A study of the microwave dielectric permittivity of liquid crystals in electric and magnetic fields

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A microwave detector based on a self-sustained oscillator circuit is proposed as a means to investigate the real and imaginary components of the dielectric permittivity of liquid crystals in external electric and magnetic fields. Results are given for measurements of a 500 MHz oscillator frequency for two types of nematic crystals, 5CBP and MBBA. Fundamental regularities are identified in the behavior of the microwave dielectric permittivity of samples in electric and magnetic fields. It is shown that the minimum of the high-frequency dielectric loss in liquid crystals correspond to a situation in which the long axes of the molecules are oriented parallel to the direction of the microwave electric field. © *1998 American Institute of Physics.* [S1063-7842(98)02201-6]

INTRODUCTION

As a rule, the dielectric constants of liquid crystals are studied in the frequency range $f=0-10^7$ Hz, where the dispersive properties of the materials arising from the orientational mechanisms for polarizing the molecules are most strongly manifest. In contrast, in the higher frequency decimeter wavelength range ($f=10^8-10^9$ Hz) the behavior of the dielectric permittivity of liquid crystals and its interrelation with molecular orientation processes in electric and magnetic fields have been practically unstudied. This is mostly due to experimental difficulties, since traditional detectors based on lumped elements no longer work in this frequency range, while electrodynamic structures with distributed parameters are too cumbersome, and hence possess inadequate sensitivity.

In order to study liquid crystals in the decimeter wavelength range, we developed a new and original miniature microwave detector structure built around microstrip circuits.[1] We showed that microstrip measurement cells can be used to accurately identify changes in both the real and imaginary components of the dielectric permittivity of liquid crystals subjected to an external electric field, even for quite small liquid crystal samples (with volumes $\simeq 10^{-3}$ cm^{-3}). The thickness of the sample layer in the microwave detector is determined by a gap between measurement plates, and ordinarily is $\simeq 100$ μm. This is comparable to the thickness of real liquid crystal cells used in various practical devices. Gaps of this size allow us to obtain important information about the influence of surfaces that bound the liquid crystal sample on the behavior of the dielectric characteristics of the material.

The microstrip detectors described in Ref. 1 were constructed for operation in tandem with standard devices for measuring amplitude-frequency characteristics. In this paper we discuss complete microwave detector structures in which the microstrip measurement resonator is included as part of the frequency-defining loop of a microwave self-sustained oscillator circuit. This arrangement significantly increases the sensitivity of the system and the accuracy of the measurements. The operation of the system is illustrated by the results of experimental studies of the behavior of the dielectric constant of two typical nematogens: 4-n-pentyl-4- cyanobiphenyl (5CBP) and 4-methoxybenzinidene-4- butylaniline (MBBA) under the action of dc electric and magnetic fields. Discussion of the results obtained is based on descriptions of processes whereby electric and magnetic fields orient the dipole molecules of the liquid crystals and change the value of the microwave dielectric loss connected with them.

SELF-SUSTAINED OSCILLATOR DETECTOR CIRCUIT

The frequency-defining loop of the microwave detector self-sustained oscillator circuit (Fig. 1) is a microstrip ‘‘ring’’ resonator that includes the vertical measurement plates.[1] The oscillator is placed in a metal package with dimensions 30×24×15 mm. The resonator structure itself, which serves as the top cover of the package, is made on a substrate of polycor ($\varepsilon=9.8$) with thickness $h=1$ mm, dimensions 24×30 mm. In this case the lower metallized side of the substrate, which we referred to as the ground plane, was soldered to the walls of the metal package of the detector along its entire perimeter. The picture shown in Fig. 1 also includes the wires on the upper side of the substrate. Gold-plated measurement plates (a) with dimensions 2.5 ×2.5 mm were soldered to the metal pads at the ends of the striplines of the microstrip resonator vertical to the plane of the substrate so that the measurement gap had a value of 100 μm. A liquid crystal placed in the gap is subjected to surface tension forces. In order to decrease the effect of the capacitance between the edges of the detector package and the measurement plates, the latter were located close to the center of the substrate, at the expense of bending the strip lines. To do this, a rather small portion of the metallized screen was removed from the backside of the substrate directly under the detector measurement plates by chemical etching.

The self-sustained oscillator circuit was tuned for exci-

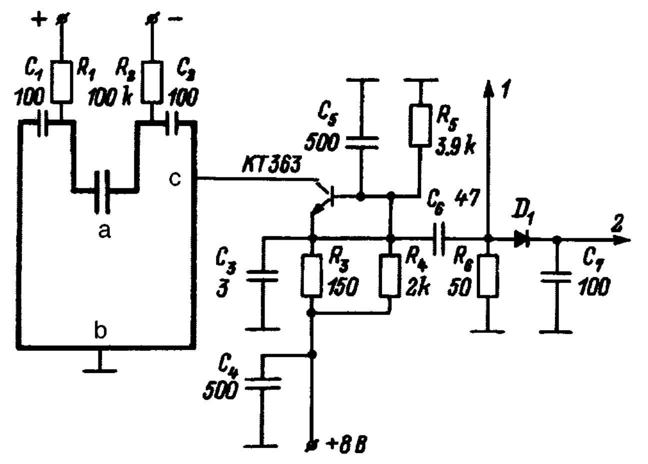

FIG. 1. Basic circuit for the self-sustained oscillator detector. *1*—output (frequency), *2*—output (amplitude).

tation of the fundamental half-wave mode of oscillation of the microstrip resonator. In order to feed the transistor with a constant current, the upper strip of the resonator is short-circuited by a jumper to the package at a point of minimum high-frequency voltage (point *b* in Fig. 1). In this case the collector of the transistor was connected directly to the strip of the microstrip resonator at point (c) in order to create conductive (autotransformer) coupling with the resonator at high frequencies. An orienting electric field was created across the measurement plates by applying a dc voltage to the latter through the decoupling resistors R_1 and R_2. In order to galvanically decouple the power supply we used divider capacitors C_1 and C_2. The microwave oscillator was designed to have two outputs: one for measuring frequency, and the other for measuring the amplitude of the self-sustained oscillations. The circuit was tuned by setting up optimum conductive coupling between the resonator and the microwave transistor, i.e., we looked for an optimum point to connect the transistor collector to the strip line of the resonator (c), and carefully chose point (b) near the midpoint of the stripline of the microwave resonator as ground. Pouring liquid crystal into the measurement gap significantly reduced the Q-factor of the resonator half- wave mode, and we found that the oscillator spontaneously switched to higher-Q ''parasitic'' resonances. In order to avoid this instability we needed to position the circuit components compactly near the resonator and choose a capacitance C_3 to shunt the higher resonances to ground. The working frequency of the self-sustained oscillator microwave detector, which was measured using a digital frequency meter, was around 500 MHz when the sample of liquid crystal was poured into the gap. In this case the microwave oscillation amplitudes measured by a digital volt meter at the output to the microwave detector were found to be about 0.1 V.

Note that all our experiments on liquid crystals were carried out at a temperature $T=24\pm0.1$ °C. Therefore, the microwave detector was placed in a miniature thermostat, which was placed between the poles of an electromagnet. The magnetic field could be varied up to a value $H=2.3$ kOe. The self-sustained oscillator package could be rotated between the end-face poles of the magnet so that the direction of the field H was either parallel or perpendicular to the direction of the high-frequency and external dc electric fields.

SAMPLES AND METHODS OF INVESTIGATION

As we have already mentioned, as samples for our investigations we used two widely known nematogens—the compound 5CBP, with a relatively high positive dielectric anisotropy,[2] and the weakly anisotropic liquid crystal MBBA with negative anisotropy.[3–5] Our measurement method was the following: the liquid crystal was placed in the gap between the measurement plates of the self-sustained oscillator microwave detector, and was kept there for an hour in order for it to reach its equilibrium state. Within this time, the temperature of the detector stabilized and initial values of the frequency and amplitude of the self-oscillations were established. For each sample of liquid crystal a magnetic field H was applied, and the dependence of the relative frequency change $F(H)$ was measured along with the relative change in amplitude $P(H)$ of the detector self-sustained oscillations in the steady-state regime. Analogous functions $F(U)$ and $P(U)$ were measured when an electric potential U was applied to the measurement plates of the detector. We also investigated the dependence of the changes in the microwave parameters of the self-sustained oscillator under the combined action of electric and magnetic fields on the liquid crystal.

For an optimal choice of the structural coupling between the microstrip resonator and the microwave transistor, the level of self-oscillations in the oscillator depended linearly on the Q-factor of the frequency-determining resonator, which in turn was related in this experiment to the dielectric loss tangent of the liquid crystal sample. As a result, the observed change in the microwave oscillation amplitude $P(U,H)$ was inversely proportional to the change in the imaginary component of the dielectric permittivity of the sample $\Delta\varepsilon''$. As is well known, changing the real component of the dielectric permittivity of the liquid crystal sample $\Delta\varepsilon'$ leads to a change in the resonant frequency of the self-sustained oscillator detector: the frequency increases as the dielectric permittivity ε' decreases, and conversely. Consequently, $F(U,H)\sim(\Delta\varepsilon')^{-1}$.

RESULTS OF EXPERIMENTAL INVESTIGATIONS

Figure 2 shows the dependence of the relative change in the level of microwave oscillations of a detector containing a sample of 5CBP. For convenience of comparison the dependences on electric potential $P(U)$ and on magnetic field $P(H)$ are shown on the same figure. Analogous curves plotted for a sample of MBBA are shown in Fig. 3. Curves *1* on these figures were plotted for no magnetic field $(H=0)$, while curves *2* and *3* were plotted for no electric field $(U=0)$. The dependences 2 were plotted for the case where the direction of the dc magnetic field H was parallel to the direction of the high-frequency field e in the measurement gap of the detector, while the curves *3* were opposite, i.e., the direction of the field H was perpendicular to the direction of

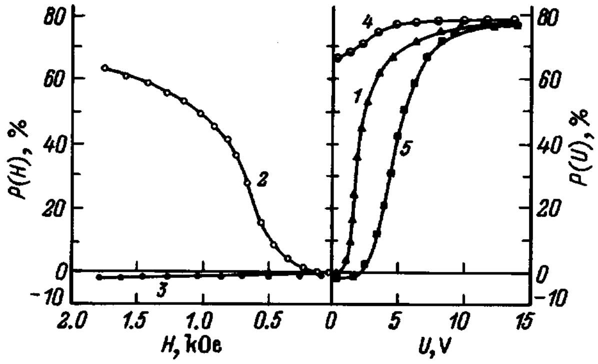

FIG. 2. Dependence of the relative level of microwave oscillations in a detector with a sample of 5CBP on the electric potential $P(U)$ and magnetic field $P(H)$.

e. Moreover, the functions $P(U)$ were plotted while a magnetic field $H=2.3$ kOe acted simultaneously on the sample; this field was directed either parallel to e (curves *4)* or perpendicular to e (curves *5).*

The features observed in the behavior of the curves $P(U)$ and $P(H)$ are connected with the well-known second-order orientational phase transitions that have been given the name Fredericks transitions.[5] These transitions are a consequence of the competition between two forces that act on molecules of the sample, one exerted by the walls of the cell that bound the liquid crystal and the other by the external field. The primary feature of a Fredericks transformation is the presence of critical fields above which the initial orientational configuration of the molecules created by the walls becomes unstable and the director of the molecules shifts to a new state determined by the magnitude and direction of the external fields. For liquid crystals of nematic type with anisotropies in the dielectric permittivity $\varepsilon_a=\varepsilon_\parallel-\varepsilon_\perp$ and magnetic susceptibility $\chi_a=\chi_\parallel-\chi_\perp$, the relation between the critical electric fields E_c and magnetic fields H_c can be described by the following expression[6]:

$$\frac{1}{2}\chi_a H_c^2=\frac{\varepsilon_a E_c^2}{8\pi}. \tag{1}$$

From Figs. 2 and 3 it is clear that the critical fields are easy to determine from the experimental results. For the 5CBP samples the field $H_c\approx 120$ Oe, and the field $E_c\approx 3$ V/mm. Since the anisotropy of the diamagnetic susceptibility of the crystal χ_a is a quantity of order 10^{-7}, Eq. (1) allows us to infer that the anisotropy of the dielectric permittivity $\varepsilon_a\approx 2$. This value of ε_a is in good agreement with the results of other measurements on liquid crystals of this type.

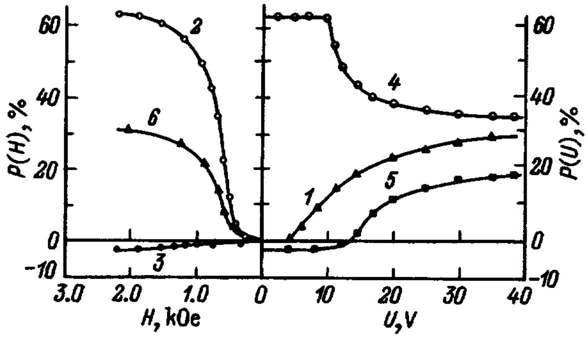

FIG. 3. Dependence of the relative level of microwave oscillations of a detector with a sample of MBBA on the electric potential $P(U)$ and magnetic field $P(H)$.

The measurements on the samples of liquid crystal MBBA (Fig. 3) show that the electric and magnetic critical fields are almost an order of magnitude larger in these samples than in the crystal 5CBP. This is entirely natural, since it is well known that the crystal MBBA is only weakly anisotropic.

By analyzing the behavior characteristics of the electric and magnetic dependences $P(U)$ and $P(H)$ in fields above the critical fields we can obviously determine qualitatively the initial configuration of the molecular states and the processes that orient them under the action of these competing forces. Note that the orienting action of the magnetic field is connected with the diamagnetic component of the susceptibility caused by the presence of benzene rings in the composition of the molecules. The minimum in magnetic energy corresponds to a state where the plane of the benzene rings lies in the direction of the magnetic field. This implies that in this type of liquid crystal the long axis of the molecule is oriented parallel to the magnetic field.

Let us first discuss the effect of dc magnetic and electric fields on the value of the microwave dielectric loss for the liquid crystal 5CBP. As is clear from Fig. 2, when the external magnetic field is parallel to the high-frequency field e, the amplitude of the self-sustained oscillations of the detector increase monotonically with increasing H up to saturation (curve *2).* In this case the long axes of the dipole molecules are rotated normal to the plane of the measurement plates, i.e., parallel to the high-frequency field e, which appreciably decreases the microwave dielectric loss in the liquid crystal sample.

However, in the opposite case, when the direction of the magnetic field $H\perp e$, the amplitude of the self-sustained oscillations, and consequently the dielectric loss in the liquid crystal sample, are almost unchanged (curve *3* in Fig. 2). Obviously this direction of the magnetic field should orient the long axes of the molecules along the measurement plates, that is the long axes of the molecule should rotate perpendicular to the direction of the high-frequency field. As a result, the dielectric loss should increase and the amplitude of the self-sustained oscillations should decrease. The fact that experiments do not reveal any significant change in $P(H)$ in this situation is evidence that all the dipoles are already oriented along the walls of the measurement plates due to the orienting forces exerted on the molecules by the surface.

Since ε_a is positive for the liquid crystal 5CBP, the parallel orientation of the director along the microwave field should be established even when the external field applied to the measurement plates acts on the sample as well. And, in fact, as is clear from Fig. 2, the behavior of the function $P(U)$ (curve *1)* shows practically no difference from that of the corresponding ''magnetic'' curve *(2).* Based on what we have said above, it is not difficult to explain the behavior of the "electric" curves *4* and *5*, which were plotted with the maximum magnetic field $H = 2.3$ kOe acting on the sample at the same time. Obviously the strong magnetic field applied parallel to the high-frequency field in this experiment almost completely orients the liquid crystal, rotating the long axes of the molecules along the direction of e. As a result, as the additional electric field applied in this direction increases we observe only a rather small rotation of the molecular axes; therefore, there is only a slight increase in the oscillation amplitude $P(U)$ (curve *4)*.

However, for a perpendicular applied magnetic field the long axes of the molecules are oriented almost perpendicular to the direction of e. As a result, we require a considerably larger dc electric field to rotate the molecular axis by 90°, because it is necessary to overcome not only the orienting forces exerted by the surfaces of the measurement plates but also the orienting forces exerted by the magnetic field. Therefore, in these experiments we observe a significant increase in the value of the critical field E_0 (Fig. 2, curve *5)*.

The liquid crystal MBBA differs from the crystal 5CBP by the negative anisotropy of its dielectric permittivity ($\varepsilon_a = -0.59$); however, the anisotropy of the diamagnetic susceptibility $\chi_a = 1.23 \times 10^{-7}$ in these materials is positive. As a result, the long axes of the MBBA molecules are oriented along the direction of the magnetic field, just as in the crystal 5CBP, while the "magnetic" functions shown in Fig. 3 (curves *2, 3)* are very similar to the analogous curves for 5CBP (Fig. 2). The magnetic field parallel to the high-frequency field orients the axis of the molecules along e; therefore, the dielectric loss decreases to a minimum (curve *2)*. Keeping in mind the fact that increasing the magnetic field $H_\perp$ causes only a slight change in of $P(H)$ in this experiment (Fig. 3, curve *3)*, we may assert that the orienting forces exerted by the walls of the measurement plates are primarily directed in the planes of the plates for MBBA, just as for 5CBP.

However, in contrast to the previous cases, when we simultaneously apply the maximum magnetic field $H_\parallel$ to the sample of MBBA crystal and increase the voltage U, we observe a slight decrease in the value of $P(U)$, i.e., an increase in the dielectric loss in the crystal (curve *4)*. This implies that in this case the electric field causes the direction of the molecular axes to deviate from the direction of the parallel orienting magnetic field. This anomalous behavior of $P(U)$ is explained by the fact that the parallel and perpendicular components of the dielectric permittivity in the MBBA crystal differ only slightly from one another: $\varepsilon_\parallel = 5.17$, $\varepsilon_\perp = 4.58$.[5] In this case the direction of the resulting polarization vector is determined as the vector sum of these two quantities. Since the polarization vector at large U is oriented along the dc electric field, the long axis of the molecule turns out to be rotated relative to its original state at $U = 0$. As a result, the microwave dielectric loss increases accordingly. The angle through which the long axes of the molecules rotate can be determined from the relation

$$\varphi = \arctan(\varepsilon_\perp / \varepsilon_\parallel) = 42.34°. \tag{2}$$

As a proof of what was said above, in Fig. 3 we show curve *6*, which was plotted for a magnetic field oriented at the computed angle $\varphi \approx 42°$. It is clear that the observed change in the quantity $P(H)$ is in rather good agreement with the maximum change in $P(U)$, which confirms that an oblique orientation of the director forms in the electric field. The rather small values of the dielectric anisotropy in the liquid crystals MBBA, as we already mentioned above, causes the critical field to increase significantly to $E_c = 50$ V/mm (curve *1)*, and to increase to $E_c = 140$ V/mm (curve *5)* when a perpendicular magnetic field acts simultaneously on the sample.

Note that in all the experiments the relative change in the self-sustained oscillator frequency did not exceed a value of $F \approx 0.6\%$. Decreasing the microwave loss in the liquid crystal samples under the action of electric or magnetic dc fields, as a rule, led to a decrease in the frequency of self-sustained oscillations of the detector, i.e., to an increase in the real part of the dielectric permittivity, and conversely. Thus, these experiments show that even in the decimeter wavelength range the processes that orient the molecules are accompanied by rather large changes not only in the imaginary part but also the real part of the components of the microwave dielectric permittivity of these liquid crystal samples.

CONCLUSIONS

The self-sustained oscillator detector scheme described in this paper for studying the properties of liquid crystals at microwave frequencies allows us to identify operationally and with good accuracy the relative changes in both the real and imaginary components of the high-frequency dielectric permittivity of samples subjected to external fields. The high sensitivity of this method reveals even greater potential for performing new and very precise experiments connected with the study of orienting processes in liquid crystals, and also distinctive features of the interaction between liquid crystal molecules and surfaces made from various materials that bound the sample.

In this paper we have shown that the changes in microwave dielectric characteristics of the liquid crystals 5CBP and MBBA are uniquely related to processes that orient the dipole molecules. We have also shown that the case where the microwave electric field is parallel to the longitudinal component of the dielectric permittivity of the liquid crystal corresponds to a minimum dielectric loss. In this situation we established experimentally that the value of the dielectric losses in liquid crystal samples at microwave frequencies are comparatively large, although, as follows from other papers (see Refs. 2–4,6) the dielectric permittivity of liquid crystals has the tendency to decrease rapidly as the frequency increases due to the large inertia of the polarization oscillations of the molecules. From this point of view, in the microwave range a liquid crystal should have only insignificant dielectric losses approaching those of the optical absorption coefficient in magnitude.

It would be logical to assume that strong absorption of microwave power observed in these experiments is caused by charging of ion complexes or impurities that are present in liquid crystals,[7,8] whose concentration can be quite high. However, this assumption does not explain the fact that dielectric loss in the experiments changes significantly when the molecules of these crystals are oriented in dc electric and magnetic fields. Therefore, it is most likely that the nature of the observed high losses in liquid crystal samples at microwave frequencies are connected with the broad spectrum of relaxation oscillations of individual ring molecular groups connected with the benzene rings, the so-called ''tails.''

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