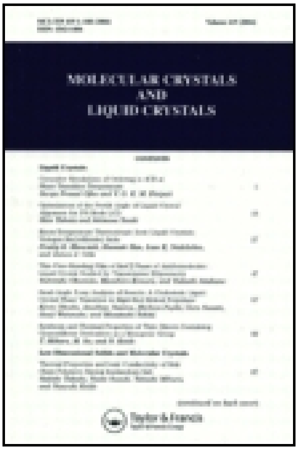

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Model for Light Scattering and Lasing in Dye-Doped Nematic Liquid Crystals

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Phenomena associated to lasing in random or quasi-random media are of growing interest both from theoretical and experimental points of view. Recent studies show that nematic liquid crystals are good candidates as scattering host for these lasing systems.

We present theoretical analysis and Monte Carlo simulations for different modelizations of scattering in nematics coupled with a two-level system which describes the dye action. We also show that a simple description of light diffusion in nematics is inadequate to describe the localization process that is at the basis of the laser action; therefore, different physical phenomena have to be taken into account.

Keywords: liquid crystals; random-lasing; scattering

INTRODUCTION

Since the first appearance of random lasing enormous interest has been focused in trying to understand the mechanisms driving this interesting and unexpected phenomenon. Random laser action in fully disordered systems has been demonstrated in a varieties of materials such nano-powdered dye solutions, colloidal suspensions, etc [1]. In fact a fascinating situation occurs when a gain mechanism is added to a disordered dielectric. Multiple scattering of light, with optical gain can lead to various forms of lasing [2–4]. Lasing in random systems is generally referred to as the random lasing, which is the topic of this paper. Recently the first observation of random laser action in

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partially ordered nematic liquid crystals (NLC) has been reported [5]. Laser emission in such systems is due to the combined action of multiple light scattering induced by random fluctuation of dielectric constant in space and light amplification produced by the gain medium.

In order to further understand this mechanism we reported in this work a theoretical model coupling a model for scattering in NLC with a description of light propagation in gain medium.

EXPERIMENTS

Several systems have been investigated by varying the dye molecules and the geometry with the aim to study the emission properties of weakly localized light in nematic liquid crystals [7]. The gain media consist of a nematic liquid crystal (NLC) mixture (BL001 by Merck) doped with 0.3 wt% of Pyrromethene PM 597 dyes (Exciton). The liquid crystal bulk phase sequence is (-10°C) Crystal-Nematic (63°C) Nematic–Isotropic. The mixture was confined in a wedge cell constituted by two glass plates separated by Mylar spacers, with a thickness of 200 μm at one edge and 2 μm at the other one. The inner side of the plates were treated with rubbed polyimide alignment layers in order to induce a privileged molecular alignment. Then, the wedge cell was filled by capillarity with the flow direction along the rubbing direction and normal with respect to the wedge. Upon observing the sample under a polarized microscope, it shows a planar alignment with the optical axis which lies in the plane of the cell parallel to the rubbing direction. The wedge sample was 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 the thick region of the sample with a spherical lens ($f = 100$ mm) yielding a beam waist of about 30 μm at the focus position. The experimental set-up presents a combination of optical elements in order to select all the states of polarization of the pump beam. A multichannel CCD spectrometer with a high spectral resolution (0.5 nm) and with a fiber termination was used to capture the emission spectra within a limited cone angle of 0.05 rad. The speckle-like pattern of the emission spot was imaged on a screen while simultaneously the emission spectrum was captured by means of the CCD spectrometer (see Fig. 1)

At low pump power, the emission spectra show the typical spontaneous emission curve of dye molecules, indicating that NLC does not considerably modify the fluorescence spectrum. Upon increasing the pump power above a given threshold value discrete sharp peaks emerge from the residual fluorescent spectrum. The line width of these

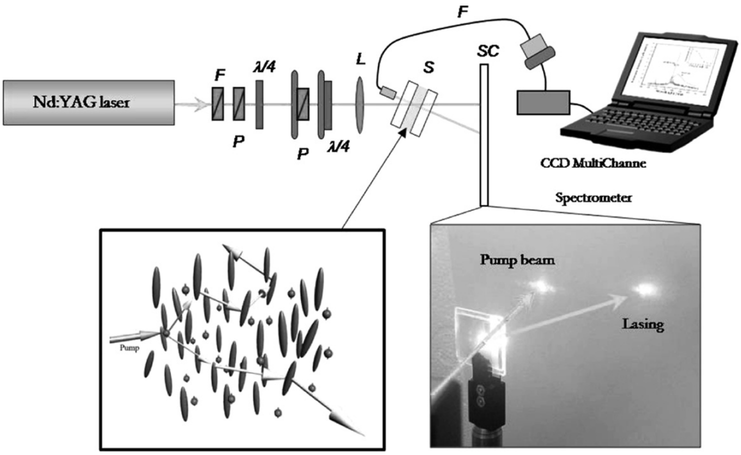

FIGURE 1 Experimental apparatus for measuring the emission features of a random laser in nematic liquid crystal sample. The laser was a frequency doubled ($\lambda = 532\,\text{nm}$) Nd:YAG. It operated at 20 Hz repetition rate and had a pulse width of 5 ns. The emitted light was collected by a spectrometer that allowed a resolution of 0.5 nm. The pump beam was focused on the sample, resulting in a spot size of 40 mm of diameter.

sharp peaks were less than 0.5 nm, yielding a quality factor Q of this random cavities larger than 1000.

THEORETICAL MODEL

In order to describe laser action in dye doped NLC it is necessary to couple a suitable description of photon gain in an active media, with an exhaustive depict of scattering in nematics. Mechanisms besides positive feedback in random lasing was widely studied considering scattering in turbid, isotropic media. In the case of NLC, they present interesting complications for light diffusion that extend beyond isotropic, random media. To describe the effect of NLC as a scattering host we have chosen as a reference the work of Stark *et al.* [7] were they investigate the nature of light diffusion and correlation transport through orientationally ordered turbid materials.

Our system is represented by a sample of width S filled with dye-doped NLC whose planar alignment respect to the glass slabs is defined by the equilibrium director $\mathbf{n}_0$. A sequence of scattering events define a light trajectory until it proceed inside the sample. Every trajectory is characterized by its position $\mathbf{r}$ calculated in the laboratory frame defined with the z-axes parallel to $\mathbf{n}_0$ and the y- axes perpendicular to the glass slabs.

The NLC is characterized by tree elastic constants *k_{1, 2, 3}*, and by the dielectric constants $\varepsilon_\perp$ and $\varepsilon_{||}$.

A single scattering event is characterized by an incoming wavevector $\mathbf{k}$ with polarization a and an outcoming wavevector $\mathbf{q}$ with polarization b. In order to define a structure factor $B_{\alpha\beta}(\mathbf{k}, \mathbf{q})$ measuring correlation between $\mathbf{k}$ and $\mathbf{q}$ we need to define a new frame of reference ($\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$) for describing scattering: $\mathbf{u}_1$ and $\mathbf{u}_2$ are orthogonal unit vectors perpendicular to $\mathbf{n}_0$, $\mathbf{u}_1$ lies in the $\mathbf{k}$–$\mathbf{n}_0$ plane, and $\mathbf{u}_2$ is perpendicular to this plane, $\mathbf{u}_3$ is along $\mathbf{n}_0$

$$\hat{\mathbf{u}}_2 = \frac{\mathbf{n}_0 \times \hat{\mathbf{k}}}{|\mathbf{n}_0 \times \hat{\mathbf{k}}|}$$

$$\hat{\mathbf{u}}_1 = \hat{\mathbf{u}}_2 \times \mathbf{n}_0$$

Thus we get:

$$B_{\alpha\beta}(\hat{\mathbf{k}}, \hat{\mathbf{q}}) = \Delta\varepsilon^2 K_B t \frac{\omega^4}{c^4} \sum_{\delta=1}^{2} \frac{N(\hat{\mathbf{e}}^\alpha, \hat{\mathbf{e}}^\beta, \hat{\mathbf{u}}^\delta)}{k_\delta(\hat{\mathbf{q}}s)}, \tag{1}$$

where $k\delta(\mathbf{q}) = \kappa_\delta q_\perp^2 + \kappa_3 q_{||}^2$ while $N(\hat{\mathbf{e}}^\alpha, \hat{\mathbf{e}}^\beta, \hat{\mathbf{u}}^\delta)$ is given by

$$N = [(\mathbf{n}_0 \cdot \mathbf{e}_\beta)(\mathbf{u}_\delta \cdot \mathbf{e}_\alpha) + (\mathbf{u}_\delta \cdot \mathbf{e}_\beta)(\mathbf{n}_0 \cdot \mathbf{e}_\beta)]^2 \tag{2}$$

w is the light frequency and $\hat{e}^1$ and $\hat{e}^2$ are electric fields related to *extraordinary (1)* and *ordinary (2)* polarization. By utilizing the structure factor defined in (1) is possible to calculate all physical quantities involved in the scattering event. In this treatment, the free main path is [7,8]:

$$l_s = \frac{\kappa_3 c^2}{\Delta\varepsilon^2 \kappa_B T \omega^2} l_\alpha, \tag{3}$$

where l_α is defined by:

$$\frac{1}{l_\alpha(\hat{\mathbf{k}})} = \frac{n_\alpha(\hat{\mathbf{k}})}{4\pi} \sum_{\beta=1}^{2} \int \frac{d\Omega_q}{4\pi} B_{\alpha\beta}(\hat{\mathbf{k}}, \hat{\mathbf{q}}) n_\beta^3(\hat{\mathbf{q}}) \tag{4}$$

It is important to underline that l_s depends on w.

Every light trajectories is also characterized by the occupation number of photons N^σ, while a trajectory propagate inside the sample, this value depends on the effect of the gain media.

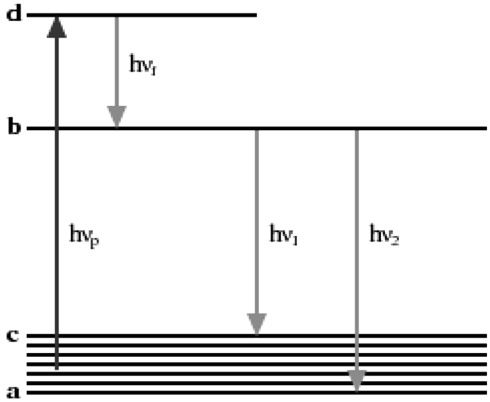

FIGURE 2 Typical tree level scheme for laser action. Level d is populated by the pump h$\nu_{p,}$ b is the meta-stable level while a-c are roto-vibrational levels thaking the role of the base levels.

The energy spectrum of a dye molecules, present a complicated structure of roto-vibrational levels. Taking into account that in our model we can deny the effect of the following transitions:

- Internal conversion
- Inter System crossing
- Triplet triplet absorption
- Phosporescence

become possible to sketch our system with usual tree level stigmatization for laser action presented in Figure 2

By considering that transition d-b is very fast and that dealing whit a frequency at time, we can considerate a single a- level at time, the equation describing the evolution of N^{σ} is given by [9]:

$$\frac{dN^{\sigma}}{ds} = \alpha[1 + \gamma N^{\sigma}] \tag{5}$$

where ds is the spatial step along the trajectory, $\gamma = 1 - n_a/n_b$ ($n_{a,b}$ are the level populations) and $\alpha = A^{\sigma}_{ab} n_b/v_g$ (v_g is the module of the group velocity and A^{σ}_{ab} related to the interaction hamiltonian).

ANALYTICAL SOLUTIONS

Equation (5) can be solved analytically giving the following expression for N_{σ}:

$$N^{\sigma} = \left(N_0 + \frac{1}{\gamma}\right)\exp(\alpha\gamma s) - \frac{1}{\gamma} \tag{6}$$

Expression 6 is exponential in γ, in order to understand how γ depends on the wavelength in the considered range, we analyse the experimental fluorescence curves presented in Fig. 3 the red curve refers to a sample of thickness 25 µm, while the yellow one to a curve relative at a sample of thickness 50 µm, both pumped by the same laser power.

It is easy to identify a right region in wavelength in which a longer path correspond to a greater photonic gain ($\gamma > 0$) and a left region in which a longer path correspond to absorption ($\gamma < 0$).

In order to understand how the combination of scattering in NLC and photonic gain in active media can generate a laser peack at a certain wavelength we have to consider that la given by expression (3) is inversely proportional to ω^2. A longer free main path means less scattering events and consequently a shorter total path. This mean that longer paths in the active region are more probable for lower wavelengths. Taking into account expression (6) this correspond to an exponential growth for N_s in the region characterized by $\gamma > 0$ (higher wavelengths) and to an exponential absorption in the region characterized by $\gamma < 0$ (lower wavelengths). The absolute value of γ is strictly related to the populations inversion (and then

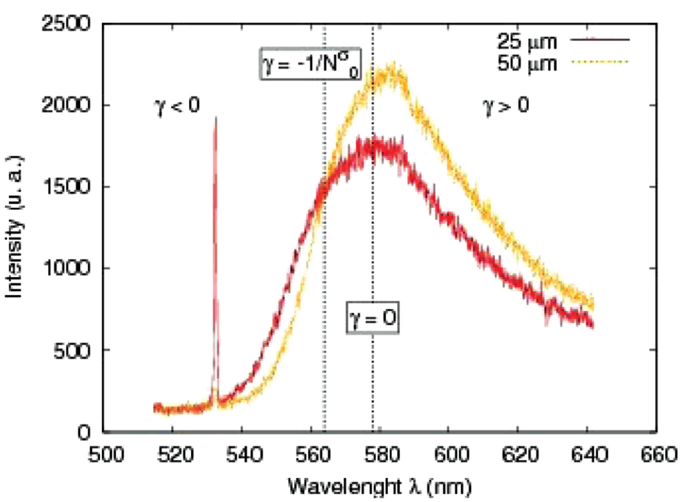

FIGURE 3 Fluorescence curve presenting output intensity as a function of wavelength. Dashed line correspond to a sample of thickness 25 µm, while the solid one refers to a sample of 50 µm. Both samples are pumped at the same power by a Nd:YAG laser.

to the pump intensity). Over a certain value of the pump intensity a sharp peak of diffusive origin can be produced.

MONTE CARLO SIMULATIONS

We also realized a Monte Carlo code describing a sequence of scattering events in NLC.

By utilizing the structure factor presented in (1), for every scattering event is possible to calculate

- the *path to walk* until next scattering events R:

$$R = \frac{l_\alpha}{\cos\delta \ln\left(\frac{1}{1-x_r}\right)}$$

where δ is the angle between the group velocity $\mathbf{v}_g$ and the wavevector $\mathbf{k}$, while x_r is a random number chosen in the interval [0:1]

- the *polarization* β after the scattering event, calculated by defining

$$C_{\alpha 1} = \frac{n_\alpha(\mathrm{k})}{4\pi}\int \frac{d\Omega_q}{4\pi} B_{\alpha 1}(\hat{\mathrm{k}}, \hat{\mathrm{q}}) n_\beta^3(\hat{\mathrm{q}})$$

$$C_{\alpha 2} = \frac{n_\alpha(\mathrm{k})}{4\pi}\int \frac{d\Omega_q}{4\pi} B_{\alpha 2}(\hat{\mathrm{k}}, \hat{\mathrm{q}}) n_\beta^3(\hat{\mathrm{q}})$$

from which we calculate:

$$R_{\alpha\beta} = \frac{C_{\alpha 1}}{C_{\alpha 1} + C_{\alpha 1}}$$

$R_{\alpha\beta}$ represents the probability that a beam with polarization α is scattered into a beam with polarization 1. Generating a random number in the interval [0:1] is possible to calculate final polarization β

$$\beta = \begin{cases} 1, & \text{if } g \leq R_{\alpha\beta} \\ 2, & \text{if } g > R_{\alpha\beta} \end{cases}$$

- the *outcoming wavevector* $\mathbf{q}$ defined by

$$\int_{\hat{\mathrm{q}}} B_{\alpha\beta} d\hat{\mathrm{q}} \geq C_{\alpha\beta} z_r$$

were

$$C_{\alpha\beta} = \begin{cases} C_\alpha 1, & \text{if } \beta = 1 \\ C_\alpha 2, & \text{if } \beta = 2 \end{cases}$$

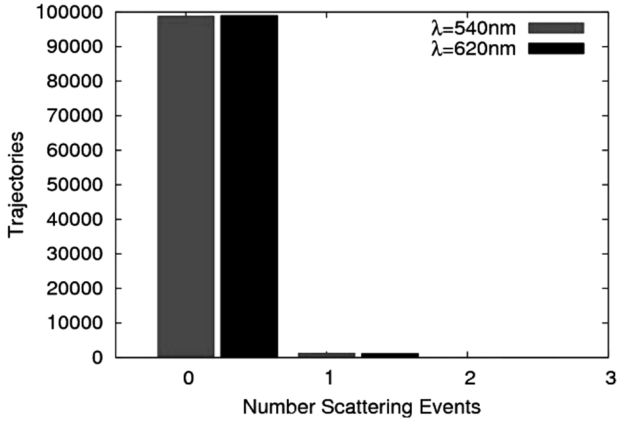

FIGURE 4 Scattering event statistic in pure NLC for $\lambda = 540\,\text{nm}$ (gray histogram) and $\lambda = 620\,\text{nm}$ (black histogram) relative to a sample with a thickness of 100 µm

A sequence of scattering events draw a light trajectory inside the sample. By utilizing in our simulations typical experimental values for the dielectric and elastic constants of E7 liquid crystal we generated 10^5 trajectories inside our 100 µm thick sample realizing a scattering events statistic for two wavelengths (540 nm and 620 nm) at the boundary of the considered range. Results of this characterisation are presented in Fig. 4 in that picture red histogram is relative to $\lambda = 540\,\text{nm}$, while black histogram is relative to $\lambda = 620\,\text{nm}$

It is evident that considering the scattering in pure NLC and a sample of thickness 100 µm the regime is balistic and there could be no difference between the two extreme wavelengths. In order to take into account that the presence of the dye molecules could change the distribution of nematic fluctuations, we introduced a correction factor C_{R} (operatively estimated 10^{-3}) applying to the free main path l_α becoming $C_{\text{R}} l_\alpha$. Results of the new characterisation are presented in Figure 5.

In the new characterisation is possible to see a difference between the two extreme wavelengths, this correspond to a different permanence in the active region and consequently to the formation of the peak over a certain pump power.

Results presented in Fig. 3 are relative to pure NLC, therefore the correction factor arise from interaction between NLC and dye-molecules. A wide range of possibility have to be investigated to understand the nature of this interaction. Reasonably it can be due to the Janossy

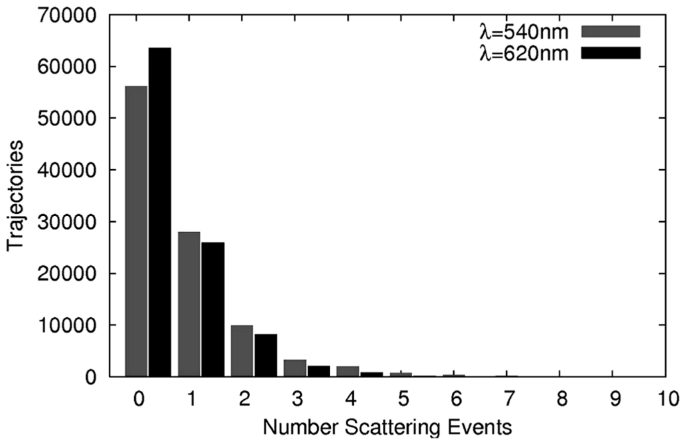

FIGURE 5 Scattering event statistic applying correction factor *CR* to the free main path $l\alpha$. Presented for $\lambda = 540\,\text{nm}$ (gray histogram) and $\lambda = 620\,\text{nm}$ (black histogram).

effect [10] but also sterical interaction between NLC and dye have to be taken into account.

CONCLUSIONS

Laser emission in dye-doped NLC is due to the combined action of multiple light scattering induced by random fluctuation of molecular director reorientation and light amplification produced by the gain medium. The combination of this effects provides an alternative feedback mechanism for lasing oscillations in respect to the feedback mechanism that characterizes a conventional cavity laser. Free main path dependence on wavelength increases the total path length of light in the active medium for lower wavelength, light amplification produced by stimulated emission identify a precise region in which the peak can be produced.

Results of a Monte Carlo simulation show that a correction factor of 10^{-3} have to be applied to the calculation of the free main path in pure NLC in order to have a diffusive regime in samples with a thickness like the ones utilized in experiments. This is due to the interaction of NLC with dye molecules that give rise to a modification in the director fluctuations distribution. Some physical effects that reasonable could be responsible of this modification has been reported.

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