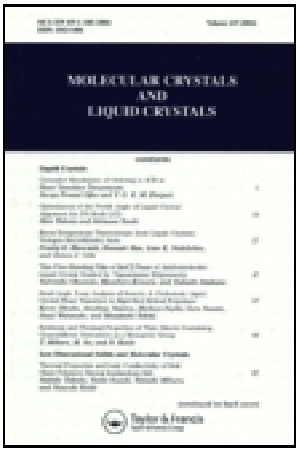

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The Homogeneous Behaviour of Absorption and Dispersion of Ultrasound in the Isotropic Phase of a Nematic Liquid Crystal

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A detailed study of acoustic properties of MBBA and BMOAB in isotropic phase has been carried out. A universal dependence of the excess absorption and the dispersion of sound velocity has been obtained in the range $\omega\tau \sim 10^{-2} - 10^{2}$, which is in full agreement with the predictions of Imura and Okano theory.

1. INTRODUCTION

It is known that the relaxation time for nematic fluctuations increases considerably in the isotropic phase of nematic liquid crystals as one approaches the transition point.[1] Ultrasonic measurements near the nematic–isotropic phase transition showed pronounced anomalies in the propagation of sound.[2] So one may expect the fluctuation parts of absorption and dispersion of the sound velocity to be the homogeneous functions of frequency and temperature as has been obtained for the critical phenomena in simple objects.[3]

$$\left(\frac{C_\omega}{C_0}\right)^2 - 1 = \frac{A(T)}{(1 + A(T))} \cdot \varphi_1(\omega\tau), \tag{1}$$

$$\alpha'\lambda = \pi A(T) \cdot \varphi_2(\omega\tau), \tag{2}$$

where C_ω is the sound velocity at the frequency ω, C_0 is the sound velocity at zero frequency, α' is the fluctuation part of absorption, $\alpha' = \alpha - \alpha_0$, α is the measured absorption and α_0 is the regular part of absorption, λ is the sound wavelength. T is the temperature, φ_1 and φ_2 are the functions of the reduced frequency $\omega\tau$ only (i.e. dispersion and absorption are the homogeneous functions). Here τ is the temperature dependent relaxation time for nematic fluctuations. The presence of biaxial fluctuations seems not to break the homogeneity.[4]

Imura and Okano[5] produced the expressions for the functions $A(T)$, $\tau(T)$, φ_1 and φ_2 using the relaxing heat capacity model.

Until now it has been common practice to compare experiments and theories by fitting the experimental data to the relevant types of the functions $A(T)$, $\tau(T)$, φ_1 and φ_2. Moreover, the power laws are presumed for the temperature dependencies of quantities in (1) and (2) although this item is not clear yet and needs special discussion. The agreement with predictions of Imura and Okano obtained in ref. 2, is qualitative and concerns $(\alpha'\lambda)_{max}$ and $\tau(T)$ only.

The experimental checkup of the main theoretical prediction for homogeneity of absorption and dispersion was not made up to now, as well as that for the degree of universality of the functions φ_1 and φ_2.

2. EXPERIMENTAL

We have carried out the detailed investigation of absorption coefficient and sound velocity in MBBA (4-methoxybenzylidene-4′-*n*-butylaniline) and BMOAB (4-*n*-butyl-4′-methoxy-azoxybenzene). We have chosen MBBA, the classic object in liquid crystal research, because there were numerous data on its properties as well as the investigations of acoustic relaxation.[2] The other substance BMOAB is notable for its high chemical stability and low impurity content. But it is particularly essential that the precise data on heat capacity have been obtained for our specimens lately.[6] Measurements were performed at five frequences ranging from 0.9 to 26.6 MHz and at temperatures 0.1 to 25 K higher than the clearing temperature T_c. Such a region of temperature and frequency gives us an opportunity to test the validity of the formulae (1) and (2) when $\omega\tau$ is being varied by a factor of ten thousand.

The velocity of sound was measured by means of phase-pulse technique using the interferometer with variable acoustic base. The accuracy is better than 0.1%. Absorption was measured by comparing the amplitudes of radio-wave pulse at two distances in liquid. The error in absorption measurements was from 2 to 5% but at 0.9 MHz it was about 20% due to uncertainty in diffraction corrections. Thermal stability was better than 1 mK. The details of technique are published elsewhere.[7]

3. DATA ANALYSIS

The homogeneity of absorption and velocity dispersion is the main question. The forms of functions are of more particular interest. So we have chosen the data processing technique which did not require the forms of functions to be given *a priori*. The procedure is as follows. The plots of $\lg(\alpha'\lambda)$ versus $\lg \omega$ are made for each temperature (Figure 1). If the relationship (2) is to be valid, then $\lg(\alpha'\lambda) = C_1 + \lg \varphi_2(\omega\tau)$ and noting that $\lg \omega = \lg(\omega\tau) + C_2$ one can join all the plots into a single curve by shifting C_1 and C_2 of each plot.

The results of the procedure described above are plotted on Figure 2. Quite analogous procedure was performed for the dispersion data, and the results are on Figure 3. The values $C_1 = \lg A(T)$ and $C_2 = -\lg \tau(T)$ are the same for absorption and dispersion data and they are considered to be adjustable parameters for each temperature point. The criterion is the best coincidence of plots, that is, the scatter along

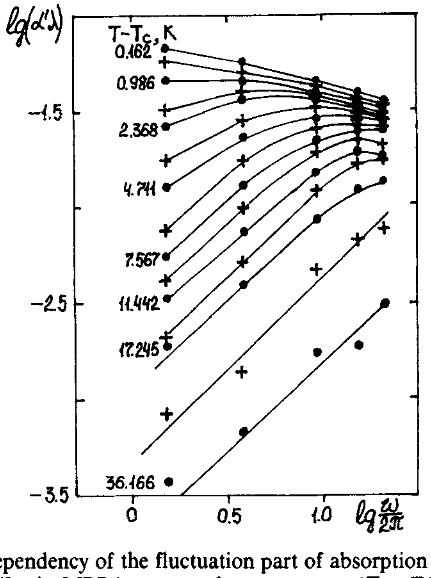

FIGURE 1 The dependency of the fluctuation part of absorption per wavelength $\alpha'\lambda$, versus frequency $\omega/2\pi$ in MBBA at several temperatures $(T - T_c)$. $T_c = 314 \cdot 453$ K.

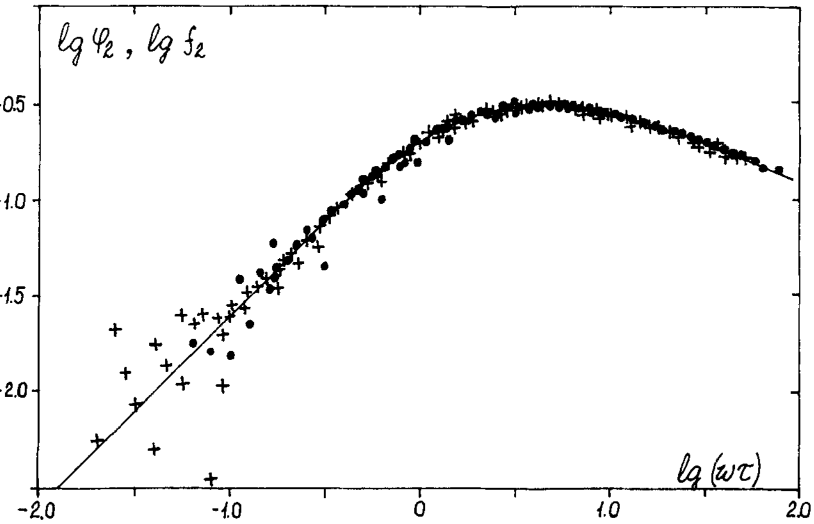

FIGURE 2 The homogeneous behaviour of the fluctuation part of absorption $\alpha'\lambda$ versus the reduced frequency $\omega\tau$ for MBBA (the dots) and BMOAB (the crosses). The solid curve and the values on axes correspond to the results of Imura and Okano.[5]

the joined curve must be as small as possible. The special item is the determination of regular parts α_0 and C_0. They were treated as adjustable parameters and they had been determined from the condition that must be fulfilled for every theory in the hydrodynamic region $\omega\tau \ll 1$; these are $\alpha'\lambda \sim \omega\tau$ and $(C_\omega/C_0)^2 - 1 \sim (\omega\tau)^2$. Figures 2 and 3 demonstrate not only the homogeneity (which is the possibility of making the joined curve from whole array of data) but also the universality of functions φ_1, φ_2 because the data both on MBBA and BMOAB form the same curves. The obtained dependencies are in good agreement with those of Imura and Okano.[5] The connection between $\varphi_1(\omega\tau)$, $\varphi_2(\omega\tau)$ and $f_1(x)$, $f_2(x)$ from ref. 5 is $\varphi_1(\omega\tau) = 1 - f_1(x)$ and $\varphi_2(\omega\tau) = f_2(x)$, where $x = (\omega\tau)^{-1}$.

The scales on Figures 2 and 3 regards to the calculations of Imura and Okano because the ultrasonic data contain no information about the absolute values, of functions φ_1 and φ_2. If the fit to Imura and Okano's predictions is supposed to be adequate, then one could obtain the absolute values of $A(T)$ and $\tau(T)$. Figure 4 shows temperature dependencies of the relaxation time of nematic fluctuations in MBBA and BMOAB and the linewidth of Rayleigh scattering.[1] The agreement is obvious with the exception of the fact that Litster's data are systematically 1.6 times lower than ours. The linewidth is known to differ from the inverse relaxation time by $(\sim (kz)^2)$.[8] However this

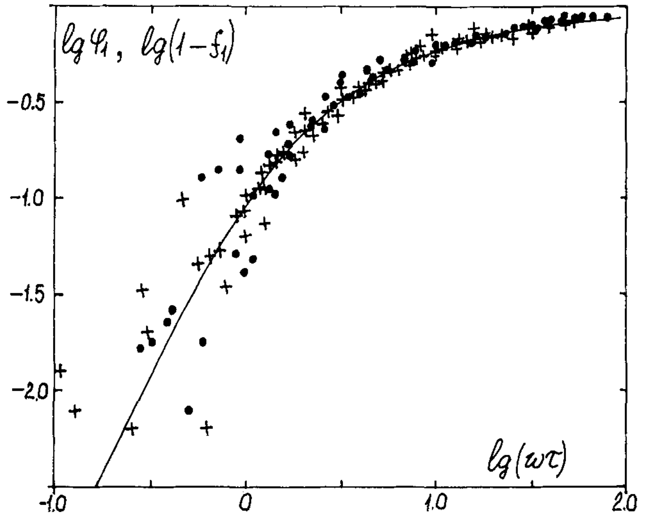

FIGURE 3 The homogenous behaviour of the dispersion of sound velocity $(C_\omega/C_0)^2 - 1$ versus the reduced frequency $\omega\tau$ for MBBA (the dots) and BMOAB (the crosses). The solid curve and the values on axes correspond to the results of Imura and Okano theory.[5]

correction does not exceed 10% for the investigated substances according to our evaluations. On the other hand, our absolute values of τ have been obtained by means of expressions of Imura and Okano, and not by straightforward method. So there still is the question about the correspondence of the "acoustic" and "spectral" values of relaxation time. Here we do not discuss the real form of the temperature dependence of τ. It is not difficult to fit it to the formula $\tau = \nu/a(T - T^*)$, where ν is the shear viscosity, and T^* is the adjustable parameter. Nevertheless, the latest and more accurate data on light scattering[9] as well as the results of heat capacity research[6] indicate that the temperature dependence of inverse susceptibility is non-liner in the whole range and the real type of the τ dependence should be discussed under this context.

In the relaxing heat capacity model the function $A(T)$ is strictly connected to the fluctuation part of the heat capacity:[5]

$$A(T) = g \cdot \frac{\Delta C_p}{C_p^0},$$

where ΔC_p is the anomalous part of isobaric heat capacity, C_p^0 is the regular part of heat capacity, $g \approx$ const. Dividing the acoustic values

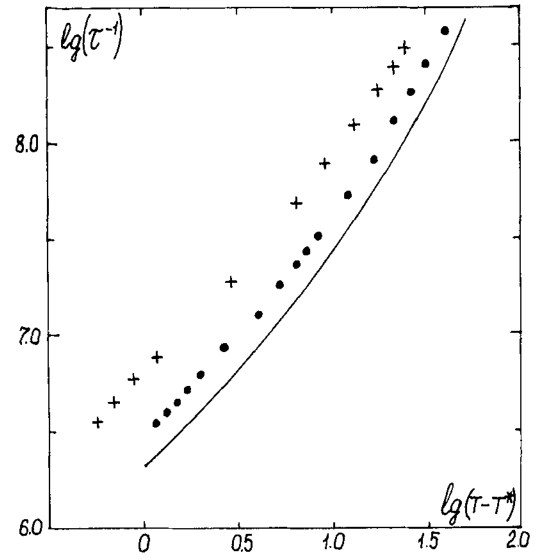

FIGURE 4 The dependency of the inverse acoustic relaxation time, versus temperature ($T - T^*$). $T^* = T_c - 1$ K for MBBA (the dots), and $T^* = T_c - 0.5$ K for BMOAB (the crosses). The solid curve is the linewidth of Rayleigh light scattering in MBBA[1], which is calculated by means of approximation $\tau^{-1} = 1.69 \cdot 10^{11} (T - T^*) \cdot e^{-3570/T}$, $\sec^{-1}$ for our value of T^*.

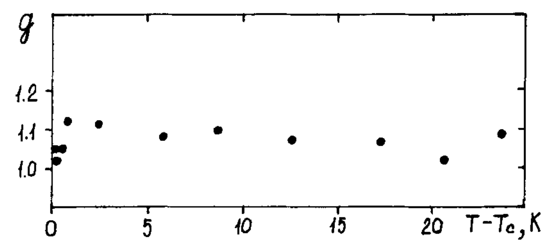

FIGURE 5 The connection of the function $A(T)$ with the fluctuation part of heat capacity $\Delta C_p/C_p^0$[6] for BMOAB. $T_c = 344.897$ K.

$A(T)$ by experimental data of $\Delta C_p/C_p^0$ obtained in[6] at corresponding temperatures we have found g to be a constant really (Figure 5); $g = 1.07 \pm 0.08$.

4. NEMATIC PHASE

We have carried out the measurements of ultrasound absorption and velocity dispersion in the nematic phases of MBBA and BMOAB, too, and in the same range of frequences. The preliminary results of the data processing indicate the existence of homogenous dependencies on ω and τ for the excess absorption and the dispersion. This will be published.

5. CONCLUSION

Our study points out reliably the existence of homogeneity (the common dependency from $\omega\tau$) in dispersion and absorption of sound. This result does not depend on the specific theoretical model. The homogenous functions turned out to be the same for both the two liquid crystals investigated. The use of independent data on the thermal dependency of heat capacity for these substances[6] showed that absorption and dispersion were described quantitatively by the theory of Imura and Okano.[5]

Acknowledgment

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