Inter-particle interactions of weak homeotropic anchoring with electric field in a homogeneous nematic cell

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ABSTRACT
We investigate the inter-particle interactions of weak homeotropic anchoring when electric field is applied parallel to far-field director in a homogeneous nematic cell. Two particles interact with the dipole–dipole and quadrupole–quadrupole interactions in the cell. A model of far-field solution dependent on the cell gap and the electric field with weak anchoring is considered to explain the experimental results. The effective radius \( R_{\text{eff}}(=R) \) is introduced to adapt the anchoring strength effect using the radius \( R \) and a coefficient \( \zeta \). The model was experimentally proved with varying electric field and cell gap and it was reasonable for weak electric field. The coefficient \( \zeta \) was measured to be \( \zeta_D = 0.70 \) – 0.74 and \( \zeta_Q = 0.78 \) – 0.85 for the dipole–dipole and quadrupole–quadrupole interactions, respectively, within the experimental range of the weak electric field. The anchoring strength was calculated as \( \sim 10^{-6} \) J/m\(^2\)

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Introduction
A nematic liquid crystal (NLC) colloid is a substance in which microparticles are dispersed and suspended throughout an NLC host. The inter-particle interactions depend on the shape and the surface anchoring conditions of the particles.\cite{1–10} Spherical particles with strong homeotropic anchoring embedded in the NLC bulk display hyperbolic hedgehog or Saturn-ring defects. In these cases, the interactions vary as \( r^{-4} \) or \( r^{-6} \) (where \( r \) is the distance between the centres of mass of two particles), which is consistent with the electric-dipole or electric-quadrupole behaviour, respectively. \cite{11–13} They are therefore referred to as the dipole configuration (DC) and the quadrupole configuration (QC), correspondingly.

NLC colloid systems are analysed in terms of the director orientation (\( \mathbf{n} \)) of the liquid crystal (LC) molecules. The inter-particle interactions are derived from the deformation energy and can be interpreted by analysing the director orientation. The effect of an external electric or magnetic field constitutes the dielectric and diamagnetic response of the LC director and affects the inter-particle interactions.\cite{14} In the vicinity of a particle, the deformation energy varies because the local variation in the director orientation is modified by the external field.

Typically, NLC colloid systems are confined between two substrates that functionally define the boundary conditions.\cite{15} These boundary conditions influence the inter-particle interactions. Inter-particle interactions

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with strong homeotropically anchored particles [16,17]. Not only the system sizes such as particle radius, cell gap and inter-particle distance but also electric field modify the interactions. The interactions were determined to change dramatically according to the relative direction between the far-field director and the electric field. When the electric field was applied parallel to the far-field director in a homogeneous nematic cell, the interactions decreased. Additionally, the interactions weakened for small cell gaps. However, the study did not include the effect of weak homeotropic anchoring on the particle surface.

A study reported on the inter-particle interactions with weak homeotropic anchoring, which were described in terms of the effective radius and the mirror-image method in the absence of electric field.[1] Weak anchoring explains the weakening of the inter-particle interactions. Particle-wall interactions were repulsive as predicted by the mirror-image method. However, it did not explain the interactions depending on electric field. A study of the modification of the elastic interaction by electric field with weak homeotropic anchoring on a particle surface is required for the control of nematic colloid systems.

Here, we present the dependence of inter-particle interactions on weak homeotropic anchoring, the cell gap and electric field in homogeneous nematic cells. We suggest a model and confirm experimentally by varying the experimental parameters for the dipole–dipole and quadrupole–quadrupole configurations.

### Experiment

We used 4-cyano-4′-pentylbiphenyl (5CB, from Merck) as the LC, which is nematic phase in the temperature range of 24°C – 36°C. However, this range changed to 20°C – 36°C after mixing with microspheres. The experiments were performed at 24°C. The effective LC shear viscosity \( \gamma_{eff} \) and elastic constant \( K \) were \( 60 \times 10^{-3} \text{ Pa s} \) and \( 7 \times 10^{-12} \text{ N} \), respectively.[18] The mass density of the LC \( \rho_{LC} \) was 1.01 g cm\(^{-3} \). The effective anisotropic dielectric constant \( \Delta \varepsilon \) was 12.0 and the average dielectric constant \( \varepsilon_{LC} \) was 11.5,[19]

Grey polyethylene microspheres (GRYPMS, from Cospheric) were dispersed in 5CB. Their mass density and diameter were \( \sim 1.00 \text{ g cm}^{-3} \) and \( 27 - 32 \mu \text{m} \), respectively. The dielectric constant \( \varepsilon_p \) of polyethylene is 2.3.[20] The buoyancy acting on the GRYPMS was not critical owing to the small difference in mass density between 5CB and GRYPMS, which prevented them from approaching the substrates too closely. The particles were nearly spherical and had a rugged surface. Atomic force microscope measurement has suggested that the anchoring energy may decrease because of the surface roughness. However, this decrease is negligible compared with the intrinsic anchoring of the material.[1] After the injection of a nematic colloid to the homogeneous cell, the hyperbolic hedgehog defect or the Saturn-ring defect is accompanied around a spherical particle.

Figure 1 illustrates the coordinates of the cell and electrodes. Two electrodes are located on a substrate with a separation to apply in-plane electric field. Both the upper and bottom substrates were coated with polyimide and rubbed uniformly along the z-axis. The far-field director and in-plane electric field are parallel to the z-axis. Cell gap \( L \) is 40 μm or 80 μm. The separation \( d \) between electrodes is about several hundred micrometres. For two parallel electrodes on a substrate, the electric field is not just \( V/d \). Here, \( V \) is the voltage applied between two electrodes. In this experiment, as the electrodes are a flat-plate capacitor structure and the electric field decreases about 40% than that of parallel electrode structure at the middle of electrodes in the experimental condition. We used the values as the electric field. The positions of two particles are defined as \( P_i(x_i, y_i, z_i) \) and \( P_j(x_j, y_j, z_j) \). In this experiment, we restricted the motions of particles to be parallel with the z-axis; thus, the inter-particle interactions are considered to have only \( z \)-component. The \( x \)-positions of particles were approximately \( x \sim 1/2 \) [1] owing to the small density difference and the interaction between particle and substrates in these cell gap conditions.

The inter-particle distances were monitored with a polarising optical microscope (Eclipse E600, from...
Nikon). A function generator (8023, from Tabor electronics) and amplifier (609E-6, from Trek) were used to apply the electric field. Because of the intrinsic limitations of the amplifier, the sinusoidal signal frequency was constrained to 5 kHz.

### Theoretical equations

We propose a model in which the inter-particle interaction was dealt with the cell gap and the electric field with weak anchoring energy. The model is based on a theoretical study reported by S. B. Chernyshuk et al. [17] The elastic dipole–dipole potential energy $U_{DD}$ and elastic quadrupole–quadrupole potential energy $U_{QQ}$ are well known and are described to the dipole moments ($p_1 = aR_1^3$, $p_2 = aR_2^3$) and quadrupole moments ($c_1 = \beta R_1^3$, $c_2 = \beta R_2^3$), where $R_1$ and $R_2$ are the radii of the spherical particles and $a$ and $\beta$ are 2.04 and 0.72, respectively. When the electric field is parallel to the far-field director in a homogeneous nematic cell, the interactions are described as [17]

$$U_{DD} = \frac{16\pi K_p p_2}{\mu_0} \left( F_1 - F_2 \cos^2 \theta \right)$$

$$F_1 = \sum_{n=1}^{\infty} \frac{2\mu_0^2 n \pi x_1}{\sin \left( \frac{n\pi x_1}{L} \right)} \sin \left( \frac{n\pi x_2}{L} \right) \left[ K_0(\mu_0 \rho) + K_2(\mu_0 \rho) \right]$$

$$F_2 = \sum_{n=1}^{\infty} \frac{2\mu_0^2 n \pi x_1}{\sin \left( \frac{n\pi x_1}{L} \right)} \sin \left( \frac{n\pi x_2}{L} \right) \left[ K_0(\mu_0 \rho) \right]$$

$$U_{QQ} = \frac{16\pi K_4 \xi_6}{\mu_0^2} \left( D_1 + D_2 \cos^2 \theta + D_3 \cos^4 \theta \right)$$

$$D_1 = L^2 \left( \frac{2F_2}{\rho^2} - \frac{F_{\rho \rho}'}{\rho} \right),$$

$$D_2 = L^2 \left( -10F_2 + \frac{5F_{\rho \rho}'}{\rho} + \frac{F_{\rho \rho}'}{\rho} - F_{\rho \rho}' \right),$$

$$D_3 = L^2 \left( \frac{8F_2}{\rho^2} - \frac{5F_{\rho \rho}'}{\rho} + F_{\rho \rho}' \right)$$

where $n$ is a positive integer, and $\xi_6$ is the electric coherence length, expressed as $\sqrt{4\pi K/\Delta E^2}$. $K_0(\mu_0 \rho)$ is the modified Bessel function of the second kind and $j$ is an integer. Equation (1) and (2) include the exponential decay of the potential energy due to the behaviour of $K_0(\mu_0 \rho)$ as a function of the cell gap $L$ and the electric field, which provide a screening. However, these equations do not account for the effect of the weak homeotropic anchoring on the particle surface.

To consider the weak homeotropic anchoring ($w$), we introduce the effective radius ($R_{eff}$), which includes the weakening of interactive potential energy that is induced by weak anchoring on the particle surface.[1] The radius and effective radius are related by $R_{eff} = \xi R$. Here, $\xi$ is a coefficient smaller than 1, which is defined as $1 - K/wR$. The elastic dipole–dipole and quadrupole–quadrupole interaction energies are expressed as $U_{DD}^{eff} = \xi_D^4 U_{DD}$ and $U_{QQ}^{eff} = \xi_Q^4 U_{QQ}$, where $\xi_D$ and $\xi_Q$ are coefficients related with the effective radius for the dipole and quadrupole configurations, respectively.

From the elastic interaction energies and electric dipole–dipole interaction force $F_E = 6\pi \varepsilon_0 \varepsilon_{LC} \left( [\varepsilon_E - \varepsilon_{LC}] / (\varepsilon_E + 2\varepsilon_{LC}) \right)^2 \left( R_1^2 R_2^2 \rho^4 / \rho^2 \right)$, [17] we can derive the forces between two particles. Because we restricted the motion of the particles along the $z$-axis, the separation distance $\rho$ is equal to $z_2 - z_1$. The elastic dipole–dipole and quadrupole–quadrupole interaction forces are expressed as $F_{DD}^{eff} = -\partial U_{DD}^{eff} / \partial z$ and $F_{QQ}^{eff} = -\partial U_{QQ}^{eff} / \partial z$, respectively. We defined an average radius $\bar{R} = (R_1 + R_2) / 2$. When single spherical particle having radius $R$ moves at the speed of $v$, then the Stokes drag force $F_{Stokes} = 6\pi \gamma_{eff} R v$ acts on the particle. For inter-particle motion, the speed is due to a relative motion between two particles and a particle’s speed is a half of $v$. And the Stokes drag force is $F_{Stokes} = 3\pi \gamma_{eff} \bar{R} v$.[12] Then, the total force acting on a particle of the dipole–dipole configuration (DDC) is $F_{DD} = F_{DD}^{eff} + F_E + F_{Stokes}$ and on one of the quadrupole–quadrupole configuration (QQC) is $F_{QQ} = F_{QQ}^{eff} + F_E + F_{Stokes}$. Because the particles move slowly, we can assume that the motion has no acceleration, $F_{DD} \sim 0$ and $F_{QQ} \sim 0$.[12] Additionally, only the $n = 1$ mode is considered because the higher modes decay rapidly at distance larger than $2\bar{R}$. Then, the velocities are expressed as
Equations (3) and (4) represent the velocities of the dipole–dipole ($\dot{z}_{DD}$) and quadrupole–quadrupole ($\dot{z}_{QQ}$) configurations, respectively.

**Results**

To confirm the validity of the model, we examined the coefficient $\zeta$. If the model functions correctly, then $\zeta$ must be measured to have a specific value for varying experimental parameters. Figure 2 shows the inter-particle motions of (a) antiparallel DDC and (b) parallel QQC. The top images of Figure 2(a) and (b) represent the initial arrangement of the two particles which are prepared in an electric field. Two particles gradually move away from each other as soon as the electric field is turned off or is decreased. The reference time $t = 0$ is determined to start of movement of particles. The separation of the DDC is faster than that of the QQC because the dipole interaction is stronger than the quadrupole one. The separation $\tilde{z}$ is the distance between the centres of the two particles. In Figure 2 (a), the motion is slightly tilted from the far-field director. This tilt was neglected because it was tiny.

Figure 3 shows the separations $\tilde{z}$ and the separating velocities $\dot{\tilde{z}}$ of DDC for two different cell gaps. In strong electric field, the two particles are aligned along the electric field, as shown in the first image of Figure 2(a). The inter-particle interactions were...
obtained by observing $\bar{z}$ as function of time ($t$). The reduction of the electric field leads to the decrease of the attractive electric force, while the repulsive elastic force does not decrease and causes the separation. Figure 3(a) and (b) shows $\bar{z}$ plotted as a function of the electric field and $t$ for two different cell gaps. As both the repulsive elastic and the drag forces act without electric field, the separation is largest without electric field.

The inter-particle interactions can be analysed to compare the experimental and calculated values of $\bar{z}$. Figure 3(c) and (d) represents the experimental (black dots) and calculated (coloured mesh grid) $\bar{z}$. The velocity is expected to be larger for short separation and with the absence of electric field. The experimental results are consistent with this prediction, while the velocity decreases for a large separation and strong electric field. The insets of Figure 3(c) and (d) show the tendencies of the calculated $\bar{z}$ (solid red line) and the experimental $\bar{z}$ (black dots), which are in good agreement with $\zeta_D = 0.74$ and $\zeta_D = 0.70$. At $E = 0 \text{ V}/\mu\text{m}$ and $E = 0.007 \text{ V}/\mu\text{m}$, $\zeta_D$ is fitted well by a constant value whereas smaller $\zeta_D$ value is observed in stronger electric fields. The experimental and calculated velocities exhibit a difference in strong electric field.

Figure 4 displays $\bar{z}$ and $\bar{z}$ values of QQC for two different cell gaps. The interactions of QQC are also weakened for large $\bar{z}$ and in strong electric field. $\bar{z}$ decreases according to the increasing electric field because both the drag and electric forces obstruct the separation. We compared $\bar{z}$ from the experiment and the calculation using Equation (4). In Figure 4(c) and (d), the experimental $\bar{z}$ is denoted by the black dots and the calculated $\bar{z}$ is plotted as coloured mesh grid. The trends of the experimental and calculated $\bar{z}$ are presented in the insets of Figure 4(c) and (d) and are consistent with $\zeta_Q = 0.85$ and $\zeta_Q = 0.78$, respectively. At $E = 0 \text{ V}/\mu\text{m}$ and $E = 0.007 \text{ V}/\mu\text{m}$, $\zeta_Q$ is fitted well by a constant value whereas larger $\zeta_Q$ is observed in stronger electric field. The experimental and calculated velocities exhibit differences in the short separation range, as mentioned above for the DDC experiment.

We used the experimental data of DDC and QQC displayed in Figure 3(d) and Figure 4(d) to estimate the
appropriate separation range in the model. In Figure 5, (a–c) show the experimental and calculated velocities for Figure 3(d). The calculations for weak electric field are in good agreement with the experimental data provided in Figure 5(a) and (b). In Figure 5(c), the calculation of $\zeta_D = 0.66$ is consistent with the experimental data up to 35 μm, but velocity trend deviates from the model with increasing separation.

To investigate the difference between the experimental data and the calculation, we introduced the
relative length ratio \((z_0 + 2R)/\xi_E\), where \(z_0\) is the distance between the stationary particles. \(z_0 + 2R\) indicates the farthest distance between the surfaces of the two particles. The length ratio for Figure 3(d) is plotted as a function of the electric field in Figure 5(d) and the inset shows the fitted \(\zeta_D\). When the electric field strength reaches 0.010 V/\(\mu m\), \(\zeta_D\) varies. From this result, the model is unsuitable for electric fields stronger than 0.007 V/\(\mu m\). The change occurs near a length ratio of 0.6. Figure 5(e–g) shows the experimental and calculated velocities for Figure 4(d). As mentioned above, the calculations agree well with the experimental data for \(\zeta_Q = 0.78\) in Figure 5(e) and (f). The trend deviates from the model when \(\zeta_Q = 0.78\) and it reasonably agrees with the results for \(\zeta_Q = 0.80\) in Figure 5(g). The relative length ratio is plotted in Figure 5(h) as a function of the electric field and the inset shows the change of \(\zeta_Q\). \(\zeta_Q\) also changes at 0.010 V/\(\mu m\).

The model is expected to be suitable for long electric coherence length, which corresponds to weak electric field. The elastic interactions strongly depend on the director field positioned between two particles and the director field starts to be further distorted when the relative length ratio is smaller than about 0.6. That qualitatively coincides with the expectation. That is, stronger electric field causes the rearrangement of the director field and reduces the elastic interaction that leads to the difference between the model and the experimental results. From this estimation, the electrical control of a nematic colloid system may be advantageous for small-sized particles because the electric coherence length is controllable.

Furthermore, we can show both the decrease of \(\zeta_D\) and increase of \(\zeta_Q\) displayed in the two insets of Figure 5(d) and (h). The models suggest that the elastic interactions decrease with the electric field. However, the quadrupole–quadrupole interactions slightly increase with the electric field. We consider that such variations are due to the director field of the two particles and the electric field of two induced electric dipoles. The electric field is similar to the director field of the QQC; therefore, the director field is strengthened by the electric field because 5CB has positive dielectric anisotropy. In the case of the DDC, however, the director field is weakened by the electric field, leading to a decrease of \(\zeta_D\). The similarity between the electric field and the director field could be a reason for the variation of the elastic interaction.

\(\zeta_D\) and \(\zeta_Q\) values obtained from the inter-particle interactions with the effective radius and mirror image method are not very different from this report for the same physical parameters.[1] The difference seems to be due to the different approach to the interaction between particles. In particular, the relative size of \(\zeta_D\) and \(\zeta_Q\) in this experiment is reversed to the previous results. We consider that this result is due to the limitation of the far-field solution. In the case of the DDC, three different types of pairs exist, one parallel (\(\rightarrow \rightarrow\)) and two antiparallel (\(\leftarrow \rightarrow\), \(\rightarrow \leftarrow\)) configurations. The arrow represents direction from the centre of a particle to the hyperbolic hedgehog defect. These interactions that are based on the far-field solution have the same dependence on the separation. However, the interactive forces of three types of dipole–dipole pairs are different, which has been reported in experimental and numerical studies.[21,22] The solution does not explain such a difference. The interaction of the parallel DDC (\(\rightarrow \rightarrow\)) is strongly dependent on a hyperbolic hedgehog defect positioned between two particles, and hence, the deformation is determined by the hyperbolic hedgehog defect and the particle surfaces near the defect rather than by the boundaries of substrates. In the case of the antiparallel configuration (\(\leftarrow \rightarrow\)), the deformational region depends on the boundaries of substrates. Furthermore, the director field of the QQC is more similar to the far-field director \(n_0\) than that of the antiparallel configuration (\(\leftarrow \rightarrow\)). Therefore, we estimate that the cell gap effect is larger for the antiparallel configuration (\(\leftarrow \rightarrow\)) than for the QQC, leading to the measurement of a smaller value for \(\zeta_D\) than for \(\zeta_Q\).

As mentioned earlier, the model considers the effect of the cell gap and the electric field, which acts as a screening effect to the elastic interactions. This screening effect can be confirmed indirectly from the experimental data and the calculated anchoring strength. The anchoring strength \(w = K/(1 - \eta)R\), calculated using \(\zeta_D\) and \(\zeta_Q\), is \(1.9 \times 10^{-6}\) J/m\(^2\) and \(2.9 \times 10^{-6}\) J/m\(^2\), respectively. These similar results indicate that the model is appropriate. The anchoring strength coincides with weak interaction between the amorphous polyethylene surface and the 5CB.[23]

**Conclusions**

The inter-particle interactions of the DDC and QQC in a homogeneous nematic cell are explained using a model. The proposed model includes the influence of the cell gap, electric field and the anchoring strength. Effect of cell gap and electric field is combined each other and modify the interaction exponentially. They obstruct the elastic interaction providing screening. Weak anchoring reduces the effective radius of the particles. For experimental parameters of electric field and the cell gap, the
model is consistent with experimental data in the range of $\zeta_D = 0.70 - 0.74$ and $\zeta_Q = 0.78 - 0.85$ in weak electric field. The model is reasonably effective when the relative length ratio is smaller than ~0.6 owing to the rearrangement of the director with the electric field. Both a decrease of $\zeta_D$ and an increase of $\zeta_Q$ are observed in strong electric field and the deviation between the experimental and calculated results increases. A variation of $\zeta$ with increasing electric field is expected because of the similarity between the electric field of two electric dipoles and the director field between two particles; namely, the screening effect is weakly dependent on the electric field created by the two induced electric dipoles. The particle has weak anchoring of approximately $10^{-6}$ J/m$^2$, according to the experimental data of $\zeta$.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

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