Thermo-recurrent nematic random laser

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Abstract: This experimental work is aimed to investigate the thermal behavior of random laser action in dye doped nematic liquid crystals. The study evidenced an important temperature dependence of the random lasing characteristics in the nematic phase and in close proximity of the nematic-isotropic (N-I) phase transition. A lowering of the laser emission intensity as the temperature increases is strictly related to the shift of the lasing threshold as function of the temperature even though the pump energy is kept fixed. The optical losses increasing owing to the thermal fluctuation enhanced scattering drive the input-output smoother behavior until the system stops to lase, because below threshold. The unexpected reoccurrence of random lasing at higher temperature, in proximity of N-I transition is found to be related to a different scattering mechanism, the micro-droplets nucleation and critical opalescence.

OCIS codes: (140.0140) Lasers and laser optics; (140.6810) Thermal effects; (290.4210) Multiple scattering; (160.3710) Liquid crystals.

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1. Introduction

Lasers were invented some 40 years ago and they are still currently used in a variety of application fields. Lasing materials can be very different and range from periodic systems such as photonic crystals to partially ordered [1] and totally disordered dielectric materials that scatter light diffusively [2]. Since the development of the liquid crystals display (LCD) technology, a significant interest was devoted to the development and characterization of these materials as they can be easily manipulated and exhibit many interesting optical properties. Liquid crystals (LCs) are extremely promising materials for engineering photonic nano-structures, either as stand-alone devices, or as part of innovative integrated systems. Their use in the application fields can range from photonics to the bio-medical arena where miniaturized tunable laser sources may find a vast area of uses, such as lab-on-chip and optical tweezers, while recently, random lasers have known a relevant impact in the physics of soft-matter and even biological tissues examination [3]. A fundamental feature of these confined systems is that optical and geometrical parameters can be modified by applying external fields (e.g., temperature, electric field, mechanical stress), therefore resulting in a direct control of the emission characteristics (wavelength tunability, emission bandwidth, directionality).

Laser action in disordered media has been a subject of intense theoretical and experimental studies in the last few years. The propagation of the light waves in random systems is quite different from the conventional case, as optical scattering may induce a phase transition in the photon transport behavior. Initially, the light scattering events in a laser system were considered detrimental, as such regime could drain photons from the lasing mode, causing an increase in optical losses (in a conventional laser cavity). It was later established that a strong enough scattering regime could be the main responsible for the lasing process itself [4]. The most important parameter of any lasing medium is the gain coefficient that includes both the positive and negative absorption and scattering losses. This is also true for LCs, that retain absorption and refraction anisotropy and scatter light due to the strong fluctuations in the local optical axis. For very weak scattering, the propagation of light could be described as a normal diffusion process [5]. When increasing the scattering intensity, recurrent light scattering events arise and the interference between the counter-propagating waves leads to enhanced backscattering, namely weak localization of light. Furthermore, a system reaching a critical scattering level makes a transition into a strongly localized state as light transmission is inhibited, leading to the so-called Anderson localization state [6]. The later manifests itself as a phase transition in the diffusion constant for a certain value of the mean free path and it is exactly this parameter to determine the lasing threshold of a random laser. In the presence of a gain medium inside the system, the recurrent multiple scattering and amplification can substitute for the distributed optical feedback of a regular laser cavity [7]. Due to a random walk with optical gain, a photon may induce the stimulated emission of other photons in the system and diffusive laser action is expected [8].

2. Experimental set-up of random lasers

Random lasers have several properties reminiscent of common cavity lasers (*i.e.,* above threshold the spectrum narrows and the emission can present a spiking behavior). Diffusive lasing has a very peculiar behavior, as randomness of laser emission is observed in time, space and frequency [9]. Light localization and interference that survive to multiple scattering events have been invoked to explain the random lasing observed in a variety of exotic and complex systems such as powdered laser crystals, micro-particles in laser dye solution, and recently even in dye doped nematic liquid crystals (NLC) [10]. Several systems have been investigated by varying different parameters (*i.e,* the active medium type and the confinement

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geometry) with the aim to study the emission properties of weakly localized light in nematic liquid crystals [11].

In this manuscript, we report the thermal behavior and characteristics of random laser action in a dye doped nematic liquid crystalline system, by varying the temperature in the range 28 °C (Room Temperature) up to the nematic-isotropic transition.

Several liquid crystalline phases are opaque, meaning the light waves are multiply scattered. In particular, LCs in the nematic phase are optically anisotropic and strongly scattering materials, exhibiting turbidity and coherent backscattering due to fluctuations of the nematic director [12]. The spontaneous fluctuations of the nematic director $n(r,t)$ (the average molecular orientation over a small volume), given by $n(r,t) = n_0 + n(r,t)$, lead to fluctuations in the local dielectric function $\varepsilon_{\alpha\beta} = \varepsilon_\perp \delta_{\alpha\beta} + (\varepsilon_{//} - \varepsilon_\perp)\, n_\alpha n_\beta$. These phenomena are the main responsible of the recurrent multiple scattering events [13].

Anisotropic light diffusion in nematics was initially observed by Kao et all [14] in static conditions and then studied also by Wiersma et all [15] in time-resolved experiments. For high enough temperatures, the liquid crystal goes into the isotropic phase and behaves as an ordinary liquid. The refractive index of a liquid crystal is highly temperature dependent, creating interesting possibilities for studying the scattering properties in different phases by changing the respective system temperature.

Experimentally, our gain medium consisted of a nematic liquid crystalline mixture (BL001 by Merck) doped with 0.3% by weight dye molecules (Pyrromethene 597 provided by Exciton). The Pyrromethene dye molecules were dissolved in the NLC at very low concentrations (0.3% – 0.5% by weight) and proved to be completely miscible as evidenced by the almost complete absence of micro-droplets (as observed by means of an optical microscope). The LC bulk phase sequence is Crystal - (10 °C) Nematic - (63 °C) Isotropic, while the principal refraction indices of BL001 are $n_{//}$=1.73 and $n_\perp$=1.52 (at 25 °C).

The mixture was then confined in two different geometries: wedge cells and cylindrical capillaries. The wedge cells were constituted by two glass plates separated by Mylar spacers having a variable thickness between 20 and 200 μm. The plates were covered with rubbed polyimide alignment layers for inducing a homogeneous (planar) alignment of the NLC molecules at the interface. Cylindrical capillaries, presenting no surface treatment, having inner diameters ranging from 50 to 200 μm respectively were used as alternative confinement structures for our mixture as well. The pump light originated from a frequency-doubled neodymium doped yttrium aluminum garnet laser (NewWave, Tempest 20). It operated at a frequency of 20Hz and the 3ns long pulses were focused into a 50 μm spot on our samples by means of a spherical lens (Fig. 1). Various input pump energy values were experimented while the beam was set at small incidence angle with respect to the plane of the sample. For analyzing the lasing emission properties as a function of temperature we placed the samples inside a purposely built teflon oven (CalcTec). We spectrally analyzed the resulting light emission from our systems by using a high resolution optical multi channel charge coupled device CCD spectrometer (Jobin-Yvon) having a resolution of about 0.3 nm.

By pumping the systems, at low pump energy, an isotropic fluorescence profile typical of dye molecules was detected. Upon increasing the pump energy above a given threshold, the emitted light emerged along the excitation volume and sharp bright tiny spots appear in the far field zone as numerous small lasing speckles. (Fig 1, inset (a)). The experimental study of the spectral and spatial properties of the emission patterns clearly revealed a random lasing behaviour characterized by narrow banded (FWHM = 0,5 nm) radiant spikes (spectrum in Fig. 1 – inset (b)) which rapidly fluctuate in frequency and spatial position (Fig. 1 - inset (a)). This mechanism is based in principle on the loss-gain balance theory: for a strong optical scattering, light may return to a diffuser from whom it was speckled before forming a closed loop path. When the amplification along such a loop exceeds the losses and the gain–loss balance become positive laser oscillation is highly expected.

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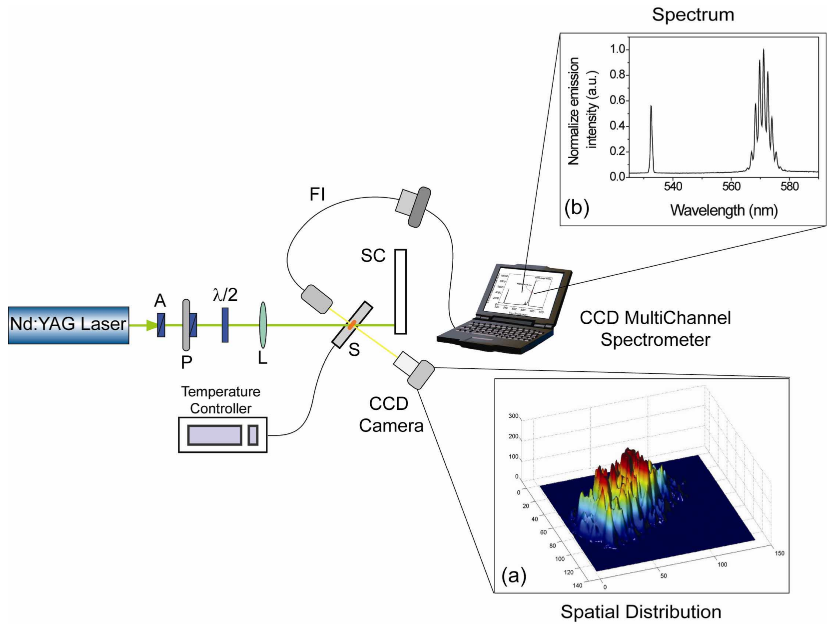

Fig. 1. Schematic diagram of the experimental set-up. A=Attenuator, P=Polarizer, S=Sample, SC=Screen and FI=Fiber. Inset (a) represents the lasing spectrum for a wedge cell maintained at fixed temperature. (b) The intensity spatial distribution is acquired by a high resolution CCD camera, demonstrating a speckle like behavior.

3. Results and discussion

In order to expand our understandings on the mechanisms behind laser action in partially ordered systems, an experimental study was performed dealing with the investigation of the input-output light emission characteristics for different system temperatures.

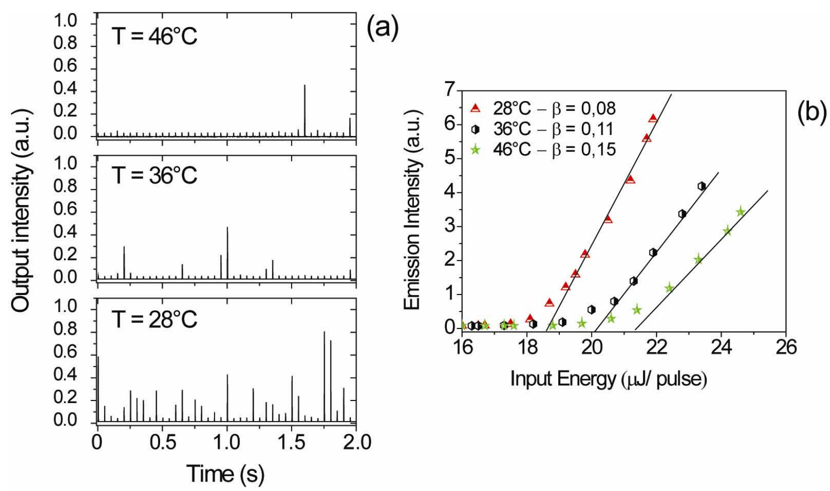

Fig. 2. (a) Time resolved emission intensity behavior in a wedge cell at different temperatures (T = 28 °C, 36 °C, 46 °C). (b) Emission intensity from a wedge cell versus pump energy. With increasing temperature, a smoother lasing threshold behavior is obtained, as shown by the black lines used as guide to the eye.

The light emission intensity was recorded as a function of the pump energy for both types of systems (wedge cell and capillary). The sharpness of the lasing threshold is determined by the *β*-factor, defined as the ratio between the rate of spontaneous emission radiated into the lasing modes and the total rate of spontaneous emission [16]. Figure 2(b) shows that in our systems *β*-factor can be tuned by temperature. In fact, the enhancement of thermal fluctuations leads to an increase of the scattering intensity. This results in a lowering of the volume of random cavities because of mean free path shortening at higher temperature.

This temperature sensitive behaviour is shown in Fig. 2(a), where we report the time resolved emission intensity as a function of temperature in a wedge cell (28, 36 and 46 °C). We notice that for T = 28 °C we have a lasing event roughly corresponding to each pump pulse (frequency = 20 Hz), while increasing temperature, a gradual lowering in the output light intensity and an intermittent and irregular emission prevails, until laser action completely fades to zero when approaching the nematic-isotropic phase transition. In Fig. 2(b) we observe that, upon increasing sample temperature, the diminished nematic order parameter results in a higher lasing threshold, approaching the behaviour of the totally disordered systems [17]. For a fixed pump energy value, the gain-loss balance is regulated by temperature. In fact only for a certain temperature region the chosen input energy is above threshold, then sufficient for triggering the lasing process. Whereas, at higher temperatures, the change of the scattering strength (coming from thermal fluctuations) produces a shift of the lasing threshold towards higher values, leading the system below threshold for a fixed pump energy (Fig. 2(b)). In fact, in this same region, a gradual attenuation of the lasing intensity is observed, until the emission vanishes around 55 °C for the wedge cell (Fig. 3(a)) and 45°C for the capillary (Fig. 3(b)).

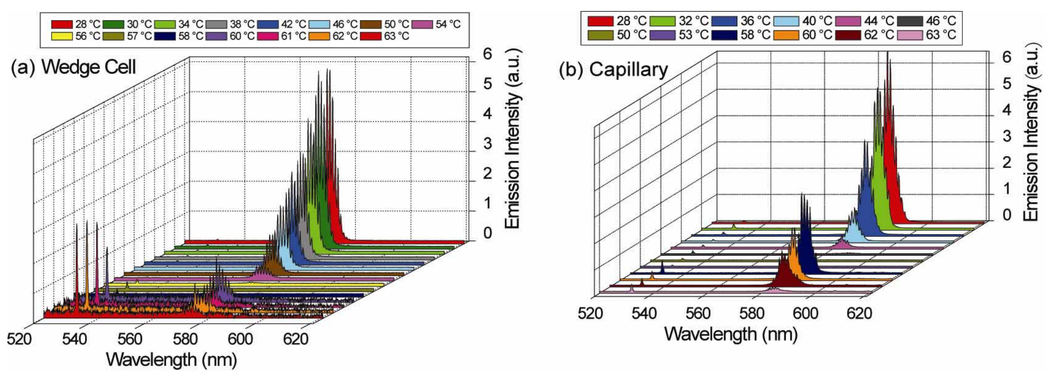

Fig. 3. (a) Emission intensity dependence on temperature for a wedge cell. (b) Emission intensity dependence on temperature for a capillary with a transverse section of about 200 µm.

This different behavior of the two samples can be explained by considering dissimilar experimental conditions (pump energy, sample thicknesses, confinement geometry, symmetry constraints, etc.). When further increasing the temperature and approaching the nematic-isotropic transition, surprisingly, laser emission reappears accompanied by a strong ligthwaves scattering as evidenced by the enhanced pump signal in the acquired spectra (Fig. 3(a)). This new effect survives for a limited temperature range, while the emission intensity progressively decreases until completely switches off when reaching the isotropic phase. Optical microscope investigations were performed in proximity of the transition by varying temperature from isotropic towards nematic phase (Fig. 4).

Just below the isotropic-nematic phase transition is evidenced the nucleation of nematic micro-droplets immersed in an isotropic environment, appearing as a poly-dispersion (see Fig. 4(a)). The concentration of liquid-crystalline micro-droplets increases with decreasing temperature, then they coalesce in larger droplets until give rise to the homogeneous nematic phase. In the range {63 – 58°C} (Fig. 4) the size of the micro-droplets and the environmental conditions result appropriate for enhancing the multiple scattering process [18], while optical gain exceeds internal losses and the system starts to lase again.

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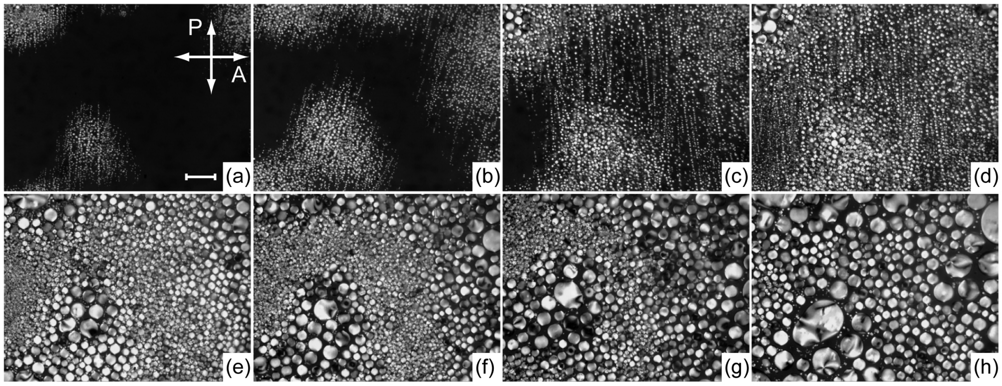

Fig. 4. Crossed polarizers optical microscopy images of a wedge cell filled by our mixture, taken for temperatures below the isotropic/nematic transition. (a) 62.5 °C; (b) 61 °C; (c) 60 °C; (d) 59 °C; (e) 58 °C; (f) 57.5 °C; (g) 57 °C; (h) 56.5 °C. Size bar in (a) is 300 μm.

This process is demonstrated to be temperature reversible (i.e. when going from isotropic to nematic and viceversa). The colossal increase of the scattering regime in this region is reported in literature [19]. These observations clearly suggest that the main responsible of laser action here remains the feedback provided by the enhanced scattering. The difference with respect the laser action obtained within the pure nematic phase consists of the origin of the scattering phenomena that here are due to poly-dispersed micro-droplets of nematic liquid crystals in an isotropic complex fluid solution. Therefore, the critical opalescence, resulting in very efficient scattering of light, lies at the basis of the observed diffusive random laser action [19].

4. Conclusion

The thermal analysis of the emission properties of nematic liquid crystals doped with fluorescent guest molecules and confined in diverse symmetry boundaries emphasizes a striking temperature behaviour. Remarkable is the change of the input-output curve found experimentally as the temperature is varied within the nematic phase range. The lasing threshold usually extrapolated by the input-output curve overcomes an important shifting towards higher values as the temperature is increased. This is directly responsible for the monotonic decreasing of the random lasing intensity even though the pump energy is kept fixed. The laser emission generated by the feedback provided by thermal fluctuations scattering shows clearly the characteristics of random emission by varying stochastically in time, space and frequency. Even more interesting is the reoccurrence of random lasing as the nematic-isotropic phase transition is approached. The already reported critical opalescence effects manifested as a colossal increase of the scattering regime at the N-I transition suggested that a new feedback mechanism is behind the observed lasing effect. Here, the mechanism at the basis of the amplification process is due to the multiple scattering yielding weak localization of light waves, but now the scattering is generated by poly-dispersed nematic micro-droplets immersed in an isotropic environment which nucleated as the N-I transition is approached. This mechanism is referable to that responsible for random lasing in aggregated nanoparticles dye solutions [20]. The scientific aspects presented in this manuscript overlook features of great interest characteristic of laser physics and material science.

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