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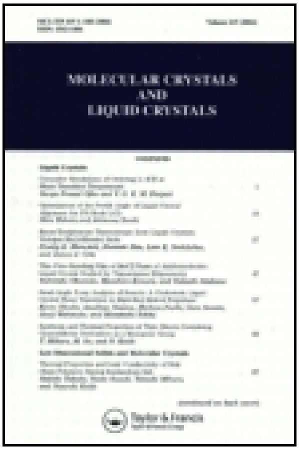

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Dielectric Properties of Smectic C* Liquid Crystals

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Dielectric Properties of Smectic C* Liquid Crystals

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We show that the existence of four order parameter relaxation modes in the dielectric spectrum of smectic C* ferroelectric liquid crystals follows directly from the symmetry properties of the chiral smectic C*—smectic A phase transition. Four macroscopic dynamic variables are thus needed to describe the dynamics of this transition and not three as suggested recently in a letter published in this journal.

In a recent Letter[1] H. R. Brand and H. Pleiner criticize the Landau—Khalatnikov dynamics of ferroelectric smectic C* (SmC*) liquid crystals as developed by Blinc and Žekš.[2] Specifically, they state that one has only three dynamic variables to characterize the orientational degrees of freedom in the SmC* phase and *not* four as used in Reference 2. Therefore only three dynamic order parameter modes should be observed by dielectric spectroscopy in the SmC* phase and not four as predicted by Reference 2. According to their opinion[1] the redundant variable is the rotation of the macroscopic polarization which is already described by the rotation of the tilt direction. The only dynamic variable involving the in-plane spontaneous polarization is thus the fluctuation in the absolute value (i.e., the magnitude) of the polarization.[1]

Here we would like to point out that the existence of four dynamic variables[2] and four order parameter modes[2] in the SmC* phase follows from symmetry arguments and involves no approximations or arbitrary assumptions. The order parameters of the SmA (point group D_∞) – SmC* (point group C_2) phase transition[3] are the two-component tilt vector

$$\vec{\xi} = (\xi_1, \xi_2) \tag{1a}$$

and the two-component in-plane polarization vector

$$\vec{P} = (P_x, P_y) \tag{1b}$$

which are bilinearly coupled in lowest order. In the SmC* phase we have a spontaneous tilt $\vec{\xi}_0$ and a spontaneous polarization $\vec{P}_0$. The fluctuations around the equilibrium value

$$\vec{\xi}(\vec{r}, t) = \vec{\xi}_0 + \delta\vec{\xi}(\vec{r}, t) \tag{2a}$$

$$\vec{P}(\vec{r}, t) = \vec{P}_0 + \delta\vec{P}(\vec{r}, t) \tag{2b}$$

are also coupled and give rise in the SmC* phase to two "in-phase tilt angle—polarization" fluctuation modes and two "out of phase tilt angle—polarization" fluctuation modes. Whereas $\vec{\xi}_0$ and $\vec{P}_0$ are perpendicular, this is not the case for $\vec{\xi}(\vec{r}, t)$ and $\vec{P}(\vec{r}, t)$. To a rather good approximation, the eigenvectors of the two low frequency "in-phase" modes can be described as a fluctuation in the magnitude of the tilt angle

$$\delta\vec{\xi}_1 \parallel \vec{\xi}_0 \tag{3a}$$

giving rise to a "soft" amplitudon-like mode, and a fluctuation in the direction of the tilt vector

$$\delta\vec{\xi}_2 \perp \vec{\xi}_0 \tag{3b}$$

giving rise to a zero frequency symmetry recovering Goldstone (phason) mode for the critical wave vector $\vec{q} = \vec{q}_0$. Obviously these two modes are degenerate in the SmA phase where there is no spontaneous tilt, $\vec{\xi}_0 = 0$.

The two "out of phase" tilt angle—polarization fluctuation modes will be of much higher frequency and thus not critical. To a rather good approximation, the eigenvectors of these two modes can be described as a fluctuation in the magnitude of the polarization

$$\delta\vec{P}_3 \parallel \vec{P}_0 \tag{4a}$$

giving rise to a "polarization" amplitudon like mode, and a fluctuation in the direction of the polarization

$$\delta\vec{P}_4 \perp \vec{P}_0 \tag{4b}$$

giving rise to a polarization "phason-like" mode. Physically this amounts to a rotation of the polarization around the molecular long axis. Since the spontaneous polarization is zero in the SmA phase, $\vec{P}_0$, these two modes are degenerate above the SmC*—SmA and close in frequency just below the SmA—SmC* transition.

The above theory thus predicts four order parameter modes in the SmC* phase: two low frequency ones (below 1 MHz), which have been already observed,[4] and two high frequency ones, which are still a matter of some controversy.[4,5,6] These last two modes should coincide in frequency as the SmC*—SmA transition is approached from below.

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Any attempt[1] to reduce the number of dynamic order parameter variables (and thus degrees of freedom) from four to three thus amounts to a description of the in-plane polarization by a scalar quantity and not a vector, violating basic symmetry principles.

It should be stressed that the number of modes observed in dielectric spectroscopy should be in fact larger and not smaller than four. This is so as in addition to the four order parameter modes one expects to see in the SmC* phase also rotations around the short molecular axis (amounting to head-tail flipping) as well as a number of internal molecular rotations. These last contributions are due to the fact that the molecules are flexible and far from rigid rods.

One should also add a word of caution about the point raised in Reference 1 concerning the biasing of molecular rotation in the SmC* phase. Magnetic resonance experiments[7,8] have clearly demonstrated that the tilting of the directors at the SmA—SmC* and SmA—SmC transitions induces a small, but non-zero biasing of the molecular rotation around the long axis which is absent—on the time average—in the SmA phase. There is however of course no rotational freeze-out at the SmA—SmC* transition.

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