Magnetically controllable random laser in ferromagnetic nematic liquid crystals

Takuya Naruta,[1] Takuya Akita,[1] Yoshiaki Uchida,[1,][*] [iD] Darja Lisjak,[2] [iD] Alenka Mertelj,[2] [iD] and Norikazu Nishiyama[1]

[1]*Graduate School of Engineering Science, Osaka University, Osaka, Japan*
[2]*Jožef Stefan Institute, Ljubljana, Slovenia*
[*]*yuchida@cheng.es.osaka-u.ac.jp*

Abstract: This paper first reports random laser action in dye-doped ferromagnetic nematic liquid crystals, which act as a randomly distributed cavity. The random laser intensity of the ferromagnetic nematic liquid crystals can be controlled by a weak magnetic field (~1 mT). Moreover, the magnetic switching of random laser is attributed to the direction and polarization dependent emission of light in the ferromagnetic nematic liquid crystals in an external magnetic field.

1. Introduction

Random laser action occurs from multiple scattering of light in disordered systems [1], in which light propagating in a closed loop path in an active medium is amplified [2,3]. Since random lasers do not require any external optical resonators, they have raised attention as easy-to-make, low-cost and speckle-free lasers advantageous for applications in imaging [4], photonics [5] and biomedicine [6,7]. Meanwhile, in general, the light paths of random lasers are so chaotic that their mode control is difficult [8]. Therefore, several methods have been developed to solve the control problems of random lasers for future applications since the random laser action was first reported by Letokhov *et al.* [9]; in particular, the random laser action in nematic liquid crystals (NLCs) has been vigorously investigated [10], whose anisotropic scattering behavior is different from the scattering behavior of colloidal suspensions as follows. Because the multiple scattering of light is driven by nematic director fluctuations [10,11], the random laser emission from NLCs is bidirectional [11] and polarized [10,12,13], unlike the omnidirectional behaviors in most of random laser resonators [14]. Because the laser action is correlated with the orientational direction of liquid crystals [15], it could respond to the molecular reorientation by a variety of external stimuli. In fact, the wavelength, intensity and polarization of the random laser in NLCs can be controlled by electrical [16–19], thermal [13,20] and magnetic [19] stimuli, and the alignment of LC cells [15,21]. In particular, the optical switching owing to the molecular reorientation in a magnetic field [22] has some advantages in remote operability [8]. However, to reorient the nematic director using a weak magnetic field, high magnetic anisotropy $\Delta\chi$ of the LC molecule is needed [23,24], which is defined as the subtraction of the molar magnetic susceptibility component ($\chi_\perp$) perpendicular to the molecular long axis from that ($\chi_\parallel$) parallel to the axis [25].

$$\Delta\chi = x_\parallel - x_\perp \tag{1}$$

The magnetic anisotropy of liquid crystalline molecules is not sufficiently high not only for diamagnetic liquid crystalline molecules ($10^{-5} \sim 10^{-4}$ emu/mol), but also for paramagnetic liquid crystalline molecules containing organic radical moieties ($10^{-5} \sim 10^{-6}$ emu/mol) or paramagnetic metal ions ($10^{-6} \sim 10^{-2}$ emu/mol) [26].

Recently, ferromagnetic nematic liquid crystals (FNLCs) have been reported [27]. In the FNLCs, ferromagnetic nanoplatelets (MNPs) with a spontaneous magnetization (***M***) normal to the

plate surface are well dispersed in NLCs, and the nanoplatelets are oriented in the same direction owing to the interaction between the plate surface and the nematic director (***n***). Therefore, ***n*** of FNLCs can be reoriented by extremely weak magnetic fields owing to the coupling between ***M*** and ***n***, and moreover, the response rate depends linearly on the strength of the field [28]. In addition, MNPs dispersed in NLCs did not affect the light propagation in FNLCs seriously. Therefore, the laser action of whispering gallery mode (WGM) occurs in FNLCs, and the wavelength of WGM laser from a droplet of dye-doped FNLC can be tuned by a very weak magnetic field (~10 mT) [29]. It implies that the random laser action in FNLCs could be controlled by much weaker magnetic fields than that in the other NLCs.

Here, we first report the observation of the random laser action in FNLCs, and demonstrate the magnetic-field-induced change of the intensity of random laser emission using homeotropic and planar alignment cells; it can be applied for both magnetically controlled normally open (NO) and normally closed (NC) switching modes of the random laser action. Moreover, we discuss the magnetic switching of the random laser in terms of the polarization of light in dye-doped FNLCs in an external magnetic field.

2. Experimental

A dye-doped NLCs (DNLCs) was prepared by doping a well-known NLC mixture, E7 [30], which shows the following phase sequence *Cr. – 10°C – NLC – 58°C – Iso*, with 0.3 wt% of Pyrromethene 597 as a laser dye. Sc-substituted barium hexaferrite nanoparticles were synthesized hydrothermally and dispersed in 1-butanol using dodecylbenzenesulfonic acid as previously in Ref. [31]. A suspension of 0.6 wt% MNPs in 1-butanol was mixed with DNLCs, where the concentration of MNPs was 0.1 wt%. The mixture was kept at 70°C, where E7 shows the isotropic phase, for 24 h to evaporate 1-butanol completely to give a dye-doped ferromagnetic NLCs (DFNLCs).

The random laser experiments were carried out in LC cells consisting of two parallel glass plates. We prepared two kinds of cells: planar alignment cells and homeotropic alignment cells. The planar alignment cells were prepared as follows. The glass plates were spin-coated with AL1254 (JSR), which induces planar alignment, at 3000 rpm for 30 s. The substrates were prebaked at 60°C for 10 min, to evaporate solvents, and then baked at 180°C for 180 min to complete imidization. They were rubbed in an antiparallel configuration, and were assembled into cells with 145 μm gap using two 145 μm thick mylar films as spacers and glue. Similarly, the homeotropic alignment cells were prepared. The glass plates were spin-coated with JALS204 (JSR), which induces homeotropic alignment, at 3000 rpm for 30 s. The substrates were prebaked at 60 °C for 10 min, to evaporate solvents, and then baked at 180 °C for 180 min to complete imidization. The substrates were assembled into cells with 145 μm gap using two 145 μm mylar films as spacers and glue. These cells were filled with DNLCs and DFNLCs to give four samples: DNLCs in a planar alignment cell (pDNLC) and in a homeotropic alignment cell (vDNLC), and DFNLCs in a planar alignment cell (pDFNLC) and in a homeotropic alignment cell (vDFNLC). The alignments of these samples were checked using a polarized microscope.

Our samples were optically pumped with 6-9 ns pulses at a wavelength of 532 nm with a repetition rate of 10 Hz, produced by the second-harmonic light of a Q-switched Nd:YAG laser (Quanta-Ray INDI-40-10-HG, Spectra-Physics). The pump beam was focused with a lens with the focal length of 35 mm. The configuration of the optical elements for controlling the pump energy and polarization is shown in Fig. 1, in which P is polarizer and $\lambda/4$ is quarter-wave plate. For detection, a fiber spectrometer (HR2000+, Ocean Optics) with a spectral resolution of 0.23 nm was used.
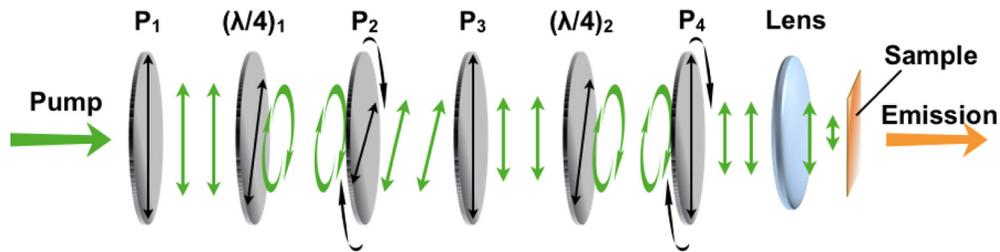
Fig. 1. Schematic of the optical setup for the laser characterization. Four plates labelled with P_n denote polarizers and two plates labelled with $(\lambda/4)_n$ denote quarter wavelength plates. The pump intensity focused on the sample was controlled by using P_1, $(\lambda/4)_1$, P_2 and P_3. The polarization of the pump beam was controlled by using $(\lambda/4)_2$ and P_4.

3. Results and discussion

First, we show the emission spectra for pDNLC and pDFNLC to a direction normal to the cell plane in Fig. 2(a). At low pump energy, no sharp peak was observed for both samples. When the pump energy increases, pDNLC and pDFNLC show discrete sharp peaks, and the intensities and wavelengths of the peaks are independent of each other and change pulse by pulse. Therefore, the discrete sharp peaks shown in Fig. 2(a) can be identified as a random laser action. The emission intensity begins to increase rapidly at almost the same threshold of pump energy (~ 10 nJ) both for pDFNLC and for pDNLC as shown in Fig. 2(b); the light scattering by the MNPs dispersed in NLCs could be ignored. The random laser action in planar alignment cells emits only two opposite directions normal to the cell plane as previously reported [10]. Meanwhile, vDNLC and vDFNLC did not exhibit any sharp peaks even when the pump energy was 15 nJ. The random laser emission can be observed only in the directions normal to the cell plane of the planar alignment cells. It implies that random laser in DFNLCs only occurs in the direction normal to the local nematic director.
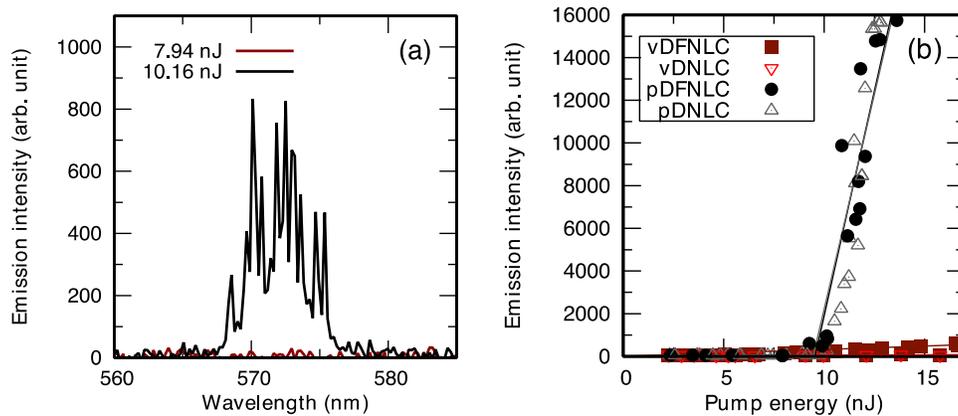

Fig. 2. Lasing characteristics. (a) The emission spectra for pDFNLC at different pump energies are shown. Discrete sharp peaks emerged when the pump energy is stronger than the lasing threshold (10.16 nJ, black) whereas they could not be observed when the pump energy is weaker than the threshold (7.94 nJ, red). (b) The dependences of emission intensity on pump energy in vDFNLC (red, square), vDNLC (red, reverse triangle), pDFNLC (black, circle) and pDNLC (gray, triangle) are shown.

We observed the reorientation of the nematic director (*n*) of pDFNLC in an external magnetic field (*B*) to realize the magnetic switching of the random lasing under polarizing microscopy as shown in Fig. 3. Without any magnetic fields (*B* = 0 mT), *n* is along the rubbing direction of the LC cell (*R*). Under a magnetic field along *R* (*B*~50 mT), ferromagnetic domain walls, the dark lines, could be observed because of the magnetic reorientation of the local nematic director. This implies the controllability of the random laser action in FNLCs by an external magnetic field.
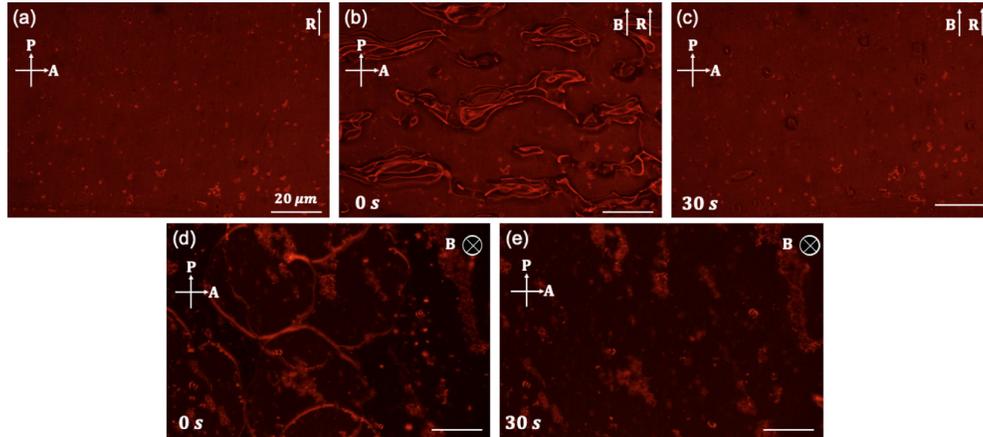

Fig. 3. Switching of ferromagnetic domains in pDFNLC in external magnetic fields as seen by polarizing microscopy. The images seem to be bright because of the fluorescence of the dye. (a) Without a magnetic field (*B* = 0 mT). Polarizing micrographs were taken under a magnetic field (*B*~50 mT) along *R* from (b) 0 s to (c) 30 s after application of the field. Polarizing micrographs under a magnetic field (B~50 mT) normal to the cell plane from (d) 0 s to (e) 30 s after application of the field.

We recorded the emission spectra for pDFNLC under an external magnetic field normal to the cell plane. The stronger the applied magnetic field was, the lower the emission intensity was, and the random laser finally was extinguished as shown in Fig. 4(a). The plot of the random laser intensity vs. the applied magnetic field shown in Fig. 4(b) suggested that the magnetic threshold of the random laser action is lower than 1 mT for pDFNLC, whereas that is 80 mT for pDNLC; the magnetic responsivity of random laser action in pDFNLC is more than 100 times as high as that in pDNLC. The origin of the magnetic NC switching of the random laser action can be explained in terms of the transition dipole moment of the dye molecule. Pyrromethene 597 is rod shaped, and therefore the transition dipole moment is aligned along the long axis in the rod-shaped LC molecules [32]. The dye molecule dissolved in an NLC phase emits linearly polarized light along the long axis of the dye molecule, and the emitted light propagates through the direction normal to the long axis of NLC molecules because NLC molecules also have an electric anisotropy. And the multiple scattering of light in the plane normal to the long axis of the dye molecules occurs so that the direction of the random laser emission is normal to the nematic director. In our results, an external magnetic field normal to the LC cell plane induced the reorientation of the dye molecule alignment along the direction normal to the cell plane by the reorientation of the nematic director. In this case, the coherent loop becomes parallel to the cell plane. In addition, the dye absorption also depends on the angle between the polarization direction of the pumping laser and the local nematic director [10], and therefore, the absorption decreases by applying a magnetic field normal to the cell plane. Although we cannot quantitatively discuss the origin of this phenomenon, the anisotropy of the coherent loop and/or that of the dye absorption are likely to contribute the extinction of the random laser emission in a magnetic field.
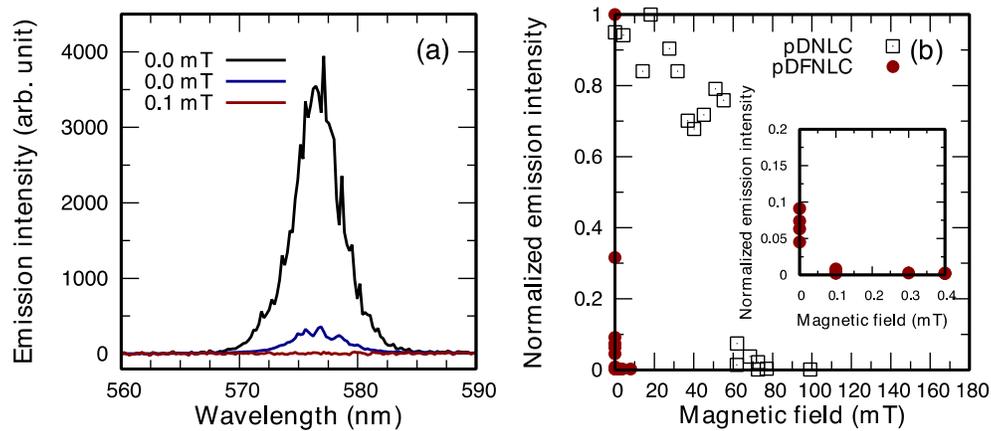
Fig. 4. Lasing characteristics for pDNLC and pDFNLC in an external magnetic field normal to the cell plane at the pump energies of 124 nJ and 119 nJ, respectively. Only for this measurement, instead of the lens with a focal length of 35 mm used in the other experiments, a lens with a focal length of 250 mm is used to prepare the optical setup inside an electro-magnet. (a) The emission spectra under different magnetic field intensities. (b) The dependence of emission intensity in an external magnetic field for pDNLC (black, square) and pDFNLC (red, circle), respectively. The emission intensities are normalized based on the maximum intensity for each sample being set to 1. The inset is the magnification in the magnetic field range from 0 to 0.4 mT and in the range of the normalized emission intensity from 0 to 0.2.

We observed the magnetic switching of the random laser action in pDFNLC. Without any magnetic fields, the random laser emission is spotted on the screen as shown in Fig. 5(a). When a magnetic field (~ 10 mT) was applied normal to the cell plane, the spotted random laser was quickly extinguished in less than 1 second as shown in Fig. 5(b).
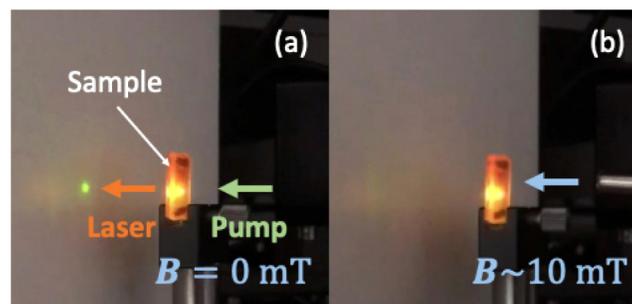

Fig. 5. Magnetic switching of random laser action in pDFNLC. (a) The random laser emission is spotted on the screen. (b) The random laser emission was extinguished when the magnetic field is applied normal to the cell plane.

We measured the emission spectra for vDFNLC and vDNLC in an external magnetic field parallel to the cell plane. As we described before, the random laser action could not be observed in this configuration without any magnetic fields. When an external magnetic field parallel to the cell plane increased, the emission intensity increased and reached the highest value as shown in Fig. 6(a). The magnetic field thresholds of the random laser switching are likely to be about 20 mT and 140 mT for vDFNLC and vDNLC, respectively. The pump energy threshold of the random laser action under external magnetic field of 250 mT for vDFNLC was about 5 nJ as shown in

Fig. 6(b). The origin of the magnetic switching of the random laser action can be also explained as we have done before. The homeotropically aligned FNLCs are reoriented homogeneously by the external magnetic field parallel to the cell plane, and then, the light emission from the dye molecule directed normal to the cell plane increases. When the amplification of light propagating normal to the cell plane exceeds the loss, laser action starts. These results represent the first report of magnetic NO switching of random laser emission using the magnetic reorientation of nematic director from homeotropic (off) to planar (on) alignment.
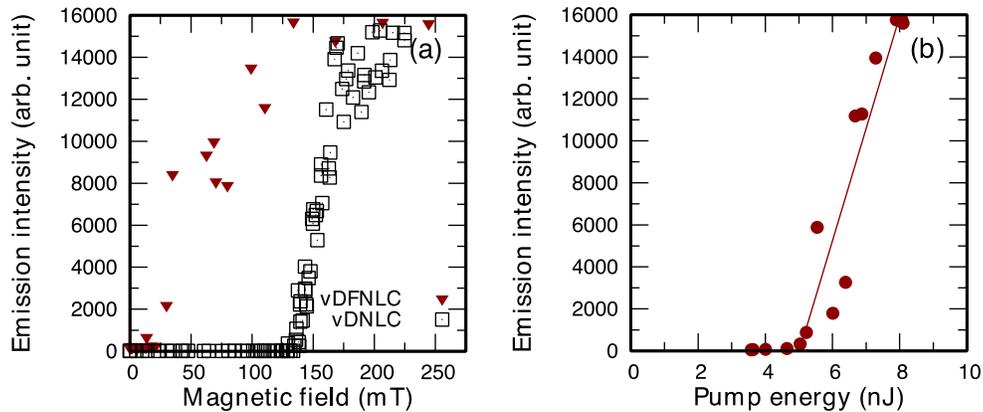

Fig. 6. Lasing characteristics under an external magnetic field. (a) The emission intensities for vDNLC (black, square) and vDFNLC (red, reverse triangle) in different magnetic field intensities at the pump energy of 20.2 nJ and 14.1 nJ, respectively. The emission intensities are normalized based on the maximum intensity for each sample being set to 1. (b) The dependence of the emission intensity for vDFNLC in an external magnetic field parallel to the cell plane of 250 mT.

To discuss the effect of the director field on the polarization of the laser action, we recorded the polarization of random laser emission from vDFNLC in an external magnetic field of about 250
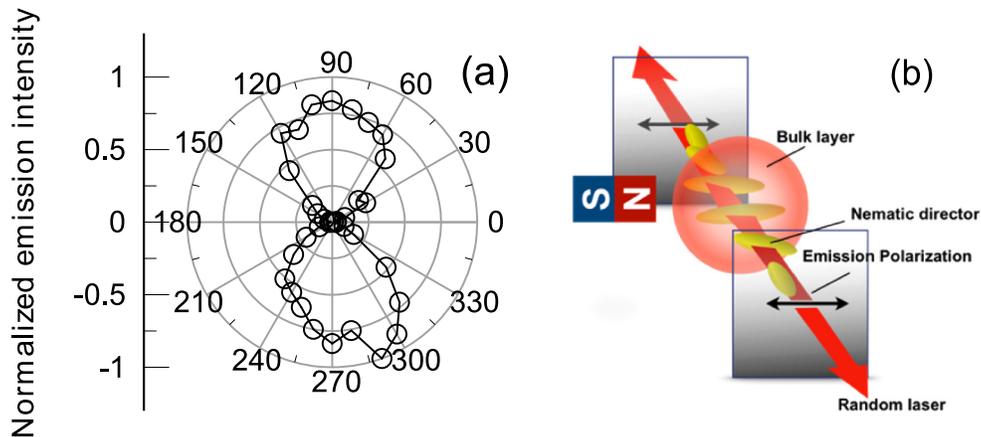

Fig. 7. The polarization of random laser for vDFNLC under a magnetic field parallel to the cell plane. (a) Polar plot of the polarization of the random laser. The emission intensities are normalized based on the maximum intensity for each sample being set to 1. The direction of the external magnetic field was along 90°. (b) Sketch of the random laser emission in vDFNLC in an external magnetic field parallel to the cell plane.

mT parallel to the cell plane as shown in Fig. 7(a). If the DNLC is well aligned homogeneously, the random laser emission from DNLC is generally polarized linearly with the same direction as the nematic director, because of the orientationally coupling between the transition dipole moment of the dispersed dye molecule and the nematic director [10,12]. This coupling forms the polarization-preserving backscattering [33], resulting in the linearly polarized random laser emission. In this case, the random laser emission in vDFNLC under a magnetic field parallel to the cell plane showed the linear polarization. It is worth pointing out that the polarization angle was the same as the external magnetic field direction ($\theta=90°$). At the bulk layer in our sample, the random laser polarization is along the local nematic director whose direction is the same as the magnetic field and emitted without being rotated as shown in Fig. 7(b) because the local nematic director is not twisted as previously explained the random laser action in twisted nematic liquid crystals [15,21]. In this way, the random laser polarization for vDFNLC was parallel to the magnetic field direction.

4. Conclusion

In conclusion, the intensity of random laser emission in dye-doped FNLCs can be controlled in NO and NC switching modes by a weak magnetic field ($\sim$1 mT). To observe a random laser action in LC cell, light scattering normal to the cell plane is essential [34]. The external magnetic field reorients the nematic director of FNLCs, resulting in the reorientation of the fluorescent light scattering direction and a closed coherent loop is generated in the plane normal to the nematic director. Therefore, the random laser emission can be switched by an external magnetic field. The intensity of the random laser in NLCs is highest when the polarization of the pump beam is parallel to the nematic director [12]; if a circularly polarized beam is used for the pumping, the polarization of the random laser in dye-doped FNLCs can be controlled by an in-plane magnetic field. This magnetically highly-sensitive random laser makes it easier to apply the random laser to magneto-optic devices in the future. Moreover, this is the first example of the magnetic switching of random laser emission from off to on state using the reorientation of the nematic director. These results could deepen the understanding of the mechanisms of random laser action in LC phase.

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