Collective excitations in 2D atomic layers: Recent perspectives ()

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Yujin Cho,^{a)} 🝺 Jiahui Huang,^{a)} 🝺 and Chee Wei Wong^{a)}

AFFILIATIONS

Fang Lu Mesoscopic Optics and Quantum Electronics Laboratory, Department of Electrical and Computer Engineering, University of California, Los Angeles, California 90095, USA

^{a)}Electronic addresses: yujincho@ucla.edu; jiahuihuang@ucla.edu; and cheewei.wong@ucla.edu

ABSTRACT

The strong advantage of two-dimensional (2D) materials is that they exhibit interesting physical properties down to the single unit layer (typically less than 1 nm). Such emergent properties are highly dependent on thickness and can be tunable via external parameters such as electric field, pressure, and/or chemical treatment. In addition, most 2D materials have hexagonal crystal structures; thus, it is possible to stack with other 2D materials to enrich the properties. This tunability and compatibility makes 2D materials and their heterostructures promising candidates for optoelectronic devices.

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Collective behavior of electrons or holes, such as excitons or charge-density-waves, is present in many crystalline structures. These collective excitations are pronounced in two-dimensional (2D) materials, contributing strongly to electronic and optical properties. Those behaviors can be tailored and designed to optimize for desired applications as well as to realize new physical phenomena which were previously not expected. Such efforts are enhanced by the development of spectroscopic and microscopic techniques to probe these phenomena reliably. In this article, we review the collective behaviors, such as excitons and charge-density-waves, on the recently synthesized 2D materials and heterostructures and describe the process of tailoring their physical properties.

Two-dimensional (2D) materials refer to crystalline structures whose atoms are positioned on a plane, like a sheet of paper, lacking out-of-plane chemical bonding. Each layer has weak van der Waals (vdW) force between the layers; therefore, it is relatively easy to control the overall stacked thickness without sacrificing the layered physical properties. As opposed to conventional bulk materials in threedimensional structures, 2D materials exhibit extraordinary physical properties owing to the reduced dimensionality. A popular example in 2D materials is graphene, consisting of carbon atoms in a honey-comb lattice structure on a 2D plane. It is not only atomically thin (\approx 3 Å) but also has good electrical and mechanical properties.^{1,2} The discovery of graphene expands the search of similar materials to quasitwodimensional (2D) materials, whose thickness of a unit layer is typically around 1 nm. They consist of several chemically bonded atomic layers along the out-of-plane direction and vdW force holds each unit layer together, as in graphene. A well-studied quasi-2D material class is that of transition metal dichalcogenides (TMDs), whose material composites are transition metals, such as Ti, Mo, or W, and chalcogen atoms, such as S, Se, and Te, in the form of MX_2 (M: transition metals, X: chalcogens). Most TMD materials have hexagonal structures from the top view and threefold crystal symmetry (2H-phase). The similarities in the structure make it possible to combine different TMD materials to tune the properties of interest. A 2D material can also have 1T-metallic phase (twofold symmetry) through a post-treatment which will be discussed later.

In terms of controlling and engineering the physical properties, these materials are adaptable to many applications. The physical properties can be easily tuned via external parameters such as strain,³ thickness,⁴ electric or magnetic fields,⁵ chemical doping,⁶ and laser pulses.⁷ For example, in MoS₂, the bandgap transitions from indirect to direct when the thickness changes from bilayer to monolayer. Its crystallographic symmetry alternates between noncentrosymmetric and centrosymmetric on the odd and even number of layers, respectively.⁸ In addition, stacking a 2D material with another 2D material allows the formation of different electronic band structures, mainly due to interlayer coupling which also depends on the relative layer-to-layer planar angle in the 2D heterostructure. As such, 2D materials, either by themselves or in a stacked structure, provide a platform to tailor the optical and electronic properties for focused applications while unveiling new and interesting phenomena.

Collective behaviors of electrons and holes in 2D materials not only reveal information about the underlying band structure but also can be applied for optoelectronic applications. In this review article, we will focus on the collective excitations and oscillations of carriers in recently synthesized 2D materials and their heterostructures, as well as on how to engineer them. This paper is organized as follows: first, we will describe recent efforts in charge density waves (CDW), a collective order of electron density. Second, we will discuss the excitonic response in twisted bilayer graphene (BLG) and TMD heterostructures. Third, we will introduce defects as a quantum emitter and phase engineering of TMD materials. Finally, we will conclude the paper with discussions about current challenges to overcome for optoelectronic devices.

Charge density wave (CDW) is a periodically modulated electron density, accompanied by lattice distortion. It not only exhibits interesting electric properties but also interacts with superconductivity (SC) in a complicated manner. When the wavelength of the CDW does not match with the underlying lattice constant, they can flow on the surface and we call this state as the incommensurate CDW (IC-CDW). When the wavelength becomes the same as the lattice constant so that the CDW is locked in the parent lattice, we call it commensurate CDW (C-CDW), as shown in the right panel of Fig. 1(a). The transition from IC-CDW to C-CDW is called nearly-commensurate CDW (NC-CDW), the middle panel of Fig. 1(b). When the CDW becomes C-CDW state, we can observe the "star of David" shape as shown in Fig. 1(a), the left panel.⁹ This is a unit cell of the CDW superlattice structure, which can be an indicator of the C-CDW state. In TaS₂, the David star includes 13 Ta atoms. We can tune the CDW states with the external temperature; it goes from IC-CDW to NC-CDW and eventually to C-CDW as temperature decreases. Often, the transition



FIG. 1. Charge density waves on TMD materials: (a) superlattice on the nearlycommensurate (NC, middle) and commensurate (C, right)-CDW phases. Left cartoon shows the imposed superlattice, called star of David. Blue dots indicate Ta atoms and the red S. (b) An electronic device prototype with 1T-TaS₂. (c) Phase transition diagram between charge-density-wave and superconducting phases as a function of the doping and the temperature on ZrTe₃. (d) Tunability of the CDW phases on 1T-TaS₂ with an applied electric field. Panel (a) and (c) are from Refs. 9 and 27, respectively, reprinted with permission from Wang *et al.*, Sci. Rep. 9, 7066 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution 4.0 International License. Zhu *et al.*, Sci. Rep. 6, 26974 (2016). Copyright 2016 Author(s), licensed under a Creative Commons Attribution 4.0. International License. Panel (b) and (d): Reprinted (adapted) with permission from Geremew *et al.*, ACS Nano **13**, 7231 (2019). Copyright 2019 American Chemical Society.

temperature depends on the temperature history, and this can be understood in a simple picture of free energy. For example, when the temperature decreases, the minimum energy of the C-CDW becomes lower than the NC-CDW state and the energy barrier separating them decreases as well. When the thermal energy is enough to excite electrons over that barrier, the CDW transitions to C-CDW state. The same mechanism is applied on heating.¹⁶ This explains why we see different transition temperatures upon cooling and heating.^{11–13,15,16,19} The different states of CDW can be observed by several methods such as electrical measurement²⁰ or scanning tunneling electron microscopy.¹⁸ Apart from temperature, pressure,¹⁰ electric field,^{11,12} chemical doping,¹⁰ cooling rate,^{19,20} or thickness¹³ can also control the states.

Control of charge density waves: For instance, $1T-TaS_2$ is a wellknown layered 2D material exhibiting CDW states.¹⁵ In a bulk, this material goes from IC-CDW to NC-CDW around 350 K, NC-CDW to C-CDW around 180 K upon cooling. This transition temperature highly depends on the thickness and the surface oxidation. From a bulk to a monolayer, the transition temperature from NC to C-CDW states decreases by \approx 70 K, while the temperature between IC to NC-CDW remains almost unchanged.¹³ In addition, when a thin 1T-TaS₂ is exposed to air, \approx 2 nm thick oxide layer is formed on the surface, which destroys the CDW order.¹⁶ Protecting the surface from oxidation with a capping layer, such as *h*BN, could extend the critical thickness down to a monolayer.^{15,17}

In addition to temperature, an external field can also control the CDW states. When an electric field is applied to the material, it slides the CDW modes and increases the conductivity.^{14,22} Moreover, it also causes joule heating which triggers the transition between different CDW states. For example, Fig. 1(d) shows the change of the CDW states on 1T-TaS₂ from the device shown in Fig. 1(b), which is advantageous for a potential electronic device operating at room temperature.^{12,21} Different CDW states have different electron mobilities; therefore, changing the states can act as a switch for a field-effect-transistor¹² or a memristive memory.¹⁹

Interplay of 2D charge density waveguides with superconducting states: Another importance of understanding CDW state is its interplay with the SC state. Both of them exhibit collective order at low temperature and interact in a complicated way. They are often known to compete because both are sharing the electrons to realize their collective orders.²³ However, sometimes the CDW state can assist the SC state through electron-phonon coupling.²⁴ Those two states can be controlled via external parameters such as chemical doping,^{25–27} as shown in Fig. 1(c), or pressure.²⁸ For detailed reviews, please see Ref. 24.

Apart from the CDW, collective behaviors can be observed in a few layers of 2D materials such as bilayer graphene or TMD heterostructures. In this section, we will present recent studies of collective excitons in bilayer graphene. Bilayer graphene (BLG) is a stack of two single-layer graphene (SLGs), but their electronic band structures are quite different.²⁹ Instead of a linear dispersion near the Dirac point, BLG has a parabolic shape as shown in Fig. 2(a), left. Therefore, the effective mass of the electrons is no longer zero. This may make BLG less intriguing due to its relatively low electron mobility, comparing with SLG, but a great advantage of BLG is the existence and tunability of the bandgap. When an external electric field is applied to the BLG, the bandgap opens as wide as 250 meV,^{30,31} as shown in Fig. 2(a). This large and gradual tunability of the bandgap is useful for electronic devices, such as a photodetector, operating at room temperature.



FIG. 2. Collective excitations in bilayer graphene: (a) Schematics of the electronic band structure of BLG near Dirac points without (left) and with (right) an external electric field. (b) One-photon (green) and two-photon (pink) absorption spectrum on 8°-twisted BLG (tBLG). (c) One-photon absorption map and (d) two-photon absorption on tBLG. Panel (a): Reprinted (adapted) with permission from Collins *et al.*, Phys. Rev. B **33**, 4378 (1986). Copyright 1986 American Chemical Society. Panel (b)–(d): Reprinted (adapted) with permission from Patel *et al.*, Nano Lett. **15**, 5932 (2015). Copyright 2010 American Chemical Society.

Interesting excitonic behaviors have been observed by photocurrent spectroscopy,^{32,33} transient absorption (TA) microscopy,^{34,35} and photoluminescence (PL) spectroscopy.^{34,36} Excitons in BLG could create narrow exciton peaks, originating from 1s and 2p states of the excitons and can linearly shift with the applied electric field as the bandgap increases.³³ Among excitonic states, optically accessible excitons are called bright excitons, while those forbidden states are called dark excitons.³⁶ The dark excitons are particularly interesting because they could have long valley relaxation time due to suppressed electron-hole exchange interactions.³⁶ Since the dark excitons require a spin flip to be accessible, it is hard to access with one-photon excitation. However, if the rotational symmetry around K and K' points is slightly broken, the dark excitons are still weak but not completely dark.³³

Control of excitons in 2D heterostructure twist angle: The physical properties of BLG can be manipulated by changing the stacking angle between the two layers, such as topological³⁷ and optical properties. We call this system as a twisted bilayer graphene (tBLG). At 1.1° between the top and the bottom SLGs, which people call it as "magic angle," superconductivity³⁸ and ferromagnetism³⁹ emerge, which was unexpected. Photoluminescence is also enhanced by a factor of 5 when the two layers are aligned so that the laser excitation resonates with the van Hove singularities.⁴⁰ This surprising discovery triggers a lot of research in the twisted bilayer system and the corresponding properties. The relative angle also changes the excitonic behaviors. The bright or the dark exciton states are selectively accessible by one or twophoton absorptions, respectively. While one photon can access bright exciton states [Fig. 2(c)], two-photon resonance can access the dark exciton states due to the change in the spin state.³⁵ Figure 2(b) shows the absorption spectrum from twisted bilayer graphene (tBLG) with

one (green) and two-photon (pink) resonant absorptions. δ is the energy difference between the bright and the dark exciton states and Δ is energy splitting owing to the antiband crossing of BLG. Figures 2(c) and 2(d) show absorption maps on the tBLG with one-photon and two-photons, respectively. Between 8° and 6.5° angle, one-photon absorption is much stronger at 6.5°, while two-photon absorption is stronger at 8°.³⁵ The drastic change of the electronic band structures with a small angle difference gives a great tunability, but at the same time, it indicates that very precise engineering is required to observe a property of interest.

As in the tBLG system, we can also expect emergent phenomena arising from the combination of two TMD materials. Most of TMD materials have hexagonal lattice structures (2H-phase), which makes it relatively possible to stack two different TMD materials. In TMD heterostructures, we observe a new exciton peak, arising from interlayer interactions—interlayer excitons can be formed between the highest valence band of a material and the lowest conduction band of the other material. Creating a trilayer structure can further tune the properties, which will be discussed in the following paragraphs.

Controlling excitonic radiative light emission in 2D materials and heterostructures: TMD materials can be used as photonic devices, operating in a broad range of light; the radiative excitonic recombination emits photoluminescence (PL) in the range of visible and nearinfrared. However, low PL quantum yield (QY) at room temperature hinders their direct application as light-emitting sources. For example, the QY of ML-WS₂ is around 20% while ML-MoS₂ is less than 1%.⁴¹ There have been numerous efforts to increase PL QY in ML-TMD materials by introducing local surface plasmons and Purcell effect through plasmonic nanoparticles,^{42–45} defect passivation,^{46,47} or constructing heterostructures.⁴⁸ Here, we will introduce the enhancement of PL QY by defect passivation and interlayer PL QY by heterostructure construction.

Defects, mainly residing on the surface, decrease PL QY due to defect-mediated nonradiative recombination. The PL QY of an ML-MoS₂ is improved to 95% when the defects are passivated by a chemical, such as a non-oxidizing organic superacid [bis(trifluoromethane) sulfonimide; TSFI], as shown in Fig. 3(a).⁴⁶ The chemical treatment further enhances the carrier lifetime by a factor of 36.⁴⁶ The hypothesis for the underlying mechanism is the protonation of the surface contaminants and the reduced vacancy defects via surface reconstruction. Moreover, encapsulation with lithographical amorphous perfluorinated polymer (CYTOP) enhances the QY, close to 100% on sulfur-based TMD materials,⁴⁹ and protects the chemically treated samples against exposure to ambient environment or chemicals, such as acetone, distilled water, and methanol, as shown in Fig. 3(b). This stabilization suggests a key figure of merit on practical photonic applications.⁴⁹ However, TSFI treatment is only effective for sulfur-based TMDs, not on selenium (Se)-based TMDs⁴⁶ because the defects in Se-based materials are mainly donor impurities.⁴¹ Therefore, the non-radiative recombination on the Se-based materials can be minimized by tailoring the promoter-to-precursor weight ratio (QY \approx 60%), solvent evaporation-mediated decoupling of the substrate and the monolayer, or strain relaxation of grown monolayers.⁴⁷

The PL QY can be also enhanced by creating a three-layer heterostructure. In Fig. 3(c), interlayer excitons are formed on TMD heterostructures (WSe₂-MoSe₂) with type-II band alignment, which feature prolonged lifetime compared with intralayer excitons. However, in



FIG. 3. Photoluminescence quantum yield (QY) enhancement in monolayer TMDs and their heterostructures. (a) PL spectrum of the untreated (blue) and chemically treated monolayer MoS₂ (red). Inset: Normalized PL spectrum. (b) Normalized PL QY of CYTOP encapsulated MoS₂ before and after exposure to various chemical and physical conditions. (c) Schematic of the band structures with intralayer and interlayer transitions in WSe2-MoSe2-WSe2 vertical trilayer heterostructures. Inset: Atomic layer configuration of the trilayer heterostructures. (d) Simulated electron and hole wavefunctions of WSe2-MoSe2-WSe2 vertical trilayer heterostructures and the inset is for the bilayer heterostructure (WSe2-MoSe2); valence band maximum (blue) and conduction band minimum (red). Panel (a): Reprinted (adapted) with permission from Amani et al., ACS Nano 16, 2786 (2016). Copyright 2016 American Chemical Society. Panel (b): Reprinted (adapted) with permission from Kim et al., ACS Nano 11, 5179 (2017). Copyright 2017 American Chemical Society. Panel (c) and (d): Reprinted and adapted from Choi et al., Nat. npj 2D Mater. Appl. 2, 30 (2018). Copyright 2018 Author(s), licensed under a Creative Commons Attribution 4.0 International License.

this two-layer heterostructure, due to the spatially indirect interlayer excitons, the recombination rate is low, which leads to low PL QY. Adding one more layer and making them a trilayer structure can enhance the interlayer PL QY.48 The extra layer tunes the electron/ hole wavefunction distribution, which, in turn, modifies the interlayer photoluminescence. As shown in Fig. 3(d), the trilayer heterostructure has a significant amount of overlap between the electron (conduction band minimum; CBM) and hole (valence band maximum; VBM) wavefunctions, enhancing the strength of Coulomb interaction and the QY, as opposed to the bilayer system whose VBM and CBM barely overlap [see inset of Fig. 3(d)]. In a vertical trilayer vdW heterostructure (WSe2-MoSe2-WSe2), QYs of both interlayer neutral and charged excitons (trions) are increased by a factor of 3 at 77 K and 5 at 4 K.48 The binding energy of trions in the trilayer system (27 meV) is comparable to that in the bilayer system (28 meV), and the lifetime of interlayer excitons in the trilayer structure $(2.5 \text{ ns})^{48}$ is slightly longer than that of the bilayer (1.8 ns).⁵⁰ Significant improvement on the interlayer QY, while maintaining other properties, may lead to the new generation of excitonic devices.4

Defects are known to be detrimental to the optical properties. However, when it is well-controlled and positioned, the spatially localized exciton at the defect states can act as a quantum dot, emitting a narrow-linewidth (μ eV) and nanoseconds-lifetime light.⁵¹ ⁴ To use it as a quantum light emitter in 2D materials, it is necessary to characterize a single defect and correlate it with the electronic properties. Single defects were directly characterized on a chemically grown monolayer WS₂ by Schuler et al. in 2019.⁵⁵ The defect density on the as-grown WS₂ is quite low; therefore, sulfur vacancy defects were intentionally created through extra annealing at an elevated temperature, as shown in Fig. 4(a).⁵⁵ In Fig. 4(b), scanning tunneling spectroscopy (STS) revealed two in-gap states induced by large spin-orbit coupling and separated by 252 meV.56 Chemical doping can also create in-gap defect states. Oxygen atoms, substituting chalcogen sites, create defect states at 300 meV below valence-band maximum (VBM).⁵⁷ Figure 4(c), left panel, depicts the STS spectra of a Cr substituted W-site of WS2, showing three in-gap states and two of them are separated by 80 meV.⁵⁶ However, Mo substituted W-site of WS_2 [right panel of Fig. 4(c)] lacks the in-gap states. The difference stems from the energy differences between Cr-3d and Mo-4d electrons and the local strain distribution at the defect sites.⁵⁶ Apart from these substitutions, two types of negatively charged defects can independently induce two or three in-gap levels, but the origin of such defects needs further investigation.5

Single defects and nanoscopic imaging: The deep in-gap states are from single defects, suggesting a possibility as a high-quality solidstate quantum light source with high fidelity and purity, which is ideal for quantum communication and information processing. However, due to technical limitations, single-photon emission from an isolated single defect has not yet been observed. Future effort can be put into directly measuring the luminescence from the localized excitons, bound to a single defect state. State-of-art atomic-scale imaging and spectroscopy such as near-field optical microscopy or scanning tunneling microscopy-induced luminescence may be able to address the luminescence.



FIG. 4. Defects in monolayer TMDs and their electronic properties. (a) STM topography of a CVD grown ML-WS₂. (b) STS spectrum of two in-gap states on an ML-WS₂. (c) STS spectrum of a chromium substituted tungsten site and pristine ML-WS₂ (left) and that of a molybdenum substituted tungsten site and pristine ML-WS₂ (right). Panel (a) and (b): Reprinted with permission from Schuler *et al.*, Phys. Rev. Lett. **123**, 076801 (2019). Copyright 2019 by the American Physical Society. Panel (c): reprinted (adapted) with permission from Schuler *et al.*, ACS Nano **13**, 10520 (2019). Copyright 2019 American Chemical Society.

As briefly mentioned earlier, TMD materials have various crystallographic phases such as semiconducting (H), metallic (T), or semimetallic (T') phases.^{58,59} The most common structure at room temperature is 2H phase which is a hexagonal structure from the top view. 1T phase has twofold symmetry which can be observed in MoS_2 after post-treatment or TaS_2 . 1T' is the same as 1T phase but distorted. They are semimetallic and can be observed in WTe_2 or $MoTe_2$. We can stabilize a certain phase of a material by controlling the growth process, laser irradiation with a short laser pulse,^{60,61} or a chemical treatment.⁵⁸

Controlled laser switching of crystal symmetry and phase: A laser pulse can switch the crystallographic symmetry and reveal a hidden state. This has been demonstrated on layered topological materials, WTe₂ and MoTe₂, whose phases are non-centrosymmetric T_d at room temperature,^{60,62} as shown in Fig. 5(a). The other phases are known to be metastable at room temperature but the irradiation of a laser pulse can stabilize it. The short pulse excitation changes the material's crystal symmetry which was confirmed with time-resolved second-harmonic generation microscopy, in Fig. 5(b).^{60,61} The change in the symmetry was further confirmed by second harmonic generation (SHG) microscopy. When the laser pulse was incident on the sample, the SHG intensity is sharply decreased, meaning that the material acquired centrosymmetry and turns into a topologically trivial phase. Similar behavior was also observed from WTe₂.⁶⁰

In addition, the phase change is also observed in MoS_2 . 2H- MoS_2 transitions to 1T- MoS_2 when treated with n-butyl lithium solution at room temperature⁵⁸ or with an electron beam.⁵⁹ The conversion rate of the chemical treatment reaches 60%–70% of the target area and it forms atomically sharp interfaces between the two phases, observed by high-resolution transmission electron microscope and *x*-ray photoelectron spectroscopy.⁵⁸ Figure 5(c) shows a patterned 2H and 1T phases by electron beam irradiation.⁵⁹ This phase engineering combines two different electronic structures with an extremely high-quality interface and therefore, can greatly improve the device performance. For example, patterning 1T phase under the electrodes decreases the contact resistance by a factor of 5, increases the performance by twice (mobilities, transconductance, etc.), and enhances the on/off ratio by one order of magnitude.⁵⁸ Creating 1T-phase islands on the 2H-phase material can also mimic quantum dots.⁵⁹ When the



FIG. 5. Phase engineering of ML-TMDC materials. (a) The crystal structure of T_d -MoTe₂ and (b) time-resolved second-harmonic generation intensities at different temperatures and time delay. The inset shows the SHG intensity with a longer time delay, up to 80 ps. (c) A quantum dot patterning using an electron beam on the MoS₂ layer. The irradiated region turns into a 1T phase while the rest of the area remains in 2H. Panel (a) and (b) is reprinted with permission from Zhang *et al.* Phys. Rev. X **9**, 021036 (2019). Copyright 2019 Author(s), licensed under a Creative Commons Attribution 4.0 International License and panel (c) is reprinted with permission from Xie *et al.*, Sci. Rep. **7**, 9965 (2017). Copyright 2017 Author(s), licensed under a Creative Commons Attribution 4.0 International License.

e-beam dose increases, the 1T-phase quantum dot-like islands may attract nonradiative charges from defects, which, in turn, increases the PL intensity. Interestingly, instead of the PL peak shifting gradually with increasing electron dose, the original PL peak at 1.8 eV disappears while a new one appears around 1.4 eV, indicating that the 2H phase turns into 1T without an intermediate structural phase.⁵⁹ The PL intensity of a new peak at 1.4 eV becomes stronger than the original one over 100 fC, which may be due to the quantum dots absorbing nonradiative charges from defects.⁵⁹ Although the stability in the long term and the repeatability over a large area of 1T states are required, the enhanced PL on the quantum dots sheds light on a quantum light source or quantum computation.

In this perspectives article, we highlighted the recent collective behaviors on 2D materials, such as charge density waves or excitons arising from twisted bilayer system and TMD heterostructures. The excitonic response can be further engineered by positioning defects and phase engineering. The combinations of 2D heterostructures and the tuning parameters provide a large range of potential for optoelectronics and quantum applications and an interesting playground to explore exotic phenomena such as Majorana modes⁶³ or quantum Hall effect.⁶⁴

Even though these materials are promising, protecting the materials' intrinsic properties from the environment still remains a challenge. In particular, oxidation often plays an important role in 2D materials thinner than 10 nm due to increase in the surface-to-volume ratio. For example, charge-density-wave order in 1T-TaS₂ is suppressed under oxidation. In addition, Te-based materials, e.g., ZrTe₃, MoTe₂, or WTe₂, are extremely sensitive to oxidation. In the case of a thin WTe₂, within a minute in ambient condition, the absorption changes significantly and becomes almost undetectable under an optical microscope.⁶⁵ Its magnetoresistance is also significantly decreased.⁶⁶ A capping layer can protect the samples from oxidation, but the interlayer interaction may affect the intrinsic material properties or obscure the data analysis. Therefore, preserving the properties and understanding the environmental effects need to be well-characterized for future applications.

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