POLYMERS
AND LIQUID CRYSTALS

Reconstruction of the Distribution Function of Relaxation Times for 7CB and 7OCB Liquid Crystals from Dielectric Spectra

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Abstract—A technique for reconstructing the distribution function of relaxation times from the dielectric spectra measured over a wide range of frequencies is proposed and tested using 7CB and 7OCB liquid crystals as examples. The objective function, which is the sum of the squares of the differences between the calculated and measured permittivities, is minimized with the Mathcad program package. It is demonstrated that, in the case of parallel orientation of the molecular director with respect to the polarization of the ac electric field, the distribution function of relaxation times is consistent with the Debye relaxation model; however, there are insignificant deviations in the short-time interval corresponding to the high-frequency portion of the dielectric spectrum. For perpendicular orientation of the director, the distribution function of relaxation times exhibits specific features that are most pronounced for the 7OCB liquid crystal and probably reflect intramolecular motions of the rigid core and fragments of the alkyl chain.

1. It is known that the orientation mechanism of dielectric polarization (which is referred to as relaxation) is decisive for many materials, including liquid crystals. The frequency dependence (dispersion) of the permittivity $\varepsilon(\omega)$, as a rule, is described by the Debye equation in which the relaxation time τ is a characteristic accounting for the dynamics of molecular motion in a particular compound. The relaxation time τ can be easily determined in the experiment from the position of a maximum in the frequency dependence of the imaginary component of the permittivity $\varepsilon''(\omega)$.

For a material with two or several relaxing subsystems, the dispersion of the permittivity $\varepsilon(\omega)$ is adequately described by the sum of the Debye equations with different relaxation times and the contribution of each subsystem is taken into account through the relevant weighting factor [1]. In the case when the number of subsystems is relatively large, the Debye dispersion relation should include a continuous distribution function of relaxation times [2]. It is of considerable interest to determine the continuous distribution function of relaxation times directly from the experimental dielectric spectra. With knowledge of the true distribution functions of relaxation times, it is possible to gain a deeper insight into the nature of the specific features observed in the polarization of materials, in particular, due to intramolecular motions. As a rule, the above problems have been solved using numerical methods.

It is important to note that the reconstruction of the distribution functions of relaxation times from experimental data involves severe problems associated not only with the choice of an adequate model and its mathematical description for a particular material but also with the choice of a numerical method ensuring a stable solution. In some cases, the distribution function of relaxation times can be analytically specified on the basis of simplified model concepts regarding the structure and molecular motion of matter. However, this approach has often led to an unacceptable disagreement between theory and experiment. In this respect, the search for methods providing for precise reconstruction of the distribution functions of relaxation times directly from the experimentally measured spectra $\varepsilon(\omega)$ is of the utmost importance.

In our previous works [3, 4], the dispersion of the permittivity of liquid crystals belonging to cyano derivative compounds was studied by approximating the dielectric spectra $\varepsilon(\omega)$ with the use of trial analytical distribution functions of relaxation times. In these studies, the continuous spectra of relaxation times were described by symmetric or asymmetric functions with one maximum and a monotonic behavior to its right and left. A broad spectrum of relaxation times is a characteristic property of liquid crystals, which is most likely associated with the specific features observed in the molecular and intramolecular motions due to the excitation of collective vibrations of the rigid core and atomic groups of the flexible alkyl chain. It is obvious that the relaxation times of molecular rotations and intramolecular motions should differ significantly from each other and that the function describing a continuous
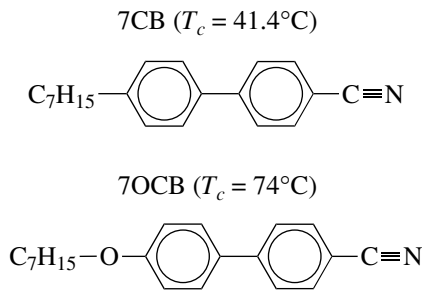
7CB (T_c = 41.4°C)

$$C_7H_{15}{-}\langle\bigcirc\rangle{-}\langle\bigcirc\rangle{-}C{\equiv}N$$

7OCB (T_c = 74°C)

$$C_7H_{15}{-}O{-}\langle\bigcirc\rangle{-}\langle\bigcirc\rangle{-}C{\equiv}N$$

Fig. 1. Structural formulas of the liquid-crystal compounds under investigation and temperatures T_c of the transition from the nematic phase to the isotropic liquid phase.

distribution of these times can have fairly complex behavior.

In this paper, we report on a relatively simple technique for reconstructing the distribution function of relaxation times from the experimentally measured spectra of the real component of the permittivity. This technique was developed with the use of the widely accepted mathematical program package Mathcad. As is known, the accuracy in the measurement of the real component of the permittivity by resonance methods is considerably higher than that of the imaginary component. The proposed technique was tested using two liquid crystals, namely, 7CB and 7OCB. The structural formulas of these liquid crystals are presented in Fig. 1. The measurements were performed both in the nematic liquid-crystal phase for parallel and perpendicular orientations of the molecular director with respect to the polarization of the microwave electric field and in the isotropic phase.

2. The real and imaginary components of the dispersion of the permittivity $\varepsilon(\omega)$ for media with a continuous distribution of relaxation times $g(\tau)$ can be represented in the form [2]

$$\varepsilon'(\omega) = \varepsilon_\infty + (\varepsilon_0 - \varepsilon_\infty)\int_0^\infty \frac{g(\ln\tau)}{1 + \omega^2\tau^2}d(\ln\tau), \qquad (1)$$

$$\varepsilon''(\omega) = (\varepsilon_0 - \varepsilon_\infty)\int_0^\infty \frac{g(\ln\tau)\omega\tau}{1 + \omega^2\tau^2}d(\ln\tau). \qquad (2)$$

Here, ε_0 is the static permittivity and ε_∞ is the high-frequency permittivity. In this case, the normalization conditions for the distribution function of relaxation times are given by

$$\int_0^\infty g(\ln\tau)d(\ln\tau) = 1. \qquad (3)$$

In the proposed technique for reconstructing the distribution function of relaxation times from the experimental data on the real component of the permittivity

$\varepsilon'(\omega)$, it is necessary to change over from integral equations (1) and (2) to a discrete system of nonlinear equations under the assumption that $g(\tau) = 0$ at frequencies outside the measured interval. For this purpose, the experimental dielectric spectrum constructed on a common logarithmic scale in the range ω_{min}–ω_{max} should be represented in the form of a histogram by dividing the entire range into a specified number N of intervals with a width given by the formula

$$\Delta S = \frac{\log(\omega_{max}/\omega_{min})}{N-1}. \qquad (4)$$

For each rectangle in the histograms, there is a particular relaxation frequency ω_i with the corresponding relaxation time $\tau_i = 1/\omega_i$.

Then, the dispersion relation for the ith component of the permittivity can be written as

$$\varepsilon'_{calcd}(\omega_i) = \varepsilon_\infty + k\Delta S(\varepsilon_0 - \varepsilon_\infty)\sum_{m=1}^{N}\frac{g(\log\tau_m)}{1 + \omega_i^2\tau_m^2}. \qquad (5)$$

Here, $\varepsilon'_{calcd}(\omega_i)$ is the calculated permittivity; $g(\log\tau_m)$ is the weighting factor, which is equivalent to the discrete distribution function of relaxation times; and $k = 1/\log e$.

It should be noted that a similar discrete form of the dispersion, but for the imaginary component of the permittivity, was used to develop the numerical algorithm for reconstructing the distribution function of relaxation times from the experimental frequency dependence of the imaginary component of the permittivity $\varepsilon''(\omega)$ for a 5CB liquid crystal in our recent work [5]. This numerical algorithm is based on the histogram method described in [6], which, however, is not applicable to calculations with the use of the real component of the permittivity because of the instability of the obtained solutions.

In expression (5), the weighting factors $g(\log\tau_m)$ can be calculated by a conventional least-squares procedure, according to which the difference between the experimental permittivity $\varepsilon'_{exp}(\omega_i)$ and the calculated permittivity $\Delta\varepsilon'_{calcd}(\omega_i)$ is minimized for each frequency ω_i. In essence, this difference is an objective function of the problem to be solved. However, in the framework of this approach, the problem of determination of the distribution function of relaxation times is practically unsolvable because of the poor stability of the derived solutions, which is determined by the experimental errors $\Delta\varepsilon'_{exp}(\omega)$. In order to eliminate this difficulty, the original mathematical and methodical techniques for obtaining regularized solutions with the use of a nonlinear regression method and special algorithms were proposed by a number of authors (see, for example, [7–10]). In our study, the objective function

(the mean square error) was minimized by the nonlinear least-squares procedure with the Minerror function involved in the Mathcad program package. Despite its main disadvantage of low speed, the Mathcad program offers considerable advantages owing to the natural form of the mathematical expressions, the convenience of programming, and the presence of a large number of built-in subroutines, which make it possible to efficiently solve a great number of problems, including those with systems of linear and nonlinear equations.

Within the proposed approach to the reconstruction of the distribution function of relaxation times, the stability of the solutions is provided by the following conditions. First, the measured dielectric spectrum should be smoothed using an approximation method. Second, the experimental error $\Delta\varepsilon'$ in the measurements should be limited artificially. This is physically justified in the case of a large number of experimental points. Third, the seed form of the function $g(\log\tau_m)$ should be specified. Fourth, the optimum number N of divisions of the frequency range should be chosen for constructing the histogram. For these purposes, initially, all the experimental data on the permittivity $\varepsilon'(\omega)$ were carefully approximated and smoothed in accordance with the Havriliak–Negami equation

$$\varepsilon_{HN}(\omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{[1 + (i\omega\tau)^\alpha]^\gamma}. \tag{6}$$

Here, α and γ are adjustable parameters. In this case, the real components $\varepsilon'(\omega)$ and the imaginary components $\varepsilon''(\omega)$ of the permittivity were calculated using equations taken from [5, 9]. Then, the measurement error was simulated by generating random numbers, which were added to the smoothed spectrum $\varepsilon'_{HN}(\omega)$. Arbitrary positive or negative numbers were generated with the function runif(N, $-\sigma$, σ), which is characterized by a uniform distribution of random numbers in the interval $\pm\sigma \approx 0.05-0.15$. Therefore, the absolute error ($\Delta\varepsilon' \equiv \sigma$) of the smoothed spectrum $\varepsilon'_{HN}(\omega) \pm \Delta\varepsilon'$ could be varied in specified limits.

The initial (seed) components $g(\log\tau_m)$ of the distribution function were specified by a formula that is asymptotically exact for slowly varying dielectric loss functions [11]:

$$g(z_m) \approx \frac{2\eta}{\pi}\left[\varepsilon''_{HN}(z_m) - \frac{\pi^2}{8}\frac{d^2\varepsilon''_{NH}(z_m)}{dz_m^2} + \ldots\right]. \tag{7}$$

Here, $z_m = \log(\tau_m)$ and η is a factor that, at each fixed frequency, was determined from the best fit of the results of the calculations from formula (5) with the use of expression (7) and the approximated dependence $\varepsilon'_{HN}(\omega)$ (6). The values of $g(z_m)$ are represented in the form of a matrix containing one column formed by N components.

When solving a nonlinear iterative problem using the Minerror function, in principle, it is possible to use three special algorithms and specific settings of the program for accelerating the minimization of the objective function $\chi(g)$:

$$\chi(g) = \sum_{i=1}^{N}\{[\varepsilon'_{HN}(\omega_i) \pm \Delta\varepsilon'(\omega_i)] - \varepsilon'_{calcd}(\omega_i)\}^2. \tag{8}$$

However, the preliminary tests showed that the default settings of the Mathcad program can be used in calculations. As a rule, the conjugate-gradient method with multiple calculations of the derivatives can be applied in this case.

As was already noted, the choice of the interval $\pm\sigma$ specifying the measurement error $\Delta\varepsilon'$ and the choice of the threshold χ_{min} at which ($\chi \le \chi_{min}$) the iterative procedure is terminated are important conditions for obtaining a stable solution. In particular, at $\chi \gg \sigma$, the distribution function of relaxation times is usually monotonic, well smoothed, and resembles the Havriliak–Negami or Cole–Cole analytical model distribution functions. At $\chi \longrightarrow \sigma$, the distribution function of relaxation times can exhibit singularities and even clearly defined maxima. Finally, at $\chi \ll \sigma$, the obtained solutions can contain prohibitive negative values of $g(\tau)$.

On this basis, we can choose the values of σ and χ_{min} that will be optimum in order of magnitude. As a rule, this can be achieved by several program runs. Then, it is desirable to accumulate numerical values of $g(\tau)$ through a series of program runs and to average the results. The distribution function of relaxation times thus obtained can be considered a desired function, because it appears to be stable and reproducible with respect to the different program settings.

3. The proposed technique for determining the distribution function of relaxation times was tested with the experimentally measured frequency dependences of the permittivity over a wide range of frequencies from 1 to 10^4 MHz for the 7CB and 7OCB liquid crystals. In the frequency range 1–30 MHz, the permittivity was measured on a Tesla BM-560 standard Q-meter with the use of a conventional measuring cell in the form of a parallel-plate capacitor. In the meter and decimeter wavelength ranges, the dielectric measurements were performed with highly sensitive frequency-tuned resonance microstrip sensors [3, 4]. In the centimeter range, the measurements were carried out using special microstrip [12] and coaxial multifrequency resonators. The amplitude–frequency characteristics of the sensors were recorded on automated digital meters intended for measurements of complex transmission gain factors in the corresponding ranges.

The dielectric spectra of the liquid crystals were measured in the nematic phase at a temperature $T = T_c - 5°C$ and in the isotropic phase at a temperature $T = T_c + 5°C$, where T_c is the temperature of the transition from the nematic phase to the isotropic liquid phase (Fig. 1).
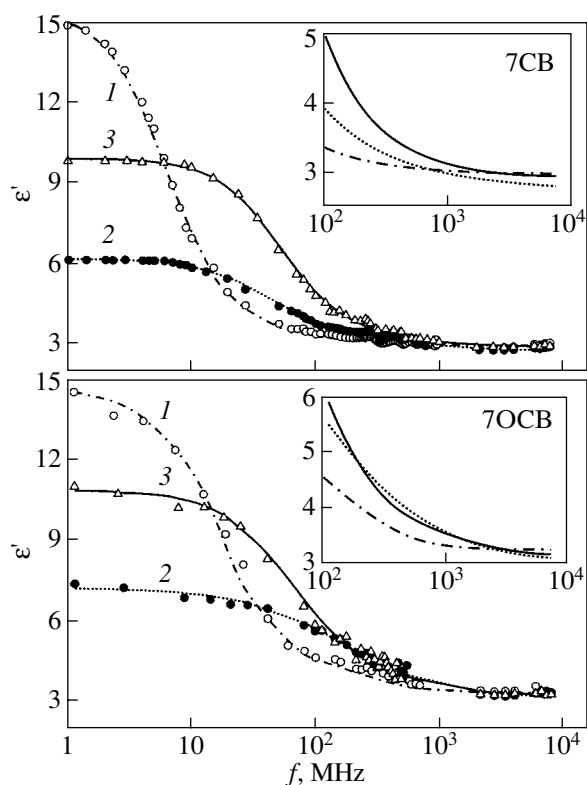
Fig. 2. Dispersion of the real component of the permittivity for the 7CB and 7OCB liquid crystals in the nematic phase for (*1*) parallel and (*2*) perpendicular orientations of the director with respect to the microwave electric field and for (*3*) the isotropic state. Points are the experimental data, and the lines correspond to approximations by the Havriliak–Negami equation.

Figure 2 shows the dielectric spectra of the 7CB and 7OCB liquid crystals both in the nematic phase with parallel and perpendicular orientations of the director and in the isotropic phase. The lines in this figure indicate the approximation of the experimental data by the Havriliak–Negami equation (6), in which the high-frequency permittivities ($\varepsilon'_{\parallel\infty}$, $\varepsilon'_{\perp\infty}$, $\varepsilon'_{is\infty}$) were determined from the measured dispersion at a maximum frequency $f \approx 10^4$ MHz. It is worth noting that, in the experimental dielectric spectra, insignificant resonance features clearly manifest themselves in the high-frequency range [13, 14]. These features were ignored in the present work when approximating the experimental results. It can be seen from the insets to the figures that the parallel component of the permittivity at high frequencies is nearly constant for both of the liquid crystals and that the high-frequency real components of the permittivity are as follows: $\varepsilon'_{\parallel\infty} = 3.05$ for 7CB and $\varepsilon'_{\parallel\infty} = 3.25$ for 7OCB. For these permittivities, we obtained the following adjustable parameters of the approximation with the Havriliak–Negami equation: $\alpha = 1$ and $\gamma \approx 0.83$–0.85 for both liquid crystals. These adjustable parameters correspond to the dielectric spec-

trum described by the conventional Debye equation with one or two relaxation times.

The reconstructed distribution functions of relaxation times $g_{\parallel}(\tau)$ for both liquid crystals are depicted in Figs. 3a. These distribution functions have the form of a Lorentzian line with a somewhat extended wing in the range of short relaxation times. The maxima in these lines correspond to the relaxation times $\tau_{\parallel 1} \approx 25$ ns for 7CB and ≈ 10 ns for 7OCB, which are close in magnitude to those obtained earlier in [3, 15]. These relaxation times characterize the low-frequency orientation range of the dispersion in terms of the Debye equation with one relaxation time. However, as was shown in [3, 15], the dispersion measured over a wide range of frequencies should be described by an equation with two relaxation times. The second relaxation times $\tau_{\parallel 2} \approx 3.2$ ns for 7CB and ≈ 6.3 ns for 7OCB, which were obtained in the present work, correspond to approximately the midpoint of the high-frequency wing $g_{\parallel}(\tau)$ and are also in reasonable agreement with the results reported in [3, 15]. Therefore, the derived distribution function of relaxation times for the parallel component of the permittivity reflects the processes of molecular polarization with two relaxation times and the corresponding weighting factors. However, in our previous paper [4], we demonstrated that the dielectric characteristics measured with high accuracy, in particular, the temperature dependence of the permittivity $\varepsilon'(\omega)$ for the 5CB liquid crystal, can be correctly approximated only with allowance made for a continuous distribution of relaxation times in the high-frequency range.

The dispersion of the perpendicular component of the permittivity $\varepsilon'_{\perp}(\omega)$ (Fig. 2, curves *2*) in the centimeter wavelength range is well pronounced and, most likely, can be observed up to optical frequencies. In this case, the perpendicular component of the permittivity $\varepsilon'_{\perp\infty}$ is often taken as the square of the ordinary refractive index n_o^2 (for example, $n_o = 1.52$ for 7CB). However, the approximation of the dispersion $\varepsilon'_{\perp}(\omega)$ by the Havriliak–Negami equation with this constant leads to unsatisfactory results. This is associated with the fact that the expressions describing the dispersion in the submillimeter and optical wavelength ranges (unlike the ultrahigh-frequency range) can differ significantly from the Debye equation due to the presence of intramolecular resonances. In this respect, the dispersion $\varepsilon'_{\perp}(\omega)$ in the ultrahigh-frequency and optical ranges should be approximated by different dispersion relations, which are given, for example, in [16]. In the present work, in order to approximate more accurately the dispersion $\varepsilon'_{\perp}(\omega)$ and to reconstruct the distribution functions of relaxation times $g_{\perp}(\tau)$, the permittivities experimentally determined at a maximum frequency $f \approx 10^4$ MHz ($\varepsilon'_{\perp\infty} \approx 2.8$ for 7CB and ≈ 3.2 for 7OCB) were
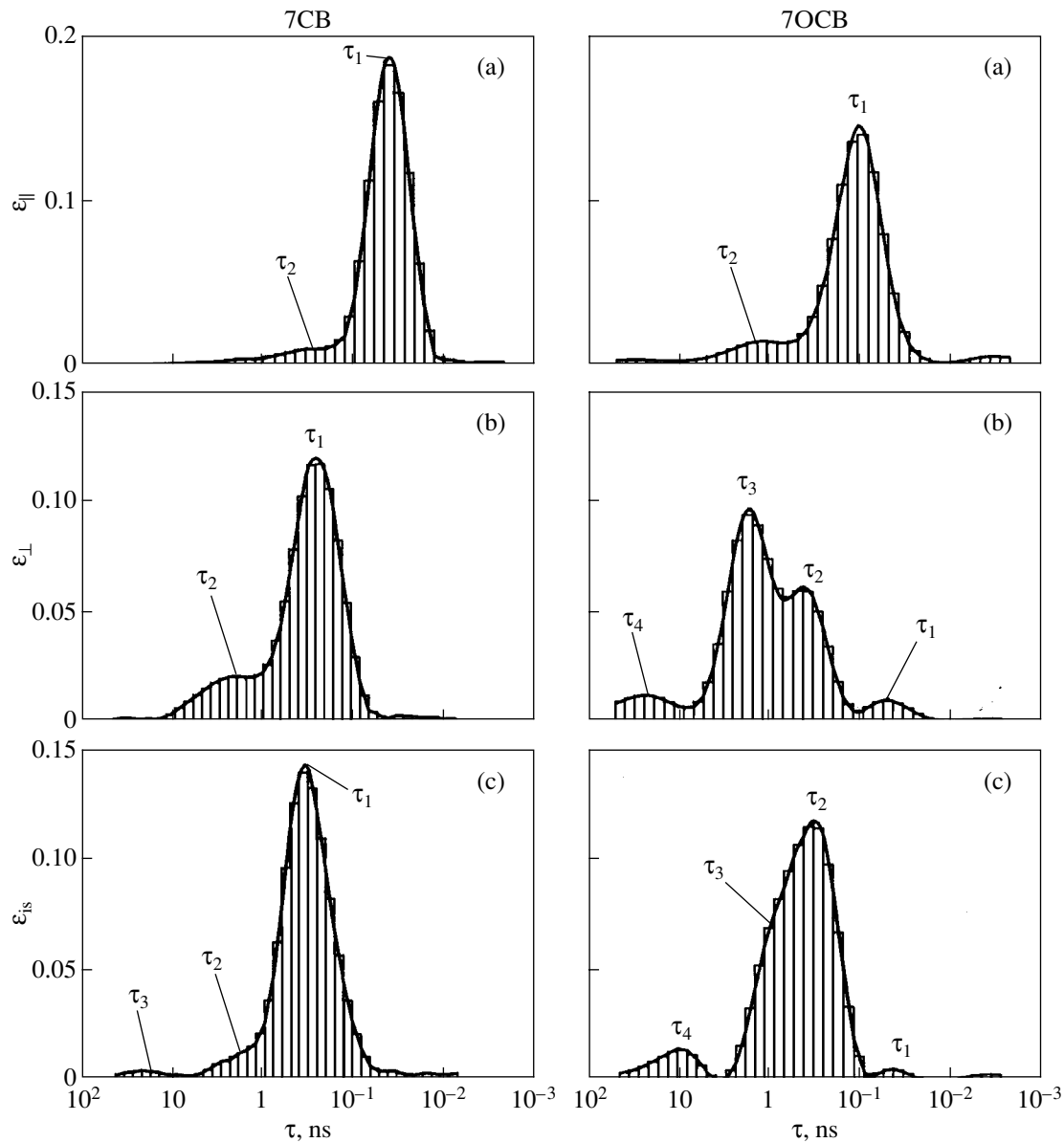
Fig. 3. Distribution functions of relaxation times for the liquid crystals in the nematic phase for (a) parallel and (b) perpendicular orientations of the director with respect to the microwave electric field and for (c) the isotropic state.

used as the constants $\varepsilon'_{\perp\infty}$. In this case, the distribution function of relaxation times is determined only at relaxation times $\tau \geq 0.03$ ns. For these constants, the adjustable parameters in the Havriliak–Negami equation are as follows: $\alpha = 0.91$ and $\gamma = 0.68$ for 7CB and $\alpha = 0.63$ and $\gamma = 0.8$ for 7OCB. The values obtained for the adjustable parameters α and γ, as a rule, correspond to broad asymmetric analytical distribution functions of relaxation times.

Figure 3b shows the reconstructed distribution functions of relaxation times $g_\perp(\tau)$ for the perpendicular component of the permittivity of the 7CB and 7OCB liquid crystals. As can be seen from Fig. 3b, the distribution functions are actually asymmetric and some-

what differ for the liquid crystals under investigation. The distribution function for the 7CB liquid crystal are characterized by a maximum at $\tau_{\perp 1} \approx 3.6$ ns and a clearly defined second relaxation range with a relaxation time $\tau_{\perp 2} \approx 4$ ns. The first relaxation time agrees well with the results obtained in [3, 15] and, apparently, can be associated with the rotation of molecules around the long axis. It seems likely that the second relaxation time can be attributed to the collective motions of the terminal alkyl group. As was noted above, the relaxation spectrum $g_\perp(\tau)$ in the short-time interval is incomplete. This spectrum does not involve the contribution from the high-frequency intramolecular motions, including intramolecular resonances. The relaxation

time $\tau_{\perp 2}$ is an averaged effective quantity, which allows us to approximate rather accurately the dielectric spectrum $\varepsilon'_{\perp}(\omega)$ over a wide range of frequencies, including ultrahigh frequencies.

For the 7OCB liquid crystal (Fig. 3b), the distribution function $g_{\perp}(\tau)$ is characterized by four well-resolved maxima at relaxation times $\tau_{\perp 1} \approx 18$ ns, $\tau_{\perp 2} \approx 2.5$ ns, $\tau_{\perp 3} \approx 0.63$ ns, and $\tau_{\perp 4} \approx 0.035$ ns. The relaxation time $\tau_{\perp 2}$ for the 7OCB compound is close in order of magnitude to the relaxation time $\tau_{\perp 1}$ for the 7CB compound. Consequently, the relaxation time $\tau_{\perp 2}$ can be attributed to the rotation of the rigid molecular core around the long axis. The relaxation times $\tau_{\perp 3}$ and $\tau_{\perp 4}$ should most likely be associated with the motions of alkyl chains, and their large weighting contribution to the relaxation process can be caused by the high intramolecular mobility due to the presence of the oxygen atom located between the core and the alkyl group (Fig. 1).

The distribution function of relaxation times for the isotropic state (Figs. 3c) is slightly asymmetric in shape. This is apparently determined by a set of orientation rotations of the molecules around the short and long axes, as well as by insignificant contributions from the intramolecular motions. It can be seen that the distribution function $g_{\perp}(\tau)$ for the 7CB liquid crystal exhibits three pronounced maxima at relaxation times $\tau_{\perp 1} \approx 2.5$ ns, $\tau_{\perp 2} \approx 0.39$ ns, and $\tau_{\perp 3} \approx 0.05$ ns. The distribution function $g_{\perp}(\tau)$ for the 7OCB liquid crystal is characterized by four maxima at relaxation times $\tau_{\perp 1} \approx 18$ ns, $\tau_{\perp 2} \approx 2.8$ ns, $\tau_{\perp 3} \approx 0.79$ ns, and $\tau_{\perp 4} \approx 0.1$ ns.

4. Thus, we proposed a technique for reconstructing the distribution function of relaxation times directly from the experimental spectrum of the real component of the permittivity. The methodology was demonstrated as applied to the 7CB and 7OCB liquid crystals with the use of the convenient mathematical program package Mathcad. The preparation procedures necessary for minimizing the objective function, which is the sum of the squares of the differences between the calculated and measured permittivities, were described. The developed program was tested for the 7CB and 7OCB liquid crystals in the nematic phase with parallel and perpendicular orientations of the director with respect to the polarization of a microwave electric field. The distribution functions and characteristic relaxation times obtained in this study are in good agreement with the analytical functions and relaxation times determined in other works by approximating the dielectric dispersion with the use of the Debye and Havriliak–Negami equations. The results of the above investigation made a significant contribution to the understanding of the mechanism of relaxation in liquid crystals. The developed technique for reconstructing the distribution function of relaxation times can be easily modified and used in different experiments not only with liquid crystals but also with other materials.

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