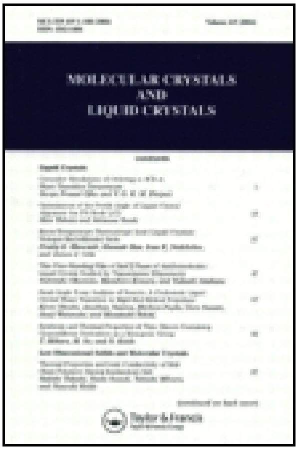

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

DIFFERENT DIELECTRIC BEHAVIORS IN THE $S^*_{C\alpha}$ PHASE: CONFRONTATION BETWEEN EXPERIMENTAL RESULTS AND THEORETICAL PREDICTIONS

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Version of record first published: 24 Sep 2006

Mol. Cryst. and Liq. Cryst., 2001, Vol. 366, pp. 637-644
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Different Dielectric Behaviors in the $S^*_{C\alpha}$ Phase: Confrontation between Experimental Results and Theoretical Predictions

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Experimental and theoretical results are compared to explain the dielectric properties in the $S^*_{C\alpha}$ phase of two homologous of the nFF series. Using the clock model and calculations developed by *N. Vaupotic and al.*, the temperature evolutions of the Goldstone and soft modes in the $S^*_{C\alpha}$ phase are studied. Two different behaviors are evidenced depending on the azimuthal angle difference α between two adjacent layers: when α is limited, the Goldstone mode is detected in all the $S^*_{C\alpha}$ phase; for higher values of α, the soft mode is predominant near the S_A- $S^*_{C\alpha}$ phase transition and the Goldstone mode is observed only at lower temperature. A good agreement is obtained with previous experimental results in considering the influence of the azimuthal angle α and its temperature evolution. These predictions are confirmed with optical period measurements performed on the same compounds.

Keywords: Dielectric relaxation; clock model; antiferroelectric liquid crystals; helical pitch

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I. INTRODUCTION

Since the discovery of the antiferroelectric, ferrielectric and so-called $S^*_{C\alpha}$ phases in liquid crystal compounds, numerous experimental and theoretical works have been devoted to explain the intermolecular interactions that stabilize these phases [1-4]. Among the different theoretical models proposed to describe antiferroelectric liquid crystal phases, the so-called clock model was found in agreement with structural observations obtained using resonant X-ray scattering [2]. Recently, this model was extended and al. to predict the dielectric properties of different chiral phases [5, 6].

In this paper, we briefly recall the different dielectric behaviours previously obtained on two homologous of a benzoate series. Using the clock model and calculations developed by N. Vaupotic et al., the temperature evolutions of the Goldstone and soft modes in the $S^*_{C\alpha}$ phase are studied. On the basis of these theoretical results, an interpretation of experimental data is proposed in considering the influence of the azimuthal angle difference α between two adjacent layers and its temperature evolution.

II. EXPERIMENTAL BEHAVIORS

The experimental results presented below were obtained on the two homologous 11FF and 9FF of a benzoate series exhibiting the following rich polymorphism [7] :

$$K - S^*_{CA} - S^*_{CFI1} - S^*_{CFI2} - S^*_{C} - S^*_{C\alpha} - S_A - I$$

These two compounds present different $S^*_{C\alpha}$ phase temperature range : 1°C and 5.2°C for 11FF and 9FF respectively. From our previous studies [8, 9], one dielectric relaxation process was evidenced in the $S^*_{C\alpha}$

phase with two different temperature behaviors (Figures 1 and 2) [1, 2]. For the 11FF compound with a small $S^*_{C\alpha}$ phase temperature range (1°C), the dielectric relaxation presents monotone evolutions versus temperature : increase of the dielectric strength and linear decrease of the critical frequency on cooling. The effect of a DC bias showed that this process is linked to the helicity of this phase (Goldstone mode) [8]. When the helix is unwound, a soft mode behavior is observed at the S_A-$S^*_{C\alpha}$ phase transition.

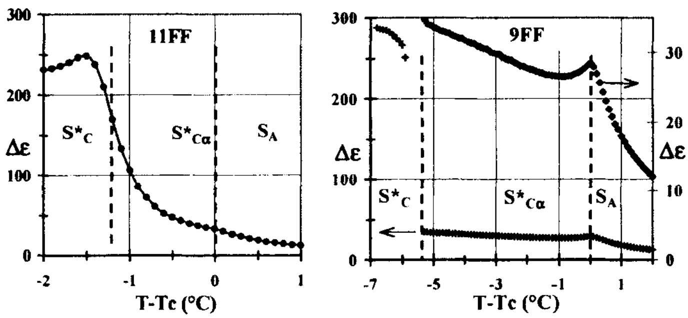

Figure 1: Experimental evolutions versus temperature of the dielectric strength in the $S^*_{C\alpha}$ phase of the homologous 11FF and 9FF.

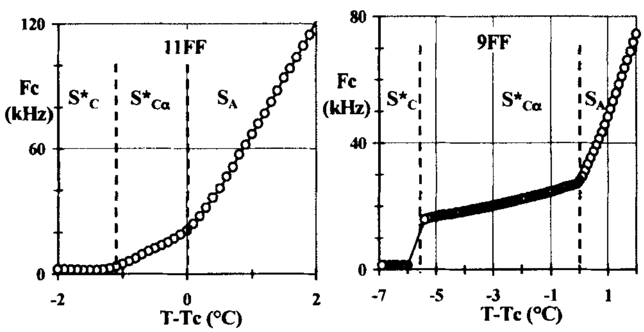

Figure 2: Experimental evolutions versus temperature of the critical frequency in the $S^*_{C\alpha}$ phase of the homologous 9FF and 11FF.

For the 9FF compound, a minimum of the dielectric strength is observed near the S_A-$S^*_{C\alpha}$ phase transition. At lower temperatures, the dielectric strength and the critical frequency slightly increase and discontinuities of these parameters are observed at the $S^*_{C\alpha}$-S^*_C phase transition. In our previous paper, these different behaviors have been attributed to the $S^*_{C\alpha}$ temperature range difference.

III. THEORETICAL MODEL AND SIMULATIONS

We discuss our experimental results on the basis of the clock model recently extended to dielectric properties prediction by N. Vaupotic et al. [5, 6]. This model takes into account ferroelectric or antiferroelectric interactions of one smectic layer with the nearest and next nearest adjacent layers respectively via the parameters a_1 and a_2 [3]. We present simulations results on the relaxation process connected with the soft mode ($\Delta\varepsilon_S$, Fc_S) and the Goldstone mode ($\Delta\varepsilon_G$, Fc_G) predicted in the $S^*_{C\alpha}$ phase. In this phase, the conditions $a_2>0$ and $|a_1|<a_2$ have to be respected and the azimuthal angle difference between two adjacent layers is $\alpha \approx \arccos(-a_1/a_2)$.

To illustrate the influence of the azimuthal angle α on the dielectric properties, we present in figures 3 and 4 the temperature dependences of the soft and Goldstone modes contributions for different values of the parameter a_1. The other parameters have been chosen equal to those previously used [5, 6].

For a limited value of the angle α (30°), the amplitude of the Goldstone mode is much higher than the soft mode in all the $S^*_{C\alpha}$ phase temperature range. So, the soft mode is masked and can't be detected.

When the azimuthal angle is larger (150°), we observe two different dielectric behaviors versus temperature in $S^*_{C\alpha}$ phase temperature range. Near the S_A-$S^*_{C\alpha}$ phase transition, the soft mode is predominant and its amplitude decreases on cooling contrary to the Goldstone mode. At lower temperatures, the Goldstone mode becomes larger than the soft mode.

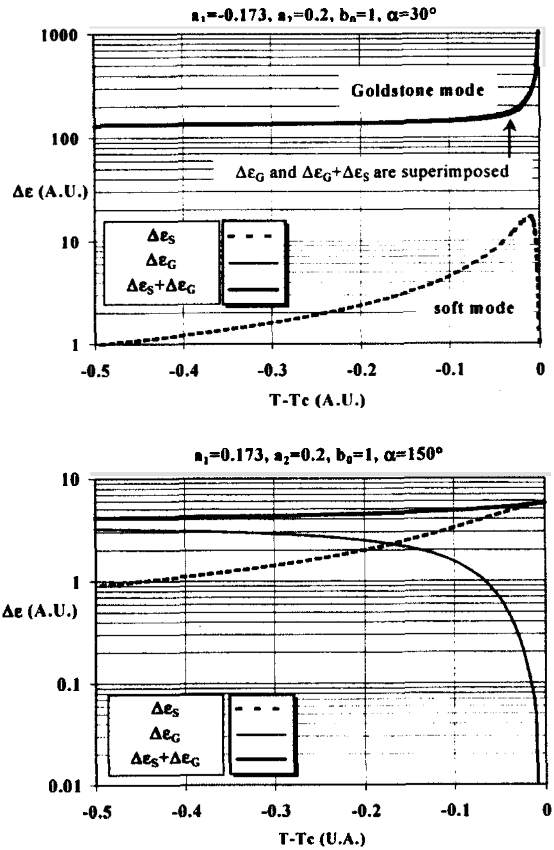

Figure 3: Dielectric strength evolutions versus temperature in the $S^*_{C\alpha}$ phase (soft and Goldstone modes).

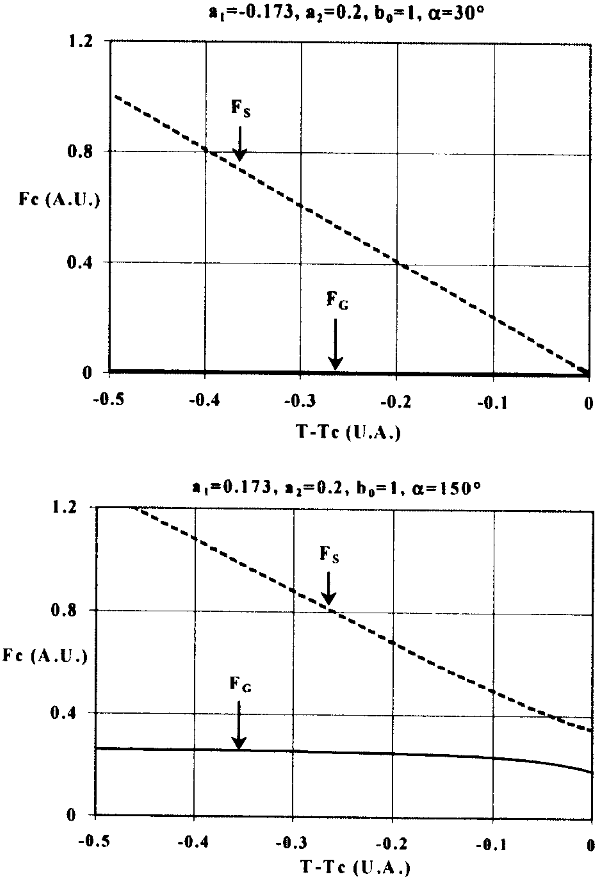

Figure 4: Critical frequency evolutions versus temperature in the $S^*_{C\alpha}$ phase (soft and the Goldstone modes).

IV. DISCUSSION

The two cases presented above can explain the experimental behavior difference mentioned in section II.

The homologous 11FF presents a monotone evolution of dielectric properties. This behavior corresponds to the case of a limited value of the azimuthal angle α. In this case, the relaxation process in this compound is linked to the Goldstone mode as it was experimentally demonstrated in previous papers [8, 9]. The dielectric strength increase and the critical frequency decrease observed on cooling can be attributed to a decrease of α with temperature.

The non monotone evolution of the dielectric properties in the 9FF compound can be explained by the predominance of the soft mode near the S_A-$S^*_{C\alpha}$ phase transition and of the Goldstone mode at lower temperatures. According to previous simulations, the observation of the soft mode in the $S^*_{C\alpha}$ phase of the 9FF compound means that this homologous presents a large azimuthal angle near the S_A-$S^*_{C\alpha}$ phase transition. Contrary to the 11FF compound, the dielectric properties discontinuities detected in the 9FF compound at the $S^*_{C\alpha}$-S^*_C phase transition are linked to the strong decrease of the azimuthal angle α at this transition.

The temperature evolutions of the azimuthal angle α experimentally deduced from optical period measurements [10, 11] confirm the previous interpretations. The optical period was found to increase (α decrease) with temperature in the $S^*_{C\alpha}$ phase of the two compounds. A divergence of the optical period was only observed near the S_A-$S^*_{C\alpha}$ phase transition of the compound 9FF showing a α angle value of 90° at this temperature. Large increase of the optical period was also evidenced at the $S^*_{C\alpha}$-S^*_C phase transition of this compound.

V. CONCLUSION

Using clock model simulations, we have shown the influence of the azimuthal angle α on dielectric properties of the $S^{*}_{C\alpha}$ phase and explained the different dielectric behaviors experimentally observed in the $S^{*}_{C\alpha}$ phase of two homologous of the nFF series. The predicted values of the azimuthal angle α and its temperature evolutions were confirmed with optical period measurements performed on the same compounds.

Acknowledgments

We would like to thank Pr. N. Isaert (Université de Lille I) and Dr. V. Laux (Université du Littoral-Côte d'Opale) for the interesting discussions.

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