Amplification of light and random laser action in partially ordered dye-doped nematics

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Experimental evidence of random laser action in a partially ordered, dye doped nematic liquid crystal with long-range dielectric tensor fluctuations is presented. When exceeding a certain threshold pump power value, the fluorescence curve in the system collapses and distinct sharp peaks emerge above the residual spontaneous emission spectrum. Unlike distributed feedback mirror-less lasers, this nematic system can be regarded as a cavity-less microlaser where the partially ordered bulk surprisingly plays the amplifying role, acting as a randomly distributed feedback laser. Here, the unforeseen surviving of constructive interference in repeated multiple scattering events of the dye emitted photons provide the necessary optical feedback for lasing in our liquid crystalline material. The random laser emission has attractive features, such as an extremely narrow banded linewidth (0.5 nm FWHM), low emission threshold (30 µJ/mm^2) and a very high efficiency (>20%). Different confining geometries are experimentally investigated, while obtaining intensity fluctuations of the spatially overlapping speckle–like emission patterns clearly demonstrates the typical spatio-temporal randomness of diffusive laser emission. These interesting inorganic lasing systems could represent an exceptionally promising route for fundamental prospective studies with strong technological implications for integrated optical systems, nanophotonics and optoelectronic fields.

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1. Introduction

Interest in structured organics and more complex dielectric materials is pervading many areas of the physics community, not only by spurring interest in further clarifying its conceptual relationships with more usual arrangements of matter, but also by offering some promising possibilities for scientific and technological applications. Diffusion, simultaneously with propagation phenomena of light waves inside complex dielectric structures have led to a wide series of experimental and theoretical investigations in the field, uncovering one of the most challenging, yet exciting, scientific area of the past decade. The study of spontaneous emission, light amplification and lasing properties in ordered and periodic systems acknowledged an amazing boost in the last couple of years, even because of the remarkable progress of various techniques allowing for the production of photonic crystal structures as low as nanoscale ranges [1]. Unpredictably, active random media repeatedly confirmed to be the appropriate contenders for achieving diffusive laser action, mainly based on the resonant feedback mechanisms in multiple scattering, thus eliminating the requirement for an external cavity (as for regular lasers). Light localization and interference effects which survive the multiple scattering events have been frequently invoked to explain the random laser action observed in many exotic and complex systems [2-7]. The optical scattering phenomenon inside a random medium is capable of inducing a phase transition in the photons transport behaviour. The interference between the counter propagating waves in a partially or totally disordered structure gives rise to the enhanced backscattering, also called weak localization [8,9]. When the scattering level is increased beyond a threshold critical value, the system suffers a transition into a localized state. The Anderson localization is fully based on the interference effect [10], which is a regular characteristic of all wave phenomena. It is in some way natural to extend the electron localization issue to photon localization (with some variance), while in disordered dielectric media. An interesting phenomenon, which would never occur in an electronic system, is represented by the laser action in a disordered gain medium [11-13]. In case of strong scattering and high gain, the multiple recurrent scattering events can provide the coherent feedback required for lasing [14,15]. In this situation, if the amplification along a closed loop path exceeds the loss in the system, laser oscillation could develop while the loop performs as a laser resonator. All of the backscattered light waves interfere and establish the lasing frequencies. Such a laser system is called a "random laser". Random lasers represent a non-conventional laser whose feedback is mediated by random fluctuation of the dielectric constant in space. Since the original predictions of Letokhov and co-workers in 1968 [16], many theoretical and experimental investigations have been performed in disordered media [17-22]. Random lasers have been fabricated in various media, spanning from semiconductor nanoparticles, ceramic powder to polymers, organic materials and even biological tissues. Their reduced manufacturing costs, sample specific lasing frequencies, small size, flexible shape and substrate compatibility could lead to vast potential applications.

First experimental confirmations of random lasers are dated 1993, while later on remarkable progress in the field was done by A. Lagendijk [19,20,23,24], D. Wiersma [4,17,25-27] and H. Cao [11,13, 28-30]. They showed the various ways to obtain a random amplifying medium: nanoparticle suspension of TiO_2, laser crystals, Ti:Sa, ZnO powders for high scattering and gain and many others. The random laser not only becomes the newest member of the laser family, but also establishes a new ground for investigating localization phenomena [31,32].

Coherent backscattering experiments revealed weak localization of light even in tensorial systems characterized by increased optical anisotropy, such as nematic liquid crystalline materials [33,34]. Nematic liquid crystals (NLCs) are uniaxial fluids with rod-like molecules aligned on average along a local anisotropy axis which is represented by the unit vector n(r,t), the molecular director [35]. The spontaneous fluctuations of the director represented by $n(r,t) = n_0 + \delta n(r,t)$ lead to fluctuations in the local dielectric tensor, $\varepsilon_{\alpha,\beta} = \varepsilon_{\perp}\delta_{\alpha\beta} + (\varepsilon_{//} - \varepsilon_{\perp})n_{\alpha}n_{\beta}$, which represents the effect responsible for the recurrent multiple scattering events as a light wave is propagating through the NLC medium. Recently, the first experimental investigations to show random laser action in a partially ordered and highly anisotropic nematic liquid crystals doped with fluorescent guest molecules were presented [36-42]. The studies of laser generation and emission this system emphasized the peculiar behaviour of diffusive laser action, randomness of lasing was observed in time, space and frequency.

In this manuscript, we present the important features of random lasing emission properties in nematic systems. Experiments confirm a very low energy threshold, high emission yield, wavelength in yellow region of the spectrum, narrow spectral line, speckle spatial profile and nanosecond pulse duration.

2. Materials and methods

The investigated active mixture is represented by the BL001 nematic liquid crystal (provided by Merck) having the following bulk phase sequence Cr. -10 °C – Nematic – 63 °C – Iso, subsequently doped with 0.3 – 0.5 wt% of fluorescent dye. Different commercially available dyes (by Exciton) have been employed: Pyrromethene 597, Pyrromethene 650 and DCM. The dye molecules dissolved in the NLC were checked to be completely miscible, as evidenced by the almost complete absence of micro-droplets of dye embedded in the nematic phase.

The experimental set-up used to perform the optical emission analysis on these systems is presented in Fig. 1. The samples (S) were optically pumped with 3-5 ns pulses produced by a frequency-doubled (532 nm) Nd:YAG laser (NewWave, Tempest 20). The pump beam was focused onto a region of the sample (about 150 μm thick) by means of a spherical lens (L, f = 100 mm) yielding a beam waist of about 20 μm at the focus position. The experimental set-up presents a combination of optical elements (filters (F), a Glan-Thompson polarizer (P), a half wave plate (λ/2)), for allowing the control upon the input beam intensity and polarization state. A high spectral resolution (0.5 nm) CCD multichannel spectrometer (Jobin-Yvon) having a fiber termination was used to characterize the emission spectra, within a limited cone angle of approximately 0.05 rad. The speckle-like pattern of the random emission spot was imaged on a screen while simultaneously the emission spectrum was captured by means of the CCD spectrometer.

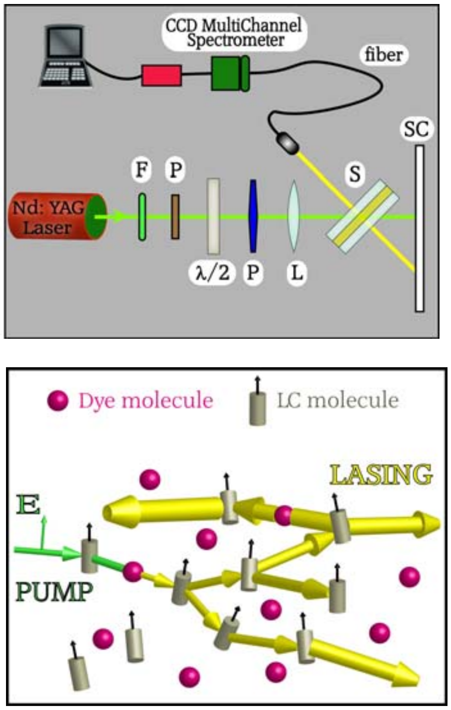

Fig. 1. (Top) Schematic representation of the experimental set-up: filter (F), polarizers (P), λ/2 – wave plate, lens (L), sample (S), observation screen (SC). (Bottom) Recurrent multiple light scattering in dye doped nematics provides the feedback for laser oscillations.

3. Experimental results and discussions

The active mixtures were enclosed in various confinement geometries, such as a wedge cell (Fig. 2a). Moreover, boundary-free systems were fabricated, where the mixture could be either free-standing (Fig. 1b) or freely suspended (Fig. 1c).

The first system (wedge cell) is made up by two glass-ITO plates separated by Mylar spacers, with a thickness of 150 μm at one side and a few μm at the other one. The inner surfaces of the ITO plates are covered with rubbed polyimide layers for inducing a predetermined homogeneous alignment for the NLC molecules at the interface. In order to exclude any cavity effect because of the multiple reflected light waves at the boundaries, the empty wedge cells were analyzed by measuring the transmission and reflection spectra with a high sensitivity spectrophotometer (UV-visible-near-infrared Cary 500 by Varian). The experiments revealed the absence of constructive interference in these cells, either by using a large aperture (2 mm × 10 mm) or small pinhole aperture (75 μm diameter) of the illuminating spot or tiny spot. In addition, the free spectral range was calculated by assuming a cavity effect and it was found to be absolutely incompatible with the linewidth of the observed lasing modes and with the spectral spacing of the modes.

The wedge cell was filled by capillarity, by directing the flow along the polyimide rubbing direction and normal with respect to the wedge. By analyzing the samples with a crossed polarized microscope a planar alignment was observed, with the optical main axis situated in the plane of the cell - parallel to the polymer rubbing direction.

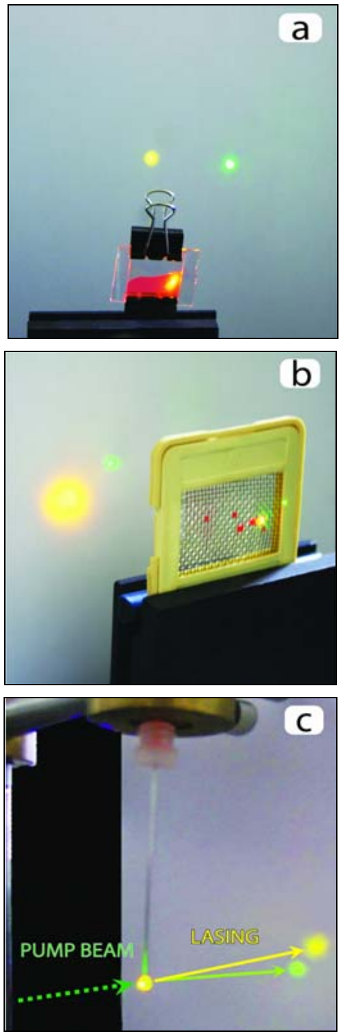

Fig. 2. Different random lasing confinement geometries: (a) wedge cell; (b) freely suspended film; (c) droplet system.

Interesting configurations were designed for creating the condition of almost complete absence of boundaries for the dye doped nematic liquid crystalline film. Thus, we were able to investigate the role played by various order degree levels in unbounded partially organized fluids, leading to fascinating organic multi-mode lasers. The reason for creating these systems is represented by the opportunity to explore the mechanisms and the interplay of amplification and localization in the absence of confinement. By freely suspending organized fluids, it is possible to fully take advantage of the unique optical properties of these materials which can be modified by external parameters, i.e. magnetic, electric, thermal, and mechanical stimuli.

The mixtures were freely suspended on a PVC net (Fig. 2b), creating squared combs (800 μm × 800 μm) while presenting a film thickness of about 300 μm. The suspended films observed at the optical microscope show a multi-domain structure where the single domain possesses a residual birefringence. A partial order could be obtained due to the flow induced by the spreading process over the comb. The profile of menisci driven by competitive forces (gravity, viscosity and surface tension) across the medium present a thinner central region of about 300 μm. Another lasing system in shown in Fig. 2c: the anisotropic mixture was suspended in a boundary-free condition. A droplet (having the diameter of approximately 2 mm) was shaped by gravitational strength by means of a special pipette syringe.

In all cases, the systems were optically pumped by means of the Nd:YAG laser and, above a given energy threshold value the random lasing effect was achieved (as seen in Fig. 2).

Subsequently, a spectral analysis versus input pump energy was performed. At low pump energy density (cca. 20 μJ/mm^2), the emission spectra shows the typical spontaneous emission curve of PM597 dye molecules, signifying that the nematic does not significantly modify the fluorescence spectrum (Fig. 3a). Upon increasing the pump energy density above a given threshold value (30-45 μJ/mm^2), discrete sharp spikes materialize from the residual fluorescence spectrum (Fig. 3b). The spectral line widths of these peaks were measured to be less than 0.5 nm, yielding a cavity quality factor, Q, larger than 1000 for our random system.

When the incident pump energy density exceeds the threshold value, the lasing peak intensity increases much more rapidly with the pump power and several more spikes appear on the spectrometer chart (Fig. 3c). Now, the gain in the system becomes clearly positive and other medium efficiency laser modes arise from the residual fluorescence.

A measure for the gain narrowing is the narrowing factor (NF), defined as the ratio between the Full Width Half Maximum (FWHM) of the emitted light below threshold ($\mathrm{FWHM_{below}}$) and the FWHM of the emission spectrum of the random laser above threshold ($\mathrm{FWHM_{above}}$):

$$NF \equiv \frac{FWHM_{below}}{FWHM_{above}}.$$

In the case of the wedge cell spectra, the narrowing factor was calculated to be around 50. Once above threshold, random laser emission is obtained in both forward and backward directions (with almost identical intensities). Thus, the total output energy (corresponding to a given input energy) of the laser emission results as twice the value obtained in one direction (i.e. forward). The net efficiency was experimentally determined to be more than 20%, a relatively high value for this category of systems.

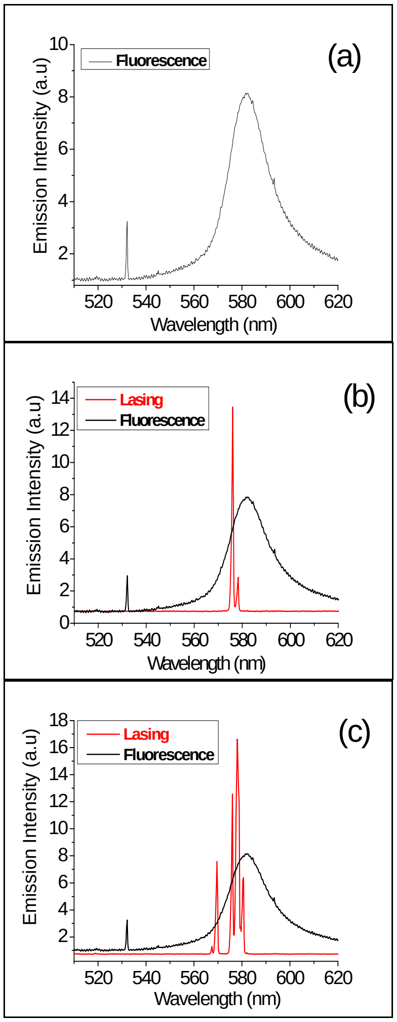

Fig. 3. Fluorescence and random lasing spectra measurements, in the case of a wedge cell when increasing the pump energy (a-b-c). Discrete sharp peaks emerge from the residual spontaneous emission for a pump energy density above 30-40 μJ/mm^2 (b). At higher pump values multiple lasing spikes are generated (c). The 532 nm diffuse pump signal is also present in the spectra. Residual fluorescence signal was multiple times magnified, for clarity reasons.

Another set of measurements was performed for characterizing the far field spatial distribution of the emitted random laser light. Lasing emission was captured in a limited cone angle (~ 0.1 rad) by means of a high resolution and sensitivity CCD camera (1390 × 1024 12bit PixelFlyQe by PCO). Fig. 4 shows an interesting scenario of the random lasing emission pattern from dye doped nematics confined in a wedge cell geometry.

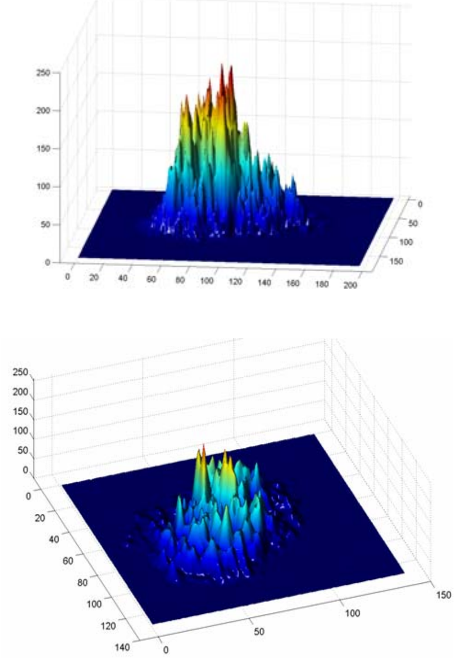

Fig. 4. Far field spatial distribution of the lasing intensity profile for a wedge nematic sample show an irregular intermittent behaviour of the emission, both spatial and temporal (top, bottom).

The intensity profile is continuously fluctuating in time, space and position demonstrating once more the randomness characteristic of the laser action in the investigated media. The intensity profile is formed by a series of various spikes that spatially overlap, generating a richly structured pattern. This is because of the emission mechanism being based on the diffusive process of spontaneously emitted photons by fluorescent guest molecules launched at random directions from random positions.

4. Conclusions

We characterized the process of random laser action in confined dye doped nematics, for various boundary restraining conditions. We tailored different wedge cells, freely suspended films and droplet shaped systems. The analyzed random lasing emission spectra peaks are extremely narrow banded (about 0.5 nm) and occur at a relatively low energy density threshold (of about 30 μJ/mm^2). The net efficiency was also determined experimentally to be more than 20% (both forward and backward directions), which is a relatively elevated value for this class of systems. The underlying mechanism of obtaining random lasing is mainly based on interference effects which survive recurrent multiple scattering driven by nematic director fluctuations. Weakly localized light waves in dye doped nematic sample are responsible for amplification while the resonance frequencies are selected through interference phenomena of the counter-propagating light waves within the localized loops. Therefore, this random system can be considered as a cavity-less microlaser where the disorder unexpectedly plays the most important role, behaving as a randomly distributed amplifier. A detailed analysis for the far field modal profile of the emission intensity shows a series of spatially overlapped emission intensity spikes, generating a changing speckled pattern which fluctuates in time, position and intensity. Further studies are in progress for fully understanding the diffusive laser action in these interesting dye-nematic liquid crystalline materials. The presented scientific results trace a very promising route for future fundamental studies and innovative applications in the fields of condensed matter physics, opto-electronics and photonics.

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