Academic Journal of Engineering and Technology Science
ISSN 2616-5767 Vol.7, Issue 6: 7-14, DOI: 10.25236/AJETS.2024.070602

Mid-infrared fiber laser based on graphene-like heterojunction modulation

Tianlong Bai[a,*], **Danqing Long**[b]

Hunan University of Science and Technology, Xiangtan, China
[a]15128910694@163.com, [b]18773906864@163.com
[*]Corresponding author

Abstract: *Graphene-based materials have the advantages of ultra-high carrier mobility, broad spectral absorption and tunable bandgap, which enable efficient optical modulation and optical signal processing and can significantly improve the performance and stability of lasers. This paper summarises the research progress of mid-infrared fibre lasers based on graphene-like heterojunction materials. The basic principles and key technologies of mid-infrared fibre lasers, the properties and advantages of graphene-like heterojunction materials, the latest research results, and the future research directions and challenges are presented.*

Keywords: *Two-dimensional materials; Heterojunction; Saturable absorber; Mid-IR fibre lasers*

1. Introduction

Mid-infrared (2.0-5.0 μm) wavelength light has high penetrability and low scattering, and can penetrate smoke, haze and human tissues, which makes mid-infrared fibre lasers show great potential for applications in environmental monitoring, medical imaging and therapy, and military long-range detection [1-4]. In the context of gas detection, fibre lasers operating in the mid-infrared band have the potential to facilitate the identification of trace gas components present in the atmosphere. This has the capacity to contribute to the advancement of environmental monitoring and climate research. [5-7]. In the medical field, mid-infrared lasers can be employed for non-invasive diagnostic techniques, including photoacoustic imaging and spectroscopic diagnosis[8]. Additionally, fibre lasers within the mid-infrared wavelength band possess significant strategic value in military applications such as optical communications, infrared countermeasures and laser weapons[9].

In recent years, with the development of materials science, graphene and its derived heterojunction materials have attracted wide attention in the field of photonics due to their unique optical and electrical properties. Graphene is a two-dimensional material composed of a single layer of carbon atoms with excellent electron mobility, thermal conductivity and mechanical strength, and more importantly, graphene has broadband absorption properties and ultrafast nonlinear response, which makes it an ideal saturable absorber material[10-11]. Nevertheless, graphene exhibits a relatively low absorption efficiency in the mid-infrared band, and its structure and thickness must be further optimised to enhance performance. In light of these limitations, researchers have initiated investigations into the integration of graphene with other materials to form heterojunction structures, with the objective of enhancing its optical performance in the mid-infrared band[12-13]. The combination of graphene with other two-dimensional materials, including transition metal disulfides (TMDs)[14], topological insulators[15], and black phosphorus[16], results in the emergence of a range of nonlinear optical effects, such as saturable absorption, optical modulation, and optical tuning. These properties render graphene-based heterojunction materials a valuable tool in fibre laser research.

In this paper, we will investigate the fundamental properties of these materials, present the saturable absorber mechanism and the working principle of fibre lasers, recent experimental findings on graphene-like materials and graphene-based heterojunction modulation, and discuss potential future avenues of research and applications.

2. Mid-infrared fibre laser with saturable absorber design

Mid-infrared fibre lasers are typically employed to emit laser light at wavelengths of 2.0-5.0 μm. The laser gain medium, which is typically doped with rare earth elements (Er^{3+}, Tm^{3+}-Er^{3+}-, Tm^{3+}-Yb^{3+}doped), is employed to achieve the desired laser output wavelength band[17-18]. The pump source is a laser of a suitable wavelength (e.g. 790nm or 1570nm), which is used to excite the rare earth ions in the fibre to generate the laser light; in order to achieve a stable and highly efficient laser output, a saturable absorber is introduced as a passive modulator, which generates and modulates the laser pulse through its nonlinear absorption characteristics at high light intensities.

The resonant cavity, which consists of a highly reflective mirror and a partially reflective mirror, is a key element in the generation of laser light. The precise design of the laser greatly affects its performance, including output power, beam quality and wavelength stability. The modulation unit consists of a Q-modulation and mode-locking device, which allows the generation of ultrashort and high peak power pulses by adjusting the laser loss and locking the longitudinal mode in phase. The output coupler is used to modulate the final output of the laser and usually consists of a partially reflective lens or a fibre coupler. Saturable absorption devices provide an essential aid to mode-locking and Q-modulation operation of mid-infrared fibre lasers with nonlinear absorption characteristics.

Before the application of heterojunctions to mid-infrared fibre lasers, researchers mainly explored the application of two-dimensional materials as saturable absorbers, which were one of the first technologies used to achieve mode-locked lasers, and as early as in the 1970s, researchers discovered that certain materials were able to compress the width of pulsed lasers[19]. The basic principle of the saturable absorber is that when low-intensity light is incident on the material, the electrons of the material will jump from the ground state to the excited state, and at the same time, the electrons in the excited state will relax back to the ground state, resulting in the continuous absorption of low-intensity light; and when high-intensity light is incident, the electrons will jump to the conduction band, and the photogenerated carriers fill in the valence band and the conduction band, resulting in the cessation of the material's absorption of light. The relaxation of excited electrons within and between bands corresponds to the fast and slow recovery times of the saturable absorber, respectively. The performance of a saturable absorber is determined by the following factors: operating bandwidth, unsaturated absorption loss, modulation depth, saturation absorption threshold and damage threshold. To achieve saturable absorption in the mid-infrared band, the material needs to have a narrow bandgap (<0.46eV@2700nm). With the exception of undoped graphene, which itself has a zero bandgap, studies of other materials have shown that the requirements can be met by optimising the process. For example, the band gap of single-walled carbon nanotubes decreases significantly with increasing diameter[20]; transition metal sulfides can reduce the bandgap by introducing defects and adjusting the elemental ratios[21]; black phosphorus has a gradual decrease in bandgap with increasing number of layers due to its unique electronic structure[22];

3. Modulation of two-dimensional materials such as graphene for mid-infrared fibre lasers

In recent years, significant progress has been made in the application of 2D materials with saturable absorbers in the mid-infrared wavelength band.In 2012, Zhang et al[23] successfully realised for the first time a thulium-doped mode-locked fibre laser using a graphene saturable absorber, which is shown in Figure 1. Its output laser has a central wavelength of 1.94 μm, a pulse width of 3.6 ps, a FWHM of 2.1 nm, and a time-bandwidth product of 0.59, and the experimental data indicate that there is a slight chirping of the pulse. The laser has an average output power of 2 mW, a repetition frequency of 6.46 MHz, and a pulse energy of about 0.4 nJ, validating the potential of graphene for applications in the mid-infrared band. The experimental all-fibre design supports the advantages of low-noise operation and small footprint, which is suitable for use in compact unit systems that can further improve stability and noise characteristics [24].

The first application of graphene in thulium-doped fibre lasers has driven its development as a saturable absorber in the 2 μm band[25]. Currently, the main fibre used to generate fibre lasers in the visible and near-infrared wavelength bands is quartz fibre. Quartz fibres cannot be used for mid-infrared fibre lasers due to their large resonance absorption coefficient in the mid-infrared band. Therefore, new matrix fibres with low phonon loss in the mid-infrared band are required. At present, the optical fibers used in mid-infrared fiber lasers are mainly heavy metal oxide fibers (such as sulfite, bismuthate, etc.), halide fibers (such as fluoride ZBLAN, chloride, etc.) and sulfide fibers (such as As2S3,As2Se3). These fibres have low phonon energy in the mid-infrared band, good solubility for rare earth ions and high refractive index. A variety of rare earth ions such as Tm3+, Ho3+, Er3+ and Dy3+ are capable of emitting light in the mid-infrared band[26].

The extensive utilisation of fluoride optical fibres in the mid-infrared band has facilitated the investigation of materials such as graphene, transition metal sulphides and topological insulators in this spectral region. In 2015, Zhu et al. reported the first demonstration of mode-locked laser pulse output in the mid-infrared band, employing Er^{3+}-doped:ZBLAN optical fibres with a multilayer graphene saturable absorber[27]. As illustrated in Figure 2, the experimental setup employs a 975 nm fibre-coupled semiconductor laser with 470 mW pump power as the pump source. The mean output power of the laser cavity is 18 mW, with a central wavelength of approximately 2784.5 nm and an FWHM of 0.21 nm. This corresponds to a Sech2 shaping and transform-limited pulse width of 39 ps. The experimental autocorrelation curve indicates a pulse width of 65 ps, while the sech2 function, fitted to the autocorrelation curve, yields a FWHM pulse width of 42 ps. The time-bandwidth product of this mode-locked laser is 0.342, indicating that the pulses are nearly transform limited at a repetition frequency of 25.4 MHz. Ultimately, the experiments validate the remarkable application of graphene as a saturable absorber in a mode-locked fibre laser in the 3 μm wavelength band, which has the potential for significant technological advancement.

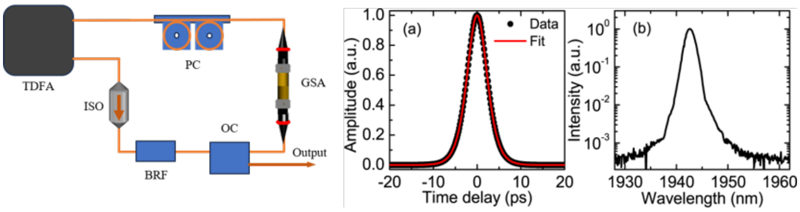

Figure 1: 1.94 μm laser experimental setup and (a) autocorrelation curves, (b) spectra[23]

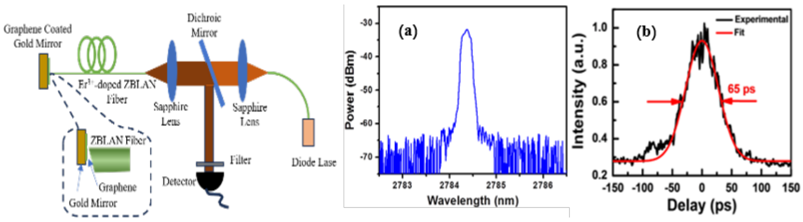

Figure 2: 2.78 μm laser experimental setup and (a) autocorrelation curves, (b) spectra[27]

Based on the above observations, it can be concluded that fibre lasers based on graphene saturable absorbers are capable of generating laser pulses in the wavelength range of 2 μm to 3 μm. However, the zero-bandgap structure of graphene usually results in relatively weak saturation intensity at long wavelengths. In contrast, phosphene (BP) materials control the bandgap by adjusting the number of layers, and multilayer phosphene has a narrow bandgap that decreases with increasing thickness, resulting in a strong saturation intensity at mid-infrared wavelengths. Phospherene is more prone to produce laser light in the mid-infrared band compared to graphene[28].Zhipeng Qin et al[29] prepared a 238-layer phosphorene sheet with a band gap of 0.33 eV by mechanical exfoliation.In their experiment, they successfully implemented a mode-locked Er:ZBLAN fibre laser, and the experimental setup is shown in Figure 3. The pulse width is 42 ps, the centre wavelength is 2783 nm, and the FWHM is 2.8 nm; the average output power is 613 mW, the pulse energy is 25.5 nJ, and the repetition frequency is 24 MHz.The experimental results show that black phosphorus has a wide range of application potentials as a saturable absorber in the mid-infrared spectral region.In a 2015 study by Sotor et al. the first demonstration of a black phosphorus-based (BP) saturable absorber in a thulium-doped mode-locked fibre laser. [30]. As shown in Fig. 4, the central wavelength is 1910 nm and the pulse width is 739 fs. Qin et al. prepared a 104-layer BP sheet with a bandgap of 0.357 eV, and achieved ultrashort pulse output based on a BP saturable absorber in the 3.5 μm band. In Q-switching operation, the average power in the 120 nm band was 1 mW, the pulse energy was 83.2 μJ, the pulse width was 5.66 μs, and the repetition frequency was 33.3462 kHz; in mode-locking operation, the average power in the picosecond pulse in the 40 nm band was 28 mW, and the repetition frequency was 91.3489 MHz. [31].

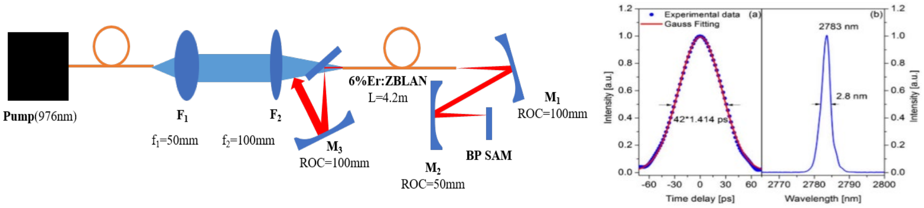

Figure 3: 2.78 μm laser experimental setup and (a) autocorrelation curves, (b) spectra[29]

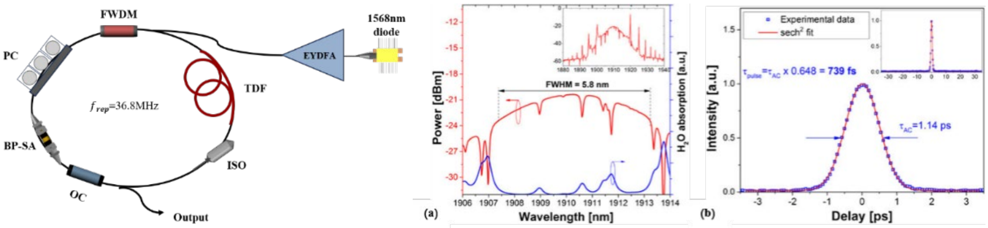

Figure 4: Laser experimental setup and (a) autocorrelation curves, (b) spectra[30]

In addition to graphene and black phosphorus (BP), other two-dimensional (2D) materials including topological insulators (TIs) and transition metal disulphides (TMDs) have shown potential applications in mid-infrared (MIR) fibre lasers.Ke Yin and co-workers proposed an ultrashort mode-locked fluoride fibre laser using a novel 2D topological insulating material, where the saturable absorbing material was prepared by drop-casting uniform Bi2Te3 nanosheets on a gold mirror,where the saturable absorbing material was prepared by drop-casting uniform Bi2Te3 nanosheets on a gold mirror[32]. Q-switched mode-locking and continuous-wave mode-locking were experimentally observed.Chen Wei et al. demonstrated a passively Q-switched Ho3+/Pr3+ co-doped fluoride fibre laser using a tungsten disulphide (WS2) saturable absorber (SA) with a central wavelength of 2865.7 nm[33]. As shown in Fig. 5, the laser was prepared using a multilayer WS2 film by sulfide growth method and then transferred to a gold mirror as a linear cavity in as feedback and SA device. Stable Q-switched pulses with an average output power of 48.4 mW were obtained at a pump power of 318.5 mW. The centre wavelength was 2865.7 nm, the FWHM 2.3 nm, the signal-to-noise ratio was 40.5 dB, the shortest pulse duration was 1.73 µs, the maximum repetition frequency was 131.6 kHz, and the energy of a single pulse was 0.37 µJ. The experimental results confirm the use of tungsten disulphide (WS_2) as a nonlinear modulator for generating pulses in the 3 µm band.

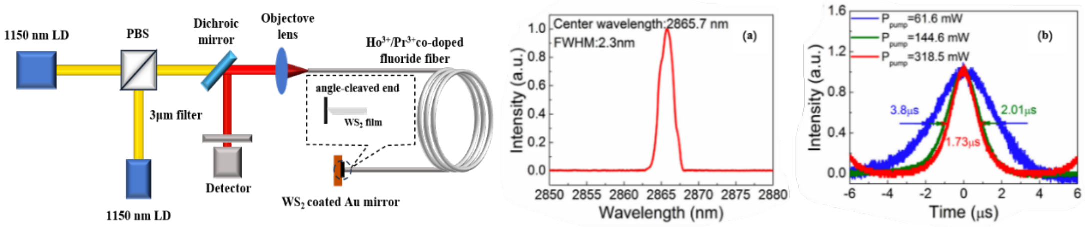

Figure 5: Ho3+/Pr3+ co-doped fluoride fibre laser and (a) autocorrelation curves, (b) spectral[33]

Although two-dimensional materials such as graphene, black phosphorus and transition metal sulphide compounds exhibit excellent optical properties in the mid-infrared band, they are still limited by their stability and absorption efficiency. Therefore, in order to further improve the performance of these materials in the mid-infrared band, researchers have begun to investigate the optical complementary effects that can be achieved by constructing heterostructures consisting of the aforementioned materials, with a view to improving the optical properties and stability of heterojunctions.

4. Mid-infrared fibre laser based on graphene and its heterojunction saturable absorber

In the mid-infrared band, graphene and transition metal disulphides (TMDs) were combined for the first time to form heterojunctions and were used as saturable absorbers in fibre lasers. In 2016, researchers Cong Liu et al. synthesised graphene and molybdenum disulphide (MoS2) van der Waals heterojunctions using chemical vapour deposition (CVD) on SiO2/Si substrates[34]. The heterojunction was employed as a saturable absorber in a passively mode-locked erbium-doped fibre laser, resulting in the generation of stable soliton pulses with a central wavelength of 1571.8 nm and a spectral width of 3.5 nm. The pulse duration was found to be 830 fs, with a repetition frequency of 11.93 MHz. The results demonstrate that graphene/MoS_2 heterojunctions are capable of supporting ultrashort pulses in fibre lasers as saturable absorbers.

In 2018, Xihu Wang et al. published a report on the use of MoS_2/graphene heterojunctions as saturable absorbers for the realisation of passively Q-modulated fibre lasers in the 2 µm and 3 µm bands[35]. The heterojunction was prepared via a hydrothermal method, which involved the combination of a monolayer of graphene and a monolayer of MoS_2 to form the heterojunction. As illustrated in Fig. 6, the gain media employed in the experiments were Tm:YAP crystals and Er:YSGG crystals, respectively. For the 2 µm laser, the shortest pulse width was 473 ns, the output power was 553 mW, and the pulse energy was 5. The laser pulse was found to be stable, with a repetition frequency of 105 kHz and an output of 267 μJ. For the 3 μm laser, a relatively stable pulse output was obtained with a pulse duration of 355 ns, an average output power of 112 mW, a pulse energy of 0.889 μJ, and a repetition frequency of 126 kHz. These results indicate that MoS2/graphene heterojunctions have significant potential for application in mid-infrared pulsed lasers.

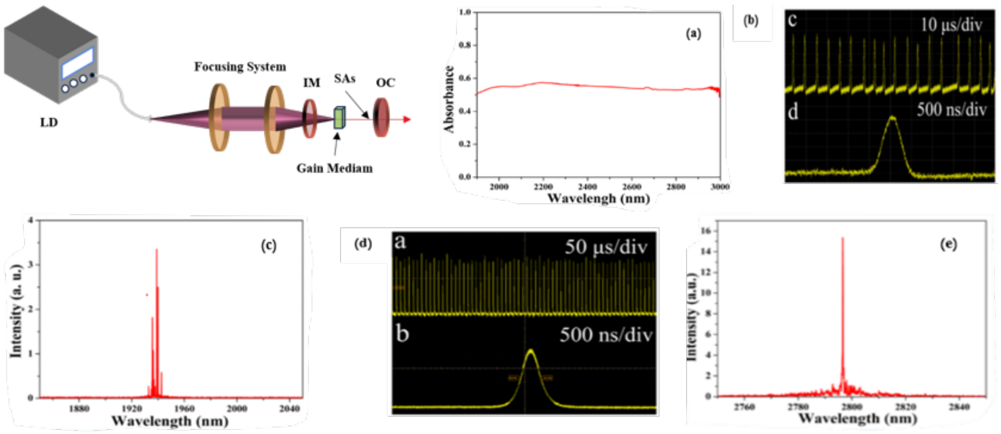

Figure 6: Fibre laser and (a) spectrum, (b) 2 μm pulsed train, (c) 2 μm pulsed output spectrum, (d) 3 μm pulsed train, and (e) 3 μm pulsed output spectrum[35]*.*

In the same year, Pinghua Tang and his team successfully prepared a graphene/MoS_2 heterojunction using a hydrothermal method and investigated its potential application in nonlinear optics[36]. They demonstrated the applicability of the heterojunction in a nonlinear optical modulator in the mid-infrared region through its nonlinear absorption parameters. The results show that the graphene/MoS_2 heterojunction is a stable optical modulator with potential applications in the mid-infrared spectral range with a narrow pulse width of 1.9 μs and a repetition frequency of 45 kHz.

Black phosphorus (BP) is an optimal material for use in saturable absorbers within the mid-infrared band, due to its pronounced anisotropic absorption and high charge mobility within the mid-infrared band. Ruicao et al. have demonstrated that BP-based composites are particularly effective in mid-infrared applications. Applications. In a typical synthesis process, black phosphorus (BP) is alloyed with arsenic (As) and carbon (C) to form β-AsP and β-PC. Under β-PC conditions, the room temperature carrier mobility and maximum absorbance spectra are suitable for mid-infrared optoelectronics[37].

Despite the great potential of BP, its weak light absorption and difficulty in tuning have been limiting its application in optical devices.In 2019, researchers introduced graphene into BP-based absorption devices and successfully obtained BP-graphene tunable perfect absorbers with ultra-wide absorption peaks by tuning the period of nanoribbons. The introduction of graphene not only enhanced the absorption of BP, but also made the absorption peak tunable by adjusting the Fermi energy level of graphene. Researchers designed a grating structure consisting of BP and graphene based on mid-infrared critical coupling, which achieved perfect absorption over a wide angular range (0°-45°), resulting in a wide-angle tunable perfect absorption[38]. Fig. 7(a) shows that the absorption peaks become wider with the increase of the gating period, and Fig. 7(b) shows that the width of the absorption peaks becomes narrower with the increase of the Fermi energy level without changing the structural parameters. The effect of spacer (dielectric) thickness t on absorption is investigated in Fig. 7(c), where the absorption peak is first red-shifted and then remains constant with increasing absorption; Fig. 7(d) shows that the position of the absorption peak is independent of the angle of incidence, and this excellent property suggests that the graphene-BP structure has a great potential for application in light absorption devices. Compared with various previously proposed structures, this design showed superior optical performance in terms of achieving tunability, easier fabrication and wider absorption peaks. Later, scientists Naixing Feng et al. investigated a three-layer infrared absorption structure with broadband absorption effect based on hybrid graphene-BP metamaterials[39], where the absorption effect of the heterojunction structure could be effectively tuned by adjusting the geometrical parameters of the structure as well as the doping levels of graphene and BP, and the structure achieved more than 87.5% of the absorption effect in the wavelength range of 10.1-24.5 μm after optimisation of the angle of incidence. An absorption of more than 87.5% was achieved. These results demonstrate the potential of graphene-BP structures as saturable absorbers in the infrared band.

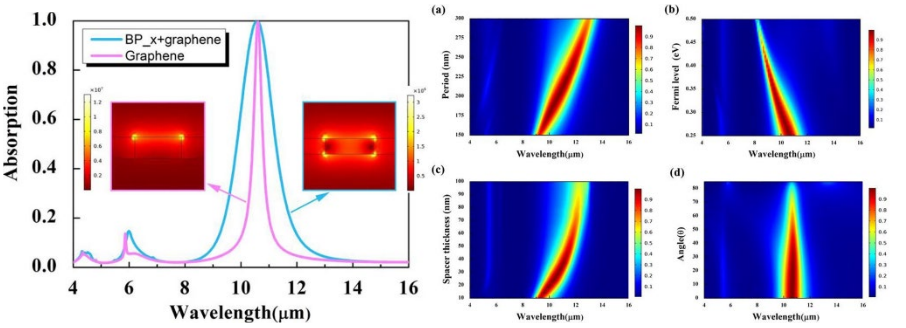

Figure 7: The left panel shows the absorption spectra for the p = 140 nm period (pink curve) and the p = 200 nm period (blue curve). The right panel illustrates the variation of the absorption peak positions with (a) the grating period, (b) the Fermi energy level of graphene, (c) the spacing thickness, and (d) the incident angularity and incident wavelength [38]

Topological insulators have ultra-high specific surface area, easy modulation of energy bands, and simple preparation, and the heterojunction formed by combining with graphene can achieve tunable relaxation time and tunable optical modulation depth.Mu et al. demonstrated a novel graphene/bismuth telluride (Bi_2Te_3) heterojunction saturable absorber, which was prepared by chemical vapour deposition method. Graphene/bismuth telluride heterojunctions with different bismuth telluride coverage enable tunable optical properties. As shown in Fig. 8, the graphene and bismuth telluride heterojunction exhibits a wide range of optical absorption properties in the visible to infrared wavelength bands, making it a promising saturable absorber device in the mid-infrared band.

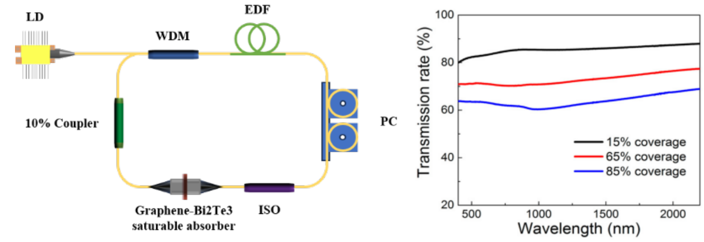

Figure 8: Fibre laser and absorption spectra[40-42]

As illustrated in the figure above, the researchers integrated a heterojunction saturable absorber into a fibre laser, resulting in the generation of stable Q-modulated and mode-locked pulse outputs. The findings demonstrated that the absorption characteristics of the heterojunction can be tailored to encompass different wavelength ranges within the mid-infrared band, due to the adaptable material coverage of the graphene/bismuth telluride heterojunction saturable absorber. Furthermore, the absorption properties of the heterojunction can be regulated by modifying the number of layers and other pertinent factors[40-42].

5. Conclusions and outlook

Graphene, black phosphorus, TMDs and TIs and their heterojunctions exhibit excellent saturable absorption characteristics in the mid-infrared band and can be used to develop high peak power and short pulse width lasers. These lasers can be used in a range of applications such as communications, spectroscopy, biomedicine, and material processing. It is recommended that future research focus on exploring new two-dimensional materials to achieve more efficient laser output and wider wavelength tuning by carefully designing different heterostructures and adjusting the number of layers. In summary, mid-infrared fiber lasers based on graphene, black phosphorus, TMDs and TIs and their heterojunctions represent a highly promising research field that is expected to achieve major breakthroughs in photonics and laser technology, providing new solutions and innovations for a variety of applications.

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