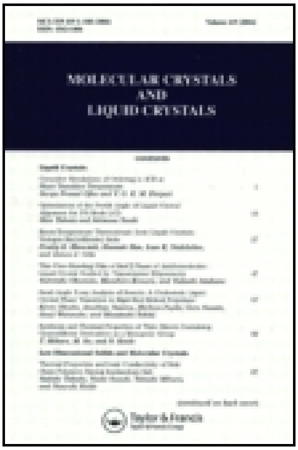

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Cholesteric Liquid Crystal Smart Reflectors

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CHOLESTERIC LIQUID CRYSTAL SMART REFLECTORS

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Abstract We present results concerning the development of liquid crystal smart reflectors, consisting of a cholesteric liquid crystal with temperature sensitive helix pitch, in combination with a light absorbing dye. Light entering the liquid crystal is absorbed by the dye, generating heat which raises the temperature of the liquid crystal. The resulting change in the helix pitch of the cholesteric causes an increase in its reflectivity, reducing the intensity of light that can be absorbed by the dye. This negative feedback stabilizes the reflector for a given light intensity. The smart reflector thus achieves a reflectivity which increases with increasing intensity of incident light. We report on two configurations of the device, with both experimental measurements and mathematical models of the system.

INTRODUCTION

Smart materials are ones that can change their properties in response to appropriate stimuli. They are one kind of realization of the general concept of a smart system. In the general view of a smart system, there are three major functions that must be coordinated to achieve a desired result. First a sensor must detect the signals that are needed. Second, a computational element must interpret the signals and control the third element, the actuator, which changes the system in an appropriate way to respond to the signal detected. This general model describes many kinds of automated devices. Within the general model, one can also distinguish between open-loop systems, in which the changes produced by the actuator element do not affect the detected signal directly, and closed-loop systems, in which the output of the actuator directly affects signals detected by the sensor. Closed loop systems are normally designed to be stabilized by negative feedback.

A smart material is one in which the sensor, computational, and actuator elements are all incorporated in the structure and composition of the material, often at the molecular level. A material with this complexity of design may not necessarily be a single homogeneous substance, but usually contains different components that must be combined and structured in the appropriate way. A layered structure is often useful, with neighboring layers interacting in a way that achieves the desired result. A bimetallic strip that changes its curvature is an example of such a smart material; it is the heart of most thermostats. In the optical control field, photo-chromic sunglasses are good examples of smart materials, in which sub-visible particles dispersed in the glass or plastic matrix change their optical absorbance in response to changes in light intensity. The bimetallic strip is an example of an open-loop system, since the eventual temperature control loop is closed by external elements in a larger smart system. The photo-chromic glass could be employed as a closed-loop device, since the actuation function can directly affect the sensor function, the absorption of light.

Self assembling materials are good candidates for smart material systems, since the correct internal structure of the smart material is usually crucial to its function. In some sense, the ultimate smart material is both fully smart and self assembling. By combining the right components, and processing them in the right conditions, one constructs a functioning smart system, without having to put it together by some laborious process.

Liquid crystals are ideal starting materials for the development of smart materials and systems for optical applications. Because they are birefringent and fluid, with an internal structure that is sensitive to changes in many parameters, such as temperature, chemical composition, and external fields, they offer the possibility to serve as sensors, computational elements and actuators, all in one, for optical systems. The general scenario is as follows: Light acts as a signal, which is sensed by appropriate dye molecules dissolved in the liquid crystal. The properties of the liquid crystal are adjusted so that it responds in a particular way to the energy absorbed by the dye molecules; this is the computational function. Finally, the change in liquid crystal structure, and thus in its optical properties, influences the propagation of light through it, which constitutes the actuator function. The resulting change in optical properties can affect the absorption of light by the dye, providing a negative feedback function to stabilize the system. It is also important to note that in such a system, light acts as both the signal and the power supply for the device.

Moreover, the liquid crystal system can be self assembling, so that by simply achieving the right composition, and containing the liquid crystal with the right boundary conditions, one can construct a functioning smart system.

The engineering of the liquid crystal material into a smart system can take place at several levels. At the molecular level, enough is now known so that particular molecules optimizing special properties can be constructed. Because liquid crystals are fluid, solutions of different components can be made to fine-tune properties, and to incorporate specialized guest molecules, such as dyes. Finally, with a combination of specialized containment and alignment methods, and the use of external fields, the macroscopic structure of the device can be controlled.

In this paper, we present the current status of development of such a liquid crystal device, the smart optical reflector. This is an optical reflector that can change its reflectivity in response to changes in the intensity of light incident on it. We start with a discussion of the properties of liquid crystals that are needed for this application. We then show how they can be used for the desired sensing, control and actuation functions, with the crucial negative feedback coupling needed to achieve stable operation. We then present results of numerical modeling of the system, to illustrate its ideal function. Next we present results of experiments on several realizations of the system, with comparisons to the modeling results. Finally, we discuss the future direction of the research program.

A cholesteric liquid crystal (CLC) is a modification of the nematic liquid crystal in which, due to the presence of optically active molecules, the local optical axis, or director, of the nematic is twisted into a helicoidal structure. The pitch P of this helicoidal structure is often comparable to the wavelength of visible light. The optical properties of CLC's have been widely studied for many years.[1] One of the most interesting properties is the selective total reflection of light in a band of wavelengths centered on the pitch of the helix, a phenomenon similar to the Bragg reflection of x-rays by a periodic crystal structure. Consider light approximately normally incident on a thin layer of CLC in which the axis of the helicoidal structure is oriented normal to the plane of the layer. One circular polarization of light in the reflection band, which matches the sense of rotation of the helix, will be totally reflected as a circularly polarized beam of the same sense, while the opposite circularly polarized component of the incident beam will be transmitted by the CLC.

The spectral width $\Delta\lambda$ of the total reflection band is given approximately by an equation derived by de Vries:[2]

$$\Delta\lambda = P\Delta n \tag{1}$$

Δn is the birefringence of the CLC, the difference between the indices of refraction parallel and perpendicular to the local nematic director. This reflection band can be typically a few tens of nanometers in width. Its finite width arises from the strong interaction of light with the periodic dielectric structure of the cholesteric, in analogy with the finite band gap for electrons propagating in a semiconductor. The basic physics of this effect has been well studied in both theory and experiment.[2–4]

The second interesting property of a CLC which we utilize in this device is the strong temperature dependence of the pitch of its helicoidal structure near the phase transition from the cholesteric phase to the smectic A phase. As temperature T is lowered toward the transition temperature T_{AC}, the pitch diverges toward infinity, with a power law dependence,[5] $P \; \alpha(T - T_{AC})^{-0.67}$. In some materials, this means that the pitch can change from the ultraviolet to the infrared in a range as small as one degree Celsius. The resulting dramatic color change finds application in the well known liquid crystal thermometers and mood rings.

We now show how these properties can be used to produce a liquid crystal smart reflector, in particular, a device which increase its reflectivity in response to an increase of the intensity of light incident upon it.

The first realization of the device is a layered system in which the sensing function has been separated from the liquid crystal, which performs all other smart functions. The device consists of a thin oriented layer of CLC with a highly temperature dependent helix pitch, backed by a solid layer containing a light absorbing dye, as illustrated in Fig.1. It works in the following way. At ambient temperature, the liquid crystal is in the smectic A phase, and incident light passes through it to the absorbing dye layer, where the light is converted to heat. This raises the temperature of the liquid crystal, converting it to the cholesteric phase, and the pitch of the helicoidal structure of the CLC is initially in the infrared. With further heating, the pitch grows shorter, until the reflection band of the CLC begins to reflect part of the incident light. The resulting reduction of optical power reaching the absorbing layer constitutes a negative feedback mechanism, controlling the temperature of the CLC and allowing just enough light to reach the absorbing layer to maintain the device at its correct operating temperature. Any increase in light intensity results in a compensating increase in temperature and reflectivity, and conversely, any decrease in incident light intensity results in a corresponding decrease in temperature and reflectivity.

As a second realization of the device, we have incorporated the dye into the liquid crystal, and eliminated the absorbing backing layer, so that the sensing function is integral to the liquid crystal material, as well as the control and actuation functions. This offers potential problems of stability, since the negative feedback function is partially compromised; even at the highest achievable reflectivity, some light is absorbed, so that reflectivity is always less than perfect and overheating could occur. However, the liquid crystal is now a complete smart material system, which can function in a more flexible environment than the layered system described above.

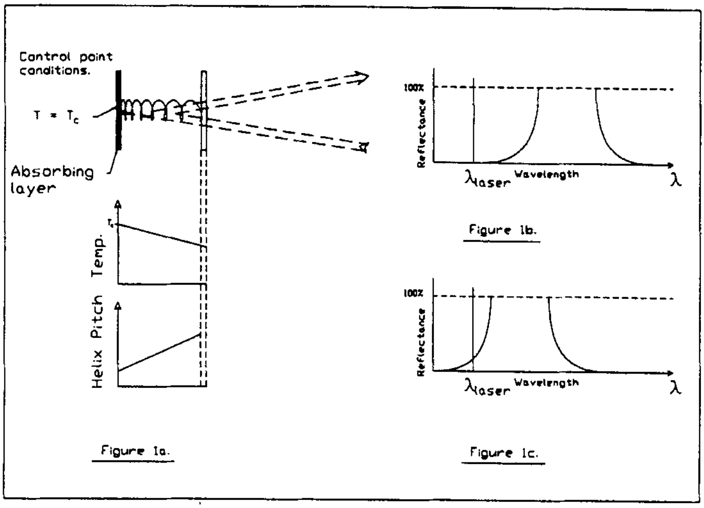

Figure 1a.

Figure 1b.

Figure 1c.

FIGURE 1: Schematic description of a smart reflector. a) In the feedback control mode, light enters from the right, and part of it reaches the absorbing layer, generating heat. This raises the temperature and shortens the helix pitch of the CLC layer. The reduced helix pitch results in increased reflectivity. b) At low light intensity the helix pitch is too long to reflect the laser beam. c) In the control mode, the laser beam is partially reflected.

MODEL CALCULATIONS

We address first the optical properties of the cholesteric helical structure. The exact solution of the wave equation for the propagation of light along the helical axis of a CLC was presented by Mauguin,[6] Oseen[7] and de Vries.[2] Nityananda and Kini[8] applied this theory and used continuity at the boundary to get the reflectivity when a CLC is sandwiched between two isotropic media. Berreman[9] used a 4 × 4-matrix formulation to solve the transmission problem numerically, and Hajdo and Eringen[10] proposed a multilayer model to explain the effect of a pitch gradient in the CLC on the reflectivity.

The reflectivity of a CLC for light of wavelength 0.5 μm, as a function of nP, the mean index of refraction times the pitch of the helix, is shown in Fig.2. We plot long pitch at the left for comparison with experiments below. Notice the steeply rising reflectivity and the total reflection band of finite width, as indicated by the de Vries formula given earlier. The negative feedback mechanism of our device is active on the steeply rising part of this spectrum.

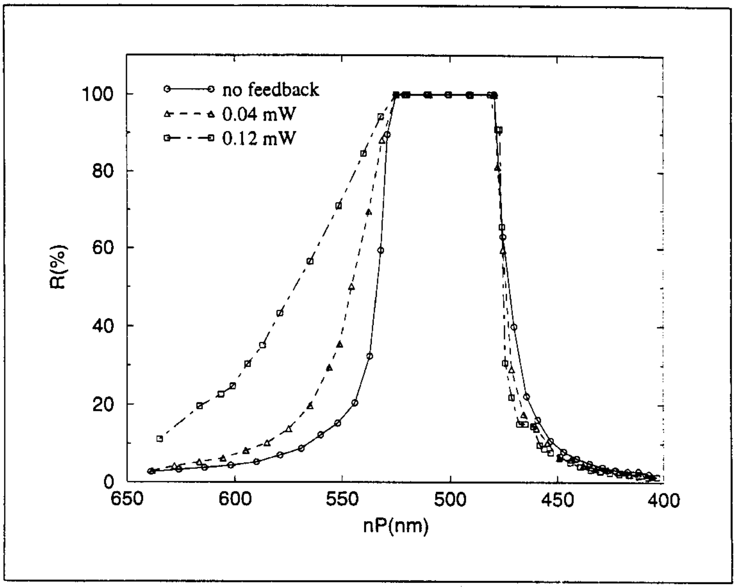

FIGURE 2: Calculated reflectivity R of the two layer model system as a function of refractive index n times ambient helix pitch P, for three incident laser powers.

A full model of the first realization of the device requires the interaction of three variables within the CLC layer: the light intensity, the temperature, and the helix pitch, as functions of distance, from the front to the back of the layer. Light enters the front of the layer, and part of it is transmitted. The light reaching the back of the layer is absorbed and converted to heat. This heat source raises the average temperature of the CLC, and also creates a temperature gradient that conducts heat back to the front of the CLC layer. The temperature change and the gradient produce corresponding changes in the mean pitch and pitch gradient in the helicoidal structure of the CLC. This change in pitch structure changes the transmission of light through the CLC layer. For a CLC in which dP/dT, the change of pitch with temperature, is negative, when operating on the long pitch side of the reflectivity spectrum shown in Fig.2. This produces a negative feedback, which leads to a stable equilibrium among the three variables in the CLC layer. This is summarized by the equation

$$tW = KA\,\frac{\Delta T}{\Delta x}, \qquad (2)$$

in which there is a temperature difference ΔT across the sample of thickness Δx, with area A, characterized by thermal conductivity K, and optical power transmission coefficient t, illuminated by laser power W.

In our model, we assume for simplicity a constant negative dP/dT. We fix the temprature at the front of the CLC layer at the ambient level. Then for a given temperature gradient, we calculate the pitch structure and the optical transmission coefficient of the CLC layer, using the multilayer model to account for the effects of the pitch gradient. From Eq. 2, we then know what incident laser power is required to maintain this temperature gradient. In Fig.2, we show two curves for the reflectivity to the device at higher power levels, as a function of the pitch of the CLC at the ambient temperature at the front of the CLC layer. As optical power W is increased, the pitch at the back of the CLC layer decreases, due to increased temperature at the back, and the reflection coefficient increases. Note that when the pitch at the front of the CLC is such that the reflection coefficient there, is nearly 100%, the relative increase in reflectivity is smaller, since very little light is transmitted in any case. This means that the point at which the reflection coefficient goes to 100% does not shift to longer pitch values. Rather, the reflectivity at longer pitch values increases with increasing power. This is precisely the effect desired for the smart reflector. Notice that on the short pitch side of the reflectivity curves, where the feedback effect is positive, rather than negative, the reflectivity of the CLC decreases with increasing indicent light power. This makes the device unstable, and for a smart reflector, operation in this region must be avoided.

For the second realization of the device, with the dye dissolved in the liquid crystal, we modified the calculations. We used a multilayer model and calculation described above to determine the optical properties of the CLC. Because of the guest-host effect, we can treat this mixture as an "absorptive cholesteric liquid crystal", which is circular dichroic in this case. The first intuitive modification is that the indices of refraction $n_{\|}$ and $n_{\perp}$ become complex where the imaginary parts of $n_{\|}$ and $n_{\perp}$ represent the absorption parallel and perpendicular to the direction of the dye molecule, which is also the direction of the director.

The two glass substrates were put into consideration in the calculation because the heat absorbed in the cholesteric liquid crystal is transferred to the glass substrates, which means that the heat raised the average temperatures of both the liquid crystal and the glass substrates. Here the heat conduction equation in equilibrium

$$\frac{d^2T_i}{dx_i^2} + \frac{Q_i}{KA\Delta x} = 0 \tag{3}$$

is used in each liquid crystal layer with the same thickness Δx, where A is the area of the sample, K the thermal conductivity of the material, and T_i and Q_i the temperature and the heat source at each layer, respectively. For each layer, the heat source is calculated from the optical propagation equation. As another new feature of this model, the cut-off reflectivity was set at 60% to allow for the effect of the defects in the sample (See the experimental section below). The reflectivity of the cholesteric liquid crystal for laser of wavelength $0.515\mu m$ as a function of ambient temperature is shown in Fig.3. The no-feedback curve corresponds to the case where the absorption is ignored. With absorption, the feedback mechanism becomes active and the reflectivity increases with increasing optical power. At high power, the curves shift in the low temperature direction due to optical heating.

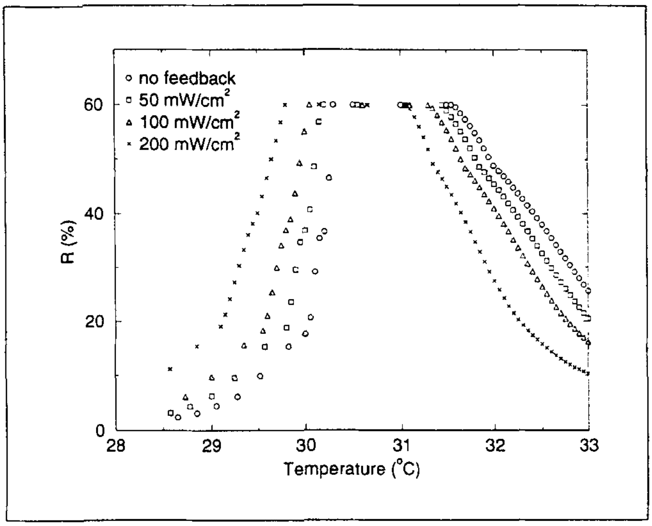

FIGURE 3: Calculated reflectivity R of the dissolved dye model as a function of ambient temperature.

EXPERIMENTS

In our experiments, the choice of the CLC was crucial. It had to have a T_{AC} above room temperature, a large birefringence to give a large reflectivity, and a pitch that varied from the infrared to the ultraviolet in a small temperature range. We used a mixture of the nematics[11] K24 and K30 (EM Chemicals) to get the correct T_{AC}. We used these materials for their high birefringence ($\Delta n \sim 0.2$). Three chiral dopants, R1011, R811, and CB15 (EM Chemicals) were used to achieve a short enough high temperature pitch. The dopants tend to lower T_{AC}, and each dopant has only a limited solubility in the nematic mixture. This is why we were led to this rather complex composition, in which the three dopants, all with right handed twisting power, work together to reduce the pitch of the CLC to the proper range. The resulting pitch of the CLC as a function of temperature is shown in Fig.4.

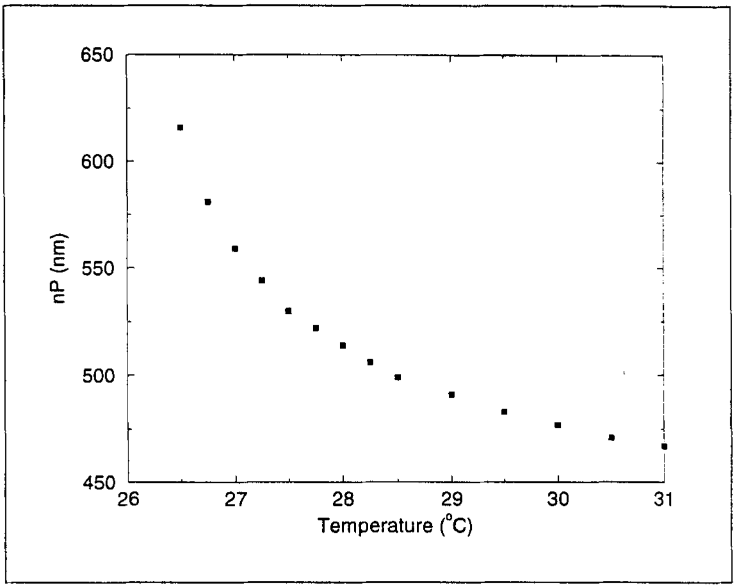

FIGURE 4: Temperature dependence of the helix pitch P times index of refraction n for the CLC used in our device.

For the first device, the CLC was sandwiched between two glass substrates separated by a $19\mu m$ Mylar spacer. One substrate was coated with polyimide and the other with an orange dye, covered with a layer of UV-cured Norland optical adhesive, to isolate it chemically from the CLC layer. Both substrates were then rubbed to induce homogeneous alignment of the CLC molecules at the surface. This helps keep the helix oriented with its pitch axis normal to the plane of the device, which is necessary for achieving high reflectivity. The device was placed in a temperature controlled oven to set the ambient temperature, determining the pitch of the helical structure in the absence of incident optical power.

An Argon ion laser (Spectra Physics model 165) with linear polarizer and quarter wave plate, produced an intense circularly polarized light source. We used a power meter to monitor the reflected light intensity from the device at near normal incidence.

Results of our experiments with the layered device are shown in Fig.5. We plot reflection coefficient as a function of the ambient temperature of the device. This is qualitatively similar to the plots to reflectivity *vs.* helix pitch in Fig.2, for the model calculations, since pitch decreases with increasing temperature. At the lowest power, the reflection coefficient is essentially that of the CLC with no heating or feedback effect. As power is raised, the reflection coefficient on the low temperature side of the curves increases, just as seen in the model calculations. On the high temperature side, the reflection coefficient at high power drops suddenly below its low power level, indicating the positive feedback that makes the device unstable in this region.

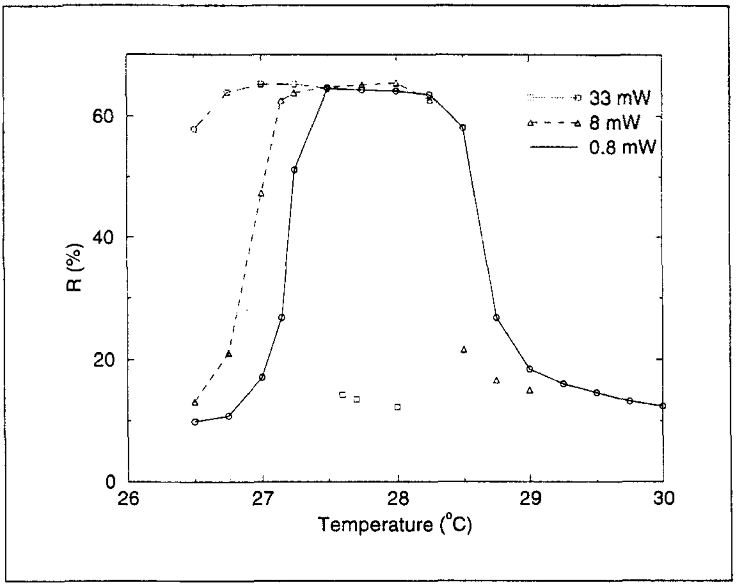

FIGURE 5: Reflectivity of the two layer device as a function of ambient temperature for three incident laser powers.

In Fig.6, we show the increase of reflectivity with power at constant ambient temperature, the essential desired behavior for the smart reflector.

Comparing the experiments with the model calculations, we see several significant differences. First, the reflection coefficient never reaches 100%. This is because the CLC layer is not perfect. There are many disclinations distributed through the CLC layer that scatter light, so the reflection coefficient is less than one in the plateau region of the curves in Fig.5. Part of the scattered light is back scattered, but some of it is forward scattered, so it reaches the absorbing layer and generates heat, even in the plateau region. This means that the feedback effect does not go to zero at the beginning of the plateau region, so the beginning of the plateau shifts to lower temperature at high power. This could not happen in the model calculations. Moreover, as temperature is increased, for high incident power, the sample overheats due to transmitted light, even in the plateau region. This results in a sudden drop in the reflection coefficient at some point in the plateau region, as the positive feedback effect drives the sample to higher temperature. This effect is made more evident because the laser power can increase the temperature of the whole CLC layer, rather than just creating a temperature gradient. In the model calculations, the temperature at the front surface of the CLC layer was held at the ambient level, making it more stable than the real device.

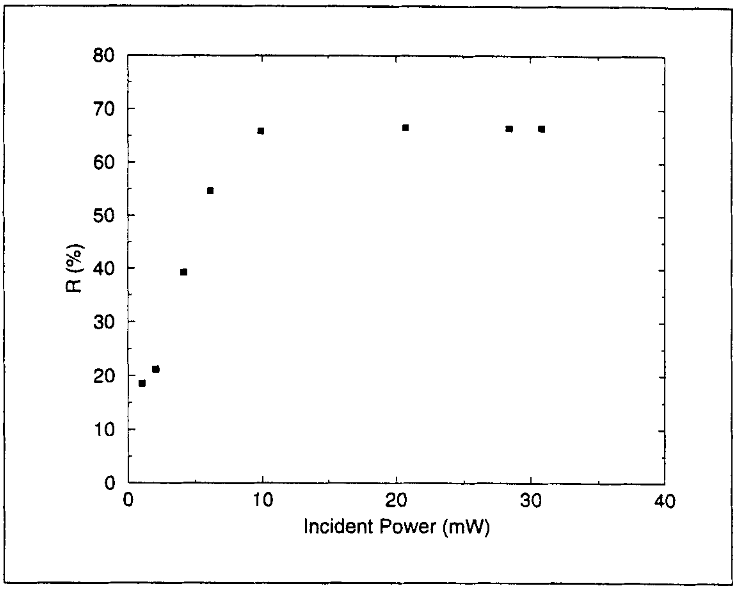

FIGURE 6: Reflectivity of the two layer device as a function of laser power at an ambient temperature of 27°C.

For the second realization of the device, a dichroic dye is dissolved in the liquid crystal mixture. The dichroic dye, which has an elongated structure similar to liquid crystal molecules, has been studied and used in liquid crystal applications for many years. Because of the similar shapes, all the molecules are compatible with each other, and the dye molecules are expected to follow the alignment of liquid crystal molecules, a relationship known as the guest-host effect. Dichroic dye D37 (2.7%,EM Chemicals) is put in the liquid crystal material as the absorptive component. The rest of the sample preparation is similar to the first device.

Results of our experiments on this device are shown in Fig.7. At the lowest power, the reflection coefficient is essentially similar to that of the CLC with no heating or feedback effect. As power is raised, the reflection coefficient on the low temperature side of the curves increases, which indicates the negative feedback that stabilizes the system. On the high temperature side, the reflection coefficient at high power drops suddenly below its low power level, indicating the positive feedback that makes the device unstable in this region. The reflectivity is generally low because light is always absorbed by the dye molecules. Besides that, there are many disclinations distributed throughout the CLC layer which scatter light. Notice that the maximum reflectivity at high power is a little bit larger than that at the lowest power. The reason might be that at high power, the heat can shorten the pitch length slowly enough to avoid creating disclination lines; at the lowest power, the pitch length is shortened by adjusting the ambient temperature, thus causing more defects. It may also be that at high power, the optical **E** field has a significant aligning effect.

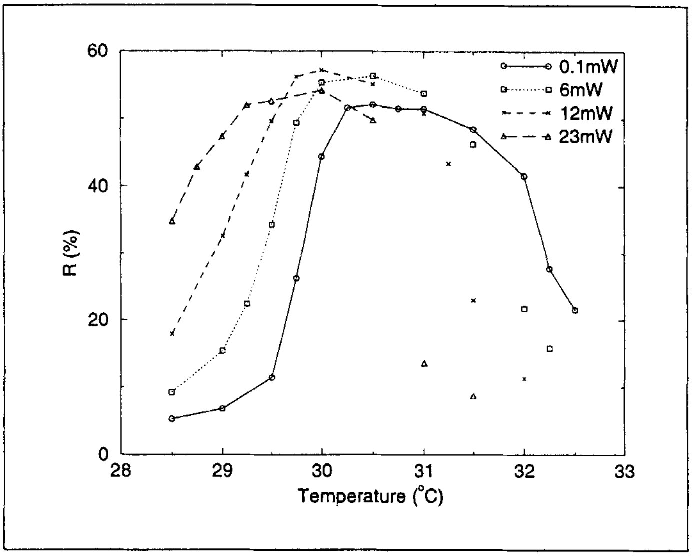

FIGURE 7: Reflectivity of the dissolved dye device as a function of ambient temperature for four incident laser powers.

The reflectivity as a function of incident power at fixed ambient temperature is shown in Fig.8. Here it is clear that the reflectivity of the device can increase with the increasing incident power, and then reach its maximum. The slope of this reflectivity change depends on the concentration of dye and $|\frac{\Delta P}{\Delta T}|$.

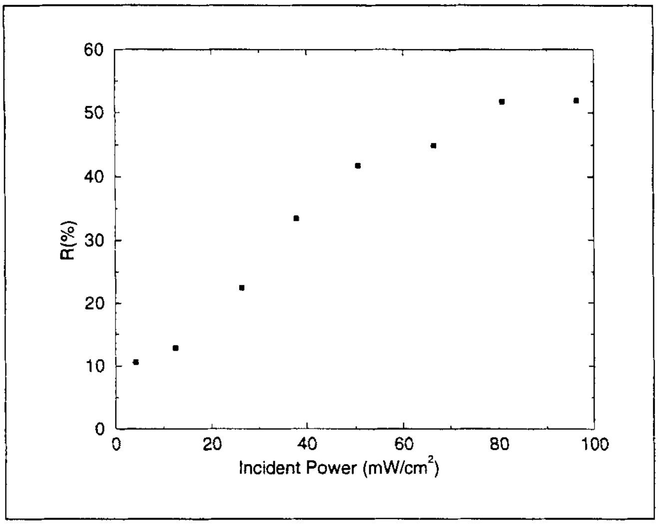

FIGURE 8: Reflectivity of the dissolved dye device as a function of laser power at a fixed ambient temperature.

We have also undertaken experiments on the response time of these devices. In general, because of the thermal part of the control process, the response time can be governed by the thermal mass of the system. To minimize this effect, we constructed devices on thin Mylar films, rather than glass slides. We found a modest decrease in response time, not proportional to the change in thernal mass. Typical response times are indicated in Fig.9. We conclude that the temperature change is not the limiting step in the response, but rather the change in the helix pitch, which is achieved by the motion of dislocations in the helix. The rate of change of helix pitch is proportional to the density and the mobility of the dislocations, which are for the most part nematic disclinations, each changing the twist in the cell by π as it sweeps across an area. Thus, although these defects limit the reflectivity of the device by scattering light, they also play an important role in controlling the response speed of the device. Research on the precise mechanism of the device response, and attemps to control it or circumvent the role of the disclinations, is under way.

In summary, we have demonstrated the liquid crystal smart reflector, in which the reflectivity is controlled, via a negative feedback mechanism, by the intensity of the incident light. The experimental results we have obtained are in good qualitative agreement with model calculations of the behavior of two realizations of the device. We can attribute important limitations of the device to the presence of defects in its structure. For a temperature of 27°C, an optical intensity of about 50 mW/cm^2 was sufficient to drive the two layer device to its maximum reflectivity. For reference, this is within the power range of incident sunlight.

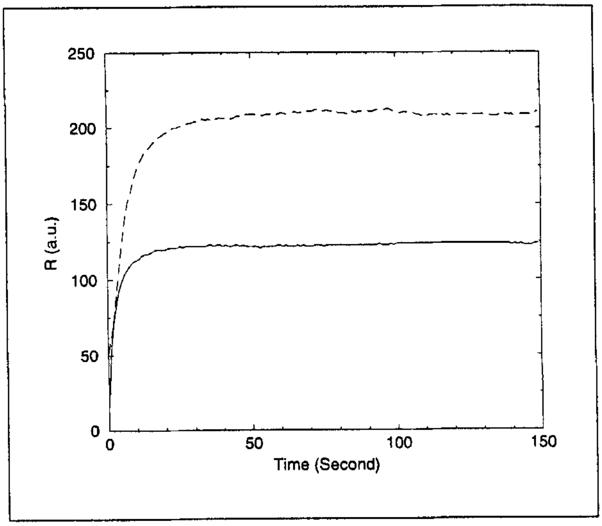

FIGURE 9: Reflectivity (in arbitrary units) as a function of time, for a thin film reflector, in response to step functions in light intensity.

For further development of this device, especially for a wide range of possible applications, one must address the questions of broad band reflection, and reflection of unpolarized light. Another important goal is removing the dependence of the system response on temperature, in fact finding a completely different mechanism for the change of helix pitch, not involving the cholesteric - smectic phase transition. We are exploring the use of dyes that interact directly with the torsion of the helix. All these issues are being pursued in ongoing research.

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