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Determination of phase transition from nematic to isotropic state in carbon nano-balls' doped nematic liquid crystals by electrical conductivity-dielectric measurements

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Abstract

The phase transition in carbon nano-balls' doped nematic liquid crystals has been investigated by electrical conductivity-dielectric measurements. The results of electrical conductivity and dielectric constant as a function temperature and frequency indicate that a phase transition, which is of the first order, takes place from nematic state and isotropic state. The phase transition for E7 and E7/C60 samples takes place at 326 and 321 K temperatures, respectively. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

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Composites based on liquid crystals (LCs) have been extensively investigated because of their unique electro and magneto-optic properties and novel display applications. Typical examples of the systems are polymer dispersed liquid crystals [1,2]. Fullerene doped LC systems were extensively studied due to their positive effects in possible technological appli-

cations [3,4]. Photorefractive like reorientation is a famous molecular mechanism in which photo excited dopants bring about the formation of space-charge complexes and their induced fields force the molecules to reorient by enhancing the photoconductivity. Effect of fullerene on reorientation process is explained by this approach indeed [3–5]. Molecular orientation of LC molecules determine the electro-optical behavior of the system and because laser molecule interaction causes molecules to reorient in these systems, our aim was to examine electro-optical measurements so that we could demonstrate the molecular reorientation

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Fig. 1. Chemical formulas of: (a) nematic host, E7; (b) fullerene, C60.

based changes in capacitance, impedance and dielectric coefficients and refractive index dispersion. Such works have been performed on dye doped and polymer doped LC previously [6–8].

Liquid crystals (LCs) are highly nonlinear optical materials due to their susceptible property activating under even relatively low optical fields and are a class of materials exhibiting thermodynamically stable but only partially ordered phases. Each mesophase is described by its own order parameter, such as a second rank tensor for orientational order (nematic) phase or a two-component tensor vector for one-dimensional translational order (smectic phase) [9]. The dielectric and electrical conductivity measurements provide about phase transition in the investigated temperature.

The aim of our study is to investigate the determination of the phase transition by dielectric-electrical measurements.

2. Experimental

2.1. The preparation of the nematic liquid crystal cell

High purity fullerene (C60) was purchased from Dr. Andrei Ponomarev (St. Petersburg, Russia). Measurement cells were made up of two glass slides separated by Mylar sheets having $\sim 8.5\,\mu m$ thickness. Before the construction of the cells, indium tin oxide (ITO) coated glass substrates were spin coated with polyvinyl alcohol (PVA) at 2000 rpm and they were cured at 50 °C for ~ 2 hours. The thickness of the coating is ~ 100 nm and these coating layers were exposed to surface treatment of unidirectional rubbing with velvet in order to obtain preliminary molecu-

lar orientation. The ultimate form of the constructed cell is planar with 2 degree rubbing tilt. The volume fraction of C60 in the E7 sample studied was taken as 1% wt/wt. Fullerene (C60) was dissolved within toluene and toluene fraction was evaporated to eliminate fullerene powders, which later mixed to LC under the reinforcement of ultrasonic effect. Once the sample cell was filled with homogenous LC/C60 content, which was prepared in the ultrasonic mixing machine, it is almost impossible to face with any colloids in such a thin medium. Even if such fullerene groups were suspected to be formed in the sample, it was going to be easily understood by color spots on the cell. Color of sample cell is still homogenous and there is no any different color spot on it after more than 1 years. Chemical formulas of fullerene and nematic host are depicted in Fig. 1.

2.2. Measurements

Electrical conductivity and dielectric constant measurements were performed by impedance analyzer. HP 16089B Kelvin clip leads were used with the Hawlett–Packard (4194A) impedance/gain-phase analyzer at various spot frequencies 10², 10³, 10⁴ and 10⁵ Hz as a function of temperature range (285–345 K). A closed-cycle helium cryostat system and Lakeshore 340 model temperature controller were employed in the system, whereby it allowed to scan the temperature with a rate of about 2 K/min. Data were recorded in a PC under isothermal conditions and temperature was stabilized with an accuracy better than 0.05 K. GaAlAs diode sensor having an accuracy of 0.01 K was utilized in the determination of temperature values.

3. Results and discussion

3.1. Electrical conductivity

Fig. 2(a) and (b) show electrical conductivity dependence of the temperature at different frequencies. The conductivity of the E7 and E7/C60 samples increases with increasing temperature, which provides an evidence of negative temperature coefficient of resistance (NTCR). In the investigated temperature range, the conductivity plots exhibit one anomaly. This anomaly in curves corresponds probably to a structural phase transition, which transforms liquid crystal from the nematic to isotropic (N–I) phase. The anomaly is more prominent for the high frequencies. The conductivity curves are defined by three regions (I, II and III). But, region II is the transition temperature region. The electrical conductivity regions (I and III) obey to the well-known equation,

$$\sigma = \sigma_0 \exp(-\Delta E/kT),\tag{1}$$

where σ_0 is the pre-exponential factor and ΔE is the activation energy. These regions are fitted by to the relation and total conductivity is written as

$$\sigma = \sigma_{01} \exp(-\Delta E_{\rm N}/kT) + \sigma_{02} \exp(-\Delta E_{\rm I}/kT), \quad (2)$$

where σ_{01} and σ_{02} are constants and $\Delta E_{\rm N}$ for nematic phase and $\Delta E_{\rm I}$ for isotropic phase are the activation energies for each region, as shown in Fig. 2(a) and (b). The activation energy values were obtained from

the slope of $\ln \sigma - 1000/T$ curves. The frequency dependence of activation energy is shown in Fig. 3. The activation energies are decreased with increasing frequency. The increase in frequency of the applied field enhances the electronic jumps between atomic sites. Consequently, the activation energies are decreased with frequency. The abrupt change in the activation energy from $\Delta E_{\rm N}$ to $\Delta E_{\rm I}$ is due to phase transition in the structure.

3.2. The variation of dielectric constant with temperature

The plots of dielectric constant ε versus temperature at 10², 10³, 10⁴ and 10⁵ Hz frequencies for the samples are shown in Fig. 4(a) and (b). From the curves, we observe an anomaly attributed to the phase transition. The dielectric constant increases up to a temperature, but afterwards, a sharp increase in the dielectric constant is observed. This is in agreement with contribution of the conductivity. It is evaluated that the anomaly observed in figures can be attributed to the presence of the nematic-isotropic phase transition. The increase of the temperature in the nematic phase and its rapid change at the transition to the isotropic state shows that this process is strongly affected by the nematic potential. The phase transition is accompanied by the rapid jump in the ε values. Thus, it should be stressed that the dielectric constant values change suddenly, this might be related to continuous

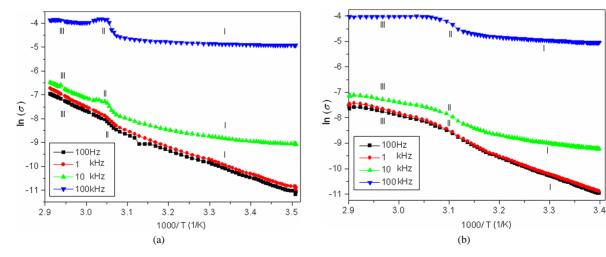


Fig. 2. Electrical conductivity dependence of the temperature at different frequencies (a) E7 sample; (b) E7/C60 sample.

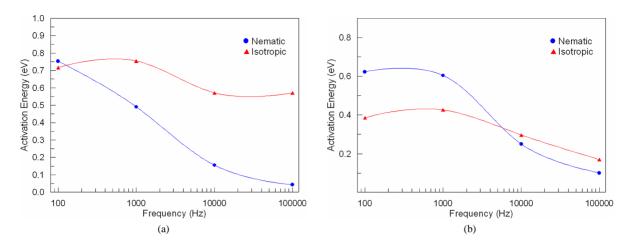


Fig. 3. Activation energy dependence of the frequency (a) E7 sample; (b) E7/C60 sample.

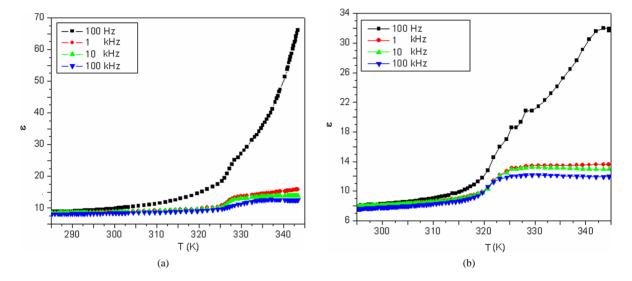


Fig. 4. The plots of dielectric constant ε versus temperature at different frequencies (a) E7 sample; (b) E7/C60 sample.

release of the reorientation of the dipole groups. In order to determine phase transition temperature, $d\varepsilon/dT$ vs. T plots are plotted. Fig. 5(a) and (b) show the plots of the $d\varepsilon/dT$ vs. temperature. $d\varepsilon/dT$ values increase with temperature and goes through a maximum and then decrease with temperature. This maximum is attributed to the phase transition and its position corresponds to phase transition temperature, T_c . The position of the peaks did not change by different frequencies, i.e., the phase transition temperature does not change with increasing frequency. This suggests that the disordering effect caused by the random occu-

pancy of the particles in the structure is not sufficient to cause the relaxor nematic phase. Thus, it is evaluated that the phase transition is a diffuse phase transition rather a relaxor phase transition. The T_c values for E7 and E7/C60 samples were found as 326 and 321 K, respectively. The phase transition temperature was suppressed by 5 K for volume fraction studied. It is evaluated that this is a very strong disordering effect.

The variation of the dielectric constant with temperature is described by the Curie–Weiss relation,

$$\varepsilon = \frac{C}{T - T_0},\tag{3}$$

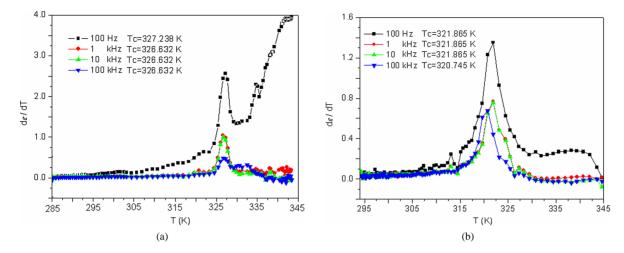


Fig. 5. $d\varepsilon/dT$ vs. T plots at different frequencies (a) E7 sample; (b) E7/C60 sample.

where C is the Curie–Weiss constant and T_0 is the Curie–Weiss temperature. The order of the phase transition can be determined by this equation. If T_0 is equal to phase transition temperature, T_c , the phase transition is of the first order. When $T_0 \neq T_c$ it is of second order type [10]. The Curie–Weiss constants and temperatures were obtained from the slope and intercept of $1/\varepsilon$ versus temperature plots. The obtained values suggest that phase is of the first order.

4. Conclusions

The phase transition in carbon nano-balls' doped nematic liquid crystals has been investigated by electrical conductivity-dielectric measurements. The electrical conductivity plots of the samples exhibit one anomaly which corresponds probably to a structural phase transition from the nematic to isotropic phase. T_c values for E7 and E7/C60 samples are respectively obtained as 326 and 321 K. The activation energies of the electrical conductivity of the samples are decreased with frequency of electric field applied. The dielectric constant of the samples at different frequencies changes with temperature. The variation of dielectric constant with temperature was investigated by the Curie-Weiss relation. Curie-Weiss constants and temperatures suggest that phase transition is of the first order. Consequently, the primary goal of this work is on the materials science by bringing about a hybrid LC composite, which is appropriate to several applications

from switches to holographic storage mediums. Actually most of these applications require the optimization of novel systems in terms of their electro-optical properties and dependency of these properties on external effects as well as phase transitions.

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