# Two-Dimensional Crystals: Managing Light for Optoelectronics

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ABSTRACT Semiconducting two-dimensional (2D) crystals such as MoS_2 and WSe_2 exhibit unusual optical properties that can be exploited for novel optoelectronics ranging from flexible photovoltaic cells to harmonic generation and electro-optical modulation devices. Rapid progress of the field, particularly in the growth area, is beginning to enable ways to implement 2D crystals into devices with tailored functionalities. For practical device performance, a key challenge is to maximize light−matter interactions in the material, which is inherently weak due to its atomically thin nature. Light management around the 2D layers with the use of plasmonic nanostructures can provide a compelling solution.
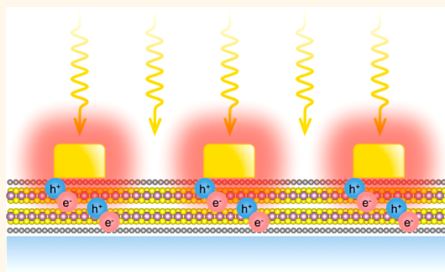
The discovery of materials with unusual physical properties often inspires exploration of the novel applications that they enable. Graphene is an excellent example: its extraordinary properties have opened up prospects for a number of emerging technologies in areas of flexible electronics, photonics, and energy storage, to name a few.[1] A recent surge of interest in two-dimensional (2D) crystals beyond graphene, such as ultrathin layers of transition metal dichalcogenides (TMDs), is justified given their newly found remarkable properties that establish them as a distinct class of materials with unique technological potential.[2−6]

> Ultrathin layers of transition metal dichalcogenides are a distinct class of materials with unique technological potential.

The family of layered TMDs thus far consists of about 40 different compounds with a chemical structure of MX_2, where M represents the transition metal and X is the chalcogen such as S, Se, or Te.[7] There are insulators like HfS_2, semiconductors such as $MoSe_2$, semimetals such as WTe_2, and metals such as NbS_2. Among many other layered compounds, including some metal oxides and halides, TMDs have been the subject of study for over half a century because of the rich physics of their quasi-2D properties. It is worth noting that some of the first examples of what we refer to as "2D crystals" today were routinely produced and studied by researchers in the 1960s.[8] These ultrathin crystals, having a thickness of just a few nanometers, led to early observations of exciton confinement effects.[8,9]

Researchers are now revisiting the subject with a fresh perspective and inspiration from graphene. Recent studies focusing on 2D crystals of semiconducting TMDs such as MoS_2 and WSe_2 have demonstrated their potential for novel optoelectronics as partly discussed in recent reviews.[2−6] Here, we focus on the potential of 2D TMDs in optoelectronics and discuss a key area of future investigation involving plasmonics.

Unique Optical Properties. One of the most distinct features of single layers of group 6 TMDs is that they are direct gap semiconductors, unlike their bulk counterparts, which are indirect gap semiconductors. This means that single layers absorb and emit photons efficiently *via* transitions at the fundamental gap. Photoluminescence and electroluminescence are therefore unique signatures of single layers.[10,11] Above the fundamental band gap, strong interband transition peaks appear in absorption.[12] This reflects a heavy effective mass of d electrons and van Hove singularity peaks

in the electronic density of states, which ensure relatively strong light—matter interactions.[13,14]

Owing to geometrical confinement and weak dielectric screening, excitons and trions, or charged excitons, are strongly bound. Features of these quasi-particles show up in the photoluminescence spectra even at room temperature.[15,16] It has also been shown that the charge on trions can be tuned by electrostatic or chemical doping.[15,16] Lack of inversion symmetry in single layers (and odd number layers) allows optical control over valley and spin polarization,[17−19] and observation of second harmonic generation.[20] The large elastic strain that single layers can accommodate permits extensive strain engineering of the band structure and corresponding optical properties of the material.[21−24]

Heterostructured TMDs for Optoelectronics. Semiconducting TMDs, particularly MoS_2, $MoSe_2$, WS_2, and WSe_2, have long attracted interest for their use in photovoltaic devices.[24] In their bulk forms, these materials exhibit band gaps in the near-infrared frequency range that are suitable for absorption of the solar spectrum. Simple solid-state photovoltaic cells based on Schottky junctions, for example p-WSe_2/Al, yielded decent efficiencies (>5%) in early studies.[26] Photoelectrochemical solar cells based on a WSe_2 photoanode achieved an impressive power conversion efficiency of 14%.[27] However, further development in this area of research was hampered in part by the absence of techniques that enable precise control over the material structure and implementation into complex devices. For example, growth of large-area, high-quality, and oriented TMD thin films has remained a key challenge. Conventional growth techniques, such as chemical vapor transport, are also not suitable for realizing heterostructures such as p–n junctions (although it is possible[28,29]), which are an essential building block of optoelectronic devices.

Recently, progress in growth and synthesis has opened up new

avenues for the implementation of TMDs into devices. For example, successful growth of centimeter-scale MoS_2 single- and few-layer films by chemical vapor deposition (CVD) has enabled fabrication of thin film photodetectors with promising photogains, fast optical responses, and detectivity.[30,31] Controlled deposition of exfoliated TMD sheets from the solution phase is also a viable route to realizing heterostacks for thin film devices[32−34] and composites with photoelectrochemical functionalities.[35] With the current rapid progress of the field, it would not be surprising to witness scalable growth and synthesis of complex functional 2D heterostructures in the near future.

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Simple stacking of 2D crystals offers an unconventional route to realizing heterojunctions and artificial crystals. Because of the flat and dangling-bond-free surfaces of 2D crystals, interfaces between different layers are atomically sharp and nearly defect-free.[36] This ensures electronically clean and abrupt interfaces between metallic, semiconducting, or insulating 2D layers. Stacking allows on-demand fabrication of semiconductor junctions with the desired band alignment (i.e., type I or II) using a library of numerous semiconducting 2D crystals.

Britnell et al.[37] recently reported that simple 2D heterostructures consisting of a semiconducting layer (WS_2, MoS_2, and GaSe) sandwiched between graphene sheets exhibit photovoltaic effects with external quantum efficiencies as high

as 30%. Because all key components of the device are 2D crystals, these devices can be fabricated on flexible substrates without severe degradation in performance. This work motivates realization of more complex heterostructures with improved performance or other functionalities such as lighting.

Two-Dimensional Dilemma. Light—matter interactions are strong for TMDs, but it is inherently weak in 2D crystals in absolute terms because the optical absorption length is extremely short (<1 nm for single layers). Single-layer MoS_2 absorbs as much as 10% of incident photons at the excitonic resonances (615 and 660 nm) and even more strongly at shorter wavelengths (430 nm) where interband transitions occur.[11] However, absorption is substantially weaker in nonresonant conditions. Trends in absorption spectra are similar for other semiconducting TMDs except that resonances lie in different spectral regions (see Figure 1).[38,39]

Weak absorption is a critical issue for photovoltaic applications of single- and few-layer TMDs. Recent calculations by Bernardi et al.[40] have shown that a power conversion efficiency of 1% can be achieved with single- and bilayer devices, which corresponds to an exceptionally high power density. However, significantly higher efficiencies need to be demonstrated for practical applications. Additional layers can help in absorbing a higher fraction of incident photons, but absorption exceeding 90% is only achieved in the thickness regime where the benefits of 2D crystals are compromised. How can one harvest sunlight with a slab of semiconductor that is 1000 times thinner than the active layer of conventional thin film solar cells?

Managing Light around an Atomically Thin Slab. There are several approaches for enhancing light—matter interaction in thin films.[41,42] Ideas so far demonstrated for graphene (summarized in recent review articles[43,44]) include use of localized surface plasmons,[45,46] surface plasmon polariton
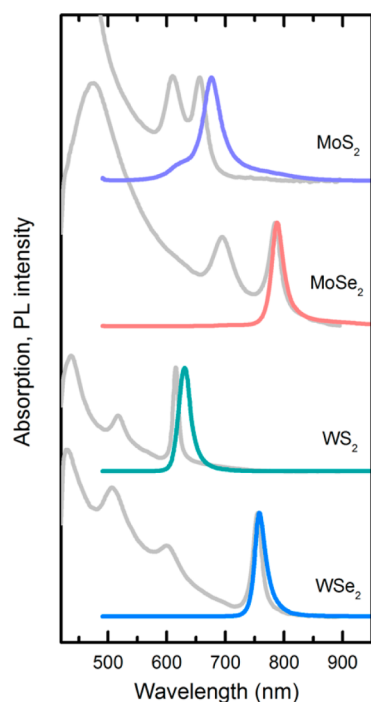
Figure 1. Absorption (gray lines) and photoluminescence (color lines) spectra of single-layer semiconducting transition metal dichalcogenide crystals (MoS$_2$, MoSe$_2$, WS$_2$, WSe$_2$). Measurements were made on layers produced by mechanical exfoliation of bulk crystals. The photoluminescence peak coincides with the band gap exciton absorption peak. The absorbance at the band gap excitonic resonance is roughly 10% for all the materials. Absorption spectra were obtained by differential reflectance measurement as described in ref 35. Photoluminescence spectra were obtained with excitation wavelength of 472 nm.

> How can one harvest sunlight with a slab of semiconductor that is 1000 times thinner than the active layer of conventional thin film solar cells?

(SPP) states,[47] integrated Si waveguides,[48] reflector microcavities,[49] planar photonic crystals (PCCs),[50] and quantum dots.[51] All of these strategies are viable for boosting the light—matter interaction in semiconducting 2D crystals.

Plasmonic nanostructures can achieve dramatic changes in the optical response of a nearby semiconducting layer while being minimally invasive in terms of device structure and integrity. These structures improve optical absorption in the semiconductor layer by (i) scattering the incident ray of light, which increases the interaction length; (ii) trapping light by excitation of localized surface plasmons, which enhances the absorption cross section; or (iii) trapping and guiding light *via* SPPs at the metal—semiconductor interface, which increases the interaction length and cross section. As summarized in the recent review articles, the benefits of using plasmonic nanostructures for graphene-based devices have been demonstrated in a number of studies,[52,53] also including the possibility of nanostructuring the graphene itself to support localized surface plasmons in the mid-infrared.[47,54]

In the work by Britnell et al.,[37] a thin layer of gold was deposited on the upper graphene electrode and subsequently annealed. This process led to coagulation of the gold film into nanoparticles that are 5—10 nm in size (see Figure 2 for device schematic). The authors observed enhancement in the photocurrent by a factor of 10 when illuminated at visible frequencies. Since gold nanoparticles of such sizes have surface plasmon resonances between 500 and 600 nm, it is possible that the enhancement is at least partly due to an improved absorption cross section.

Somewhat similar observations were made in independent work by Lin et al.[55] studying the effects of 15 nm gold nanoparticles deposited on MoS$_2$ phototransistors. They found a 10-fold enhancement in the photocurrent with gold nanoparticles. The authors show that the photocurrent spectra are significantly modified with a peak near the surface plasmon resonance of the gold nanoparticles, again suggesting the potential effect of the near field at the metal surface.

At this stage, it is early yet to claim the benefits of plasmonic effects in these devices. Conclusive evidence for the contribution of plasmonic effects is often difficult to obtain because metal nanoparticles introduce multiple effects and complicate the device operation mechanism. For example, resonantly excited charges of the metal nanoparticles can inject into the semiconductor layer,[56] leading to enhancement in the photocarrier concentration. Energy transfer, space-charge layer at the metal—semiconductor interfaces, ohmic losses, and diffuse scattering by the particles can also modify the way light is harvested. Overall effects do not guarantee improved device performance. Nevertheless, it is encouraging that in the above two studies, simple postdeposition of nanoparticles on either vertical or planar devices led to significantly positive effects. It is also worth noting that the use of graphene electrodes in vertical heterostack devices allows plasmonic nanostructures and inner semiconductor layers to be separated at a desired distance, which is a critical parameter for realizing plasmon enhancement (Figure 2).

Future Outlook. Use of plasmonics and metamaterials to boost the
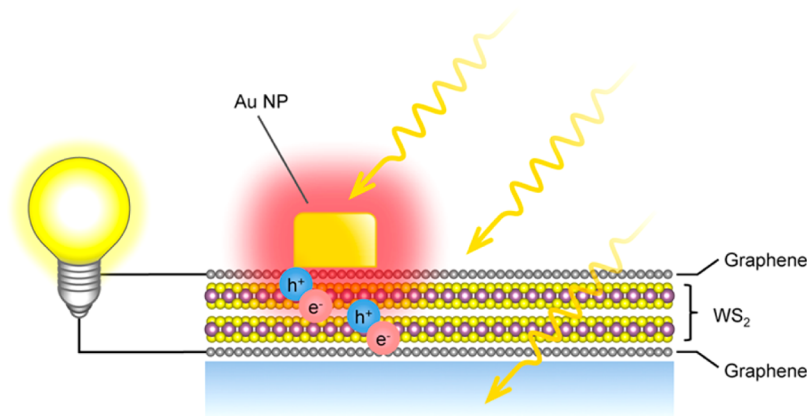
Figure 2. Schematic illustration of a light-harvesting device consisting of a vertical heterostructure of two-dimensional crystals.[37] The device consists of a semiconducting transition metal dichalcogenide (WS$_2$) as the active layer sandwiched between two graphene transparent electrodes. A plasmonic nanoparticle (NP) is depicted as trapping the incident light through localized surface plasmon resonance and enhancing exciton generation in its vicinity.

light−matter interaction in 2D crystals is a viable route to enabling practical devices as well as fundamental studies. Besides the simple use of metal nanoparticles, there are many potential strategies to guide light efficiently into and out of ultrathin semiconductor layers.

Along with the work on graphene thus far, much can be learned from ideas demonstrated with quantum-dot thin films and quantum-well devices where the active region is typically below 100 nm. For example, a 20-nm-thick layer of CdSe quantum-dot layer deposited on a Ag film has been found to absorb light confined into SPPs efficiently.[57] Similarly, photoluminescence[58] enhancement in In-GaN quantum wells *via* SPP coupling has been demonstrated. These ideas can be also applied to 2D crystals, not only for improving optical absorption and photoluminescence intensity but also for enhancing harmonic generation[59] and electroluminescence[60] intensities.

At this point, we have only seen a glimpse of what can be achieved with plasmonic nanostructures and 2D crystals. However, there is no doubt that the intersection between these two rapidly growing fields of research has much to offer to the scientific community and future technology.

Conflict of Interest: The authors declare no competing financial interest.

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