

Electrodynamics study and surface conductance of fullerenes in liquid crystal medium

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ARTICLE INFO

Keywords:

Liquid crystal
Fullerenes
Surface conductance
Zeta potential
Dukhin number

ABSTRACT

Investigation of electrodynamic parameters are interesting not only from pure scientific point of view, but they are promising parameters for optimizing the quality of electronic displays. The experiment on fullerenes-nematic liquid crystal (LC) colloid was performed in planar aligned liquid crystal cell with in-plane electric field. The super-fluorinated nematic LC mixture (MJ951160), was purchased from Merck and used as received. It has typical trifluorophenyl-2 (TFP2) structure. The fullerenes in liquid crystal were attracted in the direction of positive electrode. Different electrodynamic parameters such as zeta potential, charge, surface conductance and Dukhin number were determined only by experimentally measuring the velocity of colloidal fullerenes in liquid crystal and utilizing the theory of dielectrophoretic and electrophoretic forces.

1. Introduction

The most challenging and unique properties of condensed matter physics are to control its electric field induced phoresis by means of appropriately chosen external electric field [1–10].

Colloidal particles suspended in nematic liquid crystal is known by the term nematic colloids [1]. In ac or dc electric field induced phoresis, the particles velocity is reported to vary linearly [2,3] and nonlinearly [4–10]. The advantage of using ac over dc is that the ac field does not cause electrochemical deposition [1].

Colloidal particle in host liquid crystal (LC) medium when they are exposed to ac electric field finds numerous application in electrophoretic displays [3,11,12], genetic [13] biomedicine [14] lab on chip devices [15] and colloidal segregation [16]. Electrophoretic parameters such as zeta potential, charge on colloidal particles and mobility are required to be tailored to get the desired performance of electrophoretic display. Among them, zeta potential is one of the most important parameters and is defined as electrical potential between the fixed and portable stages in digressive progression of the fluid as for the surface [17]. The zeta

potential gives fundamental data about the electrochemical properties of colloidal particles in a suspension. This local electrical potential is determined tentatively by utilizing electrokinetic estimations including electrophoretic velocity, value of Clausius-Mossotti function and gradient of electric field. Surface conductance is defined as conductance of the colloidal particles around its surface [4]. Surface conductance is generally the conductance tangential to the surface charge and arises from the concentration of additional counterions in the electrical double layer (EDL) region near the solid-liquid interface. Surface conductance play the major role in the conduction current for colloidal particles. Another important parameter, known as “Dukhin number” can be defined as the ratio of the conductance of EDL and conductance of host dielectric medium [7,18,19]. Dukhin number describes contribution of surface conductance to polarize the thin double layer under an electric field and can be written as

$$Du = \frac{K^{\sigma}}{\sigma_1 r} \quad (1)$$

where K^{σ} is the surface conductance of double layer, and σ_1 is the

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<https://doi.org/10.1016/j.elstat.2025.104084>

Received 24 December 2024; Received in revised form 22 April 2025; Accepted 23 April 2025

Available online 13 May 2025

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conductivity of LC and r is the radius of the colloid particle.

Under an electric field, when particles are suspended in any host dielectric medium then there are two types of forces which cause the motion of colloidal particle: one is an electrophoretic force (EP) and another is dielectrophoretic force (DEP). In an EP, particle responds actively to the polarity of the applied electric field. In EP, the movement of the molecules is dictated by net positive/negative electric charges and they migrate towards one polar (negative/positive) electrode. The motion of polarized particles in dielectric host medium due to non-uniform electric field is known as dielectrophoretic (DEP) force. In DEP, the initiated movement is controlled by net dielectric strength of the colloidal particles. Bert and Smet first reported significant high value of zeta potential due effect to of DEP force [11,12]. In previous article, we investigated the different electro-dynamical parameters of colloidal carbon nanotubes in nematic LC [4]. In this paper, different electro-dynamical parameters such as zeta potential, charge, surface conductance and Dukhin number were determined only by experimentally measuring the velocity of colloidal fullerenes in LC and utilizing the theory of DEP and EP forces. In order to calculate the electro-dynamical parameters, the required other physical parameters such as viscosity, dielectric permittivity and conductivity of the LC and fullerenes were taken by the datasheet provided by Merck Company and our previously published paper.

2. Experimental

The fullerenes (C60s) of one weight percentage were doped into nematic LC. For fabricating a LC cell, an electrode-patterned glass substrate was first spin-coated to a thickness of 800 Å with homogenous alignment layer (AL-16139) from Japan Synthetic Rubber Co. The electrode width of interdigitated electrodes was 10 µm and the distance between them was 30 µm. The homogeneous alignment along the field direction was performed by rubbing through cotton. Another glass plate having no electrode, the rubbing process in antiparallel direction was performed. Then, we assembled the cell in the form of capacitor and the cell gap (d) of 9 µm was maintained with the help of polymer (NOA65) and plastic balls. Superfluorinated nematic LC was purchased from Merck Company (birefringence $\Delta n = 0.088$ at $\lambda = 589$ nm, dielectric anisotropy $\Delta\epsilon = +7.4$ at 1 kHz, flow viscosity = 18 mm²/s at 20 °C) was injected at room temperature by the capillary action. Optical polarizing microscopy (Nikon DXM1200) was used to observe the fabricated test cells. The sine wave voltage of different fields and frequencies were employed to study the electrokinetic behavior of C60s in nematic LC. The C60s velocity was calculated by observing their textures dynamics under 30 frames/second.

3. Results and discussion

The optical textures of the C60s in host LC medium are shown in Fig. 1. In the absence of an electric field, the C60s remains stable in LC medium. In the region of low frequencies of less than 5 Hz, C60s oscillate in between the electrodes and its amplitude decreases with increasing frequency. The amplitude of translation motion of C60s at different frequencies is given in Table 1. At the frequency of 3 Hz, the amplitude of translation motion becomes very small and unmeasurable and eventually the oscillation of C60s stop at the frequency of 5 Hz.

When, we apply DC field, C60s move towards the positive electrode which might be due induced polarization on colloidal C60s along the direction of electric field (DEP force) or net negative charge on colloidal C60s (EP force) [4,5]. Electro-dynamical behavior C60s in nematic LC indicates that C60s may act like para-electric materials having positive dielectric susceptibility ($\chi_e = \epsilon_r - 1$; for vacuum $\chi_e = 0$). Therefore, the relative dielectric permittivity (ϵ_r) of the C60s would be greater than one.

An expression for zeta potential (ζ) and charge (q) by incorporating both EP and DEP forces [4,5] may be expressed as

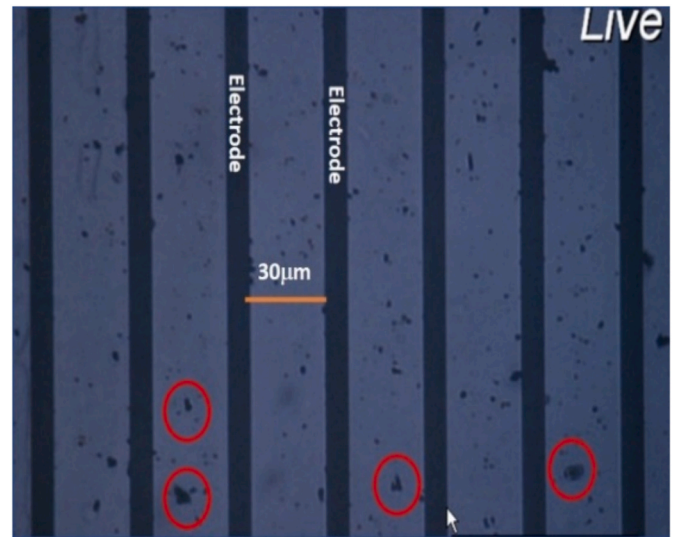


Fig. 1. Fullerene's oscillation between the electrodes at 1 Hz and 0.38 V/µm of electric field. Some of the fullerenes are shown in the circle.

Table 1

Amplitude of translation motion of C60s at different frequencies.

Frequency (Hz)	Amplitude (µm)
1.0	8.3
1.4	3.9
2.2	2.0
2.6	1.6

$$\zeta = r^2 \text{Re}\{K(\epsilon_2^*, \epsilon_1^*)\} \nabla E - \frac{3\eta v}{2\epsilon_1 E} \quad (2a)$$

$$q = \frac{1}{E} [4\pi\epsilon_1 r^3 E \text{Re}\{K(\epsilon_2^*, \epsilon_1^*)\} \nabla E - 6\pi\eta r v] \quad (2b)$$

Where r is the radius of C60s, E is an applied electric field, v is the velocity of the C60s between the electrodes and η is the viscosity of the LC. The ϵ_1 and ϵ_2 are the dielectric permittivity of LC and C60s respectively. We measured the velocity (v) of the C60s by observing their translation motion between the electrodes at 1.0 Hz of frequency and an applied electric field (E) of 0.38 V/µm and its value was 300.0 µm/Sec and corresponding radius of (r) of colloidal C60s was 2.23×10^{-6} m. At ambient temperature, the internal fluid viscosity (η) and dielectric permittivity (ϵ_1) of LC were 19.3×10^{-3} kgm⁻¹s⁻¹ and 5.4×10^{-11} F/m respectively. The Clausius-Mossotti function $\text{Re}\{K(\epsilon_2^*, \epsilon_1^*)\}$ used in the above equations can be expanded as

$$\text{Re}\{K(\epsilon_2^*, \epsilon_1^*)\} = \frac{\omega^2(\epsilon_2 - \epsilon_1)(\epsilon_2 + 2\epsilon_1) + (\sigma_2 - \sigma_1)(\sigma_2 + 2\sigma_1)}{\omega^2(\epsilon_2 + 2\epsilon_1)^2 + (\sigma_2 + 2\sigma_1)^2} \quad (3a)$$

. At low frequency region the value of Clausius-Mossotti function tends to be a function of conductivity only

$$\text{Re}[K(\omega)] = \frac{\sigma_2 - \sigma_1}{\sigma_2 + 2\sigma_1} (\omega \rightarrow 0) \quad (3b)$$

where σ_1 and σ_2 are the conductivities of LC and C60s respectively. The conductivity of the C60s ($\sim 10^{-4}$ S m⁻¹) [42] was very large than that of the LC host medium ($\sim 10^{-9}$ S m⁻¹). The value of Clausius-Mossotti function estimated from equation (3) was found to be 1.0. We also estimated the electric field gradient (∇E) by simulation method by keeping the same initial and boundary condition as that of experimental cell. Commercially available software "LCD Master" (Shintech, Japan)

was used for simulation. The simulated value of electric field gradient was $3.27 \times 10^{10} \text{V/m}^2$. The value of ζ and q were determined by substituting the value of gradient of E (∇E), v , E , r , η , and $\text{Re}\{K(\epsilon_2^*, \epsilon_1^*)\}$ in Eq. (2a) and Eq. (2b). The calculated ζ and q were -160.9 mV and $-2.4 \times 10^{-16} \text{C}$ respectively.

It has been found that the velocity of C60s depends upon the cubic power ($v = \mu^3 E^3$) of the applied electric field [5] as plotted in Fig. 2. The calculated value of nonlinear mobility coefficient μ^3 was equal to $4.0 \times 10^{-21} \text{m}^4/\text{V}^3 \text{s}$. The dimensionless parameter Dukhin number ($Du = K^\sigma / (r \cdot K_{LC})$) was calculated using the following equation [10,20].

$$\mu^3 = -r^2 \frac{e}{k_B T} \frac{\epsilon_1}{\eta} \frac{Du[23 + Du(269 + 464Du)]}{252(1 + 2Du)^3} \quad (4)$$

The Du values calculated from Eq. (4) were -0.48 , $-0.07-0.06i$ and $-0.07 + 0.06i$. As Du must be a real number, therefore ignoring the complex numbers and taking the unique real, the value of $Du = -0.48$. Substituting the value of Du , r and σ_1 in Eq. (1), the K^σ of electrical double layer was $-1.07 \times 10^{-15} \text{S}$. For glass and electrolyte solution, the surface conductance value is reported in the range of 10^{-10} to 10^{-7}S [21–25]. The reported value of surface conductance of CNTs/LC colloidal solution was of the order of 10^{-17}S . The magnitude of calculated value of surface conductance of C60s/LC colloid is far less than glass and electrolyte solution.

The value of charge density ($\frac{q}{4\pi r^2}$) in case of C60s in LC medium was $-3.8 \times 10^{-4} \mu\text{C}/\text{cm}^2$, whereas in case of CNTs, the value of charge density was $9.8 \times 10^{-5} \mu\text{C}/\text{cm}^2$, due increased charge density in C60s/LC colloids, the magnitude of surface conductance of C60s in LC was obtained larger as compared to CNTs in LC medium. The surface conductance K^σ can also be expressed as

$$K^\sigma = K_s^i + K_s^d \quad (5)$$

where K_s^i and K_s^d are conductance of stern and diffusion layers. The conductance in these layers is due to motion of charge. The surface conductance of C60s was also determined from the following relation

$$K_s^i = \left(\frac{q}{4\pi r^2} \right) \mu \quad (6)$$

Typically, the diffusion layer conductance is 100 times less that of Stern layer. Therefore, for Eq. $K^\sigma = K_s^i + K_s^d$ the conductance of electrical double layer can be approximated as $K_s^i \approx K^\sigma$. The value calculated K^σ from Eq. (6) was $-2.3 \times 10^{-15} \text{S}$ which is almost same value as estimated from Eq. (1). The different electrodynamic parameters of C60s in LC obtained from above analysis are listed in Table 2

It is important to mention here that all the electrodynamic values determined for C60s were found to be negative causing the main reasons to move towards the positive electrode, whereas in case of CNTs [4], all the electrodynamic parameters were reported positive [4].

4. Conclusions

We have investigated the electrodynamic of C60s/LC colloidal solution. Our experimental observation shows that C60s in LC are attracted towards the positive electrode, which might be due to induced polarization along the direction of electric field (DEP force) or negative net charge on the colloidal C60s. The estimated value of zeta potential, Dukhin number, charge, and surface conductance of electrical double layer were found to be negative.

CRediT authorship contribution statement

Nidhi Pandey: Formal analysis, Data curation. **Anoop K. Srivastava:** Writing – original draft, Supervision, Investigation, Conceptualization. **Sudhanshu Pandey:** Supervision, Formal analysis. **Sudhir**

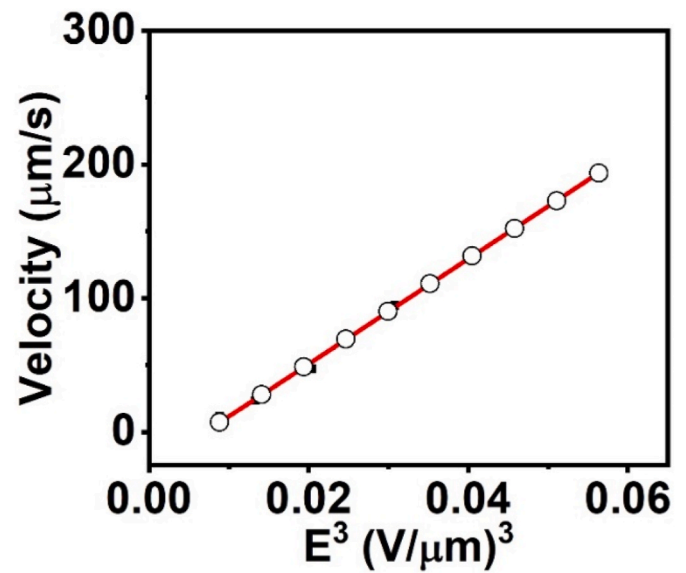


Fig. 2. Velocity variation of C60s in the LC medium with the cubic power of the applied electric field at the frequency of 1 Hz.

Table 2

Different electrodynamic parameters of C60s in LC.

Zeta Potential (ζ)	Charge (q)	Mobility (μ)	Dukhin No. (Du)	Charge Density ($\frac{q}{4\pi r^2}$)	Surface Conductance (K^σ)
-160.9 mV	$-2.4 \times 10^{-16} \text{C}$	$1.6 \times 10^{-7} \text{m}^4/\text{V}^3 \text{s}$	-0.5	$-3.8 \times 10^{-4} \mu\text{C}/\text{cm}^2$	$-1.1 \times 10^{-15} \text{S}$

Prakash: Formal analysis. **Anil Kumar:** Formal analysis. **Ramesh Manda:** Formal analysis. **Seung Hee Lee:** Writing – review & editing, Resources, Formal analysis.

Declaration of competing interest

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

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.elstat.2025.104084>.

Data availability

No data was used for the research described in the article.

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