Review

Qijie Ma, Guanghui Ren*, Arnan Mitchell and Jian Zhen Ou*

Recent advances on hybrid integration of 2D materials on integrated optics platforms

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Abstract: The burgeoning research into two-dimensional (2D) materials opens a door to novel photonic and optoelectronic devices utilizing their fascinating electronic and photonic properties in thin-layered architectures. The hybrid integration of 2D materials onto integrated optics platforms thus becomes a potential solution to tackle the bottlenecks of traditional optoelectronic devices. In this paper, we present the recent advances of hybrid integration of a wide range of 2D materials on integrated optics platforms for developing high-performance photodetectors, modulators, lasers, and nonlinear optics. Such hybrid integration enables fully functional on-chip devices to be readily accessible researchers and technology developers, becoming a potential candidate for next-generation photonics and optoelectronics industries.

Keywords: 2D materials; integrated optics; hybrid integration; optoelectronic devices.

1 Introduction

It has been more than 150 years since the scientific research was first carried out on layered materials. However, at the beginning of the 21st century, the rediscovery of graphene by Novoselov et al. formally opened

the door to the scientific studies and innovations of two-dimensional (2D) atomic- thin crystals [1, 2]. As inspired by the accessible realization of truly atomically thin graphene, a wide variety of other 2D materials have been successfully synthesized, such as black phosphorus (BP), transition metal dichalcogenides (TMDs), and hexagonal boron nitride (hBN) [3, 4]. These ultrathin 2D materials symbolize a rising category of materials at the nanoscale, whose thickness is only one or a few atomic layers [5, 6]. Their extremely thin structure reduces the dimensionality of the material resulting in unprecedented features, including mechanical, physical, optical, and electronic properties, which have the potential to deliver ultra-strong composites, fast low-power electronics, and highly responsive sensors for military, biomedical, and industrial applications among others [7, 8]. A host of recent scientific studies have shown that the thin-layered crystalline form of some 2D nanomaterials can result in unique optical and optoelectronic properties [9, 10]. It is anticipated that these emerging optoelectronic 2D materials will form the basis of efficient, compact, high-speed, and broadband optoelectronic devices opening new avenues for cost-effective manufacture. In addition, their properties can be widely tuned through chemical doping, geometrical tailoring, external electromagnetic fields, or environment varying, which provide great opportunities to improve the performance of the optoelectronic devices [11–13].

Traditionally, the optoelectronic devices are based on a homogeneous material system chosen for a particular functionality such as low loss guidance or optical activity. For example, the most popular platform currently is silicon photonics which has emerged as a mature technological platform allowing multiple optical functions to be integrated onto the same chip. Electro-optic modulators [14] and low-loss silicon waveguides [15] are now an industrial reality. The ability to epitaxially grow Si/Ge on silicon has also enabled low noise and high-speed photodetectors to be added to this system [16]. However, light emission and nonlinear functionality in silicon turns out to be intrinsically limited. As a potential solution, photonic technology researchers have explored hybrid integration where proven

***Corresponding authors: Guanghui Ren**, School of Engineering, RMIT University, Melbourne, VIC 3000, Australia; and Integrated Photonics and Applications Center (InPAC), RMIT University, Melbourne, VIC 3000, Australia, e-mail: guanghui.ren@rmit.edu.au. <https://orcid.org/0000-0002-9867-8279>; and **Jian Zhen Ou**, School of Engineering, RMIT University, Melbourne, VIC 3000, Australia, e-mail: jianzhen.ou@rmit.edu.au. <https://orcid.org/0000-0002-6971-2634>

Qijie Ma: School of Engineering, RMIT University, Melbourne, VIC 3000, Australia

Arnan Mitchell: School of Engineering, RMIT University, Melbourne, VIC 3000, Australia; and Integrated Photonics and Applications Center (InPAC), RMIT University, Melbourne, VIC 3000, Australia

optically active materials such as III–V semiconductors are introduced to established silicon photonic platforms [17]. Techniques include epitaxial growth of III–V materials on selective areas of a silicon wafer, molecular bonding of small pieces of material, or even simple adhesive bonding of pieces of material or even whole processed devices onto the silicon platform [17, 18]. These approaches can break with the mass manufacture philosophy of silicon microtechnology, but offer flexibility to rapidly adapt platforms which is attractive in modern manufacturing models [19, 20]. Researchers in 2D materials are also exploring hybrid integration. Conceptually, hybrid 2D materials, also referred to as “Van de Waals (vdWs)” heterostructures, involve stacking of atomic planes layer by layer in a specific sequence [21]. Different from the conventional heteroepitaxy, arbitrary creations of vdWs heterostructures can be delivered thanks to the considerable freedom provided by the dangling-bond-free surfaces of atomic sheets. This characteristic allows the integration of highly disparate 2D materials but can be extended to the integration of mixed-dimension materials (e.g. 2D materials with 0-dimensional quantum dots (QDs), with 1-dimensional nanowires, or with 3-dimensional bulk materials) [22]. 2D-2D vdWs heterostructures enable the manipulation and control of material properties, either to circumvent certain disadvantages of 2D materials or to enhance the advantages of distinct materials. The development of a mixed-dimensional hybrid structure has increased the possibility of novel optoelectronic devices with multiple functions and high performance [23]. While they are not as mature, 2D materials are far more accessible and flexible than their III–V counterparts, and thus many researchers are exploring harnessing 2D materials to enhance more mature integrated photonic platforms [24]. Indeed, since 2D materials can in principle offer many desired functionalities, they may prove to be more adaptable for integration onto silicon chips using simple, cheap, and scalable post-processing techniques [25, 26].

In this paper we review the state of the art of science, technology, and applications of integrated optoelectronic devices and systems harnessing hybrid integration of single- or few-layer 2D materials onto integrated optics platforms. A concise overview on the properties of the mainstream 2D materials is introduced in Section 2. In the following section, different optical properties and the latest experiment methods of 2D materials are classified and compared in four application fields (photodetector, modulator, laser, and optical nonlinearity). Finally, a conclusion on the recent progress toward 2D materials-based

optoelectronic applications is provided, followed by a prospect of 2D materials and their potential applications.

2 Basic properties of 2D materials

In this section, the properties of example 2D materials are summarized in the order of graphene, BP, TMDs, and others.

2.1 Graphene

In 2004, Novoselov and his team retouched graphite and became the first to isolate high-quality monolayer graphene with the assistance of scotch tape, introducing the so-called mechanical exfoliation method. Compared to conventional semiconductor materials, graphene demonstrates outstanding properties in its tuneable electronic structure, strong light-matter interaction, high third-order optical nonlinearity, and robust mechanical ductility [27].

The crystal structure of the monolayer graphene is a hexagonal lattice of carbon atoms that is arranged in a single atomic-thin sheet [27, 28]. Single-layer graphene possesses a gapless and semi-metallic band structure and ultrabroad optical interaction (from FIR to UV, see Figure 1B) which can be attributed to the meeting points, or Dirac points, linking the conduction and valence bands. In addition, owing to the tunability of the electronic structure of graphene, the Fermi level can be tuned up to 1 eV through electrical gating although graphene has a zero bandgap, presented in Figure 1A. This feature enables ultra-broadband light modulation across the visible to infrared spectral range [2, 12, 29]. Moreover, the electronic structure of graphene can be engineered through other methods including chemical doping, geometrical tailoring, or mechanical stretching [13, 29]. The low-defect crystallinity that can be achieved with graphene leads to high mobility, $\sim 10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature [30]. Graphene also exhibits its unique nonlinear optical properties, associated with its extremely high third-order susceptibility [13, 29]. For example, it can serve as a saturable absorber where the absorption of a monolayer graphene can reach up to 2.3% for vertical incident light at visible wavelengths [8, 10, 28]. Another attractive property of graphene is that it can remain stable under an extreme chemical environment and has a ductile structure under mechanical stress [12]. All these excellent features have led graphene to be the most intensely studied 2D material

Figure 1: Bandgap of 2D materials and their corresponding operation wavelength.

(A) A broad library of two-dimensional layered materials (2DLs) with varying chemical composition, atomic structures, and electronic properties, with an increasing bandgap from left to right. (B) Radiation wavelength from terahertz to ultraviolet. The 2D material-based applications cover a wide range of radiation wavelengths from the THz through UV range (A reproduced from [23] with permission from Nature Review Materials).

in various application fields such as modulators, photo-detectors, biomedical sensing, and many other nanoscale devices [28].

2.2 Black phosphorus

With intense studies on the properties of graphene, BP was reported as a new member of 2D material family in 2014 [31]. Different from graphene, BP is a layered orthorhombic crystal within which the phosphorus atoms are arranged in a puckered lattice, leading to strong in-plane anisotropy in electrical and optical conductivity [32]. In a monolayer BP, each phosphorus atom leverages sp^3 hybridized orbitals to bond with its three neighboring atoms [8, 31].

One of the fascinating properties of BP is its tuneable direct bandgap [11, 33], ranging from 0.3 to 2 eV (see Figure 1A), by decreasing the number of layers. BP also demonstrates high mobility reaching $1350 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature, exceeding the electron mobility of TMDs [13, 34]. Therefore, these features together can provide new opportunities to develop optoelectronic devices at the telecommunication wavelength range [32].

Besides, the intrinsic anisotropic crystalline structure of BP leads to high anisotropy in optical absorption and photoluminescence [8, 34, 35]. Its nonlinear absorption was observed to increase with the enhancement of the incident light. It has also been demonstrated that the nonlinear absorption of BP can be affected by light polarization and thickness variation. One potential application

for BP is as a saturable absorber [31]. The response can be sufficiently fast that it can enable passive mode locking of ultrafast pulsed lasers [36].

On the other hand, the stability of few-layer and nanosheet BP is a barrier hindering extensive applications [8, 12]. Recently, it was reported by researchers that a simple and effective approach has been found to fabricate BP from bulk to ultrathin multilayers which can remain stable for up to 2 weeks. Hence, a deliberate fabrication process and appropriate packaging are thought to be an important solution to the stability issue [31, 37].

2.3 Transition metal dichalcogenides

Within the 2D material family, TMDs have significantly expanded the scope of characteristics that can be achieved [38]. These atomically thin semiconductors typically have the chemical formula of MX_2 , where M denotes a transition metal coming from groups IV–X in the periodic table, while the element X represents a chalcogen within which sulfur, tellurium, or selenium is often used. The similarity of the layered structure between graphene and TMDs is that the layers in the bulk TMDs are stacked in the form of the weak van der Waals force, leading to the use of a cost-effective production method, namely mechanical exfoliation, to obtain a monolayer TMD [39, 40].

One of the most remarkable features of TMDs is their sizable bandgap, covering the energy range from 1 eV to 2.5 eV and beyond; refer to Figure 1A [29, 35, 38]. According to the band structure of TMDs, the positions of the

conduction band and the valence edges can change with the variance in thickness. By reducing the thickness of the TMDs from few-layer to monolayer it is possible to transform the band structure from indirect to direct [41, 42]. This offers great opportunities for realizing light generation functions including lasing and light-emitting diodes (LEDs) [8, 11, 33]. More importantly, monolayer TMDs have been found to have a spin-orbit coupling intrinsically as they lack inversion symmetry [43]. Their asymmetry in the crystal structure leads to spin splitting of the electronic bands that are driven by the spin-orbit interaction. This property renders them a good candidate for spintronic applications [44].

Speaking of their optical properties, monolayer TMDs possess particularly strong photoluminescence, where the single-layer emission is more than three orders of magnitude brighter compared with that of the bulk [45, 46]. Unlike the broadband optical absorption due to conductivity of graphene, the optical absorption of monolayer TMDs can reach ~10% from near infrared (NIR) to UV frequencies, which is mainly caused by the excitonic transitions [12, 39]. Besides, TMDs demonstrate a high nonlinear optical response caused by Pauli blocking and hot electron excitation [13]. Accordingly, their excellent nonlinear optical absorption can be well applied in photonic devices including mode-locking and all optical switching among others [39]. Interestingly, TMDs are found to possess electron valley relaxation and picosecond-level polarization, meaning atomically-thin TMDs including MoS_2 , MoSe_2 , WS_2 , and WSe_2 are deemed to be the prospective 2D materials in a recently emerging field – valleytronics. This property allows the development of data storage or manipulation by discrete values from the crystal momentum to be practical [47, 48].

2.4 Other 2D materials

Other 2D materials have also been widely explored by researchers as the aforementioned materials have their limitations such as zero bandgap of graphene, strong optical absorption at resonances, a relatively large bandgap of TMDs, and intrinsic ambient instability of phosphorene [49]. Therefore, it is necessary to give an overview on the versatile properties of other emerging 2D materials in the 2D ultrathin family [10].

First, hBN, a graphene-like 2D material, is a crystal and layered compound semiconductor. Its properties including low dielectric constant, high piezoelectricity, and stability at high temperature are similar to those of

graphene. It has become an ideal 2D material for deep UV optical devices due to the large bandgap, reaching 5.6–6 eV (Figure 1A) [50, 51]. Except hBN, graphene analogues including topological insulators (i.e. Bi_2Se_3 , Bi_2Te_3 , and Sb_2Te_3) fill up the lacuna of broadband photonic devices [39]. On the other hand, BP analogues have been extensively investigated as well, such as SnS, tellurene, bismuthine, and antimonene. These 2D materials are featured with high nonlinear optical response, good stability, and sufficiency in supply [49].

Secondly, metal-halide perovskite semiconductors have not attracted significant attention until its unusual optical and electronic properties had been applied in the solar cells with super high efficiency [52]. With the high demand of energy efficiency, the ability of the device hybridization in forms of plasmonics and nanocavity structure is notable to facilitate the development of more efficient devices [53, 54]. Their unique optical properties also show the tunability in exciton binding energy, ultralong carrier lifetime, the prominence in nonlinear optical gain, etc. Compared with traditional semiconductors, halide perovskites stand out as a competitive optical media for the integration with optical cavities [55].

Thirdly, metal oxides have been extensively studied for their excellent biocompatibility, chemical stability, and electronic and optical properties. This group contains, but is not limited to, ZnO , SnO_2 , WO_3 , TiO_2 , and Fe_2O_3 [56]. Specifically, TiO_2 -based 2D nanosheets possess a larger bandgap due to the size quantization compared to their bulk counterparts such as rutile and anatase TiO_2 , where, for instance, the bandgap of $\text{Ti}_{0.91}\text{O}_{2.036}$ -nanosheets (3.8 eV) is larger than that of anatase TiO_2 [57]. Thanks to their unique physicochemical characteristics, they have attracted researchers' interests in various energy applications (e.g. energy storage, energy conversion, etc.) [58].

The aforementioned 2D materials are only a few among many, many more. It is necessary to explore more novel and achievable 2D materials to accelerate the development of optoelectronic devices in the future [59, 60].

3 Applications of 2D materials in optoelectronics

This section reviews the applications of 2D materials in the categories of photodetectors, modulators, lasers, and nonlinear optics.

3.1 Photodetectors

The combination of 2D materials and photodetectors provides a promising future for the interface between optical and electronic domains. Basically, the working mechanism behind the photo-detection devices involves converting light signals to electric signals, where three processes occur in sequence: light harvesting, exciton separating followed by charge carrier transport to corresponding electrodes [22]. So far, the reported operation mechanisms for hybrid photodetectors contain the photovoltaic effect, photo-thermoelectric effect, photo-gating effects, photo-bolometric effects, photoinduced charge tunneling as well as field effect doping [61]. The operation wavelength regimes of the photodetectors can involve multiple spectral regimes [34, 62]. For those photodetectors working among the short-wavelength range (from UV to NIR), photovoltaic and photoconductive effects are often employed to obtain exceptional performance regarding responsivity, noise level, and response speed. For the photodetectors operating in the long-wave infrared (LWIR) and THz range, they are dependent on the photo-thermoelectric effect behind which temperature-difference-driven voltage is the dominant factor [63]. Also, 2D material-based photodetectors should take several critical parameters into account,

including photoresponsivity, response speed, detectivity, and fabrication cost. Generally, photodetectors can be categorized into photoconductor, photodiode, and phototransistor [31, 47]. The following subsections will review these three photodetector categories based on device structures including Schottky barrier (SB), p-n junction, metal-semiconductor-metal (MSM). A brief summary of the demonstrated photodetectors is illustrated in Table 1.

3.1.1 Photoconductors and photodiodes

Photoconductors and photodiodes are two-terminal devices in which incident light leads to an electrical current flow caused by excitation of numerous electrons. Photoconductors can find application in high-speed optical communications and high-resolution imaging [79, 80]. For example, a photoconductor with the hybridization of methylammonium lead triiodide ($\text{CH}_3\text{NH}_3\text{PbI}_3$) perovskite thin films and TMD WS_2 monolayers (Figure 2A) has been first fabricated by Ma et al. Combining the optical advantages of WS_2 and $\text{CH}_3\text{NH}_3\text{PbI}_3$, this simple bilayer structure showed a high responsivity of $\sim 17 \text{ A W}^{-1}$ with a photodetectivity of $\sim 10^{12}$ Jones (Figure 2B), and significant suppression in dark current because of the interfacial charge transfer between the bilayer structure [81].

Table 1: 2D materials-based photodetectors with key parameters.

2D materials	Active layer	Spectral range	Responsivity (A W^{-1})	Response times (ms)	Detectivity (Jones)	Ref.
Graphene	SL graphene	NIR	0.085	—	—	[64]
	PMMA overlayer on SL graphene	Visible	95	0.04	2.1×10^{12}	[65]
	HfO ₂ capped graphene nanoribbons	Visible-MIR	1.75 at 632 nm 1.5 at 1.47 μm 0.18 at 10 μm	—	—	[66]
BP	FL BP/MoS ₂	Visible-NIR	22.3	0.015	3.1×10^{11}	[67]
	FL BP	Visible-MIR	82	—	—	[68]
	FL BP	NIR telecom band	0.675	—	—	[69]
	FL BP	Visible-NIR	7×10^6 (at 20 K)	5	—	[70]
	hBN/black arsenic phosphorus/hBN	MWIR	0.19	—	—	[71]
TMDs	WS ₂ /MoS ₂ /gold nanoparticles	Visible	1090	—	3.5×10^{11}	[72]
	FL ReS ₂	Visible	16.14	—	—	[73]
	PdSe ₂ /MoS ₂	LWIR	42.1	0.065	8.21×10^9	[74]
	Bilayer PdtSe ₂	LWIR	4.5	—	7×10^8	[75]
Others	Bilayer WS ₂ /CH ₃ NH ₃ PbI ₃	UV-visible	17	2.7	10^5	[76]
	SL graphene/CH ₃ NH ₃ PbI ₃	UV-visible	180	87	10^9	[77]
	SL graphene/CH ₃ NH ₃ PbI ₃ /gold nanoparticles	Visible	2.1×10^3	1500	—	[78]

BP, black phosphorus; FL, few layers; LWIR, long-wave infrared; PMMA, poly(methyl methacrylate); SL, single layer; SLG, single-layer graphene.

In contrast to a photoconductor, a photodiode is configuration to operate under a reverse bias potential and is typically characterized by low dark current and rapid response time [80]. Moreover, to overcome the bottlenecks in the development of Si-based photodetectors, the exploitation of the internal photoemission in a Schottky diode has been proposed. This configuration, with the metal-Si interface, allows photoexcited carriers from the metal to be emitted to Si over a potential Φ_b , namely SB. The advantage of this structure is facile fabrication and integration with complementary metal-oxide-semiconductor (CMOS) technology, broadband operation, and material structure without complexity [64, 76]. Accordingly, graphene-semiconductor heterostructures, as well as quantum dot integration [65] and plasmonic nano-antenna integration [82], have been proposed to realize high-responsivity photodetectors for the telecommunication applications. Goykhman et al. demonstrated a metal-graphene-Si hybrid structure which facilitated surface plasmon polariton guiding and benefited from optical confinement due to the Schottky interface, as can be seen in Figure 2C. This led to an increase in the responsivity to 85 mA W^{-1} (Figure 2D), corresponding to 7% internal quantum efficiency, at $1.55 \mu\text{m}$ with 20 nA dark current, which is one order of magnitude higher than the conventional photodetector based on the metal-Si structure. The on-chip waveguide-integrated metal-graphene-silicon plasmonic Schottky photodetector also paves the way for silicon photonic integration [76]. Most recently, Park et al., however, criticized the current graphene-semiconductor heterojunction with high Schottky barrier height (SBH) for the less than 100% quantum efficiency of the minority carrier photodiodes which cannot fulfill the advanced applications for high internal signal amplification. They demonstrated a graphene-insulator-silicon (GIS) hybrid structure to achieve photocurrent amplification while keeping low SBH, low dark current, and high responsivity. This is mainly due to the fact that a thin oxide layer can lower the leakage current in the dark state and photo-induced SBH while the silicon substrate acts as a light absorber and a light-induced carrier re-distributor. Their as-fabricated GIS diode demonstrated a high responsivity reaching 95 A W^{-1} and a detectivity of $2.1 \times 10^{12} \text{ cm Hz}^{1/2} \text{ W}^{-1}$ at an optical power density of $35 \mu\text{W cm}^{-2}$ [64].

Beyond the Schottky diode, the p-n junction diode is another structure offering improved sensitivity, low dark current, and reduced carrier transit time and diode capacitance under reverse bias [83]. Ye et al. fabricated a heterojunction diode by taking the synthetic effects of BP and MoS_2 , which can detect the light from the visible to NIR range. Using p-type BP and n-type MoS_2 , the photodiode

demonstrated highly electrically gate-tuneable current-rectifying features with the forward-to-reverse bias current ratio of more than 10^3 . Moreover, the BP/ MoS_2 heterojunction diode-based photodetector exhibited a photoresponsivity of 22.3 A W^{-1} at 532 nm wavelengths and 153.4 mA W^{-1} at 1550 nm wavelengths, which is two to three orders of magnitude higher than few-layer BP photodetectors reported before. The detectivity of this device reached $2.13 \times 10^9 \text{ Jones}$ at 1550 nm at room temperature [67].

3.1.2 Phototransistor

Compared with other types of photodetectors, phototransistors possess many advantages including high gain, responsivity, and signal-to-noise ratio. The channel conductance of phototransistors depends on the absorption of light. Moreover, the merits of phototransistors such as responsivity and photoconductive gain are found to be largely dominant by the properties of the channel materials [80, 84]. For 2D thin-layered materials, coupling with nanostructures (e.g. antenna arrays [85], plasmonic nanoparticle arrays [86], and Au nanoparticle plasmon-induced hot electrons for vertical charge transport [87]) has been used to circumvent the low absorption, absence of gain mechanism, and other drawbacks to achieve phototransistors with enhanced photoluminescence, ultralow current leakage, and high detectivity. Gong et al. proposed a heterostructure by stacking WS_2 and MoS_2 , with gold nanoparticles (AuNPs) sandwiched between the tunneling and blocking dielectric layers, to achieve a high-sensitivity and suppressed dark current phototransistor. Electrons trapped in the AuNPs have successfully depleted the intrinsic carrier concentration. At 520 nm wavelengths, a high photoresponsivity of 1090 A W^{-1} , ultralow dark current of 10^{-11} A , and a high detectivity of $3.5 \times 10^{11} \text{ Jones}$ have been observed under a low source/drain and a zero-gate voltage [72]. In addition to TMDs, perovskites are considered to have the ability of strong light absorption. Therefore, a few hybrid structures by stacking perovskites and other 2D materials or nanostructures (e.g. perovskite/ MoS_2 phototransistor [88], graphene/ WSe_2 /perovskite/graphene phototransistor [89], perovskite/Au nanosquares/ SiO_2 spacer/Au film [90]) have been demonstrated for obtaining superior photoconductive gain and responsivity. Specifically, Lee et al. experimentally demonstrated a photoresponse-enhanced graphene-methylammonium lead halide (e.g. $\text{CH}_3\text{NH}_3\text{PbI}_3$) perovskite heterojunction photodetector. This enhancement comes from the efficient charge transfer from the graphene to the $\text{CH}_3\text{NH}_3\text{PbI}_3$. As to the phenomenon of a

Figure 2: Demonstrations of 2D material-based photodetectors.

(A) Schematic device structure of the hybrid WS_2 /perovskite photoconductor fabricated on a C-plane (0001) sapphire substrate. (B) WS_2 /perovskite photoconductor's photoresponsivity and detectivity performance (A and B reproduced from [76] with permission from Advanced Materials). (C) Si-single layer Schottky photodetectors integrated with local oxidation of Si (LOCOS) waveguides. (D) The responsivity of metal-single layer graphene-Si and reference metal-Si photodetectors as a function of reverse bias for different optical powers coupled to the Schottky region (C and D reproduced from [64] with permission from Nano Letters). (E) Schematic of the $\text{CH}_3\text{NH}_3\text{PbI}_3$ -graphene-Au-nanoparticle photodetector hybrid architecture (E reproduced from [78] with permission from Nano Scale). (F) The cross-sectional schematic of the as-fabricated hBN/b-As_{0.83}P_{0.17}/hBN heterostructure photodetector (F reproduced from [71] with permission from Nano Letters). (G) Illustration of the BP metal-semiconductor-metal photodetector operating at 3.39 μm . The polarization of the incident laser and its incident direction are represented by yellow and green arrows, respectively (G reproduced from [68] with permission from Nano Letters). (H) Top panel: schematic image of the PdSe_2 - MoS_2 infrared photodetector. Bottom panel: optical photograph of the PdSe_2 - MoS_2 device, scale bar 5 μm (H reproduced from [75] with permission from ACS Nano).

dramatic quenching of the photoluminescence intensity in the graphene-perovskite heterostructure, the recombination of the photoexcited electron-hole pairs in the perovskite is constrained because electrons in the graphene layer transfer to the proximal perovskite layer to fill the empty states in the perovskite valence band while the photoexcited electrons remain in the perovskite conduction band. The hybrid photodetector demonstrated a photoresponsivity (180 A W^{-1}) with a $5 \times 10^4\%$ effective quantum efficiency, a photodetectivity of 10^9 Jones, and a broad detection spectrum from the UV to visible regime [77]. Another demonstration by Sun et al., utilizing the surface plasmonic effect of metal nanostructures, is that they integrated AuNPs with surface plasmon resonance into graphene/ $\text{CH}_3\text{NH}_3\text{PbI}_3$ perovskite to form a hybrid photodetector (Figure 2E). The AuNPs were designed with unique configuration that the AuNPs were physically separated from the light harvesting perovskite layer by the graphene layer, resulting in a significant enhancement in responsivity of $2.1 \times 10^3 \text{ A W}^{-1}$. The device also showed an 80% increase in response time (1.5 s), compared to other hybrid photodetectors without nanoparticle plasmons [78].

Different from phototransistors, the amount of current flowing (i.e. the drain current) in the accumulated channel of field-effect transistors (FETs) is governed by the gate voltage at a given source to drain bias [80].

Under the SB structure, FET-based photodetectors integrating with low-dimensional materials have been demonstrated. 2D materials in the TMD family are worthy of broad studies in FETs. ReS_2 , contrary to its counterparts such as MoS_2 , has been found to have a unique band structure, retaining a direct bandgap of 1.5 eV from bulk to an atomic layer. Consequently, Zhang et al. fabricated a few-layer ReS_2 -based FET back-gate photodetector, which showed a remarkable responsivity of 16.14 A W^{-1} and proved ReS_2 to be a competitive material for high-efficiency photodetectors [73]. Besides TMDs, Yu et al. designed a 10-nm-wide graphene nanoribbon (GNR) encapsulated in a high-k dielectric material (HfO_2), since an appropriate GNR size configuration significantly influences the performance of the photodetector. The device was designed with a pair of metal pads (Ti/Au) as the metal contacts, heavily p-doped silicon as the back-gate and 285-nm-thick SiO_2 as the gate dielectric. Their experimental results showed that the responsivity is ~ 8 – 10 times higher from the visible to MIR wavelength range. Correspondingly, it showed 1.75, 1.5, and 0.18 A W^{-1} responsivity at the visible, NIR, and MIR range, respectively. This dielectric environment engineering method further manifests a novel strategy to develop the MIR

photodetectors [66]. Although the properties of graphene have been highly appraised, the limitations of graphene-based photodetectors still exist. In order to achieve high responsivity, a bias voltage is adopted, whereas the device will suffer from high dark current and high $1/f$ noise (which stands for the spectral density phenomenon and f denotes frequency) and shot noise. Consequently, BP has been found to be the ideal candidate to remedy this issue because of its narrow but invariably direct bandgap and intrinsic puckered structure [68]. Youngblood et al. incorporated a gated few-layer BP photodetector on a silicon photonic waveguide, operating in the NIR telecommunication range. The results showed that their designed device has only 220 nA dark current and can reach high responsivity up to 135 mA W^{-1} and 657 mA W^{-1} in 11.5-nm- and 100-nm-thick devices at room temperature, respectively [69]. In terms of BP photodetector operating at a wide temperature range, Huang et al. reported a high-responsivity few-layer BP photodetector based on back-gate FET configuration which operated in a broad range from 400 to 900 nm at a temperature range from 300 to 20 K. The responsivity exhibited a peak value of $7 \times 10^6 \text{ A W}^{-1}$ at 20 K [70]. Alternatively, heterostructures is another option to improve device performance with regard to efficiency and small footprint. Chen et al. capitalized on an optical approach by hybridizing silicon waveguide, plasmonic nanostructures, and BP flake. This hybrid design presented a responsivity of the photodetector as high as 10 A W^{-1} and a 3 dB roll-off frequency of 150 MHz [91]. The hybrid architecture stacking various 2D materials also has the potential to expand the capacity of photodetectors such as the broaden detection wavelength range [67]. Yuan et al. demonstrated a hybrid structure made of pre-fabricated black arsenic phosphorus alloy ($\text{b-As}_x\text{P}_{1-x}$) and hBN encapsulation, as shown in Figure 2F. $\text{b-As}_x\text{P}_{1-x}$ expands the photodetection range to MIR wavelength, and hBN encapsulation provides long-term stability for the device at room temperature. This work not only expanded BP operation wavelength up to $7.7 \mu\text{m}$ but also exhibited an exterior photoresponsivity of 1.2 mA W^{-1} for $7.7 \mu\text{m}$ incident light [71].

As to the MSM architecture, Guo et al. demonstrated a BP mid-infrared MSM photodetector benefiting from low dark current and high photoconductive gain (Figure 2G). They further optimized the optical absorption and carrier mobility by fabricating the BP FET with quadruple electrodes, leading to a photodetector with a high responsivity of 82 A W^{-1} at $3.39 \mu\text{m}$ MIR wavelength. The thickness of BP thin films was only 10 nm, and this chip scale BP photodetector was also reported to operate in a picowatts power detection range [68].

In another architecture, the p-n junction has been adopted in an FET to lower the high dark current and noise power density. Long et al. fabricated a p-n heterojunction by stacking n-type MoS₂ and p-type PdSe₂ (Figure 2H). Thanks to the 2D heterostructure, both dark current and noise power density have been significantly suppressed in the built-in electrical field at the junction. The PdSe₂/MoS₂ FET-based photodetector has shown a strong photoresponsivity of 42.1 A W⁻¹ in the LWIR (10.6 μm) with high stability at room temperature. The photoresponsivity is one order of magnitude higher than that of PdSe₂ [74]. Another noble TMD PtSe₂ has been investigated for an FET-based photodetector operated in the LWIR. Yu et al. demonstrated a bilayer PtSe₂ FET device with designed defect engineering, for the first time, operated as an MIR photodetector at room temperature. It was observed to have a high responsivity (~4.5 A W⁻¹) and millisecond-level response speed at 10 μm wavelength. The detectivity was calculated to be ~7 × 10⁸ Jones under 10 μm quantum cascade laser illumination, arriving at the same level as that of MIR photodetectors for commercial use [75].

3.2 Modulators

The optoelectronic properties and flexibilities of 2D layered materials have demonstrated to be valuable for optical modulators. Modulators based on ultrathin materials rely on applying external fields (i.e. electrical gating, optical excitation, and thermal heating) to realize the manipulation of the phase, polarization, or intensity of a light beam. This leads to numerous optical modulator demonstrations on different mechanisms including electro-optic, all-optical, and thermo-optic that are all showing competitive performances, see Table 2 [11, 46].

3.2.1 Electro-optic modulators

Electro-optic modulators (EOMs) rely on refractive index variations with the application of an electric field to electrically modulate light. This is generally a very rapid response effect, and thus they are ideal candidates for data communication link applications [11]. Furthermore, the combination of 2D material and EOMs has also revealed

Table 2: Reported works of electro-optical (E-O), all-optical, and thermo-optical (T-O) modulators.

2D materials	Scheme	Structure	Bandwidth or response time	Insertion loss (dB)	Modulation depth (dB)	Modulation speed	Power consumption (fJ bit ⁻¹)	Ref.
Bilayer graphene	E-O	Waveguide	7 μs	–	~1.1	–	–	[92]
SL graphene	E-O	Waveguide	2 kHz	–	9.5	5 GHz	–	[93]
SL graphene	E-O	Surface-emitting concentric-circular grating	–	–	10	110 MHz	–	[94]
SL graphene	E-O	MZI	5 GHz	–	35	10 Gbit s ⁻¹	1	[95]
SL graphene	E-O (electro-absorption)	Waveguide	2.6–5.9 GHz	3.8 at 1580 nm	5.2	10 Gb s ⁻¹	350	[96]
SL graphene	E-O (plasmonic)	Waveguide	–	1.7 dB cm ⁻¹	5	–	–	[97]
SL graphene	E-O (plasmonic)	Deep-sub-λ plasmonic metal-insulator-metal waveguide	260 fs	1.2 dB	3.5	–	35	[98]
Bilayer graphene	E-O (plasmonic)	Waveguide	–	10 dB	9	–	–	[99]
SL graphene	All-optical	Waveguide	235 ns	~1.5	10	–	–	[100]
SL graphene	All-optical	Silicon photonic crystal cavity	–	–	–	–	–	[101]
SL graphene	T-O	Waveguide coupled to a race-track ring resonator	750 ns	–	7	–	–	[102]
SL graphene	T-O	Slow-light silicon photonic crystal waveguide	750 ns	–	5	–	3.99 mW	[103]
FL BP	E-O	p-doped silicon substrate	–	–	5	–	–	[104]
SL WSe ₂	All-optical	Silicon substrate	–	–	9.4	–	–	[105]
Graphene/hBN	E-O	Silicon photonic crystal nanocavity	1.2 GHz	–	3.2	–	1	[106]

MZI, Mach-Zehnder interferometer.

Figure 3: Demonstrations of modulators based on different operation mechanisms.

(A) Schematic illustration of the quantum cascade laser-integrated graphene modulator. Only the central few rings of the CCG (orange region) are connected together with the spoke bridges to allow electrical pumping of the quantum cascade laser (QCL) over a small active region. Light is emitted vertically from the surface and is modulated by the electrically gated graphene. (B) V_g dependence of the output power of the CCG QCL with (circles) the graphene. The pumping current of the QCL is 2.80 A (A and B reproduced from [94] with permission from ACS Photonics). (C) Optical micrograph of the Mach-Zehnder interferometer (MZI) modulator. The top arm has a 400 μm single-layer graphene on Si, the bottom arm has a 300 μm single-layer graphene on Si. (D) Cross-section of the graphene phase modulator through the dashed line A-A' of (C) (C and D reproduced from [95] with permission from Nature Photon). (E) Schematic of the cavity-graphene electro-optic modulator. The dual-layer graphene capacitor on the quartz substrate is optically coupled to the planar photonic crystal cavity (E reproduced from [106] with permission from Nano Letters). (F) A three-dimensional schematic drawing of a single-layer graphene electro-absorption modulator (EAM) integrated on the SOI platform. Cross-section of the graphene EAM (F reproduced from [96] with permission from Laser & Photonics Reviews). (G) Schematic of the electro-optic modulator, where a single Au nanodisk with a radius of 60 nm is deposited on the MoS_2 monolayers. The coupling strength between a single Au plasmon and MoS_2 exciton can be effectively tuned by the gate voltage (G reproduced from [107] with permission from ACS Nano). (H) Schematic cross-section of a graphene-cladded silicon photonic crystal cavity structure built in a silicon layer with a thickness of 220 nm and a lattice constant of 420 nm (H reproduced from [101] with permission from ACS Photonics). (I) A thermo-optic microring modulator based on graphene. Three-dimensional illustration of a thermo-optic microring modulator based on graphene; monolayer graphene is on top of a ring resonator without any separation layer (I reproduced from [102] with permission from Nanoscale). (J) Schematic of the sample and of the experimental arrangement. An array of split-ring resonators (SRRs) is patterned on the surface of a gallium arsenide substrate. A graphene monolayer is transferred on the surface and isolated by a silicon oxide layer from the SRR (J reproduced from [108] with permission from Applied Physics Letters).

competitive advantages in broadband operation, low operation voltage, CMOS technology, and miniaturization. Lin et al. demonstrated a first graphene-based waveguide EOM in the MIR region, where the waveguide was made of $\text{Ge}_{23}\text{Sb}_7\text{S}_{70}$ glass functioning as a gate dielectric and then it was sandwiched between two graphene sheets. A broadband optical modulation for the TE mode has been exhibited with a modulation depth of 8 dB mm^{-1} across the wavelength range of 2.05–2.45 μm [92]. Meanwhile, BP with its bandgap size of around 0.3 eV in its few layers is also regarded as a promising option for MIR range application. Peng et al. demonstrated a multilayer BP-based EOM in the MIR region, where the quantum confined Franz-Keldysh effect was the primary physical mechanism in their BP flake samples. They suggested that if the current device is integrated with a waveguide, a modulation depth of 5 dB can be obtained with a 100- μm -long device [104].

More importantly, although 2D layered materials exhibit a strong light-matter interaction, their absolute value is small when the multilayers are reduced to one single layer, resulting in low modulation depth. Consequently, various approaches (e.g. evanescent coupling, interference enhancement, cavities, stacked 2D materials, etc.), as well as photonic integration, have been used to increase the absorption in order to improve the modulation depth [11]. For example, graphene is also known for its broad optical response at the THz region, but it exhibits limited absorption when a terahertz signal passes through a monolayer graphene. Therefore, Mittendorff et al. employed a passive silicon dielectric waveguide to incorporate with a graphene sheet where the evanescent

field penetrated the graphene sheet so that the fundamental limitation in graphene is removed. It has been shown that a modulation depth of 50% was achieved via applying a gate voltage to the graphene sheet [93]. More importantly, to achieve both high-speed modulation and a high modulation depth that are the bottleneck in the THz modulator field, Liang et al. first proposed a graphene THz modulator integrated monolithically with surface-emitting concentric-circular-grating (CCG) THz quantum cascade lasers (Figure 3A). This idea is rooted in three benefits: first, the monolithic integration gives the miniaturization that will reduce parasitic capacitance and resistance of the device and further increase the modulation speed; second, a strong interaction between the THz radiation and graphene layer will lead to a larger modulation depth (Figure 3B); and third point is that the integration avoids one stage of optical alignment and the related bulky mirrors. The as-proposed device achieved 100% modulation depth with a fast modulation speed of more than 100 MHz [94].

Moreover, integrating graphene with silicon photonics offers a brand-new opportunity for the development of high modulation efficiency, low loss, and compact phase modulators that outperform state-of-the-art silicon devices. Sorianello et al. presented a hybrid silicon waveguide (Figure 3C and D) that was composed of a Si-insulator-single-layer graphene (SLG) capacitor and the Si waveguide served as a gating electrode to the SLG. The phase modulator based on this architecture reached a modulation efficiency of 0.28 V cm at 1550 nm, one order of magnitude higher than current depletion p-n junction

Si phase modulators. It possessed 5 GHz electro-optical bandwidth and operated at 10 Gb s⁻¹ with 2 V peak-to-peak driving voltage in a push-pull configuration for binary transmission of a non-return-to-zero data stream over 50 km of single-mode fiber [95]. Except the graphene-based phase modulators applied in the telecom or datacom field, Kakenov et al. demonstrated a graphene-based THz phase modulator that can be used in THz imaging applications. The phase modulator was composed of single-layer graphene, polyethylene membrane, and the gold reflector that was placed quarter wavelength distance from the electrolyte-gated graphene. The monolayer graphene acted as a tuneable impedance surface to yield an electrically controlled reflection phase. The voltage-controlled phase modulation of π and the reflection modulation of 50 dB have been revealed according to Terahertz time domain reflection spectroscopy. They also have been able to present a multipixel phase modulator array operating as a gradient impedance surface [109].

As low-dimensional materials can be stacked in van der Waals heterostructures to extend their functionality, 2D materials represent an exciting new platform that may be key to unlocking applications which are unsolved with conventional technologies and traditional materials [110]. Here, Gao et al. demonstrated a high-speed graphene EOM (Figure 3E) that was based on a graphene-boron nitride (BN) heterostructure integrated with a silicon photonic crystal nanocavity. Owing to the submicron cavity, the light-matter interaction with graphene has been strongly enhanced, leading to efficient electrical tuning of the cavity reflection. A high modulation depth of 3.2 dB and a cut-off frequency of 1.2 GHz have been obtained [106].

Other than the above-mentioned methods to improve the performance of the electro-optic modulation, Hu et al. illustrated an optical electro-absorption modulator (EAM) based on graphene integrated with a silicon rib TM waveguide, where it realized both ultra-high bandwidth and broadband operation that are comparable to best-in-class Si/Ge modulators for the future on-chip optical interconnects. The waveguide design, as shown in Figure 3F, aimed at maximizing the interaction between the optical field and graphene layer and minimizing the capacitance. The stronger evanescent field of the TM-like mode increased the overlap with graphene. This, therefore, led to high-speed operation and a broadband modulation speed of 10 Gb s⁻¹. A low insertion loss of 3.8 dB at 1580 nm and a low drive voltage of 2.5 V have been obtained under a broadband and athermal operation [96].

Last but not least, the plasmonic structure built in EOMs is another interesting avenue that deserves experimental explorations for miniaturized device size, intrinsic

capacitance, and on-chip photonic circuits with power efficiency and high operation speed. As graphene has been demonstrated as an ideal plasmonic material, the graphene-based plasmonic EOMs have been reported in the IR and THz regime [97, 98]. More specifically, in recent research, Ding et al. found that leak plasmonic modes provide a brand-new opportunity in the graphene-based electro-optic (E/O) modulation. Dependent on the graphene-plasmonic leaky-slot-mode waveguides, a graphene plasmonic waveguide which was fully incorporated onto the silicon-on-insulator platform was designed for effective E/O modulation. Thanks to the leaky-mode-based waveguide, it provided better modulation depth that was a tunability of 0.13 dB μm^{-1} with an extremely low insertion loss of 0.68 dB μm^{-1} . These results outperformed the graphene-plasmonic devices reported before [99]. In addition, monolayer MoS₂ has been found to have a strong exciton-plasmon interaction with metallic nanostructures, which may lead to the realization of nanoscale electro-optic devices. Li et al. presented a nanoplasmonic electro-optic modulation in the visible spectrum, based on a hybrid structure of a single Au nanodisk fabricated on the MoS₂ monolayers (Figure 3G). A deep Fano resonance attributed to the narrow MoS₂ excitons coupled with broad gold plasmons was observed, where the resonance can be further tuned by applying a gate voltage [107].

3.2.2 All-optical modulators

2D materials offer a perfect platform to realize all-optical signal processing in the photonic domain. Accordingly, the light modulation can be obtained in a simple configuration (e.g. optical fiber or waveguides) with low loss, high speed, and broadband optical signal processing [11, 111].

Saturable absorbers by using 2D materials have been studied quite extensively for all-optical signal process applications. 2D materials typically serve as a passively self-amplitude modulator that allows ultrafast pulse to generate efficiently with low power cost [112, 113].

For the all-optical integrated circuits based on silicon platforms, 2D materials revealed their advantages in micro-scale light modulation with ultrashort response time and extremely low energy consumption [100]. Previously, Shi et al. demonstrated a graphene-coated photonic crystal cavity (PCC) AOM, as shown in Figure 3H. There was a 3.5 nm resonance wavelength shift along with a quality factor change of 20% when a continuous-wave (CW) laser at 1064 nm targeted at the cavity. The special design offered a two orders of magnitude lower laser power to reach the saturated absorption state of

graphene when compared with monolayer graphene on silica [101]. Beyond graphene-based all-optical demonstrations, Xia et al. explored the effectiveness of TMDs to enhance all-optical modulation in the THz regime. They took advantage of the tuneable transmission properties of the WSe₂-silicon hybrid structure, and the results showed an 87% modulation depth with the enhanced photo-doping capabilities. This is 40% higher than the modulation depth from a bare silicon substrate [105]. Although the silicon platform has its own advantages for all-optical operation, silicon and III–V semiconductors with the bandgap close to the operation wavelength in the telecom band have intrinsic free-carrier absorption and the two-photon absorption effects. These two effects limit power handling on these platforms and place restrictions on all optical effects that can be achieved. Qiu et al., therefore, projected a hybrid structure composed of a graphene-on-silicon nitride (Si₃N₄) chip. Unlike silicon, Si₃N₄ is an insulator with the bandgap in the ultraviolet, far from the operation wavelength, and has emerged as an excellent waveguide material for high power handling and low propagation loss. Qiu et al. showed experimentally that a switching response time of 253 ns under a 50 nJ switching energy has been obtained [100].

3.2.3 Other modulation mechanisms

Other modulators including thermo-optic modulators (TOMs), magneto-optic modulators, acoustic-optic modulator, etc. are also employed for light modulation. For TOMs, the refractive index of waveguide materials changes at different temperatures [11]. Graphene, known for its distinctive thermal conductivity, is favorable for thermally tuneable photonic applications [114]. Gan et al. demonstrated a graphene-coated microring resonator integrated TOM (Figure 3I) by capitalizing on the thermal energy and electrical conductivity in a monolayer graphene. Under a 28 mW electrical power the resonant wavelength of the ring resonator has shifted by 2.9 nm, which led to a large modulation depth of 7 dB and a wide operation wavelength range of 6.2 nm [96]. Furthermore, graphene, as a transparent heater, has been integrated onto various silicon photonic crystal waveguides to demonstrate enhanced tuning efficiency and fast response time, which outperform the conventional metallic microheaters [115]. Yan et al. experimentally presented an energy-efficient graphene microheater by incorporating a monolayer graphene onto a slow-light silicon photonic crystal waveguide. With the support of the slow light, the

tuning efficiency was enhanced attributing to the fact that the large group index can be achieved in the photonic crystal waveguide which further increased the interaction length between the waveguide and the microheater. Consequently, a tuning efficiency of 1.07 nm mW⁻¹ with a low power consumption (3.99 mW per free spectral range) has been obtained. The rise and decay times (10%–90%) are 750 and 525 ns, respectively, which are so far the fastest reported response time in the microheaters integrated with silicon photonics. The proposed concept of incorporating a graphene microheater with the photonic crystal waveguide provides a prospective pathway to reduce power consumption in modulators and the potential in photonic integrated circuits [103].

In addition, Zanotto et al. found a magneto-optic response from a monolayer graphene that was strongly affected by the meta-surface of a split ring resonator. This meta-surface that demonstrated a strong electric dipole resonance in the THz region can be manipulated via magnetic and electric biasing. This hybrid meta-surface where a monolayer graphene was spaced a few tenths of nanometers (Figure 3J) can be further developed into thin optical modulators based on the magneto-optic effect in the THz spectral regime. It could also be the platform for the investigation of the cavity electrodynamics of Dirac fermions in the quantum range [108]. Light modulation based on the acousto-optic effect is another research area that makes a plethora of photonic technologies possible including acousto-optic-based devices in optical communications, photo-acoustic imaging, etc. Tadesse et al. presented an acousto-optic modulation using Lamb waves to reach a modulation width up to 19 GHz frequency in the microwave K band. By integrating a suspended piezoelectric aluminum nitride (AlN) membrane, the Lamb wave mode with very high acoustic velocity would incur as the membrane thickness was less than the acoustic wave. A nanoscale inter-digital transducer and a photonic crystal nanobeam were then deposited onto the suspended AlN membrane. This structure was demonstrated to realize acousto-optic devices with unprecedentedly high modulation efficiency and frequencies [116].

3.3 Lasers

The advent of 2D materials with the exploitation of their ultrafast and broadband nonlinear optical response and facile processability resulted in real saturable absorbers (SAs), which play an indispensable role in the ultrafast photonic devices. In the following section, 2D

Figure 4: Demonstrations of waveguide lasers based on 2D materials.

(A) Prototype and spatial configuration of the monolithic mode-locked waveguide laser based on the ReSe_2 saturable absorber. The total cavity length of the monolithic laser resonator is around 11 mm. (B) The average output power as a function of the launched power and the emission spectrum of the waveguide laser modulated by ReSe_2 . The inset is the measured near-field modal profile of the output laser (A and B reproduced from [120] with permission from APL Photonics). (C) Schematics of a microdisk cavity, consisting of a microdisk with a radius of r_d and height of h_d , supported by a PMMA (poly(methyl methacrylate)) post. The disk-shaped scatterer with a radius of r_s is located at the edge of the microdisk (C reproduced from [126] with permission from ACS Photonics). (D) Design of the waveguide-integrated LED. In the LED mode, the light emitted from the p-n junction propagates through the waveguide and is coupled out at the grating coupler. Cross-sectional schematic of the encapsulated bilayer MoTe_2 p-n junction on top of a silicon photonic-crystal waveguide. The carrier concentration in MoTe_2 is controlled by the split graphite gates, the separation of the two gates is 400 nm, the dielectric layer is h-BN on top of the MoTe_2 , and the thickness is 80 nm. The source (s) and drain (d) electrodes are thin graphite flakes connected to Cr/Au leads. (D reproduced from [127] with permission from Nature Nanotechnology.) (E) Crystal structure of MoTe_2 . Schematic of the device (the Si photonic crystal nanobeam laser structure suspended in air with a monolayer MoTe_2 on top) (E reproduced from [122] with permission from Nature Nanotechnology). (F) Schematic of a MoTe_2 -on-Si nanolaser, where the far-field optimized silicon photonic-crystal cavity is the resonator, and the trilayer MoTe_2 is the gain material (F reproduced from [123] with permission from Journal of the Korean Physical Society). (G) 3D schematic image of the fabricated $\text{MoS}_2/\text{WSe}_2$ heterobilayer-photonic crystal cavity nanolaser (G reproduced from [128] with permission from Science Advances). (H) Schematic diagram of the hBN- MoTe_2 -hBN sandwich placed on the silicon single-mode cavity. A few-layer-thick flake of MoTe_2 provides the optical gain. The MoTe_2 flake is sandwiched between two layers of hBN and is placed on the cavity (H reproduced from [125] with permission from Advanced Optical Materials).

material-enabled light sources integrated on photonic platforms will be summarized.

The waveguide laser as an active device for photonic circuit integration has been continuously gaining attention in the optoelectronic field [117]. It normally includes a resonant cavity and the gain medium. The light intensity within the waveguide cavity could significantly increase to a high level thanks to the small volume of waveguide and light enhancement of the resonant cavity. Consequently, the performance of a waveguide laser is superior to fiber or solid-state lasers in terms of low lasing threshold and enhanced slope efficiency [118, 119]. Table 3 illustrates the performance of the waveguide lasers.

Optical waveguide fabricated by femtosecond laser writing could confine light field in a microscale volume of the materials. This fabrication technique also allows arbitrary figures of compact waveguides to be achieved to further integrate with photonic devices. Among various structures, depressed cladding waveguide, featured with ambient low refractive index tracks, stands out for its easy integration with other optoelectronics and better light field confinement [120, 121]. For example, Li et al. applied ReSe_2 as an SA into a monolithic Nd:YVO_4 waveguide platform (with 50 μm diameters and 11 mm long), as presented in Figure 4A, where the depressed cladding configuration was adopted. This waveguide laser could operate in the CW mode-locking mode, reaching a high repetition rate of 6.5 GHz and a short pulse duration (29 ps) at 1 μm , as shown in Figure 4B [120]. Additionally, the same waveguide configuration was adopted to achieve a high-performance Q-switched mode-locked waveguide laser. Graphene-, MoS_2 -, and Bi_2Se_3 -based SAs have been separately demonstrated to operate at the fundamental

repetition rate (6.5 GHz) with 52, 43 and 26 ps pulse duration, respectively. It was reported that the maximum slope efficiency reached 53% in the case of MoS_2 SA. This is also the first Q-switched mode-lock waveguide laser using MoS_2 and Bi_2Se_3 SAs in the waveguide platform, which pave the way for the compact ultrafast photonic applications [121].

Besides these two micron-sized waveguide laser demonstrations featured with a Fabry-Pérot (F-P) cavity, other geometric designs of resonant cavities have been broadly investigated to realize miniature size, low insertion loss, or a strong light-matter interaction [55]. Furthermore, different modes can be employed to enhance the quality factor (Q) of the microcavities, namely the loss of the cavity, which may determine the performances of nano lasers. For example, gallery whispering modes in a micro disk can obtain a higher Q factor than that of an F-P cavity, as they allow light to travel around for many more times [29]. By leveraging the optical advantage of TMDs, WS_2 and the dielectric advantage of hBN, Ren et al. designed a heterostructure micro disk optical cavity characterized by whisper gallery modes (Figure 4C). It showed a 2D trion (negatively charged exciton) light source with a high Q factor (1200) [126].

PCC is an alternative way to integrate the ultrathin 2D materials to form a laser [129]. Bie et al. demonstrated a silicon waveguided-integrated light source based on a p-n junction of bilayer MoTe_2 (Figure 4D). The waveguide with PCC mode realized point-to-point optical links [127]. Similarly, a monolayer MoTe_2 has been integrated onto a silicon nanobeam cavity (Figure 4E) by Li et al. in 2017 [122]. They are the first to report a room-temperature laser operating in the NIR region (1132 nm) with the threshold power

Table 3: Performance summary of waveguide laser based on 2D materials.

Gain materials	Scheme	Integration platform	Central wavelength (nm)	Pulse duration (ps)	Repetition rate (GHz)	Pulse energy (nJ)	Output power	Minimum threshold (W cm^{-2})	Ref.
ReSe ₂	CWML laser	Nd:YVO ₄ crystal waveguide	1064	29	6.5	–	259 mW	–	[120]
Graphene	Q-switched mode-locked	Nd:YVO ₄ crystal waveguide	1064	52	6.436	51	622 mW	–	[121]
MoS ₂			1064	43	6.48	75	598 mW		
Bi ₂ Se ₃			1064	26	6.556	28	387 mW		
MoTe ₂	Semiconductor nanolaser	Silicon photonic crystal nanobeam cavity	1132	–	–	–	–	6.6 (RT)	[122]
Tri-layer MoTe ₂	Silicon nanolaser	Silicon photonic crystal L3 nanocavity	1080	–	–	–	1.8 mW cm^{-2}	30000 (3.5 K)	[123]
MoTe ₂	Laser-like emission	Silicon photonic crystal L3 nanocavity	1305	–	–	–	20 pW	1500 (300 K)	[124]
hBN-MoTe ₂ -hBN	Laser-like emission	Silicon single-mode cavity	1319	–	–	–	–	4200 (RT)	[125]

CWML, continuous-wave mode-locked; K, Kelvin temperature scale; RT, room temperature.

density of 6.6 W cm^{-2} , where the single-layer MoTe₂ acted as a gain medium. Also, the linewidth of 0.202 nm at room temperature was the narrowest with the corresponding Q factor of 5603. Li et al., afterward, reported an on-chip TMD-based laser source (Figure 4F) at 1080 nm at 3.5 K. One aspect worthy of note is that they finely tuned that the PCC structure through far-field optimization so that an efficient output power of 1.8 mW cm^{-2} (which is the highest lasing output from TMD-based lasers) can be achieved for a real application in silicon photonic platforms [123]. More recently, a research team demonstrated lasing operation at a longer wavelength with the same material and waveguide structure. They chose few-layer MoTe₂, served as a gain material, to integrate onto an optimized silicon photonic crystal nanocavity. This device achieved a laser-like emission at 1305 nm, which provided a future potential for an integrated electrically pumped nanoscale silicon light source with low cost [124]. The heterostructure of 2D materials is another critical building block for improving light source integration with photonic devices. In 2019, Liu et al. reported the first van der Waals heterostructure-based nanocavity laser, as shown in Figure 4G. By stacking monolayered MoS₂/WSe₂, the light emission from the hybrid material was with interlayer excitons. Then, the MoS₂/WSe₂ heterostructure has been served as a gain material to integrate on a vertically coupled, free-standing PCC where the peak optical mode field was in excellent

spatial overlap with the gain medium. The measurement results showed that the laser can be excited at 1122.5 nm, with $54 \mu\text{W}$ power threshold [128].

To effectively obtain lasing, the conventional PCC designs are criticized for their airbridge geometry where the band-edge states are unable to be suppressed. Hence, the resonance-based lasing is highly demanded. To address this, Fang et al. investigated hybrid integration with recently introduced nanocavity design – a single mode cavity which provide more degrees of freedom to obtain single-mode operation (Figure 4H). More specifically, a heterostructure consisting of a layer of MoTe₂ sandwiched between thin films of hexagonal boron nitride was proposed to integrate onto the silicon single-mode resonator. This setup enables a TMD-based light source to operate over a wider free spectra range and has improved structural robustness and heatsinking. A room-temperature laser-like emission with a high Q factor of 4500 and a threshold of 4.2 kW cm^{-2} at 1319 nm wavelength has been successfully achieved [125]. A multiresonance silicon grating-waveguide structure has been experimentally found by Chen et al. that the multiple waveguide structures coupling to a monolayer WSe₂ can simultaneously provide excitation and emission enhancement with an optimal photoluminescence. The manipulation of the intensity and directionality of the WSe₂ emission has also been realized via customizing the resonant frequencies

and dispersion. With the support of their time-resolved measurements, the structure was indicated to reduce the lifetime of the radiation decay, potentially allowing the realization of ultrafast modulation of 50 Gbps.

This experiment also demonstrated TMDs to be a versatile nanoscale light source for silicon-based chip-integrated applications [130]. Besides the above photonic cavity modes, Li et al. tailored the spin-orbit coupling (SOC) effect to manipulate the photoluminescence intensity of MoS₂ so that the issue that the weak light-matter interaction in monolayer MoS₂ is addressed. They designed plasmonic spiral rings with subwavelength dimensions and fabricated the metal-dielectric-metal hybrid structures. 2D light-emitting devices based on the SOC effect were obtained and can be actively controlled through circular light polarization. The results of their experiment can be further implemented in the spin-dependent light-emitting applications at the nanoscale [131].

3.4 Nonlinear optics

Besides making use of the nonlinear optical (NLO) properties of 2D materials to act as saturable absorbers for mode-locked lasing, these excellent properties have revealed its unparalleled merits in nonlinear optical devices. They are widely applied in industrial and medical material processing, frequency converters for bioimaging, soliton devices for telecommunication networks, etc. [2]. Compared to the conventional NLO materials, 2D layered materials with an atomically thin structure have great potential to tackle current challenges such as on-chip photonic integration and low NLO susceptibility [8].

According to the fundamentals of nonlinear optics, when the optical field is high enough, the response of materials will cease to be linear and nonlinear optical effects occur, where the higher-order ($n \geq 2$) terms give rise to radiation at frequencies different from the frequency of the incident light. In the following section,

the state of the art of 2D material-based nonlinear optics and its practical applications will be discussed. Table 4 summarizes typical NLO processes in 2D materials and their key parameters (e.g. the NLO susceptibility and conversion efficiency).

3.4.1 Second-order nonlinear effects

As to second-order nonlinear effects, it involves second-harmonic generation (SHG), sum frequency generation (SFG), difference frequency generation (DFG), optical rectification, and Pockels effect [8].

A strong SHG signal pumped by an intense light comes from the lack of inversion symmetry in the 2D materials. Recent research has focused on 2D TMDs as they have an inherently asymmetric crystal structure. In order to address the weak light-matter interactions for atomically thin 2D materials in the SHG process, various methods including microcavities [134] or 1D nanostructure integration (Figure 5A and B) [135], optomechanical platforms [136] have been proposed to enhance SHG from 2D materials. To illustrate, Chen et al. reported a single-layer MoSe₂ integrated on a silicon waveguide for SHG, as shown in Figure 5C and D. This proposed device demonstrated a 5-fold enhancement of SHG from the excitation of monolayer MoSe₂ by the evanescent field of the guide mode in a 220 nm silicon waveguide. The experimental results demonstrated that the SHG strength increases with the longer 2D TMD and light interaction length, which can be realized by integrating 2D TMD onto integrated long waveguides [140].

3.4.2 Third-order nonlinear effects

Third-order nonlinear effects include, but are not limited to, the third-harmonic generation (THG), four-wave mixing (FWM), and intensity-dependent refractive index

Table 4: NLO processes in 2D materials.

	Material	Emission wavelength (nm)	Fabrication	NLO coefficient ($\chi^{(n)}$)		η	Thickness	Substrate	Ref.
SHG	MoS ₂	780	CVD	$2.0 \times 10^{-20} \text{ m}^2 \text{ V}^{-1}$		–	ML	Glass	[132]
THG	MoS ₂	520	CVD	$1.7 \times 10^{-28} \text{ m}^3 \text{ V}^{-2}$		–	ML	Glass	[132]
	Antimonene	532	Electrochemical exfoliation	$(3.98 \pm 0.3\%) \times 10^{-9}$	$(2.88 \pm 4.5\%) \times 10^{-5}$		FL	Si/SiO ₂	[133]

η , conversion efficiency; CVD, chemical vapor deposition; FL, few layers; ML, monolayer.

change (i.e. Kerr effect and saturable absorption which is usually applied in ultrafast pulsed lasers) [8].

Different from SHG, THG can be free from the restriction on the centrosymmetric structure of the

materials. The generation process, however, is the same as SHG that three incident lights at the frequency ω can create a light radiation at 3ω frequency. Woodward et al. reported third-harmonic generation in monolayer MoS_2 for

Figure 5: NLO processes in 2D material-based devices.

(A) Schematic of local strain formed in monolayer MoS_2 . (B) The schematic setup for polarized second-harmonic generation (SHG) measurements. Inset: φ in the laboratory coordinate is the rotation angle of the sample and only the parallel component of the SHG signal is measured (A and B reproduced from [135] with permission from Nano Letters). (C) Schematic design of the hybrid integration of MoSe_2 onto a Si-waveguide. (D) Schematic side view of the grating coupler for both in- and out-coupling, where the numbers mark the physical dimensions in nm; bottom: SEM top view of the two grating couplers (C and D reproduced from [136] with permission from Nano Letters). (E) Schematic illustration of the proposed nonlinear metasurface. (F) Computed third-harmonic (TH) power outflow and TH generation conversion efficiency as functions of the incident fundamental frequency wave intensity. The TH power outflow is fitted by using a cubic power function presented by the blue dotted line (E and F reproduced from [137] with permission from Journal of Optics). (G) Schematic illustration of hybrid waveguides integrated with graphene oxide (G reproduced from [138] with permission from APL Photonics). (H) Optical microscope image of a set of waveguides. The extent of the graphene (under the contacts) is shown by the dashed lines. On top of this structure the polymer electrolyte is spin-coated (not in this image) (H reproduced from [139] with permission from ACS Photonics).

the first time. The reported $\chi^{(3)}$ value under 1560 nm excitation is $1.7 \times 10^{-28} \text{ m}^3 \text{ V}^{-2}$, which is 3.4 times stronger than that in graphene [132]. Moreover, the need for the development of multifunctional nanoscale optoelectronic devices is the momentum for research to investigate the NLO properties of not yet investigated 2D materials. Lu et al. investigated the few-layer antimonene and antimonene QDs, where their third-order nonlinear susceptibilities were $3.98 \times 10^{-9} \text{ m}^3 \text{ V}^{-2}$ and $2.83 \times 10^{-9} \text{ m}^3 \text{ V}^{-2}$ under 532 nm wavelength, respectively. These 2D materials exhibit broadband NLO response with high stability at ambient conditions, outstanding their counterpart phosphorene [133]. Research effort has also been focused on measures to enhance harmonic frequency generation since the nonlinear effects that are weak in nature usually require high input power to excite. Jin et al. patterned graphene into micro-ribbons and then integrated it with a thin dielectric spacer layer and a metallic substrate to form an ultrathin nonlinear metasurface, as shown in Figure 5E. This novel design exhibited a significantly enhanced THG at the FIR and THz region (Figure 5F). It is also worth mentioning that the resonant frequency of this structure can be adjusted through tuning the Fermi energy of graphene via electrical or chemical doping; consequently, the third-harmonic generated wave can be improved at different frequencies without changing the nonlinear metasurface configuration [137].

FWM, another third-order NLO phenomenon, occurs when two or three different wavelength incident lights interact; one or two new wavelengths will be generated. This effect has been extensively utilized for a wide variety of practical applications, for example, optical signal amplification, wavelength conversion, etc. [8]. Wu et al. demonstrated an enhanced FWM phenomenon in nanomaterial graphene oxide films and silica glass waveguide structure, as shown in Figure 5G. Thanks to a strong mode overlap between the hybrid waveguide and high Kerr nonlinearity in the graphene oxide films, along with low nonlinear and linear loss,

a significant 9.5 dB enhancement in FWM conversion efficiency has been obtained [138]. FWM can also be employed to characterize the NLO properties of 2D materials. Alexander et al. designed a degenerate FWM experiment on a graphene-based silicon nitride waveguide, as shown in Figure 5H. They have observed a strong relationship between FWM conversion efficiency and the signal-pump detuning and Fermi energy of graphene, which indicates that the optical nonlinearity is electrically tuneable [139].

Another third-order nonlinear phenomenon is the Kerr effect which manifests as an intensity-dependent refractive index change. This is an elastic nonlinear effect, where no energy is absorbed by the material. Nonlinear effects can also be inelastic (e.g. saturable absorption), where energy is transferred from the light into the material where it is absorbed [36].

4 Conclusion

This paper gives a review not only on the basic properties of both well-established and recently emerging 2D materials but also on the optoelectronic devices based on hybrid integration of 2D materials, which has not yet been comprehensively touched by existing reviews.

The advances in 2D materials lay the essential foundation for the development of next-generation photonic and optoelectronic devices. First, the versatility of the physicochemical, optical, and electronic characteristics rendered 2D materials, either being stacked monotonically or heterogeneously, facilitates the development of a wide range of optoelectronic and photonic devices including modulators, detectors, light sources and nonlinear optics. Secondly, thanks to their robust properties and flexible methods for deposition 2D materials, new methods of low-cost manufacture such as direct printing of active materials can potentially be obtained.

With the increasing studies in 2D materials, practical applications have witnessed the corresponding development. For photodetectors, not only have graphene-based photodetectors been found to have an ultrabroad operation range, but photodetectors based on other 2D materials such as BP, TMDs, and the heterostructures have been experimentally demonstrated to have high detectivity, broadband detection region with low dark current. 2D materials also provide a new pathway for the development of optical modulators. No matter what the modulation scheme (either electro-optic or all-optical) is, 2D material-based optical modulators exhibit more competitive performances in modulation speed and facile integration onto silicon photonic circuits or optical fibers. This is because silicon optical modulators are restricted by its carrier mobility, leading to low modulation speed. The on-chip light sources also see integrating with ultrathin materials a great solution for the development of optoelectronic systems and sensors. Beyond graphene, the excellent nonlinear optical response of TMDs, topological insulators and other emerging 2D materials (including antimonene and bismuthene) is believed to play an important role in the compact ultrafast photonic devices as SAs or gain media. A research trend in heterostructure has been witnessed in this field because stacking different 2D materials can not only provide synthetic benefits but also deliver improved output performances of pulsed lasers. Lastly, although the advances in 2D material-based NLO properties are at a very early stage, small advances can represent significant breakthroughs setting the direction for the future of nonlinear photonics. Current advances may push a large number of related technological devices to come into effect. A recent demonstration shows that parametric oscillations can be excited in a nanocavity incorporating multilayer TMDs as nonlinear media under conventional pump intensities in a phase-matching-free regime. Such novel oscillator-embedded nanomaterials highlight new technologies including environmental detection, biophysics, and spectroscopy [101].

5 Outlook

The above discussion in 2D materials and their practical applications has unambiguously demonstrated the great potential of novel optoelectronics integrated with 2D materials. However, there are still challenges awaiting researchers to address. The research direction in 2D materials and photonic applications will be respectively summarized as follows.

A controllable and foreseeable fabrication process of 2D materials is crucial for device production with better

performances as different synthesis methods lead to the variation of surface and physiochemical features and eventually electronic and optical properties. Furthermore, the categories of 2D materials for integrating into the photonic platform were still limited. Special attention can be paid to the newly discovered 2D candidates including MXenes and metal oxides.

For photodetectors, integration with other photonic structures (e.g. plasmonics, QDs, and Schottky junction) offers another route to improve the responsivity or extend the spectral range of the photodetectors. For modulators, there is still room for the current research to develop toward practical applications. vdW heterostructures assembled by stacking different 2D materials will continue its focal position in the current research community because they bring quantum heterostructures with the tunability in electric and optical features into reality [12]. Moreover, introducing new configurations (e.g. nanoplasmonics integration) and mechanisms (e.g. magneto-optic and acoustic-optic effects) is expected to realize optical modulators equipped with faster response and miniature footprint. In terms of lasers, the improvements in cavity design is one of the most crucial topics for the future study. Last but not least, in the NLO field, it is still difficult to accurately characterize the NLO properties of new 2D materials or the existing ones such as graphene. The large discrepancy in the measured parameters of 2D materials calls for more precise and reliable calculation methods and variable controls.

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