



## High concentration ferronematics in low magnetic fields



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### ABSTRACT

We investigated experimentally the magneto-optical and dielectric properties of magnetic-nanoparticle-doped nematic liquid crystals (ferronematics). Our studies focus on the effect of the very small orienting bias magnetic field  $B_{bias}$ , and that of the nematic director pretilt at the boundary surfaces in our systems sensitive to low magnetic fields. Based on the results we assert that  $B_{bias}$  is not necessarily required for a detectable response to low magnetic fields, and that the initial pretilt, as well as the aggregation of the nanoparticles play an important (though not yet explored enough) role.

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### 1. Introduction

The control of the orientational order of liquid crystals (LCs) by magnetic field is much less wide-spread in practise than the control by electric field. The reason for this is the relatively small anisotropy of the diamagnetic susceptibility of liquid crystals. In order to overcome this difficulty, doping of LCs with magnetic nanoparticles has been proposed theoretically long time ago [1]. After the first experimental realization [2], the idea has been extensively tested in ferronematic suspensions of various compositions – see e.g., [3–5], review articles [6,7], and references therein. During these experiments an important difficulty has arisen: the aggregation of the nanoparticles [8].

A measurable optical response to low (potentially important for applications) magnetic field has been reported only lately. A linear response has been detected in planarly oriented ferronematic samples far below the threshold of the magnetic Fréedericksz transition  $B_F$ , however, in the presence of a weak orienting bias magnetic field ( $B_{bias} \approx 2$  mT) [9]. More recently, it has been shown that a similar response can be obtained even in the absence of  $B_{bias}$  [10].

The motivation of this paper was to explore the role of  $B_{bias}$ , of the initial pretilt, and that of the aggregation of nanoparticles on the response of ferronematics to low magnetic fields (below  $B_F$ ).

### 2. Experimental

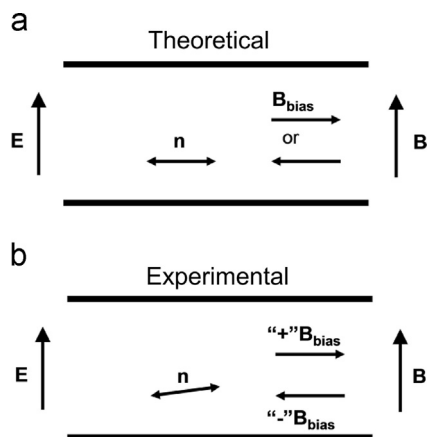
The thermotropic nematic 4-(trans-4'-n-hexylcyclohexyl)-isothiocyanatobenzene (6CHBT) was used as the LC matrix, which was doped either with spherical  $Fe_3O_4$  nanoparticles having a mean diameter of about 12 nm [10] or with single-wall carbon nanotubes functionalized with  $Fe_3O_4$  nanoparticles (SWCNT/ $Fe_3O_4$ ) [11] in a relatively high volume concentration of  $2 \times 10^{-3}$ .

The ferronematics have been filled into  $d \approx 50 \mu\text{m}$  thick, planarly oriented cells. The planar orientation was ensured by the anti-parallel rubbing of the polyimide layers coated on the inner surfaces of the two glass plates constituting the cell. The experimental setup was similar to that described in Refs. [9,12]. The cells were placed in a custom-made hot-stage having a thermal stability better than 0.05 °C. The cells could be exposed simultaneously to a magnetic induction  $B$  (up to 1 T), to an electric field  $E$ , and to an orienting bias magnetic field of  $B_{bias} = 2$  mT in an experimental geometry shown schematically in Fig. 1. The capacitance  $C$  and the conductance  $G$  were monitored by a Hioki 3522 impedance analyzer. Additionally, the setup allowed for optical studies in which the intensity of the transmitted light  $I$  was measured with crossed polarizers at an orientation of  $\pm 45^\circ$  with respect to the initial director  $\mathbf{n}$ . A laser diode emitting at  $\lambda = 657.3$  nm was used as a light source. The measurement control as well as the data collection was ensured by a LabVIEW program.

In the theoretical description of the planar orientation it is usually assumed that the nematic director  $\mathbf{n}$  (the unit vector describing the orientational order of the LC) is parallel with the bounding glass plates (Fig. 1(a)). In real cells, however,  $\mathbf{n}$  encloses a small pretilt-angle with the glass plates, as shown in Fig. 1(b). For cells with antiparallel rubbed polyimide layers, the pretilt-angle  $\theta_0$

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**Fig. 1.** Schematic representation of the experimental setup: (a) the pretilt angle is neglected (theoretical); (b) the pretilt angle is nonzero (experimental). Notations:  $\mathbf{n}$  – the nematic director,  $\mathbf{B}$  – the direction of the magnetic field,  $\mathbf{E}$  – the direction of the electric field, “+” and “-”  $\mathbf{B}_{bias}$  – direction(s) of the orienting bias magnetic field.

is typically between  $1^\circ$  and  $3^\circ$  [13]. A nonzero  $\theta_0$  breaks the symmetry and therefore, one has to distinguish between the “+” and “-” directions of the bias magnetic field  $B_{bias}$ , as indicated in Fig. 1(b).

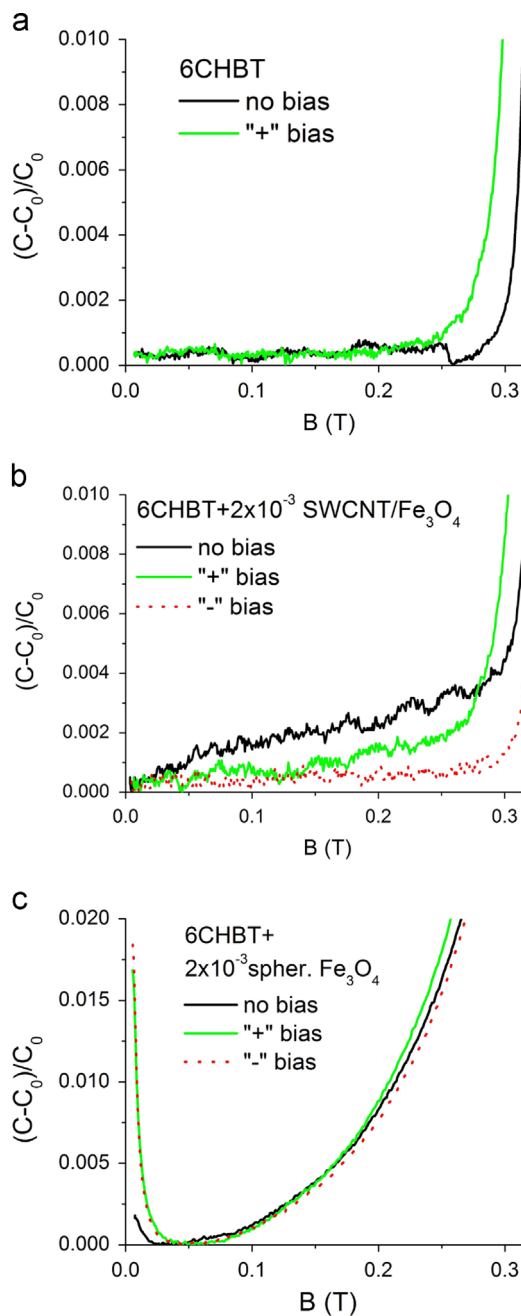
### 3. Results and discussions

The magnetic field dependence of the relative capacitance variation  $(C - C_0)/C_0$  is shown in Fig. 2 ( $C_0$  is the smallest value of the capacitance) with and without a bias magnetic field of  $B_{bias} = 2$  mT. For undoped 6CHBT neither  $B$  nor  $B_{bias}$  gave rise to a change of  $(C - C_0)/C_0$  below  $B_F$  (see Fig. 2(a)). Note that because of the presence of the pretilt, the Fréedericksz transition is not sharp; it becomes continuous in all experiments and therefore, one can define an apparent value of  $B_F$  only – see e.g., Ref. [14]. The application of “+”  $B_{bias}$  slightly decreases this apparent  $B_F$ . This is rather surprising, since naively one would expect that  $B_{bias}$  stabilizes the initial planar alignment (because of the positive anisotropy of the diamagnetic susceptibility of 6CHBT), and therefore, slightly increases  $B_F$ . We will come back to this question in a later discussion.

For 6CHBT doped with SWCNT/ $\text{Fe}_3\text{O}_4$ , a linear dependence of  $(C - C_0)/C_0$  on  $B$  has been detected below  $B_F$  in the absence of  $B_{bias}$  (see Fig. 2(b)). The application of  $B_{bias}$  of either “+” or “-” directions suppresses this dependence (especially for the “-” direction). This conclusion has also been confirmed by optical measurements of the phase shift  $\Delta\varphi$  between the ordinary and extraordinary waves to be discussed later. Note that “+”  $B_{bias}$  slightly decreases  $B_F$  again (as in 6CHBT), while on the contrary, “-”  $B_{bias}$  slightly increases  $B_F$  compared to that detected in the absence of  $B_{bias}$ .

In 6CHBT doped with spherical  $\text{Fe}_3\text{O}_4$  nanoparticles the dependence  $((C - C_0)/C_0)(B)$  is qualitatively different: it is not a monotonic function, but it has a minimum below  $B_F$  (see Fig. 2(c)). The Fréedericksz transition becomes “smoother”, i.e., the transition is much more continuous than in 6CHBT or in 6CHBT doped with SWCNT/ $\text{Fe}_3\text{O}_4$  (cf. Fig. 2(a)–(c)). On the other hand, “+” and “-”  $B_{bias}$  decreases and increases  $B_F$ , respectively, in a similar manner as in 6CHBT or in the ferromagnetic with SWCNT/ $\text{Fe}_3\text{O}_4$ .

The decrease or increase of the apparent  $B_F$  depending on the application of “+” or “-”  $B_{bias}$ , respectively, can be understood by taking into account the pretilt angle. From the schematic representation in Fig. 1(b) it becomes obvious that



**Fig. 2.** The magnetic field dependence of the relative capacitance measured at  $T = 30^\circ\text{C}$  for 6CHBT (a), 6CHBT doped with SWCNT/ $\text{Fe}_3\text{O}_4$  (b), and 6CHBT doped with spherical  $\text{Fe}_3\text{O}_4$  nanoparticles (c).

when both  $B$  and “+”  $B_{bias}$  are applied, the direction of the net magnetic field encloses a smaller angle with  $\mathbf{n}$  compared to the situation when only  $B$  is applied. That leads to a slight decrease of  $B_F$  in the former case. On the contrary, when  $B$  and “-”  $B_{bias}$  are applied simultaneously the direction of the resulting magnetic field encloses a larger angle with  $\mathbf{n}$  (closer to  $90^\circ$ ) leading to an increase of  $B_F$ .

In the case of the nematic 6CHBT, the effect of the pretilt angle  $\theta_0$  can also be discussed more quantitatively if one considers the basic magnetic properties of LCs. The magnetic moment  $\mathbf{M}$  per volume induced in the nematic LC by an external magnetic field  $\mathbf{H}$  is

$$\mathbf{M} = \chi \mathbf{H}, \quad (1)$$

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