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Liquid Crystals

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Dielectric and electro-optical properties of a photosensitive liquid crystal

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The temperature dependencies of the dielectric and electro-optical properties of a pure photo-ferroelectric liquid crystal have been investigated, in the chiral smectic C (SmC*) phase, with and without ultraviolet (UV) illumination. The SmC* dielectric Goldstone mode characteristics, the spontaneous polarization, the tilt angle and the rotational viscosity are affected by UV irradiation. Under increasing UV light intensities the dielectric strength $\Delta\epsilon_G$ increases whereas the relaxation frequency f_G , the spontaneous polarization P_s , the electro-optical rotational viscosity γ_{∞} and the tilt angle θ decrease. The twist elastic constant K_3 and the Goldstone mode rotational viscosity γ_G of the studied compound are deduced from dielectric experimental results. Good agreement was found between both viscosities. The observed dielectric behaviours are controlled by the elastic constant and the rotational viscosity variations.

Keywords: liquid crystal; ferroelectric; photosensitive; electro-optic; dielectric

1. Introduction

The photophysical properties of azobenzene derivatives, in which the light can be acting as a stimulus and control parameter (I), have attracted a significant amount of attention during the last decade because of the promising applications of these molecules in the development of various photonic devices, such as erasable optical data storage and optical switch components (2–5). The azobenzene systems are characterized by reversible photoinduced shape transformation of the molecules containing the photochromic azo groups from the *trans* form to the *cis* form (6). Upon absorption of ultraviolet (UV) light (wavelength $\lambda \simeq 365$ nm, corresponding to the π – π^* band of the azo group), the energetically more stable *trans* (*E* configuration) isomer with an elongated rodlike molecular form changes into a bent-shape less-stable *cis* (*Z* configuration) isomer. The reverse transformation (*cis* to *trans*) can be brought about either by irradiation of visible light ($\lambda \simeq 440$ nm corresponding to the n – π^* band) or spontaneously by the thermal back relaxation process in the dark. While the *trans* isomers are linear in shape and tend to stabilize the liquid crystal order, the *cis* isomers are bent and destabilize the mesophases to consequently modify the material properties (7).

Most of the studies are focused on phase transition temperatures and optical properties of these photosensitive systems, as these characteristics are critical

for their applications in different devices. Some research groups have studied the controlled photoinduced mesophases for many systems exhibiting different phase transitions: nematic–isotropic (8, 9), chiral smectic C (SmC*)– smectic A (SmA) (10, 11) and nematic–SmA (12) transitions. They demonstrated that, in addition to the photoinduced shifts of phase transition temperatures, reentrant or additional liquid crystalline phases can also be induced. The effects of UV light on electro-optical behaviours of azobenzene systems have also been reported by some groups (13–15) as electro-optical studies provide vital information regarding possible applications.

One more physical property that changes with the UV light is the dielectric relaxation of the azobenzene systems. However, modest efforts have been made to study this aspect (16, 17). The dielectric measurements of the SmC* phase give useful information about the dynamical properties of systems. The dielectric relaxation behaviour of the SmC* phase can be described mainly by two relaxation processes (18). The Goldstone mode appears owing to the phase fluctuations whereas the soft mode is connected to the amplitude fluctuations of the tilt angle. The dominant Goldstone mode is characterized by a high dielectric strength and low relaxation frequency whereas the soft mode is characterized by a lower dielectric amplitude and higher relaxation frequency. The latter is only observable when the Goldstone mode amplitude becomes weak either at the

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vicinity of the SmC*–SmA type phase transition or when a bias electric field is applied to unwind the helical structure of the SmC* phase. Therefore, we have concentrated our investigations on changes in the Goldstone mode under UV light irradiation of a pure photosensitive ferroelectric liquid crystal.

In this work, we report measurements of the dielectric strength and relaxation frequency of the Goldstone mode for the photo-ferroelectric liquid crystal with and without UV illumination. To understand the effect of the UV irradiation on the Goldstone mode and the physical parameters responsible for experimentally observed changes, we have also reported electro-optical measurements of the spontaneous polarization P_s , the tilt angle θ and the electro-optical rotational viscosity γ_{eo} . A well-known theoretical approach is used to deduce the Goldstone mode rotational viscosity and the twist elastic constant of the studied material. These investigations have been realized as a function of temperature and for different UV light intensities. A comparison of the obtained results under UV irradiation and in a dark state allows to analyse and interpret the observed behaviours.

2. Materials and methods

The influence of the illumination has been studied for the chiral photosensitive liquid crystal (S)-4-(1-methylheptyloxy)-3-fluoro-4'-(4-octadecenyloxybenzoyloxy) azobenzene (nA in short, for $n = 18$). The synthesis details and the phase sequence have already been published for compounds $n = 17$ to 18 (see (19)). The structural formula of the compound 18A is drawn in Figure 1. The polymorphism obtained in a dark state by differential scanning calorimetry (DSC; TA Instrument-Q2000) at 1°C min^{-1} is Cr (50.1°C)–SmC* (112.2°C)–twist grain boundary A phase (TGBA; 113.0°C)–I.

The liquid crystal material was introduced into commercial planar cells (EHC Inc., Japan, $15\ \mu\text{m}$ cell gap and $16\ \text{mm}^2$ indium tin oxide (ITO) electrodes) by means of capillary action in the isotropic phase. For all measurements, temperature control was achieved with a Linkam LTS120/PE94 hotstage. A well-aligned cell was obtained by applying an electric field in the slow cooling cycle from the isotropic to the SmC* phase. The temperature stability has been maintained within the range of $\pm 0.1^\circ\text{C}$. Checking of the

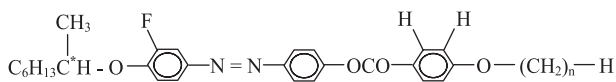


Figure 1. Chemical formula of the studied photosensitive liquid crystal namely 18A (for $n = 18$).

sample orientation and textures were performed using a polarizing optical microscope (Olympus BX60), under a low illumination from the lamp of the microscope to avoid unnecessary exposure of the sample that is susceptible to induce photo-isomerization of the azobenzene.

Trans–cis photochemical isomerization was induced by irradiation of the liquid crystal cell with a high-pressure mercury lamp or a halogen lamp of a polarizing microscope (Olympus BX60). A monochromator for selecting a wavelength ($\lambda \simeq 365\ \text{nm}$) was arranged between lamp and sample. The actual power of the UV radiation was controlled by a modification of the diaphragm. The corresponding UV measurements were performed with a power spectrometer (VLX-3W).

The dielectric measurements were carried out with a HP4192A impedance analyser generating a 100 mV sinusoidal voltage in the frequency range 5 Hz–13 MHz. The dielectric measurements were computer controlled. The real (ϵ') and the imaginary (ϵ'') parts of the complex dielectric permittivity (ϵ^*) were deduced from the impedance measurements. The dielectric dispersion data were analysed by fitting the complex dielectric permittivity, ϵ^* , with the Cole–Cole formula corresponding to m distributed relaxation processes and a static conductivity σ ,

$$\epsilon^* = \epsilon_\infty + \sum_{i=1}^m \frac{\Delta\epsilon_i}{1 + (jf/f_{ci})^{1-\alpha_i}} + \frac{\sigma}{j2\pi f\epsilon_0},$$

where $\Delta\epsilon_i$ is the dielectric amplitude or strength and f_{ci} is the critical relaxation frequency of the i th dielectric mode. Here ϵ_∞ is the high-frequency limit of the dielectric permittivity and α_i the distribution parameter of the i th dielectric mode.

Helical pitch measurements have been performed microscopically. Measurements were carried out by the Grandjean–Cano method with a prismatic cell. Pseudo-homeotropic orientation was used in the SmC* phase (20). Helical pitch measurements were performed on cooling and heating across the whole range of the SmC* phase. The average spontaneous polarization P_s and the electro-optical rotational viscosity γ_{eo} were determined by the reverse current method (21) with triangular wave voltage at 50 Hz frequency. The driving voltage, applied to the liquid crystal cell, was supplied from a function generator (HP33120A). A digital oscilloscope (HP54645A) was used to record simultaneously the applied voltage and the electro-optical response of the cell. The tilt angle (θ) variations were determined microscopically by the field reversal method and applying a low-frequency and high-amplitude square wave voltage (22).

3. Results and discussions

In this section, we report measurements of the dielectric strength and relaxation frequency of the photo-ferroelectric liquid crystal 18A in the SmC* phase with and without UV illumination. The experimental dielectric spectra display two relaxation modes. The high-frequency mode ($f > 2$ MHz) is related to the ITO electrodes of the cells. The low-frequency mode ($100 \text{ Hz} < f < 1.5 \text{ kHz}$) disappears when the helical structure of the SmC* unwinds under a DC bias voltage, consequently this mode is identified as the Goldstone mode (23, 24). No other mode was detected in the temperature range of the SmC* phase. The temperature dependencies of the average dielectric strength $\Delta\epsilon_G$ of the Goldstone mode, obtained without illumination, show that for temperatures lower than 106°C , the dielectric strength is almost temperature independent. After that, $\Delta\epsilon_G$ decreases until the SmC*–TGBA phase transition temperature T_c equal to 112.2°C . The corresponding relaxation frequency f_G varies from about 500 Hz at 84.5°C to 700 Hz at 106°C and then it quickly changes in the vicinity of T_c to reach a value close to 1.1 kHz.

Under UV illumination, the average dielectric strength and the relaxation frequency show two distinguishing features. The former is a reduction in the transition temperature T_c which is quite common in the area of photoinduced phase transitions (25, 26). Indeed the photoisomerization (*trans* to *cis*) reduces the SmC* phase stability, the result is a diminution in the phase transition temperature. The T_c values observed for different UV light intensities are plotted in Figure 2. The SmC*–TGBA phase transition temperature is continuously shifted toward lower values when the UV intensity increases. The latter feature is a

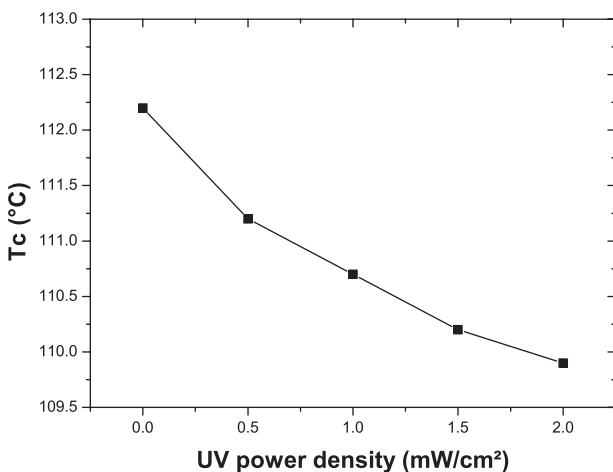


Figure 2. SmC*–TGBA phase transition temperature T_c versus UV light intensity.

growth of the Goldstone mode dielectric strength magnitude and a decrease of relaxation frequency under increasing UV illumination intensity. Curves of $\Delta\epsilon_G$ and f_G versus temperature show that the UV effect becomes relevant for UV light intensities larger than 0.5 mW cm^{-2} but becomes smaller when the temperature rises. Similar results were observed by Prasad *et al.* in the study of the soft mode of a liquid crystal material exhibiting the SmC* α phase (27).

It is commonly accepted that the influence of the UV light irradiation on photo-ferroelectric materials can be separated into primary and secondary effects (28). The former is connected to the isomer transformation from *trans* to *cis*. The resulting changes in the molecular geometry disturb the polar order of the SmC* phase leading to a reduction in the spontaneous polarisation. The latter effect is due to the presence of *cis* isomers which act as impurities by reducing the phase transition temperature. To separate the primary effect from the secondary effect (thermal effect), experimental data of the average dielectric amplitude $\Delta\epsilon_G$ and critical relaxation frequency f_G are plotted in Figures 3 and 4 versus reduced temperature with and without UV light intensity. On the one hand, these curves confirm that the UV effect is less important at high temperatures than at low temperatures. This behaviour could be due to the *cis*–*trans* thermal isomerization. On the other hand, they also highlight the existence of UV light intensity threshold effect. Indeed, the experimental curves obtained in the dark state and for 0.5 mW cm^{-2} are superposed perfectly.

In order to understand which are the relevant parameters in this problem, we used the theoretical

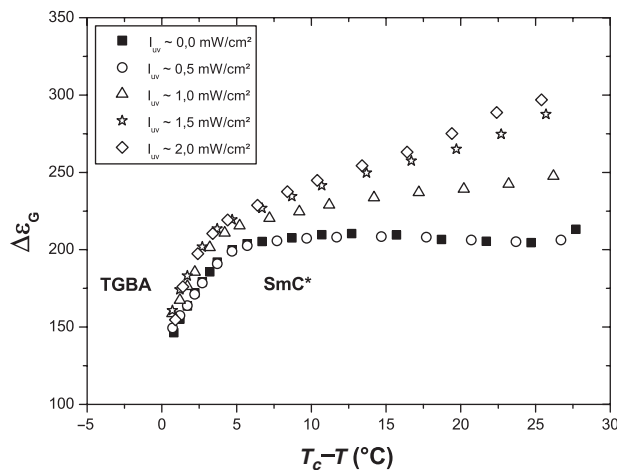


Figure 3. Dielectric amplitude of the Goldstone mode versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

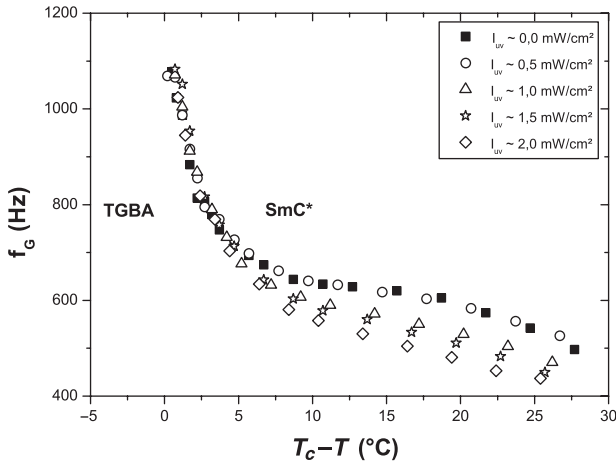


Figure 4. Dielectric relaxation frequency of the Goldstone mode versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

expressions of the dielectric amplitude and relaxation frequency of the Goldstone mode (18):

$$\Delta\epsilon_G = \frac{1}{2\epsilon_0 K_3 q^2} \left(\frac{P_s}{\theta} \right)^2, \quad (1)$$

$$f_G = \frac{K_3 q^2}{2\pi\gamma_G}, \quad (2)$$

where P_s is the spontaneous polarization, θ is the tilt angle, K_3 the twist elastic constant, $q = 2\pi/p$ is the wave vector of the helical pitch p and γ_G is the rotational viscosity of the Goldstone mode.

We have measured the helical pitch, the spontaneous polarization, the tilt angle and the electro-optical rotational viscosity with and without UV illumination. Measured values of the helical pitch vary between 0.81 and 0.89 μm and will be taken as constant in this work ($p \simeq 0.85\mu\text{m}$). The experimental data of the spontaneous polarization and the tilt angle versus reduced temperature, without and under UV illumination, are reported in Figures 5 and 6, respectively.

The general behaviour of the spontaneous polarization versus temperature is common. However, decreasing of P_s under the effect of the UV illumination is more important far from the SmC*–TGBA phase transition temperature than close to it. As previously stated, the diminution of the P_s is due to a change in the polar order under the applied UV intensity and thus is related to the concentration of the *cis* and *trans* isomers after UV irradiation. According to Langhoff and Giesselmann (28), in the SmC* phase, the change in the polar order and/or the transverse

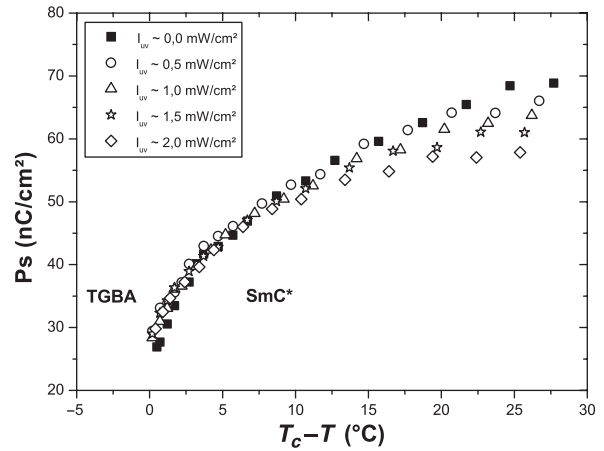


Figure 5. Spontaneous polarization versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

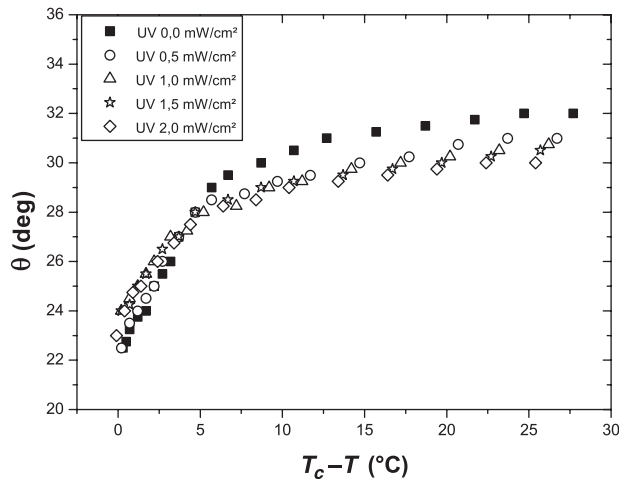


Figure 6. Tilt angle versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

molecular dipole moment is related directly to the modification of the bilinear coupling coefficient C , which is a measure of the coupling between the spontaneous polarization and the tilt angle. According to Landau theory, the magnitude of P_s depends linearly, in a first approximation, on the tilt angle ($P_s = \epsilon_0 \chi C \theta$). Langhoff and Giesselmann (29) showed that the tilt angle of a photo-ferroelectric liquid crystal mixture with low dopant concentrations remains unchanged when the sample is irradiated by low UV intensities. Nair *et al.* reported that in their systems the tilt angle changes could be only explained by the shift of the phase transition temperature (30). According to Beyer *et al.* the reduction of P_s mainly takes place in photo-induced changes of the tilt angle of the studied

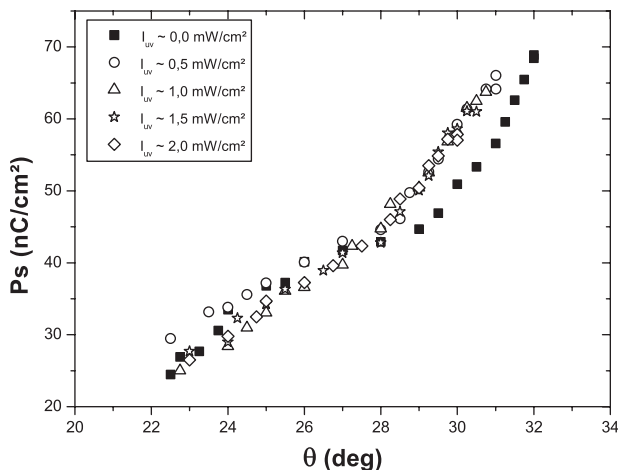


Figure 7. Spontaneous polarization versus tilt angle of the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

photosensitive ferroelectric liquid crystalline polymers (15). Our measurements of the tilt angle versus temperature (Figure 6), exhibit a modest reduction of the tilt angle values under increasing UV light intensity. Figure 7 shows that the spontaneous polarization as function of tilt angle is not linear in the dark state and for all applied UV light intensities. These results seem to indicate that the observed reduction in the spontaneous polarization P_s is a consequence of changes at the same time of the tilt angle θ and the coupling coefficient C under UV illumination.

The rotational viscosity in the SmC* phase is an important physical parameter associated with the motion of the tilted molecules along the smectic cone. This datum is needed to understand the dynamics of the ferroelectric electro-optic switching. Several electro-optical methods have been used to measure this parameter (21, 31, 32). These methods give values in good agreement with each other. In this study, we used the electro-optical transient current response to a triangular wave voltage (21, 31, 32). In Figure 8, we plot the electro-optical rotational viscosity (γ_{eo}) variations versus reduced temperature under increasing UV light intensity. These curves show clearly that the rotational viscosity varies according to the temperature and UV illumination. Under constant illumination, rotational viscosity exhibits an usual behaviour. By increasing the UV intensity, the rotational viscosity decreases. This reduction is more important far from the transition temperature T_c and attenuates with increasing the temperature. These variations must be connected to the transformation of the molecules from elongated shape (*trans* isomer) to a bent shape (*cis* isomer) under UV illumination. These shape changes could result in more important interactions between neighbouring molecules.

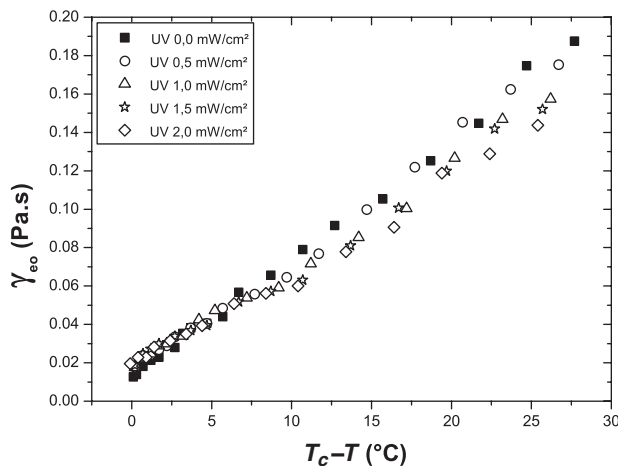


Figure 8. Electro-optical rotational viscosity versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

By using Equations (1) and (2), we deduced the theoretical expressions of the Goldstone mode rotational viscosity γ_G and the elastic constant K_3 :

$$\gamma_G = \frac{1}{4\pi\epsilon_0\Delta\epsilon_G \cdot f_G} \left(\frac{P_s}{\theta} \right)^2, \quad (3)$$

$$K_3 = \frac{1}{2\epsilon_0\Delta\epsilon_G q^2} \left(\frac{P_s}{\theta} \right)^2 = \frac{2\pi f_G \gamma_G}{q^2}. \quad (4)$$

From these formulae and the experimental data presented in Figures 3–6 we have calculated γ_G and K_3 . Deduced values of the Goldstone mode rotational

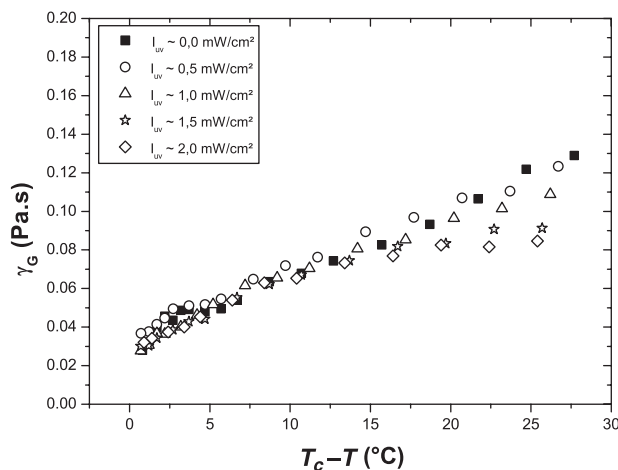


Figure 9. Goldstone mode rotational viscosity as a function of the reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

viscosity γ_G are plotted in Figure 9. These curves show a behaviour similar to that observed in the electro-optical studies. However, values of the rotational viscosity obtained with the dielectric method are slightly higher than those obtained by the electro-optical method. This difference has been reported in the literature (33, 34). The principal advantage of the dielectric method is that only small oscillations of the director are excited. Thus, problems such as disclination formation are avoided (32).

Figure 10 indicates on the one hand that the elastic constant K_3 decreases with the UV light intensity (27) and on the other hand that far from the phase transition temperature these variations are substantial. This reduction combined with the diminution in the Goldstone mode viscosity under UV illumination would be at the origin of the observed behaviours, particularly the increase of the dielectric strength and the decrease of the relaxation frequency of the Goldstone mode.

In conclusion, the Goldstone mode dielectric relaxation of a pure photo-ferroelectric liquid crystal 18A in the SmC*, is strongly affected by UV irradiation. Under increasing UV light intensities, the dielectric strength $\Delta\epsilon_G$ increases whereas the relaxation frequency f_G decreases. In addition, a SmC*–TGBA phase transition temperature shift linked with *trans* to *cis* isomerization is observed. The variations of the Goldstone mode dielectric strength and relaxation frequency are due to combined effects of the spontaneous polarization, the tilt angle, the rotational viscosity and the twist elastic constant of the material. A modest reduction of the tilt angle was measured under UV illumination. The reduction in the spontaneous polarization P_s is a consequence of the changes at the same

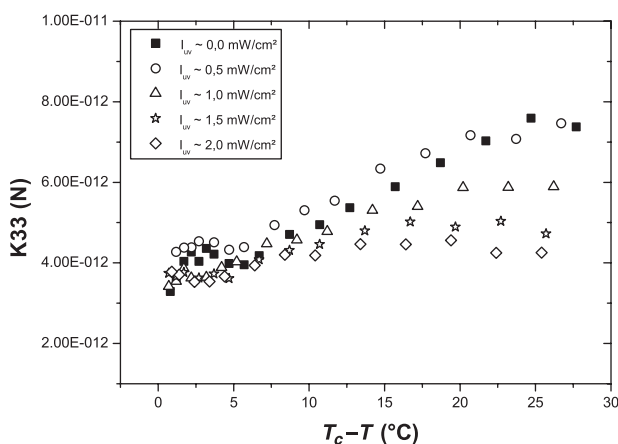


Figure 10. Twist elastic constant versus reduced temperature in the SmC* phase of the photosensitive liquid crystal 18A with and without UV light illumination.

time of the tilt angle θ and the coupling coefficient C under UV illumination. Both the twist elastic constant and the rotational viscosity are affected by UV irradiation. A good agreement was found between the experimental Goldstone mode and the electro-optical rotational viscosities. Finally, if we consider that far from the SmC*–TGBA phase transition temperature the ratio Ps/θ remains approximately constant, then the rotational viscosity and the twist elastic constant explain well the observed behaviours under UV illumination.

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