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Effects of Magnetic Field on Domain Formation, in Nematic Liquid Crystal MBBA

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Magnetic field effects on a threshold voltage of domain formation and domain width are studied for nematic liquid crystal MBBA. A diffraction pattern appearing when the domains are illuminated with laser beam is observed to study the effects. Magnetic field perpendicular to the electric field increases the threshold voltage and decreases the domain width. The magnetic field has a stabilizing effect on the electrohydrodynamic instability. Experimental results obtained in this case are compared with the Helfrich theory. On the other hand, a magnetic field parallel to the electric field decreases the threshold voltage and increases the domain width. It is found that a critical magnetic field exists for this destabilizing effect. Above the critical field, the diffraction pattern is not observed at any electric field. Optical transmission also is measured.

1 INTRODUCTION

It is well known that orientation pattern domains can be observed when a dc or ac voltage above a threshold is applied to a nematic liquid crystal which has negative dielectric anisotropy ($\varepsilon_{\parallel} < \varepsilon_{\perp}$) and positive conductivity anisotropy ($\sigma_{\parallel} > \sigma_{\perp}$).[1–8] The ac excitation of low frequency is called the "conduction regime."[2] Domains appearing under this regime are called Williams domain. The Helfrich theory predicted that the threshold voltage V_{th} of domain formation and the spatial wave vector[9] q vary when liquid crystal is placed in a static magnetic field. The voltage change has been confirmed experimentally by Orsay Group[2,10] and Teaney *et al.*,[8] for magnetic field(H) perpendicular to electric field(E). That is, domain formation occurs at a higher voltage for a higher magnetic field. In this case, magnetic field has the effect of stabilizing the electrohydrodynamic instability associated with the

occurrence of domains. Furthermore, Teaney *et al.*, found a noticeable reduction in the basic cycle width for $H \perp E$, although not mentioned in detail. For $H \parallel E$, experimental results of V_{th} and q have not been explicitly given. We have observed for $H \parallel E$ that the domain formation occurs at a lower voltage and the spatial wave vector q decreases. Therefore, in this case, the magnetic field has the effect of destabilizing the electrohydrodynamic instability.

In a present paper, we have quantitatively studied the threshold voltage V_{th} and the domain width d (approximately π/q) as a function of magnetic field by observing the diffraction pattern due to domain periodicity with laser beam. Another experiment also was performed on relation between optical transmission and magnetic field. As a result, some effects that were not, so far, reported have been found.

2 EXPERIMENTS AND RESULTS

In this experiment, we used nematic liquid crystal MBBA which has negative dielectric anisotropy ($\varepsilon_{\parallel} < \varepsilon_{\perp}$), positive conductivity anisotropy ($\sigma_{\parallel} > \sigma_{\perp}$) and positive permeability anisotropy ($\chi_{\parallel} > \chi_{\perp}$). We tested a sandwich cell with a layer of MBBA between two transparent Nesa coated glasses. The preferred orientation of molecules was performed by rubbing the glass surface coated with surfacant prior to the cell construction. Two plates of glasses were arranged so that the rubbing direction was parallel. The magnetic field was applied as shown in Figures 1 and 2. The spacing was determined by Mylar spacers from 12.5μ to 100μ, which were fixed onto the glass surface with binder.

When an ac electric field of 50 Hz was applied, domains were microscopically observed like uniform stripes perpendicular to the rubbing direction. A He–Ne laser beam (6328A) was normally incident on the liquid

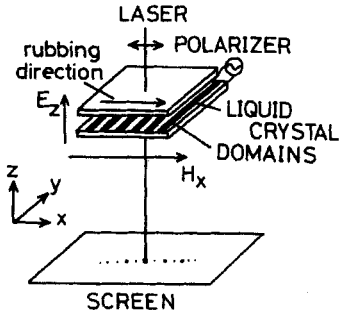

FIGURE 1 A schematic diagram for observing the diffraction pattern when $H \perp E$.

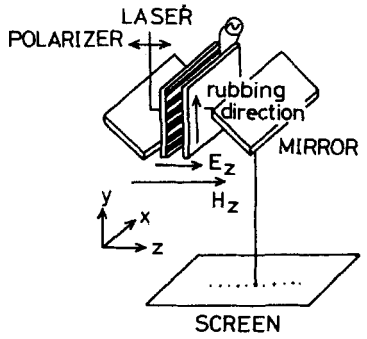

FIGURE 2 A schematic diagram for observing the diffraction pattern when $H \parallel E$.

crystal through the glass surface. A farfield diffraction pattern could be observed on the backward screen which was placed far from the cell. The magnetic field was applied perpendicular (Figure 1) or parallel (Figure 2) to the electric field. In the case of $H \parallel E$, two parallel mirrors were employed to observe the diffraction pattern.

The relation between fringes of diffraction pattern and the domain width can be considered as follows.

The fringe maxima occur when a following equation can be satisfied.[11]

$$\left| \frac{\sin(N\pi ks/2q)}{\sin(\pi ks/2q)} \right|^2 = 1 \tag{1}$$

where $s = \sin\theta$ with the diffraction angle θ, N is the slit number, k the wave vector of incident light and q the spatial wave vector of orientation pattern. The fringes of extremely strong intensity appear where

$$ks/2q = n,\ n;\ \text{integer} \tag{2}$$

Since $k = 2\pi/\lambda$ and $s = x/r$,

$$q = \frac{ks}{2n} = \frac{\pi x}{n\lambda r} \tag{3}$$

where λ is the wavelength of incident light, r the distance between the cell and screen, and x the spacing between the 0th order fringe and the nth order fringe. There is a relation between spatial wave vector q and the wavelength of spatial periodicity λ_t, $q = 2\pi/\lambda_t$.[6] Therefore, the wavelength can be determined as follows.

$$\frac{\lambda_t}{2} = \frac{n\lambda r}{x} \tag{4}$$

We have taken the domain width d as $\lambda_t/2$, since we know $\lambda_t/2$ from microscopic observations of the stripe spacing.[12] The diffraction pattern is observed as a superposition of contributions from gratings with π/q and $2\pi/q$.[12] So, we must take even values of the fringe number, n, to calculate the domain width. From Eq. (4), the fringe spacing is found inversely proportional to the domain width.

The domain widths obtained from Eq. (4) are compared with those obtained from the microscopic observation as shown in Table I, where the threshold voltage were applied for zero-magnetic field. For the microscopic observation, the domain width was calculated from the number of domains which came within the field of vision. Thus, there is an accidental error of about $\pm 2\mu$. Table I shows that the determination of the domain width from Eq. (4) is of high accuracy. Under the application of magnetic field, the direct observation of domains with the microscopy was not available. In our experiment, therefore, the fringes were observed in order to determine the domain width.

TABLE I

Comparison of domain widths by two methods

SPACER THICKNESS (μ)	DOMAIN WIDTH(μ)	
	a	b
12.5	10.6	10.4
12.5	10.2	9.5
25	19.2	20
25	18.9	20
50	38	40
100	73.5	80

a : domain width obtained from diffraction pattern

b : domain width obtained from microscopy

When no magnetic field was applied, the threshold for the domain formation was 6–7 V and almost independent on the sample thickness, although the value fluctuated within 0.5 V with the sample thickness. This was already reported.[1,6]

In the following section, we will describe the dependence of V_{th} and d on magnetic field.

In the case of $H \perp E$

Application of the voltage V_{th} to the liquid crystal caused the formation of domains. The diffraction pattern due to the domain periodicity is repre-

sented in Figure 3(a), where no magnetic field is applied. When a magnetic field of 2.6 K Gauss was applied parallel to the X axis (see Figure 1), the diffraction pattern disappeared. However, at a higher voltage, the diffraction pattern appeared again as shown in Figure 3(b). In this case the fringe spacing is wider than that in Figure 3(a). As the magnetic field strength was further increased, both the threshold voltage and the fringe spacing increased. The diffraction patterns for a higher magnetic field and accordingly for a higher threshold voltage are shown in Figures 3(c) and (d). The spacing of fringes becomes gradually wider with increasing the magnetic field as reported

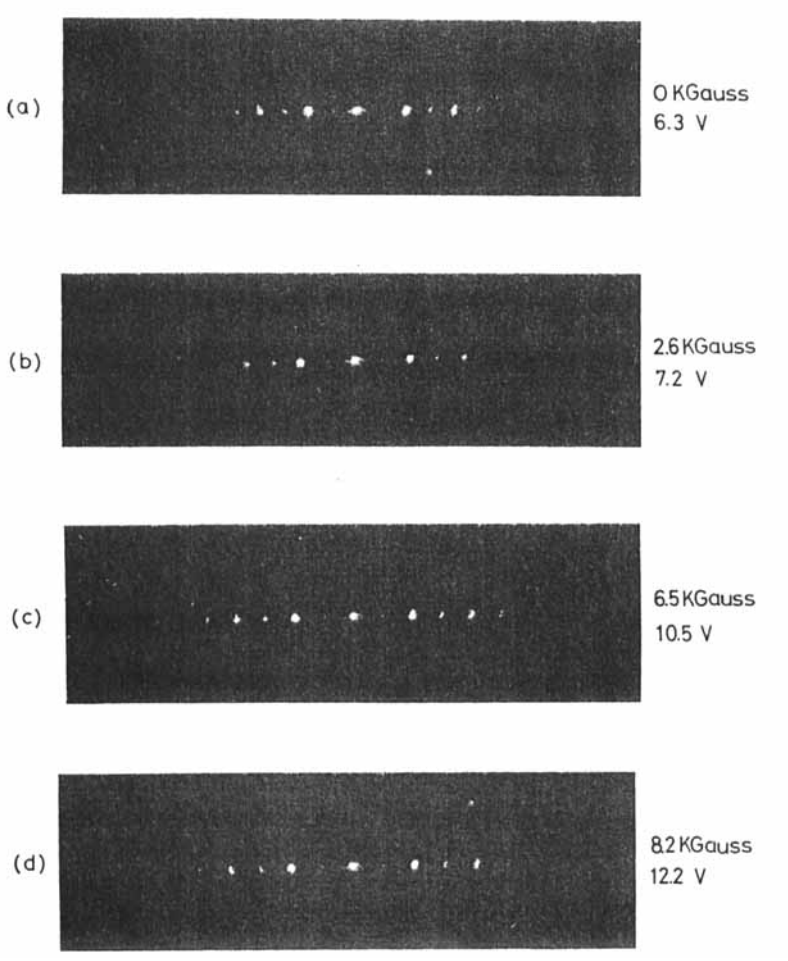

FIGURE 3 Diffraction pattern at threshold voltage when $H \perp E$. Magnetic field intensity and threshold voltage at that time are denoted. MBBA : 25 μm.

previously.[8] Relation between the magnetic field H and the domain width d is shown in Figure 4. The domain width changes greatly for the thick sample

When the initial preferred alignment of molecules was adjusted paralle to the Y axis, the observed diffraction pattern rotated around the Z axis with increasing the magnetic field as already reported.[13] The fringe spacing also increased compared with that observed under no magnetic field. With further increasing the magnetic field, the direction of the pattern was parallel to the magnetic field and thereafter the case was similar to one described above.

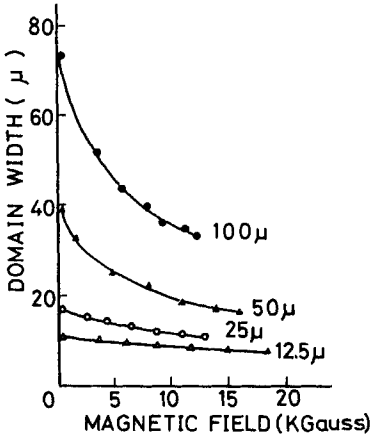

FIGURE 4 Domain width (μm) as a function of magnetic field (K Gauss) for different thicknesses of MBBA when $H \perp E$.

In the case of $H \parallel E$

When a magnetic field was applied parallel to the electric field (see Figure 2), domains were formed at a lower threshold voltage than that required for no magnetic field as shown in Figure 5. In other word, the magnetic field introduces a preferable condition of forming domains. However, it should be noted that there is an upper limit of the magnetic field strength, H_c, for this condition. Diffraction patterns for $0 \leq H \leq H_c$ are shown in Figure 6, where the threshold voltages also are represented. This shows that the field has the destabilizing effect in this region. Also, it is found that the spacing of fringes decreases compared with that observed under no magnetic field. The domain width increases with the magnetic field as shown in Figure 7. Moreover, the domain width d is approximately same to the sample thickness L near the critical field H_c. At a higher field $H > H_c$, well aligned domains do not appear for any voltage.

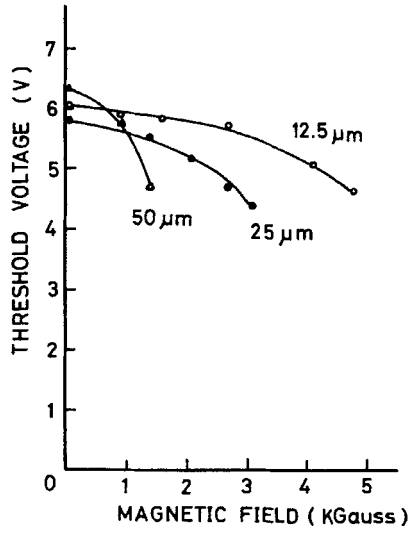

FIGURE 5 Relation between magnetic field H_z and threshold voltage V_{th} for different thicknesses of MBBA when $H \parallel E$. Threshold voltage is decreased until a critical field with increasing the field.

We have also observed another phenomenon. When an applied voltage was slightly lower than the threshold, the diffraction pattern, of course, was not observed. However, when a magnetic field was applied, the pattern appeared under the same voltage. With increasing the field, the pattern disappeared again. With further increase in the field, liquid crystal became transparent since all molecules aligned in the direction of the magnetic field.

Change in optical transmission due to magnetic field

The light intensity transmitted through the liquid crystal was rather strong (clear for eyes) at a lower voltage. With increasing the voltage, onset of decrease in the light intensity took place at a critical voltage V_c. With further increase by 2 or 3 Volts, liquid crystal turned to exhibit the dynamic scattering mode as well known and the intensity ratio decreased by about 70% (turbid). The voltage V_c changed with the applied magnetic field as shown in Figure 8.

In the case of $H \perp E$, V_c increases with H as shown previously.[8] In the case of $H \parallel E$, V_c decreases until a certain value H_c' and increases above H_c'. So far, the latter case of $H \parallel E$ has not been reported and discussed.

When the threshold voltage V_{th} for domain formation is compared with V_c in this section, it is found that both values have approximately the same value for $H \perp E$ and $H < H_c (H \parallel E)$. This leads to a conclusion that V_c is related to the domain appearance. Moreover, the value of H_c' is smaller for

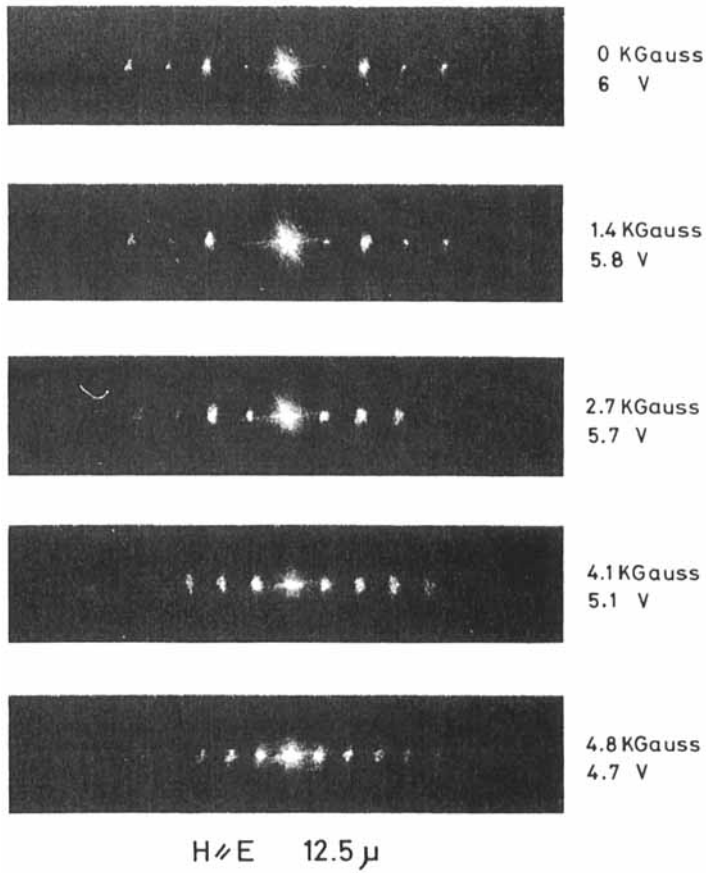

FIGURE 6 Diffraction pattern at threshold voltage when $H \parallel E$. Magnetic field intensity and threshold voltage at that time are denoted. MBBA : 12.5 μm.

a thicker sample and is found to be the same with H_c described in the preceding section.

As we described above, any distinguishable diffraction pattern did not appear for $H > H_c$ ($H \parallel E$). However, for $H > H_c'$, there still exists a critical voltage, at which the liquid crystal begins to change from clear to turbid state as shown in Figure 8. Teaney *et al.*,[8] have discussed the transverse fluctuations of the nematic director from their conductivity measurements in this region ($H > H_c$). Our result observed also may be associated with the fluctuations. That is, initial alignment of the liquid crystal molecules is oriented perpendicular to the glass surface for the higher magnetic field. The dielectric effect ($\varepsilon_{\parallel} < \varepsilon_{\perp}$) by the electric field and a subsequent effect of the conduction anisotropy cause a turbulence to the initial orientation. The

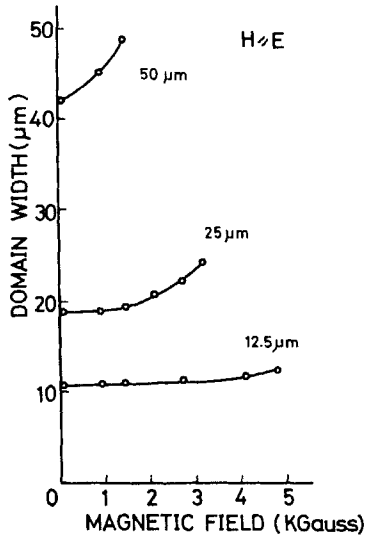

FIGURE 7 Domain width (μm) as a function of magnetic field (K Gauss) for different thicknesses of MBBA when $H \parallel E$.

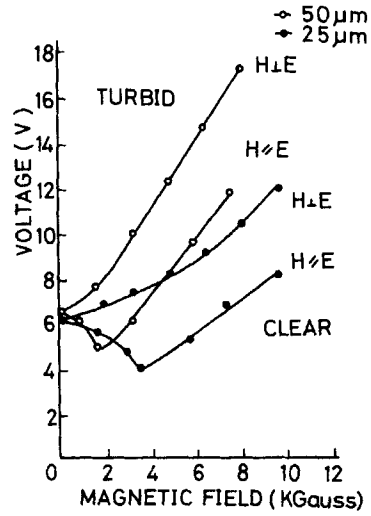

FIGURE 8 Relation between optical transmission and magnetic field for MBBA when $H \perp E$ and $H \parallel E$. Voltages at which the transmitted light intensities begin to decrease sharply are plotted. On the upper side of each curve liquid crystal is turbid and on the under side clear. It is characteristic of $H \parallel E$ that the curve has a dip.

required voltage for it may be V_c. So, the higher electric field is required for the higher magnetic field.

When a magnetic field is increased gradually under a constant voltage V (for example $5V$ in Figure 8) in the case of $H \parallel E$, the liquid crystal changes form clear → turbid → clear. This observation was described in the preceding section.

3 DISCUSSION

We have carried the work to quantitative measure of threshold and domain spacing. As a result, comparison between the theoretical prediction[9] and experimental results can be made.

In the case of $H \perp E$, the threshold voltage increased and the domain width decreased with the magnetic field. Helfrich has theoretically approached this stabilizing phenomenon. He has reduced a following equation concerning the threshold for domain formation in the case of $H \perp E$.[9]

$$-AE_z^2 + \chi_a H_x^2 + k_{33} q^2 = 0 \tag{5}$$

where A is a constant associated with the conductivity anisotropy and the dielectric susceptibility anisotropy, $\chi_a (= \chi_{\parallel} - \chi_{\perp})$ the permeability anisotropy and k_{33} the Frank's elastic constant of bend. Since $q = 2\pi/\lambda_t$,

$$V_{th}^2 = V_0^2 \frac{L^2 H_x^2}{\frac{k_{33}}{\chi_a}\pi^2} + V_0^2 \left(\frac{2L}{\lambda_t}\right)^2 \tag{6}$$

where $V_0 = (k_{33}\pi^2/A)^{1/2}$ is the threshold voltage for no magnetic field and L the sample thickness. Orsay group has treated q to be constant as $q \simeq \pi/L$.[10] In fact, for a higher magnetic field the second term $V_0^2(2L/\lambda_t)^2$ in Eq. (6) may be neglected compared with the first term. Then, the threshold voltage V_{th} may be proportional to H_x. The experiment by Orsay group seems to be made under this condition. In our experiment, however, the domain width d even in a range of higher magnetic field decreased with increasing the magnetic field. In order to compare our result with the Helfrich theory, we rewrite Eq. (6),

$$\lambda_t^2 V_{th}^2 = \frac{V_0^2 L^2}{\frac{k_{33}}{\chi_a}\pi^2} \lambda_t^2 H_x^2 + V_0^2 (2L)^2 \tag{7}$$

where the second term $V_0(2L)^2$ is a constant. When the relation between $\lambda_t^2 V_{th}^2$ and $\lambda_t^2 H_x^2$ is checked, a linear relationship is found as shown in Figure 9. It should be noted that λ_t is not constant but variable. Thus, our experimental

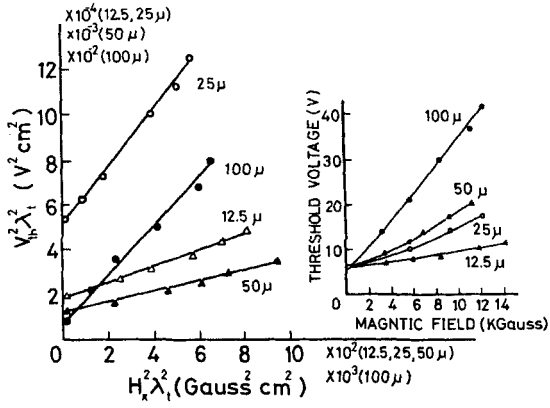

FIGURE 9 Relation between magnetic field H_x and threshold voltage V_{th} for different thicknesses of MBBA. Relation is linear when variable factor λ_t is taken into account. Note that the scales are differently taken for the thickness.

result for $H \perp E$ can be well explained by the Helfrich theory. We also calculated k_{33}/χ_a from the slope of straight lines in Figure 9. The value is 8–18 c.g.s., which is a little larger than that reported previously.[13,14]

On the other hand, the magnetic field parallel to the electric field ($H \parallel E$) caused the decrease in the threshold voltage and the increase in the domain width; this effect was not dealt with in the Helfrich theory. Helfrich reduced an equation for $H \parallel E$ on the assumption that the magnetic field has only the stabilizing effect.[9]

$$-BE_z^2 + \chi_a H_z^2 + k_{11}q^2 = 0 \tag{8}$$

where B is a constant associated with the conductivity anisotropy and the dielectric susceptibility anisotropy and k_{11} is the Frank's elastic constant of spray. We find this effect for the magnetic field $H \geq H_c$. So, Equation (8) may be modified to be applicable in this region.

$$-BE_z^2 + \chi_a(H_z - H_c)^2 + k_{11}q^2 = 0 \tag{9}$$

For the lower magnetic field, $H < H_c$, the destabilizing effect of the field can be explained to a certain extent with modification of the Helfrich theory applicable for $H \perp E$ by putting $-H_z^2$ in place of H_x^2 in Eqs. (5), (6), and (7). In this extended equation, it is easily shown that a critical value H_c should exist for V_{th}^2 to take a positive value. At the field H_c, Freederick's transition probably occurs. Freederick's transition occupies at the field $H_c = \pi/L\sqrt{k_{11}/\chi_a}$.[15,16] This formula predicts that the critical magnetic field H_c is proportional to the inverse sample thickness. The experimental result shows this relationship (Figure 10). The calculated k_{11}/χ_a is 7.8 c.g.s., which is in good agreement with those of other workers.[13]

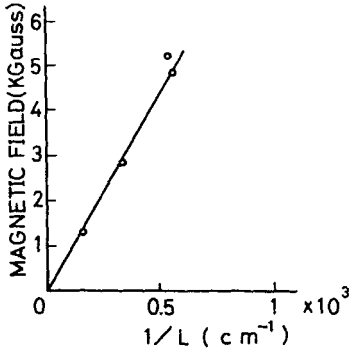

FIGURE 10 Relation between H and L^{-1} (inverse sample thickness).

Moreover, there are relations, $d \propto V_{th}^{-\beta}$ (for both cases $H \perp E$ and $H \parallel E$) where $\beta = 0.4 \sim 0.5$, between the domain width d and the threshold voltage V_{th} as shown in Figure 11. Also, it is known [3] that ac field frequency changes V_{th} and d and those are related to each other as $d \propto V_{th}^{-0.4}$. It should be noted that the relations are strikingly similar, though both physical origins differ from each other. This seems to indicate that the domain spacing change depends essentially upon the threshold voltage change due to the magnetic field. The domain width may be closely related with the molecular velocity of the vortex movement;[9,17] the higher velocity corresponds to the larger spatial wave vector. At a fixed voltage, the vortex velocity may be changed

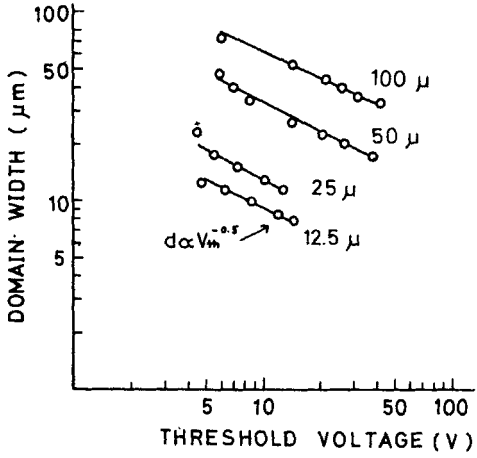

FIGURE 11 Dependence of domain width d on threshold voltage V_{th} for different thicknesses of MBBA. The threshold voltage is changed with magnetic field as shown in Figures 5 and 9.

under magnetic field as a result of the stabilizing ($H \perp E$) or destabilizing ($H \parallel E$) effect. Grueubel *et al.*, experimentally showed that $d \propto V^{-1}$ under no magnetic field.[18] If the velocity were reduced superlinearly by the field, the domain width is related to the voltage with power smaller than unity. Further study must be made to reveal the factors that determine the domain width.

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