



# High optical nonlinearity of nematic liquid crystal doped with graphene oxide



Nima Dalir<sup>a</sup>, Soheila Javadian<sup>a,\*</sup>, Zahra Dehghani<sup>b</sup>

<sup>a</sup> Department of Physical Chemistry, Faculty of Basic Science, Tarbiat Modares University, P.O. Box 14115-175, Tehran, Islamic Republic of Iran

<sup>b</sup> Department of Physics, University of Neyshabur, Neyshabur, P. O. Box 9319774400, Islamic Republic of Iran

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## ABSTRACT

Colloidal suspensions of nematic liquid crystal (NLC) doped with graphene oxide (GO) were prepared and their third-order nonlinear optical (NLO) response  $\chi^{(3)}$ , nonlinear refractive index (NLR), and nonlinear absorption coefficient (NLA) were investigated using Z-scan technique employing He: Ne laser at 632.8 nm. GO doped system exhibits a significantly different nonlinear optical response compared to pure NLC and the highest nonlinearity was observed at the system including 0.75% GO.

The results of GO doped NLC show that the external field changes the NL parameter are dependent upon the electrical field direction since it affects the director axis,  $\vec{n}$ , compared to their initial orientation. The external field is applied perpendicular to the cell surface. The  $\chi^{(3)}$  change value was measured as the applied electrical field varies. The mentioned studies were carried out without and under the electric field effect using different AC voltages. The value of NLO, NLA and  $\chi^{(3)}$  in present of electric field were more significant compare without of electric field. Additionally, the optical limiting (OL) behavior of colloidal suspensions was investigated. The obtained results provide new useful information about the NLO properties of NLC's nanocomposite in particular to design optical limiting devices and also other photonic applications.

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## 1. Introduction

Liquid crystals (LCs) are widely used in modern devices such as flat panel displays, light modulators, and sensors [1,2]. Their notable properties include high optical quality and strong nonlinearity. The nonlinear index coefficient,  $n_2$ , is on the order of  $10^{-4}$  to  $10^{-3}$   $\text{cm}^2 \cdot \text{W}^{-1}$  which results in a widespread application to optical image processing, optical limiting, or recording of dynamic holographic gratings [3–5]. NLCs show high nonlinear optical responses since they are strongly anisotropic and their high correlated molecular reorientation. Their refractive index changes drastically due to collective director reorientation toward external field [6].

The nonlinear optical phenomena stem from the coupling between two or more photons resulting in a photon with a frequency which is a linear combination of the original photon frequencies. The mentioned phenomena are responsible for several significant improvements in laser-based technologies [7,8], most of which rely on phase matching of intense electromagnetic fields in extended bulk crystalline media to enable efficient frequency conversion. High availability of nanostructured materials offered by modern nanofabrication techniques has drawn the attention of many researchers in mastering nonlinear optics

on sub wavelength scales. It is noteworthy that nonlinear optical nanomaterials have a variety of applications including optical microscopy [9,10], biological imaging/detection [11,12], and signal conversion in nanoscale photonic devices [13–16].

Doping of LCs with  $\text{C}_{60}$  or dyes (i.e., Methyl-Red [17]) has resulted in intensifying of the nonlinear optical response by some orders of magnitude. Doping LCs with different inorganic nanoparticles such as semiconductor [18,19], ferroelectric [20,21], ferromagnetic [5,22], or noble metal [23,24] significantly enhances the electro-optic and magneto-optic properties of host LC materials [25,26]. The nonlinear optical effects of gold nanoparticles doped to LC have been investigated which indicated that gold nanoparticles can improve photorefractive response [5,27]. Although notable works have studied the nonlinearity of optical properties, the optical properties of LCs doped with carbonaceous nanostructures has been rarely studied which are presented here. The optical limiting (OL) properties were investigated in the visible spectral range and the switching times of the CNTs-doped NLC display element have been registered in microsecond range [28]. Additionally, an extremely large electro-optically induced photorefractive effect was reported in NLC crystal doped with single-walled carbon nanotubes (SWCNT) and fullerene  $\text{C}_{60}$  [29,30]. A wide range of nematic behavior was observed in eutectic of pentyl-cyanobiphenyl 0.55% (5CB) and heptyl-cyanobiphenyl 0.45% (7CB) or  $\text{E}_{5\text{CN}7}$ , obtained by our research group, compared to pure counterparts [31,32]. Also, the thermodynamic

\* Corresponding author.

E-mail address: [javadian\\_s@modares.ac.ir](mailto:javadian_s@modares.ac.ir) (S. Javadian).

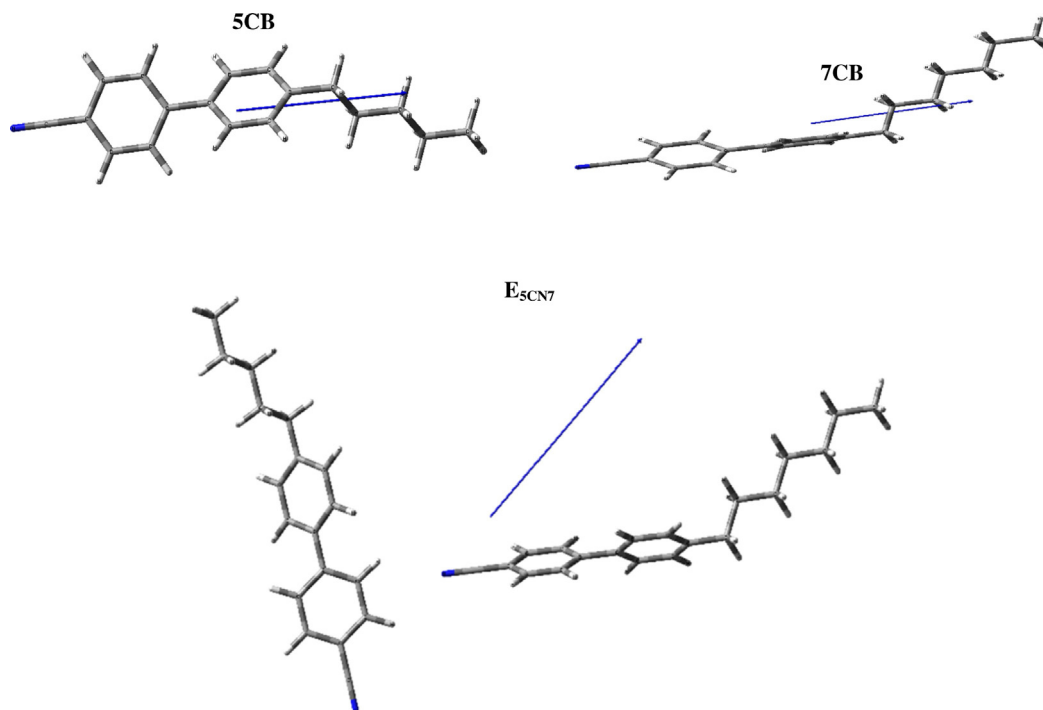


Fig 1. Geometry and optimized structure of 7CB, 5CB and E5CN7 nematic liquid crystal.

properties of mentioned mixture were studied at different ratios of GO showing that 0.75 w/w ratio of GO results in highest phase transition entropy [33].

In this work, third-order nonlinear optical (NLO) response  $\chi^{(3)}$ , nonlinear refractive index (NLR), and nonlinear absorption coefficient (NLA) of the system including E<sub>5CN7</sub> NLC doped with a graphene oxide (GO) at different GO ratios was investigated by using Z-scan technique of different voltages.

## 2. Experimental methods

### 2.1. Preparation of NLC + GO suspensions

A set of samples with different GO ratios doped to E<sub>5CN7</sub> (as shown in Fig. 1) NLC (Table 1) were prepared. Conductivity of all samples in Table 1 was determined by using multimeter (B&K 5429) and an automatic data acquisition system. GO was dispersed in CH<sub>3</sub>Cl, sonicated under continuous stirring until the solvent evaporated. Suspensions fabricated in this way were then put into a photorefractive NLC cell. The photorefractive NLC cells consisted of two glass substrates covered with a conductive layer of indium tin oxide (ITO, Scheme 1). The thickness of the gap between substrates, L, was 12 μm. Two substrates were coated with polyvinyl alcohol (PVA). Then, the cells were filled with a pure NLC or with a NLC + GO suspension by the capillary effect.

**Table 1**  
Conductivity and alignment stability of the cells containing liquid crystals or NLC + GO suspensions.

Sample	Conductivity [μS]	Cell's alignment
E <sub>5CN7</sub>	0.474	Homeotropic
E <sub>5CN7</sub> @ 0.50% GO	0.243	Homeotropic
E <sub>5CN7</sub> @ 0.75% GO	0.099	Homeotropic
E <sub>5CN7</sub> @ 1% GO	0.010	Homeotropic

### 2.2. Z-scan set up

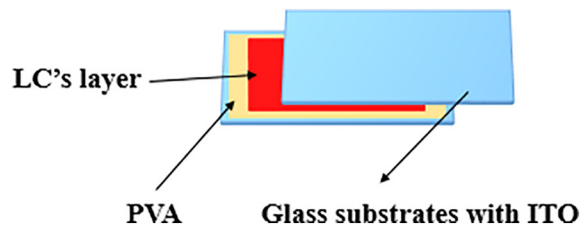
The third-order NLO properties were measured by the Z-scan technique implemented using a linear polarized GW He: Ne laser at a wavelength of 632.8 nm with a power of 50 mW as the light source as shown in (Scheme 2). The power of the laser was reduced to 10 mW by adequate filters. It is important to choose a laser of low intensity to avoid the reorientation of molecules. In the Z-scan method, a converging lens with a focal length of 8 cm was used to focus the incident laser beam on the NLC cell held in a 12 μm quartz cuvette and translated across the focal plane in the beam direction. The order parameter, S, is the transmittance of the aperture in the linear regime and the internal power (P<sub>in</sub>) and beam radius (ω<sub>0</sub>) were obtained 0.37, 10 mW and 44 μm, respectively. The Z-scan curve and the transmittance as a function of the sample position can be obtained.

### 2.3. Z-scan theories on nonlinear process

The Z-scan method includes the decomposition of the complex electric field into a Gaussian beams. Depending upon the intensity of light source, the nonlinear refraction (NLR) and nonlinear absorption (NLA) are expressed as follows [34,35]:

$$n(I) = n_0 + n_2 I \quad (1)$$

$$\alpha(I) = \alpha_0 + \beta I \quad (2)$$



Scheme 1. Photorefractive NLC's@ GO cell rubbed by PVA.

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