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EFFICIENT RANDOM LASER EFFECT IN A NEW DYE-NEMATIC LIQUID CRYSTALLINE COMPOSITE

V. BARNA[1], V. I. VLAD[2], A. PETRIS[2], I. DANCUS[2], T. BAZARU[2], E. S. BARNA[1], A. DE LUCA[3], S. FERJANI[3] and G. STRANGI[3]

[1]*Faculty of Physics, University of Bucharest, PO Box MG-11, 077125, Bucharest, Romania*
E-mail: barnavalentin@yahoo.com
[2]*Department of Lasers, National Institute for Laser, Plasma and Radiation Physics, Bucharest-Magurele, RO-077125, Romania*
[3]*LICRYL CNR-INFM and Center of Excellence CEMIF.CAL Department of Physics, University of Calabria, I-87036 Rende (CS), Italy*

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Abstract. Random lasing effects in fully disordered systems with organic and inorganic nature have been the subject of extensive studies since the beginning of the past decade. In multiple scattering materials the optical diffusion may induce a phase transition in the photon transport behavior. Beyond a critical scattering level, the system makes a transition into a strongly localized state and light transmission is therefore inhibited. This effect can be used as a photon trapping mechanism to obtain laser action in the presence of a gain medium. In this paper we present the first experimental evidence of random laser action in a dye doped nematic liquid crystal (ZLI-2659) with negative dielectric anisotropy. This material exhibits long-range dielectric tensor fluctuations, favoring the multiple scattering. 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. The laser emission has several interesting features - linewidth of the laser peaks is extremely narrow banded (approximately 0.5nm FWHM), very low threshold (30 μJ/mm^2), high efficiency (~ 20%). 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 nematic liquid crystalline system. We analyze the main physical characteristics of these relatively novel phenomena, by also including here the case of various confinement geometries for the active medium. These scientific aspects overlook features of great interest characteristic of laser physics and material science.

Keywords: random laser, nematic liquid crystal, fluorescence, scattering, light localization.

1. INTRODUCTION

The diffusion together with interesting transport phenomena of light waves in complex dielectric structures have conducted to a wide series of experimental and theoretical investigations in the field, revealing one of the most challenging, yet exciting, scientific area of the past decade. The propagation of electromagnetic waves in periodically structured dielectric systems and the linear and nonlinear optical phenomena in completely disordered systems doped with gain media represent two opposite sides of this promising scientific domain. The literature shows that much has been done in these extreme areas, but the vast intermediate world formed by the so called partially ordered systems still remains almost unexplored. The study of fluorescence phenomena, light amplification and laser emission properties in ordered and periodic systems acknowledged an amazing boost in the last couple of years, even because of the remarkable development of experimental techniques allowing for the design of photonic crystal structures as low as nanoscale ranges [1]. Unexpectedly, active random media frequently confirmed to be the appropriate candidates for attaining diffusive laser action, mainly based on the resonant feedback mechanisms in multiple scattering, thus eliminating the necessity for an external cavity - as in the case of regular lasers. Light localization and interference effects which survive the multiple scattering events have been invoked to explain the random lasing observed in many exotic and complex systems [2-9]. The optical scattering phenomenon inside a random medium is capable of inducing a phase transition in the photons transport behaviour. For weak scattering, the propagation of light can be explained by a normal diffusion process. Following an increase in the amount of scattering, recurrent light scattering events arise. The interference phenomena between the counter propagating waves in a partially or totally disordered structure give rise to the enhanced backscattering, also called weak localization [10,11]. When the scattering level is increased beyond a threshold critical value, the system suffers a transition into a localized state. Light propagation is now inhibited because of the interference in multiple scattering. Since the Anderson localization is fully based on the interference effect [12-14], and interference is a common property of all wave phenomena, it is somehow normal to extend the electron localization to photon localization while in disordered dielectric media. Nevertheless, apart from the notable similarities, we encounter several differences between electron transport and the photon transport in such media. For instance, the number of electrons is permanently conserved, while the number of photons is not in an amplifying (or absorbing) random medium.

A very interesting phenomenon that would never occur in an electronic system is the laser action in a disordered gain medium [15-18]. In the case of strong scattering and gain, recurrent scattering events can provide coherent feedback and results in lasing [19-21]. In this case, the scattering mean free path becomes comparable to the light wavelength. Light may return to a scatterer from which it was scattered before, and thereby forming closed loop paths. If the amplification along such a loop path exceeds the loss in the system, laser oscillation could occur and the loop behaves as a laser resonator. The prerequisite of the phase shift along the loop equal to a multiple of 2π establishes the oscillation frequencies. Nevertheless, the light may come back to its initial position throughout many different paths. All of the backscattered light waves interfere and determine 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 pioneering work of Letokhov and co-workers in 1968 [22], which predicted that the combination of multiple scattering and light amplification would lead to a form of laser action, lasing in disordered media has been a subject of intense theoretical and experimental studies [23-30]. Random lasers have been developed in various material systems, ranging from semiconductor nanoparticles, ceramic powder to polymers, organic materials and biological tissues. Their low fabrication cost, sample specific lasing frequency, small size, flexible shape, and substrate compatibility could lead to plenty of potential applications. There are two possible types of feedback for a random laser: one is intensity or energy feedback, the other is field or amplitude feedback. The field feedback is phase sensitive (i.e. coherent), and therefore frequency dependent (i.e. resonant). The intensity feedback is phase insensitive (i.e. incoherent) and frequency independent (i.e. non-resonant). Based on these different feedback mechanisms, random lasers are split into two main classes: (1) random laser with incoherent and non-resonant feedback, (2) random laser with coherent and resonant feedback. Lasing with nonresonant feedback arises in the diffusive regime. In a disordered medium, light is scattered and undergoes a random walk before leaving the system. In the presence of gain, a photon may induce the stimulated emission of a second photon. When the gain length is equal to the average length of light path in the bulk, the probability that a photon generates a second photon before leaving the gain medium approaches one. Thus the photon density is expected to increase. From the theoretical point of view, the solution to the diffusion equation, including optical gain, diverges. This phenomenon is similar to neutron scattering in combinations of nuclear fission. A random laser with coherent feedback can be considered a randomly distributed feedback laser. The feedback in this case is provided by disorder induced scattering.

The first experimental evidences of this phenomenon are dated 1993-1994, while later on outstanding research in this field was carried out by A. Lagendijk [24,28,30-33], D. Wiersma [4,8,24,34-38] and H. Cao [13,14,16-18,39-41].

In particular, random lasers exhibit threshold behaviour. When the gain overcomes the losses the system goes above threshold. The losses are proportional to the total sample surface and the gain to its volume, so the threshold criterion can be expressed in terms of a critical volume V_{cr} above which the system is lasing [22].

There are many 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 [22-41]. The random laser not only becomes a new member of laser family, but also provides a new path to study localization phenomena [42-44]. Because random lasing modes come from the eigenstates of disordered systems [42], these fascinating systems open a special door to study the interplay between localization and amplification in various media.

Coherent backscattering experiments revealed weak localization of light even in tensorial systems characterized by high optical anisotropy, such as nematic liquid crystals [45]. It is demonstrated how the recurrent multiple scattering events exactly back enhance the scattered intensity, giving rise to an anisotropic backscattering cone [46].

Nematic liquid crystals (NLC) 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 [47].

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 is the main effect responsible of the recurrent multiple scattering events as a light wave is propagating through the NLC medium. Furthermore, the scattering of visible light by nematic liquid crystalline turbid materials is higher, by a factor of the order of 10^6, than the scattering by conventional isotropic fluids [47]. The fluctuations of $\varepsilon_{\alpha\beta}$ come from two different sources: (i) fluctuations in $\varepsilon_{\perp}$ and $\varepsilon_{//}$ due to small, local, changes in the density, temperature, etc.; (ii) fluctuations in the orientation of n – this being the dominant effect, which is specific of nematic liquid crystals.

Recently, the first experimental observations were presented for random laser action in a partially ordered and highly anisotropic nematic liquid crystals doped with fluorescent guest molecules [48-54]. The study of laser emission in such a system emphasized the peculiar behaviour of diffusive laser action, randomness of laser emission was observed in time, space and frequency.

In this work, a new dye-nematic liquid crystal composite is investigated as random lasing system. Important features of this composite are the negative dielectric anisotropy and the long range dielectric tensor fluctuations, which can contribute to optical amplification. Our experiments confirm that the laser action occurs indeed at a low threshold of approximately 30 μJ/mm^2. This laser system has also very interesting characteristics: emission efficiency of approximately 20% in both directions (forward and backward), wavelength in yellow region of the spectrum, narrow spectral line, good spatial profile and nanosecond pulse duration. Moreover, the new dye-LC laser is compatible with nowadays miniaturization technologies.

2. EXPERIMENTS WITH THE NEW DYE-NEMATIC LIQUID CRYSTAL COMPOSITE

We studied the random lasing properties in negative dielectric anisotropy nematic liquid crystals doped with gain molecules. Our proposed systems consist of ZLI-2659 nematic liquid crystal provided by Merck that was doped with 0.5 wt% of Pyrromethene 597 dye (Exciton). The dye molecules dissolved in the NLC proved to be completely miscible, as evidenced by the almost complete absence of micro-droplets of dye embedded in the nematic phase.

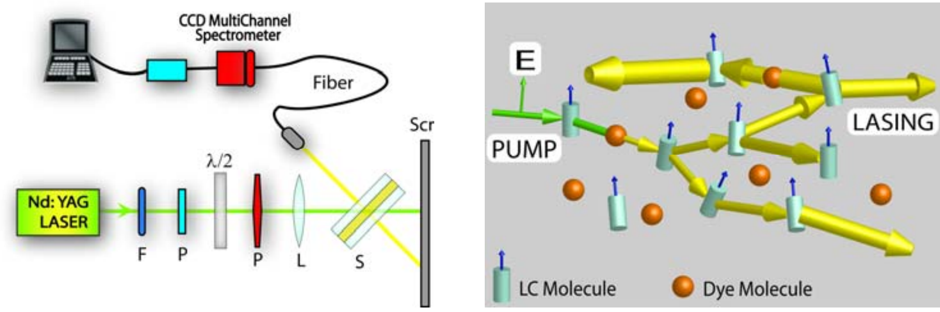

Fig. 1. – Schematic diagram of the experimental set-up (left): F is a filter, P – polarizers, λ/2 – wave plate, L – lens, S – sample, Scr – observation screen. Recurrent multiple light scattering in dye doped nematics provides the feedback for laser oscillations (right).

The samples were optically pumped by means of the second harmonic of a Nd:YAG laser (EKSPLA NL 131/SH/TH) (532 nm, pulse duration 3 ns).

The mixture of nematic and dye was enclosed in various confinement geometries, such as a sandwich cell (Fig. 2a). In addition, boundary-free systems were designed, where the mixture was freely suspended (Fig. 2b).

The first system is constituted by two glass-ITO plates separated by Mylar spacers having thickness of 75 μm in the case of the classic parallel walls sandwich cell type. The inner surfaces of the plates were covered with rubbed SiO_2 layers in order to induce a homogeneous alignment of the NLC molecules at the interface. The cell was filled by capillarity with the flow direction along the rubbing direction. Upon observing the sample under a polarized microscope, it showed an average planar alignment with the optical axis situated in the plane of the cell, parallel to the rubbing direction.

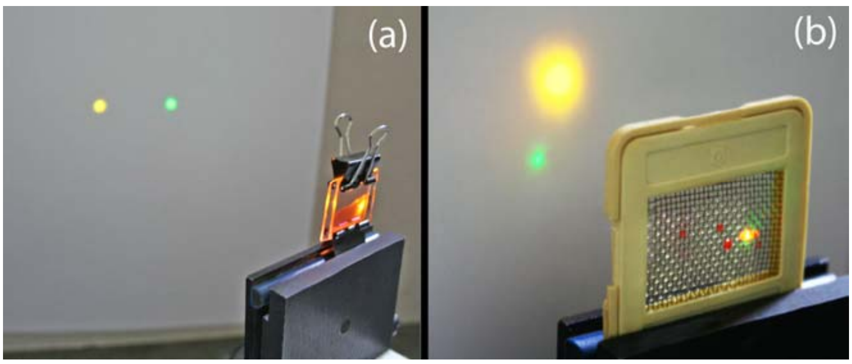

Fig. 2 – Different random lasing confinement geometries: a) sandwich cell; b) freely suspended film.

For optically exciting our system the 532 nm pump beam was focused onto the sample by means of a spherical lens (f = 500 mm), yielding a beam waist of about 80 μm at the focus position. The experimental set-up (Fig.1) also included a series of optical elements (optical filters, wave plates, polarizers, lenses) in order to better select the input energy value as well as different states of polarization for the pump beam.

3. LASER EFFECT CHARACTERIZATION AND DISCUSSION

A multichannel CCD spectrometer (Ocean Optics USB4000HR) with a high spectral resolution (0.5 nm) and with a fiber termination was used to capture the emission spectra.

At low pump energy density (20 $\mu J/mm^2$), the emission spectra depicts the typical spontaneous emission curve of dye molecules for PM597, indicating that the nematic liquid crystal does not considerably modify the fluorescence spectrum (Fig. 3a). Upon increasing the pump energy density above a given threshold value (about 30-40 $\mu J/mm^2$), discrete sharp peaks emerge from the residual fluorescence spectrum (Fig. 3a). The line width of these sharp peaks was measured to be less than 0.5nm. When the incident pump energy density exceeds the threshold value, the peak intensity increases much more rapidly with the pump power and more sharp peaks appear, because now the net gain experienced by these modes in reduced mode competition situation, becomes positive since the strong lasing mode do not saturate the gain for the others to a level which prevents them from lasing (Fig. 3b).

The narrow peaks also show a decrease in the spectrum width of the emitted light triggered by the increase of the pump energy. 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 ($FWHM_{below}$) and the FWHM of the emission spectrum of the random laser above threshold ($FWHM_{above}$):

$$\mathrm{NF} \equiv \frac{\mathrm{FWHM}_{\mathrm{below}}}{\mathrm{FWHM}_{\mathrm{above}}}. \tag{1}$$

In the case of Fig. 3a, the narrowing factor (NF) is about 40.

The speckle-like pattern of the emission spot was also imaged simultaneously on a screen (Fig. 2). In addition, it was calculated the free spectral range by assuming a cavity effect in our sandwich cell and it was found to be absolutely incompatible with the wavelength region of the lasing modes and with the spectral spacing of the modes.

A small amount from the mixture was spread on top of a rectangular network (l = 1 mm, depth= 0.5 mm) by means of a spatula. In this way we tailored a simple system where we have several parallelepiped “holes” filled with a thin film of active material, as depicted in Fig. 2c. Other geometries (disk, rectangular) of various sizes could also be used for moulding the shape of thin films.

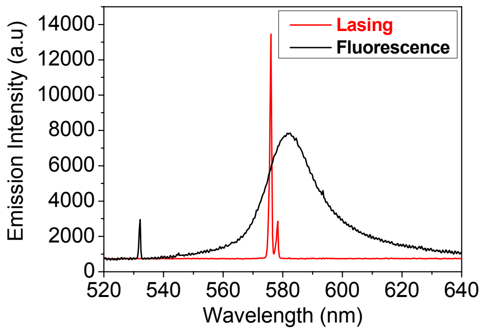

Fig. 3 – Fluorescence and random lasing spectra in the case of a sandwich cell upon increasing the pump energy. Discrete sharp peaks emerge from the residual spontaneous emission for pump energy density above 30-40 μJ/mm^2 (a). At higher pump values multiple lasing peaks are generated (b). For clarity, the residual fluorescence signal was ten times magnified.

Another experiment addressed the measuring of the energy threshold for the laser emission in our dye doped nematic liquid crystal samples. The threshold was calculated to be around 30-40 μJ/mm^2 (yet a more stable emission is obtained for pump energy values above 100 μJ/mm^2). The energy output in Fig. 4 is obtained in the forward direction (the lasing beam is perpendicular to the boundary glass plates). One important issue is that laser emission is also achieved in the completely back direction (as depicted in Fig. 1). Therefore, the total output energy (corresponding to a given input energy) should be seen as twice the value on the ordinate axis in Fig. 4b. Therefore, the efficiency of our composite system proves to be relatively high, around 20%.

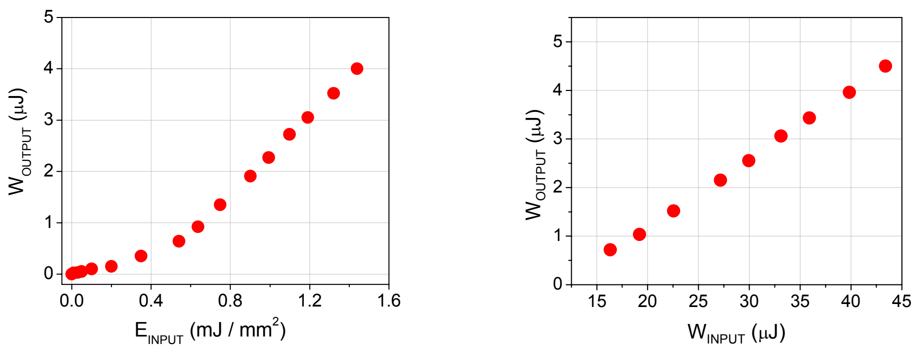

Fig.4 – a) Emission energy vs. pump energy density for the classic sandwich cell;
b) output energy vs. input energy.

This behavior has many similarities to the stimulated scattering processes [55-57]. An analytical treatment of the random laser in dye-nematic liquid crystal composites, which may explain the threshold and the high efficiency of this type of laser, is in progress.

The input polarization influence upon the lasing properties, as well as on the reflected/transmitted 532nm beam was also experimentally observed.

The input beam was linearly polarized, while we had two possibilities of orienting the main axis of the molecular director for the nematic liquid crystal cell (parallel and perpendicular to the input polarization direction). The interesting experimental results are summarized in Table 1.

Table 1

Polarization properties of the output pump and random laser beams

Input Polarization	Nematic Director Orientation	Output Polarization (Green Laser Light)	Output Polarization (Yellow Lasing Light)
↔	↔	Transmitted ↕	Transmitted ↔
↔	↔	Reflected ↔	Reflected ⤢
↔	↕	Transmitted ↔	Transmitted ↕
↔	↕	Reflected ↔	Reflected ↕

When the nematic director orientation is parallel to the polarization of the input green pump beam, the polarization of the transmitted green light is rotated with 90° and the polarization of the yellow laser light generated forward remains parallel to the pump polarization. The polarization behavior is different when the nematic director orientation is orthogonal to the input pump polarization. In this case, the polarization of both green beams, transmitted and reflected, remains unchanged, while the polarization of both yellow beams generated forward and backward, becomes orthogonal to the green pump polarization.

Another experimental investigation dealt with analyzing the far field spatial distribution of the emitted light. Lasing emission was imaged by means of a very sensitive CCD camera (640 × 480 pixels, Newport). Random lasing emission patterns from our PM 597 doped nematic liquid crystal sample confined in sandwich cell geometry can be observed in Fig. 5.

The intensity profile is fluctuating from one frame to another, being characterized by bright peaked emission regions that are spatially overlapped. We can observe, for eight successive pump pulses (Fig. 5), that the beam profile and its maximum intensity are varying, which can be related to the randomness of the laser action.

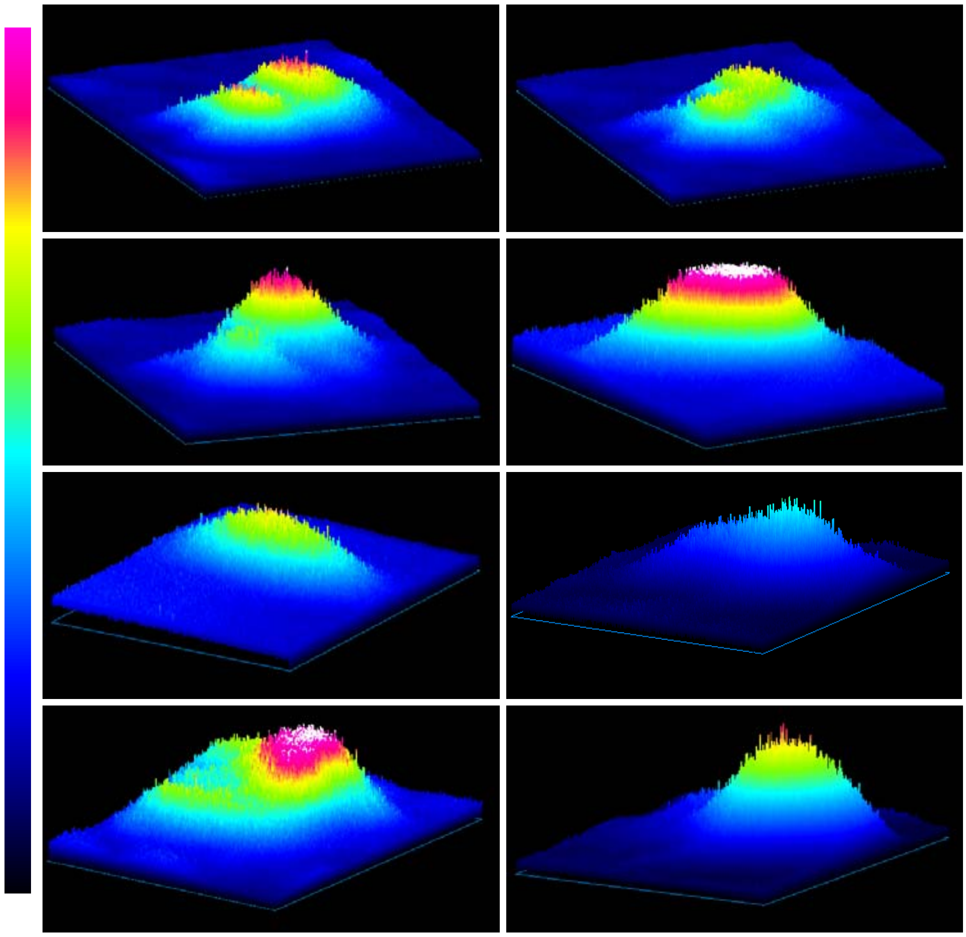

Fig. 5 – Spatial far field distribution of the intensities profile in a sandwiched ZLI 2659 doped sample for eight consecutive pump beam pulses illustrates a blinking behaviour in space and time.

4. CONCLUSIONS

We observed and characterized the process of random laser action in a new dye-nematic liquid crystal composite. We considered various boundary conditions (i.e. confinement geometry, bulk volume and surface treatment) and external field factors (i.e. pumping conditions). The underlying mechanism 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. Unlike distributed feedback mirror-less laser, this system can be considered as a cavity-less microlaser where the disorder unexpectedly plays the most important role, behaving as randomly distributed feedback laser. We obtained that the random lasing emission spectrum peaks are extremely narrow banded (about 0.5 nm), occurring at a relatively low energy density threshold of about 30 $\mu J/mm^2$.Various confining geometries can be tailored – classical sandwich cells and even freely suspended films – which own the physical characteristics of random lasers. A detailed analysis for the far field modal profile of the emission intensity illustrates a series of spatially overlapped emission peaks, generating a changing speckled pattern which fluctuates in time, position and intensity. Interesting polarization-depending features at both optical wavelengths (532 nm and 575 nm) were also observed in our systems. Further studies will be needed in order to gain full understanding of the diffusive laser action in these dye-nematic liquid crystal composites. These scientific results trace a very promising route for fundamental studies and applications in condensed matter physics, opto-electronics and photonics.

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