Low-dimensional materials as saturable absorbers for pulsed waveguide lasers

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Abstract
Low-dimensional (LD) materials, such as 2D materials, carbon nanotubes, and nanoparticles, have attracted increasing attention for light modulation in photonics and optoelectronics. The high nonlinearity, broad bandwidth, and fast response enabled by LD materials are critical to realize desired functionalities in highly integrated photonic systems. Driven by the growing demand for compact laser sources, LD materials have recently demonstrated their great capacity as saturable absorbers in pulsed (Q-switched or mode-locked) laser generation in waveguide platforms. We review the recent advances of pulsed waveguide lasers based on LD materials. A perspective is also presented in this rapidly growing research field.

1. Introduction

With the nanoscale confinement of electrons, low-dimensional (LD) materials, including zero-dimensional (0D) nanoparticles, one-dimensional (1D) nanotubes, and two-dimensional (2D) layered materials, have exhibited extraordinary physical properties that are absent from their three-dimensional (3D) counterparts [1–3]. For example, under light excitation, the near-field and optical nonlinearity of noble metallic nanoparticles could be strongly enhanced due to localized surface plasmon resonance (LSPR) [4, 5]. 2D materials have shown strong light-matter interactions due to the reduced dielectric screening [6–8]. One of the remarkable applications of LD materials is that they could be used as efficient and broadband optical modulators [9–11], which is essential for the development of various devices in nonlinear optics and ultrafast photonics. Optical waveguides are basic components in integrated photonics [12]. The integration of LD materials in optical waveguides could enable highly-desired ultrafast laser sources that are compatible with miniaturized photonics systems. Jia et al have recently presented a comprehensive review of pulsed waveguide lasers [13]. While the field still growing rapidly, a number of new research works are emerged based on LD material-based saturable absorbers (SAs) and numerous opportunities are ready to boom in the next few years. In this Perspective, we mainly focus on the recent progress and future outlook on the research of nonlinear optical properties of LD materials and their key role in solid-state waveguide platform.

This paper will be organized as follows. First, the nonlinear optical properties of LD materials as well as the solid-state waveguide systems are briefly described. Then, we demonstrate the role of the LD materials as SAs in pulsed waveguide laser operation with different operation regimes. The majority will be focused on the solid-state waveguide lasers with laser crystals as gain media. Some brief descriptions of pulsed lasers based on semiconducting III-V materials and quantum dot layers are presented as well. Finally, we highlight key challenges and possible solutions for future research in LD material-based SAs and pulsed waveguide lasers.

2. Fundamentals of LD saturable absorbers and optical waveguides

Owing to the diverse electronic structures, LD materials offer versatile options to realize wide applications based on the nonlinear optical response. Currently, semiconductor saturable absorber mirrors (SESAMs) are the most frequently used commercial SAs, processing high damage threshold and prominent stability. However, a variety of LD materials retain a number of advantages to be nonlinear SAs, of which SESAMs...
cannot fulfill, such as broadband operation, ultrafast recovery time, low cost, and better compatibility with compact devices such as fibers and waveguides. Recently, an increasing number of research efforts have been made to unfold the nonlinear optical properties as well as its corresponding applications of the emerging LD materials [14–23]. For semiconducting or semimetallic LD materials, the main mechanism for nonlinear saturable absorption is Pauli blocking. After photoexcitation, non-equilibrium carrier state could be created, in which valence-band electrons can be excited to the conduction band, and a hole can be formed on the valence band. As the increase of the incident light intensity, the photon absorption processes of LD materials experience a transition from linear absorption to saturable absorption. When the light intensity continues increasing and reaches the saturation intensity, the extra photons will no longer be absorbed by the electrons in the valence band, and the transmittance of the SA will not increase but gradually tend to a stable value. For metallic nanoparticles, the dominant mechanism is the LSPR effect arising from the interaction between the electric field of incident light and the surface electrons in the conduction band. The optical nonlinearity could be significantly enhanced while maintaining the ultrafast recovery time. These nonlinear effects are widely used in passively Q-switched or mode-locked lasers. In order to characterize the nonlinear optical properties, femtosecond Z-scan spectroscopy is typically employed by moving the sample along the Z direction through a focused Gaussian beam. The laser intensity can be varied continuously with the maximum at the focal point. With either open-aperture or close-aperture configuration, we can obtain the nonlinear parameters, such as modulation depth, saturation intensity, and nonlinear refractive index. The carrier lifetime is also an essential factor for SAs. The carrier dynamics and relaxation time could be investigated by ultrafast pump-probe measurements and subsequent exponential fitting of the experimental data.

Since the groundbreaking invention of ruby (Cr$^{3+}$:Al$_2$O$_3$) laser in 1960, lasers have attracted widespread attention and played an important role along with the modern technological evolutions [24]. Optical waveguides are the basic components in integrated photonics, in which the light field could be confined in a very small volume [12, 25]. Waveguiding structures with different structures can be fabricated by a few well-established techniques, such as femtosecond laser writing, ion beam implantation, ion exchange, pulsed laser deposition, and molecular beam epitaxy, in which the first two techniques are most commonly used for solid-state waveguides. The femtosecond laser direct writing is one of the most effective methods for preparing complex three-dimensional (3D) microscale waveguide structures [12, 26]. Due to the high energy density at the focal point, the energy of femtosecond laser pulses can be deposited inside the crystal through non-linear absorption processes (including excessive photon absorption, tunneling ionization, avalanche ionization, etc), triggering a localized lattice structure (refractive index) change. Ion beam irradiation is also a commonly used method for the preparation of crystal optical waveguides [1, 17, 18]. The deposition of ion energy causes localized lattice distortion, thereby inducing corresponding changes in the refractive index of the crystal. Based on the fabricated optical waveguides, miniature photonic devices could be achieved, such as beam splitters, directional couplers, and frequency converters [27–31]. Compact laser sources based on the waveguide platform have recently shown its great potential for the development of integrated photonics [13]. Lasers could be experimentally achieved based on different waveguiding configurations such as straight, curved, and Y-branch structures [32–38]. With the microscale waveguiding volume, waveguide lasers are expected to have better laser performances, such as higher slope efficiency and lower lasing thresholds [39]. According to the operation regime, lasers based on waveguide platforms can be classified into continuous-wave and pulsed waveguide lasers. As discussed earlier [13], LD nanomaterials could be integrated with the solid-state waveguide in two configurations, i.e. transmission absorption (directly attached in the end face) and evanescent field absorption (near field interaction with propagating waves). The combination of LD materials and the optical waveguides could enable the design and optimization of ultrafast laser sources toward compact laser sources.

3. Q-switched waveguide lasers

Passively Q-switched laser operation is an efficient way to generate nanosecond-level laser pulses with relatively high pulse energy. The LD materials could serve as optical modulators to switch the quality (Q) factor of the laser resonant cavity from a low to a very high value. Therefore, the stored cavity energy will be suddenly released and transmitted in the form of a short and intense light pulse. Based on the waveguide platform, compact Q-switched laser sources have been achieved modulated by various LD materials with a broadband spectral coverage. Table 1 summarizes the recent advances of LD materials-based Q-switched waveguide lasers. The majority of these works are focused on the waveguide laser in the near-infrared using Nd, Yb, or Er-doped gain media. Choi et al realized a diode-pumped Q-switched Yb:YAG waveguide laser modulated by single-walled carbon nanotubes [40]. With different output couplers, the highest achieved pulse energy is 39.2 nJ and the shortest pulse duration is 78 ns. Ma et al report on the passively Q-switched...
laser at $\sim 1024$ nm based on Yb:YSGG ridge waveguide [41]. Based on the femtosecond-laser-written Nd:YAG cladding waveguides, Cheng et al realized the 1 $\mu$m Q-switched waveguide laser modulated by tin diselenide (SnSe$_2$) as a new SA [42]. The Q-switched laser based on MoSe$_2$, MoSe$_3$, and WSe$_2$ is also demonstrated with a minimum pulse duration of 203, 80, and 52 ns, respectively [43, 44]. Tan et al demonstrated the ultrafast nonlinear properties of WS$_2$ and black phosphorous and their applications in Q-switched waveguide laser operation [45]. Q-switched Nd:YAG ridge waveguide laser has been demonstrated based on WS$_2$, in which the waveguide is fabricated by a combined technique of swift heavy ion irradiation and laser ablation [46]. Jia et al recently show the passively Q-switched operation in a few-mode depressed-cladding waveguide [47]. High-power Q-switched waveguide laser at 1 $\mu$m is demonstrated with peak power up to 5.6 W and maximum pulse energy of 1 $\mu$J [48]. The slope efficiency is estimated to be above 74%. The nonlinear absorption of graphene can be electrically controlled to realize gate-voltage-dependent Q-switched waveguide laser pulses [49]. With the increase of the applied voltage, the Fermi level of graphene could be tuned with the optical absorption ratio changes from 29.2% to 15%, and the modulated depth varied from 1.2% to 0%. Therefore, the pulse duration could be tunable from 32 ns to 260 ns by controlling the electric signals. Based on graphene/WS$_2$ van der Waals heterostructure, Q-switched waveguide laser has been achieved with a high slope efficiency of 37% and a maximum output power of 275 mW [50].

In the mid-infrared region, LD materials also demonstrated their capability as nonlinear optical modulators. In this regime, the waveguides are typically fabricated within Ho or Tm-doped laser gain media, such as Ho:YAG, Tm:KYW, and Tm:KLuW. Kifle et al realized $\sim 2$ $\mu$m passively Q-switched laser based on carbon nanotube and femtosecond-laser-written Tm:KLu(WO$_4$)$_2$ waveguide. The pulse duration can be as short as 50 ns and the repetition rate is as high as 1.48 MHz [59]. The passive Q-switching has also been demonstrated at 1837.1 nm with Tm surface waveguide through the evanescent field coupling of single-walled carbon nanotubes [61]. Kim et al experimentally compared the $\sim 2$ $\mu$m Q-switched waveguide laser performance under both direct- and evanescent-field interactions with carbon nanotubes [62]. The intensity required for the SA in evanescent-field interaction mode is 1500 times lower than that of direct-attached configuration. With graphene SA, Q-switched waveguide laser has been achieved at wavelength of 1831.8 nm and repetition frequency of 1.13 MHz [55]. Based on MoS$_2$ thin film and circular cladding Tm:KLuW waveguides, 1.58 MHz Q-switched laser have been demonstrated at 1845.0 nm with pulse duration down to 66 ns [57]. Sb$_2$Te$_3$ thin film has also been employed as a SA for the passive Q-switched laser at $\sim 2$ $\mu$m based on evanescent field interaction. The pulsed waveguide laser could be generated with high slope efficiency (37%) and high pulse energy (3.5 $\mu$J) [68].

The ion beam offers a powerful tool to create atomic-scale modifications and manipulate the physical properties of 2D materials toward their applications in photonics and optoelectronics [71]. As shown in figure 1, Ma et al report on the tailoring of the nonlinear optical response of WS$_2$ via Ar$^+$ (argon) ion irradiation [64]. The composition evolution before and after ion irradiation could be confirmed with Raman spectroscopy, x-ray photoelectron spectroscopy (XPS), and first-principle calculations. The linear absorption of the sample is found enhanced with the increase of the ion fluence. With the femtosecond Z-scan technique, the saturation intensity gets lower and the modulation depth gets higher with the increase of ion fluence. Improved Q-switched lasing performance is obtained with irradiated samples, with a reduction of the pulse duration from 265 ns to 52 ns as the increase of the ion fluence. Similar work has been reported for Bi$_2$Se$_3$, in which the nonlinear properties could be modified by N$^+$ ion irradiation [67]. For 2D van der Waals heterostructures, the interlayer coupling could be effectively manipulated by ion beam technique. The nonlinear optical response of graphene/WSe$_2$ heterostructure before and after ion irradiation was systematically studied by Tan et al [69]. The C$^{3+}$ (carbon) ion irradiation was employed with a fluence of $10^{12}$–$10^{13}$ ions cm$^{-2}$. From the atomic force microscopic images, the surface of the irradiated sample gets smoother after irradiation and the interlayer distance gets smaller. According to the Z-scan measurement, enhanced nonlinear responses, such as lower saturation intensity and higher modulation depth, were obtained for the irradiated van der Waals heterostructure. The relaxation time after modification was significantly decreased compared with the pristine ones according to the pump–probe measurement.

Noble metallic nanoparticles have shown LSPR effect that could significantly enhance the nonlinear optical response. Notably, embedded plasmonic nanoparticles could be fabricated within various dielectric matrices that meet the practical needs of high integration and high stability. The composite structure with embedded nanoparticles has shown a broadband nonlinear optical response from the visible to the near-infrared band. Recently, Pang et al realized Q-switched waveguide laser at near-infrared and bulk laser at visible modulated by Ag nanoparticles embedded in LiNbO$_3$ [70].
Table 1. Low-dimensional materials as SAs for Q-switched waveguide lasers.

| Low-dimensional materials | Parameters | Gain media | Wavelength (nm) | Pulse width (ns) | Frequency (MHz) | Pulse energy (nJ) | Ref. |
|---------------------------|------------|------------|-----------------|------------------|-----------------|------------------|------|
| Graphene                  | CVD        | Nd:YAG     | 1064            | 70               | 4.3             | 55               | [51] |
| Graphene                  | CVD        | Nd:YAG     | 1064            | 40               | —               | —                | [49] |
| Graphene                  | CVD        | Nd:YVO₄    | 1064            | 30               | 5.3             | —                | [52] |
| Graphene                  | CVD        | Nd:YVO₄    | 1064            | 31.2             | 14.5            | 26.8             | [47] |
| Graphene                  | CVD        | Yb₂O₃      | 1064            | 121              | 1.47            | 330              | [53] |
| Graphene                  | CVD        | Yb,Na:CaF₂ | 1013.9/1027.9   | 103.4            | 0.2693          | 130              | [54] |
| Graphene                  | CVD        | Tm:KYW     | 1831.8          | 195              | 1.13            | 5.8              | [55] |
| Graphene                  | CVD        | Tm:KYW     | 1917            | 136              | 0.37            | 1200             | [56] |
| Graphene                  | CVD        | Tm:KYW     | 1846.1          | 72               | 1.45            | 13.1             | [57] |
| Graphene oxide            | —          | Nd:YAG     | 1064            | 179              | 0.93            | 221              | [58] |
| Carbon nanotube           | LPE        | Yb:YAG     | 1029            | 78               | 1.59            | 37.7             | [40] |
| Carbon nanotube           | —          | Tm:KYW     | 1912            | 50               | 1.48            | 7                | [59] |
| Carbon nanotube           | arc-discharge | Tm:KYW | 1846.8          | 98               | 1.42            | 105.6            | [60] |
| Carbon nanotube           | arc-discharge | Tm:KYW | 1837.1         | 83               | 1.39            | 33               | [61] |
| Carbon nanotube           | arc-discharge | Yb:KYW | 1026            | 215              | 1.103           | 22               | [62] |
| Carbon nanotube           | arc-discharge | Yb:KLuW | 1040            | 88.5             | 1.16            | 613              | [63] |
| WS₂                       | CVD        | Nd:YVO₄    | 1064            | 51               | 2.3             | —                | [52] |
| WS₂                       | LPE        | Nd:YAG     | 1064            | 70               | 6.10            | —                | [45] |
| WS₂                       | CVD        | Nd:YVO₄    | 1064            | 52               | —               | —                | [64] |
| WS₂                       | LPE        | Nd:YAG     | 1064            | 125              | 0.36            | —                | [41] |
| MoS₂                      | CVD        | Nd:YAG     | 1064            | 203              | 1.10            | 112              | [44] |
| MoS₂                      | LPE        | Nd:YAG     | 1064            | 80               | 3.334           | 36               | [65] |
| MoSe₂                     | CVD        | Nd:YAG     | 1064            | 52               | 2.938           | 19               | [43] |
| WSe₂                      | CVD        | Nd:YAG     | 1064            | 129              | 2.294           | 44.5             | [42] |
| black phosphorous         | LPE        | Nd:YAG     | 1064            | 55               | 5.6             | —                | [45] |
| Bi₂Se₃                    | CVD        | Nd:YAG     | 1064            | 46               | 4.7             | 31.3             | [66] |
| Bi₂Se₃                    | CVD        | Nd:YAG     | 1064            | 45               | —               | —                | [67] |
| Sb₂Te₃                    | PVD        | Tm:GdVO₄   | 1913            | 223              | 0.2             | 3500             | [68] |
| Graphene/WS₂              | CVD        | Nd:YAG     | 1064            | 43.4             | —               | —                | [69] |
| Graphene/WS₂              | CVD        | Nd:YVO₄    | 1064            | 66               | 7.777           | 33.1             | [50] |
| AgLN                      | Ion implantation | Nd:YVO₄ | 1064          | —                | —               | 38               | [70] |
4. Mode-locked waveguide lasers

Ultrashort pulses with pulse duration down to picosecond or femtosecond regime can be realized in the waveguide platform through passive mode-locking configuration. Typically, the employed modulators based on LD materials are fast SAs with ultrashort upper state lifetime that much shorter than the mode-locked pulse duration. Recently, mode-locked waveguide lasers operating at ultrahigh repetition rates up to multi-GHz regime have gained increasing research interest. The repetition rate \( f_{\text{rep}} \) of the mode-locked laser could be determined as follows

\[
f_{\text{rep}} = \frac{c}{2nl}
\]

where \( c \) is the light speed, \( n \) represents the refractive index of the waveguide, and \( l \) is the cavity length. The frequency of mode-locked lasers is usually very high due to the very short waveguide laser cavity. The GHz laser sources have potential applications in areas such as ultrafast optical sampling, frequency combs generation, and high-speed optical communication. Table 2 summarizes the recent works of LD materials-based mode-locked waveguide lasers. So far, the majority of mode-locked waveguide laser research are related to the Q-switched mode-locking (QML) operation, with graphene SA as the main research focus. For examples, Mary et al reported a 1.5 GHz QML waveguide laser operation with graphene SA, delivering laser pulses with 1.06 ps pulse duration centered at 1039 nm [72]. Ren et al demonstrated a graphene-based passively QML laser at 2 \( \mu \)m based on a laser-written Tm:YAG waveguide cavity [73]. The laser features \( \sim 7.8 \) GHz repetition rate and 6.5 mW average output power. Thorburn et al realized 5.9 GHz QML operation at mid-infrared wavelength of \( \sim 2.1 \) \( \mu \)m based on graphene and Ho:YAG waveguide [74].

Most recently, beyond graphene, Li et al have systematically compared the mode-locked waveguide laser performances modulated by different 2D materials (e.g. graphene, MoS\(_2\), WSe\(_2\), and Bi\(_2\)Se\(_3\)) [79, 83], and achieved 6.5 GHz QML laser on femtosecond-laser-written Nd:YVO\(_4\) waveguide. With different nonlinear optical properties (e.g. saturation intensity and modulation depth), Bi\(_2\)Se\(_3\) has exhibited shortest pulse duration down to 26 ps while maintaining high stability with signal-to-noise ratio up to 59 dB, as shown in figure 2. Based on Nd:YAG waveguide cavity, platinum diselenide (PtSe\(_2\)) has been firstly applied as a SA in the waveguide system to achieve 8.8 GHz QML operation [85, 98]. The nonlinear optical response of graphene is able to be enhanced by Ag nanoparticle modification [80]. The modified graphene has exhibited an enhanced ultrafast saturable absorption response with 19 times higher modulation depth and 56 times lower saturation intensity. Consequently, superior lasing performances have been achieved based on Ag modified graphene with a much shorter 33 ps pulse duration in comparison to 52 ps with pristine graphene. Recently, Jiang et al have made a comparison of QML waveguide laser performance at a wavelength of 2 \( \mu \)m based on several LD materials [99].
| Low-dimensional materials | Fabrication method | Gain media | Operation regime | Wavelength (nm) | Pulse width (ps) | Frequency (GHz) | Ref. |
|---------------------------|-------------------|------------|-----------------|----------------|----------------|----------------|-----|
| Graphene                  | CVD               | Ho:YAG     | QML             | 2091           | ∼100           | 5.9            | [74]|
| Graphene                  | LPE               | Yb: BG     | QML             | 1039           | 1.06           | 1.5            | [72]|
| Graphene                  | CVD               | Yb:Er-doped glass | QML | 1535 | ∼70 | 6.8 | [75]|
| Graphene                  | CVD               | Ti:sapphire | CWML            | ∼800           | 0.0414         | 21.25          | [76]|
| Graphene                  | CVD               | Nd:YAG     | CWML            | 1064           |                | 16.7           | [77]|
| Graphene                  | CVD               | Nd:YAG     | CWML            | 1064           | 20             | 9.8            | [78]|
| Graphene                  | CVD               | Nd:YVO₄   | QML             | 1064           | 52             | 6.5            | [79]|
| Modified graphene         | CVD               | Nd:YVO₄   | QML             | 1064           | 33             | 6.5            | [80]|
| Carbon nanotube           | -                 | Yb:YAG     | CWML            | 1030.3         | 1.89           | 2.08           | [81]|
| ReSe₂                     | CVD               | Nd:YVO₄   | CWML            | 1064           | 29             | 6.5            | [82]|
| MoS₂                      | CVD               | Nd:YVO₄   | QML             | 1064           | 43             | 6.5            | [79]|
| WSe₂                      | CVD               | Nd:YVO₄   | QML             | 1064           | 47             | 6.5            | [83]|
| Bi₂Se₃                    | CVD               | Nd:YVO₄   | QML             | 1064           | 26             | 6.5            | [79]|
| Bi₂Te₃                    | hydrothermal exfoliation | TmZBLAN   | QML             | ∼1875           | ∼700           | 0.436          | [84]|
| PtSe₂                     | CVD               | Nd:YAG     | QML             | 1064           | 27             | 8.8            | [85]|
| Au:LN                     | Ion implantation  | Nd:YVO₄   | QML             | 1064           | 74.1           | 6.5            | [86]|
| Ag₂SIO₃                   | Ion implantation  | Nd:YVO₄   | QML             | 1064           | 27.4           | 6.5            | [87]|
| Cu₂LN                     | Ion implantation  | Nd:YAG     | QML             | 1064           | 55             | 8.6            | [88]|
| Ag:YAG                    | Ion implantation  | Nd:YAG     | QML             | 1064           | 29.5           | 10.53          | [89]|
| Cu:LT                     | Ion implantation  | Nd:YAG     | QML             | 1064           | 23.5           | 8.6            | [90]|
| InAs QD                   | MBE               | AlGaAs    | CWML            | ∼1250          | 0.393          | 21             | [91]|
| InAs QD                   | MBE               | AlGaAs    | CWML            | ∼1312          | 10             | 5              | [92]|
| InAs QD                   | MBE               | AlGaAs    | CWML            | ∼1268          | 5              | 20             | [93]|
| InAs QD                   | MBE               | AlGaAs    | CWML            | 1300           | 2              | 8              | [94]|
| InGaAs QD                 | —                 | GaAs      | CWML            | ∼1300          | 4.5            | 10             | [95]|
| InAs QD                   | MBE               | GaAs      | CWML            | ∼1312          | 1.3            | 9.1            | [96]|
| InAs/InGaAs QD            | MBE               | GaAs      | CWML            | 1310           | 1.7            | 9.4            | [97]|
| InAs QD                   | MBE               | GaAs      | CWML            | ∼1312          | 1.3            | 9.1            | [96]|

Table 2. Low-dimensional materials as SAs for mode-locked waveguide lasers.
Figure 2. Q-switched mode-locked waveguide laser performance modulated by Bi$_2$Se$_3$. (a) The Q-switched envelope containing mode-locked pulses on a nanosecond scale. The inset is on the µs timescale. (b) The recorded mode-locked pulse trains on a ps timescale. (c) Single mode-locked pulse profile (d) The measured RF spectrum. The signal-to-noise ratio (SNR) is up to 59 dB. Adapted from [79].

The embedded plasmonic nanoparticles could significantly enhance the optical nonlinearity of the surrounding matrix. Pang et al systematically investigated the ultrafast nonlinear response of Au nanoparticles embedded in lithium niobate (LiNbO$_3$) crystal fabricated by direct Au ion implantation [86]. The composite material has demonstrated its potential to be SAs with a saturation intensity of 98.14 GW cm$^{-2}$. Based on Nd:YVO$_4$ waveguide, 6.4 GHz QML laser has achieved with the pulse duration of 74.1 ps. Cu nanoparticles embedded in LiTaO$_3$ crystal have also demonstrated as an effective SA in 8.5 GHz QML waveguide laser operation [90]. As shown in figure 3, the embedded plasmonic nanoparticles could enable the highly integrated design of pulsed waveguide laser sources. Through the evanescent-field interaction modulation, QML waveguide laser operating at 1 µm is obtained with 10.53 GHz repetition rate and 29.5 ps pulse width [89]. Another intriguing example is that the giant enhancement of optical nonlinearity can be achieved in fused silica by tuning the interparticle spacing of embedded nanoparticle array. The fused silica embedded with 2D-like Ag nanoparticle array has been demonstrated as a low-cost ultrafast SA for excellent QML performance at 1064 nm [87].

Continuous-wave mode-locked (CWML) waveguide lasers have also been reported in a few works based on LD materials as SAs. Okhrimchuk et al reported a CWML Nd:YAG waveguide laser pulses with graphene SA [77]. The pulse duration is 16 ps pulses and the repetition rate is up to 11 GHz. Choi et al demonstrate carbon nanotube-based 2.08 GHz CWML laser operation with type II Yb:YAG waveguide [81], delivering 2-ps-short pulses with 322 mW average output power. The dispersion of the waveguide is compensated by Gires-Tournois interferometer (GTI) and multi-functional output coupler. Ponarina et al demonstrated a dual-wavelength waveguide laser generation of picosecond pulses with 9.8 GHz [78]. Zhang et al presented a new simulation approach to understanding the role of nonlinearity, dispersion, and gain in dual-wavelength CWML waveguide lasers [100]. Recently, as shown in figure 4, Grivas et al experimentally realized femtosecond CWML laser operation based on graphene SA and femtosecond-laser-written Ti:Sapphire waveguide [76]. By introducing Gires-Tournois interferometer, the group delay dispersion within the waveguide cavity can be controlled. The output laser could operate steadily with a fundamental repetition
rate up to 21.25 GHz and pulse duration down to 41.4 fs. Furthermore, the frequency can be doubled to 42.5 GHz with optical fiber interleaver filters. Li et al. studied the ultrafast nonlinear optical properties of a newly-developed TMDs named ReSe$_2$ [82], exhibiting a broadband saturable absorption from the visible to near-infrared range. In addition, the saturation intensity and modulation depth of this material are also superior to achieve CWML in waveguide platform according to the stability criterion. CWML within the miniature and integrative waveguide platform was achieved with ReSe$_2$ SA without external placement of complex components, indicating the promising applications in future on-chip photonics. The output laser could operate in the stable CWML regime while retaining the high fundamental repetition rate of 6.5 GHz.

In addition to the solid-state waveguide lasers based on laser crystals, electrically pumped semiconductor laser diodes based on the III–V materials also receive wide applications in silicon photonics [91, 95–97, 101–109]. The most commonly used SAs in III–V systems are InAs and InGaAs quantum-dot (QD) layers fabricated by molecular beam epitaxy (MBE). The pioneering work was carried out by Huang et al., achieving 7.4 GHz mode-locked pulses at 1.3 µm based on InAs QD and GaAs waveguide [110]. The reported shortest pulse duration was 312 fs based on InAs/InP QD [107]. Recently, Liu et al. reported on the first mode-locked laser operation on Si substrates through the evanescent interaction with two section QD layers, delivering laser pulses with 9-GHz repetition rate and 1.3-ps pulse duration [96]. The active region is the
unintentionally doped GaAs waveguide that sandwiched by p- and n-AlGaAs QD layers. More recently, Auth et al also demonstrated 9.4 GHz III–V laser based on a five-stack InAs/InGaAs QD layers [97]. The superb compatibility with the existing CMOS logic enables III–V laser systems to be an important platform for the generation of ultrashort pulses.

5. Summary and perspectives

In contrast to conventional bulk materials, the high nonlinearity and broadband response enabled by LD materials are highly desirable for pulsed lasing in compact waveguide systems. In this review, we focus on the recent advances in Q-switched and mode-locked waveguide laser devices based on LD materials as SAs. However, related research is still in the early stage with graphene SA as the main research object and most of these works are focused on Q-switched and QML operation with limited spectral coverage. To further advance the development of LD material-based solid-state pulsed waveguide lasers, there are several challenges remain to be overcome and numerous new opportunities to be further explored.

5.1. Enrich the material options of LD materials in pulsed waveguide lasing

LD materials exhibit a rich variety of optical properties due to their diverse electronic structures [111]. With abundant and diverse nonlinear optical properties, the exploration of new LD materials will provide exciting new opportunities for pushing the waveguide laser performance into a higher level. For 0D materials, the LSPR peaks and nonlinear optical properties can be tuned by changing the species, size, shape, spacing, and surrounding dielectrics. Since graphene was first mechanically exfoliated in 2004, the layered 2D materials, including graphene, transition metal dichalcogenides (TMDs), and topological insulators, offer a vast variety of material options and have attracted great interest of researchers. Several newly-developed 2D materials have shown its feasibility for pulsed lasing in fiber systems, such as TiS₂, MXene, antimonene, and bismuthene [20, 22, 112, 113]. Besides transition metal dichalcogenides (TMDCs), pulsed fiber lasers have also been demonstrated with layered transition-metal monochalcogenides (TMMCs) [23]. In addition, there is still plenty of room for pentagonal 2D materials and 2D V–V binary materials, which is recently emerged and predicted with superior properties by theoretical calculations [114–117]. The LD material systems could be further enriched by stacking distinct LD materials into 2D or mixed-dimensional van der Waals heterostructures [118–120].

5.2 Tailor and control the nonlinear optical response of LD materials

To date, there are several well-established techniques for the modification of LD materials, such as ion/electron beam irradiation [71, 121–124], laser processing [125, 126], chemical functionalization [127–129] and plasma treatment [130–133]. The prominent roles of modification to LD materials are defect formation, doping effect, structural variation. Therefore, the nonlinear optical properties of LD materials could be effectively tuned. For van der Waals heterostructures, the interlayer coupling of adjacent layers could also be modified, thus influence their nonlinear optical response. Some preliminary works have been carried out to reveal the role of ion beam modification in tailoring the optical properties [64, 67, 69, 134]. Moreover, the nonlinear optical properties are expected to be manipulated with electrical or magnetic control. It is exciting to see further results of pulsed waveguide lasing based on LD materials with tailored or controlled nonlinear optical properties.

5.3 Improve the stability of LD materials in the ambient environment

In many cases, LD materials will degrade under ambient conditions. For example, black phosphorus will encounter rapid degradation with the presence of oxygen and water. Therefore, effective passivation or encapsulation strategies are highly demanded for long-term steady operation of pulsed waveguide lasers. To address this issue, researchers have recently demonstrated the effective passivation method of layered black phosphorus and phosphorene [135–141]. Particularly, passivation can be achieved by creating van der Waals structures or combing into composite layers with 3D inorganic or organic materials. Possible techniques are organic functionalization, inorganic coatings, Al₂O₃ encapsulation, and hybrid Al₂O₃/BN encapsulation. For nanoparticles, the stability could be greatly enhanced through embedding into dielectrics with the absence of air. From another perspective, the high fluence of photons could also induce irreversible degradation or damage of LD materials. A deeper understanding of the damage mechanisms would be helpful for the design and applications of LD materials SAs. To ensure stability, it is also necessary to keep the laser operation power below the damage thresholds of the specific LD materials.
5.4 Push mode-locked waveguide lasers to stable CWML operation

The mode-locked lasers include Q-switched mode-locked (QML) and continuous-wave mode-locked (CWML) laser. Currently, based on LD materials, the CWML laser operation is only realized in limited articles [76, 77, 81, 87]. CWML laser operation features constant pulse energy output, which is superior to meet practical applications such as laser materials processing, optical communication, and nonlinear spectroscopy. However, the pulse energy required for CWML is typically lower than the critical value which can be verified with the stability criterion [142]

\[ E_{P,c} = (E_{\text{sat},L}E_{\text{sat},A}\Delta R)^{\frac{1}{2}} \] (2)

\[ E_{\text{sat},L} = E_{\text{sat},A} \cdot A_{\text{eff},L} \] (3)

\[ E_{\text{sat},A} = \frac{h\nu}{m\sigma_L} \] (4)

\[ A_{\text{eff},L} = \pi w^2 \] (5)

where \( E_{\text{sat},L} \) represents the saturation energy of the gain medium, \( E_{\text{sat},A} \) is the saturation energy of the SA, \( h\nu \) is the photon energy, \( \sigma_L \) is the emission cross-section of the waveguide, \( m \) is the round-trip number through the waveguide, and \( w \) is laser beam radius. In order to realize CWML in the waveguide platforms in future works, the Q-switching instabilities should be suppressed by a number of possible procedures as follows:

(a) The waveguides are developed on laser crystals with large laser emission cross-sections;
(b) The waveguide losses are required to be adequately low, which can be performed by optimizing the fabrication parameters;
(c) The radius of the waveguide may be as small as possible to achieve relatively higher intracavity light intensity;
(d) The SAs possess relatively low saturation intensity;
(e) The dispersion compensation elements (e.g. Gires-Tournois interferometer) may be included in the systems of laser resonators. The rapid advancement of waveguide and LD material fabrication will undoubtedly facilitate the upcoming boom of CWML laser in the waveguide platform.

5.5 Pulsed waveguide lasers are expected to be realized in a wider spectral coverage

At present, the operation wavelength of pulsed waveguide laser is limited to the near-infrared or mid-infrared band. Recently, visible lasers have gained great research interest and are considered to have a wide range of potential applications, such as high-speed wireless communication (Li-Fi), biomedical imaging, medicine, and underwater detection. Until now, the pulsed visible laser has been studied in few works based on fiber and bulk systems [143–145]. In the waveguide platform, only continuous-wave lasers have been achieved in the visible spectrum based on praseodymium (Pr\(^{3+}\))-doped crystals as the gain media [146–150]. To achieve visible pulsed waveguide lasers, it is critical to find suitable SAs that can be applied in the visible range. Conventional semiconductors are rarely reported to be worked in the visible, and only one SESAM is available at a specific wavelength of 639 nm. In contrast, due to the Pauli blocking of 2D materials or LSPR effect of nanoparticles, the majority of LD materials have photobleaching features in the visible band. Therefore, LD materials have great potential as optical modulators in visible pulsed lasing. In addition, to improve the compatibility of the laser device to the existed fiber systems, more related works could be done at 1.3 \( \mu \)m and 1.5 \( \mu \)m telecom windows. Moreover, stable CWML operations are expected to be achieved at the mid-infrared band (2–25 \( \mu \)m) to meet the practical applications related to the atmospheric windows (3–5 \( \mu \)m and 8–12 \( \mu \)m) and molecular fingerprints. 2D materials have shown excellent saturable absorption properties at mid-infrared [151, 152]. Recently, nanoparticles have also demonstrated its potential for tunable SA towards mid-infrared regimes [18, 153]. Therefore, it is of great significance to realize the pulse laser output in the visible and CWML in the mid-infrared band based on the waveguide platform, which would also be a research hotspot in the future works.

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