Optical Gain and Photostability of Different Laser Dyes, Quantum Dots and Quantum Rods for Liquid Crystal Micro Lasers

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ABSTRACT

Since the discovery of dye lasers, constant efforts are made towards developing optical gain materials with improved performance. Choosing an optical gain material for application in high-performance cholesteric liquid crystal micro-lasers requires a dedicated screening method. We employ Amplified Spontaneous Emission (ASE), and time-dependent fluorescence under pulsed excitation to study the optical gain parameter and photo-stability of laser dyes pyrromethene 567, pyrromethene 580, pyrromethene 597, pyrromethene 650, DCM, and Nile Red, dissolved in a nematic liquid crystal 5CB. Optical gain of CdSe/ZnS coreshell quantum dots dispersed in dodecene and CdSe/CdS coreshell quantum rods, dispersed in toluene was also measured and found to be much lower compared to the gain of fluorescent dyes. Pump energy and polarization dependence of ASE from dye molecules incorporated in the planar-aligned liquid crystal matrix is addressed. We determine the optical gain coefficient for different laser dyes by using the variable laser stripe illumination method using the small-signal gain model. The polarization dependence of ASE is determined in various geometries. We depict photostability from the half-life of emission intensity decay over a large number (10^6-10^7) of repeated laser pulses for fluorescent dyes, quantum rods and quantum dots. We find that quantum rods and quantum dots are much more stable compared to fluorescent dyes. Based on the photophysical characterization, we select pyrromethene 567 and pyrromethene 597 as efficient laser dyes for nematic liquid crystal micro-lasers.

Keywords: Amplified spontaneous emission, optical gain materials, VSL, Dyes, Photostability, Dichroic order parameter, microdroplet laser

1. INTRODUCTION

In the last two decades liquid crystal (LC) micro-lasers attracted considerable scientific interest owing to their flexibility and self-shaping properties, tuneability, and a good number of novel photonic applications.[1–4] Of particular interest are photonic systems that operate exclusively on light and are made of soft matter, including liquid crystals. To this aim, much of the efforts have been focused on optically pumped low-threshold lasing from the photonic band edge of a laser dye-doped cholesteric liquid crystal (CLC) with enhanced photostability. Towards achieving this goal, advances are made in developing new optical gain materials, optimizing the pumping parameters.[5–13] Numerous organic, inorganic and biological optical gain materials have been reported, however there is a need for a characterization method to screen optical gain materials for specific applications in microdroplet liquid crystal lasers. Among the standard experimental techniques to characterize the optical gain of particular material, the amplified spontaneous emission (ASE) is by far the most popular method. In a typical ASE experiment, the illuminating light is shaped into a thin stripe of variable length, hence the method is known as variable stripe length method (VSL). The stripe of light is illuminating the optical gain material that is usually deposited in a form of a thin layer on a substrate. The fluorescent light that emerges from the edge of the illuminated stripe is measured as a function of the stripe illumination intensity and length.

In this article, we present optical gain measurements from fluorescent dyes dissolved in planar aligned 5CB liquid crystal matrix, under different polarization of the excitation light. In addition we compare the optical gain of various fluorescent dyes with optical gain of quantum rods (QRs) and quantum dots (QDs) dissolved in different solvents or organic matrices. In addition to ASE, we perform extensive monitoring of photobleaching of different organic dayes in comparison to bleaching of QDs and QRs. We conclude that while QDs and QRs

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are superior in terms of photobleaching, they underperform in terms of optical gain, which is much lower for QDs and QRs compared to fluorescent dyes.

2. EXPERIMENTAL

The ASE measurement setup is schematically shown in Figure 1. The setup uses two different lasers to illuminate the optical gain materials: (i) a 30 ps laser operating at 10 Hz repetition rate and 532 nm wavelength (EKSPLA) obtained by frequency doubling the fundamental Nd:YAG 1064 nm laser line, (ii) 300 femtosecond laser ORIGAMI XP (NKT Photonics) that can be tuned in the UV, visible and IR, and provides variable repetition rates from single shot up to 100 kHz. In both cases, laser beam is first expanded using a combination of a plano-concave lens L1 and double convex lens L2 to get a 5 mm diameter Gaussian beam. A cylindrical lens L3 with a focal length of 150 mm is used to create a thin stripe of light in the focal plane of this lens, i.e. along the cylindrical axis. The direction of the light stripe is horizontal, i.e. parallel to the surface of the optical table, where the experiments have been performed. The divergence of this strip of light is several degrees. A fixed slit FS of dimensions 2×15 mm is placed in the focal point of lens L3 to create a fixed stripe of light with length of 10 mm. This fixed slit is used to select the nearly uniform intensity region of the stripe from the Gaussian profile of the incident beam. A variable slit AS, mounted on a precision linear stage is placed at a separation of ~3 mm from the FS to control the strip length in horizontal direction. The length of this stripe could be controlled with a precision of 5 μm. A combination of convex lenses L4 and L5 is put at a distance of 195 mm from FS to minimize the diffraction effects from the slits. This set of lenses produces an image of the light strip on the surface of the measuring sample. The dimensions of this light stripe is 1.2 mm (maximum length) × 50 micrometers (constant width). In this way the thin stripe of light illuminates the sample and excites the fluorescence. The stripe is set horizontally (parallel to the optical table) and the plane of polarization of the illuminating stripe is changed either parallel or perpendicular to the stripe using a half-wave plate HWP. Another polarizer P is used to precisely define the plane of polarization. A combination of two convex lenses is used to focus the fluorescent emitted light into the tip of the collecting fiber (step-index multimode, with 105μm silica core, 0.22 NA, wavelength range 400 – 2400 nm (Low OH)).

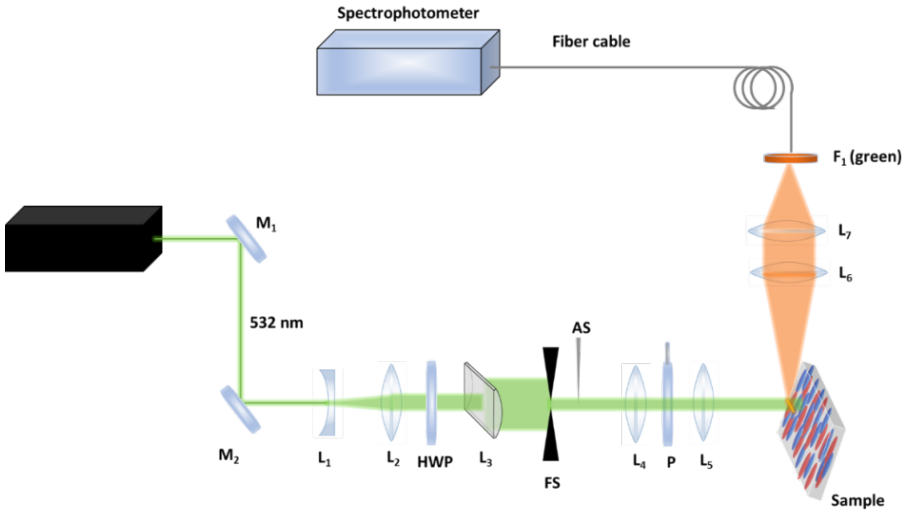

Figure 1. Schematic diagram of the experimental setup for ASE orientation measurements. The plane of polarization is rotated with a half-wave plate (HWP). M1, M2 dielectric mirrors, L1-L7 lenses, HWP – halfwave plate (@532nm), P-polarizer, FS-fixed slit, AS- Adjustable slit, F1- green filter @532 nm.

A green filter F1 (notch filter, 532 nm) is used to remove the signal of the excitation beam, which is reflected from the glass edges of the LC cell. In the experiment, ASE light is collected from the edge of the fluorescent layer, i.e. the axis of the collecting optics is parallel to the illuminated stripe of fluorescent molecules. The ASE signal is coupled to CCD imaging spectrophotometer (Andor, Newton EMCCD, Czerny-Turner spectrogaph) through a multimode fiber optic cable (step-index multimode, with 105 micorn silica core,0.22 NA, wavelength range 400 – 2400 nm (Low OH)).

Figure 2(a) shows images of the fluorescent stripe with a variable length. For maximum length of 1.4 mm, the illuminated surface area on the sample is ~0.0014 cm^2. For a typical pulse energy of ~1 μJ, the surface energy density at the sample is ~710 $\mu J cm^{-2}$. The two lasers used have very different pulse durations and hence the peak power dissipated during pulsed illumination are quite different. The single pulse duration of Ekspla laser is 30 *ps*, which gives peak power density of ~23 MW/cm^2 for a ~1 μJ pulse. Single pulse duration of Origami OPA is only 200 *fs*, which gives more than hundred times higher peak power density of ~3.6 GW/cm^2 during ~1 μJ pulse illumination. The light is collected from the left-hand side edge of the stripe. Figure 2(b) shows the spectra of collected ASE light from the ~3µm thick film of 5CB mixed with Nile red.

Laser dyes Pyrromethene 567, Pyrromethene 580, Pyrromethene 597, Pyrromethene 650, 4(Dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran (DCM), and Nile Red are chosen as optical gain media. Nematic liquid crystal 4-Cyano-4'-pentylbiphenyl (5CB, Nematel GmbH & Co. KG, Mainz)) is used as a host. The mass concentration of dyes in 5CB is typically 0.25wt%. Dyes are dissolved directly in the isotropic phase of the nematic LC at a temperature of 50°C for 15 minutes while mixing with mechanical vertex/ultrasonication. The LC-dye dispersion is centrifuged before the experiment to sediment the undissolved dye crystals, and care was taken not to introduce dye particles into the LC cells. LC-dye mixture is filled in the planar cell used in all the experiments. The cells were made of two 0.7 mm glass slides coated with polyimde and unidirectionaly rubbed to obtain good planar alignment of 5CB. The thickness of all LC cells was kept constant at 19±1 µm for all ASE experiments and 3±1 µm for all photostability and dichroic order parameter measurements.

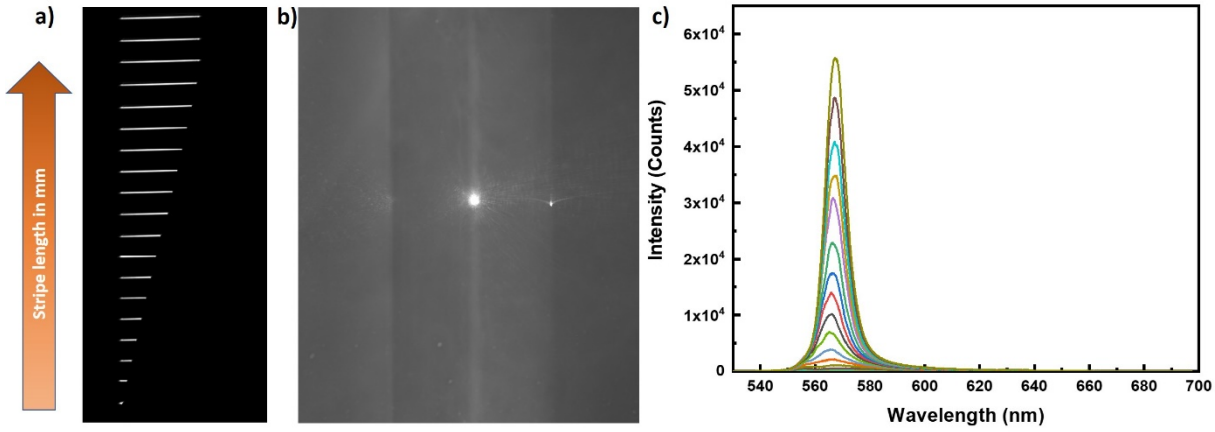

Figure 2: Typical ASE measurement. (a) Images of different stripe lengths. The ASE intensity is measured at different stripe lengths of the excitation beam. The width of the stripe is 50±5µm. (b) Side-view of the ASE light, the bright spot in the center is the ASE light exiting the stripe. On the left and right of this bright spot there are two glass slides (vertical on this picture) of thickness 700 µm, placed parallel at a separation of 20µm. This gap between the two plates is filled with LC and fluorescent dye. (c) The ASE spectra collected from the edge of the LC cell for different stripe lengths. The fluorescent dye Pyrromethene 567 is dissolved in 5CB.

Quantum dots CdSe/ZnS core-shell quantum dots, octadecyl amine ligand functionalized, from Aldrich and CdSe/CdS core-shell type quantum rods (Aldrich) were used in VSL experiments by dispersing them in PDMS, SYLGARD™ 184 two-part Silicone Elastomer (Dow chemicals Europe GmbH) is used. The silcon elastomer base and curing agent is mixed in 10:1 ratio, the mixture is degassed before adding the QD/QR dispersions.

PDMS mixture is added to a mould to make 0.5 mm thick layer, then QD/QR dispersion is added and mixed manually. The PDMS-QD/QR mixture is then cured in oven at 60° for 4-5 hours . The mass concentration of QRs in PDMS was ~ 0.16 wt% and QDs concentration was ~0.66 wt%.

3. OPTICAL GAIN

Optical gain coefficient (g) is a vital parameter for designing and optimization of lasers, and it quantifies the amount of light amplified per unit length, as it propagates through excited gain medium. ASE emission from different dyes in planar aligned Nematic LC host 5CB is measured by the Variable Stripe Length method (VSL). [14–16] This method involves the detection of single-pass amplified light emitted along the thin slab of an optically excited matter and was developed by Shaklee and Leheny in 1971 [16]. In the VSL method, a stripe-shaped beam is illuminating the surface of optical gain material, such as a crystal surface or a cell filled with solution of fluorescent dyes. The depth of the excited region is usually quite small, i.e. of the order of few micrometers. Spontaneous fluorescence is amplified by stimulated emission, as the emitted light passes through the excited volume to the edge of the optical gain material. The stripe therefore acts as an optically pumped waveguide, which amplifies spontaneous emission from the optical gain material by stimulated emission. If the spontaneous emission is isotropic and uniform in the excited volume, the intensity of the amplified light in the small gain model[17], exiting from the edge of the illuminated stripe is:

$$I_{ASE} = \frac{AI_s}{g}(e^{gL} - 1) \qquad (1)$$

Here, A is the cross-sectional area of the excited volume, I_s is the spontaneous emission rate per unit volume, and g is the net gain experienced by light, $g=g'-\alpha$, where g' is the gain due to stimulated emission and α is the optical loss. L denotes the stripe length. From the ASE intensity as a function of stripe length at constant surface energy density of the excited light, the optical gain can be determined [16] . The stimulated emission is detected from the nearly exponential increase in the detected intensity, when only the length of the excited region is increased. This is most easily seen in log-lin plot of I_{ASE} as a function of L, where the slope of the linear part of the curve indicates the optical gain parameter g, see Fig. 2 in Ref. 16. It is also clear that by increasing the surface energy density of the excitation light, the optical gain parameter increases. It is therefore of vital importance to keep all experimental parameters constant when comparing optical gain of different materials in different experiments.

The choice of stripe length is very important in the VSL method, and, based on the literature data,[14] we chose 2 mm as the maximum stripe length. Apart from the dependence of stripe length, the ASE-VSL measurements are carried out with different stripe beam polarization, and pump power density. Polarization of the excitation beam is important for the ASE experiments on fluorescent dyes dissolved in nematic liquid crystals. It is well known that the majority of dye molecules align their radiating dipoles with respect to local direction of LC molecules. Most of dyes align their radiative dipoles along the direction of LC alignment, also called the director. There are some exceptions, which align their radiative dipoles perpendicular to the director. Polarization of the excitation light should not be relevant for effectively spherical Quantum Dots (QDs), while it could be relevant for Quantum Rods (QRs).

3.1 Dependence of stripe beam polarization and orientation with respect to LC alignment

In the VSL method, the stripe beam orientation is kept horizontal and the emission intensity is collected along the stripe, i.e. perpendicular to the illuminating beam. The polyimide rubbing on the surfaces of the measuring cells gives planar orientation for the nematic 5CB liquid crystal, which acts as a host (template) for the dye molecules. All the dye molecules used in the work are linear, so dye molecules are expected to align along with the director. The emission from the transition dipole of the dye molecule is maximum in a perpendicular direction to the radiative dipole. The direction of dye molecule alignment with the pump beam therefore determines the emission intensity. The dye molecule is most strongly excited, if the polarization of the excitation light is parallel to the radiative electric dipole. In this case the light emission is maximum.

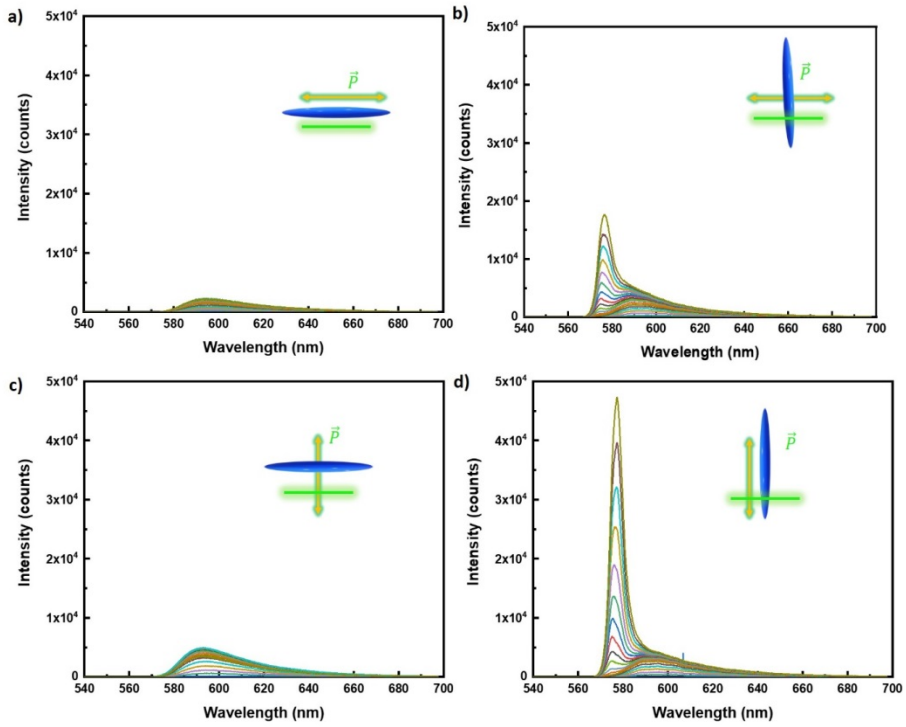

Figure 3. Orientation dependence of ASE emission spectra from 5CB+ PM 580 dye as a function of stripe length in 19-micrometer thick planar cell. Here the green line represents the stripe beam, the yellow arrow represents the direction of polarization and the blue ellipsoid represents the LC orientation (director of LC or the rubbing direction). The excitation wavelength is 532 nm, 30 ps pulse with energy of 30 μJ per pulse and the ASE light is collected from 40 pulses at a repetition rate of 10 Hz. Different colour spectrum corresponds to different stripe lengths from 0 - 2mm.

To understand the transition dipole direction of dye, orientation dependence of the pump beam polarization with respect to rubbing direction on emission intensities was investigated. As shown in the insets to Fig.3, four different orientations were used: (a) rubbing direction and direction of polarization parallel to stripe, (b) rubbing direction is perpendicular to the stripe and the direction of polarization (c) the direction of polarization is perpendicular to rubbing direction and the stripe, and (d) both rubbing direction and the direction of polarization are perpendicular to the stripe.

Figure 3 shows the polarization-dependent ASE spectra for two different orientations of PM580 with respect to the light stripe direction. As evident from Figure 3, for the three orientations in panels (a-c), the ASE intensities are quite low, and the emission spectra appear broad. When the pump beam polarization is parallel to the director and both are perpendicular to the stripe, a strong ASE peak appears, as shown in Fig. 3d. By incresing the stripe length (shown by different colours), the emission peak narrows, which is a clear indication of optical amplification by stimulated emission. Namely, the optical gain depends on the wavelength, g(λ), and by increasing amplification, certain wavelengths are strongly amplified, which results in narrowing of the emitted spectrum of ASE. A similar response is obtained for PM 567, PM 580, PM 650, DCM, and Nile Red. The strong ASE emission when the pump beam is polarized along the director can be understood in terms of the transition dipole moment direction of dyes. Emission is maximum in the perpendicular direction of emitting dipole; hence we obtain maximum light amplification, when this emitted light travels along the stripe and amplifies itself. However, there is one exception, as the isomeric dye pyrromethene 597 shows different polarization dependence on ASE emission. Figure 4 shows polarization and orientation-dependent ASE spectra for PM597. Strong ASE is obtained nearly for all polarizations and director orientation with respect to the stripe. This can be attributed to the symmetric molecular structure of this dye, for a given polarization direction some of the dipoles are contributing to the ASE signal. The maximum emmison polarization orientation, i.e. the direction of polarization is along the director/ transition dipole of dye and perpendicular to stripe ASE peak intensity is used to exctract the optical gain parameter.

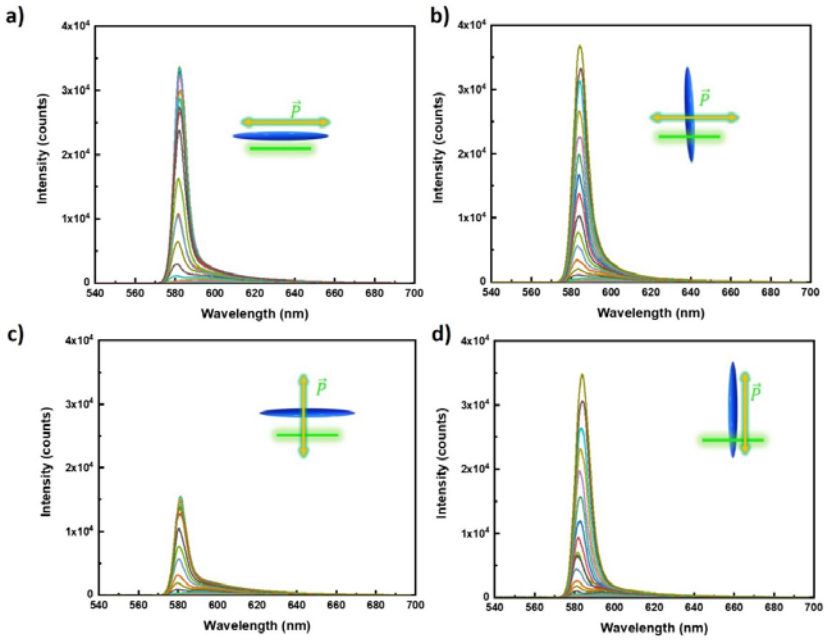

Figure 4. Orientation dependence of ASE emission spectra from 5CB+PM 597 dye in 19-micrometer thick cell for different stripe lengths. Here the green line represents the stripe beam, the yellow arrow represents the plane of polarization and the blue ellipsoid represents the LC orientation (director of LC or the rubbing direction). The different single pulse energy of 14 μJ the illuminating light stripe is shown with different colours. The excitation wavelength is 532nm, 30 ps pulses at repetition rate of 10 Hz, and the ASE intensity was accumulated for 40 pulses. Different colour of the line indicating spectrum corresponds to different stripe length from 0 - 2mm (dark yellow line).

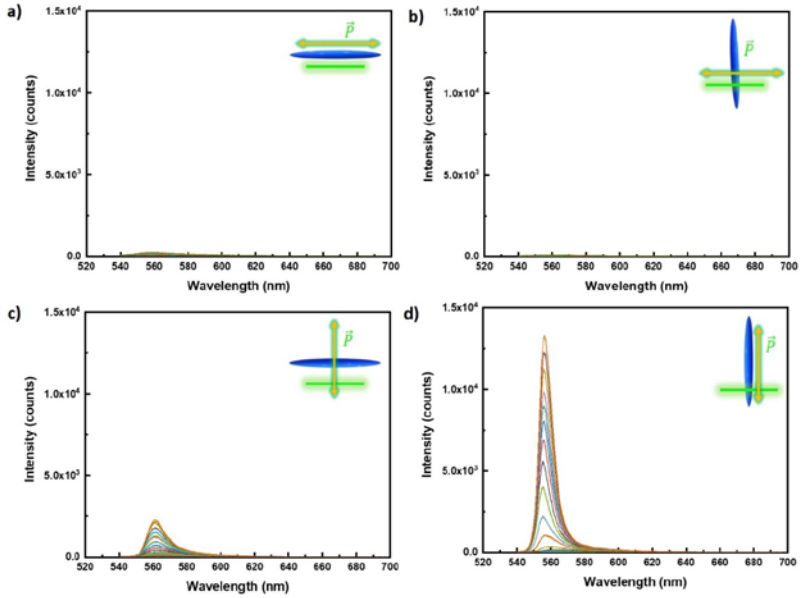

Figure 5. Orientation dependence of ASE emission spectra from 5CB+PM 567 dye in 19-micrometer thick cell cell for different stripe lengths. The excitation wavelength is 532nm, the repetition rate is 1 kHz, 200 fs pulses with pulse energy of 0.41μJ per pulse. The ASE intensity was accumulated for 1000 pulses. Different colour spectrum corresponds to different stripe length from 0 - 2mm.

Figure 5 shows the polarization-dependent ASE spectra for two different orientations of PM567 dye dissolved in 5CB with respect to the light stripe direction. In this experiment, a femtosecond OPA Origami XP OPA was used with 200 femtosecond pulses and repetition rate of 1 kHz.

3.2 Pump energy and repetition rate dependent ASE measurements

The single pulse pumping energy is another vital parameter for the VSL method of optical gain measurements, because the optical gain depends on the energy that is absorbed by the dye molecules. Figure 6 shows pump beam energy dependence of amplified spontaneous emission intensities for PM 580 and PM 597 dyes. ASE intensity increases rapidly after a threshold pump power. When the sample is excited well below the threshold, a longer stripe length is needed for amplified spontaneous emission, whereas above threshold already small stripe length is enough to get amplified spontaneous emission. For the excitation wavelength of 532 nm, the characteristic sharp and intense ASE peak appears after a threshold pump pulse energy of ~ 14 µJ for PM 597 as shown in Figure 4. The choice of stripe length is important for the optical gain measurement since pump pulse energy below threshold results in longer stripe length ASE which may underestimate the optical gain value. In all ASE experiments, the samples are excited with pump power to obtain maximum intensity, which ensures the pumping is well above the threshold. It is interesting to note that, when the pump beam polarization is parallel to dye alignment and perpendicular to the illuminating stripe, maximum ASE intensities are obtained with lower pump pulse energy, for instance, ~ 30 µJ single pulse energy for PM 650 dye. To check any possible influence of various repetition rates of the laser pulses on the measured optical gain, we performed stripe-dependent ASE experiments for various repetition rates. Figure 7 shows such a measurement for PM 567 dye in 5CB, taken at different repetition rates from 1 kHz to 5 kHz, with pulse energy 0.25 µJ. Each ASE data was taken for 1000 laser pulses and one can see that all the data nicely collapse to a single curve. This makes us confident that variable repetition rates of the lasers used in the experiments has no significant influence on the measurement of the optical gain.

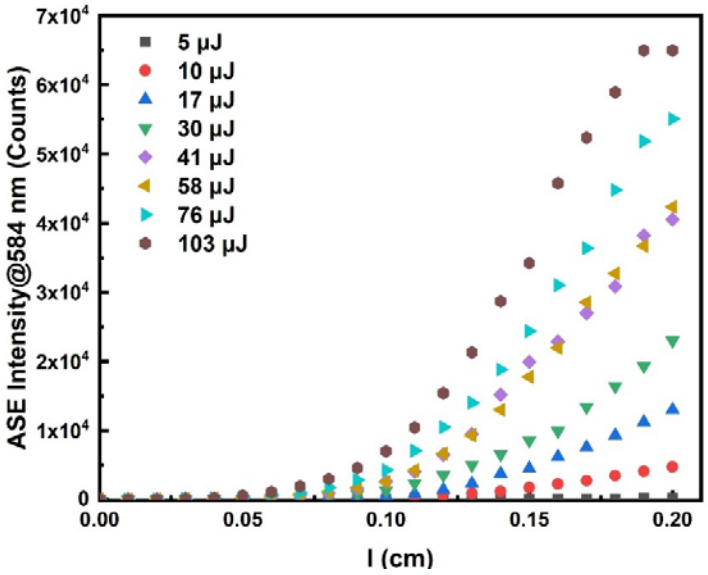

Figure 6. (a) Pump energy dependence of ASE peak intensity from 5CB+ PM597 dye in 19-micrometer thick cell. Different symbols correspond to different pulse energies. The excitation wavelength is 532 nm, the repetition rate is 10 Hz. The ASE intensity was accumulated for 40 pulses. The ASE intensity was taken at the wavelength, where the emitted spectrum shows the peak value.

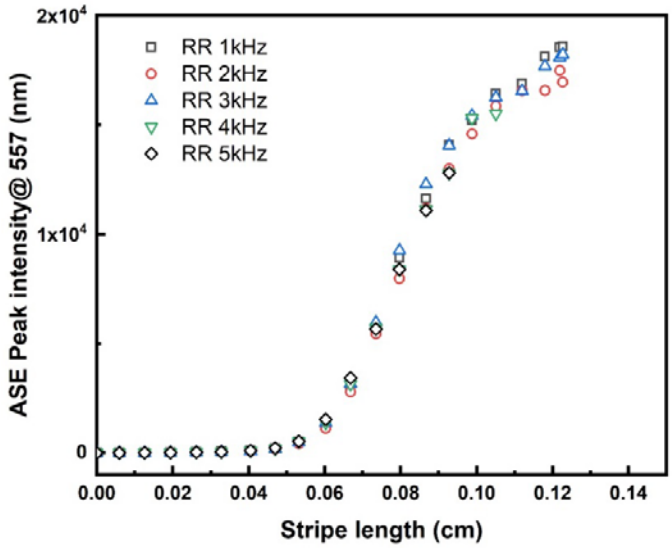

Figure 7. Stripe length dependence of ASE peak intensity for 5CB+Pyrromethene 567 dye in 19-micrometer thick cell at different repetition rates. Pump laser is Origami XP OPA 532 nm, 300 fs pulse with energy 0.25μJ per pulse, with different repetition rate. Each data collected after 1000 pulses for given stripe length at a peak emission wavelength of 557 nm.

After the parameters of ASE experiments have been set, we performed a series of stripe length dependence measurements for various dyes dissolved in 5CB and CdSe/ZnS core-shell quantum dots, and CdSe/CdS core-shell quantum rods dispersed in 0.5 mm thick layer of PDMS. The results are shown in Figure 8 for 0.41μJ energy per pulse, 532 nm illumination wavelength with 300 fs duration. The repetition rate was 1 kHz and the ASE signal was collected from 1000 pulses. The results show without doubt that the DCM dye is the brightest one. Surprisingly, it appears that quantum dots and quantum rods are quite dark compared to organic dyes. However, this has to be considered in view of the concentration of light emitters, which is quite high for fluorescent dyes.

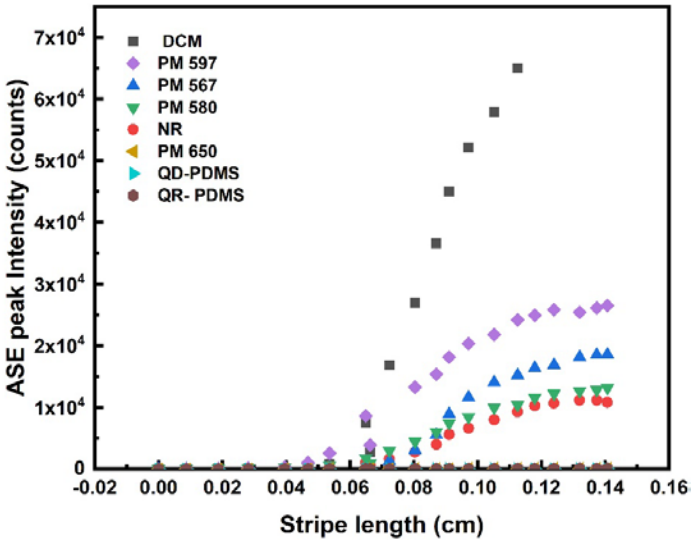

Figure 8. Stripe length dependence of ASE peak intensity for different dyes (5CB+dye) in 19-μm thick cell and CdSe/ZnS core-shell quantum dots, CdSe/CdS core-shell quantum rods in 0.5 mm thick PDMS layer. Pump laser wavelength is 532 nm, 200 fs pulse with energy 0.25 μJ per pulse, with repetition rate of 1 kHz. Each data collected after 1000 pulses for maximum stripe length.

3.3 Extraction of optical gain constant from ASE experiments

Because the ASE intensity from dyes in LCs depends upon the orientation of the director with respect to light stripe and polarization, the stripe length-dependent ASE intensity was measured in a configuration, where the ASE intensity is maximum. For all dyes except PM 597, the director is set perpendicular to the light stripe and the polarization oof excitation light is set parallel to the director. In this case the radiating dipole of dye is most strongly coupled to the incomming light and the direction of fluorescent emission is directed along the light stripe, i.e. along the direction of maximum stimulated emission. The single pulse pump energy was set well above the threshold, which made the measurement of the optical gain value reliable and accurate. All the measured ASE data follow an exponential increase in intensity with increasing stripe length, which is clear indication of ASE mechanism [16]. The optical gain values for different dyes are obtained by fitting the Equation (1) to the ASE peak intensity for different stripe lengths. In all our experiments, the ASE peak intensities grow exponentially without saturation at the maximum stripe length used, hence the small-signal gain model describes the experiments well.

Figure 9 shows the fitting of the measured ASE intensity as a function of the stripe length for the Pyrromethene 567 dye in 5 CB. When fitting the stripe length-dependence of ASE intensity, such as shown in Fig. 8, it is of vital importance to take care of the measurements taken at very short stripe lengths. These measured intensity have a large error due to: (i) inaccurate determination of the zero of L, i.e. stripe length, (ii) innacuracy in positioning the stripe with respect to the edge of the glass cell, and (iii) scattering of light of the stripe on imperfections at the cut edge of the glass. This makes the measured ASE intensity not following the Eq.1, because the origin of this light is nott entirely due to ASE. For this reason, the first measuring points up to the length of 0.02 cm (i.e. 200 μm) are not taken into account when fitting. For larger L the measured ASE intensity clearly shows exponential increase with increasing L, as shown by the fitting line in log-lin graph in Figure 9a. This means that the exponential factor in Eq. 1 is dominating with respect to all other stray light that could eventually be collected in the experiment.

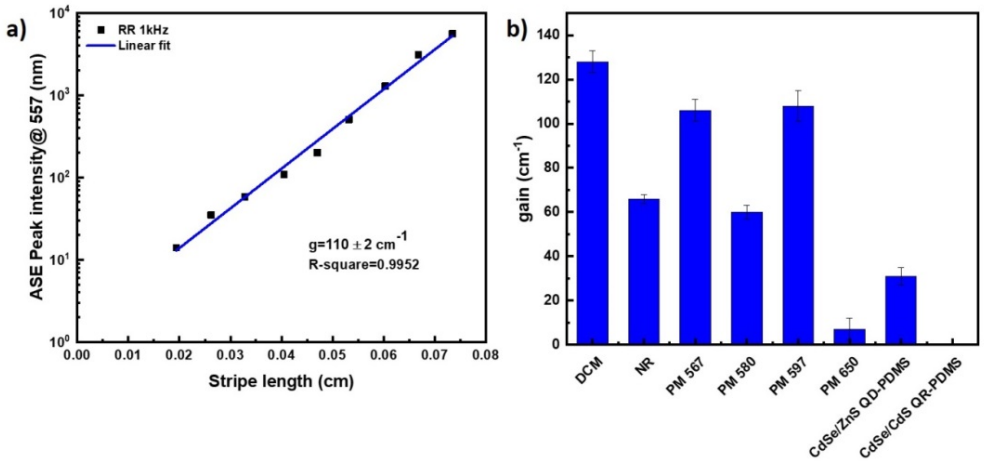

Figure 9. Optical gain parameter extraction from stripe length-dependent ASE peak intensity for (a) 5CB+Pyrromethene 567 dye in 19-micrometer thick cell at 1kHz repetition rate. The solid line corresponds to the linear fit in the log-lin scale and thus presents an exponential increase with increasing length. The scattered data points are ASE peak intensity measured for 1000 pulses taken at a wavelength of 557 nm. Extracted optical gain parameter is 110 ± 2 cm^{-1}. (b) Optical gain parameter extracted by small signal gain model for different dyes (5CB+ dye) in 19-micron thick cell and CdSe/ZnS core-shell quantum dots, CdSe/CdS core-shell quantum rods in 0.5mm thick PDMS layer. ASE intensities were measured for 1000 pulses of 532 nm, 300 fs pulses with energy 0.25 μJ per pulse and the repetition rate of 1 kHz. The concentration of dyes in 5CB were 0.25 wt% and were equal for all measured dyes, whereas the concentration of QDs and QRs in PDMS were 0.66wt% and 0.16 wt% respectively and therefore quite small compared to the concentration of dyes.

The optical gain parameters were extracted from ASE experiments for different organic dyes dissolved in 5CB, and and CdSe/ZnS core-shell quantum dots, CdSe/CdS core-shell quantum rods, dispersed in 0.5mm thick PDMS layer. The comparison of gain parameters is presented in Figure 9 b. It should be noted that the concentration of dyes in 5CB were equal for all measured dyes, whereas the concentration of QDs and QRs were quite small compared to the concentration of dyes.

4. PHOTOSTABILITY MEASUREMENTS

To investigate the photostability of different dyes in planar aligned 5CB matrix, the experimental setup used for ASE studies was employed. To quantify the photostability, photodegradation rate or photobleaching 'half-life' of light-emitting materials, we measured the time dependence of the peak ASE fluorescence at constant pulse energy of the excitation light and constant repetition rate. From these data, we determined the life time of the fluorescent material by measuring the number of laser pulses required to reduce the initial intensity by 50%.

Figure 10 shows the emitted fluorescence as a function of the number of excitation laser pulses at constant energy of pulses, for different materials.

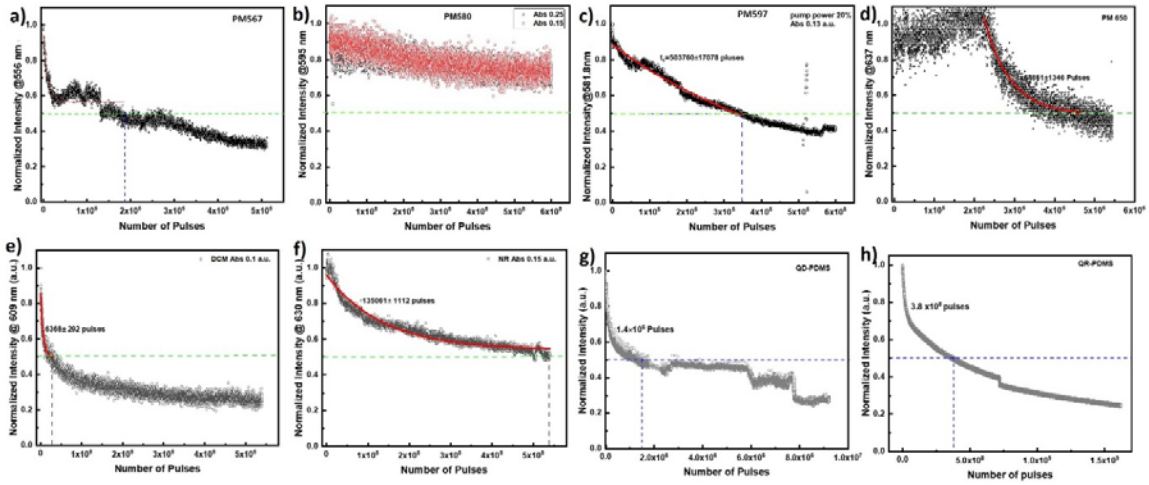

Figure 10. Fluorescent intensity from different types of dyes in 5CB, QDs and QRs in PDMS, as a function of the number of excitation laser pulses. Pulse energy is 20μJ, excitation wavelength is 532 nm, repetition rate is 10 Hz, taken with Ekspla laser. The thickness of 5CB was 4 micrometers, the thickness of PDMS was 0.5mm.

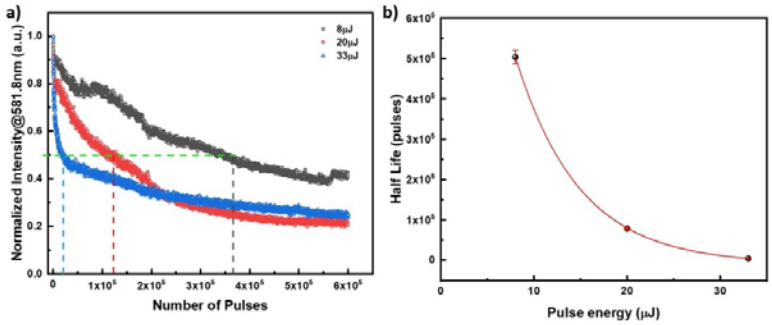

Figure 11. Photostability of pyrromethene 597 at different pump pulse energy. (a) photo decay profile measured with different pump pulse energy of 8μJ (black), 20 μJ (red), and 33 μJ (blue). The green dotted line corresponds the 50 % of the initial intensity. (b) Photo-half life is measured (in the number of pulses), which follows exponential decay with pump pulse energy.

The photostability of the dye depends on the pump power. In all the photostability experiments, pump power is set the same in respective ASE measurements. Figure 11 shows, pump power-dependent photostability of pyrromethene 597. As expected the photostability is higher with low pump power (8μJ for PM 597). With increasing pump power, the photostability decreases exponentially shown in figure 11b.

Table 1. Ranking of Laser Dyes

	PM 567	**PM 580**	**PM 597**	**PM650**	**DCM**	**NR**	**CdSe/ ZnS QD**	**CdSe/ CdS QR**
Fluorescence life time[5,18,19]	213 ns	218 ns	203 ns	220 ns	2 ns	2.8 ns		
Quantum efficiency[5,18–20]	0.99 (in methanol)	0.90 (in methanol)	0.77(in methanol)	0.54 (in methanol)	0.43 (in methanol)	0.7 (in dioxane)	> 50 %	> 70%
Dichroic order parameter	0.25	0.30	0.29	0.36	0.32	0.28		
Optical Gain	106 ± 5 cm^{-1}	60 ± 3 cm^{-1}	108 ± 7 cm^{-1}	7 ± 5cm^{-1}	128±5 cm^{-1}	66 ± 2 cm^{-1}	31±4 cm^{-1}	0 cm^{-1}
Pump Power for ASE	0.255 μJ	0.255 μJ	0.255 μJ	0.255 μJ	0.255 μJ	0.255 μJ	0.255 μJ	0.255 μJ
ASE Peak wavelength	557 nm	555 nm	576 nm	650 nm	602nm	621 nm	567 nm	557 nm
Photo-stability (Half-life)	~10^5 pulses	>6×10^5 pulses	~5×10^5 pulses	~6×10^4 pulses	6×10^3 pulses	1×10^5 pulses	1×10^8 pulses	3×10^8 pulses

Table 1 summarizes key parameters for characterizing the optical gain medium, such as optical gain, maximum emission wavelength, dichroic order parameters, and photostability in terms of half-life due to bleaching. Fluorescence lifetime and quantum efficiency were taken from literature. As shown in Table 1, in addition to the optical gain other photophysical properties are to be considered for the selection of laser dyes.

5. CONCLUSION

Emission, absorption, and photostability characteristics of dyes in planar 5CB matrix were studied for laser dyes pyrromethene 567, pyrromethene 580, pyrromethene 597, pyrromethene 650, DCM, and Nile Red. The alignment of transition dipole of dye, polarization direction, and orientation of pump laser beam significantly influences the photophysical properties of optical gain media. Emission from dye molecules is maximum when the pump polarization is parallel to the transition dipole. Pyrromethene dyes offer wider emission wavelengths and are better in terms of optical gain and stability compared to DCM and Nile Red dyes.

Quantum rods and quantum dots are of particular interest, because they show 1000 times better photostability compared to organic fluorescent dyes. On the other hand, the optical gain for CdSe/ ZnS Quantum dots is nearly for a factor of 4 lower compared to the brightest dye, DCM. However, one has to take into account the number concentration of light emmiting units in the lasing material. It turns out that molar masses of dyes, QDs, QRs are comparable. By taking into account the molar mass of 5CB (249.3 g/mol) and that of PDMS (6650-12000 g/mol), it then follows that the number concentration of CdSe/ ZnS Quantum dots compared to number concentration of DCM is 4.963 $x10^{20}$ molecules/100ml and 5 $x10^{19}$ particles/100ml, respectively. Even though the number concentration of quantum dots is an order of magnitude higher compared to dye, DCM dye provides higher optical gain. We conjecture this is due to orientational ordering of DCM dye molecules with respect to nematic director, which provides good orientational ordering of radiating dipoles. This results in a rather unidirectional emission of light from DCM (and other) dye, whereas the QD emission is isotropic.

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