Remote control of liquid crystal elastomer random laser

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***Abstract*—We demonstrate random lasing in a composite system fabricated from elastomeric liquid crystals and TiO_2 nanoparticles. By moving this random laser near the focal plane of the pump beam, we achieve control of its emission properties.**

Keywords—random lasing; liquid crystal; tunable random laser

I. Introduction

Random lasers are unique optical sources that resemble the emission properties of a conventional laser. In random lasers, the optical feedback is provided by the path length enhancement of photons due to multiple scattering. Upon excitation, the inverted medium and random scattering centers can realize laser-like emission. Ease of fabrication raises their demand as fascinating optical sources for various applications [1, 2]. In the recent past, the random lasing phenomenon has been demonstrated in several systems like dielectric nanopowders, dye-doped scatterers, biological tissues, etc. [1-6]. However, for practical application, a major drawback is the inherent fluctuations in the emission arising from the structural disorder of a random laser. For any practical implementation, it is necessary to control these fluctuations. In this regard, attempts to tackle this issue can be broadly classified into two types, either involving a modulation of the scattering mean free path using resonant scatterers or by controlling the gain properties of the amplifying medium. The concept behind using the resonant scatterers is to minimize the scattering mean free path at its resonant frequencies and thus achieving wavelength sensitive scattering [7-10]. In another case, the spatial distribution of pump profile was modified to excite a pre-selected random lasing mode [11, 12].

Direct control of the lasing threshold and output intensity has also been attempted in many random lasers. Among them, liquid crystal based random lasers offer distinctive features since their scattering properties can be changed by the orientation order and phase transition temperature, etc. For example, temperature-tunable random lasing has been demonstrated using sintered glass and 7CB liquid crystals wherein the temperature-dependent phase transition was responsible for modifying the scattering strength [13]. The sharp phase transition allows to switch the system above and below the lasing threshold by a small variation in its temperature. The orientation order of liquid crystal was utilized in dye-doped nematic liquid crystals wherein the polarization of pump light affects the output lasing intensity [14]. However, to our knowledge, the physical motion of random laser has never employed to control or trigger the random lasing emission so far.

In this paper, we demonstrate random lasing in a composite system consisting of liquid crystal elastomer (LCE) and LCE with added TiO_2 nanoparticles in the presence of an amplifying medium. This composite LCE random laser can move in and out of the focal plane of a 25 ps Nd:YAG laser. When the LCE random laser is away from the focal plane of the picosecond laser, a typical profile of gain medium appears in the emission. However, as this random laser moves towards the focal plane, narrow bandwidth emission with a definite threshold is observed. Thus, the movement of this composite random laser is exploited as a mean to control its emission.

II. Sample Fabrication

The composite LCE random laser has two parts which are attached to each other to form a single structure. The LCE random laser was fabricated between a cell made of two glass coverslips which were spin-coated with a thin layer of PVA and PI-1211. The PVA coated coverslip was scratched on the surface using a cotton cloth. The liquid crystal (LC) solution (79 mol % of the LC monomer, 20 mol % of the crosslinker, and 1 mol % of the photoinitiator) containing 4 x 10^{-3} M of DCM dye was infiltrated into the cell from one side [15]. From the other side, we infiltrated the same LC solution having TiO_2 nanoparticles to provide additional scattering.

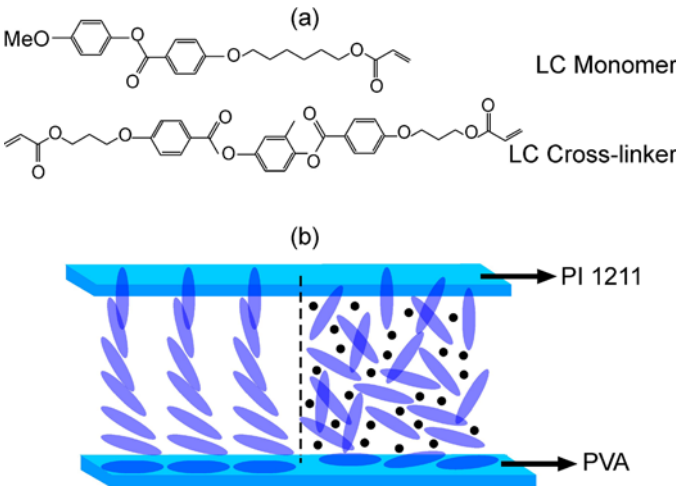

Fig. 1. (a) Chemical structure of the monomer and cross-linker molecule used to fabricate the composite LCE random laser. (b) The schematic diagram of molecular alignment. In the left half region, the liquid crystals align in the splayed fashion and in the right half, the alignment is random.

The chemical structure of liquid crystal mixture used to fabricate the LCE random laser is depicted in Figure 1(a). Once the mixture was infiltrated in the cell, the liquid crystal

starts to align as illustrated in Figure 1(b). In the vicinity of PI 1211 coating, the LC molecules settle in the vertical alignment and near the PVA coated region the LC molecules align along the rubbing direction. Overall, in the first half region, the LC molecules align in the splayed fashion. From the opposite side, LC mixture with the embedded TiO_2 nanoparticles is infiltrated. In this part, the presence of the nanoparticles prevents the long-range alignment of LC, which remains disordered. The temperature of the mixture in the cell was maintained at 80 ^oC for 1 hour and then the sample was exposed to the UV radiation for 30 minutes resulting in a single 200 µm thick composite LCE random laser. The transport mean free path (l_t) in the first half of LCE random laser was measured by the width of the coherent backscattering cone, corresponding to a l_t of ~735 µm. The critical thickness to excite random lasing modes in the LCE sample is estimated to be around 310 µm assuming a total inversion of 4 x 10^{-3} M dye molecules. Therefore, in the first half, the random lasing modes cannot be excited [16]. However, in the second half region, the LC mixture also contains TiO_2 nanoparticles to support random lasing. The average diameter and volume fraction of nanoparticle were around 280 nm and 0.005, respectively. These additional scatterers reduce the scattering mean free path to around 65 µm. Hence, this part of composite LCE laser is capable of supporting the random lasing modes.

III. Experimental Setup

Following fabrication, we cut the resulting composite film into several strips of about 2 mm width and 1.5 cm length. The experimental setup to move and simultaneously excite the random laser emission is schematically shown in Figure 2 (a).

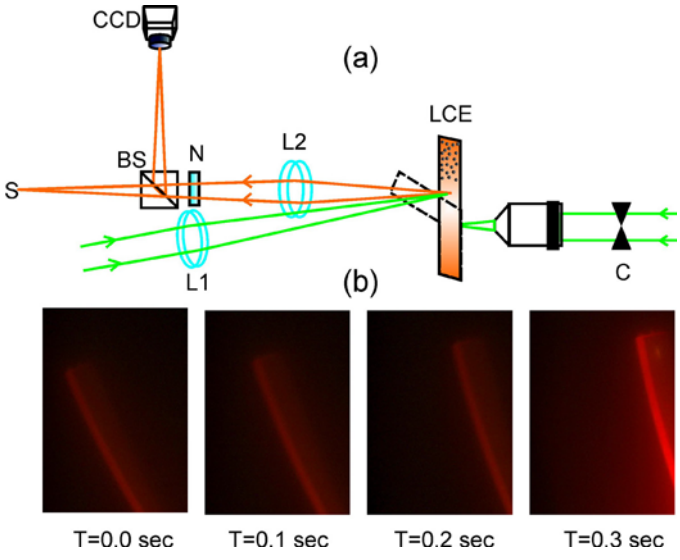

Fig. 2. (a) Experimental setup to move and excite the random lasing in a composite LCE stripe. L1, L2: lens, N: notch filter, BS: beam splitter, C: chopper, S: spectrometer. (b) Image of moving LCE stripe at t = 0.0, 0.1, 0.2 and 0.3 seconds.

The light-driven movement in the elastomer strip was induced using a CW laser (532 nm, 7 W/mm^2) chopped at 1.5 Hz using a chopper C. The laser was loosely focused on the stripe using a 5X objective lens and the motion was monitored using a CCD camera. The random lasing modes in the top part of LCE stripe are excited by another 25 ps Nd:YAG laser (λ = 532.8 nm, E_p = 13 µJ) operating at 10 Hz. Using a 50 mm biconvex lens L_1, the ps laser pulses were focused to a focal spot of 100 µm. We then parked the LCE composite random laser near the focal plane of ps laser. The shot-to-shot emission spectrum was collected by a 150 mm biconvex lens L_2 placed at an angle of 20 degree from the excitation beam and the spectra were analyzed using a 0.5 m spectrometer S having a spectral resolution of 0.1 nm.

IV. Results and Discussion

The chopped CW laser induces a light driven movement in the LCE stripe. When the LCE stripe is exposed to the CW laser, the elastomer expands in the perpendicular direction of the local LC alignment, resulting in an overall bending of the LCE stripe. Figure 2(b) shows the snapshot of a moving LCE random laser at an interval of 0.1 seconds. At t = 0 s, the LCE stripe is in a relaxed position. As the LCE stripe experience the optical power it starts to bend away from the objective lens. The maximum movement of about 3 mm is observed around t = 0.3 s. Next, when the chopper blade obstructs the CW laser, the LCE stripe returns to its initial relaxed position. The movement of composite LCE random laser follows the chopper period up to ~ 100 Hz.

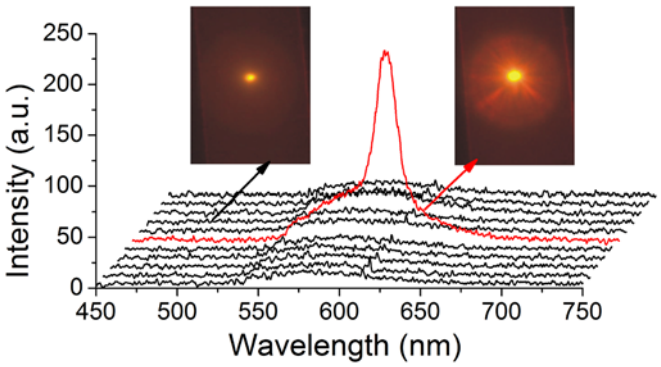

Fig. 3. Emission spectra from LCE random laser. The black curves correspond to the emission spectrum when the LCE stripe is away from the focal plane of ps laser. The red curve shows the emission spectrum close to the focal plane. The inset depicts the images of the emission spot corresponding to the black and red spectrum.

Figure 3 shows the single shot emission spectra from a moving LCE random laser. The inset depicts the simultaneously captured image of the lasing spot. The black curve corresponds to the emission when the top part of LCE stripe is out of the focal plane of the ps laser. Typical emission spectra of the DCM dye are observed, the intensity of the lasing spot is mild. At these positions, the stripe resembles the behavior of a typical random laser below threshold. As the LCE stripe approaches the focal plane of the ps laser, the system experiences a significant inversion and the output emission intensity shoots up significantly as shown by the red curve. Furthermore, the bandwidth of the emission collapses to nearly 10 nm. As shown in the inset, at this location the LCE stripe exhibits a bright lasing spot with many streaks. Occasionally, close to the focal spot, we also found ultra-narrow band emission having resolution-limited modes. These

observations confirm the excitation of random lasing modes in the top part of LCE stripe wherein the added scattering provided by the TiO_2 nanoparticle plays a crucial role and assists lasing.

V. CONCLUSION

In conclusion, we have demonstrated random lasing in liquid crystal elastomer wherein the lasing is being switched on/off by the induced bending movements of the material. The optical excitation triggers a controllable and reversible motion of the elastomeric material. Driving the elastomer laser drives the gain medium in/out from the critical threshold region and therefore allows to control its emission.

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