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A composite LC/polymer structure, patented as POLIPHEM, when properly doped with a dye is a promising material for compact tunable laser sources. We analyze the properties of distributed feedback system in a form of volume periodic grating in a medium with gain. The conditions required for lasing threshold achievement are considered for second-order Bragg diffraction regimes. First experimental results on the lasing in POLIPHEM DFB oscillator are reported.

Keywords Bragg grating; DFB laser; liquid crystals-polymer composite

Introduction

Materials showing varying electromagnetic features with periodicity at length scale of the optical wavelengths (visible or IR light) attract now great attention in photonics. Soft materials, and in particular liquid-crystal based composites, are among the best candidates because of their ease of processing, high flexibility and tuning possibility. Structures like Holographic-Polymer Dispersed Liquid Crystals (H-PDLC) being realized as Bragg gratings can also be considered for the realization of distributed feedback (DFB) lasers. However, they exhibit a number of drawbacks, ranging from high scattering losses, to high driving fields, to a poorly controlled morphology. A very promising solution is based on a uniform stripe-like grating morphology characterized by alternating lines of polymer and LC. This approach is similar to the polymer cell-walls idea formation for LC display application [1]. Based on this idea, a prototype was first developed and patented at the University of Calabria (Italy) [2] and after at Fraunhofer Institute in Potsdam (Germany) [3].

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It is a new kind of LC-polymer based diffraction grating, named POLICRYPS, (POlymer-LIquid CRYstal-Polymer Slices) or POLIPHEM (POlymer LIquid Crystal Polymer Holograms Electrically Manageable), by the Calabria and German teams respectively. As suggested by the name, they consist of an alternate sequence of homogeneous liquid crystal films and polymer slices. The absence of a droplet dispersion has a number of attractive advantages. It makes them transparent both in the reoriented and in the non-reoriented state. It reduces the electric field necessary to change the director orientation. It permits a good control on the morphology, yielding sharp interfaces between the LC and the polymer, at a nanometric scale. DFB laser and micro-laser arrays have already been realized based on a POLICRYPS structure [4]. However in that case the grating diffractive features of the structure were not exploited, and it was just employed as an ensemble of independent micro-channels hosting dye-doped cholesteric LC.

In our previous work [5] we analyzed the oscillations conditions in a distributed feedback (DFB) system for exact first-order Bragg diffraction scheme and demonstrated the absence of the oscillation regime for pure phase grating with a material gain. In contrast, DFB periodical structure with pure gain modulation was shown to reach oscillations at the exact Bragg resonance. The goal of the present publication is to consider the features of the second-order Bragg configuration and report about the experimental observation of lasing in a POLIPHEM [3] dye-doped DFB oscillator.

Second-Order Bragg Diffraction Regime

A detailed theoretical analysis of the second-order Bragg diffraction regime was presented in Ref [6]. General features of the second-order diffraction effects are similar as expected in the first-order Bragg regime [7], except the strength of the wave coupling is proportional to the square of the modulation magnitude. For pure phase modulation, the formation of pre-oscillation peaks in the grating with gain also occurs at both sides of the central maximum of the diffraction efficiency contour (edge modes [8]). The position of the contour maximum, however, does not coincide with the exact second-order Bragg resonance. In contrast to the first-order Bragg regime, pure amplitude (gain) modulation results also in the edge modes appearance. A specific and attracting features of the second-order Bragg configuration is the possibility to obtain a central oscillation mode with a proper combination of phase and amplitude modulations in a mixed grating.

Let us consider the wave interaction within a spatially modulated medium, as shown in Figure 1. The grating is supposed to be recorded in a thin film between the substrates, thus producing a kind of waveguide. The propagation angle θ is assumed to be close to the second-order Bragg angle θ_B satisfying the condition $n\Lambda\sin\theta_B = \lambda$, where Λ is the grating spacing, n is the average refractive index and λ is the light wavelength in vacuum.

In the following, we shall concentrate mostly on the DFB properties and let the analysis of the waveguide effects for further consideration.

We use the scalar wave equation for y-polarized monochromatic wave, written as

$$\frac{\partial^2 E(x,z)}{\partial x^2} + \frac{\partial^2 E(x,z)}{\partial z^2} = -k_0^2 \varepsilon(z), \tag{1}$$

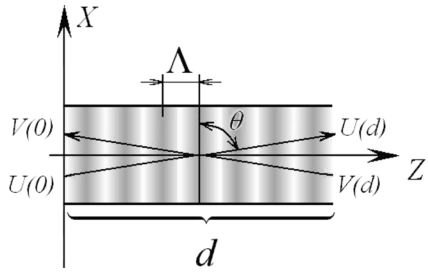

Figure 1. Schematic of the waves in the modulated medium: U is the incident wave, V is the diffracted wave, θ is the propagation angle, Λ is the grating spacing, d is the interaction length.

where k_0 is the wavenumber in free space, $\varepsilon(z)$ is the dielectric permittivity and the field in the modulated medium is

$$E(x,z) = [U(z)\exp(iKz) + W(z) + V(z)\exp(-iKz)]\exp(i\sigma_x x). \tag{2}$$

The direct propagating wave $U(z)$, intermediate-order wave $W(z)$, and reflected wave $V(z)$ are sought in a form $u\exp(i\sigma_z z)$, $w\exp(i\sigma_z z)$ and $v\exp(i\sigma_z z)$, respectively. The dielectric permittivity can be written as $\varepsilon(z) = \varepsilon_0 + \Delta\varepsilon\cos(Kz)$, where ε_0 is the dielectric permittivity of the material without modulation, $\Delta\varepsilon$ is the magnitude of modulation and $K = 2\pi/\Lambda$.

The solution of Eq. (1) results in the determination of the root [3]

$$\sigma_z = [(\delta - i\gamma)(\delta - i\gamma - \chi)]^{1/2}, \tag{3}$$

where $\delta = k\sin\theta – K$ (detuning parameter), $\gamma = \alpha K/k$ (uniform gain parameter), $\chi = (\pm i\kappa_\alpha - \kappa_n)^2/K^3$, and $\kappa_n = \frac{k^2\Delta n}{n}$ (parameter of the refractive index n modulation), $\kappa_\alpha = k\Delta\alpha$ (parameter of the gain modulation), $k = nk_0$ (wavenumber in the medium).

For the second-order Bragg resonance the expression for the amplitude diffraction efficiency (the ratio of the diffracted wave amplitude $V(0)$ and the incident wave amplitude $U(0)$) was obtained as [6]:

$$\eta = \frac{i\chi\sin(\sigma_z d)}{2i\left(\delta - i\gamma - \frac{\chi}{2}\right)\sin(\sigma_z d) - 2\sigma_z\cos(\sigma_z d)}\exp\left(\alpha\frac{k^2 - K^2}{kK}d\right), \tag{4}$$

Self-oscillations (lasing) occur in the grating when the gain (or distributed gain) reaches a threshold value. An equation for the threshold determination can be derived from Eq. (4), in the condition of the equality

$$\sigma_z\cosh(\sigma_z d) = \left(\gamma + i\delta - i\frac{\chi}{2}\right)\sinh(\sigma_z d), \tag{5}$$

which turns the denominator of Eq. (4) to zero.

The threshold condition strongly depends on mutual overlapping of the phase and amplitude gratings. The sign $\pm$ before the gain modulation term indicates two

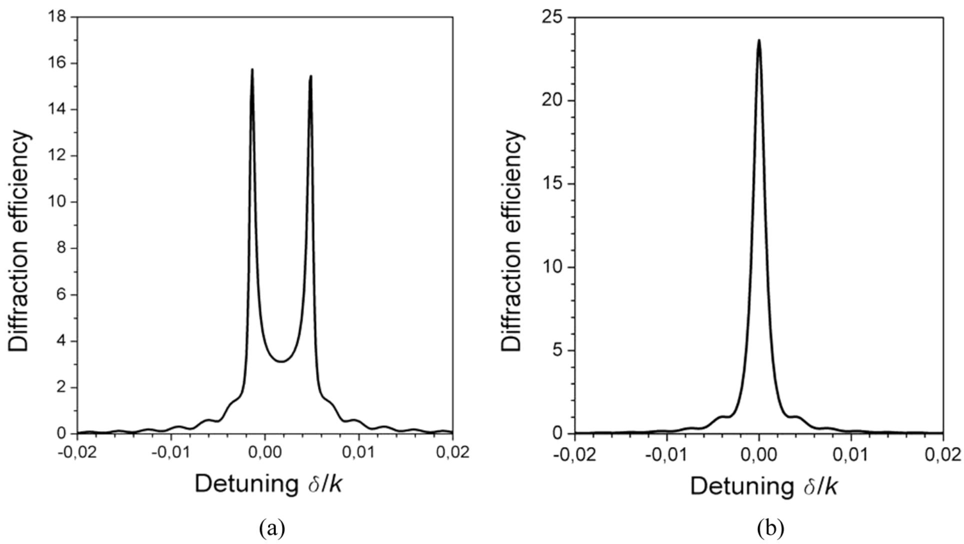

Figure 2. (a) Calculated contour of the diffraction efficiency for pure phase grating with gain in second-order Bragg diffraction regime. (b) The diffraction efficiency contour for mixed grating with parameters $\kappa_n = \kappa_\alpha = 0.02$, $\alpha/k = 8 \cdot 10^{-4}$.

possibilities for the gratings being in phase or out of phase (π-shifted). The out of phase configuration possesses much lower threshold [6].

Figure 2a shows calculated pre-oscillation peaks for pure phase grating with gain in second-order Bragg diffraction regime. In the calculations we used a normalized value $k = 1$ to generalize the results. The calculated contour in Figure 2a was plotted as a function of the detuning δ/k for an interaction length $d = 10^3/k$ (this choice corresponds, using realistic values $k_0 = 10\,\mu\text{m}^{-1}$ and $n = 1.6$, to a value $d = 62.5\,\mu\text{m}$), and the other grating parameters $K/k = \sin\theta_B = 0.9$, $\alpha/k = 8 \cdot 10^{-4}$ and $\kappa_n = 0.05$.

Similar calculations were performed for mixed-type grating. In the optimal situation, corresponding to $\kappa_n = \kappa_\alpha$, the oscillation mode appears with a minimum threshold at the exact second-order Bragg resonance, as seen in Figure 2b.

Experiment

In our experimental study a composite POLIPHEM material was used for DFB structure creation [3]. We used a reactive mixtures prepared with 35–40 wt.% of solution of nematic liquid crystal 5CB(70%) + 7CB (30%) and optical glue NOA 68 from Norland Inc as photocurable base. The mixture was doped with 1% of Perylene as a lasing active media. All components were thoroughly mixed together to get a homogeneous, isotropic and optically transparent blend. The photosensitive films were prepared by drop-filling between ITO coated glass substrates. Mylar spacers were used to obtain desired thickness in the range of 20 μm. The initial films showed an excellent optical quality. All experiments have been carried out at room temperature (20–22°C).

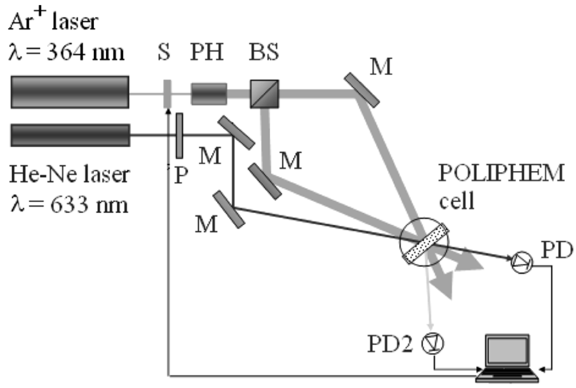

Figure 3. Experimental scheme for grating recording in the dye-doped POLIPHEM cells. M are mirrors, P is the polarizer, PH is the pinhole expander, PD1 and PD2 are photodetectors.

The scheme of grating recording is sketched in Figure 3. UV radiation from a cw Ar^+ laser was spatially cleaned and expanded by the pinhole element PH and split by the beamsplitter BS to create recording beams interfering at the sample plane.

In order to obtain necessary modulation strength, measured through the diffraction efficiency of a probe He-Ne laser beam, the recording setup was accomplished with the photodetectors PD1 and PD2. The holographic exposure was terminated upon reaching the saturation value of the diffraction efficiency. The signal for stopping the recording comes to the shutter S from a control system.

We have recorded gratings with different spatial periods. For the sake of easy direct microscopic observation of the grating fringes, also a large enough spacing was realized (20 μm). The grating structure was analyzed with the aid of crossed polarizers, which blocked the light transmission through the isotropic polymer

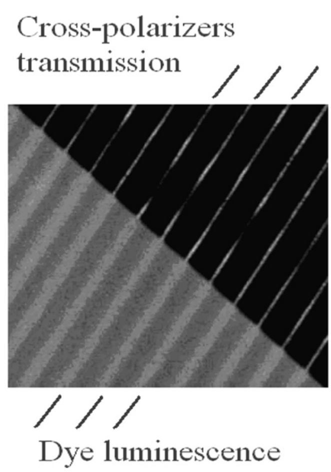

Figure 4. Microscope image of a POLIPHEM sample with a recorded grating (spacing about 20 μm). The left-down part shows the luminescence of the dye dopant; the right part is seen through crossed polarizers. Bright lines correspond to the birefringent LC regions, indicated outside with dashes. Clearly, the maxima of luminescence correspond to the LC regions.

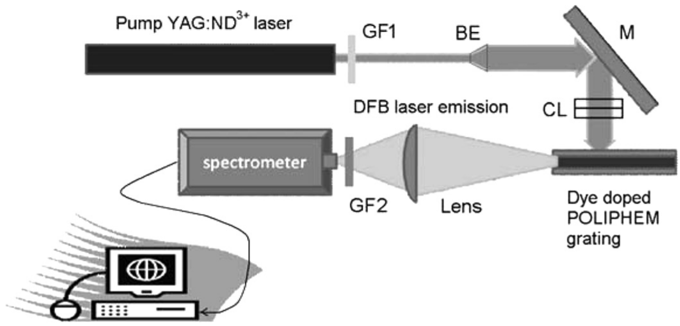

Figure 5. Sketch of the experimental setup for luminescence measurements of the dye-doped POLIPHEM samples. Beam expander BE with the mirror M and cylindrical lens CE forms the pump zone. Glass filter GF1 cuts off the fundamental harmonic of Nd:YAG laser (1.064 μm), glass filter GF2 cuts off the pump light scattering at 0.532 μm. The spectrum of the dye luminescence is analyzed with the spectrometer and recorded with the computer.

stripes. However, LC regions possessing birefringence can be visualized in transmitted light. Another part of the cell with the dye-doped grating observed through red glass filter showed also periodic variation of luminescence (Fig. 4). As seen, the maximum of luminescence, corresponding to the maximum of dye concentration, coincides with the LC location. This observation gives the confirmation on the mutual orientation of the phase and gain gratings.

The grating spacing for the DFB oscillator was chosen to satisfy the second-order Bragg resonance condition: $\Lambda = \lambda/n$. The spacing measured with the aid of He-Ne laser diffraction amounted to 0.38 μm, thus the expected wavelength of oscillation correspond to about 0.6 μm.

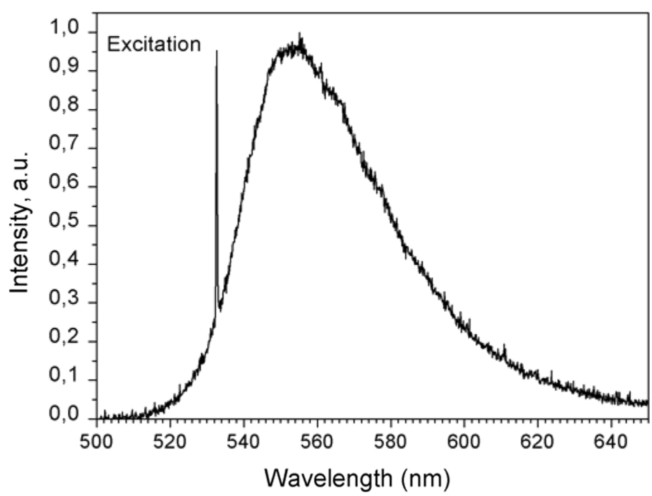

Figure 6. Luminescence spectrum of the dye observed under excitation of the dye-doped POLIPHEM film by second-harmonic pulse radiation of YAG:Nd^{3+} laser ($\lambda = 532$ nm). The peak at the excitation wavelength 532 nm is also seen.

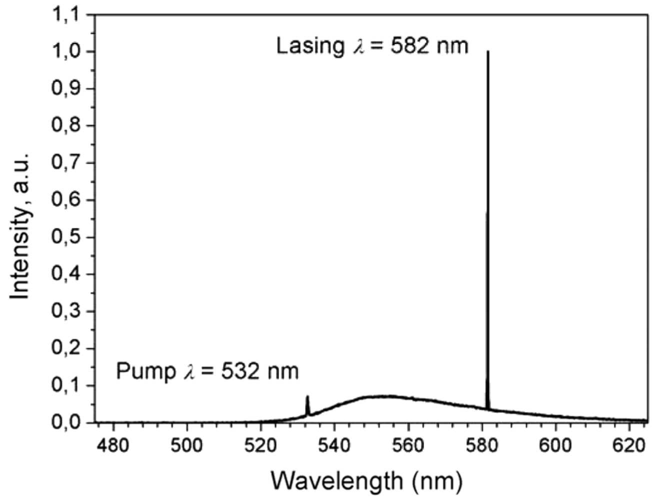

Figure 7. Experimentally detected laser line in a POLIPHEM DFB oscillator.

The experimental scheme for lasing detection is shown in Figure 5. The second harmonic radiation of a pulsed Nd:YAG laser (10 ns pulse, 12.5 Hz repetition rate) was used as pumping source. The pump beam was expanded and then focused by a cylindrical lens onto a cell containing the recorded grating. The length of the illuminated zone was about 10 mm. The light detection scheme was the same both in the case of measurement of luminescence and when lasing oscillations were generated. The emitted light, propagating in the cell plane and in the direction perpendicular to the grating vector of the POLIPHEM, was collected by a lens and analyzed by a spectrometer.

First, pumping the sample with a cw laser at the same wavelength as the pulsed laser ($\lambda = 532$ nm), we have observed the luminescence spectrum, as shown in Figure 6. The dye luminescence possesses a broad spectrum with the maximum at $\lambda = 554$ nm.

Then, pumping the sample with the pulsed laser, a sharp laser emission line was detected at $\lambda = 582$ nm, as shown in Figure 7. The spot of the laser emission, at the exit of the POLIPHEM structure, was a typical one for a waveguide mode, exhibiting a quite large divergence in the grating plane. The linewidth of the emitted radiation was within the resolution of the spectrometer, namely 0.2 nm. A systematic set of measurements is currently in progress for a complete characterization of the observed lasing effect.

Conclusions

Our theoretical analysis of the DFB oscillator in the second-order Bragg diffraction regime has demonstrated specific properties of the grating structure required for efficient generation of laser oscillations. In particular, when, in addition to the refractive index, also the gain is periodically modulated, the second-order Bragg resonance represents the most effective oscillation condition. Extending this theoretical study to take into account the variation of the LC refractive index under applied voltage, and the corresponding lasing wavelength tuning is a subject of further investigation.

In our experimental realization of a DFB oscillator, through a dye-doped POLIPHEM grating, we certainly have a mixed phase-amplitude grating. In fact, the refractive index modulation is inherent to the POLIPHEM structure, due to the difference between the LC and polymer indices. On the other hand, as Figure 4 shows, the distribution of the dye dopant is non-uniform, and therefore also the gain is spatially modulated. The observation, even though preliminary and without a full characterization, of a spectrally narrow laser line at the wavelength corresponding to the expected second-order Bragg resonance, confirms, at least qualitatively, the main result of the theory.

In summary, we succeeded in the demonstration that laser emission from POLIPHEM-based DFB oscillator is viable and promising for the realization of an efficient, tunable, soft-material micro-laser.

References

[1] West, J., Rouberol, M., Ji, Y., & Francl, J. (1999). Method for Forming Liquid Crystal Display Cell Walls Using a Patterned Electric Field, U.S. Pat. 5,929,960.

[2] Caputo, R., Umeton, C., Veltri, A., Sukhov, A., & Tabiryan, N. Italian Patent TO2003A000530 of 07/09/2003.

[3] Sakhno, O., Slussarenko, S., & Stumpe, J. (2004). POLIPHEM – new type of nanoscale polymer-LC switchable photonic devices. *SPIE Proceeding*, *5521*, 38.

[4] Barna, V., Caputo, R., De Luca, A., Scaramuzza, N., Strangi, G., Versace, C., Umeton, C., Bartolino, R., & Price, G. N. (2006). Distributed feedback micro-laser array: Helixed liquid crystals embedded in holographically sculptured polymeric microcavities. *Opt. Express*, *14*, 2695–2705.

[5] Vasnetsov, M. V., Bazhenov, V. Yu., Slussarenko, S. S., Sakhno, O., Stumpe, J., & Abbate, G. (2008). First approach to the analysis of the lasing conditions in POLIPHEM© structures. *Mol. Cryst. Liq. Cryst.*, *488*, 135–147.

[6] Vasnetsov, M. V., Bazhenov, V. Yu., Slussarenko, S. S., & Abbate, G. (2009). Coupled-wave analysis of second-order Bragg diffraction II. Threshold conditions for distributed-feedback laser oscillation. *JOSA B*, *26*, 1975–1983.

[7] Kogelnik, H. (1969). Coupled wave theory for thick hologram gratings. *Bell System Technical Journal*, *48*, 2910–2947.

[8] Kogelnik, H. & Shank, C. V. (1972). Coupled-wave theory of distributed feedback lasers. *J. Appl. Phys.*, *43*, 2327–2335.