

New pulsatory instability in cholesteric mixtures

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(Received 8 October 1982)

A new pulsing electrohydrodynamic instability has been noticed in a compensated cholesteric mixture, as a "host" in which a cumylphenol azo compound was introduced as a "guest." The liquid-crystal sample was placed in a steady dc electric field. Observations of the microstructural aspects and measurements of the transmitted light were simultaneously performed. Pulsing instabilities of 0.02–0.05 Hz were revealed. Upon an increase of the electric field under constant temperature the frequency of the instabilities increased.

INTRODUCTION

Interest in the field of electrohydrodynamic instabilities in liquid crystals has increased in recent years. Besides the papers of the Orsay group^{1,2} others predicted the possibility of oscillatory electrohydrodynamic instabilities in nematics,^{3,4} smectics *A*, and cholesterics.⁵ The instabilities occur under either a temperature gradient^{4,5} or a steady dc electric field.³ Some of the theoretical predictions were experimentally proved.⁶

Concerning the influence of the dc electric field, Laidlaw³ established the conditions for the oscillatory instabilities to occur in nematics. Kapustin *et al.*,⁷ and Hirata and Tako⁸ have noticed that patterns of nematics oscillate coherently in a dc electric field.

In this paper we report observations of pulsing electrohydrodynamic instabilities in a compensated binary cholesteric mixture with a pleochroic dye as guest. To our knowledge, this is the first time pulsing instabilities are reported to occur under a steady dc electric field at constant temperature. Thus, we prove that recurrent instabilities induced by a steady dc electric field can also develop in mixtures of two cholesterics to which a dye has been added.⁹

For cholesterics the instabilities are experimentally found to recur at roughly 20 sec. As for the value of the electric field it is smaller than the critical value^{10,11} (which causes the cholesteric-to-nematic phase transition) irrespective of the temperature.

EXPERIMENT

The pulsing instabilities we discuss in the present paper have been recorded for the dye cumylphenol azo compound from paratoluidine dissolved in the host mixture. The dye was introduced as guest (1:100 weight ratio) in the cholesteric host mixture: cholesteryl chloride + cholesteryl laurate (65:35% weight ratio). We investigated the samples with the azo compound at constant temperature¹² and a dc steady electric field applied to the sample. The puls-

ing instabilities of electrohydrodynamic nature were found to be sudden changes of the microstructural array.

Figure 1 shows photographs of the microstructure of a sample for which the evolution in time of the pulsing instabilities was recorded. Before applying the electric field, the sample had a focal conic texture^{11,13} [Fig. 1(a)]. Sequences of "elongated" fingerprints appeared when the corresponding 30-V dc voltage was applied [Fig. 1(b)]. 30 V and 35°C were afterwards maintained constant. The fingerprint texture suddenly changed, the structure became strongly birefringent, and the visual field of the microscope was homogeneously lighted for a time interval of the order of hundreds of seconds [Fig. 1(c)]. After the flash of the structure appeared, during its relaxation, a focal conic texture with a preferential orientation was noticed again [Fig. 1(d)]. During the relaxation, the texture color varied continuously from yellow to blue. Before another flash appeared, elongated fingerprints were seen again and the phenomenon recurred.

In order to record the variation of the light intensity transmitted by the sample, a detection system, consisting of a photomultiplier, an amplifier, and a recorder, was attached to the polarizing microscope.

Figure 2 shows the variation of the light intensity transmitted by the sample, during the microstructural changes previously described (Fig. 1). Figures 2(a) and 2(b) show the variation of the light intensity transmitted at the temperatures of 25 and 47°C, starting at the moment of the application of the electric field ($t=0$). In Figs. 2(c) and 2(d), the variation of the light intensity transmitted at 35°C is plotted starting at $t=0$ [Fig. 2(c)] and at 1 h 15 min after the moment of application of the same voltage, 30 V [Fig. 2(d)].

Figure 3(a) shows the transmission variations corresponding to pulsing instabilities recorded for 25, 30, 35, 40, 45, and 50 V. Each voltage value was kept constant for 1 h and the temperature was kept constant throughout. The mean value of the rate of

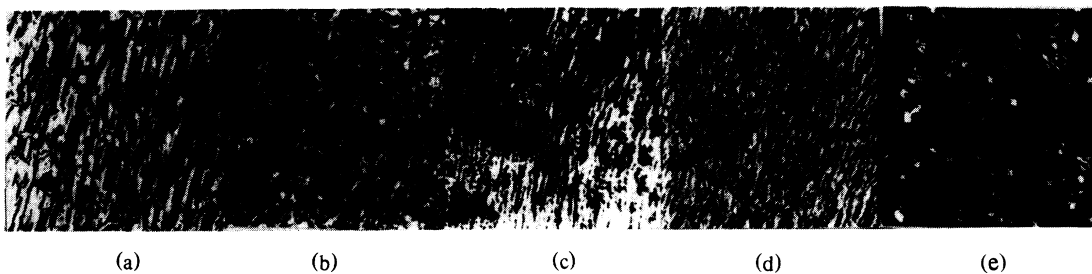


FIG. 1. Textures of the liquid crystal, ($A \perp P$), $t = 35^\circ\text{C}$. (a) Focal conic texture before applying the electric field. (b) Fingerprint texture; after setting $U = 30\text{ V}$ ($200\times$). (c) The birefringent texture during the flash; $U = 30\text{ V}$, ($200\times$). (d) Focal conic texture during relaxation; $U = 30\text{ V}$ ($200\times$). (e) The nematic droplets; $U = 80\text{ V}$ ($200\times$).

recurrence (reportedly also called period) of the instabilities as a function of the applied field is shown in Fig. 3(b). Table I gives the mean periods corresponding to the plots in Fig. 2.

Investigations on a series of samples of different thickness and with various types of dyes made us conclude that the cell thickness is the critical parameter on which the occurrence of this type of pulsing instability depends.¹⁴ For thicknesses under $18\ \mu\text{m}$ and above $30\ \mu\text{m}$ the nematization process takes place as already described in the literature.^{11,13} The pulsing instabilities reported in this paper are very in-

tense for samples with thicknesses about $24\ \mu\text{m}$. The instabilities were also noticed in $20\text{-}\mu\text{m}$ samples containing the same host mixture with xylenazo- β -naphthol as a guest, with guest:host weight ratios 1:100 and 1:200, respectively.¹⁵ For a $24\text{-}\mu\text{m}$ -thick cell a much fainter recurring flash was also noticed in the host mixture without dye.

The values of the applied electric field at which the pulsing effect takes place are always under the critical ones. We observed that this effect is produced when

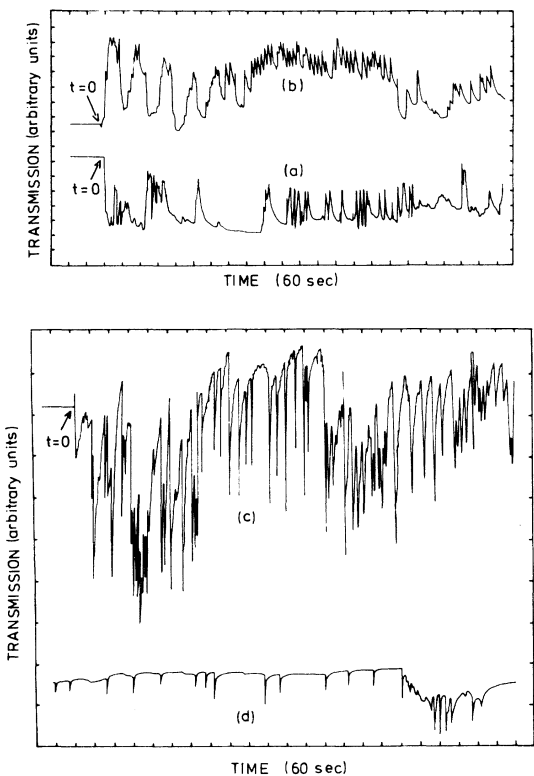


FIG. 2. The recordings of transmission in time ($A \perp P$): (a) $t = 23^\circ\text{C}$, $U = 20\text{ V}$; (b) $t = 47^\circ\text{C}$, $U = 40\text{ V}$; (c) and (d) $t = 35^\circ\text{C}$, $U = 30\text{ V}$.

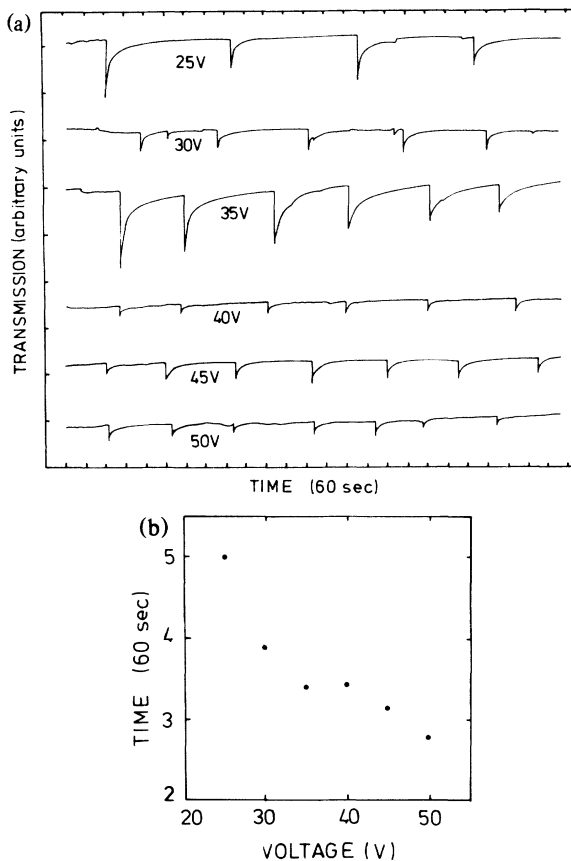


FIG. 3. (a) The recordings of transmission in time ($A \perp P$), $t = 35^\circ\text{C}$. (b) The mean value of the effect period vs the applied field.

TABLE I. Mean value of the rate of recurrence of the instabilities as a function of applied field.

t (°C)	U (V)	Mean period (sec)
23	20	21
35	30	37
47	40	50

Malta-cross-shaped nematic droplets^{16,17} were seen on the background of the homeotropic nematic texture [Fig. 1(e)]. These nematic droplets disappear only when applying an electric field much higher than the critical one. It has been noticed that the position and the orientation of the droplets do not change in cells with a thickness smaller than 18 μm or greater than 30 μm . The pulsing instabilities were obtained only in 20–25- μm samples. In these, the nematic droplets oscillate between, respectively, the parallel and the perpendicular positions with respect to the plane of electrodes. In 25–30- μm samples the droplets rotate around an arbitrary axis, appearing and disappearing in the visual field, when the applied field is higher than the critical one. For lower-than-critical electric fields, whirls are produced round the nematic droplet, the mass of the liquid crystal is set in a turbulence motion, and a dynamic scattering mode is established.^{15,18}

We consider that the introduction of some pleochroic dyes modifies the distribution of electric charges in the liquid crystal. In the presence of the electric field the system undergoes recurrent (quasi-periodic) changes of structure which also point to processes of transfer of energy; the nematic droplets are a support for the coherently oscillating system.

The temperature dependence of the phenomenon results in the decrease of the mean period when the temperature increases.

When the pulsing instabilities were measured for a long time in progressively higher electric fields, a stability of the investigated phenomenon was observed [Fig. 3(a)]. The effect period decreases with the increase of the applied field, as it was theoretically predicted for oscillatory instabilities in nematics in dc steady electric fields.³

In conclusion, we can state that the recurrent (pulsing) instabilities in compensated cholesterics depend mainly on the thickness of samples; the applied electric field and the temperature determine only the amplitude and the evolution in time of this effect. The frequency of pulsing instabilities in a steady dc electric field of $\geq 10^6$ V/m was measured to a range between 0.02 and 0.05 sec^{-1} for the 20–25- μm samples. The measured frequency is of the same order of magnitude as the calculated value, $\omega = 0.05 \text{ sec}^{-1}$, for the convective oscillations due to the temperature gradient $\Delta T = 1$ K/cm in cholesteric samples.⁵ But the thickness of our cells is quite different and, as we have stated, it conditions the occurrence of the pulsing instability.

The values of the electric fields which trigger the pulsing instabilities (10^6 V/m for cholesterics) are not far from other characteristic cholesterics values (the critical field, the field governing the storage effect and so on).

The recurrent pulsing instabilities reported here did not occur in the absence of the electric field.

We think that the occurrence of this type of pulsing instability in a steady dc electric field might be accounted for by the variety of stabilizing and/or destabilizing effects provided by the dielectric anisotropy and conduction anisotropy³ of compensated cholesterics in which the dyes are likely to modify these properties.

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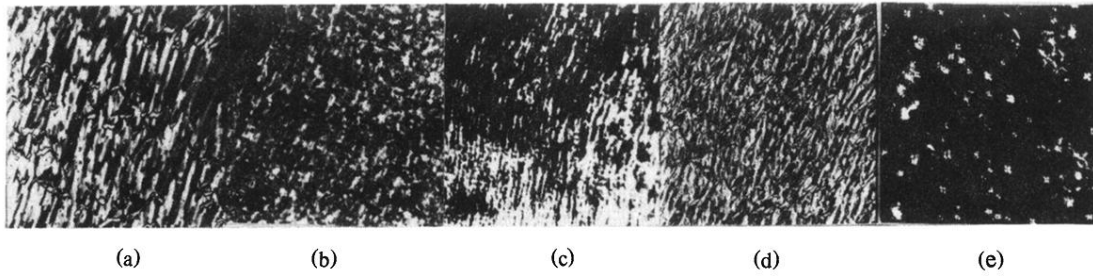


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