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The high frequency dielectric response of a ferroelectric liquid crystal

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The temperature and frequency dependences of the complex dielectric susceptibility of a ferroelectric liquid crystal near the smectic C*–smectic A phase transition have been calculated using the classical and generalized Landau models. It is shown that although the dielectric response of the S_C^* phase consists generally of four modes (soft, Goldstone, and two high frequency polarization modes) only three bands appear in the dielectric loss spectrum of ferroelectric liquid crystals at the S_A–S_C^* phase transition. These results are in agreement with recent experimental data.

1. Introduction

The Landau model [1] and its generalizations (see, for example, [2–4]) are most frequently used to explain the thermodynamic properties of ferroelectric liquid crystals. In particular it has been explicitly shown [2–4] that the generalized Landau model can qualitatively explain the main experimental features of the dielectric behaviour of these materials.

The dynamic dielectric response of a ferroelectric liquid crystal in the context of the Landau model (both classical and generalized) consists generally of four modes—the two high frequency polarization modes and the two low frequency soft and Goldstone modes. In the S_A phase, there are only two modes—the high frequency polarization mode and the soft mode.

In recent experimental investigations of the broad band dielectric spectra (up to 1 GHz) of ferroelectric liquid crystals in the vicinity of the S_A–S_C^* phase transition, three bands in the dielectric loss spectra of S_C^* have been observed [5, 6]. In the low frequency region, the soft and Goldstone modes were observed as expected. However, in the microwave (high) frequency range, only one (β-relaxation) band was observed. At the phase transition S_A–S_C^*, this band did not split or broaden, and its dielectric strength did not strongly decrease [5, 6]. It was concluded [6] that these results did contradict the Landau model [1] in its generalized form [2–4]. Moreover these results have led Pleiner and Brand [7, 8] to suggest a model which predicts only three modes, in contrast to the four predicted by the Landau model.

2. Results and discussion

In the present paper we show that there is really no contradiction between the experimental observations [5, 6] and the predictions of the Landau model. We show that for the Landau model, we indeed expect only three peaks in the dielectric loss

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spectrum of a ferroelectric liquid crystal in the frequency range up to 1 GHz and not four as has earlier been suggested.

Calculations of the dielectric susceptibility of both the S_C^* and S_A phases have been made using the values of the material parameters introduced by Carlsson *et al.* [2] as a standard set of parameters for the S_C^* phase. We have not considered any temperature dependence of the rotational viscosities.

Results of the calculation of the imaginary part of the normalized complex dielectric susceptibility $\tilde{\chi}(\omega)=\tilde{\chi}'(\omega)-i\tilde{\chi}''(\omega)$ near the S_A–S_C^* phase transition are shown in figure 1. As expected, in the low frequency region, we can see the soft and Goldstone modes for $T<T_c$ (see curves 1 and 2 of figure 1), and only the soft mode for $T>T_c$ (see curves 4 and 5). The high frequency modes observed appear to merge into one band. Similar dependences for the classical Landau model [2] are shown in figure 2.

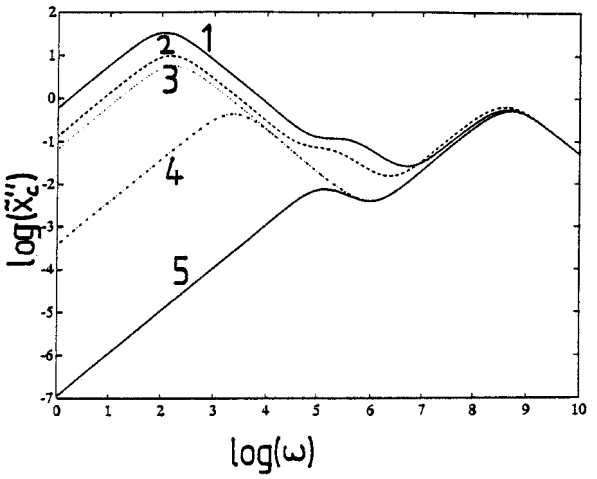

Figure 1. The imaginary part of the complex susceptibility $\tilde{\chi}''(\omega)$ for the generalized Landau model, near to the S_C^*–S_A transition. Curve 1, $T=326{\cdot}04$ K; curve 2, $T=326{\cdot}51$ K; curve 3, $T=T_c=327{\cdot}0$ K; curve 4, $T=327{\cdot}015$ K; curve 5, $T=327{\cdot}96$ K.

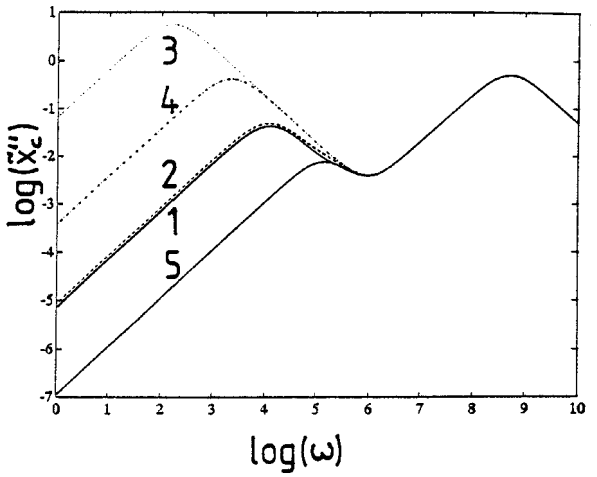

Figure 2. $\tilde{\chi}''(\omega)$ for the classical Landau model near the S_C^*–S_A transition. Curve 1, $T=326{\cdot}08$ K; curve 2, $T=326{\cdot}52$ K; curve 3, $T=327{\cdot}0$ K; curve 4, $T=327{\cdot}015$ K; curve 5, $T=328$ K.

In the context of the generalized Landau model, the high frequency dielectric susceptibility is given by the following equations [4]:

for smectic C* phase

$$\tilde{\chi}_C(\omega)=\tilde{\chi}'_C(\omega)-i\tilde{\chi}''_C(\omega)\cong\Delta\tilde{\chi}_{PC}\left[\frac{G_S}{1+i\omega\tau_{PS}}+\frac{G_G}{1+i\omega\tau_{PG}}\right],\quad (G_S+G_G=1), \tag{1}$$

for smectic A phase

$$\tilde{\chi}_A(\omega)=\tilde{\chi}'_A(\omega)-i\tilde{\chi}''_A(\omega)\cong\frac{\Delta\tilde{\chi}_{PA}}{1+i\omega\tau_{PA}}. \tag{2}$$

As we can see from equation (1), $\tilde{\chi}_C(\omega)$ consists indeed of the two high frequency polarization modes. (We denote the first and second terms in the right hand side of equation (1) as the S- and G-modes, respectively).

For the generalized Landau model, in the limit $T\to T_c$, when the normalized spontaneous polarization $\tilde{P}_0$ and tilt $\tilde{\theta}_0$ tend to zero, the dielectric strength $\tilde{\Delta}\chi_{PC}$ may be represented as (in the designations of [2])

$$\Delta\tilde{\chi}_{PC}\cong\Delta\tilde{\chi}_{PA}+\frac{1}{2}\frac{\tilde{\theta}_0^2-4\tilde{P}_0^2}{(1+v^2)^2}. \tag{3}$$

v is a material parameter, $v=\mu[\tilde{\epsilon}/k_3]^{1/2}$, μ is a coefficient of the flexoelectric coupling, k_3 is the elastic modulus and $1/\tilde{\epsilon}=1/\epsilon-\mu^2/k_3$; ϵ is a generalized susceptibility. As follows from equation (3), the model may predict both small increases and decreases of the dielectric strength $\Delta\tilde{\chi}_{PC}$. This depends on the ratio $\tilde{\theta}_0^2/4\tilde{P}_0^2$, which is determined by the material parameters of the model. Also we have a similar behaviour of the relaxation times τ_{PS} and τ_{PG}, namely

$$\tau_{PS}\cong\tau_{PA}\left[1+\frac{\tilde{\theta}_0^2-3\tilde{P}_0^2}{(1+v^2)}\right],\qquad \tau_{PG}\cong\tau_{PA}\left[1-\frac{\tilde{P}_0^2}{(1+v^2)}\right]. \tag{4}$$

Equations (3) and (4) show clearly that at the phase transition temperature $\tilde{\chi}_C(\omega)=\tilde{\chi}_A(\omega)$, where both $\tilde{\theta}_0$ and $\tilde{P}_0$ are zero.

As follows from equations (1)–(4), in the vicinity of the phase transition temperature, the deviations of the high frequency part of the susceptibility $\tilde{\chi}_C(\omega)$ from that of $\tilde{\chi}_A(\omega)$ are of the order $\{\tilde{\theta}_0^2,\tilde{P}_0^2\}$. Hence there is no splitting or broadening of the band at the phase transition, in full agreement with the comments of Pleiner and Brand [7, 8]. The frequency of the band maximum varies smoothly across the S_A–S_C^* transition, not showing any jumps, cusps or kinks (see figures 1 and 2).

Such a type of high frequency dielectric behaviour of a ferroelectric liquid crystal is not surprising in view of the fact that the imaginary part of the complex susceptibility, $\tilde{\chi}(\omega)$, for the two relaxation processes with the relaxation times $\tau_1=\tau_{PS}$ and $\tau_2=\tau_{PG}$ defined by equation (1) may have two maxima, if the ratio τ_1/τ_2 is sufficiently large [9]. The minimum value of τ_1/τ_2 for the occurrence of two maxima depends on the relative weight of the two relaxation process: for $G_1=G_2=1/2$, $\tau_1/\tau_2>3+2(2)^{1/2}\cong 5{\cdot}8$. When $G_1\neq G_2$, the ratio τ_1/τ_2 must be larger to obtain a separation of the maxima.

For ferroelectric liquid crystals we usually have $\tau_1/\tau_2<2$ [3]. In such a case, equation (6) (or (1)) predicts always only one maximum in the spectrum of dielectric loss and the frequency dependence can be approximated by a single relaxation process with a single relaxation time $\tau=(\tau_1\tau_2)^{1/2}$.

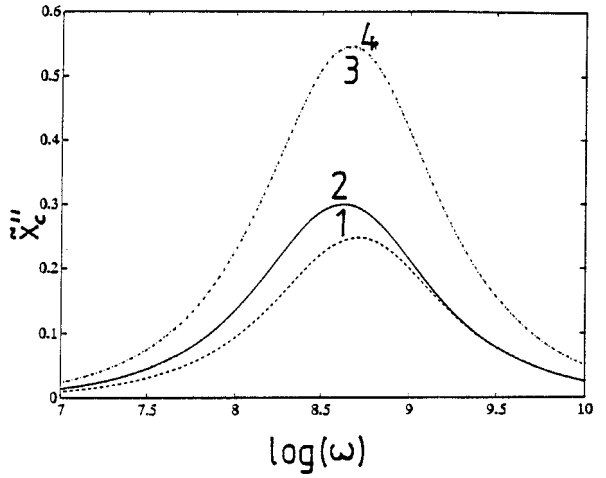

Figure 3. The high frequency behaviour of $\tilde{\chi}''_C(\omega)$, for the generalized Landau model, for the S_C^* phase, $T = 326{\cdot}81$ K. Curve 1, the high frequency G-mode; curve 2, the high frequency S-mode; curve 3, the sum of the first two curves; curve 4, the relaxation band with the single relaxation time $\tau = (\tau_1\tau_2)^{1/2}$ (in this case $\tau_1/\tau_2 \cong 1{\cdot}212$).

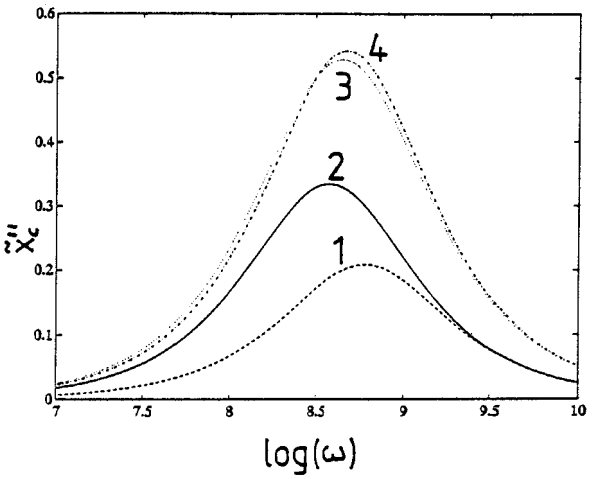

Figure 4. The high frequency behaviour of $\tilde{\chi}''_C(\omega)$, for the generalized Landau model, for the S_C^* phase, $T = 326{\cdot}04$ K. Curve 1, the high frequency G-mode; curve 2, the high frequency S-mode; curve 3, the sum of the first two curves; curve 4, the relaxation band with the single relaxation time $\tau = (\tau_1\tau_2)^{1/2}$. For this case $\tau_1/\tau_2 \cong 1{\cdot}60$.

Typical examples are illustrated by figures 3 and 4 and these support the above conclusions. We can see by inspection of these figures that the high frequency band of a ferroelectric liquid crystal resembles that for a single relaxation process.

The two relaxation times τ_{PS} and τ_{PG} in equation (1) are determined by the two rotational viscosities γ_{PS} and γ_{PG} which are related to the rotation of the molecules around their long axes. We can expect $\gamma_{PS} \cong \gamma_{PG} \cong \gamma_{PA}$. In this case, for the classical Landau model [1] we have

$$\tilde{\chi}_A(\omega) = \tilde{\chi}_C(\omega) = \frac{\Delta\tilde{\chi}}{1 + i\omega\tau}. \tag{6}$$

Equation (6) is valid for all temperatures in the vicinity of the S_C^*–S_A transition (see figure 2), that is to say, in the case under consideration, the classical Landau model predicts the three models only—the soft, Goldstone, and degenerate high frequency polarization modes.

It should be noted that the Landau model versions under consideration do not explain details of temperature dependences of experimental dielectric spectra, because it has been assumed that most of the model parameters (in particular, the rotational viscosities) are temperature independent. For example, if we accept in the context of the classical Landau model (a particular instance of the generalized case), an Arrhenius behaviour of the rotational viscosities γ_{PS} and γ_{PG} (relaxation times), i.e. γ_{PS} and $\gamma_{PG} \sim \exp(A/T)$, we could explain the temperature dependence of the frequency of the β-relaxation band maximum. Also, temperature variations of the dielectric strength observed in [6] may be explained by the temperature dependences of the intermolecular correlations, density, high frequency dielectric constant, etc.

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