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Electrostatic Force on a Moist Particle Near a Ground Plane*

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The effect of surface moisture on the electrostatic force of attraction between an uncharged particle and a ground plane is investigated. Describing a moist sphere with an effective complex permittivity allows the image force to be calculated using complex linear multipoles. The number of multipoles needed for an accurate image force calculation is determined as a function of particle-plane separation. Adsorbed moisture introduces a relaxation in the electrostatic image force. For particles exposed to a range of relative humidities, the model predicts relaxation times that vary by several orders of magnitude. In addition, at small particle-plane separations adsorbed moisture can increase the image force by several orders of magnitude. Finally, multipolar moments are used to calculate the electric field in the gap separating the particle and ground plane to determine the onset of breakdown or field emission.

KEY WORDS multipole; dipole; moisture; surface conductivity, sphere; image force; relaxation; complex permittivity; breakdown; field emission; particle; moment; water; humidity; adhesion.

INTRODUCTION

Moisture, adsorbed in humid environments, greatly increases the surface conductivity of dielectric particles.¹⁻³ This change in a particle's electrical properties alters its electrostatic force of attraction and adhesion to surfaces. For several technologies, such as electrorheology and electrophotography, the electrostatic force on particles near surfaces needs to be controlled. Therefore, it is important to understand the effect of surface moisture on electrostatic forces.

An ohmic surface layer affects the electrical properties of a particle by preventing electric fields from entering the particle's bulk. For water layers, the conduction mechanism is ion migration. Ions migrate along the particle surface in response to an applied field causing an otherwise insulating particle to polarize as if it were lossy. In addition, the finite response time of the ions introduces a time dependence to the attractive force between the particle and ground plane. By examining a particle's effective dipole and multipole moments, the effect from adsorbed water on the electrostatic attraction of a particle to a nearby grounded surface can be determined.

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Evidence of the effect from adsorbed water on the electrical properties of particles and on electrostatic particle forces appears in the literature. One notable area is electrorheology, where particle surface moisture activates the mechanism controlling the liquid-to-solid transformation.⁴ Research on single particles by Tombs and Jones⁵ illustrates the time dependence brought on by adsorbed moisture which manifests as a relaxation in the particle's dipole moment. Using atomic force microscopy, Mizes⁶ measured the force on a single particle in air at a fixed distance above a ground plane. When an external field was applied, the force increased over time by an order of magnitude. He attributed the increase to surface conductivity. Schaefer *et al.*,⁷ also using atomic force microscopy, measured the removal force of individual particles from surfaces in air and vacuum. With air in the chamber the force increased over time, suggesting an effect from adsorbed moisture. However, it is unknown whether the increase in removal force is due to a capillary effect.

This article concentrates specifically on the electrostatic image force, which is generated by an external electric field, on an uncharged spherical particle near a ground plane. Of course, other particle forces can be substantial and at times dominate, especially when a small particle is in contact with a surface. For details on other forces, such as van der Waals, capillary, double layer, magnetic, and chemical, the reader is referred to the extensive body of literature that exists on these topics.^{8,9}

In this article, the magnitude and time dependence of the attractive force between a moist spherical particle and a ground plane is examined by extending Jones's¹⁰ method of images to include lossy particles with concentric layers. The effective complex permittivity of the moist particle is used to calculate the image force. Next, the condition for breakdown or field emission between the particle and ground plane is derived. The importance of multipolar images is then examined as a particle approaches a ground plane and the number of multipoles that need to be considered for an accurate force calculation is determined. Next, estimates of the image force on a moist dielectric particle are given. Finally, the dependence of the image force on the frequency of the applied field is shown and discussed.

THEORY

A dielectric particle polarizes in the presence of an electric field. If the field is non-uniform the particle experiences a force, known as the dielectrophoretic force.¹¹ The polarization of a spherical particle in a non-uniform field can be described by an expansion of linear multipoles. Typically, only the dipole moment is considered because the influence of the higher order moments are often insignificant.¹⁰ However, as a particle nears a ground plane the higher order moments become important and, for small separations, must be considered. In addition, when a neutral particle nears a ground plane there is a dielectrophoretic force that acts on the particle even in a uniform applied field because the particle distorts the field.

Jones¹⁰ derived the dielectrophoretic force on a single lossless sphere in a nonuniform axially symmetric field. He used linear multipoles to describe the field from any axially symmetric electrode structure, then calculated the multipoles induced in the dielectric sphere. Summing the forces acting on each multipole yields the total force on the sphere. The same technique was used by Fowlkes and Robinson¹² to calculate the image force on a single lossless spherical particle near a ground plane. They derived the field produced by multipolar images in the ground plane that results from a uniform field interacting with a sphere. Using symmetry arguments to calculate the multipolar moments of the particle, the image force was then found by summing the force on each multipole. The theory can be extended to include lossy dielectric particles in lossy media by describing the electrical properties of the particle with a complex permittivity. Sancho *et al.*,¹³ employed complex permittivities to calculate the interactions between 2 lossy spheres in a similar manner but only used 10 multipoles in their calculation. Here, calculations of the force between a lossy multi-layered sphere and a ground plane are presented. In addition, the number of multipoles needed to achieve acceptable precision is determined for a range of conditions.

Consider a lossy dielectric sphere having conductivity, σ_2 , and dielectric constant, κ_2 , in a lossy medium of conductivity, σ_1 , and dielectric constant, κ_1 (see Figure 1a). For wavelengths large compared with the size of the particle, the complex permittivity describing the particle is

$$\underline{\varepsilon}_2 = \kappa_2 \varepsilon_0 - j\sigma_2 / \omega \tag{1}$$

while the complex permittivity of the surrounding medium is

а

<u></u>22

 (κ_2,σ_2)

$$\underline{\varepsilon}_1 = \kappa_1 \varepsilon_0 - j\sigma_1 / \omega \tag{2}$$

where ω is the radian frequency of the applied electric field, $\varepsilon_0 = 8.854 \times 10^{-12}$ F/m, and $j = \sqrt{-1}$. Effective complex permittivities can also be used to model multilayered particles, such as moist particles and the coated carrier particles found in some two-component electrophotographic developers. A sphere of radius *R*, coated with a single layer of thickness δ , conductivity σ_3 , and dielectric constant κ_3 (Figure 1b) has an effective complex permittivity¹⁴ of

$$\underline{\varepsilon}_{2eff} = \underline{\varepsilon}_{3} \left[\frac{((R+\delta)/R)^{3} + 2(\underline{\varepsilon}_{2} - \underline{\varepsilon}_{3})/(\underline{\varepsilon}_{2} + 2\underline{\varepsilon}_{3})}{((R+\delta)/R)^{3} - (\underline{\varepsilon}_{2} - \underline{\varepsilon}_{3})/(\underline{\varepsilon}_{2} + 2\underline{\varepsilon}_{3})} \right]$$
(3)

b

δ



FIGURE 1 The electrical properties of a spherical particle can be described by an effective complex permittivity: a) uniform lossy sphere; b) sphere with an outer layer of thickness δ .

$$\underline{\varepsilon}_3 = \kappa_3 \varepsilon_0 - j \sigma_3 / \omega \tag{4}$$

Note that Eq. (3) does not depend on the electrical properties of the medium.

If the effective complex permittivity of a multi-layered particle and a uniform particle are the same, the two particles respond equivalently to any external field. Therefore, once the effective complex permittivity of a particle is known, the particle can be treated as if it were uniform when calculating the electrostatic force on the particle. Effective permittivities can also be identified for spheres with more than one layer, such as biological cells.¹⁵

Equation (3) describes the effective complex permittivity of a moist particle, but the equation can be simplified when $\sigma_2 \ll \sigma_3$ and $\delta \ll R$ to

$$\varepsilon_{2\text{eff}} = \kappa_2 \varepsilon_0 - j(\sigma_2 + 2\lambda/R)/\omega, \tag{5}$$

where λ is the particle's surface conductivity in Siemens (1/ Ω):

$$\lambda = \sigma_3 \delta \tag{6}$$

Miles and Robertson¹⁶ and later O'Konski^{17,18} derived Eq. (5) directly by solving the potentials inside and outside a sphere having a thin conductive surface film.

Once the effective permittivity of the particle is identified, the dipole and multipole moments of the particle can be calculated. Both the multipolar moments and the image force were derived by following the method outlined by $Jones^{10}$ and Fowlkes and Robinson¹² except, here, complex permittivities describe the medium and particle. For a neutral particle near a ground plane (see Figure 2) the moments induced by a uniform, alternating electric field, E(rms), are described by the following infinite set of complex linear equations:

$$\underline{p}^{(n)} = \frac{(-1)^{n} R^{2n+1}(\underline{\varepsilon}_{2} - \underline{\varepsilon}_{1})}{(n-1)!(n\underline{\varepsilon}_{2} + (n+1)\underline{\varepsilon}_{1})} \sum_{k=0}^{\infty} \left[\frac{(n+k)!(-1)^{k} \underline{p}^{(k)}}{k!(2R+2s)^{n+k+1}} \right] + \frac{4\pi\kappa_{1}\varepsilon_{0} ER^{3}(\underline{\varepsilon}_{2} - \underline{\varepsilon}_{1})}{\underline{\varepsilon}_{2} + 2\underline{\varepsilon}_{1}} \delta(n-1)$$
(7)



FIGURE 2 Image force calculations are made for a mosit neutral dielectric sphere spaced a distance, s, from a ground plane.

where n = (1, 2, 3, ...), s is the distance separating the particle from the ground plane, and the delta function, $\delta(n-1)$, equals 1 when n = 1 but otherwise equals 0. The monopole moment $p^{(0)}$ equals zero for the system studied here because the sphere has no net charge. Once the multipolar moments are found, the image force between the particle and group plane can be calculated. In an alternating field the time averaged force was found to be

$$\langle F_{image} \rangle = \frac{1}{8\pi\kappa_1 \varepsilon_0} \sum_{n=0}^{\infty} \sum_{k=0}^{\infty} \frac{(-1)^{n+k+1}(n+k+1)! p^{(n)} p^{(k)}}{n! k! (2R+2s)^{n+k+2}}$$
(8)

The image force increases quadratically as the applied electric field rises. However, the field is ultimately limited by breakdown and field emission. Breakdown is initiated in the gap between the particle and ground plane because the particle focuses the field. While this limitation is not represented in the equations above, the field in the gap is readily calculated by summing the applied field with the field produced by the multipoles of the particle and multipolar images in the ground plane. Using Jones's¹⁰ derivation of the field produced by a set of multipoles, the field along the z-axis of the particle was found to be

$$\underline{E}_{\text{total}}(z) = \underline{E} + \frac{1}{4\pi\kappa_2\varepsilon_0} \sum_{n=0}^{\infty} (n+1)\underline{p}^{(n)} \left[\frac{1}{(z-R-s)^{n+2}} \frac{1}{(-z-R-s)^{n+2}} \right]$$
(9)

where z = 0 corresponds to the ground plane. Breakdown or field emission resulting from excessive fields alters the particle charge thereby changing both the image force and Lorentz force (particle charge multiplied by the applied field) on the particle.

To calculate either the image force or the electric field, the set of complex linear equations (Eq. (7)) for the multipolar moments of the particle are first solved. Equations (7), (8), and (9) cannot be solved exactly, thus the number of multipoles used in the calculation determine the accuracy. After calculating the coefficients of the complex linear equations, the set of equations is simultaneously solved using standard algorithms of linear algebra. Finally, the image force and field are found by substituting the moments of the sphere into Eqs. (8) and (9), which must also be appropriately truncated.

Some conditions require significantly more multipoles than others to achieve acceptable precision, thus it is important to identify the conditions that require the inclusion of higher order moments. In general, conducting particles require the largest number of multipoles for a given accuracy and separation distance. Figure 3 illustrates the contributions from higher order multipoles at a range of separation distances $(R = 10 \,\mu\text{m}, \kappa_1 = 1, \kappa_2 = 3, E = 0.1 \,\text{V}/\mu\text{m})$. When the sphere is far from the ground plane, relative to its size, the image force depends solely on the dipole moment of the particle, as expected. However, the importance of the multipolar images increases greatly as the particle approaches the ground plane. At small separations higher order multipoles cannot be ignored. For example, Figure 3 indicates that, for a 1 nm separation, the total force is more than 2 orders of magnitude larger than the force due only to the dipole moment.

Figure 4 shows the number of multipoles needed to achieve 0.1% accuracy for a uniform conducting particle and also for uniform dielectric particles with dielectric constants of 10 and $3(R = 10 \,\mu\text{m}, \kappa_1 = 1)$. For dielectric particles with surface moisture



FIGURE 3 The image force on a spherical conducting particle versus the number of multipoles used in the calculation ($R = 10 \mu m$, $\kappa_1 = 1$, $\kappa_2 = 3$, $E = 0.1 V/\mu m$). Higher order multipoles become increasingly important as the separation distance, s, decreases.



FIGURE 4 The number of multipoles required to achieve 0.1% accuracy in the force calculation as a function of the particle-plane separation distance ($R = 10 \,\mu$ m, $\kappa_1 = 1$). For moist dielectric particles in the low frequency limit the curve for the conducting particle is used, while in the high frequency limit the dielectric constant of the particle's bulk determines the correct curve.

the number of multipoles needed for an accurate calculation depends on the frequency of the applied field. In the low frequency limit a moist dielectric particle has the same behavior as a conducting particle because the surface layer completely screens the electric fields from the bulk of the particle. As the frequency increases, the response of the surface layer lags behind the alternating field. In the high frequency limit the field alternates too fast for the surface layer to affect the image force. The dielectric constant of the bulk then determines the image force and also the number of multipoles needed for an accurate calculation. For example, in Figure 4 the curve for the conducting particle also applies to moist dielectric particles in the low frequency limit, while in the high frequency limit the appropriate curve is determined by the dielectric constant of the bulk.

Although Figure 3 indicates that the force increases without bound as a conducting particle (or moist dielectric particle in the low frequency limit) nears a ground plane, short range repulsive forces limit the separation distance to approximately 0.4 nm (for this reason the curves in Figure 4 are truncated at 0.4 nm). Because the calculation only diverges when the separation distance is zero, a finite number of multipoles can be used to calculate accurately the image force for any realistic condition. For a conducting particle with a 10 μ m radius spaced 0.4 nm from a ground plane, 620 multipole moments are needed to achieve 0.1% accuracy. In practice, the numerical calculation of more than 500 multipolar moments becomes difficult on a typical computer workstation because: 1) the magnitude of the coefficients becomes excessive (due to the factorial, (n + k)!/k!, in Eq. (7)) and 2) a large amount of memory is required to store the coefficients and intermediate results when solving the set of linear equations.

RESULTS

The remainder of this article concentrates solely on moist dielectric particles, particularly on the time dependence associated with the ohmic surface layer. Consider a moist dielectric sphere in air separated from a ground plane by a distance, s. If the surface layer behaves ohmically, the particle exhibits a Debye relaxation in the image force when an electric field is applied. At low frequencies the surface layer relaxes completely and expels all electric fields, while at high frequencies, where the response time of the ohmic surface layer is much slower than the alternating field, the force depends entirely on the bulk. The difference between the image force at high frequencies and low frequencies depends largely on the separation distance between the particle and the ground plane and on the dielectric constant of the particle (see Figure 5). Of course, it should be realized that the behavior of particles as a function of frequency also describes the time dependence of the image force when a DC field is suddenly applied.

A particle's surface conductivity depends on the amount of adsorbed water and the bulk conductivity of that water layer. Surface conductivity is affected by many factors, such as the relative humidity that the particle is exposed to, surface contamination, the particle's composition, and surface morphology.^{3,19} Figure 6 illustrates the strong dependence of the response time on the surface conductivity of a sphere spaced 0.1 µm from a ground plane ($R = 10 \,\mu\text{m}$, $\kappa_1 = 1$, $\kappa_2 = 3$, $E = 0.1 \,\text{V}/\mu\text{m}$). The curve for a lossless particle is also shown and, as expected, has no frequency dependence. The range of



FIGURE 5 The difference between the image force at high frequencies compared with low frequencies depends on the dielectric constant of the particle and on the separation distance between the particle and the ground plane ($R = 10 \,\mu$ m, $\kappa_1 = 1, \lambda = 10^{-13} S$, $E = 0.1 \,\text{V/}\mu\text{m}$).



FIGURE 6 An ohmic surface layer causes a relaxation in the image force on a particle. The frequency at which the relaxation occurs depends on the surface conductivity and the radius of the particle $(R = 10 \,\mu\text{m}, s = 0.1 \,\mu\text{m}, \kappa_1 = 1, \kappa_2 = 3, E = 0.1 \,\text{V}/\mu\text{m}).$

surface conductivities chosen for the calculations presented in Figure 6 are based on measurements made on individual glass particles exposed to relative humidities (RH) ranging from 15% to 90%: $\lambda \leq 10^{-14}$ S roughly corresponds to a particle exposed to 15% RH and $\lambda = 10^{-9}$ S corresponds to a particle exposed to 90% RH.²⁰ At high relative humidities the image force on a particle can relax very quickly. At low humidities the same particle takes much longer to relax. Figure 6 shows the enormous difference in the relaxation time between moist particles and dry particles: at least 5 orders of magnitude. Note that the response time of the image force depends not only on the surface conductivity but also on the size of the particle, which is evident from Eq. (5).

In Figure 7 the effect that a layer of surface moisture has on the image force is shown as the sphere approaches the ground plane ($R = 10 \,\mu\text{m}, \kappa_1 = 1, \kappa_2 = 3, E = 0.1 \,\text{V/}\mu\text{m}, \lambda = 10^{-13} \text{ S}$). In the high frequency limit the force is relatively small at any separation. As the frequency of the applied field decreases, the force increases. For small separations the image force increases by more than 3 orders of magnitude. When considering particles that are placed in DC electric fields or when a DC field is suddenly applied, it is evident from Figure 7 that the image force can increase dramatically over time for moist particles.

As previously mentioned, the image force increases quadratically with the applied field. The image force, however, is limited by electric field breakdown. Because the particle focuses the field, breakdown initiates in the particle-plane gap. In all of the plots previously described the applied field was fixed at 0.1 V/ μ m, which is too small to induce breakdown. Figure 8 demonstrates the effects of spacing and frequency on the



FIGURE 7 The image force is a strong function of the separation distance and increases drastically with time ($R = 10 \,\mu\text{m}, \kappa_1 = 1, \kappa_2 = 3, E = 0.1 \,\text{V}/\mu\text{m}, \lambda = 10^{-13} \text{S}$).



FIGURE 8 The electric field magnitude at the particle's pole versus the frequency of the applied field $(R = 10 \,\mu\text{m}, \kappa_1 = 1, \kappa_2 = 3, \lambda = 10^{-13} \text{ S})$. The external field is magnified by the particle, thus excessive fields may induce breakdown or field emission at small separations.

magnitude of the electric field when the applied field is $10 \text{ V/}\mu\text{m}$. The field at the particle's pole nearest to the ground plane is shown for $R = 10 \,\mu\text{m}$, $\kappa_1 = 1$, $\kappa_2 = 3$, and $\lambda = 10^{-13} \text{ S}$. The field is largest at the low frequency limit, where it increases as the separation distance decreases. The solid line indicates the onset of field emission in small air gaps ($< 5 \,\mu\text{m}$). Field emission occurs at approximately 75 V/ μ m but depends somewhat on the composition of the surface.²¹ When the separation distance is 10 nm, the field exceeds the 75 V/ μ m limit, but only at low frequencies. In electrophotographic devices it is not uncommon for toner particles to experience fields as high as 40 V/ μ m, thus field emission and air breakdown may be a concern. It should be noted that the image force calculation, Eq. (8), is no longer valid when field emission or other forms of breakdown occur.

CONCLUSION

The multipole image theory^{10,12} has been extended to include lossy multilayered spheres near a ground plane. The dipole approximation is clearly inadequate when calculating the image force on a particle spaced less than a few diameters from a ground plane. The number of multipoles needed for an accurate image force calculation was determined as a function of particle-plane separation and only diverges when the separation equals zero, a physically impossible situation.

Surface moisture was shown to cause a relaxation in a particle's image force. The relaxation time depends on the surface conductivity and particle radius and can vary by at least 5 orders of magnitude for particles exposed to a range of relative humidities. At small particle-plane separations, adsorbed moisture was shown to increase the image force by several orders of magnitude. Multipolar moments were also used to calculate the electric field in the gap separating the particle and ground plane. Electric field breakdown or field emission can limit the image force when applied fields are large. Experiments, geared specifically toward measuring the relaxation effects predicted here, are currently being explored using an atomic force microscope to examine the effects on individual particles.

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