

Review

Interfacing 2D Semiconductors with Functional Oxides: Fundamentals, Properties, and Applications

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Abstract: Two-dimensional semiconductors, such as transition-metal dichalcogenides (TMDs) and black phosphorous (BP), have found various potential applications in electronic and opto-electronic devices. However, several problems including low carrier mobility and low photoluminescence efficiencies still limit the performance of these devices. Interfacing 2D semiconductors with functional oxides provides a way to address the problems by overcoming the intrinsic limitations of 2D semiconductors and offering them multiple functionalities with various mechanisms. In this review, we first focus on the physical effects of various types of functional oxides on 2D semiconductors, mostly on MoS₂ and BP as they are the intensively studied 2D semiconductors. Insulating, semiconducting, conventional piezoelectric, strongly correlated, and magnetic oxides are discussed. Then we introduce the applications of these 2D semiconductors/functional oxides systems in field-effect devices, nonvolatile memory, and photosensing. Finally, we discuss the perspectives and challenges within this research field. Our review provides a comprehensive understanding of 2D semiconductors/functional oxide heterostructures, and could inspire novel ideas in interface engineering to improve the performance of 2D semiconductor devices.

Keywords: two-dimensional semiconductors; functional oxides; interface; heterostructures

1. Introduction

Dielectric materials are essential components in modern electronics. In field-effect transistors (FETs), the electric on/off states of semiconducting channel materials are switched by a gate voltage applied through dielectrics. This has been a fundamental use of various FET-based devices in large-scale logic circuits. Most traditional dielectrics are made of metal or non-metal oxides. Their properties could drastically influence the gating efficiency of FET-based devices, especially when the channel is a nanometer-thick material.

Monolayer or few-layer two-dimensional (2D) semiconductors are promising channel materials in field-effect devices due to their very large surface-to-volume ratio and ultrathin body thickness. They usually have novel electronic and optical band structures, high carrier mobility, and extremely strong light-matter interactions that enable their applications in electronic and optoelectronic devices. They also possess an atomic level of surface roughness and anisotropic physical properties (in-plane versus out-of-plane) because of strong in-plane covalent bonds and weak van der Waals (vdW) interlayer interactions [1–3]. Because of their ultrathin nature, however, monolayer or few-layer 2D semiconductors must be placed on a solid substrate in electronics. Currently, most of these substrates are dielectric oxides, such as SiO₂, Al₂O₃, and mica. Interface interactions between 2D

semiconductors and substrates must be considered. Besides the traditional dielectric screening effect, several new effects of oxide substrates, including charge transfer, energy transfer, and strain, have emerged and may separately or co-functionally dominate the properties of overlayer 2D semiconductors [4,5].

Challenges still exist in the present 2D semiconductor devices. For example, molybdenum disulfide (MoS_2) and black phosphorous (BP) have already been integrated into nano-electronic devices, but problems like low carrier mobility and low photoluminescence (PL) efficiency limit their further applications in electronics and opto-electronics. As Moore's law is still proven to be effective, FET devices scale down in size very rapidly. When the leakage current through dielectric oxides caused by quantum mechanical tunneling becomes unacceptable, high- κ oxide materials, such as ZrO_2 , HfO_2 , and Ta_2O_5 , are expected to replace traditional SiO_2 as insulating gate layers [6,7]. Such high- κ materials also prove to be successful in enhancing the carrier mobility of channels because their strong dielectric screening effectively reduces the interface impurity scattering. On the other hand, multiple functions are also desirable in electronic devices for their miniaturization. This purpose could be achieved by either a multiple functional channel material or a functional substrate that modulates the channel in multiple ways. In this regard, many functional oxides may offer unique properties such as piezoelectricity, strong polarization, and spin injection. When 2D semiconductors interface with functional oxides, not only can the band gap be manipulated, but also multi-functional, co-operative devices could be built to output new features through coupling. These aspects may open up a new route to modulate novel 2D devices with interface engineering by functional oxides.

In this review, we first focus on the physical effects of various types of functional oxides on 2D semiconductors. Insulating, semiconducting, conventional piezoelectric, strongly correlated, and magnetic oxides are discussed. Then we introduce the applications of these 2D semiconductors/functional oxide systems in FET devices, nonvolatile memory, and photosensing. Finally, we discuss the perspectives and challenges in this research field.

2. Physical Effects of Oxides on 2D Semiconductors

2.1. 2D Semiconductors/Traditional Dielectric Material Heterostructures

Most traditional dielectrics are made of metal oxides and non-metal oxides. The dielectric layers in transistors are indispensable and control the electric on-off states of channel materials. In 2D semiconductor devices, different dielectric environments will influence the electrical and optical properties of the 2D channels. At the same time, charge traps at the interface and doping effect also have a significant impact on their physical properties.

Carrier mobility is a key parameter in determining the electrical properties of a channel material. When the channel length gets smaller, the thickness of a traditional silicon channel in a FET must also be reduced. The surface roughness and dangling bonds in silicon will affect the electrical transport and cause a degradation in mobility [8]. In 2D semiconductors, it is believed that the interlayer vdW force is weak and small defects will lead to less scattering and better electrical conduction. However, the observed mobility is only $0.1\text{--}10\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ after exfoliation [9,10], compared with $200\text{--}500\text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ in bulk MoS_2 [11]. One possible explanation is that the intrinsic phonon scattering is the main limiting factor in bulk materials. In ultrathin 2D layers, however, charge traps and impurities at the interface may become additional scattering centers, hindering their thermal and electrical transport [12,13]. Charge traps are found to have a strong relationship with the carrier mobility and the series resistances of an electronic device [14]. According to the study of Ayari et al., the concentration of trap states changes the overall conductance, and calculations show that interfacial charge traps, instead of bulk charge traps, play a dominant role [15]. When charge traps exist at the interface, electrons might be accumulated or reduced to form a screening layer [16]. This suggests that the carrier type and screening length would affect the electrical conduction at the same time. High

dielectric materials, such as HfO_2 , have been used to reduce the Coulomb force by dielectric screening to improve carrier mobility [9,11].

Several factors may function together to determine the overall carrier mobility. Besides intrinsic phonons, the so-called ‘remote’ phonons also exist in dielectric materials. They are polar-optical-phonon modes that can be excited by electrons in semiconductors through long-range Coulombic interactions [17]. As semiconductors become thinner, the interactions become stronger. For 2D semiconductors, intrinsic phonons and remote phonons in the dielectrics and impurities together influence the mobility [18]. The remote phonons become dominant at room temperature although they have minor impacts at low temperature. In the case of high-density impurities at the interface, impurity scattering can hinder the carrier transport of the semiconducting channel. In ultra-clean materials, however, intrinsic phonons usually dominate the mobility. Through theoretical calculation, the mobility in $\text{HfO}_2/\text{MoS}_2/\text{ZrO}_2$ structure will be even lower than in the SiO_2/air caused by remote phonons [19]. Currently, electron mobility in 2D semiconductors is mostly limited by high-density impurities, while remote phonons usually play a secondary role. Dielectrics are also found to have an impact on carrier transport direction. Calculations show that a smaller scattering angle will cause a stronger screening in a low- κ environment. Thus the electron transport will be more directional with high- κ surrounding oxides [19].

The photoluminescence (PL) properties of 2D semiconductors are also closely related to the dielectric environment. The Coulombic interaction between negatively charged electrons and positively charged holes will be modulated by dielectric screening, which further influences the binding energies and the stabilities of excitons and trions. Lin et al. studied the behaviors of these quasiparticles under different dielectric environments by using various organic solvents [20]. The whole PL peak can be divided into three peaks, representing the A^- trion, A exciton, and B exciton, respectively (Figure 1a). As the dielectric constant increases, the intensities of PL peaks are rapidly enhanced, almost exponentially. As depicted in Figure 1b, the peak positions all have blue shifts in high- κ solvents, where the position of A^- trion is closer to that of the A exciton, suggesting that the binding energy of trions becomes smaller. Thus the intensity ratio of trions to excitons can also be regulated by the dielectric environment. The experimental data fit well with the theoretical model. In this work, organic solvents are used to exclude the possible doping or strain effect. It is reasonable to expect that oxide dielectric substrates will have similar effects on the PL properties of 2D semiconductors.

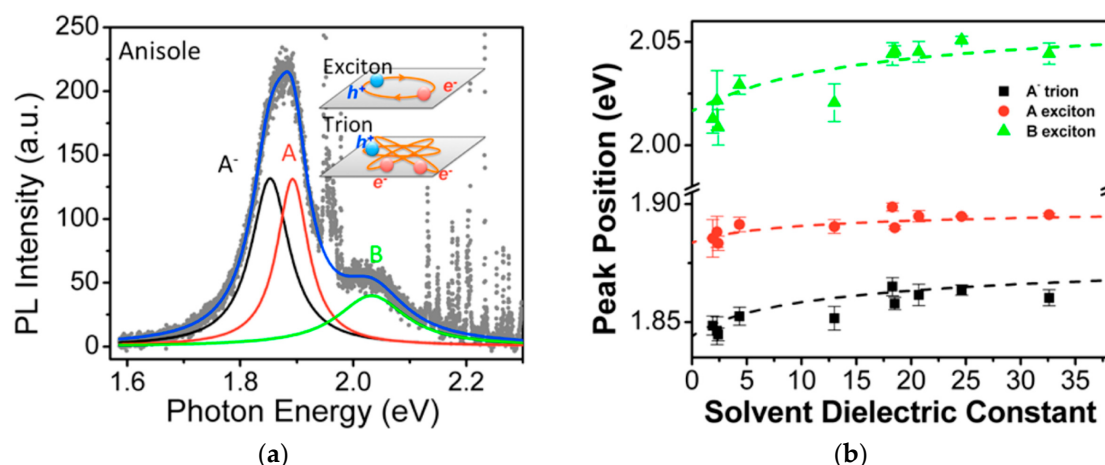


Figure 1. Cont.

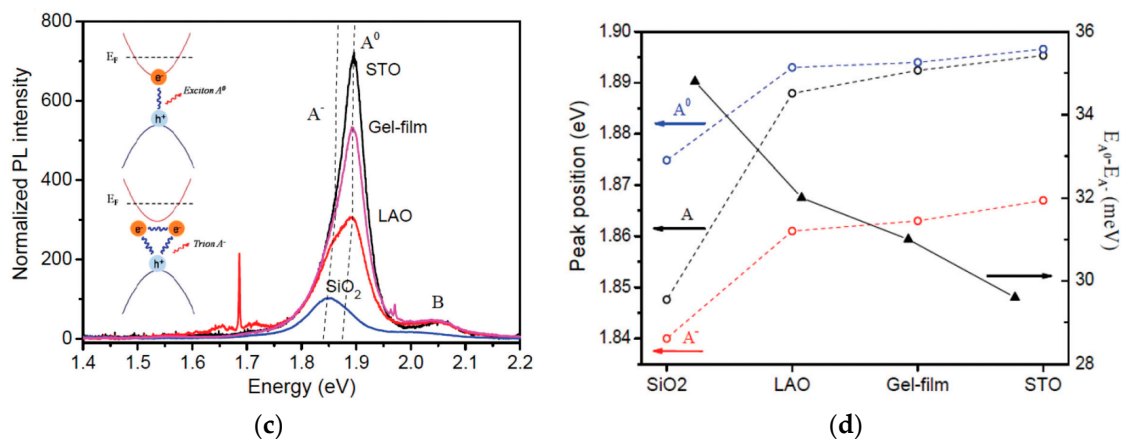
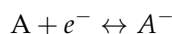


Figure 1. MoS₂ PL properties influenced by dielectric environment and charge doping. (a) A typical PL spectrum of single-layer MoS₂ that is fitted with three peaks. The inset schematics demonstrate classical pictures of a trion and an exciton; (b) PL peak positions as a function of surrounding solvent dielectric constants (Yuxuan Lin et al. [20]); (c) PL spectra of monolayer MoS₂ on SiO₂, LaAlO₃, gel-film, and SrTiO₃ substrates; (d) Peak positions of PL peak (A), neutral excitons emission (A⁰), and trion emission (A⁻), and energy difference between neutral excitons emission (A⁰) and trion emission (A⁻) (Yuanyuan Li et al. [21]).

Other effects, such as charge transfer and charge doping, also have many influences on the electrical and optical properties of 2D semiconductors [22]. Charge doping can change the types of semiconductors depending on the doping of electrons or holes. For example, at the MoS₂/glass interface, the first principle calculations show that Na atoms in the glass might become donors to make the system an n-type semiconductor, while the O dangle bonds could do the opposite because of the opposite charge polarity [16]. Li et al. utilized different substrates including LaAlO₃, SrTiO₃, and SiO₂ to control the charge transfer between a substrate and a monolayer of MoS₂ [21]. A model is set up to explain how the Fermi level and work function of the substrate influence the charge transfer. Compared to a SiO₂ substrate, there is a lower transfer of electrons and therefore a lower free electron density for MoS₂ on a SrTiO₃ substrate. As a chemical equilibrium exists between the charged and neutral excitons [23,24], the ratio of negatively charged trions to neutral excitons will decrease with a lower doping level.



Because of the larger binding energy of neutral excitons, the PL peak will have blue-shifted as there is a higher concentration of neutral excitons [25]. Studies also show that the relative proportion and intensities of neutral to charged excitons can be tuned by a gate voltage and the ambient temperature [23]. Another experiment focused on the band offset values of MoS₂/high- κ interfaces, where substrate charge transfer has a similar effect on PL properties [26]. Interference has an impact on the absorption and emission intensities but has no relation to the peak position. It is also found that Raman modes are more related to the strain caused by the substrate instead of doping and dielectric screening [21]. Research on other transition-metal dichalcogenide (TMD) materials, such as MoSe₂ and WS₂, has shown similar properties. Currently, most reports are focused on MoS₂, and thereby we mainly discuss MoS₂ in this review [27–29].

Dielectric oxides have some other effects on 2D semiconductors, such as influencing the electron-hole recombination process. The Auger Carrier Capture Model is usually utilized to explain how defects assist the recombination of electrons and holes. Theoretical model shows that in atomic layer materials, Auger scattering is related to the strong Coulomb force inside the material [30]. Thus dielectric screening will affect the correlations of electrons and holes. Besides electrical mobility and optical luminescence, the Coulombic potential may have a further influence on electronic

structures of 2D semiconductors. At higher gate voltages in MoS₂, a metal-insulator transition (MIT) is observed. The electrical conductance shows a metallic behavior when the charge density is beyond 10^{13} cm^{-2} [12]. In another experiment, the results of electrical transport measurements are consistent with a variable range hopping (VRH) model. In this case, randomly distributed charges and their mutual Coulomb interaction may cause disorders in the material [10]. The top insulating layer has a dielectric capping effect on 2D semiconductors [31]. When an Al₂O₃ top dielectric layer is added to the black phosphorous channel, positive charges will accumulate and fix at the interface. This will result in band bending and effectively reduce the Schottky barriers at the contact.

2.2. 2D Semiconductors/Semiconducting Oxide Heterostructures

When 2D semiconductors interface with semiconducting oxides, their band alignment must be considered. Some novel phenomena, such as exciton magnetic polarons and two-dimensional electron gas, emerge in the case of large band offsets. The overall mobility is influenced by both carrier injection and scattering.

The band gap and work function are two of the most important parameters for semiconductors. The relative positions of the valence and conduction bands of two semiconductors determine the behaviors of the heterojunction. Anderson's rule [32], quantum dipole theory [33], and other models are utilized to predict band offsets. Based on the band alignment, semiconductor heterojunctions can be classified into three types: type I (straddling or symmetric), type II (staggered gap), or type III (broken gap), as shown in the Figure 2. The heterostructure is type I if $\text{VBM}_A < \text{VBM}_B < \text{CBM}_B < \text{CBM}_A$, type II if $\text{VBM}_A < \text{VBM}_B < \text{CBM}_A < \text{CBM}_B$, and type III if $\text{VBM}_A < \text{CBM}_A < \text{VBM}_B < \text{CBM}_B$ [34,35].

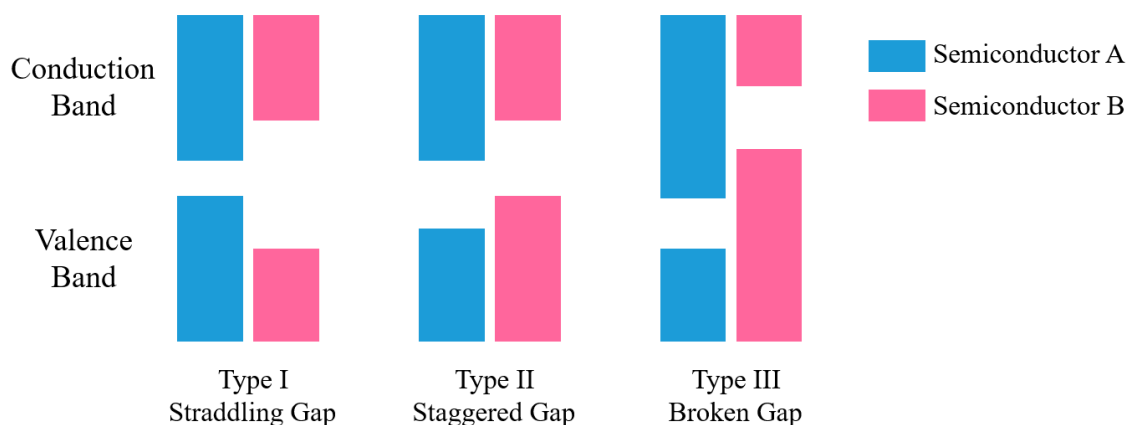


Figure 2. Illustration of type I, II, and III semiconducting heterostructures.

The three types of heterojunctions have been used in various devices owing to their different properties. Type I heterojunctions can well confine electrons and holes in quantum wells for more efficient electron-hole recombination [36], and therefore they are often used to generate light in optical devices such as LEDs and lasers [37]. Type II heterojunctions have very large band offsets on either conduction band or valence band and can easily confine electrons in one layer. They have been used to fabricate heterojunction field-effect transistors with high carrier mobility [38]. Both staggered gaps and broken gaps can regulate the energy difference between the valence band and the conduction band. This can affect the photon absorption and electron transition process and is applicable in photo detectors [39,40] and mid-infrared lasers [41]. In tunneling transistors [42], the type II heterojunction is utilized to enhance the tunneling current. However, in a type II junction, a finite tunneling barrier is not high enough to hinder the carrier transmission. A bigger band offset (type III heterojunction) is desirable if the carrier transmission needs to be totally reduced.

Besides these traditional applications, 2D semiconductor heterojunctions can also enable lots of novel properties including two-dimensional electron gas (2DEG), dilute magnetic oxides,

and interlayer excitons. When 2D semiconductors and semiconducting oxides form heterojunctions, they will interface with each other through band alignments and their properties can be modulated by external fields. In InGaPN/GaAs heterostructures, the addition of nitrogen concentration lowers the conduction band. The original type I heterostructure changes to type II. Simultaneously, 2DEG will form at the interface [43]. In dilute magnetic semiconductors, where magnetic ions and carriers both exist in comparable strength, spin splitting and optical properties are closely related to each other. In such heterojunctions, when spin splitting of the valence induced by magnetic field exceeds the band offset in the zero field, there will be a band alignment transition. In one spin state, holes are confined to the quantum wells (type I behavior), while in the other spin state, holes will be confined to the barriers, showing type II behavior [44]. Another interesting phenomenon is interlayer excitons and polarons. At the interface of MoSe_2 – WSe_2 , bound electrons and holes are localized and stabilized in respective layers. Interlayer excitons are found to have a long lifetime of ~ 1.8 ns in these circumstances [45,46]. In $\text{ZnSe}/(\text{Zn,Mn})\text{Se}$ quantum wells, exciton magnetic polarons have been observed to have much longer lives (20ns) than in type I because of the larger band offset [47]. In the same system, calculations show that interlayer excitons have smaller binding energies and lifetimes than quantum well excitons [48].

Besides the band alignment, oxides may also inject carriers towards 2D semiconductors at the interface. This will regulate interfacial Schottky barriers as well as electrical contacts. The regulation is essential, particularly in the case of p-type MoS_2 devices where holes need to be provided for electrical conduction. Chuang et al. used MoO_x as contacts to fabricate p-type field effect transistors [49]. As Figure 3a shows, the substoichiometric compound MoO_x ($x < 3$) has a high work function and its Fermi level is pinned at the valence band of MoS_2 . Thus holes can transport easily from MoO_x to MoS_2 by overcoming small Schottky barrier heights. Figure 3d shows the on and off states of the PFET device. MoO_x functions as an efficient hole injector to exhibit appropriate work function and improve the contact at the interface. Different from the case of MoO_x used as contacts, when MoO_3 is deposited on the monolayer MoS_2 as a capping layer, electron densities in the MoS_2 channel will decrease because of the charge transfer effect and its overall transport properties change [50].

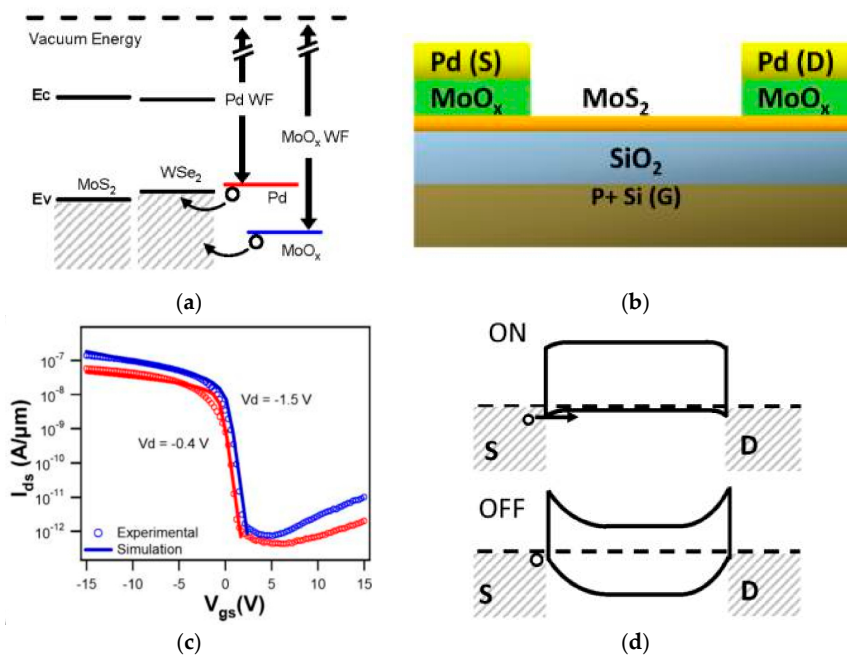


Figure 3. $\text{MoS}_2/\text{MoO}_x$ heterojunction. (a) Valence and conduction band positions with respect to vacuum level for MoS_2 , WSe_2 , Pd, and MoO_x ; (b) Schematic of the device structure; (c) I_{ds} vs. V_{gs} characteristics for a representative MoS_2 PFET with MoO_x contacts; (d) Qualitative band diagrams for the ON (top panel) and OFF (bottom panel) states of the MoS_2 PFET (Steven Chuang et al. [49]).

2.3. 2D Semiconductors/Conventional Piezoelectric Oxide Heterostructures

Piezoelectric ceramics are also usually oxides in which electric charges can be accumulated in response to external mechanical fields, or, conversely, mechanical strains can be generated in response to external electric fields. ZnO and lead zirconate titanate (PZT) are both conventional piezoelectric oxides. When piezoelectrics and 2D semiconductors form heterostructures, strain and electronic properties can have a mutual effect and one can be utilized to modulate the other property.

Piezoelectric charge accumulation at a 2D semiconductor/piezoelectric ceramic heterojunction will influence the carrier density and the electrical conductance in the 2D semiconducting channel. Chen et al. have utilized a piezoelectric ZnO nanowire to function as a gate to modulate the electrical transport behavior of MoS₂ by external mechanical stimulus [51]. In FET devices, the source-drain current is usually modulated by a gate voltage. In this MoS₂/ZnO system, however, environmental air pressure is instead applied for gating. When the ZnO nanowire is grown on top of an exfoliated MoS₂ flake, the increase of pressure is significantly lower the source-drain current, where the ZnO nanowire functions as an effective back gate voltage: the drain current curve when $V_{gs} = -7$ V with 6.25 MPa pressure is similar to that when $V_{gs} = -15$ V without pressure. Another device is fabricated with an Al₂O₃ interlayer. In this device, a larger pressure induces a larger source-drain current, which is in sharp contrast to the former device. The mechanism is further explained by the piezotronic effect. At the interface of an n-n heterojunction, electrons accumulate and form a charged barrier for the balance of Fermi levels. In the ZnO–MoS₂ heterostructure, free electrons accumulate to screen positive polarization charges of ZnO. Pressure will induce stronger polarization in ZnO and deplete electrons in the MoS₂ channel. With an interlayer, Al₂O₃ functions as a capacitor dielectric layer where holes accumulate at the ZnO–Al₂O₃ interface and electrons accumulate at the MoS₂–Al₂O₃ interface. Thus a larger electron concentration leads to a larger source-drain current when the polarization is enhanced by the applied pressure. In a reverse case, piezoelectricity has also been utilized in touch sensors to detect pressure based on a change in the channel current difference [52]. In ferroelectrics, strong spontaneous polarization exists and can be tuned by temperature. Fang reported a MoS₂/PMN-PT composite structure, in which infrared light (IR) is used to generate heat and reduce previous polarization in ferroelectrics [53]. The charge accumulates at the interface and the carrier concentration is therefore modulated. Under such circumstances, the IR works as a negative ‘gate’ in the FET devices [53,54].

The piezoelectricity also has a great impact on the photoelectric response of 2D semiconductors. Xue et al. fabricated a p-MoS₂ and an n-ZnO diode and measured its photoresponse performance under different pressures [55]. MoS₂ is pretreated by SF₆ to become p-type and acrylic is used to apply a pressure and simultaneously transmit the ultraviolet. The highest external quantum efficiency (EQE) is 52.7% with six layers of MoS₂ at a -2 V drain voltage. The photoresponse time is much longer at low power densities, mainly because the separation of excitons in the heterojunction is slow. The barrier formed at the p-n junction interface and depleting area is crucial to stabilizing the separated photogenerated carriers [56]. When applying pressure, charges generated by piezoelectricity accumulate at the interface and lower both the valence and conduction bands of ZnO. Thus a lower barrier results in a larger depletion region, which is beneficial to optoelectronic performance. As shown in Figure 4b, the photo current and EQE both increase quickly as external pressure increases. The EQE almost quadruples at 23 MPa external mechanical stimulus. ZnO proves to be a good ‘gate’ to modulate the separation, transport, and recombination process of photocarriers. A similar experiment has been conducted at the graphene/ZnO junction with a higher Schottky barrier and a wider depletion area induced by strain, which resulted in better device performance [57].

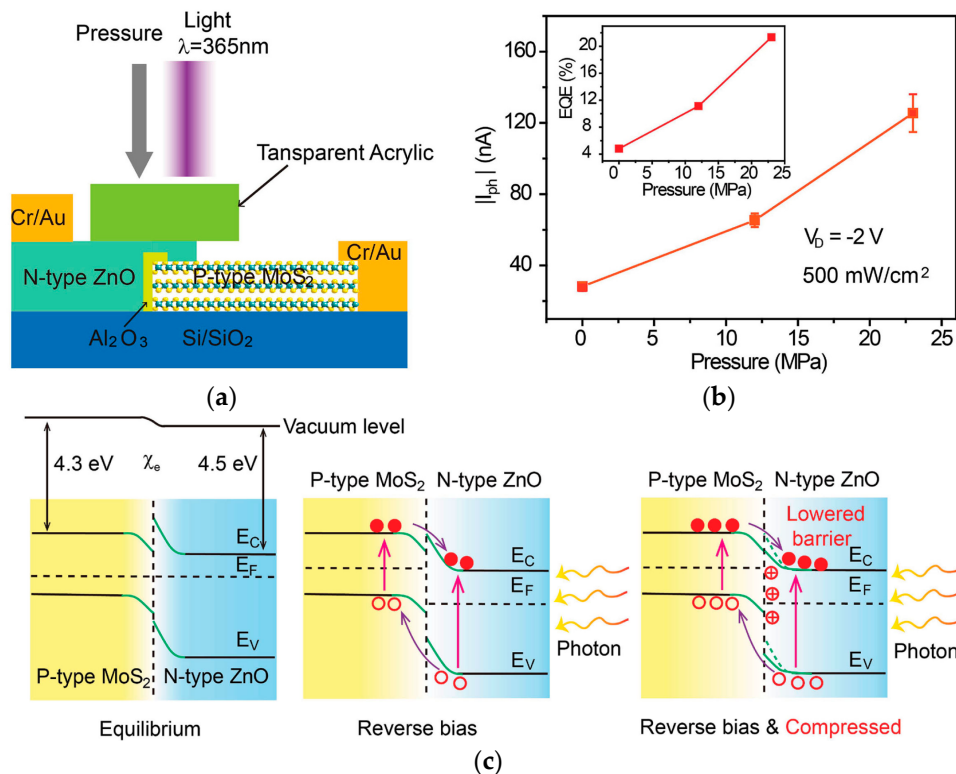


Figure 4. Photoelectric response of a 2D semiconductor/piezoelectric ceramic heterostructure. (a) Schematic of experimental set-up for testing the diode; (b) Pressure dependence of I_{ph} at a drain bias of -2 V and an incident power density of 500 mW cm^{-2} . The inset shows the increased EQE with the increased applied pressure; (c) Band diagrams interpreting photogenerated carriers and piezophototronic effect enhanced photocurrents (Fei Xue et al. [55]).

2.4. 2D Semiconductors/Strongly Correlated Oxide Heterostructures

Strongly correlated systems are a wide range of materials where their electronic structures, magnetic properties, spin, and mechanical strain are strongly correlated with each other. The traditional band theory is based on independent electron approximation, i.e., only electron-ion interaction is considered and electron-electron interaction is ignored. However, when the interaction between electrons or spinons is not negligible, some novel phenomena such as metal-insulator transitions, spin-charge separation, and high-temperature superconductivity may appear in such strongly correlated systems. Many transition metal oxides are classified into strongly correlated materials. For example, the concept of strongly correlated Mott insulators was first proposed in NiO because its properties cannot be explained by the traditional band theory. Compared with traditional piezoelectric materials, more degrees of freedom can be utilized in strongly correlated systems. The diverse properties of strongly correlated oxides provide a variety of possibilities to modulate the properties of ultrathin 2D semiconductors; equally, 2D semiconductors can be utilized as indicators of the phase changes in strongly correlated oxides.

Electronic structures and band gaps of 2D semiconductors can be modulated by strongly correlated materials through strain transfer [58]. When interfacing MoS₂ with PMN-PT, a new type of ferroelectric oxides, the band gap of MoS₂ can be regulated through strain by applying an external voltage bias on the PMN-PT [59]. Raman spectroscopy confirms that there is a strain transfer from PMN-PT to MoS₂. As shown in Figure 5c, the MoS₂ PL peak blue shifts as the strain increases. A 0.2% compressive strain (corresponding to 500 V bias on PMN-PT) can shift the PL peak by 60 meV ($\sim 300\text{ meV}$ per 1% strain) and double the PL intensity during the process. Previous researchers have tried to explain the strain-induced change of electronic structures in MoS₂ by first principle

calculations [60–62]. As depicted in Figure 5d, both valence and conduction bands shift to higher energies with a compressive strain, and the energy levels of conduction band changed to a larger extent. At 0.2% strain, MoS₂ remains an indirect semiconductor but with a larger band gap. Another study shows that the surface potential and work function of MoS₂ can also be influenced by the strong polarization of ferroelectrics [63]. Because of the induced dipole effect, a work functional variation is found at the edge and interior region of PbTiO₃ surface.

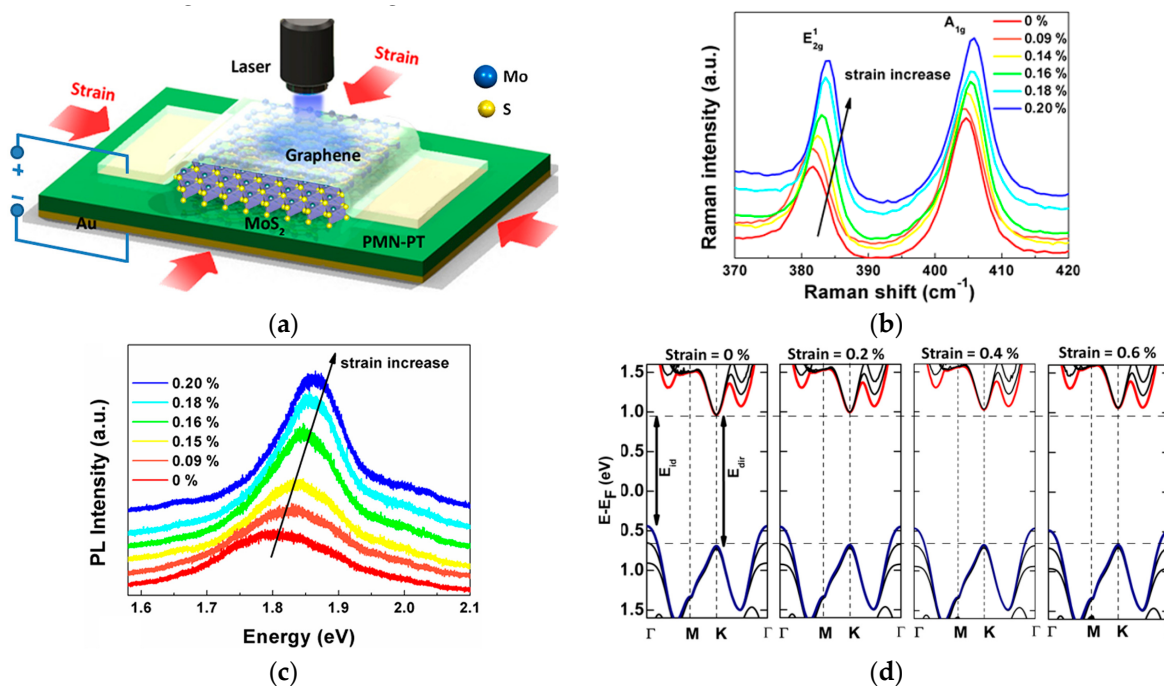


Figure 5. Electronic structure and band gap of a 2D semiconductor influenced by strongly correlated materials through strain transfer. (a) Schematic diagram of Raman and PL measurements on MoS₂. MoS₂ is sandwiched between a piezoelectric PMN-PT substrate and a graphene top electrode; (b) Raman spectra of a trilayer MoS₂ under various applied strains; (c) PL spectra of MoS₂ under various strains; (d) Band structures of ABA-BAB stacking trilayer MoS₂ under 0.0%, 0.2%, 0.4%, and 0.6% biaxial compressive strain (Yeung Yu Hui et al. [59]).

Strongly correlated materials have also been utilized to modulate the optical properties of 2D semiconductors. When interfacing MoS₂ with VO₂, which is a textbook Mott insulator and exhibits a metal-insulator transition (MIT) at around 68 °C, both the Raman and PL properties of MoS₂ are strongly modulated by the VO₂ substrate [64,65]. Both the E_{2g} and A_{1g} modes of MoS₂ are thus found to have red shifts when heating the VO₂ substrate to metal phase [65]. The PL position also shifts to a lower energy due to increased non-radiative electron-hole recombination. Meanwhile, the PL intensity is drastically enhanced because the excitons and trions are enhanced during the MIT process. However, Hou et al. attributed this enhancement to an optical interference effect due to the different refractive indexes of the two VO₂ phases [64].

2.5. 2D Semiconductors/Magnetic Oxide Heterostructures

Ferromagnetic insulators and multiferroic oxides are two types of important magnetic oxides. Some 2D materials including graphene and MoS₂ have the advantage of long diffusion lengths of spin electrons. When interfaced with functional magnetic oxides, 2D semiconductors may offer new functions and open up novel applications.

In the EuO_x/graphene heterojunction, the Eu atoms will generate proximity induced ferromagnetism in graphene. Proximity-induced exchange splitting and spin orbit coupling have been

revealed by first principle calculations [66]. They could also lead to a nonlinear dispersion at Γ point through inter-valley interactions [67]. Multiferroics shows the coupling between ferroelectricity and magnetism and is promising for spintronic devices. In BaMnO_3 /graphene structures, a distinct spin polarization is caused by the interaction between π electrons from graphene and d electrons from Mn atoms. An evident spin splitting as large as 300 meV is observed in the system [68]. A high-mobility electronic region is achieved by acceptor-doping in graphene. Meanwhile, theoretical calculations have proven that multiferroic oxides can be utilized to generate 2DEG at the surface, which provides a good platform to conduct research on spin interaction and spin field effect devices [69–71].

It is worth noting that all of these devices are based on the advantages of the long spin diffusion length and high carrier mobility in graphene. Some other 2D semiconductors such as MoS_2 possess similar advantages. Therefore, similar devices are expected to develop in 2D semiconductors with a high performance and low energy dissipation.

3. Applications of 2D Semiconductors/Functional Oxide Heterostructures

3.1. Volatile Field-Effect Transistor (FET) Devices

In modern computers, most calculations are done by a Boolean operation, which is carried out by logic gate circuits. Logic gates are mainly realized by diodes and transistors to perform as electronic switches. Compared with traditional bipolar transistors, field effect transistors (FETs) have become mainstream in practical applications because of several advantages. Bipolar transistors have majority carrier and minority carrier conduction, while in MOSFET devices only majority carriers conduct the electricity. Complementary Metal-Oxide-Semiconductor (CMOS) logic utilizes both n-channel and p-channel MOSFETs to achieve a high speed and low power consumption. Several parameters are crucial in logic operation and integrated circuits. Smaller transistor size can enhance the integration per unit area and reduce the production cost. Faster switching and response speed calls for higher mobility in channel materials and lower subthreshold swing. A high on–off ratio guarantees high performance and low leakage current. Low leakage current is also beneficial to less power dissipation.

Continuously scaling down the size of FET devices relies on the development of advanced lithography techniques and a new structure design (e.g., FinFET). As we have discussed in Section 2, short channel effect and leakage current will become big problems when the size of FETs becomes smaller. Thinning channel materials is a way to solve these problems, but in this case the existence of dangling bonds and charge traps of traditional silicon channels will hamper the mobility. Thus, novel ultrathin materials are still desired to improve the performance. A theoretical calculation based on non-equilibrium Green's function has predicted the ultimate performance limit of MoS_2 transistors [72]. This shows several merits: (i) Because the band gap of MoS_2 is larger than that of silicon, the minimum current can be very small when ignoring the leakage current. The on/off ratio can reach more than 10^{10} ; (ii) Because of wave function reflection at contacts, a reduced density of states in the channel can result in a high intrinsic device transconductance ($4.4 \text{ mS}/\mu\text{m}$); (iii) Subthreshold swing can be as low as 60 mV/decade (the theoretical limit) with the optimal dielectrics. For a 15-nm gate, the drain-induced barrier lowering is 10 mV/V, showing good short-channel behavior. What is more, most of the III–V semiconductors have larger mobility than MoS_2 and provide better choices for high-performance transistors.

Several methods have been adopted to enhance device mobility. High- κ materials are often used as gate dielectrics [73,74]. Radisavljevic et al. used 30 nm HfO_2 as a top gate and single layer MoS_2 as a channel (Figure 6) [9]. At a 500 mV source drain voltage, more than 10^8 on–off current ratio and $1 \mu\text{S}/\mu\text{m}$ transconductance are achieved. The mobility is around $200 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and the subthreshold slope is 74 mV/decade [75]. Al_2O_3 is also employed as a back gate to regulate multilayer MoS_2 . From the transfer curve shown in Figure 6d, high mobility ($>100 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$), a ~ 70 mV per decade subthreshold swing is also observed. Li et al. systematically studied the effect of the channel

thickness on the overall mobility [13]. For either Ti or Au contact, 14–15 layers of MoS₂ shows the best performance. The ultrathin FET channels may be strongly affected by short-range Coulombic interactions [11], and impurity density should also be considered in the devices. Oxygen molecules, water molecules, or other chemical contaminants in the ambient environment may be absorbed onto the surface and become scattering impurities. Hydrophobic substrates and dry environments are usually required for obtaining good performance of devices [76].

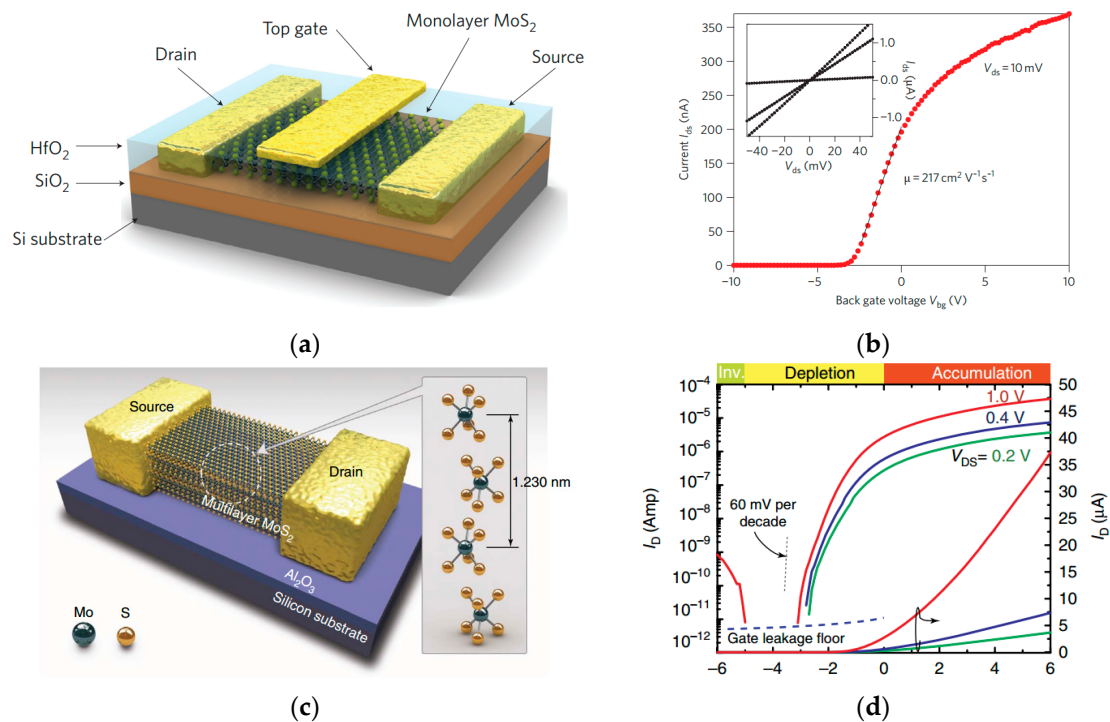


Figure 6. Schematics and performance of monolayer and multilayer MoS₂ transistors. (a) Schematic of MoS₂ monolayer transistors; (b) Room-temperature transfer characteristic for an FET with a 10 mV applied bias voltage V_{ds} . Inset: I_{ds} – V_{ds} curve acquired for V_{bg} values of 0, 1 and 5 V (Branimir Radisavljevic et al. [9]); (c) Schematic of a MoS₂ thin film transistors with a multilayer MoS₂ crystal; (d) Drain current versus back gate voltage, showing a $\sim 10^6$ on/off ratio and ~ 80 mV per decade subthreshold slope (Sunkook Kim et al. [75]).

Interfacial contact is one of the key issues for achieving ideal transistor performance. Tunneling contacts have low Schottky barriers, but the Fermi level is likely to pin at the energy level of defects, which causes a large tunneling resistance. An ideal circumstance for MOSFETs is Ohmic contact with low resistance and matched energy levels between channels and electrodes. However, it is difficult to find metals with a suitable work function to form Ohmic contacts with channel semiconductors [72]. Some metals, such as Au, Ti [13], and Sc [77], have been employed as electrodes in the 2D semiconducting transistors, but contact problems usually exist. Many types of other non-metal electrode materials have thus been explored to solve the contact problem. For example, the high work function of MoO_x pins its Fermi level at the valence band of MoS₂, and holes can transport through a thin Schottky junction [49]. The situation is different in metal-semiconductor field effect transistors (MESFET), where a Schottky barrier should be apparently formed to decrease the leakage current [78]. Experiments show that higher barriers induce larger on–off current ratios and better subthreshold slopes. Meanwhile, the drain current should not be leaked to the gate. An NiO_x electrode is fabricated and demonstrates a good performance, including high mobility (500 – 1200 cm²V^{−1}s^{−1}), low threshold voltage (~ -1 V), low subthreshold swing (83 mV) and quick photoswitching speed (2 ms).

Compared with TMD materials, black phosphorus has a lot advantages as a volatile FET channel material. Much effort has been made to improve the performance of BP devices. BP has a moderately large band gap and much higher mobility of $\sim 1000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, as reported [79]. The type of contact metal and alternation of thicknesses can modulate p-type BP into n-type, with hole mobility still as high as $\sim 950 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ [80]. BP transistors with a 20 nm channel length show a small short channel effect and still preserve an on/off ratio of 100 [81]. However, BP is unstable in air compared with other typical 2D semiconductors, which has limited its further application. The current solution is mainly to deposit an Al_2O_3 top gate to encapsulate the device [81–83]. By this method, BP can be isolated from air and current fluctuation of its devices has been effectively suppressed.

Traditionally planar thin film transistors (TFTs) are susceptible to the cracking of the used materials during bending because of the insufficient mechanical flexibility. In flexible electronics, both channel and electrode materials should have good flexibility. Liu et al. fabricated vertically aligned TFTs with indium gallium zinc oxide (IGZO) as channel materials and graphene as flexible contacts [84]. The on–off current ratio is larger than 10^5 and there is no evident degradation over 1000 cycles of bending. The techniques can easily be extended to 2D semiconductors. A heterojunction between BP and ZnO is used as junction FET [85]. The mechanical flexibility of BP is crucial for good interfacial contact and also has great potential in flexible devices. BP FETs on flexible polymer substrates can sustain 5000 bending cycles with unchanged properties [86]. Bessonov et al. show the potential for printed flexible electronics of $\text{MoO}_x/\text{MoS}_2$ and WO_x/WS_2 heterostructures [87]. Strong electrical nonlinearity and low operating voltage (0.1–0.2 V) are demonstrated with 3 nm thick ultrathin oxide dielectric layers. Compared with traditional electrostatic doping, ionic liquid gating can simultaneously provide p-doping and n-doping with a surface charge density as high as 10^{13} cm^{-2} [88]. A p–n junction is likely to form inside the semiconducting channel in the cation/anion transition region. In practical applications, transistors have to be assembled together to form complementary integrated logic circuits, which have been also demonstrated with 2D materials [89]. Wang et al. fabricated 2–12 transistors in one circuit to offer a group of functions, such as inverter, static random access memory, etc. CMOS circuits are demonstrated by Yu et al. in WSe_2 with a voltage gain of about 38 and a small static dissipation [90]. Although the performance of the present devices seems good, further work still needs to be done to increase operating speeds and decrease the power dissipation for practical applications.

3.2. Nonvolatile Memory and Data Storage

Nonvolatile memory is a type of information storage mostly used in computers. Compared with volatile memory, nonvolatile memory can retain data even when power is turned off. Hard disk drives, magnetic tapes, optical discs, and flash memory are types of nonvolatile memory.

Ferroelectric random access memory (FeRAM) is a typical class of nonvolatile memory. Spontaneous polarization exists in ferroelectrics. When a coercive voltage is applied, the polarization can be triggered so as to change the on/off state of the channel. FeRAMs have many advantages such as low radiation resistance, low power consumption, and fast writing speed. Evidence supports the idea that the length of ferroelectrics can still scale down to nanometers and new device capabilities can be studied and put into use [91]. In industry, however, the difficulties lie in how to integrate FeRAM compatibly with present CMOS processes and how to produce them more effectively. Much research has been done to improve the switching speed and extend the degradation time. Ferroelectric polymer [P(VDF-TrFE)] has been integrated with MoS_2 and BP to make nonvolatile memory devices and fabricate inverter circuits [92,93]. Graphene has been utilized as transparent electrodes to improve the electrical contact. The device with monolayer MoS_2 possesses a mobility as high as $220 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, a subthreshold swing of 300 mV/decade, and a $\sim 5 \times 10^3$ write and erase current ratio [94]. The memory inverters also show a good performance and high output voltage efficiency [95].

These organic FeFETs are still limited by low operation speed, high work voltage, and short degradation time. In contrast, inorganic metal oxide ferroelectrics can produce faster memory switching

and increase mechanical and thermal durability. Ko et al. utilized inorganic PZT ($\text{Pb}[\text{Zr}_{0.2}\text{Ti}_{0.8}]\text{O}_3$) as gate electrodes [96]. Due to the high dielectric constant (~ 200) of PZT, low coercive voltage and V_G (2–3 V) is observed. The low threshold voltage has the advantage of low power consumption in electronic applications. The switching curve (Figure 7c) shows a quick response and a stable performance, even under a voltage wave of 1 ms duration and 10 μs ramping time. Relaxation dynamics reveals that the RC charging effect may be the reason for the switch relay, calling for a better device contact to further improve the response properties.

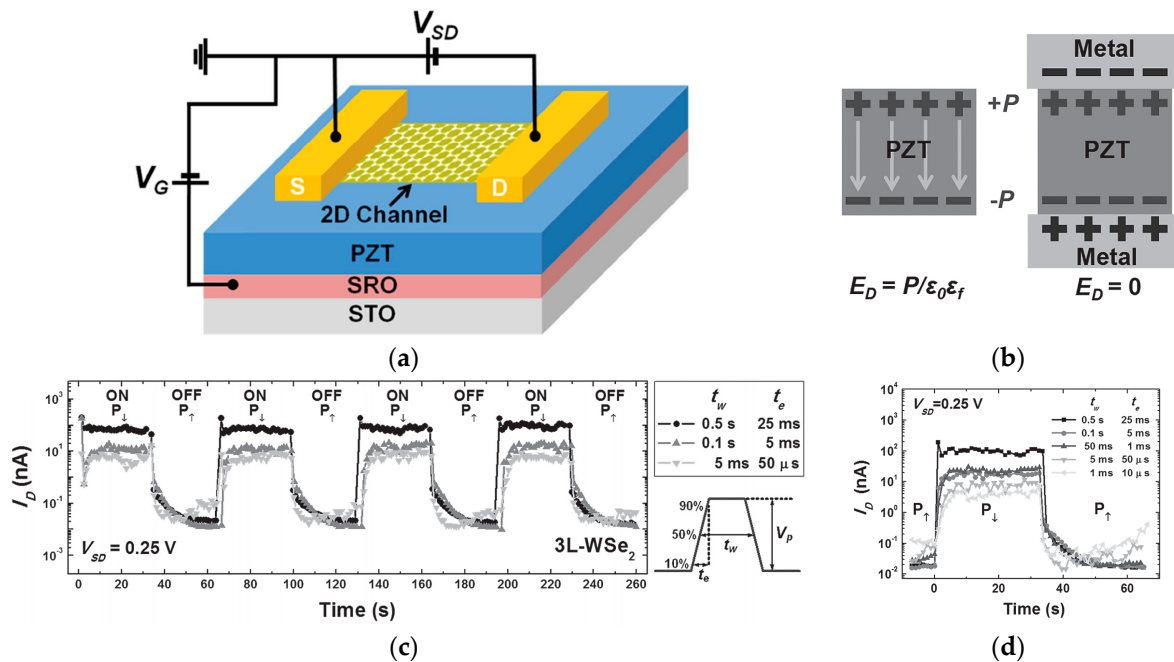


Figure 7. Nonvolatile memory produced by inorganic FeFETs. (a) Device schematic of the 2D TMD/PZT heterostructure; (b) E_D in an isolated PZT thin film and in a sandwich structure with metal electrodes; (c) I_D versus time curves of on and off states with different sets of voltage pulse parameters: peak voltage (V_P), width (t_w), and edge (t_e); (d) A series of relaxation curves immediately after pulsed gating at on (down-polarized, $P_{\downarrow}$) states and off (up-polarized, $P_{\uparrow}$) states at various t_w and t_e (Changhyun Ko et al. [96]).

The duration time is a very important parameter in nonvolatile memory. As shown in Figure 7b, in the opposite direction of PZT polarization, E_D , a depolarization field will be set up to suppress the polarization and lead to the instability of the memory device. High-temperature annealing and microscopic particle collision will also accelerate the process. If the dielectrics are in direct contact with metal, the internal electrical field will be fully eliminated. When the semiconductor channel is contacted with ferroelectrics, charges are mainly accumulated near the interface of metal contacts and the total depolarization field can be described as $E_{D0}/((C_S/C_f) + 1)$, where E_{D0} , C_S , and C_f are, respectively, the original depolarization field, the capacitances of the semiconductor, and the ferroelectrics. Thus, when interfacing with ferroelectrics, 2D semiconductors have more charges accumulated at the interface compared to traditional silicon channels. As a result, the 2D semiconductor-ferroelectrics structure has a longer retention time. Its duration surpasses 2×10^4 s, much better than traditional FET devices.

Spintronics opens a new dimension to explore internal properties of electrons and has a wide range of prospects in nonvolatile memory devices [97]. Rather than direct spin injection due to the tunneling effect at junctions, two-dimensional materials have the advantage of long-distance spin transport to produce nonlocal spin injection. With the help of the long diffusion length in MoS_2 , the spin flow can travel for a long distance without being scattered. Calculations have been done to

prove that the spin transport in such structures can be electrically regulated by a gate voltage, which widens its range of applications [98]. However, a 2D semiconductor/ferromagnetic interface may still hinder the device performance because of the large Schottky barrier resistance. Chen et al. added a thin layer of MgO between Co and MoS₂ [99]. The Schottky barrier height is reduced by 84%. Meanwhile, carrier depletion at the interface is alleviated and the MgO tunnel junction actually decreases the junction resistance. TiO₂ is also used as the interlayer between Co and MoS₂ and is found to have similar effects [100]. TiO₂ of 1 nm thickness significantly lowers the Schottky barrier from 121 meV to 27 meV. The on-state current increases by ~2 orders and the device mobility increases by ~6-fold. TiO₂ is also fabricated with BPs to verify the universality of this method [101]. In all three devices, the contact resistance can be regulated by gate voltage to optimize for spin injection and detection.

Theoretically, all materials with a hysteresis loop could find potential applications in nonvolatile memory devices. For instance, VO₂ also possesses a hysteresis during its MIT. When interfacing MoS₂ with VO₂, the PL intensity of MoS₂ will be enhanced or weakened during the MIT of VO₂ due to an interference effect [64]. Lasers can be used to introduce additional heat at controlled positions, and the phase change can be maintained at the ambient temperature even if the laser is turned off. An optical contrast between insulating and metallic VO₂ can be distinguished by the naked eye (Figure 8). Through this method, designed patterns can be written on the VO₂ canvas by a laser, and the patterns would also be erased by a simple cooling-down process. The transition temperature of VO₂ can be further lowered down to reduce power consumption by W-doping or other methods [102]. In this way, a nonvolatile, rewritable, contactless method of pattern writing is achieved, which opens a new way for convenient and graphic nonvolatile information storage and transfer.

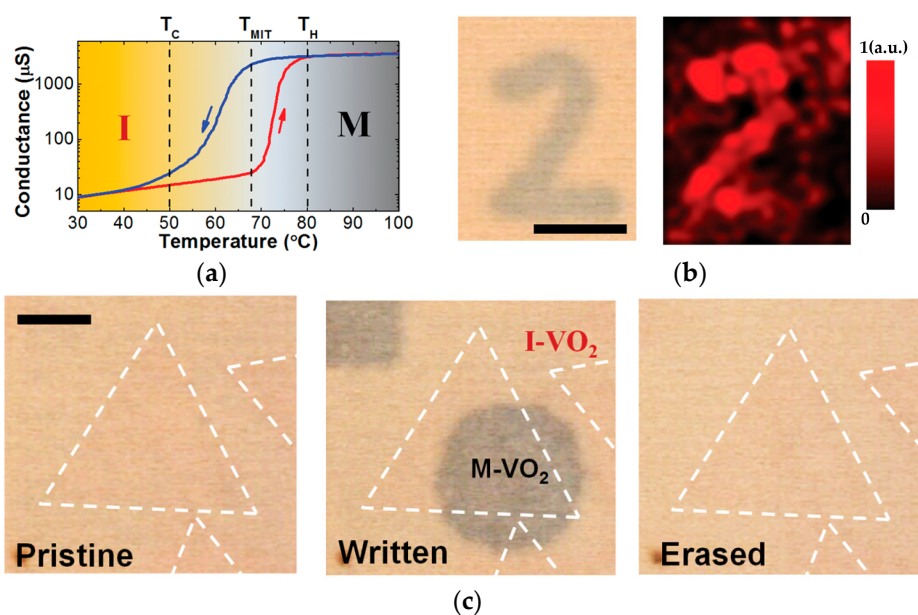


Figure 8. Writing and erasing desired patterns on VO₂ to modulate the PL intensity of MoS₂. (a) Temperature dependence of electrical conductance of a typical VO₂ film; (b) Laser-writing induced metallic regions with a shape of “2” on an insulating VO₂ film covered by a MoS₂ monolayer; (c) Laser writing a metallic VO₂ circle, which enhances the PL intensity of the overlayer MoS₂ (Jiwei Hou et al. [64]).

3.3. Sensing and Detection

Different kinds of devices are integrated into micro-electromechanical systems. The processor is the central unit that processes data, and the transducer can transform one type of energy into another. A sensor is often used to detect a signal in the environment and turn it into an available signal, usually an electrical signal. Two-dimensional semiconductors and oxides are two good platforms for

the purpose of sensing and detection. Here, based on the mechanisms of electron–hole separation in p–n junctions and photocurrent generation in phototransistors [56,103], we mainly discuss the research progress on photodetection and -conductivity in such heterojunctions. The following parameters are crucial in describing the performance of photo sensors. A sensor’s sensitivity refers to the ratio of output changes to input changes. High responsivity requires a relatively high current output and a low leakage current, guaranteeing a high on–off ratio for phototransistors. Fast electron hole separation and carrier conduction are essential to increase the switching speed. Wide sensing band and high photoelectric efficiency indicate a larger photo sensing range and a high energy conversion efficiency.

It is found that strong light–matter interaction occurs in the graphene/TMDs/graphene heterostructures [104,105]. Significantly strengthened photon absorption and electron–hole generation are also observed. Previous graphene-based photodetectors have several advantages such as wide absorption band and high speed, but weak absorption and small built-in potential limit their efficiency [106,107]. In contrast, the graphene/ZnO heterostructure can detect an ultraviolet band with a fast response in milliseconds [108]. A wide band range from ultraviolet to far-infrared and a high responsivity are also produced based on hot carrier tunneling of two graphene layers [109]. In TMD devices, vertically stacked structures provide a large response area and the intrinsic Schottky barriers guarantee an effective carrier separation. According to band diagram analysis, gate voltage can regulate the intensities and direction of photocurrents. The maximum external and internal quantum efficiency are measured to be 55% and 85%, respectively [105]. The performance can be further enhanced by improving the light absorption—for example, by adding optical resonators or plasmonic nanostructures to enhance the ambient electric fields of light [104]. Compared with 2D materials, oxides can be easily deposited with few processes and at a low cost. In SnO and at the MoS₂ p–n junction, the electron–hole separation depends on the depletion area [110]. The diode can be utilized to perform a rectification (high forward to reverse current ratio as 9.3×10^3) with a good laser response. Under different gate voltages, the junction type changes from n/p⁺ to n/p, and even n⁺/p[−], thus influencing both the dark current and the photon-induced carrier separation (Figure 9d). It is worth noting that the same electron–hole separation mechanism applies to both photosensing and photovoltaic applications, so the generated photocurrent also counts in energy harvesting.

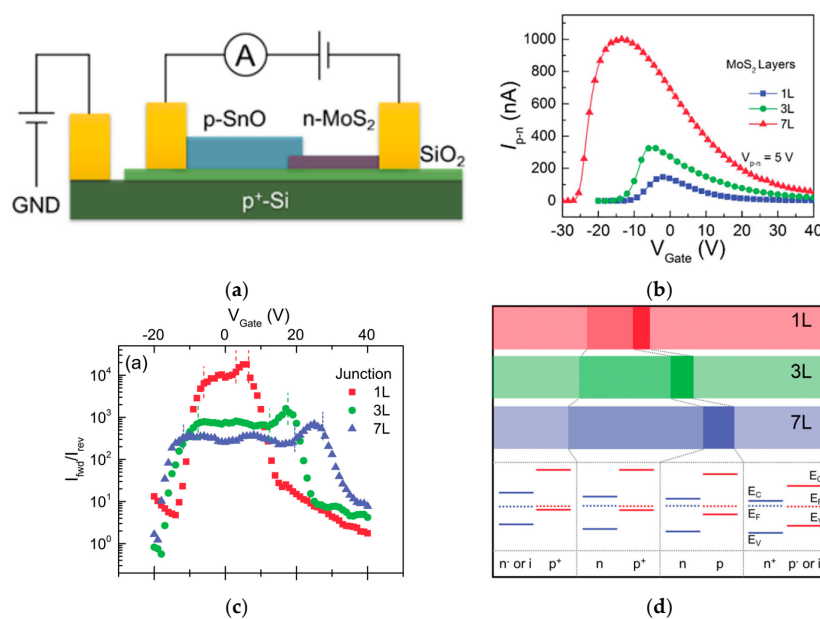


Figure 9. Rectification effect in MoS₂/SnO p–n diodes. (a) Schematic illustration for the setup of gate-tunable I–V characterization; (b) Anti-ambipolar transfer curves for all three junctions; (c) Rectification ratio of the 1 L, 3 L, and 7 L heterojunctions as a function of gate voltage; (d) Illustration of the band alignments according to the change in rectification ratio (Zhenwei Wang et al. [110]).

Other methods are used to further improve photodetector performance. Owing to the strong polarization and local electrostatic field ($\sim 10^9$ V/m) of ferroelectrics, the dark current of a channel is suppressed and the power dissipation is therefore reduced [111]. At the MoS₂/ferroelectrics junction, a high responsivity of 2570 A/W is obtained. The detectivity, defined as $D^* = RA^{1/2}/(2eI_{\text{dark}})^{1/2}$, is 2.2×10^{12} Jones. Another device based on MoS₂/ferroelectrics heterostructures works at low temperatures. A responsivity of over 300 A/W is reached. The photoresponse range is visible to 1550 nm [112]. When the channel material is replaced by In₂Se₃, the photoresponse time is as fast as 200 μ s [113]. To minimize environmental influence on the device performance, the MoS₂ photodetector is encapsulated with atomic layer deposited HfO₂ [114]. Free from ambient doping and contaminants, a lower noise and a better sensitivity are achieved. The maximum responsivity and sensitivity are 10^4 A/W and 7.7×10^{11} Jones respectively.

Another novel application based on a photoelectric process is photoconductive switches [110]. Under a 2-mW light, the photoresponsivity (the ratio of generated photocurrent to the light power absorbed by the device channel) of BP/SrTiO₃ is 2.5 A/W. The photocurrent is not volatile and the decay time is as long as several days. Red light can turn on the switch and increase the current by increasing its intensity, while UV light can turn off the switch immediately. The responsivity reaches as high as 1.1×10^5 A/W, several tens of times larger than previous devices. Such a huge responsivity is attributed to the intrinsic defects in SrTiO₃. Lights of different wavelengths have different optical penetration depths and could excite different donor and acceptor states in defects. UV light can generate carrier separations of different energy levels but only at the surface, which leads to the recombination of electrons and holes during the conduction. In contrast, red light can excite electrons from acceptor states through the entire sample. Instead of accumulating charges at the surface, holes can recombine at the top layer in the case of red light; as a result, the intrinsic electrical field declines and the entire conductivity increases. Therefore, lights with different wavelengths can be utilized to control the on/off states of the switch.

Similar photodetector devices have been fabricated with other 2D semiconductors. MoSe₂ can be changed to p type by Nb doping to form a p–n homojunction [115]. WS₂ light emitting transistors have also been demonstrated [116]. Black phosphorus could generate a greater photocurrent owing to its narrow band gap. An intrinsic 657 mA/W responsivity in the near-infrared band is reported for a 100 nm thick device [117]. In the mid-infrared light range, external responsivity is measured to be 82 A/W [118].

4. Perspectives and Challenges

So far, we have discussed lots of fundamental properties of 2D semiconductors/functional oxides heterojunctions, as well as some recent research progress on their applications. The various types of interface interactions benefit the heterojunctions in forming novel structures and providing multiple functionalities. In the future, with the miniaturization and integration of nanodevices, their applications in electronics and optoelectronics will be further enriched.

Although some novel 2D semiconductors and functional oxides have been studied for many years or even decades, there are still several challenges in the routes of their heterojunctions to practical applications. As to device fabrication, for example, the most commonly used and reliable methods are still based on the transfer technique to form vdWs heterogeneous structures. The process is facile but time-consuming, cost-ineffective, and not scalable. Further development of in situ, large-scale, and direct growth of 2D semiconductors on novel functional oxides (e.g., strongly correlated oxides) is much desired to lower the production cost and improve the performance. This is very challenging because the ambient condition in the growth of 2D semiconductors may damage the surface of these novel oxide substrates, thereby affecting the interfaces in the formed heterojunctions. Some delicately designed protecting processes should be adopted here. In the future, when more reliable and high-volume preparation of the heterojunctions is realized, their potential applications will be fully exploited.

The other challenge is to answer the following question: Are 2D semiconductors/functional oxides heterojunctions really unique or indispensable for electronic applications evaluated in the conventional figure of merits? So far most related studies have been focused on 2D electronic devices such as FET devices and p–n junctions. Lots of effort has been made to improve the mobility and speed of the devices as well as reduce their energy consumption. Promising performance has indeed been shown in several lab-made heterojunction systems. However, it will also introduce several problems in these systems, such as the instability of many 2D semiconductors (e.g., BP) and functional oxides (through losing oxygen) and susceptibility to ambient conditions (e.g., absorption of molecules in air). Overcoming these cons usually requires additional processes, which may make the 2D systems not so competitive as compared with conventional devices. In this regard, the development of 2D semiconductors/oxide systems with novel properties is a more hopeful research direction because it could provide multiple new functions that conventional devices cannot offer. For example, more dimensions of freedom in 2D semiconductors such as valley electronics have been found to be capable of regulating and carrying the information. The coupling of multiple degrees of freedom of 2D semiconductors with novel functional oxides would be a hot topic in future research.

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