Review

Huanhuan Liu, Zilong Li, Ye Yu, Jincan Lin, Shuaishuai Liu, Fufei Pang* and Tingyun Wang

Nonlinear optical properties of anisotropic two-dimensional layered materials for ultrafast photonics

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Abstract: The discovery of graphene has intrigued the significant interest in exploring and developing the two-dimensional layered materials (2DLMs) for the photonics application in recent years. Unlike the isotropic graphene, a number of 2DLMs possess the in-plane anisotropic crystal structure with low symmetry, enabling a new degree of freedom for achieving the novel polarization-dependent and versatile ultrafast photonic devices. In this review article, we focus on the typical anisotropic 2DLMs including BP, ReS₂, ReSe₂, SnS, and SnSe and summarize the recent development of these anisotropic 2DLMs in the pulsed laser and the optical switch applications. First, we introduce the fabrication methods as well as the material characterization of the anisotropic 2DLMs by analyzing the polarized Raman configuration. Second, we discuss the anisotropic nonlinear optical properties of the anisotropic 2DLMs and concentrate on the anisotropic nonlinear absorption response. Next, we sum up state of the art of the anisotropic 2DLMs in the application of pulse lasers and optical switches. This review ends with perspectives on the challenge and outlook of the anisotropic 2DLMs for ultrafast photonics applications.

Keywords: anisotropic two-dimensional layered materials; nonlinear optical properties; saturable absorption; Q-switched laser; mode-locked laser.

1 Introduction

The discovery of graphene marks an important milestone and has opened a new era in the exploration and development of the two-dimensional layered materials (2DLMs) in recent years. In addition to graphene, various 2D materials have attracted much attention including topological insulators [1–7], transition metal dichalcogenides (TMDCs) [8–18], black phosphorus (BP) [19–31], MXenes [32–39], IV–VI chalcogenides [40–44], group VA material [45–47], metal phosphorus trichalcogenides (MPTs) [48], 2D metal–organic frameworks (2DMOFs) [49], 2D metal–halide perovskite [50], 2D nonlayered materials [51–54], and so on. Graphene has zero bandgap, whereas TMDCs have the bandgap with a range of 1–2.5 eV [55–59]. Black phosphorus has a direct bandgap between 0.3 and 1.5 eV, depending on material thickness, filling the space of bandgap between graphene and TMDCs. For 2DLMs, atoms are strongly bonded within the same plane but weakly attached to the above and below layers by van der Waals forces [60, 61]. The weak interlayer interaction leads to the feasible extraction of monolayer or a few layers of atoms. Recently, the research of 2D materials has made a rapid progress in the optoelectronic and photonic applications including photodetectors [62–75], optical modulators [76–78], optical switches [79–82], pulsed lasers [83–89], and so on. Such 2DLMs could enable high-performance nanophotonic devices equipped with the highly integrated, miniaturized, and portable features [90, 91].

When an incident light interacts with 2DLMs, either the intensity or the phase of incident light could be altered because of the linear and nonlinear optical responses of 2DLMs [92]. Indeed, the polarization is also an important degree of freedom for the light to be controlled. As the example shown in Figure 1A, it is much more attractive

***Corresponding author: Fufei Pang**, Key laboratory of Specialty Fiber Optics and Optical Access Networks, Joint International Research Laboratory of Specialty Fiber Optics and Advanced Communication, Shanghai Institute for Advanced Communication and Data Science, Shanghai University, 99 Shangda Road, Shanghai 200444, China, e-mail: ffpang@shu.edu.cn.
<https://orcid.org/0000-0002-8925-6947>

Huanhuan Liu, Zilong Li, Ye Yu, Jincan Lin, Shuaishuai Liu and Tingyun Wang: Key laboratory of Specialty Fiber Optics and Optical Access Networks, Joint International Research Laboratory of Specialty Fiber Optics and Advanced Communication, Shanghai Institute for Advanced Communication and Data Science, Shanghai University, 99 Shangda Road, Shanghai 200444, China

Figure 1: Anisotropic 2DLMs for ultrafast application.

(A) Schematic diagram of polarization-dependent pulse shaping by anisotropic 2DLMs. The crystal structure of typical anisotropic 2DLMs. (B) Orthorhombic black phosphorus (BP). Reproduced with permission from Ling et al. [93]. (C) Triclinic ReSe_2 . Reproduced with permission from Meng et al. [94]. (D) Triclinic ReSe_2 . Reproduced with permission from Echeverry and Gerber [95]. (E) Orthorhombic SnSe. Reproduced with permission from Ye et al. [40]. (F) Orthorhombic SnS. Reproduced with permission from Tian et al. [96].

to achieve the polarization-dependent pulse shaping by 2DLMs. Unlike the in-plane isotropic graphene, a number of 2DLMs possess the in-plane anisotropic crystal structure with low symmetry. For example, because of the different bond angles, BP exhibits the strong in-plane anisotropy between the armchair and the zigzag directions [93, 97–99]. Figure 1B plots schematically the bulk BP with the orthorhombic crystal structure, showing that each layer of BP has puckered lattice structure. Other emerging anisotropic 2DLMs contain ReS_2 [94, 95, 100, 101], ReSe_2 [95, 102], SnS [96, 103], SnSe [104, 105], and so on. As shown in Figure 1C and D, ReSe_2 and ReS_2 have a distorted 1T structure with triclinic symmetry, forming Re chains aligned along the b-axis. Similar to the orthorhombic crystal structure of BP, SnSe and SnS consist of puckered honeycomb layered crystal structure as shown in Figure 1E and F, and thus they are named as the BP analogy materials. Because of the anisotropic feature, the crystal orientation-dependent carrier mobility [106], optical absorption [94, 105, 107], and even valence bands [108] of these 2DLMs are found to be anisotropic. Such unique advantage of anisotropic feature brings a new degree of freedom

for exploring the 2DLMs to be novel polarization-dependent photonic devices that are impossible with the in-plane isotropic 2DLMs.

Currently, the nonlinear optical properties of the anisotropic 2DLMs have been studied, exhibiting the polarization dependence. Wu et al. [109] have experimentally investigated the power, polarization, and thickness dependence of the third-harmonic generation in the multilayer BP. The anisotropic optical properties of the layered ReS_2 and ReSe_2 have been theoretically investigated [95], and its polarization-dependent second- and third-harmonic generations have been demonstrated [100, 110]. Moreover, a number of anisotropic 2DLMs have been found with the polarization-dependent saturable absorption (SA) properties, such as BP [107], SnSe [105], SnS [111], ReS_2 [94], ReSe_2 [95], and so on. However, the review of the polarization-dependent optical properties of 2DLMs for the ultrafast photonic applications has rarely been reported.

Herein, we focus on the polarization-dependent optical properties of anisotropic 2DLMs and present the recent development of anisotropic 2DLMs for the pulsed

lasers and optical switches application. For different crystal structures, the representative anisotropic 2DLs attempted in ultrafast photonics application have been summarized: orthorhombic BP, orthorhombic SnSe and SnS, triclinic ReS₂, and ReSe₂. In the first part, the fabrication methods and the material characterization of the anisotropic 2DLs by the polarized Raman configuration are discussed. In the second part, we summarize the anisotropic nonlinear optical properties of the anisotropic 2DLs and focus on the anisotropic nonlinear absorption response. In the third part, the recent progress of the photonic application of the anisotropic 2DLs in pulse lasers and optical switches is included. This review offers outlook and perspectives for the potential applications of the anisotropic 2DLs. It is expected that this review can stimulate further interest in exploring the potential of anisotropic 2DLs for ultrafast photonics applications.

2 The preparation and characterization

2.1 Preparation of anisotropic 2DLs

The difference between the anisotropic and the isotropic 2DLs lies mainly in the in-plane structure and properties. Because the layers of 2DLs are stacked by van der Waals force, the fabrication methods of the anisotropic 2DLs are similar to that of the isotropic 2DLs. The typical fabrication methods are mainly classified into two schemes: the top-down method and the bottom-up method. The former commonly includes mechanical exfoliation (ME), liquid-phase exfoliation (LPE), and lithium ion intercalation exfoliation (LIE); the latter commonly includes chemical vapor deposition (CVD), physical vapor transportation (PVT), and so on. The key procedures of the above methods are briefly introduced as follows.

2.1.1 Top-down methods

By overcoming the van der Waals force between the layers, it is possible to exfoliate the monolayer or multilayer 2DLs from the bulk crystal. Among the top-down methods, the ME method is relatively convenient to achieve monolayer or multilayer samples, because it only requires tape to exfoliate bulk crystals repeatedly. Moreover, the ME method allows for clean surface and high quality, which benefits for the material investigation as well the

as the device application. For example, two high-quality BP flakes with different thicknesses of ~15 and 25 layers are prepared by the ME method and further explored as optical saturable absorbers in the fiber laser application [112]. However, the sample fabrication efficiency by the ME method is low, and the control of sample thickness is difficult, limiting the application of ME method in large-scale industrialization [61, 113].

Different from the ME method, both LPE and LIE methods need solution processing. The LPE is a pure physical process, requiring the dispersion of the crystal powder through a liquid-phase dispersion solvent. Common solvents include ethanol, NMP (*N*-methyl-2-pyrrolidone), DMF (dimethyl formamide), IPA (isopropyl alcohol), acetone, and surfactant sodium cholate. In order to break the van der Waals force between the layers of 2DLs, the strong ultrasonic treatment is required to create microbubbles in the material. Then the centrifuging force is applied to separate the exfoliated materials from the residual unexfoliated material flakes by mass difference. For example, in virtue of the LPE method, Zhang et al. [114] have fabricated the high-quality BP nanosheets with ~5 nm (~8 layers) as saturable-absorber mirrors (SAMs). Even though the LPE method can prepare abundant samples with the atomic-layer thickness, the layer number of samples is not evenly distributed, and thus the sample quality is relatively poor. To some extent, the LIE method is similar to the LPE method. The LIE method also needs ultrasonication and centrifugation. However, the main difference is that the LIE method needs lithium ions as intercalation to expand the interlayer separation of the bulk material in solvent, which is widely used to prepare single-layer 2DLs [115, 116]. However, the LIE method usually causes defects such as structural changes in the sample, and sometimes it requires reprocessing to improve the quality of samples.

2.1.2 Bottom-up methods

The bottom-up methods can directly synthesize the molecular-level 2DLs, which are completely different from the top-down methods by breaking the van der Waals force between the layers of bulk materials. The CVD method is commonly used to fabricate high-quality 2DLs. The principle is to use redox between reactants to generate solid depositions to form thin films. During the CVD process, the temperature, the air pressure, and the size of air flow all influence the formation quality. The film-formation status can be controlled by changing these parameters. As an example, the large-scale ReS₂

with the thickness of ~ 2.2 nm and ReSe_2 with the thickness of ~ 3.7 nm can be directly synthesized on a sapphire substrate by the CVD method [117]. Generally, the quality of 2DLMs prepared by the CVD method is high, but the corresponding cost and complexity is also high.

Unlike the CVD method, the PVT method does not need to undergo the redox process but the physical processes such as the evaporation and deposition. The PVT method is suitable for preparing the materials such as transition metal sulfides and metals with lower sublimation temperatures. For example, Tian et al. [96] have synthesized the SnS film with the thickness of 6 nm (~ 10 layers) by PVT method. Because the accessible 2DLMs are the base for any further device application, it is hoped that the advanced fabrication method can be developed to prepare the anisotropic 2DLMs with high quality, large scale, and low cost.

2.2 Characterization of anisotropic 2DLMs

In order to characterize the crystal orientation and the anisotropy of 2DLMs, the high-resolution transmission microscopy (HRTEM) images and the diffraction patterns are widely utilized [93, 102, 103]. Besides the optical image, a powerful tool is the angle-resolved polarized Raman spectroscopy.

2.2.1 Raman spectrum

By taking the BP crystal as an example, in theory, the Raman tensors for the A_g and B_{2g} modes can be given by [118, 119]

$$R(A_g) = \begin{pmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{pmatrix} \quad (1)$$

$$R(B_{2g}) = \begin{pmatrix} 0 & 0 & f \\ 0 & 0 & 0 \\ f & 0 & 0 \end{pmatrix} \quad (2)$$

in which the elements a , b , c , and f are complex values, determined by the light absorption of material. The intensity of the Raman modes can be calculated by

$$I = |\hat{e}_i^T R \hat{e}_s|^2 \propto \left| (\sin\theta, 0, \cos\theta) R \begin{pmatrix} \sin\gamma \\ 0 \\ \cos\gamma \end{pmatrix} \right|^2 \quad (3)$$

where $\hat{e}_s$ and $\hat{e}_i$ refer to the polarization vectors of the scattered and the incident light, respectively. θ is the angle between the incident polarization and the zigzag direction; and γ is the angle between the scattered light polarization and the zigzag orientation.

For the parallel backscattering configuration, $\theta = \gamma$, so

$$I_{Ag}^{//} = |a|^2 \left[\left(\sin^2\theta + \left| \frac{c}{a} \right| \cos\Phi_{ca} \cos^2\theta \right)^2 + \left| \frac{c}{a} \right|^2 \sin^2\Phi_{ca} \cos^4\theta \right] \quad (4)$$

where Φ_{ca} refers to the phase difference between the complex Raman tensor elements c and a . From (4), we can see that the polarized Raman intensity profile is a function of $|c/a|$ and Φ_{ca} . Whether the axis is parallel to the zigzag direction or the armchair direction is determined by $|c/a| > 1$ or $|c/a| < 1$. Typically, the measured angle-resolved intensity of vibrational mode is plot in the polar coordination, and thus the twofold or the fourfold rotational symmetry might be observed.

In the experimental measurement, the angle-resolved polarized Raman spectroscopy not only relates to the anisotropy of 2DLMs, but also the measurement configuration. According to the angle between the polarization of incident and scattered light, there are three different configurations: only the incident light (P_i); the incident light is parallel to the scattered light ($P_i // P_s$), and the incident light is perpendicular to the scattered light ($P_i \perp P_s$). Here, P_i and P_s represent the polarizations of incident light and scattered light, respectively.

The polarized Raman spectra of BP samples under the parallel configuration (the case of $P_i // P_s$) at 633 nm are shown in Figure 2A, with the angle of 0° – 180° [93]. Here, by taking the HRTEM images, the zigzag direction of BP is figured out paralleling to the incident light polarization of 0° . Three typical Raman peaks corresponding to A_g^1 , B_{2g} , and A_g^2 modes are observed at 361, 438, and 466 cm^{-1} , respectively. The minimum intensity of B_{2g} mode shows a change period of 90° , in which the angles at 0° and 90° are aligned with the armchair and the zigzag directions. From the obtained results, for a given thickness of BP sample, we can use the angle-resolved polarized Raman spectroscopy to figure out the crystal orientation.

The polarized Raman spectra of a 328-nm-thick ReS_2 sample under the parallel configuration ($P_i // P_s$) at 532 nm are plotted in Figure 2B, showing that the V-mode at ~ 212 cm^{-1} changes periodically from 0° to 360° [94]. By extracting the angular-dependent Raman intensity at V-mode, we can obtain the twofold symmetry in polar plots. The direction of the maximum V mode intensity at 0° is aligned with the b-axis of exfoliated ReS_2 sample.

Figure 2: The angle-resolved polarized Raman spectroscopy of the anisotropic 2DLMs.

(A) BP flake (the insert shows the corresponding polar plots). Reproduced with permission from Ref. Ling et al. [93]. (B) ReSe_2 : the angle-resolved polarized Raman spectroscopy with V-mode highlighted in orange and corresponding polar plot of the Raman intensity of V-mode. Reproduced with permission from Meng et al. [94]. (C) ReSe_2 : polar plots of the extracted angular-dependent intensity at Raman peak 238 cm^{-1} under “only incident,” “parallel,” and “perpendicular” configurations. Reproduced with permission from Zhang et al. [102]. (D) SnS : Raman spectra of 14.6-nm SnS flake with 532-nm incident laser under parallel and perpendicular polarization configurations (left figure); the polar plots of intensity of A_g and B_{3g} under parallel and perpendicular polarization (right figure), respectively. Reproduced with permission from Xia et al. [120]. (E) SnSe : the angle-resolved polarized Raman spectroscopy (left figure); angular dependence of the Raman intensities of $A_g(1)$, B_{3g} , $A_g(2)$, and $A_g(3)$ modes (right figure). Reproduced with permission from Zhang et al. [105].

The extracted angle-resolved Raman intensity of 7-nm ReSe_2 at Raman peak of 238 cm^{-1} under three configurations of P_i , $P_i // P_s$, $P_i \perp P_s$ is shown in Figure 2C [102]. Under the first configuration, the polar plot presents twofold symmetry. Under the configurations of $P_i // P_s$ and $P_i \perp P_s$, it is observed that the two corresponding polar plots present the fourlobed shape, which are orthogonal to each other and decomposed of the twofold symmetry polar plot.

The polarized Raman spectra of SnS with the two configurations of $P_i // P_s$ and $P_i \perp P_s$ are shown in Figure 2D

[120]. There are two typical Raman modes: A_g mode (190.7 cm^{-1}) and B_{3g} mode (162.9 cm^{-1}). The extracted Raman intensities of A_g and B_{3g} modes are obviously angle-dependent. The intensities of B_{3g} mode become zero along the armchair and the zigzag directions under $P_i // P_s$, but achieve the maximum under $P_i \perp P_s$.

Similarly, the two configurations of $P_i // P_s$ and $P_i \perp P_s$ of angle-resolved polarized Raman spectroscopy are explored to characterize SnSe [105]. As shown in Figure 2E, the polarized Raman spectra present the four

typical Raman peaks corresponding to $A_g(1)$, B_{3g} , $A_g(2)$, and $A_g(3)$ modes at 70.9, 110.1, 134, and 151.5 cm^{-1} , respectively. The A_g mode shows a clear 180° periodic variation against angle. On the contrary, a period of 90° is found for the B_{3g} mode with a fourfold symmetry. Under the configuration of $P_i // P_s$, when the incident light polarization is parallel to the zigzag direction of SnSe, the maximum Raman intensity of the $A_g(3)$ mode always takes place regardless of the excitation wavelength or the sample thickness. Note that the angle-resolved polarized Raman spectra might be affected by the thickness of 2DLMs, so that one needs to combine both the angle-resolved polarized Raman spectroscopy and optical images to identify the crystal orientation and properties.

2.2.2 Polarized absorption

When the vector light irradiates an anisotropic material, the optical absorption depends on the structure of the material [101, 102, 105, 121]. In theory, based on Fermi's Golden rule, the optical absorption probability can be modeled by the absorption coefficient α [93, 105]:

$$\alpha(E_L) \propto \sum_{f,i} |\langle f | H_{op} | i \rangle|^2 \delta(E_f - E_i - E_L) \quad (5)$$

where E_L refers to the incident photon energy; E_f and E_i are the energies of the final and the initial states. $\langle f | H_{op} | i \rangle$ represents the electron-photon matrix element of perturbation H_{op} between the final and initial states. The element of $\langle f | H_{op} | i \rangle$ determines an optical transition from state i to f , which can be estimated within the dipole approximation:

$$\langle f | H_{op} | i \rangle \propto P \cdot D_{fi} \quad (6)$$

where P refers to the polarization vector of the incident photon; D_{fi} is dipole vector. From (5) and (6), we can see clearly that the absorption coefficient α is polarization-dependent.

In the optical measurement, the polarized optical absorption spectra of the anisotropic 2DLMs are believed to be thickness-dependent. Figure 3A shows the transmittance of two BP samples with the thicknesses of 25 and 1100 nm at 1.55 μm [121]. When the polarization changes, both 25- and 1100-nm BP samples exhibit a period change in transmission, showing a period of 180° . On the one hand, the transmission of the 25-nm BP sample is always higher than that of the 1100-nm BP sample, because the former has the shorter interaction length with the less absorption than that the latter does. On the other hand, the

transmission modulation of 1100-nm BP is $\sim 29.6\%$, which is much higher than that of 25-nm BP with a modulation of $\sim 4.8\%$. The estimated sixfold ratio clearly shows that the thickness of BP plays an important role in the anisotropic optical absorption. The results indicate that an increase in the layer thickness can enhance the anisotropic strength. But in turn, the high transmission is sacrificed. One needs to balance between the transmission and the anisotropic strength in the practical application.

ReS_2 also exhibits the anisotropic absorption due to the distorted 1T crystal structure. The angle-resolved linear absorption spectra of the seven- to eight-layer ReS_2 samples are plotted in Figure 3B [101]. Here, the angle represents the polarization direction with respect to the b -axis. Two resonance bands at ~ 1.53 eV (~ 810.5 nm) and ~ 1.56 eV (~ 794.9 nm) labeled as X_1 and X_2 are polarization-dependent. The two resonance peaks originate from the two lowest, energetically nondegenerate direct excitation states near Γ point [101, 122]. Notably, the absorption peaks arise at different polarization directions: $\sim 19^\circ$ for 810.5 nm and $\sim 87^\circ$ for 794.9 nm. The results indicate the feasibility of switching the individual excitation states by adjusting the light polarization.

Different from ReS_2 presenting the switchable resonant absorption peaks under different polarization, ReSe_2 demonstrates a relatively broadband polarized absorption. As shown in Figure 3C, both the theoretical and the experimental investigations reveal the polarized absorption of ReSe_2 over a broadband waveband [102], especially in a range of 1.7–2.4 eV. By computing the dielectric function, the calculated optical absorption spectra indicate a linear dichroism behavior in ReSe_2 [122, 123]. In the experimental investigation, beyond the photon energy of 1.32 eV, the transmission spectra present a large decrease with incident light polarization paralleling to the b -axis of the ReSe_2 . The results show the great potential of ReSe_2 to be broadband polarized devices in the infrared waveband.

The polarization-dependent absorption spectra of SnSe flake sample are shown in Figure 3D [105]. By changing the polarization direction, the absorption rate at 850 nm can be reduced from 33.7% to 30.8%, whereas the absorption rate at 800 nm can be reduced from 35% to 28%. The polar plots of the linear absorption of SnSe flake against polarization at 800 and 850 nm are provided for comparison. The linear absorption reaches the maximum and the minimum along the armchair and the zigzag directions, respectively. Obviously, different anisotropies are observed at 800 and 850 nm against the polarization of the incident light. Because more energy bands contribute to different anisotropies, the polarization-dependent transmission possibility could vary with the photon energy [105].

Figure 3: The angle-resolved linear absorption spectra of the anisotropic 2DLMs.

(A) The transmittance of the BP films with the thickness of 25 nm (the blue curve) and 1100 nm (the black curve), showing the anisotropic absorption behaviors. Reproduced with permission from Li et al. [121]. (B) Left figure: the absorption spectra of seven- to eight-layer ReSe₂; right: the corresponding spectral intensities of X₁ (blue dots) and X₂ (red dots) in polar plot. Reproduced with permission from Sim et al. [101]. (C) The absorption spectra of bulk ReSe₂. Top figure: the theoretical calculation of angle-resolved optical absorption spectra of the bulk ReSe₂ (the insert shows the calculated optical absorption spectra of monolayer ReSe₂); bottom figure: the measured polarization-dependent optical absorption spectra of ReSe₂ (the insert shows the experimental transmission spectra in a wider range with unpolarized light). Reproduced with permission from Zhang et al. [102]. (D) Top figure: the polarization-dependent linear absorption spectra of SnSe flake sample; bottom figure: the polar plots of the linear absorption rate of SnSe flake sample at 800 and 850 nm, respectively. Reproduced with permission from Zhang et al. [105].

3 Anisotropic nonlinear optical properties

Under the high intensity of incident light, the nonlinear effects could take place. The optical response of an optical medium is typically determined by the polarization $P(t)$ in terms of the optical field $E(t)$ [124, 125]:

$$P(t) = \varepsilon_0 (\chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \dots) \quad (7)$$

where ε_0 is the vacuum permittivity; $\chi^{(n)}$ is the n th-order nonlinear optical susceptibility; and $\chi^{(1)}$ refers to the linear optical effects including the absorption and the refraction. When the optical light is intense or the nonlinear optical susceptibility is large, the higher order ($n > 2$) terms becomes significant. $\chi^{(2)}$ responds for the two-frequency effects including second-harmonic generation [41, 42, 110, 126–129], sum- and different- frequency generation [124, 125, 130, 131], the Pockels effect [124, 125], and optical

rectification [132]. However, the presence of second-order and other even-order nonlinear optical effects needs a material having no center of inversion symmetry [124, 125]. On the contrary, the third-order nonlinear effects can take place in all materials [43, 100, 126, 133, 134]. The $\chi^{(3)}$ responds for the three-frequency effects, such as third-harmonic generation and four-wave mixing. The real part of $\text{Re}(\chi^{(3)})$ represents the optical Kerr effect that is the intensity-dependent refractive index change, while the imaginary part of $\text{Im}(\chi^{(3)})$ refers to the nonlinear absorption under the strong incident light, known as SA. The mechanism of SA is related to the Pauli-blocking principle [135]. At high incident power, because of the state filling effect at conduction band minimum (CBM) or the deletion in valence band maximum (VBM), additional photons cannot induce additional carrier excitation, leading to a reduction in absorption.

Taking into account the SA, the absorption coefficient (α) of an optical medium can be modeled by [136–138]:

$$\alpha = \frac{\alpha_s}{1 + \frac{I}{I_s}} + \alpha_{ns} \quad (8)$$

where α_s and α_{ns} are the SA coefficient and the non-SA coefficient, respectively; I is the input light intensity; and I_s is the saturation intensity. Thus, the intensity-dependent transmittance T can be given as [136, 139]:

$$T = \exp \left(-\frac{\delta T}{1 + \frac{I}{I_s}} - \alpha_{ns} L \right) \quad (9)$$

where $\delta T = \alpha_s L$ is modulation depth.

3.1 Anisotropic nonlinear absorption response

Because of the reduced symmetry of anisotropic 2DLMs, not only the linear optical absorption but also the nonlinear absorption response is polarization-dependent. In order to investigate the impact of the polarization on the nonlinear absorption response, the polarization azimuth of the measurement beam irradiating on the anisotropic 2DLMs is adjusted. Figure 4A gives an example of the measured transmittance of the exfoliated BP samples, showing the nonlinear power-dependent transmission and the polarization-dependent SA [107]. The modulation depth varies in the range of 0.6%–4.6%. When the polarization is parallel to the y -axis (zigzag direction) of BP lattice structure, the highest transmittance is obtained. A roll-off effect is observed as the input fluence

increases, named as the reserved SA (RSA). Here, the RSA is explained to be originated from the two-photon absorption (TPA) [140, 141]. Generally, the TPA response is determined by the third-order nonlinear susceptibility term, which is typically observed at high peak power due to small TPA absorption cross-section [94]. When the TPA response exists, (8) can be modified by

$$\alpha = \alpha_{ns} + \frac{\alpha_s}{1 + \frac{I}{I_s}} + \beta I \quad (10)$$

in which β is the TPA coefficient.

The RSA response against polarization angles has been found in ReS_2 [94]. Instead of the power-dependent transmission, the absorption coefficients are plotted as a function of laser peak power. As shown in Figure 4B, at polarization angle of 100° , the absorption drops monotonically with laser peak power, indicating a strong SA. On the contrary, at 180° , the absorption starts to rise at low peak power, indicating the RSA effect. The originality of the RSA effect is complicated that may involve distinct mechanisms: the free carrier absorption (FCA), the excited-state absorption (ESA), and the TPA. The FCA belongs to an intraband absorption process, requiring preexisting electron at CBM or holes at VBM. The FCA needs the assistance of phonons or impurities to conserve the momentum. If the 2DLMs are undoped with negligible purities, the FCA can be safely omitted. To some extent, the ESA is similar to the TPA, but the ESA can occur along with the single-photon absorption, whereas the TPA usually does not. In Meng et al. [94], as the RSA is observed at low peak power, it is believed that the RSA originated from the ESA. Previous reports have suggested that such RSA effect can benefit for ultrafast high-repetition rate lasers [142]; thereby, the RSA allows anisotropic BP for femtosecond-GHz pulsed lasers.

Recently, researchers have investigated the polarization-dependent SA of SnSe at dual wavelength as shown in Figure 4C [105]. The SnSe samples exhibit different anisotropies at 800 and 850 nm. By fitting the transmission results according to (9), δT can be extracted and plotted in a polar diagram as shown in Figure 4C. Compared with the SA response at 850 nm, the anisotropic ratio of δT at 800 nm can be achieved to be 6.7 between the armchair and the zigzag directions. It is observed that the saturation intensity is also polarization-dependent at 800 and 850 nm. According to the optical transition selection rules, the polarization and the photon energy of the incident light can induce a variation to the permitted transition rate. The final excited states density is thus changed, leading to the polarization-dependent saturation intensity.

Figure 4: The measured power-dependent transmittance against different polarization.

(A) The exfoliated BP. Reproduced with permission from Sotor et al. [107]. (B) ReS_2 : the angle-resolved absorption coefficient against power and the involved mechanisms including the saturation absorption and excited saturation absorption. Reproduced with permission from Meng et al. [94]. (C) SnSe : the normalized transmission against intensity at 800 and 850 nm; the corresponding polar plots of the absolute modulation depth as well as saturation intensity in terms of polarization. Reproduced with permission from Zhang et al. [105].

3.2 Polarization-dependent transient absorption

In order to study the dynamical response of the anisotropic 2DLs, time-resolved pump-probe spectroscopy is widely used. The anisotropic response of 2DLs is related to the convolution of pump-probe pulse as well as the polarization of incident light. Figure 5A shows the polarization dependence of the carrier dynamic process of BP with thickness of 16 nm [98]. A 730-nm linear polarized laser pulse is used as pump to induce photocarriers in BP, with the pulse duration of 100 fs and the peak fluence of $10 \mu\text{J}/\text{cm}^2$. The 810-nm and 100-fs probe pulse arrives at different delay time with respect to the pump pulse. The differential reflection ($\Delta R/R_0$) of signal refers to the pump-induced relative change in the probe reflection, which is given by [105, 143]

$$\Delta R/R_0 = (R - R_0)/R_0 \quad (11)$$

where R_0 and R are the probe reflection of the sample modulated without and with the pump pulse, respectively. Generally, the biexponential function can be used to fit the decay of the signal:

$$\Delta R/R_0 = A_1 \exp(-t/\tau_1) + A_2 \exp(-t/\tau_2) \quad (12)$$

The short-time constant (τ_1) is related to the motion of carriers from the detection window to the band edge in the process of thermalization. The long-time constant (τ_2) is attributed to the carrier lifetime. As shown in Figure 5A, the $\Delta R/R_0$ achieves the maximum around zero probe delay. After time delay of ~ 20 ps, the carrier lifetime can be fitted with $\tau_2 = 100$ ps. The peak $\Delta R/R_0$ is found to be linearly proportional to the pump fluence.

Figure 5: Time-resolved differential reflection signal from BP and SnSe samples and its angular dependence.

(A) BP. Top figure: the measured differential reflection signals as a function of probe delay (the red line indicates the fitting with a time constant of ~ 100 ps, and the inset shows the peak signal against the pump influence); bottom figure: the peak differential reflection signals as a function of angle with the pump-probe polarization orientation of horizontal-vertical (H-V), horizontal-horizontal (H-H), and vertical-vertical (V-V), respectively. Reproduced with permission from He et al. [98]. (B) SnSe. The measured differential reflection signals as a function of probe delay at 800 and 850 nm; the corresponding polar plots of the peak differential reflection signals and carrier relaxation time. Reproduced with permission from Zhang et al. [105].

The bottom figure in Figure 5A shows the $\Delta R/R_0$ of BP sample under different pump-probe polarization configurations: horizontal-vertical (H-V), horizontal-horizontal (H-H), and vertical-vertical (V-V) directions [98]. The results not only indicate the anisotropic response of BP sample, but also enable us to figure out the crystalline direction. Figure 5B shows the polarization-dependent transient absorption of the anisotropic SnSe flake [105]. The polar plots of peak $\Delta R/R_0$ against polarization at both 800 and 850 nm are given. When the polarization direction of the pump is parallel to the armchair direction, obviously, the peak $\Delta R/R_0$ reaches the maximum value. Especially, with respect to the 800-nm probe pulse, by comparing the peak $\Delta R/R_0$ values, it is deduced that the pump-injected carrier density along the armchair direction is approximately 4.6 times that along the zigzag direction. However, the carrier relaxation time exhibits little anisotropy, which is 380 and 320 ps for the probe at 800 and 850 nm, respectively. Such anisotropic optical absorption as well as the isotropic relaxation time confirms the remarkable ultrafast, polarization-dependent properties of anisotropic 2DLMs, showing the great potential of the anisotropic 2DLMs for the functionally optical modulation.

In addition to BP and SnSe, the differential reflection signal of bulk ReSe_2 has been investigated [144], measured with a 620-nm pump and 820-nm probe pulses. The photocarrier light is obtained at approximately 80 ps and drops slightly by increasing the carrier lifetime.

By rotating the sample orientation, the peak signals take place when the long edge of the sample is either parallel or perpendicular to horizontal direction. This happens because the absorption coefficient of ReSe_2 is the largest when the Re chains are along the light polarization. An interesting phenomenon of fourfold symmetry in the polar plots is observed. The authors attribute the fourfold symmetry to the potential compensation between the maximum absorption coefficient and the minimum transient absorption response in the parallel and the vertical directions [144]. Similar to BP and SnSe, although the magnitude of the signal is dependent on angle, its time evolution is angle-independent.

The key parameters used to evaluate the anisotropic properties of 2DLMs are summarized in Table 1. By rotating the incident light polarization, the modulation depth, the saturation intensity, and the absorption coefficient of anisotropic 2DLMs vary. Even the switch from SA to RSA can be observed for BP and ReSe_2 by adjusting the polarization. The polarization-dependent nonlinear optical absorption of the anisotropic 2DLMs is totally different from that of the isotropic 2DLMs including graphene [146, 147], NiPS_3 [48], TiS_2 [17], antimonene [45, 47], bismuthene [46], and 2DMOFs [49], whose nonlinear properties are insensitive to the incident light polarization. Such polarization-dependent nonlinear optical absorption response of anisotropic 2DLMs allows for the pulse shaping with a new degree of freedom.

Table 1: Summary of the SA properties of anisotropic 2DLMs.

Anisotropic 2DLMs	Thickness	Fabrication method	Anisotropic properties of 2DLMs under different polarizations					SA to RSA	Peak $\Delta R/R_0$ ($\times 10^{-4}$)
			Modulation depth	Saturation intensity (GW/cm ²)	Absorption coefficient (cm ⁻¹)	Carrier lifetime			
BP	16 nm [98]	ME [98, 107]	0.6%–4.6% [107]	Varied [107]	–	100 ± 5 ps [98]	Y [107]	13–72 [98]	
	300 nm [107]								
ReS ₂	328 nm [94]	CVD [94, 117]	–	500 ± 71–182,000 ± 16,717 [94]	27,000 ± 75–30,200 ± 480 [94]	–	Y [94]	–	
ReSe ₂	2.2 nm [117]	ME [145]	–	–	–	–80 ps [144]	–	0.3–1.65 [144]	
	21 nm [145]								
SnSe	1.5 nm [144]	LPE [144]	2.4%–12.9% at 800 nm [105]	5.858–14.137 at 800 nm [105]	1383–7384 at 800 nm [105]	380 ps at 800 nm [105]	–	5.4–25 at 800 nm [105]	
	175 nm [105]	ME [105]							
	9.5 nm [40]	LPE [40]	15.4%–19.6% at 850 nm [105]	4.577–15.249 at 850 nm [105]	8784–111,129 at 850 nm [105]	320 ps at 850 nm [105]		17–27 at 850 nm [105]	
	6 nm [40]								
	2.5 nm [40]								

4 Ultrafast photonics applications

The application of 2DLMs in pulsed laser systems has become a hot topic in recent years because of the advantages of 2DLMs including the wide bandwidth operation, the fast carrier dynamics, compactness, and so on. [61, 91, 92, 105, 148, 149]. The pulse formation in laser systems incorporating 2DLM-based saturable absorber mainly involves two mechanisms: passive Q-switching and passive mode locking. For the passively Q-switched lasers, the cavity energy is constantly accumulated by pumping when the saturation of saturable absorber is not reached. Once the saturation of SA reaches, a giant pulse could be lasing at the threshold that the cavity gain is larger than the cavity loss. The separation between the consecutive pulses or the repetition rate of the Q-switched pulse train is mainly determined by the lifetime of gain medium and the pump power. Typically, the Q-switched fiber laser has the tunable repetition rate of kilohertz. Different from the Q-switched laser, the passively mode-locked laser produces pulses with a fixed repetition rate determined by the cavity length. When the initial pulses from amplified spontaneous emission pass through the saturable absorber, the wings of pulses with low intensity experience much higher loss than the peak part of pulses with high intensity does. Based on the power-dependent optical transmission of saturable absorber, the pulse is reshaped where the single-to-noise ratio can be improved. The circulated reshaped pulse is further amplified by the gain medium, and the peak power of the pulse is enhanced. After hundreds of round trips, stable pulse trains with high repetition rate (MHz to GHz), ultrashort pulse duration (ps to fs), and high peak power can be produced from the laser cavity under the balance among the laser gain, loss, dispersion, and nonlinearity [91, 150]. By managing the net cavity dispersion, the traditional soliton, the stretched pulses, similariton, and dissipative soliton can be formed in the mode-locked lasers. The pulsed light sources have shown great potential in micromachining, laser physics, medicine, and so on [8, 9, 52, 85, 147, 151–164]. Compared with the isotropic 2DLMs, the anisotropic 2DLMs have a new degree of freedom to control the light polarization. Next, we introduce the application of the anisotropic 2DLMs in the Q-switched and the mode-locked lasers. Both the fiber lasers and the solid-state lasers are introduced. In addition, the application of anisotropic 2DLMs in optical switches has been introduced, showing the feasibility of the anisotropic 2DLMs as optical modulators.

4.1 Laser application

The incorporation of the anisotropic 2DLMs in fiber laser is expected to produce polarized pulses. In order to characterize the polarization of laser output, the degree of polarization (DOP) of laser output is given by:

$$\text{DOP} = (P_{\max} - P_{\min}) / (P_{\max} + P_{\min}) \quad (13)$$

where $P_{\min}$ and $P_{\max}$ refer to the measured minimum and maximum power, respectively.

Taking BP as an example, Li et al. [121] have reported the BP-based Q-switched and mode-locked lasers. The applied BP sample has a thickness of 1100 nm, showing transmission difference of 29.6% between the zigzag and armchair directions. The laser setup is shown in Figure 6A, which consists of 1-m-long erbium-doped fiber (EDF) pumped by a 980-nm laser diode through a wavelength division multiplexer. When the pump power is 23 mW, a stable pulse train can be achieved as shown in Figure 6B. When the pump power ranges from 23 to 55 mW, the repetition rate of the pulse train increases from ~26 to ~40 kHz, and the pulse duration reduces from ~9.5 μs to ~3.1 μs . The repetition rate and the pulse duration against the pump power are shown in Figure 6C, which is a typical Q-switching operation. The output polarization is characterized by placing a rotatable polarizer between the laser output and the power meter. The obtained Q-switched pulse is fully linearly polarized with DOP of ~99%.

The mode-locking operation can be achieved by adding an additional 3-m-long single-mode fiber (SMF-28)

in the cavity, and the laser has a net anomalous cavity dispersion of -0.25 ps^2 [121]. Figure 6E–H manifest the mode-locking performance. The spectral bandwidth is 6.2 nm at the center wavelength of 1558.7 nm. The Kelly sidebands are superimposed on the spectrum, indicating the generation of traditional soliton. The corresponding pulse duration and the repetition rate are ~786 fs and ~14.7 MHz, respectively. It is worth to mention that the obtained ultrashort pulses are also linearly polarized with the DOP of ~98%. The obtained results have proved that the anisotropic BP-based saturable absorber can contribute to the linearly polarized ultrashort pulse generation.

4.1.1 Q-switched laser

The Q-switched fiber lasers have advantages of high pulse energy, high beam quality, compactness, robustness, low cost, and so on. [29, 165, 166]. For different gain fibers, such as ytterbium-doped fiber (YDF), EDF, and thulium/holmium-doped fiber laser (THDF), the anisotropic 2DLMs can assist fiber lasers to generate Q-switched pulses at the center wavelength at 1, 1.5, and 2 μm . Compared with fiber lasers, the solid-state lasers could generate pulses at shorter and longer wavelength. For different gain mediums, such as Pr:YLF crystal, Nd:YAG crystal, Ti:sapphire, and Er:YAP crystal, the anisotropic 2DLMs can assist the solid-state lasers to generate pulses at the center wavelength ranging from 640 nm to 3 μm . The

Figure 6: The linearly polarized Q-switched and mode-locked fiber lasers incorporating BP-based saturable absorber.

(A) Experimental setup. (B) Q-switched pulse train. (C) The pulse duration as well as repetition rate against the pump power. (D) Output polarization property of Q-switched fiber laser. (E) Optical spectrum of mode-locked laser. (F) The corresponding autocorrelation trace and (G) RF spectrum. (H) Output polarization property of mode-locked fiber laser. Reproduced with permission from Li et al. [121].

state of art about the Q-switched fibers and the solid-state lasers incorporating the anisotropic 2DLMs is summarized in Table 2.

For the BP-incorporated Q-switched fiber lasers, Yu et al. [175] have prepared the BP-based saturable absorber with modulation depth of ~24% and demonstrated the stable Q-switching in THDF laser at 2 μm . The repetition rates of obtained pulse train changes from 69.4 to 113.3 kHz. The maximum pulse energy is 632.4 nJ. This is the first report of 2- μm BP-incorporated Q-switched fiber laser with the highest repetition rate, shortest pulse

duration, and largest pulse energy. For BP-incorporated Q-switched solid-state laser, the BP-based saturable absorber fabricated by LPE method has been successfully applied in a Q-switched solid-state laser at 2 μm for the first time [176]. The maximum average power of output pulses is 151 mW, corresponding to the pulse energy of 7.84 μJ , with the pulse duration and repetition rate of 1.78 μs and 19.25 kHz, respectively. Wang et al. [178] have prepared the SAM based on the multilayer BP with the modulation depth and the saturation power intensity of 10.7% and 0.96 MW/cm², respectively. Based on such

Table 2: Summary of Q-switched lasers with anisotropic 2DLMs.

Anisotropic 2DLMs	Gain medium	Center wavelength (nm)	Repetition rate (kHz)	Pulse duration (μs)	Max. pulse energy (nJ)	Ref.
BP	Pr ³⁺ -doped ZBLAN fiber	635	108.8–409.8	0.383–1.56	27.6	[167]
	Yb:LuYAG	1029	63.9	1.73	0.09	[168]
	Yb:CaYAlO ₄	1046	87.7–113.6	0.62–1.2	325.7	[169]
	Yb ³⁺ :ScBO ₃	1063.6	20–30.6	0.4955–1.393	1400	[170]
	YDF	1069.4	8.2–32.9	10.8–17.9	328	[171]
	EDF	1550	31.53–82.85	5.52–9.36	51	[172]
	EDF	1556.9	5.73–31.07	3.59–25.77	142.6	[173]
	EDF	1561.9	7.86–34.32	2.96–55	194	[174]
	EDF	1562.8	6.983–15.78	29.84–310.3	94.3	[112]
	EDF	1595	13.33–26.6	7.11–10.67	468.03	[29]
	THDF	1912	69.4–113.3	1.42–0.731	632.4	[175]
	Tm:CaYAlO ₄	1930	17.7	3.1	680	[168]
	Tm:YAP	1988	10.85–19.25	1.78–4.05	7840	[176]
	Tm:YAG	2009	6.1–11.6	2.9–9.1	3320	[177]
	Cr:ZnSe	2400	98–176	0.189–0.396	205	[178]
	Er:Y ₂ O ₃	2720	12.6	4.47	480	[168]
	Er:ZBLAN fiber	2800	39–63	1.18–2.1	7700	[179]
	Er:CaF ₂	2800	26.5–41.93	0.9548–2.38	4250	[180]
	Er:Lu ₂ O ₃	2840	60–107	0.359–0.72	7100	[181]
ReS ₂	Pr:YLF	640	80–525	0.17–1	103	[182]
	Nd:YAG	1064.5	70–165	0.834–2.6	491	[183]
	YDF	1047	52–134	1.56–3.32	13.02	[145]
	Nd:YAG	1064	275–504	0.1216–0.29	130	[184]
	Nd:YAG	1064	130–650	0.16–0.65	186	[182]
	Nd:YAG	1318	10–308.4	0.111–0.4	330	[184]
	Nd:YAG	1329	69–214	0.403–1.071	420	[185]
	EDF	1532	43–64	2.1–7.4	38	[186]
	EDF	1557.3	12.6–19	5.496–23	6280	[187]
	Tm:YAP	1991	36–68	0.41–0.985	3620	[182]
	Er:SrF ₂	2790	24–49	0.508–1.625	1210	[188]
	Er:YSGG	2796	47–126	0.324–1.1	825	[189]
	Ho,Pr:LiLuF ₄	2950.5	25.48–91.49	0.676.7–1.233	1130	[190]
ReSe ₂	YDF	1065	17.89–39.86	2.27–5.92	31	[191]
	Nd:Y ₃ Al ₅ O ₁₂	1066.5	204–274	1.08–1.53	2500	[192]
	YDF	1054.2	31.6–68.7	2.87–3.77	81.62	[193]
	EDF	1566	6.64–21.04	4.98–16.5	36	[76]
	Er:YAP	1937.8	19.5–89.4	0.9258–1.8	17,600	[194]
	Er:YAP	2796	110–244.6	0.2028–0.6375	2200	[195]

BP-SAM, a stable Q-switched pulse with the pulse energy of 205 nJ can be generated from 2.4- μm solid-state laser.

For the ReS_2 -incorporated Q-switched fiber laser, Mao et al. [187] have fabricated the few-layer ReS_2 nanosheets by LPE method. The ReS_2 -PVA film shows the modulation depth of 0.12% with the saturable intensity of 74 MW/cm^2 at 1.55 μm . They have demonstrated the passive Q-switching with the maximum pulse energy of 62.8 μJ . For ReS_2 -incorporated Q-switched solid-state laser, Su et al. [182] have prepared ReS_2 -based saturable absorber by LPE method. They have achieved the broadband Q-switched solid-state lasers at center wavelengths of 0.64, 1.064, and 1.991 μm . Meanwhile, the femtosecond pulses have been generated from the solid-state laser incorporating the ReS_2 saturable absorber at 1.06 μm for the first time.

For the ReSe_2 -incorporated Q-switched fiber laser, Wang et al. [193] have fabricated the ReSe_2 -based saturable absorber by ME method, demonstrating the saturation intensity, modulation depth, and nonsaturable loss of 9.56 MW/cm^2 , 28%, and 64%, respectively. The repetition rates of obtained pulse train change from 31.6 to 68.7

kHz. The obtained maximum pulse energy is 81.62 nJ. For ReSe_2 -incorporated Q-switched solid-state laser, Yao et al. [195] have prepared ReSe_2 -based saturable absorber by LPE method with the modulation depth and the saturation intensity of 7.5% and 14.5 $\mu\text{J}/\text{cm}^2$, respectively. By using the prepared ReSe_2 -based saturable absorber, a Q-switched Er:YAP solid-state laser near $\sim 3 \mu\text{m}$ is developed with the shortest pulse duration of 202.8 ns and the highest repetition rate of 244.6 kHz for the first time. The obtained pulse peak power and the pulse energy are 10.6 W and 2.2 μJ , respectively.

4.1.2 Mode-locked laser

Compared with the Q-switched lasers, the passively mode-locked lasers involve the soliton formation. By adjusting the cavity dispersion and nonlinearity, the ultrashort pulse could be produced [17, 19, 136, 196–200]. The state of art about performance of the mode-locked lasers incorporating the anisotropic 2DLs is summarized in Table 3.

Table 3: Summary of mode-locked lasers with anisotropic 2DLs.

Anisotropic 2DLs	Gain medium	Center wavelength (nm)	3-dB bandwidth (nm)	Repetition rate (MHz)	Pulse duration (ps)	Average power (mW)	Pulse energy (pJ)	Ref.
BP	YDF	1030.6	0.11	46.3	<400	32.5	701.9	[201]
	Nd:YVO ₄	1064.1	0.288	140	6.1	460	3290	[114]
	YDF	1085.5	0.23	13.5	7.54	10	5930	[202]
	EDF	1555	4.6	37.8	0.687	1.38	36.5	[203]
	EDF	1557.7	1	1.65	9.41	2.1	1270	[204]
	EDF	1558.1	1.25	15.59	2.18	0.0776	4.98	[205]
	EDF	1560.5	10.2	28.2	0.272	0.5	17.73	[107]
	EDF	1560.7	6.4	6.88	0.57	5.1	740	[206]
	EDF	1561	0.985	1	2.66	1.224	7350	[172]
	EDF	1562	4.5	12.5	0.635	–	–	[207]
	EDF	1564.6	5.7	3.47	0.69	–	–	[21]
	EDF	1566.5	3.39	4.96	0.94	–	–	[22]
	EDF	1569.2	9.35	60.5	0.28	–	–	[208]
	EDF	1571.4	2.9	5.96	0.946	–	–	[112]
	EDF	1579.4	5.8	12	0.686	0.77	64.17	[26]
	THDF	1898	3.9	19.2	1.58	2.5	130.2	[209]
	TDF	1910	5.8	36.8	0.739	1.5	40.7	[154]
	HDF	2094	4.2	29.1	1.3	11	379	[210]
ReS_2	Er:ZBLAN fiber	2783	2.8	24.27	42	613	25.5	[211]
	Yb:CALGO	1060	4.23	50.71	0.323	350	6903	[182]
	EDF	1563.3	8.2	1.78	3.8	–	–	[212]
	EDF	1556	1.85	5.48	1.6	0.4	72.99	[187]
ReSe_2	EDF	1565	1	1.896	2.549	–	–	[213]
	Ti:sapphire	1064	1.3	6514	29	259	20	[214]
	EDF	1561.2	3.4	14.97	0.862	0.5	33.4	[215]
SnS	EDF	1567.9	1.8	459	1.02	–	–	[216]
SnSe	EDF	1559	5.3	15.5	0.61	0.1	6.45	[217]

For the BP-incorporated mode-locked fiber laser, Sotor et al. [107] have reported an EDF laser incorporating BP-based saturable absorber prepared by the ME method to generate ultrashort pulses with 272 fs at 1560.5 nm. They have also observed the TPA effect of exfoliated layers BP; the coefficient of TPA is approximately 500 cm/MW. Zhao et al. [204] have prepared BP-based SA with the modulation depth and the saturation intensity of 12.4% and $2.16 \mu\text{J}/\text{cm}^2$, respectively. A synchronous triwavelength mode-locked EDF laser has been achieved with the repetition rate of 1.65 MHz. For mode locking at the longer wavelength, Qin et al. [211] have used Er:ZBLAN fiber as gain medium and fabricated a BP-SAM to generate pulses with the duration of 42 ps at 2.8 μm .

For the ReSe_2 -incorporated mode-locked fiber laser, the prepared ReSe_2 -based saturable absorber has a modulation depth and saturation power of $\sim 3.9\%$ and $\sim 42 \text{ W}$, respectively, for the transverse electric mode, whereas for the transverse magnetic mode the modulation depth and the saturation power are little different, which are $\sim 2.4\%$ and $\sim 53 \text{ W}$, respectively [215]. The stable pulses with the pulse duration of $\sim 862 \text{ fs}$ have been generated from an EDF laser. This is the first report of femtosecond mode-locked pulse generation based on ReSe_2 saturable absorber. For ReSe_2 -incorporated mode-locked waveguide laser, Li et al. [214] have demonstrated the continuous-wave mode-locking operation with pulse duration of 29 ps at 1 μm . As the SAMs are directly integrated on the gain waveguide, the fundamental repetition rate of 6.5 GHz can be achieved in a monolithic waveguide platform [214].

For the SnS-incorporated mode-locked fiber laser, Feng et al. [216] have prepared the SnS-based saturable absorber with the modulation depth of 5.8%. By

designing the laser cavity with the largely net anomalous cavity dispersion, the developed laser can produce the 105th harmonic soliton molecule near 1.5 μm for the first time.

For the SnSe-incorporated mode-locked fiber laser, Jhon et al. [217] have fabricated the SnSe-based saturable absorber with the modulation depth of 7.1% and demonstrated the ultrashort pulse generation with the pulse duration of 610 fs at 1560 nm for the first time.

4.2 Optical switch

The polarization-dependent SA of the anisotropic 2DLMs can be further explored in optical switches. When the anisotropic 2DLM is irradiated by ultrashort pulses, plenty of excited electrons fill the conduction band. Subsequently, the absorption of signal will decrease based on the Pauli-blocking principle. In the meantime, as the optical transition selection rules imply, the incident polarization direction will affect the energy level into which the electrons are transported [105]. Thereby, the change of polarization direction controls the transmission of the signal light. Thus, the “ON/OFF” states of the signal light can be switched by controlling the angle between the incident light and the armchair direction. As shown in Figure 7, an optical switch is demonstrated based on the polarization-dependent SA of SnSe [105]. Figure 7A shows the normalized transmittance of signal light (633 nm, CW, 13 μW) with or without switching light (800 nm, 65 fs, 1 kHz, 34 GW/cm^2), at the polarization direction that is parallel or perpendicular to the armchair direction. Compared with the case without

Figure 7: The characterization of the SnSe-based all-optical switch.

(A) The time evolution of normalized transmission of 175-nm SnSe along zigzag and armchair directions when the switch light is “on” and “off”. (B) The time evolution of normalized transmission of SnSe along zigzag and armchair directions when the polarization of switch light is $105^\circ/15^\circ$. Reproduced with permission from Zhang et al. [105].

switching light, in the presence of switching light, the transmittance of SnSe along the armchair direction as well as the zigzag direction is higher. An ON/OFF ratio is introduced, representing the difference of the normalized transmittance between the “ON” and “OFF” states. It is found that the ON/OFF ratio is 16% for incident polarization paralleling to the armchair direction and 8% for incident polarization paralleling to the zigzag direction. By keeping the switching light on and adjusting the angle of the switching light between 105° and 15° , the normalized transmission of signal light is modulated with a contrast of $\sim 45\%$. Such anisotropic 2DLM provides a new perspective for all-optical switches.

5 Conclusions and perspectives

Various investigations toward the anisotropic 2DLMs have demonstrated the remarkable polarization-dependent light modulation property of the anisotropic 2DLMs, providing a new degree of freedom for future photonics applications. In addition to the representative five anisotropic 2DLMs including BP, SnSe, SnS, ReS_2 , and ReSe_2 , there still are other kinds of anisotropic 2DLMs such as WTe_2 , GeP, GeS_2 , and so on. Song et al. [218] have reported the polarization-dependent anisotropic Raman response of few-layer and bulk WTe_2 , and scholars have demonstrated WTe_2 as saturable absorbers to generate ultra-short pulses [219–221]. GeP also exhibits low-symmetry crystal structure as a member of group IV–V compounds [222], which has been developed for saturable absorbers, optical switches, and photodetectors [223, 224]. GeS_2 , as a member of binary IV–VI chalcogenides, is another kind of low-symmetry material that crystallizes in a layered monoclinic structure. Yang et al. [225] have reported polarized photodetection from visible to UV region based on the anisotropic GeS_2 . Although we have focused on the representative five anisotropic 2DLMs, the scope of materials can be further enlarged for the 2D materials that exhibit the polarization-dependent nonlinear SA. Such unique advantage of polarization manipulation in 2D materials allows for the broadband, versatile, ultrafast, polarization-sensitive photonic devices, showing great potential in highly polarized pulse generation. However, the challenges of the anisotropic 2DLMs for ultrafast photonics include the following:

- (1) With respect to the anisotropic 2DLM-based devices, the first challenge might be short light–matter interaction length. This challenge also exists in most of 2DLMs. The short interaction length might result in

the limited polarization-dependent nonlinear optical absorption. In order to strengthen the nonlinear optical absorption, one straightforward way is to increase the number of layers. However, the stacked layers will inevitably cause an increase in nonsaturable loss. An alternative solution to balance the absorption might be heterostructure, stacking by different 2DLMs with reasonable absorption. Once the anisotropic 2DLMs are given, in order to further enhance the polarization-dependent nonlinear optical absorption, the anisotropic 2DLMs can be integrated with cavity resonators [226].

- (2) The second challenge of applying the anisotropic 2DLMs could arise from the system integration. Even though the anisotropic 2DLMs can achieve polarization manipulation, the difficulty is how the pulsed laser systems maintain the manipulated polarization state in the whole optical path. In other words, the pulsed laser systems consist of not only the saturable absorber but also the gain, the pump, and the cavity feedback. Any perturbation from the intracavity components to light polarization might deplete the DOP of the laser. In order to achieve high DOP, in addition to the anisotropic 2DLM-based saturable absorber, the rest of the laser cavity components are suggested to be polarization-maintaining.

This review is aimed to intrigue further interest in study and application of anisotropic 2DLMs in the future. Potential research directions in anisotropic 2DLM-incorporated pulsed laser include the following:

- (1) The van der Waals heterostructures: compared with the single 2DLM, the heterostructures based on 2DLMs display great properties with the suitable bandgap and high carrier mobility [227]. For example, the combinations of BP with ReS_2 [228] or graphene [229] bring the heterostructures' excellent properties while maintain its anisotropic property [228, 229]. Although the isotropic 2D materials are thought to be polarization-independent, the heterostructures by the combination of isotropic 2D materials with anisotropic 2D materials can also be ideal saturable absorbers for highly polarized pulse lasers. Such isotropic 2DLMs could include the group VA material [45–47], MPTs [48], to 2DMOFs [49]. Based on the van der Waals epitaxy growth method [230], even 2D-layered materials can be combined with 2D nonlayered materials such as selenium [51] and tellurium nanosheets [52]. Note that the combination of 2D materials is meaningful only if the performance of the device is improved.

- (2) Highly polarized pulsed laser: benefiting from the unique advantage of polarization manipulation, the anisotropic 2DLM-based saturable absorbers can assist the generation of polarized pulses from the laser systems equipped with highly integrated, miniaturized, and portable features.
- (3) Mode-locked multi-GHz lasers: because of polarization-dependent SA and even the reverse SA effect, by controlling the light polarization properly, a reasonable loss modulation from anisotropic 2DLM-based saturable absorbers can assist pulse split, leading to high-repetition-rate mode-locked laser.
- (4) All-optical switch/modulators: based on polarization-dependent SA of the anisotropic 2DLMs, by adjusting the polarization of switching light, the normalized transmission of signal light can be optically modulated, showing a great potential for all-optical switches.

Indeed, the applications of the anisotropic 2DLMs in ultra-fast photonic are not limited to the above suggestions. It is expected that the transformation of experimental research into industrial application will benefit the future photonics technology.

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