Electronic control of nonresonant random lasing from a dye-doped smectic A* liquid crystal device

S. M. Morris, A. D. Ford, M. N. Pivnenko, and H. J. Coles
Centre of Molecular Materials for Photonics and Electronics, Cambridge University Engineering Department, 26A Cambridge Science Park, Cambridge CB4 0FP, United Kingdom

(Received 8 September 2004; accepted 31 January 2005; published online 30 March 2005)

The electronic control of the excitation threshold for random lasing in a dye-doped smectic A* liquid crystal is demonstrated. Random lasing is the term given to the nonlinear amplification of light which is the result of feedback due to multiple scattering. With the application of an electric field the smectic A* phase forms a highly scattering texture for which the nonlinear amplification of light occurs at an excitation threshold of 10 μJ/pulse. In comparison, nonlinear amplification is not observed in the field induced homeotropic texture. As a result, a device has been conceived and demonstrated whereby random lasing is switched “on” or “off” with an applied electric field.


Random lasing, which results from the multiple scattering of light, has been observed in both inorganic and organic media. Such lasing also comes in two flavours: it can be coherent or incoherent depending upon whether the feedback is resonant or nonresonant, respectively.

From an applications point of view achieving control of the threshold for random lasing with electrical stimulation is highly desirable. So far, the majority of the research on random lasing performed in the laboratory has been on powder lasers and solutions of micro and nanoparticles in laser dyes, which cannot be controlled by external stimuli. However, external control over the scattering strength has been demonstrated in nematic liquid crystal solutions incorporating sintered glass and a laser dye. The sintered glass was added to enhance the scattering in the nematic medium which is otherwise a weak scatterer in thin films (~10–20 μm). The results showed that the emission from the sample could be switched off or on by heating or cooling the sample to above or below the clearing temperature so as to alter the excitation threshold. However, as yet there have been no reports demonstrating the ability to change the excitation threshold using an electric field.

In this letter, we demonstrate nonresonant random lasing in an electrically controlled focal conic scattering texture of a smectic A* phase (SmA*) doped with a laser dye and a chiral additive but without the need to include scattering particles. We examine how the different optical textures, i.e., pseudo planar, homeotropic, or electrically induced focal conic, intrinsic to these materials, influence the random lasing. We show that the feedback leading to the nonlinear amplification is nonresonant by examining closely the emission spectrum. Finally, we give a basic demonstration of how the emission changes as the excitation threshold is changed using an electric field and discuss how this feature could serve as the basis of a device.

The sample used for this study was prepared by dissolving a low concentration of the laser dye 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran (DCM, Lambda Physik) (~2% by weight) and the high twisting power chiral dopant (BDH1281, Merck NB-C) (~3% by weight) into the nematic host 4-octoxy-4′-cyanobiphenyl (M24, Merck NB-C). The sample was doped with a chiral additive so as to reduce the size of the focal conic domains and increase the scattering density. The phase sequence of the sample is: Isotropic-(76 °C)-chiral nematic-(48 °C)-Smectic A-(42 °C)-crystalline, where the numbers between the brackets denote the phase transition temperature determined by optical microscopy. The sample was capillary filled into 7.5 μm thick cell which had rubbed polyimide alignment layers and gave a pseudo-planar aligned texture in the undeformed state of the SmA* phase.

The emission properties of the sample were studied using a focused Nd:YAG laser (λ=532 nm) which emits 6 ns long pulses at a repetition rate of 1 Hz. At the sample the spot size of the pump laser is estimated to be 160 μm in diameter. A microscope objective was used to collect and couple the light emitted from the sample into an optical fiber, which was connected to a spectrometer with a resolution of 1.3 nm (USB2000, Ocean optics). In order to determine whether the type of feedback in our case was resonant or nonresonant we measured the spectral profile of random laser emission using a spectrometer with a 0.04 nm spectral resolution (HR2000, Ocean Optics).

Theoretically, a length scale which is of particular importance, when considering the excitation threshold for random lasing, is the transport mean free path, λ, the average distance a photon travels before its direction of propagation is completely randomized. This can be considered as a way of quantifying the scattering density. A low threshold for random lasing requires a small λ. There have been other reports which have shown theoretically and experimentally that the threshold for lasing can indeed be lowered by decreasing λ. In Ref. 9, the lowering of the excitation threshold by decreasing the transport mean free path was demonstrated by increasing the concentration of the scattering particles. For a Smectic A* liquid crystal we will show the same effect can be achieved by changing the texture of the mesophase using an electric field. A uniform alignment of the director such as a pseudo-planar or homeotropic texture has a large λ in comparison to a nonuniform alignment such as a focal conic texture. By employing an ac electric field (E...
strengths at a frequency of 1 kHz in Figs. 1

were also noted in Ref. 10. At 17 V\textmu m\textsuperscript{−1} these lines had

irregularities in the alignment is on a much smaller length scale than was the case for the initial pseudo-planar texture. This scattering texture begins to be formed at the threshold of 1 V \textmu m\textsuperscript{−1} [see Fig. 1(e)] and continues to evolve as the strength of the electric field is increased. Above the threshold a drop in the transmitted intensity is recorded, Fig. 1(e), and is a result of light scattering. For field strengths greater than 4 \textmu V \textmu m\textsuperscript{−1} the transmitted intensity reaches its minimum value, Fig. 1(e), and no further change in the transmitted intensity was recorded. However, the sample was not completely dark because of the presence of defect lines which were also noted in Ref. 10. At 17 V \textmu m\textsuperscript{−1} these lines had disappeared [see Fig. 1(c)]. On decreasing the \( E \) field to 2 \textmu V \textmu m\textsuperscript{−1} the sample formed a highly scattering texture which has a fine grained structure and a gridlike pattern formation [Fig. 1(d)]. A further decrease to zero field results in a return to the pseudo-planar texture. By reapplying a field of amplitude 2 \textmu V \textmu m\textsuperscript{−1} directly, the sample returns to the scattering texture of Fig. 1(b) but the response time is slow ~1 to 2 s. A much faster response, ~10 ms, is observed for a field induced change from the homeotropic, Fig. 1(c), to the scattering texture of Fig. 1(d).

The emission properties of the 7.5 \textmu m thick cell in the electrically controlled scattering state of the SmA\textsuperscript{*} phase are shown in Fig. 2. The peak intensity of the emission spectrum and the linewidth as a function of excitation energy are plotted for the controlled scattering state in Fig. 2(a). For comparison the peak intensity and the FWHM as a function of excitation energy for the controlled scattering state of a dye-doped SmA\textsuperscript{*} phase contained in a 7.5 \textmu m film. The scattering texture was prepared using an applied a.c. electric field of first 17 \textmu V \textmu m\textsuperscript{−1} to obtain a homeotropic texture and then 2 \textmu V \textmu m\textsuperscript{−1} to obtain the scattering texture upon transition from the field induced homeotropic alignment.

Optical textures of the SmA\textsuperscript{*} phase, viewed between crossed polarizers, are shown for different a.c. \( E \) field strengths at a frequency of 1 kHz in Figs. 1(a)–1(d). In addition, a plot of the transmitted intensity through crossed polarizers is presented in Fig. 1(e). For an electric field strength of less than 1 \textmu V \textmu m\textsuperscript{−1} the SmA\textsuperscript{*} phase has the pseudo-planar texture produced by the alignment layer shown in Fig. 1(a). This texture exhibits regions of nonuniform alignment which are on a large length scale. In a previous review of the realignment of a SmA\textsuperscript{*} phase with an applied electric field\textsuperscript{10} it was suggested that the initial zero-field texture is actually a partially aligned focal conic as opposed to an ideal planar alignment, i.e., a pseudo-planar. Figure 1(b) is the fine grained scattering texture observed for \( E \) field strength of 1.5 \textmu V \textmu m\textsuperscript{−1} whereby the separation of the

FIG. 1. The optical texture of the 7.5 \textmu m thick sample for different applied electric field strengths in the SmA\textsuperscript{*} phase at 46 °C. These pictures show a 400×300 \textmu m\textsuperscript{2} portion of the cell viewed between crossed polarizers. The sequence of photomicrographs are for (a) \( E = 0 \) (pseudo-planar), (b) \( E = 1.5 \textmu V \textmu m\textsuperscript{−1} \) (scattering), (c) \( E = 17 \textmu V \textmu m\textsuperscript{−1} \) (homeotropic) on increasing the field strength, and (d) \( E = 2 \textmu V \textmu m\textsuperscript{−1} \) (scattering) on reducing the applied field from the field induced homeotropic texture. (e) The transmitted intensity as a function of an applied a.c. electric field with a frequency of 1 kHz.

FIG. 2. (a) A plot of the peak intensity of the emission spectrum (closed squares) and the FWHM (open triangles) as a function of excitation energy for the controlled scattering texture of a dye-doped SmA\textsuperscript{*} phase contained in a 7.5 \textmu m film. The scattering texture was prepared using an applied a.c. electric field of first 17 \textmu V \textmu m\textsuperscript{−1} to obtain a homeotropic texture and then 2 \textmu V \textmu m\textsuperscript{−1} to obtain the scattering texture upon transition from the field induced homeotropic alignment. (b) Emission spectra measured using a high resolution spectrometer (0.04 nm) at an excitation energy of 90 \textmu J\textperp pulse.
poration the emission intensity for the homeotropic texture is indicated on the graph by the dashed line. The onset of random lasing is indicated in Fig. 2(a) by the presence of a threshold on the peak emission intensity curve at \(\sim 10 \mu J/\text{pulse}\); the dependence exhibits a partial S-shape which is typical of lasers. At the excitation threshold there is a noticeable collapse in the full width at half maximum (FWHM) from 80 to 8 nm. The shape of both the linewidth and peak emission intensity plots are reminiscent of those presented in a report dedicated to the theory of lasing in a multiple-scattering medium (cf. Figs. 5 and 6 in Ref. 8). No threshold for nonlinear amplification is observed for the homeotropic texture and the output consists of only spontaneous emission for the range of excitation energies used. We assume that for the homeotropic texture, \(\ell^*\) is greater than the thickness of the cell and, therefore, no amplification is observed because the average optical path length is equivalent to the cell thickness, i.e., the transport of light is not diffusive. Figure 2(b) is the emission spectrum from the sample for an excitation energy of \(80 \mu J/\text{pulse}\) recorded by the 0.04 nm resolution spectrometer. Overall the spectral profile extends from 610 to 630 nm and has a peak emission intensity which is centered at 619 nm, the FWHM is \(\sim 8\) nm. The shape of the emission curve appears to be relatively smooth and is free of the closely spaced sharp peaks which indicate the spatial resonance of light and, therefore, coherence. There are some small peaks located at the maximum intensity of the emission spectrum but these are far less pronounced than a laser line generated by a microcavity and the fine structure is random. Since no definite peaks were observed we conclude that the observed random lasing was due to nonresonant feedback.

Finally, to demonstrate the electronic control of the excitation threshold for random lasing, the emission spectra of the SmA\(^*\) phase for an excitation energy of \(30 \mu J/\text{pulse}\), which is necessarily greater than the excitation threshold for the controlled scattering texture, is shown for three different textures in Fig. 3. The pseudo-planar, the homeotropic, and the field induced scattering texture. In the initial pseudo-planar state, with no field applied \(E=0\), the emission spectrum includes a broad spontaneous emission peak from 560 to 660 nm and a smaller amplified spontaneous emission peak centered at 616 nm. For the pseudo-planar texture we found that an excitation threshold for random lasing occurs at \(40 \mu J/\text{pulse}\), which is a factor of 4 greater than the threshold for the controlled scattering texture. The difference in \(\ell^*\) between the pseudo planar texture and the controlled scattering texture is estimated to be approximately one order of magnitude. For an \(E\) field of \(17\) \(V/\mu m\) the contribution of amplified spontaneous emission to the overall emission spectrum is suppressed due to dampening of the director fluctuations in the homeotropic state. For the field induced scattering state, i.e., for \(E=2\) \(V/\mu m\), the output is entirely different. Figure 3(c) shows a more intense, narrower emission line centered at 632 nm for \(E\) field strength of \(2\) \(V/\mu m\) but no alteration in the excitation energy. Note that the wavelength at the center of the emission line is different from the wavelength of the amplified spontaneous emission peak in the zero-field texture. Further as shown in Fig. 2, the output intensity can be increased by increasing the excitation energy.

In summary, we have demonstrated the electronic control of the excitation threshold for nonresonant random lasing in a dye-doped smectic A\(^*\) thin film. Random lasing has been observed in a field controlled scattering texture above an excitation threshold of \(10 \mu J/\text{pulse}\) whereas nonlinear amplification, and therefore, random lasing, was not observed in the field induced homeotropic state. Based upon these findings we have demonstrated the ability to switch on and off random lasing using an electric field to change the excitation threshold by altering the scattering density of a dye-doped SmA\(^*\) phase.

We thank the EPSRC for support of this work through a COMIT Faraday Partnership CASE studentship with Dow-Corning (ADF).

\begin{thebibliography}{10}
\end{thebibliography}