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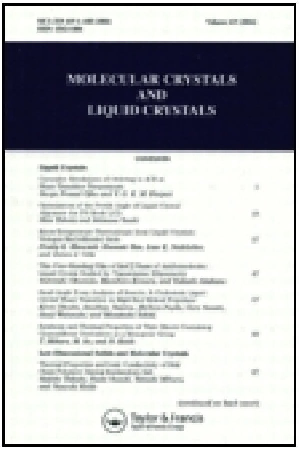

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl17

Dielectric Relaxation and the Determination of the Sm A-Sm C* Phase Transition Temperature in DOBAMBC

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Version of record first published: 19 Dec 2006.

To cite this article: A. Levstik, T. Carlsson, C. Filipič & B. Žekš (1988): Dielectric Relaxation and the Determination of the Sm A-Sm C* Phase Transition Temperature in DOBAMBC, Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics, 154:1, 259-266

To link to this article: http://dx.doi.org/10.1080/00268948808078735

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Mol. Cryst. Liq. Cryst., 1988, Vol. 154, pp. 259–266
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Dielectric Relaxation and the Determination of the Sm A–Sm C* Phase Transition Temperature in DOBAMBC

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(Received April 6, 1987; in final form September 14, 1987)

The complex dielectric constant as a function of temperature and frequency has been measured in DOBAMBC close to the Sm A–Sm C* phase transition. The relaxation frequency and the dielectric strength of the Goldstone mode and soft mode were determined and the results are compared with the dielectric strength calculated using an extended Landau type of free-energy density proposed by Žekš. By analysing the experimental data carefully we are able to establish the Sm A–Sm C* phase transition temperature to be located a few degrees above the comparatively "broad" maximum exhibited by the dielectric strength. This location is also supported by our theoretical model.

INTRODUCTION

Since the existence of ferroelectric liquid crystals was reported by Meyer *et al.*,[1] a considerable experimental and theoretical progress has been made in understanding the Sm A–Sm C* phase transition. In this context the dielectric constant is one of many studied properties[2–8], however the results of different authors are at variance with respect to some aspects. One important disparity between different authors is the location of the comparatively "broad" maximum of the low frequency dielectric constant which is generally observed. Some authors[2–4,8] locate this maximum a few degrees below the

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Sm A–Sm C* phase transition temperature T_c, while others[5–7] claim the location of the maximum to coincide with T_c. In this work we will show how a careful analysis of dielectric relaxation measurements can be used to establish that the location of the maximum a few degrees below T_c is the correct one.

EXPERIMENTAL RESULTS AND COMPARISON WITH THEORY

The frequency and temperature dependence of the complex dielectric constant has been measured on one sample of DOBAMBC in the frequency range from 20 Hz to 4 kHz close to the Sm A–Sm C* phase transition (Figures 1–3). For another sample the temperature dependence of the real part of the dielectric constant was measured at 40 Hz (Figure 4). The results of the measurements have partly been published previously[3] and also the experimental details are published elsewhere.[8] Generally two modes are expected to contribute to the dielectric strength of ferroelectric liquid crystals. One of those is connected to the phase changes (Goldstone mode) while

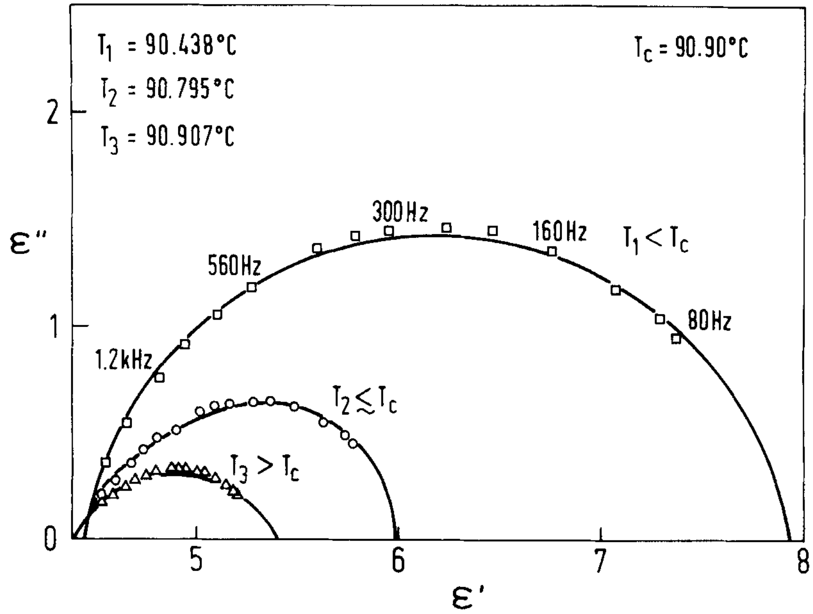

FIGURE 1 Cole–Cole diagrams of DOBAMBC at three different temperatures close to the Sm A–Sm C* transition.

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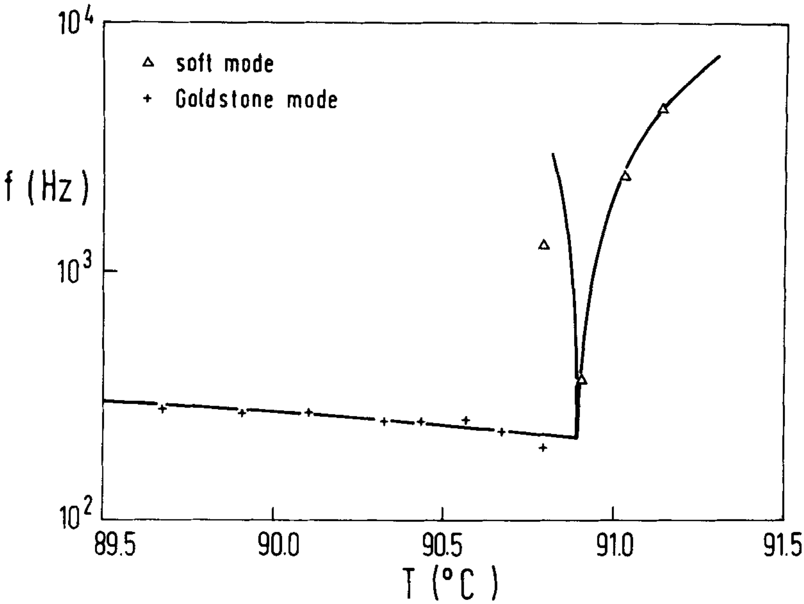

FIGURE 2 The dielectric dispersion frequency of DOBAMBC as a function of temperature.

the other is connected to the amplitude changes (soft mode) of the order parameters. We have resolved the measured dielectric strength into the contributions from these two modes. This division will enable us to locate T_c in a proper way. We also present the results of a calculation of the static dielectric susceptibility[8,9] of ferroelectric liquid crystals, which is shown to be in good agreement with the experimental data reported here.

As we expect two relaxation modes to contribute to the dielectric behaviour of ferroelectric liquid crystals we can write the total dielectric strength as $\epsilon_0 - \epsilon_\infty = \Delta\epsilon_G + \Delta\epsilon_s$, where ϵ_∞ and ϵ_0 are the infinite frequency and static dielectric constants respectively and $\Delta\epsilon_G$ and $\Delta\epsilon_s$ represent the contributions from the two modes in question. The measured data were analysed by the use of Cole–Cole plots.[10] The details of this analysis in the case of ferroelectric liquid crystals are discussed elsewhere.[8] In Figure 1 we have plotted Cole–Cole diagrams at three different temperatures. These correspond to the three distinct behaviours which are observed by analysing the data. In the Sm C* phase, except close to T_c, we observe Cole–Cole diagrams exhibiting one relaxation only ($T = T_1 < T_c$). This relaxation is the Goldstone mode. Close to T_c, in the Sm C* phase, we observe

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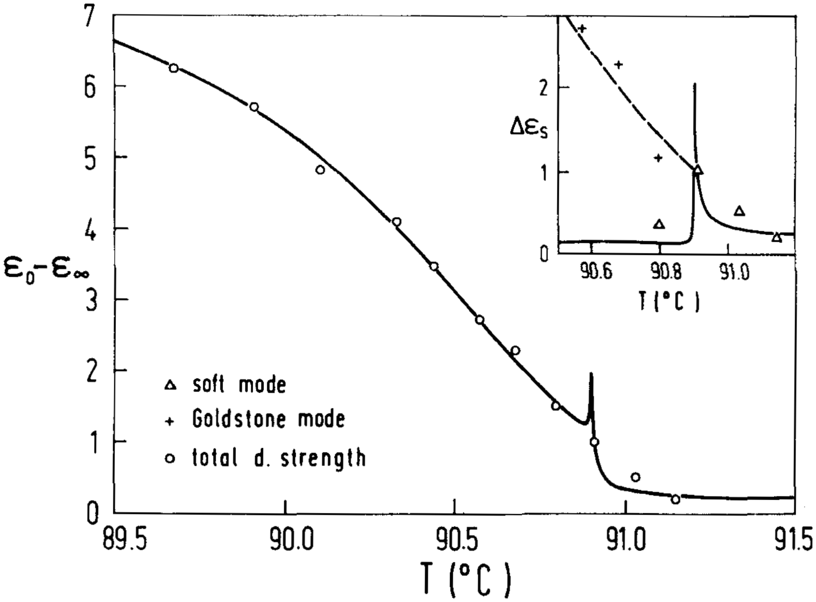

FIGURE 3 The dielectric strength $\epsilon_0 - \epsilon_\infty$ of DOBAMBC as a function of temperature (circles). The full line represents calculated results using the parameter values[8] $\gamma = 1.84$, $\beta = 0.28$, $\rho = 2.0$, $\lambda = 0.091$, $\nu = -0.010$ and $\delta = 2.6 \times 10^{-3}$. The inset shows the separation of the dielectric strength into the Goldstone mode (crosses) and the soft mode (triangles). The full line represents the calculated contribution of the soft mode while the dotted line represents the calculated contribution of the Goldstone mode.

the superposition of two relaxations ($T = T_2 \lesssim T_c$). Here both the Goldstone mode and the soft mode contribute to the dielectric strength, a behaviour which can be observed only in a narrow temperature interval, $\Delta T \approx 0.2$ K, just below T_c. In the Sm A phase only one relaxation is observed, corresponding to the soft mode ($T = T_3 > T_c$). The intensity of this is rapidly decreasing as the temperature is raised.

In Figure 2 the relaxation frequency of the Goldstone mode (crosses) and of the soft mode (triangles) obtained by the analysis of Cole–Cole diagrams are shown as a function of temperature. The cusp-like minimum of the relaxation frequency of the soft mode enables us to determine T_c. For $T > T_c$ the solid line through the experimental points is given by the expression $f_s = k\,(T - T_c)$ where we have determined k to be 17.6 kHz K^{-1}. For $T < T_c$ we have plotted the line $f_s = 2k\,(T_c - T)$. This is motivated by the fact that in solid ferroelectrics the behavior of the soft mode is generally such, that

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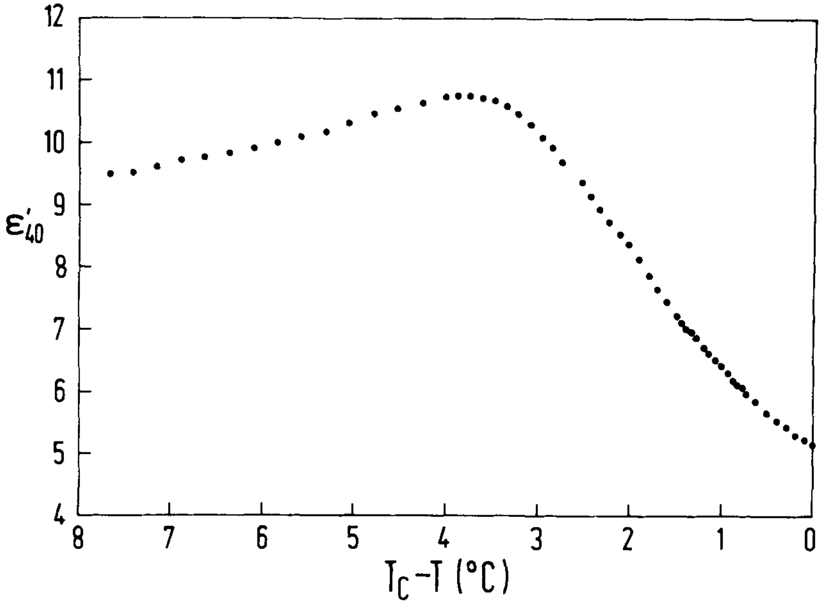

FIGURE 4 The real part of the dielectric constant of DOBAMBC obtained at 40 Hz.

the slope of the $f_s(T)$ line is twice as large in the low symmetry phase.[11] The solid line through the points of the Goldstone mode serves just as a guide to the eye.

In Figures 3 and 4 are shown the dielectric strength $\hat{\epsilon} = \epsilon_0 - \epsilon_\infty$ and the real part of the dielectric constant obtained at 40 Hz, ϵ'_{40}, for the two samples respectively. As can be seen from Figure 2, 40 Hz can be assumed to be a low frequency compared to the typical relaxation frequencies of the system. Thus the qualitative behaviour of $\hat{\epsilon}$ and ϵ'_{40} can be expected to be the same. In Figure 3, the circles show the obtained dielectric strength as a function of temperature. The dielectric strength is related to the susceptibility χ by the relation $\chi = \epsilon_s \hat{\epsilon}$, where ϵ_s is the permittivity of free space. We have developed[9,12] a theoretical model of the Sm C* phase which is based on a generalized Landau expansion of the free-energy density of the system. In such a way we are able to calculate the temperature dependence of the basic thermodynamic quantities of the Sm C* phase in accordance with their experimental behaviour. Also the dielectric susceptibility of the Sm C* phase can be calculated,[8,9] by the use of this model. The solid line through the circles in Figure 3 shows the result of such a calculation. It would however lead too far even to give a brief

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summary of the calculations here, but instead the reader is referred to the original work of references 8, 9 and 12. For completeness we just give the parameter values used in the calculations: $\gamma = 1.84$, $\beta = 0.28$, $\rho = 2.0$, $\lambda = 0.091$, $\nu = -0.010$, $\delta = 2.6 \times 10^{-3}$, $T^* = 0.50$, $\chi^* = 1.59 \times 10^{-12}$ C/Vm. The division of the measured dielectric strength into the Goldstone mode and the soft mode is shown in the inset of Figure 3, where the result of the separate calculations of the corresponding quantities is also shown. We note that the soft mode is contributing to the dielectric strength only in a narrow temperature interval close to T_c, showing a peak at T_c. This peak is one of the details which enables us to locate T_c. The Goldstone mode gives a finite contribution in all the Sm C* phase showing a drop but still being finite as T_c is approached. In the Sm A phase the Goldstone mode is absent. As the contribution of the soft mode is rapidly decreasing as $T_c - T$ is increased, it is clear that the "broad" maximum of the low frequency dielectric constant can be attributed solely to the Goldstone mode. From the theoretical considerations made elsewhere[9,12] we have been able to derive the following approximate formula, expressing the Goldstone mode part of the dielectric susceptibility χ_G in terms of polarization (P_0), tilt (θ_0) and pitch (p)

$$\chi_G = \frac{1}{8\pi^2 K_3}\left(\frac{P_0 p}{\theta_o}\right)^2$$

where K_3 is the elastic modulus of the system. The validity of this relation has also been verified experimentally.[9] Further on we note that as well the experimental data as the calculations show that both modes contribute equally to the dielectric strength at T_c. The slopes of the calculated $\Delta\epsilon_G(T)$ and $\Delta\epsilon_s(T)$ are related in such a way that we expect $\hat{\epsilon}$ to exhibit a small peak at T_c. In order to determine the experimental existence of this, high resolution temperature measurements are needed to be performed close to T_c. In the paper by Yoshino *et al.*[2] there is however an indication of this peak to exist. The same general features of the dielectric behaviour as that reported for DOBAMBC above have also been observed by us in a ferroelectric liquid crystal mixture which we have named BAHABAC.[8]

CONCLUSIONS

As mentioned in the introduction, the location of T_c in connection to dielectric measurements is somewhat diversed in the literature.

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Some authors report T_c to be (wrongly) located at what turns out to be the temperature where the Goldstone mode is giving its maximum contribution[5–7] (We denote this temperature by T_0 in the following discussion). These authors have probably not determined T_c by any independent method, but just assumed T_c to coincide with this maximum, which also by the reasons discussed above is the maximum of the total dielectric strength. Other authors[2–4,8] have located T_c a few degrees above this maximum. This is, in our opinion, the correct assignment of T_c by the following reasons:

1. Both the relaxation frequency and the dielectric strength of the Goldstone mode, as well as the total dielectric strength, are smooth and continuous functions of temperature at T_0. Generally, we would expect some kind of discontinuity of the derivative of this quantities at a second order phase transition.
2. At the temperature to which we assign the phase transition the dielectric strength of the Goldstone mode ceases to exist, while the dielectric strength of the soft mode exhibits a peak. Also the relaxation frequency of the soft mode exhibits a cusp-like minimum at this temperature as can be seen in Figure 2. This fulfills the requirement of a discontinuous behaviour of the dielectric properties at T_c which was demanded above.
3. By the simultaneous measurements at the low frequency dielectric constant and the pitch of the helix of a ferroelectric liquid crystal mixture,[12] we have confirmed our choice of T_c to be the correct one.

In conclusion we summarize that in this work we have presented dielectric measurements of DOBAMBC, close to the Sm A–Sm C* phase transition. Resolving the data into the contributions from the Goldstone mode and the soft mode, we have been able to determine that the Sm A–Sm C* phase transition temperature is located a few degrees above the temperature where the dielectric susceptibility adopts its "broad" maximum, a maximum which is solely attributed to the Goldstone mode. We have also demonstrated, that the calculation of χ, the details of which are presented by us elsewhere,[8,9] is able to describe the dielectric behaviour of DOBAMBC in good agreement with experiments. We also point out, that the same general features of the dielectric properties as those observed by us in DOBAMBC, are also exhibited by a ferroelectric liquid crystal mixture which we have studied,[8] thus suggesting this behaviour to be a general feature of ferroelectric liquid crystals.

References

1. R. B. Meyer, L. Liebert, L. Strzelecki and P. Keller, *J. Physique Lett.*, **36**, L69 (1975).
2. K. Yoshino T. Uemato and Y. Inuishi, *Japan J. Appl. Phys.*, **16**, 571 (1977).
3. A. Levstik, B. Žekš, I. Levstik, R. Blinc and C. Filipič, *J. Physique C3*, **40**, C3-303 (1979).
4. M. Glogarova and J. Pavel, *Mol. Cryst. Liq. Cryst.*, **114**, 249 (1984).
5. J. Hoffman, W. Kuczynski and J. Malecki, *Mol. Cryst. Liq. Cryst.*, **44**, 287 (1978).
6. B. I. Ostrowskii, A. Z. Rabinowich, A. S. Sonin and B. A. Strukov, *Zh. Eksp. Teor. Fiz.*, **74**, 1748 (1978).
7. L. G. Benguigui, *Ferroelectrics*, **58**, 269 (1984).
8. A. Levstik, T. Carlsson, C. Filipič, I. Levstik and B. Žekš, *Phys. Rev. A*, **35**, 3527 (1987).
9. T. Carlsson, B. Žekš, C. Filipič, A. Levstik and R. Blinc, submitted to Mol. Cryst. Liq. Cryst.
10. K. S. Cole and R. H. Cole, *J. Chem. Phys.* **9**, 341 (1941).
11. R. Blinc and B. Žekš, Soft Modes in Ferroelectrics and Anti-ferroelectrics, North-Holland Publishing Company (Amsterdam 1974).
12. T. Carlsson, B Žekš, A. Levstik, C. Filipič, I. Levstik and R. Blinc, *Phys. Rev. A*, **36**, 1484 (1987).
13. A. Levstik, B. Žekš, C. Filipič, R. Blinc and I. Levstik, *Ferroelectrics*, **58**, 33 (1984).

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