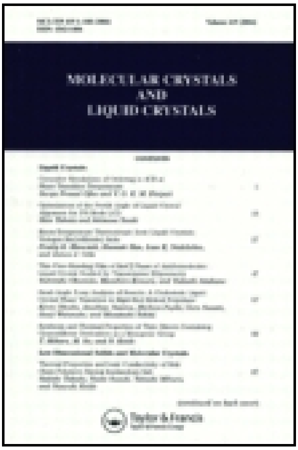

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New Molecular Alignment Models of Bubble Domains and Striped Domains in Cholesteric-Nematic Mixtures

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New Molecular Alignment Models of Bubble Domains and Striped Domains in Cholesteric-Nematic Mixtures

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Bubble domains and striped domains in cholesteric-nematic mixtures were observed carefully with a polarizing microscope, a differential interference microscope and a holographic interference microscope. Some facts that could not be explained by the conventional models of them were found. The new models which can explain the observation well are proposed. The stability of these domains was discussed from the energetic and topological points of view.

1 INTRODUCTION

It has been known that bubble domains and striped domains appear when a cholesteric liquid crystal is bounded perpendicularly (homeotropic molecular alignment).[1,2] Particularly, striped domains are called fingerprint textures and are interpreted in terms of various types of disclinations in cholesterics.

Though some molecular alignment models of them have been proposed,[1-4] they were not confirmed enough in experiments. In order to investigate the molecular alignment of these domains we carefully observed them in very large pitch cholesterics with a polarizing microscope, a differential interference microscope and a holographic interference microscope, and found some facts that could not be explained by the conventional models that have been thought as the most naive models.

In this paper we will show some defects of the conventional models from the experimental and theoretical points of view, and propose the new molecular

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alignment models of them. Those new models are studied from the energetic and topological points of view.

2 EXPERIMENTAL PROCEDURE

In order to observe the domains precisely, we used three kinds of the mixtures of *p*-methoxybenzilidene-*p*'-*n*-butylaniline (MBBA) and cholesteryl chloride (CC) in the weight ratio of 99.9:0.1, 99.95:0.05, and 99.97:0.03. These mixtures have pitches of about 150 μm, 300 μm, and 500 μm, respectively.

To compare the structure of the domains in a very large ptich cholesteric liquid crystal with that in a shorter pitch, we also use another mixture of MBBA and CC in the weight ratio of 99.5:0.5, which has a pitch of about 30 μm. The mixtures were sandwiched between two glass plates making a small angle. Each sample cell has the sample thickness to pitch ratio (d/p) which varies continuously about 0.4 to 1.2. To obtain a homeotropic molecular alignment, carboxylatochromium complexes were coated on the surface of the glass plates.

We could make the very big bubble domains of about 1 mm diameter and the striped domains of about 0.8 mm width (the weight ratio of 99.95 (MBBA):0.05 (CC)). By observing these big domains carefully with a polarizing microscope, we confirmed there were no structural differences between the very big domains and the normal size domains which is about 30 μm diameter (bubble) and the 24 μm width (stripe) (the weight ratio of 99.5 (MBBA):0.5 (CC)).

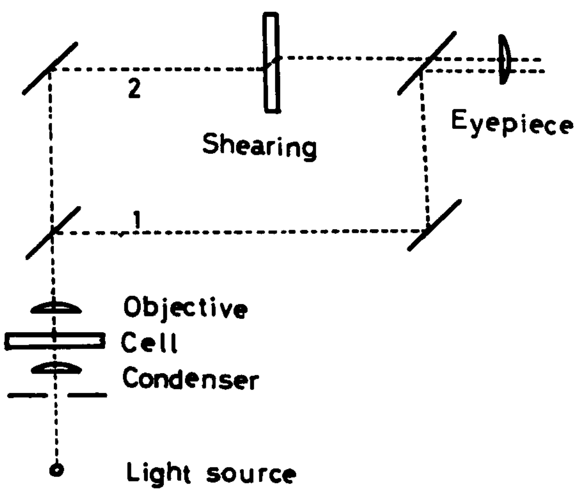

FIGURE 1 Optical system of a differential interference microscope.

We also observed these sample cells by a differential interference microscope and a holographic interference microscope. The differential interference microscope used in this investigation was "interphaco" of Carl Zeiss Co. Its optical system is the Mach-Zehnder interferometer shown in Figure 1. The light beam 2 can be sheared in a definite direction to the beam 1 by a shearing equipment. These two beams were interfered to make the differential interferogram. In order to compare the difference made by the direction of the linearly polarized light incident upon the cell, we also used the polarizer.

The schematic representation of the holographic interference microscope used in this experiment is Figure 2. We used a He-Ne laser as a light source. Before setting the sample cell, we took the hologram which would supply the reference wave. When we observed the sample cell, the diffracted wave by the cell and the reference wave from the hologram were interfered. The interference pattern thus obtained represents the distribution of the refractive index averaged along the direction which is perpendicular to the glass plate.

3 RESULTS

3.1 Observation with the polarizing microscope

i) Bubble domains

Figure 3 shows the observed result without a polarizer and an analyzer. The center is black. There is sometimes dust at the center. There is a light center ring.

Figure 4 shows the observed result without an analyzer (observation with a polarizer only). If we take the y-axis parallel to the direction of a polarizer, there are black brushes at the lower domain like the function $y = -x^3$.

Figure 5 shows the observed result without a polarizer (observation with an analyzer only). If we take the y-axis parallel to the direction of an analyzer, there are black brushes at the upper domain like the function; $y = x^3$.

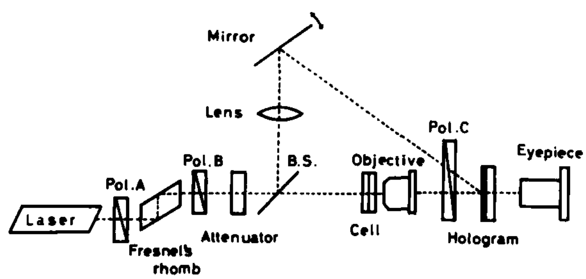

FIGURE 2 Optical system of a holographic interference microscope.

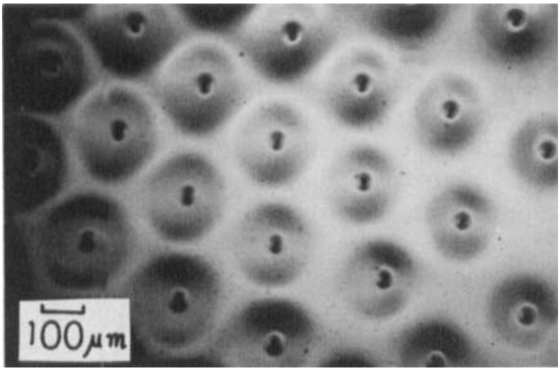

FIGURE 3 Bubble domain observed without a polarizer and an analyzer.

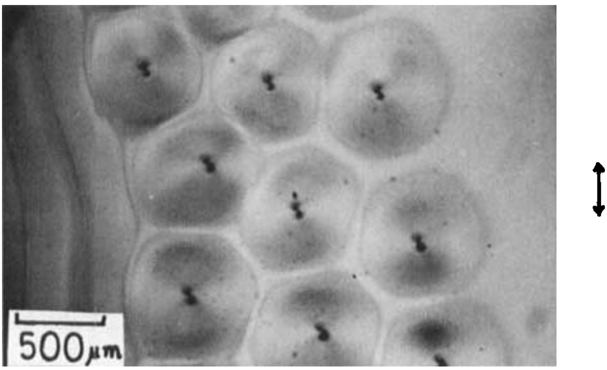

FIGURE 4 Bubble domain observed with a polarizer. The arrow shows the direction of a polarizer.

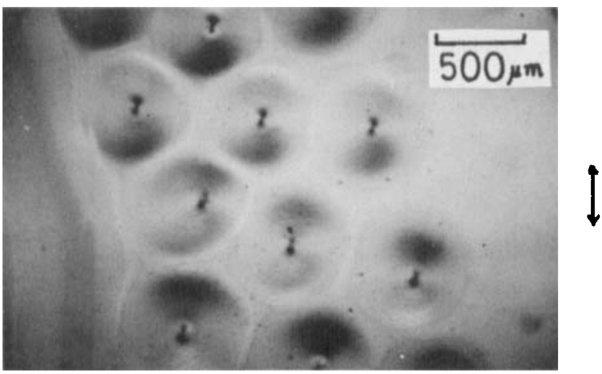

FIGURE 5 Bubble domain observed with an analyzer. The arrow shows the direction of an analyzer.

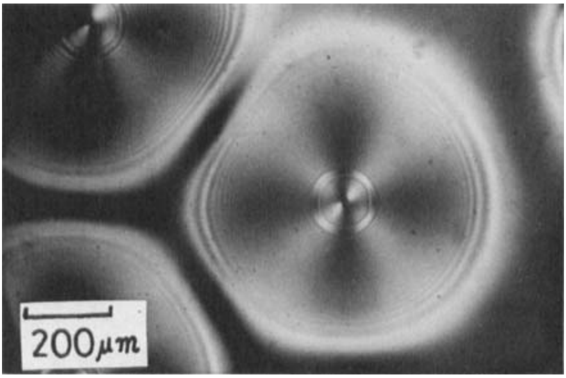

FIGURE 6 Bubble domain observed with crossed nicols.

Figure 6 shows the result with crossed nicols. There are crossed black brushes. There is no variation with the rotation of the cell between the fixed crossed nicols. By 90° rotation of the analyzer, we observed the bubble domain under parallel nicols (see Figure 7). Outside of the center light ring, the black brushes rotated by 45° and become dim compared with the case of crossed nicols. Inside the center ring, the brushes due to the analyzer rotated by 90°, but the brushes due to the polarizer were unchanged.

ii) Striped domains

There are two kinds of ends of striped domains. One has a singularity, the other has no singularity (see Figure 8). There is the inside boundary whose

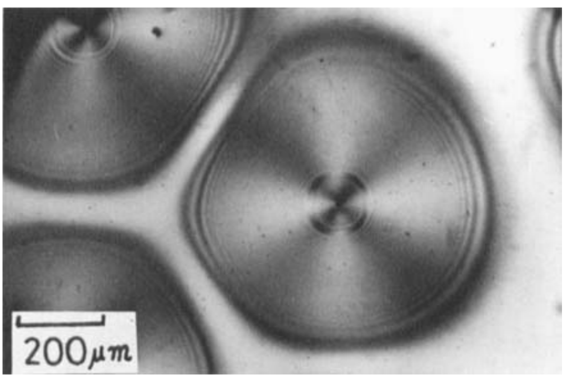

FIGURE 7 Bubble domain observed with parallel nicols.

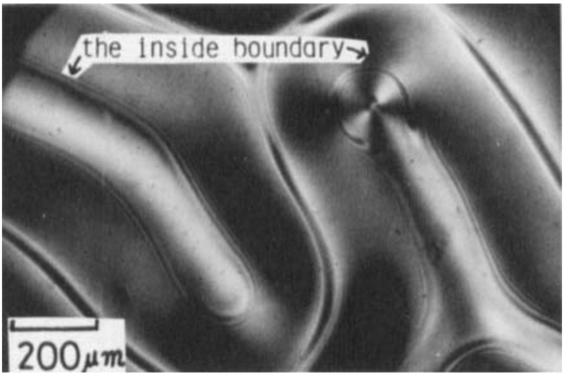

FIGURE 8 Striped domain observed with crossed nicols.

width varies depending on d/p_0 in proportion to the diameter of the light center ring of bubble domains. The black brushes under the crossed nicols are shown in Figure 8. We can see the striped interference pattern in the range between the inside boundary and the outside boundary of bubble domains but can not see that the inside of the inside boundary (see Figure 9). This suggests that the distribution of the refractive index over a period of stripes seems to be Figure 10. The dim part under the parallel nicols are shown in Figure 11.

The looped domain shown in Figure 12 is regarded as the looped striped domain. We can observe the vertical black brushes in Figure 12 under both parallel and crossed nicols. When we overturn the cell, we can observe the vertical black brushes rotated reversely.

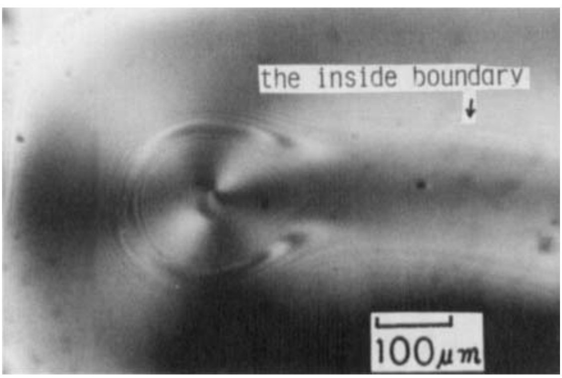

FIGURE 9 End of the striped domain with a singularity.

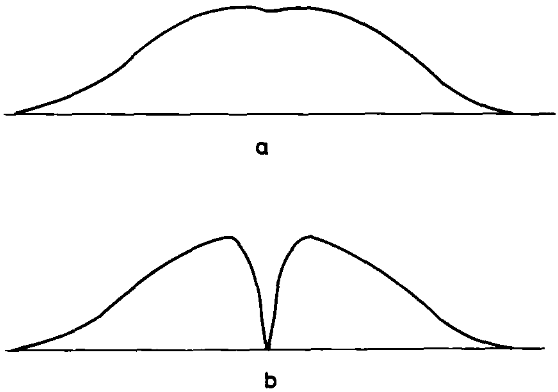

FIGURE 10 Distribution of refractive index over a period of stripes (a) and bubbles (b).

3.2 Observation with the differential interference microscope

i) Bubble domains

Figures 13(a), (b) and (c) show the differential interferogram of bubble domains in cases of the non-polarized incident rays, and the horizontally and vertically linearly polarized incident rays, respectively.

ii) Striped domains

Figures 14(a), (b) and (c) correspond to the cases of the non-polarized incident rays and the horizontally and vertically linearly polarized incident rays, respectively. We could not analyze these results because the continuity of the interference pattern were not clear.

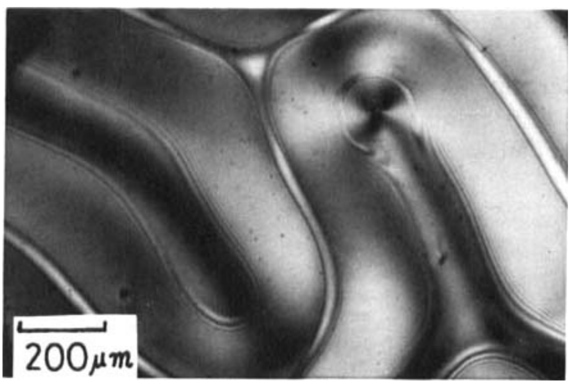

FIGURE 11 Striped domain observed with parallel nicols.

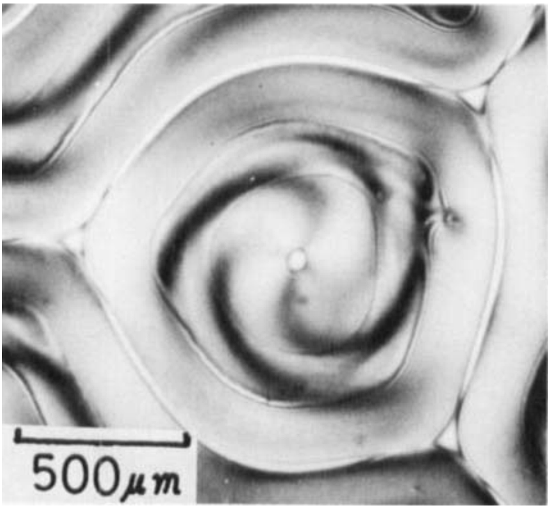

FIGURE 12 Looped striped domain observed with parallel nicols.

3.3 Observation with the holographic interference microscope

We can get the horizontal parallel interference pattern without a sample cell as shown in Figure 15. Figures 16(a) and (b) show the interferogram of the bubble domain in cases of the horizontally and vertically linearly polarized incident rays, respectively. We can see the interference pattern such as Newton's rings in Figure 16(b). This pattern also should appear on the upper side to make a pair. Because the coherence of the light having passed through the upper side of the bubble domain is disturbed, the pattern of the upper side does not appear in Figure 16(b). We can see a pair of these patterns on the left and the right sides in Figure 16(a).

Figures 17(a) and (b) show the results of striped domains in case of the horizontally and vertically linearly polarized incident rays, respectively. We recognize that the distribution of the refractive index varies rapidly in the outside of the inside boundary in Figure 17(a). These variations are not so much in Figure 17(b).

4 DISCUSSION

In this section we propose the new molecular alignment models that can explain the experimental results well, and compare the new models with the con-

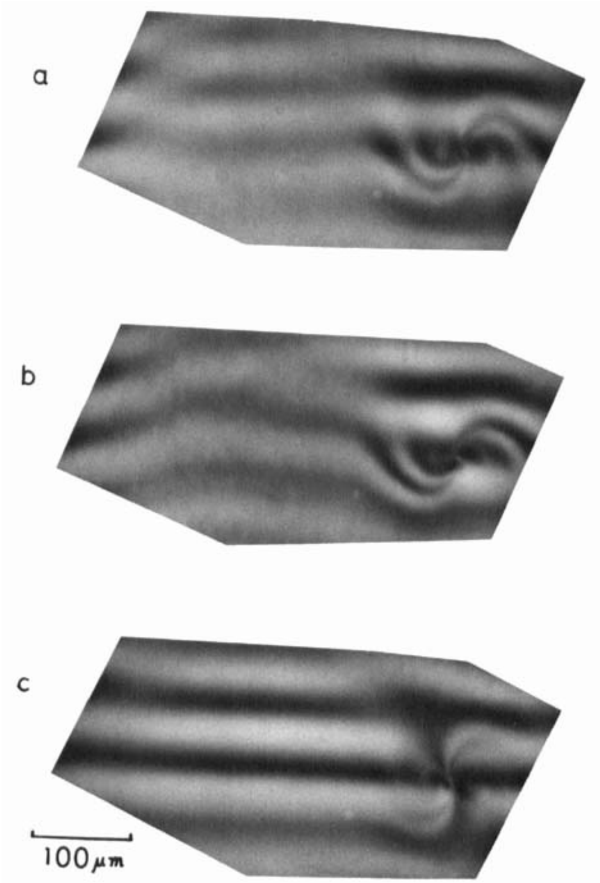

FIGURE 13 Differential interferogram of a bubble domain in case of non-polarized (a), horizontally (b), and vertically (c) linearly polarized incident rays.

ventional models, we discuss the defects of the conventional models and the superiority of the new models.

4.1 New molecular alignment models

We show the new and conventional molecular alignment models of bubble domains and striped domains.

Figures 18(a) and (b) show the conventional models for striped and bubble domains respectively. We propose the new models for striped domains as shown in Figure 19 and bubble domains as shown in Figure 20. To see the three-dimensional structure of bubble domains, we show three illustrations.

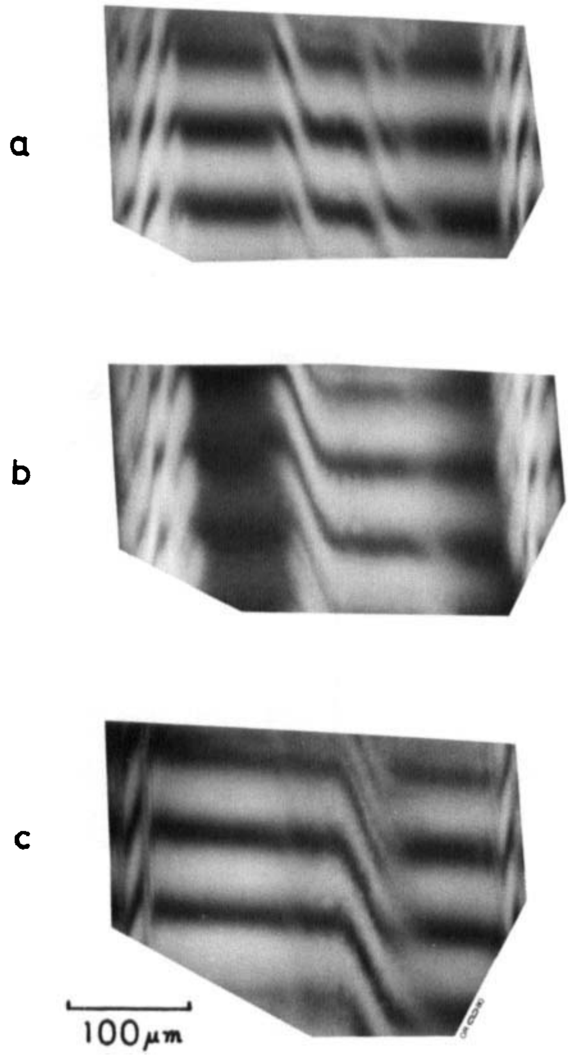

FIGURE 14 Differential interferogram of a striped oomain in case of non-polarized (a), horizontally (b), and vertically (c) linearly polarized incident rays.

Figure 21(a) is the A-B vertical section for a striped domain and Figure 21(b) is the C-D vertical section for a bubble domain.

Figure 22(a) and (b) show the conventional and new molecular alignment models of a looped striped domain, respectively. The dashed curves in these figures represent the black brushes that should be observed on each model with the polarizing microscope (crossed or parallel nicols).

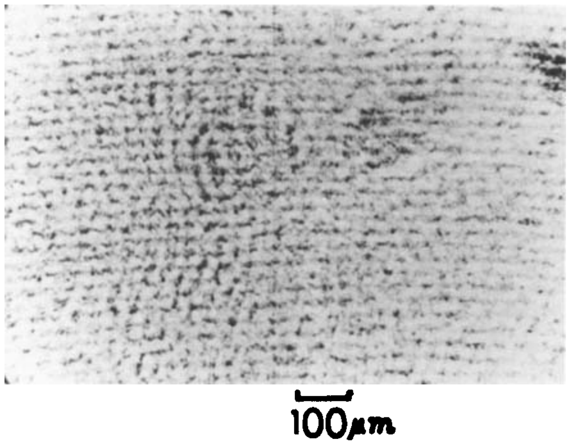

FIGURE 15 Holographic interferogram without sample cell.

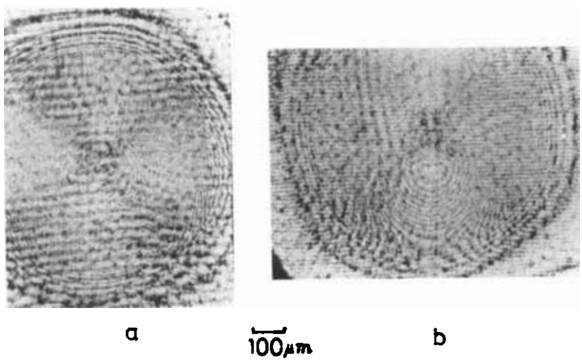

FIGURE 16 Holographic interferogram of a bubble domain in case of horizontally (a) and vertically (b) linearly polarized incident rays.

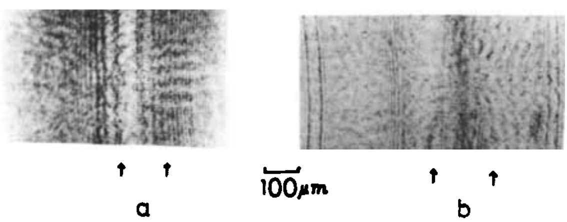

FIGURE 17 Holographic interferogram of a striped domain in case of horizontally (a) and vertically (b) linearly polarized incident rays. The arrows show the inside boundaries.

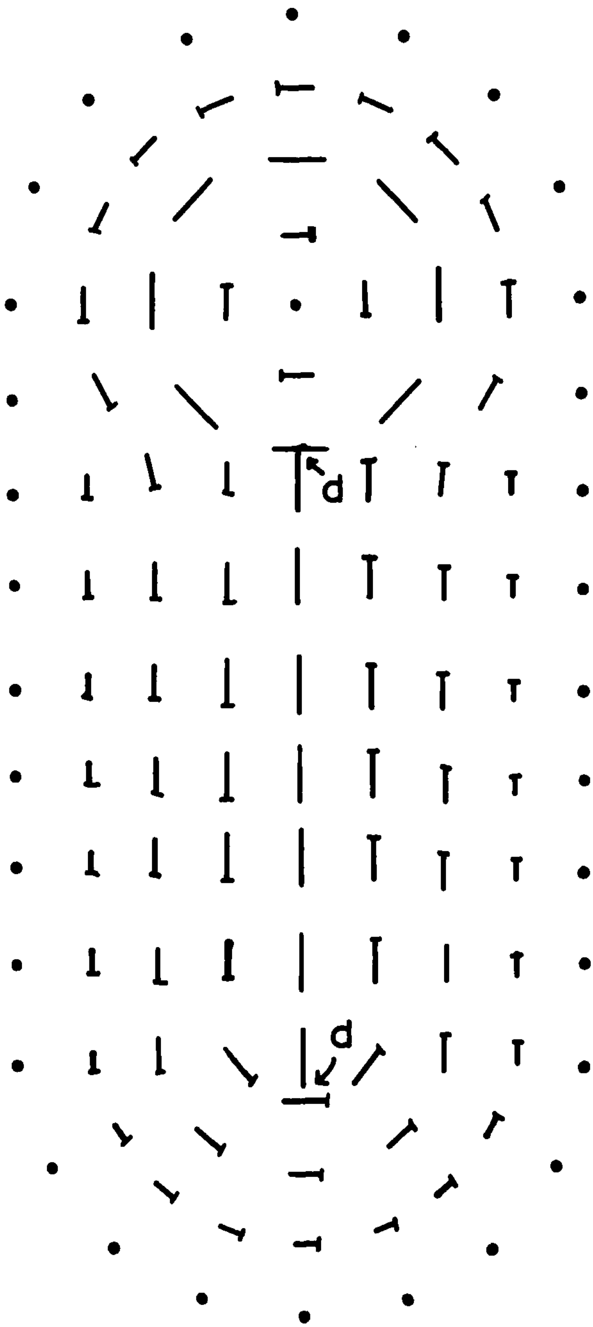

FIGURE 18a Conventional models of striped (a) and bubble (b) domains. "*d*" shows the disclination.

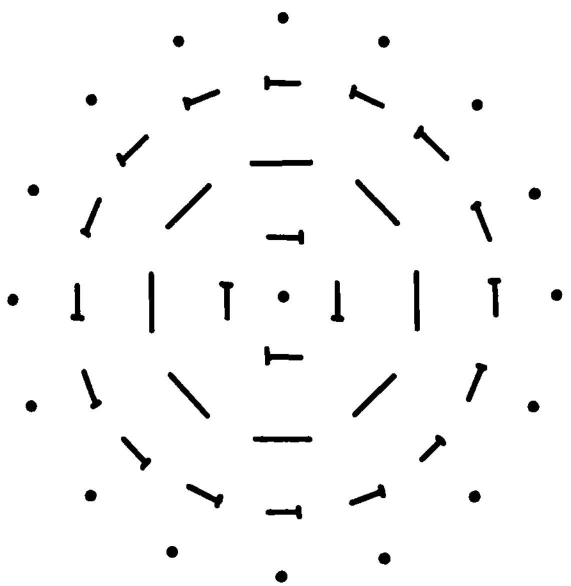

FIGURE 18b

Figure 23 shows the molecular alignment on the middle horizontal plane of the "thigh" part of striped domains. The alignment of the striped domain with the looped striped domain is shown in Figure 24.

4.2 Defects of the conventional models

We show the defects of the conventional models in order.

i) Though the inside boundary of the striped domain is light under the crossed nicols (see 3.1, ii), we should see the whole striped domain is dark on the conventional model (Figure 18a).

ii) The experimental results of the black brushes of a looped striped domain (Figure 12) is contradictory to the predicted dashed curve of Figure 22(a).

iii) The distribution of the refractive index obtained by the observation of Figure 10(a) is inconsistent with that calculated by the conventional models.

iv) We have two types of the end of the striped domains. One has a singularity and the other has no singularity. On the conventional models we can not explain the appearance of a singularity, because the end with a singularity in unstable comparing with the end without singularity from the energetic point of view.

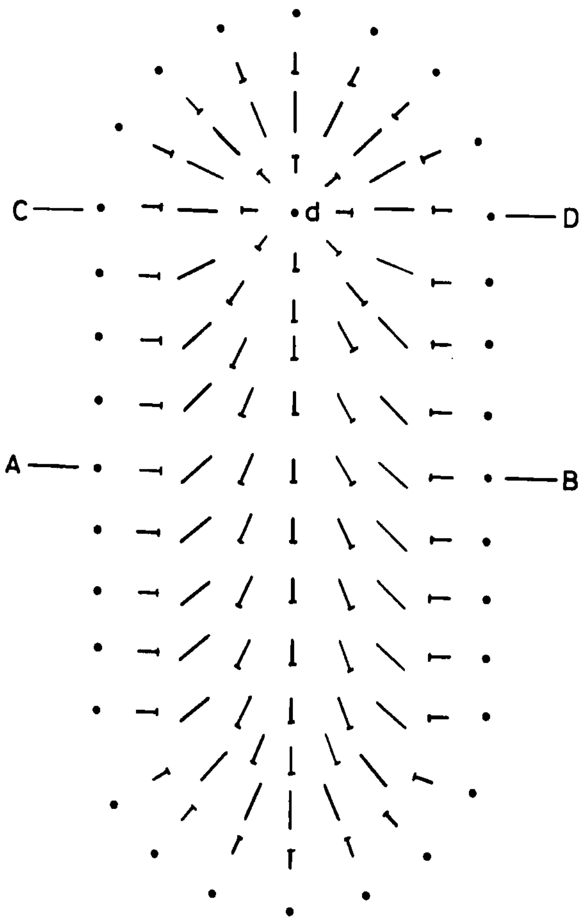

FIGURE 19 New model for striped domain.

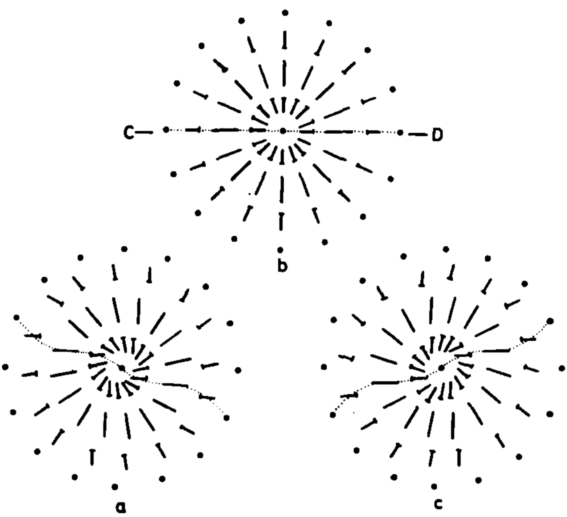

FIGURE 20 New model for bubble domain. a: upper plane, b: middle plane, c: lower plane. The dashed curve shows the same orientation of the profection of directors on the glass plane.

v) There are two disclinations at "*d*" parts in Figure 18(a). We can not observe such disclinations really. If we had such disclinations, the striped domain would be unstable from the topological point of view.

vi) Figure 25(a) shows the contour lines of the distribution of the refractive index of bubble domains in case of the vertically linearly polarized incident ray calculated by the conventional model. This is inconsistent with the experimental results with the holographic microscope, Figure 16(b).

vii) The conventional model of a bubble domain can not explain the feature of it shown in Section 3.1, i).

4.3 Estimation of new models

New models do not have the defects discussed in Section 4.2. We show the superiority of new models and estimate the stability of new models from the energetic and topological points of view.

i) Figure 25(b) shows the contour lines of the distribution of the refractive index of new models. This is consistent with the experimental results shown in Figure 16(b).

ii) We made the differential interference pattern roughly by the construction using Figure 25(b). The results are shown in Figure 26. The direction of the

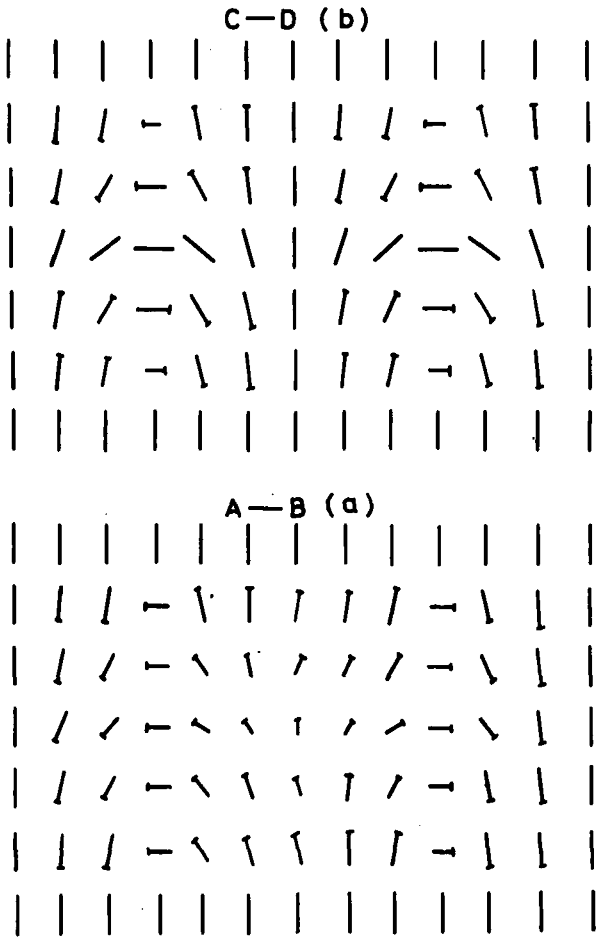

FIGURE 21 Vertical sections for a striped (a) and bubble (b) domain. (See Figures 19 and 20)

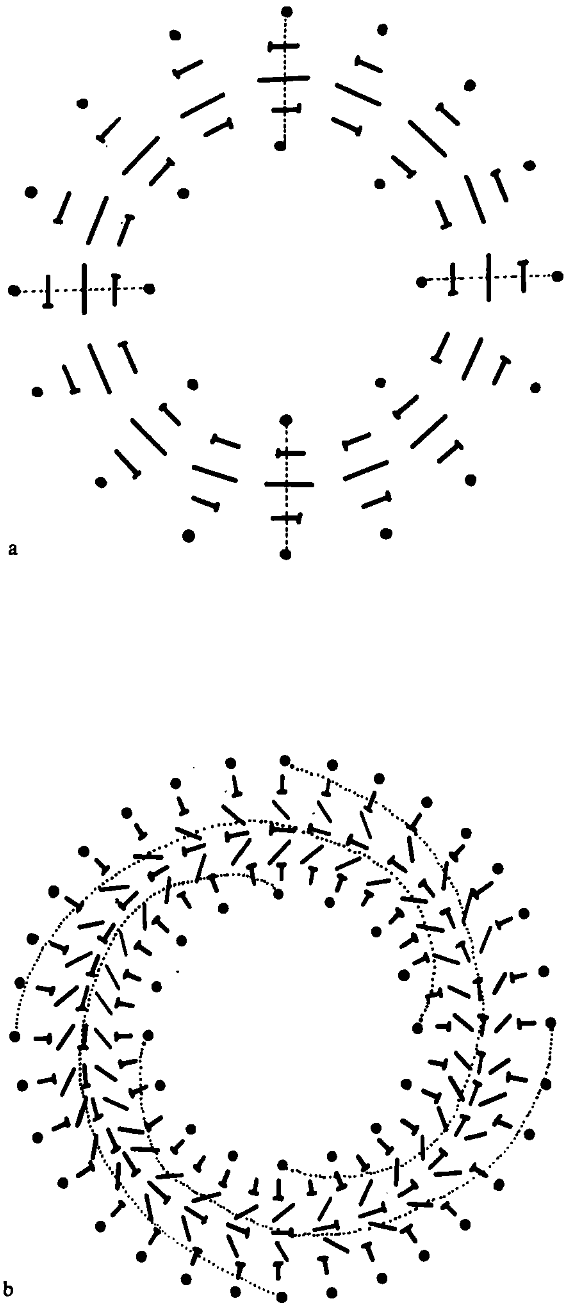

FIGURE 22 Conventional (a) and new (b) models of a looped striped domain. The dashed curve represent the black brushes that should be observed with the parallel nicols.

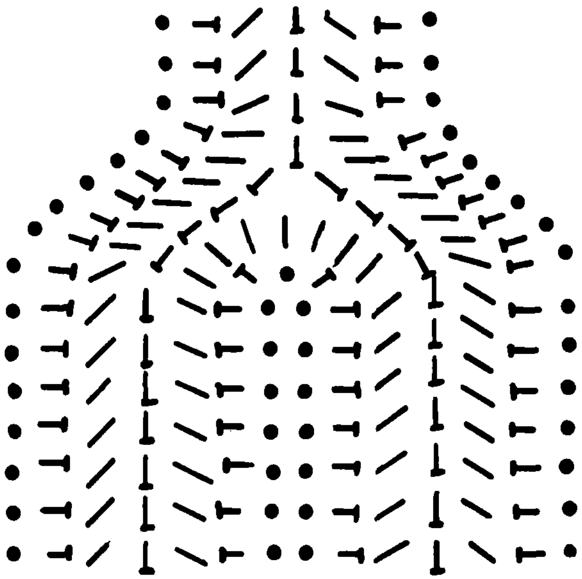

FIGURE 23 New model of a "thigh" part of a striped domain.

arrow above each figure shows the vibrational direction of linearly polarized incident ray. The dashed curves represent the light center ring and the outer boundary of a bubble domain. The results agree with the experimental results of Figures 13(b) and (c), respectively.

iii) The unknown property of a striped domain is led from the new model. This is the property concerning with the necessity of the appearance of a singularity in a striped domain as follows. "A striped domain must have a singularity in its domain or must be looped."

If a striped domain is not looped, we have two types of its end like Figure 19. In the upper end we must have a singularity at (d) but not in the lower end. Because the singularity have high energy, there is only one singularity in a striped domain so that a striped domain is stable. It does not have to have a singularity at its end. We can observe such a striped domain as has a singularity in its middle part. If it is looped, it is stable without singularities (see Figure 24).

This property is confirmed by the microscopic observation of many striped domains. Even a complicated striped domain has a singularity.

iv) The black brushes observed with the parallel nicols in a looped striped domain coincide with the dashed curve shown in Figure 22(b).

v) We discuss the twist structure of new models to estimate its energetic stability. Seeing Figure 21, we recognize that there is a little right rotational twist

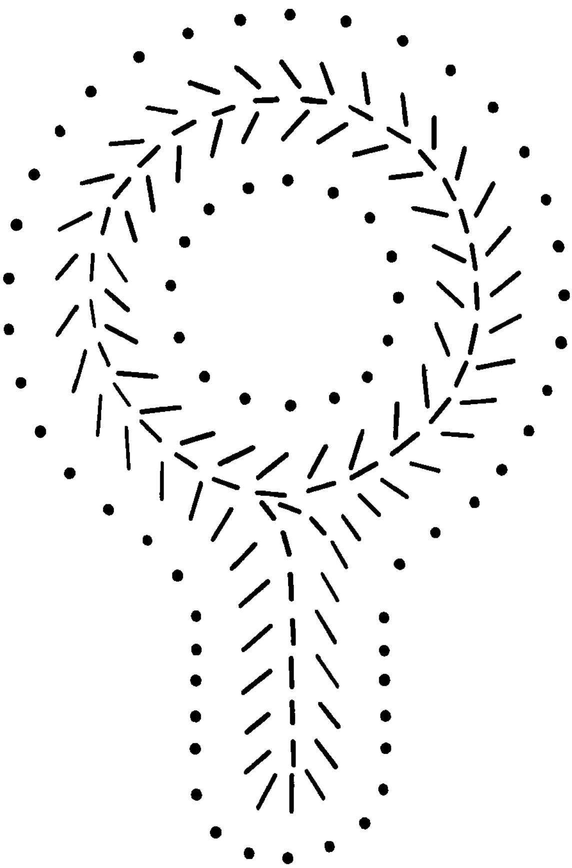

FIGURE 24 New model of a striped domain with a looped striped domain.

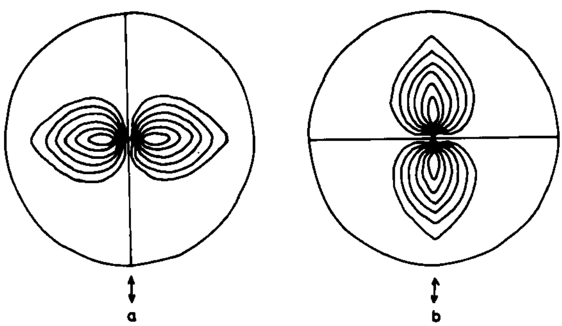

FIGURE 25 Contour lines of the distribution of refractive index of a bubble domain in case of the vertically linearly polarized incident ray calculated by the conventional (a) and new (b) models.

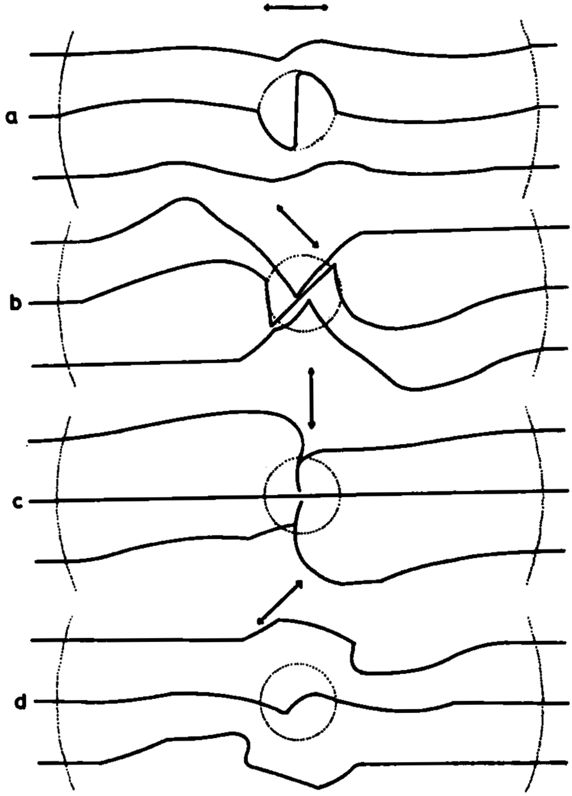

FIGURE 26 Differential interference pattern of a bubble domain estimated by the new model. The dashed curves represent the light center ring and the outer boundary. The arrow above each figure shows the direction of the polarization.

structure from top to bottom or from bottom to top in the new model of a bubble domain. There is also such twist structure in the new model of a striped domain shown in Figure 21(a). We recognize that the helical axis of a striped domain and a bubble domain is perpendicular to the cell boundary. This is the reason why the new models are stable from an energetic point of view.

Comparing Figure 18(a) with Figure 19, we can see that there is no disclination which is the cause of the topological instability in the new model. In addition to this, it is natural for a striped domain to have a bubble domain in the new model as shown in Figure 19. This reason has been already mentioned at iii) in this section.

After all, we can recognize that the new models are stable from energetic and topological points of view.

5 CONCLUSIONS

Observing carefully the striped domain and the bubble domain with three kinds of microscopes, we found that the conventional molecular alignment models are inconsistent with the observation.

We proposed the new models that could explain well the observation, and recognized their stability in the energetic and topological points of view.

Acknowledgment

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