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Effect of magnitude of voltage and magnetic field on memory effect in nematic phase of liquid crystal and their composites with aerosil and goethite nanoparticles

Katarína Kónyová [a], Dmytro Miakota [a], Veronika Lacková [a], Shie-Chang Jeng [b], Dorota Węgłowska [c], Filippo Agresti [d], Peter Kopčanský [a], Natália Tomašovičová [a,*]

[a] Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, Košice, 040 01, Slovakia
[b] Institute of Imaging and Biomedical Photonics, College of Photonics, National Yang Ming Chiao Tung University, Tainan, Taiwan
[c] Institute of Chemistry, Military University of Technology, Warsaw, Poland
[d] Institute of Condensed Matter Chemistry and Technologies for Energy (ICMATE), National Research Council of Italy (CNR), Corso Stati Uniti 4, Padova, 35127, Italy

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ABSTRACT

In liquid crystals doped with aerosil nanoparticles, the state induced by applying voltage or a magnetic field stays remembered after removing the power. The ability to remember the induced state after removing the power is known as non-volatile memory effect. The present paper describes how the magnitude of voltage and magnetic field affects the memory effect in the nematic phase of liquid crystal 4-cyano-4′-pentylbiphenyl doped with aerosil nanoparticles and with a combination of aerosil and magnetic goethite nanoparticles. Capacitance measurements revealed increasing in memory when the magnitude of voltage and magnetic field is increased. Applying fields with various magnitudes provides the possibility of fabrication a multilevel memory device based on the response to electric or magnetic field. The memory can be erased, and the initial state can be restored by heating the composites to the same temperature.

1. Introduction

Liquid crystals are liquids with anisotropic properties, for which long-range molecular arrangement is responsible. During the past few decades, significant efforts have been made to improve their already outstanding properties by doping them with various nanoparticles as for example carbon based nanomaterials [1,2], zinc oxide nanoparticles [3], cobalt ferrite nanoparticles [4], gold nanoparticles [5,6] or iron oxide nanoparticle [7]. Aerosil nanoparticles dispersed in a liquid crystal affect the phase transition between isotropic and nematic phase [8–10], dielectric properties [11,12], and optical properties [12]. Additionally they form agglomerates whose size and shape have been studied [13,14], they affect electro-optical properties of liquid crystal polymers [15,16], but probably the most interesting property of the system is the presence of a memory effect.

The memory effect in nematic liquid crystal systems was first observed by Eidenschink and De Jeu in 1991 [17]. They observed a transition from scattering state to transparent after the application of voltage to nematic filled with aerosil nanoparticles. This transparent state remained stable even after removing the voltage, although a relatively high voltage (approximately 150 V) was necessary for switching to and preserving the transparent state. Weak hydrogen bonds among aerosil nanoparticles and between the aerosil nanoparticles and the liquid crystal matrix were found to be responsible for the presence of the effect [18,19]. When the concentration of the nanoparticles is within a range of approximately 1% to 10% [20,21], aerosil nanoparticles dispersed in nematic matrix form a network where nematic domains are present. The liquid crystals in these nematic domains respond to external stimuli similar to pure nematics. For instance, when electric field is applied to the composite, the molecules of liquid crystal are realigned either parallel or perpendicular to the applied voltage, depending on the sign of anisotropy of the dielectric permittivity. When aerosil nanoparticles are absent, the original orientation is reached after removing the voltage in order of milliseconds. However, when an aerosil network is present, the new orientation induced by the electric field stays remembered due to hydrogen bonds, which prevents relaxation back to the initial arrangement. To switch off the transparent state, Eidenschink and De Jeu [17] proposed using small mechanical shear or applying

* Corresponding author.
E-mail address: nhudak@saske.sk (N. Tomašovičová).

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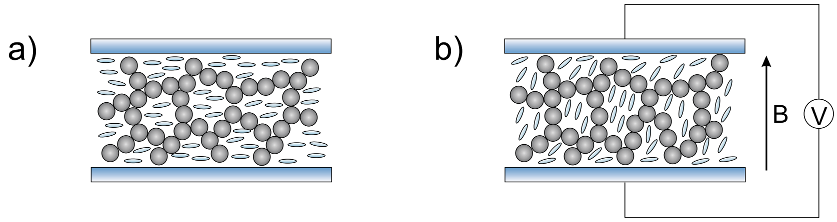

Fig. 1. The geometry of the experiment is illustrated as follows: Elongated shapes represent molecules of the liquid crystal and spheres represent aerosil nanoparticles. a) Surface treatment of the cell ensures planar alignment. b) Application of an electric or magnetic field realigns liquid crystal molecules perpendicularly to the cell surface.

ultrasound. However, the most practical solution was using a nematic liquid crystal with anisotropy of dielectric permittivity changing from positive to negative with varying a frequency of the applied voltage. This allowed switching between a transparent homeotropic alignment, reached by applying voltage with a frequency in the region of positive dielectric permittivity anisotropy, and a planar alignment, which represents the scattering state, achieved by applying voltage with a frequency in the region of negative dielectric permittivity anisotropy. Kreuzer et al. [22,18] showed a laser-writing process in the same composite. After switching to the transparent state by application of voltage, they locally reached the scattering state by irradiation with a laser beam. Thermal effects induced by laser irradiation caused the disordered scattering state. Numerous papers devoted to nematic liquid crystals doped with either aerosil [23–25] or other particles, such as carbon nanotubes [26], $Cd_{1-x}Zn_xS/ZnS$ core/shell quantum dots [27], $Bi_4Ti_3O_{12}$ [28], $Bi_2Ti_2O_7/Bi_4Ti_3O_{12}$ [29] or $SrTiO_3$ [30] were published since then. In addition, $BaTiO_3$ nanoparticles in nematic liquid crystal lead to the presence of memory effect, but only in the isotropic phase [31,32]. In the nematic phase the effect was not present. In the isotropic phase a different mechanism is responsible for the memory effect. Ferroelectric $BaTiO_3$ nanoparticles have a spontaneous permanent polarization due to which the molecules of the liquid crystal surrounding these $BaTiO_3$ nanoparticles are aligning into pseudonematic domains. Pseudonematic domains are reoriented by an electric field and as thermal energy is not sufficient to induce a random alignment and long-range elastic interaction is not present in the isotropic phase the pseudonematic domains do not recover the original alignment after switching off the voltage.

The memory effect was explored in various systems, in both the nematic as well as isotropic phase, but a pronounced memory effect at reasonably low voltage or other stimuli required for the technical applications was missing. Although the memory effect was observed mostly after applying an electric field, magnetic field or in principle other stimuli have the same potential. The presence of the memory effect in the nematic phase of liquid crystal 4-cyano-4′-pentylbiphenyl (5CB) doped with aerosil nanoparticles or a combination of aerosil and magnetic nanoparticles induced by relatively low voltages and by magnetic field was shown in our previous papers [33,34]. The addition of magnetic nanoparticles to the mixture of 5CB and aerosil nanoparticles allowed tunning the memory effect. Initially [33] spherical nanoparticles were used in our first work, which were replaced by elongated goethite magnetic nanoparticles in the following work [34]. Goethite nanoparticles have interesting magnetic properties, aligning their long axis parallel to a small magnetic field and perpendicular to a large magnetic field [35,36]. The combination of goethite nanoparticles with liquid crystal may lead to new fascinating effects. With the aim of erasing the effect and reaching the initial state, the samples were heated to the isotropic phase and then cooled back to the nematic phase. Heating the sample before performing the experiment revealed a great impact of temperature on the hysteresis and the final capacitance of the remembered state [34].

The aim of the present paper is to describe how the magnitude of voltage and magnetic field affects the memory effect in liquid crystal 5CB doped with aerosil nanoparticles and a combination of aerosil and goethite nanoparticles. The magnitude of external stimuli is one of the key parameters which can enhance the performance of the system. In addition, the possibility of using the composite as a base for a multilevel memory device, which can be erased by heating to a specific temperature, is described.

2. Material and methods

The composite of 5CB liquid crystal and aerosil (SiO_2) nanoparticles was prepared by mixing aerosil R812 nanoparticles (Evonik Industries, Essen, Germany) in powder form with 5CB liquid crystal in isotropic phase. The mean diameter of the aerosil nanoparticles was 7 nm. The mixture was placed to a sonicator for 1 hour to break up aggregates and achieve a homogeneous sample. The composite of 5CB and a combination of aerosil and goethite nanoparticles was prepared by adding goethite nanoparticles in powder form to a part of the composite containing aerosil nanoparticles, followed by an additional 1-hour sonification. The goethite nanoparticles had a lath-like shape, with a length of 350±100 nm, a width of 25±7 nm, and a thickness of 10±5 nm. TEM images of the goethite nanoparticles were published in our previous article [34]. The concentration of aerosil nanoparticles was 1 wt% and the concentration of goethite nanoparticles was 0.04 wt% (the volume fraction 10^{-4}).

The samples were filled in cells with a thickness of 50 μm (MWAT, Warsaw, Poland) for performing the experiments. These cells consisted of two glass plates with indium-tin-oxide layers and polyimide layers. The indium-tin-oxide layers serve as transparent electrodes and polyimide layers are responsible for a planar orientation of liquid crystal in a cell. The size of the electrode area was 5 mm × 5 mm. The samples were sonicated before filling the cells to ensure a homogeneous sample without aggregates. The cells were filled at room temperature (23 °C), while the sample was in the nematic phase. Voltage and magnetic field were applied to the samples in cells perpendicular to the cell surface (see the geometry of experiment in Fig. 1). A sinusoidal voltage with a frequency of 1 kHz was applied with an oscilloscope (TiePie Handyscope HS5), and a magnetic field was applied with an electromagnet (GMW, model 5403). The capacitance of the samples was acquired by oscilloscope (TiePie Handyscope HS5) and a capacitance bridge with high precision (Andeen Hagerling) when voltage and magnetic field were applied, respectively. Before performing each measurement the composites were heated to the isotropic phase using a temperature controller (Linkam TMS93). The heating rate was set to 5 °C/min to heat the sample to approximately 2 °C below the target temperature and then a rate of 0.1 °C/min was used to achieve the desired target temperature. When the target temperature was reached, the sample was rapidly cooled by water-cooling system set to 25 °C.

3. Results

Voltage and magnetic field with various maximal values were applied to the composites to explore how the magnitude of voltage and magnetic field affect the electromechanical and magnetomechanical memory effects, i.e. the hysteresis in capacitance-voltage and capacitance-magnetic field dependencies. The measurements were performed as follows. The cell was filled with the sample in the nematic

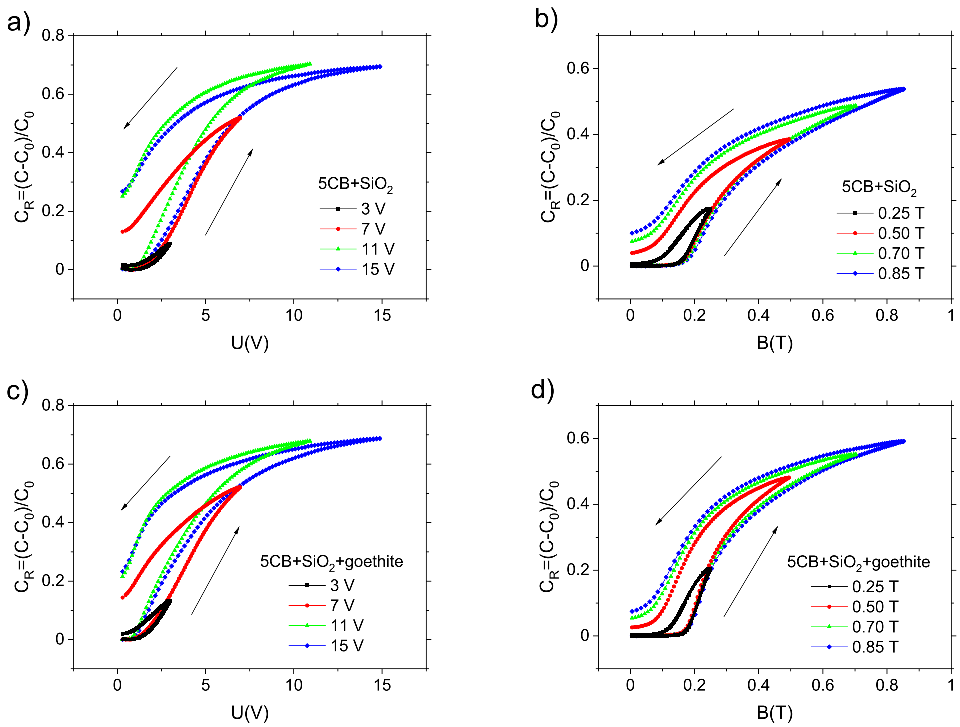

Fig. 2. The influence of the magnitude of voltage and magnetic field on the memory effect. Figure a) and b) show the effect of the magnitude of the electric and magnetic fields on the composite of 5CB with aerosil nanoparticles, while c) and d) demonstrate the effect of the magnitude of the electric and magnetic fields on the composite of 5CB with a mixture of aerosil and goethite nanoparticles. The arrows denote the direction of the measurement. Voltage and magnetic field in the legend label the maximal values applied to the composites (V_{max}, B_{max}).

phase. After filling the cell and before each experiment it was heated to the isotropic phase - to 36 °C for measuring voltage dependencies and to 41 °C for measuring magnetic field dependencies. These temperatures were selected because after heating to these temperatures the hysteresis in capacitance-voltage and capacitance-magnetic field dependencies were the most significant and concurrently reproducible within the same sample [34]. Then the sample was cooled back to 25 °C, at which the sample is in the nematic phase. At 25 °C the composites were far below the transition temperature from the isotropic to the nematic phase T_{IN}. The T_{IN} of the studied composites decreased by less then 0.1 °C from pure 5CB liquid crystal, which has a T_{IN}~33.7 °C [34]. The voltage or magnetic field was applied to the sample in the nematic phase while capacitance was measured. The voltage or magnetic field was slowly increased up to the maximal values, V_{max} and B_{max}, and subsequently slowly decreased. During increasing and decreasing of the voltage or magnetic field, the capacitance was monitored and hysteresis was observed. After the initial experiment was finished, the sample was heated to the same temperature in isotropic phase again, and a second measurement this time using larger values of V_{max} and B_{max} was performed. The same procedure was applied to the composite with only aerosil nanoparticles and to the composite with aerosil and goethite nanoparticles.

The capacitance-voltage and capacitance-magnetic field dependencies are shown in Fig. 2. C_R denotes the reduced capacitance given by $C_R = (C - C_0)/C_0$, where C is the actual capacitance, and C_0 is the minimum capacitance. Fig. 2 a) and b) show the results for the composite with only aerosil nanoparticles, while Fig. 2 c) and d) present the results for the composite with a combination of aerosil and goethite nanoparticles. An increase in V_{max} and B_{max} cause an increase in the final capacitance $C_{R final}$ (the reduced capacitance after switching off voltage and magnetic field), which is the most critical parameter for a memory device as the final capacitance represents the remembered state. The application of an electric or magnetic field causes the reorientation of liquid crystal molecules towards the direction of the applied voltage or magnetic field. This occurs due to the positive dielectric permittivity anisotropy and positive diamagnetic susceptibility anisotropy of 5CB. As the field is applied, the capacitance increase, as the angle between the long axis of the molecule with respect to the cell surface is increasing. A larger field cause reorientation by a larger angle, and as result a reorientation by a larger angle is retained due to the presence of the aerosil nanoparticles network.

To explore the influence of the magnetic field on the electromechanical memory effect, the following experiments were performed. Before conducting the initial measurement, the sample was heated to 36 °C, i.e. to the temperature at which the hysteresis for the electromechanical effect was the largest, and then cooled back to 25 °C. When temperature was stabilized at 25 °C, the first measurement was conducted. The voltage was slowly increased and decreased while capacitance was monitored (Fig. 3 b) black curve). Afterwards, a magnetic field (0.85 T) was applied to the composite. After switching off the magnetic field, the voltage was slowly increased and decreased again while capacitance was measured (red curve in Fig. 3 b)). Since the magnetic field of 0.85 T has a weaker effect compared to the voltage of 15 V, it did not significantly impact the capacitance. Before conducting the last measurement, the sample was heated to 36 °C and cooled back to 25 °C. The process erased the effect, and the identical curve resembles the one observed after the first heating (see blue curve overlaying the black curve in Fig. 3 b)). Similarly the effect of voltage on the magnetomechanical effect was explored. The composite was heated to 41 °C, the temperature at which the hysteresis for the magnetomechanical effect was the largest. Then it was cooled to 25 °C, the temperature at which the measurements were performed. At 25 °C, the magnetic field was applied, and hysteresis in the capacitance-magnetic field dependency was observed (black curve in Fig. 3 a)). When the measurement finished, en electric voltage (15 V) was applied. After switching off the voltage, the magnetic field was slowly increased and decreased again, and the capacitance was moni-

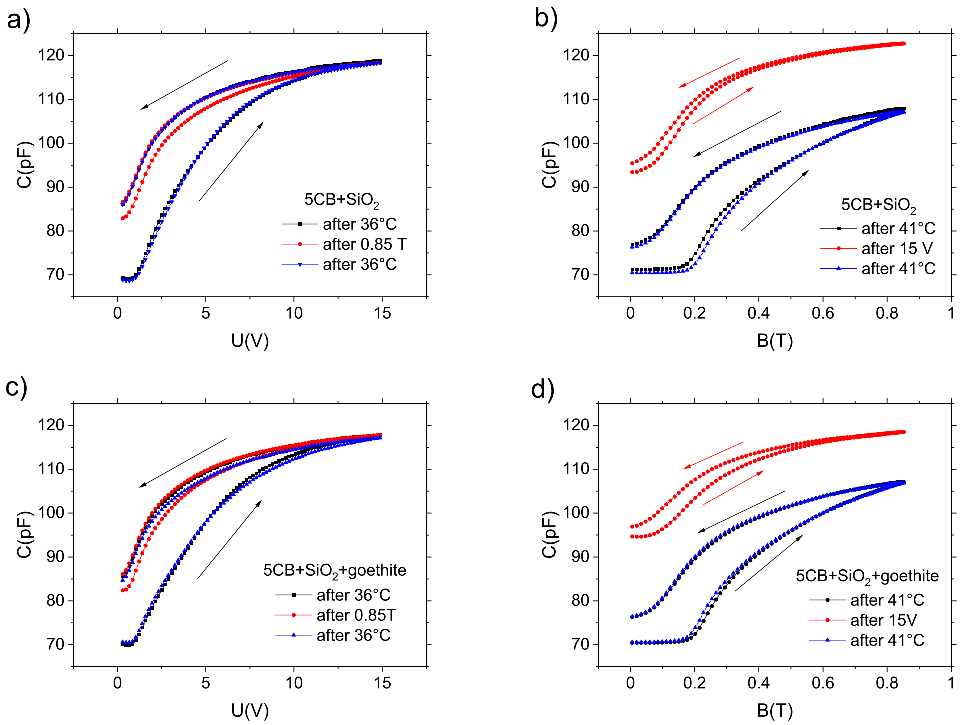

Fig. 3. The effect of the magnetic field on the electromechanical memory effect and the effect of voltage on the magnetomechanical memory effect for a) and b) 5CB doped with aerosil nanoparticles, while c) and d) illustrate 5CB doped with aerosil and goethite nanoparticles. The arrows denote the direction of the measurement.

tored (red curve in Fig. 3 a)). The capacitance of the composite after applying 15 V was higher as the voltage caused additional reorientation of the nematic domains, which remain stable after removing the voltage. The magnetic field of 0.85 T in the first measurement, which is the maximal magnetic field that could be reached in our apparatus, was not high enough to induce the same degree of reorientation as the 15 V. Subsequent heating of the composite to 41 °C and cooling to 25 °C led to erasing the memory resulting in a curve identical to that observed after the first heating (see Fig. 3 a) - blue curve overlay the black curve). The same set of experiments were performed on the composite containing aerosil and goethite nanoparticles (see Fig. 3 c) and d)) with the qualitatively similar results.

The presented results provide a base for preparing a multilevel memory device. Various states can be achieved either by applying various magnitudes of voltage or magnetic field or by a combination of electric and magnetic fields. The remembered state can be erased by heating the composite. Heating the composites to the same temperature results in the same capacitance-voltage or capacitance-magnetic field dependencies, i.e. with setting the identical conditions, the results can be easily reproduced. Goethite magnetic particles can reduce or increase the threshold magnetic field of Fréedericksz transition [37]. The potential enhanced performance of the goethite nanoparticles might be reached by cooling the composite containing both aerosil and goethite nanoparticles in a magnetic field. We supposed the addition of goethite nanoparticles can influence the memory effect significantly. However, the addition of goethite nanoparticles did not notably alter the composite response to the magnetic field.

4. Conclusions

The effect of the voltage and magnetic field magnitudes on the memory effect in 5CB liquid crystal doped with aerosil nanoparticles and a combination of aerosil nanoparticles and magnetic goethite nanoparticles was studied. Increased voltage and magnetic field led to a higher capacitance of the system remembered after removing the voltage and magnetic field. The higher capacitance reflects the higher degree of liquid crystal molecule reorientation, which remain stable due to the hydrogen bonds among the aerosil nanoparticles and between aerosil nanoparticles and liquid crystal molecules. Based on the results the principle of a multilevel non-volatile memory device was proposed. Several states can be reached by the application of voltage or magnetic field with various magnitudes or by a combination of voltage and magnetic field. The remembered state can be erased by heating the composite to the selected temperature and then cooling it back to the working temperature of the device. Maintaining a consistent heating temperature provides good reproducibility of the results. The memory effect is present at reasonably low or moderate voltages (in order of Volts), but for practical applications, the magnetic field intensity needs to be reduced.

CRediT authorship contribution statement

Katarína Kónyová: Writing – original draft, Methodology, Investigation, Conceptualization. **Dmytro Miakota:** Visualization, Formal analysis. **Veronika Lacková:** Writing – review & editing, Visualization, Conceptualization. **Shie-Chang Jeng:** Resources. **Dorota Węgłowska:** Resources. **Filippo Agresti:** Resources. **Peter Kopčanský:** Project administration, Funding acquisition. **Natália Tomašovičová:** Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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