Expanding the flexibility of dynamics simulation on different size particle–particle interactions by dielectrophoresis

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Abstract

In this paper, we perform flexible and reliable dynamics simulations on different sizes of two or more particles' interactive motions, where they encounter positive or negative dielectrophoresis (DEP) forces. The particles with identical or non-identical size are in close proximity suspended freely in a solution under a homogeneous electric field. According to the description of classic dipole moment, DEP forces make the particles form a straight chain. Therefore, dynamics simulation based on Newton's laws is utilized to understand AC DEP phenomena among multiple particles. To solve the relevant governing equations, Stokes drag and repulsive forces (including wall and particles) are combined with DEP forces to obtain the trajectories of particles. Results show that particles with the same sign of the Clausius–Mossotti (CM) factor revolve clockwise or counterclockwise to attract each other parallel to the electric field direction. Conversely, the particle chain is perpendicular to the field. This programmable advantage is of great benefit to the study of three or four particle motions. Meanwhile, the pearl chain consisting of three or four particles is related not only to an individual CM factor but also to initial spatial configuration. Both the cluster and short chain are dependent on symmetry between the geometric distribution and electric field, while it implies different size particles easily cause the chain structure with less time.

Keywords Dielectrophoresis · DEP · Pearl chain · Programming flexibility · Dynamics simulation

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1 Introduction

Dielectrophoresis (DEP), as initially defined by H. A. Pohl, was of extreme interest for the recent development of a lab-on-a-chip device for biological investigations and applications, such as concentration, separation, sorting, and transportation for micro/nano-sized cells, protein, DNA, and colloid particles [1, 2]. To manipulate these polarizable electroneutral particles, relying on DEP forces towards or away from electric field maxima has gradually become a valuable approach for pathologic analysis and clinic diagnosis. Furthermore, DEP technology has provided a new insight into the set-up of clusters or chains from many particles in biological morphology research. The biomimetic heterogeneous pattern, which stems from tissue engineering, proposes the high demand of an inerratic array structure from randomly distributed cells exerted by DEP forces. Hence, Ho et al. [3] presented a novel microfluidic chamber to generate the DEP effect, manipulating the original randomly distributed hepatocytes and leading them into the desired pearl-chain array pattern. The reconstructive tissues can gain considerable economic benefits, for example, an artificial regenerated organ [4, 5]. Meanwhile, micro/nano-particle manipulation by using a DEP force can be adopted for fabricating a high-quality multifunctional biosensor and electronic device. For example, alignment of carbon nanotubes (CNTs) can be used in the field of humidity measurement [6, 7].

In general, the magnitude of a non-uniform electric field is an important factor for DEP manipulation. Different from a non-uniform electric field generated by asymmetric electrodes, assembling some of the free particles, chain form of closely spaced particles, can be attributed to a uniform electric field. Currently, there are two primary physical interpretations for attractive or repulsive particle–particle interactions. One approach mainly considers the distortion of the applied electric field in the vicinity of each particle. Hossan et al. [8] and Ai et al. [9, 10] pointed out that charges unevenly accumulate in the interface between the particle and suspending media under a uniform field due to their different dielectric properties. Equivalently, the presence of each particle in close proximity results in the distortion of the electric field, so that local non-uniform fields induce these particles to form pearl chain structures. On the other hand, the description from Jones et al. [11] focused on dipole–dipole interactions, which stems from the ‘action-at-a-distance’ physical interpretation of electrostatics. In fact, there are many direct numerical models to calculate and simulate the chain formation in terms of particle–particle motions, as shown in Table 1.

The results of the particle chain, including geometric shape, applied frequency, and particle number, have been further studied. For instance, Xie et al. [16] and Liu et al. [20] combined the flexibility of effective dipole moment with the accuracy of the Maxwell stress tensor (MST) to simulate double or abundant particle trajectories. Taking into account the iterative dipole moment, the mutual DEP force for any two particles is also in good agreement with the MST method. In addition, Gutierrez et al. [19] proposed a novel numerical model for the help of both the electromagnetism and the solid mechanics modules from COMSOL finite element software, which provided insight into chain formation from such elliptical particles, including size, aspect ratio, and heterogeneity. In contrast to either ALE or IIM numerical models, both dynamics simulation (DS) and Monte Carlo (MC) methods can be used for an overwhelming amount of particles suspended in an aqueous solution [1, 15, 22]. In our previous work [23], the MC method was used to describe the behavior of carbon nanotubes and their arrangement when the total system energy decreases and becomes stable. However, a study is not available on the time-dependent evaluation of particle trajectories. Although Hossan et al. [8], Ai et al. [9, 10], and Kang et al. [14] attempted to obtain highly reliable DEP forces by using the MST method, this decreases the universalism of the numerical model. Since the mesh generation with regard to existed particles from Multiphysics

Table 1 Numerical models and DEP forces used in previous studies to interpret the behaviors of particles suspended in aqueous solution

Researchers	DEP force	Algorithms
Juarez et al. [12]	PD	MC
Hossan et al. [8]	MST	IIM + FDM
Ai et al. [9, 10]	MST	ALE+FEM
Camarda et al. [13]	PD	DS
Kang et al. [14]	MST	SIM + FVM
Hu et al. [15]	PD	MC
Xie et al. [16]	MST	ALE+FEM
Xie et al. [17, 18]	IDM	FEM
Gutierrez et al. [19]	PD	ALE+FEM
Liu et al. [20, 21]	IDM	FEM
Belijar et al. [22]	PD	DS

PD point dipole, *MST* Maxwell stress tensor, *IDM* iterative dipole moment, *ALE* arbitrary Lagrangian Euler, *FEM* finite element method, *FVM* finite volume method, *SIM* sharp interface method, *MC* Monte Carlo, *DS* dynamics simulation

COMSOL or other finite element software (FLUENT and ANSYS) is still ambiguous, a re-mesh error resulting from a moving mesh in the computational domain occurs owing to an inverted mesh element near coordinates. According to Belijar's group's results, the simulated particle distributions are consistent with experimental measurement. In the literature, an intensive systematic numerical model based on classical Newton law has rarely been conducted on particle trajectory with a few particles in close proximity under the AC electric field. Therefore, we numerically built three-dimensional DEP motions from two to four particles. Compared to the ALE model with time-consuming calculations, the programmable DS model flexibly adjusts the size and position of any particle, simultaneously studying the particle chain.

2 Theory and model

Dynamics simulation has become a mainstream form of understanding particle behaviors and structure in the microscopic world. As mentioned above, the term DEP relates to a ponderomotive force exerted by a non-uniform electric field on polarizable neutral particles. As far as the effective dipole moment is concerned, the mutual DEP force between two particles can be expressed in Eq. (1) [24] as:

$$\mathbf{F}_{DEP,ij} = \frac{3}{4\pi\epsilon_m r_{ij}^5} \left(\mathbf{r}_{ij} (\mathbf{p}_i \cdot \mathbf{p}_j) + (\mathbf{r}_{ij} \cdot \mathbf{p}_i) \mathbf{p}_j + (\mathbf{r}_{ij} \cdot \mathbf{p}_j) \mathbf{p}_i - \frac{5}{r_{ij}^2} \mathbf{r}_{ij} (\mathbf{p}_i \cdot \mathbf{r}_{ij}) (\mathbf{p}_j \cdot \mathbf{r}_{ij}) \right) \quad (1)$$

where ϵ_m is the permittivity of the medium, $\mathbf{r}_{ij}$ is a distance vector between the center of particle i and the center of particle j , and scalar quantity r_{ij} equals $|\mathbf{r}_{ij}|$. The effective dipole moment $\mathbf{p}_i$ for particle i can be expressed in Eq. (2) [25] as:

$$\mathbf{p}_i = 4\pi a_i^3 \epsilon_m \text{Re}[f_{CM}] \mathbf{E} \quad (2)$$

where a_i is the radius of particle i , $\mathbf{E}$ is the external applied electric field. The parameter f_{CM} is the well-known Clausius–Mossotti (CM) factor, which determines the direction of the DEP force. The CM factor, dependent on inherent dielectric properties from both the particle and medium, is given in Eq. (3) [25] as:

$$f_{CM} = \left(\varepsilon_{p,i}^* - \varepsilon_m^* \right) / \left(\varepsilon_{p,i}^* + 2\varepsilon_m^* \right) \quad (3)$$

where the complex permittivity ε^* is $\varepsilon + j\sigma/\omega$ (the subscripts p and m stand for particle and medium, respectively) and ω is the angular frequency equal to $2\pi f$ (f is applied frequency). In the same way, the effective dipole moment $\mathbf{p}_j$ for particle j is also obtained by Eqs. (2) and (3). In this paper, the objective particle is considered as the polystyrene (PS) bead. Since the induced field mobility of counterions in the electric double layer brings about the change of surface conductance K_s , the conductivity of the particle is expressed as the sum of the bulk conductivity σ_{bulk} through the particle and the surface conductance K_s surrounding the particle [26]:

$$\sigma_{p,i} = \sigma_{bulk} + 2K_s/a_i \quad (4)$$

where the bulk conductivity σ_{bulk} is 10^{-14} S/m and the surface conductance K_s is 2.56 nS. The relative permittivity of the PS bead and medium is 2.6 and 80, respectively, and the conductivity of the medium is 4.5×10^{-4} S/m. The dielectric properties with respect to particle and medium are constant unless otherwise mentioned. In terms of Eqs. (3) and (4), the real value of the CM factor derived from the different radius PS particle is shown in Fig. 1.

The real part of CM factors for all particles are completely negative numbers at higher frequencies. In addition, it has been illustrated that the CM value is strongly dependent on particle size. A particle with a radius of 6 μm only experiences a negative DEP force. As for the microfluidic device, there is a relatively small Reynolds number ($Re = \rho u_0 L / \eta$, where u_0 is the average fluid velocity, L is a length characteristic of the geometry and ρ and η are aqueous density and viscosity, respectively). In our simulation, the parameters ρ and η are 10^3 kg/m³ and 10^{-3} Pa·s, respectively. Furthermore, a typical microparticle with a radius of 10 μm with a moving velocity of 10 mm/s yields a Reynolds number on the order of 0.1. Neglecting inertia terms from Navier–Stokes equations, the hydrodynamic force exerted on the particle can be simplified as expressed in Eq. (5):

$$\mathbf{F}_{drag,i} = -6\pi\eta a_i \mathbf{u}_i \quad (5)$$

Fig. 1 The real value of the CM factor for particles with different radii in a range of frequencies

where $\mathbf{u}_i$ is the velocity vector of particle i . Furthermore, the short-range repulsive force to prevent particle overlap acts over distances ranging from Ångströms to nanometers, as expressed in Eq. (6) [27]:

$$\mathbf{F}_{rep,ij} = -F_0 \exp\left[-\kappa\left(\frac{r_{ij}}{\delta} - 1\right)\right] \frac{\mathbf{r}_{ij}}{|\mathbf{r}_{ij}|} \quad (6)$$

where κ^{-1} , approximately equal to 0.01, characterizes the range of the repulsive force and δ is the minimum distance between two particles ($a_i + a_j$). The prefactors F_0 are chosen as the electrostatic force scale. In our simulation, force F_0 is assumed to be 2.3×10^{-9} N.

However, the gravity effect is neglected in the ALE model presented from Ai and Kang researchers [10, 14] due to their quasi-two-dimensional model. In order to make our model more rigorous, the sedimentary phenomena from gravity effects are introduced into the model:

$$\mathbf{F}_{g,i} = \frac{4}{3} \pi (\rho_p - \rho_m) a_i^3 \mathbf{g} \quad (7)$$

where $\mathbf{g}$ is gravitational acceleration as a constant (9.8 m/s^2), ρ_p and ρ_m are density of the PS bead ($\approx 1300 \text{ kg/m}^3$) and medium ($\approx 1000 \text{ kg/m}^3$), respectively. The motion of PS beads is generally governed by Newton's equation of motion in classical theory. It is well-known that the velocity Verlet method is significantly superior in regard to the stability and accuracy of a simulation. Taking into account these forces, the position and velocity of ambient particles can be evaluated by the velocity Verlet method, as expressed in Eq. (8):

$$\begin{cases} \mathbf{r}_i(t+h) = \mathbf{r}_i(t) + h\mathbf{u}_i(t) + \frac{h^2}{2m_i} \mathbf{f}_i(t) \\ \mathbf{u}_i(t+h) = \mathbf{u}_i(t) + \frac{h}{2m_i} (\mathbf{f}_i(t) + \mathbf{f}_i(t+h)) \end{cases} \quad (8)$$

where $\mathbf{f}_i$ is the net force acting on particle i and $\mathbf{r}_i$ is the position of particle i . Repeating Eq. (8), both positions and velocities of particles can be evaluated at every time interval h in the model.

3 Results and discussion

When the basic forces have been introduced one by one, the particle trajectory can be calculated based on classical Newton method. However, there is a highly complicated morphological structure for many particles. In our 3D model, we mainly focus on the infrastructural chain from two to four particles.

3.1 Dielectrophoretic motion of two particles

Firstly, the distribution of two particles is similar to Ai's model. In the x - y plane, the center of the square fluid coincides with the midpoint of the connecting line of both particles. Two particles are initially separated with a center-to-center distance of r_{12} , presenting an angle of θ

Fig. 2 Schematic of two particles suspended in aqueous solution

with respect to the x -axis. An AC electric field E ($[E_x, 0, 0]$) is applied along the x -axis direction to induce DEP forces, as shown in Fig. 2.

We assume that the strength order of electric field $|E|$ is 6×10^6 V/m and that there is uniform distribution along the x -axis component. The connection of such two particles perpendicular to the applied electric field is first studied for two particles with equal or unequal radius. In terms of Eqs. (3) and (4), the real part of the CM values with regard to two particles a_1 and a_2 with a 2- μm radius is 0.38 at a frequency of 2 kHz, reflecting that their polarized directions are identically parallel to the

Fig. 3 Two particles perpendicular to a uniform electric field: **a** initial position of the two particles with the same radius of 2 μm at an applied frequency of 2 kHz ($\text{Re}[f_{\text{CM}}(a_1)] = \text{Re}[f_{\text{CM}}(a_2)] = 0.38$); **b** the two particles are separated from each other at 0.3 s; **c** initial position of the two particles with different radii (its radius of red and cyan particles is 2 and 4 μm , respectively) at an applied frequency of 100 kHz, $\text{Re}[f_{\text{CM}}(a_1)]$ and $\text{Re}[f_{\text{CM}}(a_2)]$ is 0.25 and -0.03 respectively; **d** the two particles are attracted to form a chain perpendicular to the applied electric field

electric field. For both $\text{Re}[f_{\text{CM}}] > 0$, the two particles with distance r_{12} equal to $5 \mu\text{m}$ repel each other, as shown in Fig. 3 (a)–(b). In contrast, an attractive movement of them, separated by $7 \mu\text{m}$ of distance, occurs if the values $\text{Re}[f_{\text{CM}}]$ of the particles a_1 and a_2 of $2 \mu\text{m}$ and $4\text{-}\mu\text{m}$ radius are 0.25 and -0.03 at a frequency of 100 kHz , as shown in Fig. 3c, d. This demonstrates that the particle–particle interactions are either attractive for parallel alignment or repulsive for perpendicular alignment relying on the sign of themselves for the $\text{Re}[f_{\text{CM}}]$ factor.

Furthermore, particles are randomly suspended in the aqueous solution with any arbitrary orientation (θ is non-zero). As shown in Fig. 4, the trajectory of two particles having either equal or unequal radius are clearly consistent with Ai's results [10]. The particles with the same size ($a_1 = a_2 = 2 \mu\text{m}$) move at an identical velocity toward each other at 2-kHz frequency, which offers same value of $\text{Re}[f_{\text{CM}}] = 0.38$. Due to Newton's third law, the mutual forces are actually antisymmetric with respect to the y -axis. When the applied frequency is 2 MHz , the trajectories of different size particles break down the antisymmetric characteristics when the radii of such particles a_1 and a_2 are $2 \mu\text{m}$ and $6 \mu\text{m}$, respectively. In this case, the real part of the CM factor of them is -0.46 and -0.47 , i.e., they have the same sign. The center of the chain gravity generated by different particles is obviously below the line of $y = 0$. Especially the trajectories of their motion are not smooth; different from Hossan and Ai's results, as shown in Fig. 4c and f. The uneven trajectories are mainly attributed to the repulsive force. Ai and Hossan et al. considered hydrodynamic pressure force to prevent the particle overlap. On the other hand, the repulsive force acting between particles in our model is a short-range force ranging from Ångströms to nanometers. The DEP force exponentially increases as the distance decreases. The chain form appears along with the emergence of much interactive collision. Before the steady state, the repulsive force repeatedly overcomes the DEP force, forming a chain, and adjusts its arrangement parallel to the electric field direction (Movies 1 and 2 in the

Fig. 4 Two particles with an initial orientation of θ equal to 45 degrees: **a** and **d** are the initial positions of two particles with either equal or unequal size at 0 s ; **b** and **e** are the equilibrium states of the particle chain for **a** and **d** in 0.3 s , respectively; **c** is the trajectory of two particles with the same radius at a frequency of 2 kHz ; **f** is the trajectory of two particles with different radii at an applied frequency of 2 MHz

Supplementary Material). In Figs. 5 and 6, the x -axis velocity components of two particles with equal and unequal radius show a damped wave that gradually decreases to zero. In addition, the x -axis and y -axis velocity components of the same particles have identical magnitude. However, particle a_1 moves faster than the larger particle a_2 in the DEP particle–particle interaction since particle a_2 has great inertia, as shown in Fig. 6. It is imperative to point out that the z -axis velocity component is non-zero due to consideration of the gravitational effect in our model. Once the gravitational force is neglected, the z -component velocity becomes zero.

3.2 Dielectrophoretic motion of three particles

In this section, we attempt to extend the understanding of three particles. The mass center of a triangular pattern consisting of three particles is ideally selected at the original point, as shown in Fig. 7. Meanwhile, the angle θ between the vector $\mathbf{r}_1$ for particle a_1 and the x -axis direction is defined.

Fig. 5 Velocity component from two particles with the same radius of $2\ \mu\text{m}$ in Fig. 4(a)–(c): the first and second columns correspond to particles a_1 and a_2 , respectively. In addition, the first, second, and third rows are the x , y and z velocity components of particles a_1 and a_2 , respectively

The three particles with the same radius of $2\text{ }\mu\text{m}$ are studied at the frequency of 2 MHz , so that all of their CM factors are the same ($\text{Re}[f_{\text{CM}(a1)}]$, $\text{Re}[f_{\text{CM}(a2)}]$, and $\text{Re}[f_{\text{CM}(a3)}]$ are -0.46). In comparison with Kang's results, three particles are assumed initially located in a configuration of an equilateral triangle, where the center of mass is at the original point and the rotation angle θ is 10° , 30° , 45° , 60° , 80° , 110° . Similar to the chain of two particles, most cases imply that three particles finally align in a chain with the electric field direction, as shown in Fig. 8. However, the generation of the chain is strongly dependent on their initial location. It is obvious that particles firstly attract each other and form a chain if they are located near another particle. And then a group of two particles continue to attract the third particle e.g., particles a_1 and a_2 for $\theta = 10^\circ$ and 45° (Fig. 8a, c) and particles a_2 and a_3 for $\theta = 80^\circ$ and 110° (Fig. 8e, f). Consistent with Kang's result, the particles mutually revolve either clockwise or counterclockwise depending on how the line joining their centers is inclined relative to the electric field direction (Movie 3 in the

Fig. 6 Velocity component from two particles with different radii of $2\text{ }\mu\text{m}$ and $6\text{ }\mu\text{m}$ in Fig. 4d, e: the first and second columns correspond to particles a_1 and a_2 , respectively. In addition, the first, second, and third rows are the x , y , and z velocity components of particles a_1 and a_2 , respectively

Fig. 7 Schematic of three particles suspended in an aqueous solution

Supplementary Material). However, it is worthwhile mentioning that rotation angle θ equal to 30° is a vertically symmetric structure with respect to the electric field direction. When particles a_1 and a_2 become one chain, particle a_3 is right below them. The same polarized moments keep them from moving away from each other without a revolved motion similar to Fig. 3 (b). In contrast, the three particles as a horizontally symmetric structure ($\theta = 60^\circ$) form a crystal structure different from Kang's chain in the long term. The cluster is balanced at the equilibrium state by the DEP force and repulsive force. Particle a_2 simultaneously attracts both particles a_1 and a_3 . Both a_1 and

Fig. 8 Three particles with the same radius of $2\ \mu\text{m}$ under an applied electric field at a frequency of 2 MHz. Thus, all of their $\text{Re}[f_{\text{CM}}]$ values are -0.46 . The distances (r_{12} , r_{23} , and r_{13}) among them are $12\ \mu\text{m}$ and the rotation angle θ starts with **a** 10° , **b** 30° , **c** 45° , **d** 60° , **e** 80° , **f** 110° . Their initial position is *black* and the triangle line is *green*. The particles a_1 , a_2 and a_3 are *red*, *cyan*, and *magenta*, respectively

a_3 particles repel each other forming the chain perpendicular to the electric field direction. Actually, the cluster is not at a steady state. When a slight perturbation from an external vibration or another particle collision occurs, a cluster composed of any two particles gradually becomes one chain and attracts the other particle.

To the best of our knowledge, little attention has been paid to chain studies from many particles with different radii. Owing to programming flexibility in our model, three particles with different radii have already been simulated, as shown in Fig. 9. Here, the radius of particle a_1 is 4 μm greater than the particles a_2 and a_3 with a radius of 2 μm . Considering a frequency of 2 MHz, the values of each CM factors are negative ($\text{Re}[f_{\text{CM}(a_1)}]$ is -0.47 but both $\text{Re}[f_{\text{CM}(a_2)}]$ and $\text{Re}[f_{\text{CM}(a_3)}]$ are -0.46). The results demonstrated that all particles subsequently form a chain parallel to the electric field, no matter what value the rotation angle θ is. By close examination of Eq. (2), the magnitude of polarization for such a particle a_1 is greater than both the particles a_2 and a_3 . The strong DEP force generated by particle a_1 breaks down either the vertical or horizontal symmetry, for example, Fig. 4e reflects that the orthocenter of a chain composed of two particles with different radii is not located at the original point. Considering rotation angles 30° and 60° , the asymmetric structure from one large and small particles could lead to further attract the other particle (Movie 4 in the Supplementary Material).

Figure 10, on the other hand, shows trajectories taken by three particles with different CM factors when the applied frequency decreases to 0.15 MHz. Meanwhile, both the electric and fluidic conditions are the same. The real parts of the CM factor for particles a_1 , a_2 , and a_3 are -0.138 , 0.14 and 0.14 , respectively. As mentioned above, there are two particles having an opposite sign of the CM factor attracting each other perpendicular to the electric field direction.

Fig. 9 Three particles (the radius of particle a_1 is 4 μm but both a_2 and a_3 particles are 2 μm) at an applied frequency of 2 MHz. The real parts of the CM factor for a_1 , a_2 , and a_3 are -0.47 , -0.46 and -0.46 , respectively. The distances (r_{12} , r_{23} , and r_{13}) among them are 16 μm and the rotation angle θ starts with **a** 10° , **b** 30° , **c** 45° , **d** 60° , **e** 80° , and **f** 110° . Their initial position is in black and the triangle line is green. Particles a_1 , a_2 and a_3 are red, cyan, and magenta, respectively

Fig. 10 Three particles with different CM values ($\text{Re}[f_{\text{CM}(a1)}]$ is -0.138 but $\text{Re}[f_{\text{CM}(a2)}]$ and $\text{Re}[f_{\text{CM}(a3)}]$ are 0.14) at the applied frequency of 0.15 MHz. The rotation angle θ starts with **a** 10° , **b** 30° , **c** 45° , **d** 60° , **e** 80° , and **f** 110° . The other basic conditions, such as distance and particle radius, are similar to Fig. 9

However, an exclusive chain composed of three particles occurs under the condition of rotation angle θ equal to 10° . Since particles a_2 and a_3 have the same sign of $\text{Re}[f_{\text{CM}}]$, they mutually attract each other. On the other hand, the largest particle a_1 repels the others to move it in the horizontal direction and towards its either top or bottom location. In fact, particle a_2 is difficult to move towards the top of particle a_1 due to the attractive force of particle a_3 , as show in Fig. 10b–f. Finally, the two small particles are underneath particle a_1 (Movie 5 in the Supplementary Material).

Fig. 11 Schematic of four particles suspended in aqueous solution

3.3 Dielectrophoretic motion of four particles

We continued to study DEP phenomena of four PS particles with either identical or non-identical size and different signs of the CM factor (real number) when the applied frequency changed. It is essential to understand that the distribution of four particles is much more complicated than three particles. At the x - y plane, they group into various patterns, including trapezoid, rhombus, and rectangle. Therefore, we mainly focused on the square shape, which has the same boundary length (i.e., $r_{12} = r_{23} = r_{34} = r_{41}$). As shown in Fig. 11, the rotation angle θ is still between the position vector r_1 for particle a_1 and the x -axis direction is similar to Fig. 7. The center of mass for this square is assumed to locate at the original point.

As for same-size particles, the real part of the CM factors are -0.46 at the frequency 2 MHz. Considering four different rotation angles, for example 0° , 30° , 45° , and 60° , the calculated results indicate that the particle chain also remains if the particle initial distribution and electric field could not satisfy geometrical symmetric characteristics, as shown in Fig. 12b, d. In accordance with three particles' motion, the existence of a chain needs to satisfy asymmetric structure between particles spatial location and electric field direction. Conversely, the classical pattern from either a cluster or short chain is attributed to their geometric symmetry. Considering the rotation angle to be zero, particle a_1 simultaneously attracts the particles a_2 , a_3 , and a_4 . That each particle is

Fig. 12 Four particles with the same radius of $2\ \mu\text{m}$ under a uniform electric field at a frequency of 2 MHz. Thus, all of their $\text{Re}[f_{\text{CM}}]$ are -0.46 . The distances (r_{12} , r_{23} , and r_{13}) between them are $12\ \mu\text{m}$ and the rotation angle θ starts with **a** 0° , **b** 30° , **c** 45° , and **d** 60° . Their initial position is *black* and the square line is *green*. The particles a_1 , a_2 , a_3 and a_4 are *red*, *cyan*, *magenta*, and *blue*, respectively

interactive tends to make them combine into a cluster similar to Fig. 8d. However, DEP forces only perform in either the vertical or horizontal direction due to distance vector r_{ij} being parallel or perpendicular to the electric field. For example, particle a_1 attracts particle a_2 , as well as repelling particle a_4 . At last, there is only the short length chain suspended in the solution. It should be noted that these symmetric patterns are rare in a real experimental circumstance, since particle distance and azimuth correlation with regard to electric fields have much more irregular geometrical characteristics (Movie 6 in the Supplementary Material).

Mixture particles composed of large and small particles are studied for different rotation angles, as shown in Figs. 13 and 14 (Movies 7 and 8 in the Supplementary Material). Assuming that only one greater size particle a_1 is equal to $4\text{ }\mu\text{m}$ instead of $2\text{ }\mu\text{m}$ in Fig. 13, mutual DEP forces form a cluster at the rotation angle to be zero. On the other hand, a large-size particle is able to capture the near other particles to align themselves parallel or perpendicular to the direction of the electric field. When particle a_3 is introduced into the diagonal location of particle a_1 , it is found that the chain structure becomes more complicated. Two large particles, a_1 and a_3 , capture the small particles a_2 and a_4 . Compared with a single greater particle suspending in solution, two large particles contribute to the generation of the pearl chain. Two greater particles should lie at the diagonal apex, otherwise, they will attract each other and repel two small particles below them, similar to Fig. 12c.

Fig. 13 Four particles (the radius of particle a_1 is $4\text{ }\mu\text{m}$ but the other particles a_2 , a_3 , and a_4 is $2\text{ }\mu\text{m}$) at the applied frequency of 2 MHz . For the real part of the CM value, the largest particle a_1 is -0.47 , whereas the small-size particles a_2 , a_3 , and a_4 are -0.46 . Each of the square distances are $16\text{ }\mu\text{m}$ and the rotation angle θ starts with **a** 0° , **b** 30° , **c** 45° , and **d** 60° . Their initial position is black and the triangle line is green. The particles a_1 , a_2 , a_3 , and a_4 are red, cyan, magenta, and blue, respectively

Fig. 14 Four particles (the radius of particles a_1 and a_3 is $4\ \mu\text{m}$ but particles a_2 and a_4 is $2\ \mu\text{m}$) at the same applied frequency similar to Fig. 13. Particle a_1 (a_3) is -0.47 and particle a_2 (a_4) is -0.46 . The rotation angle θ starts with **a** 0° , **b** 30° , **c** 45° , **d** 60° . The particles a_1 , a_2 , a_3 and a_4 are red, cyan, magenta, and blue, respectively

Furthermore, different values of the CM factor with regard to four particles (both a_1 and a_3 particles are -0.138 but both a_2 and a_4 particles are 0.14) have been investigated when the applied frequency is $0.15\ \text{MHz}$. Figure 15 shows that two large particles with an identical sign of the CM factor are aligned in parallel to the electric field direction (Movie 9 in the Supplementary Material). According to the aforementioned results, two small particles, a_2 and a_4 , move towards the top and bottom of large particles a_1 and a_3 . The distance between the small and large particles is equal. It is important to note that particle a_1 , simultaneously trapping two small particles a_2 and a_4 , is dependent upon the perturbation of external heating. Meanwhile, many particles near the four particles also affect their motions analogous to the correlation phenomena. In addition, they could not move into the gap between particle a_1 and a_3 because they do not have an equivalent CM factor.

4 Conclusion

Overall, this dynamics simulation provides a new insight to describe the motion of particles exerted by a homogeneous AC electric field. Possible advantages compared to other methods are a flexible programming, low-time consumption relying on Pohl's classical formula due to the neglect of high-order moments. For this reason, the calculated results of particle interactive motion are relatively less accurate than the rigorous MST method, especially for smaller

Fig. 15 Four particles with different CM values ($\text{Re}[f_{\text{CM}(a1)}]$ and $\text{Re}[f_{\text{CM}(a3)}]$ are -0.138 , whereas $\text{Re}[f_{\text{CM}(a2)}]$ and $\text{Re}[f_{\text{CM}(a4)}]$ are 0.14). The rotation angle θ starts with **a** 0° , **b** 30° , **c** 45° , **d** 60° . The other simulated conditions, such as distance and particle radius, are similar to Fig. 12

distances between two particles. In addition, the motions of many particles also limit applicability as a much longer chain leads to a significant error in the case of mutual interactions between closely spaced particles. The non-uniformities of the induced field due to the particles themselves are comparable to the particle's size, so high-order terms must be retained.

We numerically studied three-dimensional DEP motions from two to four particles. Since the CM value depends on particle size, either identical or non-identical radius particles experienced by different DEP forces are completely introduced. In order to extend the model application for a pearl chain, the DEP force relying on effective dipole moment was coupled into the model to simulate each particle trajectory at the different applied frequencies. The other forces, including Stokes drag and repulsive forces (hard sphere system), were coupled into the model to depict the two-, three-, and four-particle arrangement. In order to help understand and compare with Kang and Ai's work better, all particles were assumed to be initially at the same height for the z -axis plane.

The trajectory of the two particles is basically consistent with Ai et al. [9], Kang et al. [14], and Xie et al. [16] results by using the rigorous MST method. When both particles are of the same sign, they revolve clockwise or counterclockwise to attract each other and are parallel to the electric field direction, whereas, they are perpendicular to the field. With this in-depth study, the pearl chain from three or four particles is related not only to the individual CM factor but also to initial spatial configuration. Either a cluster or short-chain

occurs if the initial particle distribution and electric field direction do not satisfy spatial symmetrical characteristics. Moreover, the two particles having opposite electric polarization form a chain perpendicular to the electric field. However, it is difficult to evaluate a large particle and a small particle, as shown in Fig. 10b. Since a large particle with a radius of 4 μm firstly repels a small particle with a radius of 2 μm at the same level, the small particle far away from the large particle may be attracted again when it reaches the top of the large particle. Otherwise, it could be attracted by other particles to form another chain. In addition, it should be noted that large size particles suspended in solution form a chain structure easily. The present study potentially leads to further understanding of a variety of AC DEP effects for particle–particle interactive motion.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflicts of interest.

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