sensitive to the strain.$^{48,49}$ The electronic properties of the strained transition metal dichalcogenides have been invested both experimentally and theoretically in which the monolayer MoS$_2$ can deform up to 11%. The flexible property gives an engineering so-called “straintronics”. The direct to indirect gap transition in MoS$_2$ occurs at 2.7% of the biaxial strain.$^{50}$ Wang et al.$^{51}$ found that the band gap monotonically decreases with increasing the strain. Moreover, the conduction band minimum shifts toward the Fermi level, leading to semiconductor to metal transformation at 10% of the biaxial strain.$^{52}$ To investigate the effect of strain on the properties of the heterostructure, the graphene/MoS$_2$ heterostructure under $xy$–plane biaxial compressive and tensile strain is studied.

The biaxial strain is applied along the $xy$–plane by varying the lattice constant of the heterostructure and can be defined as follows:

$$\varepsilon = (a-a_0)/a,$$

where $a$ and $a_0$ are the equilibrium and strained lattice constant, respectively. One can observe that under the varying biaxial strain from $-4\%$ to $+6\%$, the interlayer distance and bond lengths of C–C in graphene layer and Mo–S in MoS$_2$ layer increase. Particularly, the interlayer distances of graphene/MoS$_2$ heterostructure are 3.246 Å, 3.386 Å, 3.423 Å, and 3.453 Å under the biaxial strain of $-4\%$, $-2\%$, $+2\%$, and $+6\%$, respectively. Moreover, when biaxial compressive strain is applied the binding energy maintains negative value. On the contrary, when biaxial tensile strain is applied the binding energy changes to positive value, indicating the weaken interaction between graphene and MoS$_2$.

Fig. 6 shows the charge density difference of the graphene/MoS$_2$ heterostructure under the biaxial strain. It is found that the fluctuation of differential charge density curve decreases when the biaxial strain changes from $-4\%$ to $+6\%$, indicating the less charge transfers from graphene layer to MoS$_2$ layer [Fig. 6 (a)]. The same phenomenon can observe through the three dimensional isosurface of the charge density difference of the graphene/MoS$_2$ heterostructure as shown in Fig. 6.
According to Mulliken charge analysis, 0.062 e, 0.059 e, 0.055 e, and 0.051 e are transferred from graphene layer to MoS$_2$ layer under the biaxial strain of $-4\%$, $-2\%$, $+2\%$, and $+6\%$, respectively. Thus, by applying biaxial strain, the interlayer distance and charge density difference change slightly. While there is a obviously change for the electronic structure of the graphene/MoS$_2$ heterostructure by applying the biaxial strain. Although the electronic band structures of pristine graphene is still preserved, the band structure of MoS$_2$ substantial changes under the biaxial strain. In particularly, the band gaps of the MoS$_2$ layer in heterostructure are 1.669 eV, 1.806 eV, 1.352 eV, and 0.618 eV under the biaxial strain of $-4\%$, $-2\%$, $+2\%$, and $+6\%$, respectively (Fig. 7).

It is obvious that the graphene/MoS$_2$ heterostructure is characterized the metal/semiconductor heterostructure. In the graphene/MoS$_2$ heterostructure, the n-type Schottky barrier heigh ($\Phi_{SB,N}$) is $\Phi_{SB,N} = E_{CBM} - E_F$, whereas the p-type Schottky barrier height ($\Phi_{SB,P}$) is $\Phi_{SB,P} = E_F - E_{VBM}$. The $E_{CBM}$, $E_{VBM}$, and $E_F$ represent the conduction band minimum, valence band maximum, and Fermi level, respectively. Fig. 8 displays the evolution of the n− and p−type Schottky barrier height ($\Phi_{SB,N}$ and $\Phi_{SB,P}$) of the graphene/MoS$_2$ heterostructure at the various interlayer distance and under the biaxial strain. When the interlayer distance is reduced, the $\Phi_{SB,N}$ gradually increases, but $\Phi_{SB,P}$ gradually decreases. Moreover, the value of $\Phi_{SB,P}$ is always larger that that of $\Phi_{SB,N}$, indicating n−type Schottky contact. The $\Phi_{SB,N}$ are 0.647 eV, 0.568 eV, 0.509 eV, and 0.418 eV when the interlayer distances are 3.209 Å, 3.346 Å, 3.482 Å, and 3.755 Å, respectively [Fig. 8 (a)]. For applying the biaxial strain from $-4\%$ to $+6\%$, the $\Phi_{SB,P}$ gradually increases for the graphene/MoS$_2$ heterostructure, while the $\Phi_{SB,N}$ increases initially and then decreases [Fig. 8 (b)]. Furthermore, the $\Phi_{SB,N}$ is only 0.080 eV under the biaxial strain of $+6\%$, indicating the Ohmic contact nearly forms.
4. Conclusions

First principles calculations are performed to study the effects of the biaxial strain on electronic properties and Schottky barrier of graphene/MoS\(_2\) heterostructures. It is found that the interlayer interaction is weakened and the charge transfer from the graphene layer to MoS\(_2\) layer is reduced with increasing interlayer distance, resulting in shift of Fermi level to a high energy state. The n-type Schottky barrier is formed with \(\Phi_{SB,N}\) are 0.647 eV, 0.568 eV, 0.509 eV, and 0.418 eV when the interlayer distances are 3.209 Å, 3.346 Å, 3.482 Å, and 3.755 Å, respectively. The interlayer distance and charge density difference change slightly, while there is a obviously change for the electronic structure of the graphene/MoS\(_2\) heterostructure by applying the biaxial strain. For the biaxial strain from −4% to +6%, the \(\Phi_{SB,P}\) gradually increases for the graphene/MoS\(_2\) heterostructure, while the \(\Phi_{SB,N}\) increases initially and then decreases. Furthermore, the \(\Phi_{SB,N}\) is only 0.080 eV under the biaxial strain of +6%, indicating the Ohmic contact nearly forms. Our results provide a detailed understanding of the interfacial properties of graphene/MoS\(_2\) heterostructures and help to predict the performance of 2D materials-based devices.

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Tables

Table 1. Interlayer distance $d$ (Å), bond lengths of $C-C$ $L_{C-C}$ (Å) and $Mo-S$ $L_{Mo-S}$ (Å), and binding energy $E_b$ (meV/C) of the graphene/MoS$_2$ heterostructure under the biaxial strain.

| Strain (%) | $d$ (Å) | $L_{C-C}$ (Å) | $L_{Mo-S}$ (Å) | $E_b$ (meV/C) |
|------------|---------|---------------|---------------|--------------|
| -4         | 3.246   | 1.402         | 2.381         | -10          |
| -2         | 3.386   | 1.430         | 2.393         | -55          |
| 0          | 3.414   | 1.460         | 2.406         | -39          |
| +2         | 3.423   | 1.489         | 2.421         | 47           |
| +4         | 3.437   | 1.518         | 2.436         | 195          |
| +6         | 3.453   | 1.547         | 2.453         | 397          |
Figure Captions

Fig. 1. Optimized geometries for top view of (a) graphene and (b) MoS$_2$ monolayers, and (c) graphene/MoS$_2$ heterostructure. (d) Binding energy of graphene per C as a function of the interlayer distance between graphene and the topmost S atom of MoS$_2$ for graphene/MoS$_2$ heterostructure.

Fig. 2. Band structures of (a) graphene and (b) MoS$_2$ monolayers, as well as (c) graphene/MoS$_2$ heterostructure. The Fermi level is indicated by the green line.

Fig. 3. (a) Schematic representation of the band alignment for graphene and MoS$_2$ monolayers, as well as graphene/MoS$_2$ heterostructure. (b) Plane averaged charge density differences, (c) three dimensional isosurface of the charge density difference, and (d) plane averaged effective potential of the graphene/MoS$_2$ heterostructure. The red and green areas represent electrons accumulation and depletion, respectively, and the isosurface value is 2×10$^{-4}$ e/Å$^3$.

Fig. 4. (a) Plane averaged charge density difference and (b) three dimensional isosurface of the charge density difference of the graphene/MoS$_2$ heterostructure at the various interlayer distance.

Fig. 5. Band structures of the graphene/MoS$_2$ heterostructure at the various interlayer distance: (a) 3.209 Å, (b) 3.346 Å, (c) 3.482 Å, and (d) 3.755 Å.

Fig. 6. (a) Plane averaged charge density differences and (b) three dimensional isosurface of the charge density difference of the graphene/MoS$_2$ heterostructure under the biaxial strain.

Fig. 7. Band structures of the graphene/MoS$_2$ heterostructure under the biaxial strain: (a) −4%, (b) −2%, (c) +2%, and (d) +6%.

Fig. 8. The evolution of the n– and p–type Schottky barrier height ($\Phi_{SB,N}$ and $\Phi_{SB,P}$) of the graphene/MoS$_2$ heterostructure: (a) at the various interlayer distance and (b) under the biaxial strain.
Fig. 1
Fig. 2
Fig. 3
Fig. 4
Fig. 5
Fig. 7
Fig. 8