Bistable cholesteric liquid crystal displays (LCDs) are characterized by high driving voltages. This makes the application of cholesteric LCDs to high information displays difficult. In this study we examine the possibility of reducing the unwinding voltage by partial destruction of the order parameter, which was accomplished by the use of additives to the liquid crystal. In addition, we also examined the physics of adding such dopants by fitting the data to empirical theories.

Helix unwinding of cholesteric liquid crystals was first studied theoretically by de Gennes and Prost and by Meyer. Experiments on long-pitch cholesteric liquid crystals subsequently confirmed the predictions that the pitch diverged logarithmically at the critical field and the cholesteric–nematic phase transition was observed. The influence of the anchoring energy and the cell thickness on the helix unwinding process was also revealed in the cano-Grandjean disclination experiment. The minimization of free energy favors the nematic phase if the thickness is less than the unperturbed pitch. For the parallel-plates configuration, the unwinding actually occurs below the critical d/p ratio. Above this ratio, the cholesteric will develop a periodic pattern or isolated fingers. At an ac electric field, the problem becomes complicated since ion segregation cannot be neglected, especially for liquid crystals of negative dielectric anisotropy. The critical voltage exhibits a dispersive characteristic in the frequency domain and it is lower than the unperturbed pitch. For the parallel-plates configuration, the unwinding actually occurs below the critical d/p ratio. Above this ratio, the cholesteric will develop a periodic pattern or isolated fingers. At an ac electric field, the problem becomes complicated since ion segregation cannot be neglected, especially for liquid crystals of negative dielectric anisotropy. The critical voltage exhibits a dispersive characteristic in the frequency domain and it is lower than the unperturbed pitch.

In this letter, we show that, contrary to Baesseler et al.’s results, the helix unwinding voltage saturates at high frequency, with a smooth maximum at several hundred Hz. This characteristic cannot be simply extended by Hurault’s treatment. The spectral reflectivity and the frequency dependence of the unwinding field were studied as a function of nonmesogenic dopant concentration. We shall use de Gennes and Prost’s result adapted for the electric counterpart, and the corresponding dc unwinding field is given by

$$E_U(0) = \frac{\pi^2}{p_0} \sqrt{\frac{4\pi k_{22}}{\Delta \epsilon}} \ , \quad (2)$$

where $p_0$ is the unperturbed pitch, $k_{22}$ is the twist elastic constant, and $\Delta \epsilon$ is the dielectric anisotropy. Since the space charge effects are central to the electrohydrodynamic instabilities and the helix unwinding, the following empirical relation that has the Lorentzian term is suggested to fit the experimental data:

$$V_U(\omega) = \frac{V_U(0)}{\zeta} \left( \frac{\zeta + \omega^2 \tau^2}{1 + \omega^2 \tau^2} + \frac{\omega \kappa}{\sqrt{1 + \omega^2 \kappa^2}} \sqrt{\frac{\zeta + \omega^2 \tau^2}{1 + \omega^2 \tau^2}} \right) , \quad (3)$$

where $\zeta$ is a dimensionless quantity and it depends on the material properties of the liquid crystal mixture whereas $\tau$ and $\kappa$ are empirical time constants associated with the unwinding. These are consistent with the hydrodynamic relaxation of the helix unwinding at which at least two time constants are necessary for the experimental fitting. Since the pitch is independent of the order parameter, the elastic constant and dielectric anisotropy expressed in order parameter $S$ are

$$k_{22} = L_1 S^2 , \quad (4)$$

$$\Delta \epsilon = \Delta \epsilon_{max} S , \quad (4)$$

where $L_1$ is a constant determined by the Landau–de Gennes equation and $\Delta \epsilon_{max}$ is the anisotropy at the perfect alignment stemming from the polarizability of molecules. They do not depend on the driving ac frequency. By combining Eqs. (2) and (4), the dc unwinding field becomes
The effect of varying order parameter on the helix unwinding voltage was studied experimentally using standard sample cells. The LC cell was made with un rubbed polyimide, which was spin coated on ITO glass with a pretilt of about 2°. The cell gap was approximately 4 μm. The undoped cholesteric mixture was made by dissolving 24.8 wt % of benzophenone from Merck. Benzoylbenzene (benzophenone) of 99% purity was mixed with the cholesteric as the dopant.

First, the effect of the dopant on the reflectance spectrum was measured. For this measurement, the doped cholesteric cell was aligned at an oblique angle to the incident rays. A commercial xenon lamp system with good collimation and a PR650 spectrophotometer were used. The spectral data were collected at a small angle offset from the principal reflection direction. This was important in order to minimize the effect of Fresnel reflections at air–glass interfaces. The incident angle was about 10°. A high voltage pulse at 1 kHz was used to reset the samples to the planar state first. A long delay time was allowed to make sure that there was no slow long time constant relaxation before the measurement was taken.

The results are plotted in Fig. 1. It can be seen that the reflection peak for the undoped case is at about 550 nm. This peak shifts to shorter wavelengths as the doping concentration increases. The reflectivity and full width at half maximum (FWHM) spectral width are also reduced. However, at about 5% doping concentration, the reflectivity ceases to change before it drops abruptly at higher concentration. The peak in the latter case shows only a slight variation. The percentage change in the peak reflectance wavelength is given by

\[ E_U(0) = \frac{\pi^2}{p_0} \sqrt{\frac{4 \pi L_1}{\Delta \varepsilon_{\text{max}}}} \sqrt{S}. \]  

Equations (3) and (5) are the main results that we want to confirm experimentally. In addition to being physically interesting, Eq. (5) provides a means of reducing \( E_U \). One can reduce the helix unwinding voltage simply by reducing the order parameter. However, the order parameter should not be reduced so much that the liquid crystal property is also destroyed. So there should be an optimal value of \( S \).

Figure 2 shows the relative spectral shift as a function of doping concentrations. A systematic trend can be observed. On the other hand, since the variations of reflectivity and reflection peak follow a similar pattern from 0% to 5%, it can be deduced that the apparent pitch actually changes with the doping concentration. This implies that the average refractive index plays a minor role in comparison to the apparent pitch. Therefore, it is direct confirmation that the birefringence is influenced by the dopant.

The frequency dependence of the transmittance in the cross polarization geometry was also measured. A laser beam at 543.5 nm was used to monitor the transmittance every second as the ac voltage was gradually increased. The null point in transmittance was determined as the unwinding voltage and was confirmed under an optical microscope. The behavior of the cholesteric–nematic phase transition observed under the microscope at different frequencies was quite interesting. At low frequency, the distorted helical structures are observed as vibrating energetically as the field approaches the critical value. The helical axes lie effectively in plane. Helix unwinding begins at the spacers and the nematic phase grows from there. At several hundred Hz, the motion of domain boundaries becomes nonuniform. In this case, the helix unwinding undergoes two stages. The initial stage is similar to that at low frequency but the nematic phase is not homogeneous. Distorted helical structures that resemble an isolated fingerprint texture coexist and are unwound from the finger-like tips.

At high frequency, the helical structure vibration and the motion of domain boundaries are not noticeable. Stable topological textures such as closed-loop and isolated fingerprint-like textures coexist and are readily unwound as the field increases. Saturation in unwinding voltage was obvious at high frequency. The actual cause of this saturation and the smooth bump were not clear although we believed that it should be due to the ion segregation and the non-Newtonian viscous flow gradient in the deformed elastic me...
uum. Usually, they are significant at low and intermediate frequencies. At the unwinding voltage, the deformation and flow gradient are so large that the perturbation approaches breakdown and the interplay of these effects will make the frequency dependence intricate.

In Fig. 3, the rms unwinding voltage was plotted as a function of the ac field frequency with different concentrations of benzophenone. The lines (solid, dotted, and dashed) were theoretical fits using Eq. (3). It can be seen that the agreement with data and theory is good. The values of the parameters used in the fitting procedure are listed in Table I. The shapes of the curves in Fig. 3 are very similar. Physically, the reduction in the unwinding voltage can be explained as follows. The nonmesogenic dopant, such as benzophenone, which does not exhibit the nematic phase but has a rod-like structure, effectively screens the intermolecular coupling between liquid crystal molecules and thus reduces the long-range order. This effectiveness depends on the structural and chemical properties of the dopant. For benzophenone, it was shown that a small concentration of 4% could produce a sizeable change in the unwinding voltage.

The authors thank Professor Vladimir G. Chigrinov of Shubnikov Institute of Crystallography, Russian Academy of Sciences, Russia, for critical comments. This research was supported by the Hong Kong government Industry Department.