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Electro-optical and dielectric characterizations of the Goldstone mode relaxation in ferroelectric chiral smectic C liquid crystals

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Abstract

We report structural (helical pitch), electro-optical (tilt angle and spontaneous polarization) and dielectric (Goldstone mode) investigations of ferroelectric liquid crystals (FLCs) exhibiting the chiral smectic C phase (SmC*). All these characterizations were performed on two pure FLCs showing the SmC*–SmA–N* phase sequence and having small pitch, high spontaneous polarization and a large relaxation frequency. We have determined the Goldstone rotational viscosity and the twist elastic constant in the SmC* phase from the helical pitch, tilt angle, polarization, dielectric strength experimental data and from the relaxation frequency of the Goldstone-mode relaxation. An Arrhenius-type behaviour of the Goldstone rotational viscosity was obtained and the corresponding activation energies were evaluated.

1. Introduction

The study of ferroelectric liquid crystal (FLC) materials exhibiting the chiral smectic C (SmC*) phase has attracted increasing interest in recent years. Therefore, many investigations

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Table 1. Chemical structure, phase sequences and transition temperatures ($^{\circ}\text{C}$) for the homologous biphenyl benzoate series. (Note: Cr = crystalline phase; Sm = smectic phases A, C*; N* = cholesteric phase; BP = blue phase; I = isotropic phase; () = monotropic transition.)

n	Cr	Sm	SmC*	SmA	N*	BP	I
7	●	100 ● (52)	●	134	—	●	166 ●
8	●	88 —	●	138	—	●	165.5 ●
9	●	89 —	●	142	—	●	162 ●
10	●	88 —	●	143 ●	144 ●	●	159 ●
11	●	88 —	●	146 ●	149 ●	●	157 ●
12	●	81 —	●	145.5 ●	150 ●	●	154.5 ●

into the synthesis and the experimental and theoretical aspects of FLC have been published. These different studies were carried out in an effort to understand the ferroelectric properties of the SmC* phase, thus enabling the development of electro-optical devices. Taking into account the reduced symmetry in SmC*, Meyer has shown that the FLC SmC* [1] possesses a ferroelectric polarization. The development of a spontaneous helical structure due to the chirality of the molecules causes the director to precess around the tilt cone so that the macroscopic polarization of the sample vanishes to zero. Nevertheless, the helical structure can be unwound by the surface effects if the samples are sufficiently thin. This leads to surface stabilized ferroelectric liquid crystal (SSFLC) reported for the first time in 1980 by Clark and Lagerwall [2].

The periodic structure can also be deformed by applying an external electric field to the samples parallel to the smectic layers (perpendicular to the helical axis). Blinc has shown that the dielectric relaxation response is linked to four dielectric relaxations [3]. Two of these modes are related to fluctuations of the polarization and are shown as a 'polarization mode' [4]. The other two modes [3, 5] are connected with the director's reorientational motions and have relaxation frequencies lower than those observed for the polarization modes. The first of these modes is attributed to the Goldstone mode, which corresponds to the phase fluctuations in the azimuthal orientation of the tilt direction. This dielectric process appears in the SmC* phase with a low relaxation frequency and a high amplitude. The second contribution appears to be due to the fluctuations in the tilt angle amplitude and is the so-called soft mode [3, 6]. The dielectric strength and relaxation frequency in the SmC* phase are available by using a simple Landau theory [7]. Indeed, the expression for this dielectric response shows the relationship between the different parameters, namely helical pitch, tilt angle, spontaneous polarization, dielectric amplitude and critical frequency.

In previous investigations [8–10], we studied C8, C10 and C11 compounds of the homologous biphenyl alkyloxy benzoates series gathered in table 1. In this work we extend the previous investigation to the study of the pure chiral C12 compound, which displays the SmC*–SmA–N* phase sequence near the suitable SmC*–SmA–N* multicritical point [8, 9]. We measured the helical pitch in the SmC* and N* phases by using a structural study. The electro-optical properties of the SmC* phase have been studied, including tilt angle and spontaneous polarization. To determine some characteristic parameters in the SmC* phase, we focused our dielectric investigation on the contribution of the Goldstone mode. From the dielectric analysis spectra, we report the dielectric response in the SmC* phase via the dielectric amplitudes and their relaxation frequencies. From a combination of the theoretical model and the experimental

data, we determined the Goldstone-mode rotational viscosity and the twist elastic constant. Finally, Arrhenius-type behaviours of the rotational viscosity were obtained and the activation energies determined. Both the dielectric measurements of the soft mode (without and with bias) in the SmC* and SmA phases, and the electroclinic effect in the N* phase near a SmC*–SmA–N* multicritical point (for C12 material) were reported in our recent paper [11]. All the results obtained for C12 compound are compared with those previously found for C11 [12] of the same homologous series.

2. Experimental method and procedures

Ferroelectric properties were studied for the C11 ($n = 11$) and C12 ($n = 12$) compounds of the homologous biphenyl alkyloxy benzoate series. The synthesis procedures for this homologous series (C7–C12) have already been published [9]. The phase identifications and the transition temperatures at atmospheric pressure were determined by both thermal microscopy (Mettler FP 5) and differential scanning calorimetry (Perkin-Elmer DSC 7).

The helical pitch was measured by the Grandjean–Cano method [13, 14] using the defect lines. Prismatic-shaped cells with a wedge angle of less than 0.5° were used. In the N* phase, a prismatic planar anchoring of the samples was obtained. In this configuration, the molecules are parallel and the helical axis is perpendicular to the glass surface. In order to measure the helical pitch in the SmC* phase, ‘pseudo-homeotropic’ samples were prepared, which led to an orientation of both the molecular long axis and the helical axis perpendicular to the glass plates. The orientations of the Grandjean–Cano lines and optical properties of the sample were observed by means of a polarized optical microscope in the transmitted mode. During these optical studies, the FLC cells were then placed in a heating stage for temperature control. With this method we obtained an excellent orientation formed with regular Grandjean–Cano steps in the N* and SmC* phases, allowing measurements of helical pitch.

The tilt angle θ and the spontaneous polarization P_S , were measured as a function of temperature. For these studies, surface stabilized ferroelectric liquid crystal switching (SSFLC) planar geometry [2] was used. The sample was aligned in the SSFLC bookshelf with a thickness of about $3 \mu\text{m}$. In order to measure the tilt angle temperature variations, the linear regime of light detection between crossed polarizers was used. A high-amplitude and low-frequency electric field ($E = 5 \text{ V } \mu\text{m}^{-1}$; $F = 0.2 \text{ Hz}$) was applied to the samples. The tilt angle was measured from the value of the microscopic stage rotation between extinction states, which can be obtained by reversing the applied electric field ($+E/-E$). To measure P_S , the well-known reverse current technique with a triangular wave electric field was used [15]. The applied voltage to saturate the polarization is around 18 V and its frequency is about 1 kHz.

The dielectric measurements were made with a planar orientation of the sample: smectic layers are perpendicular to the electrodes in the frequency range of 5 Hz–1 MHz, using a previously described experimental procedure [16]. The electric field was applied perpendicular to the SmC* helical axis and parallel to the smectic layers (figure 1). Planar orientation of the sample was achieved with a polyvinyl alcohol (PVA) coating and rubbing. In order to obtain a good alignment, the cell was slowly filled by capillary action from the isotropic phase into the SmC* phase, under an applied electric field. The cell thickness of $30 \mu\text{m}$ was chosen to be much higher than the pitch value to obtain a planar-wound geometry in the SmC* phase. Checking of the sample orientation and the transition temperatures was performed using a polarizing microscope equipped with a heating stage. These measurements were made without adding a DC bias to the measurement electric field. The complex dielectric constant $\varepsilon^*(f, T)$ for a distribution of the Cole–Cole type may be written as:

$$\varepsilon^* - \varepsilon(\infty) = \frac{\Delta\varepsilon_G}{1 + (jf/f_G)^{1-\alpha_G}} + \frac{\Delta\varepsilon_S}{1 + (jf/f_S)^{1-\alpha_S}}. \quad (1)$$

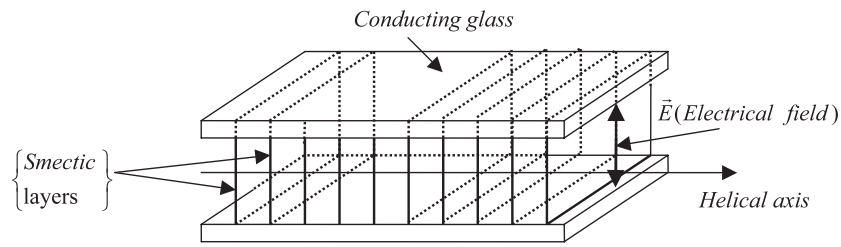


Figure 1. Schematic drawing of the cell with smectic layers, helical axis and applied electrical field.

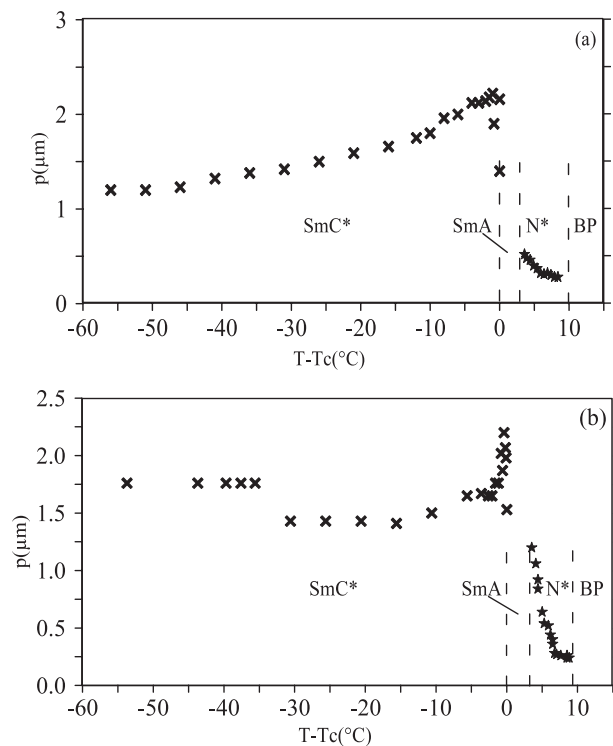


Figure 2. Pitch temperature dependence in the SmC* and N* phases for C11 (a) and C12 (b).

In this expression, $\varepsilon(\infty)$ represents the limit of dielectric permittivity at high frequencies, f_G and f_S are the Goldstone and soft mode relaxation frequencies, α_i is the corresponding distribution parameter and $\Delta\varepsilon_i$ is the dielectric strength of the domain i ($i =$ Goldstone or soft mode).

3. Results and discussion

3.1. Structural and electro-optical studies

Figure 2 shows the temperature dependence of the pitch in the N* and SmC* phases. In the N* phase, this pitch is very short ($p \sim 0.25 \mu\text{m}$ for C11 and C12) near the N*-BP. In this

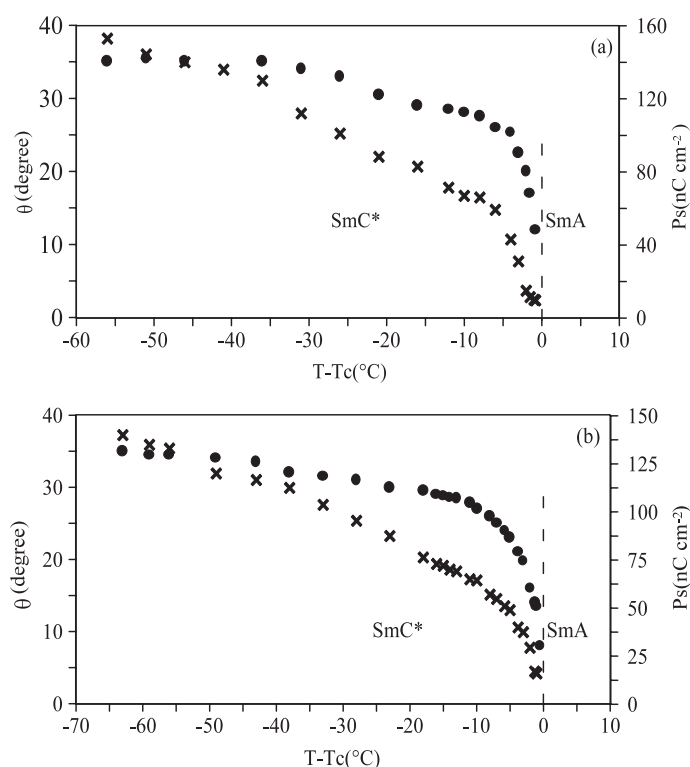


Figure 3. Temperature dependences of tilt angle (●) and spontaneous polarization (x) for C11 (a) and C12 (b).

region, the selective reflection of the wavelength range of visible light was observed for both materials. Near the $\text{N}^* - \text{SmA}$ transition, the N^* pitch increases regularly and diverges at the $\text{N}^* - \text{SmA}$ transition temperature. In the SmC^* phase, for C12, we have obtained two temperature ranges where the helical pitch is very small and constant: $p \sim 1.75 \mu\text{m}$ in the temperature range $35.6 \leq T_C - T \leq 53.7$ $^{\circ}\text{C}$ and $p \sim 1.4 \mu\text{m}$ for $15.6 \leq T_C - T \leq 30.6$ $^{\circ}\text{C}$, where T_C is the $\text{SmC}^* - \text{SmA}$ transition temperature. When approaching the SmA phase, the pitch increases up to $2.2 \mu\text{m}$ for C11 and C12 compounds. Close to T_C , the Grandjean–Cano defects may become invisible because both the rotatory power and the tilt angle nearly vanish. The ‘flat drop’ method [17] has been used and gives the limit value of the pitch at T_C . We have estimated this value as $1.5 \mu\text{m}$ for both compounds (see figures 2(a) and (b)).

In figure 3 we present the temperature dependence of the tilt angle and the spontaneous polarization, respectively, for the C11 (a) and C12 (b) materials. At low temperatures, a saturated tilt angle of 35° was observed. When the temperature is increased, this angle decreases continuously up to T_C , where it is almost equal to 12° and 8° (for C11 and C12, respectively). This is due to the coupling between the applied electric field and the electroclinic effect. The resulting temperature dependences for the spontaneous polarization are also shown, indicating that our compounds display high values of P_s . At a reduced temperature, $T_C - T = 50$ $^{\circ}\text{C}$, values of $P_s = 160$, 150 and 140 nC cm^{-2} were found for C10 [9], C11 and C12, respectively. For these materials, the alkyl chain length was changed, and an increasing polarization was measured as the chain length shortened. Hence, this may be quite understood by the fact that the C12 compound and its longer alkyloxy chain hinders rotational motion

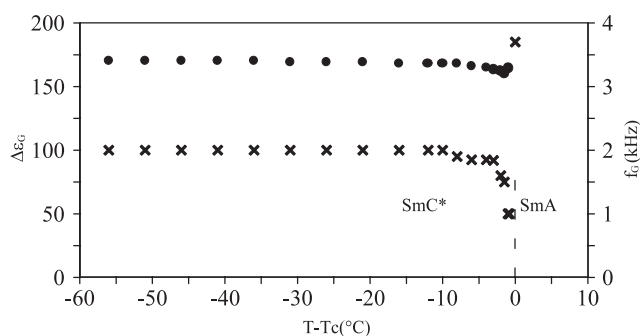


Figure 4. Temperature dependences of the dielectric strength (●) and relaxation frequency (x) of the Goldstone mode in the SmC* phase for C11.

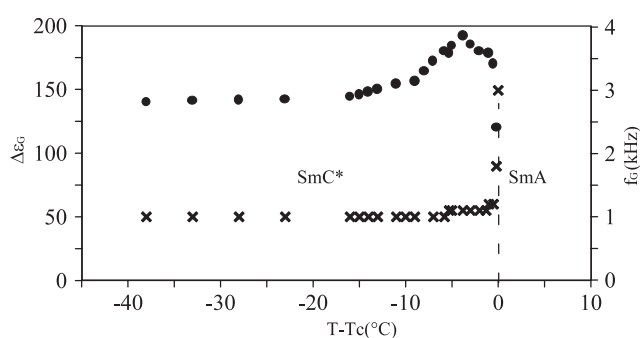


Figure 5. Temperature dependences of the dielectric strength (●) and relaxation frequency (x) of the Goldstone mode in the SmC* phase for C12.

of the molecules [18] which is not the case for C10 and C11 materials. In other words, this means that this fact contributes to decreasing the amplitude of spontaneous polarization. We can also notice that for these series the chiral alkyloxy chain is linked to the biphenyl core by the ester group $-\text{COO}-$. The presence of a halogen substituent (Cl) in the vicinity of the ester group $-\text{COO}-$ and the biphenyl core favours high values of spontaneous polarization for our compounds.

3.2. Dielectric study

Dielectric relaxation spectroscopy in the SmC* phase shows a single relaxation domain. This can be eliminated by unwinding the helicoidal structure under DC bias [19] and is connected to the classical Goldstone mode. The variations in dielectric amplitude $\Delta\varepsilon_G$ and critical relaxation frequency f_G versus temperatures for C11 and C12 are given in figures 4 and 5, respectively. At lower temperatures and far from T_C , $\Delta\varepsilon_G$ and f_G for the C12 material are rather temperature independent as the helical pitch and have relatively high values: $\Delta\varepsilon_G = 140$ and $f_G = 1$ kHz, compared to those observed for other compounds [20, 21]. With increasing temperature, $\Delta\varepsilon_G$ increases slowly to its maximum value $\Delta\varepsilon_G = 190$, which occurs slightly below T_C . At $T = T_C$, the dielectric strength decreases to reach $\Delta\varepsilon_G = 120$, whereas the critical frequency increases and diverges to a high value of $f_G = 3$ kHz. Such behaviour of $\Delta\varepsilon_G$ and f_G is due to the soft-mode contribution, which becomes predominant near T_C .

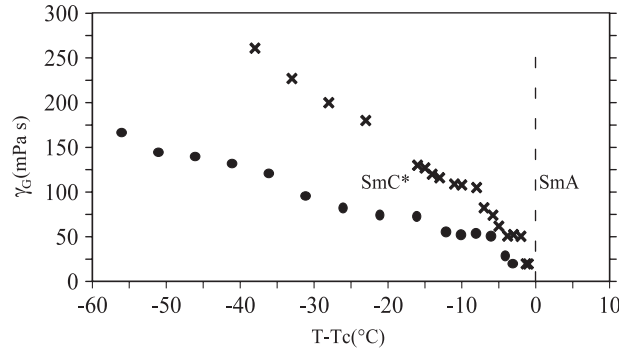


Figure 6. Experimentally determined temperature dependence of the Goldstone rotational viscosity γ_G in the SmC* phase for C11 (●) and C12 (x).

From structural, electro-optical and dielectric data, some characteristic parameters for the Goldstone-mode relaxation can be determined using the predictions of the generalized Landau models [7] around the SmC*–SmA transition. The Goldstone-mode dielectric strength and the relaxation frequency in the SmC* phase are given by

$$\varepsilon_0 \Delta \varepsilon_G = \frac{1}{2K_{33}q^2} \left(\frac{P_S}{\theta} \right)^2 \quad (2)$$

$$f_G = \frac{K_{33}q^2}{2\pi\gamma_G}, \quad (3)$$

where P_S is the spontaneous polarization, θ is the tilt angle, $q = 2\pi/p$ is the wavevector of the modulated SmC* phase, while K_{33} and γ_G are, respectively, the twist elastic constant and the Goldstone rotational viscosity; ε_0 is the free space permittivity. Thus we can see from equation (2) that the Goldstone-mode dielectric strength $\Delta \varepsilon_G$ is closely related to the helical pitch ($q = 2\pi/p$). Close to the SmC*–SmA transition, the helical pitch (for C12) varies swiftly from about $p = 2.2 \mu\text{m}$ at $T_C - T = 0.5 \text{ }^\circ\text{C}$ to $1.53 \mu\text{m}$ at $T \cong T_C$ (see figure 2); so the quick decrease of $\Delta \varepsilon_G$: $\Delta \varepsilon_G(T = T_C)/\Delta \varepsilon_G(T_C - T = 0.5 \text{ }^\circ\text{C}) \cong 0.65$ can be easily understood by the decreases of p : $p^2(T = T_C)/p^2(T_C - T = 0.5 \text{ }^\circ\text{C}) \cong 0.5$.

From the two equations, we can derive the following expression:

$$\gamma_G = \frac{1}{4\pi\varepsilon_0} \left(\frac{1}{\Delta \varepsilon_G f_G} \right) \left(\frac{P_S}{\theta} \right)^2. \quad (4)$$

From this equation and from the experimental values of P_S , θ , $\Delta \varepsilon_G$ and f_G , γ_G can be evaluated in the SmC* phase. The temperature dependences of this parameter thus obtained for C12 and C11 are reported in figure 6. As we can see from expression (4), the viscosity is proportional to the reverse of the product of dielectric strength and relaxation frequency $1/(\Delta \varepsilon_G \times f_G)$. The temperature evolution of γ_G does not seem to be solely governed by $1/(\Delta \varepsilon_G \times f_G)$ but also by the ratio P_S/θ , which rather varies with temperature, especially when getting close to T_C . Far from the SmC*–SmA phase transition, the experimental values of γ_G can be compared for C11 and C12 materials. The experimental values of p , P_S , θ , $\Delta \varepsilon_G$ and f_G for these compounds in the SmC* phase, measured at $T_C - T = 40 \text{ }^\circ\text{C}$ are listed in table 2.

At $T_C - T = 40 \text{ }^\circ\text{C}$ we obtained, respectively, for C12 and C11: $\gamma_G = 260$ and 131 mPa s ; therefore, γ_G for C12 is twice the value found for C11: $\gamma_G(\text{C12}) = 2\gamma_G(\text{C11})$. This result for γ_G can be explained by the values of the dielectric strength and relaxation frequency

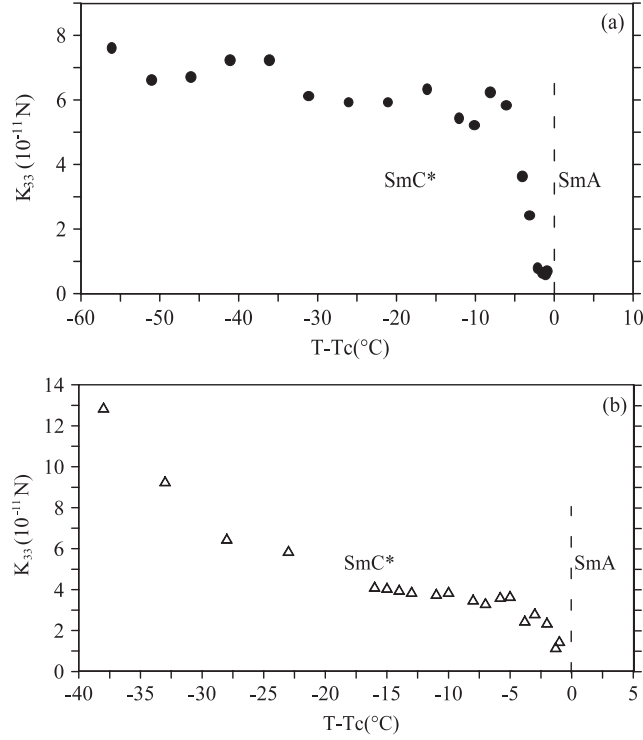


Figure 7. Experimentally determined temperature dependence of the twist elastic constant K_{33} in the SmC* phase for C11 (a) and C12 (b).

Table 2. Experimental values of helical pitch, polarization, tilt angle, dielectric strength and relaxation frequency in the SmC* phase for C11 and C12 at $T_c - T = 40^{\circ}\text{C}$.

Compound	p (μm)	P_S (nC cm^{-2})	θ (deg)	$\Delta\varepsilon_G$	f_G (kHz)
C11	1.31	135	35	170	2
C12	1.75	115	32	150	1

(see table 2), which are closely related to the helical pitch of the SmC* phase (see equations (2) and (3)).

From equation (2), we can also deduce the twist elastic constant:

$$K_{33} = \frac{1}{8\pi^2\varepsilon_0} \left(\frac{p^2}{\Delta\varepsilon_G} \right) \left(\frac{P_S}{\theta} \right)^2. \quad (5)$$

We thus see that we can estimate the elastic constant K_{33} of the SmC* phase from the measured values of p , P_S , θ and $\Delta\varepsilon_G$. The variations of this characteristic parameter with temperature (for C11 and C12) are plotted in figures 7(a) and (b), respectively. At low temperatures ($T_c - T = 40^{\circ}\text{C}$) we obtained $K_{33} = 7 \times 10^{-11}$ and 13×10^{-11} N, respectively, for C11 and C12. The order of magnitude of this parameter is the same as that found by Kocot *et al* for other ferroelectric liquid crystals [22]. As can be seen, the determined value of K_{33} for C12 is twice the value obtained for C11. This result can be understood from this brief analysis: from our experimental studies (see table 2), we have measured a similar ratio of P_S/θ , for both materials. This means that the experimental ratio of the elastic constant (see equation (5)) of

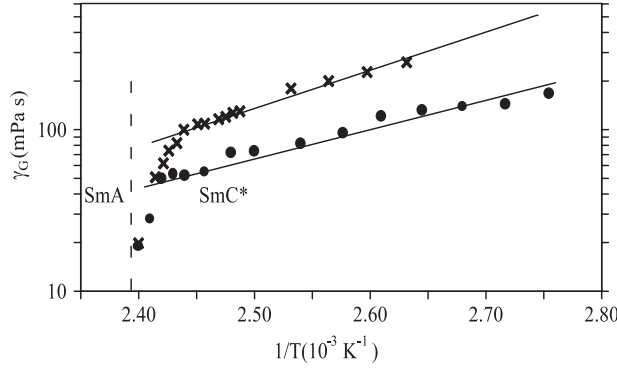


Figure 8. Arrhenius plot of the experimentally determined Goldstone-mode rotational viscosity for C11 (●) and C12 (x).

C12 and C11 is almost equal to the reduced expression:

$$\frac{K_{33(C12)}}{K_{33(C11)}} \approx \left(\frac{p^2}{\Delta \varepsilon_G} \right)_{C12} \times \left(\frac{\Delta \varepsilon_G}{p^2} \right)_{C11}. \quad (6)$$

If we take into account this expression and the experimental values of p and $\Delta \varepsilon_G$ (see table 2), this experimental ratio can easily be evaluated; as expected we then obtain $K_{33(C12)}/K_{33(C11)} \approx 2$.

Finally, our interest will focus on the evaluation of the rotational viscosity activation energy with the Arrhenius equation:

$$\gamma_G = \gamma_0 \exp\left(\frac{E_a}{k_B T}\right), \quad (7)$$

where E_a is the molecular rotation activation energy on the cone, when the applied electric field is reversed ($+E/-E$) and k_B is the Boltzmann constant. The reverse temperature $1/T$ dependence of $\ln \gamma_G \sim E_a/(k_B T)$ of the obtained rotational viscosity, for C11 and C12 is plotted in figure 8. Far from the SmC*–SmA phase transition, we observe a linear behaviour versus reverse temperature. The slopes of the Arrhenius plots give the activation energy values in the SmC* phase; we thus obtained, $E_a = 0.32$ and 0.42 eV, respectively for C11 and C12. Such values were found to be in quantitative agreement with those obtained by other authors using the same methods [23–25].

4. Conclusion

Structural, electro-optical and dielectric studies of FLCs exhibiting N*–SmA–SmC* phase sequence have been investigated as a function of temperature. All characterizations have been performed in the ferroelectric chiral SmC* phase, in which small pitch and high polarization have been measured. Using dielectric relaxation spectroscopy without bias voltage, we have studied the Goldstone-mode relaxation, which appears with a high relaxation frequency. Using structural (helical pitch), electro-optical (tilt angle and spontaneous polarization) and dielectric studies (dielectric strength and relaxation frequency), we show how the Goldstone-mode rotational viscosity and twist elastic constant in the SmC* phase can be determined from a combination of theoretical models and experimental data. The activation energies were also determined by applying the Arrhenius behaviour of the Goldstone-mode rotational viscosity.

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