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How to Enhance Sensitivity of Liquid Crystals to External Magnetic Field?

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Liquid crystals, due to their large dielectric anisotropy, respond very sensitively to application of an external electric field, whereas they are only weakly sensitive to the magnetic field. A possible way of improving that sensitivity is doping liquid crystals with magnetic nanoparticles. As a result, stable colloidal suspensions of liquid crystals with relatively low concentrations of magnetic nanoparticles (called ferronematics, ferrocholesterics, ferrosmectics, etc.) can be produced. We illustrate some examples of the influence of the magnetic field, as well as of a superposition of magnetic and electric fields on the structural transitions (e.g. on the Fréedericksz transition) in ferronematics based on the calamitic liquid crystal 4-(*trans*-4'-*n*-hexylcyclohexyl)-isothiocyanatobenzene (6CHBT). It is shown that the samples respond to the applied magnetic field of low strength. The effects of the magnetic particles and magnetic field on the nematic to isotropic phase transition temperature are discussed as well.

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1. Introduction

Liquid crystals (LCs) are a class of soft condensed matter characterized by fluidity, a long-range orientational order and a resulting anisotropy of the physical properties. This anisotropy makes liquid crystals behave differently compared to ordinary fluids when subjected to external stresses, and thus serves as the basis for their successful commercial exploitation. Typical devices, which utilize the anisotropic optical properties of LCs, are the widely used liquid crystal displays.

In the past decades many liquid crystalline mesophases have been identified [1]. The simplest of them is the nematic phase, where the long axes of individual molecules are oriented in about the same direction (characterized by a unit vector $\boldsymbol{n}$ called the director), however, without any positional order of their centers of mass.

One of the most important early findings related to LCs was that they can be controlled by external fields. Their reorentational response typically has a threshold behaviour — an effect described by Fréedericksz [2] and named after him as the "Fréedericksz transition". Liquid crystals can be (re)oriented with electric or magnetic fields due to the anisotropy of their dielectric permittivity (ε_a) or diamagnetic susceptibility (χ_a), respectively [1]. As the dielectric anisotropy of liquid crystalline materials is of the order of unity, thus in conventional devices the required driving voltages are of the order of a few volts. In the case of applied magnetic field, however, LCs are much less sensitive: due to their very low anisotropy of the diamagnetic susceptibility ($\chi_a \approx 10^{-7}$), realignment of liquid crystals may require a large magnetic field strength $\boldsymbol{B}$ of the order of 1 T.

Ferronematics are stable colloidal suspensions of fine magnetic particles in nematic liquid crystals. They are a manifestation of the idea of Brochard and de Gennes [3], who suggested that doping liquid crystals by fine magnetic particles may enhance their sensitivity to magnetic fields. The most essential feature of these systems is a strong orientational coupling between the magnetic particles (their magnetic moment $\boldsymbol{m}$) and the liquid crystal matrix (the director $\boldsymbol{n}$). Based on the experiments, which excluded the presence of parallel orientation of $\boldsymbol{m}$ and $\boldsymbol{n}$ in some thermotropic ferronematics, Burylov and Raikher improved the original idea [4–6].

Their theory considers the finite value of the surface density of the anchoring energy W at the nematic–magnetic particle boundary. W and the parameter ω, which is defined as the ratio of the anchoring energy to the elastic energy of the liquid crystal ($\omega = Wd/K$, where d is the size of the magnetic particles and K is Frank's orientational elastic modulus), determines the type of anchoring of the nematic molecules on the surface of magnetic particles.

For $\omega \gg 1$ there is a rigid anchoring, where $\boldsymbol{m} \parallel \boldsymbol{n}$. Soft anchoring is characterized by $\omega \leq 1$ and, unlike rigid anchoring, permits both $\boldsymbol{m} \parallel \boldsymbol{n}$ and $\boldsymbol{m} \perp \boldsymbol{n}$ boundary conditions.

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The paper presents an overview of the experimental observations on structural transitions in ferronematics based on the liquid crystal 4-(*trans*-4'-*n*-hexylcyclohexyl)-isothiocyanatobenzene (6CHBT) doped with magnetic nanoparticles of different size and shape. We describe the influence of the magnetic field, as well as that of superposed magnetic and electric fields, on the structural transitions (e.g. on the Fréedericksz transition), discuss the capacitance variations due to an applied magnetic field of low strength, as well as address the effects of magnetic particles and magnetic field on the nematic to isotropic phase transition temperature.

Experimental results are divided into six subsections. In Sect. 3.1 we discuss the basics of the field induced reorientational (Fréedericksz) transitions in a liquid crystal and in ferronematics, as they provide the easiest way to observe the change in the magnetic response due to the doping. It is followed by Sect. 3.2, where the effect of doping of the same host LC with nanoparticles differing in size and shape is described. In the next Sect. 3.3 we discuss the response of ferronematics to the low magnetic fields, far below the magnetic Fréedericksz transition. Section 3.4 describes the influence of the size of rodlike nanoparticles on the magnetic Fréedericksz transition, as well as on the low magnetic field response. In Sect. 3.5 we present the magnetic-field-induced shift of the temperature of the isotropic-nematic phase transition in a calamitic liquid crystal doped with rodlike magnetic nanoparticles. The last Sect. 3.6 is devoted to structural phase transitions in a composite system, in which the phase transition from isotropic to nematic phase is via a two-phase droplet state.

2. Materials and methods

All studied ferronematic samples were based on the thermotropic nematic 6CHBT which is an enantiotropic liquid crystal with high chemical stability [7]. The nematic-to-isotropic phase transition temperature (the clearing point) of the studied nematic is $T_{NI} = 42.8\,^{\circ}$C. The liquid crystal was doped with magnetic particles of various shapes (spherical, rod-like, chain-like), or with single-wall carbon nanotubes (SWCNTs). Synthesis of the spherical magnetic nanoparticles was based on the co-precipitation of Fe^{2+} and Fe^{3+} salts by NH_4OH. Magnetite nanorods were synthesized through hydrolysis of $FeCl_3$ and $FeSO_4$ solutions containing urea. The chain-like particles have been produced by biomineralization, extracted from the magnetotactic bacteria Magnetotacticum Magnetospirillum (AMB-1) [8]. SWCNTs are commercially available from Cheap Tubes Inc. Ferronematics were finally prepared by adding the surfactant covered magnetic nanoparticles to the thermotropic nematic liquid crystal in required concentrations in the isotropic phase under continuous stirring. The homogeneity of the resulting suspension is ensured by ultrasonic agitation.

Structural transitions in ferronematic samples have been monitored by capacitance measurements in a capacitor made of indium-tin-oxide (ITO) coated glass electrodes. The capacitor with the electrode area of approximately 1×1 cm^2 has been placed into a thermostat system, regulated with a temperature stability of $0.05\,^{\circ}$C. The distance between the electrodes (the sample thickness) was $D = 5$ μm. The capacitance was measured at the frequency of 1 kHz and voltage of 0.1 V by a high-precision capacitance bridge Andeen Hagerling (the accuracy at 1 kHz is 0.8 aF). During the measurements an external magnetic field was applied parallel with the capacitor electrodes.

3. Experimental results

One of the most important questions solved in the theory of ferronematics is the problem of the equilibrium orientation of a magnetic particle, i.e. the direction of its magnetic moment $\boldsymbol{m}$ in the nematic matrix. Burylov and Raikher derived in their theory [6] an approximate relation between the critical field H_{CLC} of the nematic host and the critical field H_{CFN} of the ferronematic

$$H^2_{\mathrm{CFN}} - H^2_{\mathrm{CLC}} = \frac{2W\Phi}{\mu_0 \chi_a d}. \tag{1}$$

Here d is the mean diameter of the magnetic particles, Φ is their volume concentration in the liquid crystal, μ_0 is the permeability of vacuum, and χ_a is the anisotropy of the diamagnetic susceptibility of the host liquid crystal. Both H_{CFN} and H_{CLC} can be measured experimentally. Using $\chi_a = 4.805 \times 10^{-7}$ for 6CHBT at $35\,^{\circ}$C, and knowing the volume concentration of magnetic particles Eq. (1) yields $W \approx 10^{-5}$ N m^{-1} for the anchoring energy density. Using $K_1 = 6.71$ pN as the splay elastic constant, one obtains $\omega \approx 10^{-1} < 1$ [9]. This value corresponds to soft anchoring, which allows both initial orientations of the director and the magnetic moment of magnetic particles: parallel as well as perpendicular.

3.1. Magnetic Fréedericksz transition in 6CHBT doped with spherical magnetic nanoparticles

To investigate the influence of doping with magnetic nanoparticles the ferronematics with volume concentrations $\Phi_1 = 2 \times 10^{-4}$, $\Phi_2 = 5 \times 10^{-4}$ and $\Phi_3 = 1 \times 10^{-3}$ of the spherical magnetic particles were prepared. From capacitance measurements one can calculate a reduced capacitance $(C - C_0)/(C_{\max} - C_0)$ as the function of the applied magnetic field; C_0, $C_{\max}$ and C are the capacitances at zero, at the maximal available and at the actual magnetic field, respectively. This reduced capacitance is presented in Fig. 1 for the undoped 6CHBT as well as for the ferronematics. The obtained results show that the Fréedericksz threshold field decreases with the increasing volume concentration of the magnetic nanoparticles [9].

The decrease of the critical field with the increase of the volume concentration of magnetic nanoparticles is an evidence that the director and the magnetic moment of magnetic nanoparticles are parallel to each other.

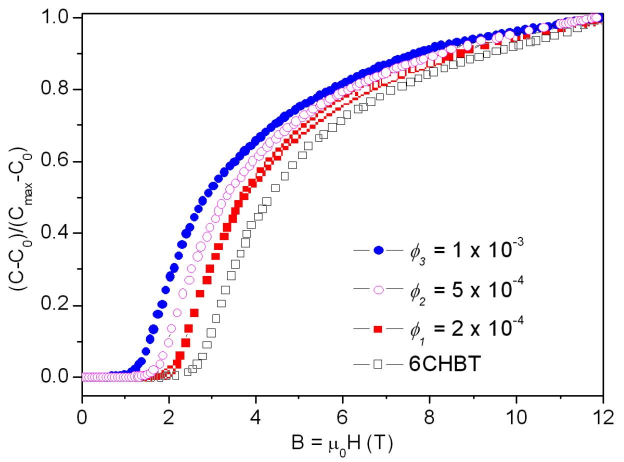

Fig. 1. Reduced capacitance versus magnetic induction for undoped 6CHBT and for 6CHBT doped with different volume concentrations of spherical magnetic nanoparticles.

3.2. Structural changes in 6CHBT doped with spherical, rodlike, and chainlike magnetic particles

In our work [8], the influence of the shape of magnetic particles on the structural transition in the 6CHBT liquid crystal was studied. Spherical, rod-like, and chain-like magnetic particles of volume concentration $\Phi = 2 \times 10^{-4}$ were used.

Figure 2 shows how the reduced capacitance of the undoped 6CHBT and 6CHBT doped with spherical, chain-like, and rod-like magnetic particles depend on external magnetic fields H measured at a bias voltage of $U_{\text{bias}} = 7$ V (applied perpendicular to H).

From these results it can be seen that the shape and the size of the magnetic particles significantly influence the strength of anchoring of the nematic molecules on the surface of magnetic particles and thus the behaviour of ferronematics in the external magnetic field. Doping with magnetic particles reduced the critical magnetic field. The highest reduction was observed for the liquid crystal doped with rod-like magnetic particles. It can be concluded that doping with magnetic particles shaped similarly to the liquid crystal molecules is advantageous for ferronematics in applications where a magnetic field is necessary to control the orientation of the liquid crystal. The experimental results again indicated mutually parallel initial orientation of the magnetic moment of the magnetic particles and the director.

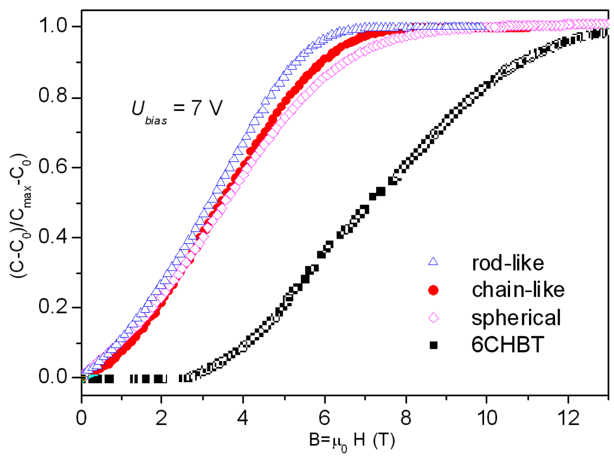

Fig. 2. Dependence of the reduced capacitance of undoped 6CHBT and 6CHBT doped with spherical, chain-like and rod-like particles on the external magnetic field measured at a bias voltage of $U_{\text{bias}} = 7$ V.

3.3. Capacitance changes in ferronematic liquid crystals induced by low magnetic fields

In recent works by Podoliak et al. [10] and Buluy et al. [11] both experimental and theoretical investigations have been reported about the optical response of suspensions of ferromagnetic nanoparticles in nematic liquid crystals on the imposed magnetic field (which finally leads to a Fréedericksz transition). The authors have measured an additional, linear response in ferronematics at low magnetic fields (far below the threshold of the Fréedericksz transition). These results inspired us to perform the experimental study of the capacitance response to low external magnetic fields (below 0.1 T) of suspensions of spherical magnetic nanoparticles, single-wall carbon nanotubes (SWCNT), SWCNT functionalized with carboxyl group (SWCNT-COOH), and SWCNT functionalized with Fe_3O_4 nanoparticles in the nematic liquid crystal 6CHBT [12]. 6CHBT was doped in two different volume concentrations of magnetic particles or SWCNT: $\Phi_1 = 10^{-4}$ and $\Phi_2 = 10^{-3}$.

In the undoped 6CHBT the magnetic Fréedericksz transition starts at 2.63 T. Due to doping, the Fréedericksz threshold is shifted to lower values, but it is still higher than 1 T (see Fig. 1).Figure 3 shows the variation

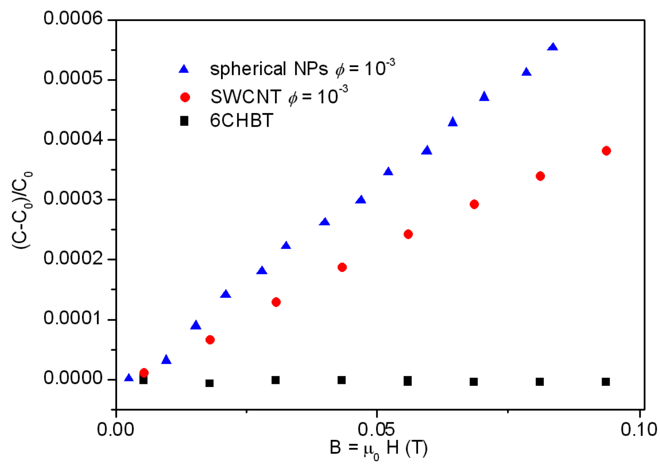

Fig. 3. Relative capacitance variation vs. magnetic field for undoped 6CHBT and 6CHBT doped with spherical Fe_3O_4 nanopaticles or with SWCNT.

of the relative capacitance of the 6CHBT liquid crystal doped with spherical Fe_3O_4 nanoparticles and SWCNT (volume concentration of $\Phi_2 = 10^{-3}$) as a function of the magnetic induction B in the low magnetic field range (up to 0.1 T), far below the threshold of the magnetic Fréedericksz transition.

Figure 3 provides a clear evidence for a linear magnetic field dependence of the capacitance in this low magnetic field region. The obtained results confirm that the ferronematic suspensions may show well measurable response in capacitance to the applied magnetic field, even much below the magnetic Fréedericksz threshold. The 6CHBT doped with "nonmagnetic" SWCNT has a low magnetic field response comparable to those of ferronematics obtained by doping 6CHBT with spherical magnetic particles.

3.4. Increasing the magnetic sensitivity of liquid crystals by rod-like magnetic nanoparticles

In work [13] the magnetic Fréedericksz transition was studied in ferronematics based on the nematic liquid crystal 6CHBT that was doped with rod-like magnetic particles of different size.

Figure 4 shows the magnetic Fréedericksz transition in undoped 6CHBT and in ferronematics doped with larger (diameter $d_A = 18 \pm 3$ nm, mean length $L_A = 400 \pm 52$ nm) and smaller (diameter $d_B = 10 \pm 1$ nm, mean length $L_B = 50 \pm 9$ nm) rod-like particles of volume concentration $\Phi = 10^{-3}$.

Using Eq. (1), the density of the anchoring energy as well as the parameter ω were estimated. The obtained values are in the order of 10^{-5} N m^{-1} and 10^{-2} for W and ωrespectively, which correspond to soft anchoring.

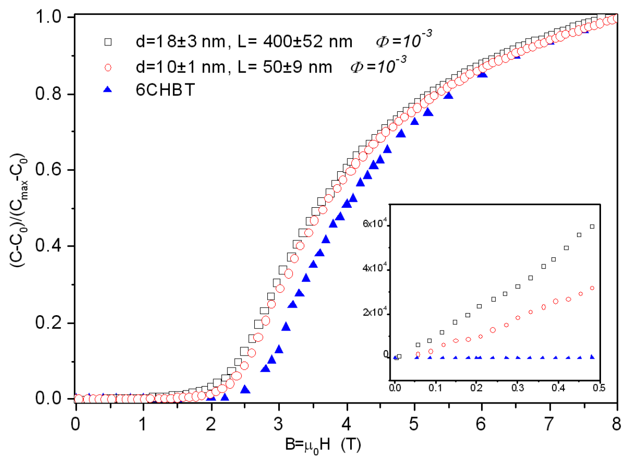

Fig. 4. Reduced capacitance versus magnetic field for undoped 6CHBT and 6CHBT doped with rod-like particles of different size. The inset represents the blowup of the low magnetic field range.

We have demonstrated that both the threshold of the magnetic Fréedericksz transition and the dielectric response to low magnetic fields (far below the Fréedericksz transition) depend not only on the volume concentration of the magnetic particles, but also on the size of the particles. According to our results, the larger is the particle, the bigger are the effects (i.e. larger decrease of the threshold of the Fréedericksz transition and a more pronounced linear response to low magnetic fields).

3.5. Magnetic-field induced isotropic to nematic phase transition in ferronematics

It has long been known that there exists a possibility in liquid crystals for an external field to substantially alter the nematic-isotropic transition temperature [14–16]. Nevertheless, the effect could not be induced by magnetic-field [17] until recently [18].

The principal reason is that the estimated critical fields are well over 100 T for traditional liquid crystal materials. The first experimental observation of the predicted magnetic-field dependence of the nematic-isotropic phase transition temperature has occurred recently [18] using a powerful electromagnet (B up to 30 T). To demonstrate the effect, besides the high field, the proper choice of a "non-conventional" (bent-core) nematic liquid crystal material was also necessary.

The magnetic field induced isotropic-nematic phase transition was also studied in the "conventional" calamitic liquid crystal 6CHBT doped with spherical magnetic particles and rod-like magnetic particles in volume concentration of $\Phi = 2\times10^{-4}$ [19]. A shift in the transition temperature from isotropic to nematic phase was observed only in the sample doped with rod-like magnetic particles. Figure 5 depicts how the capacitance of the sample varies with the temperature during the isotropic to nematic phase transition of 6CHBT doped with rod-like magnetic particles at various magnetic fields.

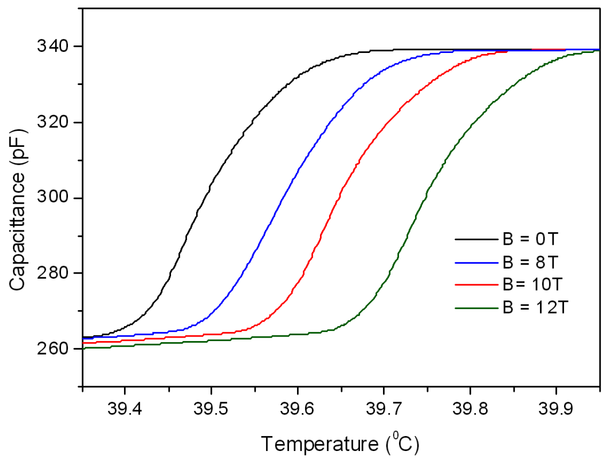

Fig. 5. Capacitance vs. temperature for 6CHBT doped with rodlike magnetic particles measured at different magnetic fields.

Figure 6 exhibits the magnetic field dependence of the capacitance at a fixed temperature. In accordance with the behaviour in Fig. 5, a reduction of the capacitance was detected upon increasing the magnetic field.

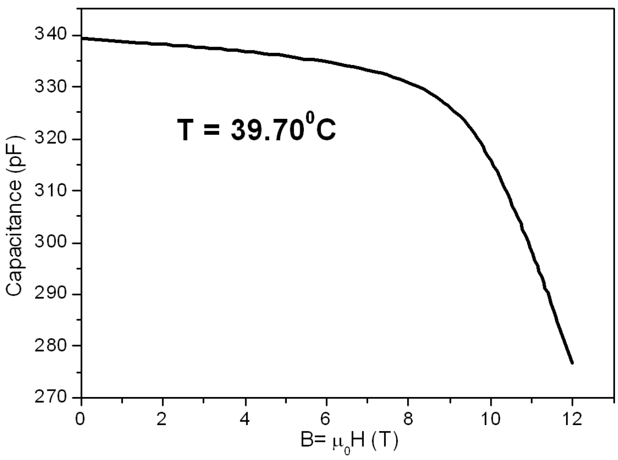

Fig. 6. Capacitance vs. magnetic field for 6CHBT doped with rodlike magnetic particles measured at constant temperature.

Our results have confirmed that the shape of the magnetic particles affects the phase transition. In the undoped 6CHBT as well as in 6CHBT doped with spherical magnetic particles no measurable field induced shift of the isotropic-nematic phase transitions temperature was observed in magnetic fields up to 12 T. On the contrary, in 6CHBT doped with rodlike magnetic particles (diameter of 10 nm, length of 50 nm and volume concentration of 2×10^{-4}) a shift of 0.25 °C was found in the phase transition temperature at 12 T. Therefore, our results have proven that ferronematics composed of calamitic liquid crystal and rodlike magnetic nanoparticles can be just as effective in demonstrating the magnetic field induced isotropic-nematic phase transition as bent-core nematics [19].

3.6. The structural transitions in 6CHBT-based ferronematic droplets

Another study [20] was inspired by the work of Kedziora et al. [21], where a coexistence of nematic and isotropic phases was observed in 6CHBT dissolved in a nonpolar medium (benzene) in the vicinity of the isotropic-to-nematic phase transition temperature. Due to the existence of a short-range orientational order of the mesogenic molecules (6CHBT), pseudonematic domains (droplets of 500–1000 nm in size depending on the temperature) were formed in the isotropic phase. The size of these droplets increased, as the temperature of the liquid decreased. We performed a similar experiment with the nematic liquid crystal 6CHBT mixed with phenyl isocyanate (that is less volatile than benzene) doped with spherical magnetic nanoparticles. The observations of the ferronematic droplets by polarizing microscope showed a similar behaviour as found for undoped droplets.

The transition from isotropic to nematic phase via this two-phase droplet state was observed by capacitance measurements. The mixture of 6CHBT and phenyl isocyanate was doped with fine magnetic particles in volume concentration of $\Phi = 5 \times 10^{-4}$. With increasing the external magnetic field the temperature of the phase transition increases as is seen from Fig. 7, which shows the temperature dependence of the capacitance of the sample measured at a bias voltage of $U_{\mathrm{bias}} = 10$ V for various external magnetic fields ($B = 0$, 2, 4, 7, 10, and 13 T).

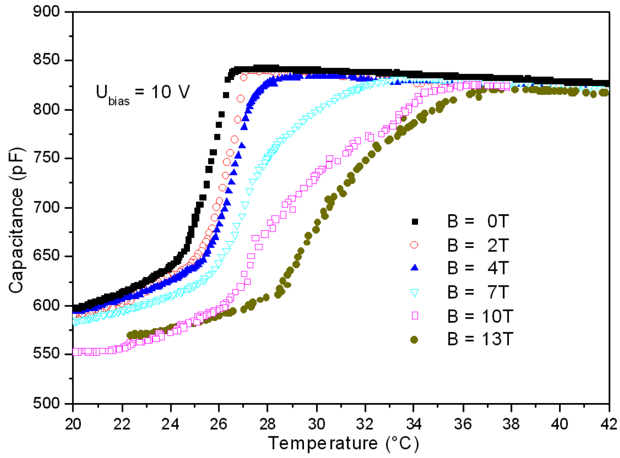

Fig. 7. Temperature dependence of the capacitance of the sample (molar fraction of 6CHBT is 0.906) doped with magnetic particles at volume concentration of $\Phi = 5 \times 10^{-4}$, for $U_{\mathrm{bias}} = 10\ \mathrm{V} < U_{\mathrm{c}}$ and external magnetic fields $B = 0$, 2, 4, 7, 10 and 13 T. (The magnetic field is applied perpendicular to the electric field.).

Embedding magnetic particles in a nematic liquid crystal causes an effective orientational coupling between the magnetic moment of the magnetic particle and the director of the nematic. This coupling may come from the anisotropy of the anchoring of the nematic molecules on the particle's surface. The results presented above show for the case of ferronematics based on the liquid crystal 6CHBT that though the anchoring of the liquid crystal molecules on the surface of magnetic particles is soft, their orientation is parallel to the magnetic moment. However, the director of the liquid crystal at some distance from the surface of the magnetic particles in the droplets already depends on the strength of the applied external electric and magnetic fields, and $\boldsymbol{m}$ may be either parallel or perpendicular to $\boldsymbol{n}$. Moreover, the temperature of the phase transition increases with the increasing value of the external magnetic field due to a field induced nematic order [20].

4. Conclusions

We reviewed our experimental work on ferronematics, namely on the liquid crystal 6CHBT doped with various magnetic nanoparticles. It was demonstrated that addition of the nanoparticles has a substantial influence on the sensitivity of ferronematics to external magnetic fields. Due to bounding between magnetic particles and molecules of the liquid crystal, the magnetic particles help to turn the molecules toward the direction of magnetic field. These results correspond to a soft anchoring, with parallel orientation of the director and the magnetic moment of magnetic nanoparticles. We have shown that the shape and the size of the magnetic nanoparticles play a significant role in structural transitions. We have observed that the magnetic particles as well as SWCNTs can influence the response of liquid crystals also in the low magnetic field range, far below the Fréedericksz transition. Moreover, an increase of the isotropic-nematic phase transition temperature could be observed in ferronematics based on a calamitic liquid crystal doped with magnetic nanoparticles at magnetic fields of ≈ 10 T.

Our experimental results obtained from samples mixed with phenyl isocyanate suggest that the temperature interval, in which the isotropic-to-nematic phase transition occurs via droplet state (i.e. the width of the two-phase region), increases as the external magnetic field increases.

Acknowledgments

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