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Field Induced Unwinding of Cholesteric and SmC* Helix: Expectations and Reality

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Introduction. Due to a periodic helical structure, cholesteric (CLC) and SmC* liquid crystals represent convenient models for studying optical properties of 1D photonic bandgap systems, particularly lasing effects [1,2]. Their stop-band corresponds to the Bragg reflection band whose spectral position and width are determined by helical pitch P_0 and principal refractive indices $n_{\|}$, $n_{\perp}$. Due to soft nature of these materials such factors as chemical dopants, temperature, UV light, mechanical stress, etc. enable tuning P_0 and, consequently, the position of laser emission over a wide spectral range. Moreover, both theoretical calculations and early experimental observations [3-5] have shown possibility for the tunability of the helical pitch by an external electric or magnetic fields. However, despite numerous publications on lasing in CLC, to our knowledge, there is no report on field induced laser line tuning. On the contrary, as shown in [6] the electric field applied to a CLC structure only suppressed the lasing line without spectral shift. As to the SmC* phase, spectral tuning the lasing emission in a limited range was recently reported by Yoshino's group [2].

The reason for such a disappointing situation with CLC is very simple: despite the fact that field unwinding of the helix is thermodynamically profitable there is a strong topological limitation on the unwinding process. It is easily seen from Fig.1. In zero field we have a helical structure of the director **n**. We assume that the helix is either infinite or limited by two boundaries with infinitely weak azimuthal anchoring at least at one of them. Therefore, there is no confinement, which would prevent a free rotation of the non-anchored director at that boundary.

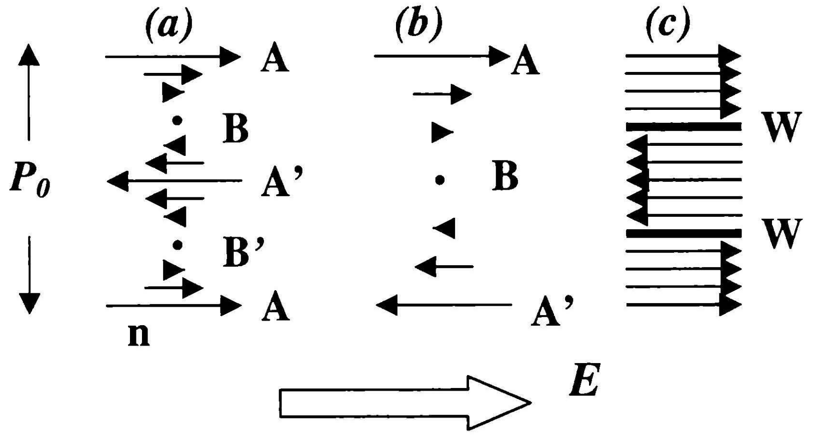

Fig.1. Field behaviour of a cholesteric helix (ε_a >0).
a) zero-field structure (the helical axis **h** is vertical)
b) unfavorable structure with a larger field induced pitch
c) Favorable wall structure with unchanged pitch

Now, imagine that we apply an electric field **E**⊥**h** to structure (*a*) with equilibrium pitch P_0 and want to increase the pitch twice, $P_E=2P_0$, as shown in sketch (*b*). To do this we must turn the director from the orientational state A' (**n**||**E**, favorable due to positive dielectric anisotropy ε_a assumed) to unfavorable state B, where **n**⊥**E.** Similarly, the director from the initial state A (at the bottom) must make a full turn against the field to new state A' and this situation takes place along the whole helix. In reality, due to this topological limitation, structure (c) forms with favorable orientation of the director everywhere. The positions of the walls separating areas with **n** differed by $\Delta\phi=\pi$ are fixed and, of course, the energy of structure (c) with the initial pitch P_0 is larger than the energy of more profitable structures with an enhanced pitch. The same consideration is valid for the SmC* but in that case $\Delta\phi=2\pi$.

Calculations. Our numerical modelling confirms this picture. The corresponding techniques have been described earlier [7]. Figs.2 and 3 show the calculated distribution of the azimuthal angle φ and the optical transmission T for the planar cholesteric structure of thickness d=10P_0. Both the zenithal and azimuthal anchoring strength at the first substrate is strong, $W_{z1}=W_{a1}$=0.1 mJ/m^2. At the second substrate W_{z2} is also that strong but the azimuthal energy is negligibly small $W_{a2}=10^{-4}$ mJ/m^2. This provides easy sliding of the director along the second substrate. The voltage is applied between in-plane interdigitated electrodes with a gap of l=20 μm.

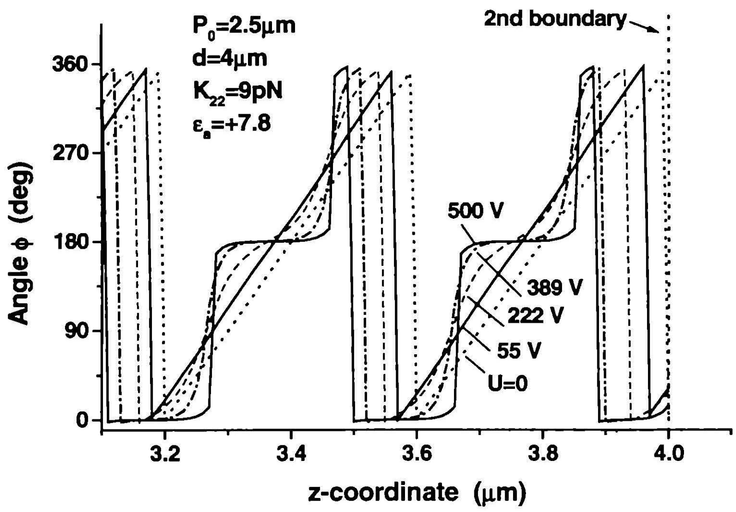

Fig.2. Director azimuth angle ϕ for the last two periods of the helix adjacent to the second boundary. Angle ϕ repeatedly increases from 0 to 360° within each period P_0. The in-plane voltage is applied to the helix, $E\perp z$. The total number of periods (10) between the two boundaries (over cell thickness d=4 μm) remains the same. In the middle of each period the director is progressively reoriented along the field (ϕ→π).

From Fig.3 it is clear that with increasing field the Bragg minimum is only slightly shifted to *shorter* wavelengths due to a distortion of the helix seen in Fig.2 and then disappeared at the field of 25V/μm. This field is considerably larger than the critical field calculated from the free energy approach [3,4] for the infinite helix with parameters of our model, $E_c= (\pi^2/P_0)(4\pi K/\varepsilon_a)^{1/2} \approx 7$V/μm. In fact, there is no distinct criterion for unwinding the planar helical texture (see Fig.2) other than the optical one (Fig.3). Our modelling, however, shows that, after unwinding, upon decreasing voltage, the helix forms at a lower field, much closer to E_c i.e. the whole process manifests a remarkable hysteresis. A variation of the cell thickness d does not influence our main result, which seems to be valid for the infinite helix as well.

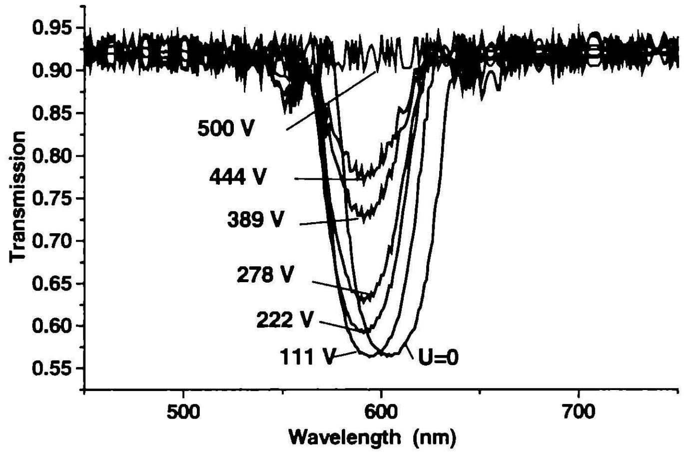

Fig.4. Calculated optical transmission spectra of the planar cholesteric texture as functions of the electric voltage applied. The "optical threshold" for helix unwinding is between 450 and 500V (unpolarized light, $n_{\|}$=1.550, $n_{\perp}$=1.474, for other parameters see the text and Fig.3).

Experiment. In reality, other processes such as the formation of defects result in the transition from the planar helical texture to the uniform nematic one. Fig.4 shows the result of our experiment specially carried out to check the results of the modelling. We used a chiral mixture MLC6601 + 21.7% ZLI-811 (both from Merck) with P_0 =0.42μm ($\partial P_0/\partial T$<0), $\varepsilon_a \approx 6$, $K_{22} \approx 6$pN, $n_{\|} \approx 1.55$, $n_{\perp} \approx 1.47$ and expected $E_c = (\pi^2/P_0)(4\pi K/\varepsilon_a)^{1/2} \approx 7$V/μm. The 20μm wide chromium stripes with a gap l=20μm in between were deposited on the bottom substrate. These interdigitated electrodes were covered by polyimide but not rubbed to reduce the azimuthal anchoring energy W_{a2} to a minimum. The top glass substrate was covered by polyimide and rubbed to provide good quality of the planar helical texture. The gap between glasses (d=12μm) was filled by the mixture in the isotropic phase. To avoid any thermal and hydrodynamic processes an optimum frequency of an external field was chosen to be 500Hz, a voltage U(rms) was varied from 0 to 120 V.

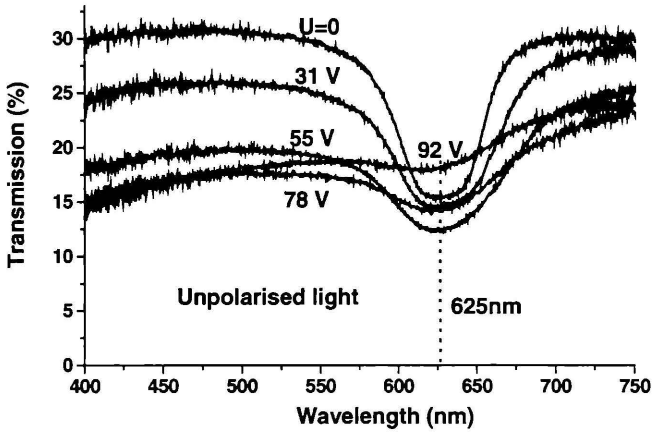

Fig.4. Experimental transmission spectra of the planar cholesteric texture as function of the external field (unpolarised light). The absolute value of the transmission is reduced by the system of the non-transparent interdigitated electrodes. Note a small shift of the Bragg minimum to *shorter* wavelengths before its complete disappearance at U≈120 V.

As seen from Fig.4, with increasing field the Bragg minimum broadens and finally disappears. In this experiment, the voltage of the full disappearance of the Bragg reflection (about 120V) approximately corresponds to the estimated critical field E_c. However, again no shift to longer wavelengths is seen, which would be indicative of helix unwinding. On the contrary, there is very small shift to shorter wavelengths predicted by modelling. Under a microscope the field induced defect grid is seen with the voltage exceeding 70 V, which can be erased only by some texture shearing. This defect structure is mediating the cholesteric-nematic transition.

In conclusion, we insist that, due to topological limitations, the ideal planar cholesteric or SmC* structure, even infinite, cannot be unwound continuously by transverse electric or magnetic field. Of course, the strong field always unwinds the helix, however we believe that in all successful experiments (in the sense of continuous process) helix unwinding has been mediated by defects (fingerprint texture in [4,8], Grandjean in [6], probably some other defects in [2]). In paper [9] the planar texture seems to be also non-ideal (with some random tilt of the helical axes) and the field could improve it with a subsequent slight shift of the Bragg minimum to longer wavelengths.

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