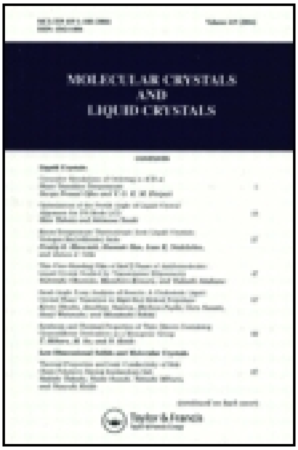

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 21 Mar 2007.

To cite this article: H. Koelmans & A. M. Van Boxtel (1971): Electrohydrodynamic Flow in Nematic Liquid Crystals, Molecular Crystals and Liquid Crystals, 12:2, 185-191

To link to this article: http://dx.doi.org/10.1080/15421407108082771

Molecular Crystals and Liquid Crystals. 1971. Vol. 12, pp. 185–191

Printed in Great Britain

Electrohydrodynamic Flow in Nematic Liquid Crystals†

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Received October 16, 1970

Abstract—Electrohydrodynamic instabilities in nematic liquid crystals (cellular flow in domains, turbulence) arise by the same mechanism as similar phenomena in isotropic liquids. In agreement with this view the rise time of the instability leading to turbulence in nematic liquid crystals was found to obey the same equation as was recently found for isotropic liquids.

When a thin layer of a nematic liquid crystal is subjected to an increasing voltage two electro-optical phenomena occur.

Above threshold voltage, which is usually about 5 V, the originally clear and transparent liquid crystal develops the well known domain structure first studied by Williams(1) in 1963. Up to a few volts above the threshold this domain pattern is stationary in time. On looking more closely, however, we observed that within the domains the liquid is not at rest, but rotates slowly around the domain axis. Liquid flow was deduced from the motion of tiny solid particles suspended in the liquid crystal.

Similar observations were made by Durand and co-workers of the Orsay liquid crystal group.(2)

One might doubt whether these particles are safe tracers of liquid flow. In principle, due to electrophoretic or dielectrophoretic effects, the particles might move with respect to the liquid instead of being passively dragged by the liquid. If, however, electrophoretic and/or dielectrophoretic effects were preponderant one would expect to find no correlation between the motions of different particles.

Actually a strong correlation has been found. Within a domain all

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† Presented at the Third International Liquid Crystal Conference, Berlin, August 24–28, 1970.

particles rotate in a plane perpendicular to the domain axis. The most important argument, however, is that when rotation is clockwise in one domain it is found to be anticlockwise in the next domain etc.

When the voltage is further increased the domain pattern is no longer stationary but gradually changes into the regime of turbulent flow in which the nematic liquid crystal strongly scatters light.(3) We conclude that in both the turbulent mode and the domain mode the electric field induces liquid motion.

These phenomena strongly recall the electrohydrodynamic flow known for a long time in ordinary isotropic liquids such as benzene and hexane.(4) In these low conductivity liquids a strong electric field leads to the build up of a space-charge. The force exerted on this space-charge by the electric field may cause hydrodynamic instabilities closely related to the classical Bénard instabilities where the driving force is a density difference due to a temperature gradient.

To check whether the mechanism by which electrohydrodynamic instabilities are generated in nematic liquid crystals is indeed the same as in ordinary isotropic liquids we did the following experiment:

The motion of solid particles was studied in the nematic phase slightly below the critical temperature and in the isotropic phase slightly above the critical temperature. Below the threshold voltage for domain formation the particles were at rest in both the nematic and the isotropic phase. Above threshold voltage both the nematic and isotropic phase developed slow cellular motion gradually changing into turbulent motion at higher voltages. This close correlation between the electrohydrodynamic phenomena in the nematic and the isotropic phase has been observed for a number of nematics such as *p*-methoxy benzilidene *p*-*n* butylaniline (MBBA), *p*-azoxyanisol (PAA) and Licristal N 4 (Merck).

The same type of experiment was also done in a slightly different way. A boundary between the nematic and the isotropic phase was produced by applying a small temperature gradient parallel to the surface of the sample. The isotropic liquid close to the phase boundary then usually contains small globules of the nematic phase. When this emulsion is insufficiently stable it may be stabilized by the addition of a small amount of surfactant. When a voltage below threshold is applied the nematic part of the liquid is transparent and the nematic droplets in the isotropic phase do not move. Above the threshold voltage the nematic liquid shows the domain structure and the nematic droplets show slow circular motion. At higher voltages the nematic liquid gradually develops light scattering associated with turbulent motion, whereas the nematic droplets gradually assume turbulent motion. These observations indicate that the mechanism by which the electrohydrodynamic instabilities are generated is the same in nematic and isotropic liquids. This conclusion is in conflict with a model recently proposed by Helfrich⁽⁵⁾ in which the anisotropic electrical properties of the nematic liquid crystal play a vital role.

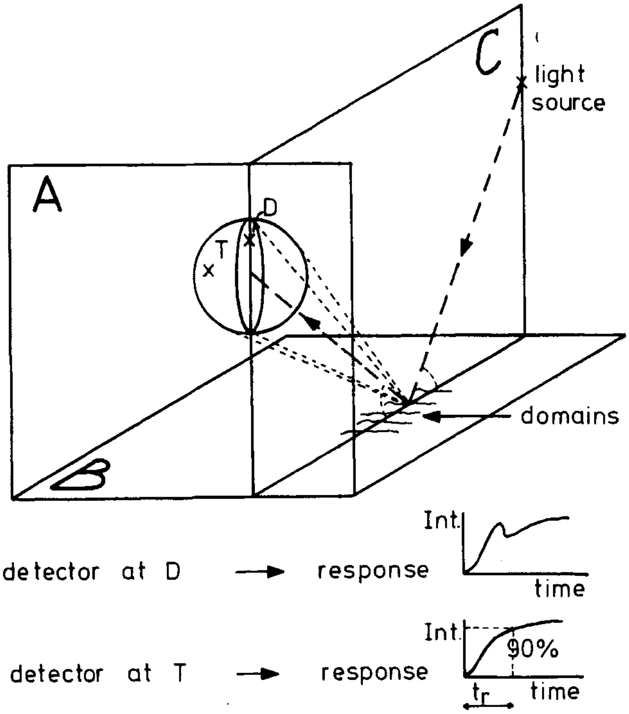

Figure 1. Geometrical arrangement for measuring the rise time of light scattering associated with electric field induced turbulence. The flat ellipse represents area in plane A where strong modulation due to domains is measured. The circle in plane A represents the area where strong light scattering due to turbulence is measured. At D light modulation due to domains obscures rise of light scattering due to turbulence. At T, outside the flat ellipse, no interference due to domains occurs.

Next we investigated the rise time of the turbulent instability in nematic liquid crystals by measuring the rise in intensity of the concomitant light scattering. The liquid crystal was excited by square voltage pulses. It was observed that during rise the turbulent instability is preceded by the domain instability. Conversely, during decay from the turbulent state the liquid crystal comes back to the unperturbed state via the domain mode. Light modulation due to the transient formation of domains may obscure the rise of light scattering associated with turbulence. Measurements of the rise of light scattering due to turbulence were made with the geometry sketched in Fig. 1. The liquid crystal (in plane *B*) is sandwiched between an upper glass plate coated with conducting transparent tin oxide and a lower glass plate coated with a vacuum-deposited aluminum mirror. The domains were uniformly oriented with the aid of the Chatelain rubbing technique.(6) With the incident light in a plane perpendicular to the domain axis (plane *C* in Fig. 1)

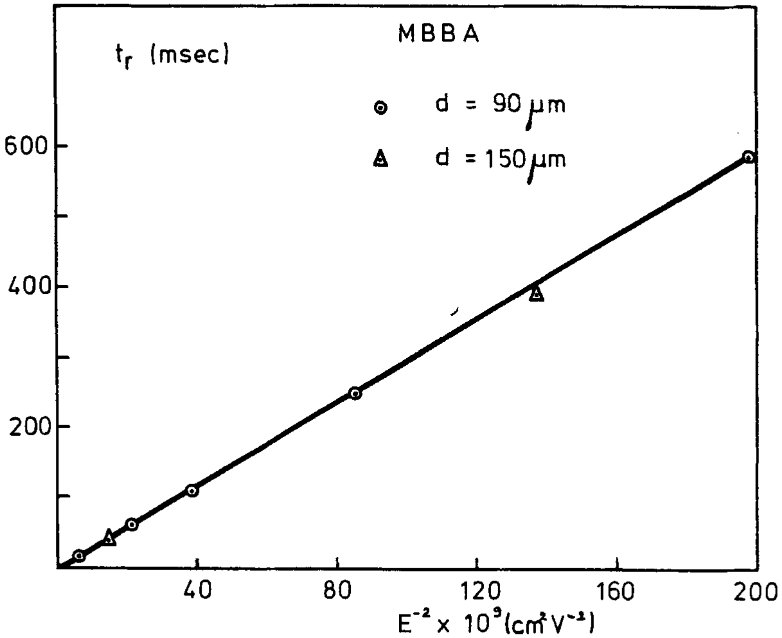

Figure 2. Rise time (t_r) of light scattering associated with turbulence as a function of the electric field. Sample: MBBA.

transient modulation of light due to domains is almost exclusively confined to this plane. In the turbulent mode the liquid crystal scatters light in all directions. When the detector is at D, a strong modulation peak due to domains obscures the rise of light scattering associated with turbulence. At T, well outside plane C, the domains do not interfere (cf. response curves at the bottom of Fig. 1).

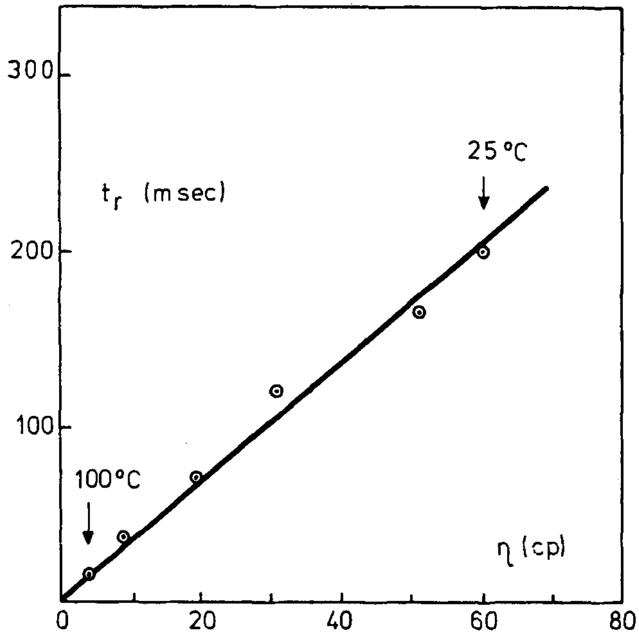

Figure 3. Rise time (t_r) of light scattering associated with turbulence as a function of viscosity (η). Sample: mixture of Schiff bases with nematic range between 20 and 100 °C.

The rise time t_r was defined as the time to reach 90% of steady state intensity of the scattered light. The dependence of t_r on the electric field E is shown in Fig. 2. In fact t_r was measured at a number of applied voltages for various distances d between the electrodes. The results for $d = 90\,\mu m$ and $d = 150\,\mu m$ are given in Fig. 2. It is seen that t_r is proportional to E^{-2}. Figure 3 shows the influence of the viscosity on t_r. The viscosity of the sample, which was a mixture of nematic Schiff bases, was changed by variation of the temperature from 20° to 100 °C close to the critical temperature of the mixture. The rise time t_r is directly proportional to η. To show that the change of temperature influences t_r exclusively via change in viscosity of the sample both t_r and η were plotted as a function of temperature in Fig. 4. The curves for t_r and η were matched in one point. Then the curves coincide over the whole range of temperatures. It is interesting to note the well known rise in viscosity on approaching the critical temperature, t_r closely following this rise.

We thus find t_r to be proportional to η/E^2. Some simple dimensional analysis leads then to the equation:

$$t_r = \frac{C \cdot \eta}{\epsilon \cdot E^2} \tag{1}$$

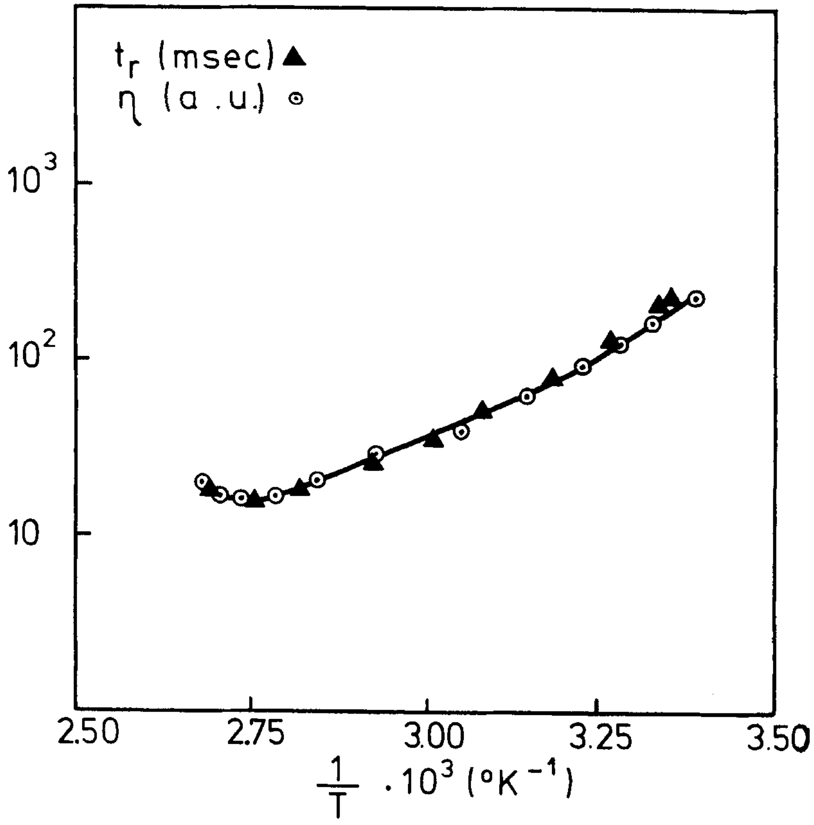

Figure 4. Rise time (t_r) and viscosity (η) as a function of temperature. Sample: mixture of Schiff bases with nematic range between 20 and 100 °C.

where ϵ is the dielectric constant of the liquid crystal and C is a numerical constant which was found to be about 100. It is interesting to note that the same equation with a numerical constant of the same order of magnitude was recently found to describe the rise of turbulent electrohydrodynamic instability in isotropic liquids.(7) In a recent paper Watson, Schneider and Till(8) showed that Eq. (1) may be derived when strong unipolar injection leading to space-charge-limited current is assumed. Their calculations gave a numerical constant of 300. This could mean that the assumption of strong unipolar injection is not too unrealistic in the nematic liquid crystals which we investigated. The electrical currents measured did not show the well known V^2/d^3 dependence of space-charge-limited currents. In fact the measured current is usually only slightly more than ohmic and even tends to increase with d, which was also observed by Heilmeier.(3) Apparently the contribution of injection to the total current is only small compared with the ohmic contribution due to impurity ions.

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