Polarizabilities and electric field-induced forces in periodic two-component linear dielectric sphere chains

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Abstract

Idealized single-stranded linear chains of dielectric spheres in a dielectric medium can be used as a model system for electric field-induced particle chaining in complex systems, such as electrorheological fluids. The polarizability of the spheres in the chains and the interparticle (bonding) forces between them determine the macroscopic (e.g. dielectric, rheological) behavior of the bulk. We have derived new analytical equations for the bonding forces in a linear, bidisperse chain, where two components with different sizes form a periodic structure. The force equations were obtained on the basis of local field strength at the particle sites by taking into account the interactions beyond the nearest neighbors within the chain. The dependence of the bonding force and the local field strength on the ratio of particle sizes was investigated. As an application the dielectric permittivity of a bulk model system with bidisperse chains was calculated using the Clausius-Mossotti equation.

Keywords: periodic chains, polarizability, force, electrorheological fluid

1. Introduction

Dielectric particles dispersed in a medium will form chain like structures when exposed to an external electric field, which is the result of interparticle forces arising from the field-induced polarization of the particles. The phenomenon of particle pattern formation driven by electrostatic forces plays a crucial role in a wide spectrum of application areas, e.g. electrorheology [1], microfluidics [2], bottom-up material synthesis [3], etc. Important technological applications are the use of electrorheological fluids (ERFs, suspensions of micrometer sized dielectric particles in a non-conducting liquid) for power transmission [4], vibration damping [5], etc. In these applications the controllable and reversible change in viscosity caused by the particle chaining is exploited.

The viscosity (and shear stress) of an activated ERF is directly connected to the mechanical strength of the chain-like structures. It is determined by the electrostatic forces between the particles in the chains, which hold them together. Therefore, the rheological behavior of an ERF can be derived by direct calculations of the electrostatic forces within the particle structures.

Starting from the treatment of isolated particles as point dipoles in their centers [6] numerous attempts have been made to theoretically describe the ER effect by means of electrostatic interactions. Beyond the point dipole approximation Jones et al. [7] investigated the role of higher order multipoles, and calculated the interparticle force in monodisperse (one-component) particle chains. Anderson [8] used several analytical and numerical methods to include the effect of all orders of multipoles and derived the breaking strength of single chains and columnar structures of aggregated monodisperse chains.

Mostly, monodisperse model systems are used for simplicity, but in this work we examine the interparticle force in bidisperse (two-component) linear

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chains, where spheres with two different diameters are arranged periodically. Motivated by that many ERFs contain two components with different sizes, we focus on such bidisperse systems. We consider the idealized case of linear bidisperse periodic chain made of touching spheres in a dielectric medium under an external electric field. Both the particles and the background medium are considered non-conductive, with different permittivities. Between the particles within the chain only dipole-dipole interaction are taken into account, all other interparticle forces are neglected. The system is presumed to be quiescent, thus Brownian motion, and hydrodynamic effects are also omitted.

We use a simple microscopic method, where the induced dipole moments of the spheres in the bidisperse chain are determined by the external field and the field from all other induced dipoles. First, we derive the induced dipole moments based on the local field strength at the particle sites, and from that an analytical equation for the field-induced force between the particles is obtained. The magnitude of the bonding force is examined at various particle size ratios. To demonstrate the application of our method we calculate the change in permittivity caused by the presence of bidisperse chains in a random dispersion of dielectric spheres using the relationship between the polarizability of the components and the dielectric permittivity of the bulk system.

2. Theory

2.1. Induced dipole moment and polarizability in a linear chain

One-component chain

Consider a linear chain of N_C uniformly sized touching dielectric spheres of diameter σ and dielectric permittivity ϵ_p immersed in a dielectric fluid of permittivity ϵ_f, where $\epsilon_\mathrm{p} > \epsilon_\mathrm{f}$. Assume that there is an external macroscopic electric field $\mathbf{E}_0$, which is parallel with the chain. Due to the interaction of induced dipoles at the site j an internal field $\mathbf{E}_{\mathrm{int},j}$ appears. Therefore, at the site j the local field $\mathbf{E}_{\mathrm{loc},j}$ is the sum of the external and the internal fields:

$$\mathbf{E}_{\mathrm{loc},j} = \mathbf{E}_0 + \mathbf{E}_{\mathrm{int},j} \,, \tag{1}$$

where

$$\mathbf{E}_{\mathrm{int},j} = \sum_{i=1, i\neq j}^{N} \frac{2\mathbf{p}_i}{|i-j|^3\sigma^3} \,. \tag{2}$$

In this equation $\mathbf{p}_i$ is the dipole moment of the ith particle induced by the corresponding local field:

$$\mathbf{p}_i = \alpha \mathbf{E}_{\mathrm{loc},i} \,, \tag{3}$$

where α is the polarizability of the single spheres, given by

$$\alpha = \frac{\epsilon_\mathrm{p} - \epsilon_\mathrm{f}}{\epsilon_\mathrm{p} + 2\epsilon_\mathrm{f}} \left(\frac{\sigma}{2}\right)^3 = \alpha_\epsilon \sigma^3 \,, \tag{4}$$

where $\alpha_\epsilon = (\epsilon_\mathrm{p} - \epsilon_\mathrm{f})/8(\epsilon_\mathrm{p} + 2\epsilon_\mathrm{f})$.

In case of finite length chains Eqs. (1-4) can be solved for all $\mathbf{E}_{\mathrm{loc},j}$ and the corresponding dipole moments can be calculated from Eq. (3). For isotropic spheres the dipole moments lying along the direction of the external field, therefore Eqs. (1-4) reduce to scalar equations. The magnitude of the local field, and with that the induced dipole moments can be different along a chain, for example at the end of the chain the dipole moments are lower than in the case of inner sites [7].

Considering an infinitely long monodisperse chain (Figure 1(a)) the magnitudes of $E_{\mathrm{loc},i} = E_\mathrm{loc}$ are the same in all sites (for all i) due to symmetry. Denoting the particles according to Figure 1(a), on the basis of Eqs. (1-3) for $j = 0$ we can write that

$$\begin{aligned} E_\mathrm{loc} =& E_0 + \sum_{i=-\infty,\, i\neq 0}^{\infty} \frac{2\alpha E_\mathrm{loc}}{|i|^3\sigma^3} \\ =& E_0 + 2\sum_{i=1}^{\infty} \frac{2\alpha E_\mathrm{loc}}{i^3\sigma^3} \,. \end{aligned} \tag{5}$$

After some algebra we obtain

$$E_\mathrm{loc} = \frac{1}{1 - 4\alpha_\epsilon \zeta(3)} E_0 \,, \tag{6}$$

where $\zeta(3) = \sum_{i=1}^{\infty} 1/i^3 = 1.20205...$ is the Riemann zeta function. Eq. (6) contains the contributions by all other dipoles to the local field at each dipole site, therefore the magnitude of the induced dipole moments of the particles in the chain is

$$p = \alpha E_\mathrm{loc} = \alpha \frac{1}{1 - 4\alpha_\epsilon \zeta(3)} E_0 = \alpha f E_0 \,, \tag{7}$$

where the factor f is the ratio of the local and external field strengths:

$$f = \frac{E_\mathrm{loc}}{E_0} = \frac{1}{1 - 4\alpha_\epsilon \zeta(3)} \,. \tag{8}$$

Kim et al. introduced a similar dimensionless "local enhancement factor" in Ref. [9], which agrees

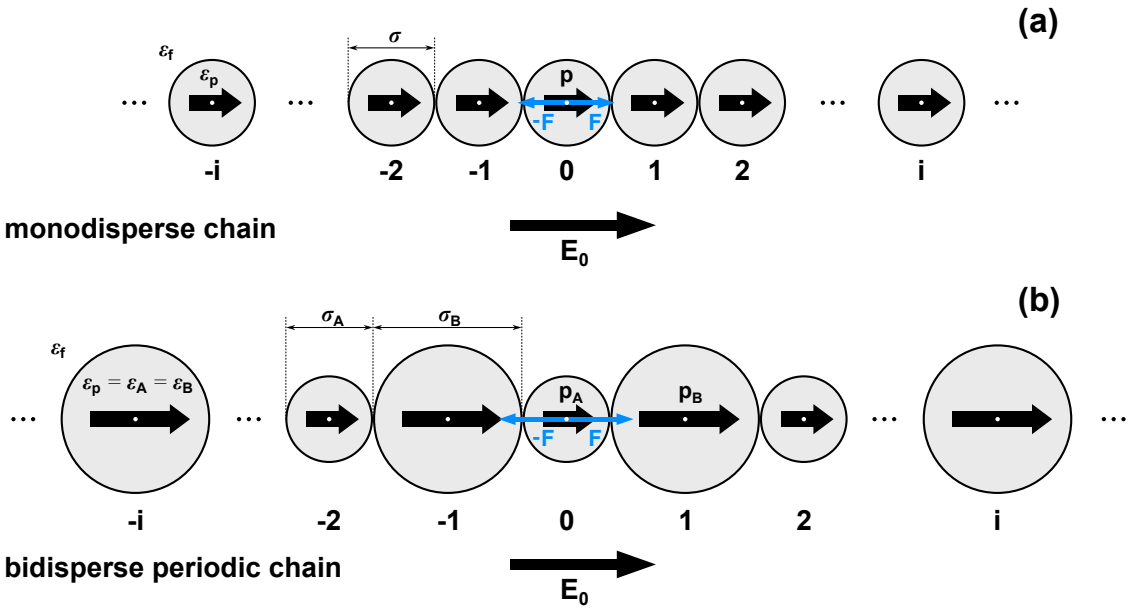

Figure 1: Idealized geometry of infinite monodisperse (a) and bidisperse periodic (b) particle chains in a medium with permittivity ϵ_f under an external parallel electric field $\mathbf{E}_0$. The dielectric spheres carry dipole moments ($\mathbf{p}$ in the monodisperse chain, $\mathbf{p}_A$ and $\mathbf{p}_B$ in the bidisperse chain). The particle chain is held together by the bonding force $\mathbf{F}$. For simplicity the force acting at the center of every spheres is marked only at the 0th sphere (together with $-\mathbf{F}$).

with Eq. (8) for linear particle clusters. For positive polarizability $f > 1$, which means that the induced dipole moment of a sphere in an infinitely long chain is higher than that of a single sphere's.

Two-component periodic chain

In the next, we calculate the induced dipole moments of the particles in a bidisperse periodic chain with the structure depicted in Figure 1(b). Denote the polarizability of component A with α_A, and same for component B with α_B. The diameter of the corresponding spheres are σ_A and σ_B. According to the symmetry of the system we can distinguish only two different local fields $E_{\mathrm{loc,A}}$ and $E_{\mathrm{loc,B}}$. Starting the summation from the 0th A-type particle for the local field $E_{\mathrm{loc,A}}$ we obtain that

$$\begin{aligned} E_{\mathrm{loc,A}} =& E_0 + \sum_{i=-\infty,\, i\neq 0}^{\infty} \frac{2\alpha_i E_{\mathrm{loc},i}}{|i|^3 a^3} \\ =& E_0 + 2\sum_{i=1}^{\infty} \frac{2\alpha_i E_{\mathrm{loc},i}}{i^3 a^3} \\ =& E_0 + 4\alpha_{\mathrm{B}}^* E_{\mathrm{loc,B}} \left(\frac{1}{1^3} + \frac{1}{3^3} + \frac{1}{5^3} + \dots\right) \\ &+ 4\alpha_{\mathrm{A}}^* E_{\mathrm{loc,A}} \left(\frac{1}{2^3} + \frac{1}{4^3} + \frac{1}{6^3} + \dots\right) \\ =& E_0 + \frac{7}{2}\alpha_{\mathrm{B}}^* \zeta(3) E_{\mathrm{loc,B}} + \frac{1}{2}\alpha_{\mathrm{A}}^* \zeta(3) E_{\mathrm{loc,A}} \,, \end{aligned} \tag{9}$$

where the characteristic distance is $a = (\sigma_A + \sigma_B)/2$, and the reduced polarizabilities are $\alpha_A^* = \alpha_A/a^3$ and $\alpha_B^* = \alpha_B/a^3$. A similar equation can be obtained for $E_{\mathrm{loc,B}}$ if we start the summation from a B-type particle. At the end, we obtain two coupled linear equations for the local electric field strengths:

$$\begin{aligned} (2 - \alpha_{\mathrm{A}}^* \zeta(3)) E_{\mathrm{loc,A}} - 7\alpha_{\mathrm{B}}^* \zeta(3) E_{\mathrm{loc,B}} =& 2E_0 \\ -7\alpha_{\mathrm{A}}^* \zeta(3) E_{\mathrm{loc,A}} + (2 - \alpha_{\mathrm{B}}^* \zeta(3)) E_{\mathrm{loc,B}} =& 2E_0 \,. \end{aligned} \tag{10}$$

The solution for the local field strengths are:

$$\begin{aligned} E_{\mathrm{loc,A}} =& \frac{2+6\alpha_{\mathrm{B}}^{*}\zeta(3)}{2-(\alpha_{\mathrm{A}}^{*}+\alpha_{\mathrm{B}}^{*})\zeta(3)-24\alpha_{\mathrm{A}}^{*}\alpha_{\mathrm{B}}^{*}\zeta^{2}(3)}E_0 \\ E_{\mathrm{loc,B}} =& \frac{2+6\alpha_{\mathrm{A}}^{*}\zeta(3)}{2-(\alpha_{\mathrm{A}}^{*}+\alpha_{\mathrm{B}}^{*})\zeta(3)-24\alpha_{\mathrm{A}}^{*}\alpha_{\mathrm{B}}^{*}\zeta^{2}(3)}E_0 \,. \end{aligned} \tag{11}$$

Of course if $\sigma_{\mathrm{A}} = \sigma_{\mathrm{B}} = \sigma$ and $\alpha_{\mathrm{A}} = \alpha_{\mathrm{B}} = \alpha$, then Eq. (11) gives back the corresponding one-component result (see Eq. (6)). The induced dipole moments of the two types of spheres in an infinite periodic chain are:

$$\begin{aligned} p_{\mathrm{A}} =& \alpha_{\mathrm{A}}E_{\mathrm{loc,A}} = \alpha_{\mathrm{A}}f_{\mathrm{A}}E_0 \\ p_{\mathrm{B}} =& \alpha_{\mathrm{B}}E_{\mathrm{loc,B}} = \alpha_{\mathrm{B}}f_{\mathrm{B}}E_0 \,, \end{aligned} \tag{12}$$

while the local enhancement factors take the

$$\begin{aligned} f_{\mathrm{A}} =& \frac{2+6\alpha_{\mathrm{B}}^{*}\zeta(3)}{2-(\alpha_{\mathrm{A}}^{*}+\alpha_{\mathrm{B}}^{*})\zeta(3)-24\alpha_{\mathrm{A}}^{*}\alpha_{\mathrm{B}}^{*}\zeta^{2}(3)} \\ f_{\mathrm{B}} =& \frac{2+6\alpha_{\mathrm{A}}^{*}\zeta(3)}{2-(\alpha_{\mathrm{A}}^{*}+\alpha_{\mathrm{B}}^{*})\zeta(3)-24\alpha_{\mathrm{A}}^{*}\alpha_{\mathrm{B}}^{*}\zeta^{2}(3)} \end{aligned} \tag{13}$$

form.

According to Eqs. (12) and (13) the polarizability of the whole linear bidisperse chain can be calculated as

$$\alpha_{\mathrm{C}} = \frac{1}{2}(f_{\mathrm{A}}\alpha_{\mathrm{A}} + f_{\mathrm{B}}\alpha_{\mathrm{B}}) \,. \tag{14}$$

2.2. Bonding forces in a periodic chain

The induced dipole moments of the spheres are in the head-to-tail configuration, and forces are acting between each sphere due to attractive dipole-dipole interactions, which hold the linear particle chain together. The force acting on the 0th sphere in an infinite long chain (see Figure 1) is the sum of the forces between the 0th and all other particles:

$$\begin{aligned} F = F_{01} + F_{02} + F_{03} + F_{04} + F_{05} + F_{06} + \dots \\ + F_{13} + F_{14} + F_{15} + F_{16} + F_{17} + \dots \\ + F_{25} + F_{26} + F_{27} + F_{28} + \dots \\ + F_{37} + F_{38} + F_{39} + \dots \\ + \dots + \dots + \dots \,. \end{aligned} \tag{15}$$

The indices denote the positions of the two particles between the corresponding force is acting. Instead of the summation of the rows it is better to add the columns as

$$\begin{aligned} F =& F_{01} + (F_{02} + F_{13}) + (F_{03} + F_{14} + F_{25}) \\ &+ (F_{04} + F_{15} + F_{26} + F_{37}) + \dots \,, \end{aligned} \tag{16}$$

to ensure that every component is accounted for only once. Using the force expression between two dipoles for a monodisperse system we obtain that

$$\begin{aligned} F =& -\sum_{i=1}^{\infty} i\frac{6p^2}{(i\sigma)^4} \\ =& -\frac{6p^2}{\sigma^4}\left(1 + \frac{1}{2^3} + \frac{1}{3^3} + \dots\right) \\ =& -\frac{6p^2}{\sigma^4}\zeta(3) \,, \end{aligned} \tag{17}$$

in agreement with Zhang and Widom [10] and Anderson [8].

For a two-component periodic system different products of dipole moments appear in the interaction forces in Eq. (15), therefore we can write that

$$\begin{aligned} F = -6\frac{p_{\mathrm{A}}p_{\mathrm{B}}}{a^4}\left(1 + \frac{1}{3^3} + \frac{1}{5^3} + \frac{1}{7^3} + \dots\right) \\ -6\frac{p_{\mathrm{A}}^2}{a^4}\left(\frac{1}{2^4} + 2\frac{1}{4^4} + 3\frac{1}{6^4} + \dots\right) \\ -6\frac{p_{\mathrm{B}}^2}{a^4}\left(\frac{1}{2^4} + 2\frac{1}{4^4} + 3\frac{1}{6^4} + \dots\right) \,. \end{aligned} \tag{18}$$

The series in the brackets can be expressed with the Riemann zeta function as:

$$\begin{aligned} &\frac{1}{2^4} + 2\frac{1}{4^4} + 3\frac{1}{6^4} + \dots = \\ &= \frac{1}{2^4}\left(1 + \frac{1}{2^3} + \frac{1}{3^3} + \dots\right) = \frac{\zeta(3)}{2^4} \end{aligned} \tag{19}$$

and

$$\begin{aligned} &1 + \frac{1}{3^3} + \frac{1}{5^3} + \frac{1}{7^3} + \dots = \\ &= \zeta(3) - \left(\frac{1}{2^3} + \frac{1}{4^3} + \frac{1}{6^3} + \dots\right) = \\ &= \zeta(3) - \frac{1}{2^3}\left(1 + \frac{1}{2^3} + \frac{1}{3^3} + \dots\right) = \frac{7}{8}\zeta(3) \,, \end{aligned} \tag{20}$$

therefore the force F takes the following form:

$$F = -\frac{6}{(2a)^4}\left(p_{\mathrm{A}}^2 + 14p_{\mathrm{A}}p_{\mathrm{B}} + p_{\mathrm{B}}^2\right)\zeta(3) \,. \tag{21}$$

For the monodisperse case, when $\sigma_{\mathrm{A}} = \sigma_{\mathrm{B}} = \sigma$ and $p_{\mathrm{A}} = p_{\mathrm{B}} = p$ Eq. (21) renders back to Eq. (17).

3. Numerical results and discussion

3.1. Applicability of the model

Our method for the calculation of the bonding forces can be applied only if the chain has a periodic

structure. But beside the -A-B-A- type chains considered here, it is valid for other periodic chains too, such as -A-A-B-B-A-A-, etc. In real bidisperse systems the chains are mostly periodic at low σ_B/σ_A, but if the size ratio is large, then longer chains of smaller particles (A) will be included between the larger particles, forming a -B-$(A)_n$-B- structure, where $n \gg 1$.

For the present calculations it is assumed that the particle structure is linear. Such structures are dominant in real systems, but clusters with more complex topology can be also present. For example, particles with permanent dipole moments can form rings, and in bidisperse systems arcs of smaller particles also appear along the local field lines of the larger particles, as it was shown by Prokopieva et al. [11]. In strongly bidisperse ($\sigma_B/\sigma_A \gg 1$) systems with induced dipole moments it is expected that most of the smaller particles will gather in the gaps of larger particles (where the local field strength is large), and form nearly linear structures, but some of the smaller particles will aggregate around the local field lines of the larger particles.

In the following we show a few results for the application of our analytical equations obtained for the bonding forces and the polarizability of a linear bidisperse chain. For the calculations we used a reference system with the following parameters. The size of the A-type spheres was fixed at $\sigma_A = 10^{-6}$ m, while the size of the B-type spheres was determined by the σ_B/σ_A ratio. The two types of spheres had the same dielectric permittivity $\epsilon_A = \epsilon_B = 4.0$, therefore $(\alpha_\epsilon)_A$ and $(\alpha_\epsilon)_B$ are the same, and it is denoted by α_ϵ. The permittivity of the background medium was $\epsilon_f = 2.7$. The external electric field strength was $E_0 = 10^5$ Vm^{-1}. When two-component chains were present, those were periodic with an -A-B-A- structure.

3.2. Local field strength and bonding force

First, let us examine how the local field strength and the bonding forces change in a bidisperse chain with the σ_B/σ_A size ratio of the components. Figure 2 shows the local enhancement factors f_A and f_B, which give the ratio of the local field strength to the external field strength at the site of A- and B-type particles, respectively. The corresponding $E_{loc,A}$ and $E_{loc,B}$ are calculated on the basis of Eq. (11), while the polarizabilities of single spheres by Eq. (4). The point where the two curves intersect (at $\sigma_B/\sigma_A = 1$) shows the field ratio in an infinitely long one-component chain. It is higher than

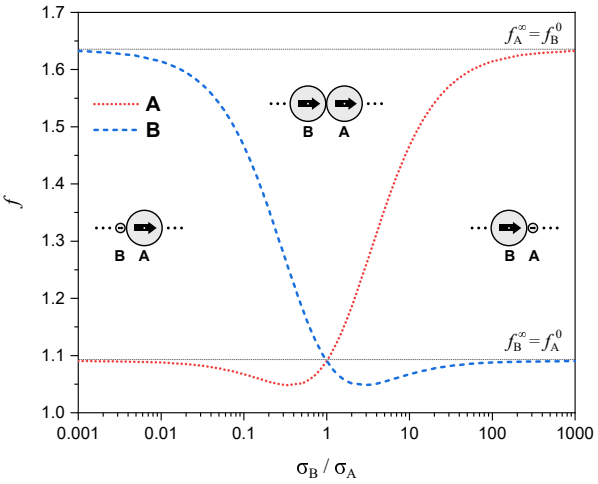

Figure 2: The enhancement factors (ratio of the local and external field strengths) within a bidisperse chain at the sites of A- and B-type particles as a function of the size ratio of the particles. The horizontal lines are the asymptotes of f_A and f_B, when $\sigma_B/\sigma_A \to \infty$.

one, and of course is in agreement with the value obtained from Eq. (8). The results show that in a bidisperse chain the local field strength is larger at the sites of the smaller particles than at the larger particles. If the quality of the particles is different ($\epsilon_A \neq \epsilon_B$), then this difference can be more significant. In the limit of $\sigma_B/\sigma_A \to \infty$ both local enhancement factors approach asymptotic values, which can be calculated as follows. For the reduced polarizabilities of the particles we can write that

$$\begin{aligned} \alpha_A^* &= \alpha_\epsilon \left(\frac{2\sigma_A}{\sigma_A + \sigma_B} \right) \\ \alpha_B^* &= \alpha_\epsilon \left(\frac{2\sigma_B}{\sigma_A + \sigma_B} \right) , \end{aligned} \tag{22}$$

and the limiting values are

$$\begin{aligned} \lim_{\sigma_B/\sigma_A \to \infty} \alpha_A^* &= 0 \\ \lim_{\sigma_B/\sigma_A \to \infty} \alpha_B^* &= 8\alpha_\epsilon . \end{aligned} \tag{23}$$

Based on that, the limits of f_A and f_B are

$$f_A^\infty = \frac{1 + 24\alpha_\epsilon \zeta(3)}{1 - 4\alpha_\epsilon \zeta(3)} \tag{24}$$

and

$$f_B^\infty = \frac{1}{1 - 4\alpha_\epsilon \zeta(3)} . \tag{25}$$

It is obvious that due to symmetry reasons these limiting values are equivalent with the corresponding limits if $\sigma_B/\sigma_A \to 0$: $f_A^\infty = f_B^0$ and $f_B^\infty = f_A^0$.

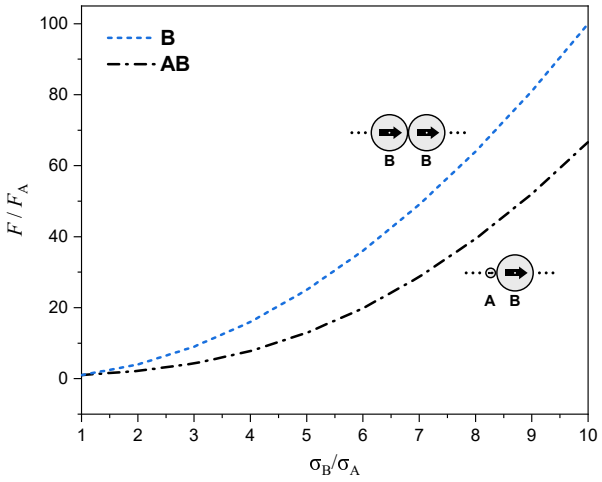

Figure 3: The bonding force between the spheres (normalized by the force in a monodisperse A-type chain) grows quadratically if the diameter of the larger B-type particles is increased. The force in a bidisperse chain is smaller than in the corresponding monodisperse chain.

Eq. (25) is equivalent with the enhancement factor of a one-component chain (Eq. (8)). Therefore, $f_{\rm B}$ at $\sigma_{\rm B}/\sigma_{\rm A} = 1$ has the same value as $f_{\rm B}^{\infty}$ (and $f_{\rm A}^{0}$), since all of these cases correspond to a one-component chain.

The bonding force F between the particles in the bidisperse chain is shown in Figure 3 for different $\sigma_{\rm B}/\sigma_{\rm A}$, which is calculated according to Eq. (21), while the induced dipole moments of the particles are obtained from Eq. (12). For comparison the force in a monodisperse chain with increasing particle size is also presented. The magnitude of F is given relative to the force acting in a one-component A-type chain ($F_{\rm A}$), where $\sigma_{\rm A} = 10^{-6}$ m. The increasing force in the one-component case shows that a chain of larger particles is stronger bonded than a linear chain composed of smaller particles. This is understandable because of the larger induced dipole moment of the larger particles. The force acting in a bidisperse chain is also increasing with $\sigma_{\rm B}/\sigma_{\rm A}$, but remains smaller than in the corresponding monodisperse chain. This can be viewed as the inclusion of smaller particles makes the chains weaker. If $\sigma_{\rm B}/\sigma_{\rm A}$ approaches infinity, then

$$\lim_{\sigma_{\rm B}/\sigma_{\rm A}\to\infty} \frac{F}{(\sigma_{\rm B}/\sigma_{\rm A})^2} = -6(f_{\rm B}^{\infty}\alpha_{\epsilon}\zeta(3)\sigma_{\rm A})^2\,. \quad (26)$$

Therefore, the growth of F with the particle size ratio is quadratic, and asymptotically $F \to \infty$ as

Table 1: The contribution of the non-nearest neighbors to the bonding force F in an infinite particle chain at various particle size ratios.

$\sigma_{\rm B}/\sigma_{\rm A}$	$(F - F_{01})/F$
1	0.168
2	0.373
3	0.636
4	0.796
5	0.880
10	0.982
50	0.999

a function of $\sigma_{\rm B}^2$ (when $\sigma_{\rm A}$ is fixed), which can be seen in Figure 3.

As Eq. (15) shows, the bonding force in an infinite chain is the sum of the force between the nearest neighbors (F_{01}) and the long range contribution of all other particles in the chain ($F - F_{01}$). The magnitude of the long range component depends on the ratio of the particle sizes as it is shown in Table 1. In a monodisperse chain the bonding force comes largely from the interaction between the adjacent particles; the long range contribution is only 16.8%. By increasing the size of the B-type particles the contribution of the non-nearest neighbors is increasing, and above $\sigma_{\rm B}/\sigma_{\rm A} \sim 10$ it becomes orders of magnitudes larger than F_{01}.

We note, that the present method for the field-induced force calculations can be applied to the analogous magnetic case too (chains of magnetizable particles, i.e. magnetorheological fluids) by translating the equations to the magnetic language. Moreover, the model is also applicable when the particles have permanent dipole moments, like in a colloidal dispersion of magnetic nanoparticles, i.e. ferrofluids. For monodisperse systems we can compare the bonding force in a chain of field-induced dipoles with the case when the particles have permanent dipole moments. Prokopieva and co-workers [12] obtained an analytical equation for the total energy of a ferroparticle chain of N beads with m permanent dipole moments as

$$U_{\rm N} = -2\frac{m^2}{d^3}N\zeta(3)\,, \quad (27)$$

where d is the diameter of the magnetic core. In case of $N \to \infty$ the one-particle energy is

$$U = \lim_{N\to\infty}\frac{U_{\rm N}}{N} = -2\frac{m^2}{d^3}\zeta(3)\,. \quad (28)$$

From this equation the force acting on a permanent dipole in an infinite chain can be obtained as the negative distance (diameter) derivative of U:

$$F = -\frac{\partial U}{\partial d} = -6\frac{m^2}{d^4}\zeta(3)\,, \qquad (29)$$

which is in agreement with our Eq. (17). We conclude, that the single particle force in a permanent dipole chain is equal with the corresponding force in a polarized chain if the dipole moments are the same.

3.3. Effective permittivity

If we know the polarizabilities of the constituents in a multi-component system, where single particles and linear particle chains are also present we can calculate the effective dielectric permittivity of the bulk. Let us consider a bidisperse case when the system is the mixture of three components in a volume V: single A-type particles in a number of N_A (component A), single B-type particles in a number of N_B (component B) and single-stranded periodic bidisperse chains as component C (the total number of particles in the chains is N_C). According the Clausius-Mossotti equation the effective permittivity (ϵ) of the bulk system is

$$\frac{\epsilon-\epsilon_\mathrm{f}}{\epsilon+2\epsilon_\mathrm{f}} = \frac{4\pi}{3}(\rho_\mathrm{A}\alpha_\mathrm{A}+\rho_\mathrm{B}\alpha_\mathrm{B}+\rho_\mathrm{C}\alpha_\mathrm{C})\,, \qquad (30)$$

where $\rho_\mathrm{n} = N_\mathrm{n}/V$ is the number density and α_n is the polarizability of component n. The polarizabilities of the single particles and the chains can be calculated with Eqs. (4) and (14), respectively. We assume that in equilibrium the particles are either single or part of a periodic bidisperse chain, thus the total number of particles is $N_\mathrm{A}+N_\mathrm{B}+N_\mathrm{C}=N$. If the concentrations of the components are given by $x_\mathrm{n} = N_\mathrm{n}/N$ number fractions, then because the system is composed of equal number of A and B particles (which is true for the bidisperse periodic chains too) the concentration of the single particles is $x_\mathrm{A} = x_\mathrm{B} = (1-x_\mathrm{C})/2$. By substituting the reduced quantities $\rho^* = \rho a^3$ and α^*_n Eq. (30) takes the form

$$\frac{\epsilon-\epsilon_\mathrm{f}}{\epsilon+2\epsilon_\mathrm{f}} = \frac{4\pi}{3}\rho^*\left(\frac{1-x_\mathrm{C}}{2}(\alpha^*_\mathrm{A}+\alpha^*_\mathrm{B})+x_\mathrm{C}\alpha^*_\mathrm{C}\right)\,. \qquad (31)$$

In a monodisperse system Eq. (30) is reduced for two components, and the concentration of the particles is $1-x_\mathrm{C}$. A similar equation was used previously to describe the time evolution of the permittivity of ERFs during chain formation [13].

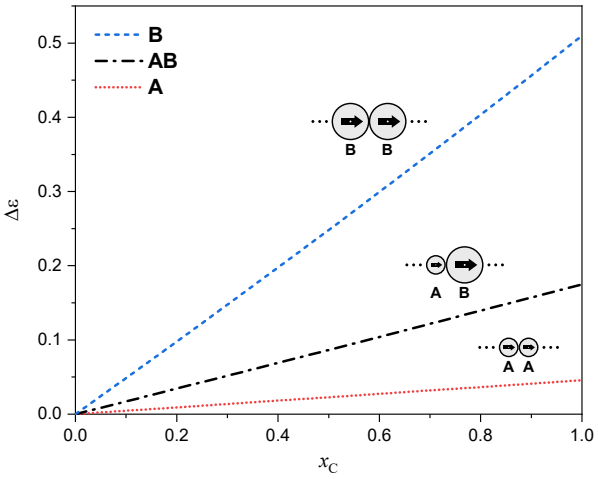

Figure 4: The change of dielectric permittivity of a bulk system as a function of the concentration of chains relative to the permittivity of the isotropic system ($\epsilon_\mathrm{A}=\epsilon_\mathrm{B}$, $\sigma_\mathrm{B}/\sigma_\mathrm{A}=2$). Chain formation increases the permittivity in cases of both types of monodisperse and bidisperse systems too, but with different magnitude.

The effective permittivity is calculated relative to the permittivity of the isotropic system ($\Delta\epsilon = \epsilon-\epsilon_\mathrm{iso}$), where only randomly dispersed individual particles are present ($x_\mathrm{C}=0$). By increasing x_C we approach the structure where all particles are part of a chain ($x_\mathrm{C}=1$). The value of $\Delta\epsilon$ is calculated for the cases of the two monodisperse structures (only A- or B-type particles are present), and for a bidisperse case when the concentrations of components A and B are equal. The B-type particles are larger than the A-type, and the ratio of the diameters is $\sigma_\mathrm{B}/\sigma_\mathrm{A}=2$ in all cases. The total concentration of the particles is 5% by volume, which corresponds to a dilute dispersion.

Figure 4 shows that the chain formation increases the permittivity of the bulk, which is in agreement with other experimental [14, 15] and theoretical [16] results. Since $f>1$ the polarizability (thus the induced dipole moment) of a particle in a chain is larger compared to the polarizability of a single particle. Therefore, as the concentration of entities with larger dipole moments (chains) is increased the effective permittivity increases linearly. The magnitude of the induced dipole moment is proportional to the cube of the particles' radius (see Eqs. (3) and (4)), so $\Delta\epsilon$ is significantly larger in case of the monodisperse system containing larger B-type particles than in case of monodisperse A-type particles. The permittivity increment of the bidisperse

system is between the $\Delta\epsilon$ of the B-type and A-type monodisperse case.

4. Conclusions

In the present work bidisperse dielectric spheres aligned by an external electric field into linear periodic chain have been studied. The main results are the following.

- We have derived new analytical equations for the induced dipole moments and polarizabilities of dielectric spheres in a periodic chain. According these equations the polarizability of a sphere in a one-component chain is also given, which agrees with previous results of Kim et al. [9].
- Using the expressions for the local field strength at the sites of the spheres analytical equations were obtained for the electric field-induced bonding forces in two-component chains. The equations render back to the force expressions in Refs.[10] and [8] for monodisperse chains.
- We have presented calculations for the dependence of the field-induced force in a two-component chain on the size ratio of the spheres. The results showed that the bidisperse chain becomes stronger as the size of the larger component is increasing, but it is weaker than the corresponding monodisperse chain.
- As an application of the microscopic model the dielectric permittivity of a bulk bidisperse system have been calculated, which contains smaller and larger single particles and bidisperse periodic chains. The effective permittivity of the bidisperse system is linearly proportional with the concentration of the chains, and lies between the permittivity of the two corresponding monodisperse systems.

The method used here is applicable not just for linear chains, but for bidisperse periodic clusters with higher dimensions too, such as periodic monolayers and cubic lattices. The latter could be used as a model for the columnar structures in an ERF under higher electric field strength, where aggregates of multiple linear chains are present. Furthermore, as it was pointed out, the calculation method can be applied for bidisperse periodic chains made of magnetizable particles under a magnetic field (magnetorheological fluids) by translating the equations to the analogous magnetic language.

We believe that the analytic expressions derived here could be useful to predict the dielectric and rheological properties of bidisperse ERFs.

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Data availability statement

All data generated and analyzed during this study are available from the corresponding author on reasonable request.

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