Enhanced laser performance of cholesteric liquid crystals doped with oligofluorene dye

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We propose a new highly efficient organic dye, oligofluorene, which has great potential for lasing in cholesteric monomeric and glassy liquid crystal oligomers. We perform a detailed comparative experimental study of the laser characteristics of monomeric cholesteric liquid crystals (CLCs) doped with oligofluorene and a well-studied dye, 4-(dicyanomethylene)-Z-methyl-6-(4-dimethylaminostryl)-4*H*-pyran (DCM), commonly used for lasing in CLCs. Oligofluorene-doped CLCs yield a total output energy in the transverse single-mode regime five times that of DCM-doped CLCs with superior temporal and spatial stability. © 2008 Optical Society of America

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1. INTRODUCTION

Dye-doped cholesteric liquid crystals (CLCs) are self-assembling, mirrorless, distributed-feedback, low-threshold laser structures. The idea of lasing in CLCs was first proposed by Goldberg and Schnur in 1973 [1]. Independently, Kukhtarev proposed the idea and developed a theory of CLC distributed feedback lasers in 1978 [2]. The first experimental observation of lasing action in CLCs and a number of follow-up experiments were performed by Ilchishin *et al.* [3,4] and Ilchisin [5] in 1980, almost 20 years before the nature of lasing in CLCs at the photonic band edge was explained by Kopp *et al.* [6]. Taheri *et al.* [7] reported the observation of laser action in CLC structures shortly after Kopp *et al.* [6]. CLC lasers became a subject of a great interest over the past decade. They combine the tunability of dye lasers together with the compactness and robustness of semiconductor lasers. Because of their small size, tunability, and low-cost fabrication these lasers have a great potential to be used in medicine and LCD technology.

CLC structures are produced by mixing a nematic liquid crystal with a chiral additive that causes the nematically ordered molecules to arrange themselves into a helical structure. In the planes perpendicular to the helical axis of the structure the molecules have a nematiclike order, aligning along some preferred direction. This direction can be characterized by a unit vector called the local director. The local director rotates from plane to plane as we look along the helical axis of the CLC structure. The distance along the helical axis needed for the director to complete a full circle is called the helical pitch P.

Alignment of the rod-shaped molecules along a preferred direction causes the CLC structures to exhibit local birefringence. The refractive indices "seen" by the light polarized along the local director and in a perpendicular direction are called extraordinary (n_e) and ordinary (n_o), respectively. When a circularly polarized light with the same handedness as that of the helical structure propagates along the CLC helical axis it sees a periodic modulation of the refractive index and a selective reflection if the wavelength of the light is in a certain range. This range is defined by the pitch of the CLC according to

$$\Delta\lambda = P\Delta n, \tag{1}$$

where

$$\Delta n = n_e - n_o. \tag{2}$$

Thus, a CLC can be regarded as a 1D photonic crystal with the center of the photonic band gap defined by the wavelength

$$\lambda_c = nP, \tag{3}$$

where

$$n = \frac{n_o + n_e}{2}, \tag{4}$$

is the average of the refractive indices of the CLC. Propagation of circularly polarized light with the handedness opposite to that of the CLC is unaffected by the structure and experiences merely the average refractive index ($n_o + n_e$)/2. In this paper we call a circularly polarized wave right-handed if its electric field vector appears to rotate clockwise as the wave propagates towards an observer and left-handed if its electric field vector rotates counterclockwise. This is a standard definition of handedness of circularly polarized light [8].

If one dopes a CLC structure with an organic dye that has the emission spectrum overlapping with the CLC photonic bandgap, one can observe changes in photoluminescence of the dye. The emission will be enhanced at the low- and high-frequency photonic band edges and suppressed at the bandgap for the circularly polarized component with the same handedness as that of the CLC

structure. The sharp rise of the photoluminescence at the band edges is due to the fact that the density of states is very high at these spectral ranges. The circularly polarized photoluminescence with the opposite handedness does not experience any changes in the CLC.

A CLC host can serve as a resonator for a laser dye doped into it. The enhancement of the dye photoluminescence at the band edges, caused by the high density of electromagnetic states, leads to easily achievable low-threshold lasing [6]. The two lasing modes having the lowest threshold are situated at the band edges. The mode on the low-frequency band edge is comprised of two circularly polarized counterpropagating waves, resulting in a standing wave with the electric field vector aligned along the local director (provided that $n_o < n_e$) [9]. The mode at the high-frequency band edge is similar to that at the low-frequency edge, except in this case the electric field vector is perpendicular to the local director. Depending on the mutual alignment of the dye transition dipole moment and the local director, lasing oscillations can occur at the low-frequency, high-frequency, or at both band edges [9,10].

There are several ways to characterize the orientation of the dye dipole moment with respect to the local director [9–12]. The most widely used characteristic is the dye emission order parameter S_{em}, given by the expression

$$S_{em} = \frac{I_\parallel - I_\perp}{I_\parallel + 2I_\perp}, \tag{5}$$

where $I_\parallel$ is the fluorescence intensity of the nematic liquid crystal phase doped with the dye, measured for the radiation with the electric field parallel to the director; and $I_\perp$ is the fluorescence intensity for the radiation with the electric field polarized perpendicular to the director. Obviously, the case $S_{em}=1$ corresponds to a perfect alignment of the dye dipole moment along the liquid crystal director, the case $S_{em}=-1/2$ corresponds to a perfect alignment of the dye dipole moment perpendicular to the director, and the case $S_{em}=0$ corresponds to an isotropic orientation. We use the characteristic given by Eq. (5) to describe the dye dipole moment alignment in this paper.

Dyes with high-order parameters are believed to have a number of advantages over the dyes with lower-order parameters [10,13]. As most of the molecules's dipole moments of such dyes are well-aligned with respect to the local director, the threshold for lasing oscillations at low-frequency band edge is lower and the efficiency is higher as compared to the dyes with low-order parameter (under the condition that all other characteristics of the dyes are similar). As there is little or no competition between the modes corresponding to the low- and high-frequency band edges for the energy of the pump radiation, the highly oriented dyes can exhibit highly stable single-mode oscillations, generating laser radiation only into the mode at the low-frequency edge. As opposed to the highly oriented dyes, the dyes with lower-order parameters can yield two lasing peaks, corresponding to the low- and high-frequency band edges. It causes the output radiation to be multimode and unstable, as the two frequency modes compete for the pumping energy. Therefore, to achieve better performance in CLC lasers, including lower thresh-

old, higher efficiency, and higher stability, it is important to search for new laser dyes with high-order parameters. In reality, one never deals with two identical dyes with the only difference in their optical properties being the value of the emission order parameter. That is why, comparing two laser dyes with different order parameters, one has to take into account other optical properties that can influence laser threshold and efficiency, such as the radiative lifetime, quantum yield, and the characteristics of the triplet state typical to all organic dyes. The above characteristics are important if one wants to establish the influence of the order parameter on the laser threshold and efficiency. The latter is beyond the scope of our paper.

A generation of low-threshold, mirrorless, distributed-feedback, self-assembling lasers is being actively developed. Since the work published by Kopp *et al.* [6] brought light into the origin of lasing in CLCs, there were a great number of reports on lasing in dye-doped [7,10,12–22] and undoped [23] CLCs, as well as in liquid crystal elastomers [24], polymer network devices [25,26], glassy liquid crystals [27], and cholesteric blue phase [28]. Various methods of tunability in such kinds of structures have been addressed by many researchers: electrical [26], temperature [3,12,29]; mechanical [24], optical [30–33], and through concentration [34]; and pitch [35] gradients. Fluorescence behavior in the CLC structure has been extensively investigated both theoretically [9,36,37] and experimentally [9,11,37–40]. For optimization of the laser performance of dye-doped CLCs their laser characteristics as functions of different parameters, such as the CLC structure thickness and the dye concentration [15], temperature [12], excitation rate [17], incident angle and polarization of the pump radiation [21], and the size of the pump spot [22] have been studied.

The dye 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostryl)-4*H*-pyran (DCM) is very popular among researchers working in the field on CLC lasers [12,14–16,21,22,24–26] as this dye can be well-incorporated into most of the CLC hosts. On the other hand, DCM has a low-order parameter with the value around 0.4 [10]. To the best of our knowledge, there are very few publications reporting an attempt to find a better dye with higher-order parameter for use in CLC lasers [10,13,41].

Here we report on a new laser dye, oligofluorene OF2, developed in our laboratory [42]. We refer to this dye as OF later on in this paper. The emission order parameter of OF is 0.60, which is significantly higher than that of DCM. We perform a comparative study of the new oligofluorene dye and the commonly used DCM doped into the same CLC material. For both dyes, we find the optimal concentrations at which their laser performances in CLC are the best. We demonstrate that oligofluorene has a higher laser output and stability than does DCM.

2. SAMPLE PREPARATION

We mixed the nematic liquid crystal (ZLI-2244-000, Merck) with the chiral twisting agent (CB15, Merck) and fluorescent dyes to produce right-handed helical structures. We filled 22 μm thick glass cells with the dye-doped mixture. The walls of the cells were coated with polyimide

and rubbed to align the helical axis perpendicular to the substrates. The cells were prepared in clean room conditions.

The new fluorescent dye OF, an oligomer consisting of a central red-emitting segment end-capped by tetrafluorenes with aliphatic pendants [42], was used as a highly oriented candidate for lasing in CLC in our studies. We prepared six samples with different OF concentrations: 1.00, 1.25, 1.50, 2.00, 2.50, and 3.00 wt. %. For the purpose of comparison, we prepared the samples doped with 0.50, 0.75, 1.00, 1.25, and 1.50 wt. % of DCM dye. We also prepared a CLC sample with 1.75 wt. % DCM, but the clusters of DCM crystals were observed in this sample under a microscope, which indicates that films with high concentration of DCM encounter phase separation due to limited solubility of DCM in our CLC host. Nevertheless, the ranges of the weight percent of the dyes used in our studies cover the optimal dye concentrations for the best performances of both DCM and OF, and these concentrations for the two dyes are different. Even though the weight percent concentrations of DCM used in our samples are slightly lower than those of OF, the corresponding molar concentrations of DCM are almost an order of magnitude higher. For example, 1.00 wt. % of DCM corresponds to a molar concentration 2.64×10^{-2} M/l, while 1.00 wt. % of OF corresponds to a molar concentration 2.76×10^{-3} M/l. Observation under a microscope showed that the CLC structures had large monodomain areas (the areas without pitch fluctuations). The reflection spectra of the samples revealed an excellent alignment, displaying sharp interference fringes (see Fig. 1). The depressed baseline on the short-wavelength side of the stop band in Fig. 1 is because of the dye's light absorption in the visible region that diminishes the observed reflection from the dye-doped CLC film.

The pitch length of a CLC structure depends on the concentrations of both the chiral agent and the fluorescent dye. We have done a regression calculation to evaluate the helical twisting power of the chiral agent in the absence of a dye. We then calculated how much the presence of 1.00 wt. % of a dye increases the pitch length. Following these calculations, one can match the low-frequency band edge to the dye's maximum fluorescence wavelength with the precision of ±10 nm. As an example,

the reflection and fluorescence spectra of CLCs doped with 1.25 wt. % DCM and 3.00 wt. % OF are presented in Fig. 1.

To evaluate the emission order parameters of DCM and OF dyes we measured the emission intensities polarized parallel and perpendicular to the director in the nematic liquid crystals doped with the dyes. Then we used Eq. (5) to find the values of the emission order parameters to be 0.36 for DCM and 0.60 for OF. These values were obtained for 22 μm thick samples. Additional measurements have shown that the emission order parameters of 2.5 μm thick DCM- and OF-doped nematic liquid crystals are 0.41 and 0.70, respectively. We also measured the absorption order parameters of the samples and found that their values for DCM- and OF-doped nematic liquid crystals are 0.43 and 0.76, respectively, and do not change with the sample thickness. Note that the absorption order parameter can also be defined by Eq. (5) if one replaces the fluorescence intensities by corresponding intensity absorption coefficients. The values that are significant for our analysis are the emission order parameters for 22 μm thick samples (this thickness corresponds to the thickness of our CLC samples for laser measurements).

3. EXPERIMENTAL SETUP

Our experimental setup is depicted in Fig. 2. We used the second harmonic frequency of a Nd:YAG laser EKSPLA (Altos) to obtain 532 nm 35 ps pulses (FWHM) with the 10 Hz repetition rate as a pump source for the dye-doped CLC structures. The laser produced nearly Gaussian pulses with the diameter 3.3 mm FWHM with respect to the intensity at the position of the focusing lens. A lens of 20 cm was used to focus the laser beam onto the sample into a spot of 28 μm FWHM. We used a half-wave-plate and a linear polarizer to control the pump energy. To ensure maximum absorption and minimum scattering loss of the pump radiation, we converted the linearly polarized light exiting the polarizer to the left circularly polarized (LCP) light by means of a quarter-wave-plate. The LCP pump radiation penetrated inside a right-handed CLC sample and got efficiently absorbed by the dye molecules without losses due to reflection off the CLC structure.
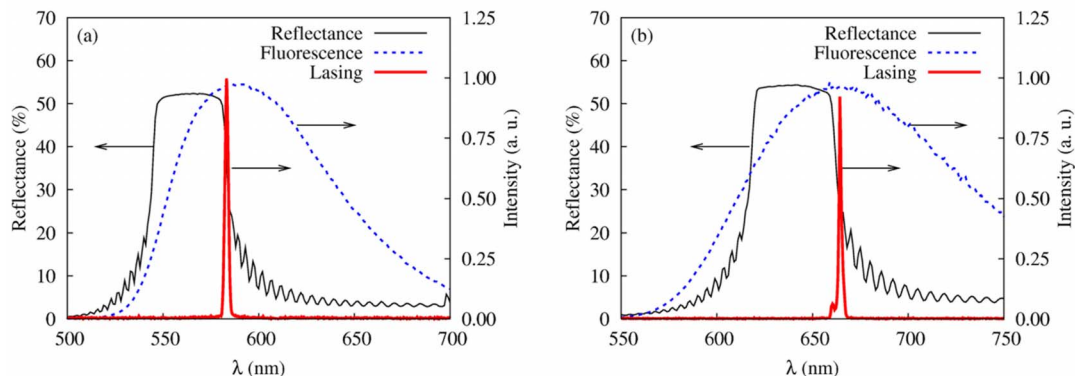

Fig. 1.   (Color online) Reflectance (thin solid curve) and lasing (bold solid curve) spectra of (a) 1.25 wt. % DCM-doped CLC sample and (b) 3.00 wt. % OF-doped CLC sample plotted together with the fluorescence spectra of the dyes (dashed curve).
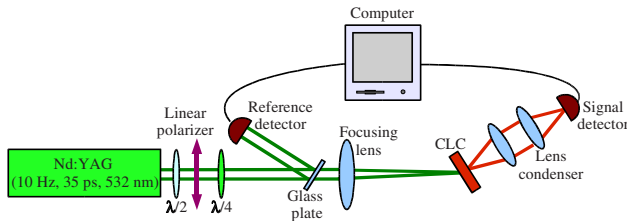
Fig. 2.   (Color online) Experimental setup.

The laser radiation originating from the sample was collected by a lens condenser composed of two lenses with focal lengths of 5 cm each. The collected laser radiation was sent to an energy meter or Ocean Optics spectrometer USB-2000 with 1 nm resolution. The spectrometer was used to record the laser spectra of the samples. A sample was mounted on a 3D translation stage (not shown in Fig. 2) to provide fine adjustment of focusing and optimization of alignment.

For measuring the energy of the CLC laser radiation a reference-signal setup configuration was used. We reflected 10% of the pump beam by a microscopic glass plate to the reference energy meter, while the signal energy meter was used to measure the energy of the CLC laser output. The energy meters were connected to a computer. Using software, we set a range of acceptable values of the reference energy for each data point. This way we eliminated the influence of shot-to-shot fluctuations in the pump laser and significantly increased the signal-to-noise ratio in our data.

4. STABILITY AND FREQUENCY MODE COMPETITION

Using an electron-multiplied cooled EM-CCD camera (Andor Technologies), we recorded the intensity distributions of DCM- and OF-doped CLC laser outputs. The patterns recorded at the pump fluence approximately twice the threshold are presented in Fig. 3. It can be seen from the figure that DCM has a highly nonuniform intensity distribution in its output while OF displays a much more

uniform intensity distribution. The spatial pattern of DCM-doped CLC was found to change significantly in time while that of OF remained stable.

We attribute the highly unstable behavior of DCM-doped CLC to the strong degree of competition between the low- and high-energy photonic band edge modes. The nature of the competition between the frequency modes is as follows. The molecules of a dye with an order parameter $S_{em} > 0$ tend to align with their transition dipole moments along the local director of a CLC structure. However, there are nonzero components of the dye molecules's dipole moments perpendicular to the local director, as S_{em} is typically less than 1. This implies that the dye's emission contributes to both frequency modes of the CLC situated at the low- and high-energy band edges. The two band edge frequency modes compete with each other for the use of the pump energy. When the CLC structure doped with the dye is pumped at the wavelength of the dye's absorption, laser generation is most likely to occur at the low-energy band edge frequency mode, as most of the dye's emission contributes to that mode. At low pump energies the low-energy band edge mode suppresses the high-energy edge mode as the preferred orientation of the dye's dipole moments is along the local direction. As the pump energy grows higher, the competition between the low- and high-energy band edge frequency modes for the use of the pump energy becomes stronger. At a certain level of the pump the high-energy edge mode can reach its threshold and the resulting output spectrum will contain two peaks: a stronger peak corresponding to the low-energy edge mode and a weaker peak of the high-energy edge mode.

Typical lasing spectra of our samples are shown in Fig. 1, together with the reflectance of the CLC structures and fluorescence spectra of the dyes. Both DCM- and OF-doped CLCs displayed lasing at the low-energy band edge. As the order parameter of DCM is much smaller than that of OF, it is supposed to be relatively easy to observe the second peak in the lasing spectrum of DCM, corresponding to the high-energy band edge frequency mode [10,13,41]. Nevertheless, the second peak in the laser out-
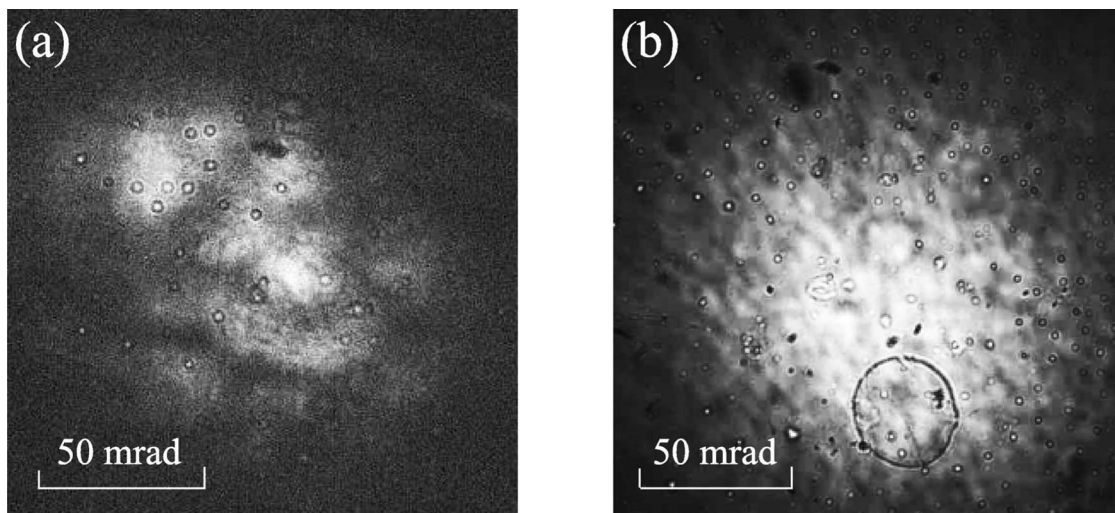

Fig. 3.   Intensity distribution of the laser output measured using a CCD camera in (a) 1.00 wt. % DCM-doped CLC and (b) 2.00 wt. % OF-doped CLC. The dark ring at the bottom of the picture is a camera artifact.

puts of DCM and OF failed to appear, as the lasing degradation and damage of our CLC host occurred with the increase of the pump fluence before the high-frequency mode reached its threshold. However, DCM would experience a stronger competition between the low- and high-energy band edge frequency modes, regardless of whether its lasing spectrum does or does not contain the second peak. We believe that this competition between the band edge frequency modes caused the temporal and spatial instabilities that we observed in DCM-doped CLC laser output.

5. LASING OUTPUT

We measured the lasing output characteristics of OF- and DCM-doped CLC samples in two different regimes. The first regime corresponds to the sample position precisely at the focal point of the lens used to focus the pump radiation. In this regime we observed laser output of a sample in the form of a single spot corresponding to the single *transverse* fundamental spatial mode [see Fig. 4(a)]. For this reason, we call this regime "transverse single mode." We positioned the sample precisely to the focal point of the lens by translating it laterally on the micrometer stage while measuring the laser output at a very low pump level (slightly higher than the threshold). By doing it this way we optimized for the maximum output energy, which helped us to ensure that the sample is precisely at the focus.

In the second regime we defocused the pump radiation by longitudinally translating the sample 14 mm away from the focal point of the lens. In this case, we observed a ring pattern at the sample's output, corresponding to the generation of several transverse spatial modes [see Fig. 4(b)]. We call the second regime "transverse multimode." The pump spot diameters in transverse single-mode and multimode regimes were 28 and 230 μm FWHM, respectively. We have done a comparative study of the laser performances of DCM- and OF-doped CLCs in both regimes.

A. Transverse Single-Mode Regime

Laser output characteristics in the transverse single-mode regime, in which the samples were positioned precisely at the focal point of the lens focusing the pump radiation, are presented in Fig. 5. The way we obtained the data points in Fig. 5 is by averaging over 30 measurements for each pump energy setting and evaluating the standard deviation. Different curves correspond to different concentrations of DCM and OF, as reflected in the legends. The ranges of the pump energies, shown on the X axes of the graphs, are different, as the output characteristics of DCM-doped CLCs saturated much more rapidly with increasing the pump energy. The Y axes of the graphs, representing the output energy of the samples, have the same scale and it is obvious that OF produces 5 times more output energy in the transverse single-mode regime, as compared to DCM. The output versus pump energy characteristics in the transverse single-mode regime were reproducible within 10%. The standard deviation of the measured output energy was less than 10%. The lasing threshold fluences of all DCM and OF samples are approximately the same and are around 7 mJ/cm^2. The slope efficiencies derived from the linear parts of the output characteristics as the ratios of the output energy changes to the changes in the incident pump energy are presented in Fig. 6. It is clear from Figs. 5 and 6 that there is an optimal concentration for each laser dye at which the CLC laser demonstrates the top performance, yielding the highest output energy and slope efficiency. At the dye concentrations lower than the optima, there are not enough dye molecules in CLC to produce much energy and efficiency. At high dye concentrations the pump radiation gets absorbed within several micrometers from the front surface of the sample and cannot get inside the CLC structure far enough for the dye molecules all through the length of the CLC to be excited. Besides, in the case of high dye concentrations the dye molecules are spaced so closely that the parasitic reabsorption and triplet quenching effects characteristic to all organic dyes [43] get much stronger. The maximum slope efficiency achievable in the transverse single-mode regime was around 5% for both
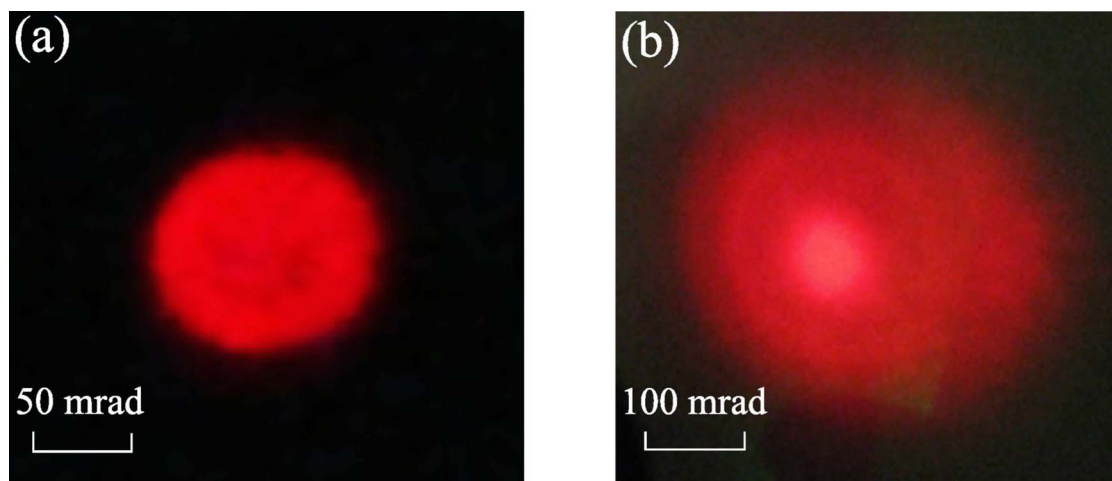

Fig. 4.   (Color online) (a) Photograph of a single transverse mode observed in the lasing output of the 2.00 wt. % OF-doped CLC in transverse single-mode regime. (b) A photograph of a ring pattern observed in the lasing output of the 2.00 wt. % OF-doped CLC in transverse multimode regime.
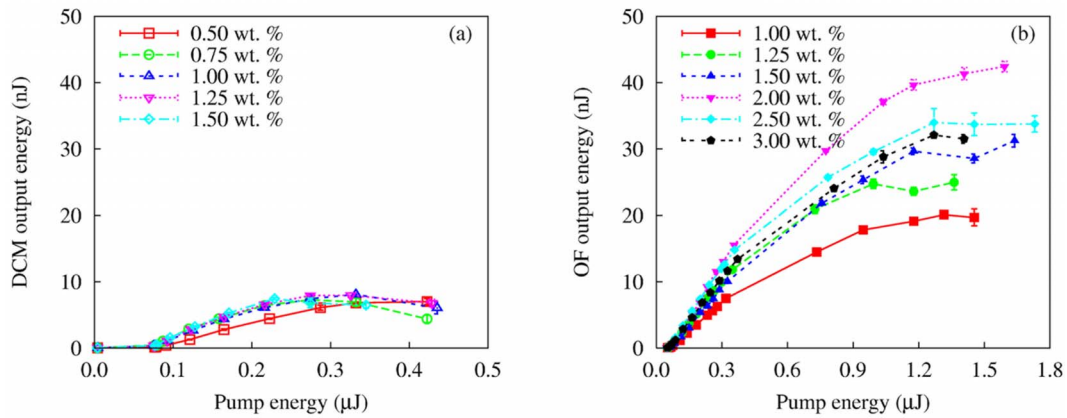
Fig. 5.   (Color online) Laser output energy plotted as a function of the incident pump energy of (a) DCM- and (b) OF-doped CLC samples in transverse single-mode regime.

DCM and OF dyes. It can be seen from Fig. 6 that the best laser performance was achieved with 1.25 wt. % DCM and 2.00 wt. % OF samples. The slope efficiency data were reproducible and accurate within 10%, which is reflected in the size of the error bars in Fig. 6. Within this publication we report the slope efficiency values based on the measurement of the CLC laser output collected only in one direction.

B. Transverse Multimode Regime
Laser output characteristics measured in the transverse multimode regime, corresponding to the position of the samples 14 mm away from the focal spot of the lens, are shown in Fig. 7. The reproducibility of the data obtained in the transverse multimode regime is within 20%, while the standard deviation of the output energy is around 10%. The maximum output energy of OF in the transverse multimode regime was measured to be 1.6 times greater than that of DCM. The lasing threshold fluences of all OF and DCM samples were around $0.7 \, \text{mJ/cm}^2$, which is an order of magnitude lower than that in the transverse single-mode regime. The reason for the higher

threshold in the transverse single-mode regime can be that the pump is focused so tightly that the radiation with such a small beam diameter cannot efficiently pump the transverse fundamental spatial mode. This makes it more difficult to achieve the threshold on that mode, and makes it impossible for higher-order spatial modes to appear at the output of the CLC laser. The latter is good when it is crucial to obtain a single transverse fundamental spatial mode in the CLC output.

The slope efficiencies of the samples in the transverse multimode regime are shown in Fig. 8. Unlike in the transverse single-mode regime, where the slope efficiencies of DCM and OF were comparable, in the transverse multimode regime OF displayed an almost twice larger maximum slope efficiency. A possible explanation to this is as follows. In the transverse multimode regime there are many spatial modes in the laser output. They all contribute to the total laser output energy and, therefore, influence the overall slope efficiencies of the output characteristics. As it has been shown in the transverse single-mode regime, the saturation of the output energy with the increase of the pump energy occurs much more rapidly in DCM- than in OF-doped CLCs. In the transverse multimode regime there are many modes contributing to the total output of DCM, but each of those modes saturates more rapidly than a similar mode of an OF-doped CLC. That is why the overall slope efficiency of DCM-doped CLCs is significantly lower in the transverse multimode regime.

Based on the above picture, we can also explain the reason why in the transverse single-mode regime OF-doped CLCs produce a 5 times higher maximum output energy as compared to DCM-doped CLCs, while in the transverse multimode regime they produce an only 1.6 times higher maximum output energy. In the transverse single-mode regime we measure the output only of the fundamental spatial mode. That is why the maximum output energies of the dye-doped CLCs are limited by the saturation of that mode. In the multimode regime, the pump spot is an order of magnitude larger and it efficiently pumps several spatial modes. As the intensity distribution at the pump spot on the sample is Gaussian, the first mode to lase would be the fundamental mode. The other modes switch on as we increase the pump energy, so
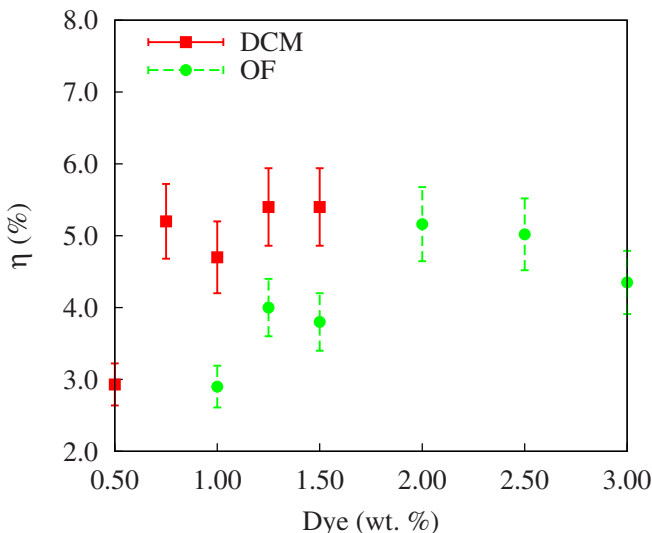

Fig. 6.   (Color online) Slope efficiency of the laser output of DCM- (squares) and OF-doped CLC samples (circles) as a function of the dye weight percent in transverse single-mode regime.
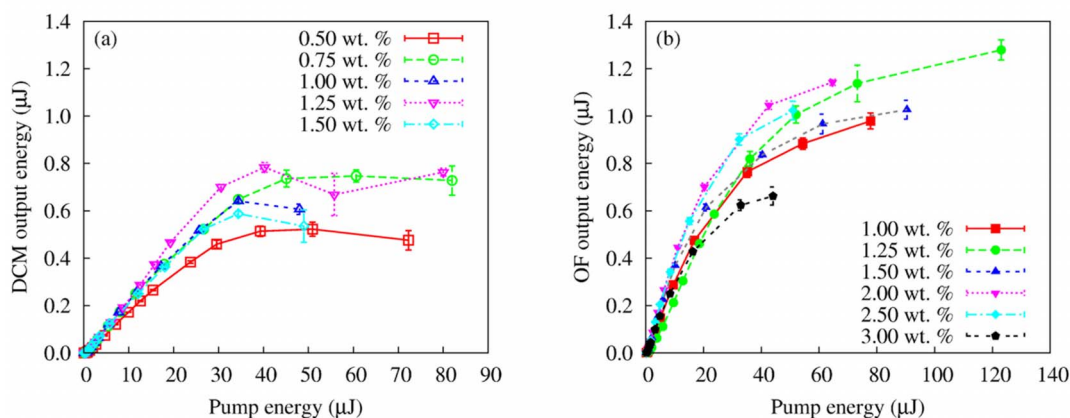
Fig. 7.   (Color online) Laser output energy plotted as a function of the incident pump energy of (a) DCM- and (b) OF-doped CLC samples in transverse multimode regime.

that the intensity at the edge of the pump spot is high enough for them to meet the threshold conditions. As we increase the pump energy, more and more spatial modes appear in the laser output, and DCM-doped CLCs saturate not as rapidly as they would if there were only one fundamental mode in the output. That is why the difference in the maximum output energy between DCM- and OF-doped CLCs is smaller in the transverse multimode regime.

Comparing the laser performances of DCM- and OF-doped CLC samples we found that the laser thresholds are similar for all samples. All DCM- and OF-doped CLCs display the thresholds 7 and 0.7 mJ/cm^2 in the transverse single-mode and multimode regimes, respectively. The slope efficiencies of DCM- and OF-doped samples are similar in the transverse single-mode regime. In the transverse multimode regime OF-doped samples display a slope efficiency almost twice as high as that of DCM-doped samples. The maximum laser output obtained with OF-doped CLCs was found to be 5 times greater than that obtained with DCM-doped CLCs in the transverse single-mode regime, and 1.6 times greater in the multimode re-

gime. In addition, the spectral purity and the temporal and spatial stability of the laser output of OF-doped CLCs is much higher than that of DCM-doped CLCs. Based on the above results one can conclude that OF is a better choice for lasing in CLC structures.

C. Laser Output Degradation Issues in Cholesteric Liquid Crystals
Working with dye-doped CLC in liquid phase requires extra care. Even a slightest mechanical stress that one accidentally applies to a CLC sample can cause the loss of alignment, the appearance of multidomain regions, and a change in the pitch and the lasing wavelength. Besides, as the host is in a liquid state the period of the structure is sensitive to the heating from the pump radiation and degradation of the CLC laser output, caused by the heating, can occur. Even though glassy liquid crystal hosts are more robust, it is the CLCs in a liquid state that allow one to achieve tunability of the laser wavelength. Therefore, it is important to investigate CLC lasers with both liquid and glassy hosts. The experiments with glassy liquid crystal hosts doped with OF dye are in progress and will be reported in a later publication.

We observed a difference in the behavior of DCM- and OF-doped CLC lasers. For example, in the transverse single-mode regime at the pump energies in the range between 200 and 500 nJ, we observed a significant degradation of laser output with time in DCM-doped CLCs, while OF-doped CLCs displayed stable lasing in this range of pump energies. More work on the temporal stability in dye-doped CLCs is in progress.

6. CONCLUSIONS
We have performed a detailed comparative study of the laser output characteristics of CLC structures doped with a commonly used laser dye, DCM, with the emission order parameter 0.36 and with a new laser dye, OF, with the order parameter 0.6. The study of the laser spectra showed that only a single laser peak, corresponding to the low-energy band edge frequency mode, can be observed in the output of all DCM and OF samples. We expected to observe a second peak, corresponding to the high-energy band edge mode in the output spectra of DCM-doped
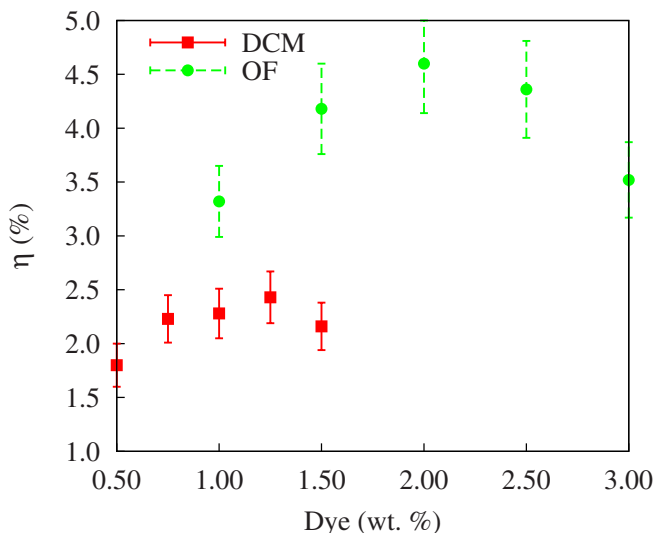

Fig. 8.   (Color online) Slope efficiency of the laser output of DCM- (squares) and OF-doped CLC samples (circles) as a function of the dye weight percent in transverse multimode regime.

CLCs, but it appears that the threshold for the second frequency mode is higher than the damage threshold of our CLC host.

OF-doped CLCs displayed a much higher temporal and spatial stability in the output radiation. We attribute this to the higher value of OF's order parameter that prevents strong competition between the high- and low-energy edge frequency modes. Strong competition between the modes, taking place in case of a dye with a low-order parameter, degrades the laser performance, creating temporal and spatial instabilities, which we observed with DCM-doped CLCs.

We measured the laser output characteristics of DCM- and OF-doped CLCs in two different regimes corresponding to generating a single fundamental spatial mode and a multimode ring pattern. This study itself is interesting as, to the best of our knowledge, spatial transverse single-mode and multimode regimes have not been discussed in the CLC structures before. The value of the transverse single-mode regime is that one can obtain a single fundamental spatial mode at low pump energy. The transverse multimode regime is interesting because it yields much higher output energy. The way one can control the number of the spatial modes appearing in the output is through changing the size of the pump spot in the sample. We found that the laser thresholds and the slope efficiencies of DCM- and OF-doped CLCs are similar in the transverse single-mode regime, but OF-doped CLCs produce a maximum output energy 5 times greater. The laser thresholds of the samples doped with both dyes in the transverse multimode regime are similar, but the slope efficiency of the OF-doped samples is almost twice as high.

The results of our study invite the conclusion that OF is an excellent laser dye for use in CLC structures, demonstrating a better laser performance in many aspects than that of the popular dye DCM. More experiments with OF-doped liquid and glassy CLCs are under way.

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