Review of laser-diode pumped Ti:sapphire laser

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Abstract
Ti:sapphire laser has an important position in ultrashort pulse generation and wavelength tuning. The use of expensive frequency-doubled diode-pumped solid-state laser as the pump source limits the promotion of its application to a certain extent. With the maturity of high-power blue-green laser diodes (LDs), low-cost LD directly pumped Ti:sapphire laser becomes possible. After more than 10 years of development, the output parameters of LD pumped Ti:sapphire lasers have been greatly improved. Moreover, the lasers have been applied to optical frequency combs, multiphoton microscopy imaging, and so forth, both of which have achieved impressive results.

KEYWORDS
diode-pumping, low-cost, Ti:sapphire laser, ultrashort pulse generation

1 | INTRODUCTION

In 1982, a novel laser material was born in MIT Lincoln Laboratory which called Ti:sapphire (Ti:Al₂O₃). Crystal of Al₂O₃ doped with Ti³⁺ are pink, and perhaps its special pink appearance is destined for an extraordinary life. At present, Ti:sapphire lasers still occupy an important position in scientific and industrial applications. The characteristics of laser materials have been described in detail from earlier publications showing broadband absorption centered in the blue-green wavelength region (400–600 nm) and associated broadband emission (650–1150 nm) at red-near IR wavelengths, which make it one of the best materials for ultrashort pulses generation and wavelength tuning.

The pump sources of Ti:sapphire lasers include flashlamp, Argon-ion laser, frequency-doubled diode-pumped solid-state laser (DPSSL), frequency-doubled fiber laser, optically-pumped semiconductor laser (OPSL), and frequency-doubled laser diode. At one time, Argon-ion lasers were also the mainstream pump source for Ti:sapphire lasers, which slowly withdrew from people’s view due to their large size, complexity, and maintenance difficulties. The flashlamp pumped Ti:sapphire laser could yield large amounts of energy, its operating life and conversion efficiency are limited. However, the flashlamp pumping technology still used in extremely large devices. In addition, DPSSL, frequency-doubled fiber laser, OPSL and frequency doubled laser diode all use frequency conversion technology to obtain blue-green laser, which undoubtedly increases the complexity and reduces the reliability of the system. Today, the most widely used pump sources are DPSSLs, which could emit tens of watts of output power with high beam quality. Everything is not so perfect, the prices are too expensive, which has become its criticism. Therefore, employing an inexpensive and reliable pumping method also become a trend for Ti:sapphire lasers, just like the laser diode direct-pumped solid-state lasers.

Semiconductor laser diodes (LDs) are widely used as pump sources because of low costs, small sizes, high efficiencies and long lifetimes. With the development of gallium nitride (GaN) semiconductor materials, it is possible to pump Ti:sapphire directly by GaN-based LDs. At present, multi-mode blue and green LDs can reach watt-level output power, while the maximum output power of blue (448–472 nm) LDs is up to 5 W and that of green (518–532 nm) LDs is still limited at 1.5 W. In 2009, Roth et al. used a 1 W 452 nm GaN LD to pump the Ti:sapphire crystal and realized 19 mW continue wave (CW) laser. At this point, the research of LD directly pumped Ti:sapphire officially kicked off.
off. In this article, we review the progress of LD-pumped Ti:sapphire lasers in the past 12 years.

The structure of this work is as follows. The current status of blue-green LDs and Ti:sapphire materials are introduced in Section 2. Section 3 mainly introduces the development status of LD-pumped Ti:sapphire lasers, including blue LD pumped Ti:sapphire oscillators, green LD pumped Ti:sapphire oscillators, dual-color LDs pumped Ti:sapphire oscillators, and LD directly pumped Ti:sapphire regenerative amplifier. In Section 4, some related applications of LD pumped Ti:sapphire femtosecond lasers are presented, such as optical frequency combs, multi-photon imaging. Finally, the development of LD directly pumped Ti:sapphire lasers is summarized and further prospected.

2 CURRENT STATUS OF BLUE-GREEN LASER DIODE AND TI:SAPPHIRE LASER MATERIALS

The three main elements of a conventional laser are pump source, gain medium and resonant cavity. There’s no doubt that the current status of the pump source and gain medium can greatly affect the output of the laser system. As the change in pumping method, it will certainly bring new opportunities to the Ti:sapphire laser. The performance of these early LD pumped Ti:sapphire laser systems was moderate. In fact, they were still limited by the output power of blue-green LDs. Therefore, the development status of GaN-based LDs is very important. We will give a brief introduction on it. Meanwhile Ti:sapphire is considered to be a very mature laser crystal and is well known for its various optical properties. However, when they encountered blue LDs, some novel phenomena appeared, which cannot be fully explained yet. These may be due to the inherent defects introduced in the growth process of Ti:sapphire crystal. It’s also possible that blue LD pumping amplifies such inherent defects in the crystal, rather than green pumping.

2.1 Blue-green laser diode

In 2001, Nichia Chemical Corporation invented the first GaN-based blue LDs with a wavelength of 450 nm, which had a lifetime of approximately 200 h at a constant output power of 5 mW. In the process of optimizing the structure of epitaxial layers, device structures and packages, the performance of GaN-based blue LDs had been greatly improved. In 2008, GaN-based 1-W blue LD at 445 nm wavelength was fabricated successfully. Today the output power of blue LDs is already up to more than 5 W with a much longer lifetime.

However, the challenges of fabricating longer wavelength GaN-based LDs are numerous and become more severe as the laser wavelength increases, which means that it is more difficult to manufacture high power GaN-based green LDs than blue LDs. The peak gain of GaN-based LDs decreases as the emission wavelength increases from violet to green range, which will result of the much lower efficiency of the green LDs than that of blue LDs. Therefore, the output power of the green LDs has been relatively low for a long time. Till 2013, watt-level green LDs at 525 nm came out. Now their output power has been increased to 1.5 W.

At present, Nichia Corporation can provide a variety of blue-green LDs, which represent the top level of the industry. If only considering the price and output power, it is a good choice to use blue LDs to pump the Ti:sapphire laser. Of course, it also involves some other issues, which will be explained later. Even some research institutions can provide high-power fiber-coupled LDs, such as Frankfurt Laser Company, DILAS, Institute of Semiconductors (Beijing). Such high-power LDs can provide much powerful pump power for Ti:sapphire lasers.

As the power of blue-green LDs gradually increases, they can be widely used in displays, lighting, quantum technology, materials processing, and so forth. The large market demand will also further promote the development of GaN-based LDs.

2.2 Ti:sapphire laser materials

The main growth methods for Ti:sapphire laser crystals include vertical-gradient-freeze (VGF) technique, heat-exchanger method (HEM), Czochralski method (CZM), flame-fusion method (FFM), horizontally directed crystallization method (HDCM), and Kyropoulos (Ks) technique. The largest Ti:sapphire crystal of 235 mm diameter was currently produced by HEM and was successfully used in the 10 PW system. In addition to general quality problems such as scattering particles, doping uniformity, optical uniformity and stress, laser crystals grown based on various methods have a common important quality parameter-figure of merit (FOM) value, which acts an important role for Ti:sapphire crystal quality. The FOM value is strictly defined as $\alpha_{490}/\alpha_{800}$, where $\alpha_{490}$ and $\alpha_{800}$ are the absorption coefficient at the pump wavelength of around 490 nm and absorption coefficient associated with the parasitic absorption by $\text{Ti}^{3+} - \text{Ti}^{4+}$ pairs at about 800 nm, respectively. In some cases, the FOM could be also defined as $\alpha_{394}/\alpha_{820}$ or $\alpha_{532}/\alpha_{800}$ by the crystal suppliers. At present, the relatively mature doping concentration is 0.03–0.25 wt%, which can ensure a high FOM value. When the doping concentration increases, the pump absorption coefficient and the parasitic absorption coefficient increase simultaneously, which will lead to a decrease in the effective FOM value of Ti:sapphire crystal and the optical properties will deteriorate. However, the parasitic absorption can be further reduced through the improvement of the growth process and the later annealing treatment. Ti:sapphire with high doping concentration will
reduce the difficulties of laser cavity design and help to obtain higher output power. The growth of Ti:sapphire crystals with high doping and high FOM value is also a challenging task.

3 | LASER-DIODE PUMPED Ti: SAPPHIRE LASERS

Advances in materials have led to opportunities for the development of lasers. GaN-based LDs were introduced into the Ti:sapphire lasers for the purpose of reducing the costs, simplifying the system and further expanding their applications. The beam quality of LD is extremely poor and requires the necessary beam shaping to improve the mode matching. Moreover, all current works have done based on such a model. Since 2009, the performance of LD-pumped Ti:sapphire lasers has been gradually improved, approaching the performance of traditional Ti:sapphire lasers in terms of high power, wide wavelength tunability, and short pulse duration.

3.1 | Blue-laser-diode pumped Ti:sapphire oscillators

In Reference 19, initially determined the keynote of the entire LD-pumped Ti:sapphire research work. A simple three-mirror cavity was used to realize the CW operation. However, as shown in Figure 1(A), the CW optical power obtained using the 452 nm LD pumping is much lower than that of 532 nm pumping and exhibiting a higher pump threshold. It is understandable that the crystal (5 mm long, 0.25 wt% doping concentration and FOM = 400) has a lower absorption fraction (65%) to 452 nm LD pump light than that of 532 nm pumping (87%), due to deviation from the absorption peak. Over a few minutes, the laser output power continued to decline (from 60 mW down to 19 mW), and then kept stabilized. What’s the most interesting is that once output power stabilizes, it will remain a constant for a long time. When the oscillator is turned on next time, the output power will immediately reach the previous stable value (19 mW). However, the laser performance could be restored to the initial level by 532 nm pumping, which required exposure for tens of minutes. Such laser behavior implies an underlying loss mechanism and is not caused by thermal and mechanical reasons. In order to further study the loss mechanism, the combined beam of a wavelength-tunable Argon-ion laser and a 532 nm DPSSL was used to pump the Ti:sapphire laser. At first, it was further confirmed that such a loss was dependent on the pumping wavelength (Figure 1(B)). This laser behavior was considered to be caused by charge transfer interactions in Ti$^{3+}$ – Ti$^{4+}$ pairs.19 Subsequently, based on this work, they achieved passive mode-locked operation with output power of 101 mW and pulses duration of 111 fs using semiconductor saturable absorber mirror (SESAM).21

Durfee et al. achieved LD-pumped KLM femtosecond Ti:sapphire laser for the first time in 2012.64 For a KLM laser directly pumped by LDs, the challenge is the low brightness of the pump source and poor mode matching. They innovatively combined the two LDs in space and focused them inside the crystal, which increased the brightness of pump beam and the power density of the crystal to a certain extent.69 The emitting structure of LD determines that the pump light can only be matched with the cavity mode in the vertical direction, which will lead to strong gain differences.70 The spectral range of the LD-pumped KLM Ti:sapphire laser was approximately from 725 to 875 nm, and the spectral shape was controlled by the prisms and curved mirror positions. Pulses as short as 15 fs were obtained measured by FROG.

Five years later, Backus et al. used a 3.1 W 465 nm LD instead of the previous complicated combined beam pump structure to pump the Ti:sapphire laser.65 As shown in

![Figure 1](image-url)
Figure 2(A), the mode-locked spectrum was slightly wider than previous results. A pulse duration of 13.5 fs was obtained with 140 mW output power by external compression (Figure 2(B)). In the following year, Kopylov et al. achieved comparable output parameters, namely a pulse duration of 15 fs and an average power of 170 mW.

It is not easy for a mode-locked laser to obtain high power and short pulses simultaneously, especially in the case of LD pumping. In 2017, Rohrbacher et al. employed two 2.9 W 450 nm blue LDs to bilaterally pump a 4 mm long Ti:sapphire crystal, and used a SESAM to start and maintain the mode-locking operation. The mode-locked oscillator could obtain a maximum output power of 460 mW with 82 fs pulse width. The optical-to-optical conversion efficiency of blue LD directly pumped Ti:sapphire mode-locked laser is close to 8%. If the mode matching can be further optimized, the efficiency will be higher. The full width at half maximum (FWHM) spectral bandwidth was 9.4 nm at the central wavelength of 784.5 nm. The mode-locked spectrum was limited by the bandwidth of the SESAM.

Coyle et al. used two 3.5 W 450 nm blue LDs to pump Ti:sapphire oscillator and achieved SESAM passive mode-locking and KLM with wavelength tunability. The tunable wavelength bandwidths of SESAM mode-locked laser and KLM laser were 37 nm and 120 nm, respectively. The maximum output power of SESAM mode-locked laser (433 mW) was higher than that of KLM laser (382 mW). The pulse width of SESAM mode-locked Ti:sapphire laser was 85 fs at the maximum average power of 433 mW. In KLM operation, the pulse width was as short as 66 fs at the maximum average power (382 mW). In Refs. 60,61, fully demonstrated that blue LDs pumped Ti:sapphire oscillator had the potential for short pulses generation and high power output.

In 2019, we reported a blue LD directly pumped wavelength tunable KLM Ti:sapphire laser. A 3.5 W 450 nm LD was used to pump a four-mirror cavity with the intra-cavity dispersion compensation by prisms. The tuning bandwidth of the LD-pumped KLM Ti:sapphire laser was 89 nm which was from 736 to 825 nm. In this wavelength tuning process, the spectral width and output power varied with the central wavelength. The shortest pulse duration of 17 fs was obtained at the central wavelength of 736 nm.

Next, we used double-chirped mirrors (DCMs) and prism pairs to achieve fine compensation of intra-cavity dispersion, as shown in Figure 3(A). A broad spectrum covering from 650 to 950 nm (Figure 3(B)) was obtained, which could support 7.6 fs Fourier transform limited (FTL) sech²-shaped pulses (inset in Figure 3(B)). Through the extracavity dispersion compensation, 8.1 fs pulse duration was realized (Figure 3(C)). It was the first sub-10 fs Ti:sapphire oscillator directly pumped by LD.

We also conducted research on output power scaling. Two 3.5 W 465 nm LD were used to pump Ti:sapphire simultaneously, while the CW output power exceeded 600 mW. Then a SESAM (BATOP, SAM-800-1-5 ps) was inserted in the cavity to initiate passive mode-locking. The pulse duration was measured to be 12 ps (assuming sech² pulse shape) with average power of about 220 mW. The power reduction is mainly due to the loss introduced by the SESAM.

### 3.2 Green-laser-diode pumped Ti:sapphire oscillators

In contrast to blue LDs, the emission wavelength of green LDs is closer to the absorption peak of Ti:sapphire and thus are more suitable as the pump sources. In 2014, the first green LD pumped Ti:sapphire femtosecond oscillator was realized by Sawai et al. Laser pulses as short as 62 fs and output power up to 23.5 mW were obtained, which opened the door for green LD pumping. K. Gürel et al. used two 520 nm 1 W green LD as the pump sources for Ti:sapphire laser, and adjusted the output power of each LD to 1.5 W by means of overdriven current. Figure 4 shows that a standard X-fold cavity with tight focusing. Using Gires-Tournois interferometer (GTI) dispersion compensation mirrors made the LD pumped KLM Ti:sapphire laser more compact. The maximum KLM output power was 450 mW with 58 fs pulse width and 16.7 nm spectrum bandwidth. Moreover, the
mode-locked laser ran very stably at a repetition rate of 420 MHz. Muti et al. designed a single green LD pumped femtosecond multipass-cavity (MPC) Ti:sapphire laser and increased the pulse energy up to 5 nJ. Miao et al. achieved watt-level CW laser output from a Ti:sapphire oscillator directly pumped with 21 W 517 nm green LD module in 2019. The optical-to-optical efficiency was about 6.3% and the pump threshold exceeded 5 W.

3.3 Dual-color laser diodes pumped Ti:sapphire oscillators

The purpose of dual-color LDs pumped Ti:sapphire laser was to weaken the pump-induced loss introduced by blue LDs pumping and to increase the output efficiency. Sawada et al. used 451, 478, and 520 nm LDs to successfully verify the theory in Ref. 19 that pump-induced loss depends on the pump wavelength. As shown in Figure 5(B), the charge transfer process of Ti$^{3+}$–Ti$^{4+}$ pairs was described in detail, and the threshold of charge transfer of the ground states Ti$^{3+}$–Ti$^{4+}$ and the ground states Ti$^{4+}$–Ti$^{3+}$ were about 4.7 and 4.17 eV, respectively. They also assumed the existence of excited state absorption (ESA) to further explain the pump-induced loss. However, based on the previous works, there is no expected ESA for Ti:sapphire, due to its simple 3d$^1$ configuration. So its accuracy remains to be verified. Moulton et al. believed that the model of the charge transfer of Ti$^{3+}$–Ti$^{4+}$ pairs have not been perfect enough to fully explain related problems of pump-induced loss. As shown in Figure 5(A), a dual-color pump beam (4 W in total) composed of LDs with wavelengths of 478 nm and 520 nm was used to pump an X-

FIGURE 3 (A) Experimental setup of the blue LD-pumped Ti:sapphire oscillator. (B) Mode-locked spectrum with multi-peaks. Inset shows the FTL pulse in the time domain. (C) Interferometric auto-correlation (IAC) trace of the pulse generated from the Ti:sapphire laser (blue curve) and theoretical IAC envelope of a sech² pulse of 8.1 fs (red curve). Reprinted with permission from Ref. 68, © Chinese Laser Press [Color figure can be viewed at wileyonlinelibrary.com]

FIGURE 4 Cavity picture with pump beam drawn in green and the oscillating laser beam in red. Laser setup for KLM operation. Reprinted with permission from Ref. 59, © The Optical Society [Color figure can be viewed at wileyonlinelibrary.com]
shaped cavity with a 2.5 mm long Ti:sapphire, and a CW output power of 593 mW was obtained. On the basis of this work, SESAM mode-locking (315 mW, 126 fs) and KLM (360 mW, 48 fs) were both realized. The dual-color blue-green LD pump structure has significant advantages. First, 478 and 520 nm pumping can reduce the pump-induced loss in a LD pumped Ti:sapphire laser. Secondly, it can provide a higher and more effective pump power. The complex kinetic process caused by dual-color pump remains to be studied. Further optimization of the mode matching, crystal parameters, and the transmission rate of the output coupler is expected to achieve higher mode locked output power through such a dual-color pump structure.

3.4 | Blue laser diode module pumped Ti: sapphire regenerative amplifier

Researchers are eager for the laser output power to be further scaled. In 2017, KMLabs reported on a direct LD pumped Ti:sapphire ultrafast regenerative amplifier laser system. The regenerative amplifier was directly pumped by a 50 W fiber-coupled LD and achieved 3 W CW laser output power by removing the polarization components (polarizer, Pockels cell). In order to further test the 100 W pump (Figure 6(A)), they also built a double focus CW cavity, and then obtained a CW output power of 11 W corresponding to a slope efficiency of 26%. Figure 6(B) reveals an important message that when using the high-power blue LD pumping, cryogenic cooling of Ti:sapphire crystal is necessary, otherwise there is no laser action. When the Ti:sapphire crystal was at ~188 K, the fluorescence intensity of 450 nm LD pumped crystal exceeded that of 532 nm pumping, and continued to rise as the temperature decreased. At present, this laser action could be for two reasons that the thermal lens effect and non-radiative decay channels. A KLM Ti:sapphire oscillator was pumped by a green 520 nm LD as the seed source, while the amplifier was pumped by two 50 W, 450 nm fiber-coupled LD. 32 nm FWHM spectral bandwidth was obtained from the amplifier system, which supported 30 fs pulse width. The laser amplifier system produced 3.8 μJ pulse energy at a repetition rate of 250 kHz.
FIGURE 6  (A) Double focus CW cavity for high power 100 W tests. (B) Marked quenching of fluorescence from Ti:sapphire pumped with high-power 450 nm diodes. Left: 50 W input power, Ti:sapphire crystal at 295 K. Note that the cryocell window is dark. Right: same, but crystal at 93 K. Note that this behavior is not as marked with 532 nm pumping, suggesting new gain dynamics in the system. Reprinted with permission from Ref. 79, © The Optical Society [Color figure can be viewed at wileyonlinelibrary.com]

4 | NOVEL APPLICATIONS

Since the birth of LD pumped Ti:sapphire lasers, researchers have begun to explore advanced applications and have achieved a series of progress. As early as 2012, Young et al. applied LD pumped KLM Ti:sapphire laser to multiphoton imaging for the first time. This work was known as reducing the “entry” cost of the multiphoton microscope system. With the advancement of light sources, LD pumped Ti:sapphire laser with better performance had also been used in two-photon excited fluorescence (TPEF) microscopy and second harmonic generation (SHG) microscopy.

K. Gürel et al. realized the carrier-envelope offset frequency stabilization of green LDs pumped Ti:sapphire femtosecond laser and built the first LD pumped Ti:sapphire optical frequency comb (OFC). In 2019, Pablo Castro-Marín et al. also achieved a low-cost Ti:sapphire OFC based on a dual-wavelength LDs pumped Ti:sapphire femtosecond oscillator. LD directly pumped Ti:sapphire lasers can not only reduce system cost, but improve the time jitter and the signal to noise ratio (SNR) of the system.

As the output performance of LD-pumped Ti:sapphire lasers is further improved, they may be widely used in optical transfection, cellular microsurgery, THz radiation generation, spectroscopy, coherent high-harmonic EUV generation and so on. In the near future, low-cost and high-performance LD-pumped Ti:sapphire lasers will certainly blow a storm in many applications.

5 | CONCLUSION AND OUTLOOK

With the increasing maturity of GaN-based blue-green LDs, low-cost LD directly pumped Ti:sapphire lasers have become reality. Since 2009, the first LD directly pumped Ti:sapphire oscillator came out. After more than 10 years of development, the output parameters of LD directly pumped Ti:sapphire lasers have been significantly improved and are gradually approaching those of traditional DPSSL pumped Ti:sapphire lasers. LD directly pumped Ti:sapphire femtosecond lasers have been applied to OFC and multiphoton imaging, and have shown excellent performance.

At present, the performance of fiber coupled LD is improving rapidly, namely output power increases and fiber diameter decreases. These developments are all creating conditions for low-cost Ti:sapphire lasers. There are still many scientific and engineering problems to be solved for Ti:sapphire laser with high output power and short pulse duration. To further challenge the output limit of femtosecond Ti:sapphire laser and expand its application range is the motivation and research significance of LD directly pumped Ti:sapphire laser.

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are openly available in The Optical Society at https://www.
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