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An experimental study of the effect of pump pulse duration on liquid crystal laser performance

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Much work has been done to understand the factors that impact photonic band-edge liquid crystal (LC) laser threshold and slope efficiency; two parameters often stated to quantify performance. Conventionally, LC lasers are optically pumped using Q-switched lasers with a fixed pulse duration, and thus the effect of pump pulse duration on LC laser performance has received little attention. Whilst some studies have been published at different pump pulse durations, these use different laser sources and experimental conditions, making the data incomparable. By exploiting a recent breakthrough in laser diode pumping, our experimental results prove and quantify the detrimental effect of an increase in pump pulse duration on LC laser performance. We also show that the dependency of threshold on pump pulse duration depends upon how threshold is defined, due to an ambiguity in the definition of pulse energy in systems where peak power and pulse duration can be independently controlled. For improved comparison within the literature of LC laser device performance, we thus propose an alternative convention whereby threshold is stated in units of peak power density.

Liquid crystal (LC) photonic band-edge lasers have received much attention over the past few decades and recent advances in the field are continuing to bring applications of this technology closer to realization [1–3]. Chiral nematic LCs spontaneously self-organize to form a one-dimensional photonic band-gap, which, when doped with an appropriate gain medium (such as organic dye), can act as a mirrorless resonant microcavity in a photonic band-edge laser [4–6]. Their microscale size, simplicity to fabricate, and ability to deliver wavelengths spanning the visible spectrum and beyond [7,8], exemplify their benefits over alternative tuneable laser technologies, such as large and expensive optical parametric oscillators or supercontinuum lasers. LC laser emission at the photonic band edge is produced through optical stimulation from a pulsed laser pump source. Typically, this has been achieved with nanosecond (or picosecond) scale, high peak power pulses from passively Q-switched lasers. However, a recent breakthrough in LC laser research demonstrated laser diode (LD) pumping of LC lasers; significantly reducing the size and cost of the pumping architecture [2,9]. An additional advantage and novelty presented by an LD pump source is the ability to independently vary the pump pulse peak power (through varying the current supplied to the LD), repetition rate, and duration. While several studies have investigated the effect of repetition rate on LC laser performance [10–13], very little work has been done to determine the effect of pump pulse duration, as this has previously been difficult to achieve with Q-switched lasers. The threshold and slope efficiency are common figures of merit for quantifying the performance of LC lasers, whereby the former is desirably minimized, and the latter maximized. Several studies have shown how different materials, pump and alignment geometries, and environmental conditions affect threshold and efficiency [14–19]. With regards to pump pulse duration, work by Cao *et al.* showed an order of magnitude reduction in LC laser threshold using a 40 ps pump laser compared to a 7.5 ns source [20], but their study was limited to these two arbitrary data points, and no insight was provided into the effect on slope efficiency. Theoretical work by Shtykov *et al.* showed a reduction in LC laser threshold and an increase in slope efficiency with a decreasing duration of the rising edge of a hypothetical trapezoidal pump pulse [21], and Sanz-Enguita *et al.* derived an equation showing the proportionality between threshold and pump pulse duration [18]. However, such work has not been possible to experimentally verify with conventional (fixed pulse duration) pump sources. It is only with the advent of LD pumping that a more rigorous investigation of the effect of pump pulse duration can now be conducted.

This work exploits the incremental pulse duration control of an LD pump source to measure the resultant effects on LC laser threshold and slope efficiency. Furthermore, the temporal characteristics of the pump are investigated to gain a deeper understanding of the key pump parameters that determine LC laser threshold and slope efficiency. We also propose an alternative method of defining LC laser threshold to enable comparison between different pump regimes.

An in-house fabricated LC laser cell, comprising two glass substrates (*Laser2000*) separated by 10 µm spacer beads (*Sinochem Nanjing Corporation*), was capillary-filled with a dye-doped chiral nematic mixture of LC (*BL006, Merck)* with 4.5 wt% chiral dopant (*BDH-1281, Merck*) and 0.5 wt% organic dye 4-(dicyano- methylene)-2-methyl-6-(4-dimethylaminostryl)-4H-pyran (*DCM, Exciton*). DCM was chosen as the gain medium for this work due to its high quantum efficiency [22] and its absorption spectra coinciding with the emission wavelength of the LD pump source [9].

A 445 nm LD (*NUBM44, Nichia*) was integrated with driver electronics (*PicoLAS GMBH*) to control the LD pulse duration, repetition rate, and peak power. The optical arrangement is shown in Fig. 1. A motorized piezo rotation stage (*AG-PR100, Newport*) containing a half-waveplate rotated the plane of linear polarization of the LD relative to a Glan-Taylor linear polarizer. This allowed further control of the LD peak power (in addition to diode current control), and enabled automated data acquisition using in-house developed software for measuring threshold and slope efficiency for different pump pulse durations. A quarter-waveplate converted the pump beam to circular polarization of opposite handedness to the chiral nematic, ensuring the pump was not rejected by the LC photonic band gap. A 10 mm focal length lens focused the pump beam onto the LC laser cell, producing a spot size of 78.4 (± 4.5) µm^2 (measured by knife-edge technique), and had the dual purpose of collimating the resulting LC laser emission. A dichroic mirror (*ZT442rdc, Chroma*) separated the pump and LC laser emission. Two energy meters (*PD10-pJ-C, Ophir*), simultaneously measured pump and output laser pulse energies

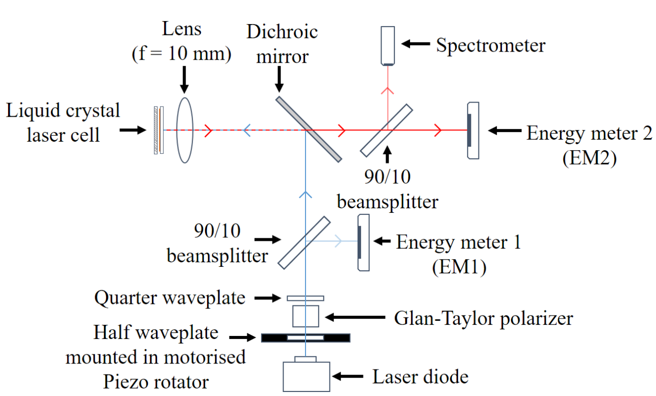

Fig. 1. Optical arrangement for threshold and slope efficiency measurements.

A range of pump pulse durations was used (approximately 10 ns apart), from 16 ns up to a maximum of ~ 100 ns (longer pulse lengths resulted in rapid degradation of output emission as a consequence of triplet state population of the gain medium depleting the population of lasing singlet states [23]). The LD was set to a repetition rate of 10 Hz to prevent LC laser performance degradation associated with high repetition rates [13]. Input and output pulse energy data were recorded at each half-waveplate position (i.e. different peak powers) over an interval of 1 second, and averaged over the corresponding 10 pulses), enabling a slope efficiency plot. This was then repeated for different pulse durations. The software plotted the LD pulse energy vs. LC laser pulse energy (accounting for optical losses in the delivery and collection optics), and a least squares fit algorithm calculated two linear trendlines indicating pre-threshold spontaneous emission and post-threshold stimulated emission. The slope efficiency was given by the gradient of the post-threshold data and the threshold was measured from the point of intersection of the two trendlines. The LC laser threshold was additionally verified by noting the pump energy at which narrow-linewidth laser emission was detected by a USB spectrometer (*CCS100/M, Thorlabs*, *resolution < 0.5 nm*).

Measurements of the temporal profile of the pump pulse were conducted by replacing EM1 with a photodiode (*DET025A/M, Thorlabs*) connected to an oscilloscope (*WavePro 735Zi, LeCroy*).

The threshold and efficiency data over the range of pump pulse durations is shown in Fig. 2. An increase in threshold and decrease in the slope efficiency can clearly be seen as the pump pulse duration is increased.

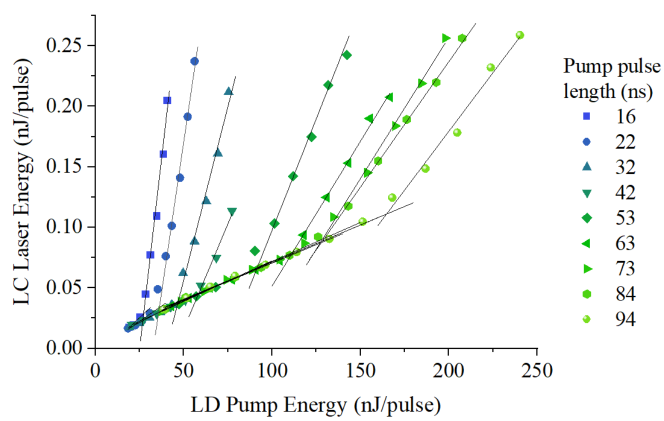

Fig. 2. The slope efficiency of an LC laser cell for nine different pump pulse durations. An increase in threshold and decrease in LC laser slope efficiency can be seen as the pump pulse duration is increased.

Forty-five individual efficiency measurements, comprising nine pulse durations recorded at five cell positions were analyzed separately to calculate the average threshold and slope efficiency values of the LC laser at each pump pulse duration. Fig. 3 reveals a clear pattern of an approximately linearly increasing LC laser threshold, and decreasing slope efficiency, as the pump pulse length is increased.

Our results are in good agreement with theoretical predictions and other incidental experimental data [20,21,24]. Moreover, this investigation is believed to be the first in which a controlled, incremental increase in pump pulse duration shows the adverse effects on LC laser performance, and provides compelling evidence for minimizing the pump pulse duration to optimize LC laser performance. The inverse relationship between pump pulse duration and slope efficiency can be attributed to the increasing population of the triplet states of the dye molecules with increasing pump pulse length [23]. An investigation into the temporal characteristics of the LD pump was also conducted to gain a better understanding of the trends observed in Fig. 3.

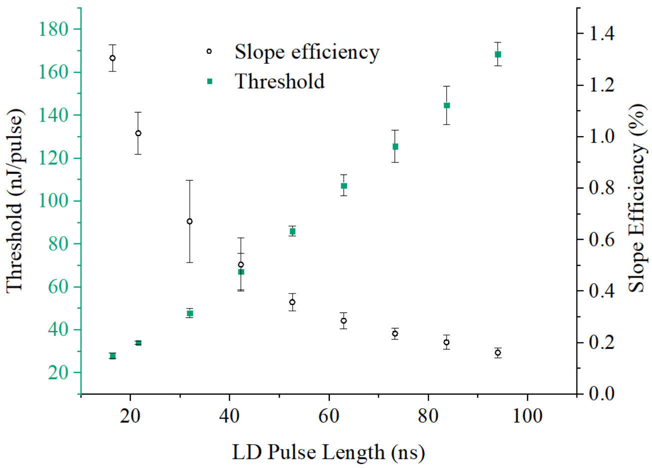

Fig. 3. Effect of pump pulse duration on LC laser threshold (green squares) and slope efficiency (black hollow circles). An increase in threshold and decrease in slope efficiency is seen as the pump pulse duration is increased.

Control of the pump pulse energy is usually achieved with polarization optics or absorbing filters to modulate beam intensity. In such instances the energy is controlled through varying the peak power, although this is rarely explicitly stated. The energy can also be controlled through varying the pulse duration. As this cannot typically be controlled with Q-switched lasers, this distinction has not been necessary in previous works. With the ability to now pump LC lasers with LDs, the term "pulse energy" becomes ambiguous, as the energy can be controlled both by peak power variation (through either LD current control or by intensity modulation) and by the duration of the electrical signal delivered to the LD (i.e. pump pulse duration). To determine if peak power or pulse duration determines LC laser threshold, the temporal characteristics of the pump pulse were measured.

Fig. 4 shows the temporal profile of the LD when set to pulse lengths of 11 ns (black data) and 32 ns (red data). In Fig. 4a, the 11 ns pulse illustrates the minimum peak power required to induce LC laser emission. The area under the peak is the threshold energy. The 32 ns pulse has the same energy (i.e. the area under both temporal profiles is equal) but this longer pulse did *not* produce LC laser emission.

The energy of the 32 ns pump pulse was then increased by increasing the peak power, through the rotation of the half-waveplate (equivalent to increasing the LD current) until threshold was reached. LC laser emission occurred when the peak power of the 32 ns pulse coincided with the peak power of the 11 ns pulse, as can be seen in Fig. 4b.

Clearly, there has to be sufficient energy in a pump pulse to overcome LC laser threshold, but these data show that the duration over which this energy is delivered is also a crucial factor in determining if threshold can be overcome. It is therefore not simply the energy of the pump pulse that determines if threshold can be reached, but a combination of the energy and the time over which the pulse is delivered. Therefore, as shown in Fig. 2 and Fig. 3, the process of converting pump light to stimulated emission becomes *less efficient* as the pump pulse length is increased.

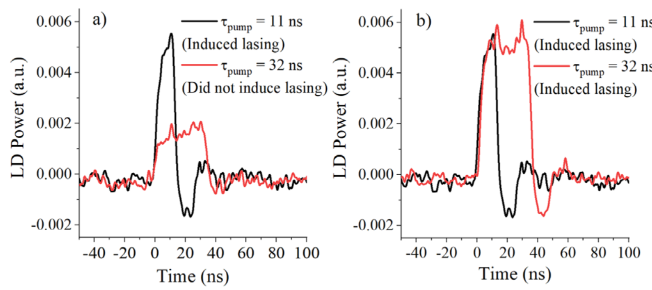

Fig. 4. Temporal profiles of pump pulses with durations of 11 ns (black line) and 32 ns (red line). a) Shows the two pulses with the same energy, but only the 11 ns pulse induced LC laser emission. In b), the energy of the 32 ns pulse was increased through increasing the peak power until LC laser threshold was reached. The negative signal is an electrical effect that did not affect the optical signal.

The threshold of a laser is an inherent feature of that particular system. When stating threshold, there should ideally be sufficient information to compare one pumping regime to another without ambiguity. Previous studies have highlighted the importance of pump beam spot size in determining the pump's ability to overcome LC laser threshold [11], but unfortunately it is often overlooked in the literature. When included, LC laser threshold can be better defined in terms of fluence (energy density), rather than pulse energy. When omitted, comparisons between LC lasers with different focusing optics cannot easily be made. However, fluence provides no information regarding the temporal delivery of this energy, which our data above has demonstrated to also be of crucial importance.

Optical power density is a common unit of threshold in CW laser systems, but for pulsed lasers there remains further ambiguity regarding whether this refers to average power density (which is dependent on pulse energy and repetition rate) or peak power density (which is dependent on pulse energy and duration). In Fig. 5 we plot the same threshold data from Fig. 3 in four separate data series, distinguished only by their units: the first (as per Fig. 3) in units of pulse energy (black squares); the second in units of fluence (orange diamonds); the third in units of average power density (purple triangles); and the fourth in units of peak power density (green circles). The first three data sets show a linear increase in threshold with pump pulse duration. However, if one instead defines threshold in terms of peak power density, by dividing the fluence by the pulse duration, we then obtain thresholds that are independent of input pulse duration, as required. Only when pump pulse duration is included can a pulse length independent threshold be obtained. It can therefore be concluded that when making comparisons between different LC laser systems, the way in which threshold is defined is vitally important, and that all pump parameters ought to be accounted for. For this data, the LC laser threshold can be newly defined in terms of peak power density as 2.2 (± 0.2) MW/cm^2/pulse (or mJ/pulse/cm^2/ns).

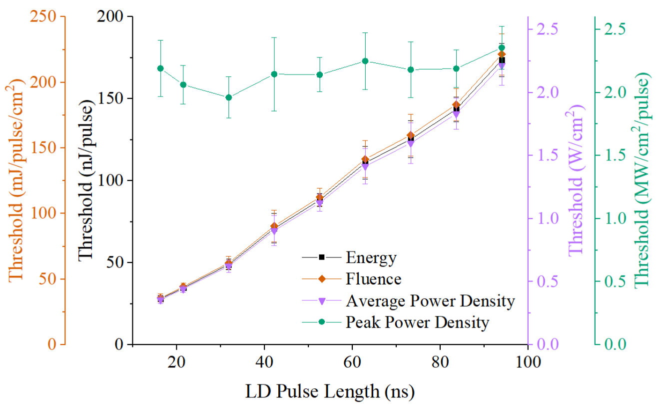

Fig. 5. LC laser threshold in terms of pulse energy (black data, left inner axis), fluence (orange data, left outer axis), average power density (purple data, inner right axis) and peak power density (green data, outer right axis). The lines between data points are a guide to the eye only.

It is important to also note that the above proposed definition of threshold assumes the same pump wavelength. Also, as previously mentioned, the theoretical work by Shtykov *et al.* showed a further threshold dependency with the pump pulse rise time [21]; a study that was verified by Herrnsdorf *et al.* in an LED-pumped organic solid state laser [25]. In the case of our work, it can be seen in Fig. 4b that the rising edge for both pump pulses is near-identical, thus enabling a fair and direct comparison of LC laser threshold.

Identification of the full range of pump pulse lengths that can successfully induce LC lasing was beyond the capabilities of the LD driver circuitry used in this investigation, but would be worthy of future exploration.

This work provides the first comprehensive experimental evidence that increasing pump pulse duration detrimentally affects the performance of an LC laser. Through temporal analysis, we have additionally shown that it is not just the pump pulse energy (nor indeed its fluence) that determines LC laser threshold, but also the duration over which the pulse is delivered (i.e. the peak power density). We propose that threshold definitions ought to incorporate *both* spatial *and* temporal information of the pump pulses, in addition to pulse energy. Adoption of this convention will then enable accurate performance comparisons to be made between different LC laser devices (i.e. materials, architectures, etc.), which in the literature are frequently excited by different pumping conditions. The results from this study are therefore important to the development and optimization of LC lasers (and LD driver technology), and of wider organic laser technology.

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Data availability. Data underlying the results presented in this paper are available in Ref. [26].

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