Near-Infrared Switching of Light-Guided Random Laser

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Abstract: We report on all-optical modulation and switching of light-guided random laser emission in optically pumped dye-doped nematic liquid crystals, whereby a continuous-wave near-infrared beam at 1.064 μm, nonresonant with the medium and collinear with the pump source, forms a reorientational spatial soliton. Such soliton-assisted cavity-less laser exhibits a beam character with directional emission and smooth transverse profile and can be either switched-ON or made more efficient by injecting a weak input. We achieve energy amplification so slow phase emission in excess of 18 dB by injecting a 6 mW near-infrared beam.

Index Terms: Spatial solitons, random lasing, nematicons, nonlinear optics, liquid crystals.

1. Introduction
Random lasers are cavity-less light sources, commonly pumped by a pulsed beam at a wavelength within the absorption spectrum of the gain medium. In such systems, the optical gain is often provided by suitable guest molecules in a host medium with scattering centers, such that the positive feedback necessary for laser oscillations can be obtained by recurrent scattering of the emitted photons [1]–[3]. Random lasers in dye-doped nematic liquid crystals (NLC), in particular, were reported in thin cells and shown to exhibit good features, despite a poor and spiky transverse profile as well as the lack of predetermined direction for the emitted light [4]–[6]. Several approaches have been explored specifically to resolve the latter issue [7], including, e.g., the use of optical fibers [8], hollow micro-channels [9], shape-tailored pump beams [10] and planar nanostructures [11], [12]. Earlier, a soliton-assisted configuration was introduced in reorientational NLC, where a beam-induced channel waveguide could effectively collect the amplified spontaneous emission in a pump-soliton configuration with orthogonal wave-vectors [13]. More recently, adopting a collinear geometry, we presented a soliton-assisted random laser, in which improved slope efficiency and narrower spectra were obtained as a result of the interaction between the non-resonant light-induced waveguide and the optical pumping [14], [15], [29]. In this Paper we show that the collinear geometry with visible pump and near-infrared (NIR) soliton launched with parallel propagation wavevectors (k_{pump} / / k_{NIR} / / k) not only solves the major issues mentioned above yielding both
smooth emission profile and well-defined beam direction, but also enables tuning the laser threshold versus pump energy, allowing the all-optical transistor operation of a random laser.

Nematic liquid crystals are positive uniaxial dielectrics in a fluid state, characterized by an optic axis $n$ along the main axes of the anisotropic molecules and a substantial optical birefringence, with refractive index eigenvalues (for electric fields parallel/orthogonal to the optic axis) typically differing by $\Delta n \geq 0.2$ in the visible/near-infrared [16], [17]. When NLC are excited by an intense light beam with extraordinary polarization (electric field linearly polarized in the principal plane $n - k_{\text{NIR}}$), the optic axis tends to reorient against the elastic intermolecular forces and the background boundary conditions, giving rise to a stable graded-index waveguide which prevents diffraction and supports self-confined light propagation [17], [18], i.e., forms a reorientational soliton or nematicon [19]–[21]. While several phenomena have been reported in conjunction with such self-guided beams [21], their use in synergy with an active light-matter response such as optical amplification and lasing brings about a wealth of yet unexplored effects.

2. Experimental Details

We adopted a planar configuration with light propagation in the bulk of a thick PM597-doped NLC sample (planar glass cell with 100 $\mu$m spacing) with optic axis pre-oriented (by suitable mechanical rubbing of the Polyimide-coated glass interfaces) at $\pi/4$ in the principal plane $yz$ with respect to the input wavevector $k_{\text{NIR}} / // z$, as sketched in Fig. 1(a) [22]. A mixture of the commercial E7 (NLC host) and 0.3 wt% Pyrromethene 597 dye (guest) filled the cell by capillarity. The refractive index values of E7 along the principal axes are $n_\parallel = 1.71$ and $n_\perp = 1.52$, respectively. An ordinary-wave 532 nm laser beam (electric field oscillating along $x$) comprising 6 ns pulses at 20 Hz and a NIR (1064 nm) continuous-wave extraordinary-wave beam were co-launched in the sample along $z$, both gently focused to a waist radius of 3 $\mu$m at the input of the cell. The NIR beam formed a nematicon, whereas the visible pump provided gain by populating the laser levels of the guest-host. The generated fluorescence, as well as the amplified spontaneous and random laser emissions, were extraordinary-waves [14], [23], [29] and, therefore, could be confined within the spatial soliton and propagate as guided-waves along the 2 mm long cell. Fluorescence and laser emission, as well as the soliton beam could be imaged versus propagation by means of the photons scattered out of the plane $yz$, and also at the sample output, using high-resolution cameras.
3. Results

Because of the optical amplification and the scattering inherent to the dynamically fluctuating NLC molecules, when pumping at high enough energy the system turned into a random laser, oscillating in the interval 570–590 nm with slope efficiency and threshold energy clearly depending on the power injected with the NIR soliton [14], [29]. The nematicon, in fact, not only improved the collection efficiency [13], but also enhanced the pump interaction with the active medium and the emitted photons in the waveguide volume [14], [15], [29]. Using a time-gated (50 ms) spectrometer (resolution 0.15 nm) we collected the generated light at the exit of the cell and acquired single-shot spectra while varying both the pump energy and the nematicon power. Fig. 1(b) displays the output peak intensity $I_{\text{max}}$ versus pump energy/pulse around the emission wavelength (582 nm), averaging over a number of single-pulse acquisitions, i.e., $I_{\text{max}} = \frac{1}{N} \sum_{k=1}^{N} I_k(\lambda)$, with $I_k(\lambda)$ the individual emission intensity spectra and –typically- $N = 200$. It is apparent that, above a nematicon-dependent pump energy threshold, the input-output characteristics exhibit lasing with significant slope efficiency. The latter increases with NIR power, whereas the threshold value correspondingly decreases, indicating that the soliton indeed enhances the overall operation of the laser.

By setting the pump at given energies/pulse, the nematicon power modifies the spectral outcome of the system, as shown in Fig. 2 for single-shot spectral realizations: various sharp lasing peaks versus NIR power appear over the pedestal of fluorescence and amplified spontaneous emission spikes, consistently with the higher index contrast associated with reorientation and soliton confinement.

The observations above led us to explore the possibility to all-optically modulate/switch this laser (active) system, at variance with transistor action in (passive) quadratically nonlinear systems [24], [25]. To this extent, we set the pump energy quite close but slightly below the threshold value for a given nematicon power and then switched the nematicon on and off. The main results are summarized in Fig. 3 for the sample case of a 6 mW nematicon and spectra $I(\lambda) = \frac{1}{N} \sum_{k=1}^{N} I_k(\lambda)$ averaged over $N = 200$ realizations. They show that -below threshold - the spatial soliton has little influence on the output spectrum, dominated by fluorescence and amplified spontaneous emission. Above threshold, conversely, more numerous lasing peaks appear, with sharper features and higher intensities. Close but still below threshold, the noisy spectral emission associated with amplified
spontaneous emission (without collinear soliton) evolves in the presence of the soliton into the characteristic random laser spectrum with well-defined peaks. This phenomenon, associated with the threshold adjustments illustrated above in Fig. 1(b), pinpoints the resulting all-optical modulation, whereby the random laser can be switched off/on when introducing a collinear mW continuous-wave beam into the actively-pumped reorientational system.

When averaging over 200 single pulses, with appropriate optics the RL emission output (above threshold) can be measured in energy units, in order to evaluate the overall conversion efficiency. With reference to the soliton-driven transitions indicated in Fig. 3(d), switching takes place near threshold for pump pulses of energy 0.43 \( \mu \text{J} \) between output emission energies of 5 and 32 pJ, i.e., with efficiency increasing by almost an order of magnitude from 0.001% to 0.008%; for the transition at pump pulses of 0.51 \( \mu \text{J} \) the RL energy switches from 0.083 to 5.20 nJ, with an (external) efficiency going from 0.017% to a 1.062%, quite a large gain increase in random lasers based on organic materials. The resulting energy amplification at this latter operation point is as high as 18 dB, with bigger figures obtainable when pumping with larger energies.

Noteworthy, the all-optical transistor effect occurs for every pump pulse despite the randomness of the phenomenon versus time, i.e., in each individual realization. Fig. 4 illustrates the transistor action as in Fig. 3, for the same pump energy and soliton power, but for single-shot acquisitions and the corresponding input-output characteristics. As expected, the intensity counts are larger
Fig. 4. Transistor operation of random laser for a given soliton power. Single-shot emission spectra without (black, lower line) and in the presence of a 6 mW NIR nematicon (red, upper line), when the pump energy per pulse is (a) 0.35 $\mu$J below threshold, (b) 0.51 $\mu$J, above threshold (c) 0.43 $\mu$J, below threshold without soliton but above threshold with the 6 mW soliton. (d) Sketch of the transitions (a)-(c) in the single-shot input-output characteristics $I_{\text{max}}$ versus pump energy (Fig. 1(b)).

Fig. 5. Single-shot photographs (sample individual realizations) of the random laser output (plane $xy$) at the cell exit (a) without co-launched NIR beam, (b) in the presence of a collinear 6 mW nematicon. The NIR was filtered out. In both cases (a-b) the pump was injected at pulse energies of 0.55 $\mu$J, above RL threshold (see inset in Fig. 1). The spots in (b) correspond to the exit position of the nematicon due to walk-off $\delta$ in the positive uniaxial medium.
and the observed features are sharper and sparser versus wavelength when compared to Fig. 3, with similar enhanced features brought about by the presence of a collinear NIR soliton. It is worth underlying that, at variance with previous reports [26], [27], the investigated all-optical switching and amplification are neither based on absorptive and photo-thermal responses nor on trans-cis light-induced isomerization, but exclusively on the non-resonant effects stemming from guided-wave confinement by way of a reorientational soliton.

Finally, in Fig. 5(a)–(b) we display the acquired photographs of emitted RL light at the cell output. In the absence of a soliton waveguide, the random laser produces a diffused pattern without specific emission direction and profile; conversely, in the presence of a collinear nematicon, the system acquires beam-like features, with a smooth nearly bell-shaped distribution at the exit of the cell and a well-defined direction corresponding to the walk-off angle $\delta$ of extraordinary wave solitons in the birefringent guest-host [28]. While further analyses of this intriguing aspect are underway, we attribute the smooth transverse profile of this nematicon-aided random laser to the propagation and intensity superposition of the various guided-wave modes excited by (non-orthogonal) RL localized spikes in the actively pumped region near the sample input, where the pump is most intense.

4. Conclusions

In conclusion, we have investigated the transistor-like operation of a random laser which, based on the interplay between near-infrared spatial solitons and optical pumping in dye-doped nematic liquid crystals, can be switched off/on or made more efficient by injecting a collinear polarized non-resonant cw near-infrared beam at low powers. Amplifications of RL emitted energy well in excess of 63 were obtained when pumping with about 0.5 $\mu$J pulses and injecting 6 mW near-infrared nematicons. This soliton-aided random laser yields directional emission along the trajectory of the nematicon, with a smooth transverse profile as compared to the most common RL systems. The demonstration of a random laser with beam characteristics and all-optical modulability definitely paves to way to more applications of these cavity-less lasers.

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References


