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# Electrically switchable random to photonic band-edge laser emission in chiral nematic liquid crystals

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Using a chiral nematic liquid crystal with a negative dielectric anisotropy, it is possible to switch between band-edge laser emission and random laser emission with an electric field. At low frequencies (<1 kHz), random laser emission is observed as a result of scattering due to electro-hydrodynamic instabilities. However, band-edge laser emission is found to occur at higher frequencies (>5 kHz), where the helix is stabilized due to dielectric coupling. These results demonstrate a method by which the linewidth of the laser source can be readily controlled externally (from 4 nm to 0.5 nm) using electric fields. © *2012 American Institute of Physics*. [doi:10.1063/1.3684804]

Both photonic band-edge and random laser emissions have been observed in liquid crystal (LC) media in recent years.[1–7] Band-edge laser emission from dye-doped chiral LCs is characterized by a single-mode, narrow linewidth laser line that appears at the edge of a photonic band gap (PBG) due to enhancement in the density of the photon states. Random laser emission, on the other hand, occurs in LCs when the multiple scattering of light is combined with a component that can generate optical gain through stimulated emission.[8]

Generally, dye-doped LC media exhibits either band-edge or random laser emission but not both. This is because the disorder-induced scattering required for random lasing acts as a significant loss for band-edge laser emission since it negatively affects the quality of the helical structure and, consequently, the photonic band-gap. However, using dye-doped LC emulsions,[9] it has recently been shown that by altering the droplet dimensions in the polymer dispersed LC, it is possible to change the feedback mechanism from one of intense multiple scattering (small droplets) to distributed feedback from a periodic macroscopic structure (large droplets).[7] The result is a system whereby the emission characteristics can be altered from a broad linewidth to a narrow linewidth by simply changing the mixing speed during the preparation of the emulsion without having to adjust the mixture composition.

One drawback with the emulsion system, however, is that the mechanism has to be selected before preparation and it cannot be modified *in-situ*. It would be highly desirable if the feedback mechanism, and consequently the emission linewidth, could be controlled using external electric fields. Therefore, the purpose of this investigation is to demonstrate that it is possible to control the feedback mechanism by using a dye-doped chiral nematic LC with a negative dielectric anisotropy that is subjected to an electric field of different frequencies.

The sample studied consisted of the nematogen LC mixture, MLC-7029 (Merck KGaA), that was doped with a low concentration (3.5 wt. %) of high twisting power chiral dopant (BDH1281, Merck KGaA) and a high quantum efficiency laser dye (PM597, Exciton) at a concentration of 1 wt. %. The nematogen host was selected as it has a negative dielectric anisotropy ($\Delta\varepsilon = -3.5$ at 25 °C) and the concentration of chiral dopant was chosen so as to position the long-wavelength band-edge at the gain maximum, which in LC media is at $\lambda = 590$ ($\pm 10$) nm for the PM597 laser dye.[10] A chiral nematic LC with a negative dielectric anisotropy is required to ensure that the macroscopic helical structure, a prerequisite for band-edge laser emission, can be recovered using an applied electric field. The components were thermally mixed at 100 °C for a period of 24 h to ensure that a chiral nematic LC had been formed and that the dye was uniformly dispersed throughout the bulk mixture. Finally, the mixture was capillary filled into a 12 $\mu$m-thick cell whose inner surfaces consisted of 1 cm$^2$ indium tin oxide electrodes and unidirectionally rubbed polyimide layers. Upon filling, these alignment layers ensured planar LC alignment at the surfaces and thus induced the helical axis to align perpendicular to the plane of the substrates (Grandjean texture). An electric field was applied across the LC cell using a signal generator (TG1304, Thurlby Thandar) and a high voltage amplifier (in-house). The amplitude of the applied electric field was 13 V/$\mu$m in all cases.

To illustrate the change in alignment of the LC, photographs of the optical texture recorded on a polarizing microscope are presented in Fig. 1 for a range of electric field conditions at a temperature of 25 °C. In the absence of an electric field (Fig. 1(a)), the sample has a uniform appearance (red color) which is unchanged when the sample is rotated between crossed polarizers (indicative of a Grandjean texture). When subjected to an electric field with a low frequency of 10 Hz, a scattering texture is observed consisting

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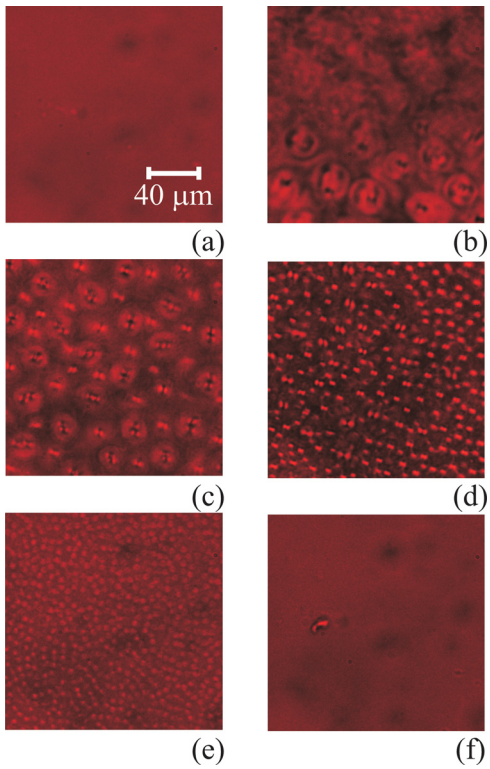


FIG. 1. (Color online) Photographs of the optical texture observed on a polarizing microscope with a 20× objective for the dye-doped sample when subjected to different electric field conditions. (a) 0 V/$\mu$m, (b) 13 V/$\mu$m, $f = 10$ Hz, (c) 13 V/$\mu$m, 100 Hz, (d) 13 V/$\mu$m, 500 Hz, (e) 13 V/$\mu$m, 1 kHz, and (f) 13 V/$\mu$m, 5 kHz.

of microdomains ($\sim$40 $\mu$m in diameter) that oscillate in a random manner as a result of electrohydrodynamic instabilities (EHDIs) (Fig. 1(b)). As the amplitude of the field is maintained and the frequency is increased to 100 Hz, the size of the microdomains ($\sim$20 $\mu$m in diameter) decreases and the turbulence begins to reduce (Fig. 1(c)). With a further increase in the frequency up to 1 kHz, the size of the domains continues to decrease and EHDI disappears (Figs. 1(d) and 1(e)). At frequencies above 3.5 kHz, a uniform monodomain is formed that is similar to that observed at zero-field (Fig. 1(f)). In this regime, dielectric coupling dominates and the sample adopts a Grandjean alignment as a result of the negative dielectric anisotropy of the chiral nematic host. Measurements of the transmission spectrum for white light revealed that there was no macroscopic PBG for frequencies of the applied electric field ranging from 1 Hz to 3.5 kHz but the PBG did reappear at frequencies above 3.5 kHz.

Emission characteristics for three different frequencies, low (10 Hz), intermediate (560 Hz), and high (5 kHz) of the applied electric field, are shown in Fig. 2 along with a pictorial representation of the alignment of the LC. For this experiment, we used the second harmonic (532 nm) of an Nd:YAG laser to optically excite the samples with an excitation energy of 5 $\mu$J/pulse (pulse length of 4–5 ns), an excitation rate of 1 Hz, and a pump spot with a diameter of 80 $\mu$m. The emission spectra were recorded using a high resolution (0.3 nm) universal serial bus spectrometer. When a 10 Hz electric field is applied, the emission spectrum consisted of a number of closely spaced narrow linewidth modes ($\Delta\lambda \approx 1$ nm) that are superposed upon the unsaturated fluorescence spectrum (Fig. 2(a)). In this example, the maximum intensity occurs at $\lambda = 571$ nm; however, from pulse-to-pulse, the emission spectrum is found to vary in accordance with the chaotic nature of a random laser. It is believed that the feedback is due to light scattering from the EHDI-driven fluctuations in the orientation of the domains combined with the localization of light from the randomly oriented micro-helices.

On the other hand, between frequencies of 100 Hz and 1 kHz, the spectrum is markedly different and is of the form of a random laser consisting of a smooth emission profile with a peak wavelength of 577 nm. For the example shown in Fig. 2(b), the feedback is the result of multiple scattering from the focal conic texture, and the narrow linewidth modes that were visible at 10 Hz are no longer observable. Here, the spectrum consists of closely spaced modes, which overlap to result in a smooth peak similar to that observed in a previous report.[11] The overall linewidth of the laser mode in this case is $\Delta\lambda = 4$ nm. Finally, increasing the frequency above 3 kHz (Fig. 2(c)) results in the transformation to band-edge laser emission at a wavelength of 582 nm ($\Delta\lambda = 0.5$ nm) as dielectric coupling promotes a Grandjean alignment of the chiral nematic LC.

The data show that the emission characteristics of the dye-doped LC can be varied significantly from the broader linewidth ($\Delta\lambda = 4$ nm) of a random laser to the single mode,

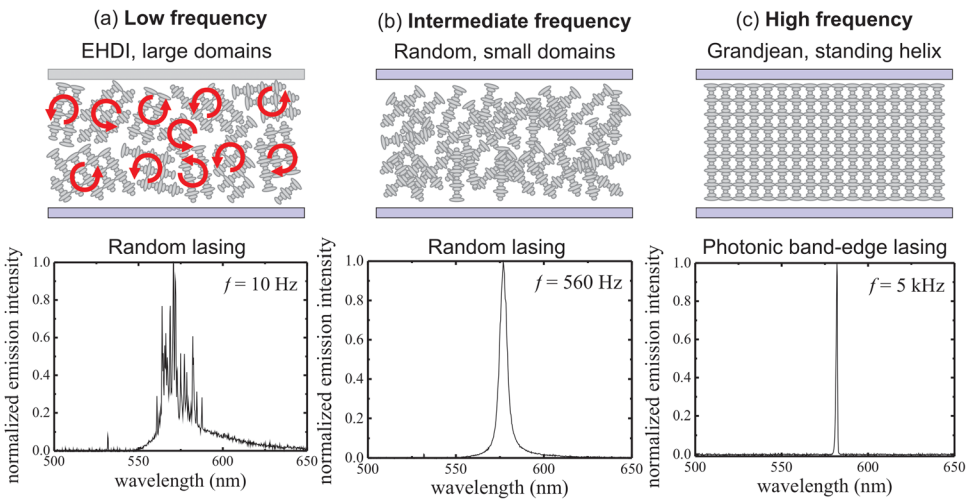


FIG. 2. (Color online) Normalized emission from the dye-doped sample for different electric field conditions when optically excited at $\lambda = 532$ nm. (a) 13 V/$\mu$m, 10 Hz, (b) 13 V/$\mu$m, 560 Hz, and (c) 13 V/$\mu$m, 5 kHz.

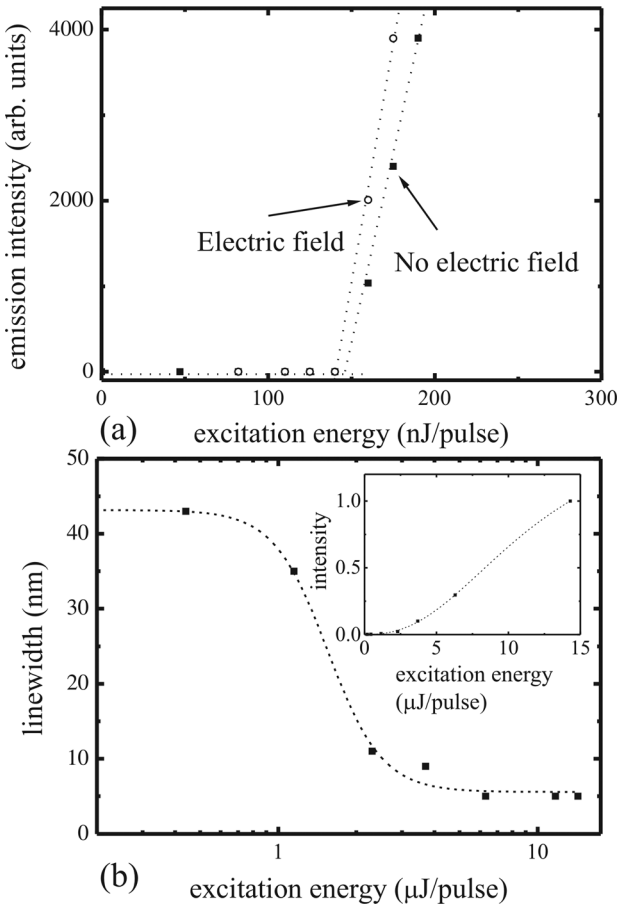


FIG. 3. Threshold measurements for (a) band-edge laser with and without the applied electric field (13 V/$\mu$m, 5 kHz) and (b) a random laser generated using an electric field of 13 V/$\mu$m, 560 Hz. The solid line in (b) is a sigmoidal fit to the data and the inset shows the relative emission intensity as a function of the excitation energy.

narrow linewidth ($\Delta\lambda = 0.5$ nm) output of a band-edge laser by simply adjusting the frequency of the external electric field. At low frequencies (e.g., 10 Hz), random laser emission consisting of discrete narrow emission peaks is also observed. To compare the different laser mechanisms, we have recorded the excitation threshold ($E_{th}$) for both the band-edge and and random laser that was generated using intermediate frequencies. These results are presented in Fig. 3. Due to the chaotic nature of the random laser with the discrete localized modes, it was not possible to measure the excitation threshold for this regime. This is because the formation of individual lasing modes are varying on timescales that are faster than the pump repetition rate, resulting in a variation in the position and intensity of laser emission from one pulse to another.

For the band-edge laser, data are presented for the output intensity as a function of excitation energy both with and without an electric field applied (Fig. 3(a)). It can be seen that the excitation threshold is approximately $E_{th} \approx 140$ nJ/pulse in both cases and is actually slightly lower when the electric field is applied; the efficiency, albeit qualitatively, also appears to increase. There is a small blue-shift ($\approx$4 nm) of the long-wavelength band-edge with the application of the field, but this is likely to have a minimal effect on the gain curve. The reduction in the threshold is considered to be due to a stabilization of the helix and dampening of the thermally induced director fluctuations. The slope efficiency of the band-edge laser is found to be $\sim$15%, which is less than that observed for the same dye in other LCs and is considered to be due to the lower birefringence of the nematic host.[10] Figure 3(b) is a plot of the full width at half maximum (FWHM) of the random laser emission spectrum as a function of the excitation energy. For such lasers, the excitation threshold is typically defined as the point of inflection on the sigmoidal plot through the data. In this case, the excitation threshold is found to be approximately $E_{th} \approx 1.5$ $\mu$J/pulse, which is a factor of two lower than that recently observed for emulsion-based polymer dispersed LC lasers.[7] For the random laser, the slope efficiency is found to be $<2\%$ in the forward direction parallel to the normal of the glass substrates although the emission occurs over a much greater cone angle than that of the band-edge laser. The results show that, in order to observe a direct control of the emission linewidth from this laser sample, it is necessary to use excitation energies that are greater than 1.5 $\mu$J/pulse.

To summarize, we have demonstrated a single laser source that can exhibit two fundamentally different types of lasing mechanisms. Depending upon the frequency of the applied electric field that is used to address the liquid crystal, one can observe random lasing or band-edge lasing. Each type of laser exists within a different frequency regime. The ability to fabricate micron-sized laser devices, which can change the linewidth of the emission spectrum *in-situ*, opens up new avenues in the creation of laser devices that can be optimized for different environments. This is of particular importance when dealing with the issue of speckle in imaging systems.

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