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Citation for published version:
Brown, C, Dickinson, D & Hands, P 2021, 'Diode pumping of liquid crystal lasers', *Optics and laser technology*, vol. 140, 107080. https://doi.org/10.1016/j.optlastec.2021.107080

Digital Object Identifier (DOI):
https://doi.org/10.1016/j.optlastec.2021.107080

Link:
Link to publication record in Edinburgh Research Explorer

Document Version:
Publisher's PDF, also known as Version of record

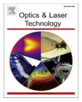
Published In:
Optics and laser technology

Optics & Laser Technology 140 (2021) 107080

Contents lists available at ScienceDirect

Optics and Laser Technology

journal homepage: www.elsevier.com/locate/optlastec



Full length article

Diode pumping of liquid crystal lasers

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ARTICLE INFO

Keywords:
Liquid crystal lasers
Band-edge lasing
Organic lasers
Laser diode pumping
Dye lasers
Liquid crystals

ABSTRACT

Liquid crystal lasers are typically pumped with Q-switched lasers, which provide the short pulse durations necessary to produce band-edge liquid crystal laser emission. However, the current cost and bulk of the pump laser, relative to the liquid crystal component, limit the application potential of liquid crystal laser technology. A low-cost and compact alternative pump source has often been discussed within the liquid crystal laser community but has not yet been demonstrated. In this paper, a 445 nm laser diode was integrated with carefully selected electronics in an attempt to reproduce equivalent pump conditions to that achieved from a Q-switched laser, and then used to pump a liquid crystal laser. Successful band-edge lasing was achieved, demonstrating, for the first time, a clear threshold and single mode, narrow linewidth emission. Red, green and blue outputs from different liquid crystal lasers were also realised when pumped with the same laser diode, demonstrating the broad wavelength selectivity of liquid crystal lasers using a single low-cost, compact device.

1. Introduction

Since the first patent in the 1970s [1], and subsequent developments [2–4] leading to unequivocal experimental evidence in 1998 [5], photonic band-edge liquid crystal (LC) lasers have become established as an exciting research field within photonics and materials science. Their small footprint [6,7] and large wavelength selectability, from the UV to the near-IR [8,9], combined with low fabrication costs, low thresholds (typically nJ - μJ/pulse) and self-organising structure, have great potential in offering solutions in applications such as medical imaging and displays.

Conventionally, photonic band-edge LC lasers comprise a chiral nematic (N*) LC which forms a self-organised resonant cavity when optimally aligned in a “cell” consisting of two parallel glass substrates. A periodic change in refractive index of the N* due to the chiral helix, results in a photonic band-gap (PBG). The PBG reflects normally incident light of the same handedness of circular polarisation as the helix across a wavelength range determined by the birefringence and helical pitch [10]. Wavelengths outside the PBG, and light with an opposite handedness of polarisation to that of the helix, are transmitted. When a gain medium, in the form of an organic dye, is added to the N*LC and optically pumped with an external source, lasing is possible. This is contingent upon the dye having an absorption spectrum corresponding to the wavelength of the pump source and a fluorescence spectrum that overlaps with the (typically long) edge of the PBG [11]. The output from an LC laser is characterised by circularly polarised light emitted through two counter propagating emission cones parallel to the helical axis, (normal to the cell for substrates with planar surface-alignment treatment). If one of the inner surfaces is coated with a reflective material, one of the emitted laser beams undergoes a phase change upon reflection and both emission cones emerge from the same side of the cell. The net output is therefore linearly polarised as a result of the superposition of left-handed and right-handed circularly polarized light [12]. Photonic band-edge LC lasers also exhibit narrow linewidth emission, and a clear gradient change in the slope efficiency upon lasing, defined as the laser threshold [13]. The position of the band-edge, and hence the LC laser emission wavelength, is determined by the degree of chirality of the N*LC and choice of laser dye.

LC lasers are typically pumped with Q-switched lasers (most frequently at 532 nm) as they provide the peak energies required to overcome the LC laser threshold. (Although LC laser thresholds are typically described in terms of pulse energy, it has been shown that the energy *density* (energy per pulse per unit area) provides a more accurate description of the laser threshold specifications, as it incorporates the spot size of the pump source [14].) Moreover, Q-switched lasers are capable of producing nanosecond (or shorter) pulse durations. This is essential to prevent triplet state formation, molecular reorientation and thermal damage that all lead to degradation in LC laser performance (increased threshold and reduced efficiency) caused by longer pulse lengths [15–18]. However, the size and cost of LC lasers are currently

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https://doi.org/10.1016/j.optlastec.2021.107080
Received 30 October 2020; Received in revised form 22 February 2021; Accepted 9 March 2021

limited by the relatively bulky and expensive Q-switched pump source. The limited available wavelengths of Q-switched pump lasers (especially in the blue-violet region of the spectrum) also severely hinder the ability to make LC lasers that are tuneable across the full visible spectrum. A more versatile, compact and lower cost pump could offer a significant advantage, as well as commercially scalable fabrication, and opportunities for integration into future portable optical systems. Laser diodes (LDs) or light emitting diodes (LEDs) would offer an ideal solution [18–22], especially in the blue-violet region of the spectrum. While LD pumping has been successful with conventional dye lasers [23–28], the few attempts at semiconductor-based pumping of LC lasers have yet to demonstrate single-mode, narrow linewidth and stable emission. An attempt at LC laser pumping using high intensity pulsed blue and green LEDs as pump sources was conducted by Coles *et al.*, resulting in some evidence of weak multimodal stimulated emission [29]. Munoz *et al.* attempted pumping with continuous wave sources from an LED and a Nd:YAG laser in which they claimed narrow linewidth LC laser emission [30]. However, in both studies, the output spectra were dominated by dye fluorescence upon which were superimposed multimodal narrow linewidth features. With reference to [30], Ortega et al. concluded the results did not sufficiently prove the presence of lasing and were more consistent with amplified spontaneous emission [22]. While both studies highlight the potential of semiconductor pump sources, neither address the requirement of the short pump pulse durations necessary for LC laser operation, as previously discussed. As such, narrow linewidth single-mode LC laser emission has yet to be demonstrated with a semiconductor pump source. The required combination of high pulse energy and short pulse duration is limited by the capabilities of semiconductor devices and electronics. However, with recent improvements in both technologies, we hypothesised that narrow linewidth single-mode LC laser emission ought to be possible using a diode laser pump source, potentially achieving comparable LC laser emission properties to that of a Q-switched pumped system. We based this hypothesis on the following assumptions:

1. A laser cell can be fabricated, for which a low lasing threshold (of the order of 10 mJ/pulse/cm^2 [14,15,31]) is optimised by implementing previously published results, such as: the use of a reflective cell geometry [32,33]; optimised cell spacing between 10 and 20 µm [18]; and the use of a high quantum efficiency laser dye at optimal concentration [34].
2. High power laser diode (LD) technology is implemented, capable of producing >1 W optical output from a single source, enabling nJ/pulse energies in nanosecond pulse durations.
3. Diode driver electronics with the ability to deliver the diode's maximum current in 10 s of nanoseconds (or less) can be sourced.

This paper aims to test the hypothesis that narrow linewidth single mode emission is achievable through LD pumping of a LC laser. Low threshold behaviour is first confirmed by fabricating an optimised LC laser cell with high quality alignment and pumping it using a 532 nm Q-switched laser source. A high power, pulsed LD is then used to pump the same cell and the emission characteristics are compared to the previous Q-switched pump results to confirm equivalent operation. Multiple output wavelengths ranging from the blue to red regions of the spectrum are also presented by pumping different cells with the same LD pump source, to further demonstrate the versatility of the system.

2. Method

2.1. Liquid crystal laser fabrication

The fabrication of the LC lasers was carried out in a Class 10 semi-industrial cleanroom facility to enable high quality cell construction. The commercial nematic LC BL006 (*Merck*) was selected for its high birefringence [12] and used throughout the investigation. This was doped with different concentrations of the high twisting power chiral dopant BDH1281 (*Merck*) to deliver long photonic band-edges at the desired emission wavelengths in close proximity to the fluorescence maxima of the laser dyes. Different organic laser dyes (*Exciton*) were selected to produce the desired output wavelength. 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostryl)-4H-pyran (DCM) is a well-understood four-level laser dye [35] and was chosen because of its mutual absorption at the two pump wavelengths of interest: 532 nm for the Q-switched laser and 445 nm for the LD. This was experimentally confirmed, with the absorption coefficient of DCM at 445 nm being 20% higher than at 532 nm. The two further dyes, Coumarin 504 (C504) and Coumarin 540A (C540A), were chosen based on their absorption efficiency at 445 nm and their output spectra in the blue and green regions of the spectrum respectively [26,36]. The relative concentrations were based on best available data and in-house experimental evidence of optimum performance for DCM [34], C540A [37] and C504.

A reflective cell geometry was chosen based on the evidence published in [33] in which the laser threshold was reduced by 25% when compared to transmissive cells. The laser cells comprised two $15 \times 15 \times 3$ mm glass substrates *(Laser 2000)* with an aluminium coating on the inside surface of one of the substrates (Fig. 1). The inner surfaces of each cell were coated with polyimide *(SE1410, Nissan Chemical Industries)* and antiparallel rubbed to promote homogenous planar surface alignment of the LC director, and thus form a self-organising standing helix [38–40]. The substrates were bonded together using a UV curable adhesive *(NOA68, Thorlabs)* mixed with 10 µm diameter silica spheres *(Nanjing Jianzun Glass Microsphere Plant Company Ltd)*, the closest available spacing to the published optimum of 14 µm [18]. Cells A, B and C were capillary filled with the corresponding mixtures shown in Table 1. The cells were thermally annealed on a hotplate at 100 °C, just below the chiral nematic-isotropic transition temperature, and slowly cooled to room temperature to promote a high quality monodomain texture. The edges of the cells were later sealed with Torr Seal (*Thorlabs*).

Once made, these cells were visually inspected to verify the presence of a well-aligned, planar texture necessary for high performance lasing.

2.2. Optical experiments

The pump lasers used were a 532 nm Nd:YAG Q-switched laser *(Crylas FDSS532-Q2, pulse duration* $\leq$ *1.3 ns)* and a 445 nm LD *(Nichia, NUBM44)*. Initially, a LD with the same wavelength as the Q-switched laser was considered, in order to directly compare the different pump methods. However, the 445 nm LD was found to have the highest power rating, 3.5 W, and was within the absorption spectrum of the DCM dye. This was therefore chosen to maximise the chances of overcoming the LC laser threshold. The LD was soldered onto a driver board *(PicoLAS GmbH, LDPV_50100_V3.3)* and connected to a signal generator *(PicoLAS GmbH, PLCS-21)* that produced a pulse duration of 16 ns, and was measured to deliver a maximum pulse energy of 51.6 nJ. Both lasers were set to a repetition rate of 1 Hz for consistency and to minimise possible thermal heating and optical reorientation effects within the LC laser.

The optical arrangement for pumping the LC laser is shown in Fig. 2. The energy of the linearly polarised pump beam incident upon the LC laser cell was controlled by rotating the half waveplate (with antireflective coatings designed for each pump source) relative to the static Glan-Thompson linear polariser. A quarter waveplate was then used to produce circularly polarised light of the opposite handedness of the chiral helix to ensure maximum transmission of the pump laser by the LC and hence maximum absorption of the pump beam by the laser dye. The focussed pump laser spot size at the plane of the LC laser cell was measured using a knife edge technique [41], and found to be approximately circular with a diameter < 14 µm for the Nd:YAG pump, and slightly elliptical for the LD pump with major and minor axes of <12 and <9 µm respectively. Long-pass dichroic mirrors *(Thorlabs*

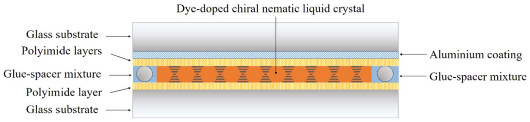

Fig. 1. LC laser cell cross-section, with reflective geometry for unidirectional emission.

Table 1
Relative dopant and dye concentrations for three different laser dyes and their resultant output laser wavelengths.

Cell	Chiral Dopant Concentration ($\%_{wt}$)	Dye	Dye Concentration ($\%_{wt}$)	Output Laser Wavelength (nm)
A	4.35 ± 0.05	DCM	1.50 ± 0.05	610
B	5.37 ± 0.05	C504	0.89 ± 0.05	480
C	4.89 ± 0.05	C540A	1.00 ± 0.05	530

DMLP550R for the 532 nm Q-switched pump; Chroma ZT442rdc for the 445 nm LD pump) allowed separation of the input and output beams. The output wavelength and linewidth were determined from the data recorded by a spectrometer *(Ocean Optics, USB4000, resolution ~1.5 nm)*. An energy meter *(Ophir, PD10-pJ-C)* was used to measure the pulse energy from the LC laser, having first calibrated the waveplate for the pump energy in order to calculate the threshold and efficiency. An additional long-pass filter was positioned after the dichroic mirror to remove any extraneous pump light which could cause erroneous energy measurements.

3. Results and discussion

3.1. Verification of band-gap and liquid crystal alignment quality

Photonic band-gap spectra were obtained to confirm that the long band-edge of the band-gap was located close to the fluorescence maximum of the DCM dye (~600 nm [9]). Reflective geometry cells reflect not only the band gap from the chiral nematic LC, but also reflect light outside the band gap from the aluminium layer. A transmissive cell geometry (i.e. with no reflective aluminium layer) is therefore required to carry out these specific measurements. A commercial transmissive cell *(Instec LC2-20.0)* was filled with the chiral nematic LC mixture (without the dye) described for Cell A in Table 1. The solid black lines in Fig. 4a and c show the transmission spectra of a white light source *(Ocean Optics HL-2000-CAL)* and shows the expected 50% reflectance from the PBG region between approximately 500–600 nm. The long band-edge was confirmed to be close to 600 nm, as required.

After the DCM dye was added to the N*LC and the mixture was filled into our reflective cells, further verification of the quality of the cell and alignment of the LC was carried out using white light microscopy. Fig. 3

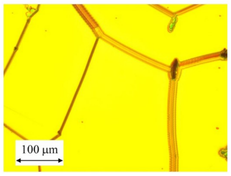

Fig. 3. Microscopy image of reflective chiral nematic LC laser cell, showing large monodomain structure with characteristic oily streak defect lines between the well-aligned chiral nematic domains.

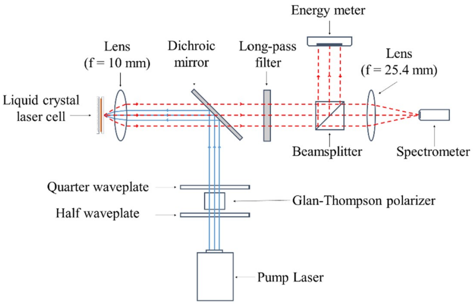

Fig. 2. Experimental set-up used to optically pump reflective cells A, B & C.

shows a uniform texture with large monodomains (>200 μm; significantly larger than the focussed pump spot size), with the characteristic oily streak defect lines, often found between well aligned neighbouring chiral domains [42]. The cell was therefore considered to have been well prepared and (if appropriately excited) was predicted to yield lasing emission with low threshold and narrow linewidth.

3.2. *Q-switched pumping*

The lasing threshold, wavelength, linewidth and polarisation output of the DCM-doped LC laser cell were first measured when pumped with the 532 nm Q-switched laser.

Cell A was positioned at the focal point of the lens. A narrow linewidth (<1.5 nm FWHM, limited by the resolution of the spectrometer) LC laser output at 610 nm was observed, close to the fluorescence maximum and coincident with the measured band-edge position, as shown in Fig. 4a. The output was confirmed to be mostly linearly polarised, as expected for a cell with reflective geometry. The output energy was plotted as a function of the input energy to determine the slope efficiency and threshold (Fig. 4b). A clear change in the gradient of the slope efficiency was observed, and above threshold a slope efficiency of 8.1% was measured, similar to those presented in [14,34] for equivalent 532 nm Q-switch-pumped DCM-doped LC lasers. As expected, a low threshold was confirmed, and calculated to be 7.4 nJ/pulse (6.93 mJ/pulse/cm^2). Overall, these data confirm that photonic band-edge lasing was occurring within our LC laser cell, and that LD pumping ought to be hypothetically possible with our hardware, as the threshold was below the maximum pulse energy measured for the laser diode.

3.3. *Laser diode pumping*

As before with the Q-switch-pumped system, the LC laser linewidth and wavelength were recorded on the spectrometer and the input and output energies measured on the energy meter. It was found that LD pumping successfully produced a LC laser output beam with near-identical characteristics to results obtained with the same cell when pumped with the Q-switched laser. Once again, a narrow linewidth (<1.5 nm FWHM) single-mode output at 610 nm was produced, coincident with the long wavelength band-edge of the PBG (Fig. 4c). The same degree of linear polarisation as the results obtained with the Q-switched-pump was confirmed, as was the presence of a distinct gradient change of the slope efficiency at the point of threshold (Fig. 4d).

From the point of intersection of the two linear fit lines in Fig. 4d, the threshold of the LD pumped LC laser was found at 12.3 nJ/pulse (15.7 mJ/pulse/cm^2), with the slope efficiency above threshold of 3.1%.

The Q-switch-pumped LC laser efficiency data in Fig. 4b, with a clear gradient change at threshold, is typical behaviour for a band-edge laser. The near-identical behaviour of the LD-pumped case in Fig. 4d is therefore the first unequivocal evidence of successful diode-pumped LC lasing. Furthermore, the narrow linewidth output, identical for both pumping regimes (Fig. 4a and b) is additional verification that the LC laser is operating above threshold. The nanosecond-scale pulsing of the laser diode is what distinguishes this work from previous attempts [29,30] and is likely the principal reason for our successful LC laser emission. The higher power output of the laser diode compared to the LEDs used in previous work will also be a contributing factor.

Given the slightly higher absorption of DCM at 445 nm compared to 532 nm, it might be reasonable to expect a lower threshold and higher

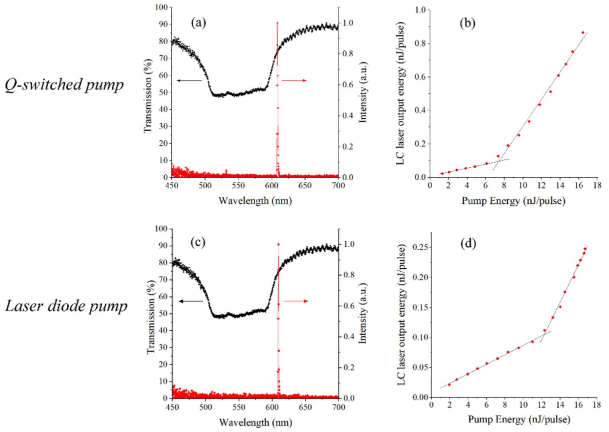

Fig. 4. LC laser output characteristics: a) narrow-linewidth lasing at the long band-edge at 610 nm when pumped with the 532 nm Q-switched laser and b) associated distinct gradient change indicating threshold energy. c) Near-identical narrow laser emission at 610 nm when pumped at 445 nm with the laser diode with d) distinct gradient change at a slightly higher threshold energy attributed to the longer pulse duration of the laser diode. Each data point is the average of 60 sequential pulses at 1 Hz repetition rate.

efficiency with the LD than with the Q-switched pump. However, we observe the opposite trend here. We attribute this to the LD's longer pulse length (16 ns compared to 1.3 ns), which has previously been shown to negatively impact LC laser thresholds [15–18], and appears to be the dominant effect here.

The LD-pumped LC laser displayed a lower maximum output pulse energy compared to the Q-switch-pumped system. This is to be expected due to the higher peak powers associated with Q-switching, relative to laser diodes. With continued improvements in laser diode technology, it is expected that the discrepancy between the two pump source energies and pulse widths will be reduced, leading to further improvements in LC laser performance. We therefore anticipate that laser diode pumping will become a viable alternative to larger, more complex and expensive Q-switched pump laser sources.

To further validate the results and demonstrate the capabilities of this pump source, Cells B and C (as described in Table 1) were prepared and tested alongside Cell A. All were pumped with the same 445 nm LD. The results in Fig. 5 successfully show narrow linewidth LC laser emission at 480 nm, 530 nm and 610 nm. Intermediate wavelengths are also possible through the use of different chiralities and (if necessary) other dyes [9]. Whilst red, green and blue lasing from an LC laser have been demonstrated previously [29,43,44], these works required the use of a tuneable OPO pump laser, a nitrogen pump laser and a frequency tripled Q-switched pump laser, respectively, which are comparably much larger and more expensive than our laser diode pump source. This work therefore represents an exciting new opportunity for achieving wavelength-tuneable (or selectable wavelength) lasers within a compact package.

4. Conclusions

A laser diode has, for the first time, been successfully used to pump a LC laser to achieve single mode, narrow linewidth emission. The 445 nm laser diode enabled LC laser emission at 610 nm, with a clearly defined laser threshold at 12.3 nJ/pulse, and an identical output laser spectrum to the conventional Q-switch-pumped LC laser system. Despite these similarities, current achievable peak powers differ. However, LD power is expected to improve with continued advances in semiconductor-based technology. Furthermore, their size and cost means LDs could be combined into arrays and clusters to facilitate higher power LC laser devices [45,46].

Two further LC lasers with emission wavelengths at 480 nm and 530 nm were also achieved by pumping with the same laser diode source, demonstrating the broad wavelength range of our laser system, not otherwise achievable without the use of large and expensive pump sources. This development represents an opportunity to maximise the accessibility of tuneable (or selective) wavelength technology to a wider range of users.

Future work will investigate the performance limitations of the laser diode with a view to increasing LC laser output powers. Examples include increasing the LD current, optimising the pulse parameters and improving the LC laser fabrication process with a view to better understanding performance limitations and optimising capabilities. Future work will also explore the opportunities for LD-pumped LC lasers in applications including portable biomedical imaging.

Funding

This work was made possible by the Scottish Universities Physics Alliance (SUPA), Scottish Imaging Network: A Platform for Scientific Excellence (SINAPSE) & Scottish Universities Life Sciences Alliance (SULSA) [grant number H18034].

This work was also supported by the UK Engineering and Physical Sciences Research Council (EPSRC) [grant EP/R513209/1], and the EPSRC and the Medical Research Council [grant EP/L016559/1].

In accordance with funding requirements, data from this research can be accessed at https://doi.org/10.7488/ds/2809.

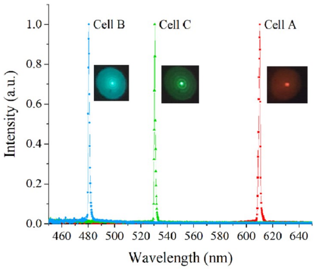

Fig. 5. Output intensity for three different cells when pumped with a 445 nm laser diode (lines overlaid as a guide) with inset images of each beam spot as illustrative examples.

CRediT authorship contribution statement

Calum M. Brown: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing - original draft, Writing - review & editing, Visualization, Funding acquisition. **Daisy K. E. Dickinson:** Validation, Investigation, Resources, Writing - review & editing. **Philip J.W. Hands:** Conceptualization, Resources, Supervision, Writing - review & editing, Project administration, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The authors would like to thank Richard Scott of the School of Engineering Electronics Workshop (University of Edinburgh) for his assistance with the construction of the laser diode electronics driver enclosure. Thanks also to Jason Norman and Danial Chitnis (University of Edinburgh) for their feedback and discussion.

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