Random Lasing Control with Optical Spatial Solitons in Nematic Liquid Crystals

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Keywords: Random Laser, Nematic Liquid Crystals, Optical Spatial Solitons.

Abstract: We discuss the synergy of reorientational self-focusing and random lasing in a dye- doped nematic liquid crystalline material. The laser emission resulting from amplification and multiple scattering inside the medium can be either modulated or triggered depending on the energy of the visible pump beam and the power of the near-infrared spatial soliton, respectively exciting the two nonlinear responses. Moreover, the presence of the self-induced waveguide improves the properties of the emitted beam, i. e., directionality and profile. Finally, the laser light can be re-directed by steering the spatial soliton with the aid of an external low-frequency electric field.

1 INTRODUCTION

Random lasing occurs when multiple recurrent scattering inside an optically active medium provides the necessary feedback for the stimulated emission to reach gain higher than losses (Wiersma, 2008). Since its prediction and the first experimental evidence (Letokhov, 1967; Lawandy, 1994), random lasers have attracted great attention, letting researchers envision the realization of low-cost tunable coherent light sources. As a drawback, their emission is randomic in direction, and the resulting profile has poor quality with respect to standard sources (Cao, 2003). During the last decades, a number of materials have been employed to generate and control random laser emission, including powders (Leonetti, 2011), biological tissues (Polson, 2004), conjugated polymers (Tulek, 2010), semiconductor polycrystalline films (Cao, 1998), perovskites (Safdar, 2018), and nematic liquid crystals (Strangi, 2006). The latter provide light scattering due to the thermal oscillations of their weakly linked molecules. At the same time, the low binding forces allow reorientation of the anisotropic molecules by optical beams, allowing nonlinear self-focusing and the generation of optical spatial solitons (Peccianti, 2003).

In this work, we combined the reorientational nonlinearity with the strongly scattering behaviour of a nematic liquid crystal mixture doped with a dye - acting as the active medium -, generating a nematicon and random laser emission at the same time (Perumbilavil, 2016), and studied their mutual interaction. The paper is organized as follows: first, we illustrate the principle of both random laser emission and propagation of spatial solitons in nematic liquid crystals. Then we expose the experimental results, while the last part is dedicated to the discussion of the results and the conclusion.

2 RANDOM LASING AND NEMATICONS IN DYE-DOPED NEMATIC LIQUID CRYSTALS

Nematic Liquid Crystals (NLC) are anisotropic materials featuring high reorientational nonlinearity owing to the torque induced rotation of the elongated molecules due to electric fields at either low or optical frequencies (De Gennes, 1993). Non uniform molecular reorientation generated by finite size beams compensates for diffraction, allowing for self-focusing and the formation of spatial solitons (Stegeman, 1999), i. e., optical wavepackets with

Piccardi, A., Perumbilavil, S., Kauranen, M., Strangi, G. and Assanto, G.
Random Lasing Control with Optical Spatial Solitons in Nematic Liquid Crystals.
DOI: 10.5220/0007575102890293
In *Proceedings of the 7th International Conference on Photonics, Optics and Laser Technology (PHOTOPTICS 2019)*, pages 289-293
ISBN: 978-989-758-364-3

invariant profile in propagation (Segev, 1992; Peccianti, 2000; Leo, 2004; Rotschild, 2006)
The nonlocal character of the response prevents catastrophic collapse - typical of local media -, supporting the stable propagation of 2D spatial solitons, operating as self-induced waveguides for optical signals at different wavelengths (Conti, 2003). The generation and control of spatial solitons in nematic liquid crystals, namely nematicons, have been demonstrated in a number of configurations, together with their possible applications in devices for signal addressing and processing (Piccardi, 2010; Piccardi, 2016; Izdebskaya, 2017; Laudyn, 2018) .

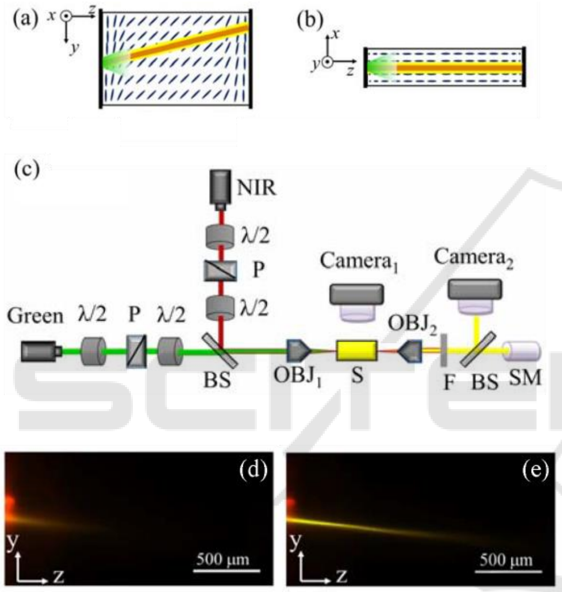

Figure 1: Geometry of the NLC sample where both the visible pump and the self-confined near-infrared beams are indicated: (a) top and (b) side view of the sample. The blue ellipses represent the NLC molecules. (c) Experimental set-up. P: polarizer, S: sample, OBJ: microscope objective, BS: beam splitter, SM: spectrometer. Acquired images of the emitted radiation without (d) and with (e) a 6mW nematicon. The near-infrared radiation has been filtered out.

Due to their unique optical and mechanical properties, NLC can provide various kinds of nonlinear behaviors, including random laser emission when samples are doped with a fluorescent dye (Ferjani, 2006). In fact, the strong scattering provided by thermal molecular oscillations combined with dye fluorescence can provide spontaneous and eventually stimulated emission when properly pumped, allowing amplification and random lasing (Bolis, 2016; Perumbilavil, 2016; Perumbilavil, 2018-1; Perumbilavil, 2018-2).

Moreover, this class of materials demonstrated to be highly versatile, since the emission can be easily controlled by exploiting temperature variations or applied voltages (Wiersma, 2001; Lee, 2011), as the properties depend on molecular distribution.

3 EXPERIMENTAL RESULTS

The set-up employed for the experiments is sketched in Fig. 1. The sample, sketched in fig.1(a)-(b) - top and side view, respectively - is a 100µm thick cell whose top and bottom surfaces have been rubbed in order to obtain a uniform molecular reorientation at 45° with respect to the z axis, thus maximizing the reorientational response. The sample is filled with a mixture of E7 ($n_{\parallel}$=1.71 $n_{\perp}$=1.52 the refractive indices at λ=1064nm for extraordinary and ordinary polarization, respectively) doped with 3%wt of Pyrromethene 597 dye. As visible in fig. 1(c), a near infrared cw beam at λ=1064nm (out of the absorption band of the dye) is injected into the sample with wave vector along z, a waist of about 3µm and polarization along *y* - extraordinary polarization -, and it generates a nematicon. A second beam from a pulsed source at λ=532nm (close to the absorption peak of the dye), with pulse duration of 6ns and repetition rate 20Hz, is focused with wave vector collinear with the near infrared beam, and yields random lasing.

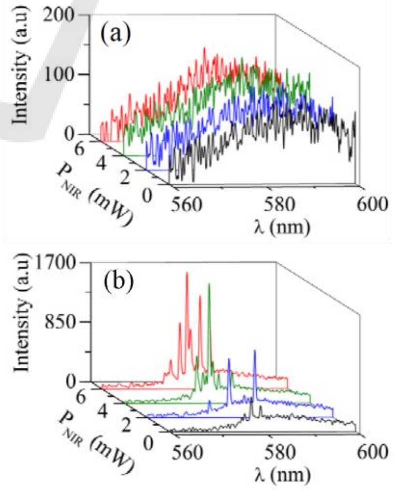

Figure 2: Acquired single shot spectra of output intensity - arbitrary units, a.u. - (a) below (E=0.40µJ) and (b) above (E=48µJ) threshold for various nematicon powers.

Preliminary measurements showed that the ordinary pump polarization (along *x*) maximizes the emitted radiation.

The evolution of the beams is observed by collecting the light scattered out of the propagation plane (yz) with a CCD camera, while the output profile is recorded by a second camera imaging the output with a microscope objective. Finally, a spectrometer detects the output spectrum with a resolution <1nm.

We excited the random laser with the green beam and varied the near infrared power to investigate the role of the self-induced waveguide on the laser light. Fig. 1(d) and (e) show the propagation of the green beam alone and when co-propagating with a near infrared beam of power P close to 6mW, respectively. As it can be seen, the emission is confined within the nematicon waveguide. Fig. 2 shows single shot spectra from the output of the sample at various pump energies and nematicon powers.

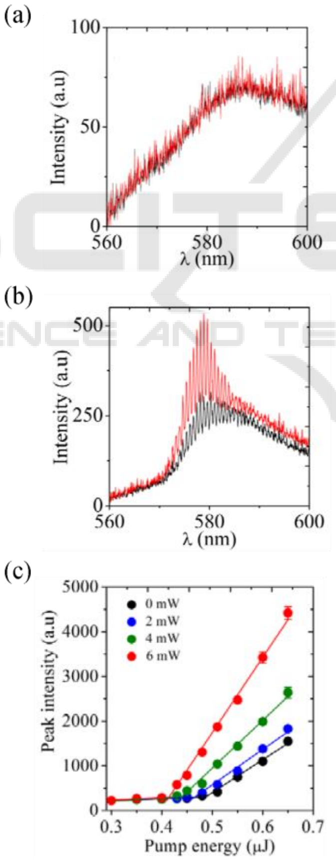

Figure 3: Acquired spectra averaged over 100 ms (a) below (E=40μJ) and (b) above (E=48μJ) threshold. (c) In-out characteristic versus pump energy at several nematicon powers.

When the visible beam is at low energy (around 0.40μJ) the nematicon has negligible effects irrespective of its power. Conversely, when the energy overcomes the threshold value (0.48μJ) the effect of the near infrared beam is to enhance the emission, with the occurrence of a number of lasing peaks of random wavelength and amplitude.

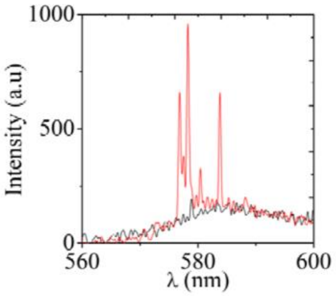

Figure 4: Single shot acquisition slightly below threshold: without nematicon (black line) the system does not lase. With a 6mW nematicon (red line) the system switches- to the random lasing regime.

To rid of the stochastic character of the lasing emission we gated the acquisition over 100ms (200 acquired spectra), obtaining the results shown in Fig. 3. The enhancement due to the nematicon is still apparent, and the in/out characteristic (Fig. 3(c)) shows the typical lasing features, with a threshold and a slope efficiency increasing with nematicon power.

As shown in Fig. 4, when the pump energy is close to (but under) the lasing threshold, the nematicon favours the transition to the lasing regime, demonstrating the possibility of switching on the laser emission by optical means. Thus, the nematicon does not only guide the emitted photons, but enhances the lasing process, improving its efficiency when increasing its near-infrared power and the corresponding guided-wave confinement.

Being an effective waveguide for the emitted photons, the nematicon also affects the transverse profile of the emission. We collected both the emitted radiation backscattered at the input and the output signal. Fig. 5 compares the two typical profiles: the speckled beam of the backscattered radiation (Fig. 5(a)) and the smoother beam after confined propagation within the nematicon (Fig. 5(b)).

Finally, the laser emission can also be re-addressed by steering the nematicon waveguide. We apply a voltage across the NLC sample, i. e. across *x*. In this way the electric torque lifts the molecules out of the propagation plane *yz* and the nematicon walk-off chenges according to the voltage

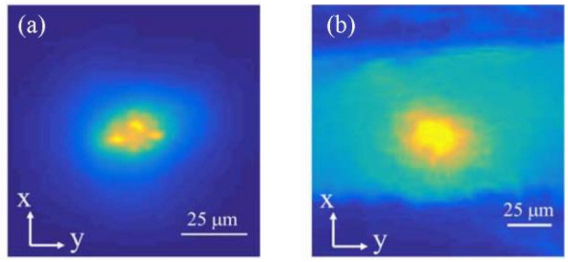

Figure 5: Acquired photographs of the laser beam profiles taken from (a) backscattered emission at the input and (b) forward emission after propagation within the nematicon.

dependent orientation of the optic axis. Remarkably, the laser emission follows the power flow of the near infrared beam. We stress that the observable quantity is the apparent walk-off, i. e., the projection of the actual walk-off on the propagation plane. Fig. 6 shows both the output laser profiles without and with applied voltage and the plot of the resulting apparent walk-off versus voltage, with a simple numerical fit.

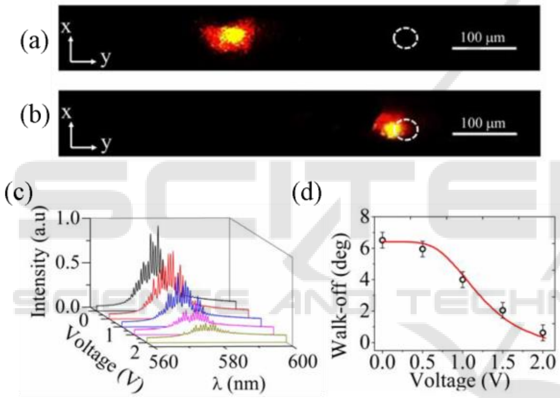

Figure 6: Voltage-driven steering of random laser. Photographs of output laser profiles for (a) V= 0V and V= 2V. (c) Acquired spectra averaged over 100ms as a function of applied voltage. (d) Measured (black circles) and calculated (red line) apparent walk-off versus applied voltage.

4 CONCLUSIONS

We presented an innovative approach to random lasing control in nematic liquid crystals, exploiting the self-induced waveguide and the corresponding molecular distribution of a spatial soliton to improve the emission properties in terms of profile and directionality, demonstrating also the possibility to electrically control the emission direction.

The effects of the nematicon are not only to guide the emitted photons, but also to enhance the conversion efficiency. The corresponding guided-wave random laser profile results smoother, and the electric control of the nematicon walk-off allows controlling the direction of the laser emitted photons.

We believe this opens new perspectives on application-oriented random lasing, introducing a low cost source with electro-optic control.

Some open questions still request deeper investigation: the actual role of the nematicon-induced refractive index profile on the random laser emission and on its bell-shape profile must be addressed; the degree of coherence of random lasing modes has to be verified, as well as their wavelength dependence; moreover, the correlation between spectral and spatial components of the random laser emission must be investigated. Future studies will thus tackle a model to account for the interaction between the two nonlinearities, addressing the modulation of the guest-host parameters (doping percentage, sample geometry, etc.) for the optimization of the laser. Other strategies for direction control could also be implemented, in either two- or three-dimensional geometries.

ACKNOWLEDGEMENT

Academy of Finland, FiDiPro grant no. 282858.

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