2D Layered Materials: Synthesis, Nonlinear Optical Properties, and Device Applications

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In recent years, 2D layered materials, including graphene, topological insulators, transition metal dichalcogenides, black phosphorus, MXenes, graphitic carbon nitride, and metal-organic frameworks, have attracted considerable interest due to their potential applications in the fields of physics, chemistry, biology, and energy. Their rise in the field of nonlinear photonics began around 2009 and has become an important research direction. Here, the synthesis techniques, nonlinear optical properties, integration strategies, and device applications of layered materials are reviewed. In terms of nonlinear optical properties, the focus is on saturable absorption and Kerr nonlinearity. On this basis, their applications in various pulsed lasers, including fiber lasers, solid-state lasers, waveguide lasers, and related nonlinear optical phenomenon, are summarized. In addition, novel optical devices using layered materials, such as optical modulators, optical polarizers, optical switchers, and even all-optical device, are also involved. It is believed that the development of 2D layered materials in the field of photonics will continue to deepen, thus laying a good foundation for its practical application.

1. Introduction

In recent years, interdisciplinary development has gradually evolved into a distinct feature of scientific research, that is, through the cross-integration of different fields, many frontier directions have emerged.[1] For example, in 2004, A. Geim and K. Novoselov at Manchester university succeeded in stripping a layered material, graphene, which consists of a single carbon atom.[2–4] This discovery, like the first domino, has aroused a worldwide upsurge in graphene research in various fields due to its rich physical properties. Over the past decade, scientists around the world have found a large number of physical phenomenon of graphene and designed related devices, thus making it a good link in different fields.[5–9] More importantly, stimulated by the success of graphene, other layered materials such as topological insulators,[10–12] transition metal dichalcogenides (TMDCs),[13–18] black phosphorus,[19–23] MXenes,[24] graphitic carbon nitride (g-C_3N_4),[25–27] and metal-organic frameworks (MOFs),[28–30] as shown in **Figure 1**, have also been discovered and developed rapidly, thus enriching the family of 2D layered materials.

Nonlinear photonics is one of the most brilliant fields in the application of layered materials.[31–35] This is because, physically speaking, most layered materials have excellent nonlinear optical properties; in terms of practical demand, finding suitable optical materials has always been an eternal theme in the field of nonlinear photonics. Around 2009, graphene has been found to have excellent saturable absorption characteristics and can be used to design pulse-shaping devices.[5,36–40] For example, scientists from two different research teams, Cambridge University and Nanyang University of Technology, demonstrated ultrafast fiber lasers based on graphene saturable absorbers, respectively. In addition, the nonlinear optical property of graphene have been also confirmed by other researchers.[41] These early explorations promoted the rapid development of graphene in the field of ultrafast photonics. Inspired by the study of graphene, researchers have found more layered materials for lasers, and achieved many exciting results, which makes this direction flourish.

Meanwhile, layered materials-based novel optical devices, including optical modulators, optical switchers, optical polarizers, and optical sensors, have also developed rapidly.[42,43] For example, researchers developed a broadband optical polarizer based on the evanescent field interaction between graphene and D-shaped fiber.[44] The wide application of layered materials in these photonic devices has become one of the research hot-spots in recent years, and a large number of important works have emerged, which greatly promotes the development of optical devices. However, there is still a lack of in-depth summary of this aspect.

Here, we review the latest research status of nonlinear photonics based on layered materials, with a particular emphasis on their synthesis techniques, nonlinear optical properties,

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The ORCID identification number(s) for the author(s) of this article can be found under https://doi.org/10.1002/lpor.201800327

DOI: 10.1002/lpor.201800327

integration strategies, and applications in pulsed lasers and other new optical devices. We hope that layered materials will play a key role in future photonics development.

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2. Fundamental of 2D Layered Materials

It is well known that the preparation and physical properties of materials are the basis of their applications. To this end, we will briefly introduce the synthesis techniques, nonlinear optical properties, and integration strategies of layered materials as follows.

2.1. Synthesis Techniques

Because layered materials are one kind of nanomaterials, the common physical and chemical methods for preparing nanomaterials can be used to synthesize 2D layered materials. In general, they can be synthesized by two techniques: top-down exfoliation (mechanical or solution exfoliation) and bottom-up growth (chemical vapor deposition, pulsed laser deposition, or wet chemical method).[45–49] Using these methods, researchers synthesized a variety of layered materials. As we know, each method has its merits and drawbacks. For the application of optical devices, we hope that the synthesized layered materials have large area, uniform thickness, and smooth surface. Next, we focus on several common synthesis methods of layered materials widely used for optical devices.

In 2004, a team from the University of Manchester, A. K. Geim and K. S. Novoselov, prepared monolayer graphene from graphite by tape method.[2] Since then, the study of graphene and other layered materials has become a very hot research topic. More importantly, this simple preparation method of graphene provides a new way for the synthesis of layered materials and has been applied in the early research of related optical devices. Although this method can synthesize high-quality multi-layer or even single-layer nanomaterials, it is not suitable for large-scale use because of its poor scalability and low production efficiency. In addition, the chemical traces left by the tape after stripping are another problem to be solved.

Liquid phase exfoliation is another effective way for preparing high-quality layered materials. For this method, there are several critical stages: oxidation treatment, ion intercalation/exchange, and surface passivation of solvents.[50–52] It is one of the most commonly used layered material preparation methods in photonic device applications. This is because it has many advantages, such as simplicity, efficiency, low cost, high scalability, and standardized production process. In addition, it also has unique advantages in composite materials and film preparation. Notably, from the application point of view, the layered materials prepared by this method can be easily transferred to optical devices, which has been widely developed, and will be described in detail later. However, it is difficult to prepare large-area layered materials, which affects its application in high performance devices.

Physical or chemical deposition is also a kind of the most commonly used methods to synthesize layered materials.[53] Among them, chemical vapor deposition has a long history, and its technology is relatively mature. It is a common bottom-up method to prepare nanomaterials on preset substrates through the reaction of precursors at high temperatures. In addition, pulsed laser deposition has also attracted wide attention. Compared with other methods, layered materials with high crystalline quality, controllable thickness, large area, and smooth surface can be prepared by physical or chemical deposition. In the latter part, we will focus on discussing that the layered materials prepared by this method have good optical properties and show great potential in device applications.

2.2. Third-Order Optical Properties

Third-order optical nonlinearity plays a key role in nonlinear photonic devices.[31,36–40] In this section, we will briefly discuss two important nonlinear properties of layered materials, namely Kerr effect[54–56] and nonlinear absorption,[57–61] and review their recent research history.

Physically, under the action of an external electric field, the total polarization consists of two parts: the linear and nonlinear response, which can be described by

$$\boldsymbol{P} = \varepsilon_0 (\chi^{(1)} \cdot \boldsymbol{E} + \chi^{(2)} : \boldsymbol{E} + \chi^{(3)} \vdots \boldsymbol{E} + \cdots) \tag{1}$$

where ε_0 describes the dielectric constant of vacuum. The first-order susceptibility $\chi^{(1)}$ describes the linear part of layered materials. The second-order susceptibility $\chi^{(2)}$ describes

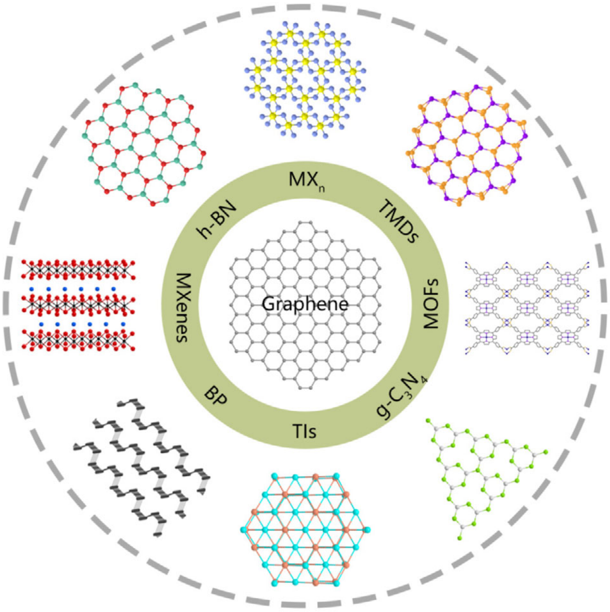


Figure 1. Schematic diagram of typical 2D layered materials.

dual-frequency phenomenon. The third-order susceptibility $\chi^{(3)}$ is a complex number. Its imaginary part describes triple-frequency phenomenon, including nonlinear absorption,[62–66] third-harmonic generation,[67–71] parametric process,[72–74] and Raman effect, while its real part describes the Kerr nonlinearity.

In fact, under the action of laser field, we usually use another way to express the effective refractive index n of layered materials under laser intensity I as follows:

$$n = n_0 + n_2 I \tag{2}$$

where n_0 describes the linear part. The Kerr nonlinearity n_2 depends on the real part of $\chi^{(3)}$. The imaginary part of $\chi^{(3)}$ describes the nonlinear absorption of layered material with the input laser intensity. The saturable absorption and multiple-photon absorption are two kinds of nonlinear absorption. It is worth noting that if the laser intensity is very high, multi-photon absorption becomes very important.[75–79] If two-photon absorption is considered, the saturable absorption formula can be derived from

$$\alpha = \alpha_{\mathrm{ns}} + \frac{\alpha_S}{1 + I/I_{\mathrm{sat}}} + \beta I \tag{3}$$

Among them, β describes the two-photon absorption, α_{ns} describes the unsaturable absorption, that is, intensity-independent loss, α_S describes the saturable absorption, I describes the input laser intensity, and I_{sat} describes the saturable intensity. If the laser intensity is small, the absorption equals $\alpha_{\mathrm{ns}} + \alpha_S$, if the laser intensity is strong, the absorption equals α_{ns} the maximum displacement of absorption α_S.

Moreover, we can also use the figure of merit (FOM) to describe the nonlinear optical effects of layered materials. Specifically, it can be expressed as follows:

$$FOM = \left|\mathrm{Im}\chi^{(3)}\right| / \alpha_0 \tag{4}$$

The larger the value of FOM, the stronger the nonlinear optical effect of layered materials. Therefore, it can help researchers compare the samples used with previous layered materials.

Under the guidance of the above theory, the researchers began to explore experimentally the nonlinear optical properties of layered materials. Z-scan measurement is a common method for studying nonlinear optical processes, including saturable absorption, multi-photon absorption, reverse saturable absorption, and Kerr effect. Using this method, the nonlinear optical coefficients of many layered materials are obtained, as illustrated in **Table 1**.

Surprisingly, experimental work on the third-order nonlinear optical properties of layered materials began around 2009, before the Nobel Prize in Physics for graphene research.[41,80–83] During this period, graphene[84–89] and its derivatives were widely studied as 2D materials, including graphene oxide,[90–93] reduced graphene oxide,[94] and graphene composite materials.[95–98] Several early studies on their nonlinear optical properties are worth mentioning. For example, the researchers used Z-scan technology and pump-probe spectroscopy to study saturable absorption and carrier dynamics in graphene suspensions,[41] as shown in **Figure 2**a–c. In addition, the nonlinear optical properties of graphene oxide under nano/picosecond laser pulses were also explored,[80] as shown in Figure 2d–f. Meanwhile, another group of researchers studied the parametric process of few-layer graphene by four-wave mixing method,[81] as illustrated in Figure 2g–i. They found that graphene exhibits a very large third-order index and is basically non-dispersive in the wavelength range. These early works have greatly promoted the exploration of graphene-based nonlinear optics.[99–104]

Based on the successful exploration of the nonlinear properties of graphene and its derivatives, researchers began to study other 2D materials, such as topological insulators, layered TMDCs, black phosphorus, and MXenes. Among them, topological insulators including Bi_2Se_3, Bi_2Te_3, and Sb_2Te_3, characterized by a robust metallic edge or surface state and a narrow band-gap bulk insulating state, have gained great attention in the field of physics, chemistry, and material.[10–12] Interestingly, around 2012, the researchers found that topological insulator exhibits saturable absorption behavior and can be used to develop pulsed lasers,[105–107] as illustrated in **Figure 3**. Since then, many groups around the world have begun to study the nonlinear optical properties of topological insulators in depth, and obtained many important results.[108–112] These studies show that topological insulators not only have saturable absorption, but also have a very large nonlinear refractive index, suggesting that they have great application potential in pulse-shaping and optical modulation.

Following the topological insulators, layered TMDCs are another type of 2D materials which has been widely studied.[13,14] TMDCs can be expressed with the formula MX_2 (M = Mo, W, Ta, V, Nb, Re, Ti, etc; X = S, Se, Te). Due to the specific 2D confinement of electron motion and the absence of interlayer coupling, layered TMDCs possess a direct bandgap, making its nonlinear optical performance dramatically better than that of its bulk counterpart.[113] As early as 2013, researchers discovered saturable absorption properties of few-layer MoS_2, which caused a sensation in the field of nonlinear photonics.[114] For example, the researchers revealed the saturable absorption

Table 1. Summary of nonlinear optical properties of layered materials.

2D Mater.	Laser parameters	NLO response	T [%]	I_S [GW cm^{-2}]	α_0 [cm^{-1}]	α_{NL} [cm GW^{-1}]	Im $\chi^{(3)}$ [esu]	Ref.
GO	532 nm, 1 Hz, 25 ns	SA	–	1.5	426.55	1.44	–	[91]
rGO	532 nm, 1 Hz, 25 ns	SA	–	2	880.67	2.67	–	[91]
graphene	800 nm, 1 kHz, 100fs	SA	16.8	583 ± 127	17.85	$-(1.52 \pm 0.4) \times 10^{-2}$	$-(8.7 \pm 2.4) \times 10^{-15}$	[115]
	1030 nm, 100 Hz, 340fs		48	88.9 ± 27.3	53.6	-0.66 ± 0.3	$-(5 \pm 2.3) \times 10^{-13}$	
	1064 nm, 10 Hz, 6 ns		44.87	3.9 ± 0.83	58.5	-15.8 ± 3.8	$-(1.23 \pm 0.3) \times 10^{-11}$	
Bi_2Te_3	1550 nm, 35 fs	–	–	–	–	–	0.2×10^{-17} (n_2)	[107]
Bi_2Se_3	800 nm, 1 kHz, 25 ns	SA	46.7	1199	–	–	0.0097 (n_2)	[106]
	1562 nm, 20.8 MHz, 1.5 ps		30.55	1.26×10^{-3}			0.86 (n_2)	
	1930 nm, 32.3 MHz, 2.8 ps		26.71	7.09×10^{-3}			2.12 (n_2)	
WSe_2	1064 nm, 20 Hz, 25ps	2PA	–	–	–	1.9 ± 0.57	$-(6.35 \pm 1.35) \times 10^{-12}$	[123]
MoS_2	800 nm, 1 kHz, 100fs	SA	32.6	381 ± 346	11.22	$-(2.42 \pm 0.8) \times 10^{-2}$	$-(1.38 \pm 0.45) \times 10^{-14}$	[115]
$MoSe_2$	800 nm, 1 kHz, 100fs	SA	45.3	590 ± 225	7.93	$-(2.54 \pm 0.6) \times 10^{-2}$	$-(1.45 \pm 0.34) \times 10^{-15}$	[115]
$MoTe_2$	800 nm, 1 kHz, 40fs	SA	86.3	217 ± 11	1.47	$-(3.7 \pm 1.2) \times 10^{-2}$	$-(2.13 \pm 0.66) \times 10^{-15}$	[115]
WS_2	800 nm, 1 kHz, 40fs	SA	35.75	156	7.22×10^5	-397 ± 40	$-(1.78 \pm 0.16) \times 10^{-9}$	[75]
BP	800 nm, 10 kHz	SA + TPA	42.1	–	8.7	-1.38×10^{-2}	-7.85×10^{-15}	[135]
	1330 nm, 50 kHz, fs		45.9 ± 2.2		7.8 ± 0.5	$-(1.9 \pm 0.3) \times 10^{-2}$	$-(1.83 \pm 0.29) \times 10^{-14}$	
	1420 nm, 50 kHz, fs		55.5 ± 0.3		5.9 ± 0.1	$-(1.5 \pm 0.9) \times 10^{-2}$	$-(1.05 \pm 0.22) \times 10^{-14}$	
	1550 nm, 50 kHz, fs		59.1 ± 0.1		5.3 ± 0.1	$-(1.8 \pm 0.9) \times 10^{-2}$	$-(1.98 \pm 0.95) \times 10^{-14}$	
	1972 nm, 50 kHz, fs		55.3 ± 1.5		5.9 ± 0.3	$-(10 \pm 2.8) \times 10^{-2}$	$-(1.41 \pm 0.4) \times 10^{-13}$	
	2100 nm, 50 kHz, fs		60.6 ± 0.5		5 ± 0.1	$-(5.7 \pm 1.4) \times 10^{-2}$	$-(8.49 \pm 2.1) \times 10^{-14}$	
Si	1540 nm, 220 fs	–	–	–	–	–	0.45×10^{-17} (n_2)	[106]

mechanism of MoS_2 dispersions, as shown in **Figure 4**a–c. It is found that they exhibit better saturable absorption than that of graphene under the same conditions.[115–119] In addition, the two-photon absorption response of WS_2 and WSe_2 films was also discovered,[76] as shown in Figure 4d–f. Recently, more TMDCs including MS_2,[120–125] MSe_2,[126–129] MTe_2,[130,131] have been also deeply studied. These explorations lay a foundation for their applications in nonlinear photonics.

Another 2D material that should be mentioned is black phosphorus. It is found that black phosphorus has a singular direct band-gap, which varies between 2 eV (single layer) and 0.3 eV (bulk) with the number of layers and lies between zero-bandgap graphene and relatively large-bandgap TMDCs, thus making up for the shortcomings of these 2D materials in the field of photonics.[19–21] In 2015, the researchers discovered the broadband nonlinear optical response of multilayer black phosphorus,[132] as shown in **Figure 5**. Subsequently, this optical property was confirmed by other research groups.[133–136] Thus, black phosphorus has huge application potential in broadband laser and passive optical devices, especially in near infrared and mid-infrared photonic systems.[137–140]

In the above studies, an important issue is the formation mechanism of saturable absorption of layered materials. It is found that most layered materials, such as graphene, topological insulators, and black phosphorus, have an energy-band structure of symmetry Dirac-cone type. Physically, any electron can be excited into the conduction band when the intensity of incident light is larger than the bandgap of layered materials. Then, the distribution rapidly thermalizes and cools down to form a hot Fermi–Dirac distribution. Through a dynamic process, electrons and holes recombine until the equilibrium distribution is restored. This describes the linear optical transition under low excitation intensity. However, as the light intensity increases to a higher level, the photocarriers increase instantaneously and fill the energy states near the edge of conduction and valence band, the absorption is blocked due to the Pauli-blocking principle. Eventually, the photons at specific wavelength can transparently transmit the layered materials without absorption. This mechanism plays a key role in pulse-shaping and has been widely used in pulsed lasers. In fact, whether a material has saturable absorption at specific wavelength is a precondition for its application in pulse-shaping devices. In the latter part, we will find that the saturable absorbers in passively mode-locked/Q-switched lasers take advantage of this mechanism.

As we know, for different materials, there are obvious differences in their band-gap. Interestingly, besides conventional saturable absorption, sub-bandgap absorption is also a common phenomenon in device applications, which may be attributed to material defects, two-photon absorption, or edge-mode absorption.[141] In addition, unlike graphene, TMDCs and black phosphorus have band-gaps that vary with the number of layers, and their absorption mechanism is more complex. These results indicate that our understanding of saturable absorption of layered materials is only the tip of the iceberg and needs further study.

Meanwhile, other optical properties, including third-harmonic generation,[142–147] parametric process,[148–151] and stimulated Brillouin scattering,[152,153] have been also explored, which promotes the development of layered materials-based nonlinear optics.

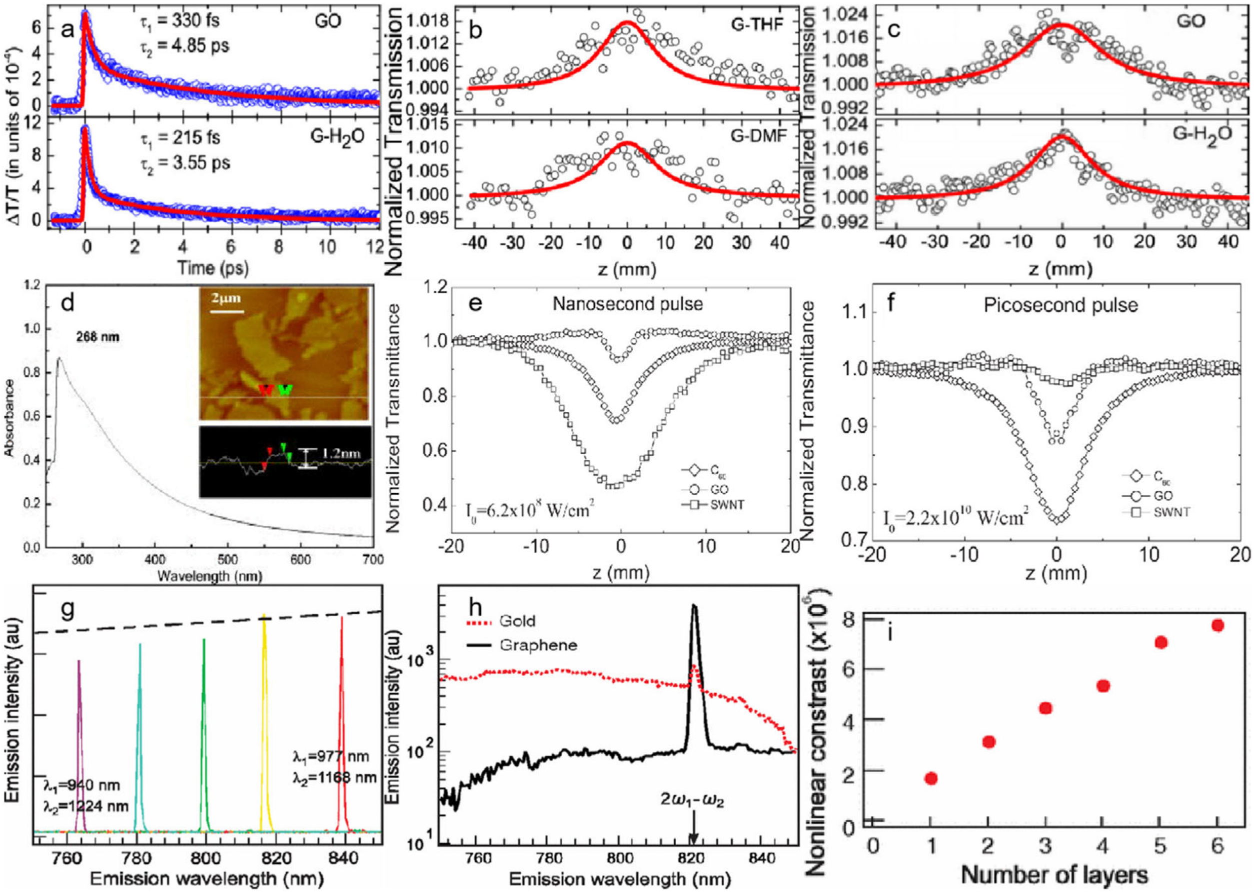


Figure 2. Nonlinear optical properties of graphene and graphene oxide. a) Transient differential transmission spectra of graphene nanosheets in pump-probe experiment. b,c) Z-scan curves. d) Ultraviolet absorption of graphene oxide in *N,N*-dimethyl formamide. The inset: AFM images of graphene oxide nanosheets. e,f) Z-scan curves of various materials for different laser pulses. g) Optical spectra of graphene stimulated under different pump pulses. h) Optical spectra of graphene and gold film stimulated under the laser beam (969 nm, 1179 nm). i) The relation between the nonlinear constrast and the number of graphene layers. Reproduced with permission.[41] Copyright 2009, AIP Publishing. Reproduced with permission.[80] Copyright 2009, AIP Publishing. Reproduced with permission.[81] Copyright 2010, APS Publishing.

2.3. Integration Strategies for Pulse-Shaping Devices

After layered materials are synthesized, how to integrate them into waveguide devices to form pulse-shaping devices or optical modulators is a key issue affecting their subsequent applications. For optical fiber devices, several mature technologies have been proposed and developed,[154] as shown in **Figure 6**. For example, the researchers can sandwich them between two optical connectors (Figure 6a). In this scheme, layered materials are usually transferred to the end surface of one of the connectors by optical deposition or polymer film. In addition, in-fiber microfluidic channel is also proposed (Figure 6b). Another alternative is integrate layered materials into microstructured optical fibers, such as photonic crystal fibers (Figure 6c), D-shaped (Figure 6d), and tapered fibers (Figure 6e), has also been widely developed. Among these schemes, sandwich devices have simple structure, low-cost but having short-range interaction length; photonic crystal fibers filled with layered materials have strong interaction between the laser and layered materials but larger insertion loss and distortion of regional guidance mode; the tapered or D-shaped fibers coated with layered materials have stronger strength tolerance and longer interaction length, but uneasy to achieve flat material thickness. Recently, the fully integrated monolithic fiber laser (Figure 6f) has become a research hotspot. It can be predicted that with the development of micro-/nano-fabrication technology, its performance will be greatly improved. For space transmission or solid-state laser applications, layered materials are generally transferred to optical lenses using spin-coating or direct-dropping to form saturable absorption mirrors.

3. Versatile Pulsed Lasers using 2D Layered Materials

According to the operation state, there are two types of lasers: continuous wave and pulse. In order to turn the continuous-wave into a train of pulses, a nonlinear optical element, called saturable absorber (SA), is usually needed in the pulsed lasers.[5,155] As described in Section 2.2, 2D layered materials have excellent saturable absorption property and can be used as SAs in the lasers to obtain mode-locked/Q-switched pulses. Since 2009, versatile

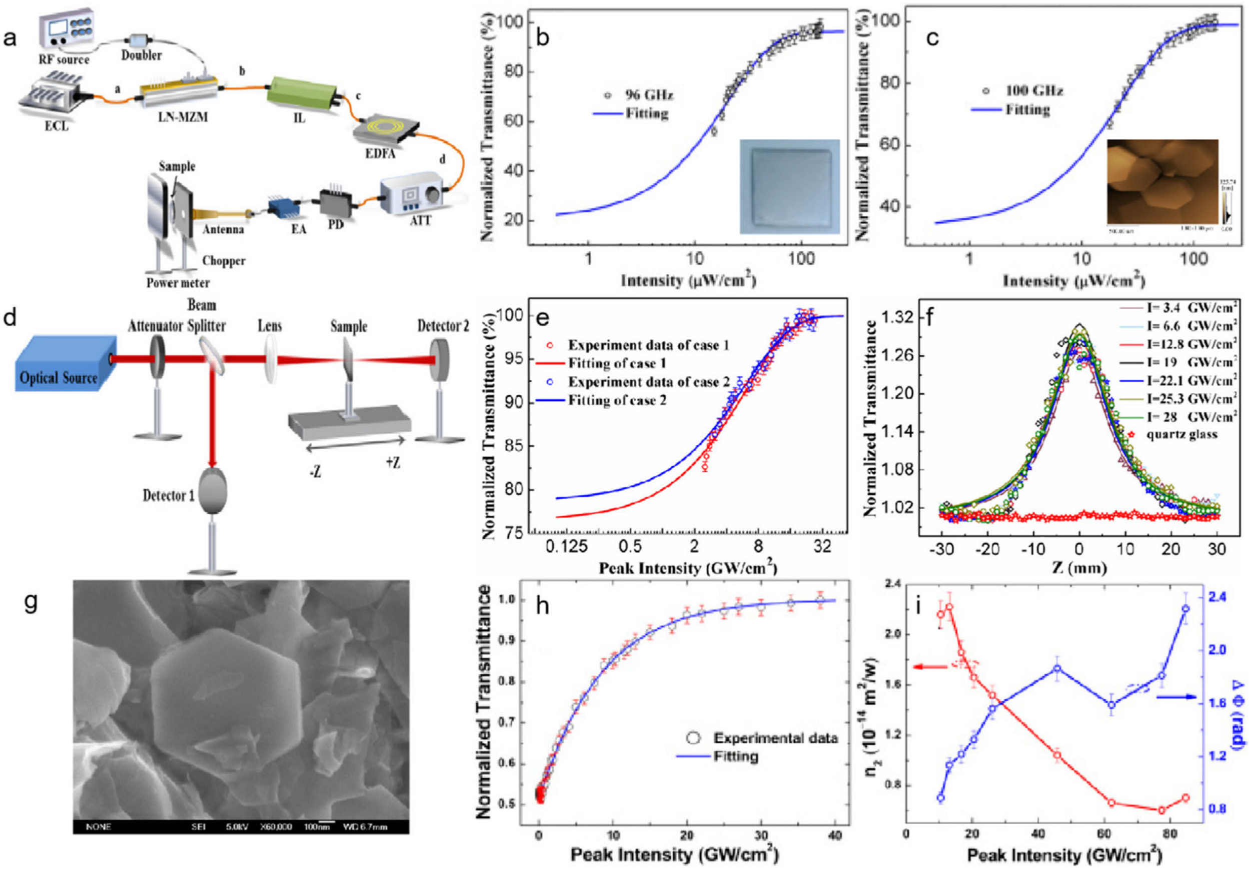


Figure 3. Nonlinear optical properties of topological insulators. a) Schematic of the microwave generation and saturable absorption measurement. b,c) Saturable absorption of Bi_2Te_3 at different microwave frequencies. d) Schematic of the Z-scan experimental setup. e) Relation between normalized transmittance and input peak intensity. f) Open Z-scan curve at 800 nm. Reproduced with permission. [107] Copyright 2014, Optical Society of America. g) SEM image of Bi_2Se_3. h) Relation between normalized transmittance and input peak intensity. i) Dependence of nonlinear phase ($\Delta\Phi$) and Kerr refractive index (n_2) on peak intensity. Reproduced with permission.[106] Copyright 2013, Optical Society of America.

pulsed lasers (fiber, solid-state, disk, and waveguide) based on layered materials have been developed rapidly.[156–160] In this section, we will review the application of layered materials in these pulsed lasers and look forward to its future development trend.

3.1. Fiber Lasers

Fiber laser is a very important kind of laser, which has attracted much attention because of its miniaturization, compact structure, no adjustment, and high reliability. In terms of performance, its ultrashort pulse-width and ultrahigh peak power operation have always been the goal of researchers in the field of laser.[161–163] In order to obtain these pulses, two pulse-shaping mechanisms—mode-locking and Q-switching—are mostly used. As mentioned earlier, in either case, an SA is needed as a pulse-shaping device. Over the past decade, SAs made of layered materials have been widely used in passively mode-locked/Q-switched lasers, and a lot of important results have been achieved.[164–166] In this section, we will briefly review the research history and recent achievements of these lasers and related nonlinear optical phenomenon.

3.1.1. Mode-Locking Operation

Graphene is the first 2D material used for mode-locked lasers. In 2009, the researchers from Cambridge University and Nanyang University of Technology found that graphene exhibits saturable absorption property and can be used as a pulse-shaping device.[36–38,167,168] In their experiment, they demonstrated the soliton mode-locked fiber lasers based on graphene SA, respectively, as shown in **Figure 7**. These early studies show that graphene is an excellent pulse-shaping device with wide operation range and fast response time due to its zero band-gap and ultrafast carrier dynamics. In addition, compared with traditional SAs, including semiconductor saturable absorber mirrors or carbon nanotubes, graphene does not require bandgap optimization and diameter or chiral adjustment, thus greatly simplifying the preparation process. With these advantages, graphene and its derivatives, such as graphene oxide, reduced graphene oxide, and graphene composite materials, have been widely developed in mode-locked fiber lasers,[168–235] as shown in **Table 2**. In terms of the performance of these mode-locked lasers, some exciting results have been obtained, including minimum pulse-width, maximum output power, and repetition rates of 29 fs,[211] 520 mW,[226] and 162 GHz,[232] respectively. In the aspect of graphene

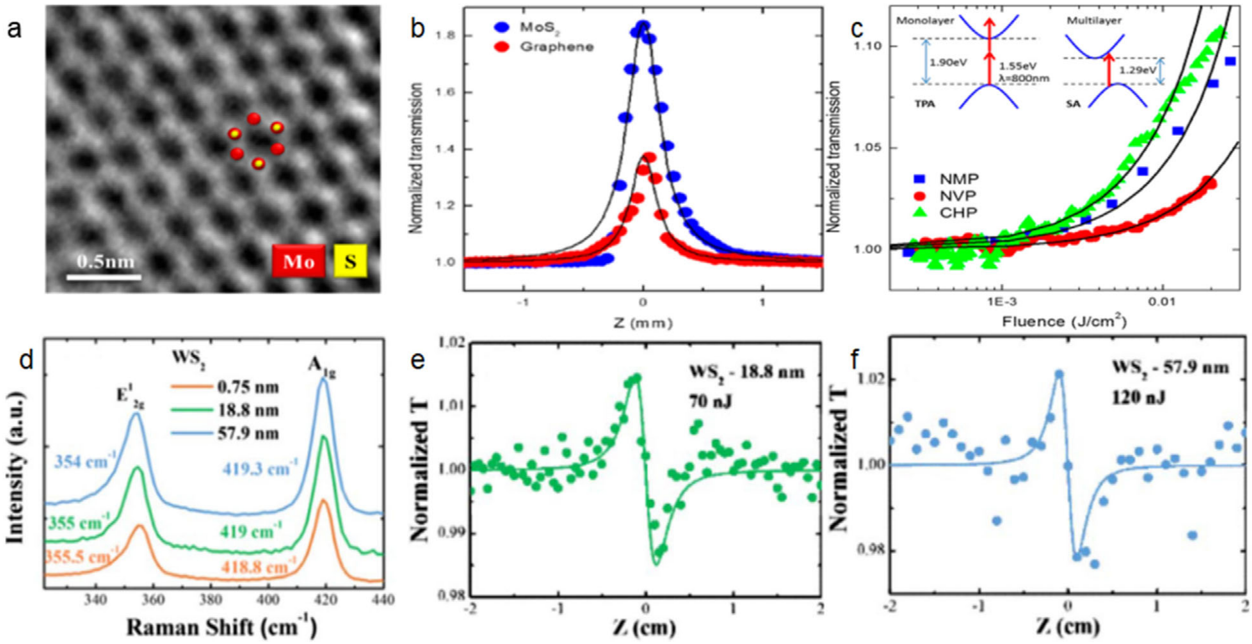


Figure 4. Nonlinear optical properties of few-layer MoS$_2$, WS$_2$. a) High-resolution TEM image of MoS$_2$ nanosheets. b) Open-aperture Z-scan curves of few-layer MoS$_2$ and graphene under the pump pulse (800 nm, 100 fs). c) Relation between the transmission and the energy density for different materials. d) Raman spectrum of WS$_2$ film. e,f) Z-scan curves of WS$_2$ films. Reproduced with permission.[114] Copyright 2013, ACS Publishing. Reproduced with permission.[76] Copyright 2016, Optical Society of America.

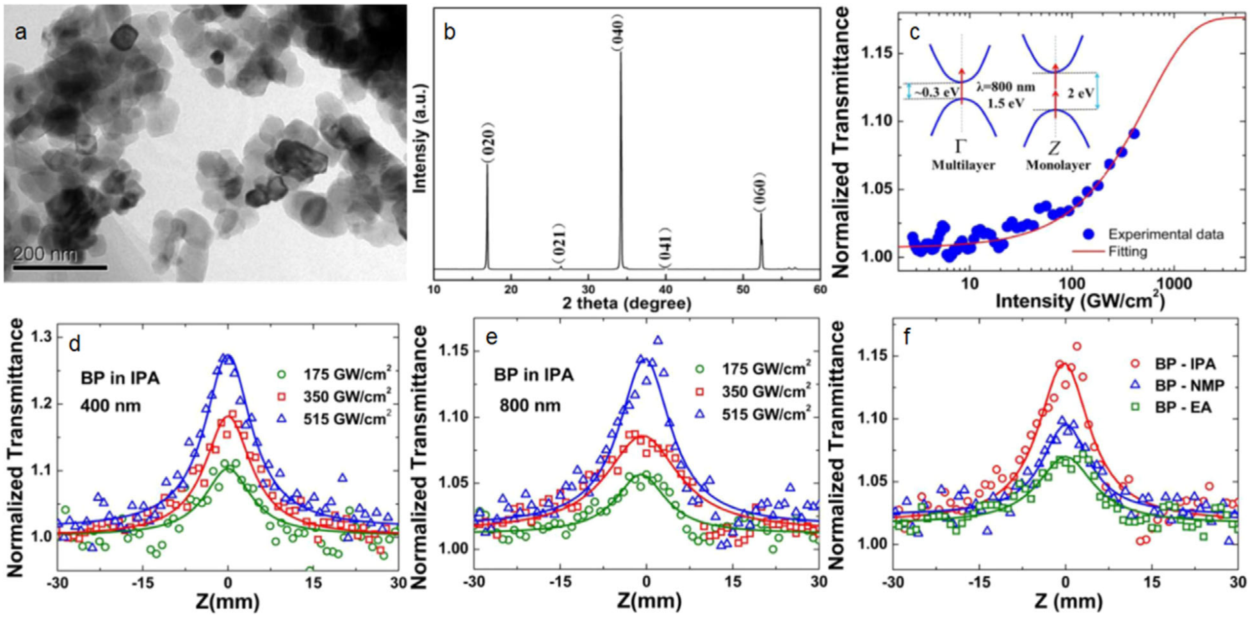


Figure 5. Nonlinear optical properties of few-layer black phosphorus. a) TEM image of black phosphorus nanosheets, b) XRD of black phosphorus powder, c) Normalized transmittance as a function of input intensity for black phosphorus nanosheets, d–f) Z-scan curves of few-layer black phosphorus at different wavelengths and solvents, respectively. Reproduced with permission.[132] Copyright 2015, Optical Society of America.

integration, besides sandwiched structure, the combination with microstructured optical fibers (tapered fibers, side-polished fibers, or photonic crystal fibers) is also an effective method to fabricate graphene mode-locker. These efforts not only deepen our understanding of graphene, but also promote the development of mode-locked lasers. Nevertheless, graphene also shows some weaknesses in laser applications. For example, it is found that graphene has no bandgap and its optical modulation depth is very weak (≈2.3%/layer), which limits its application in flexible tunable operation and in situations requiring strong

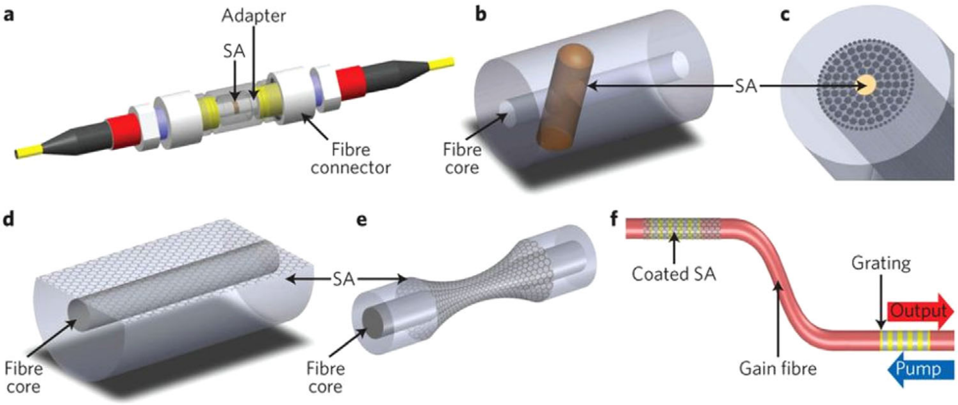


Figure 6. Various layered materials-SA integration strategies for fiber devices. a) Sandwiched device. b) In-fiber microfluidic channels, c) Photonic-crystal fibers, d) D-shaped fibers, e) Tapered fibers, and f) Fully integrated monolithic fiber laser. Reproduced with permission.[154] Copyright 2013, Springer Nature.

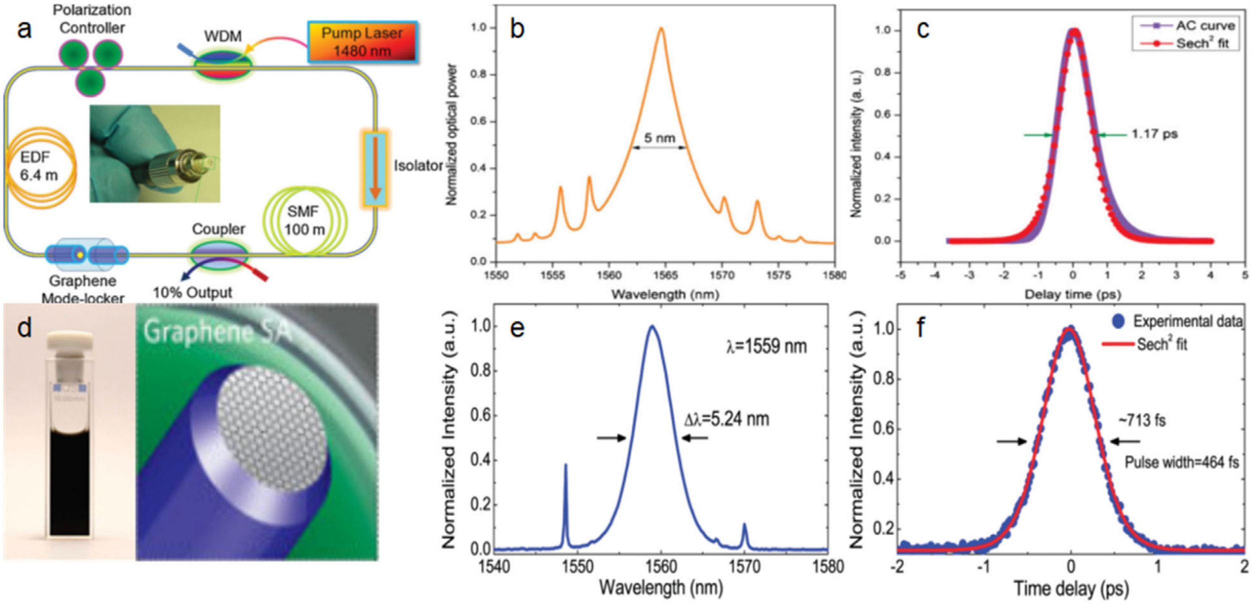


Figure 7. Mode-locked fiber lasers with graphene SA and its performance. a) Experimental setup. Inset: Photographs of mode-locker. b,e) Optical spectra. c,f) AC traces of mode-locked pulses. d) The photograph of graphene sample and mode-locker. Reproduced with permission.[36] Copyright 2009, Wiley Publishing. Reproduced with permission.[38] Copyright 2010, American Chemical Society.

laser-material interaction.[42–44] Thus, it is still a long-term goal for researchers to explore new SAs.

The successful application of graphene in mode-locked fiber lasers has greatly inspired researchers to explore other layered materials. Like graphene, topological insulators have an energy-band structure of symmetry Dirac cone due to their strong spin-orbit interaction, which implies that they can be developed into a new kind of SAs. Inspired by this, Bernard and his collaborators experimentally discovered the saturable absorption behavior of topological insulators in 2012.[105] Subsequently, the researchers demonstrated a soliton mode-locked fiber laser based on the topological insulator SA,[236] as shown in **Figure 8**a–c. It is found that, unlike graphene, topological insulators have a non-zero bandgap and large modulation depth (up to 95%), which are more conducive to the development of mode-locked lasers. In view of these advantages, various mode-locked fiber lasers based on topological insulators including Bi_2Se_3,[237,239–242] Bi_2Te_3,[238,243–251] Sb_2Te_3,[252–256] have been developed, as shown in Table 2. Through these efforts, some exciting results such as maximum repetition rate, maximum output power, and minimum pulse width of 3.125 GHz,[241] 45.3 mW,[243] and 195 fs[255] have been obtained, respectively. These results indicate that, besides electrical and thermal properties, topological insulators may also have attractive application prospects for nonlinear photonics.

Almost simultaneously, the application of layered TMDCs and their derivatives in mode-locked lasers has also attracted wide attention of researchers in the field of ultrafast photonics. It is found that TMDCs are a very large material system,[25] which

Table 2. Summary of mode-locked fiber lasers with layered materials.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	$f_{\rm rep}$/MHz	τ/ps	$P_{\rm ave}$/mW	E/nJ	Ref.
graphene	Sandwiched	EDF	1565	1.79	0.756	2	–	[36]
graphene	Sandwiched	EDF	1589.68	6.95	0.694	13.1dBm	3	[37]
graphene	Sandwiched	EDF	1559	19.9	0.463	–	–	[38]
graphene	Sandwiched	EDF	1576.3	6.84	0.415	–	7.3	[167]
graphene	Sandwiched	EDF	1570–1600	6.95	1.08	–	–	[168]
graphene	Deposited into SPF	EDF	1561.6	6.99	1.3	21.4 dBm	7.25	[169]
graphene	Sandwiched	EDF	1555	27.4	0.174	1.2	0.044	[170]
graphene	Sandwiched	EDF	1576	10.9	3.2	4.8 dBm	–	[171]
graphene	Sandwiched	EDF	1525–1559	8	1	1	0.125	[172]
graphene	Sandwiched	EDF	1532	5.27	0.85	–	–	[173]
graphene	Sandwiched	EDF	1572.6	91.5	–	–	–	[174]
graphene	Sandwiched	EDF	1566	6.22	0.88	–	–	[175]
graphene	Sandwiched	EDF	1565	42.8	0.19	0.4	0.09	[176]
GO	Filled into HC-PCF	EDF	1561.2	7.68	4850	4.3	0.56	[177]
graphene	Monolayer	EDF	1561	5.5	1.23	3	20	[178]
graphene	Sandwiched	EDF	1560.5	2.22 GHz	0.9	9.6	–	[179]
graphene	Deposited into back mirror of F-P cavity	EDF	1562	9.67 GHz	0.865	–	–	[180]
graphene	Sandwiched	EDF	1557	114.1	0.57	–	–	[181]
graphene	Deposited into microfiber	EDF	1545.5–1550	27	14	–	–	[182]
graphene	21-layer	EDF	1559.12	25.67	0.43247	–	0.09	[183]
graphene	Filled into HOF	EDF	1555	506.9	0.51	80	5.2	[184]
GO	Deposited on fused silica windows	EDF	1558	58	0.39	92	0.0337	[185]
			1559	58	0.39	82		
rGO	SAM	EDF	1556.9	25.6	0.6	3.3	0.13	[186]
			1560	22.9	0.2	5.8		
graphene	Sandwiched	EDF	1564	57.96	0.315	1.9	0.033	[187]
rGO	Deposited into microfiber	EDF	1560	7.47	18	1.2	–	[188]
graphene	Sandwiched	EDF	1550	332.5	600	–	–	[189]
graphene	Sandwiched	EDF	1560	490	0.97	–	–	[190]
graphene	Sandwiched	EDF	1556–1560	46.126	0.57	–	0.0228	[191]
graphene	Deposited into microfiber	EDF	1557.56	3.33	15.7	4.2	1.26	[192]
graphene	Sandwiched	YDF	1180	0.4	200 ns	–	–	[193]
graphene	Filled into PCF	EDF	1567.6	25	0.65	–	–	[194]
graphene	Deposited into SPF	EDF	1560	14.64	0.78	–	–	[195]
graphene	Sandwiched	EDF	1562	–	–	–	–	[196]
graphene	Sandwiched	EDF	1560	54.9	0.81	1	20	[197]
graphene	Deposited into microfiber	EDF	1559	312.5	0.679	–	–	[198]
graphene	Sandwiched	Zr-EDF	1551–1570	10.9	0.73-0.78	1.4	0.128	[199]
graphene	Deposited into microfiber	EDF	1555	–	0.765	–	–	[200]
graphene	Super-capacitor	YDF	1255	4.54	0.084	–	–	[201]
graphene	Sandwiched	EDF	1561.4	9.9	1	10.5	1	[202]
graphene	Sandwiched	YDF	1035	16.29	6500	–	0.81	[203]
		EDF	1564	19.3	0.87		0.0104	
		THDF	1908	1.82	65		16.2	

(*Continued*)

Table 2. Continued.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	f_{rep}/MHz	τ/ps	P_{ave}/mW	E/nJ	Ref.
graphene	Sandwiched	Zr-EDF	1563	69.3	0.73	1.6	0.0231	[204]
GO	Sandwiched	YDF EDF	1029.5 1560	–	190 0.7505	–	–	[205]
graphene	Deposited into microfiber	EDF	1565	33	0.494	–	–	[206]
graphene	Sandwiched	Er:Yb-DCF	1610	5.882 GHz	2.9	–	–	[207]
graphene	Deposited into microfiber	EDF	1550	22.34–111.7	2.32–9.24	3	1.34	[208]
GO	SAM	EDF	1555.92	48.2	0.502	2.7	–	[209]
graphene	Gate-controlled	EDF	1609	30.9	0.423	3.1	–	[210]
graphene	Sandwiched	EDF	1550	18.67	0.029	52	2.8	[211]
graphene	Sandwiched	EDF	1560	28.5	1.027	6.9	0.24	[212]
GO	Sandwiched	EDF TDF	1565.9 1961.6	37.2 27.37	0.613 1.36	0.83	0.0223	[213]
graphene	Deposited into SPF	EDF	1558.2	4.77	1.07	21.2 dBm	3.08	[214]
GO	Sandwiched	EDF	1561.8	62.2	0.735	0.82	–	[215]
graphene	Deposited into microfiber	EDF	1559.74/1560.54	100 GHz	1.63	–	–	[216]
graphene	Deposited into microfiber	EDF	1550	–	3.5	–	–	[217]
graphene	Sandwiched	EDF	1557	48.14	0.216	1.3	–	[218]
graphene	Sandwiched	EDF	1545	21.15	0.088	1.5	0.071	[219]
graphene	Deposited into SPF	EDF	1607.7	37.7	0.377	–	–	[220]
graphene	Deposited into tapered fiber with inner air-cavity	EDF	1557	8.655	0.674	6.77 dBm	0.55	[221]
GO	Sandwiched	EDF	1559	40.15	0.9538	0.299	–	[222]
graphene	Deposited into SPF	EDF	1557	12.29	0.256	–	–	[223]
graphene	Deposited into microfiber	EDF	1531.3	1.89	1.21	0.45	–	[224]
rGO	Filled into PCF	EY-DCF	1560	222.9	0.791	1.14	5.1	[225]
graphene	Deposited into microfiber	EY-DCF	1559	366	7	520	–	[226]
graphene	Sandwiched	EDF	1560	25.8	0.364	15	2	[227]
graphene	Sandwiched	EDF	1574	–	0.33	–	–	[228]
GO	Sandwiched device	EDF	1559.6	22.7	1.5	1	–	[229]
graphene	Sandwiched	EDF	1560	8.22	1.12	–	–	[230]
graphene	Deposited into SPF	EDF	1563	11.53	0.713	–	–	[231]
graphene	Microfiber knot	YDF EDF	1044.8 1550	162 GHz 106.7 GHz	6.17 9.37	–	–	[232]
graphene	Fiber device	EDF	1560	–	–	–	–	[233]
graphene	Coated to micro-CFBG	EDF	1550.238 1551.026	17.8	30.71 38.36	–	–	[234]
graphene	Coated to micro-CFBG	EDF	1550.196	12	–	–	–	[235]
Bi_2Se_3	SAM	EDF	1557–1565	1.21	1.57	–	–	[237]
Bi_2Se_3	Sandwiched	EDF	1557.5	12.5	0.66	1.8	–	[239]
Bi_2Se_3	Sandwiched	YDF	1040	16	380	17.1	1.06	[240]
Bi_2Se_3	Deposited into microfiber	EDF	1560.88	3.125 GHz	1.754	6.4	4.5 pJ	[241]
Bi_2Te_3	SAM	EDF	1554–1564	1.21	1.21	–	–	[236]

(Continued)

Table 2. Continued.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	f_{rep}/MHz	τ/ps	P_{ave}/mW	E/nJ	Ref.
Bi_2Te_3	Deposited into microfiber	EDF	1558.5	2.04 GHz	2.49	5.02	–	[238]
Bi_2Te_3	Deposited into microfiber	EDF	1564.1	2.95 GHz	0.92	45.3	–	[243]
Bi_2Te_3	Deposited into SPF	EDF	1547	15.11	0.543	–	–	[245]
Bi_2Te_3	Deposited into SPF	EDF	1555.9	773.85	0.63	1.4	–	[246]
Bi_2Te_3	Filled into HC-PCF	YDF	1065.4	28.73	575.8	–	–	[247]
Bi_2Te_3	Sandwiched	EDF	1557	8.635	1.08	0.25	–	[249]
Sb_2Te_3	Sandwiched	EDF	1558.6	4.75	1.8	0.5	–	[252]
Sb_2Te_3	Sandwiched	EDF	1558.2	304	2.2	4.5	–	[253]
Sb_2Te_3	Deposited into microfiber	EDF	1561	34.5	0.27	1	–	[254]
Sb_2Te_3	Deposited into SPF	EDF	1568.8	33.07	0.195	9	–	[255]
Sb_2Te_3	Deposited into SPF	YDF	1036.7	19.28	5.3	4	–	[256]
MoS_2	Sandwiched	YDF	1054.3	6.58	800	9.3	–	[257]
MoS_2	Sandwiched	EDF	1568.9	8.288	1.28	5.1	–	[258]
MoS_2	Deposited into microfiber	YDF	1042.6	6.74	656	2.37	–	[259]
MoS_2	Deposited into microfiber	EDF	1558	2.5 GHz	3	5.39	–	[260]
MoS_2	Deposited into SPF	EDF	1560	14.53	0.2	3	–	[261]
MoS_2	Sandwiched	EDF	1569.5	12.09	0.71	1.78	–	[262]
MoS_2	Sandwiched	YDF	1029.78	22.44	13.8	34.6	1.54	[265]
WS_2	Deposited into microfiber	EDF	1561	24.93	0.369	1.93	–	[270]
WS_2	Deposited into microfiber	EDF	1558.5	19.58	0.675	0.625	–	[269]
WS_2	Deposited into microfiber	EDF	1540	135	0.067	–	–	[273]
WS_2	Deposited into microfiber	EDF	1557	8.86	1.32	110	–	[266]
WS_2	Filled into SM-PCF	EDF	1563.8	19.57	0.808	2.64	0.1336	[267]
WS_2	Deposited into microfiber	EDF	1557	460.7	0.66	6.23	–	[268]
WSe_2	Deposited into microfiber	EDF TDF	1556.42 1886.22	14.02 11.36	0.477 1.18	–	–	[282]
ReS_2	Sandwiched	EDF	1558	5.48	1.6	0.4	–	[288]
$MoTe_2$ WTe_2	Sandwiched Deposited into SPF	EDF	1561 1555	5.26 5.34	1.2	–	–	[279]
$MoTe_2$	Deposited into microfiber	EDF TDF	1559.57 1934.85	26.6 15.37	0.229 1.3	57 212	2.14 13.8	[280]
WTe_2	Deposited into SPF	EDF	1556.2	13.98	0.77	0.04	–	[281]
WS_2	Sandwiched	EDF	1568.3	0.487	1.49	62.5	–	[275]
SnS_2	Sandwiched	YDF	1062.66	39.33	656	2.23	–	[285]
$MoSe_2$	Sandwiched	EDF	1558.25	8.028	1.45	0.4	–	[277]
$MoSe_2$	Deposited into SPF	EDF	1557.3	3.27 GHz	0.688	22.8	–	[278]
ReS_2	Deposited into microfiber	EDF	1563.3	1.78	3.8	–	–	[287]
$ReS_{2(1-x)}Se_{2x}$	Sandwiched	EDF	1561.15	2.95	0.888	–	0.275	[289]
In_2Se_3	Deposited into microfiber	EDF TDF	1565 1932	40.9 15.8	0.276 1.02	83.2 112.4	2.03 7.1	[290]

(*Continued*)

Table 2. Continued.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	$f_{\rm rep}$/MHz	τ/ps	$P_{\rm ave}$/mW	E/nJ	Ref.
In_2Se_3	Sandwiched	EDF	1532.5	0.5863	389.2 ns	11.96	20.4	[291]
InS	Sandwiched	YDF	1033.3 1038.4	1.02	486.7	1.91	–	[292]
TiS_2	Sandwiched	EDF	1569.5	5.34	1.04	–	5.05	[293]
TiS_2	Deposited into microfiber	EDF	1563.3	22.7	0.812	–	25.3 pJ	[294]
PtS_2	Sandwiched	EDF YDF	1572 1072	15.04 11.2	2.1	1.1	–	[295]
$PtSe_2$	Deposited into microfiber	EDF	1563	23.3	1.02	–	0.53	[296]
$PtSe_2$	Deposited into SPF	EDF	1567.07	8.24	0.861	–	–	[297]
$PtSe_2$	CVD	Nd:YAG	1064	8.82 GHz	27	–	–	[298]
HfS_2	Deposited into microfiber	EDF	1561.8	21.45	0.2217	89.4	4.17	[299]
BP	Sandwiched	EDF	1571.45	5.96	0.946	–	–	[300]
BP	Deposited into microfiber	EDF	1532–1570	4.69	0.94	5.6	–	[301]
BP	Sandwiched	EDF	1545–1579	60.5	0.28	–	–	[302]
BP	Sandwiched	EDF	1558.7	0.786	14.7	–	–	[303]
BP	Sandwiched	EDF	1560.5	28.2	0.242	0.5	–	[304]
BP	Sandwiched	EDF	1562	12.5	0.635	–	–	[305]
BP	Sandwiched	YDF	1085.5	13.5	7.54	80	–	[306]
BP	Sandwiched	EDF	1555	23.9	0.102	1.7	0.071	[307]
BP	Deposited into microfiber	YDF EDF	1064.4 1576.1	16.77 34.27	51 0.4037	18.9 1.9	1.13 0.055	[308]
BP	Sandwiched	EDF	1562	5.426	1.236	–	–	[309]
graphene	Sandwiched	EDF	1570–1600	1.5	49	–	2.3	[310]
graphene	Sandwiched	EDF	1570–1600	–	70-150	–	–	[311]
graphene +SWCNT	Sandwiched	EDF	1560	7.05	9.15	–	–	[312]
graphene	Deposited into SPF	EDF	1565	16.99	13.8	174	10.2	[313]
graphene	Sandwiched	YDF	1069.8	0.9	580	0.37	0.41	[314]
rGO	Deposited into microfiber	EDF	1564.6/1567.4	7.9	–	8	–	[315]
GO	Sandwiched	YDF	1064.9	2.99	520–60800	147.8	159.4	[316]
GO	SAM	EDF	1531	19.5	11	23.3	1.2	[317]
GO	Sandwiched	YDF	1029	–	191–1680	539	–	[318]
Bi_2Se_3	Sandwiched	YDF	1031.7	44.6	47	–	0.756	[319]
Bi_2Te_3	Sandwiched	EDF	1548–1570	10.71	4.5	–	2.8	[320]
Sb_2Te_3	Deposited into SPF	EDF	1565	22.32	0.128	–	0.0448	[321]
Sb_2Te_3	Deposited into SPF	EDF	1558	25.38	0.167	–	0.21	[322]
Sb_2Te_3	Deposited into SPF	YDF	1065.3	19.28	5.9	–	0.81	[323]
MoS_2	Deposited into SPF	EDF	1568	26.02	4.98	–	0.08	[324]
WS_2	Deposited into SPF	YDF	1052.45	23.26	0.713	–	1.29	[325]
WS_2	Deposited into SPF	YDF	1063.6	630	5.57	–	13.6	[326]
MoC_2	Sandwiched	YDF	1061.8	3.23	418	–	–	[554]
graphene	Sandwiched	TDF	1940	6.46	3.6	2	0.4	[333]
Bi_2Se_3	Sandwiched	EDF	1561.6/1562.1	3.54	13.6–25.2 ns	–	0.593-2.824	[328]
graphene	Sandwiched	TDF	1953.3	16.937	2.1	130	0.08	[335]

(*Continued*)

Table 2. Continued.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	f_{rep}/MHz	τ/ps	P_{ave}/mW	E/nJ	Ref.
graphene	Sandwiched	TDF	1884	20.5	1.2	1.35	–	[336]
graphene	Sandwiched	TDF	1897.7–1930.3	0.964	122–134 ns	–	10.6-35.2	[337]
graphene	Sandwiched	HDF	2066.8	38	1.07	44	1.2941	[338]
graphene	Deposited into SPF	TDF	1910	19.31	0.773	115	6	[341]
graphene	Deposited into microfiber	TDF	1880–1940	19.7	1.9	1.96	–	[342]
graphene	Sandwiched	TDF	1945	58.87	0.2	13	0.22	[343]
Bi_2Te_3	Deposited into SPF	TDF	1935	27.9	0.795	–	0.72	[344]
Bi_2Te_3	Deposited into microfiber	TDF	1909.5	21.5	1.26	–	–	[345]
graphene	Sandwiched	TDF	2060	20.98	0.19	54	2.55	[346]
MoS_2	SAM	TDF	1905	9.67	843	–	15.5	[347]
WS_2	Deposited into SPF	TDF	1941	34.8	1.3	–	0.0172	[348]
WTe_2	Deposited into microfiber	TDF	1915.5	18.72	1.25	–	2.13	[349]
WSe_2	Deposited into microfiber	TDF	1863.96	11.36	1.16	32.5	–	[350]
$MoTe_2$	Deposited into microfiber	TDF	1930.22	14.353	0.952	36.7	2.56	[351]
BP	Sandwiched	TDF	1910	36.8	0.739	1.5	0.0407	[352]
BP	Sandwiched	TDF	2094	29.1	1.3	11	0.379	[353]
graphene	Sandwiched	Er^{3+}-ZBLAN	2784.5	25.4	42	100	0.7	[354]
BP	SAM	Er^{3+}-ZBLAN	2783	24	42	–	25.5	[355]
BP	SAM	Ho^{3+}/Pr^{3+}-ZBLAN	2866.7	13.987	8.6	–	6.2	[356]
BP	Sandwiched	Er^{3+}-ZBLAN	2771.1	27.4	–	–	–	[357]
BP	SAM	Er^{3+}-ZBLAN	3489	28.91	–	40	–	[358]
graphene	Deposited into fiber taper	EDF	1530/1531.5/1533/1534.5	8.034	8.8	32.6	4.06	[435]
graphene	Deposited into fiber taper	YDF	1031.43/1034.94/1038.43	0.5515	74.6	3.53	6.4	[436]
graphene	Sandwiched	SOA	1544.7/1545.9	210	210	–	–	[437]
GO	Sandwiched	YDF	1056.5/1062.3/1069.5	14.2	–	–	–	[438]
graphene	Deposited into fiber taper	YDF	1061.8/1068.8	1.78	4.41-4.23	3.05	1.713	[439]
graphene	Sandwiched	EDF	1533.4/1556.1	8.4 9.1	0.9 0.94	11.63 14.27	1.38 1.57	[440]
Bi_2Se_3	Sandwiched	EDF	1547.6–1548.4 1549.2–1550 1551.4–1552.2	8.95	30	–	1.12	[441]
Bi_2Se_3	Sandwiched	EDF	1567.2/1568/1568.8/1569.2	8.83	22	–	1.1	[448]
Bi_2Te_3	Deposited into microfiber	EDF	1559.4/1557.7	239/388	1.3	–	–	[451]
WS_2	Deposited into microfiber	EDF	1558.5/1566	8.83	0.605/0.585	–	1.14	[458]
WS_2	Deposited into microfiber	EDF	1568.55/1569	2.14	11	–	6.64	[459]
BP	Sandwiched	EDF	1557.2/1557.7/1558.2	1.65	9.41	–	–	[461]
BP	Sandwiched	EDF	1533/1558	20.8	–	–	–	[462]

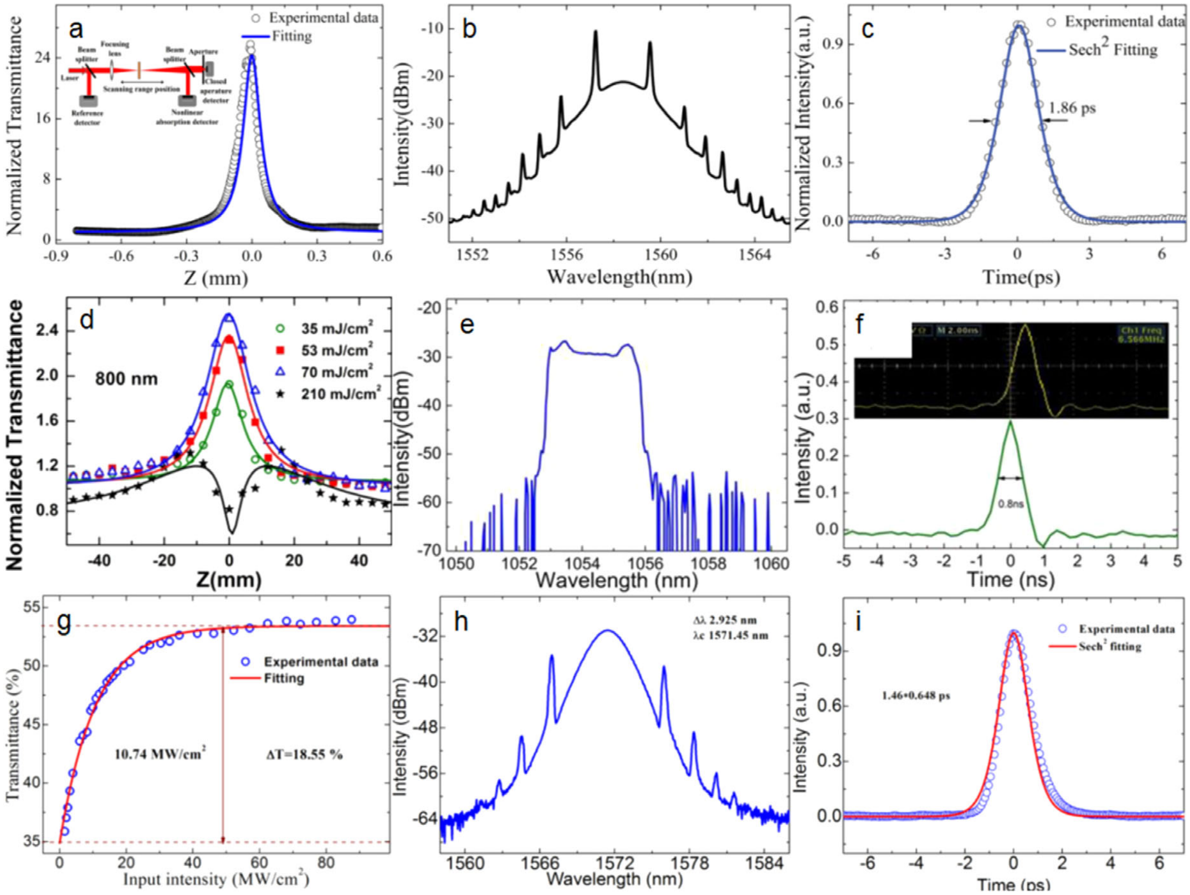
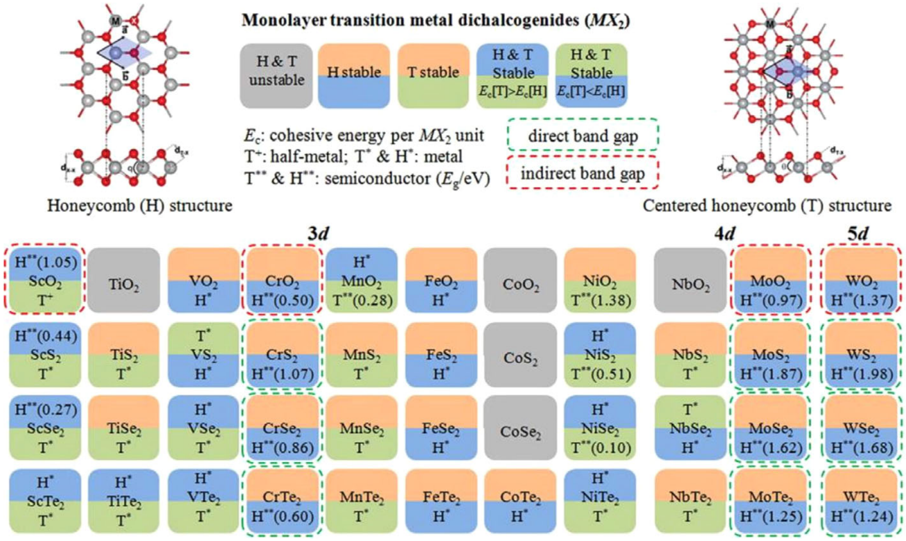


Figure 8. Mode-locked fiber lasers with different layered materials. a) Z-scan curves of few-layer topological insulator. Insert: Z-scan experimental setup. b,e,h) Optical spectra, and c,i) AC traces of mode-locked pulses. d) Z-scan curve of MoS_2 dispersion with different input fluences. f) oscilloscope trace of mode-locked pulses. g) Relation between the transmittance and input intensity for few-layer BP. Reproduced with permission.[236] Copyright 2012, AIP Publishing. Reproduced with permission.[257] Copyright 2014, Optical Society of America. Reproduced with permission.[300] Copyright 2015, Optical Society of America.

contains dozens of materials, as shown in **Figure 9**. Among them, layered MoS_2 was first studied. In 2013, the researchers experimentally discovered the saturable absorption behavior of few-layer MoS_2.[114,115] In 2014, another group of researchers further revealed its broadband saturable absorption and applied it to fiber lasers, realizing soliton mode-locking,[257] as shown in Figure 8d–f. These efforts have greatly promoted the development of few-layer MoS_2 in mode-locked lasers and achieved a lot of important results.[258–265] Similar to MoS_2, layered WS_2 also has a direct bandgap and exhibits excellent nonlinear optical properties.[266–276] For example, the researchers demonstrated the soliton mode-locked fiber lasers using WS_2 nanosheets, which were prepared by the liquid phase exfoliation[266] and pulsed laser deposition method,[267] respectively, as shown in **Figure 10**a–f. Moreover, a series of laser pulses with a pulse width of 67 fs from a single-cavity were also realized,[273] as illustrated in Figure 10g–h.

Driven by these efforts, mode-locked fiber lasers based on other layered TMDCs and their derivatives including $MoSe_2$,[277,278] $MoTe_2$,[279,280] WTe_2,[281] WSe_2,[282] $MoSe_2$,[279,283] SnS_2,[284–286] ReS_2,[287–289] In_2Se_3,[290,291] InS,[292] TiS_2,[293,294] PtS_2,[295] $PtSe_2$,[296–298] and HfS_2,[299] have also developed rapidly, as shown in Table 2. It can be seen that for these lasers, the minimum pulse width is 67 fs,[273] maximum output power is 212 mW,[280] and maximum repetition rate is 8.82 GHz.[298] Interestingly, the number of layers, oxidative and defective surfaces of layered TMDCs do not degrade their saturable absorption performance. For example, the researchers achieved the femtosecond mode-locking by using the near-infrared saturable absorption of defective bulk-structured WTe_2.[281] These studies indicate that besides graphene and topological insulators, TMDCs are another promising layered materials for mode-locked lasers.

Since 2014, black phosphorus (BP), a new 2D material, has attracted worldwide attention and rapidly become a star material.[19] This is because, its narrow bandgap can build a bridge between zero-bandgap graphene and wide-bandgap TMDCs, which has important applications in near-, and middle-infrared range. Meanwhile, its lattice is composed of two atomic layers, each of which consists of a twisted chain of phosphorus atoms. This wonderful feature makes it easy for BP to combine with many atmospheric molecules and biomolecules, so it has a very rich application in the fields of physics, chemistry, biology, and energy. The emergence of BP in ultrafast photonics began in 2015.[20–22,132–136] In this year, the researchers revealed the broadband saturable



3d								4d		5d
H^{**}(1.05) ScO_2 T^+	TiO_2	VO_2 H^*	CrO_2 H^{**}(0.50)	H^* MnO_2 T^{**}(0.28)	FeO_2 H^*	CoO_2	NiO_2 T^{**}(1.38)	NbO_2	MoO_2 H^{**}(0.97)	WO_2 H^{**}(1.37)
H^{**}(0.44) ScS_2 T^*	TiS_2 T^*	T^* VS_2 H^*	CrS_2 H^{**}(1.07)	MnS_2 T^*	FeS_2 H^*	CoS_2	H^* NiS_2 T^{**}(0.51)	NbS_2 T^*	MoS_2 H^{**}(1.87)	WS_2 H^{**}(1.98)
H^{**}(0.27) $ScSe_2$ T^*	$TiSe_2$ T^*	H^* VSe_2 T^*	$CrSe_2$ H^{**}(0.86)	$MnSe_2$ T^*	$FeSe_2$ H^*	$CoSe_2$	H^* $NiSe_2$ T^{**}(0.10)	T^* $NbSe_2$ H^*	$MoSe_2$ H^{**}(1.62)	WSe_2 H^{**}(1.68)
H^* $ScTe_2$ T^*	H^* $TiTe_2$ T^*	H^* VTe_2 T^*	$CrTe_2$ H^{**}(0.60)	$MnTe_2$ T^*	$FeTe_2$ H^*	$CoTe_2$ H^*	H^* $NiTe_2$ T^*	$NbTe_2$ T^*	$MoTe_2$ H^{**}(1.25)	WTe_2 H^{**}(1.24)

Figure 9. Summary of stability analysis and semiconducting properties of 44 different MX_2 compounds. Transition metal atoms indicated by M are divided into 3d, 4d, and 5d groups. MX_2 compounds shaded light gray form neither stable H ($2H-MX_2$) nor T ($1T-MX_2$) structure. In each box, the lower-lying structure (H or T) is the ground state. The resulting structures (T or H) can be half-metallic (+), metallic (*), or semiconducting (**) with direct or indirect band gaps. Reproduced with permission.[25] Copyright 2013, Chemical Society of America.

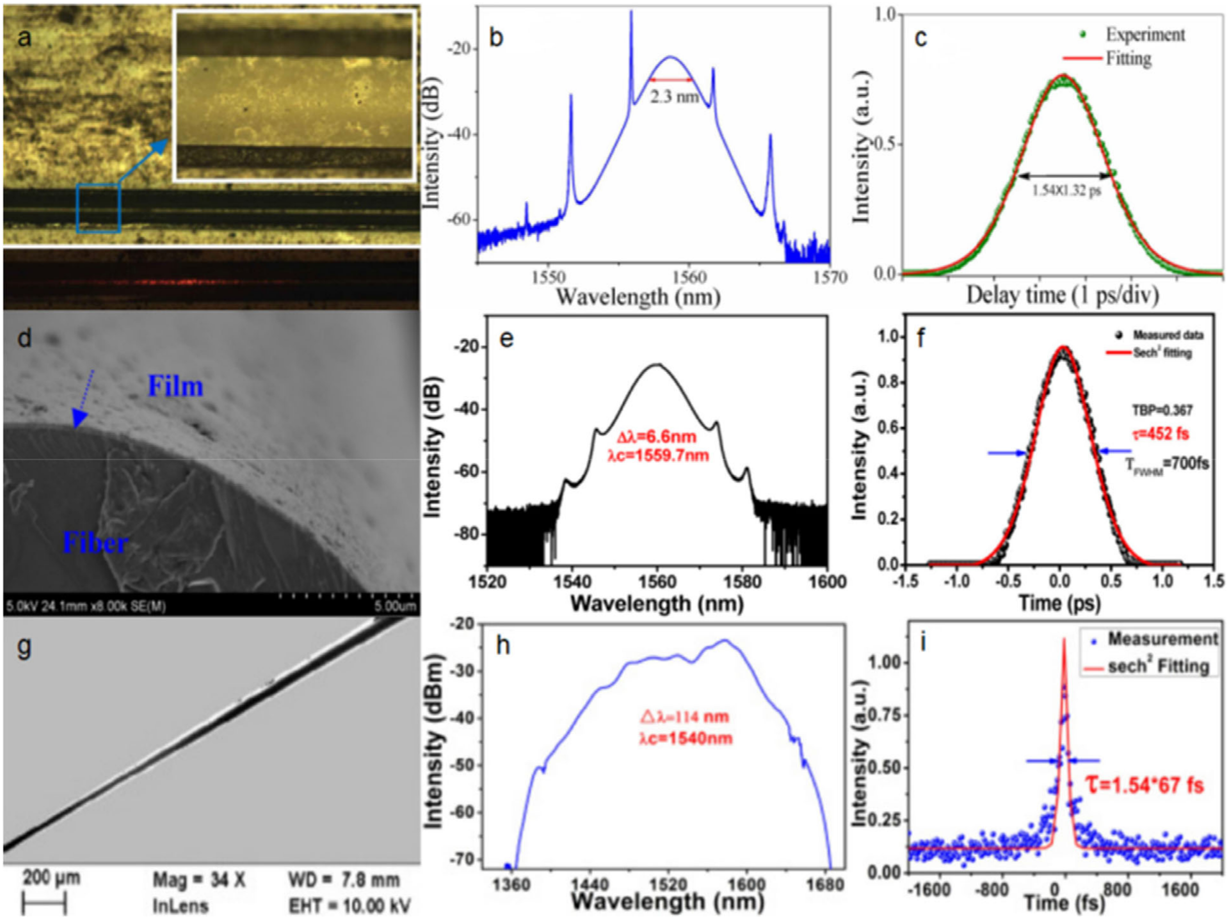


Figure 10. Mode-locked fiber lasers with few-layer WS_2. a) Photograph of mode-locker with and without the input of red light, b,e,h) Optical spectra, and c,f,i) AC traces of mode-locked pulses. d,g) SEM images of mode-locker with the microfiber. Reproduced with permission.[266] Copyright 2016, Nature Publishing. Reproduced with permission.[267] Copyright 2019, Optical Society of America. Reproduced with permission.[273] Copyright 2017, Optical Society of America.

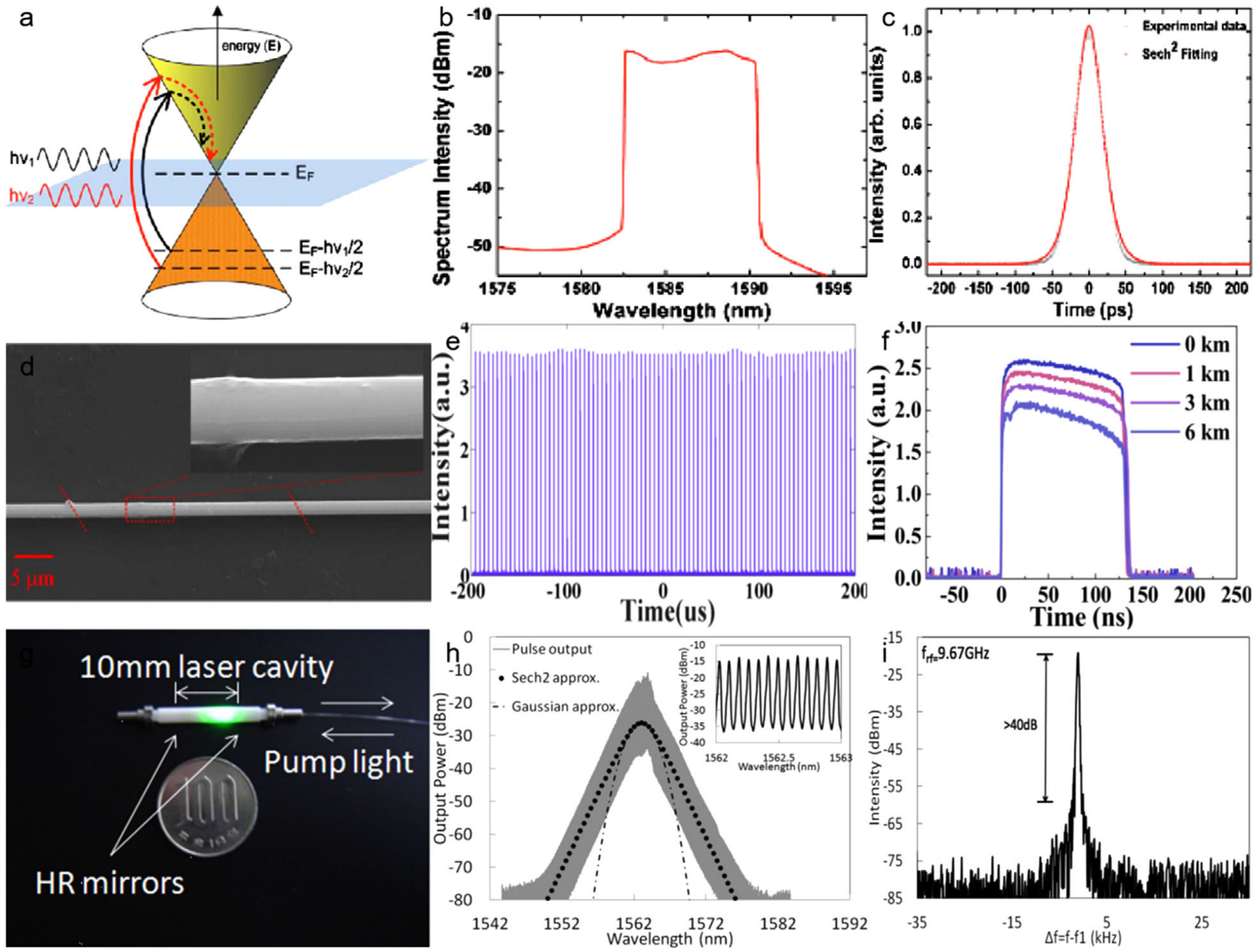


Figure 11. High performance fiber lasers based on layered materials. a) Diagram of graphene's band structure and photon absorption. b,h) Optical spectra, and c) AC trace of mode-locked pulses. d) SEM image of WS_2-deposited microfiber. e) Oscilloscope trace, and f) AC trace of rectangular pulses. g) Experimental setup of Fabry-Perot cavity laser. i) RF spectrum of mode-locked pulses. Reproduced with permission.[310] Copyright 2010, AIP Publishing. Reproduced with permission.[327] Copyright 2018, IEEE Photonics Society. Reproduced with permission.[180] Copyright 2012, AIP Publishing.

absorption characteristics of BP in the wavelength range of 1–2 μm and quickly applied it to pulsed fiber lasers,[300] as shown in Figure 8g–i. This work has attracted great attention of many research groups and has obtained many important results,[301–309] as shown in Table 2. For these mode-locked lasers, some exciting results have been obtained, including maximum repetition rate, maximum output power, and minimum pulse width of 60.5 MHz,[302] 80 mW,[306] and 102 fs,[307] respectively. These efforts not only enrich our understanding of the nonlinear optical properties of BP, but also find suitable application for it, which greatly promotes the simultaneous development of advanced materials and laser photonics.

Besides the achievements mentioned above, mode-locked fiber lasers based on layered materials have greatly improved in pulse-energy, repetition-rate, and mid-infrared waveband. The summary of these aspects will help us better understand the application of layered materials in fiber lasers.

For a long time, the realization of high pulse-energy mode-locked lasers has been a hot topic in the field of ultrafast photonics due to its potential applications in optical frequency measurement, optical sensing, and data query. To this end, researchers have proposed and developed several kinds of high pulse-energy pulses, such as dissipative solitons, self-similar pulses, rectangular pulses, and noise-like pulses. Among them, dissipative solitons have attracted special attention because their pulse energy may increase by several orders of magnitude over conventional solitons. In 2010, the researchers first introduced graphene into the study of dissipative solitons and realized it in fiber lasers,[310] as shown in **Figure 11**a–c. Thereafter, dissipative soliton fiber lasers using layered materials, including graphene,[311–316] graphene oxide,[317–319] Bi_2Te_3,[320] Sb_2Te_3,[321–323] MoS_2,[324] and WS_2[325–327] have developed rapidly, and great progress has been made in pulse energy and pulse width. Specifically, the maximum pulse-energy and minimum pulse-width are 159.4 nJ[316] and 128 fs,[321] respectively. Interestingly, rectangular pulse fiber lasers based on layered materials were also developed. For example, the researchers obtained the rectangular pulses with pulse energy of 810 nJ in a fiber laser based on WS_2 SA,[327] as shown in Figure 11d–f. Meanwhile, another group of researchers achieved the rectangular pulses with pulse energy of 2.8 nJ in a fiber laser based on topological insulator SA.[328] These efforts have strongly

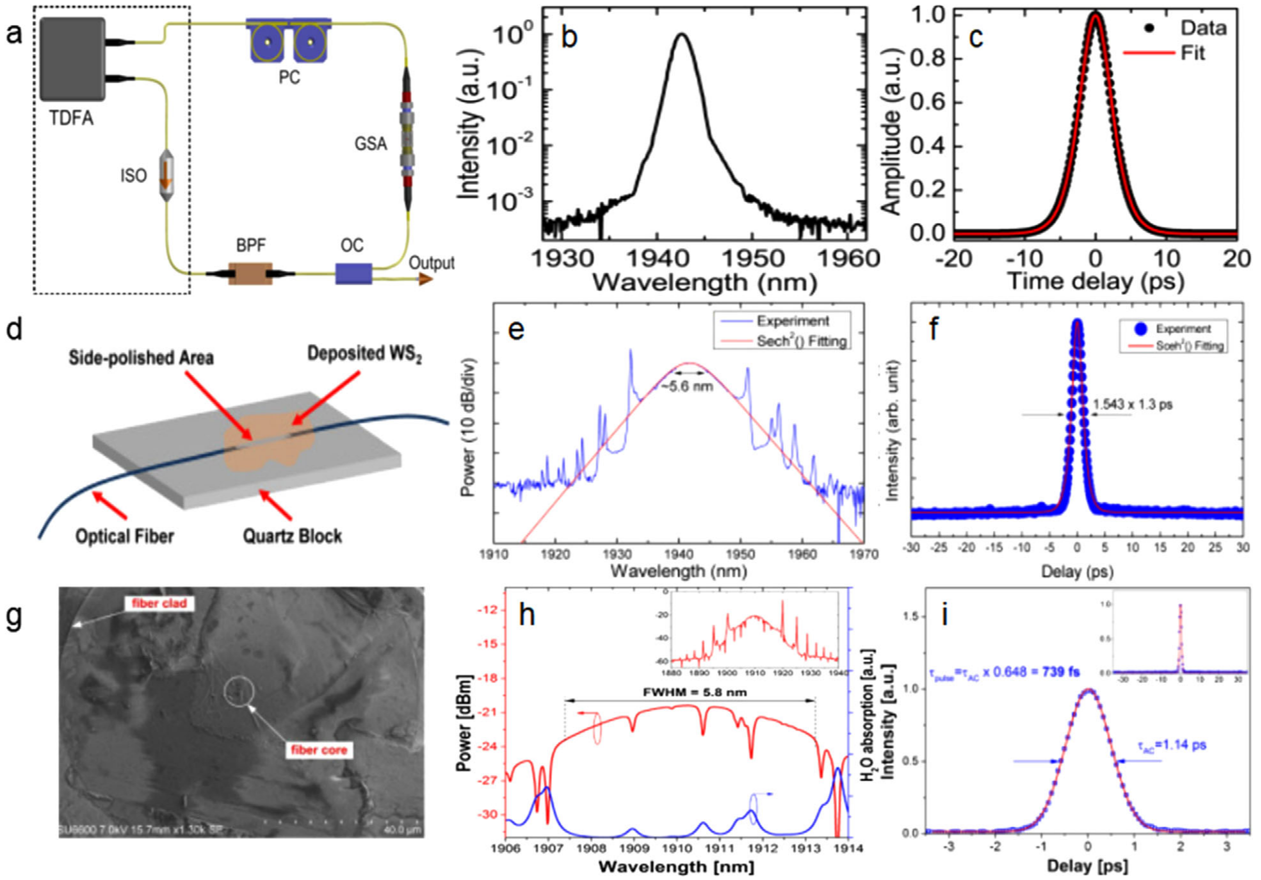


Figure 12. 2 μm mode-locked fiber lasers with layered material-based SAs. a) Experimental setup. b,e,h) Optical spectra, and c,f,i) AC traces of mode-locked pulses. d) Schematic diagram of mode-locker. g) SEM image of sandwiched mode-locker. Reproduced with permission.[333] Copyright 2012, Optical Society of America. Reproduced with permission.[348] Copyright 2015, Optical Society of America. Reproduced with permission.[352] Copyright 2015, Optical Society of America.

promoted the development of layered materials in high-energy laser pulses.

The application of layered materials in high repetition-rate fiber lasers with several GHz should also be mentioned. In theory, shortening the length of laser cavity is one of the effective ways to improve the pulse repetition-rate. For example, the researchers obtained a series of laser pulses with a repetition rate of 9.67 GHz in a Fabry–Perot cavity fiber laser based on graphene SA,[180] as shown in Figure 11g–i. In addition, harmonic mode-locking is also a common technique. With this scheme, a large number of high repetition-rate lasers based on layered materials have been developed.[179,180,207,208,216,232,238,241,243,260,278,298] Among them, the maximum repetition rate is 106.7 GHz.[232] These works paved the way for the practical application of high repetition-rate mode-locked lasers.

It is noteworthy that most of the aforementioned fiber lasers operate in the near-infrared band. Meanwhile, visible[329–332] and mid-infrared[333–358] pulsed lasers based on layered materials have also developed rapidly. Especially for the latter, this is a great breakthrough. This is because, for common SAs, such as semiconductor saturable absorber mirrors and carbon nanotubes, it is not easy to operate in mid-infrared, especially in the band above 2.5 μm. For this reason, researchers in the field of ultrafast lasers have been seeking for suitable pulse-shaping materials, and the emergence of layered materials provides such a new opportunity. The application of layered materials in mid-infrared fiber lasers began in 2012. For example, the researchers realized a 2-μm mode-locked fiber laser based on graphene SA,[333] as shown in **Figure 12**a–c. Additionally, researchers from other groups also demonstrated the 2 μm mode-locked lasers based on WS_2 SA[348] and BP SA,[352] respectively, as shown in Figure 12d–i. Up to now, 2 μm mode-locked fiber lasers have been achieved with layered materials, such as graphene,[333–341,344] Bi_2Te_3,[345,346] MoS_2,[347] WS_2,[348] WTe_2,[349] WSe_2,[350] $MoTe_2$,[351] and BP,[352,353] respectively. Among them, several important works should be emphasized, such as the maximum pulse energy of 35.2 nJ,[335] the repetition rate of 58.87 MHz,[341] and the minimum pulse width of 190 fs.[344]

Quite recently, the 3-μm pulsed laser using layered materials such as graphene[354] and BP,[355–358] has also been developed. For these mode-locked lasers, some exciting results have been achieved, including maximum output power, minimum pulse width, and maximum repetition rate of 25.5 nJ,[355] 8.6 ps,[356] and 28.91 MHz,[358] respectively. For example, the researchers prepared a mid-infrared saturable absorption mirror by transferring the few-layer BP to a gold mirror and further obtained a

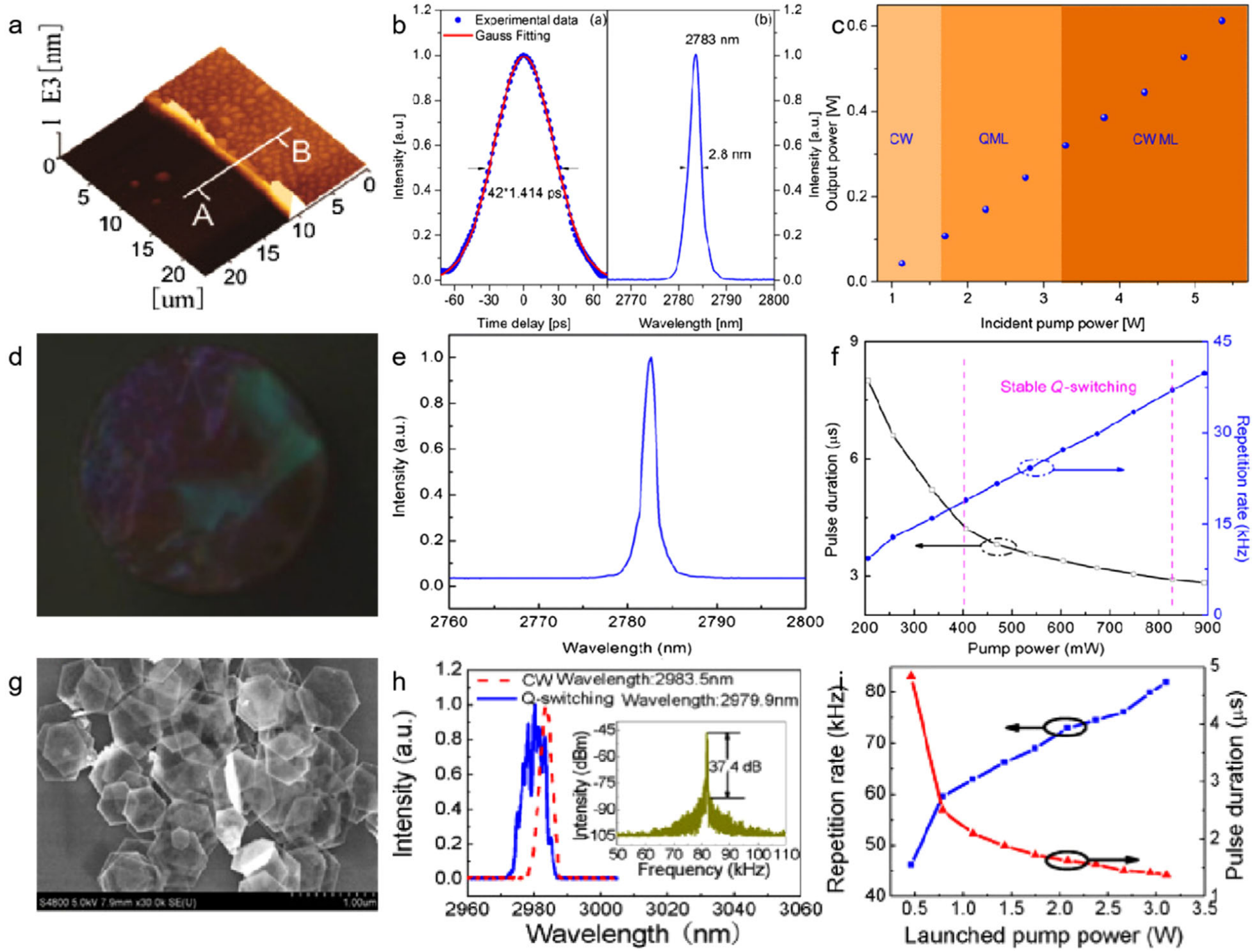


Figure 13. 3 µm mode-locked/Q-switched fiber lasers with layered material-based SAs. a) SEM image of BP flakes. b) AC trace and the corresponding spectrum. c) Relation between the output power and the pump power. d) SEM image of sandwiched mode-locker, e) Optical spectrum of Q-switched pulses. f,i) Relations between the pulse duration and repetition rate and the pump power. g) SEM image of Bi_2Te_3 samples. h) Optical spectra of cw and Q-switched pulses, inset: RF spectrum. Reproduced with permission.[355] Copyright 2016, Optical Society of America. Reproduced with permission.[371] Copyright 2015, Optical Society of America. Reproduced with permission.[389] Copyright 2013, Optical Society of America.

mode-locked Er: ZBLAN fiber laser at 2783 nm,[355] as illustrated in **Figure 13**a–c. These efforts have strongly promoted the development of mid-infrared pulsed lasers.

3.1.2. Q-Switching Operation

Shortly after the realization of the graphene mode-locked lasers, the researchers began to introduce layered materials into Q-switched fiber laser to obtain high peak power laser pulses. In theory, if a material has saturable absorption, it can be used as a Q-switcher, which turns the continuous wave into a series of Q-switched pulses, and most layered materials just have this property. Based on this consideration, the researchers have achieved Q-switched fiber lasers based on graphene,[359] topological insulator,[360] and few-layered MoS_2,[361,362] respectively. These early efforts have promoted the rapid development of layered materials in Q-switched lasers. For example, Q-switched fiber lasers based on layered materials including graphene,[363–380] topological insulators,[381–392] TMDCs,[393–414] and BP,[415–420,422,423] have been obtained, as illustrated in **Table 3**. It can be seen that these lasers have excellent performance in output power, pulse width, repetition frequency, and peak power, which is not inferior to the traditional lasers based on semiconductor saturable absorption mirrors and carbon nanotubes. Among them, the maximum pulse energy is 877 µJ[379] and minimum pulse width is 155 ns,[405] respectively.

More importantly, these Q-switched lasers cover a wide wavelength range from visible, near-infrared to mid-infrared, and have several distinct characteristics. First, besides near-infrared operation, the visible-light Q-switched fiber lasers using TMDCs,[329,330] Bi_2Se_3,[331] and BP[332] have been also achieved, respectively. For example, the researchers realized a visible-light Q-switched Pr^{3+}-doped fiber laser based on TMDCs SA,[329] as shown in **Figure 14**. Second, layered materials-based mid-infrared Q-switched fiber lasers at 2 µm[361,372–374,379,382,406] and 3 µm[371,389,418] have been also developed rapidly. For example, the researchers achieved the Q-switched fiber laser near 3 µm using graphene[371] and topological insulator[389] saturable absorption mirrors, respectively, as shown in

Table 3. Summary of Q-switched fiber lasers with layered materials.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	f_{rep}/kHz	τ/μs	P_{peak}/W	E/nJ	Ref.
graphene	Sandwiched	EDF	1566.17/1566.35	3.3–65.9	3.7	1.1	16.7	[359]
Bi_2Se_3	Sandwiched	YDF	1060	8.3–29.1	1.95	–	17.9	[360]
MoS_2	Sandwiched	YDF	1066.5	6.4–28.9	5.8–17	–	32.6	[361]
		EDF	1560	6.5–27	5.4–23.3		63.2	
		TDF	2030	33.6–48.1	1.76–2.5		1000	
MoS_2	Sandwiched	EDF	1520–1568	10.6–34.5	5–9	–	160	[362]
graphene	Sandwiched	EDF	1522–1555	36–103	2	2.4	40	[363]
graphene	SAM	YDF	1064.2	140–257	70	12	46	[364]
graphene	Sandwiched	YDF	1029–1037.4	3–3.9	39.8–56.1	0.58	10.3	[365]
		EDF	1554–1560.23	2.8–63	2.5–51	4.57	72.5	
graphene	Sandwiched	EDF	1519.3–1569.9	8.5–29.05	4.6	0.2225–2.4	82.61	[366]
graphene	Sandwiched	EDF	1564/1566	104–116	1.85–3.85	14.6	125	[367]
graphene	Sandwiched	EDF	1531.12/1556.79	12.6–22.8	8.2–26.5	–	70	[368]
GO	Sandwiched	EDF	1550–1570	22–61	6.6–13.7	9.3	63.9	[369]
graphenen	Sandwiched	PM YDF	1027	28.9–110	1.3–3.2	15.6	141.8	[370]
graphene	SAM	Er-ZBLAN	2783	7–37	2.9–7	62	1.67 μJ	[371]
GO	Deposited into microfiber	TDF	2032	20–45	3.8–9	302	6.71	[372]
graphene	SAM	TDF	1957	103–252	0.76–1.4	96	0.38	[373]
graphene	SAM	TDF	2005	73–280	0.32–0.47	5.2 W	18	[374]
graphene	Sandwiched	YDF	1066.83	5.93–20.03	3.1–15	106.2	5.3 μJ	[375]
		EYDCF	1535.56	8–29	3.9–25	33.5	1.2 μJ	
graphene	Deposited into microfiber	YDF	1060	30.32–101.29	2.61–5.21	0.99	–	[376]
graphene	In-line	EDF	1570	6.2–11.8	–	–	–	[378]
GO	Sandwiched	TDF	1950.27	33.5–83.2	1.1–1.5	73W	0.877 mJ	[379]
graphene	Wrapped around DFB	EDF	1544	0.7–3.8	1–7	–	10	[380]
graphene	Deposited into SPF	Ho-ZBLAN	1192.6	24–111	800–5730	1125	440	[377]
Bi_2Se_3	Sandwiched	EDF	1545–1565	4.5–12.88	13.4–36	–	13.3	[381]
Bi_2Se_3	Sandwiched	EDF	1980	8.4–26.8	4.18–18.5	–	313	[382]
Bi_2Se_3	Sandwiched	EDF	1530	6.2–40.1	4.9	–	39.8	[383]
Bi_2Se_3	Sandwiched	EDF	1565	459–940	1.9–7.76	–	23.8	[384]
Bi_2Te_3	Sandwiched	EDF	1510–1589	2.15–12.8	13–49	–	1525	[385]
Bi_2Te_3	Deposited intoSPF	EDF	1562.9	7.5–42.8	2.81–9.36	–	12.7	[386]
Bi_2Te_3/BP	Deposited into SPF	EDF	1550	4.43–18	9.35–31	–	28.3	[387]
		TDF	1832	20–25.5	4–6.67		75	
Bi_2Te_3	Deposited into SPF	EDF	1559.5	8.74–21.24	4.88–8.46	–	3.8	[388]
Bi_2Te_3	SAM	Ho- ZBLAN	2979.9	46–81.96	1.37–4.83	–	3.99	[389]
Sb_2Te_3	SAM	EDF	1530–1570	98–338	0.4	–	18.07	[390]
Bi_2Se_3	Sandwiched	EDF	1550.5	63.2–68.9	1.49–2.54	–	0.797	[391]
Bi_2Te_3	Sandwiched	EDF	1550	31.54–49.4	3.7–5.2	5.5	125	[392]
MoS_2	Sandwiched	YDF	1030–1070	65.3–89	2.68–4.4	–	1.1	[393]
MoS_2	SAM	EDF	1549.83	116–131	0.66–0.76	–	152	[394]
MoS_2	Sandwiched	EDF	1550–1575	22	6–35	–	150	[395]
MoS_2	Sandwiched	EDF	1549.91	10.6–173.1	1.66–6.11	–	27.2	[396]
MoS_2	Sandwiched	EDF	1560.5	28.6–114.8	1.92–3.7	–	8.2	[397]
MoS_2	Sandwiched	EDF	1560	36.8–91.7	3.2–5.1	–	0.029	[398]
$Mo_{0.5}W_{0.5}S_2$	Sandwiched	EDF	1560	34.1–98.5	1.95–3.42	–	–	[399]
$ReSe_2$	Sandwiched	EDF	1566	16.64	4.98	–	36	[400]
WS_2	Sandwiched	EDF	1027–1065	65.3–106.2	1.57–2.11	–	28.8	[401]
WS_2	Sandwiched	YDF	1030	24.9–36.7	3.2–6.4	–	13.6	[402]
		EDF	1558	79–97	1.1–3.4		179.6	

(*Continued*)

Table 3. Continued.

Device integration		Laser parameters						
2D Mater.	Integration method	Gain medium	λ/nm	f_{rep}/kHz	τ/μs	P_{peak}/W	E/nJ	Ref.
WS_2	Sandwiched	EDF	1547.5	80–120	1–3.1	–	0.05	[403]
WS_2	Sandwiched	Pr-ZBLAN	635.1	232.7–512.8	0.207	–	0.04	[329]
MoS_2			635.5	240.4–438.6	0.227		0.03	
$MoSe_2$			635.4	357.1–555.1	0.24		0.02	
WS_2	Sandwiched	Pr-ZBLAN	604	67.3–127.9	0.435–1.101	–	6.4	[330]
MoS_2			602	50.8–118.4	0.602–1.955		5.5	
WS_2	Deposited into SPF	EDF	1567.8	82–134	0.92–2.82	–	19	[404]
WS_2	SAM	EDF	1560	29.5–367.8	0.155–1.27	–	68.5	[405]
$MoSe_2$	Sandwiched	YDF	1060	60–74.9	2.8–4.6	–	116	[406]
		EDF	1566	26.5–35.4	4.8–7.9		825	
		TDF	1924	14–21.8	5.5–16		42	
MoS_2	Sandwiched	EDF	1560	7.758–41.45	9.92–13.534	–	184.7	[407]
$MoSe_2$				60.72–66.85	4.04–6.506	–	365.9	
WS_2				47–77.925	3.966–6.707	–	1179.4	
WSe_2				46.28–85.36	4.063–9.182	–	484.8	
SnS_2	Sandwiched	EDF	1532.7	172.3–233	510–1010	9.33	–	[408]
WSe_2	Sandwiched	EDF	1562	77–242	1.2	26.7	110	[409]
$MoSe_2$			1558	64–122	1.53	17.16	140.7	
$TiSe_2$	SAM	EDF	1530	70–154	1126	11.54	–	[410]
MoS_2	Sandwiched	EDF	1552	26.6–40.9	3.9–5.4	3.5	–	[411]
$MoSe_2$	Sandwiched	EDF	1557.6	47.5–105.7	1.09	23.2	224	[412]
WS_2	Sandwiched	EDF	1559	16.15–60.88	2.396–7.6	9.5	195	[413]
PtS_2	Sandwiched	EDF	1568.8	24.6	4.2	1.1	45.6	[414]
BP	Sandwiched	EDF	1562.87	6.983–15.78	10.32–39.84	–	94.3	[415]
BP	Sandwiched	EDF	1561.9	7.86–34.32	2.96–55	–	194	[416]
BP	Sandwiched	EDF	1912	69.4–113	0.731–1.42	–	632.4	[417]
BP	SAM	Dy-ZBLAN	2970–3230	47–86	0.74–1.8	–	–	[418]
BP	Sandwiched	Pr-ZBLAN	635.4	108.8–409.8	0.383–1.56	–	27.6	[332]
BP	Sandwiched	EDF	1563–1567	64.51–82.64	1.36–3.39	–	148.63	[419]
BP	SAM	Er-ZBLAN	2779	39–63	1.18–2.1	–	7.7	[420]
graphene/WS_2	SAM	$Nd:YVO_4$	1064	3528–7777	66–149	275	33.1	[421]
BP	Deposited into microfiber	EDF	1542.4	3–18.5	10–40	1.4	90	[422]
			1543.2	2.6–9.4	12–50	1.35	135	
BP	Deposited into microfiber	YDF	1064.7	26–76	2–5.5	1.4	–	[423]
WSe_2	Sandwiched	EDF	1560	4.5–49.6	3.1–7.9	1.23	33.2	[463]

Figure 13d–i. Third, the broadband tunable operation of these lasers[362,363,365,366,369,381,385,390,393,395,401,402,418,419] have also been widely studied. It is worth mentioning that the Q-switched pulse with a maximum tunable range of 78.2 nm was obtained in a Q-switched laser using topological insulator.[385] Interestingly, besides single-wavelength operation, layered materials-based multiwavelength lasers have been also developed.[359,367,368,422] These efforts have injected vitality into the research of Q-switched fiber lasers and provided new choice for their commercialization.

3.1.3. Nonlinear Optical Phenomenon

In the above two sections, we briefly reviewed the applications of layered materials in mode-locked/Q-switched fiber lasers. For these lasers, the saturable absorption property of layered materials play a key role. Interestingly, their Kerr nonlinearity is also very important in mode-locked fiber lasers, which has attracted great attention of researchers in nonlinear optics. As described in Section 2.2, most of layered materials have strong Kerr nonlinearity. Thus, if the layered materials-based nonlinear optical devices are introduced into the fiber lasers, it will become a good platform for the study of nonlinear optical phenomenon.[157–159,424]

Since 2010, versatile soliton pulses, including conventional soliton, dark solitons, dissipative soliton, and even rogue waves, have been observed in mode-locked fiber lasers based on layered materials such as graphene,[425–440] topological insulators,[441–451] TMDCs,[452–460,463] and BP.[461,462] For example, the researchers studied the dynamic of soliton pulses in graphene mode-locked fiber lasers and obtained the disordered multiple solitons and

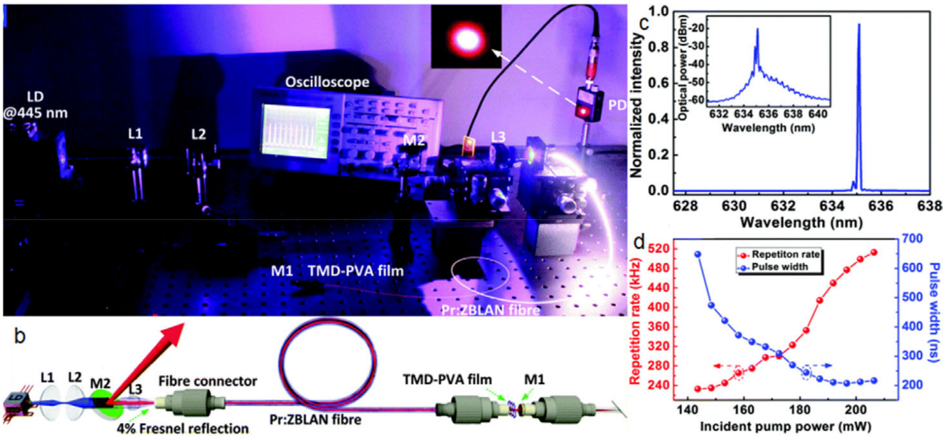


Figure 14. Visible-light Q-switched fiber laser based on TMDCs SA. a) Photograph and b) image of experimental setup. c) Optical spectra of Q-switched pulses, d) Relation between the repetition rate and pulse width and the pump power. Reproduced with permission.[329] Copyright 2016, RSC Publishing.

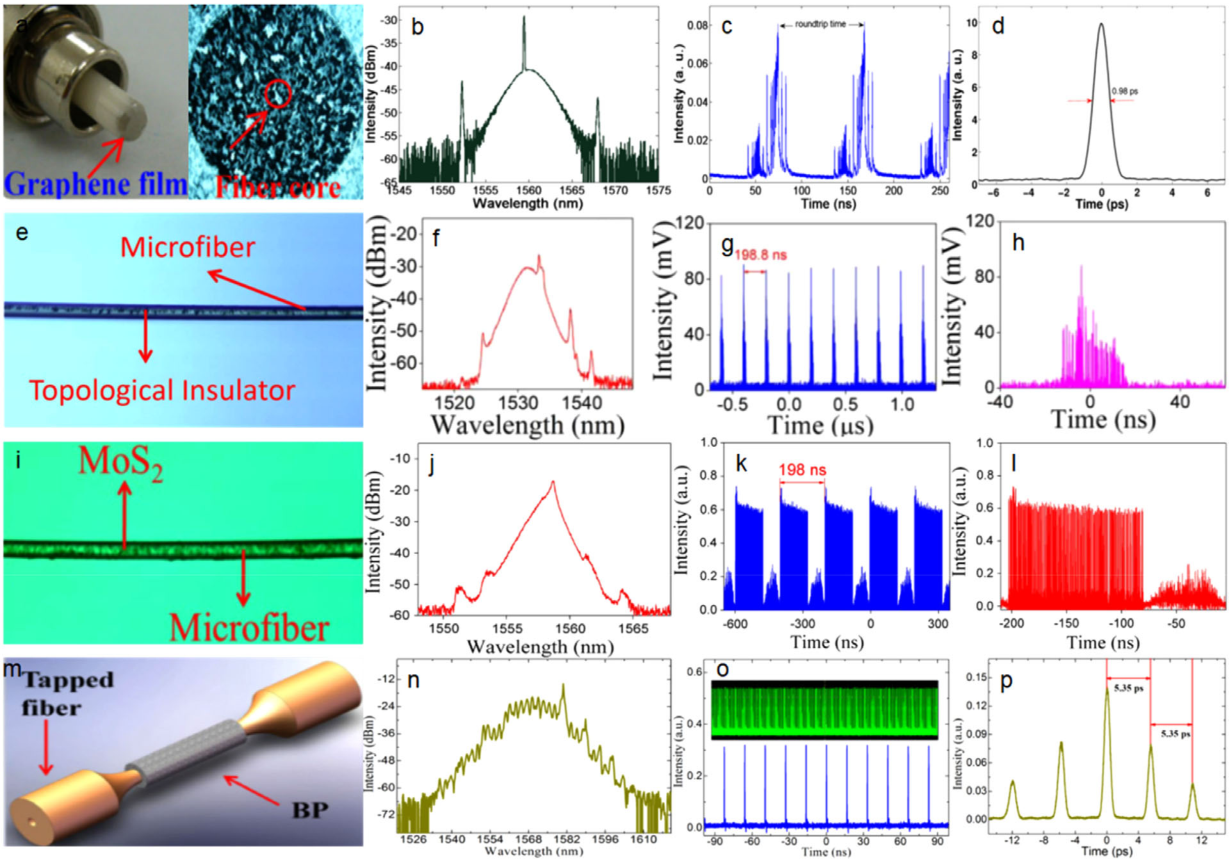


Figure 15. Versatile soliton pulse generated from mode-locked fiber lasers with layered materials. a) Image of sandwiched mode-locker. b,f,j,n) Optical spectra, c,g,k,o) Oscilloscope traces, d) AC trace of soliton pulses. e) Image of mode-locker, and h) single pulse of mode-locked chaotic multi-pulse bunch. i) Image of mode-locker, and l) single pulse of double-scale soliton clusters. m) Image of tapered fiber-based SA. p) AC trace of bound solitons. Reproduced with permission.[425] Copyright 2012, Optical Society of America. Reproduced with permission.[428] Copyright 2015, Optical Society of America. Reproduced with permission.[442] Copyright 2015, Optical Society of America. Reproduced with permission.[302] Copyright 2016, Optical Society of America.

bound solitons at low pump strength and soliton flow at higher pump strength,[425] as shown in **Figure 15**a–d. In addition, the researchers from another group observed the dissipative rogue waves[428] and even more soliton patterns,[442] including multi-soliton molecules, soliton clusters, and chaotic multi-pulse in a mode-locked fiber laser based on layered materials, as shown in Figure 15e–l. Furthermore, bound solitons and noise-like states were also trapped in a fiber laser based on BP SA,[302] as shown in Figure 15m–p. Interestingly, another group also found six kinds of dip sidebands in the WS_2 mode-locked fiber laser,[456] as

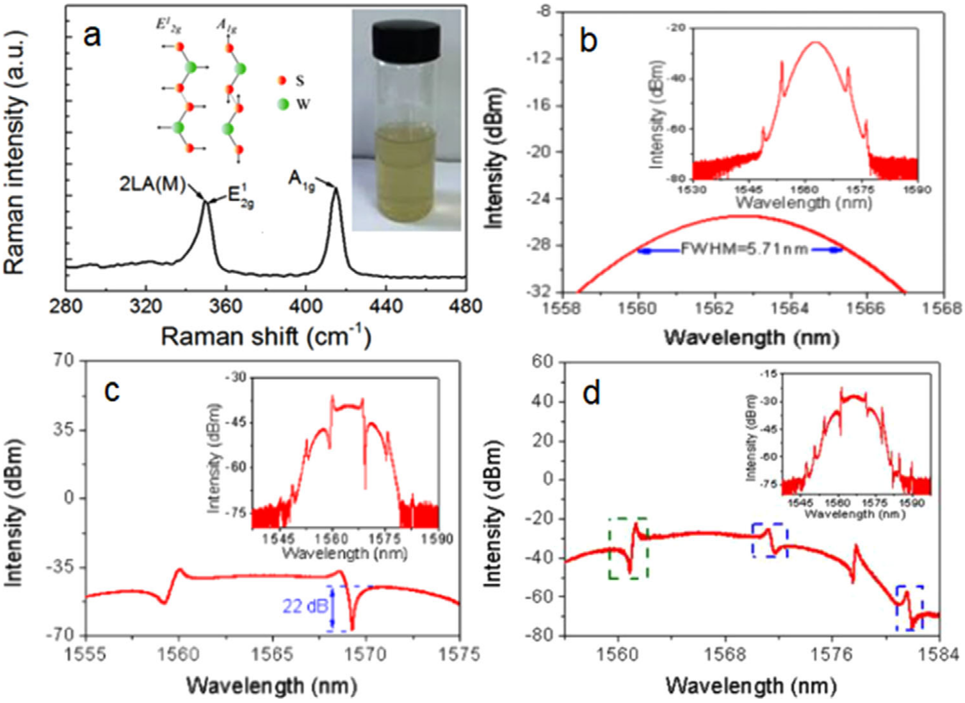


Figure 16. Dip-type sidebands pulse in a soliton laser based on WS_2 SA. a) Raman spectrum and photograph of WS_2 sample. b) Optical spectrum of conventional soliton pulses. c,d) Optical spectra of dip-type sideband pulses with the maximum depth and second-order form, respectively. Reproduced with permission.[456] Copyright 2016, Optical Society of America.

illustrated in **Figure 16**. These discoveries not only greatly enrich our understanding of layered materials, but also promote the study of nonlinear optics.[154–166,464]

3.2. Solid-State Lasers

Besides fiber lasers, layered materials have also been widely used in solid-state lasers. It is well known that solid-state lasers are generally composed of free-space resonators, and rare earth-doped glass or crystal matrix materials are used as gain media. Similar to fiber lasers, solid-state lasers also operate in two modes: continuous wave and pulse. In order to achieve pulse operation, a suitable SA is usually needed, which has been the pursuit of researchers in the field of solid-state lasers. The emergence of layered materials offers such a good opportunity. Next, we will briefly review the applications of layered materials in mode-locked and Q-switched solid-state lasers, respectively.

3.2.1. Mode-Locking Operation

As described in Section 2.2, most of the layered materials have excellent saturable absorption properties and can be used to prepare SA. However, its fabrication process is different from that of fiber integrated devices. In the experiment, the researchers transferred it to the optical mirrors by deposition or spin-coating method, made a saturable absorption mirror (SAM), and then placed it in a solid-state laser to achieve mode-locking.

In 2010, the researchers obtained the mode-locked pulse in a ceramic Nd:YAG solid-state laser based on graphene SAM.[465] This work promotes the application of graphene and other layered materials in mode-locked solid-state lasers. Up to now, plenty of mode-locked solid-state lasers using layered materials such as graphene,[466–491] TMDCs,[492–495] and BP[496–498] have been achieved, as shown in **Table 4**. It can be seen that these lasers have excellent performance in pulse width, repetition rate, and pulse energy, which is not inferior to similar lasers. Among them, the maximum pulse energy is 18.3 μJ,[482] maximum repetition rate is 6.8 GHz,[484] and minimum pulse width is 32 fs,[486] respectively. For example, the researchers achieved the femtosecond pulses from mode-locked $Yb:CaYAlO_4$[486] and Yb:YAG[492] lasers based on graphene and WS_2 SAM, respectively, as shown in **Figure 17**a–f. Recently, the researchers from another group realized the self-starting mode-locking operation from a solid-state laser based on BP SAM,[496] as illustrated in Figure 17g–i.

In order to obtain laser pulses at different wavelengths, the researchers used different gain media in the above-mentioned solid-state lasers, including Nd:YAG,[465,477] Yb:KGW,[466] $Nd:GdVO_4$,[467,498] Cr:forsterite,[468,485] Tm:YAP,[469] Tm:CLNGG,[470] Ti:sapphire,[471,488] $Tm:Lu_2O_3$,[472] Cr:ZnSe,[473] Yb:GAGG,[474] Cr:YAG,[475] $Nd:YVO_4$,[478,481,496] Cr:ZnS,[480,487] Yb:YAG,[482,492] Cr:LiSAF,[483] Yb:CYA,[486] Tm:MgW,[489] $Pr:GdLiF_4$,[493] and Yb,Lu:CALGO.[497] Using abundant gain media and layered materials, the researchers have extended the operation wavelength of mode-locked solid-state lasers from visible light to near infrared, even 3 μm mid-infrared band. These efforts provide a new idea for the research of ultrafast solid-state lasers.

Table 4. Summary of mode-locked solid-state lasers with layered materials.

Device integration		Laser parameters						
2D Mater.	SA type	Gain medium	λ/nm	f_{rep}/MHz	τ/ps	P_{ave}/mW	E/nJ	Ref.
graphene	SAM	Nd:YAG	1064	88	4	100	–	[465]
graphene	SAM	Yb:KGW	1031.1	86	0.428	504	5.9	[466]
graphene	SAM	Nd:GdVO$_4$	1065	43	16	360	8.4	[467]
graphene	SAM	Cr:forsterite	1240	74.65	0.094	230	–	[468]
graphene	SAM	Tm:YAP	2023	71.8	10	268	3.7	[469]
graphene	SAM	Tm:CLNGG	2018	99	0.729	60.2	–	[470]
graphene	SAM	Ti:sapphire	800	99.4	0.063	480	–	[471]
graphene	SAM	Tm:Lu$_2$O$_3$	2067	110	0.41	270	2.45	[472]
graphene	SAM	Cr:ZnSe	2500	77	0.226	80	–	[473]
rGO	SAM	Yb:GAGG	1041.1	45	0.643	0.8	–	[474]
graphene	SAM	Cr:YAG	1516	85	0.091	107	–	[475]
graphene	SAM	VECSEL	935–981	2.48 GHz	8	12.5	–	[476]
graphene	SAM	Nd:YAG	1064	112	15.6	2	18	[477]
graphene	SAM	Nd:YVO$_4$	531.7	71.4	0.374	117	1.6	[478]
graphene	SAM	VECSEL	1030	1.76 GHz	0.353	10.2	2.8	[479]
graphene	SAM	Cr:ZnS	2370	46	0.87	700	15.5	[480]
graphene	SAM	Cr:ZnS	2370	108	0.041	250	2.3	[481]
graphene	SAM	Yb:YAG	1048	105.7	0.367	1930	18.3	[482]
graphene	SAM	Cr:LiSAF	850	132	0.068	11.5	0.086	[483]
graphene	SAM	Yb:Er-doped phosphate glass	1535	6.8 GHz	6	27	–	[484]
graphene	Voltage-controlled gold supercapacitor	Cr:forsterite	1240	4.51	0.089	54	–	[485]
graphene	SAM	Yb:CYA	1068	113.5	0.032	26.2	–	[486]
graphene	SAM	Cr:ZnS	2120–2408	112.22	2.4	880	7.8	[487]
graphene	Voltage-controlled gold supercapacitor	Ti^{3+}: sapphire	830	131.7	0.048	113	–	[488]
graphene	SAM	Tm:MgW	2017	75.95	0.086	91	1.1	[489]
graphene	SAM	Alexandrite	740	5.56	0.065	–	1.4	[490]
graphene	SAM	Yb:CNGS	1064	85.9	0.085	23	–	[491]
WS$_2$	SAM	Yb:YAG	1064	86.7	0.736	–	3.11	[492]
MoS$_2$	SAM	Pr^{3+}:GdLiF$_4$	522.4 607.6 639.2 639	101.4 90.2 104.4 94.7	46 30 55 25	– – – –	0.1 0.2 0.21 0.49	[493]
PtSe$_2$	SAM	Nd:LuVO$_4$	1066.573	61.3	15.8	180	2.94	[494]
In$_3$Se$_2$	SAM	Yb:KYW	1064	42.4	0.352	560	13.2	[495]
BP	SAM	Nd:YVO$_4$	1064.1	140	6.1	–	3.29	[496]
BP	SAM	Yb,Lu:CALGO	1053.4	63.3	0.272	–	6.48	[497]
BP+EOM	SAM	Nd:GdVO$_4$	1340.7	58.14	9.24	–	–	[498]

3.2.2. Q-Switching Operation

Besides the mode-locked lasers mentioned above, the researchers also introduced layered materials into Q-switched solid-state lasers to obtain high peak power pulses. As we know, layered materials have saturable absorption properties and can be used to make Q-switchers. Based on this, the researchers fabricated a graphene Q-switcher and applied it to solid-state lasers to achieve Q-switched pulses,[499] as shown in **Figure 18**. Subsequently, they also realized the Q-switched solid-state lasers with topological

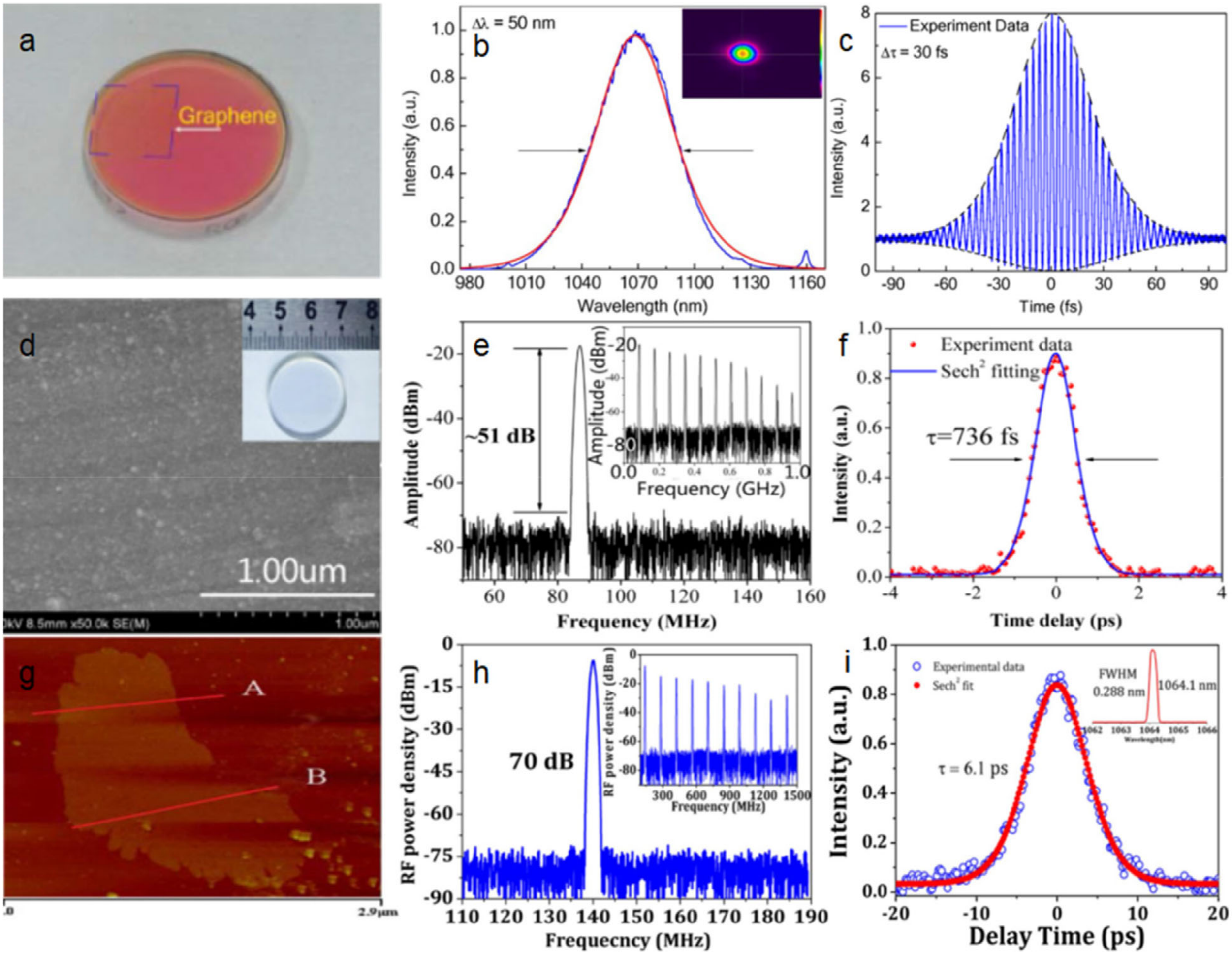


Figure 17. Mode-locked solid-state lasers with layered materials-based SAs. a) Image of the graphene SA mirror. b) Optical spectrum. Inset: its spatial beam profile. c,f,i) AC traces. d) SEM image of WS_2 film (Inset: image of the the SiO_2 substrate). e,h) RF spectra. g) AFM image of phosphorene. Reproduced with permission.[486] Copyright 2016, Optical Society of America. Reproduced with permission.[492] Copyright 2015, Optical Society of America. Reproduced with permission.[496] Copyright 2015, Optical Society of America.

insulator[500] and MoS_2,[501] respectively. These early efforts have greatly promoted the development of layered materials in Q-switched solid-state lasers.

Since 2010, Q-switched solid-state lasers based on layered materials including graphene,[502–514] topological insulators,[515–525] TMDCs,[526–561] BP,[562–570] and $g\text{-}C_3N_4$,[571–573] have been developed rapidly, as shown in **Table 5**. It can be seen that these lasers have excellent performance in output power, pulse width, repetition rate, and peak power. Among them, the maximum pulse energy (16.2 µJ,[508] 9.63 µJ,[519] 2.63 mJ,[542] 24.3 µJ[559]) and the minimum pulse width (22.9 ns,[507] 238 ps,[522] 850 fs,[533] 2.86 ps[580]) have been obtained in solid-state lasers with graphene, topological insulators, TMDCs and BP, respectively.

More importantly, these Q-switched lasers have several distinct characteristics as follows. First, they cover a very wide wavelength range from visible, near infrared to mid-infrared, that is, 0.6–3 µm. For example, the researchers achieved the visible-light Q-switched laser using topological insulators.[525] In addition, 3-µm Q-switched lasers based on layered materials have also been widely studied.[531,544,548,551–553,555,562,566,571] Second, besides single-wavelength operation, multiwavelength Q-switched lasers have been also developed.[503,512,516,518,519,527] As can be seen from Table 5, multiwavelength Q-switched lasers at 1 µm, 2 µm[563] or even 3 µm[553,562] have emerged in recent years. Third, in order to compensate for the shortcomings of single approach, the researchers have also developed a hybrid Q-switching scheme, which combines the layered material-based Q-switcher with the active Q-switcher.[533,540–542] Further study shows that this technology has more advantages than single electro-optic modulation[540,542] or acousto-optic modulation.[533,541] These work not only verify the broadband saturable absorption property of layered materials, but also greatly expand their application range.

3.3. Waveguide/Disk Lasers

Besides the aforementioned fiber- and solid-state lasers, the applications of layered materials in other types of lasers have also been developed rapidly. In this section, we focus on two types of

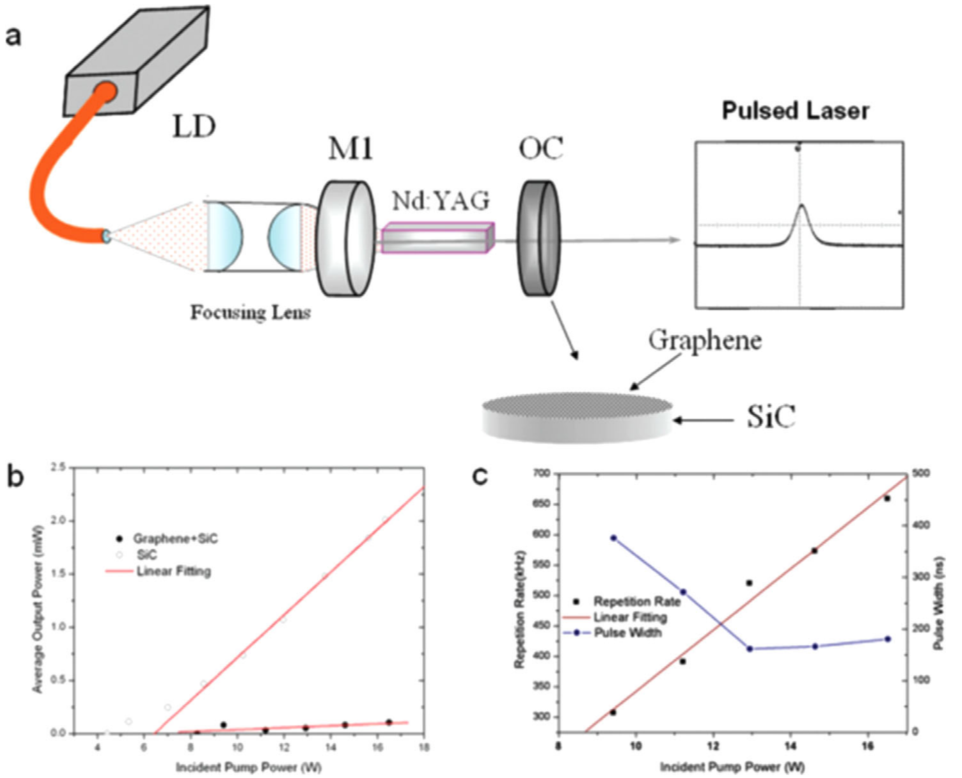


Figure 18. Q-switched solid-state laser based on graphene SA. a) Experimental setup, b) Relation between the output power and the pump power, c) Relation between the repetition frequency and pulse width and the pump power. Reproduced with permission.[499] Copyright 2010, American Chemical Society.

lasers: waveguide lasers and disk lasers. As a miniaturized and integrated laser, waveguide laser has a waveguide structure that can be used as both resonator and gain medium.[574] In view of this feature, it has lower threshold and higher slope efficiency than the fiber-/solid-state lasers. Similarly, it has two modes of operation: continuous wave and pulse. In order to achieve pulse operation, it has been a dream of researchers in the field of waveguide lasers to find suitable SAs, and layered materials just meet this demand.

In 2013, the researchers fabricated a graphene SAM and applied it to a single-chip waveguide laser to obtain the mode-locked pulses with a pulse width of 1.06 ps and a repetition rate of 1.5 GHz,[575] as shown in **Figure 19**. Since then, waveguide lasers using layered materials such as graphene,[576–585] Bi_2Se_3,[586,587] TMDCs,[588–592] and BP[593,594] have been further developed, as shown in **Table 6**. It can be seen that, for Q-switching operation, the minimum pulse width and maximum pulse energy are 25.2 ns[576] and 310 nJ,[579] respectively. For example, the researchers achieved the Q-switching operation in a Nd:YAG ceramic channel waveguide laser based on Bi_2Se_3 SA,[586] as shown in Figure 19. These works not only greatly expand the application scope of layered materials, but also promote the rapid development of waveguide lasers.

As a kind of wavelength-scale nanolasers, disk lasers have attracted great attention in recent years due to its ultrafast modulation speed, low power consumption, and high density integration. For these lasers, finding a suitable material with excellent physical properties and good compatibility with nano-lasers has always been the pursuit of researchers in the field of disk lasers. The emergence of layered materials offers such a good opportunity. In 2012, the researchers integrated graphene into an electrically driven disk laser and realized the room-temperature operation with a threshold current of ≈300 μA,[595] as shown in **Figure 20**a,b. Since then, micro-disk/cavity lasers based on layered materials such as graphene,[596–598] TMDCs,[599–604] and perovskites[605–608] have developed rapidly. For example, the researchers achieved a 2D exciton visible-light laser by introducing the single-layer WS_2 into a disk cavity,[599] as illustrated in Figure 20c,d. In addition, another group of researchers also obtained continuous-wave lasing by introducing the monolayer $MoTe_2$ into the silicon nanowire cavity.[601] Notably, unlike the aforementioned pulsed lasers, in these micro-disk lasers, layered materials are used as gain media rather than as SAs. These efforts provide new possibilities for the design of micro-disk/cavity lasers and greatly promote the application of layered materials in nano-optics.

3.4. Recent Advances in Layered Material-Based Pulsed Lasers

In the above section, we briefly review the applications of common layered materials in mode-locked/Q-switched lasers. This field has been booming all along. Quite recently, other nanomaterials derived from them, such as heterostructures, quantum dots,

Table 5. Summary of Q-switched solid lasers with layered materials.

Device integration		Laser parameters						
2D Mater.	SA type	Gain medium	λ/nm	f_{rep}/kHz	τ/ns	P_{ave}/mW	E/nJ	Ref.
graphene	SAM	Nd:YAG	1064	660	161	105	159.2	[499]
Bi_2Se_3	SAM	Nd:$GdVO_4$	1063	547	666	32	58.5	[500]
MoS_2	SAM	Nd:$GdVO_4$	1060	90–732	0.97	–	0.31	[501]
		Nd:YGG	1420	40–77	0.729	–	0.67	
		Tm: Ho:YGG	2100	110–149	0.41	–	1.38	
GO	SAM	Nd:$GdVO_4$	1063	704	105	2300	3.2 µJ	[502]
Bi_2Se_3	SAM	Nd:Lu_2O_3	1077/1081	44.3–94.7	0.7–1.8	–	0.8342	[503]
graphene	SAM	Tm:YAG	2011	27.9	2080	38	1.74 µJ	[504]
graphene	SAM	Er:YAG	1645	35.6	2340	251	7.05 µJ	[505]
graphene	SAM	Nd:$GdVO_4$	1340	43	450	260	2.5 µJ	[506]
graphene	SAM	Nd:YAG	1123	22.9–46.8	875.7–1513.1	332	–	[507]
GO	SAM	Nd:YAG	1064	40–167	192–1200	2700	16.2 µJ	[508]
GO	SAM	Nd:YAG	1064	92.9	523	–	–	[509]
graphene	SAM	Ho:YAG	1907	28–64	2.6–9 µs	264	9.3 µJ	[510]
graphene	SAM	Tm:$KLu(WO_4)_2$	1948	81–190	285–1250	310	1.6 µJ	[511]
graphene	SAM	Nd,Mg:$LiTaO_3$	1082/1092	133	176	365	2.75 µJ	[512]
graphene	SAM	Yb:YAG	1032	168–285	228–522	185	650	[513]
graphene	SAM	Cr:ZnSe	2400	108–154	157	256	1.66 µJ	[514]
Bi_2Se_3	SAM	Nd:YVO_4	1066.6/1066.8	1–135	0.25–0.55	–	0.56	[516]
Bi_2Se_3	SAM	Nd:$LiYF_4$	1313.04	36.5–161.3	0.433–0.628	–	1.23	[517]
Bi_2Te_3	SAM	Yb^{3+}:$GdAl_3(BO_3)_4$	1043.7/1045.3/1046.2	30–110	0.37–2.5	–	511.7	[518]
SAM	SAM	Yb:$YCa_4O(BO_3)_3$	1033.5/1036.4	111	96	3850	9.63 µJ	[519]
Bi_2Se_3	SAM	Tm:LuAG	2027	30–118	0.62–1.9	–	18.4	[520]
Bi_2Te_3	SAM	Tm:LuAG	2021.7	145.5	233	1740	–	[521]
Bi_2Te_3	SAM	Tm:YAP	1980	108	0.238	–	1.25	[522]
/Graphene			2796	88	0.243	–	1.25	
Sb_2Te_3	SAM	Tm:$GdVO_4$	1913	200	223	700	3.5	[523]
Bi_2Te_3	SAM	Yb:$LuPO_4$	1014.5	1670	34	5020	3	[524]
Bi_2Se_3	SAM	Pr:YLF	721	60.5–185.2	368	460	170	[525]
			640	72.6–263.1	210	730	160	
			607 + 604	78.6–192.3	263	710	190	
MoS_2	SAM	Nd:$YAlO_3$	1079.57	32–232.5	0.227–0.58	–	1.11	[526]
MoS_2	SAM	Yb:LGGG	1025.2/1028.1	94–333	0.182	–	1.8	[527]
MoS_2	SAM	Yb:$LuPO_4$	1020.8	429	83	–	4.8	[528]
			1010.5	870	61		1.8	
MoS_2	SAM	Tm:CLNGG	1979	80–110	4.84–6	–	0.72	[529]
MoS_2	SAM	Tm:$GdVO_4$	1902	25.58–48.09	0.8–2	–	2.08	[530]
MoS_2	SAM	Er:Lu_2O_3	2840	48–121	0.335–1	–	8.5	[531]
MoS_2	SAM	Tm,Ho:YAP	2129	55	0.435	–	–	[532]
MoS_2	SAM+AOM	Nd:YVO_4	1064	10	0.00085	–	18.3	[533]
MoS_2	SAM	Ho:YAP	2119.5	91.2	1640	3300	23 100	[534]
WS_2	SAM	Nd: GYSGG	1061	70.7	591	367	1050	[535]
MoS_2	SAM	Nd:$GdVO_4$	1064.3	654.7–1030	269.2	1390	1350	[536]
WS_2	SAM	YVO_4/ Nd:YVO_4	1064	100–1030	0.056–0.24	–	1.6	[537]
WS_2	SAM	Nd:YVO_4	1064	55–135	2.3–4.94	–	0.145	[538]
WS_2	SAM	Nd:YVO_4	1064	55–135	2300–4940	19.6	145	[539]
WS_2	SAM+EOM	Nd:$Lu_{0.15}Y_{0.85}VO_4$	1064	519–731	0.467	–	341.5	[540]
WS_2	SAM+AOM	Nd:YVO_4	1064	10	0.81	233	–	[541]

(*Continued*)

Table 5. Continued.

Device integration		Laser parameters						
2D Mater.	SA type	Gain medium	λ/nm	f_{rep}/kHz	τ/ns	P_{ave}/mW	E/nJ	Ref.
WSe_2	SAM+EOM	Nd:YAG	946.3	125	10.8	–	2.63 mJ	[542]
MoS_2	SAM	Er:YAG	1645	46.6	1138	1080	23 080	[543]
WS_2	SAM	Er:Y_2O_3	2710-2740	29.4	720	–	7920	[544]
WSe_2	SAM	Tm,Ho:LLF	2950	89.3	571	147	1650	[545]
ReS_2	SAM	Er:YSGG	2796	47–126	0.324–1.1	–	0.825	[546]
ReS_2	SAM	Pr:YLF	640	520	160	52	–	[547]
		Nd:YAG	1064	644	139	120		
		Tm:YAP	1991	677	415	245		
$TiSe_2$	SAM	Ho,Pr:LLF	2950	98.8	160.5	130	1320	[548]
MoS_2/WS_2	SAM	Yb:$LuPO_4$	1002.3	1270	39	1570	1240	[549]
			1001.8	1430	34	2340	1640	
$MoTe_2$	SAM	Yb:YCOB	1035.5	704	52	1580	2250	[550]
WSe_2	SAM	Ho,Pr:LLF	2950	89.3	571	147	1650	[551]
$TiSe_2$	SAM	Nd:YVO_4	1000	152	483	410	–	[552]
		Nd:$GdVO_4$	1300	224	344	360		
		Tm:YAP	2000	84	350	990		
		Er:YSGG	2800	78	160	250		
$ReSe_2$	SAM	Er:YAP	2730/2800	244.6	202.8	–	2200	[553]
ReS_2	SAM	Er:SrF_2	2790	49	508	580	12 100	[555]
BP	SAM	Tm:YAG	2009	11.6	2900	38.5	3320	[556]
ReS_2	SAM	Yb:$ScBO_3$	1063.6	30.6	495.5	–	1400	[557]
$MoTe_2$	SAM	Tm:$CaYAlO_4$	1829–1941	70.9	690	750	10 580	[558]
$PtSe_2$	SAM	Tm:YAP	1987	58	244	–	24 300	[559]
Ta_2NiS_5	SAM	Tm:YAP	1910	50	313	1100	22 000	[560]
$SnSe_2$	SAM	Nd:YAG	1300	–	323	–	610	[561]
		Tm:YLF	1900		716		2070	
BP	SAM	Er:SrF_2	2790.1/2790.9	61–77.03	0.702–1.5		2.34	[562]
BP	SAM	Tm:YAP	1969/1979	41–81	0.181–0.72	–	39.5	[563]
BP	SAM	Tm:YAP	1988	11–19.25	1.78–4	–	7.84	[564]
BP	SAM	Yb:LuYAG	1030	63.9	1.73	–	0.09	[565]
		Tm:$CaYAlO_4$	1930	17.7	3.1		0.68	
		Er:Y_2O_3	2720	12.6	4.47		0.48	
BP	SAM	Ho^{3+},Pr^{3+}:$LiLuF_4$	2950	55–158.7	0.1943–0.58	–	2.4	[566]
BP	SAM	Yb:CYA	1046	87.7–113.6	0.62–1.2	–	0.3257	[567]
BP	SAM	Er:YAG	1645	40	3.2	–	2.15 μJ	[568]
					2.9		2.4 μJ	
BP	SAM	Cr:ZnSe	2411	126–176	189–396	36	205	[569]
BP	SAM	Nd:YVO_4	1064.4	–	2.86	–	166	[570]
WS_2					3.99		150	
MoS_2					5.4		365	
g-C_3N_4	SAM	Er:Lu_2O_3	2840	48–99	0.351–1.5	–	11.1	[571]
g-C_3N_4	SAM	Nd:LLF	1320.9	112–147	0.275–1.3	–	9.51	[572]

even MXene and metal-organic framework, have also been developed rapidly, thus enriching the family of layered materials.

Although layered material-based SAs have been used to realize pulsed lasers, there are some shortcomings in spectral range, modulation depth, absorption intensity, and carrier dynamics when used alone. To this end, the researchers have attempted to design novel heterostructures using different types of layered materials to use their common advantages.[609–613] In 2015, the researchers realized a mode-locked fiber laser based on graphene-Bi_2Te_3 heterostructure SA,[614] as shown in **Figure 21**a–c. Studies found that, the SA has faster carrier dynamics and greater modulation-depth than the layered material alone. Since then, mode-locked/Q-switched lasers using layered materials heterostructures have attracted wide attention.[615–622]

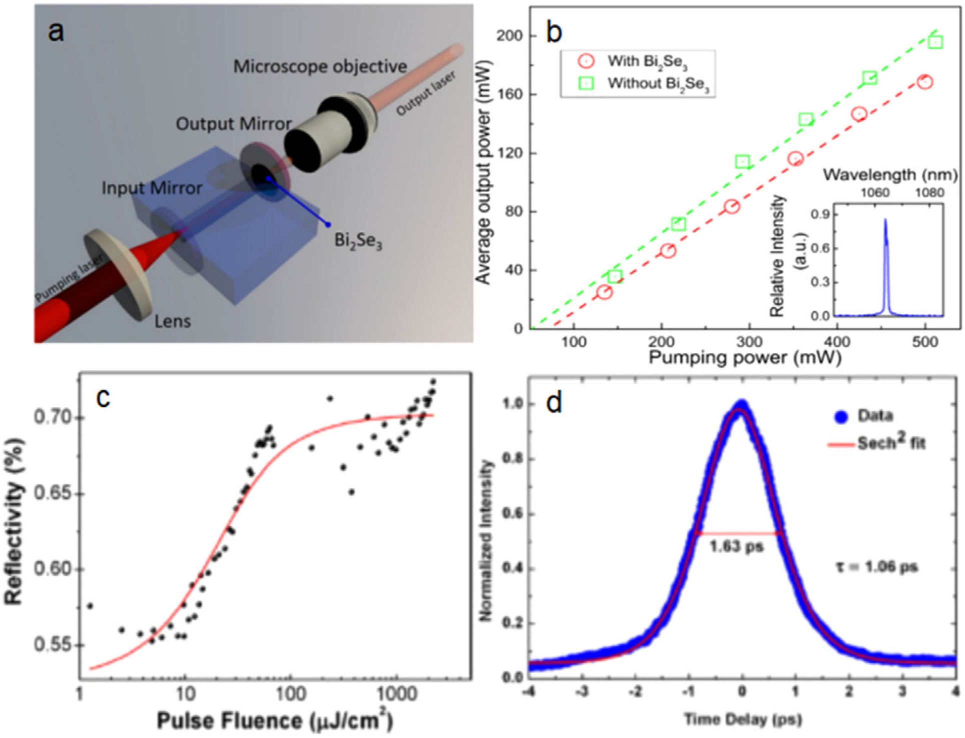


Figure 19. Mode-locked waveguide lasers based on layered materials SAs. a) Experimental setup of waveguide laser with Bi_2Se_3. b) Relation between the output power and the pumping power. Inset: the corresponding optical spectrum. c) Nonlinear reflectivity as a function of pulse fluence of graphene. d) AC trace of waveguide laser with graphene. Reproduced with permission.[586] Copyright 2015, Optical Society of America. Reproduced with permission.[575] Copyright 2013, Optical Society of America.

These efforts have injected new vitality into the research of ultrafast photonics.

Another kind of nanomaterials, quantum dots, has also attracted huge interests due to their unique quantum confinement and edge effect.[623] In 2016, the researchers fabricated a BP quantum dot SA and introduced it into a fiber laser to obtain the picosecond-level soliton pulses,[624] as shown in Figure 21d–f. This work promotes the application of quantum dots such as black phosphorus,[625–627] antimonene,[628] MXene,[629] perovskite,[630–632] carbon,[633] and lead sulfide[634–636] in mode-locked/Q-switched lasers. Currently, this direction is still developing rapidly.

Recent studies found that MXene,[637–642] antimonene,[643–648] bismuthene,[649–651] and metal-organic framework (MOF) [652] have also excellent saturable absorption property and can be used in mode-locked/Q-switched lasers. For example, the researchers achieved the soliton mode-locked fiber laser based on MXene,[638] antimonene,[644] and Ni-MOF,[652] respectively. These efforts not only promote the development of layered materials and ultrafast photonics, but also stimulate the researchers to explore other materials such as nonlayered nanosheets,[653,654] perovskites,[655–657] oxides,[658] nano-crystals,[659] and nano-composites.[660]

Interestingly, besides passive scheme, actively mode-locked/Q-switched lasers based on layered materials such as graphene and antimonene have been also proposed.[661–663] For these lasers, layered materials-based optical modulators play a key role, and we will review them in detail in the next section.

4. Novel Optical Devices using 2D Layered Materials

In the previous section, we briefly reviewed the applications of layered materials in pulsed lasers, in which the imaginary part of their third-order nonlinear susceptibility, namely the saturable absorption, plays a key role. In fact, their third-order nonlinearity has many other applications.

In recent years, optical modulators have attracted great interest due to their wide applications in optical interconnection, environmental monitoring, medicine, and security.[42,43] It is found that high nonlinearity plays an important role in modulator design. However, the widely used nonlinear optical materials, including lithium niobate, chalcogenide glass, and highly nonlinear optical fibers, have many limitations such as large absorption loss, large scattering loss, small phase shift, and vulnerability to laser damage in optical modulators. Fortunately, layered materials with large third-order nonlinearity and other excellent physical properties including strong light-material interaction, broadband optical response, fast relaxation, controllable optical properties, and high compatibility with other photonic structures, can make up for these shortcomings, thus bringing new opportunities for the development of optical modulators.

Up to now, besides the traditional modulators (electro-optic,[664–669] thermo-optic,[670] terahertz,[671–674] plasmonic[675–677]), all-optical modulators based on layered materials, including graphene,[678–681] phosphorene,[682] and

Table 6. Summary of Q-switched waveguide lasers with layered materials.

Device Integration		Laser Parameters						
2D Mater.	SA type	Gain medium	λ/nm	f_{rep}/kHz	τ/ns	P_{ave}/mW	E/nJ	Ref.
graphene	SAM	Yb: Y_2O_3	1030.8	1040	98	83	12.3	[575]
graphene	SAM	Nd:YVO_4	1064.4	16300	25.2	114	7.1	[576]
graphene	SAM	Nd:YAG	1064	2300	90	173	63	[577]
graphene	SAM	Tm:KLu$(WO_4)_2$	1948	190	285	310	1.6	[578]
$MoSe_2$	SAM	Nd:YAG	1064	0.995–3.334	80	–	36	[580]
WSe_2				0.781–2.938	52		19	
WSe_2	SAM	Yb:YSGG	1024.8	0.36	125	–	21.7	[581]
$SnSe_2$	SAM	Nd:YAG	1064	0.337–2.294	129	–	6.7–44.5	[582]
				0.438–1.865	183		6.5–43.1	
graphene	SAM	Ho: YAG	2091	5.9 GHz	170	–	–	[583]
graphene	SAM	Tm:KYW	1834.2	11–372	98	26.7	21	[585]
Bi_2Se_3	SAM	Nd:YAG	1064	2700–4700	46	168.6	31.3	[586]
$MoSe_2$	SAM	Nd:YAG	1064	995–3334	80	–	36	[588]
WSe_2				781–2938	52		19	
MoS_2	SAM	Nd:YAG	1064	510–1100	203	85.2	112	[589]
WS_2	SAM	Yb:YSGG	1024	360	125	7.8	–	[590]
$SnSe_2$	SAM	Nd:YAG	1064	337–2294	129	347	44.5	[591]
MoS_2	SAM	Tm:KLuW	1849.6	1390	88	247	18	[592]
WS_2	SAM	Nd:YAG	1064	3230–6100	24	144	–	[593]
BP				4300–5600	55	126		
graphene	SAM	Tm: ZBLAN	2000	1130	–	43.6	–	[594]
Bi_2Se_3				1250	44.9	29.1		
MoS_2				1040	68.3	4.3		
$MoSe_2$				1010	53	7.3		
WS_2				890	45.6	5.1		
WSe_2				–	46.4	7.4		
BP				810	41.2	7.1		
graphene	SAM	Yb: Y_2O_3	1030	1149	160	456	310	[579]

2D boron,[683] have also developed rapidly. In these devices, by tuning the carrier density via electrical or optical means that modify their physical properties, optical response of the layered materials can be instantly changed, thus making them versatile nanostructures for optical modulation. For example, the researchers realized the graphene-clad microfiber all-optical modulator with a response time of 2.2 ps (corresponding to a bandwidth of 200 GHz for Gaussian pulses),[678] as shown in **Figure 22**. In addition, the researchers from another group achieved the all-optical threshold and modulation device by using the strong Kerr effect of phosphorene.[682] These efforts have greatly promoted the development of all-optical photonics.

Besides optical modulators, other optical devices including optical polarizers,[44,684–686] optical filters,[687] optical isolators,[688] optical switchers,[689–692] optical parametric devices,[72–74,148–151,693–695] light-control-light device,[696–700] and even all-optical signal processing,[701–703] have been also studied. For example, the researchers developed a broadband polarizer based on graphene to support transverse mode surface wave transmission in visible and near-infrared bands,[44] as shown in **Figure 23**. In addition, another group of researchers also fabricated a graphene-deposited microfiber, which can generate the cascaded four-wave mixing effect,[72] as shown in **Figure 24**. Very recently, the researchers also explored the all-optical-signal-processing by using the topological insulator,[701] black phosphorus,[702] and antimonene,[703] respectively, as shown in **Figure 25**. It should be pointed out that the development of these all-optical devices has just started, and a lot of work needs to be further studied.

Interestingly, besides the optical devices mentioned above, the application of layered materials in optical sensors has also developed rapidly in recent years due to their abundant physical properties (e.g., humidity, temperature, mechanical, electrical, or optical absorption).[704–707] The emergence of layered materials provides new opportunities for sensor development. Up to now, various optical sensors using layered materials such as graphene, graphene oxide, reduced graphene oxide, and WS_2, have been obtained,[708–716] as shown in **Figure 26**. On the device level, these optical sensors with prisms, optical fibers, waveguides, optical flow control, and optical interferometer have the advantages of high-sensitivity, good stability, compact structure,

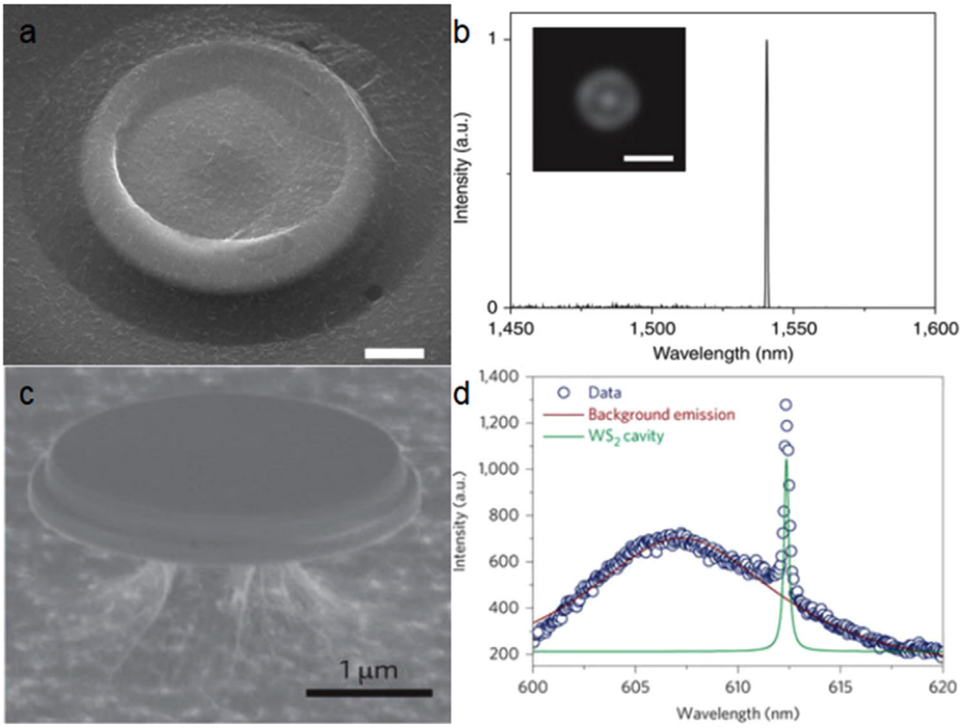


Figure 20. Microdisk lasers based on layered materials. a) SEM image of disk laser with the graphene sheets. b) Optical spectrum of single-mode lasing. Inset: image of lasing mode. c) SEM image of disk laser with WS_2 sheets. d) Optical spectrum of WS_2 cavity emission. Reproduced with permission.[595] Copyright 2012, Nature Publishing. Reproduced with permission.[599] Copyright 2015, Nature Publishing.

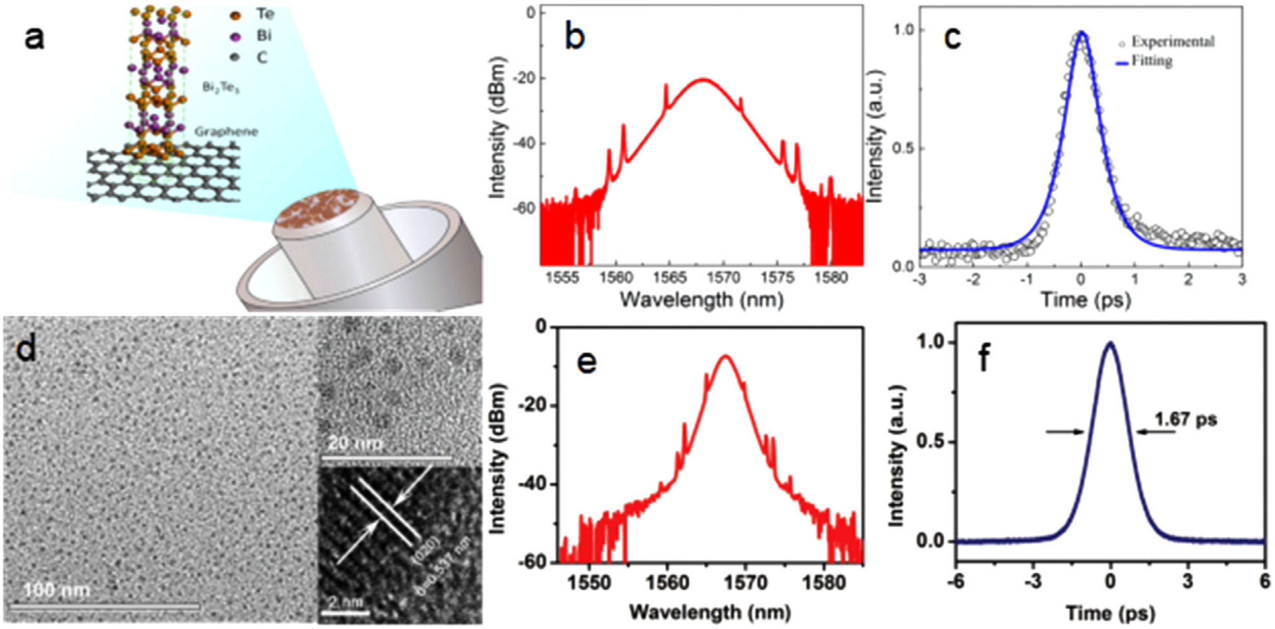


Figure 21. Mode-locked fiber lasers with layered materials-based heterostructures or quantum dots SAs. a) Schematic diagram of graphene-Bi_2Te_3 heterostructure, b,e) Optical spectra of mode-locked pulses and c,f) their corresponding AC traces. d) TEM, enlarged TEM and HRTEM images of black phosphorus quantum dots. Reproduced with permission.[614] Copyright 2016, American Chemical Society. Reproduced with permission.[624] Copyright 2016, Wiley Publishing.

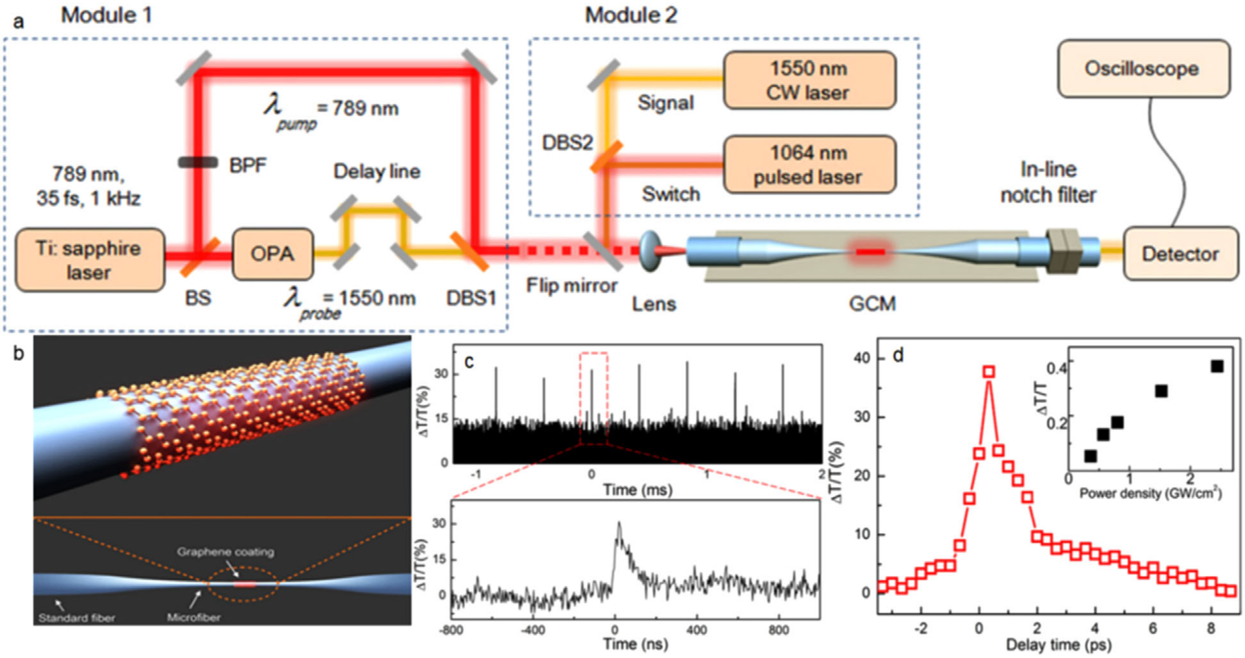


Figure 22. Demonstration of all-optical modulator with graphene. a) Experimental setup. b) Photograph of the microfiber wrapped with few-layer graphene, c) Pulse train. d) Relation between the differential transmittance of probe light and the pump-probe time delay. Inset: Relation between the modulation depth and the pump intensity. Reproduced with permission.[678] Copyright 2014, American Chemical Society.

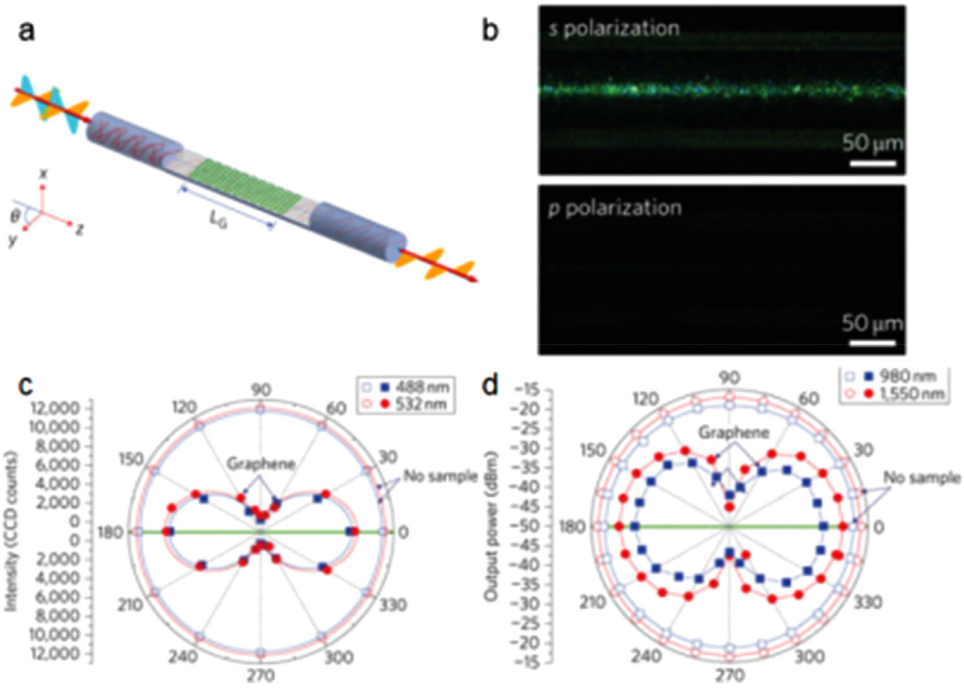


Figure 23. Broadband graphene polarizer. a) Schematic model of side-polished fiber-based graphene polarizer. L_G presents the length of graphene film. b) Polarized diagram of the laser beam along *s* and *p* polarization. c) Polar diagram at 488 and 532 nm, respectively. d) Polar diagram at 980 and 1550 nm, respectively. Reproduced with permission.[44] Copyright 2011, Nature Publishing.

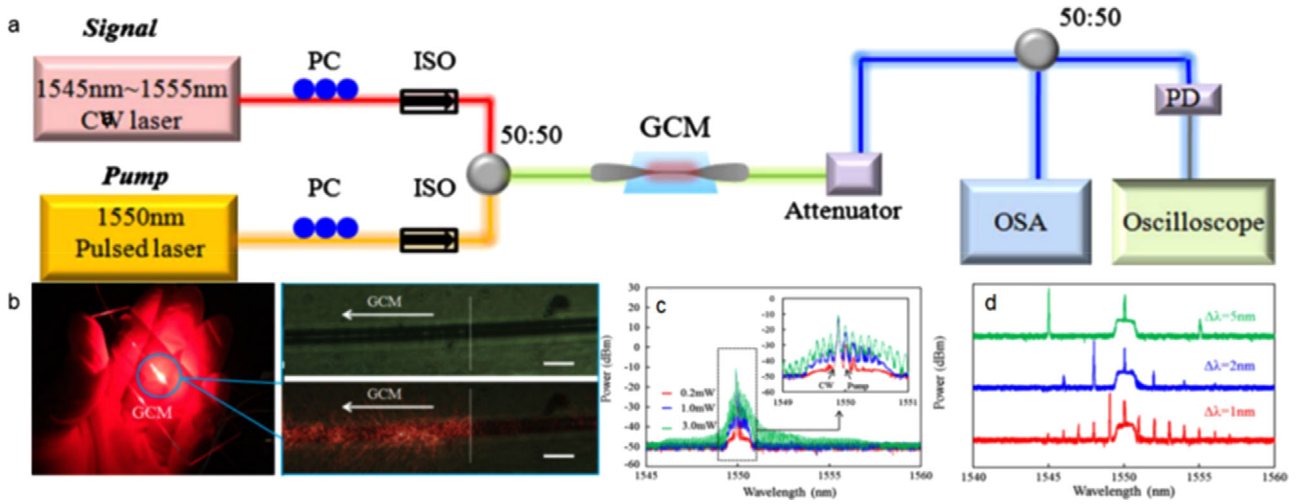


Figure 24. Cascaded FWM process generated from a graphene-deposited microfiber device. a) Experimental setup. b) Dark-field and optical photograph of graphene-based microfiber. c) Optical spectra at different pump powers. Inset: zoom-in diagram at 1549–1551 nm, d) Optical spectra of FWM process at different detuning wavelength. Reproduced with permission.[72] Copyright 2015, Optical Society of America.

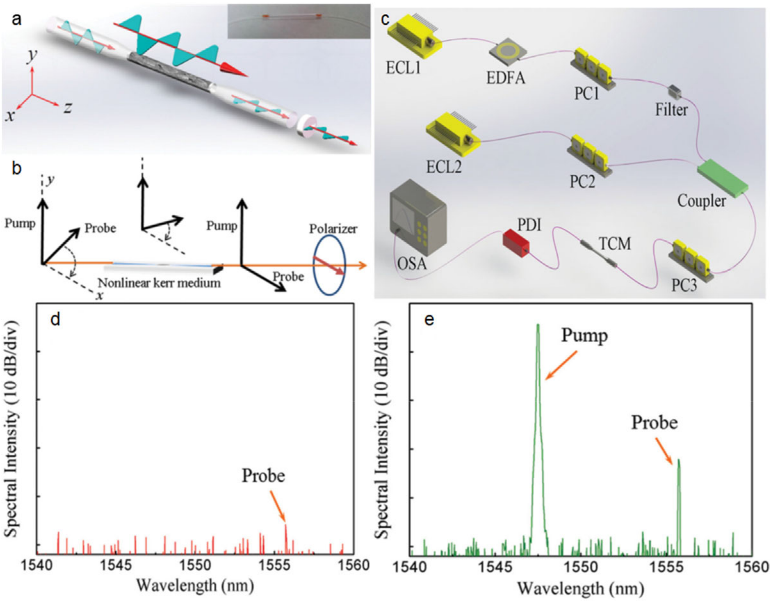


Figure 25. Demonstration of all-optical signal processing device with few-layer topological insulator-based microfiber. a) Principle of Kerr switcher, b,c) the corresponding diagram and experimental setup. d,e) Optical spectra of the light propagates after Kerr switcher and the polarizer with and without pump signal, respectively. Reproduced with permission.[701] Copyright 2015, Wiley Publishing.

light weight, and strong distributed sensing ability, and lab-on-fiber ability. These efforts greatly promote the application of layered materials in optical fiber sensors[717–723] and other sensors.[724–734]

5. Conclusions and Perspectives

Over the past decade, 2D layered materials-based pulsed lasers and novel optical devices have experienced rapid development, and a series of important results have been achieved, and they have gradually become a very hot research topic in the field of nonlinear photonics. This is mainly attributed to the maturity of layered materials preparation and integration technology and the in-depth study of their nonlinear optical properties, as well as the continuous progress of pulsed lasers and passive optical devices for decades. Their combination has spawned a new direction, namely, 2D material-based photonics. In this article, we briefly review the development of this emerging field. Fortunately, it is still developing rapidly.

As future perspective, several aspects may be considered to further develop the 2D materials-based photonics. First, the

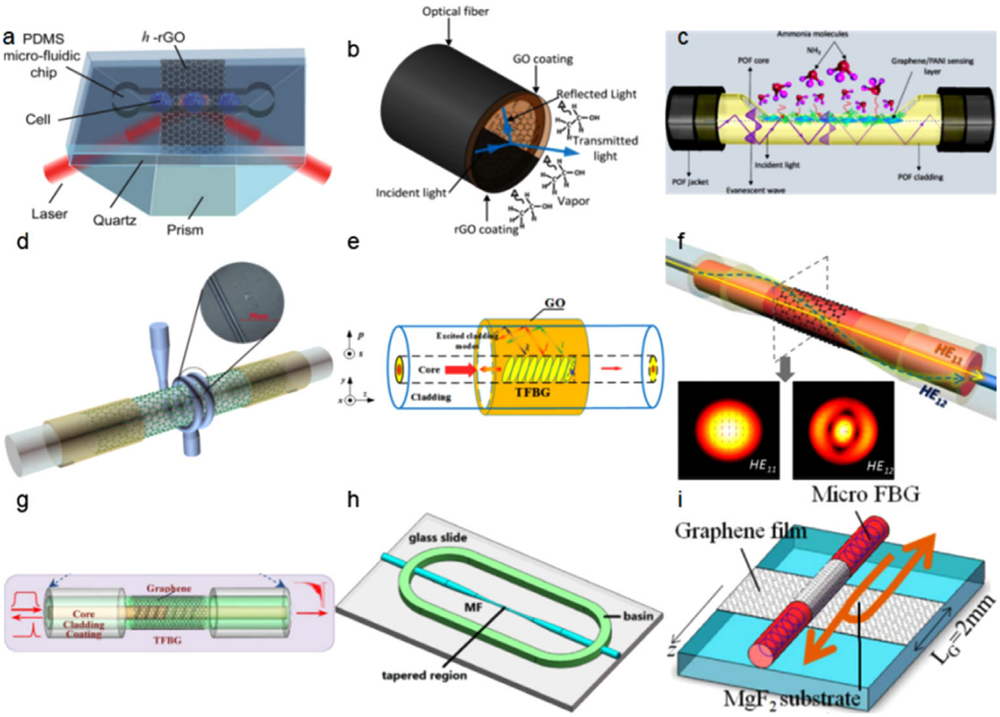


Figure 26. Schematic diagram of various optical sensors with layered materials. a) Optical single-cell sensor with graphene. Reproduced with permission.[708] Copyright 2014, American Chemical Society. b) Polymer optical fiber (POF) gas sensor with graphene oxide (GO) and reduced graphene oxide (rGO). Reproduced with permission.[709] Copyright 2013, Nature Publishing. c) Interaction mechanism between NH_3 gas molecules and graphene/polyaniline composite sensing layer. Reproduced with permission.[710] Copyright 2017, Optical Society of America. d) Electrical current sensor based on graphene. Inset: Photograph of microfiber. Reproduced with permission.[711] Copyright 2015, AIP Publishing. e) Humidity sensor with the tilted fiber Bragg grating (TFBG) and GO. Reproduced with permission.[712] Copyright 2015, AIP Publishing. f) Fiber-optic interferometer based on partially rGO and fluorescent resonance energy transfer. Reproduced with permission.[713] Copyright 2016, Nature Publishing. g) TFBG based on the graphene. Reproduced with permission.[714] Copyright 2015, Optical Society of America. h) WS_2 wrapped on microfiber for enhancing humidity sensing. Reproduced with permission.[715] Copyright 2015, Optical Society of America. i) Gas sensor with graphene micro-FBG. Reproduced with permission.[716] Copyright 2014, Optical Society of America.

performance of these pulsed lasers and novel optical devices will continue to improve. For example, mode-locked/Q-switched lasers in the ultraviolet and 2-4 μm wavebands will be deeply studied. In addition, versatile inkjet-printed lasers based on 2D crystals or heterostructures can be also realized. Second, more abundant nonlinear optical phenomenon, including multi-soliton molecules and rogue waves, may be discovered in the fiber lasers based on 2D materials. Third, more novel 2D materials will be synthesized, and exploring their nonlinear optical properties and related device applications will be the main topics in the future. We believe that 2D materials-based photonics will gradually develop from laboratory research to practical industrial applications, which will give an innovation in the fields of optics, medicine, biology, and energy.

Acknowledgements

B.G. and Q.-l.X. contributed equally to this work. This project was funded by Heilongjiang Natural Science Foundation (JJ2019LH1509), National Natural Science Foundation (NSFC) (61961136001, 61705136), Natural Science Foundation of SZU (No. 2019016, 860-000002110435), and 111 Project of Harbin Engineering University (B13015).

Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D materials, fiber laser, nonlinear optics, optical modulator, Q-switched laser, solid-state laser, ultrafast laser

Received: November 28, 2018
Revised: September 25, 2019

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