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## Walls in Nematics†

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**Abstract**—At the Freedericks transition of a nematic slab, the molecules can rotate by two different ways, giving rise to adjacent domains separated by a wall. We present observations on the structure and dynamics of these walls at magnetic fields above the transition.

The local distortion through the wall agrees with theoretical prediction. The ellipticity of a wall surrounding a closed domain allows to determine some elastic constant ratios. If the magnetic field is decreased to the Freedericks critical field, the thickness and the velocity of the walls are observed to diverge respectively as  $(H - H_c)^{-1/2}$  and  $(H - H_c)^{-1}$ . From the velocity of the walls we can deduce the twist viscosity  $\gamma_1$  in planar geometry, and a renormalized viscosity  $\gamma^*$  including backflow corrections in homeotropic geometry.

Usually, a nematic sample is made up of several domains, corresponding to different orientations of the long axis of the molecules.

The transition between each orientation can be achieved in a discontinuous way, by a common disclination line, or in some cases, by a progressive change in the orientation of the molecules, which takes place over distances much larger than the molecular length. That is what we shall call a wall.

Such walls were first discussed by Helfrich<sup>(1)</sup> for a change of  $\pi$  in the molecular orientation (quite similar to Bloch or Neel walls in a ferromagnet). More recently, the generation of  $\pi$  twist walls in a rotating nematic sample placed in a magnetic field was considered by de Gennes,<sup>(2)</sup> and observed by the present author.<sup>(3)</sup>

We describe here another simple way of nucleating walls in a nematic sample. We have already given some preliminary results on those walls.<sup>(4)</sup> In the present paper, a more complete set of experiments is presented and compared to theoretical predictions by F. Brochard.<sup>(5)</sup>

† Presented at the Fourth International Liquid Crystal Conference, Kent State University, August 21-25, 1972.

### 1. Nucleation and Observation of the Walls

If a nematic monocrystal, oriented with its molecules normal to the glass plates, is placed in a magnetic field larger than the Fredericks critical field :

$$H_c = \frac{\pi}{d} \sqrt{\frac{K_3}{\chi_a}}$$

( $d$  is the sample thickness,  $K_3$  the bend elastic constant and  $\chi_a$  the anisotropic part of the magnetic susceptibility), the molecules in the middle of the slab rotate to align parallel to the magnetic field.

Since the director  $\mathbf{n}$  is equivalent to  $-\mathbf{n}$ , this rotation can be achieved in two equivalent ways, corresponding to an angle  $(\mathbf{n}, \mathbf{n}_0) = \pm \theta_\infty$  (Fig. 1a). This degeneracy exists only if the magnetic field  $H$  is exactly normal to the plate. Inside the wall, the tilt angle goes continuously from  $-\theta_\infty$  to  $+\theta_\infty$ .

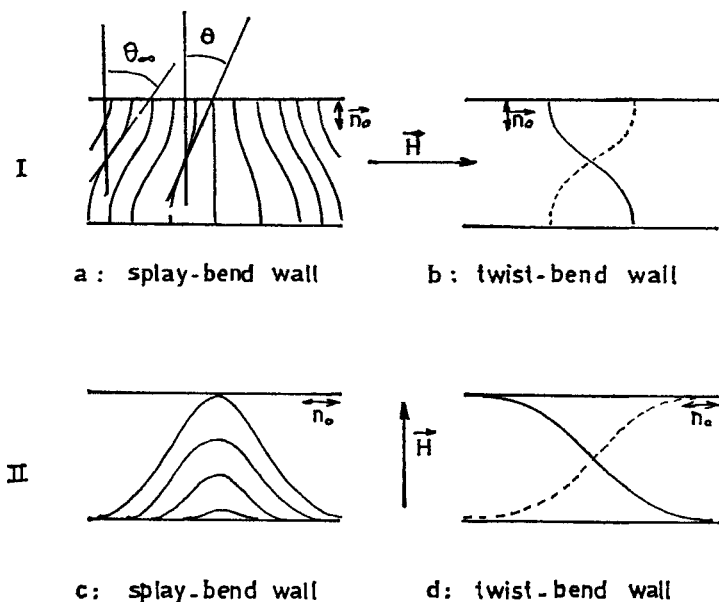


Figure 1. Schematic distortion of the molecular alignment through the wall for the two geometries we have studied.  $\mathbf{n}_0$  represent the direction of orientation of the molecules if the magnetic field  $\mathbf{H}$  is equal to 0.

Optically, this progressive distortion introduces a continuous variation of the birefringence of the slab, as can be seen on Fig. 2.

The index of refraction for a polarization normal to  $(\mathbf{H}, \mathbf{n}_o)$  is always equal to the ordinary index  $n_o$ . On the other hand, for a polarization in the plane  $(\mathbf{H}, \mathbf{n}_o)$  the effective index has a value  $n'_o$  (related to  $n_e$ ,  $n_o$  and  $\theta_\infty$ ) far from the wall and a value  $n_o$  in the middle of the wall. Between crossed polarizers, oriented at  $\pi/4$  with respect to  $\mathbf{H}$ , one can see birefringence fringes in the entire distorted area.

Similar walls can also be obtained by starting with  $\mathbf{n}_o$  parallel to the glass plates, and applying  $\mathbf{H}$  normal to the slab.

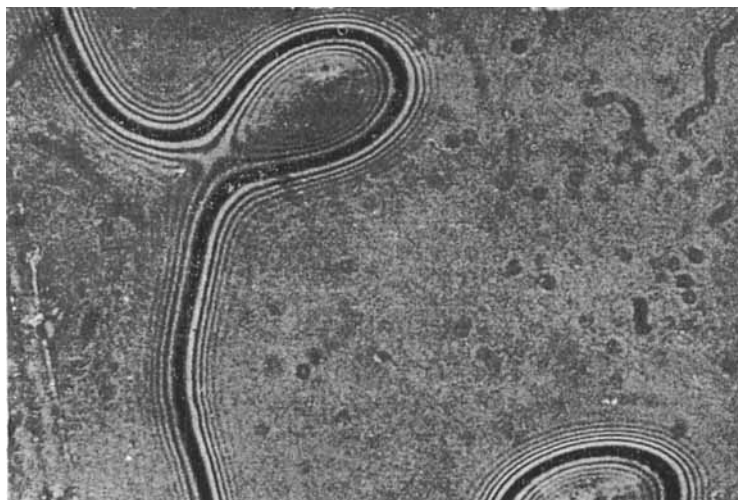


Figure 2. Typical view of a wall in geometry I, observed between crossed polarizers, at  $\pi/4$  with respect to  $\mathbf{H}$ , with parallel white light.

## 2. Static Behaviour of the Walls

As described by F. Brochard,<sup>(5)</sup> there are many possible situations, which are described on Fig. 1. One can distinguish between a twist-bend or a splay-bend wall as their thicknesses are respectively proportional to  $\sqrt{K_2}$  and  $\sqrt{K_1}$  for geometry I, or  $\sqrt{K_2}$  and  $\sqrt{K_3}$  for geometry II. ( $K_2$  is the twist elastic constant and  $K_1$  the splay elastic constant).

Closed domains are very often nucleated. The equilibrium shape

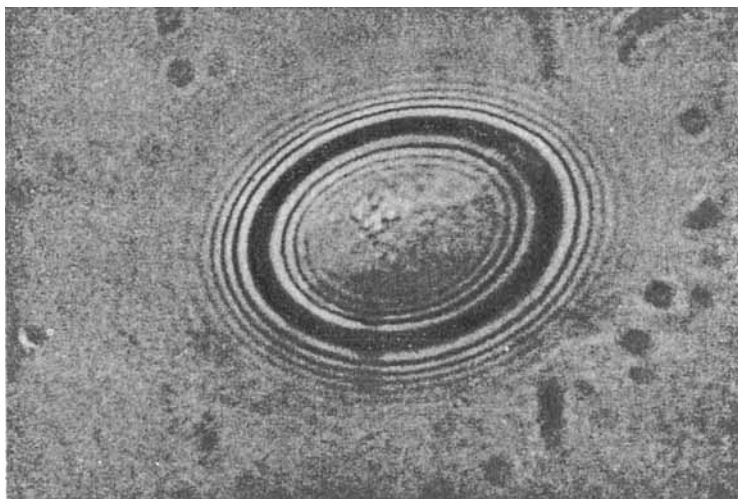


Figure 3. Equilibrium shape of a wall surrounding a closed domain. Such a ring spontaneously collapses, but, as long as the dimensions of the ellipse stay larger than the wall thickness, it remains an ellipse of fixed ellipticity.

of a wall surrounding a closed domain is elliptic (Fig. 3). The ellipticity gives a very simple evaluation of the ratio of some elastic constants: we have obtained, following the evaluations of Ref. 5,  $a/b = \sqrt{(K_1/K_2)} = 1.45$  in geometry I, and  $a/b = \sqrt{(K_3/K_2)} = 1.7$  in geometry II, for M.B.B.A. (methoxybenzylidene butylaniline) at room temperature.†

Reference 5 also predicts a divergence of the thickness of the wall, as the field  $H$  is decreased to  $H_c$ . Figure 4 gives an evidence for this divergence, in the case of a splay-bend wall (geometry I) observed with monochromatic light.

A plot of the distance between the two first birefringence fringes

† The M.B.B.A. we used had a transition temperature from nematic to isotropic of  $46.5^\circ\text{C}$ , before the preparation of the samples (vacuum distilled M.B.B.A.), which fell down to  $45^\circ \pm 0.5^\circ\text{C}$  after the samples had been prepared. The samples were completely sealed so that no further degradation could be observed during the experiments.

For geometry II, the boundary conditions were achieved by a conventional rubbing technique, while for geometry I, the glass plates were treated (after a very good chemical cleaning), with hexadecyl trimethyl ammonium bromide in solution in toluene, in a way quite similar to Haller's one.<sup>(10)</sup>

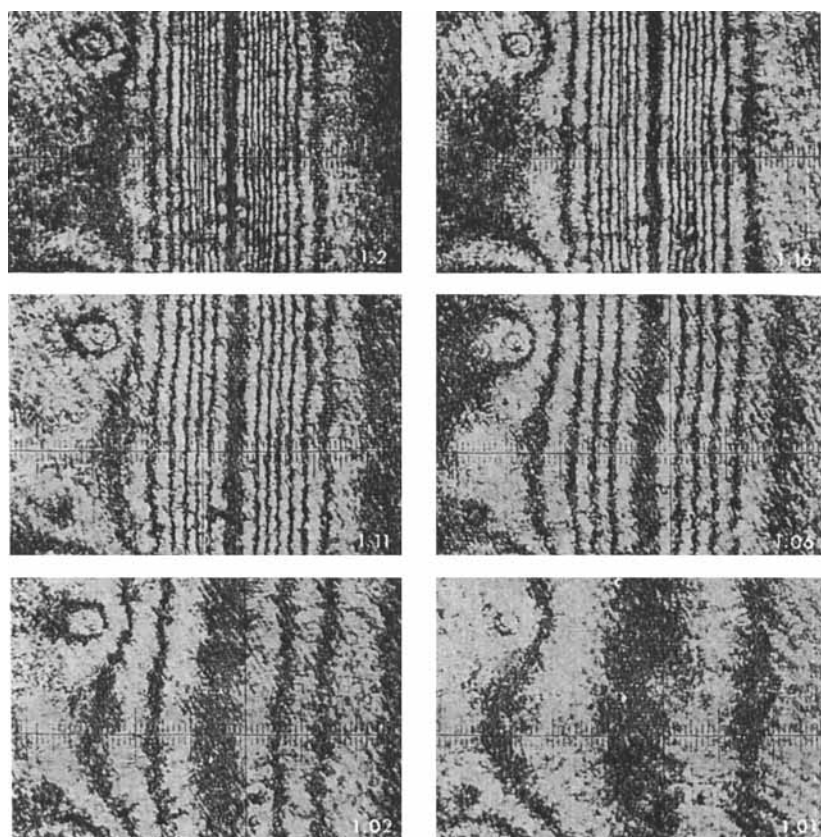


Figure 4. Divergence of a splay-bend wall thickness as  $H$  goes to the Fredericks critical field  $H_c$ , in geometry I. The wall is observed in monochromatic light ( $\lambda = 0.63 \mu\text{m}$ ) between crossed polarizers oriented at  $\pi/4$  with respect to the field.

versus the magnetic field is given on Fig. 5. The inset shows that the divergence is, as expected,<sup>(5)</sup> in  $\Delta H^{-1/2}$ .

The observation of the birefringence fringes gives a quite precise information on the mean tilt angle of the molecules at a given distance of the core of the wall. The jump in  $\theta_M^2$  (where  $\theta_M$  is the local tilt angle) is constant from one extinction to the other: we can thus construct a plot of  $\theta_M$  versus the distance through the wall. This is shown on Fig. 6; the results are in reasonable agreement with the theoretical  $(\tanh^2)$  law.<sup>(5)</sup>

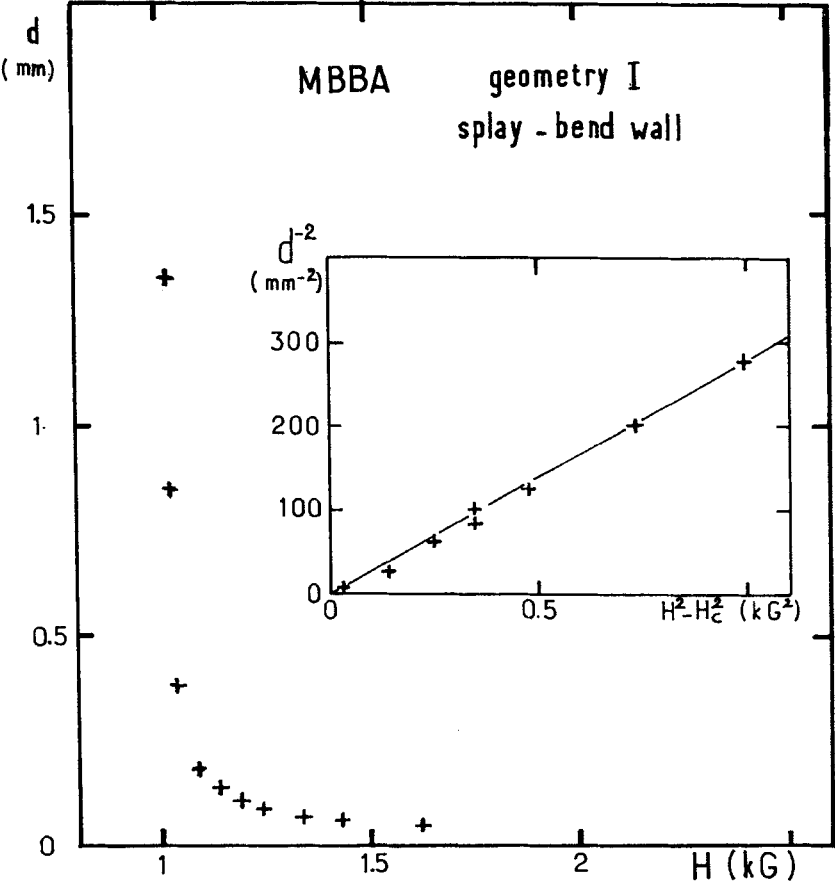


Figure 5. Plot of the distance between the first two birefringence fringes versus the magnetic field  $H$ . The inset shows that the divergence of the wall thickness is in  $\Delta H^{-1/2}$ .

Similar observations can be made with planar configurations ( $\mathbf{n}_o$  parallel to the glass plate: geometry II). But the optical aspect of the wall is slightly different in geometry II: the molecules in the core of the wall always stay parallel to  $\mathbf{n}_o$ . This gives an index of refraction larger in the core of the wall than in the domains, and the light is focused in the core of the wall. This is shown on Fig. 7a. Figure 7b gives a proof that the distortion is in the plane  $(\mathbf{H}, \mathbf{n}_o)$ : with a polarizer nearly normal to  $(\mathbf{H}, \mathbf{n}_o)$  the wall is no longer visible.

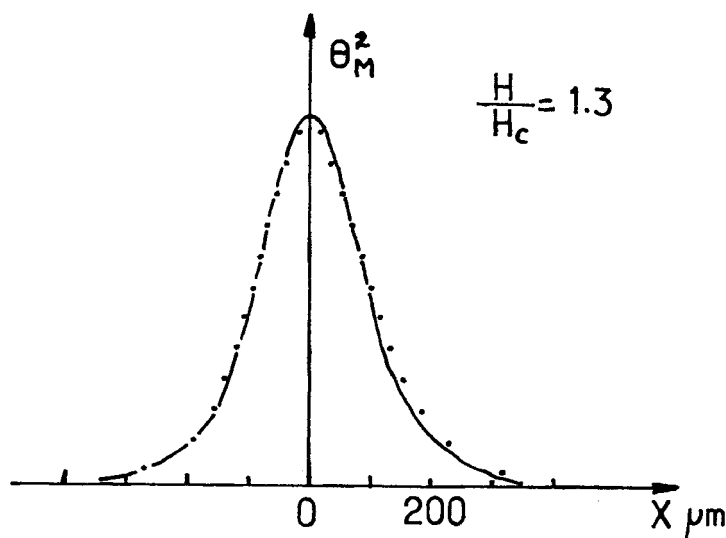


Figure 6. Plot of the mean distortion  $\theta_M$  through the wall deduced from the study of the birefringence fringes of Fig. 5. The points are experimental, the line corresponds to the theoretical tanh law. The slight dissymmetry of the distortion arises from the fact that the wall is not completely immobile.

### 3. Dynamic Behaviour of the Walls

A slight tilt ( $\epsilon \sim 5^\circ$ ) of the magnetic field favours one domain in front of the other; the walls move to enlarge the favoured domains. Figure 8 shows the experimental dependence of the velocity of the wall versus the magnetic field, for a fixed  $\epsilon$ . The divergence as  $H$  goes to  $H_c$  is observed to be in  $\Delta H^{-1}$ . The velocity of the wall is expected to be<sup>(5)</sup>

$$S = \frac{12}{\pi} \frac{\chi_a H^2}{\gamma^*} \frac{\epsilon \xi}{\theta_\infty^2} \quad (1)$$



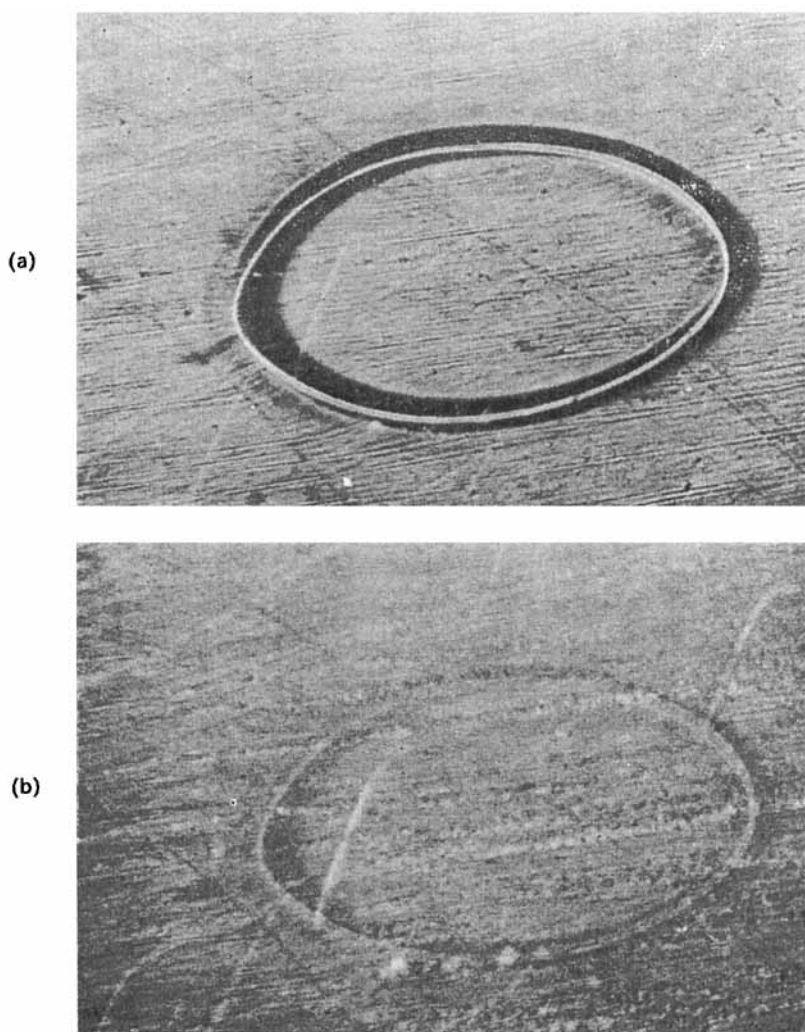


Figure 7. (a) typical view of a wall in geometry II ( $\mathbf{n}_o$  parallel to the glass plates), between crossed polarizers oriented at  $\pi/4$  with respect to  $\mathbf{n}_o$ . (b) with the polarizer normal to  $\mathbf{n}_o$ , the wall becomes no longer visible, as the molecules always stay in the plane  $(\mathbf{H}, \mathbf{n}_o)$ .

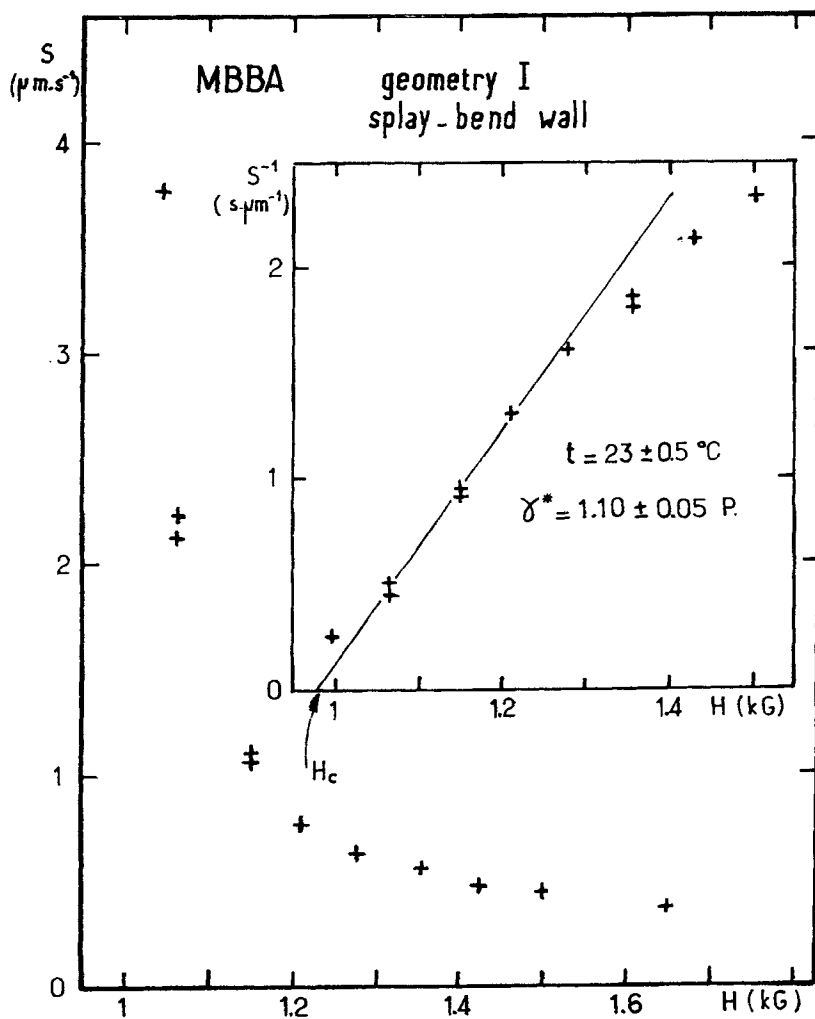


Figure 8. Velocity of a splay-bend wall,  $s$ , in geometry I, versus the magnetic field  $H$ , for a tilt angle of the magnetic field  $\epsilon = 1/10\text{rd.}$  The inset shows that this velocity diverges as  $\Delta H^{-1}$ , if  $H$  is decreased to  $H_c$ .

with  $\theta_\infty^2 = 2[(K_3/K_1)((H^2 - H_c^2)/H^2)]$ ;  $\theta_\infty$  represents the mean tilt angle of the molecules far from the wall.  $\xi$  is the magnetic coherence length  $\xi = 1/H\sqrt{(K_3/\chi_a)}$ .

The effective viscosity  $\gamma_1^*$  is not exactly equal to the twist viscosity  $\gamma_1$ , because of the coupling between flow and orientation in a nematic.<sup>(6)</sup> When the wall moves, the molecules in the core of the wall rotate from  $-\theta_\infty$  to  $+\theta_\infty$ . This rotation induces a flow in the sample, as described by F. Brochard.<sup>(7)</sup> This flow re-orientes the molecules and this tends to decrease the effective viscosity. This effect is large in geometry I, where the velocity gradient is normal to the molecules in the core of the wall, and negligible in geometry II, where it is parallel to the molecules.

From the velocity data, we can deduce the renormalized viscosity  $\gamma^*$  in geometry I, and the twist viscosity  $\gamma_1$  in geometry II. For M.B.B.A., at  $23 + 0.5^\circ\text{C}$ , we have obtained

$$\gamma^* = 1.1 \pm 0.05 P$$

$$\gamma_1 = 1.3 \pm 0.05 P$$

This gives a ratio  $\gamma^*/\gamma_1 = 0.85$  in good agreement with other measurements.<sup>(8,9)</sup>

Figure 9 gives the temperature dependence of  $\gamma^*$ , obtained by this method. It is in good agreement with other determinations, although our values are larger than those obtained by Prost and Gasparoux.<sup>(9)</sup> This discrepancy can arise from the fact that our method is completely free of nucleation of defects at the surfaces during the measurement.

The plot of Fig. 8 gives a very precise determination of the Freedericks critical field  $H_c$ , with no ambiguity due to the fact that the time response of the system diverges as  $H$  goes to  $H_c$  (which makes difficult any classical static method). It allows for an easy determination of elastic constants. A specific example is shown on Fig. 10 giving the temperature dependence of  $K_3$ , as measured by this method.

To conclude, walls in nematics provide us a useful tool to investigate not only static but also hydrodynamic properties of the nematic mesophase. They give a simple way of testing the backflow corrections predicted by Leslie's theory. For all quantitative studies,

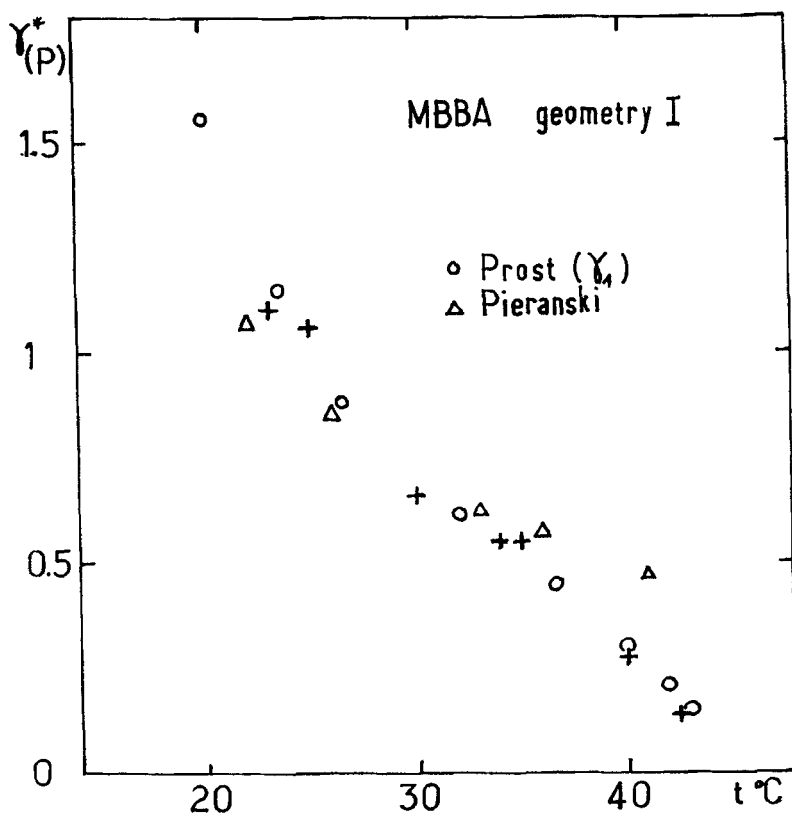


Figure 9. Temperature dependence of the renormalized viscosity  $\gamma^*$ , for methoxy-benzylidene butylaniline (M.B.B.A), deduced from the study of the velocity of a splay-bend wall, in geometry I, at different temperatures. Our experimental values correspond to the crosses, and are compared to other determinations. The values from Prost and Gasparoux are in fact determinations of  $\gamma_1$ , so they should be larger by a factor 1.3 to be in good agreement with ours, but the temperature dependence is in good agreement with ours.

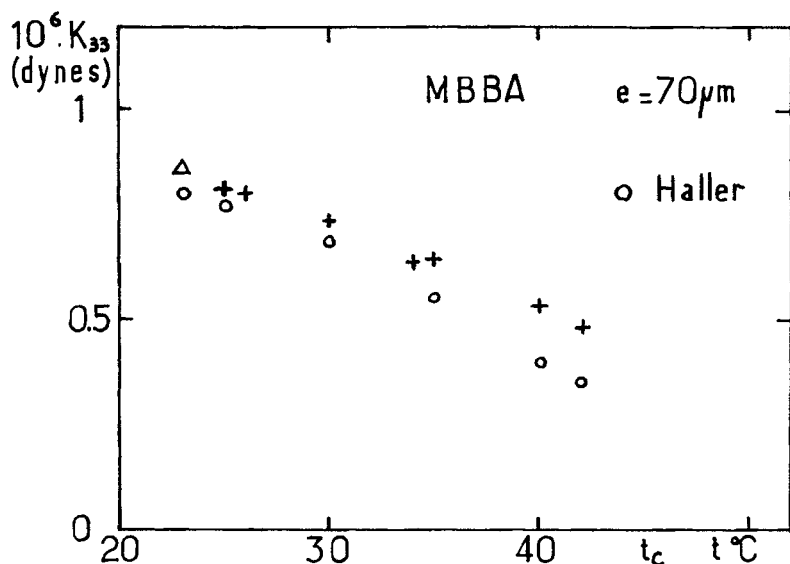


Figure 10. Temperature dependence of the bend elastic constant  $K_{33}$  for methoxy-benzylidene butylaniline (M.B.B.A), deduced from the determination of the Fredericks critical field  $H_c$  by the method indicated on Fig. 8. For comparison we have plotted on the same figure the values of  $K_{33}$  deduced from Haller's measurements<sup>(10)</sup>(0).

walls are preferable to line disclinations: with lines, the static and, more critically, the dynamic behaviour depend sensitively on the core region, for which no acceptable theory is available.

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