

## MULTIPOLAR INTERACTIONS OF DIELECTRIC SPHERES

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### Summary

The interactions of uncharged dielectric particles in an electric field can be described mathematically using multipolar expansions. Interparticle force calculations require that account be taken of the interactions of all these moments. Practical models for particle chains have been developed using a "dipole" approximation, where all moments except the dipole are neglected. To determine the accuracy of this dipole approximation, the effect of higher-order terms (quadrupole, octupole, etc.) upon the effective moment of particle chains is determined as a function of the number of particles in a chain, the relative dielectric constant  $\epsilon_p/\epsilon_m$ , and the spacing between particles  $d$ . It is found that the accuracy of the dipole model is severely compromised for closely spaced particles when  $\epsilon_p/\epsilon_m > 4.0$ , due to slow convergence of conventional multipolar expansions. Similar limits on the accuracy of the dipole model are found for short and long chains. For chains of touching spheres with  $\epsilon_p/\epsilon_m \gg 1.0$ , impracticably large numbers of terms are required to achieve convergence.

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### 1. Introduction

The subject of this paper is the strong short-range electrostatic interactions induced between small uncharged dielectric particles in an electric field. These interactions may be represented by multipoles, including the familiar dipole and quadrupole plus other higher-order terms. Interparticle forces may be calculated by accounting for multipolar interactions which become very strong when particles approach each other in the presence of an electric field. Such calculations are essential to models for the electromechanics of systems of closely spaced particles in a variety of important physical situations.

In the present work, attention is restricted to chains of particles aligned parallel to the applied field. Particle chaining, reported as early as 1927 by Muth [1], is a ubiquitous and very important phenomenon. It has been observed in biological cells suspended in aqueous media [2], and is crucial to the success of cellular electrofusion as practiced by Zimmermann et al. [3]. It is

observed in liquid high-voltage insulants, where contaminant particles form chains, increasing the likelihood of electrical breakdown under some circumstances. Electric-field-induced chain formation plays a crucial role in electroviscous media, which are suspensions of semi-insulating particles in dielectric liquids [4]. Finally, chaining is evident in a variety of electric field separation and filtration system [5].

Very similar behavior is often observed when a magnetic field is applied to magnetizable particles. A good example of magnetic particle chaining is found in the magnetostabilized bed when strong fields are applied [6]. Also, interparticle forces and chaining are crucial to magnetic brush xerography [7]. While the analysis in this paper is conducted for dielectric particles in electric fields, virtually all equations and calculated results can be converted to the case of magnetizable particles in magnetic fields by a set of simple substitutions of magnetic for electric quantities.

### *Motivation*

The examples given above illustrate the importance of electric-field-induced force interactions between uncharged dielectric particles (and the analogous case of ferromagnetic particles). Though various attempts have been made in the past to calculate these forces, a general yet practical method has not been achieved. The immediate goal of the present work is to develop reliable methods for calculating the induced multipoles that exist when chains of particles are aligned parallel to the applied electric (or magnetic) field. The limits of accuracy for the commonly used dipole approximation are explored and the convergence of higher-order moment expansions are studied in order to identify the conditions where slow convergence is encountered. Simplified approximate calculation of multipolar moments will facilitate accurate predictions of interaction forces between adjacent particles.

## **2. Background**

The first practical investigation of the mechanics of particle chaining was reported by Harpavat who had an interest in the behavior of 100- $\mu\text{m}$  developer particles in a magnetic brush xerographic copying machine [7]. He limited his investigation to highly permeable spheres of uniform size, formed into chains of from two to seven touching particles, and under the influence of a non-uniform magnetic field. Only dipole interactions were considered. His analysis and accompanying experiments revealed that the weakest joint in a chain of particles is not necessarily found in the region of the weakest magnetic field. As will be shown in Section 3 of this paper, Harpavat's analysis seriously underestimates the absolute magnitude of the moments for contacting particles. Nevertheless, his observations of the behavior of magnetic particle chains

are qualitatively accurate and illuminate some important magnetic brush phenomenology.

Yarmchuk and Janak treated the problem of magnetic particle chains more generally by allowing for arbitrary size and spacing for each of up to ten spheres [8]. They did not restrict their analysis to the high-permeability limit or to uniformly sized particles and, more importantly, admitted higher-order linear multipoles in the particle-field interactions. Their analysis, accompanied by some unique measurements of magnetic moments of single particles, showed that, for closely spaced particles of high permeability, higher-order multipoles are essential to accurate calculation of interparticle forces. Alward and Imaino generalized the analysis to account for the interactions among low-permeability toner particles formed into long chains and, for their calculation of interparticle forces and chain length, assumed a non-uniform magnetic field solution representative of developer rolls in single-component xerographic development engines [9]. They neglected all but the dipole terms, an assumption valid for their low-permeability particles, but not generally applicable.

Paranjpe and Elrod described an electromechanical model for the magnetic brush which uses the dipole approximation to account for static forces and torques exerted on individual chains by the magnetic field [10]. An energy minimization method was used successfully to calculate the average chain length. Some experimental results, obtained using a modified two-component xerographic engine as a test fixture, were reported as well.

More recently, works have appeared which consider the analogous case of interacting dielectric spheres. Miller and Jones treated infinite chains of uniformly sized dielectric spheres to calculate the effective dielectric constant of heterogeneous media consisting of filaments or chains in a homogeneous matrix [11]. Similar to Yarmchuk and Janak [8], they employed a multipolar expansion to model particle-particle interactions and to verify the importance of higher-order terms for closely spaced particles with high dielectric constant. Sancho et al. [12] have reported a computation of the interactions between lossy dielectric spheres using an expansion of linear multipoles. They evaluated the first ten moments, using them to compute estimates for the interparticle forces between closely spaced spheres in a uniform electric field.

### **3. Critique of dipole approximation**

The purpose of this section is to critique the dipole approximation used to model the interactions of dielectric spheres subject to a uniform electric field. The results should be relevant when the field is slightly non-uniform and may be transformed to the analogous case of magnetic particles by an appropriate set of substitutions of magnetic for electric quantities. In most respects, our criticism of the dipole approximation, wherein all but dipole terms are neglected in calculation of the mutual field interactions, is that of Yarmchuk and

Janak [8]. We have computed the dipole moment per particle in short and long chains as a function of the number of multipolar terms retained in the calculation to examine the convergence of this lead term for varied dielectric constant and particle spacing. In general, we find that for touching spheres with  $\epsilon_p/\epsilon_m > 4$ , the dipole approximation seriously compromises accuracy in moment calculations. The results are presented in such a way as to show how the addition of higher-order multipolar terms improves the accuracy of the dipole moment itself. The implications for accurate interparticle force calculation are evident.

### Finite chains

To formulate the dipole model, consider a linear array of  $N$  uniformly sized touching dielectric spheres of radius  $R$  and dielectric permittivity  $\epsilon_p$  immersed in a dielectric fluid of permittivity  $\epsilon_m$  and aligned parallel to a uniform electric field of magnitude  $E_0$  as shown in Fig. 1. The analysis is similar to that of Harpavat [7], except for our recognition of finite particle permittivity. According to the dipole model, for a two-particle chain ( $N=2$ ) the net external electric field experienced by particle 1 consists of  $E_0$  plus the dipole field due to particle 2. Then

$$E_1 \cong E_0 + p_2/2\pi\epsilon_m z^3|_{z=2R} \quad (1)$$

where  $p_2$  is the dipole moment of particle 2 and  $z$  is the distance between the particle centers. Due to symmetry,  $p_1=p_2$  and  $E_1=E_2$ . For convenience, the relationship between the induced linear multipoles and the axisymmetric electric field components  $E_n$  is introduced here [13]:

$$p_n^{(i)} = \frac{4\pi\epsilon_m K^{(i)} R^{2i+1}}{(i-1)!} \frac{\partial^{i-1} E_n}{\partial z^{i-1}} \quad (2)$$

where  $p_n^{(i)}$  is the  $i$ th linear multipolar moment of particle  $n$  (so that  $i=1$  for the dipole,  $i=2$  for the quadrupole, etc.) and  $K^{(i)} = (\epsilon_p - \epsilon_m) / [i\epsilon_p + (i+1)\epsilon_m]$ .

For convenience when considering the dipole approximation, the superscript  $(i)$  is dropped so that  $K^{(1)}=K$ , etc. Then

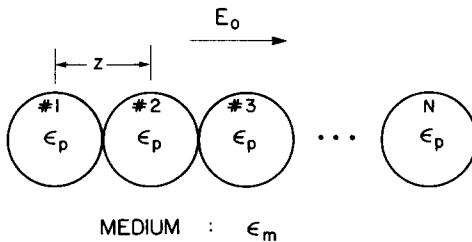


Fig. 1. Linear chain of  $N$  identical touching dielectric spheres of radius  $R$  and dielectric permittivity  $\epsilon_p$  aligned parallel to a uniform electric field  $E_0$  in medium of permittivity  $\epsilon_m$ .

$$p_1 = p_2 = \frac{p_0}{1 - K/4} \quad (3)$$

where  $p_0 = 4\pi\epsilon_m KR^3 E_0$  is the dipole moment of an isolated sphere in the uniform electric field  $E_0$ . Also for convenience, a net effective moment of the chain is defined

$$(p_{\text{eff}})_N = \sum_{n=1}^N p_n \quad (4)$$

and so, for a two-sphere chain, the effective moment is

$$(p_{\text{eff}})_{N=2} = \frac{2p_0}{1 - K/4} \quad (5)$$

For chains of more than two spheres ( $N > 2$ ), the problem becomes more complex because the dipole moments are not all equal. Consider the dipole approximation for three spheres numbered 1, 2 and 3.

$$E_1 \cong E_0 + p_2/2\pi\epsilon_m z^3|_{z=2R} + p_3/2\pi\epsilon_m z^3|_{z=4R} \quad (6)$$

$$E_2 \cong E_0 + p_1/2\pi\epsilon_m z^3|_{z=2R} + p_3/2\pi\epsilon_m z^3|_{z=2R} \quad (7)$$

From symmetry,  $E_1 = E_3$  and  $p_1 = p_3$ . Then, using eqn. (2)

$$p_1 = \left( \frac{1 + K/4}{1 - K/32 - K^2/8} \right) p_0 \quad (8)$$

$$p_2 = \left( \frac{1 + 15K/32}{1 - K/32 - K^2/8} \right) p_0 \quad (9)$$

and finally from eqn. (4)

$$(p_{\text{eff}})_{N=3} = \left( \frac{3 + 31K/32}{1 - K/32 - K^2/8} \right) p_0 \quad (10)$$

Similar methods may be employed for chains of any length.

The effective moments of chains (normalized to  $p_0$ ) for  $N=1$  through 6 are plotted versus  $K$  in Fig. 2, while Table 1 tabulates  $(p_{\text{eff}})_N$  values at the  $K=1.0$  ( $\epsilon_p/\epsilon_m \rightarrow \infty$ ) limit. The two columns at the far right side of Table 1 are from an independent computation of the effective dipole moment of conducting spheres which is based on the method of images [14]. The large differences between the two results for effective moment at  $K=1.0$  indicate that the dipole approximation is quite inaccurate for touching spheres when  $\epsilon_p/\epsilon_m \gg 1$ . This observation is supported by close examination of Stratton's analysis of the multipoles induced in a dielectric sphere by a point charge when the point charge is close to the sphere [15].

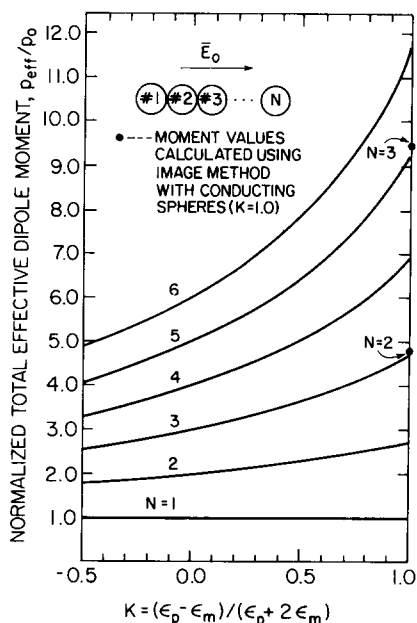


Fig. 2. Net effective dipole moment  $p_{\text{eff}}(N)$  normalized to  $p_0$  for chains of  $N$  spheres calculated using the dipole approximation.

TABLE 1

Moment calculations for finite chains of dielectric particles for  $K = 1.0$  normalized to  $p_0$

$N$	Results from Harpavat <sup>a</sup> (dipole approximation)								Image theory <sup>b</sup>	
	$p_1$	$p_2$	$p_3$	$p_4$	$p_5$	$p_6$	$(p_{\text{eff}})_N$	$(p_{\text{eff}})_N/N$	$(p_{\text{eff}})_N$	$(p_{\text{eff}})_N/N$
1	1.0						1.0	1.0	1.0	1.0
2	1.33	1.33					2.67	1.33	4.81	2.40
3	1.48	1.70	1.48				4.67	1.56	12.13	4.04
4	1.55	1.92	1.92	1.55			6.94	1.73	23.81	5.95
5	1.59	2.00	2.10	2.00	1.59		9.28	1.86	~40 <sup>c</sup>	~8.1 <sup>c</sup>
6	1.61	2.04	2.19	2.19	2.04	1.61	11.68	1.95	~63 <sup>c</sup>	~10 <sup>c</sup>

<sup>a</sup>Calculated using dipole interactions only with  $K = 1.0$ .

<sup>b</sup>Obtained from Ref. [14] for uniform conducting spheres.

<sup>c</sup>Provided by R. Meyer of Xerox Corporation, Webster, NY.

### Higher-order expansions

Higher-order multipoles may be introduced to improve the accuracy of the effective dipole calculation. As an example, consider again a chain of two spheres, but now include dipole and quadrupole terms, that is,  $p_n^{(1)}$  and  $p_n^{(2)}$ ,

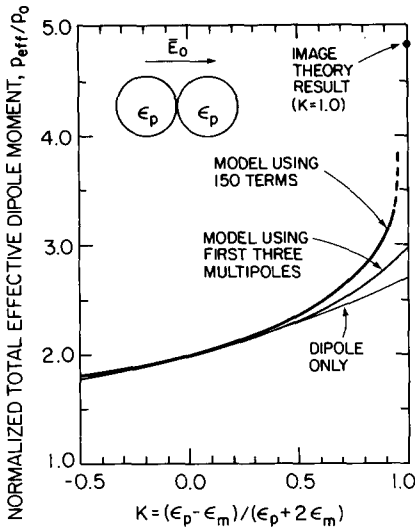


Fig. 3. Net effective dipole moment  $p_{\text{eff}}$  normalized to  $p_0$  for a chain of two touching dielectric particles calculated using 1, 3, and 150 multipolar terms.

respectively. Note from eqn. (2) that the quadrupole moment is related to the first axial derivative of the electric field.

$$E_1 = E_0 + p_2^{(1)} / 2\pi\epsilon_m z^3 \big|_{z=2R} - 3p_2^{(2)} / 4\pi\epsilon_m z^4 \big|_{z=2R} \quad (11)$$

$$\frac{\partial E_1}{\partial z} = -3p_2^{(1)} / 2\pi\epsilon_m z^4 \big|_{z=2R} + 3p_2^{(2)} / \pi\epsilon_m z^5 \big|_{z=2R} \quad (12)$$

Because of symmetry  $p_1^{(1)} = p_2^{(1)}$  and  $p_1^{(2)} = -p_2^{(2)}$ . The resulting set of equations may then be solved to obtain an improved estimate for the effective dipole moment of a chain of two spheres. Figure 3 contains plots of  $(p_{\text{eff}})_{N=2}$ , normalized to  $p_0$ , for  $-0.5 < K < 1.0$  based on the dipole approximation, a dipole plus quadrupole plus octupole approximation, and a calculation which employs the first 150 multipoles (from  $p^{(1)}$  to  $p^{(150)}$ ). The value of the moment at  $K=1.0$  obtained using the image method [14] is also plotted. It is evident that the dipole approximation fails for  $K > 0.5$  and that even 150 terms fall far short of an accurate result near  $K=1.0$ .

### Infinite chains

For an infinite chain of spheres, we must add the contributions of all the particles on either side of an arbitrary central sphere. Because all the particles are identical, the electric field experienced by an arbitrarily selected central sphere may be written

$$E_z = E_0 + E_{\text{dipoles}} + E_{\text{octupoles}} + \dots \quad (13)$$

Due to symmetry, only the odd multipolar terms contribute to the total field  $E_z$ . These terms are [13]

$$E_{\text{dipoles}} = 2 \left( \frac{p^{(1)}}{2\pi\epsilon_m(2R)^3} + \frac{p^{(1)}}{2\pi\epsilon_m(4R)^3} + \dots \right) \quad (14)$$

which simplifies to become

$$E_{\text{dipoles}} = \frac{p^{(1)}}{\pi\epsilon_m(2R)^3} \zeta(3) = \frac{1.20206p^{(1)}}{\pi\epsilon_m(2R)^3} \quad (15)$$

and

$$E_{\text{octupole}} = 2 \left( \frac{p^{(3)}}{\pi\epsilon_m(2R)^5} + \frac{p^{(3)}}{\pi\epsilon_m(4R)^5} + \dots \right) \quad (16)$$

which becomes

$$E_{\text{octupole}} = \frac{2p^{(3)}}{\pi\epsilon_m(2R)^5} \zeta(5) = \frac{2.07386p^{(3)}}{\pi\epsilon_m(2R)^5} \quad (17)$$

Note that  $\zeta(n)$  with integer  $n$  is the Riemann-zeta function [16]. The octupolar moment  $p^{(3)}$  is related to the second derivative of the electric field (cf. eqn. (2)). If all but the dipole and octupole contributions are ignored,

$$\frac{\partial^2 E_z}{\partial z^2} \cong \frac{12p^{(1)}}{\pi\epsilon_m(2R)^5} \zeta(5) + \frac{60p^{(3)}}{\pi\epsilon_m(2R)^7} \zeta(7) \quad (18)$$

$$\cong \frac{0.388848p^{(1)}}{\pi\epsilon_m R^5} + \frac{0.472664p^{(3)}}{\pi\epsilon_m R^7} \quad (19)$$

Equations (2), (13), (15), (17), and (19) may be combined to solve for the dipole moment  $p^{(1)}$  and, if desired,  $p^{(3)}$ . Calculated results versus  $K$  are shown in Fig. 4, and for purposes of comparison the result of the dipole approximation is also provided. Furthermore, Fig. 4 contains a plot of the dipole moment per particle calculated using the first 28 nonzero terms (viz.,  $p^{(1)}$ ,  $p^{(3)}$ , ...,  $p^{(55)}$ ) which is accurate to 3 places for  $-0.5 < K < 0.95$ . Failure of the dipole approximation is evident for  $K > 0.5$ .

Figure 5 contains a plot of the dipole moment per particle  $(p_{\text{eff}})_N/N$  versus  $N$  using the dipole approximation for various values of  $K^{(1)}$ . Values calculated using the image theory method for conducting particles (which corresponds to  $K^{(1)} = 1.0$ ) are also plotted [14]. It is clear that the dipole approximation seriously underestimates the moment per particle for chains of any length when  $K \cong 1.0$ . Harpavat's result [7] that  $p^{(1)} = 2.5p_0$  for each sphere in an infinite chain when  $K = 1.0$  is incorrect. In fact, the moment per particle goes to infinity for  $K = 1.0$  as  $N \rightarrow \infty$ .



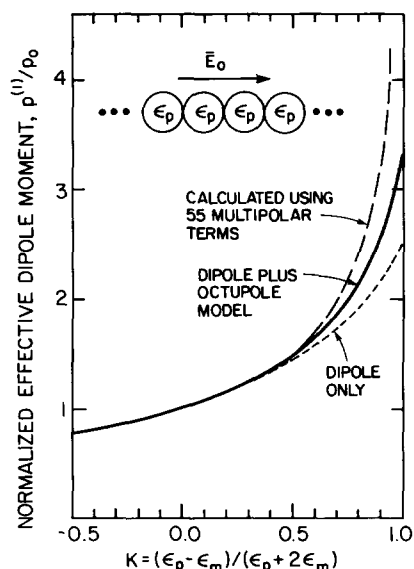


Fig. 4. Net effective dipole moment  $p^{(1)}$  per particle normalized to  $p_0$  for an infinite chain of dielectric spheres calculated using 1, 3, and 55 multipolar terms.

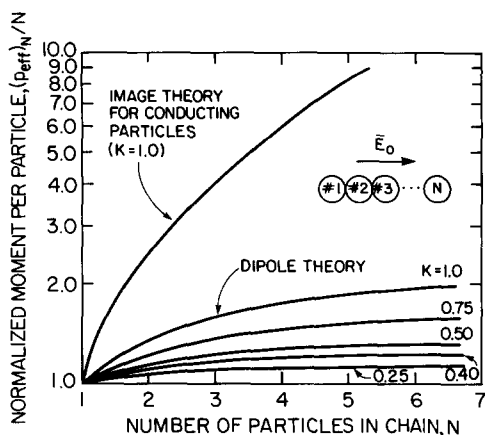


Fig. 5. Average effective dipole moment per particle normalized to  $p_0$  using the dipole approximation for linear chains of up to  $N=6$  dielectric spheres at  $K=0.25, 0.40, 0.50, 0.75$ , and  $1.00$ .

### Closure

This critique shows that the dipole approximation fails to provide accurate results for the field interactions of linear arrays of dielectric spheres subjected to a uniform electric field when  $K > 0.5$ . If the calculation of the dipole moment is incorrect, then suspicion is cast on any interparticle force calculations based on the dipole approximation. The error is significant for long chains of touch-

ing particles when  $\epsilon_p/\epsilon_m > 4.0$ . In the next section of this paper, we explore the influence of particle separation on the accuracy of truncated multipolar expansions.

#### 4. Effect of particle spacing on dipole moment

Figures 3 and 4 clearly illustrate that, for chains of touching spheres, the dipole model provides a reasonably accurate estimate for  $p^{(1)}$  in the range  $-0.5 < K < 0.5$ . For any given value of  $K$ , the accuracy improves as more multipolar terms are added, though a large number of terms may be required as  $K \rightarrow 1.0$ . It is shown here that the restriction on  $K$  is relaxed dramatically for

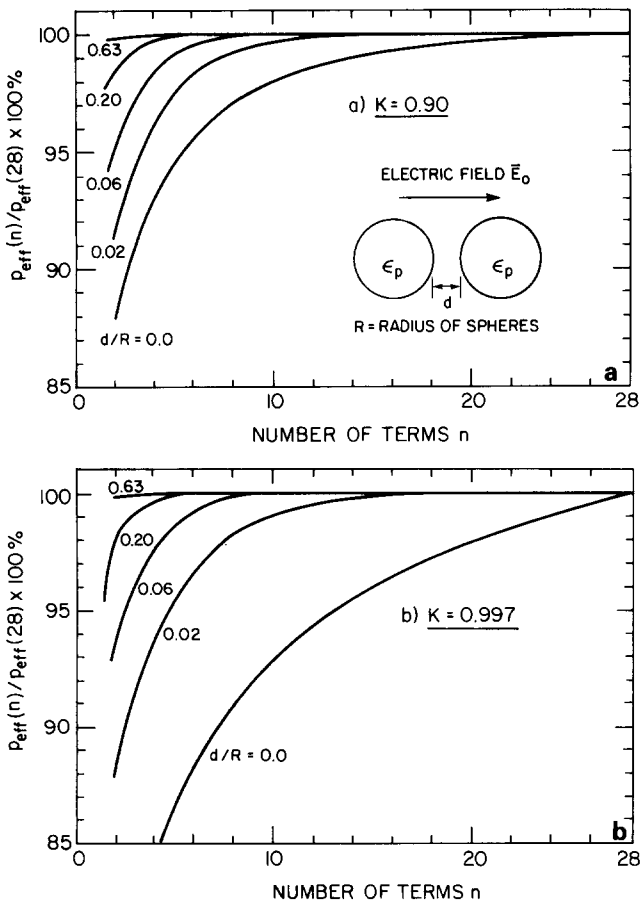


Fig. 6. Convergence plot of net effective dipole moment  $p_{\text{eff}}(n)$  normalized to  $p_{\text{eff}}(28)$  for a chain of two uniform dielectric spheres as a function of the number of multipolar terms  $n$  used in the calculation. (a)  $K=0.90$  ( $\kappa=28.0$ ), (b)  $K=0.997$  ( $\kappa=1000$ ).

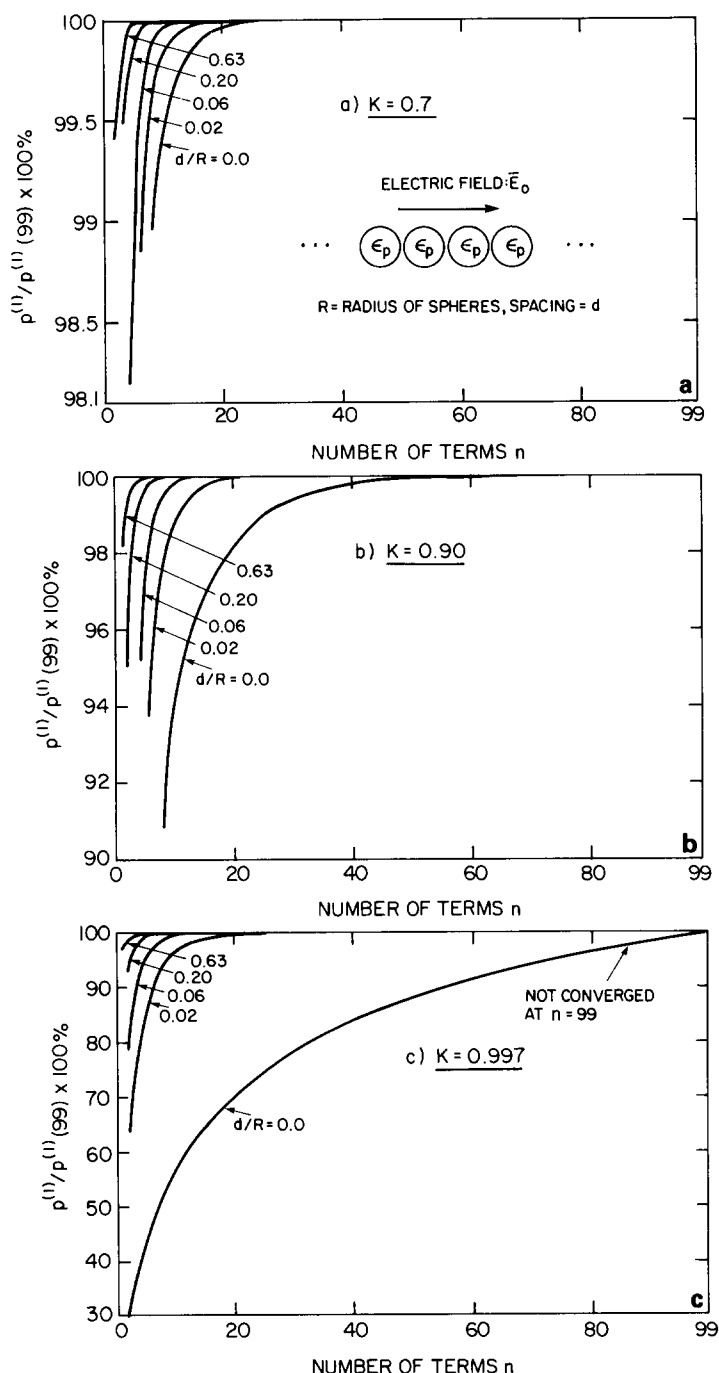


Fig. 7. Convergence plot of dipole moment per particle  $p^{(1)}(n)$  normalized to  $p^{(1)}(99)$  for an infinite chain of uniform dielectric spheres as a function of the number of multipolar terms  $n$  used in the calculation. (a)  $K = 0.70$  ( $\kappa = 8.0$ ), (b)  $K = 0.90$  ( $\kappa = 28.0$ ), (c)  $K = 0.997$  ( $\kappa = 1000$ ).

particles which are not in contact. As in section 3, attention is limited to the cases of two interacting spheres and infinite chains.

### *Two interacting spheres*

Consider first the problem of two identical dielectric spheres of radius  $R$  at spacing  $z = 2R + d$ . If the first  $L$  multipoles are used, then a system of  $L$  linear equations in the moments  $p^{(1)}, p^{(2)}, p^{(3)}, \dots, p^{(L)}$ , can be formulated and solved by iterative means [17] or matrix inversion techniques [18]. Results illustrating the sensitivity of the convergence upon  $K$  and  $d/R$  are shown in Figs. 6a and 6b. In both these figures, the normalized dipole moment  $p_{\text{eff}}(n)/p_{\text{eff}}(L)$  is plotted versus  $n$ , where  $p_{\text{eff}}(n)$  is the moment correct to  $n$  terms and  $L = 28$ . For  $K = 0.9$  ( $\kappa \equiv \epsilon_p/\epsilon_m = 28.0$ ), convergence is reasonably assured at  $n \sim 12$  when  $d/R = 0.02$ , while over twenty multipolar terms ( $n > 20$ ) are needed for  $K = 0.997$  ( $\kappa = 1000$ ) at the same spacing.

For particles in contact ( $d/R = 0$ ), convergence is not achieved when  $K > 0.9$  at  $n = 28$  terms. Attempts to gain convergence by increasing  $N$  are not necessarily successful because of truncation errors and underflow, even when double-precision variables are used. Based upon the extremely slow convergence we have found for the dipole moments  $p^{(1)}$ , it is evident that considerably more than the ten terms employed by Sancho et al. [12] would be required for accurate calculation of the interparticle forces when  $d/R \ll 0.2$ .

### *Infinite chains of spheres*

The results are qualitatively and quantitatively similar for infinite chains of equally spaced identical dielectric spheres. Due to symmetry, only odd multipoles ( $p^{(1)}, p^{(3)}, p^{(5)}, \dots, p^{(N)}$ ) are required. Using the method of Miller and Jones [11], calculations have been made of the dipole moment per particle  $p^{(1)}$  as a function of the number of terms  $n$  used in its evaluation. The results, shown in Figs. 7a,b and c, are normalized to values evaluated using  $L = 99$  multipolar terms, i.e.  $p^{(1)}(n)/p^{(1)}(99)$ . The rates of convergence seem comparable in every way to those for the two-sphere chains shown in Figs. 6a and b.

## **5. Conclusion**

Alward and Imaino use the dipole approximation in their analysis of the forces of interaction between single-component toner particles [9]. These particles are heterogeneous mixtures of small  $\text{Fe}_2\text{O}_3$  particles and pigments in a thermo-setting polymer matrix. The effective relative permeability of the toner particles is about 2, corresponding to  $K^{(1)} \approx 0.25$ . From Figs. 3 and 4, it is evident that the dipole approximation is adequate in this range of  $K^{(1)}$  values. Therefore, their calculations are expected to be accurate. However, they argue incorrectly that the dipole approximation is valid because of the heterogeneous nature of the toner particles. The fault with this argument is that the small

magnetite particles distributed through a toner particle experience significantly different magnetic field values and therefore have different induced dipole moments. This spatial distribution of dipoles with unequal moments signifies the existence of effective quadrupole and higher-order multipoles which are entirely analogous to the multipoles induced in an idealized and homogeneous toner. Strictly speaking, Alward and Imaino's argument would apply only if all the  $\text{Fe}_2\text{O}_3$  mass were located at the center of the toner particles. Use of the dipole approximation remains valid for magnetic toner particles because of their low magnetite loading and low resultant effective permeability.

In this paper, the multipolar interactions of uncharged, polarizable particles aligned parallel to an electric field have been studied. Attention has been focused on the dipole moment because (i) it is the dominant term for particles when  $K = (\epsilon_p - \epsilon_m) / (\epsilon_p + 2\epsilon_m) < 0.5$ , and (ii) the "dipole" approximation has been used almost exclusively in the literature. By scrutinizing the  $p^{(1)}$  moment, it has been possible to establish the limits under which the dipole approximation itself can be used to achieve an accurate result. We have quantified the contribution of higher-order multipoles for closely spaced particles as a function of the relative values of  $\epsilon_p$  and  $\epsilon_m$ . The availability of accurate results for dipole moments of chains of conducting spheres [14] has made it easier to test the convergence for close spacings and large values of  $\epsilon_p/\epsilon_m$ . The remaining task is to investigate the interparticle forces themselves, establishing the influence of the two important parameters (i.e., particle spacing and relative dielectric constant) on convergence and mapping out the conditions where a dipole approximation provides adequate accuracy.

### Acknowledgement

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