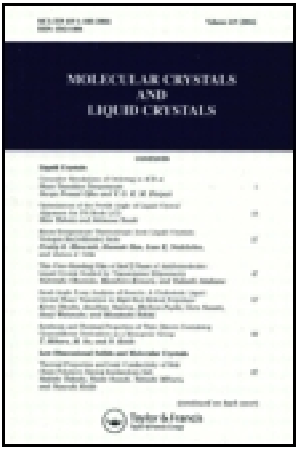

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Nonlinear Effects in Ultrasonic Propagation in a Nematic Liquid Crystal

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NONLINEAR EFFECTS IN ULTRASONIC PROPAGATION IN A NEMATIC LIQUID CRYSTAL

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Measurements of the ultrasonic second harmonic amplitude in the nematic liquid crystal MBBA have been made at fundamental frequencies of 2, 5, and 10 MHz. It is found that the isentropic derivative of the sound velocity, which is related to the nonlinear coupling coefficient C, decreases by approximately 37% as temperature decreases from 35°C above to 20°C below the nematic to isotropic transition temperature, T_c, and may be discontinuous at T_c. In the nematic phase the coupling coefficient is found to be independent, within experimental error, of the angle between the ultrasonic wave vector and the nematic director. This confirms a previous result. Here, however, we are able to place a limit of at most a 4% change in C with a change in direction of propagation. From this value we calculate an upper bound of 10^{-11} dynes/cm^2 for the isentropic derivative of the sound velocity <u>anisotropy</u> with respect to pressure.

In a previous paper[1] we reported the results of measurements of the ultrasonic second harmonic generated in the nematic liquid crystal n-(p-methoxybenzylidene)-p-butylaniline (MBBA). In those measurements the 10 MHz ultrasonic second harmonic generated by a 5 MHz ultrasonic wave was monitored as a function of temperature. Changes in the relative amplitude $A_2(T,\ell)$ of the second harmonic were measured and compared with the general phenomenological expression[2]

$$A_2(T,\ell) = C\, k^2 A_o^2 \, \frac{e^{-2\alpha_1\ell} - e^{-\alpha_2\ell}}{\alpha_2 - 2\alpha_1} \qquad (1)$$

where ℓ, k, A_o, and α_1 indicate the propagation distance, wave vector, initial amplitude, and attenuation coefficient of the fundamental ultrasonic wave, respectively, and α_2 indicates the attenuation coefficient at twice the fundamental frequency. C represents the nonlinear coupling coefficient of the material. In particular, it was found that a small systematic difference ΔA_2 existed between the measured second harmonic and that calculated from Eq. (1). This difference was attributed to a change with temperature in the nonlinear coupling coefficient C. It was also found that within experimental error this difference was independent of whether the sample was unoriented or oriented by a magnetic field. The cases $\hat{n} \parallel \vec{k}$ and $\hat{n} \perp \vec{k}$ were examined; $\hat{n}$ being the director.

In this note we present new data on ultrasonic second harmonics in MBBA. We have extended the frequency range of our previous measurements to include 2-4 MHz and 10-20 MHz (the first number in each pair refers to the fundamental frequency and the second to that of the harmonic). For the 10-20 MHz measurements we have extended the temperature range under study from room temperature to about 35°C above the nematic to isotropic transition temperature T_c. For the 10-20 MHz case, measurements have also been made as a function of propagation distance.

Most of the present measurements were performed in unoriented samples because of the lack of dependence of the coefficient C on the angle between the ultrasonic wave vector $\vec{k}$ and the director $\hat{n}$. This independence was again verified in the present case (see below).

The equipment and procedure used in making our harmonic amplitude measurements are essentially the same as those used previously[1].

Here again as in the 5-10 MHz case, we find, for 10-20 MHz, a systematic difference, $\Delta A_2(T,\ell)$ between the measured second harmonic amplitude and that calculated from Eq. (1). This is shown in Figure 1 where the measured and calculated second harmonic amplitudes have been normalized at $T-T_c$ = 35°C. The dashed line in the figure is the expected difference when the temperature dependence of $k = \omega/v$ is taken into account[3] (ω and v are the angular frequency and sound velocity, respectively). It is seen that the temperature dependence of the wave vector k is not sufficient to account for the difference ΔA_2 between measured and calculated values. Thus, within the framework of Eq. (1), this difference is attributed to a change in the coupling coefficient C(T) with temperature.

The data in Figure 1 show that C decreases uniformly as temperature is lowered in the isotropic phase ($T-T_c > 0$) and further decreases in the nematic phase ($T-T_c < 0$). In the

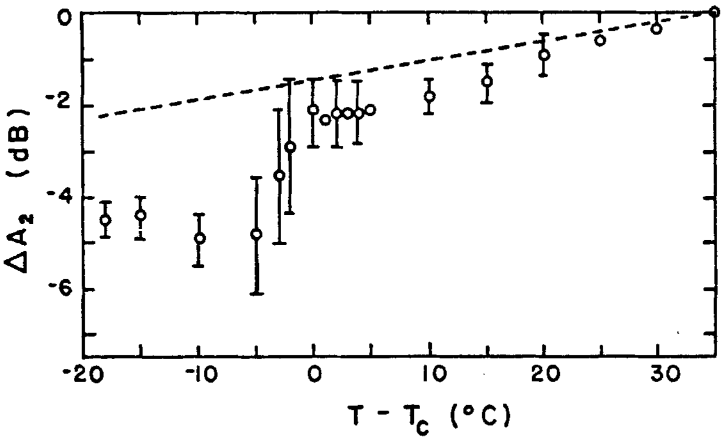

FIGURE 1 ΔA_2 versus $T-T_c$ at 10-20 MHz. The experimental and calculated values have been noramlized at $T-T_c$ = 35°C.

immediate neighborhood of T_c, the error bars of the experimental points are large and it is impossible to say whether there is a discontinuity in C(T) at the transition temperature or if C(T) simply decreases continuously, but much more rapidly with temperature in the range $T-T_c$ = 0 to -3°C. The data at 10-20 MHz are consistent with those at 5-10 MHz[1].

The quantity ΔA_2 which we attribute to a change in C, was also measured as a function of propagation distance, ℓ, at 10-20 MHz for $T-T_c$ = -15°C. The results, normalized so that ΔA_2 = 0 at $T-T_c$ = +5°C (this temperature was chosen to facilitate comparison with our previous work[1]), are shown in Figure 2 (see circles). They indicate that for values of ℓ in excess of some value which lies between 0.25 and 0.44 cm, ΔA_2 is indeed constant. For smaller values of ℓ, ΔA_2 decreases in magnitude. At present we do not know the reason for this decrease, but it may be due to an insufficient distance (in number of wavelengths) traveled by the ultrasonic wave.

Measurements made at 2-4 MHz for values of ℓ = 0.55 cm resulted in a ΔA_2 whose error bars overlap the value zero. This may be due to the presence of an effect similar to that described above where ΔA_2 decreases in magnitude for small ℓ/λ rather than to the absence of a change in C (λ is the

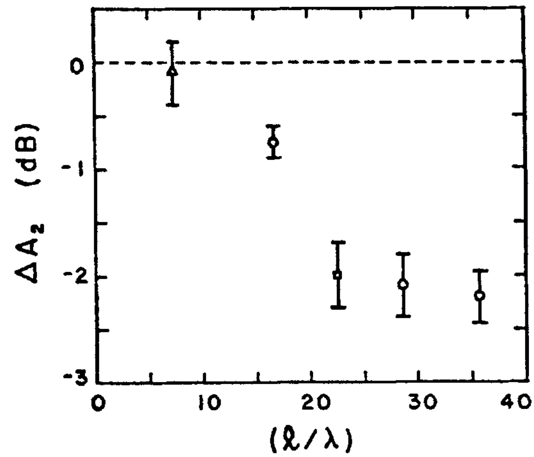

FIGURE 2 $\Delta\alpha$ versus ℓ/λ at $T-T_c = -15°C$. Experimental and calculated values have been normalized at $T-T_c = 5°C$. Triangle: 2-4 MHz. Square: 5-10 MHz. Circles: 10-20 MHz. For 10-20 MHz, the values ℓ/λ = 16.5, 28.5, and 35.6 correspond to ℓ = 0.254, 0.439, and 0.549 cm, respectively.

wavelength at the fundamental frequency). Indeed, for the 2-4 MHz case $\ell/\lambda = 7.2$, which is less than half the value of ℓ/λ at which the 10-20 MHz measurement of ΔA_2 started to decrease from its constant value for large ℓ/λ. Unfortunately, it was not possible to obtain a measurable harmonic amplitude at 2-4 MHz, with our instrumentation, at larger values of ℓ/λ. Values of ΔA_2 as a function of ℓ/λ at $T-T_c$ = -15°C are shown in Figure 2 for 2-4 MHz, 5-10 MHz, and 10-20 MHz.

As mentioned earlier, a check was performed to verify the independence of C on the angle between $\vec{k}$ and $\hat{n}$. This was done at room temperature by orienting the sample with a magnetic field and monitoring the change in amplitude of the ultrasonic second harmonic as the magnetic field was switched from the perpendicular position ($\vec{H} \perp \vec{k}$) to the parallel position ($\vec{H} \parallel \vec{k}$). The measured change in amplitude of the second harmonic was then compared to the change expected on the basis of Eq. (1). This expected change in $A_2(T,\ell)$ was calculated using previously determined values of the attenuation coefficients α_1 and α_2, and independently measured values of their changes $\Delta\alpha_1$ and $\Delta\alpha_2$ (here $\Delta\alpha$ is the difference between the value $\alpha_{||}$ of the attenuation coefficient of an ultrasonic wave with $\vec{k} \parallel \hat{n}$ and the value $\alpha_{\perp}$ of a wave with $\vec{k} \perp \hat{n}$).

A total of five measurements at 2-4 MHz and 5-10 MHz were made. A path length of 1 cm was used in three of these and 0.5 cm in the other two. The average difference in dB between the measured and calculated changes in harmonic amplitude was found to be 0.36 dB indicating a change of at most 4% in the value of the coupling coefficient C with orientation of the director (aligned by the magnetic field). For the two measurements of 0.5 cm path length, the average difference between measured and calculated changes in harmonic amplitude was only 0.16 dB indicating a change of at most 2% in C. The limit of a change in C of at most 4% (or 2% for the smaller, 0.5 cm path length) may be more indicative of errors associated with measuring α and $\Delta\alpha$ (typically 5%) rather than actual changes in the coupling coefficient.

The nonlinear coupling coefficient C can be expressed in terms of the isentropic derivative of the sound velocity v with respect to pressure[4,5].

$$C = 2[1 + \rho_o v(\partial v/\partial p)_s] \qquad (2)$$

where ρ_o is the unperturbed value of the density. Using Eq. (2) and our experimental limit of at most a 4% change in C in going from the $\vec{k} \parallel \hat{n}$ to the $\vec{k} \perp \hat{n}$ configuration, one can establish an upper bound for the anisotropy of the isentropic derivative of the sound velocity with respect to pressure. If one writes $v_{||} = v_{\perp}(1 + \delta)$, then the ratio $C_{||}/C_{\perp}$ of the coupling coefficient for the two configurations can be written, to first order in δ, as

$$\frac{C_{||}}{C_{\perp}} = 1 + \left[\frac{2\rho_o v_{\perp}(\partial v_{\perp}/\partial p)_s \delta + \rho_o v_{\perp}^2(\partial\delta/\partial p)_s}{1 + \rho_o v_{\perp}(\partial v_{\perp}/\partial p)_s}\right] \qquad (3)$$

The velocity anisotropy δ is frequency dependent[6]. For the frequencies used here, δ in MBBA is of the order of 10^{-3}. Also, $\rho \cong 1$ g/cm^3 and $v \cong 1.5 \times 10^5$ cm/sec. The quantity $(\partial v/\partial p)_s$ has not, to our knowledge, been measured for any liquid crystal. However, if one uses the value $\rho_o v(\partial v/\partial p)_s = 5$ typical of organic liquids[7], one finds that the room temperature value of $(\partial\delta/\partial p)_s$ is at most 10^{-11} dynes/cm^2. If, on the other hand, one assumes that the second term in the numerator in brackets in Eq. (3) is of the same order of smallness as the first term, then the ratio $C_{||}/C_{\perp}$ would differ from unity by a few parts per thousand. This small difference would be impossible to determine from second harmonic amplitude measurements.

From our measurements we observe a decrease in the coefficient C with decreasing temperature (see Figure 1). Since the product $\rho_o v$ increases with decreasing temperature ($\rho_o v$ increases[3,8] by ∿ 19% in going from $T-T_c$ = 35°C to $T-T_c$ = -20°C), we conclude that the isentropic derivative $(\partial v/\partial p)_s$ in Eq. (2) decreases with decreasing temperature. The overall decrease in $(\partial v/\partial p)_s$ amounts to approximately 37% over a temperature range of 55°C. Our data also suggest that this derivative may be discontinuous at the transition temperature T_c.

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