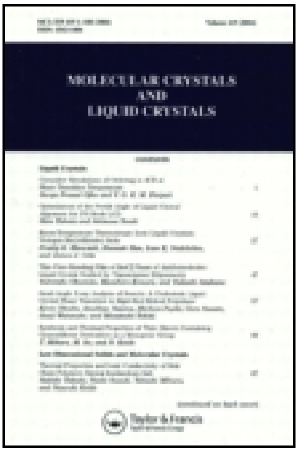

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The Role of Goldstone Mode and Electroclinic Effects in Electromechanical Responses of Chiral Smectic C Liquid Crystals

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The Role of Goldstone Mode and Electroclinic Effects in Electromechanical Responses of Chiral Smectic C Liquid Crystals†

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We studied the electromechanical effects of smectic C* sandwich cells. Some earlier results are reviewed and new measurements are reported of the motions of the cover glass as a function of frequency and voltage for different textures (chevron, striped and uniform bookshelf). The analysis of the observations show that there are two mechanisms which dominate the linear electromechanical effects: the coupling between field induced director rotation (Goldstone mode) and flow and the electroclinic effect. The Goldstone mode is most strongly excited in unwound smectic bookshelf or chevron textures when the spontaneous polarization is parallel to the substrate. It causes a horizontal motion of the cover plate. The electroclinic effect induces a vertical motion. It is most strongly excited in an unwound bookshelf or chevron texture when the polarization is vertical. The mechanical responses show resonances in the kilohertz frequency range, in particular for the vertical responses. We found that the resonances correspond to eigenmodes of the glass plates.

Keywords: ferroelectric liquid crystal, electromechanical effects, electroclinic effect, goldstone mode

I. INTRODUCTION

Linear electric field induced mechanical motions or distortions (linear electromechanical effects) and the converse, distortion induced electric polarization (piezoelectric effects) are confined to materials without a center of symmetry. This includes of course all ferroelectric materials. A mechano-electric effect in a liquid

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crystal was first observed in 1975[1] with a ferroelectric smectic C* phase in form of a shear flow induced polarization. The electroclinic effect (electric field induced tilt) was first observed in 1977 with a smectic A* phase.[2] Vibrations of the cover plate induced by a linear electromechanical (LEM) effect of a smectic C* film were first reported in 1985.[3] Since then more extensive experimental[4] and theoretical[5] work has been published.

The electromechanical response of smectic C*'s can be strong and audible. Its strength depends on the texture. Films with a homeotropic texture, where the smectic layers are in plane of the substrate, do not show a linear response for perpendicular fields, in contrast to chevron and bookshelf textures.

The "chevron" structure[6] is commonly obtained in the smectic C* films with a planar alignment coating (e.g. buffed polyimide, PVA or nylon coating). In this texture the layers are not normal to the glass plates but inclined, by an angle of typically 15°, in opposite directions on the two surfaces so that a kink forms in the mid-plane (see Figure 1b). For strong anchoring at the surfaces and with an inclination angle nearly equal to the director tilt angle, the polarization in chevron textures is nearly horizontal.

Some chevron texture can be transformed to a striped bookshelf structure by the application of strong electric fields of low frequency ($E > 10$ V/μm, $f \sim 10$ Hz), where the layers are perpendicular but zigzagging parallel to the substrate, with a periodicity that is typically in the order of the sample thickness.[7-9] The spontaneous polarization is vertical. A striped texture can also form spontaneously from the chevron texture.[10] This relaxed striped texture is different from the electrically induced striped bookshelf texture. The stripe width is about an order of magnitude larger, and the polarization alternates between "up" and "down" (see Figure 1c). Uniform bookshelf texture with vertical layers without kinks and with a nearly vertical spontaneous polarization (see Figure 1a) can be obtained with special naphthalene derivatives using a polyimide coating[11] and with an other strongly polarized material using a silane coating and enforcing the bookshelf texture by applying an electric field and simultaneous shearing.[12]

In this paper we review and report new experimental results and show that two basic molecular mechanisms are mainly responsible for the observed linear electromechanical responses of smectic C* cells.

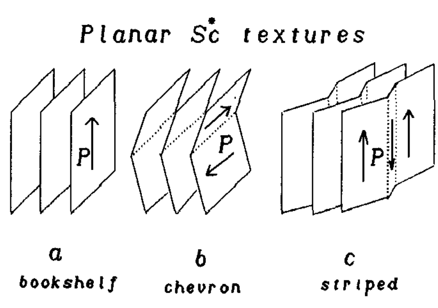

FIGURE 1 Structure of S_C^* textures.

II. THEORETICAL CONSIDERATIONS

We use the commonly accepted model of the C* phase. The director gives the preferred alignment of the molecular axes which is tilted against the layer normal. The spontaneous polarization is parallel to the layers and perpendicular to the director. There are two modes (see Figure 2) of director motion:

a) *The Goldstone mode.* Here the director rotates about the layer normal without a change of the tilt angle. The polarization P_0 rotates simultaneously without a change of magnitude. The mode may be excited by electric fields due to the torque $E \times P_0 = EP_0 \sin \phi$. The magnitudes of small oscillations are accordingly proportional to $\sin \phi$.

b) *The electroclinic effect.* It corresponds to a change of the tilt angle and of the magnitude of the permanent polarization. The polarization is proportional to the director tilt, therefore an electric field parallel to the polarization increases the director tilt angle. The induced change is proportional to the field and proportional to $\cos \phi$.

The Goldstone mode induces flow parallel to the layer planes which exerts a horizontal force on the cover plates.[13] For unwound bookshelf or chevron texture, the corresponding stress is given by

$$\sigma_{xy} = (1/2)(\gamma_2/\gamma_1 + 1)EP_0 \sin \phi \qquad (1)$$

γ_1 is the rotational viscosity and $\gamma_2 = \eta_b - \eta_c$, where η_b and η_c are the shear viscosities with the directors in the direction of the flow gradient and parallel to the flow, respectively. Note that the stress is independent of the frequency which implies that the acceleration for the horizontal motion is also frequency independent.

The electroclinic effect has a direct mechanical consequence. The change of the tilt angle is coupled to changes of the layer distance and therefore with a change of dimensions. As the layer distance changes the layers expand to keep the volume

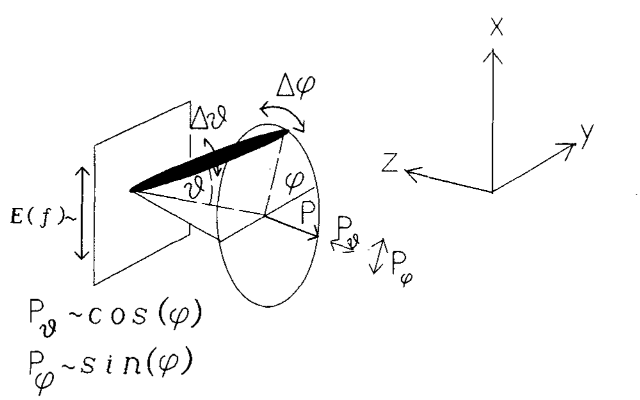

FIGURE 2 Field induced polarization changes in S_C^* liquid crystals with a vertical layer structure due to ac electric fields. P_θ magnitude change of polarization due to change in tilt angle, P_ϕ orientation change of polarization due to c director rotation.

constant. This gives a vertical force on the cover plate which causes the vertical motion.

The direction of the polarization in unwound samples determines the direction of the induced plate motion. For a horizontal polarization the induced motion is mainly horizontal and due to the viscous coupling between director rotation and flow, and for a vertical polarization the induced motion is vertical and due to the electroclinic effect. It may be noted here that perfect helical bookshelf structures should not show a linear response.

III. EXPERIMENTAL ASPECTS

The measurements were made on 5 μm thick films sandwiched between glass plates (1 mm thick and 2 by 2 cm) that had conductive ITO coatings on the inner surfaces. The plates were separated by polystyrene balls and placed horizontally. The lower plate was fixed while the upper plate was free to move. The motion of the upper plate was monitored, using accelerometers (from Bruel & Kjaer), in three orthogonal directions, in x-direction perpendicular to the film, in y-direction parallel to the film and to the smectic layers, and in z-direction parallel to the buffing direction. The accelerations were measured with an accuracy greater than 0.1 mm/s^2 in the range of 0.2–7 kHz. The sample temperatures were controlled with an accuracy of 10 mK.

We used the following FLC mixtures in various planar alignments and compared the responses.

a) ZLI 4237-000 from E. Merck. The material has a relatively small polarization (P_0 = 7 nC/cm^2) and large pitch ($p > 40$ μm), so the LC films were unwound (surface stabilized). With rubbed polyimide coated surfaces and by application of a periodic shear, a bookshelf texture is obtained in the S_A^* range that changes to a chevron texture with an approximately horizontal spontaneous polarization on cooling to S_C^*. At room temperature the chevron texture transformed to a striped bookshelf texture[10] in which the polarization alternates with the stripes, approximately between "up" and "down."

b) FLC 6430 from Hoffmann LaRoche. The material has a large polarization (P_0 = 90 nC/cm^2) and a small pitch (p = 0.43 μm). The textures are helical but can be unwound by electric fields. Samples made with rubbed polyimide coatings gave a chevron texture in S_C^* which relaxed to a striped bookshelf texture. Silane coatings (Dow Corning, X1-6136) gave spontaneously homeotropic alignments with the layers parallel to the substrates. By the simultaneous application of field and shear the homeotropic textures could be realigned to a uniform bookshelf texture.[12] The uniform bookshelf texture was also stable and it could be unwound with ac fields of 2 V/μm.

IV. EXPERIMENTAL RESULTS

The frequency dependencies of the linear responses are relatively simple for ZLI 4237-000 films with chevron textures (see Figure 3). The response in y-direction

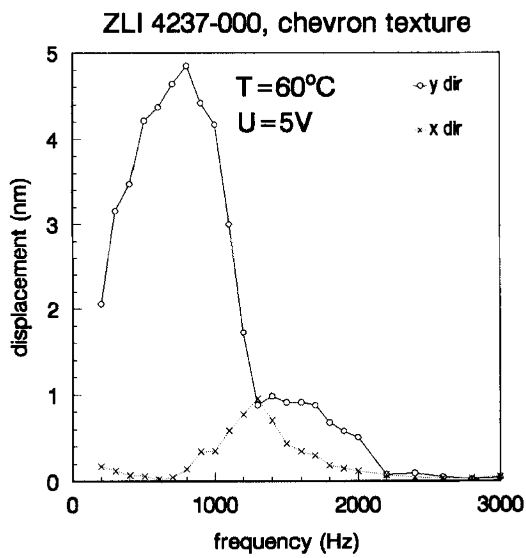

FIGURE 3 Chevron texture, ZLI 4237-000, displacement spectra in y and x-direction; $T = 60°C$ and $U = 5$ V.

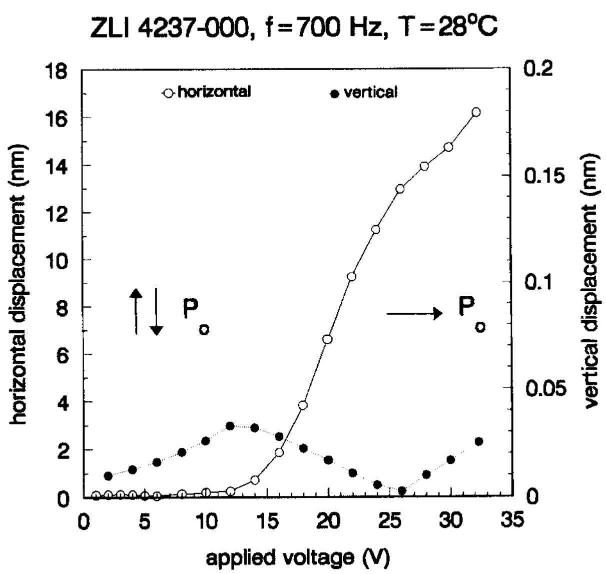

FIGURE 4 Striped bookshelf texture, ZLI4237-000, voltage dependence of x- and y-displacements $T = 28°C, f = 700$ Hz.

dominates. It is flat and small in z-direction, in x-direction it has a weak resonance-like peak around 1.1 kHz. The y-response peaks at about 800 Hz then it drops sharply to a weak minimum at 1.1 kHz. The minimum is probably due to some interference with the x-displacement modes.

The y-response decreases when the texture transforms from chevron with a horizontal polarization to striped bookshelf with a vertical polarization (see Figure 2, Reference 10). In microscopic studies[10] we found that the spontaneous polarization in the striped texture can be reoriented with ac fields. Above a frequency dependent threshold voltage the permanent polarization reorients from the nearly vertical polarization to a nearly horizontal polarization. This reorientation affects the displacements. We measured the displacements in x- and y-direction at fixed frequencies as the function of voltage. The results of such a measurement at 700 Hz is shown in Figure 4. At this frequency the reorientation takes place in the voltage range of 15–25 V. The y-response is very small up to about 10 V. In the range from 15–25 V it increases dramatically, while the vertical response drops.

We turn now to measurements made on FLC 6430 films with uniform bookshelf textures. As mentioned, these films have normally a helical structure, but the helix can be electrically unwound. The unwound texture was metastable[14] and remained unchanged typically for several minutes after removal of the field. It is therefore possible to unwind the texture with a larger voltage and measured its response later in the low voltage range.

In Figure 5 we compare the linear responses in y-direction of the helical texture and the unwound textures in the range from 0 to 10 V. The response for the unwound texture is about an order of magnitude larger than for the helical texture. More results for the unwound bookshelf texture are shown in Figure 6.

We measured the responses in the three earlier defined directions as a function of frequency and voltage. The response in z-direction is relatively small and not shown. The vertical x-response (Figure 6) has several, clearly separated resonance

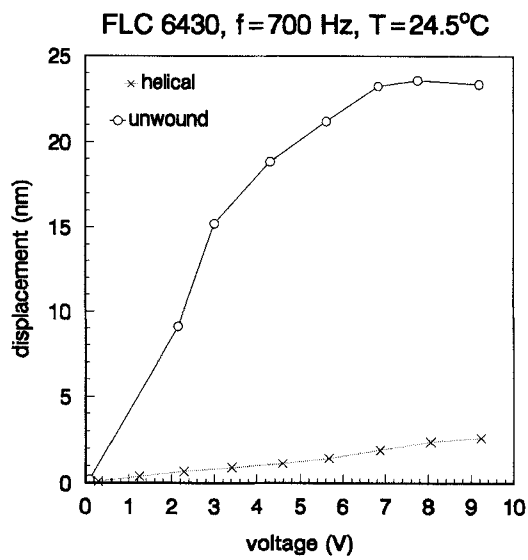

FIGURE 5 Helical and unwound uniform bookshelf texture, of FLC 6430, voltage dependences of y-displacements, T = 24.5°C, f = 700 Hz.

peaks. The horizontal response parallel to the layers, y-response, (Figure 6b) shows a complicated frequency dependence. It is relatively strong below 1.5 kHz but there is a deep dip near 600 Hz which is probably caused by interference with the very strong resonance peak at 600 Hz in the x-displacements.

The resonance-like properties of the 600 Hz peak are confirmed by phase shift measurements (Figure 7). We measured the displacement together with the phase shift. Across the peak the phase shift changes by about 180° as expected for a vibrational resonance. The resonance frequency shows no critical temperature dependence. On heating to the $S_A - S_C^*$ transition the resonance frequency decreases by less than 10%. Similarly, the variation of the width of the peak was less than 10% over the whole S_C^* range. The amplitude of the resonance peak increases gradually as the $S_A - S_C^*$ transition is approached. Passing the $S_A - S_C^*$ transition in heating the amplitude of the vertical vibration drops to zero without showing any critical behavior.

The resonance frequency does not depend much on the film area. We decreased it up to 40% by shifting the cover plate and found that the variation of the resonance frequency was also less than 10%.

To determine the nature of the resonance we measured some of the mechanical eigenmodes of the cell. We used small mechanical pulses and measured the time dependence of the resulting vibrations using the x-axis accelerometer. The Fourier analysis gave a frequency spectrum that is very similar to that of the frequency spectrum of the vertical linear electromechanical response. We made also experiments with a similar empty cell and obtained a vibration spectrum that had eigenmodes at about the same frequencies.

To compare the strength of the responses in different directions we integrated the displacements over the frequency range. The ratio of the integrated displacement in x and y directions (s_y/s_x) depends in a characteristic way on the direction of the polarization. For ZLI4237-000 the ratio decreased from 10 to 1 when the texture relaxed from chevron to striped bookshelf. During the relaxation process the polarization changes from mainly horizontal to vertical. With the same material we find that the ratio for the bookshelf texture increased from 1 to about 50 when the polarization is reoriented by electric fields from the vertical to the horizontal. Similar trends were observed with FLC 6430 by comparing the ratio for the chevron texture with the ratio for the uniform bookshelf texture. The corresponding ratios for the unwound textures are about 10 and 2.

V. DISCUSSION

The studies confirm, that the linear electromechanical effects in C* films are due to backflow and the electroclinic effect. The dependence of the response on the texture and the director alignment can be qualitatively well explained on this basis.

The backflow causes a motion of the cover plate that is mainly horizontal parallel to the smectic layers. It is strongest when the polarization is parallel to the substrate,

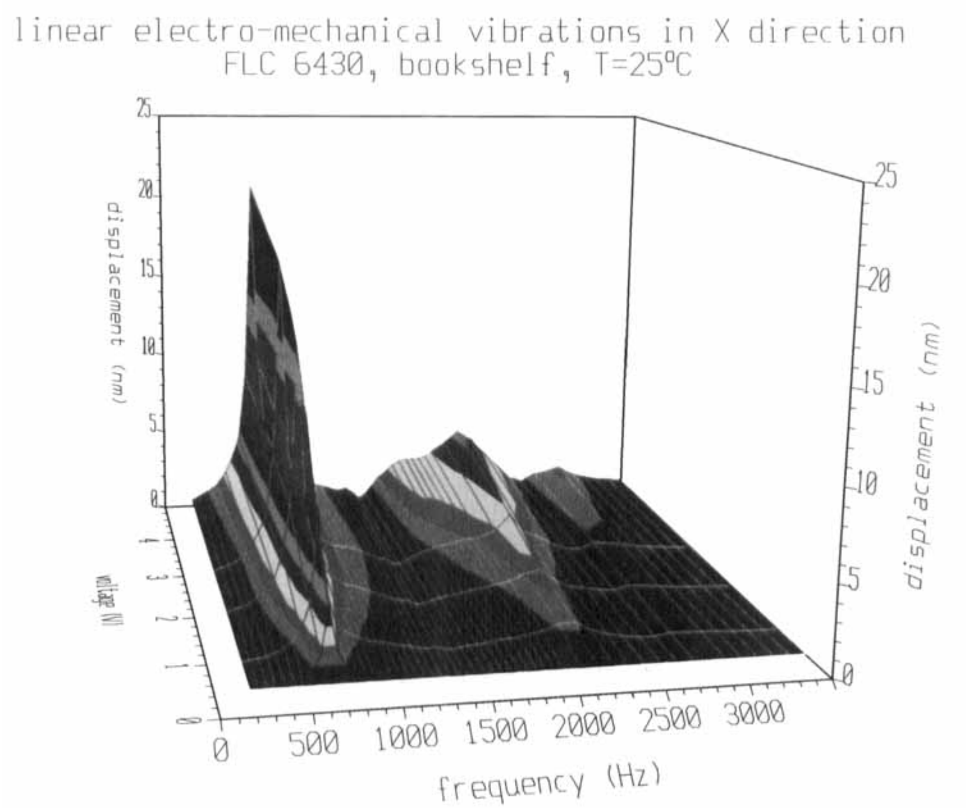

FIGURE 6 Unwound bookshelf texture, FLC 6430, frequency and voltage dependence of displacements, T = 25°C; a) x-direction; b) y-direction. See Color Plate XVIII.

as is the case for unwound chevron and reoriented stripe bookshelf textures, because at this orientation the induced director oscillations are strongest.

The electroclinic effect causes a vertical motion of the cover plate and is strongest when the polarization is vertical, as in bookshelf textures, because the induced tilt angle changes are strongest for this polarization. The absence of the critical temperature dependence of the effect near the chiral smectic A to C* transition can be explained easily. Close to the second order transition the induced change of the tilt angle is inversely proportional to the angle itself[2] and the susceptibility diverges, in the mean field approximation with a critical exponent of 1/2. The induced mechanical effect, on the other hand, is proportional to the tilt angle. Accordingly, the observed effect does not show a critical temperature dependence as the transition is approached but it should drop abruptly to zero when the transition is passed.

The results show that the vertical displacements are generally small when compared to the horizontal motion. For instance for nearly vertical orientations, where the backflow is small, the average horizontal displacement is still comparable to the averaged vertical displacement.

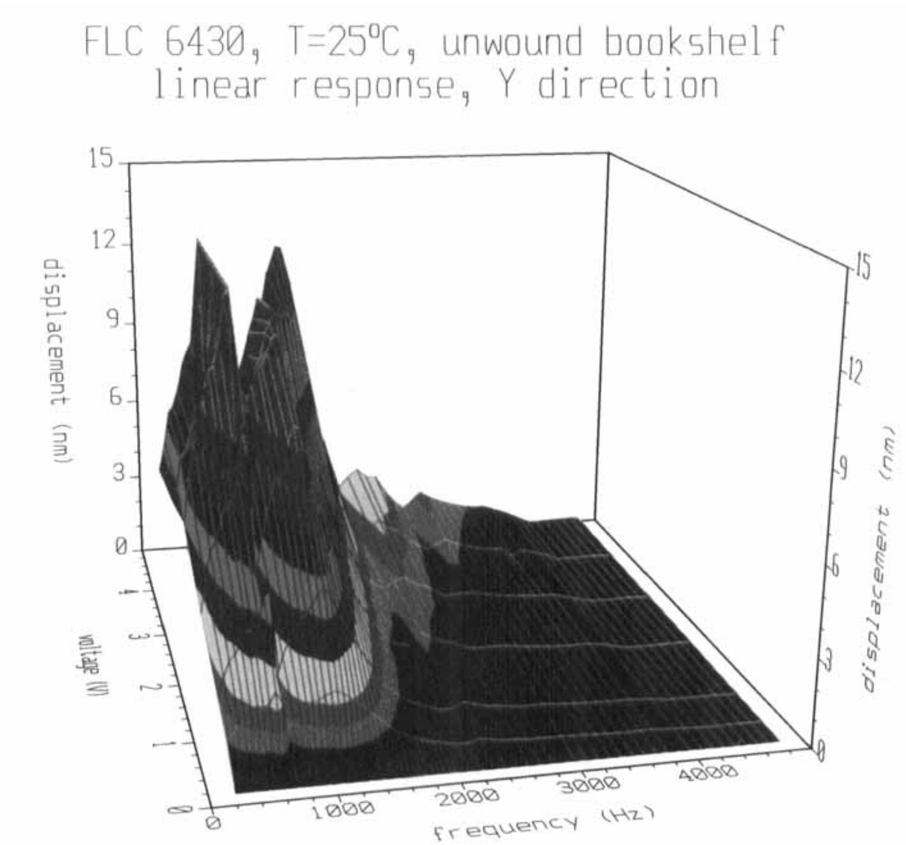

FIGURE 6 (*Continued*) See Color Plate XVIII.

The horizontal response for a helical bookshelf texture is about an order of magnitude smaller than for the unwound texture (Figure 5). We assume that the response that remains with helical bulk textures is due to the unwound surfaces films that have a thickness in the order of the pitch.[15] The helical bulk part does not contribute to the response. We can estimate therefore how much the effect should decrease by the helix formation. For a 5 μm thick sample of FLC 6430 with the pitch of 0.43 μm the effect is accordingly expected to be an order of magnitude smaller for the helical texture which is in good agreement with the experimental result.

The backflow mechanism[13] explains the decrease of the y-response for frequencies above the relaxation frequency $f_G = K(\pi/d)^2/\gamma$, (K is the relevant elastic constant, γ is the rotational viscosity and d is the sample thickness). At low frequencies however it gives a wrong dependence. Another complication is due to interference between different modes on the measured displacement. This may cause the complicated frequency dependence of the y-response, in particular around the resonance peaks, for the vertical response. These resonance peaks in x-direction are probably all due to excitations of vibrational modes of the glass plates since

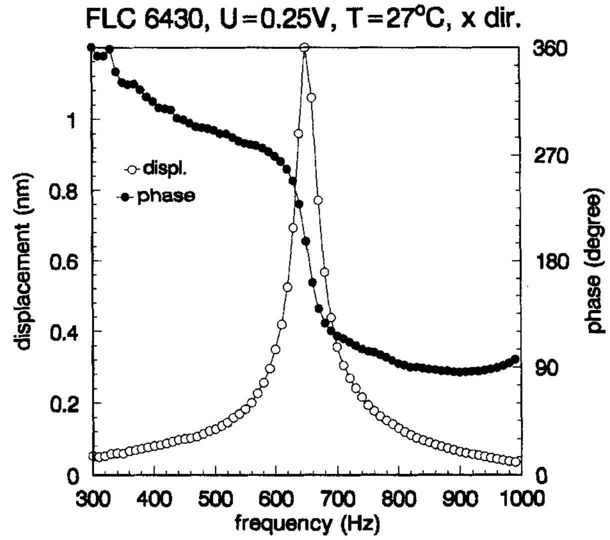

FIGURE 7 Unwound bookshelf texture, FLC 6430, frequency dependence of x-displacement and phase shift, $T = 27°C$, $U = 0.25$ V.

the resonance frequencies measured with the empty cells match fairly well some of the resonance frequencies observed in the vertical responses spectrum.

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