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Liquid crystal optical response dynamics of the flexoelectric effect in a spatially inhomogeneous electric field†

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An investigation of the flexoelectric effect in a spatially inhomogeneous field revealed the two specific features of the liquid crystal optical response. First the optical response occurs at frequencies up to 500 Hz testifying to possible response time for flexoelectric effect-devices from 2 to 5 ms. Secondly in a photosensitive structure a flexoelectric effect application allows the implementation of an optical analogue of synchronous detection as well as the subtraction of images.

1. Introduction

Electrooptical effects in a liquid crystal occurring in a spatially inhomogeneous electric field are of interest for several reasons. First, electrooptical investigations of liquid crystals in a spatially inhomogeneous electric field show a significant dependence of the optical response on the liquid crystals physical parameters and its interaction with the substrates [1]. This makes it possible to determine both the liquid crystal parameters (elastic constants, dielectric anisotropy etc.) and its surface polarization, anchoring energy and so on. Secondly, the type of deformation in such inhomogeneous field in fact determines the most important characteristics of liquid crystal image transducers, such as their resolution and sensitivity to the incident light [2], these transducers are major elements for optical and optoelectronic applications [3]. Thirdly, electrooptical effects in a spatially inhomogeneous electric field are widely used for flaw detection of thin dielectric films.

In [2, 3] the nematic optical response in a spatially inhomogeneous field, in particular, in that created in a photoconductor-liquid crystal structure has been investigated. The study shows that the electrooptical effect in a nematic represents the Fredericks transition, i.e. the change of the director orientation **n** caused by the destabilizing torque $(\varepsilon_a(\bar{\mathbf{E}}\bar{\mathbf{n}})\bar{\mathbf{E}})/4$, where $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ is the dielectric anisotropy and **E** is a spatially inhomogeneous electric field. In this case the Fredericks effect has no threshold. As a result, the observed optical response in weak electric fields is considerably below the threshold value corresponding to the Fredericks transition in a homogeneous field. In [5, 6] it was pointed out that the response to a spatially inhomogeneous field could be induced by a flexoelectric effect. In this case the destabilizing effect of the electric field is proportional to $(e_{11} + e_{33})\mathbf{n}\cdot\mathrm{grad}\,\mathbf{E}(\mathbf{r})$, where e_{11} and e_{33} are flexoelectric coefficients. When $4\pi(e_{11} + e_{33}) \gg \varepsilon_a U$, the

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flexoelectric effect must be of major importance both for liquid crystal image transducers and flaw detection in integral microcircuit dielectric films. In [5] the main characteristics of the director deformation for $\varepsilon_a \rightarrow 0$, i.e. for the case of the flexoelectric effect are analysed. The present work deals mainly with specific features of flexoelectric effect optical response dynamics in a spatially inhomogeneous electric field.

2. Samples and procedure

In the experiment we have used mixtures based on 4-*n*-butyl-4′-methoxyazoxybenzene (BMAOB) with various values of $\varepsilon_a(-0{\cdot}1 < \varepsilon_a < 0{\cdot}1)$, attained by adding a small amount, less than 1 per cent of 4-*n*-pentyl-4-cyanobiphenyl (5CB) having ε_a of 12, into the mixture. The nematic layer thickness in most cells varied from 5 to 25 μm depending on the thickness of the shims. We determined the thickness using interference transmission spectra of the empty cells.

In most samples the nematic was homeotropic. In some experiments we also used samples with a planar orientation achieved by rubbing the cell surfaces. No film orientant was applied. A homeotropic orientation was attained by one of three methods. In one it appeared spontaneously on carefully cleaned substrates after heating followed by cooling [7]. In the second the alignment was achieved by adding a small amount (less than 1 per cent) of lecithin directly into the nematic. In the third a small amount of lecithin was applied directly onto the substrate surface. According to the measurements and comparison with the calculations [5], all three methods give different surface anchoring energies: 10^{-4}, 5×10^{-4} and 3×10^{-4} erg/cm^2, respectively. The spontaneous orientation results in the nematic layers with a pronounced surface polarization [7, 8], while the lecithin orientation is unlikely to lead to such a polarization.

The experiments were carried out with two types of samples, and the procedure of optical response measurement was adapted accordingly. Samples of type 1 (used to create a spatially inhomogeneous electric field within the nematic layer) contained on one of the substrates an interdigital electrode system with a 25 μm period and 16 μm gap between the electrodes. Thus, structures of type 1 had a fixed value of the inhomogeneity of the electric field. At the same time in these structures we can easily measure the frequency dependence of the optical response (with variation of the electric voltage frequency on the electrode system).

Figure 1 shows the design of samples of type 2. A bismuth silicate ($Bi_{12}SiO_{20}$) monocrystal plate of 200–300 μm thickness was used as a photoconductor. Bismuth silicate was selected because of its good uniformity of photoelectrical and optical properties, which allowed us, practically without spatial resolution losses, to transform the initial light pattern into a required distribution of the electric field potential at the semiconductor nematic interface. As distinct from the structures of type 1, the characteristic size of the field spatial inhomogeneity in this particular case could be changed within the range from several microns to tenths of a millimeter. In investigating samples of this type, a test pattern with a harmonic intensity distribution on one of the coordinates was projected onto the structure's semiconductor. The exposition wavelength was 442 nm, i.e. within the bismuth silicate photosensitivity range. When the voltage was applied to the electrodes of the structure, the preset harmonic light intensity distribution resulted in a harmonic potential distribution at the semiconductor-nematic interface. This interface potential distribution caused the

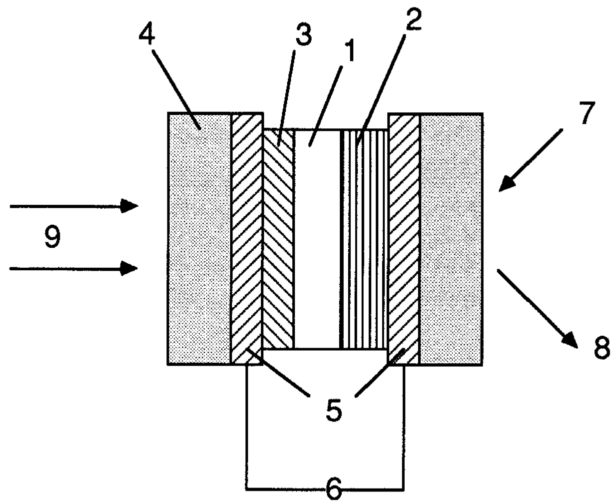

Figure 1. Design of the sample for the photosensitive experiment: 1-semiconductor; 2-nematic; 3-dielectric epoxy; 4-substrates; 5-transparent electrodes; 6-voltage supply; 7,8-read-out and modulated beams; 9-write beam.

director deformation and a spatial modulation of the refractive index of the nematic. The distribution of the refractive index obtained was equivalent to a phase diffraction lattice in a nematic layer (similar phenomena occur in the structures of type 1 as well). The read-out beam with a wavelength of 0·63 μm, usually incident normal to the structure surface, diffracts on this phase lattice. The diffraction angle measured from the propagation direction of the incident light beam is proportional to the spatial frequency of the lattice W_n:

$$\sin\theta \sim \theta \sim \lambda W_n/2\pi = \lambda/d_p,$$

where d_p is the lattice period. For photosensitive structures of type 2, the initial pattern was made using the Michelson interferometer, allowing us to vary the original test pattern period and, consequently, the lattice period in the nematic and the diffraction angle θ.

In the experiment the intensity of one of the diffraction orders was measured depending on the lattice spatial frequency, as well as the intensity-time dependence of the external AC voltage and input impulses at various frequencies. The measurements were carried out in a Fourier plane of a lens placed in the read-out beam.

3. Results

The spatial characteristics of the resultant diffraction lattices have been examined and presented in [5]. Here we would just point out that with the flexoelectric effect the diffraction lattice spatial period is twice as much as the spatial period of the potential exciting this lattice. According to [9], this is a proof of the polar nature of the flexoelectric effect. The diffraction pattern obtained can, undoubtably, be interpreted as the flexoelectric effect. Figure 2 shows photographs of the original lattice diffraction pattern with a periodicity specified by the interdigital electrode system (19 μm) and the lattice induced by the flexoelectric effect. It is seen that the flexoelectric effect diffraction orders are between those of the main lattice which is a direct

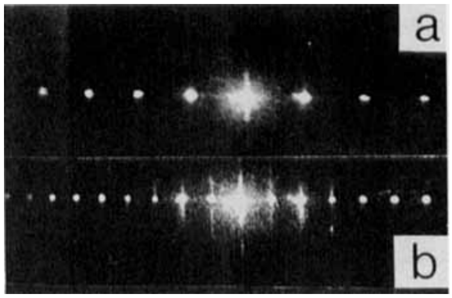

Figure 2. Photographs of a diffraction pattern on nematic-interdigitated electrode structures: (*a*) in the presence of the flexoelectric effect signal, (*b*) in the absence of the flexoelectric signal.

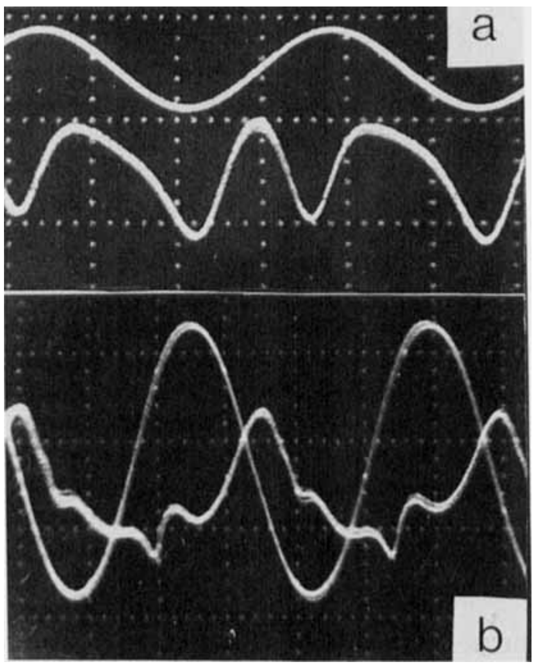

Figure 3. Oscillograms of responses at sinusoidal excitation voltage: (*a*) low voltage, (*b*) high voltage.

indication of the double period of the lattice. The intensities of the diffraction orders vary with twice the frequency of the applied voltage. It is only with large amplitudes of the external voltage that the first, third and higher odd harmonics of the applied voltage emerge (see figure 3). This, as well as other factors such as the change in the response time as the read-out light beam deviates from the normal incidence, indicate that the director oscillates around the balanced position coinciding with the normal to the surface and has a bias whose direction depends on the sign of the applied voltage. For the Fredericks transition, the director deviates only to one side and in the response signal there is always a constant component considerably higher than the alternating component, when the frequency of the applied voltage, **F**, exceeds τ^{-1}. In fact, for small angles, θ, of the director bias from the normal, the first order diffraction intensity, η, is

$$\eta \sim \theta^2.$$

With the flexoelectric effect we have $\theta \sim \cos\omega'$ and, consequently, $\eta = \eta(2\omega)$. When a flexoelectric response is superimposed on a dielectric one (the Fredericks transition), $\theta = \theta_0 + \theta_m \cos\omega t$, where θ_0 is a result of the dielectric effect, $\theta_m \cos\omega t$ is a result of the flexoelectric effect, the oscillations occur around the position $\theta_0 \neq 0$. Then $\eta \sim \theta_0^2 + \theta_1^2 \cos^2\omega t + 2\theta_0\theta_1 \cos\omega t$. Thus, the optical response contains a constant component, as well as the first and higher harmonics.

In the structure with the interdigital electrode system, the flexoelectric response optical response amplitude dependence on the frequency of the exciting voltage was studied. For low voltages (1 V_{eff}) corresponding to an electric field of the order of 0·05 V/μm, the optical response within the range investigated (0·1–1000 Hz) occurred in the second harmonic of the applied voltage. With an increase in the voltage up to 10 V_{eff} in the frequency range less than 4–5 Hz, the main harmonic component of the applied voltage is added. This causes a deviation of the linear dependence of the response amplitude on the voltage amplitude observed earlier (see figure 4). This type of signal is indicative of rapid damping of the dielectric effect as the voltage frequency increases. Figure 5 shows the frequency dependences of the r.m.s. voltage of the first and second harmonics present in the response signal. These dependences show that

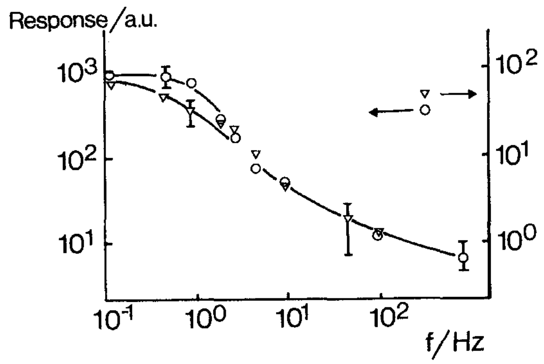

Figure 4. Frequency dependence of the response for the flexoelectric effect: 1–1 V_{eff} (▿); 2–10 eV_{eff} (○).

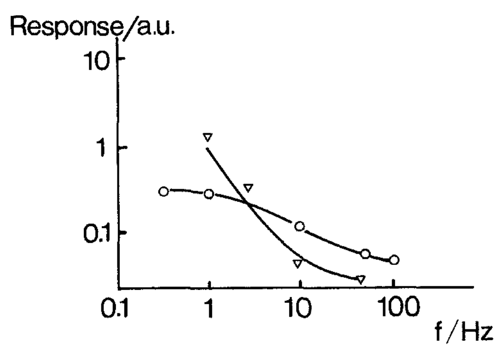

Figure 5. The frequency dependences of the first (▿) and second (○) signal harmonics for the flexoelectric effect at 10 V_{eff}.

the first and second harmonic signals have their cut-off frequencies differing by several orders of magnitude.

The check experiment for the nematic mixture with $\varepsilon_a \approx 5$ demonstrated that under the same conditions the type of signal and its dependence on the voltage amplitude are completely different. It has the constant component considerably higher than the variable one, there is no linear dependence of the response amplitude on the exciting voltage amplitude, the spatial period of the excited response in the nematic corresponds to the interdigital electrode period, i.e. it is twice as low as that of the flexoelectric effect-induced lattice period. Figure 6 illustrates these differences; the frequency dependence of the response amplitude for the case described are given for two interdigital electrode voltages—1 V_{eff} and 10 V_{eff}.

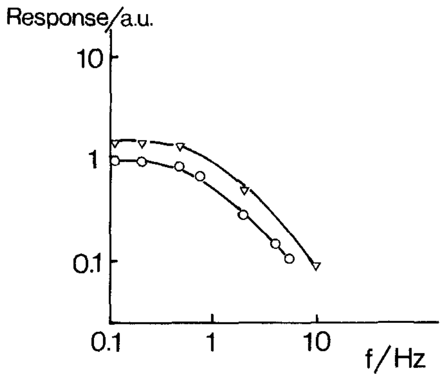

Figure 6. Frequency dependence of the response in a photosensitive structure with the nematic having $\varepsilon_a = 5$. ○ = 1 V and ▽ = 10 V.

The sensitivity of the optical response to variations of the applied voltage up to very high frequencies (500 Hz) allows us to utilize the flexoelectric effect in light modulation devices. The sensitivity of the effect to the sign makes it possible to obtain an optical response of 1 or 2 ms.

We have also observed the dynamic response of the flexoelectric effect in photosensitive structures of type 2 illuminated by light pulses whose duration was more than 1 s. The structure was fed by a sinusoidal voltage with a frequency of 20–100 Hz (the range was determined by the structure sensitivity region), and the temporal behaviour of the intensity of the first order diffraction was observed. At the same time the period of the lattice recorded was varied thus changing the size of the field inhomogeneity. The experimental results are shown in figure 7 where the measured response relaxation time is plotted versus the spatial frequency. It can be seen that the curves differ drastically. In the first case (corresponding to low voltages and low spatial frequencies) the response oscillograms indicate that the director oscillates symmetrically to the normal to the layer. In the second case such symmetry is broken. Thus, although the director motion is still oscillatory, the oscillations do not occur around the normal to the layer, but with a considerable tilt of the director. Such behaviour of the director motion is likely caused by the distortion of the homeotropic orientation in the first moment, when the moment of the force from the side of the transverse electric field near the nematic surface is higher than the surface anchoring strength. This hypothesis is partially confirmed by the fact that the behaviour of the development of the nematic response impulse in the experiments

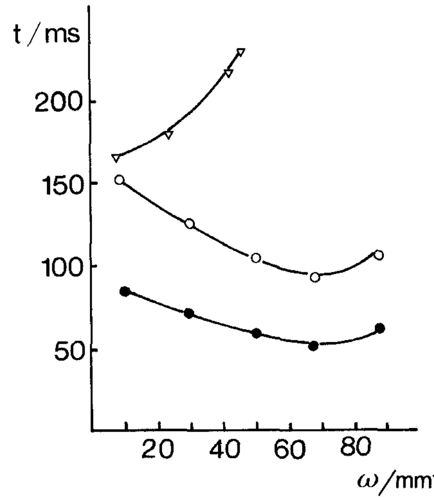

Figure 7. Dependence of the response relaxation time on the spatial frequency for various orientation types: (*a*) spontaneous orientation, ○ = 30 V and ● = 70 V, and (*b*) orientation by lecithin, ▽ = 70 V.

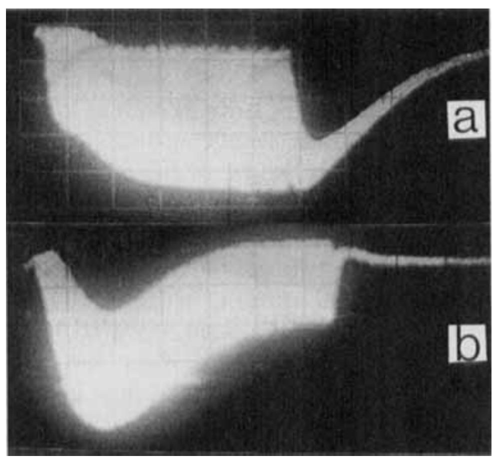

Figure 8. Structure response oscillograms for impulse excitation: (*a*) spontaneous orientation, (*b*) orientation by lecithin.

described depends upon the way in which the homeotropic orientation was achieved (see figure 8).

The check experiments with structures comprising an interdigitated electrode system were carried out in which pulses of sinusoidal voltage having a duration of more than 1 s were applied. On the one hand, the check experiments were to simulate a situation in photosensitive structures; on the other hand, they allowed us to avoid the integrating effect typical of photosensitive structures (the time of photocurrent saturation in the structures investigated was of the order of 20 to 40 ms). The experiment results demonstrate the existence of two types of the response behaviour. In particular, with the same spatial frequency the second type of behaviour occurs in such structures when the applied voltage frequency and amplitude grow (see figure 9).

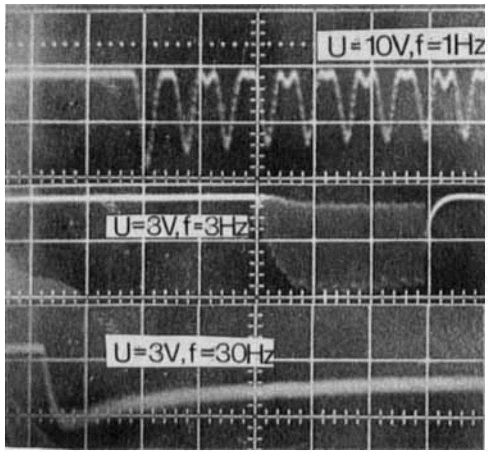

Figure 9. Oscillograms of response for structures with interdigitated electrodes at impulse excitation (the voltage and frequency are indicated on the oscillograms).

4. Practical application of the flexoelectric effect for image processing and recording

Together with the application of the flexoelectric effect for rapid light modulation, owing to its sensitivity to the sign of the exciting electric field it can also be implemented in liquid crystal image transducers for new image recording and optical data processing. First, the effect occurs at a frequency twice as high as that of the applied voltage. This allows us to distinguish the image signal contribution in the combined signal of a photoelectric detector placed at the output of the image transducer. Secondly the flexoelectric effect makes it possible to implement a selection of images with periodically variable brightness (illumination intensity).

In the latter case, a recorded image containing among other objects those that scintillate periodically, was projected onto the image transducer. On the boundaries of the image details in the nematic layer plane there occurs a sign-variable (with applied voltage frequency) transverse electric field perpendicular to the initial homeotropic orientation. As the voltage is sign-variable, the director deviates simultaneously with the applied voltage from the initial position ($\theta = 0°$) within $-\theta_m < \theta < \theta_m$, where θ_m is an angle measured from the normal. In this case the optical response is relatively small.

When the frequency of the applied voltage coincides with the frequency of the brightness variation of the selected object, especially when the brightness maximum coincides with the maximum of the applied voltage amplitude, the optical response of the structure increases many times. This is because with such a cophased illumination a pulsating (single-polar) voltage rather than a sign-variable one occurs on the semiconductor-nematic interface. The director motion in such a field (the field geometry is unchanged) is asymmetrical: $\theta''_m < \theta < \theta'_m$. Here the magnitude of the bias angle increases as compared to that in the previous case, i.e. $\theta'_m \gg \theta_m$, as the field in each subsequent temporal cycle acts in the same direction, the field effect is accumulated (the director relaxes affected rather by the anchoring strength than by the field, the time being on the order of 100 ms). At the same time the phase difference, φ, increases significantly owing to this accumulation, $\varphi \sim \theta^2$, and consequently the optical response increases as well.

In fact we can state that a structure with an optical response time constant integrates the effect of the electric field. Due to the polar nature of the flexoelectric effect the voltage pulses occurring under illumination of a semiconductor are added each with its own sign. This sum may be about zero for sign impulses of different; for instance, at a stationary illumination of a semiconductor, when both voltage half-periods affect the nematic in the same way. In case of impulses of the same polarity when the applied voltage frequency coincides with the light impulse rate, it increases drastically. Accordingly, a drastic increase of the optical response is observed.

The effect of the growth of the phase difference when the applied voltage frequency, F, approaches the frequency of variation of the object's brightness, F_c, is of a resonance type. The width of this resonance is $\Delta F \sim \tau^{-1}$, where τ is the effective time of structure integration, the main contribution being the nematic elastic relaxation time. Resonances are also observed at multiples of the frequencies of the applied voltage to the frequency of brightness variation, i.e. at $F = nF_c$, where $n = 2, 3, 4, \ldots$, with a gradual decrease of the response amplitude due to degradation of the structure photosensitivity.

An essential condition for the flexoelectric effect is a minimal dielectric anisotropy, otherwise the Fredericks transition will lead to a director reorientation to only one side of the normal to the surface. On the other hand, the Fredericks effect responds similarly both to sign-constant and sign-variable sequences of the voltage impulse.

The resonance frequency dependence of the flexoelectric effect response can be explained using a simple model of temporal integration of the structure periodical excitation. The optical response can be represented by the series

$$\sum_{h=0}^{\infty} \int_0^{\infty} A \sin(2\pi t T_2^{-1} + \varphi)(f - nT_1)\exp(-t/\tau)dt,$$

where the factor $(t - nT_1)$ corresponds to a periodic (with a period of T_1) time sequence of exciting light impulses, $A\sin(2\pi t T_2^{-1} + \varphi)$ corresponds to the applied voltage (period of T_2). These effects are added in time (light pulse-to-pulse accumulation), the relaxation of their effect being represented by a factor of $\exp(-t/\tau)$, where τ is the integration time of the structure. The result depends considerably on the T_2/T_1 ratio, i.e. on the relationship between the applied voltage and illumination frequencies, reminiscent of the transmission dependence of the Fabry–Perot reference. The transmission peaks correspond to the values $T_2/T_1 = K$, where $K = 1, 2, 3, \ldots$, i.e. integer multiples of the frequencies. Assuming that the peak halfwidth is $\Delta F/\mathrm{F} = (2\pi\sqrt{2\tau/T_1})^{-1}$, for $T_1 = 0{\cdot}01$ s and $\tau = 0{\cdot}4$ s we obtain $\Delta F/F = 0{\cdot}01$, i.e. the pulse frequency selectivity at $F = 100$ Hz is $\Delta F = 1$ Hz. The response maximum to minimum ratio is $2\tau/T_2 = 50$.

The experimental results give a smooth dependence of the structure sensitivity on the applied voltage for a stationary image (see figure 9). However, for a periodic variation of the image brightness ($T_1 \sim 1$–3 ms) the picture is disturbed: when the applied voltage frequency, F (20 Hz $< F <$ 500 Hz), coincides with the brightness fluctuation frequency of the object, F_c, then a resonance increase from 40 to 50 times in the object response amplitude is observed (see figure 10). The total resonance width characterizing the selectivity of the method is 2–3 Hz. It should be noted that these results are obtained in a structure with integration time of 300–500 ms determined by the visco-elastic relaxation of the nematic (photocurrent saturation and relaxation time being 10 ms). In the structure with the integration constant determined by

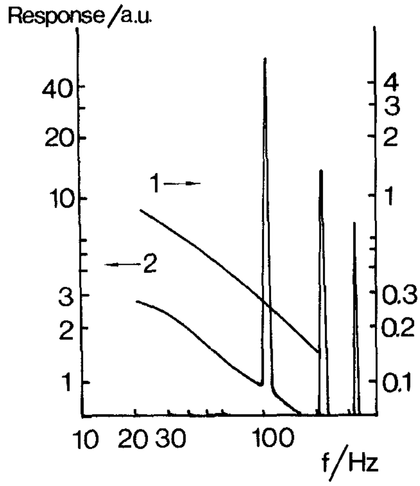

Figure 10. Experimental dependence of the signal response on the applied voltage frequency: 1-stationary light signal; 2-periodic signal with a frequency of 108 Hz.

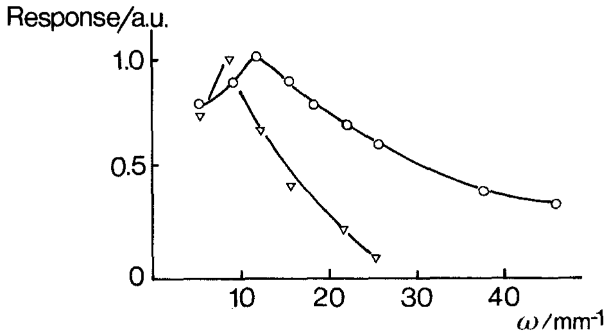

Figure 11. Dependence of the diffraction intensity on the spatial frequency for a stationary image (○) and a scintillating image under resonance conditions (▽).

saturation time of 800 ms, the peak width approached 1 Hz. Thus the response parameters are described quite accurately by this simplified model.

The results shown in figure 11 demonstrate the structure's ability to transmit spatial details of the image.

The check experiments with structures having $\varepsilon_a = 3$–15 (the Fredericks transition) showed no resonance. As distinct from the flexoelectric effect application, the Fredericks transition the response signal is maximal for a stationary case. The frequency band exhibiting this effect for structures with bismuth silicate did not exceed 10–500 Hz due to the limitation of the photoconductor speed, but it could be expended up to 100 Hz, provided photoconductors with a fast response (silicon, gallium arsenide) are used.

The phenomenon observed is an optical analogue of synchronous detection, well-known in electronics. It should be noted that liquid crystal image transducers, with the flexoelectric effect can select out of a number of objects the one with a specified frequency and phase of brightness variation. In addition, if different images are projected onto the transducer during positive and negative half-periods, then the differences between them will be revealed and emphasized by resonance. These differences can be also revealed in the temporal evolution of the two images under comparison.

5. Conclusion

The results of our investigation demonstrate specific features of the nematic optical response for the flexoelectric effect in a spatially inhomogeneous field:

(*a*) the optical signal occurs at frequencies up to 500 Hz, testifying to possible response of nematic devices based on the flexoelectric effect of 2–5 ms;
(*b*) in a photosensitive structure application of the flexoelectric effect allows us to implement an optical analogue of synchronous detection as well as to subtract images.

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