Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 21 Mar 2007.

To cite this article: C. A. Castro , A. Hikata & C. Elbaum (1974): Ultrasonic Second Harmonics in a Nematic Liquid Crystal, Molecular Crystals and Liquid Crystals, 25:1-2, 167-173

To link to this article: http://dx.doi.org/10.1080/15421407408083415

Mol. Cryst. Liq. Cryst., 1974, Vol. 25, pp. 167-173

Printed in Dordrecht, Holland

Ultrasonic Second Harmonics in a Nematic Liquid Crystal†

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(Received June 14, 1973)

The amplitude of an ultrasonic second harmonic as a function of temperature has been measured in the nematic liquid crystal MBBA. Measurements were made in the absence of a magnetic field as well as with fields parallel and perpendicular to the direction of ultrasonic propagation. The results indicate a temperature dependent change in the nonlinear coupling coefficient, in the nematic phase, on approaching the nematic-isotropic transition.

INTRODUCTION

Recent ultrasonic work in liquid crystals has been directed primarily towards measurements of the attenuation constant α and the velocity of propagation v.[1,2] From these quantities one can observe relaxation processes occurring in the medium and obtain data concerning some of the various viscosity coefficients used in describing the hydrodynamics of liquid crystals. In this note we present data regarding the relative amplitude of the 10 MHz ultrasonic harmonic generated by a 5 MHz ultrasonic wave.

Measurements were made on the nematic liquid crystal *N*-(*p*-Methoxybenzylidene)-*p*-butylaniline (MBBA). Samples were obtained from Eastman Organic Chemicals and no attempt was made to purify further the material. During measurements the liquid crystal was placed in a gold plated brass container, the bottom of which was a 10 MHz quartz transducer. A movable plunger served as mount for a 5 MHz quartz transducer. In this manner the separation between the two transducers could be varied from zero to 1.5 cm, but in most cases it was set

† Research Supported by the National Science Foundation.

between 6 and 7 mm. The whole apparatus was contained in an atmosphere of nitrogen.

Many runs were performed and in each run the sample was heated to a temperature above its clearing temperature and allowed to cool. During the cooling the relative amplitudes of the 10 MHz harmonic and the 5 MHz fundamental were recorded simultaneously and continuously as a function of temperature. For each run the amplitude of a 10 MHz fundamental wave was measured independently during cooling, again as a function of temperature. In both cases, absolute attenuation measurements were also taken at temperatures close to room temperature. The temperature of the sample was monitored using a thermistor in a branch of a *dc* resistance bridge. Standard procedures for cooling the samples were established and used throughout. The average cooling rate was about 0.4°C per minute. Fresh sample material was used every two runs although one sample was used for three runs.

RESULTS

Preliminary experiments were made to determine the magnitude of the magnetic fields necessary to align our samples. The results of these are shown in Figure 1 and on this basis a field of 1 KOe was chosen for all runs involving bulk alignment. Similar measurements were also repeated at temperatures above the clearing temperature where no effect of magnetic field on attenuation is expected, and within our accuracy none was observed.

All curves obtained show a distinct change in the slope of the relative attenuation versus temperatures. This change occurs at a temperature T_0 which

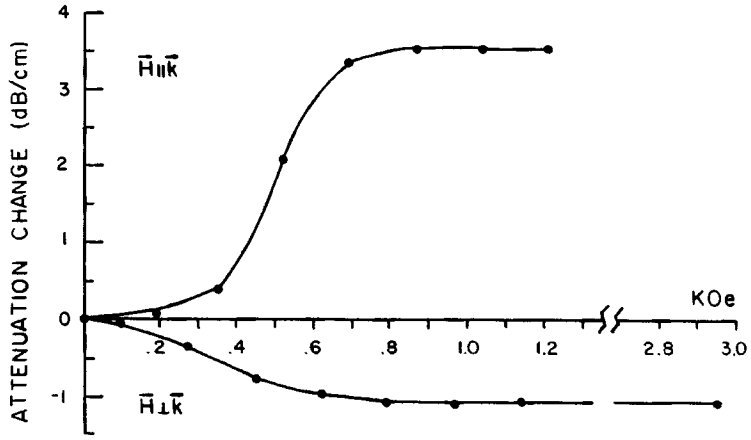

FIGURE 1 Change in ultrasonic attenuation (5 MHz) versus magnetic field, at t=23°C.

varied slightly from sample to sample and with sample history. T_0 ranged from 38.3 to 40.8°C and we interpret T_0 as indicating the onset of the isotropic-nematic transition. Subsequent visual examination of the sample during cooling also bears this out. The data obtained from three runs are shown in Figures 2 and 3. In Figure 2 we present measured values of the relative attenuation of the 5 MHz fundamental as a function of temperature. The values have been normalized to unity at 5°C above T_0. The actual values of the attenuation for $T-T_0 = 5°C$ ranged from 9.7 to 16.8 dB/cm for α_5 and from 13.5 to 22.7 dB/cm for α_{10}. Figure 3 shows similar data for 10 MHz. In both cases the open circles refer to measurements made in the absence of a magnetic field and the full circles and full squares to measurements made with a magnetic field of 1 KOe once parallel and once perpendicular to the ultrasonic wave vector k. Both these figures show that the relative attenuation is largest when H is parallel to k and smallest when it is perpendicular. This behavior is typical of all the data obtained and is in agreement with Figure 1. Similar differences in attenuation have been reported by others.[3]

Figure 4 shows the measured values of the relative amplitude of the 10 MHz harmonic generated in the sample. We compare our results to the general, phenomenological expression[4]

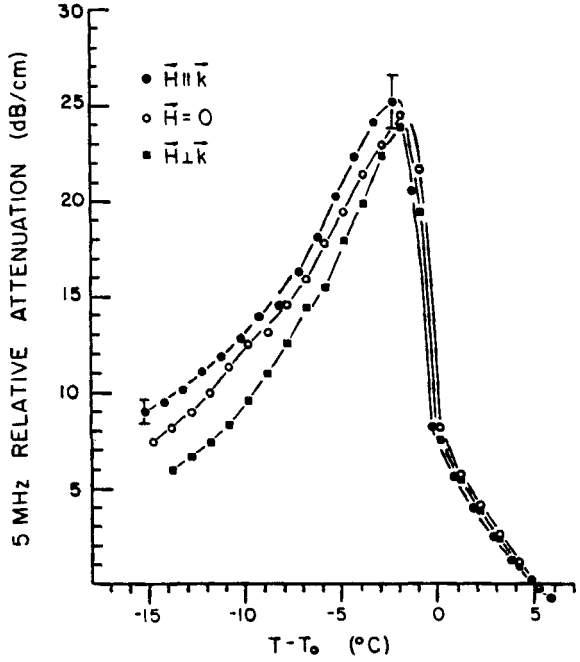

FIGURE 2 5 MHz relative attenuation versus $T-T_0$.

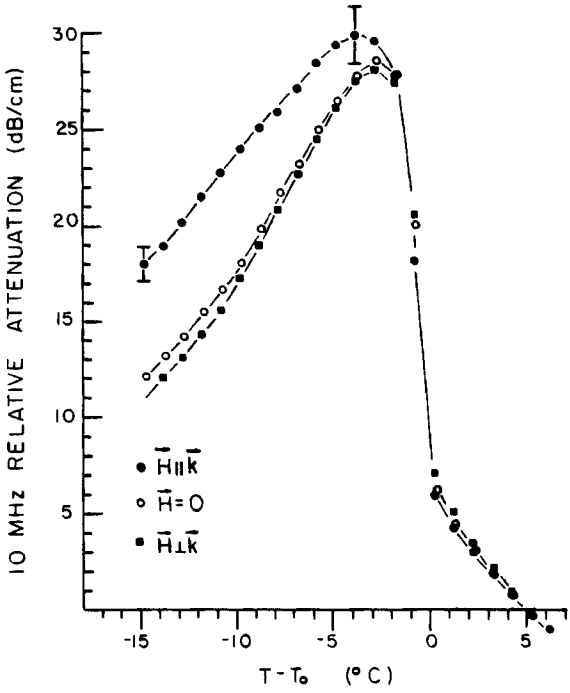

FIGURE 3 10 MHz relative attenuation versus $T-T_0$.

$$A(T,\ell) = C\,k^2 A_0^2(\ell=0)\,\frac{\exp[-2\alpha_5\ell]-\exp[-\alpha_{10}\ell]}{\alpha_{10}-2\alpha_5} \tag{1}$$

where ℓ, k and $A_0(\ell = 0)$ refer to the propagation distance, the wave vector and the initial amplitude of the 5 MHz fundamental, respectively. C represents the nonlinear coupling coefficient of the material. Formula (1) is derived by including in the stress strain relation terms containing the squares of the displacement gradients, and solving the resulting equation of motion approximately by iteration. The reader is referred elsewhere for details regarding this calculation.[4] The use of this expression implies, of course, that the physical features specific to liquid crystals are contained in the coupling coefficient C.

The function $A(T,\ell)$ was evaluated from (1) using our data for relative attenuation together with one value of absolute attenuation for each of the quantities $\alpha_5(T)$ and $\alpha_{10}(T)$. This was done for every run and the computed values were compared to those measured during that run. The solid curve in Figure 4 represents one such calculation where all values have been normalized to unity at T_0 + 5° C. The values of absolute attenuation used here were measured at $T - T_0 = -15$°C and were $\alpha_5 = 19.3$ dB/cm and $\alpha_{10} = 31.5$ dB/cm. In this

run ℓ was 6.8 mm and H was parallel to k. The points in Figure 4 represent the measured values of the amplitude of the harmonic and are typical of all runs analyzed.

Two other aspects of Eq. (1) have been examined, i.e., the dependence of $A(T,\ell)$ on the square of the amplitude of the fundamental and the behavior of $A(T,\ell)$ with respect to distance traveled ℓ. The first of these is shown in Figure 5 where the amplitude change of the harmonic is plotted against the amplitude change of the fundamental. As for the second aspect mentioned above, from the expression for $A(T,\ell)$ it is seen that the amplitude of the harmonic grows to a maximum (for constant T) at a distance ℓ_{max} given by the relation

$$\ell_{max} = \ln(2\alpha_5/\alpha_{10})/(2\alpha_5 - \alpha_{10}) \tag{2}$$

and then decays for increasing ℓ. This behavior was observed in the isotropic phase (typically at temperatures T greater than 60°C) where an increase in the distance ℓ produced at first an increase in the amplitude of the harmonic, follow-

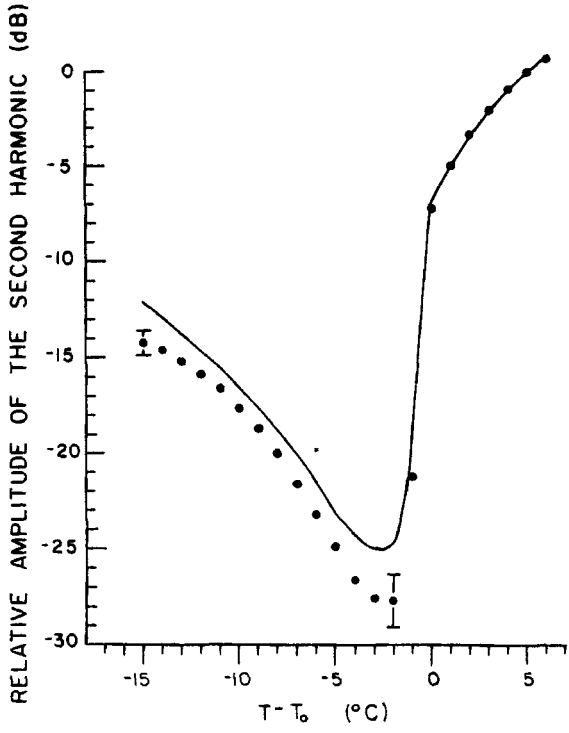

FIGURE 4 Relative amplitude of the second harmonic versus $T-T_0$. The solid curve represents calculated values of $A(T,\ell)$ using expression (1). The full circles are measured values.

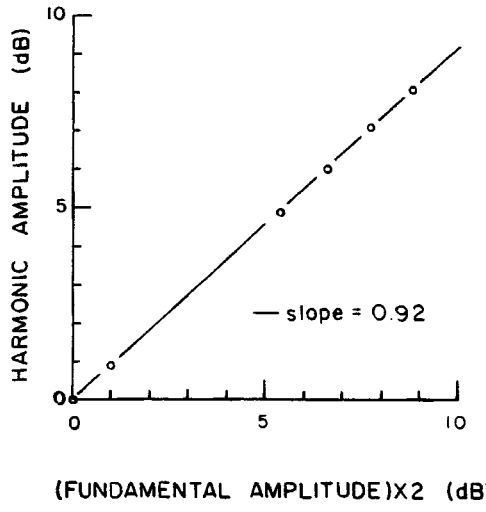

FIGURE 5 Amplitude change of the second harmonic versus the square of the amplitude change of the fundamental.

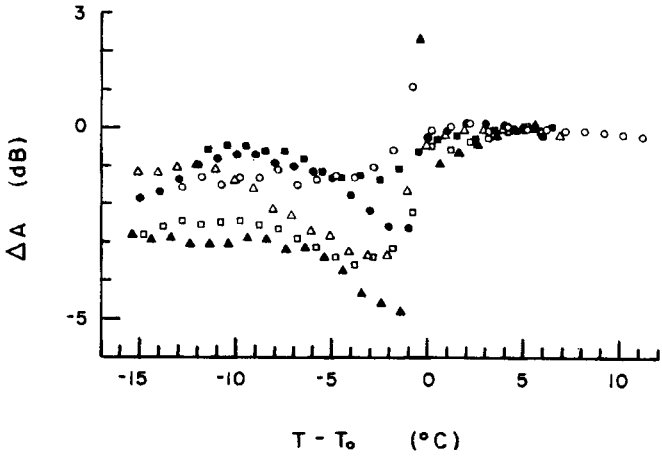

FIGURE 6 Difference between the mearured value of the second harmonic and that obtained using (1), versus $T-T_0$. Data from six runs with $\vec{H}\|\vec{k},\vec{H}\perp\vec{k}$ and $\vec{H}=0$.

ed by a decrease, in accordance with Eq. (1). In the nematic phase where the attenuations involved are much larger, one can only see the decay of the harmonic since ℓ_{max} here is of the order of a millimeter.

DISCUSSION

Our experiments show that the dependence of the amplitude of the second harmonic on temperature and magnetic field can be primarily accounted for by the behavior of the two attenuation coefficients $\alpha_5(T)$ and $\alpha_{10}(T)$. Although bulk alignment of the liquid crystal affects the values of the attenuation coefficients, it does not affect the generation of the harmonic (i.e., the coupling coefficient C) in the sense that the same type of behavior is encountered for H both parallel and perpendicular to k, as well as when no magnetic field is applied. There is, however, a small systematic difference ΔA, present in all runs, between the experimental points and the values calculated using (1). This difference is such that when the values are normalized to some value obtained in the isotropic phase (as in our data), the measured value of the harmonic is always smaller in the nematic phase than that calculated from (1).

The dependence of ΔA on temperature is given in Figure 6, which shows that a significant decrease in the amplitude of the second harmonic occurs on approaching the transition temperature T_0. The calculated results used here take into account the temperature dependence of the attenuation, which is measured independently. In order to verify whether the temperature dependence of $k = \frac{\omega}{v}$ is responsible for the observed behavior, the dependence of sound velocity on temperature[5] measured at 6 Mhz is used. It is thus found that a decrease in the amplitude of the second harmonic of at most 0.8 dB, relative to the value given by the solid curve of Figure 4, could be accounted for. Since the actual difference is larger than this value, it is concluded that the nonlinear coupling coefficient displays a temperature dependence, in the nematic phase, on approaching the nematic-isotropic transition.

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