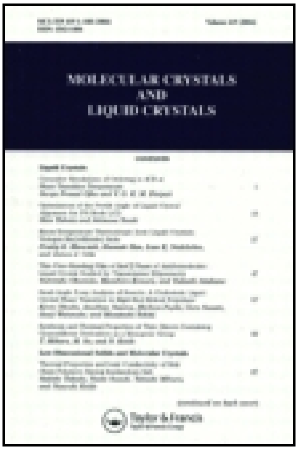

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Dielectric Degrees of Freedom in Chiral Smectic C Liquid Crystals

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Dielectric Degrees of Freedom in Chiral Smectic C Liquid Crystals

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We show that there are only three low frequency relaxation modes in the dielectric spectrum of smectic C* liquid crystals due to the symmetry of that phase. Two additional symmetry violating dielectric modes may exist at very high frequencies. We refute a recent criticism of that picture expressed in this journal.

Keywords: Dielectric effects, smectics, chirality, order parameter

Recently, we have argued[1,2] that in the dielectric response of a chiral smectic C* phase only three (and not four) modes occur at frequencies up to about 1 GHz. Now, B. Zeks and R. Blinc tried to defend[3] their traditional picture of four dielectric modes using the argument that the smectic A to smectic C* phase transition would be described by four order parameters (two 2-component vectors) due to the symmetry considerations of Reference 4, However, as we will explain below, such a four dimensional order parameter description is not appropriate for that phase transition and, indeed, nowhere in Reference 4 such a claim has been made.

At the phase transition from the smectic A to the C* phase the in-plane isotropy is lost due to the occurrence of an in-plane preferred direction $\hat{\mathbf{c}}$ (the projection of the tilted director $\hat{\mathbf{n}}$ onto the layers). This reduction in symmetry from D_∞ to C_2 locally gives rise to the existence of a polarization vector $\mathbf{P}$, which forms a mutually orthogonal triad with the in-plane director $\hat{\mathbf{c}}$ and the layer normal $\hat{\mathbf{p}}$. This symmetry change is described by a two-dimensional representation,[4] where one could choose *either* the two in-plane components of the polarization (P_x, P_y) *or* the appropriate director components $(n_z n_x, n_z n_y)$ characterizing the tilt angle (between $\hat{\mathbf{n}}$ and $\hat{\mathbf{p}}$) and the tilt direction. From the symmetry point of view both sets of order parameters are equivalent, but for physical reasons the latter is the principal one. The two-dimensional order parameter describing the symmetry change gives rise to the two low frequency modes (Goldstone and soft mode) observed in dielectric measurements. The C_2 point symmetry completely fixes the direction of the polarization ($\mathbf{P} \parallel \hat{\mathbf{p}} \times \hat{\mathbf{n}}$), which is, thus, not an independent variable. However, the absolute value of the polarization is not fixed by symmetry and can fluctuate independently of the director fluctuations. Since it relaxes more slowly in

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the vicinity of the smectic A to smectic C* phase transition than other microscopic quantities, we keep it as an additional variable. Hence, the dielectric behaviour in the vicinity of the phase transition from A to C* including the polarization effects is described by three low frequency variables, two of which are related to symmetry changes (order parameters) and the third one—being a scalar quantity—is not affected by the symmetry change. The dielectric mode associated with this scalar quantity, therefore, is an 'optical' mode occurring at higher frequencies. And this is precisely what has been found in experiments[5,6] and what has since been corroborated by numerous additional experiments with quite different types of low molecular weight and polymeric liquid crystalline materials[7-11] showing an A to C* phase transition. In addition, dielectric investigations of the A to C transition in racemates[7,9] show no change for the high frequency relaxation demonstrating that this third mode is not at all correlated with the symmetry change at the phase transition. This is further evidenced by the observation of the same mode at an isotropic to smectic C* transition,[8] which is first order and where one could expect a drastic change of this mode if it were connected with the phase transition.

If one took four independent order parameters (as was still done in Reference 3) one would describe a phase transition from a phase with local D_∞ symmetry to a hypothetical phase with local C_1 symmetry. With four independent order parameters one can construct a polarization vector, which is generally no longer parallel to $\hat{\mathbf{p}} \times \hat{\mathbf{n}}$, thus reducing the C_2 (triclinic) symmetry to C_1 (monoclinic) symmetry. Thus using independently $n_z n_x$, $n_z n_y$, P_x and P_y as order parameters, one does not describe the phase transition smectic A to smectic C* (with the conventional local C_2 symmetry), but to a hypothetical chiral smectic phase with local C_1 symmetry.

Of course, at very high frequencies (very high energies) symmetry violating fluctuations (e.g. fluctuations into a phase with C_1-symmetry locally) are possible. Then the direction of the polarization is independent of $\hat{\mathbf{p}} \times \hat{\mathbf{n}}$. The two angles by which this direction is defined then constitute two additional,[1] but ultra-high, frequency modes (in-plane rotations of $\mathbf{P}$ relative to $\hat{\mathbf{c}}$ and out-of-plane rotations of $\mathbf{P}$ relative to $\hat{\mathbf{p}}$). These modes are detectable in principle by dielectric spectroscopy at ultra-high frequencies. However, it is not unlikely that at such high frequencies (many) other modes exist due to variables not kept in the Ginzburg-Landau approach, such as for example vibrations within a single molecule, thus, invalidating this approach.

For the smectic A phase the picture we give is quite analogous to that of the C* phase. Near the phase transition C_2-symmetric fluctuations are the most likely ones for energetic reasons. Thus, for any rotation $\delta\mathbf{n}$ of the director (away from the layer normal $\hat{\mathbf{p}}$) the direction of a polarization fluctuation is fixed by symmetry ($\delta\mathbf{P} \parallel \hat{\mathbf{p}} \times \delta\mathbf{n}$) and only its absolute value is an independent variable. Hence, there are two (degenerate) low frequency modes (the two rotations of the director) and one high frequency mode. The latter is not affected by the symmetry change at the smectic A to C* phase transition and does not change its general behaviour there. Indeed this is what has been found in all dielectric experiments.[5-11] These broadband dielectric studies were corrobarated by observations using quite different techniques, like NMR[12] and four-wave mixing.[13] In both cases the third, high frequency mode is not affected by the symmetry change at the A to C* phase transition. Again, ultra-high frequency modes (which have not been detected up to frequencies f of 10 GHz in the experiments[5-11]) due to fluctuations into phases with C_1 symmetry locally or due to non-Ginzburg-Landau variables are possible.

In conclusion, we have shown that in the smectic C* phase only three dielectric modes exist up to about 1 GHz due to the local C_2 symmetry of conventional C* phases. This result has not been obtained—as is assumed in Reference 3—by erroneously describing the polarization as a scalar quantity, but on the contrary, by taking into account that the local C_2 symmetry rigorously fixes the direction of the polarization relative to the director and the layer normal, thus rendering independent only three of the four variables, $n_z n_x, n_z n_y, P_x, P_y$, used in previous attempts to set up a Ginzburg-Landau description.

Note Added in Proof

The following Comment by R. Blinc and B. Zeks shows that we still disagree on the number of components of the order parameter necessary to describe the smectic A to smectic C* phase transition. The problem of dielectric fluctuations, raised again in the following Comment, has already been discussed by us in References 1 and 2.

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