Electrokinetics Enhances Cross-stream Particle Migration in Viscoelastic Flows

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Advancements in understanding the lateral migration of particles have helped in enhanced separation in microfluidic devices. In this work, we investigate the effects of electrokinetics on the particle migration in a viscoelastic flow, where the electric field is applied parallel to the flow. Through experiments and theoretical analysis, we show that the interaction of electrokinetic and rheological effects can result in an enhancement in migration by an order of magnitude. The theoretical analysis, in agreement with experiments, demonstrates that the particles can be focused at different equilibrium positions based on their intrinsic electrical properties.

In the past two decades, biological and healthcare research has significantly benefited by the application of various particle and cell sorting techniques—inertial focusing [1–3] and viscoelastic focusing [4, 5]—in microfluidic devices. The working principle of such devices is based on the non-linearity in the flow (arising from inertial or polymeric stresses) which breaks the symmetry and generates a lateral force on the particle, pushing it towards the equilibrium positions. Since most biological fluids are non-Newtonian, it is essential to understand rheological effects on particle migration. The pioneering studies of Mason and his coworkers [6, 7] and Ho and Leal [8] clearly demonstrated that the presence of polymer chains in the fluid exerts a normal stress on the particle, pushing it towards the region of lowest shear. In biological fluids, strands of DNA enact the role of polymeric chains and help in the lateral migration of cells [9]. In recent years, this normal stress induced migration has been exploited to design tubular-viscoelastic-focusers, in which the region of lowest shear is the channel axis [10]. However, the labor-intensive fabrication process limits the accessibility of these devices [11]. The more readily available rectangular microchannels are plagued by a limitation of multiple equilibrium positions (1 center and 4 corners), which renders the downstream detection and separation difficult [12, 13]. Furthermore, the passive nature of the migration requires large focusing length which are dependent on the flowrate [10].

Unlike flows at large scale, in micro-scale flows, the short-ranged effects near the particle surface can determine their fate. One can resort to manipulation of surface effects through external fields to tune particle focusing [14]. This has potential applications in focusing, separation and detection of circulating tumor cells in the blood stream [15]. In our previous study [16], we studied the effects of electric fields on the inertial migration in pressure driven flow of a Newtonian fluid. Since the biological fluids exhibit non-Newtonian behaviour, it is of importance to understand the influence of electrophoresis on migration in complex fluids. Specifically, we want to find the role of electrical properties (such as zeta potential), flow profile characteristics (shear and curvature of the flow), and examine whether electrokinetic effects hasten or retard the cross-stream migration in viscoelastic flows.

In this letter, we experimentally and theoretically investigate the influence of electrophoresis on viscoelastic focusing of particles, where the electric field is applied parallel to the flow. A vast majority of cells and particles, when submerged in aqueous electrolytes, acquire a surface charge which attracts counter-charged ions. As a result, a thin double layer forms around the particle [17]. An external electric field causes a tangential slip on the surface of the particle and consequently generates a disturbance field around the particle which can amplify the asymmetry in normal stresses. Our experimental results demonstrate that the focusing is significantly accelerated which reduces the overall focusing length. Furthermore, it provides an external control over particle migration and enables charge-selective focusing. We explain the experimental observations qualitatively and quantitatively using a classical mathematical approach. We analyze the system in the limit of slow and slowly varying flows [8, 18] and capture the viscoelastic effects using a second order fluid (SOF) model. This allows us to predict the migration velocity through a perturbative solution. Using Lorentz reciprocal theorem we show that the electrophoretic motion of the particle generates additional polymeric stresses which enhance the migration by \(O(\kappa^{-1})\), where \(\kappa\) is the particle to channel size ratio. We show that the theoretical results are in reasonable agreement with the experiments, without having to resort to fit any parameter.

In our experiments, we use a 66 \(\times\) 54 \(\mu\)m\(^2\) and 2 cm long PDMS microchannel. We supply the electric fields by a high-voltage DC power supply using a wire connection to electrodes at the channel inlet and outlet. A dilute suspension of polystyrene spheres of 2.2 \(\mu\)m diameter is suspended into a 250 ppm PEO solution (2 \(\times\) 10\(^6\) g/mol) and is pumped at 25 \(\mu\)L/h. The particle zeta potential is -83 mV. Digital videos are recorded at a rate of 15 frames per second, which generate the snapshots and superimposed images [19].

Fig.1(a) shows the particles at the inlet and outlet of

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i.e., the channel when no electric field is applied i.e., the focusing is purely elastic [6-8]. The particles remain almost uniformly distributed at the channel exit. This weak focusing is depicted in Fig.1(d), where the particle stream width. In Fig.1(b) an application of electric field parallel to the flow shows that a significant fraction of the particles are focused at the centerline. Fig.1(d) shows that introduction of electric field results in substantial reduction in particle stream width [19]. Upon reversing the direction of the electric field (see Fig.1(c)), the particles focus near the walls. These results demonstrate the potential to efficiently separate particles and cells based on their intrinsic electrical properties. Now the question is: how can this be explained?

In what follows, we formulate a mathematical framework to predict, quantify and draw physical insights into the migration behavior observed in the experiments. Since the particle suspension in the experimental study is dilute, we analyze the migration of a single particle. Fig.1(e) shows the schematic of an electrophoretic sphere ( neutrally buoyant) suspended in an inertia-less pressure driven flow of SOF, subjected to a parallel electric field ($E_\infty^*$). The fluid moves with a maximum velocity of $U_{\max}^*$ at the centerline. The frame of reference is placed at the center of the particle. PDMS micro-channel walls and polystyrene particles usually acquire a surface charge (formally quantified as zeta-potential ($\zeta^*$) when in contact with an electrolytic solution [20], which gives rise to an electrical double layer (EDL) [17]. In comparison to the channel and particle size, the EDL can be assumed to be asymptotically thin i.e., $\lambda_\text{EDL}/a^* \ll 1$ (where $\lambda_\text{EDL}$ is the EDL thickness). Under the influence of an external electric field, the ions inside the EDL slip in the tangential direction, known as electro-osmotic slip. For low zeta potentials ($\zeta^* \sim 25$ mV), the slip at the particle surface can be represented by Smoluchowski’s relation [17, 21, 22].

We represent the system in terms of dimensionless variables, where the coordinates, potential, velocity and pressure are non-dimensionalized using $a^*$, $|E_\infty^*|a^*$, $\kappa U_{\max}^*$ and $\mu^* U_{\max}^*/a^*$, respectively ($\mu^*$ being the dynamic viscosity). The dimensionless parameters $De$ and $\delta$ govern the magnitude of normal stresses in SOF [18]. $De$ (Deborah number) is the ratio of viscoelastic time scale ($|\Psi_1^* + \Psi_2^*|/\mu^*$) to the time scale corresponding to the average shear in the background flow ($l^*/U_{\max}^*$), and $\delta$ is a viscometric parameter ($-\Psi_1^*/2(\Psi_1^* + \Psi_2^*)$). Here $\Psi_1^*$ and $\Psi_2^*$ are the steady shear viscometric coefficients [18].

We analyze the system in terms of disturbance variables (i.e. deviation from the undisturbed flow): potential ($\psi = \phi - \phi_\infty$), velocity ($v = U - V_\infty$) and pressure ($p = P - P_\infty$). The undisturbed potential distribution ($\phi_\infty$) is generated solely due to the external electric field: $-E_\infty x$. The undisturbed flow profile ($V_\infty$) is identical to that of a Newtonian fluid:

$$V_\infty = (\alpha + \beta z + \gamma z^2)e_x + Ha\zeta_w \nabla \phi_\infty - U_s.$$  (1)

Here, $\zeta_w$ is the dimensionless wall zeta potential ($\zeta^*/|E_\infty^*|a^*$) and $Ha$ is the Hartmann-type number: $Ha = \varepsilon^* E_\infty^2 a^*/4 \pi \mu^* U_{\max}^*$ (i.e. the ratio of electrical energy density to viscous shear stress), where $\varepsilon^*$ is the electrical permittivity. In (1), $Ha\zeta_w \nabla \phi_\infty$ represents the electro-osmotic slip at the wall; $\alpha = 4 s(1 - s)/\kappa$ (here $s$ represents dimensionless particle to wall distance: $d^*/l^*$);
\( \beta \) and \( \gamma \) represent the shear and curvature in the undisturbed flow: \( \beta = 4(1 - 2s) \) and \( \gamma = -4s \).

We now describe the equations governing the disturbances. Since the bulk fluid medium is electroneutral, the disturbance potential is governed by the Poisson equation: \( \nabla^2 \psi = 0 \). Far away from the particle, \( \psi \) decays to zero. Applying Gauss law at the surface of the particle and walls (i.e., slip plane of the double layer) yields the following boundary conditions:

\[
e_{r} \cdot \nabla \psi = - e_{r} \cdot \nabla \phi_{\infty} \quad \text{at } r = 1
e_{z} \cdot \nabla \psi = 0 \quad \text{at walls.}
\]

The disturbance flow is governed by continuity (\( \nabla \cdot \mathbf{v} = 0 \)) and Cauchy’s momentum equation (\( \nabla \cdot \sigma = 0 \)). Here \( \sigma \) is the hydrodynamic stress tensor which contains the Newtonian and polymeric contributions:

\[
\sigma = -p e^{(1)} + De(\Sigma).
\]

Here, \( \Sigma \) is the polymeric stress tensor [19] and \( e^{(1)} \) is the rate of strain tensor for the disturbance flow (\( \nabla \mathbf{v} + (\nabla \mathbf{v})^T \)).

The disturbance flow is subject to the following boundary conditions:

\[
(i) \quad \mathbf{v} = \Omega_s \times \mathbf{r} + Ha\zeta_p \nabla (\psi + \phi_{\infty}) - \mathbf{V}_{\infty} \quad \text{at } r = 1,
\]

\[
(ii) \quad \mathbf{v} = Ha\zeta_p \nabla \psi \quad \text{at walls},
\]

\[
(iii) \quad \mathbf{v} \to \mathbf{0} \quad \text{as } r \to \infty.
\]

Here, \( \zeta_p \) is the dimensionless particle surface zeta potential. In view of the characteristic velocity scale (\( \kappa U_{\text{max}}^* \)), \( Ha\zeta_p \) is of \( \mathcal{O}(1) \).

We seek to find the migration at \( \mathcal{O}(De) \) using a regular perturbation expansion. For small values of \( De \), the disturbance field variables (\( \xi \)) are expanded as:

\[
\xi = \xi^{(0)} + De\xi^{(1)} + \cdots.
\]

Here, \( \xi \) represents velocity (\( \psi \)), pressure (\( p \)), translational (\( U_s \)) and angular velocity (\( \Omega_s \)). Since the electrostatic potential is decoupled from the hydrodynamics, we do not perform a similar expansion for \( \psi \). The angular (\( \Omega_s \)) and translational velocity (\( U_s \)) are evaluated by balancing total force and torque on the particle with the rate of translational and angular momentum, respectively, at each order. The force (\( \mathbf{F} \)) and torque (\( \mathbf{L} \)) contains the contributions from both hydrodynamic and Maxwell stresses. Since the potential distribution is not affected by the fluid rheology, the Maxwell force on the particle in a second order fluid is identical to that in Newtonian fluid [16, 23].

In our earlier analysis [16], we found that the Maxwell force acts along the cross-stream direction alone, and Maxwell torque is zero. The contribution of Maxwell stresses to the particle velocity is:

\[
U_{sM} \approx \frac{4\pi Ha}{6\pi(1 + \mathcal{O}(\kappa))} \left( \frac{3\pi}{16} n^4 \left( \mathcal{Z}(4, s) - \mathcal{Z}(4, 1 - s) \right) \right) e_z.
\]

Here, \( \mathcal{Z} \) is the generalized Riemann zeta function. This contribution pushes the particle only in the \( z \)-direction and is always wall-repulsive in nature. Later we show that this contribution decays rapidly away from the walls and is negligible in the bulk of the channel.

To find the contribution of hydrodynamic stresses on the particle migration, we substitute (4) into the equations governing the flow and boundary conditions (3). We obtain a system of equation at \( \mathcal{O}(1) \) and \( \mathcal{O}(De) \) each. The symmetry of Stokes flow implies that the \( \mathcal{O}(1) \) field cannot produce a lateral lift. Therefore, the lift must arise from \( \mathcal{O}(De) \) field. Lorentz reciprocal theorem [8, 16] allows us to determine the migration associated with \( \mathcal{O}(1) \), without having to solve for \( \mathcal{O}(De) \) field. The reciprocal theorem relates the properties of an unknown Stokes flow to a known test flow field, provided both flow fields correspond to the same geometry. The test field (\( u^t, p^t \)) is chosen to be that generated by a moving sphere in the positive \( z \)-direction (towards the upper wall) with unit velocity in a quiescent Newtonian fluid [16]. Following Ho and Leal [8], we obtain the lateral migration velocity at \( \mathcal{O}(De) \):

\[
U_{\text{mig}} = -\frac{1}{6\pi(1 + \mathcal{O}(\kappa))} De \int_{V_j} \Sigma^{(0)} : \nabla \mathbf{u}^t \, dV.
\]

Here, subscript \( H \) denotes the hydrodynamic contribution, \( \Sigma^{(0)} \) is the shear and Maxwell stresses. Since the potential distribution is not affected by the fluid rheology, the Maxwell force on the particle in a second order fluid is identical to that in Newtonian fluid [16, 23].

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\]

Using the definitions of \( \beta \) and \( \gamma \) (see (1) et seq.), we conclude that the first term in the above expression is \( \mathcal{O}(\kappa) \), and is identical to that obtained by Ho and Leal [8], who analyzed the viscoelastic migration of a particle in the absence of electrokinetic effects.

The above expression depicts that the direction of the lateral migration is determined by the magnitude and sign of the viscometric parameter \( \delta \). For most viscoelastic fluids \( De \) is positive, and \( \delta \) is reported to be between -0.5 and -0.7 [18, 26–28]. This renders the first term in (7) strictly positive below the centerline (\( \beta > 0 \)) and negative above it (\( \beta < 0 \)) i.e. the pure elastic stresses push the particle towards the channel axis at \( \mathcal{O}(\kappa) \). In the absence of electric field, the experimental particle stream width depicted in Fig.1(a,d) shows that the particle is subject to a very weak migration towards the center.

The second term is \( \mathcal{O}(1) \); its proportionality to \( Ha\zeta_p \) indicates that this dominant contribution originates due to the electrohydroetic slip i.e., the addition of electrophoresis into the system enhances the migration by \( \mathcal{O}(\kappa^{-1}) \). For \(-0.7 < \delta < -0.5 \), a particle with positive
\( \zeta_p \) will migrate towards the walls (at \( O(1) \)), when the electric field is applied in the direction of the flow. The reverse would hold for a particle with negative \( \zeta_p \) or electric field applied in the opposite direction. These findings qualitatively agree with the experiments depicted in Fig.1.

The enhancement demonstrates that an addition of electrokinetic slip on the particle surface can drastically alter the normal stresses which consequently enhances the particle migration. The slip creates a source-dipole disturbance in the fluid which decays rapidly away from the particle (\( \sim O(1/r^3) \)). This suggests that the migration occurs from short-ranged interactions, which are unaffected by the boundary effects unless the particle is very close to the walls. Furthermore, (7) reveals that the parameters such as wall zeta potential (\( \zeta_{w} \)) and curvature of the background flow (\( \gamma \)) do not affect the migration.

Since the experimental parameters are in the regime of the theoretical analysis (\( De < 1, \ De \gg Re_p, Ha\zeta_p \sim O(1), \kappa \ll 1 \)), we now quantitatively compare the two. Theoretically, the trajectories of a single particle are calculated using (7) and converted to an equivalent particle stream-width using: \( |l^* - 2(l^* - d^*)| \), for particle focusing at the centerline (\( l^* \) being the channel height and \( d^* \) is the particle to wall distance). We use the relaxation time scale (\( (\Psi_1^* + \Psi_2^*)/\mu^* \)) corresponding to \( C_{PEO} = 250 \) ppm as 2.75 ms [29]. Fig. 2(a) shows the comparison of particle stream widths for two cases: (i) No electric field (i.e., viscoelastic focusing) and (ii) parallel electric field (i.e., electro-viscoelastic focusing). For case-(i), the theoretical prediction shows modest particle migration. The experiments also show weak focusing of particles. For case-(ii), the theoretical predictions are in good agreement with the experimental results. We define an enhancement ratio \( (\mathcal{E}) \) as the difference between the particle stream widths before and after applying the electric field, normalized by the channel width. The enhancement in migration is captured qualitatively well by the theoretical model (see Fig.2(b)).

Our mathematical framework reveals that the migration in Fig.1(a-c) arises from: [a] Maxwell stresses (5), [b] pure elastic stresses (eq.7-term1), and [c] electrophoresis induced elastic stresses (eq.7-term2). A comparison of these migration velocities, namely, Maxwell migration (MM), viscoelastic migration (VM) and electro-viscoelastic migration (EVM) is shown in Fig.3. The curves correspond to Fig.1(b): a particle with negative zeta potential and electric field applied parallel to the flow. The migration due to decoupled Maxwell stresses (MM) is dominant near the walls and decays rapidly away from it. Although this wall-repulsive force is negligible in the bulk of the channel, it helps eliminate the corner equilibrium positions [30]. The other components VM and EVM act towards the center as depicted by the negative gradient at the central equilibrium position. For conditions corresponding to Fig.1(c), where flow and electric field are opposite to each other (\( Ha < 0 \)), we see, from (7), the EVM curve reverses. Consequently, the central equilibrium position becomes unstable and particle migrates towards the walls.

To conclude, our experimental and theoretical analysis depict an enhancement of \( O(\kappa^{-1}) \) in migration of a particle in viscoelastic flow, when subjected to a parallel electric field. Using a second-order fluid model, the theoretical analysis reveals the contribution from the underlying stresses around the particle (see eq.5 and eq.7). These constitute an approximate expression for the migration velocity which agrees well with the experimental results (see Fig.2). The analytical results further reveal that the particle zeta potential and background shear determine the equilibrium positions. Wall zeta potential, Maxwell stresses, and curvature of the background flow do not affect the migration at the leading order. These findings have a direct practical relevance to applications related to Lab-on-a-chip devices used to separate cells in biological fluids which are known to exhibit non-Newtonian behavior [31, 32].

The current work addresses the particle migration in dilute polymer concentration regime. As the polymer
concentration increases, the direction of migration is reported to be opposite to that of the current work and is currently unknown [33]. In such cases, constitutive models like Oldroyd-B and Giesekus fluid can be implemented numerically to predict the migration behaviour. In our future studies we intend to address these questions.

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Electrokinetics Enhances Cross-stream Particle Migration in Viscoelastic Flows
Supplementary Material
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In this Supplementary Material we provide the details related to our mathematical framework and validation with the literature. In section I, we demonstrate the procedure for evaluating the disturbance variables using the method of reflections. Section II details the evaluation of the angular and translational velocity of the particle. In section III, we analyze the volume integral which determines the migration velocity. We demonstrate that the particle trajectory exactly matches with that in the literature (i.e., in the absence of electrokinetics). Section IV provides the details of the experimental setup.

I. EVALUATION OF DISTURBANCES

Here, we demonstrate the solution of the field variables \( \psi, \psi_{(0)} \), and \( u^i \). When the particle is not too close to the wall (\( \kappa/s \ll 1 \)), the solution to the disturbance field variables can be sought in terms of successive reflections [S37]. The first reflection represents the disturbance due to a particle in an unbounded domain. The second reflection corrects the previous disturbance by imposing the boundary condition at the walls and ignoring the particle. Subsequently, each reflection imposes the boundary condition alternatively at the particle and walls. Every successive pair of reflections increases the accuracy by \( O(\kappa) \) [S38]. This iterative process is performed until a desired accuracy is attained.

A. Electrostatic potential disturbance

We seek the disturbance potential as a sum of reflections:

\[
\psi = (1)\psi + (2)\psi + \cdots. \quad (S1)
\]

Here, \( (i)\psi \) represents the \( i^{\text{th}} \) reflection. Substituting the above equation in Poisson equation (\( \nabla^2 \psi \)), we obtain for the first reflection:

\[
\begin{align*}
\nabla^2 (1)\psi &= 0, \\
\mathbf{e}_r \cdot \nabla (1)\psi &= -\mathbf{e}_r \cdot \nabla \phi_\infty \quad \text{at } r = 1, \\
(1)\psi &\rightarrow 0 \quad \text{as } r \rightarrow \infty,
\end{align*} \quad (S2)
\]

and for the second reflection:

\[
\begin{align*}
\nabla^2 (2)\psi &= 0, \\
\mathbf{e}_z \cdot \nabla (2)\psi &= -\mathbf{e}_z \cdot \nabla (1)\psi \quad \text{at walls.}
\end{align*} \quad (S3)
\]

Solution to \( (1)\psi \): Equation (S2) suggests that \( (1)\psi \) is a harmonic function which decays as \( r \rightarrow \infty \). The boundary condition suggests that it must be linear in the driving force: \( \nabla \phi_\infty (= -\mathbf{e}_z) \). Therefore, the solution is sought in terms of spherical solid harmonics [S39] as:

\[
(1)\psi = \frac{1}{2} \frac{r}{r^3} \nabla \phi_\infty = -\frac{1}{2} \frac{x}{r^3}. \quad (S4)
\]

Solution to \( (2)\psi \): To evaluate \( (2)\psi \), we adopt the approach devised by Faxén [S35] in the context of bounded viscous flows. Using this, the disturbance around the particle is expressed in an integral form which satisfies the boundary conditions at the walls. The first reflection is characterized by particle scale \( a^* \). The second reflection \( (2)\psi \) is characterized by a length scale \( l^* \). Therefore, the coordinates for second reflection are stretched, and are termed as ‘outer’ coordinates. These outer coordinates (denoted by tilde) are defined as:

\[
\tilde{r} = r \kappa, \quad \tilde{x} = x \kappa, \quad \tilde{y} = y \kappa, \quad \tilde{z} = z \kappa. \quad (S5)
\]

Since \( (2)\psi \) depends on \( (1)\psi \) through the wall boundary condition in (S3), both reflections must be represented in the coordinates of same scale. The first reflection of potential disturbance in the outer coordinates \( (1)\tilde{\psi} \) is:

\[
(1)\tilde{\psi} = -\frac{1}{2} \frac{X}{R^3} \kappa^2. \quad (S6)
\]

Faxén [S35] represented the fundamental solution of Laplaces equation (in Cartesian space) in the form of Fourier integrals. Following Faxén, we write:

\[
\frac{1}{R} = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{i(\Theta - \frac{i\lambda}{2})} \frac{d\xi d\eta}{2\lambda}, \quad \text{and} \quad (S7)
\]

\[
R = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \left(1 - e^{i(\Theta - \frac{i\lambda}{2})}\right) \left(1 + \frac{\lambda |Z|}{2}\right) \frac{d\xi d\eta}{\lambda^3/2}. \quad (S8)
\]

Here, \( \Theta = (\xi X + \eta Y)/2 \) and \( \lambda = (\xi^2 + \eta^2)^{1/2} \). \( \xi \) and \( \eta \) are the variables in Fourier space. Using (S7) and (S8), we transform the disturbance field (S6) by taking derivatives of the above equations. For example: (S8) contains \( X/R^3 \), which is expressed using the above transformations as:

\[
\frac{X}{R^3} = -\frac{\partial}{\partial X} \left(\frac{1}{R}\right) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{i(\Theta - \frac{i\lambda}{2})} \left(-\frac{i\xi}{2}\right) \frac{d\xi d\eta}{2\lambda}. \quad (S9)
\]
This yields an integral representation for the first reflection:

\[
(1) \tilde{\psi} = \frac{\kappa^2}{2\pi} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{i(\eta - \frac{\lambda z}{2})} b_1 \left( \frac{i\xi}{\lambda} \right) \mathrm{d}\xi \mathrm{d}\eta. \tag{S10}
\]

Here, \( b_1 = 1/8 \). The boundary condition expressed in (S3) suggests a form of solution for \((2) \tilde{\psi}\) similar to the above equation:

\[
(2) \tilde{\psi} = \frac{\kappa^2}{2\pi} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} e^{i\eta} \left( e^{-\frac{\lambda z}{2}} b_2 + e^{\frac{\lambda z}{2}} b_3 \right) \left( \frac{i\xi}{\lambda} \right) \mathrm{d}\xi \mathrm{d}\eta. \tag{S11}
\]

Substituting (S10) and (S11) in the boundary condition (S3), yields \( b_2 \) and \( b_3 \) in terms of \( b_1 \):

\[
b_2 = \frac{b_1 \left( 1 + e^{(1-s)\lambda} \right)}{-1 + e^{\lambda}}, \quad b_3 = \frac{b_1 \left( 1 + e^{s\lambda} \right)}{-1 + e^{\lambda}}. \tag{S12}
\]

The next reflection \((3) \psi\) is \( O(\kappa^3) \). As we restrict \( \kappa \ll 1 \), the first two reflections capture the leading order contribution to the disturbance potential.

\[\text{\textbf{B. Velocity disturbance}}\]

We seek the disturbance \((v_0, p_0)\) as successive reflections:

\[
v_0 = (1)v_0 + (2)v_0 + \cdots, \quad p_0 = (1)p_0 + (2)p_0 + \cdots. \tag{S13}
\]

Upon substituting the above expansion in the equations governing the flow (continuity and Cauchy equation), we obtain the following set of problems:

\[
\begin{aligned}
\nabla \cdot (1)v_0 &= 0, \quad \nabla^2 (1)v_0 - \nabla(1)p_0 = 0, \\
(1)v_0 &= U_{sz}(0)e_x + \Omega_{sy}(0)e_y \times r + \mathcal{H}_\nu(\nabla \psi_1 + \nabla \psi_2) \\
-(\alpha + \beta z + \gamma z^2) + \mathcal{H}_\nu(\zeta_p - \zeta_w) e_x & \quad \text{at } r = 1, \\
(1)v_0 &\rightarrow 0 \quad \text{at } r \rightarrow \infty.
\end{aligned} \tag{S14}
\]

\[
\begin{aligned}
\nabla \cdot (2)v_0 &= 0, \quad \nabla^2 (2)v_0 - \nabla(2)p_0 = 0, \\
(2)v_0 &= \mathcal{H}_\nu(\nabla \psi_1 + \nabla \psi_2) - (1)v_0 & \quad \text{at the walls}, \\
(2)v_0 &\rightarrow 0 \quad \text{at } r \rightarrow \infty.
\end{aligned} \tag{S15}
\]

The expression for \((1)v_0\) is found by employing Lamb’s general solution [S25]:

\[
(1)v_0 = A \left( e_x + \frac{xr}{r^2} \right) \frac{1}{r} + B \left( -e_x + \frac{3xr}{r^2} \right) \frac{1}{r^3} + C \left( \frac{xe_x}{r^3} - \frac{x e_z}{r^3} \right) \frac{1}{r^3} + D \frac{zx r}{r^3} + E \left( 2e_x + xe_z - \frac{5zx r}{r^2} \right) \frac{1}{r^5}
\]

\[
+ F(e_x - \frac{2z^2e_x + xr}{r^2} + \frac{2zx e_z}{r^2}) \frac{1}{r^3} + G(e_x - \frac{5z^2 e_x + 10zx e_z + 13zx r}{r^2} + \frac{75zx^2 r}{r^4}) \frac{1}{r^5}
\]

\[
+ H(e_x - \frac{5z^2 e_x + 10zx e_z + 5zx r}{r^2} + \frac{35zx^2 r}{r^4}) \frac{1}{r^5}. \tag{S16}
\]

Here, the coefficients are:

\[
A = \frac{3}{4}(U_{sz}(0) - \alpha - \gamma/3), \quad B = -\frac{1}{4}(U_{sz}(0) - \alpha - 3\gamma/5 - 2H_\nu z_p),
\]

\[
C = \Omega_{sy}(0) - \frac{\beta}{2} D = -\frac{5\beta}{2} E = -\frac{\beta}{2} F = \frac{\gamma}{3},
\]

\[
G = -\frac{7\gamma}{120}, \quad H = \frac{\gamma}{8}. \tag{S18}
\]

Similar to previous section, \((2)v_0\) can be found using Faxen transformation [S16]. In § III, we show that the higher reflection of the velocity field are dispensable.

\[\text{\textbf{II. EVALUATION OF } U_{sz}(0) \text{and } \Omega_{sy}(0)}\]

The translational and angular velocity can be found by imposing force-free and torque-free conditions on the particle at \( O(De^0) \): \( \mathbf{F}_H(0) + \mathbf{F}_M = 0 \) and \( \mathbf{L}_H(0) + \mathbf{L}_M = 0 \).

\[\text{\textbf{A. Maxwell force and torque}}\]

The electric force and torque are expressed as a function of reflections as:
Substituting (S4) and (S11) in both (S19) and (S20), we obtain:

\[ F_M = 4\pi Ha \int \mathbf{n} \cdot \left( \nabla \left( (1)\psi + (2)\psi \right) \nabla \left( (1)\psi + (2)\psi \right) \right) \mathbf{l} \, dS, \]  
\[ L_M = 4\pi Ha \int \mathbf{n} \times \left( \mathbf{n} \cdot \left( \nabla \left( (1)\psi + (2)\psi \right) \nabla \left( (1)\psi + (2)\psi \right) \right) \right) \mathbf{l} \, dS. \]  

Substituting (S4) and (S11) in both (S19) and (S20), we obtain:

\[ F_M = 4\pi Ha \left( \frac{3\pi}{16} \kappa^4 \left( 3(4, s) - 3(4, 1 - s) \right) \right) e_z + O\left( \kappa^7 \right), \]  

\[ L_M = 0. \]  

Here, \( \zeta \) is the generalized Riemann zeta function \([S40]\). This leading order electrical force acts only in the \( z \)-direction and always away from the walls (i.e., positive below the channel centerline and vice-versa). As the particle approaches walls \((s \to 0 \text{ or } 1)\), (S21) asymptotically matches with Yariv’s single wall expression \([S23]\). We also find that the Maxwell stresses exert no torque on the particle, at the present level of approximation.

### B. Hydrodynamic force and torque

For a particle-wall system, Happel and Brenner \([S38, \text{p. 239}]\) derived the hydrodynamic force and torque on a spherical particle in terms of successive reflections:

\[ F_H(0) = (1)F_H(0) + (3)F_H(0) + \cdots, \quad \text{and} \]
\[ L_H(0) = (1)L_H(0) + (3)L_H(0) + \cdots. \]  

Since the particle is absent in the formulation of even-numbered reflections, the hydrodynamic drag and torque, due to even fields vanishes. Thus, the contribution to force and torque arises only from the odd-numbered reflections. In the current problem \((3)F_H(0)\) and \((3)L_H(0)\) are dispensable. An interested reader can refer our earlier work \([S16]\) for further details.

### C. Force and torque balance

Since the electric force \((S21)\) acts only along the \( z \)-axis and the electric torque \((S22)\) is zero, \( U_{sx}(0) \) and \( \Omega_{sx}(0) \) are found by hydrodynamic force and torque balance in \( x \) and \( y \) directions, respectively. Imposing the hydrodynamic force and torque to be zero and following Kim and Karrila \([S41, \text{p. 88}]\), we obtain:

\[ U_{sx}(0) = \alpha + \gamma/3 + Ha(\zeta_p - \zeta_w) + O(\kappa^3) \]
\[ \Omega_{sx}(0) = \beta/2 + O(\kappa^4). \]  

### III. Migration Velocity Volume Integral

Using the Lorentz reciprocal theorem, we obtain the following:

\[ U_{LH} = -\frac{1}{6\pi(1 + O(\kappa))} De \int_{V_f} \Sigma_{(0)} : \nabla \mathbf{u}' \, dV \]  

We substitute \( \mathbf{u}_{(0)} \) into \( \Sigma_{(0)} \) and evaluate the migration velocity by performing the volume integration \((S25)\).

The polymeric stress is defined as:

\[ \Sigma = \mathbf{e}^{(1)} : \mathbf{e}^{(1)} + \mathbf{w}^{(1)} + \delta(\mathbf{e}^{(2)} + \mathbf{w}^{(2)}). \]  

The different terms, \( \mathbf{e}^{(2)} \), \( \mathbf{w}^{(1)} \) and \( \mathbf{w}^{(2)} \) are the various components of the polymeric stress:

\[ \mathbf{e}^{(2)} = \mathbf{v} \cdot \nabla \mathbf{e}^{(1)} + \mathbf{e}^{(1)} \cdot \nabla \mathbf{v} + \nabla \mathbf{v} \cdot \mathbf{e}^{(1)}, \]
\[ \mathbf{w}^{(1)} = E^{(1)} \cdot \mathbf{e}^{(1)} + \mathbf{e}^{(1)} \cdot E^{(1)}, \]
\[ \mathbf{w}^{(2)} = \nabla \mathbf{v} \cdot \nabla \mathbf{e}^{(1)} + \mathbf{e}^{(1)} \cdot \nabla \mathbf{v} + \nabla \mathbf{v} \cdot \mathbf{e}^{(1)} + \mathbf{v} \cdot \nabla E^{(1)} + \mathbf{v} \cdot \nabla \mathbf{e}^{(1)} + \nabla \mathbf{v} \cdot \mathbf{e}^{(1)}. \]

Here, \( \mathbf{E}^{(1)} \) and \( \mathbf{e}^{(1)} \) are the rate of strain tensor for the undisturbed and disturbance flow, respectively; \( \mathbf{e}^{(2)} \) is the disturbance Rivlin-Eriksen (RE) tensor; \( \mathbf{w}^{(1)} \) is the ‘interaction tensor’ arising from the interaction between background flow and disturbance flow; and \( \mathbf{w}^{(2)} \) is the RE interaction tensor.

For convenience, we divide the fluid domain \((V_f)\) into two asymptotic sub-domains \([S8, S36]\):

\[ V_1 = \{ \mathbf{r} : 1 \leq |\mathbf{r}| \leq \rho \}, \quad V_2 = \{ \mathbf{r} : \kappa \rho \leq |\mathbf{r}| \leq \infty \}. \]

Here, the intermediate radius \( \rho \) satisfies: \( 1 \ll \rho \ll 1/\kappa \).

The characteristic length scale corresponding to the inner domain \((V_1)\) is particle radius \( a^*\). Whereas, channel width \( l^*\) is the characteristic length corresponding to the outer domain \((V_2)\).
The order of magnitude for the integrand $\Sigma(0): \nabla u^t$ is:

$$\sim (Ha_\beta)^2 \mathcal{O}\left(\frac{1}{r^{10}} + \delta \frac{1}{r^7}\right) + \beta \mathcal{O}\left(\frac{1}{r^6} + \frac{1}{r^6}\right) +$$

$$\gamma Ha_\beta \mathcal{O}\left(\frac{1}{r^5} + \frac{1}{r^5}\right) + \beta^2 \mathcal{O}\left(\frac{1}{r^5} + \frac{1}{r^5}\right) +$$

$$\beta \gamma \mathcal{O}\left(\frac{1}{r^4} + \frac{1}{r^6}\right) + \mathcal{O}(\kappa^4).$$  \hfill (S28)

Upon integrating the above expression in $V_1$, we find that the contribution from all the terms vanishes except the second and fifth term: Upon evaluating (6) in $V_1$, we obtain:

$$\frac{6\pi(1 + \mathcal{O}(\kappa))U_{LH}}{Dc} = \beta \gamma \left(\frac{10\pi}{3} + \delta \pi \right) - \frac{3\pi}{2} \beta Ha_\beta (1 + \delta)$$

$$+ \mathcal{O}(\kappa^4).$$  \hfill (S29)

In view of the definition of $\beta$ and $\gamma$ (see main text), the first term in the above expression is $\mathcal{O}(\kappa)$, and is identical to that obtained by Ho and Leal [S8], who reported the viscoelastic migration of a particle in the absence of electrokinetic effects. The second term is $\mathcal{O}(1)$, which dominates the migration in the current study.

We now find the contribution of the integral in the outer domain $V_2$. Since the characteristic length in the outer domain is $l^*$, the disturbances are stretched into the outer coordinates ($\tilde{r} = r\kappa$). Upon substituting $\Sigma(0)$ and $\tilde{u}^t$ (expressed in outer coordinates) into the volume integral, we find that the integrand is $\mathcal{O}(\kappa^4)$.

Since the dominant contribution arises from the inner integral (i.e. $\mathcal{O}(1)$), the contribution from the outer domain can be neglected. Hence the migration velocity due to hydrodynamic stresses is represented by (S29).

**Validation with the literature:** Fig. S1 depicts the stream width of particles suspended in viscoelastic flow, in absence of electric field. The result matches well with that reported by Ho and Leal [S8] (The particle stream width is converted from trajectory using $|l^* - 2(l^* - d^*)|$). Upon increasing the accuracy (i.e., incorporating leading order wall effects into account), we deviate slightly from Ho and Leal [S8]. In the present work, we include the wall effects.

**IV. EXPERIMENTAL DETAILS**

The particle suspension was prepared by suspending polystyrene spheres (Thermo Scientific) in the viscoelastic solution. We dissolved polyethylene oxide (PEO, molecular weight = $2 \times 10^6$ g/mol, Sigma-Aldrich) powder into a buffer solution (0.01 mM phosphate buffer mixed with 0.5% Tween 20 (Fisher Scientific)) at a concentration of 250 ppm. The particle concentration was kept low (<0.1% in volume fraction) in the fluid, and hence the particle-particle interaction and its effect on fluid viscosity can be neglected. The microchannel was primed with the particle-free suspending fluid prior to the introduction of the particle suspension. The microchannel used in the experiment was 2 cm long with a rectangular cross-section of 66 µm × 54 µm. Particle motion was visualized at the outlet of the microchannel through an inverted microscope (Nikon Eclipse TE2000U) equipped with a CCD camera (Nikon DS-Qi1MC). Digital videos were recorded at a rate of 15 frames per second, from which the superimposed images could be obtained and further processed in the Nikon imaging software (NIS-Elements AR 3.22).

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