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Electro-orientation in particle light valves

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Abstract
Electro-orientation of rod-like particles in liquids, under the application of an external AC field, is analysed. A rod shape is suitable for particle light valve (PLV) applications. When they are aligned with their long axes parallel to the electric field (and the direction of light is assumed to be parallel to the applied electric field), then it can lead to good transmission of light. Various criteria to arrive at appropriate parameters for PLV applications are proposed. It is found that good electric conductors are excellent rod materials for PLV applications. They lead to an appropriate orientation of the rods and at the same time result in maximum orientational torque. Water-like liquids with higher values of permittivity are appropriate choices as suspending liquids since the Brownian dispersion in the presence of the electric field is minimized. The time it takes the rods to fully diffuse in the orientational space, once the electric field is turned off, decreases with decreasing liquid viscosity.

1. Introduction

A suspension of rod or disc-like particles in a transparent liquid can be used to produce windows with variable transmission of light. These windows comprise a flat cell with a liquid–particle suspension layer between transparent electrode sheets. The alignment of the rod or disc-like particles, in response to an applied AC electric field (electro-orientation), causes a large change in the transmission, absorption and reflection of light (electro-optics). This is the basis of the technology referred to as particle light valves (PLVs) or suspended-particle devices (SPDs).

The key challenges in making a stable suspension of particles in a liquid, for PLVs, have been gravitational settling and agglomeration. However, with the fabrication of nanoparticles these issues can be addressed satisfactorily. The PLV technology is therefore being harnessed for commercial applications (see e.g. www.smartglass.com). The current generation of PLVs was produced empirically and with a limited range of material properties [1]. There remains a need to optimize the performance of PLVs by appropriately modelling the underlying physical mechanisms. Optimal PLVs will be relatively inexpensive, thus permitting use in place of conventional windows in residential homes, commercial buildings and transportation vehicles [1].

The theoretical modelling of PLVs can be broadly divided into light scattering and transmission studies for given particle orientations (electro-optics), and electrohydrodynamic studies. In this paper the latter problem is considered. The electrohydrodynamic problem has two components—particle orientation by an electric field (electro-orientation) and fluid dynamics. Fluid dynamics describes the Brownian motion of the particles and the time to start or shut off the device, among other issues. It is noted here that other electrohydrodynamic issues, such as electro-osmosis, electro-thermal effects [20, 21] etc, that could also arise in PLV devices are not the focus of this work and will be considered in a separate study.

The theory of electro-optics of particle suspensions has been presented by, e.g., Marks [2] and Stoylov [3]. They used a simplified theory of electro-orientation. The specific influence of the particle geometric parameters and their material properties on the electrohydrodynamic issues was not addressed in detail. A systematic study of the electrohydrodynamic problem based on the leading theory of electro-orientation [4] is required. The key goal is to find criteria that will help select appropriate parameters, during the set-up and design of a PLV device, such that the electrohydrodynamic issues to be listed in section 2.2 are adequately addressed.

Two approximate theories for electro-orientation have been proposed—the energy theory [5, 6] and the dipole moment theory [7]. Objections were raised, however, that the energy theory is invalid in dissipative media [8, 9]. The predictions of the dipole moment and energy theories have been compared with the experiments of ellipsoidal

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erythrocytes [10, 11, 4]. The results showed that the dipole moment theory provides a better explanation of the experimental observations of orientational phenomena. These conclusions were further supported by the rigorous derivations of Sauer and Schlögl [12] for spherical particles. Their rigorous theory is hard to apply to cases such as non-spherical particles. The dipole moment theory, despite some approximations involved, is a useful method in such problems [4]. Hence, in this work the dipole moment theory of electro-orientation will be used.

In section 2 the electrohydrodynamic issues in a PLV device will be discussed along with a theoretical analysis. Results will be presented in section 3 and conclusions in section 4.

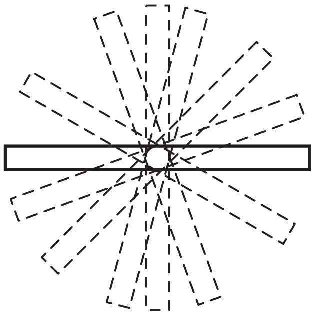

Figure 1. Various orientations of a rod with its long axis perpendicular to the applied electric field. The applied electric field is directed into the plane of the paper.

2. Theoretical modelling

In PLV applications, AC electric field is preferred to minimize the effects of electrophoretic migration. Jones [4] has given an expression for the orientational torque, due to an AC electric field, on an ellipsoidal particle. The dipole moment theory is used. It can be verified from their equations that the torque acting on an ellipsoid is zero whenever the electric field is aligned with one of its principal axes (i.e. these are the equilibrium orientations of the ellipsoid). However, all three possibilities are not stable equilibrium orientations. It is possible to deduce the stable orientation based on the signs of the three components of the torque in various orientations. Such a chart is presented by Miller [11] and reproduced by Jones [4]. The signs of the torque components depend on the electric field strength, the particle geometry, the conductivities and permittivities of the media, and the applied frequency. The stable orientations of rods and discs can be deduced by idealizing them as prolate and oblate ellipsoids, respectively. Once the stable orientation is known the optics problem can be solved to deduce the light transmission properties.

A disc suspended in a liquid can have two equilibrium orientations—electric field perpendicular and parallel to the plane of the disc. Similarly, there are two equilibrium orientations for a rod—electric field parallel and perpendicular to the long axis of the rod. Among all these cases, a rod parallel to the electric field will typically have the best light transmission properties [2, 3]. Now consider a case where the rod axis is perpendicular to the electric field. By symmetry, any orientation, obtained by rotating the rod with respect to the axis parallel to the electric field, is possible (figure 1). Thus, rotational Brownian motion will readily occur with respect to this axis. The resultant random orientations of the rods can lead to reduced transmission of light [2, 3]. Turning off the electric field will result in Brownian diffusion in the entire orientational space, which can also reduce light transmission. The rod shape is therefore viable for applications where good as well as reduced transmission of light is desired in a controllable manner. Since the key applications of PLVs have this objective, we will consider the rod geometry in this work.

2.1. Electro-orientation torque on rods

In the discussion to follow, a rod will be modelled as a prolate ellipsoid. Such an approximation is reasonable for large aspect ratios because the leading order trend remains the same [13]. Dilute particle concentrations will be assumed so that an analysis based on a single particle is adequate. The length scale of the PLV device, e.g. the distance between the electrodes, will be assumed to be much larger than the particle size so that the dipole moment theory is a reasonable approximation. It will be assumed throughout that the light is directed parallel to the applied electric field.

Consider the geometry shown in figure 2. Let an AC electric field of amplitude E_0 act along the x' axis. Let the rod be rotated about the z axis (concurrent with the z' axis) by an angle θ as defined in figure 2. The x, y and z axes are fixed along the three principal axes of the prolate ellipsoid. According to the dipole moment theory, the time averaged alignment torque with respect to the z axis, $\langle T^E \rangle^z$, is given by [4]

$$\frac{3\langle T^E \rangle^z}{2\pi a b^2 E_0^2 \varepsilon_f} = C_s \frac{\sin(2\theta)}{2}, \tag{1}$$

where

$$C_s = \mathrm{Re}[(L^x - L^y)[(E_R\Omega - jS_R)^2]\{([1 + E_R L^x]\Omega - j[1 + S_R L^x])([1 + E_R L^y]\Omega - j[1 + S_R L^y])\}^{-1}]. \tag{2}$$

C_s is the non-dimensional torque. Henceforth, in the paper it shall be referred to as the torque constant. E_R, S_R and Ω are the non-dimensional parameters defined as follows:

$$E_R = \frac{\varepsilon_p - \varepsilon_f}{\varepsilon_f}, \qquad S_R = \frac{\sigma_p - \sigma_f}{\sigma_f}, \qquad \text{and} \qquad \Omega = \frac{\varepsilon_f \omega}{\sigma_f}. \tag{3}$$

Superscripts denote the x, y or z components. The semi-axis length of the ellipsoid in the y and z directions is b while that in the x direction is a. ε and σ are the permittivity and the ohmic conductivity, respectively. The material properties are assumed to be isotropic and constant. Generalization to exclude these assumptions is straightforward [4]. Subscript f denotes the liquid and p denotes the particle. In equation (2), j is $\sqrt{-1}$ and Re denotes the real part. ω is the applied radian frequency. For a prolate ellipsoid, $a \gg b$ and $L^x \ll L^y = L^z$. Thus, we have [4]

$$L^y \approx \tfrac{1}{2}, \qquad \text{and} \qquad L^x \approx \frac{b^2}{a^2} \ln\left(\frac{2a}{b} - 1\right), \tag{4}$$

where a/b is the aspect ratio of the slender rod-like ellipsoid.

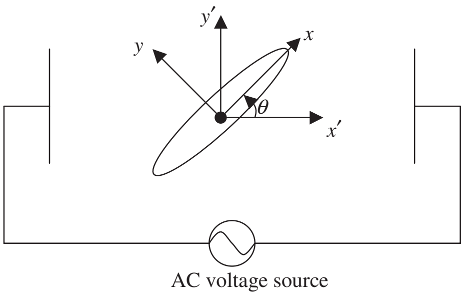

Figure 2. A prolate ellipsoid suspended in an AC electric field.

It follows from equation (1) that a negative value of C_s will ensure that $\theta = 0$ (or equivalently $\theta = \pi$) is the stable orientation while a positive value will make $\theta = \pi/2$ (or equivalently $\theta = 3\pi/2$) the stable orientation. For the objectives at hand, the parameters should be such that $\theta = 0$ is the stable orientation when the electric field is turned on (henceforth referred to as the on state). Note that, by symmetry, the equation of rotation with respect to the y' axis (made concurrent with the y axis) will be identical. The rotation about the x axis is inconsequential due to the symmetry of the rod geometry. There is also a time dependent component of the torque that acts on the particle, but the applied frequency will be assumed to be large so that the ellipsoid responds only to the time averaged torque [4].

2.2. Electrohydrodynamic issues in rod orientation

Consider the rod depicted in figure 2. The equation of rotation for a small angular displacement θ about the $\theta = 0$ equilibrium position can be approximated by

$$I^z \frac{\mathrm{d}^2\theta}{\mathrm{d}t^2} = \frac{2\pi ab^2 E_0^2 \varepsilon_f}{3} C_s \theta - 8\pi\eta a^3 K_s \frac{\mathrm{d}\theta}{\mathrm{d}t}, \tag{5}$$

where

$$K_s = \frac{4e^3(2 - e^2)}{3(-2e + (1 + e^2)\ln([1 + e]/[1 - e]))},$$

$$\text{and} \qquad e = \sqrt{1 - b^2/a^2}. \tag{6}$$

I^z is the moment of inertia about the z axis and η is the liquid viscosity. The last term in equation (5) denotes the viscous resistance to rotation about the z axis, and K_s is a shape factor [14]. Equation (1) is used in equation (5) where $\sin(2\theta)$ is approximated by 2θ. The effect of the rotational Brownian motion will be quantified later. An analysis based on the above equations is not strictly valid for large θ. However, it is commonly used to obtain order of magnitude estimates for Brownian particles in external force fields [15].

The key electrohydrodynamic issues in PLV devices will be discussed below with appropriate theoretical analyses:

(1) The stable orientation of the rods can change with the frequency. Rods aligned with their long axes parallel to the direction of light are typically desirable for good transmission of light. The torque constant C_s should be negative for this objective.

(2) In the absence of an external electric field, the rods are randomly oriented due to the rotational Brownian motion (henceforth to be referred to as the off state). The time it takes a rod to orient after the application of the electric field is called the response time of the device. The parameters should be such that the response time is short.

Since the rods are small, the inertia term (left-hand side of equation (5)) is negligible. The alignment torque is balanced by the viscous resistance. This balance determines the *order of magnitude* of the response time of the rod, which is the same as the time it takes the rod to come back to its equilibrium location after being disturbed by an angular displacement θ. It follows that

$$t_R = \frac{12\eta a^2 K_s}{b^2 E_0^2 \varepsilon_f |C_s|}, \tag{7}$$

where t_R is the order of magnitude of the response time and $||$ indicates the absolute (positive) value of the real valued torque constant C_s. $t_R \ll 1$ ensures a short response time.

(3) The inertia of the rod is negligible if

$$R_I = \frac{I^z b^2 E_0^2 \varepsilon_f |C_s|}{96\pi\eta^2 a^5 K_s^2} \ll 1, \tag{8a}$$

where

$$I^z \approx \frac{1}{12} M l^2 = \frac{4\pi}{9} \rho_p a^3 b^2. \tag{8b}$$

Equations (5) and (7) are used, in the order of magnitude analysis, to get equation (8*a*). M is the mass of the rod, ρ_p is the rod density and $l = 2a$ is the length of the rod.

(4) In the presence of an AC field, the timescale, based on the applied frequency, should be much shorter than the response time of the rods i.e. $t_R \gg 1/\omega$. If this condition is not satisfied, then the rods will oscillate about the mean orientation. This can reduce light transmission in the on state. Equation (7) together with this condition implies

$$\frac{12\eta a^2 K_s \omega}{b^2 E_0^2 \varepsilon_f |C_s|} \gg 1. \tag{9}$$

(5) The angle θ_D by which a rod will diffuse in the orientational space, due to the rotational Brownian motion, in time t is given by

$$\theta_D^2 = \frac{k_B T}{4\pi\eta a^3 K_s} t, \tag{10}$$

where the rotation is about the z axis, T is the temperature and k_B is the Boltzmann constant.

Good transmission of light in the on state will be achieved if the rods remain in their stable equilibrium orientation (i.e. $\theta = 0$ for the case considered here) at all times. However, this is not the case due to the rotational Brownian motion. We shall refer to this diffusion as the Brownian dispersion in the on state. For good performance, the parameters should be such that the Brownian dispersion in the on state is minimized.

The angle θ_D^R by which a rod will diffuse in time t_R is an *estimate* of the Brownian dispersion in the on state. Equations (7) and (10) give

$$(\theta_D^R)^2 = \frac{3k_B T}{\pi ab^2 E_0^2 \varepsilon_f |C_s|}. \tag{11}$$

A small value of θ_D^R implies less dispersion and consequently better transmission of light. Note that θ_D^R is independent of the viscosity of the liquid. This is a consequence of the fact that the effects of η in equations (7) and (10) cancel each other.

Thus, the liquid permittivity, not its viscosity, plays a key role in determining the Brownian dispersion in the on state. This result, although not intuitive, is consistent with earlier results on the dispersion of particles in force fields [16].

Equation (11) implies that the Brownian dispersion in the on state can be controlled by the strength of the electric field. The dispersion is minimized if

$$\frac{3k_BT}{\pi ab^2E_0^2\varepsilon_f|C_s|} \ll 1. \tag{12}$$

(6) Once the electric field is turned off, there is no orientational torque on the rods. Hence, the Brownian diffusion causes unhindered dispersion of the rods in the orientational space until a fully random distribution (off state) is achieved. The time it takes for the rods to go to the off state, once the electric field is turned off, is the decay time. Putting $\theta_D = \pi/2$ in equation (10) we get

$$t_{dec} = \frac{\pi^3\eta a^3K_s}{k_BT}, \tag{13}$$

where t_{dec} is the *order of magnitude* of the decay time. Equations (7), (12) and (13) imply

$$\frac{t_{dec}}{t_R} \gg \left(\frac{\pi}{2}\right)^2, \tag{14}$$

i.e. t_{dec} has to be larger than t_R to ensure that Brownian motion is not dominant in the on state. Equations (12) and (14) are equivalent.

A small value of t_{dec} is desirable to make sure that the PLV goes to the off state quickly after the electric field is turned off. This condition and equation (14) can be reasonably satisfied by appropriate choice of parameters. A low viscosity liquid will ensure small decay time (equation (13)), while a high electric field strength or large ε_f can be chosen to ensure equation (12) (i.e. equivalently equation (14)).

In summary, the following can assist in the 'designing' a PLV with rod-like particles.

(a) Equation (2) helps to identify the appropriate electrical properties of the materials and the frequency range so that the long axis parallel to the electric field is the stable orientation of the rods.
(b) Equation (8*a*) may be regarded as a criterion for the rod size so that the inertia can be neglected.
(c) Equation (9) indicates that the operating frequency of the device should be chosen large enough so that the rods respond only to the average orientational torque.
(d) Equation (12) can be regarded as a criterion for choosing the magnitude of the electric field strength or ε_f, so that the Brownian dispersion is minimized in the on state. A large magnitude of the torque constant C_s is also recommended for this objective.
(e) Equation (13) leads to a criterion on the liquid viscosity so that the decay time of the device is not large.

A similar process can be repeated for other geometries (e.g. discs) along with the required objectives to arrive at the optimal values of the parameters. Criteria (a)–(e), above, will form the basis of the presentation of the results.

3. Results

In section 3.1, plots of the torque constant C_s versus various parameters will be considered. This will lead to information about the sign and the magnitude of C_s. The sign of C_s is useful to determine the stable orientation of the rods (criterion (a)) while a large magnitude of C_s is desirable to minimize the Brownian dispersion in the on state (criterion (d)). In section 3.2 the design criteria, discussed above, will be applied to specific cases. In both these sections, calculations will be presented for an aspect ratio $a/b = 20$. The effect of varying aspect ratios will be discussed in section 3.3.

3.1. Plots of the torque constant C_s for ellipsoidal rods

The value of the torque constant C_s depends on E_R, S_R, Ω and a/b. In the results presented below, values of E_R between -1 and 100 will be considered. This range is determined, based on the typical values of permittivities of liquids and solids [17, 18]. The value of S_R can range from negative (for insulating rods) to large positive values (for good electric conductors). Hence, the entire range will be considered. The minimum possible value of S_R is -1. In our planned electro-orientation experiments, the maximum applied frequency, f_{max}, will be of the order of 100 kHz (i.e. a maximum radian frequency $\omega_{max} = 2\pi f_{max} = 0.63$ rad μs^{-1}). Similar frequency is reported by Saxe and Thompson [1] for PLV devices. It is noted, however, that electro-rotation has been measured for frequencies up to 20 MHz [20]. Although different types of liquids may be considered for this application, in section 3.2, we will specifically consider water, toluene and chloroform. Based on the typical material properties of these and other organic liquids, the maximum value of Ω of interest is 50. Although the minimum value of Ω is determined by equation (9), values of Ω from 0 to 50 will be considered in this section.

Before we depict the plots of C_s, a few remarks are in order. The value of C_s for $\Omega \to 0$ is given by

$$C_s|_{\Omega\to 0} = (L^x - L^y)\frac{S_R^2}{(1+S_RL^x)(1+S_RL^y)}. \tag{15}$$

Note that this value is independent of E_R. Similarly, the value of C_s for $\Omega \to \infty$ is given by

$$C_s|_{\Omega\to\infty} = (L^x - L^y)\frac{E_R^2}{(1+E_RL^x)(1+E_RL^y)}, \tag{16}$$

which is independent of S_R. We have verified the above by plotting C_s versus Ω for different values of E_R and S_R. In fact $C_s|_{\Omega\to 0}$ and $C_s|_{\Omega\to\infty}$ have the same limiting value for $S_R \to \infty$ and $E_R \to \infty$, respectively. This limiting value is a function of only the aspect ratio of the rod. It represents the maximum magnitude of C_s for a given aspect ratio of the rod:

$$C_{s,max} = C_s|_{\Omega\to 0, S_R\to\infty} = C_s|_{\Omega\to\infty, E_R\to\infty} = \frac{1}{L^y} - \frac{1}{L^x}. \tag{17}$$

If C_s is plotted as a function of Ω, then the value of C_s changes from that given by equation (15) for $\Omega \to 0$ to that given by equation (16) for $\Omega \to \infty$. Equations (15) and (16), thus, reveal the orientation of the rods at high and low frequencies, respectively. Both the equations have the

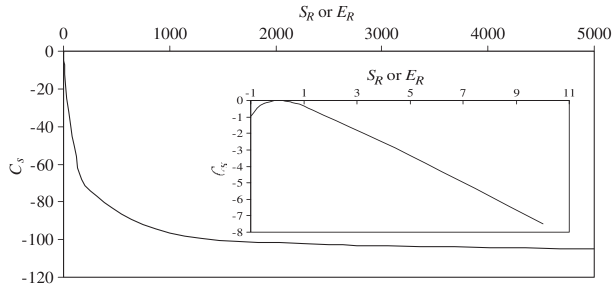

Figure 3. Plot of the torque constant C_s in the limit of $\Omega \to 0$ or $\Omega \to \infty$ (equations (15) or (16)) as a function of S_R or E_R, respectively ($a/b = 20$). The inset shows the plot at small values of S_R or E_R.

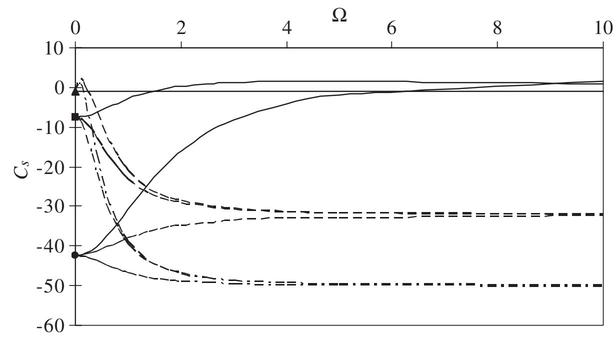

Figure 4. Plot of the torque constant C_s as a function of Ω (regime A) for $E_R = 100$ (— · —), 50 (– – –), -1 (——). All curves originating at the same point on the ordinate axis have the same value of S_R and are denoted by a symbol. The values are $S_R = -1$ (▲), 10 (■), 75 (●).

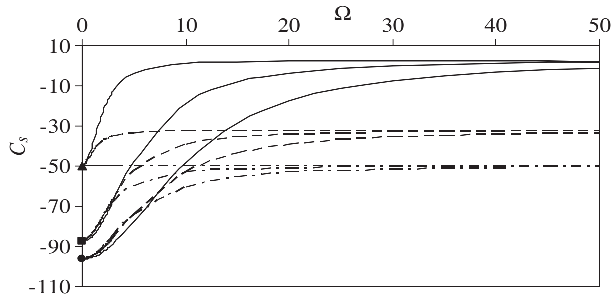

Figure 5. Plot of the torque constant C_s as a function of Ω (regime B) for $E_R = 100$ (— · —), 50 (– – –) and -1 (——) and $S_R = 100$ (▲), 500 (■) and 1000 (●).

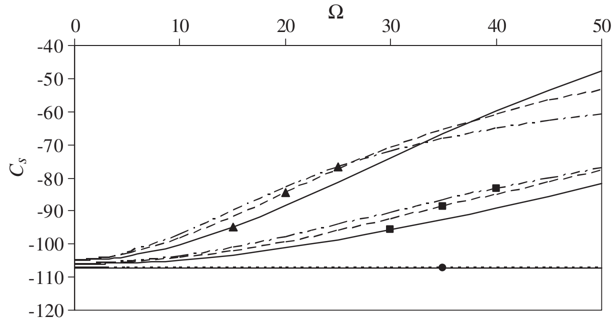

Figure 6. Plot of the torque constant C_s as a function of Ω (regime C) for $E_R = 100$ (— · —), 50 (– – –) and -1 (——) and $S_R = 5000$ (▲), 10^4 (■) and 10^6 (●).

same functional form with respect to S_R and E_R, respectively. In figure 3 this function is plotted. It can be deduced that the stable orientation at low ($\Omega \to 0$) and high ($\Omega \to \infty$) frequencies is always the long axis parallel to the electric field (the desired configuration) for large aspect ratio rods (here, we have $a/b = 20$).

Equation (2) implies that C_s is independent of Ω (i.e. constant) when $E_R = S_R$. For $E_R \neq S_R$, the value of C_s does not necessarily change monotonically from the low to the high frequency limits of equations (15) and (16), respectively. Hence, it could have positive values (i.e. the long axis perpendicular to the electric field) for some intermediate values of Ω depending on the values of E_R and S_R.

For clarity of presentation, the plots of the torque constant C_s versus Ω are categorized into three regimes depending on the value of S_R. Regime A is defined for $S_R < 100$, regime B for $100 \leqslant S_R < 5 \times 10^3$ and regime C for $S_R \geqslant 5 \times 10^3$. Each of these cases will be discussed in the following.

Figure 4 shows the plot for regime A. In this regime, the value of C_s rapidly attains the asymptotic value of equation (16). Hence, the plots are shown only up to $\Omega = 10$. The asymptotic value is reached typically for $\Omega > 5$. Since the asymptotic value is negative the stable orientation of the rod will have its long axis parallel to the electric field. A larger value of E_R is however preferred since it implies larger magnitude of C_s—a desirable feature to satisfy the condition in equation (12). At smaller values of Ω the value of C_s can be positive especially for S_R and/or E_R magnitudes of order unity or less (figure 4). Thus, the rod stable orientation will be the long axis perpendicular to the electric field. Therefore, such parameters are unfavourable for the application of interest here.

The plot for regime B is depicted in figure 5. In this case, the high frequency asymptotic value is reached at larger values of Ω. It is clear that if E_R is of order unity, then the value of C_s is positive over a considerable range of Ω (figure 5). Thus, larger values of E_R are preferred. C_s is negative and its magnitude is large at small Ω because the value of S_R is large in this regime (equation (15)).

Regime C is shown in figure 6. This is the large S_R regime where the value of C_s does not reach the asymptotic value of equation (16) in the range of Ω of interest. The values of C_s are typically closer to those given by the limiting value in equation (15). Since S_R is large, the magnitude of C_s is also comparatively large. Both the stable orientation and the magnitude of C_s are favourable in this regime for almost the entire range of Ω of interest. Thus, good electric conductors (i.e. large values of S_R) will be excellent candidate materials for PLVs.

3.2. Design criteria applied to specific cases

In this sub-section specific materials will be considered. The design criteria listed at the end of section 2 will be considered. The procedure to be presented below can be easily repeated for other materials. Our choice of materials is primarily motivated by what is likely to be used by us in our experimental investigation.

All the dimensions of interest are more than 10 nm. Hence, molecular scale effects will not be considered, except for the case of single-wall carbon nanotubes (SWCNTs). All the material properties will be the 'bulk' values (except for SWCNTs).

Water, toluene and chloroform will be considered as the suspending liquids. The rod materials being considered are gold, germanium, silicon, zinc oxide and SWCNTs. Tables 1(a) and (b) list the properties of these materials (see [17–19] and references therein), where the dielectric constant is the permittivity of the material normalized by the permittivity of vacuum (8.854×10^{-12} F m^{-1}). For SWCNTs we follow Dimaki and Bøggild [19, and references therein] for the choice of its properties. Table 2 shows the values of S_R and E_R for each liquid–rod combination. These values will be used to generate the relevant plots.

As stated before, the maximum frequency, f_{max}, is assumed to be 100 kHz ($\omega_{max} = 0.63$ rad μs^{-1}). The corresponding values of Ω_{max} for each liquid are listed in table 3. For a given liquid, we are interested in the value of the torque constant C_s only up to Ω_{max}. It is seen that the range of Ω of interest for toluene is much smaller than that for water and chloroform due to the smaller value of ε_f/σ_f of toluene.

Next, the different criteria listed at the end of section 2 will be considered. To this end, the values of certain variables will be assumed. Although a large value of E_0 is desirable to minimize Brownian dispersion in the on state, it is not practical to attain unreasonably large electric fields. Hence, a value of $E_0 = 1$ V μm^{-1} will be considered. Rods with $a = 100$ nm and $b = 5$ nm will be considered here. Temperature is set to 300 K.

Criterion (b) listed at the end of section 2 quantifies the inertia (equation (8*a*)). The values of R_I for each liquid–rod combination are listed in table 4. A small value of R_I is required for the applicability of the analysis used here. Table 4 shows that the condition is satisfied for each case considered here.

Criterion (c) (i.e. equation (9)), leads to the minimum value of Ω to ensure that the rods do not oscillate in response to the AC electric field. To obtain a conservative estimate we use the maximum value of $|C_s|$, from equation (17), for a given rod aspect ratio. The values of Ω_{min}, for the different liquids, are listed in table 3. The corresponding values of the minimum frequency, f_{min}, are also listed for each liquid. It is evident that the allowable frequency range for water (27.13–100 kHz) is smaller than that for toluene (1.26–100 kHz) and chloroform (2.69–100 kHz). This is primarily due to the larger value of the permittivity of water (this will also be obvious from equation (21) to be presented later).

Given that E_0 is fixed, criterion (d) (i.e. equation (12)) leads to the minimum value of $|C_s|$. The values of $|C_s|_{min}$, for each liquid, are listed in table 3. The actual value of $|C_s|$, for a particular rod material and in the frequency range of interest, should be much larger than this minimum value to ensure that the Brownian dispersion is not dominant in the on state. The value of $|C_s|_{min}$ is smallest for water (because it has the largest value of ε_f; see equation (9)), thus indicating a more favourable condition according to this particular criterion (table 3).

Criterion (e) (equation (13)) is regarding the decay time. The values of t_{dec} are also listed in table 3 for each liquid. A small value of t_{dec} is desirable—a condition that is adequately satisfied by all the liquids considered here. It is evident from table 1(a), equation (13) and table 3 that lower the viscosity, the smaller is the decay time. Water has the largest t_{dec} due to larger viscosity.

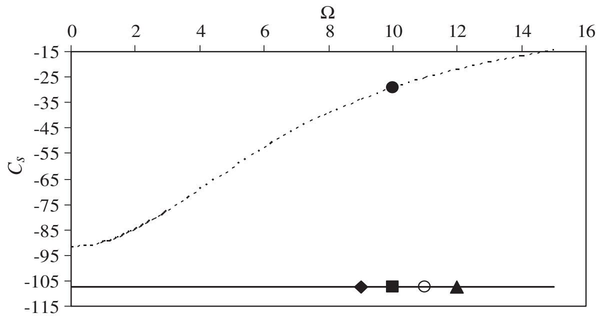

Figure 7. Plot of the torque constant C_s as a function of Ω for different materials in water. The plot of ZnO is denoted by '●', SWCNT by '■', Ge by '♦', Si by '▲' and Au by '○'.

Lastly, to address criterion (a) (equation (2)) the values of the torque constant C_s at different values of Ω for each liquid–rod case were considered. Plots were made (not shown here) for each case up to the maximum value of Ω for that liquid (table 3). It was found that the value of C_s remained close to $C_{s,max} \sim -107$, corresponding to the limit $C_s|_{\Omega\to 0, S_R\to\infty}$, for Au, Ge, Si and SWCNT in all liquids (similar to regime C). This is because of the large values of S_R for these rod materials (table 2). Thus, all these materials result in the best possible value of C_s in the chosen liquids.

In the case of ZnO, the value of S_R is not very high. The value of C_s is negative and remains close to the y intercept value given by equation (15) for ZnO in toluene ($C_s \sim -68$) and chloroform ($C_s \sim -106$). In the case of ZnO in water the value of S_R is moderate, leading to smaller magnitudes of C_s at higher frequencies (figure 7).

Simply obtaining large magnitudes of C_s is not sufficient for the best performance of PLVs. The operating condition should be such that the magnitude of C_s is as far (larger) from $|C_s|_{min}$ as possible so that Brownian dispersion in the on state is minimized. This condition is best satisfied in the case of water since $|C_s|_{min}$ is smallest (2.22, see table 3). It is seen that for ZnO in toluene $|C_s| \sim 68$, which is smaller than $|C_s|_{min} = 76.07$ (table 3) for toluene. This indicates significant Brownian dispersion in the on state for ZnO in toluene.

The value of θ_D^R according to equation (11), calculated with $C_s = C_{s,max}$, also gives an *estimate* of the Brownian dispersion in the on state. These values are listed in table 3 for each liquid. The dispersion is minimum in the case of water. This is due to the larger permittivity of water. The liquid viscosity does not play a role in determining θ_D^R (see equation (12)).

A value of Ω close to Ω_{max} is preferred so that the rods respond only to the average electric field. This condition is best satisfied in toluene since f_{min} corresponding to Ω_{min} is the least for toluene (table 3).

In conclusion, the above analysis suggests that water is the best suspending liquid among those considered due to its higher value of permittivity, resulting in less Brownian dispersion in the on state. High conductivity materials are best for the rods.

3.3. *Effect of the rod size and the aspect ratio*

In the above results, rods of fixed size and aspect ratio were considered. Here the effect of these parameters will be presented.

Table 1. (a) Properties of liquids considered in this work [17, 18]. (b) Properties of rod materials considered in this work [17–19].

(a)	Liquid	Dielectric constant	ε_f (F m^{-1})	σ_f (Ω^{-1} m^{-1})	η (Pa s)
	Water	80.42	7.12×10^{-10}	3×10^{-5}	8.9×10^{-4}
	Toluene	2.35	2.08×10^{-11}	1×10^{-4}	5.6×10^{-4}
	Chloroform	4.81	4.26×10^{-11}	2×10^{-6}	5.37×10^{-4}
(b)	Rod material	Dielectric constant	ε_p (F m^{-1})	σ_p (Ω^{-1} m^{-1})	ρ_p (kg m^{-3})
	ZnO	8.15	7.22×10^{-11}	0.02	5 660
	SWCNT	2.5	2.21×10^{-11}	10^5	1 300
	Ge	16.04	1.42×10^{-10}	188.67	5 323
	Si	11.75	1.04×10^{-10}	1000	2 328
	Au	11.97	1.06×10^{-10}	4.52×10^7	19 300

Table 2. Values of S_R and E_R for different liquid–rod combinations.

		ZnO	SWCNT	Ge	Si	Au
Water	S_R	665.66	3.33×10^9	6.29×10^6	3.33×10^7	1.52×10^{12}
	E_R	−0.899	−0.9688	−0.801	−0.8532	−0.851
Toluene	S_R	199	9.99×10^8	1.89×10^6	9.99×10^6	4.52×10^{11}
	E_R	2.471	0.0647	5.8143	4.0255	4.11
Chloroform	S_R	9999	4.99×10^{10}	9.43×10^7	4.99×10^8	2.26×10^{13}
	E_R	0.696	−0.4798	2.329	1.4553	1.50

Table 3. List of parameters calculated for different liquids.

	Ω_{max}	Ω_{min}	f_{min} (kHz)	$\|C_s\|_{min}$	t_{dec} (ms)	$\theta_D^R\|_{Cs,max}$ (deg)
Water	14.90	4.04	27.13	2.22	0.70	8.26
Toluene	0.13	0.0016	1.26	76.07	0.44	48.31
Chloroform	13.37	0.36	2.69	37.16	0.42	33.77

Table 4. Values of R_I for each liquid–rod combination.

	ZnO	SWCNT	Ge	Si	Au
Water	1.44×10^{-5}	2.94×10^{-3}	1.35×10^{-5}	5.92×10^{-6}	4.91×10^{-5}
Toluene	1.06×10^{-6}	1.37×10^{-4}	1.00×10^{-6}	4.09×10^{-7}	3.62×10^{-6}
Chloroform	2.37×10^{-6}	2.92×10^{-4}	2.22×10^{-6}	9.73×10^{-7}	8.07×10^{-6}

First, it should be noted that small rod sizes are essential to obtain a stable liquid–rod suspension where the gravitational settling is negligible. The increased Brownian motion of small rods helps in 'stabilizing' the suspension. Second, large aspect ratios can help in having high contrast in light transmission as a function of the rod orientation. A spherical particle made of isotropic material will not serve the objective of a PLV device. Thus, small size and high aspect ratios are critical desirable features in PLV devices.

In the following it is analysed how these parameters influence criteria (a)–(e) discussed in section 2. Based on the results in section 3.2, rods with large values of S_R will be considered. Thus, in each of the conditions to be considered below, the value of the torque constant C_s will be assumed to be adequately approximated by $C_{s,max}$ (equation (17)). The following approximate expressions for C_s and K_s [4, 14] for large values of a/b will be used:

$$K_s \approx \frac{1}{3\ln(2a/b)}, \tag{18}$$

$$C_s \approx C_{s,max} \approx -\frac{a^2}{b^2 \ln(2a/b)}. \tag{19}$$

Equation (19) implies that the larger the aspect ratio, the larger is the magnitude of C_s. The stable orientation is as desired because C_s is negative (criterion (a)).

Using these expressions in equation (8*a*) (criterion (b)) we get

$$R_I = \frac{\rho_p E_0^2 \varepsilon_f}{24\eta^2} a^2 \left(\frac{b}{a}\right)^2 \ln\left(\frac{2a}{b}\right). \tag{20}$$

The value of R_I is thus smaller for smaller values of a and larger values of a/b.

Criterion (c) in equation (9) becomes

$$\frac{12\eta\omega}{E_0^2 \varepsilon_f} \gg 1. \tag{21}$$

Thus, the minimum frequency is independent of the rod size or its aspect ratio. This is because the leading order variation of $a^2 K_s/b^2$ is the same as that of C_s under the current approximations. This also implies that the value of t_R, according to equation (7), does not depend on the rod size or its aspect ratio.

Criterion (d) in equation (12) becomes

$$\frac{3k_B T \ln(2a/b)}{\pi a^3 E_0^2 \varepsilon_f} \ll 1. \tag{22}$$

Smaller values of a and larger values of the aspect ratio are unfavourable for this condition. This is a direct consequence of the fact that the Brownian dispersion increases for small a and large a/b. Thus, stronger electric fields will be required to ensure the satisfaction of equation (22).

Condition (e) according to equation (13) gives t_{dec}. Smaller rods with large aspect ratios have shorter decay times.

4. Conclusions

The objective of this work was to analyse the electro-orientation in PLVs with appropriate models and to make recommendations for its optimal functioning. Both the electrical and fluid mechanics issues were considered. The dipole moment theory was used to analyse the electrohydrodynamic problem theoretically. Constant isotropic properties of the materials were assumed. A dilute concentration limit was considered so that interparticle interaction effects are not important. Further generalizations like including the effect of Joule heating, electro-osmosis, electro-thermal behaviour etc are relegated to future work.

A liquid–rod suspension was considered. Various criteria were proposed to design a PLV device. Based on the analysis, the following general conclusions can be drawn.

(i) Good electric conductors like metals (e.g. gold), semiconductors (e.g. Ge, Si) and nanotubes can be excellent rod materials for PLV applications. They lead to the appropriate orientation of the rods in the on state and at the same time have maximum orientational torque for a given rod size and aspect ratio.

(ii) For low conductivity rod materials, an appropriate choice of the operating frequency must be made (section 3.1). In general, the values of S_{R} and E_{R} should be greater than order unity to ensure that the rods do not achieve the undesirable long axis perpendicular to the electric field orientation.

(iii) The Brownian dispersion in the on state is a consequence of the competition between the rotational Brownian motion and the orientational torque due to the electric field. It is found that the Brownian dispersion in the on state is independent of the liquid viscosity but depends on the liquid permittivity (equation (12)). This result is consistent with prior results on dispersion of particles in force fields [16]. Thus, water like liquids with higher permittivities are appropriate choices as suspending liquids since the corresponding Brownian dispersion in the on state is smaller.

(iv) Once the electric field is turned off, the rods acquire random orientations by rotational Brownian motion. The time it takes the rods to fully diffuse in the orientational space depends on the liquid viscosity (equation (13))—lower values are preferred.

(v) It must be ensured that the operating frequency f is such that the rods respond only to the average orientational torque. Liquids with larger viscosity and smaller permittivities (equation (9)) offer a greater range of permissible operating frequencies. The minimum frequency and the response time are almost independent (equation (21)) of the rod size or its aspect ratio (for sufficiently large values of the aspect ratio).

(vi) As expected, the rod inertia is always negligible for the 100 nm scale rods considered here.

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