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# Dielectric RelaxationPhenomenon of New Materials Containing Cholesteryl Group: Twisted Grain Boundary Phases in Liquid Crystalline Compounds

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The Twisted Grain Boundary (TGB) phases of liquid crystals (LCs) are characterized not only by bulk properties viz. weak transition enthalpy, optical texture, dielectric permittivity, dielectric anisotropy and viscosity etc. but also from the molecular parameters (relaxation frequency, dielectric strength etc.) for their mutual association and rotation under an applied electric field. Dielectric relaxation mechanism of TGB phases of LCs are related to the response of LC molecules to the application of an electric field. In the present research article, theoretical aspect of the dielectric relaxation phenomenon of TGB phases of the liquid crystals has been discussed.

Chirality in functional molecular materials is a powerful tool for inducing properties and molecular organizations absent in non-chiral materials. This is particularly true in mesogenic materials. For instance, TGB phase is a type of frustrated phases with extraordinary physical and structural features, which are found exclusively in optically

active systems.

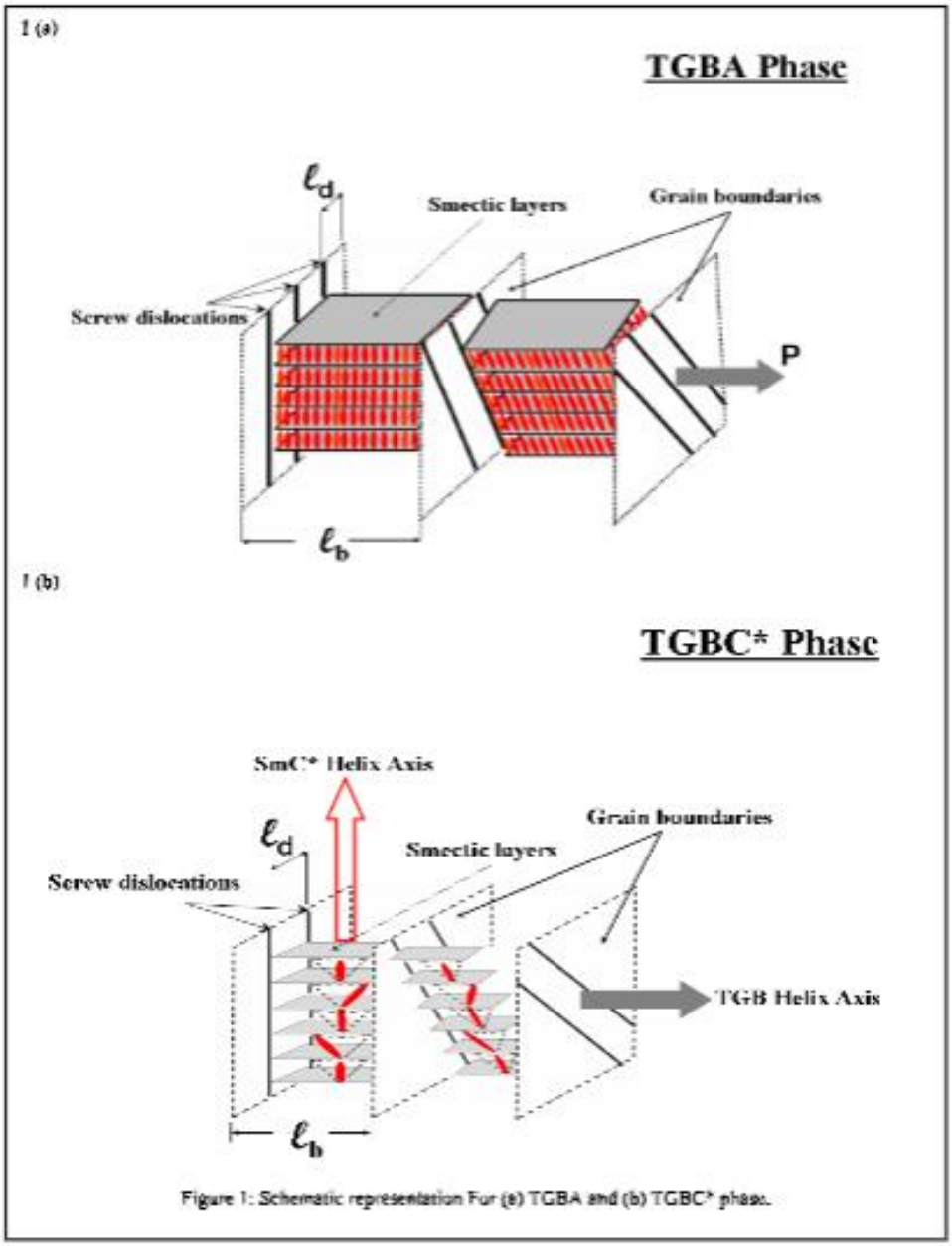


Figure 1: Schematic representation For (a) TGBA and (b) TGBC* phase.

TGB phases have been known since 1988 [**1**] and have attracted great consideration during the last two decades. Some of the chiral liquid crystals have the tendency to form a cholesteric-like helical director field along with a smectic layer structure. However, it is impossible to realize a continuous structure which exhibits both a cholesteric director field and a smectic layer structure simultaneously. The competition between these two structural features may result in frustrated structures containing a regular lattice of grain boundaries, which in turn consist of a lattice of screw dislocations. This defect structure exhibits an interesting theoretical analogy to the flux line lattice which occurs in the Shubnikov phase

of type II superconductors [**2**].However, the range of parameters determining the structure is larger in liquid crystals than in superconductors. A large variety of new phases, such as the TGBA [**1**], TGBC [**3, 4**], TGBC$^*$ [**3, 4**], melted grain boundary (MGB) phases [**5**], a defect line liquid $N^*_L$ [**6**], antiferroelectric crystals of twist grain boundaries [**7**], and smectic blue [**8**] phases have been predicted,most of theseexperimentally observed. The first experimental observation of TGBA phase was reported by Goodby et al [**9, 10**]. As shown in **Figure 1**(**a**), a TGBA phase consists of smectic slabs, separated by defect walls (grain boundaries) consisting of defect lines (twist dislocations). In the slabs, molecules are arranged in layers with their director normal to the smectic layers. Neighbouring slabs (and hence molecular director in the slabs) are twisted with respect to each other by an angle $\Delta\chi$, thereby forming a helical structure with the helix axis normal to the molecular director [**1, 2**]. The length along the helical axis corresponding to the twist of smectic slabs ($l_b$), distance between defect lines ($l_d$) and pitch (P) are related by **Equation 4**. Two more TGB phases, namely TGBC and TGBC$^*$ were predicted by Renn and Lubensky [**3, 4**]. The schematic demonstration for TGBC$^*$ phase are shown in **Figure1**(**b**). These would correspond to Abrikosov vortex lattices in a hypothetical superconductor wherein Ginzburg parameter (k) is negative and there is Bose condensation. Renn [**4**] derived the mean-field phase diagram of N$^*$-SmA-SmC$^*$ within the framework of the chiral Chen-Lubensky model, which showed the existence of TGBC and TGBC$^*$ phases. He showed that the TGBA-TGBC transition is replaced by TGBA-TGBC$^*$ transition when the cholesteric pitch length P increases beyond ~$2P_C$ /3, where $P_C$ is the pitch length of SmC$^*$. In the proposed TGBC structure [**4**], directors of the molecules in the smectic slabs are tilted with respect to the smectic layer normal. In TGBC$^*$ phase, slabs are filled by SmC$^*$ structure and thus TGBC$^*$ has two helices that are mutually perpendicular to each other. Dielectric studies of the TGB phases are scarce. So far, only a few literature data are available on the dielectric

behaviour of TGB phases [**11-32**]. This is because the mostly TGB phases exist over a very short temperature range due to their frustrated nature.Initial frequency dependent (dynamic) dielectric studies of the TGBA phase [**11-13**] show that, like those of $SmA^*$ phase, electric field induces amplitude fluctuation of tilt angle and hence soft mode of dielectric relaxation is observed in TGBA phase. Similarly in TGBC phase, electric field induces phase fluctuation of the tilt angle and hence Goldstone mode of dielectric relaxation is observed like those in $SmC^*$ phase [**14, 15**]. However experimental evidences suggest that TGB phase relaxation processes have lower amplitudes and higher relaxation frequencies as compared to those observed in the classical $SmA^*$ and $SmC^*$ phases[**33-62**]. The sameisapparent from our dielectric datathat the range of the relaxation frequency ($f_R$) i.e. ~ 40 -7700 kHz for the observed mode (presumably the soft mode) of TGBA/$TGBC^*$ phase is larger than the range of $f_R$ i.e. ~ 5 -800 kHz for the soft mode of $SmA^*$ and $SmC^*$ phases of ferroelectric systems [**33-62**]. Similarly, the dielectric strengths (~0.1-1.5) of the soft mode ($\delta\varepsilon_S$) of these TGB phases is smaller as compared to $\delta\varepsilon_S$ (~1-300) of the $SmA^*$ and $SmC^*$ phases of the ferroelectric systems [**33-62**].Ismaili et al. [**15**] have proposed a theoretical model verified by some experimental work as well, which suggests that Goldstone mode of TGBC and soft mode of TGBA phases are strongly reduced due to the existence of an elastic parameter ($H_2$) in these phases. Ismaili et al. [**15**] have obtained the dielectric strength ($\delta\varepsilon_G$) of Goldstone mode of TGBC and ($\delta\varepsilon_S$) of soft mode of TGBA phase as:

$$\delta\varepsilon_G = \frac{\varepsilon_0\chi_e^2 C^2}{H_2}\cos\theta_S^2 \qquad (1)$$

$$\text{and} \quad \delta\varepsilon_S = \frac{\varepsilon_0\chi_e^2 C^2}{\alpha(T-T_C)+H_2} \qquad (2)$$

with their respective relaxation frequencies as

$$f_G = \frac{H_2}{2\pi\gamma_G'} \quad (3)$$

and $$f_S = \frac{\alpha(T - T_C) + H_2}{2\pi\gamma_S} \quad (4)$$

Where $$H_2 = \frac{8\beta_a^2}{1-\beta_a^2/3}\frac{K_{22}}{l_b^2} \quad (5)$$

In **Equations** (**1**) to **(5)**, $\theta_S$ is the spontaneous tilt angle of TGBC phase, C expresses the linear coupling between the tilt and the polarization, $T_C = T_0+\varepsilon_0\chi_e C^2/\alpha$ with $T_0$ representing SmA-SmC transition temperature in a non-chiral compound. However this theory has not been convincingly verified for general systems possessing various TGB phases. The TGB phase relaxation processes have lower amplitudes and higher relaxation frequencies. The higher relaxation frequencies of the soft mode like relaxations are due to the presence of the elastic parameter $H_2$ (of TGB phases) in the numerator of **Equation** (**4**). Consequently, due to the presence of $H_2$ in the denominator of **Equation (2)**, the dielectric strengths of the soft mode ($\delta\varepsilon_S$) of these TGB phases is smaller as compared to $\delta\varepsilon_S$ of the $SmA^*$ and $SmC^*$ phases of the ferroelectric systems [**33-62**]. Existence of $H_2$ is connected to the elastic distortion of the director and its amplitude depends strongly on the anchoring parameter ($\beta_a$) arising due to the anchoring forces at the grain boundaries and distance between the grain boundaries ($l_b$) (see **Equations** (**5**)**)**.

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