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Dielectric investigation of the diamagnetic anisotropy and elasticity of 4-*trans*-4′-*n*-hexyl-cyclohexyl-isothiocyanatobenzene (6CHBT)

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We have measured the dielectric constants of 6CHBT. The results from studies of various alignments and thicknesses measured under different electric and magnetic fields are presented. We discuss how the dielectric properties depend on boundary conditions, sample thickness and the magnitudes of electric and magnetic fields. Experimental results and discussion in the terms of continuum theory make it possible to compute the diamagnetic anisotropy ($\Delta\chi$), as well as the splay and bend elastic constants (K_{11}, K_{33}) of 6CHBT.

1. Introduction

Measurements of the K_{ii} elastic constants of nematic liquid crystals (NLCs) are generally based on the determination of critical values of electric (E_c) and magnetic (H_c) fields for different types of Fréedericksz transitions (FTs) [1–3]. Moreover $\Delta\chi$, which must be known to calculate some of values K_{ii}, is often obtained from the relation between E_c or H_c and proper magnetic (H) or electric (E) field strengths [4]. On the other hand we know that accurate values of E_c and H_c associated with FTs may only be obtained if the boundary angle Θ_B equals 0 or $\pi/2$ exactly and does not vary with increasing fields strength. Since, under laboratory conditions it is very difficult to obtain perfectly parallel and homeotropically oriented NLC cells and to satisfy simultaneously requirements of strong anchoring at them, many controversial results have been published on elastic constants. In this paper we try to present an attempt to determine the diamagnetic anisotropy $\Delta\chi$ and splay K_{11} and bend K_{33} elastic constants from dielectric measurements on samples with tilt angles ($\Theta_B \geqslant 0$ or $\Theta_B \leqslant \pi/2$) where the assumption of strong anchoring seems to be satisfactory.

2. Experimental

The investigated compound, 4-*trans*-4′-*n*-hexyl-cyclohexyl-isothiocyanatobenzene (6CHBT), has been synthesized and described earlier [5–7]. The perpendicular $\varepsilon_\perp = 4{\cdot}0$ and $\varepsilon_\parallel = 12{\cdot}0$ components of the permittivity tensor ε at 298 K were measured with a Tesla BM 484 bridge at 1592 Hz with a measuring voltage of 3 V. The sample was studied in a double plane capacitor with silver electrodes. The thicknesses of the liquid crystal layers were 2 mm. To orient the sample a magnetic field of 1T was used. Further dielectric measurements were made with the measuring cells and set-up previously described in [8]. Cells in the form of flat condensers were made from indium tin oxide (ITO) coated glass with an electrode area of 2 cm^2 and thickness varying from 30 to 120 μm. Boundary conditions were changed by coating the electrodes with 30 nm of different kinds of polyimides followed by suitable rubbing or by lecithin. The sample alignment was checked and identified by conoscopic observation and measurements

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[9]. The dielectric constants ε at $298{\cdot}0 \pm 0{\cdot}2$ K were measured by a Precision Component Analyser WAYNE KERR 6425 with a measuring voltage from 10 mV to 5 V and a frequency of 1500 Hz. The magnetic induction was changed from 0·0 to 1·1 T.

3. Theory

Consider a nematic layer of thickness d between planes (electrodes) $z=0$ and $z=d$ of a cartesian coordinate system. The experimental geometry of this system is shown in figure 1. An external measuring field E is applied to the LC medium along the z axis. An external magnetic field H lies in the xz plane and forms an angle ψ with the z axis. The director $\mathbf{n}$ and x axis at position z is denoted by $\Theta(z)$. In the absence of magnetic and electric fields, the director everywhere forms the same angle of $\Theta(z)=\Theta_B$ with the x axis.

On the basis of the Oseen and Frank elastic continuum theory of a nematic phase in the presence of a magnetic (H) and electric (E) field, in the limit of small distortions, the balance of elastic, magnetic and elastic torques per unit volume requires [10]

$$[K_{11}\cos^2\Theta(z)+K_{33}\sin^2\Theta(z)]\frac{d^2\Theta(z)}{dz^2}=\Delta\varepsilon\varepsilon_0 E^2(z)\sin\Theta(z)\cos\Theta(z) + \Delta\chi\mu_0 H^2\sin[\Theta(z)+\Psi]\cos[\Theta(z)+\Psi], \quad (1)$$

where K_{11}, K_{33}, $\Delta\varepsilon$ and $\Delta\chi$ are the LC elastic, splay and bend constants and dielectric and diamagnetic anisotropies of the LC medium, respectively. Here ε_0 is the electric permittivity and μ_0 is the magnetic permeability of free space.

Due to the dielectric and diamagnetic anisotropies connected with local electric, $E(z)$, and magnetic, H, fields, vector fields of directors $\mathbf{n}(\Theta(z))$ are formed in the LC medium. We must notice that distortion of NLC layers by combinations of electric and magnetic fields is, in general, not uniform across the sample. In our case, when we apply a voltage U to the slab, the electric field $E(z)$ has a component only in the z direction.

If the LC-substrate anchoring force is the same for both electrodes (both electrodes have been treated in the same way), the solution of equation (1) is symmetric with respect to the $z=d/2$ plane. The angle Θ is a function of coordinate z, $\Theta(z)$ assuming the maximum value Θ_m at $z=d/2$ and the value of Θ_B at the boundaries. After multiplying

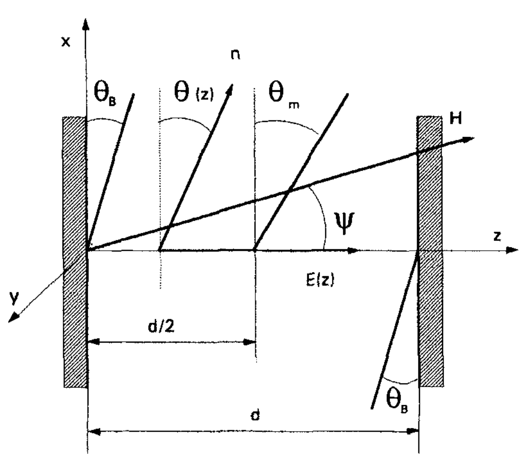

Figure 1. Schematic diagram of the system geometry.

equation (1) by $d\Theta/dz$ and then integrating it and knowing that for $z=d/2$, $\Theta(z)=\Theta_m$ and $d\Theta/dz=0$ we obtain

$$\frac{d\Theta(z)}{dz}=\left\{2\left[\int_{\Theta_m}^{\Theta}\frac{\Delta\varepsilon\varepsilon_0 E^2\sin\Theta'\cos\Theta'}{K_{11}\cos^2\Theta'+K_{33}\sin^2\Theta'}d\Theta' + \int_{\Theta_m}^{\Theta}\frac{\Delta\chi\mu_0 H^2\sin^2(\Theta'+\Psi)\cos^2(\Theta'+\Psi)}{K_{11}\cos^2\Theta'+K_{33}\sin^2\Theta'}d\Theta'\right]\right\}^{1/2}. \tag{2}$$

For the dielectric displacement vector

$$\mathbf{D}(z)=\varepsilon(z)\varepsilon_0\mathbf{E}(z), \tag{3}$$

where

$$\varepsilon(z)=\varepsilon_\perp+\Delta\varepsilon\sin^2\Theta(z). \tag{4}$$

We have the equation

$$\operatorname{div}\mathbf{D}=0 \tag{5}$$

which requires the z component D_z of $\mathbf{D}$ to be constant.

Applying a measured voltage U to the cell filled with LC material characterized by K_{11}, K_{33}, $\Delta\varepsilon$, $\varepsilon_\perp$, and $\Delta\chi$, initially formed into a layer with d and Θ_B, under a magnetic field described by H and Ψ, and the measured dielectric constant ε^Ψ can be written as

$$\varepsilon^\Psi(U,d,H,\Theta_B)=\frac{d}{\displaystyle\int_{\Theta_B}^{\Theta_m}\left[\varepsilon_\perp+\Delta\varepsilon\sin^2\Theta(z)\right]^{-1}\left[\frac{d\Theta(z)}{dz}\right]^{-1}d\Theta}, \tag{6}$$

where $d\Theta(z)/dz$ is given by equation (2), and now the required function of $E(z)$ may be calculated from equation (5) which becomes

$$\varepsilon^\Psi(U,d,H,\Theta_B)\frac{U}{d}=\{\varepsilon_\perp+\Delta\varepsilon\sin^2\Theta(z)\}E(z). \tag{7}$$

Equation (6) supplemented by equation (7) can only be solved numerically.

The typical solutions of this problem for a given material with $K_{11}=K_{33}=8{\cdot}0\times10^{-12}\,\mathrm{N}$, $\varepsilon_\perp=4{\cdot}0$, $\Delta\varepsilon=8{\cdot}0$, $\Delta\chi=44{\cdot}0\times10^{-8}$ in the planar configuration (PC) in a magnetic field B and for $\Psi=\pi/2$ are plotted in figures 2 and 3. The dashed lines represent an untilted structure with $\Theta_B=0{\cdot}00$ and dotted lines are for tilted ones with $\Theta_B=0{\cdot}03$.

If we assume that in the limit of a small deformation near the critical value U, $E^2(z)$ is nearly constant and equals $\langle E^2\rangle$, where

$$\langle E^2\rangle=\frac{1}{d}\int_0^d E(z)^2\,dz, \tag{8}$$

we will obtain results corresponding to points within regions between dashed and solid lines or dotted and solid ones. Solid lines have been plotted with an assumption that the electric field within the sample is uniform (the case with $\langle E^2\rangle=U^2/d^2$). Taking into consideration equation (8), equation (2) after some mathematical treatment takes the form of

$$\frac{d\Theta(z)}{dz}=\left[\frac{A}{K_{11}(K-1)}\ln\frac{\cos^2\Theta+K\sin^2\Theta}{\cos^2\Theta_m+K\sin^2\Theta_m}\right]^{1/2}, \tag{9}$$

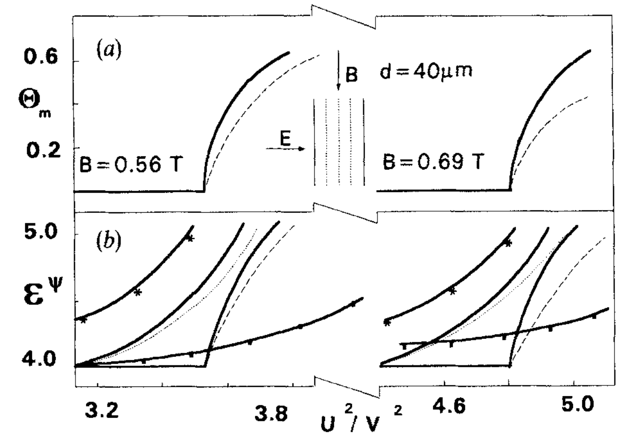

Figure 2. (a) Maximum deformation angle Θ_m (in the middle of the layer) versus U^2, calculated from equations (6) and (7). (b) Values of ε^{Ψ} dielectric constants versus U^2 calculated from equations (6) and (7). In both cases, a PC (planar configuration) cell is placed in the magnetic field. The angle between **H** and **E** vectors is $\Psi=\pi/2$ ($\varepsilon^{\Psi}=\varepsilon^{\perp}$ for $\Psi=\pi/2$), The cell parameters are as follows: $d=40\,\mu$m, $K_{11}=K_{33}=8\times10^{-12}$ N, $\varepsilon_{\perp}=4{\cdot}0$, $\Delta\varepsilon=8{\cdot}0$, $\Delta\chi=44\times10^{-8}$; (- - -) $\Theta_B=0{\cdot}00$, (. . .) $\Theta_B=0{\cdot}03$, (∗∗∗) $\Theta_B=0{\cdot}09$. (◆ ◆ ◆) Experimental values of the corresponding ε^{Ψ} constants for 6CHBT (see figure 4) have been plotted for comparison. The meaning of the solid lines is explained in the text.

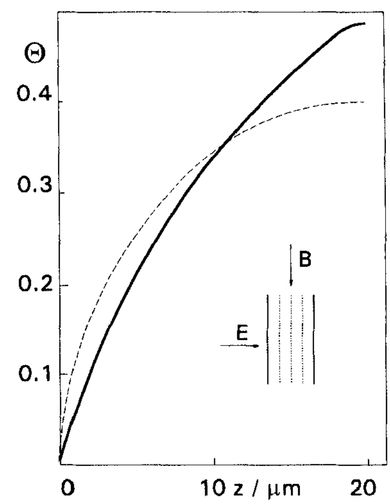

Figure 3. Plots of calculated distribution functions $\Theta(z)$ of a PC sample. $d=40\,\mu$m, $K_{11}=K_{33}=8\times10^{-12}$ N, $\varepsilon_{\perp}=4{\cdot}0$, $\Delta\chi=44\times10^{-8}$; (- - -) $\Theta_B=0{\cdot}000$, $B=0{\cdot}69$ T, $U=2{\cdot}25$ V with $E=E(z)$; (——) $\Theta_B=0{\cdot}00$, $B=0{\cdot}69$ T, $U=2{\cdot}23$ V with $E=U/d$ (the solid line has been plotted with an assumption that the electric field within the sample is uniform). In both cases the calculated ε is the same ($\varepsilon^{\Psi}=\varepsilon^{\perp}=4{\cdot}8$).

where

$$K = K_{33}/K_{11}$$
$$A = \Delta\varepsilon\varepsilon_0\langle E^2\rangle + \Delta\chi\mu_0 H^2 \quad \text{for} \quad \Psi = 0,$$

and

$$A = \Delta\varepsilon\varepsilon_0\langle E^2\rangle - \Delta\chi\mu_0 H^2 \quad \text{for} \quad \Psi = \pi/2.$$

In this situation, for $\Psi = \pi/2$, equation (6) takes the form of equation (10) ($\varepsilon^{\Psi} = \varepsilon^{\perp}$)

$$\varepsilon^{\perp}(\langle E^2\rangle, d, H, \Theta_B) = (\Delta\varepsilon\varepsilon_0\langle E^2\rangle - \Delta\chi\mu_0 H^2)^{1/2} d\left\{\int_{\Theta_B}^{\Theta_m} \frac{1}{\varepsilon_\perp + \Delta\varepsilon\sin^2\Theta}\left[K_{11}(K-1) \times \left(\ln\frac{\cos^2\Theta + \sin^2\Theta}{\cos^2\Theta_m + K\sin^2\Theta_m}\right)^{-1}\right]^{1/2} d\Theta\right\}^{-1}. \tag{10}$$

It is worth pointing out that if we deform a slab with homeotropic or planar orientation by crossed ($E \perp B$) electric and magnetic fields, the critical solutions of equations (9) ($d\Theta/dz > 0$) and (10) ($\varepsilon^{\perp} > \varepsilon_\perp$) appear only when $\Theta_B = \pi/2$ or $\Theta_B = 0$, respectively.

When we regard that, near critical conditions, the distribution function $\Theta(z)$ may be approximated by

$$\Theta(z) = \Theta_m \sin\left(\frac{\pi}{d} z\right) \tag{11}$$

for a planar configuration (PC), and by

$$\Theta(z) = \frac{\pi}{2} - \Theta_m \sin\left(\frac{\pi}{d} z\right) \tag{12}$$

for a homeotropic one (HC), equations (9) and (10) transform smoothly into well-known formulae given in [1, 2, 4, 10–12].

4. Results and discussion

We have measured the dielectric characteristics of 6CHBT at 298 K for PC and HC, for different Θ_B, d, H, U and Ψ values. The typical dielectric characteristics of PC with $d = 40\,\mu$m and $\Theta_B = 0{\cdot}02$ and for HC with $d = 95\,\mu$m and $\Theta_B = \pi/2$ are presented in figures 4 and 5, respectively.

Using the obtained relations, the plots of U^2 versus B^2 for PC and HC cells with different d and Θ_B values were drawn (see figure 6). It can be seen from equation (10) and numerical analyses of equations (6) and (7) that to keep the value of $\varepsilon^{\perp}$ constant in a given cell with a certain configuration described by d and Θ_B for different U and H values the A factor must be constant

$$\Delta\varepsilon\varepsilon_0 E^2 - \Delta\chi\mu_0 H^2 = \text{constant}. \tag{13}$$

If $E(z)$ did not change with z (as is the case only at critical points where $\varepsilon^{\perp} = \varepsilon_\perp$ or $\varepsilon^{\perp} = \varepsilon_\parallel$) the curves in figure 6 (*a*) and (*b*) should be straight lines and parallel to each other. When $\varepsilon^{\perp}$ approaches $\varepsilon_\perp$ for PC or $\varepsilon_\parallel$ for HC, the differences between $E(z)^2$ and $\langle E^2\rangle$ decrease (see the gap between the solid and dashed or dotted lines in figure 2) and finally disappear for any Θ_B.

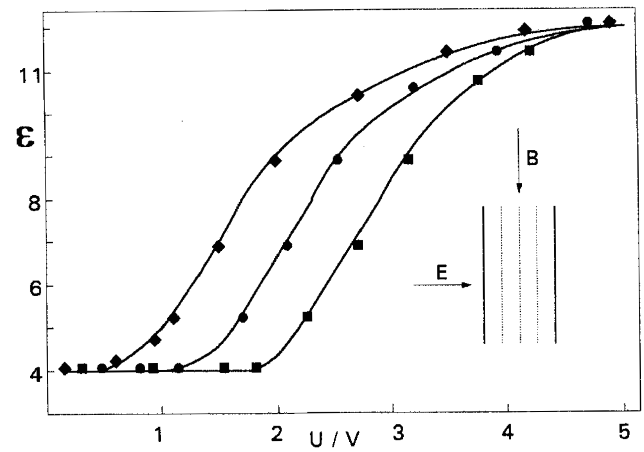

Figure 4. The dielectric characteristics of 6CHBT for a PC with $d = 40\,\mu m$ and $\Theta_B = 0{\cdot}02$ at 298 K; (◆) $B = 0{\cdot}00$ T, (●) $B = 0{\cdot}45$ T, (■) $B = 0{\cdot}69$ T.

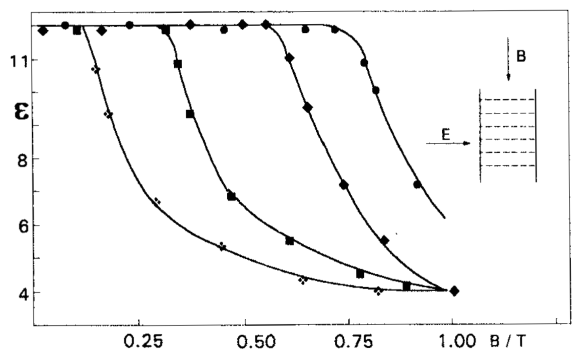

Figure 5. The dielectric characteristics of 6CHBT for a HC with $d = 95\,\mu m$ and $\Theta_B = \pi/2$ at 298 K; (◇) $U = 0{\cdot}0$ V, (■) $U = 2{\cdot}0$ V, (◆) $U = 4{\cdot}0$ V, (●) $U = 5{\cdot}0$ V.

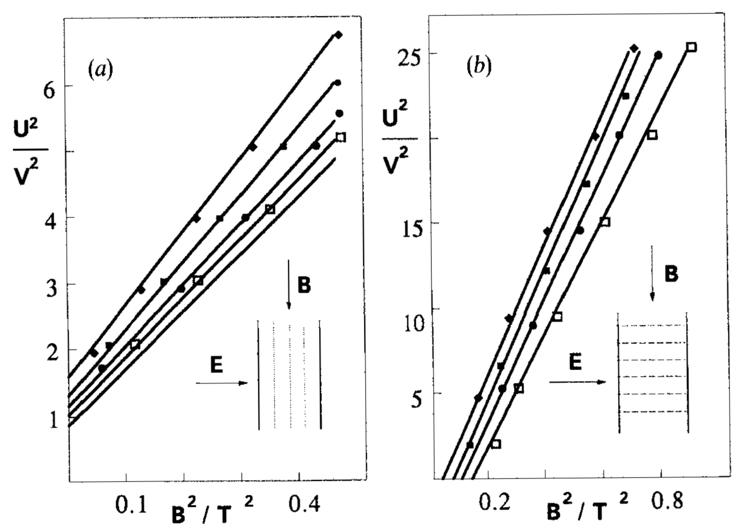

Figure 6. (a) U^2 as a function of B^2 of 6CHBT for a PC with $d = 40\,\mu m$ and $\Theta_B = 0{\cdot}02$ at $T = 298$ K; (◆) $\varepsilon_\perp = 5{\cdot}75$, (■) $\varepsilon_\perp = 5{\cdot}00$, (●) $\varepsilon_\perp = 4{\cdot}75$, (□) $\varepsilon_\perp = 4{\cdot}50$, (——) $\varepsilon_\perp = 4{\cdot}25$. (b) U^2 as a function of B^2 of 6CHBT for a HC with $d = 95\,\mu m$ and $\Theta_B = \pi/2$ at $T = 298$ K; (◆) $\varepsilon_\perp = 11{\cdot}6$, (■) $\varepsilon_\perp = 11{\cdot}3$, (●) $\varepsilon_\perp = 10{\cdot}4$, (□) $\varepsilon_\perp = 9{\cdot}5$.

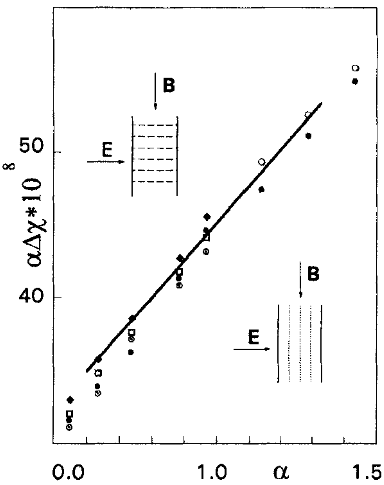

Figure 7. $\Delta\chi$ as a function of α. As for a PC: $\alpha=\varepsilon^{\perp 2}/\varepsilon_{\perp}^{2}$; (○) $d=40\,\mu$m, $\Theta_B=0{\cdot}02$; (●) $d=95\,\mu$m, $\Theta_B=0{\cdot}00$. As for a HC: $\alpha=\varepsilon^{\perp 2}/\varepsilon_{\parallel}^{2}$; (◆) $d=36\,\mu$m, $\Theta_B=\pi/2$; (□) $d=43\,\mu$m, $\Theta_B=\pi/2-0{\cdot}02$; (●) $d=95\,\mu$m, $\Theta_B=\pi/2$; (⊙) $d=108\,\mu$m, $\Theta_B=\pi/2-0{\cdot}03$.

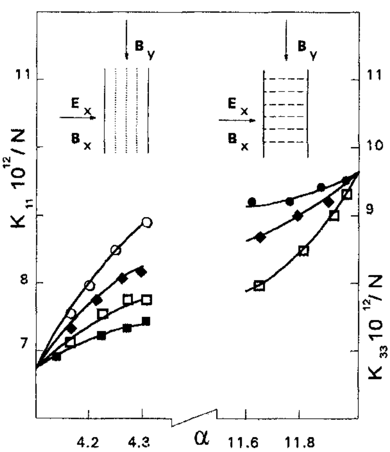

Figure 8. Plots of calculated K_{11} and K_{33} constants of 6CHBT at 298 K. For a PC, with $d=40\,\mu$m and $\Theta_B=0{\cdot}02$; (■) $E_x=0$, $B_x>0$, $B_y=0$, (◆) $E_x>0$, $B_x=0$, $B_y=0{\cdot}31$ T, (□) $E_x>0$, $B_x=0$, $B_y=0$, (○) $E_x>0$, $B_x=0$, $B_y=0{\cdot}56$ T. For a HC with $d=36\,\mu$m and $\Theta_B=\pi/2$; (●) $E_x=0$, $B_y>0$, (◆) $E_x=1$ V/36 μm, $B_y>0$, (□) $E_x=2$ V/36 μm, $B_y>0$.

Assuming that, for small deformation ($\varepsilon^{\perp}\approx\varepsilon_{\perp}$ or $\varepsilon^{\perp}\approx\varepsilon_{\parallel}$) the dielectric displacement D in the sample changes insignificantly when we go from $\varepsilon_{\perp}$ or $\varepsilon_{\parallel}$ to $\varepsilon^{\perp}$, equation (13) then takes the form of

$$U^2-\frac{\Delta\chi d}{\Delta\varepsilon\varepsilon_0\mu_0}\alpha B^2=\text{constant},\tag{14}$$

where $\alpha=\varepsilon^{\perp 2}/\varepsilon_{\perp}^{2}$ for HC and $\alpha=\varepsilon^{\perp 2}/\varepsilon_{\parallel}^{2}$ for PC.

As we can see from equation (14) $\Delta\chi$ can be determined from the slopes of the curves of U^2 versus B^2 obtained for any configuration with any Θ_B. The results of the calculations of diamagnetic anisotropy $\Delta\chi$ of 6CHBT are listed in figure 7.

As we see, the results of $\Delta\chi$ obtained both for PC and HC approach the same values of $\Delta\chi = 44 \times 10^{-8}$ when $\alpha = 1{\cdot}0$.

Having known $\Delta\chi$, K_{11} and K_{33} can be computed using procedures based on equations (2), (6) and (7). Figure 8 shows typical results of such computer calculations.

Using the previously described method, after averaging over all samples, splay K_{11}, and bend K_{33} elastic constants were estimated as $7{\cdot}5 \times 10^{-12}$ N and $9{\cdot}5 \times 10^{-12}$ N, respectively. These results are in good agreement with [7].

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