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Mechanism for the formation of bubble domains

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The mechanism for the formation and stabilization of acoustic bubble domains is investigated experimentally within the framework of the Akahane and Tako model which is based on the assumption of the formation of a system of defects in the layer of a cholestric liquid crystal, these defects pin the bubble domains. The theory takes no account of the interaction between bubble domains, this being valid in the case of a low density of domain packing. The correlation between the experimental results and theory is quite satisfactory, especially in the region of $d/P_0 < 1{\cdot}5$ where the bubble domain packing density is very low (where d is the thickness of the cholestric layer and P_0 is the pitch of the helix).

For a long-time memory in liquid crystal based acousto- and electro- optic devices to be realized, use is made of the relaxation of non-equilibrium states and the stability of some intertransformable textures in cholesteric liquid crystals. One of these methods is connected with the creation of a stable system of bubble domains in a cholestric induced electrically, thermally or acoustically. In the present work the mechanism for the formation and stabilization of bubble domains is investigated within the framework of the Akahane and Tako model [1] which is based on the assumption of the formation of defects in the layer which pin the bubble domains. The theory takes no account of the interaction of domains with one another this being valid at a low density of domain packing, and, according to preliminary observations [2], acoustic bubble domains fall under this theory.

The cholesterics investigated were solutions of cholesteryl caprinate or cholesteryl chloride in nematic liquid crystals 4O.4 and H-8, respectively. The proportions of these substances were chosen so that the equilibrium value of the pitch, P_0, varied in the range 3–48 μm. The thickness, d, of the layer was changed in the range 2–150 μm. To provide homeotropic conditions on the surfaces of the plates bounding the cholesteric layer, the plates were treated with lecithin or cleaned chemically. The acoustic conditions in the experiment were such that either compression of the cholestric (0·55 and 3 MHz) or its shearing (0·22 MHz) was induced by sound. The experimental set up is shown in figure 1, where: 1 is the acoustic transducer; 2 is the cholesteric layer; 3 is the upper plate; 4 is the lower plate; 5 is an electrically conducting coating; 6 is an electrically conducting and light-reflecting coating of aluminium; 7 is the light source; 8 is a semi-transparent beam splitter; 9 is a microscope; 10 is a photo-multiplier; 11 is a measuring voltmeter; 12 are polarizers; 13 is a peep-hole; 14 is an immersion liquid; and 15 is a temperature control system. The acoustic contact of the transducer with the cholestric layer was realized either through the immersion liquid (see figure 1 (*a*)) or through a thin adhesive layer (figure 1 (*b*)). In figure 1 (*a*) the cholesteric layer is bound on the liquid side by a lavsan film pulled over a rigid carcass.

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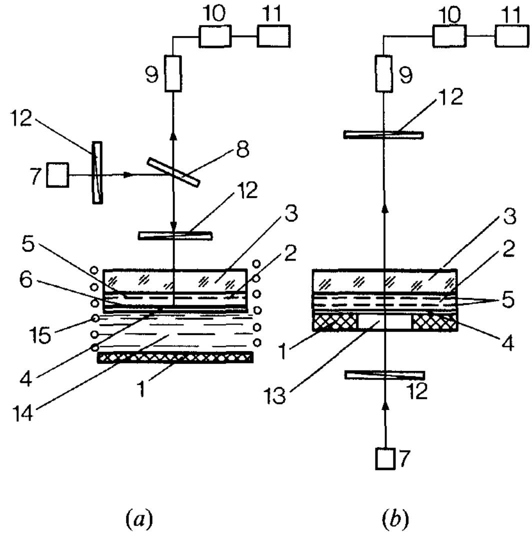
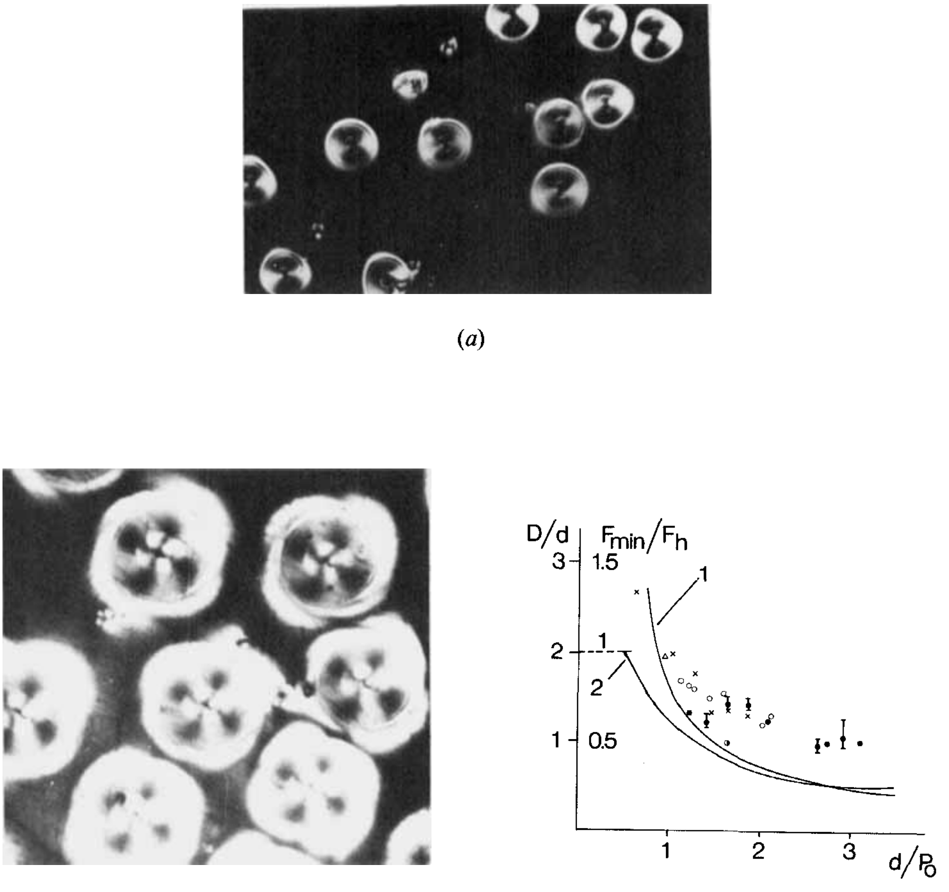


Figure 1. Experimental set-up for studying the textural transformation of the cholesteric layer under acoustic action in reflected (*a*) and transmitted (*b*) polarized light: 1, acoustic transducer; 2, cholesteric layer; 3–4, upper and lower plates; 5, coating of SnO_2; 6, coating of aluminium; 7, light source; 8, beam splitter: 9, microscope; 10, photomultiplier; 11, voltmeter; 12, polarizers; 13, peep-hole; 14, immersion liquid.

Changes in the layer macrostructure were observed in polarized transmitted and reflected light (see figures, 1 (*b*), (*a*)), respectively.

It is known that the type of macrostructure formed in a cholesteric flat layer with homeotropic boundary conditions depends on the ratio of its thickness to the equilibrium pitch of the helix [3]. Observations showed that under the action of acoustic vibrations changes in the macrostructure of the cholesteric layer for specimens in which a uniform homeotropic orientation still exists did not differ significantly from those which are characteristic of the homeotropic layer of a nematic liquid crystal under similar conditions. There exists a threshold of the sound intensity, and if this threshold is exceeded, stationary acoustic flows disturb the director orientation and cause the formation of disclinations. This new disordered state reveals itself optically in an intense diffuse scattering of light. This effect is known as acoustic dynamic scattering of light. The essential differences in the orientational states of the cholesteric and nematic layers manifest themselves when the action ceases. Figure 2(*a*), (*b*) illustrates a typical inhomogeneous texture of acoustic bubble domains which are formed in the cholestric in the relaxation stage. This new texture is stable in time, i.e. this textural transition prevents the cholesteric from relaxing to its initial macro-structure, and thus the effect of the acoustic memory is realized. For such a stable system of acoustic bubble domains to form, it is necessary that the acoustic intensity in the irradiation stage should exceed the dynamic light scattering threshold. The system of defects arising in the cholesteric at lower levels of the action disappears after irradiation ceases, and the initial homeotropic orientation is restored in the layer.

The dynamics of formation of acoustic bubble domains is illustrated by the graphs shown in figure 3(*a*). They correspond to the following conditions: $d = 40\,\mu\text{m}$, $P_0 = 48\,\mu\text{m}$, $f = 0{\cdot}2\,\text{MHz}$ (shearing) and irradiation time = 1 min. It can be seen



(a)

(b)

(c)

Figure 2. Comparison of the Akahane and Tako model [1] with the experimental data: microphotographs of acoustic bubble domains formed in the cholesteric layer in the relaxation stage at various values of the layer thickness and the helix pitch ($d/P_0 < 1{\cdot}5$ (*a*), $d/P_0 > 1{\cdot}5$ (*b*)); theoretical dependences of the bubble domain diameter and free energy normalized to the layer thickness and the free energy of the homeotropic structure, respectively, on the ratio d/P_0 (curves 1 and 2), points 1–6 are values of D/d observed in the frequency range (0·2–3) MHz. ◑ = 1, ■ = 2, △ = 3, × = 4, ○ = 5, ● = 6.

that increasing the time, t, after irradiation leads to some increase in the number of bubbles N, but practically does not influence the bubble diameter D. (The value of N corresponds to the number of bubbles within the field of vision when observing the layer under a polarizing microscope.) From the data presented in figure 3 (*b*), it follows that there is a definite relationship between the diameter of domains and the layer thickness; an increase in the frequency of the acoustic action having no influence upon the form of the function $D = f(d)$. The labels 1 and 2 refer to the frequencies 3 and 0·5 MHz (compression) respectively; 3 corresponds to 0·2 MHz (shearing); $P_0 = 48\,\mu m$; the values of D correspond to an extremum of the size distribution of BDs.

The data in figure 2 (*c*) show the influence of the helix pitch on the diameter of the acoustic bubble domains. The labels 1 to 4 correspond to the experimental values

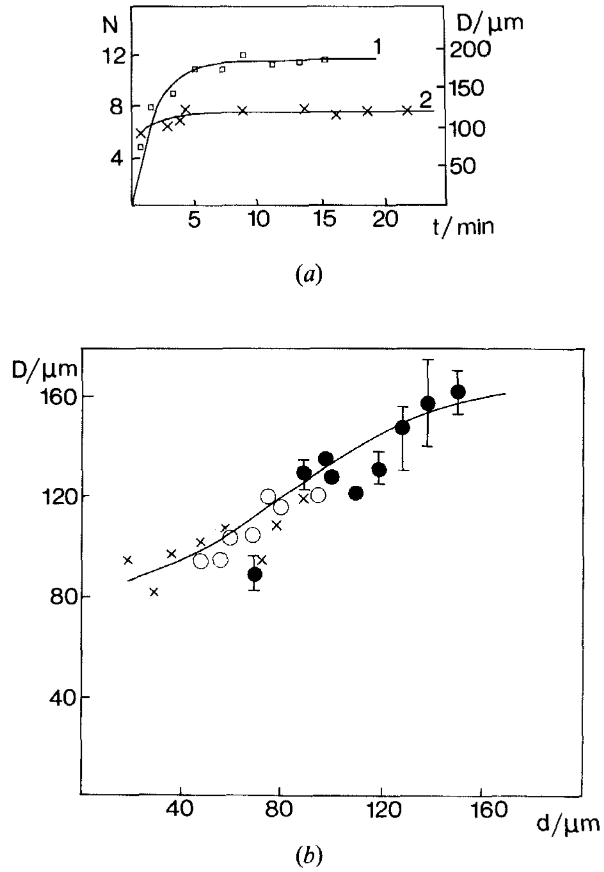


Figure 3. Acoustic bubble domains in the cholestric for the helix pitch $P_0 = 48\,\mu m$: variation with time t of the number, N, and diameter D, at $d = 40\,\mu m$, $f = 0{\cdot}2$ MHz (curves 1 and 2) (*a*) and bubble domain diameter as a function of the layer thickness (points ● and ○ refer to 3 and 0·5 MHz (compression), points x correspond to 0·2 MHz (shearing) (*b*).

of D/d at 0·2 MHz for specimens with equilibrium pitches 3; 4·2; 10·5 and 48 μm. The labels 5 and 6 refer to the specimen with $P_0 = 48\,\mu m$ and frequencies 0·5 and 3 MHz. The layer thickness was varied in the range 10–150 μm. In each of the series of experiments (P_0 = constant) observations were made with unchanged boundary conditions. The values of D/d and d/P_0 are used for convenience of comparing the data for specimens of different thickness.

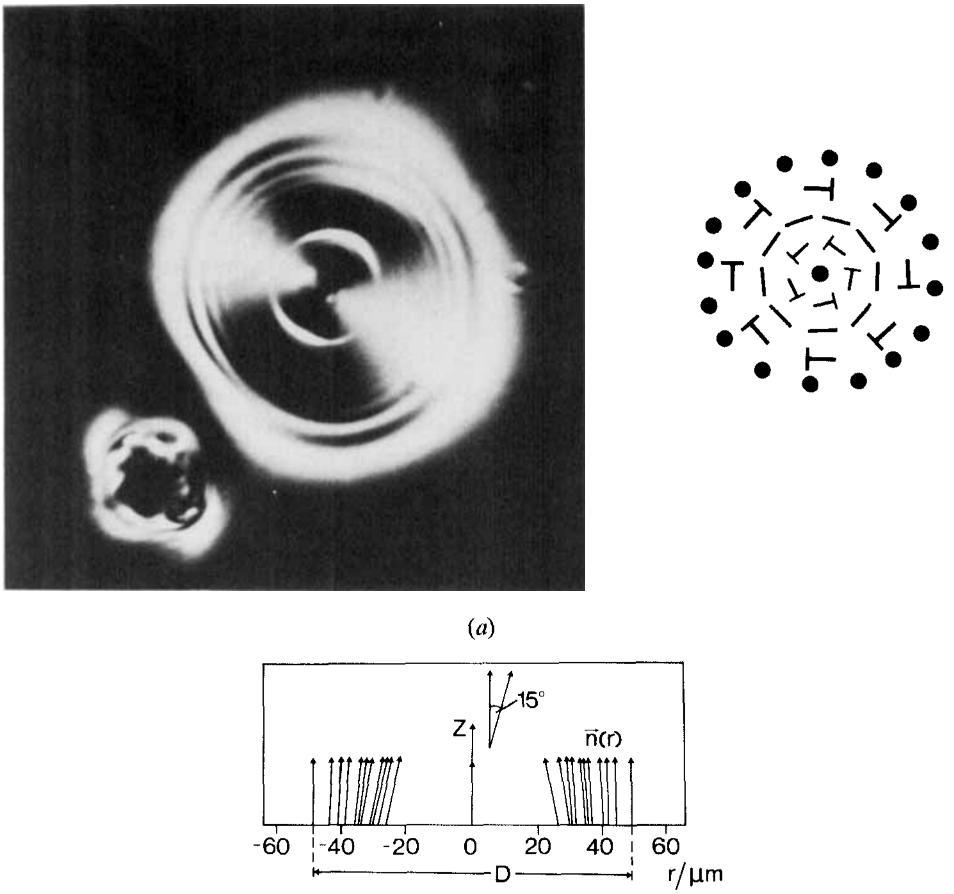
Let us compare the experimental results with the calculations made by Akahane and Tako [1]. Graph 1 in figure 2(*c*) presents the theoretical dependence $D/d = f(d/P_0)$ calculated in [1] with the following assumptions: (*a*) the director alignment inside the domain corresponds to the model proposed by Kawachi *et al.* [4], this meaning a non-uniform distribution of the director **n** (which is close to the cholesteric helix) at a distance of the order of P_0 in the layer plane; (*b*) disclinations are localized at points with coordinates $z = \pm d/2$, $r = P_0/4$ (in a cylindrical coordinate system r, θ, z; the z axis is perpendicular to the layer plane); (*c*) bubble domains do not interact with one another. It is seen that the agreement between the experiment and theory is quite satisfactory, especially in the region $d/P_0 < 1{\cdot}5$. Larger experimental values of D/d, as compared to theory, observed at $d/P_0 > 1{\cdot}5$ are likely to be due to some increase in the packing density of acoustic bubble domains; this is supported by the micrographs in figure 2 and also due to factors resulting from the interaction of the domains with one another, which is not considered by theory [1].

Graph 2 in figure 2 (*c*) shows the behaviour, according to the theory of Akahane and Tako, of free energy $F_{\min}$ of a stable bubble domain pinned by surface disclinations for the values of d/P_0 considered. The value of $F_{\min}$ is normalized to the free energy F_H of the homeotropic macrostructure. From these data it follows that at $d/P_0 < 0{\cdot}5$ the ratio $F_{\min}/F_h > 1$, i.e. the homeotropic orientation is more stable than that which forms bubble domains. Thus, the theoretical value of $(d/P_0)_{\min}$ following from the energy relations is equal to 0·5, which is in agreement with the experimental results: $(d/P_0)_{\min} \approx 0{\cdot}55$.

The idea of Akahane and Tako about the mechanism of stabilization of bubble domains by a system of orientational defects is also supported by visual observations. First, acoustic domains are formed only in the parts of the layer where the density of disclinations produced by the acoustic flows is sufficiently high [5]. Secondly, the density of packing of acoustic bubble domains increases as an increase in frequency leads to a higher non-uniformity of the acoustic field and so to a decreasing scale l of acoustic streaming responsible for orientational disturbances of the macrostructure. This results in an increase in the non-uniformity of the orientational distribution of the director in the cholestric layer; $l \sim \delta_{ac}\lambda$, where λ is the acoustic wavelength and δ_{ac} is the thickness of the acoustic boundary layer, $\delta_{ac} \sim f^{-1/2}$. Finally, the packing of the cholestric director in the bubble domains accepted by the model of Akhane and Tako is in agreement with the microscopic observations. Figure 4 (*a*) shows a micrograph of a single acoustic bubble domain in a parallel beam of polarized light (polarizers are crossed) obtained under the following conditions: $d = 40\,\mu$m, $P_0 = 48\,\mu$m and $f = 216$ kHz (shearing). The boundary of the domain separates the parts of the specimen with different director orientations and different refractive indices. Turning the object (layer) on the microscopic stage does not lead to the clearing of the layer outside the bubble domains, this indicates that the director arrangement in this part of the specimen is homeotropic. Within the bubble domain there is an interference pattern representing a system of concentric light and dark rings of double refraction the centre of which coincides with the symmetry axis of the domain. The ring width decreased towards the centre. This means that the phase difference δ_m of the interfering rays due to their passage through the layer within the bubble domain increases as the distance from its boundary increases. Figure 4 (*b*) shows, schematically, the change in the angle φ between the director **n** and the normal to the layer for the case of passing through the bubble domain boundary. It varies from $\varphi = 0$ outside the domain (uniform homeotropic orientation) up to $\varphi = \varphi_m$ inside the domain. The values φ_m are calculated for the orders m of interference within the limits of vision of the pattern using the known conditions for extrema of the optical transmissivity function

$$I(r)/I_0 = \sin^2[\delta_m(r)/2]$$

which describes the optical properties of a uniaxial-crystal plane-parallel plate (cut out perpendicular to the optic axis). Observations showed that if the composition of the solution was so changed that the pitch decreased, the width of the interference ring in the bubble domains also decreased. This agrees with the results of [6] where it was shown experimentally that a decrease in P_0 leads to a reduction of the birefringence



(a)

(b)

Figure 4. Micrograph of an isolated acoustic bubble domain in a parallel beam of polarized light (crossed polarizers) (*a*) and diagram showing the change in the angle φ between the direction **n** and the normal to the layer inside this domain (*b*) at $d = 40\,\mu$m, $P_0 = 48\,\mu$m, $f = 0{\cdot}2$ MHz.

in the cholestric. It seems that this effect causes the change in the interference pattern. The presence of the Maltese cross on the micrographs of the acoustic bubble domains confirms the validity of a non-uniform distribution of the director **n** (see figure 4 (*b*)). Such a distribution of **n** in the domains creates the conditions for a parallel polarized beam of light which are equivalent to those for a convergent beam of polarized light which arise in a layer with a uniform distribution of the director (φ = constant) (cut perpendicular to the optic axis). From this distribution, **n**(**r**), it follows that the effective refractive index of the medium inside the bubble domain must increase in the direction from its boundary to the centre. In this case the domain will behave as an optical collecting lens. Indeed, the focal line shaped as a ring is seen distinctly in figure 4 (*a*) (the microscope is focused somewhat above the cholesteric layer surface).

In conclusion, our analysis shows that the results of experiemnts on acoustic bubble domains are accounted for by the model of Akahane and Tako concerning the mechanism of formation of bubble domains and their stability. Because of a comparatively low packing density acoustic bubble domains offer the unique possibility of a quantitative test of the model.

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