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D. G. Gleed[a]; J. R. Sambles[a]; G. W. Bradberry[a]
[a] Thin Film and Interface Group, Department of Physics, University of Exeter, Exeter, England

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A method for indirectly measuring the second sound velocity in smectic A liquid crystals

by D. G. GLEED, J. R. SAMBLES and G. W. BRADBERRY
Thin Film and Interface Group, Department of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, England

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We present a simple method by which the variation of the hypersound velocity with the angle between the liquid crystal director and the scattering plane is fitted to theory. Combinations of bulk elastic constants can be obtained together with a value for the second sound velocity. This method is used to study the smectic A phase of 4-*n*-octyl-4′-cyanobiphenyl. We compare our results with previous work.

1. Introduction

In the smectic A phase of a liquid crystal two propagating phonon modes can exist [1, 2]. One corresponds to the usual sound velocity in a fluid, i.e. longitudinal compression and the other to a shear-like motion of the smectic layers at approximately constant density. This second mode which is often referred to as second sound has a much lower velocity than the first mode. Several methods have been used to study the behaviour of this mode. Acoustic techniques enable detailed investigations at lower frequencies (providing measurements in a good approximation to the hydrodynamic regime) [3, 4]. Brillouin scattering has also been used both directly [5] and indirectly [6] to measure the second sound velocity. The indirect method involves measuring the variation of the normal hypersound velocity as a function of the angle between the liquid crystal director and the scattering plane.

The angular dependence is a relatively elaborate function of three elastic constants and a large number of data points need to be acquired to allow good least squares fitting to give these constants. We present a simpler method by which a smaller number of data points can be used to fit to a reformatted theory in the form of a straight line. From this straight line fit we not only confirm the theory in its general form but obtain combinations of the elastic constants, which are then used to fit the unreduced data set by adjustment of one parameter. Results are given for the temperature dependence of the elastic constants and the second sound velocity in the smectic-A phase of the liquid-crystal 4-*n*-octyl-4′-cyanobiphenyl (8CB) and comparisons are made with existing data.

2. Background

Several theories exist for the phonon modes in the smectic A phase of liquid-crystalline materials [1, 2]. These predict that, in general, there are two sets of propagating modes with velocities given by

$$\varrho^2 V_1^2 V_2^2 = (AB - C^2)\sin^2\theta\cos^2\theta \quad (1)$$

and

$$\varrho(V_1^2 + V_2^2) = A + (B + 2C)\cos^2\theta, \tag{2}$$

where A is the bulk modulus, B and C are elastic constants related to motion of the smectic layers, and θ is the angle between the director and the scattering plane. V_1 is the velocity associated with the mainly longitudinal mode and V_2 is the second sound velocity. At $\theta = 0°$ and $90°$ this second propagating mode is overdamped. The angular dependence of the velocities of the two propagating modes are given by

$$\varrho V_1^2 \approx A + 2C\cos^2\theta + B\cos^4\theta \tag{3}$$

and

$$\varrho V_2^2 \approx B\sin^2\theta\cos^2\theta. \tag{4}$$

When Brillouin scattering is used to study the temperature dependence of the hypersound velocity in liquid crystals it is V_1 which is usually measured [7]. V_2 is a much smaller velocity ($V_1(45°) \approx 5V_2(45°)$) and hence the corresponding Brillouin signal will generally be obscured by the elastically scattered light. To observe V_2 directly the contrast of the instrument used to acquire the data must be high enough to resolve the weak Brillouin lines which are very close to the strong elastic Rayleigh line.

However, by measuring the angular dependence of V_1 and fitting the data to theory, an indirect measurement can be made of V_2. As can be seen from the form of equation (3) many data points would be needed to obtain a good least squares fit to the data. It is, however, possible to reformat equations (1) and (2) so that the theory takes the form of a straight line. This method provides combinations of the liquid crystal bulk elastic constants and gives a good fit with fewer data points.

3. Reformatting the theory

The first step is to express equations (1) and (2) in a slightly different form [8] using another set of elastic constants, C_{11}, C_{13} and C_{33}. C_{11} and C_{33} are elastic constants corresponding to longitudinal sound propagation in the x and z direction (the z axis is perpendicular to the smectic A layers, i.e. along the director). C_{13} is a measure of the elastic anisotropy. This gives us

$$\varrho^2 V_1^2 V_2^2 = (C_{11}C_{33} - C_{13}^2)\sin^2\theta\cos^2\theta \tag{5}$$

and

$$\varrho(V_1^2 + V_2^2) = C_{11}\sin^2\theta + C_{33}\cos^2\theta. \tag{6}$$

Comparing equations (1) and (2) with (5) and (6) it can be seen that

$$A \equiv C_{11}, \quad B \equiv C_{33} + C_{11} - 2C_{13} \quad \text{and} \quad C \equiv C_{13} - C_{11}.$$

Substituting for ϱV_2^2 from equation (5) in (6) gives

$$\varrho V_1^2 = C_{11}\sin^2\theta + C_{33}\cos^2\theta - \frac{1}{\varrho V_1^2}(C_{11}C_{33} - C_{13}^2)\cos^2\theta\sin^2\theta. \tag{7}$$

If we define V_{1C} by the relation $V_{1C}(\theta) = V_1(90° - \theta)$ we can then write

$$\varrho V_{1C}^2 = C_{11}\cos^2\theta + C_{33}\sin^2\theta - \frac{1}{\varrho V_{1C}^2}(C_{11}C_{33} - C_{13}^2)\cos^2\theta\sin^2\theta. \tag{8}$$

Combining equations (7) and (8) gives

$$(V_1^2 + V_{1C}^2) = \frac{1}{\varrho}(C_{11} + C_{33}) - \frac{1}{\varrho^2}(C_{11}C_{33} - C_{13}^2)\sin^2\theta\cos^2\theta\,\frac{(V_1^2 + V_{1C}^2)}{V_1^2 V_{1C}^2}. \tag{9}$$

Equation (9) has the form of a straight line if we take

$$X = \left(\frac{V_1^2 + V_{1C}^2}{4V_1^2 V_{1C}^2}\right)\sin^2 2\theta \tag{10}$$

and

$$Y = V_1^2 + V_{1C}^2. \tag{11}$$

We can, therefore, obtain two combinations of elastic constants, $(1/\varrho)(C_{11} + C_{33})$ from the intercept and $(1/\varrho^2)(C_{11}C_{33} - C_{13}^2)$ from the slope by reducing the data into pairs of velocities at complementary angles. Note that the first combination is simply given by

$$V_1^2(0°) + V_1^2(90°) = \frac{1}{\varrho}(C_{11} + C_{33}).$$

Using equation (7) the expression for the angular dependence of V_1 is

$$V_1^2 = \frac{1}{2\varrho}(C_{11}\sin^2\theta + C_{33}\cos^2\theta) + \frac{1}{2\varrho}[(C_{11}\sin^2\theta + C_{33}\cos^2\theta)^2 - 4(C_{11}C_{33} - C_{13}^2)\sin^2\theta\cos^2\theta)]^{1/2}. \tag{12}$$

The angular dependence of V_2 would be given by the right hand side of equation (12) but with a negative sign in front of the square root term. It is then possible to fit the unreduced data set to equation (12) by choosing $(1/\varrho)C_{11}$ or $(1/\varrho)C_{33}$ then using the intercept, $(1/\varrho)(C_{11} + C_{33})$ to select the other elastic constant. The final parameter required is $(1/\varrho^2)(C_{11}C_{33} - C_{13}^2)$ which is the slope obtained from the straight line fit.

When $\theta = 45°$ the equations for the angular dependence of the velocities reduce to a form which just include the combinations of elastic constants obtained from the straight line fit. Using equations (5) and (12) with $\theta = 45°$ we obtain the expression

$$\varrho V_2^2(45°) = \frac{(C_{11}C_{33} - C_{13}^2)}{(C_{11} + C_{33}) + [(C_{11} + C_{33})^2 - 4(C_{11}C_{33} - C_{13}^2)]^{1/2}}. \tag{13}$$

for the second sound velocity at 45°

4. Experimental

A study was made of 8CB in the smectic A phase. The phase transition temperatures for 8CB are

$$C \rightarrow S_A\ 21{\cdot}5°C, \quad S_A \rightarrow N\ 33{\cdot}5°C, \quad N \rightarrow I\ 40{\cdot}5°C.$$

Thin film samples (100 μm) were aligned by sandwiching the liquid crystal between glass plates. The homogeneous alignment required to study the variation in hypersound velocity as a function of the angle between the director and the scattering plane was achieved using a thin film of SiO_x, evaporated at 60° to the substrate normal, as an aligning layer.

An argon-ion laser (λ = 488 nm) in single mode operation was used for the scattering experiments. The scattered light was collected at 90° to the incident beam and frequency analysed using a Burleigh triple-pass scanning interferometer together with a low noise photon counting tube (EMI 9893), fast scaler and data store. The samples were held in a heated stage, which allowed the temperature to be controlled in 0·1°C steps and the sample temperature was monitored using thin calibrated copper-constantan thermocouples inserted in the scattering cell. Measurements were made in the temperature range 23·5 to 32·0°C for various different angles between the director and the scattering plane. The angle between the incident beam and the normal to the liquid crystal cell was set to 45°. With this geometry it is not necessary to know the refractive index of the liquid crystal in order to determine the hypersound velocity [9].

5. Results

At each temperature a set of spectra was obtained from which the angular dependences of V_l were found. These enabled a straight line plot to be made using the substitutions of equations (10) and (11). Figure 1 shows the spectra recorded at 32·0°C with the director at 45° to the scattering plane. The Brillouin lines due to the longitudinal phonons are clearly seen at channel numbers 25, 105, 145 and 225. At this angle the second sound velocity has its greatest value. The Brillouin lines corresponding to this mode however are very close to the centre of the Rayleigh line and are not directly observed. Figures 2 and 3 show the straight line fit for the data collected at

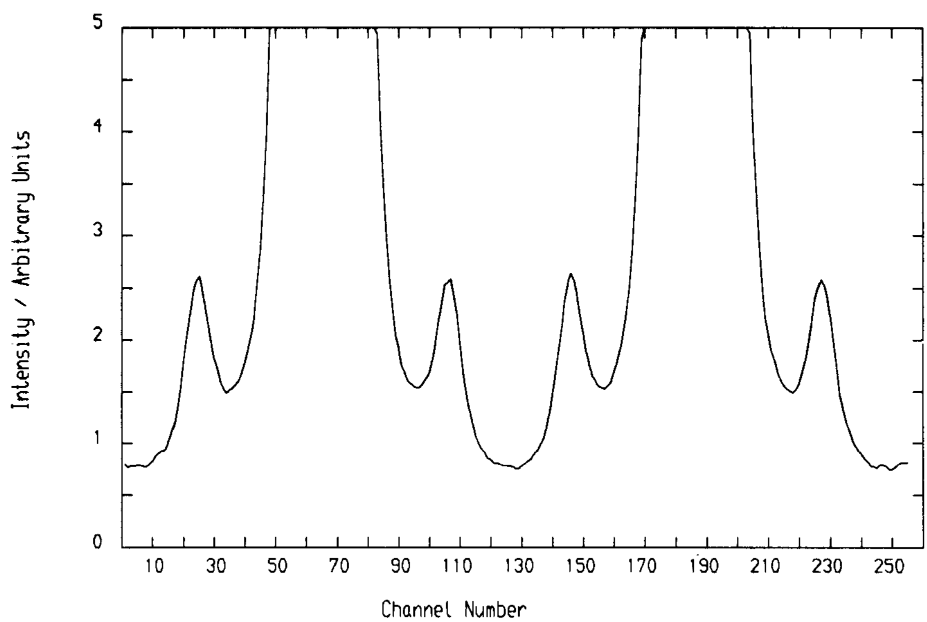

Figure 1. A typical Brillouin spectra for 8CB in the smectic A phase at 32·0°C. The angle between the scattering plane and the liquid crystal director is 45°.

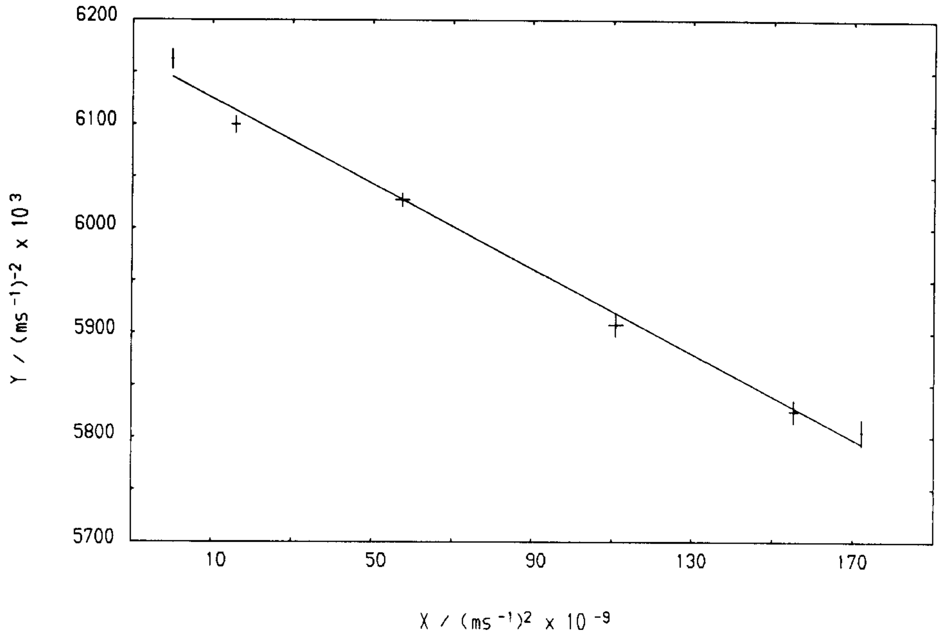

Figure 2. The hypersound velocity, V_l, as a function of the angle, θ, between the scattering plane and the director for 8CB at 23·5°C plotted with redefined variables. $X = (V_l^2 + V_{lC}^2/4V_l^2 V_{lC}^2)\sin^2 2\theta$ and $Y = V_l^2 + V_{lC}^2$ where $V_{lC}(\theta) = V_l(90° - \theta)$.

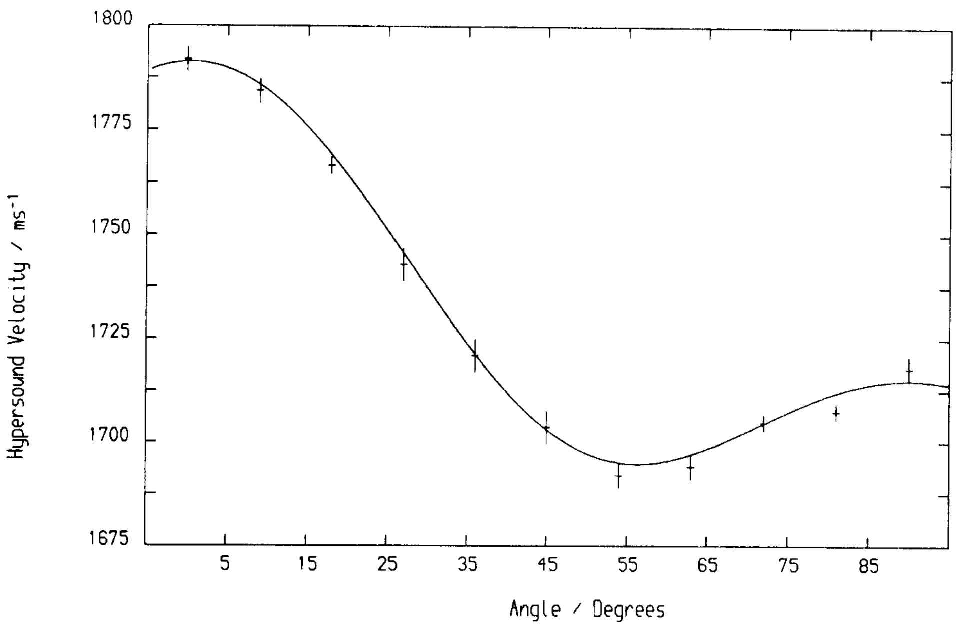

Figure 3. The hypersound velocity against the angle between the scattering plane and the director for 8CB at 23·5°C fitted to theory.

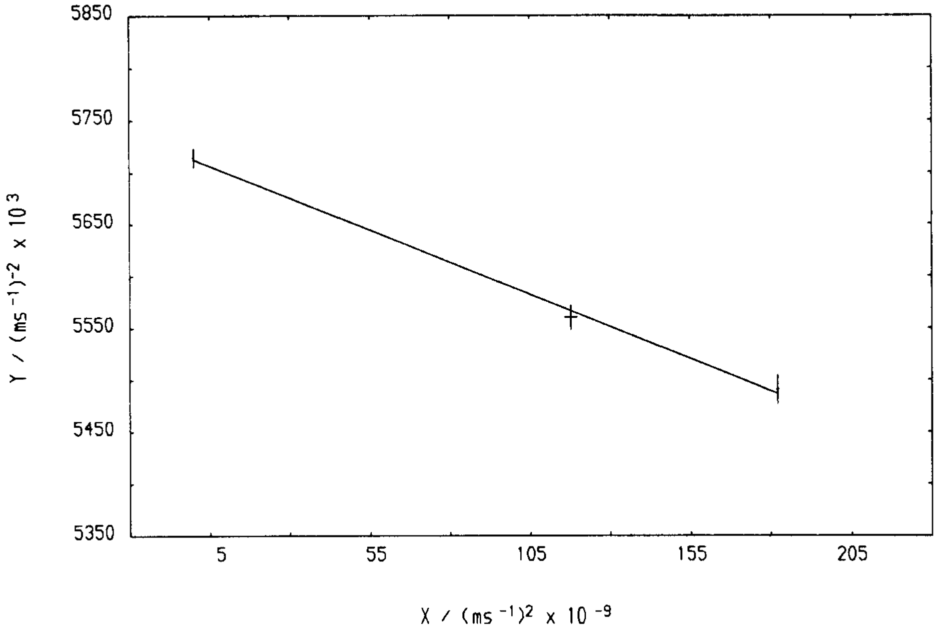

Figure 4. The hypersound velocity, V_1, as a function of the angle, θ, between the scattering plane and the director for 8CB at 30·5°C plotted with redefined variables. $X = (V_1^2 + V_{1C}^2/4V_1^2V_{1C}^2)\sin^2 2\theta$ and $Y = V_1^2 + V_{1C}^2$ where $V_{1C}(\theta) = V_1(90° - \theta)$. Only a small number of data points are used.

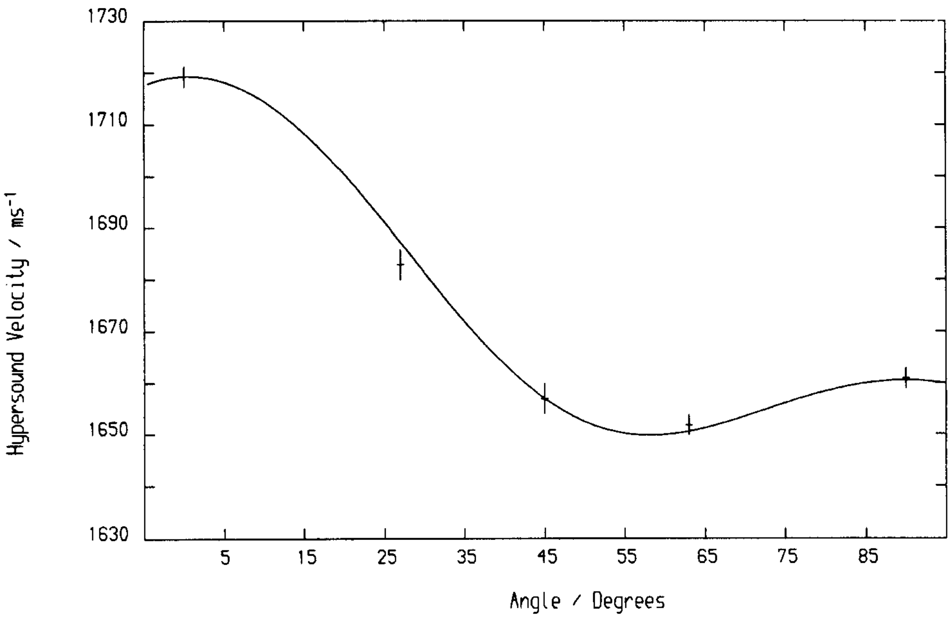

Figure 5. The hypersound velocity against the angle between the scattering plane and the director for 8CB at 30·5°C fitted to theory using a smaller number of data points.

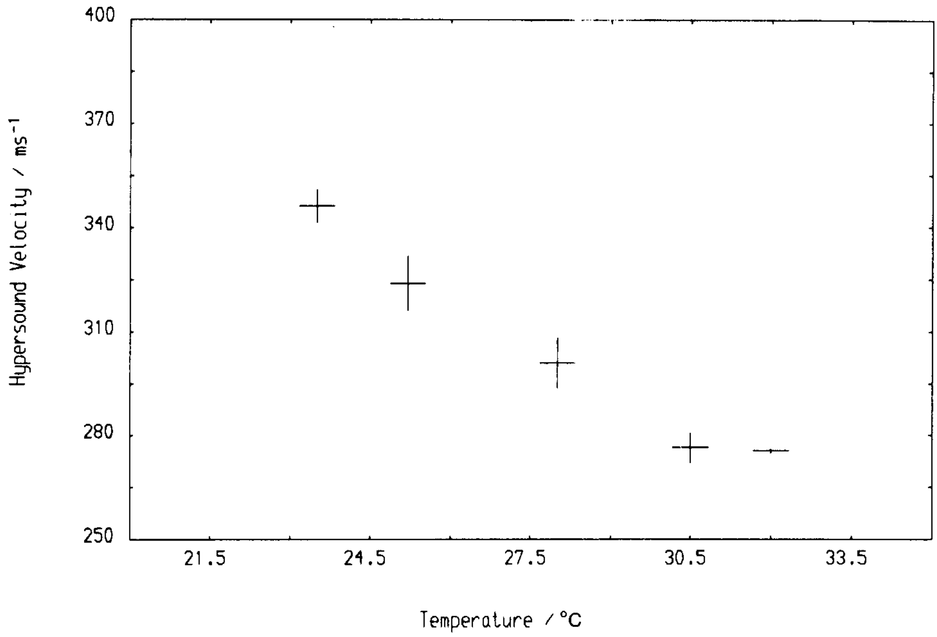

Figure 6. The temperature dependence of the second sound velocity in 8CB when the angle between the scattering plane and the director is 45°.

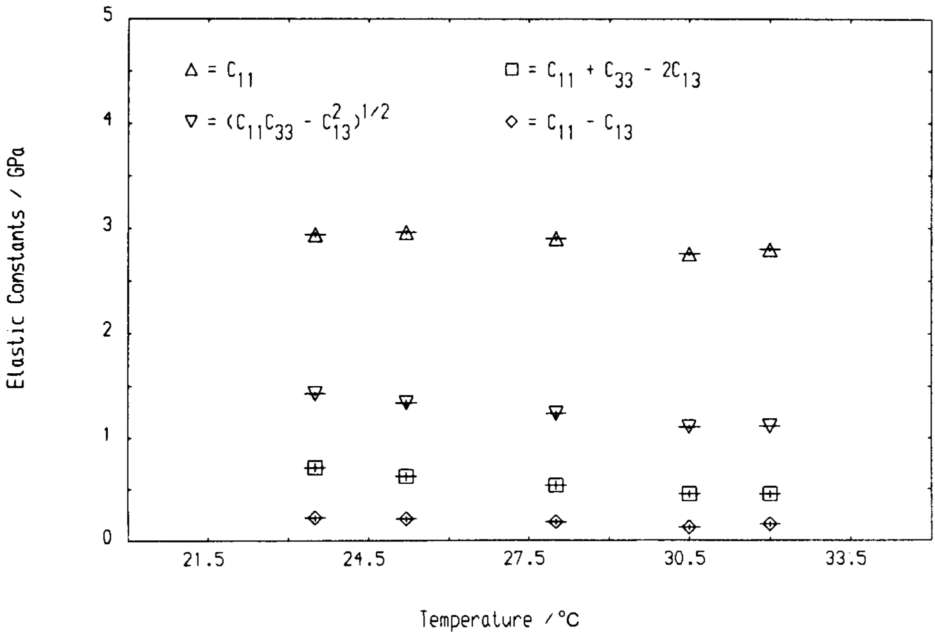

Figure 7. The temperature dependence of various combinations of bulk liquid crystal elastic constants plotted in the same form as in the paper by Bradberry and Vaughan [6].

23·5°C and the corresponding velocity against angle plot. The usefulness of this technique is illustrated in figures 4 and 5 which use fewer data points. Thus fewer spectra need to be accumulated for each temperature and hence the time required to obtain temperature dependent information is significantly reduced. Figure 6 shows the temperature dependence of the second sound velocity when the angle between the director and the scattering plane is 45°. Figure 7 shows the temperature dependence of various combinations of elastic constants, namely C_{11}, $(C_{11}C_{33} - C_{13}^2)^{1/2}$, $(C_{33} + C_{11} - 2C_{13})$ and $(C_{11} - C_{13})$. These are plotted in a similar form to figure 2 in the paper by Bradberry and Vaughan [6] except for $(C_{11}C_{33} - C_{13}^2)^{1/2}$.

6. Conclusions

The variation of the hypersound velocity in the smectic A phase with the angle between the director and the scattering plane can be explained by invoking the presence of a second phonon mode. We have presented a method by which a small number of data points can be used to determine the velocity of this mode and obtain a good fit to theory.

The longitudinal velocities in 8CB at 23·5 ± 0·3°C were found to be 1791·3 ± 4·5 ms^{-1} and 1715·0 ± 4·5 ms^{-1} for propagation parallel and perpendicular to the director. The errors quoted for the velocities indicate the accuracy of the data fitting. The data taken at 23·5°C and 30·5°C used a free spectral range of 13·82 GHz whilst the data taken at 25·2, 28·0 and 32·0°C used a free spectral range of 14·25 GHz. There is an uncertainty of 0·5% in the free spectral range. At 25·2 ± 0·3°C the values obtained for the elastic constants A, B and C are 2·962 ± 0·031 × 10^9 Nm^{-2}, 0·710 ± 0·066 × 10^9 Nm^{-2} and −0·221 ± 0·045 × 10^9 Nm^{-2}. These values are in good agreement with previous work [10] on 8CB which gave the values of A and $B + 2C$ as 2·92 ± 0·10 × 10^9 Nm^{-2} and 0·23 ± 0·04 × 10^9 Nm^{-2} at 25°C. The second sound velocity in 8CB reduces with temperature from 346·7 ± 7·1 ms^{-1} at 23·5°C but stays finite (≈ 275 ms^{-1}) as the temperature approaches the smectic A–nematic phase transition. This is in qualitative agreement with the theoretical predictions of Lui [11] who suggests the existence of a second propagating mode in the nematic phase due to phonons of higher frequency than those present in the smectic A phase.

The value of $V_2(45°)$ at 23·5°C and the temperature dependence of the elastic constants in figure 6 are similar to those obtained by Bradberry and Vaughan [6] and confirm that none of the combinations of these constants extrapolate to zero close to the smectic A–nematic transition temperature. However the values of some of the combinations of the elastic constants are slightly different. We believe that the data presented here are more reliable due to the procedures used to fit the data.

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