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### Molecular Crystals and Liquid Crystals

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F. Simoni <sup>a</sup> , G. Cipparrone <sup>a</sup> & R. Bartolino <sup>a</sup>

<sup>a</sup> UNICAL Liquid Crystal and Optics Group and GNSM, Dipartimento di Fisica, Università della, Calabria, Italy

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# Tuning of a Dye Laser By a Liquid Crystal<sup>†</sup>

F. SIMONI, G. CIPPARRONE and R. BARTOLINO

UNICAL Liquid Crystal and Optics Group and GNSM, Dipartimento di Fisica, Università della Calabria, Italy

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We describe how a cholesteric liquid crystal device can be used in a laser cavity to induce tuning of the laser line. Tuning of the order of 10Å is obtained by applying low voltages to the liquid crystals film. The problems, the advantages and the feasibility of the method is discussed, reporting several experimental data.

Keywords: cholesteric liquid crystal, laser, laser cavity, tunable laser

#### I. INTRODUCTION

The peculiar property of selective reflection due to the helical structure of cholesteric liquid crystals has been considered attractive by several scientists in order to exploit a liquid crystal device as a tunable reflector in a laser cavity. In fact, it is well known that the reflection band is quite narrow ( $\sim$ 20 nm) and its spectral position is determined by the value of the pitch of the cholesteric helix. Therefore, when external agents (such as electric or magnetic fields, mechanical strains, temperature variations) change the pitch value p, the reflection peak shifts according to the relation  $\lambda = np$ , where n is the average refraction index and  $\lambda$  the light wavelength.

This property suggests the use of cholesteric liquid crystals as tunable reflectors, where the reflectivity is driven by external factors, in order to achieve an external control of the modulation of the laser gain. Using this kind of mirror, Il'Chishin et al. 1 realized a dye laser

<sup>†</sup> Supported by CNR under contract 83.01863.11.

with an emission band of 4 nm, using temperature variations as the tuning method. A second example given by the same authors is a distributed feedback dye laser<sup>2</sup> where the feedback effect is produced by the Bragg reflection of a cholesteric liquid crystal. In this case the laser is a thin film where the liquid crystal is mixed to the active medium. One of the major limitations of these methods is the use of temperature variations to induce tuning of the laser bandwidth, which implies very slow modulation. Moreover, nobody paid much attention to the anomalous rotatory power of cholesterics within the frame of this research.

Since 1980, in our group we have studied the effects of d.c. electric fields and of mechanical strains (dilation, compressions, shears) on the optical properties of cholesteric liquid crystals. We have demonstrated that with low excitations it is possible to get dramatic variations of the optical rotation for the wavelength falling in the selective reflection band.

In the following, we describe how it is possible to exploit these effects as a tuning method for dye lasers, which are more sensitive to the external agents in comparison to systems using only the reflection property. In Section II we give the principles of the method, while the experiments are described and discussed in Section III.

#### II. THE TUNING METHOD

The basic element of our method is the Voltage Controlled Polarizer (VCP)<sup>3</sup> and the tuning effect has been briefly reported.<sup>4</sup> The VCP is realized by a mixture of cholesteric liquid crystals (cholesteryl chloride, cholesteryl oleate, cholesteryl nonanoate) and has been described elsewhere.<sup>4</sup> We recall here only how it works. The externally applied electric field induces a structure deformation in a way that the incident light "sees" the molecular layers at a slant angle. For small deformations (i.e., low-applied voltages), the effect is the same one has increasing from zero the angle of incidence of the light. Therefore, a shift of the reflection peak to shorter wavelengths in observed and the same effect can be detected on the rotatory dispersion curve.

We have demonstrated that by applying a low voltage to a cholesteric liquid crystal, it is possible to change the polarization of the light in a continuous range controlled by the applied voltage if a monochromatic light is used.<sup>6</sup> It has also been shown that the effect is strongly dependent on the wavelength and is more evident for  $\lambda$  close to the reflection peak  $\lambda_0$ .

The shift of the rotatory dispersion band induces a variation of the rotatory power for each wavelength. The rotatory power can vary by several orders of magnitude for  $\lambda$  close to  $\lambda_0$  and by a neglecting quantity if  $\lambda$  is far from  $\lambda_0$ .

The principle of our tuning method is shown in Figure 1, where the rotatory dispersion curve of a generic cholesteric liquid crystal is reported. If we fix the angle of optical rotation  $(\rho_1)$ , we get a different wavelength for each applied voltage. To obtain this effect it is necessary to place a VCP inbetween two crossed polarizers. In fact, only the wavelengths suffering an optical rotation close to  $\pi/2$  will be transmitted by such a device with low attenuation. By applying a small voltage, the rotatory dispersion curve shifts and induces a shift on the transmission band of the device.

This system seems suitable to get a gain modulation in a dye laser in order to control the laser emission band. Of course, care has to be taken in order to match the rotatory dispersion curve of the cholesteric liquid crystal with the fluorescence band of the gain medium.

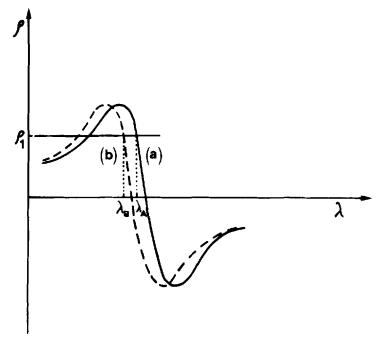


FIGURE 1 Typical rotatory dispersion curve for cholesteric liquid crystals. The angle of optical rotation vs the light wavelength is reported. (a) V=0 Volt; (b)  $V\neq 0$  Volt. We have the same optical rotation  $\rho_1$  for  $\lambda_A$  at V=0 and for  $\lambda_B$  at  $V\neq 0$ .

In such a case, the effect of inserting the device in the laser cavity will be a narrowing of the emission band. Then the described effect of the electric field will induce the tuning.

#### III. THE EXPERIMENTS

We have realized a very simple cavity depicted in Figure 2. The pump beam is given by the second harmonic ( $\lambda = 530$  nm) of a Nd-YAG Q-switched laser (pulse duration  $\tau = 20$  ns). A cylindrical lens focusses the pump beam on a spectrophotometric cuvette filled by a solution of Rd-6G in ethyl alcohol ( $5 \cdot 10^{-3}$  M/l). Between the dye cuvette and the back mirror ( $R_2 = 99\%$ ) are placed the crossed polarizers with the VCP inbetween. From the output mirror ( $R_1 = 94\%$ ) the radiation is sent to a high resolution ( $\Delta\lambda = 0.5\text{Å}$ ) Jobin-Yvon monochromator and the signal is detected by an AVP photomultiplier. Each spectrum is obtained by detecting the signal intensity at each wavelength on the scope and averaging over 20–30 pulses.

The expected narrowing effect of the laser emission band is reported in Figure 3. In this case, the liquid crystal film was 20  $\mu$ m thick. The effect of the device is to induce a narrowing by a factor of 5.

The data obtained using the same device at two applied voltages are reported in Figure 4 ((a): V = 0 volt; (b): V = 15 volt). One observes a shift of the emission peak of about 12Å with a shift of the whole emission band. Under these voltage values the effect is completely reversible. A further increase of the voltage causes a small variation in the band position but the shape becomes strongly deformed. The cause of this is presumably the permanent distortion

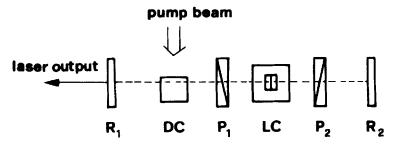


FIGURE 2 Sketch of the dye laser.  $R_1$ ,  $R_2$ : mirrors; DC: Dye Cell;  $P_1$ ,  $P_2$ : polarizers; LC: Liquid Crystal Cell.

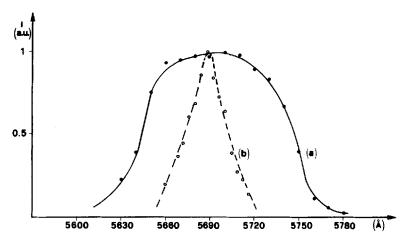


FIGURE 3 Laser emission band: (a) without Liquid Crystal; (b) with the Liquid Crystal device in the cavity.

occurring in the liquid crystal<sup>7</sup> with the onset of many structural defects. It is interesting to compare this data with those obtained using a different configuration, where only one polarizer is coupled to the VCP in the laser cavity. In this case, the device does not fix the optical rotation angle.

The observed effect is therefore due only to the shift induced by the electric field on the reflection band which influences the laser gain with a minimum of transmission.

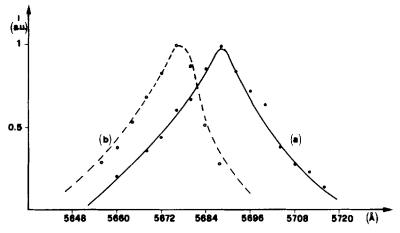


FIGURE 4 Laser emission band with the Liquid Crystal device in the cavity for a Liquid Crystal film 20  $\mu$ m thick,  $T=23^{\circ}$ C. (a) V=0 Volt; (b) V=15 Volt.

Figure 5 reports data for this configuration with a liquid crystal film  $20~\mu m$  thick. Comparing this with Figure 4, one observes only a slight change in the shape of the emission band but no detectable shift in it.

This fact demonstrates that the tuning effect shown in Figure 4 is essentially due to a field-induced shift of the rotatory dispersion curve, while the selective reflection affects only the shape of the band. Actually, depending on the particular matching of the fluorescence band with the cholesteric reflection band, we have a negative effect on the laser emission which sometimes shows a minimum and a secondary maximum close to the emission peak. We are working in order to control this effect.

A different series of measurements has been performed using VCP with liquid crystal film 5 µm thick. An example of these data is reported in Figure 6. Also, in this case, the tuning effect is evident: a peak shift of 8Å with a narrowing of the linewidth from 18Å to 12Å, with only 5 V of applied voltage.

An interesting feature of these data is the shift of the peak to longer wavelengths, while for the case of Figure 4 we have a blue shift. We don't believe this difference is dependent on the thickness of the liquid crystal film but rather on the mutual position of the dye fluorescence spectrum and the rotatory dispersion curve of the cholesteric liquid crystals.

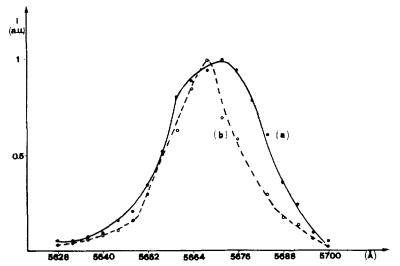


FIGURE 5 Laser emission band with the Liquid Crystal cell and only one polarizer in the cavity. Film thickness:  $20 \mu m$ ,  $T = 23^{\circ}C$ . (a) V = 0 Volt; (b) V = 15 Volt.

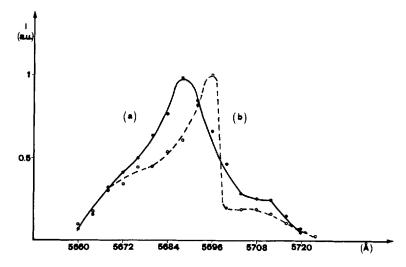


FIGURE 6 Laser emission band with the Liquid Crystal device in the cavity for a Liquid Crystal film 5  $\mu$ m thick,  $T=23^{\circ}$ C. (a) V=0 Volt; (b) V=5 Volt.

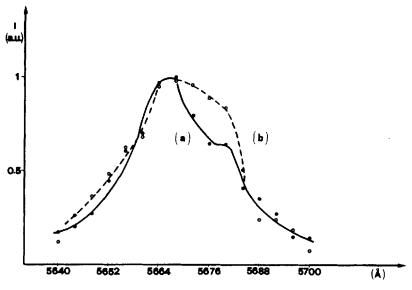


FIGURE 7 Laser emission band with the Liquid Crystal device in the cavity for a Liquid Crystal film 20  $\mu$ m thick,  $T=29^{\circ}$ C. (a) V=0 Volt; (b) V=10 Volt.

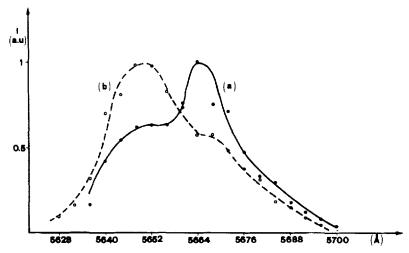


FIGURE 8 Same as Figure 7 at  $T = 22^{\circ}$ C. (a) V = 0 Volt; (b) V = 10 Volt.

In order to show how the matching between these two bands is important to get the effect of tuning, we have used a thermostatic system for the VCP in order to have a temperature control of at least 0.5°C. In this way, we have performed measurements at different temperatures, varying the cholesteric unperturbed reflection band and changing its temperature. Two examples are given in Figure 7 and Figure 8, which report data taken at 29°C and at 22°C, respectively, with a liquid crystal film 20 µm thick. In the first case we have a strong mismatch between the dye fluorescence spectrum and the rotatory dispersion curve of the liquid crystal and we can't see any detectable tuning effect. In the latter case, we have almost optimum matching and we observe a tuning effect similar to that shown in Figure 6. A further decrease of the temperature results in a new mismatch and the tuning effect disappears.

#### IV. CONCLUSIONS

We have described a system using cholesteric liquid crystals as the main medium to obtain tuning of a dye laser line. The experimental data show the effect when a small voltage is applied to the liquid crystal. The method appears reliable and easy optimum matching between the dye fluorescence band and the rotatory dispersion curve can be achieved with temperature control. The use of higher quality optical components of the cavity will lead to further narrowing of the

laser band, giving a real future to this method. Attention should be paid to the possibility of having a wavelength sweep at frequencies up to 20 Hz, which could be interesting for wavelength modulation spectroscopy.

We are studying the use of different external agents to induce the tuning effect.

#### **Acknowledgments**

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