Hydrodynamic Instabilities in Nematic Liquids Under ac Electric Fields*

Orsay Liquid Crystal Group†
Laboratoire de Physique des Solides, 91 Orsay, France
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We present some measurements on the instability threshold of *p*-methoxy-*n*,*p*-benzilidene butylaniline (MBBA) under ac fields of frequencies 10-1000 Hz. The threshold curves, in both the low- and high-frequency regimes, are interpreted by a simple extension of the Helfrich theory to time-dependent phenomena.

Since the first observations by Williams[1] a large amount of work has been devoted to the electric instabilities of nematics. It is now apparent that the onset of instability involves many physical effects: (A) With dc electric fields, recent observations[2,3] indicate that (at least for the current samples and electrodes) unipolar charge injection must play a role.[4,5] (B) With ac excitation (as soon as the period is shorter than the transit time of injected carriers), unipolar injection becomes negligible.[6] We show in this Letter that the onset of instability under ac fields may be interpreted entirely in terms of the Carr-Helfrich model,[7,8] i.e., in terms of the conductance anisotropy of the nematic.

Consider a nematic slab parallel to the x-y plane under a field $E = E_M \cos\omega t$ applied along z.[9] The dielectric anisotropy is assumed negative ($\epsilon_\parallel - \epsilon_\perp = \epsilon_a < 0$) and the conductance anisotropy positive ($\sigma_\parallel - \sigma_\perp = \sigma_a > 0$). In the unperturbed state the molecules lie along x; in the perturbed state we assume that they lie in the x-z plane making a small angle $\varphi(x)$ with x. The state variables are the charge density q and the local (bending) curvature $\psi = \partial\varphi/\partial x$. They are ruled by the equations[10]

$$\dot{q} + q/\tau + \sigma_H \psi E(t) = 0, \tag{1}$$

$$\dot{\psi} + \psi/T(t) + qE(t)/\eta = 0, \tag{2}$$

where $\tau = \epsilon_\parallel/4\pi\sigma_\parallel$ is a dielectric relaxation time, while T is the response time for the molecular alignment:

$$\frac{1}{T(t)} = \left(\frac{1}{\gamma_1} + \frac{1}{\eta_0}\right)\left[-\frac{\epsilon_\perp}{\epsilon_\parallel}\frac{\epsilon_a}{4\pi}E^2 + K_{33}k^2\right]. \tag{3}$$

In Eq. (1), σ_H is $\sigma_\parallel(\epsilon_\perp/\epsilon_\parallel - \sigma_\perp/\sigma_\parallel)$; η, η_0, and γ_1 in Eqs. (2) and (3) are viscosity coefficients, which are defined in Ref. 10 in terms of the Ericksen-Leslie coefficients; K_{33} is the bending elastic constant, and k the wave vector of the perturbation.

There are two regimes of importance:

(1) If $T \gg \tau$ (low electric fields) the frequencies ω of interest are low ($\omega\tau \lesssim 1$). Thus $\omega T \gg 1$ and, from Eq. (2), the only important Fourier component of ψ is at 0 frequency. Then, from Eq. (1), $q(t)$ is modulated at the frequency ω. We call this the "conducting regime." This creates a pattern of hydrodynamical cellular flow which, in visual observation, corresponds to the Williams domains.[1] The onset of hydrodynamic instability occurs at a field[11]

$$\bar{E}^2 = \tfrac{1}{2}E_M^2 = E_0^2\frac{1+\omega^2\tau^2}{\zeta^2 - (1+\omega^2\tau^2)}, \tag{4}$$

where $E_0/(\zeta^2-1)^{1/2}$ is the Helfrich static threshold[8] and ζ is defined by

$$\zeta^2 = \left[1 - \frac{\epsilon_\parallel}{\epsilon_a}\frac{1}{1+\eta_0/\gamma_1}\right]\left[1 - \frac{\sigma_\perp}{\sigma_\parallel}\frac{\epsilon_\parallel}{\epsilon_\perp}\right]. \tag{5}$$

Note that E_0 and hence $\bar{E}$ are inversely proportional to the sample thickness[8] e: Most experimental studies, including ours, do show that the voltage threshold V_{th} is independent of e as $\omega \to 0$.

Eq. (4) displays a cutoff frequency,

$$\omega_c = (\zeta^2-1)^{1/2}/\tau, \tag{6}$$

above which the approximation $\omega T \gg 1$ breaks down. The proportionality between ω_c and σ ($1/\tau$ is proportional to σ) has already been checked experimentally.[12,13] A comparison between Eq. (4) and our data on MBBA samples is shown in Fig. 1.

(2) If $\omega \gg \omega_c$ (or $\omega \gg 1/\tau$) the charge q is no longer able to follow the excitation; then q is static, while ψ is modulated, with components at ω, 3ω, etc. This might be called the "dielectric" regime, or the "fast-turnoff" mode as in the initial report describing it.[13] Indeed, the average relaxation rate $1/\bar{T}$ is here comparable with ω and the threshold condition can be shown to be

$$\omega\bar{T} = C(\zeta), \tag{7}$$

where $C(\zeta)$ is a dimensionless function.[14] Since the fields E are high, $1/\bar{T}$ is essentially propor-

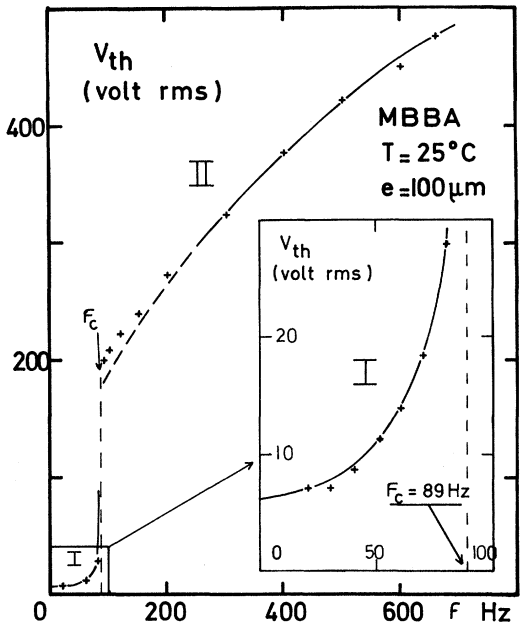

FIG. 1. Voltage threshold V_{th} of ac instabilities, versus the frequency f $(=\omega/2\pi)$ of the applied field. Region I, conducting regime (Williams striations); Region II, dielectric regime (fast turnoff mode of Heilmeier and Helfrich). Solid line: theory, from Ref. 10; $\zeta^2 = 4.5$ (see text).

tional to $\overline{E}^2$. Then Eq. (7) shows that the field threshold is proportional to $\omega^{1/2}$; this is confirmed by the data of Fig. 1. Equation (7) also shows that in this regime the field threshold is independent of sample thickness, in agreement with our measurements.

Optically, in the "dielectric" regime, the onset of instability is displayed by the occurrence of parallel striations ("chevrons") of interval much shorter (typically ~3 μm) than the sample thickness. This may also be explained in terms of Eqs. (1), (2), and (3): Although it is more expensive energetically to produce a distortion of large wave vector k, it leads to an improved coupling because large k's lead to short response times T [see Eq. (3)]. A detailed discussion of these aspects and of the effects of a magnetic field on the chevron periodicity will be published separately.[3]

On the whole, we conclude that Eqs. (1) and (2) provid a reasonable description of the hydrodynamic instabilities of MBBA under electric fields. Experimental values of ζ (derived from our measurements[15] of ω_c, σ, and ϵ) are in the range $2.5 < \zeta < 5$. However certain difficulties must be mentioned: The optimal values of ζ (fitting curves such as Fig. 1) are smaller for the thinner samples (thickness <30 μm) than for the thicker ones (>100 μm). This might be connected with the following complications: (a) For thin samples the two regimes tend to merge and our analysis is not applicable. Also, boundary effects become more serious and residual injection may play a role. (b) For thick samples the time constants $\overline{T}$ are large in the conducting regime, and the optical detection of threshold becomes inaccurate. We are beginning some systematic work to clarify these points.

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†Theorist members involved in the present work: E. Dubois-Violette, P. G. de Gennes, and O. Parodi. Experimentalists: G. Durand, M. Veyssie, F. Rondelez, and L. Leger.

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[14]For $\zeta \gg 1$, $\psi(t)$ is essentially a simple sine wave and $C(\zeta) \simeq (\zeta^2-1)^{1/2}$. For $\zeta \sim 1$, $\psi(t)$ contains many harmonics and $C(\zeta)$ has been obtained by numerical methods. Experiments measuring $\psi(t)$ directly will be described in a later publication.

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