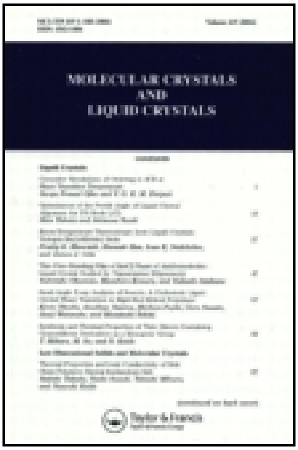

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Influence of Small Spherical Particles on the Spatial Director Distribution and Light Scattering in a Nematic Cell

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Director spatial distribution near a small spherical particle is calculated for the case of weak director anchoring with the particle surface. The differential cross-section of the light scattering by the director inhomogenuity near the particles is calculated in dipole approximation. We use the Percus - Yevick approximation for hard spheres to account for interference effects. It is studied the influence of the external ac electric field on the director profile and magnitude of light scattering cross-section.

Keywords: filled liquid crystals; light scattering, theory

I. INTRODUCTION

Heterogeneous liquid crystal (LC) systems are widely studied during the last time because of their current (possible) use in different technologies for displays applications. Some representatives of such systems are: polymer dispersed liquid crystals (PDLC)[1], filled liquid crystals[2], ferronematics - dispersion of magnetic particles in LC matrix[3], porous glass dispersed liquid crystals[4,5], dispersion of small water droplets in a nematic liquid crystal[6], dispersion of fine plate-like clay mineral in a nematic liquid crystal[7], dispersion of ultra-fine particles of carbon black and titanium dioxide in ferroelectric liquid crystals[8]. Many of heterogeneous LC systems possess strong electronically controlled light scattering effects. This permits the development of new types of display which have some advantages in comparison with traditional LC displays.

This paper is the first in the series of theoretical studies of the properties of filled liquid crystals. Even bearing in mind the widespread applications of other heterogeneous LCs, the technical applications of filled LCs seem to be extremely promising[9–18]. In particular the previous studies of the scattering type displays based on the filled LC have demonstrated their essential additional potential advantages. For instance, their contrast is independent of the viewing angle, thermostability is extremely high (which is only restricted by the thermostability of LC), there is no need for index matching and the display fabrication technology is rather simple.

Filled LCs consist of a low concentration of highly dispersed pyrogenic silica (aerosil), typically 2-3 volume percent, in a nematic LC. The primary particles form larger aggregates via hydrogen bonding[10,19]. These silica particles, of diameter $100\dot{A}$, cause the appearance of multiple defects in the LC orientational structure, which give rise to strong light scattering. The transformation and disappearance of a good proportion of these defects in an electric field causes an initially opaque medium to become transparent. When the electric field is removed the system may either return to the initial high scattering state - this is known as reversible response, or it may (partially or completely) remain transparent, in which case the phenomenon is known as memory response.

When mixed with LC, the silica aggregates may also form a three dimensional network and thus divide the LC into domains of typical dimensions in the range of $100-300nm$[10]. The actual structure of the filled LC strongly depends on the surface properties of silica particles and their concentration in a suspension and will be considered in the next papers.

In this paper we consider the liquid crystal matrix filled with small spherical silica particles which remain in the form of colloidal suspension[20]. These solid particles produce the LC director inhomogeneities around inclusions that cause intensive light scattering. Applying voltage reorients director along the electric field and sample may become transparent. Due to the small particles concentration there is practically nor overlapping of the LC director distortion areas caused by the neighbouring particles nor the influence of the particles themselves on the optical properties of a cell.

This paper is organised as follows. In subsection II A from the condition of minimum of the total free energy we write down the Euler - Lagrange equation and boundary condition to it. In subsection II B we find director field distortion caused by a small solid spherical particle in a nematic LC matrix in the switched off state. Then in subsection II C we study the influence of an applied electric field on this director distribution. Next in section III we discuss light scattering by the system under consideration. Finally in section IV we make some concluding remarks. Some technical results are relegated to the appendices.

II. DIRECTOR DISTRIBUTION IN LC MATRIX CONTAINING SMALL SPHERICAL PARTICLE

A. Free energy minimization

We consider a small spherical particle of radius R in a nematic LC subject to homeotropic boundary conditions at the particle surface. We also suppose that an external homogeneous electric field is applied to the sample. The total free energy of the system is then

$$F = F_{el} + F_E + F_S \tag{1}$$

$$\begin{aligned} F_{el} &= \frac{1}{2}K \int \left[(\operatorname{div}\mathbf{n})^2 + (\operatorname{rot}\mathbf{n})^2\right] dV - \\ &\quad -\frac{1}{2}K_{24} \int \operatorname{div}\left(\mathbf{n}\operatorname{div}\mathbf{n} + \mathbf{n}\times\operatorname{rot}\mathbf{n}\right) dV, \\ F_E &= -\frac{\varepsilon_a}{8\pi}\int (\mathbf{n}\cdot\mathbf{E})^2\, dV, \\ F_S &= \frac{1}{2}W \int (\mathbf{n}\cdot\mathbf{e}_r)^2\, dS. \end{aligned}$$

The first term in F describes the bulk elastic free energy, the second term is responsible for the contribution to the free due to the applied external electric field, and the last term describes the anchoring of LC director at the particle surface in the form of Rapini potential[21]. Here K, K_{24} are the LC elastic constants, ε_a is LC dielectric constant anisotropy at the frequency of applied ac voltage, W is so-called anchoring energy, and $\mathbf{e}_r$ is the unit vector directed along the droplet radius.

Minimization of the total free energy subject to the condition $\mathbf{n}^2 = 1$ leads to the following Euler - Lagrange equation (see Appendix A)

$$K\Delta\mathbf{n}+\frac{1}{4\pi}\varepsilon_a\mathbf{E}\,(\mathbf{n}\cdot\mathbf{E})+\lambda\mathbf{n}=0, \quad \mathbf{r}\in\mathbf{V} \tag{2}$$

and boundary condition

$$\begin{aligned}(K-K_{24})\{\operatorname{rot}\mathbf{n}\times\mathbf{e}_r+\mathbf{e}_r\operatorname{div}\mathbf{n}\}+&\\ +K_{24}(\mathbf{e}_r\cdot\nabla)\,\mathbf{n}+W\mathbf{e}_r\,(\mathbf{n}\cdot\mathbf{e}_r)-\mu\mathbf{n} &= 0, \qquad \mathbf{r}\in\mathbf{S}.\end{aligned} \tag{3}$$

Here λ and μ are Lagrange multipliers.

In mathematical sense this problem is similar to the problem of director configuration within spherical LC droplet in a polymer matrix under a weak homeotropic anchoring that we considered in[22]. In the general case of an arbitrary value of the anchoring energy W equation (2) subject to the boundary condition (3) can be solved only numerically. However if the anchoring energy is sufficiently small ($\xi = \frac{WR}{K} \ll 1$, as we shall see later), then approximate solution can be found in the analytical form.

B. Weak anchoring solution

Let the anchoring energy W is sufficiently small so that the director distribution $\mathbf{n}\,(\mathbf{r})$ may be presented in the form

$$\mathbf{n}=\mathbf{n}_0+\delta\mathbf{n}\,,$$

where $\mathbf{n}_0$ is the director orientation in the homogeneous cell without particle and $\delta\mathbf{n} \ll 1$.

In the spherical coordinate system with Oz axis directed along the vector $\mathbf{n}_0$ we have

$$\delta\mathbf{n} = \{\Psi(\theta, r)\cos\phi, \Psi(\theta, r)\sin\phi, 0\}, \quad \Psi(\theta, r) \ll 1,$$

r, θ, ϕ are the spherical coordinates.

Substituting this into the boundary condition (3) and linearizing it with respect to $\delta\mathbf{n}$ one get the following equation

$$\left(R\frac{d\Psi(\theta, r)}{dr} + (1 - k_{24})\Psi(\theta, r) + \frac{1}{2}\xi\sin 2\theta\right)_{r=R} = 0, \tag{4}$$

where $k_{24} = \frac{K_{24}}{K}$, $\xi = \frac{WR}{K}$.

The form of boundary condition (4) gives us the clue to seek solution of (2) as

$$\Psi(\theta, r) = \sin 2\theta f(r).$$

On substituting this into (2) we have the following equation for $f(r)$

$$r^2 f'' + 2rf' - f\left(6 + \frac{\varepsilon_a E^2}{4\pi K}r^2\right) = 0, \tag{5}$$

where E is the electric field in homogeneous LC cell.

First consider the case when the external electric field is absent, $E = 0$. Then solution to the equation (5) is

$$f(r) = C_1 r^{-3}.$$

The unknown constant C_1 we find from the boundary condition (4), and finally get:

$$\Psi(\theta, r) = \xi\frac{\sin 2\theta}{4\left(1 + \frac{1}{2}k_{24}\right)}\left(\frac{R}{r}\right)^3. \tag{6}$$

From (6) it is seen that $\Psi(\theta, r)$ is small if the anchoring parameter $\xi \ll 1$. For typical values of $R \sim 100$ - $1000\dot{A}$, $K \sim 10^{-6} dyn$ this condition is valid at $W \ll 10^{-1} erg/cm^2$. It is also seen that R is the characteristic length of $\delta\mathbf{n}$ decrease.

C. Filled liquid crystal in an electric field

Now we switch on the external electric field. It is convenient to introduce the non-dimensional parameter

$$x = \frac{r}{l_E},$$

where $l_E = \sqrt{\frac{4\pi K}{\varepsilon_a E^2}}$ is so called electric field correlation length[23]. Then the equation (5) takes the form

$$x^2 f'' + 2xf' - f\left(6 + x^2\right) = 0\,.$$

The solution to this equation can be written in the form

$$f(x) = C_2\sqrt{\frac{\pi}{2x}}K_{5/2}(x)\,,$$

where $K_{5/2}(x)$ is the modified spherical Bessel's function[24]

$$K_{5/2}(x) = \sqrt{\frac{\pi}{2x}}\exp(-x)\left(1 + \frac{3}{x} + \frac{3}{x^2}\right)\,.$$

Respectively

$$\begin{aligned} \delta n_x(r,\theta,\phi) &= C_2\sqrt{\frac{\pi}{2}}\left(\frac{r}{l_E}\right)^{-1/2} K_{5/2}\left(\frac{r}{l_E}\right)\sin 2\theta\cos\phi\,, \\ \delta n_y(r,\theta,\phi) &= C_2\sqrt{\frac{\pi}{2}}\left(\frac{r}{l_E}\right)^{-1/2} K_{5/2}\left(\frac{r}{l_E}\right)\sin 2\theta\sin\phi\,. \end{aligned} \quad (7)$$

In contrast to the power law of director deviation spatial decrease in a switched off state now $\delta\mathbf{n}$ goes to zero exponentially with the characteristic length scale l_E. Unknown constant C_2 in (7) we find from the boundary condition (4):

$$\begin{aligned} C_2 &= \xi\frac{1}{\pi}\frac{\exp x_R}{\left(1+\frac{3}{x_R}+\frac{3}{x_R^2}\right)+\left(\frac{1}{x_R}+\frac{6}{x_R^2}+\frac{9}{x_R^3}\right)-(1-k_{24})\left(\frac{1}{x_R}+\frac{3}{x_R^2}+\frac{3}{x_R^3}\right)} \\ &= \xi\frac{1}{\pi}\frac{x_R^3\exp x_R}{x_R^3+3x_R^2+6x_R+6+k_{24}\left(x_R^2+3x_R+3\right)}\,, \end{aligned}$$

where

$$x_R = \frac{R}{l_E}\,.$$

From the comparing of expressions (6) and (7) for the director deviation $\delta\mathbf{n}$ it is seen that threshold value of the applied ac voltage to switch the behaviour of the system from OFF to ON state can be estimated from the condition

$$l_E = R,$$

so

$$E_{th} = \left(\frac{4\pi K}{\varepsilon_a R^2}\right)^{1/2}$$

In the case of strong anchoring when the anchoring parameter $\xi \gg 1$ director deviation $\delta\mathbf{n}$ cannot be considered small, and the director distribution near the particle surface is either close to $\mathbf{e}_r$ with a disclination ring[25,26]or to a hyperbolic hedgehog with point defect[6]. Influence of an applied electric field onto director configuration near a spherical particle at strong anchoring is a subject of the next paper.

III. LIGHT SCATTERING

We confine our interest to the problems in which the single scattering problem can be solved within the dipole approximation[23]. The light scattering differential cross-section can be written in the form

$$\frac{d\sigma}{d\Omega} = \left|\frac{k^2}{4\pi}\mathbf{e}_\alpha \cdot \hat{\epsilon}\left(\mathbf{q}_s\right) \cdot \mathbf{e}_\mu\right|^2, \tag{8}$$

where $\mathbf{e}_\alpha$, $\mathbf{e}_\mu$ are the unit vectors of the polarization of the incoming and outgoing waves with wave vectors $\mathbf{k}$ and $\mathbf{k}'$ respectively, $\mathbf{q}_s = \mathbf{k}' - \mathbf{k}$ is the scattering wave vector,

$$\hat{\epsilon}(\mathbf{q}_s) = \int \left(\delta\epsilon(\mathbf{r}) - I\right) \exp\left\{-i\mathbf{q}_s{\cdot}\mathbf{r}\right\} d\mathbf{r} \tag{9}$$

is the Fourier transform of $(\delta\hat{\epsilon}\,(\mathbf{r}) - I)$.

Now we recall the relationship between liquid crystalline and optical properties. Locally at a point $\mathbf{r}$ the liquid crystalline optical properties are defined in terms of the director $\mathbf{n}(\mathbf{r})$ by the dielectric constant $\hat{\epsilon}(\mathbf{r})$ at the relevant optical frequency, where:

$$\epsilon_{ij}(\mathbf{r}) = \epsilon_{\perp}\delta_{ij} + \epsilon_a n_i n_j.$$

here $\epsilon_{\|}$ and $\epsilon_{\perp}$ are parallel and perpendicular components of the dielectric constant.

Supposing that concentration of particles is small enough, so that the regions of director distortion caused by different particles do not overlap we have

$$\epsilon_{ij}(\mathbf{r}) \equiv \epsilon_{ij}^0 + \sum_m \delta\epsilon_{ij}(\mathbf{r} - \mathbf{r}_m)\, v(a - |\mathbf{r} - \mathbf{r}_m|),$$

$$\begin{aligned} \epsilon_{ij}^0 &= \delta_{ij}(\epsilon_{\perp} + \epsilon_a \delta_{iz}), \\ v(x) &= \begin{cases} 1 & x \geq 0 \\ 0 & x < 0 \end{cases} \end{aligned}$$

$$\begin{aligned} \delta\epsilon_{ij}(\mathbf{r} - \mathbf{r}_m) &= \epsilon_a (\delta_{iz}\delta_{jx}\delta n_x(\mathbf{r} - \mathbf{r}_m) + \delta_{jz}\delta_{ix}\delta n_x(\mathbf{r} - \mathbf{r}_m) \\ &\quad + \delta_{iz}\delta_{jy}\delta n_y(\mathbf{r} - \mathbf{r}_m) + \delta_{jz}\delta_{iy}\delta n_y(\mathbf{r} - \mathbf{r}_m)), \end{aligned}$$

here we direct Oz axis of laboratory coordinate system along $\mathbf{n}_0$, a is an averaged distance between particles, and summation is taken over all N particles.

Substituting this into (8) we get

$$\frac{d\sigma}{d\Omega} = \frac{k^4}{16\pi^2} |e_\alpha \cdot \delta\epsilon_{\alpha\mu}(\mathbf{q}_s) \cdot e_\mu|^2 \left(N + \sum_{m \neq m'} \exp\{-i\mathbf{q}_s \cdot (\mathbf{r}_m - \mathbf{r}_{m'})\} \right) \quad (10)$$

Introducing the positional pair-correlation function[27] $g(\mathbf{r})$, we can write

$$\frac{1}{N} \sum_{m \neq m'} \exp\{-i\mathbf{q}_s \cdot (\mathbf{r}_m - \mathbf{r}_{m'})\} = \frac{N}{V} \int g(\mathbf{r}') \exp\{-i\mathbf{q}_s \cdot \mathbf{r}'\}\, d\mathbf{r}', \quad (11)$$

where $\frac{N}{V}$ in the number of particles per unit volume. The Fourier transform of $g(\mathbf{r})$ given by eq.(11) has a strong peak at $\mathbf{q}_s \simeq 0$ that in fact does not contribute to the scattering and usually is subtracted[28, 29]. Therefore we can rewrite eq.(10) as

$$\frac{d\sigma}{d\Omega}=\frac{k^4}{16\pi^2}N\left|e_\alpha\cdot\delta\epsilon_{\alpha\mu}\left(\mathbf{q}_s\right)\cdot e_\mu\right|^2 S\left(\mathbf{q}_s\right), \tag{12}$$

where $S\left(\mathbf{q}_s\right)$ is so-called structure factor. In liquid theory much effort is extended in the determination of $S\left(\mathbf{q}\right)$. In what follows we use the Percus-Yevick approximation[30]for direct correlation function to calculate $S\left(\mathbf{q}\right)$ and give relevant formulas in the Appendix B.

If the incident light is not polarized one has to average eq.(12) over the polarization, and finally for incoming light with the wave vector $\mathbf{k}\parallel Oz$ we get

$$\begin{aligned}\frac{d\sigma}{d\Omega} &= \frac{k^4}{16\pi^2}\frac{1}{2}N\left|\delta\epsilon_{zx}\left(\mathbf{q}_s\right)+\delta\epsilon_{zy}\left(\mathbf{q}_s\right)\right|^2 S\left(\mathbf{q}_s\right)\\ &= \frac{k^4}{32\pi^2}\epsilon_a^2 N\left|n_x\left(\mathbf{q}_s\right)+n_y\left(\mathbf{q}_s\right)\right|^2 S\left(\mathbf{q}_s\right)\ ,\end{aligned}$$

where $n_x\left(\mathbf{q}_s\right), n_y\left(\mathbf{q}_s\right)$ are the Fourier transforms of $\delta n_x\left(\mathbf{r}\right)$ and $\delta n_y\left(\mathbf{r}\right)$ which are determined by the expressions of the section IIA.

Further for simplicity we suppose that scattering occurs in the ZOY plane and scattering angle between $\mathbf{k}$ and $\mathbf{k}'$ is ϑ, then

$$\mathbf{q}_s=\left(0, k\sin\vartheta, -2k\sin^2\frac{\vartheta}{2}\right).$$

A. Light scattering in the switched off state

In the switched off state from (6) we obtain

$$\begin{aligned}n_x\left(\mathbf{q}\right) &= \int\delta n_x(\mathbf{r})\exp\left\{-i\mathbf{q}\cdot\mathbf{r}\right\}d\mathbf{r}\\ &= \frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)}\int_1^\infty dx\int_0^\pi d\theta\sin\theta\sin 2\theta\frac{\exp\left\{-iq_zRx\cos\theta\right\}}{x}\\ &\quad\times\int_0^{2\pi}\cos\phi\exp\left\{-iq_yRx\sin\theta\sin\phi\right\}d\phi\\ &= 0,\end{aligned}$$

and

$$\begin{aligned} n_y(\mathbf{q}) &= \int \delta n_y(\mathbf{r}) \exp\{-i\mathbf{q}\cdot\mathbf{r}\}\, d\mathbf{r} \qquad (13) \\ &= \frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)} \int_1^\infty dx \int_0^\pi d\theta \sin\theta \sin 2\theta \frac{\exp\{-iq_z Rx\cos\theta\}}{x} \\ &\times \int_0^{2\pi} d\phi \sin\phi \exp\{-iq_y Rx \sin\theta \sin\phi\}\ . \end{aligned}$$

These integrals can be evaluated only numerically, however in the limit of small angle light scattering when $qR \ll 1$ one obtains the following analytical expression for $n_y(\mathbf{q})$ (see Appendix C)

$$n_y(\mathbf{q}) \approx \frac{2\pi}{3} \frac{\xi R^3}{\left(1+\frac{1}{2}k_{24}\right)} \cot\frac{\vartheta}{2},$$

and thus

$$\frac{d\sigma}{d\Omega} \frac{1}{N\pi R^2 \xi^2 \epsilon_a^2} \simeq \frac{1}{72\pi} \left(\frac{1}{1+\frac{1}{2}k_{24}}\right)^2 (kR)^4 \cot^2\frac{\vartheta}{2} S\left(2kR\sin\frac{\vartheta}{2}\right).$$

For typical values of $R \preceq 1000\dot{A}$ and $\lambda = 0.5\mu$ this approximation is valid for scattering angle $\vartheta < 0.1$.

In Fig.1 we show the numerical results for the light scattering differential cross-section, $10^4 \frac{d\sigma}{d\Omega} \frac{1}{N\pi R^2 \epsilon_a^2 \xi^2}$ in the switched OFF state for different values of particle concentration η. Fig. 2. presents the influence on light scattering of the parameter kR (ratio between the particle size and light wave length $kR = \frac{2\pi R}{\lambda}$).

B. Light scattering in the switched on state

In the switched on state from (7) we obtain

$$\begin{aligned} n_x(\mathbf{q}) &= \int \delta n_x(\mathbf{r}) \exp\{-i\mathbf{q}\cdot\mathbf{r}\}\, d\mathbf{r} \\ &= \frac{l_E^3 \pi}{2} C_2 \int_{R/l_E}^\infty dx \exp(-x)\left(x+3+\frac{3}{x}\right) \\ &\times \int_0^\pi d\theta \sin\theta \sin 2\theta \exp\{-il_E x q_z \cos\theta\} \\ &\times \int_0^{2\pi} d\phi \exp\{-il_E x q_y \sin\theta \sin\phi\} \cos\phi = 0, \end{aligned}$$

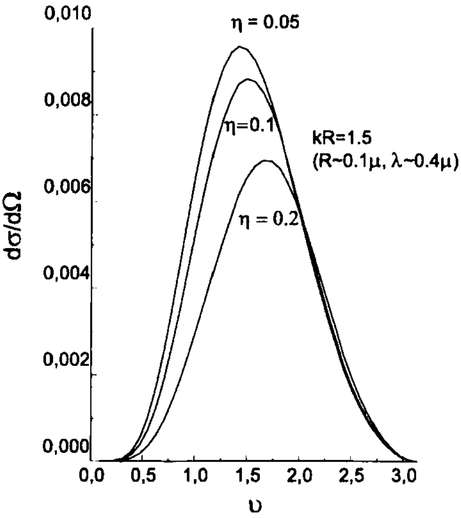

FIGURE 1 See text for explanation

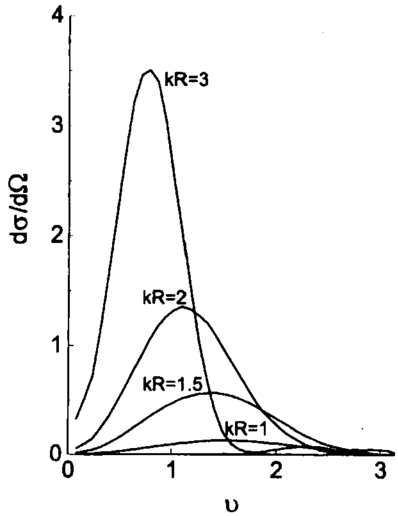

FIGURE 2 See text for explanation

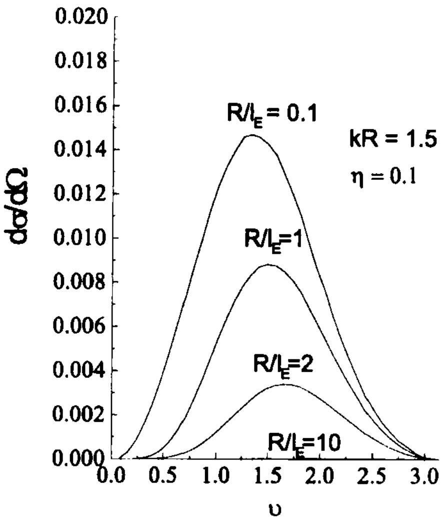

FIGURE 3 See text for explanation

$$n_y(\mathbf{q}) = \int \delta n_y(\mathbf{r}) \exp\{-i\mathbf{q}\cdot\mathbf{r}\}\, d\mathbf{r} \tag{14}$$

$$= \frac{l_E^3\pi}{2} C_2 \int_{R/l_E}^{\infty} dx \exp(-x)\left(x + 3 + \frac{3}{x}\right)$$

$$\times \int_0^{\pi} d\theta \sin\theta \sin 2\theta \exp\{-il_E x q_z \cos\theta\} \tag{15}$$

$$\times \int_0^{2\pi} d\phi \exp\{-il_E x q_y \sin\theta \sin\phi\} \sin\phi\,. \tag{16}$$

If we are interested in the small angle light scattering when $ql_E \ll 1$, then (see Appendix C)

$$n_y(\mathbf{q}) \approx -\frac{32}{15} e^{-\frac{R}{l_E}} \left(R^3 + 15l_E^3 + 15l_E^2 R + 6l_E R^2\right) l_E^2 \pi^2 k^2 C_2 \sin\vartheta \sin^2\frac{\vartheta}{2}$$

and respectively

$$\frac{d\sigma}{d\Omega}\frac{1}{N\pi R^2 \epsilon_a^2 \xi^2} \simeq \frac{2}{\pi}\left(\frac{4}{15}\frac{x_R^3 + 15 + 15x_R + 6x_R^2}{x_R^3 + 3x_R^2 + 6x_R + 6 + k_{24}(x_R^2 + 3x_R + 3)} x_R^2\right)^2$$

$$\times (kl_E)^8 \left(\sin\vartheta \sin^2\frac{\vartheta}{2}\right)^2 S\left(2kR\sin\frac{\vartheta}{2}\right).$$

For typical experimental parameters: $\varepsilon_a = 10$, $K = 10^{-6} dyn$, cell thickness $L = 10\mu$, applied voltage $U \succeq 10$ *volt* we have $l_E < 1\mu$.

For $ql_E \gtrsim 1$ integrals in (14) can be evaluated only numerically. In Fig.3 we show the numerical results for the light scattering differential cross-section per particle, $10^4 \frac{d\sigma}{d\Omega} \frac{1}{N\pi R^2 \epsilon_a^2 \xi^2}$, in the switched ON state at different values of the applied voltage (l_E).

IV. CONCLUSIONS

Under the weak homeotropic director anchoring with the particle surface the director distortion in the cell volume decreases with distance $\mathbf{r}$ to the particle centre as r^{-3}. Characteristic length of director significant change equals approximately the particle's radius R. The external ac voltage changes the law of director decrease to exponential one with the characteristic length of decreasing l_E. The switching of the director configuration from the switched OFF state to the switched ON state occurs at the value of electric field $E \geq E_{th} = \left(\frac{4\pi K}{\varepsilon_a R^2}\right)^{1/2}$

Characteristic multiplier showing the decreasing of light scattering cross-section value upon switching on of the ac electric field ($E \gg E_{th}$) has the order of magnitude $\left(\frac{l_E^2}{\lambda R}\right)^2$

In the future papers in this series we shall be addressing the problems of light scattering in diffraction regime and at higher particles concentration when silica network is formed.

APPENDIX A: FREE ENERGY VARIATION

To find director field $\mathbf{n}(\mathbf{r})$ that minimizes the total free energy (1) subject to the constrain $\mathbf{n}^2 = 1$ in the cell volume and at particle surface one has to evaluate the first variation of the functional:

$$\Phi = \frac{1}{2} K \int \left[(\operatorname{div} \mathbf{n})^2 + (\operatorname{rot} \mathbf{n})^2 \right] dV -$$

$$-\frac{1}{2}K_{24}\int \operatorname{div}(\mathbf{n}\operatorname{div}\mathbf{n} + \mathbf{n}\times\operatorname{rot}\mathbf{n})\,dV$$
$$-\frac{\epsilon_a}{8\pi}\int(\mathbf{n}\cdot\mathbf{E})^2\,dV - \frac{1}{2}\int\lambda\mathbf{n}^2 dV +$$
$$+\frac{1}{2}W\int(\mathbf{n}\cdot\mathbf{e}_r)^2\,dS - \frac{1}{2}\int\mu\mathbf{n}^2 dS.$$

Here λ and μ are Lagrange multipliers.

$$\begin{aligned}\delta\Phi &= K\int[\operatorname{div}\mathbf{n}\operatorname{div}\delta\mathbf{n} + \operatorname{rot}\mathbf{n}\cdot\operatorname{rot}\delta\mathbf{n}]\,dV\\ &-\frac{1}{2}K_{24}\int\operatorname{div}(\delta\mathbf{n}\operatorname{div}\mathbf{n} + \delta\mathbf{n}\times\operatorname{rot}\mathbf{n})\,dV\\ &-\frac{1}{2}K_{24}\int\operatorname{div}(\mathbf{n}\operatorname{div}\delta\mathbf{n} + \mathbf{n}\times\operatorname{rot}\delta\mathbf{n})\,dV\\ &-\frac{\epsilon_a}{4\pi}\int(\mathbf{n}\cdot\mathbf{E})(\delta\mathbf{n}\cdot\mathbf{E})\,dV\\ &-\int\lambda\mathbf{n}\cdot\delta\mathbf{n}dV + W\int(\mathbf{n}\cdot\mathbf{e}_r)(\delta\mathbf{n}\cdot\mathbf{e}_r)\,dS - \int\mu\mathbf{n}\cdot\delta\mathbf{n}dS\end{aligned}$$

It is easy to check the following

$$\begin{aligned}\operatorname{div}\mathbf{a}\operatorname{div}\mathbf{b} &= \operatorname{div}(\mathbf{a}\operatorname{div}\mathbf{b}) - \mathbf{a}\cdot\nabla\operatorname{div}\mathbf{b},\\ \operatorname{rot}\mathbf{a}\cdot\operatorname{rot}\mathbf{b} &= \operatorname{div}(\mathbf{a}\times\operatorname{rot}\mathbf{b}) + \mathbf{a}\cdot(\nabla\operatorname{div}\mathbf{b} - \Delta\mathbf{b})\end{aligned}$$

Applying these formulas and Gauss theorem we get

$$\begin{aligned}\delta\Phi &= -K\int[\delta\mathbf{n}\cdot\Delta\mathbf{n} + \operatorname{rot}\mathbf{n}\cdot\operatorname{rot}\delta\mathbf{n}]\,dV\\ &+K\int\mathbf{e}\cdot(\delta\mathbf{n}\times\operatorname{rot}\mathbf{n})\,dS + K\int(\mathbf{e}\cdot\delta\mathbf{n})\operatorname{div}\mathbf{n}dS\\ &-\frac{1}{2}K_{24}\int\mathbf{e}\cdot(\delta\mathbf{n}\operatorname{div}\mathbf{n} + \mathbf{n}\operatorname{div}\delta\mathbf{n} + \delta\mathbf{n}\times\operatorname{rot}\mathbf{n} + \mathbf{n}\times\operatorname{rot}\delta\mathbf{n})\,dS\\ &-\frac{\epsilon_a}{4\pi}\int(\mathbf{n}\cdot\mathbf{E})(\delta\mathbf{n}\cdot\mathbf{E})\,dV\\ &-\int\lambda\mathbf{n}\cdot\delta\mathbf{n}dV + W\int(\mathbf{n}\cdot\mathbf{e}_r)(\delta\mathbf{n}\cdot\mathbf{e}_r)\,dS - \int\mu\mathbf{n}\cdot\delta\mathbf{n}dS.\end{aligned}$$

Here $\mathbf{e}$ is the external normal to the particle surface which in the case of spherical particle coincides with $\mathbf{e}_r$.

Using the fact that $\int_{\sum} \mathbf{e} \cdot \operatorname{rot} \mathbf{b} dS = 0$ if $\sum$ is a closed surface (this follows from Stokes theorem) and equality

$$\mathbf{n} \operatorname{div} \delta\mathbf{n} + \mathbf{n} \times \operatorname{rot} \delta\mathbf{n} = \operatorname{rot}(\mathbf{n} \times \delta\mathbf{n}) + \delta\mathbf{n} \operatorname{div} \mathbf{n} - (\delta\mathbf{n} \cdot \nabla)\mathbf{n} + \nabla_{\delta n}(\mathbf{n} \cdot \delta\mathbf{n}),$$

where $\nabla_{\delta n}$ means taking derivatives of $\delta\mathbf{n}$ only, we finally obtain

$$\begin{aligned} \delta\Phi = & -K \int \delta\mathbf{n} \cdot \left(\Delta\mathbf{n} + \frac{\epsilon_a}{4\pi} \mathbf{E}(\mathbf{n} \cdot \mathbf{E}) + \lambda\mathbf{n} \right) dV \\ & + (K - K_{24}) \int \delta\mathbf{n}\left((\operatorname{rot} \mathbf{n} \times \mathbf{e}_r) + \mathbf{e}_r \operatorname{div} \mathbf{n} \right) dS \\ & + K_{24} \int \delta\mathbf{n} \cdot (\mathbf{e}_r \cdot \nabla)\mathbf{n} dS \\ & + W \int \delta\mathbf{n} \cdot (\mathbf{n} \cdot \mathbf{e}_r)\mathbf{e}_r dS - \int \delta\mathbf{n} \cdot \mu\mathbf{n} dS . \end{aligned}$$

From the necessary condition of minimum, $\delta\Phi = 0$, one get the following Euler - Lagrange equation (2)

$$K\Delta\mathbf{n} + \frac{1}{4\pi}\epsilon_a \mathbf{E}(\mathbf{n} \cdot \mathbf{E}) + \lambda\mathbf{n} = 0, \quad \mathbf{r} \in \mathbf{V},$$

and boundary condition (3)

$$\begin{aligned} & (K - K_{24})(\operatorname{rot} \mathbf{n} \times \mathbf{e}_r + \mathbf{e}_r \operatorname{div} \mathbf{n}) + \\ & + K_{24}(\mathbf{e}_r \cdot \nabla)\mathbf{n} + W\mathbf{e}_r(\mathbf{n} \cdot \mathbf{e}_r) - \mu\mathbf{n} = 0, \qquad \mathbf{r} \in S . \end{aligned}$$

APPENDIX B: PERCUS_YEVICK APPROXIMATION FOR STRUCTURE FACTOR

Suppose we treat assembly of particles in our cell as a fluid of hard spheres and employ the Percus-Yevick approximation[30]. Then

$$S(\mathbf{q}_s) = \frac{1}{1 - \rho c(\mathbf{q}_s)} - 1, \tag{17}$$

where $c(\mathbf{q}_s)$ is the Fourier transform of the Ornstein-Zernike direct correlation function[27], ρ is the particles number density,

$$c(\mathbf{r}) = \begin{cases} -\lambda_1 - 6\eta\lambda_2 \left(\frac{r}{2R}\right) - \frac{1}{2}\eta\lambda_1 \left(\frac{r}{2R}\right)^3 & r < 2R \\ 0 & r > 2R \end{cases}.$$

Here

$$\lambda_1 = \frac{(1+2\eta)^2}{(1-\eta)^4}, \quad \lambda_2 = -\frac{\left(1+\frac{1}{2}\eta\right)^2}{(1-\eta)^4},$$

and η is the parking fraction

$$\eta = \frac{4\pi}{3} R^3 \rho .$$

Taking Fourier transform we have

$$\begin{aligned}
\frac{1}{2\pi} c\left(\mathbf{q}_s\right) &= -2\lambda_1 \frac{\sin 2qR - 2qR\cos 2qR}{q^3} \\
&+12\eta\lambda_2 \frac{2\left(qR\right)^2 \cos 2qR - 2qR\sin 2qR - \cos 2qR + 1}{q^4 R} \\
&+ \left(2\left(qR\right)^4 \cos 2qR + 3\cos 2qR - 6\left(qR\right)^2 \cos 2qR\right. \\
&\left. + 6qR\sin 2qR - 4\left(qR\right)^3 \sin 2qR - 3\right)\eta\lambda_1 \left(q^6 R^3\right)^{-1}
\end{aligned} \tag{18}$$

APPENDIX C:

In this Appendix we estimate $n_y(\mathbf{q})$at small $\mathbf{q}$. From (13) we have

$$\begin{aligned}
n_y(\mathbf{q}) &= \frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)} \int_1^\infty dx \int_0^\pi d\theta \sin\theta \sin 2\theta \frac{\exp\left\{-iq_z Rx\cos\theta\right\}}{x} \\
&\times \int_0^{2\pi} d\phi \sin\phi \exp\left\{-iq_y Rx \sin\theta \sin\phi\right\} \\
&= -2\pi i \frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)} \int_0^\pi d\theta \sin\theta \sin 2\theta \\
&\times \int_1^\infty dx \frac{\exp\left\{-iq_z Rx\cos\theta\right\}}{x} J_1\left(q_y Rx \sin\theta\right) .
\end{aligned}$$

Here to find

$$\int_0^{2\pi} d\phi \sin\phi \exp\left\{-iq_y Rx \sin\theta \sin\phi\right\}$$

we introduced a new function

$$I(t) = \int_0^{2\pi} d\phi \sin\phi \exp\left\{-it\sin\phi\right\} ,$$

then it is easy to evaluate the integral

$$\int I(t)\,dt = i\int_0^{2\pi} d\phi \exp\{-it\sin\phi\} = 2\pi i J_0(t) \ ,$$

and finally to obtain

$$I(t) = 2\pi i J_0'(t) = -2\pi i J_1(t) \ ,$$

where $J_n(t)$ is Bessel's function. At small angle light scattering, when $qR \ll 1$, we have

$$\frac{1}{x}J_1(q_y Rx\sin\theta) \approx \frac{1}{2}q_y R\sin\theta$$

at $x\tilde{}1$ and $\frac{1}{x}J_1(q_y Rx\sin\theta)\,\tilde{}\,\frac{1}{x^{3/2}}$ at $x \gg 1$, therefore

$$\begin{aligned}
n_y(\mathbf{q}) &\approx -\pi i\frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)}\int_0^{\pi} d\theta\sin\theta\sin 2\theta q_y R\sin\theta \\
&\quad \times\int_1^{\infty}\exp\{-iq_z Rx\cos\theta\}\,dx \\
&= -\pi\frac{\xi R^3}{4\left(1+\frac{1}{2}k_{24}\right)}\frac{q_y}{q_z}\int_0^{\pi} 2\exp(-iq_z R\cos\theta)\sin^3\theta d\theta \\
&= -\pi\frac{\xi R^3}{2\left(1+\frac{1}{2}k_{24}\right)}\frac{q_y}{q_z}\int_{-1}^{1}\exp(-iq_z Rt)\left(1-t^2\right)dt \\
&= -\pi\frac{\xi R^3}{2\left(1+\frac{1}{2}k_{24}\right)}\frac{q_y}{q_z}\frac{1}{q_z^3 R^3} \\
&\quad \times\left(2ie^{-iq_z R} - 2e^{-iq_z R}q_z R - 2ie^{iq_z R} - 2e^{iq_z R}q_z R\right) \\
&\approx -\pi\frac{\xi R^3}{\left(1+\frac{1}{2}k_{24}\right)}\frac{q_y}{q_z}\frac{2}{3} \\
&\approx \frac{2\pi}{3}\frac{\xi R^3}{\left(1+\frac{1}{2}k_{24}\right)}\cot\frac{\vartheta}{2}\,.
\end{aligned}$$

Similarly from (14):

$$\begin{aligned}
n_y(\mathbf{q}) &= \int \delta n_y(\mathbf{r})\exp\{-i\mathbf{q}\cdot\mathbf{r}\}\,d\mathbf{r} \\
&= \frac{l_E^3\pi}{2}C_2\int_{R/l_E}^{\infty} dx\exp(-x)\left(x+3+\frac{3}{x}\right) \\
&\quad \times\int_0^{\pi} d\theta\sin\theta\sin 2\theta\exp\{-il_E xq_z\cos\theta\}
\end{aligned}$$

$$\times \int_0^{2\pi} d\phi \exp\{-il_E x q_y \sin\theta \sin\phi\} \sin\phi$$

$$= -i2\pi^2 l_E^3 C_2 \int_{R/l_E}^{\infty} dx \exp(-x) \left(x + 3 + \frac{3}{x}\right)$$

$$\times \int_0^{\pi} d\theta \sin\theta \sin 2\theta \exp\{-il_E x q_z \cos\theta\} J_1(l_E x q_y \sin\theta)$$

If we consider small angle light scattering when $l_E q \ll 1$ then

$$J_1(l_E x q_y \sin\theta) \approx \frac{1}{2} l_E x q_y \sin\theta,$$

$$\exp\{-il_E x q_z \cos\theta\} \approx 1 - il_E x q_z \cos\theta$$

at actual for integration values of $x \sim \dfrac{R}{l_E} \sim 1$ (we are interested now in light scattering near the threshold voltage)

$$n_y(\mathbf{q}) \approx -i2\pi^2 l_E^3 C_2 l_E q_y \int_{R/l_E}^{\infty} dx \exp(-x) \left(x + 3 + \frac{3}{x}\right) x$$

$$\times \int_0^{\pi} (1 - il_E x q_z \cos\theta) \sin\theta \sin 2\theta \sin\theta d\theta$$

$$= 2\pi^2 l_E^5 C_2 q_y q_z \int_{R/l_E}^{\infty} \exp(-x) \left(x + 3 + \frac{3}{x}\right) x^2 dx$$

$$\times \int_0^{\pi} \cos\theta \sin\theta \sin 2\theta \sin\theta d\theta$$

$$= \frac{8}{15} e^{-\frac{R}{l_E}} \left(R^3 + 15 l_E^3 + 15 l_E^2 R + 6 l_E R^2\right) 2\pi^2 l_E^2 C_2 q_y q_z$$

$$= -\frac{32}{15} e^{-\frac{R}{l_E}} \left(R^3 + 15 l_E^3 + 15 l_E^2 R + 6 l_E R^2\right) l_E^2 \pi^2 k^2 C_2 \sin\vartheta \sin^2\frac{\vartheta}{2}.$$

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