

University of Nebraska - Lincoln

DigitalCommons@University of Nebraska - Lincoln

Faculty Publications from the Department of
Electrical and Computer Engineering

Electrical & Computer Engineering, Department
of

12-25-2000

The Electrostatic Interaction of Charged, Dust-Particle Pairs in Plasmas

M. E. Markes

University of Nebraska-Kearney, Kearney, NE

P. F. Williams

University of Nebraska-Lincoln

Follow this and additional works at: <https://digitalcommons.unl.edu/electricalengineeringfacpub>



Part of the [Electrical and Computer Engineering Commons](#)

Markes, M. E. and Williams, P. F., "The Electrostatic Interaction of Charged, Dust-Particle Pairs in Plasmas" (2000). *Faculty Publications from the Department of Electrical and Computer Engineering*. 104.
<https://digitalcommons.unl.edu/electricalengineeringfacpub/104>

This Article is brought to you for free and open access by the Electrical & Computer Engineering, Department of at DigitalCommons@University of Nebraska - Lincoln. It has been accepted for inclusion in Faculty Publications from the Department of Electrical and Computer Engineering by an authorized administrator of DigitalCommons@University of Nebraska - Lincoln.

The electrostatic interaction of charged, dust-particle pairs in plasmas

M. E. Markes¹ and P. F. Williams²

¹Department of Physics, University of Nebraska-Kearney, Kearney, NE 68849, USA

²Department of Electrical Engineering, University of Nebraska-Lincoln, Lincoln, NE 68588-0511, USA
(Corresponding author, email: pfw@moi.unl.edu)

Abstract

We report the results of a study of the electrostatic interaction between negatively charged particles in a plasma. The goal of the study was to investigate the possibility of an attractive interaction which would make possible the formation of “molecules” of particles. For all approximations relating the positive ion density to the local electrostatic potential that we examined, we find that the interaction is repulsive for all particle separations.

Keywords: Dusty plasma particle interaction

1. Introduction

Small particles can be confined to a nearly planar region just above the driven electrode in RF-driven plasmas. Under proper conditions they will arrange themselves into a regularly spaced array called a “Coulomb crystal” [1]. Similar structures have been seen in colloidal suspensions [2], the positive column of glow discharges [3], thermal plasmas [3], and ion traps [4]. This phenomenon has attracted considerable interest because of the insight it may lend into the formation and dynamics of conventional crystals, and because of possible technological applications.

A dust particle becomes negatively charged in the plasma in order to balance the electron and ion currents impinging on the surface. In the conventional model of a Coulomb crystal, the negatively

charged dust particles are spatially constrained by electric fields associated with the electrode geometry and other factors such as momentum transfer from the ion drift current. Acting together, these forces form a trapping site, within which electrostatic repulsion between like-charged dust particles produces a lowest energy configuration having translational symmetry [5]. In this model, the interparticle force is repulsive. Condensation into a localized “crystal” is the result of the confining potential.

There is some empirical evidence that under the proper conditions an attraction may exist between charged dust particles without the presence of a confining potential. Chen et al. [6] present photographs of isolated “molecules” containing six particles, and claim these to be evidence of an attraction between the particles which is not due to

a trap-confinement mechanism. Further, there are several reports that particles in adjacent strata are aligned, with the particles in the upper stratum aligned directly above those of the lower [7, 8]. These data might also be interpreted to imply an attractive interaction. Recently, however, Melzer et al. have shown that this behavior is probably due to the attraction of the lower dust particle to an ion-rich wake formed by the upper particle [9]. The existence of a wake-mediated mechanism does not preclude an attraction between the particles themselves, but it does show that the observation of interstratum alignment does not imply it. Konopka et al., on the other hand, report a direct measurement of the interparticle potential function in directions parallel to the electrode surface. They find a repulsive potential, which is well described by a screened potential similar to the Debye–Hückel result [10].

The idea of a net attractive force between dust particles is intriguing, in that it raises the possibility of dust “molecules”. Several workers have proposed mechanisms that yield a net attraction between charged dust particles. However, most are based on ion inertial effects [11–13] rather than a purely electrostatic mechanism as in molecular bonds. An exception is a model proposed by Resendes et al. [14]. A similar model has also been proposed independently by Chen et al. [6]. These authors calculate the total electrostatic energy of a system of two particles as a function of the particle separation. The resulting curve has a minimum, reminiscent of a Morse potential. They interpret this minimum as implying a net attraction between the particles. The result of Resendes et al. has been used by Astrakharchik et al. [15] to model the formation and dynamics of particle clusters in a plasma.

In this Letter we have two objectives. First, we point out that the use of total electrostatic energy curves (Refs. [6 and 14]) in a non-closed system is not appropriate for a force determination. A negative slope in the potential energy vs. particle separation graph, for this type of system, does not necessarily imply an attraction because of an indeterminate energy flow between the plasma and the system. The force can be calculated directly from the electrostatic force on the particle in question. For the Debye–Hückel approximation, this calculation is simple, and the result is that the force is everywhere repulsive.

Because of the novelty of a classical “molecular” bond between the particles, we were interested to see if other approximations relating electron density to electrostatic potential might produce an attraction based on an electrostatic mechanism. Accordingly, we have carried out calculations for several such approximations. Our goal was to determine whether or not a net electrostatic attractive force might occur under any conditions. The second objective of this paper is to report and discuss these results. In all cases we have investigated, we find a repulsive force on the particles for all separations. All our approximations assume thermalized positive ions. This assumption is certainly not valid in the sheath of the driven electrode of a low-pressure RF plasma where the ion wind produces a force apparently attracting the downwind particle to the upwind, but not vice versa [9]. Our results complement these findings in that they show that a purely electrostatic mechanism of attraction is unlikely.

2. Debye–Hückel approximation

In the Debye–Hückel approximation the potential is a solution to the Poisson equation

$$\nabla^2 \Phi = -\rho/\epsilon_0, \quad (1)$$

where Φ is the electrostatic potential and ρ is the charge density. In addition, the charge density is taken to be a linear function of the local electrostatic potential, Φ :

$$\rho = -\frac{q_e^2 n_0}{K T_p} \Phi, \quad (2)$$

where q_e is the electronic charge, n_0 the plasma density, T_p the “parallel” combination of the electron and ion temperatures, and the ions are assumed to be singly ionized. Using Equation (2) in Equation (1) yields, for an isolated spherical particle of charge Q and radius a ,

$$\begin{aligned} \Phi(r) &= \frac{Q}{4\pi\epsilon_0(1+a/\lambda_D)r} e^{-(r-a)/\lambda_D} \\ &\approx \frac{Q}{4\pi\epsilon_0 r} e^{-r/\lambda_D}, \end{aligned} \quad (3)$$

where $\lambda_D = (\epsilon_0 K T_p / q_e^2 n_0)^{1/2}$, and the approximation is valid for $a \ll \lambda_D$.

Equation (3) applies to an isolated particle, but since Equation (2) is linear in Φ , the potential for multiple non-polarizable particles is the sum of

terms like Equation (3), one for each particle. Thus, the total electrostatic energy of two particles of charge $-Q$ immersed in the plasma is

$$U_T = \frac{1}{2} \int (\rho_1 + \rho_2)(\Phi_1 + \Phi_2) dV. \quad (4)$$

where $\rho_{1,2}$ includes the charge density corresponding to the particles and the sheath. This integral can be evaluated analytically for the case of two point particles, yielding

$$U_T(r) = \frac{Q^2}{8\pi\epsilon_0} \left[\frac{2}{r} - \frac{1}{\lambda_D} \right] e^{-r/\lambda_D}. \quad (5)$$

Equation (5) has a minimum at $(1 + \sqrt{3})\lambda_D \approx 2.73\lambda_D$. Resendes et al. [14], obtain the same result, as have we [16] and Riley [17]. Chen et al. [6] use a simpler, more approximate method to calculate the energy, but arrive at a similar conclusion.

It is tempting to calculate the force from dU_T/dr , using Equation (5). This is the procedure implied by Resendes et al. [14] and Chen et al. [6]. It is incorrect in this case, however, because the system is not isolated. In changing the separation by dr , beside the mechanical work, $F dr$, the plasma ambient also supplies or sinks energy, which must be considered. The correct result for the interparticle force can be obtained simply from $F = -QE$, where E is the electric field at one particle from all charges except that particle. Because the Poisson equation with the Debye–Hückel approximation is linear, superposition applies, and the plasma charge density is the superposition of two spherically-symmetric distributions given by (2) and (3). The result is everywhere repulsive, with magnitude (in the limit $a \ll \lambda_D$)

$$F = \frac{Q^2}{4\pi\epsilon_0} \frac{1 + r/\lambda_D}{r^2} e^{-r/\lambda_D}. \quad (6)$$

3. Other approximations to the plasma state

Two assumptions underlie the Debye–Hückel approximation: thermodynamic equilibrium, and the validity of a linearization of the resulting Boltzmann exponentials. Both assumptions are invalid for conditions typical of a Coulomb crystal experiment. In order to explore further the possibility of an electrostatic attraction between particles we have also investigated other approximations to the plasma state. These approximations are also of

limited validity. However, they do provide some insight into the range of behaviors the underlying mathematics might allow.

All cases we have investigated assume that the charge density is a function of the local potential, Φ , and this function is used in Equation (1). Since the Debye–Hückel approximation involves a linear relationship, we also investigated both sublinear and superlinear relations in an attempt to bound the likely behavior. For the calculations we describe here, we considered two particles, and assumed cylindrical symmetry along the interparticle axis. In order to investigate possible geometric effects, we have looked at two cases: (1) the particles are cylinders aligned with the interparticle axis, and (2) the particles are spheres. For the cylinders, the diameter and length were taken to be equal, and for comparison the diameter of the sphere was taken the same as that of the cylinder. For the cylinders, Equation (1) was solved using a standard, rectangular-mesh, finite difference, successive over-relaxation (SOR) method, and the force was calculated from the surface fields according to

$$\vec{F} = \frac{1}{2} \epsilon_0 \oint_S E_n^2 \hat{n} dA, \quad (7)$$

where \hat{n} is a unit vector normal to the surface. This expression assumes the surface, S , is an equipotential.

For the spheres, a standard triangular-mesh, finite elements, SOR method was used. The force was calculated by calculating the Coulomb-law force between the two particles and between one particle and the surrounding positive ion sheath. In the latter case, the surface charge density on one particle is calculated from the surface electric field, and the particle surface and the surrounding ion sheath are divided into thin, coaxial rings. The force between rings at (r_j, z_j) and (r_i, z_i) can be shown to be

$$F_{i,j} = \frac{2r_i r_j (z_j - z_i) \lambda_i \lambda_j}{\epsilon_0 [(r_j - r_i)^2 + (z_j - z_i)^2]} \times \frac{E \left(\sqrt{\frac{4r_i r_j}{(r_j + r_i)^2 + (z_j - z_i)^2}} \right)}{\sqrt{(r_j + r_i)^2 + (z_j - z_i)^2}}, \quad (8)$$

where $E(x)$ is the complete elliptic integral of the second kind, and λ_i is the linear charge density of the i th ring. The force on the particle is then ob-

tained by summing the force between appropriate pairs of rings.

In both methods the particles were taken to have the same fixed potential, and the system was placed inside a large cylinder with a grounded surface. The ion and electron temperatures were taken as 0.026 and 1.0 eV, respectively, for all calculations presented in this Letter. Several values of the plasma density and particle size were investigated in the course of this study, but here we report results only with $n_0 = 1 \times 10^8 \text{ cm}^{-3}$ (corresponding to $\lambda_D \approx 120 \text{ }\mu\text{m}$), and particle diameter and length of $33 \text{ }\mu\text{m}$ ($\approx 0.28 \lambda_D$). Several values of particle potential were also investigated, but here we report results only for -0.2 V . For both the cylindrical and spherical particles the solution volume was 0.2 cm ($\approx 16.7 \lambda_D$) in diameter and length, with $\Phi = 0$ on the volume surface. (We made use of symmetry about the $z = 0$ plane to cut the solution volume in half.)

3.1. Collisionless approximation (sublinear dependence)

A well-known approximation for the dependence of ion density on potential was developed to describe the behavior of Langmuir probes. The approximation assumes either thermalized or monoenergetic ions in a boundary region surrounding the probe (or particles in our case), and collisionless trajectories for the ions inside this boundary. Except for difficulties related to bound orbits and ion collisions with the probe (particles), the ion density is easily worked out, and turns out to be a function of the local potential, Φ . If we assume thermal ions, with temperature T_i at the boundary region, the result is

$$n_i(\vec{r}) = n_0 \left[\frac{2}{\sqrt{\pi}} \sqrt{-q_e \Phi(\vec{r}) / K T_i} + \operatorname{erfc}(\sqrt{-q_e \Phi(\vec{r}) / K T_i}) e^{-q_e \Phi(\vec{r}) / K T_i} \right], \quad (9)$$

where n_0 is the plasma density and q_e is the unsigned electronic charge. To obtain this result, we assumed the central particle to be transparent to the ions, and we neglected bound orbits.

Applying the same conditions to the electrons yields a Boltzmann-like result

$$n_e(\vec{r}) = n_0 e^{-q_e \Phi(\vec{r}) / k T_e}. \quad (10)$$

We solved Equation (1) for a range of interparticle separations with the charge density given by

$$\rho = q_e(n_i - n_e). \quad (11)$$

The result for cylindrical particles, along with the results for the other cylindrical particle cases we investigated, and the analytic Debye–Hückel result are shown in Figure 1. Similar ($\pm 12\%$) results were obtained for spherical particles over the range of separations from about 1 to $4 \lambda_D$.

We also calculated the total electrostatic energy as a function of particle separation according to Equation (4). The result was a Morse-like curve similar to, but shallower than, the Debye–Hückel result.

3.2. Boltzmann approximation (superlinear dependence)

If the system were in local thermodynamic equilibrium (LTE), the electron and ion densities would be given by Boltzmann distributions. Because of recombination at the particle surface, LTE is not realized, and, at least near the particle, the Boltzmann distribution predicts a substantially larger ion density than is likely to be present. The Boltzmann distribution does provide an interesting example for comparison with the collisionless model in that the dependence of ion density on potential is superlinear, rather than sublinear. For this reason, we have carried out numerical solutions similar to those used for the collisionless approximation, for charge density given by

$$\rho(\Phi) = n_0 q_e (e^{-q_e \Phi / k T_i} - e^{q_e \Phi / k T_e}). \quad (12)$$

The interparticle force vs. separation for cylindrical particles calculated using Equation (12) is also shown in Figure 1.

3.3. Debye–Hückel approximation (linear dependence)

Primarily as a check of our numerical method, we have carried out numerical calculations using the Debye–Hückel approximation for the charge density (Equation (2)). The results for cylindrical particles are also shown in Figure 1, along with the “analytic” result for the Debye–Hückel model. The spherical-particle results were similar.

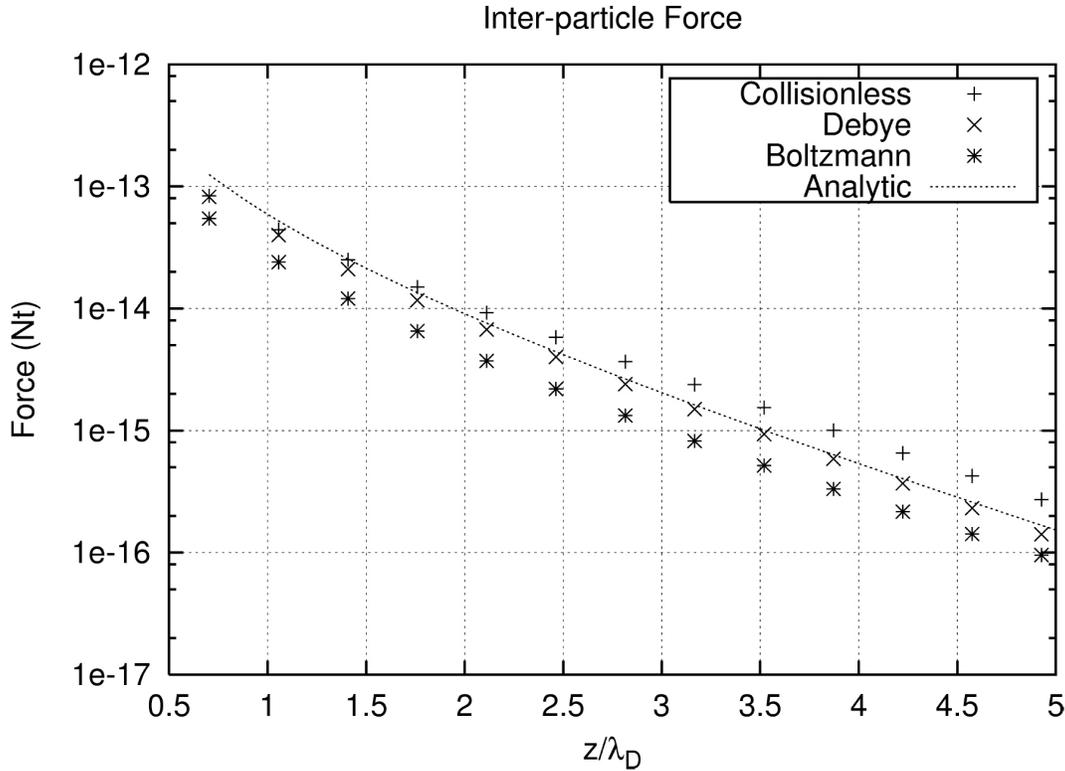


Figure 1. Plot of the cylindrical-particle interparticle force vs. particle separation, calculated numerically, for the collisionless, the Debye, and the Boltzmann approximations. Also shown is the result of the Debye approximation, calculated analytically. The particle potentials were -0.2 V, and the other parameters were as given in the text.

The “analytic” result takes the potential produced by one particle to be of the form of Equation (3), calculates the electric field by taking the gradient of that potential at the location of the second particle, and then determines the force on the second particle by multiplying the field at the second particle by the charge on the particle. The resulting force is

$$F = 4\pi\epsilon_0 V_0^2 \left(\frac{a}{x}\right)^2 \left(1 + \frac{a}{\lambda_D}\right) \left(1 + \frac{x}{\lambda_D}\right) \times e^{-(x-a)/\lambda_D}, \quad (13)$$

where a is the radius of the (assumed spherical) particle. This problem differs slightly from the problem solved numerically in three ways:

1. The particles are taken to be spherical, whereas the finite-difference numerical calculation uses cylindrical particles.
2. Taking the potential to be the superposition of isolated-particle potential functions is slightly in error because the resulting potential does not

satisfy the boundary condition that $\Phi = V_0$ on the particle surfaces.

3. The force is taken to be QE , where Q is the total charge on the particle and E is the field at the center of the particle from all sources except the particle itself. In the numerical calculations the force was obtained from the spatially varying σ and E on the particle surface.

For particles much smaller than the separation, these differences should not affect the results significantly, and good agreement is expected. As seen in Figure 1, the agreement is very good.

4. Discussion

The particle potentials for the results shown in Figure 1 were rather arbitrarily chosen as -0.2 V. This potential is approximately the potential for which the electron and ion fluxes impinging on the surface of a particle are equal for the case of a Boltzmann ion distribution function. Since the ion density, at

the particle surfaces, for other distribution functions (Debye and collisionless) is lower, the charging potential in steady state for these cases is correspondingly larger in magnitude. However, we chose to present data for a single potential for all distribution functions to facilitate comparison, and the equal-flux potential under the Boltzmann case was chosen because the Boltzmann ion density becomes unphysically large for much larger (in magnitude) potentials.

We have carried out similar calculations with assumed particle potentials around -1.0 V for the collisionless and Debye approximations. We have also examined several other approximations, as discussed below, and in all cases find only repulsive force.

In addition to the assumed forms for $n_i(\Phi)$ discussed above, we have also looked at several other, quite unphysical, relationships in an attempt to see if an attractive force could be produced by any local approximation for n_i . In all cases, the electron density was taken to be a Boltzmann distribution, except that in no case was the net charge density allowed to become negative. Three cases were investigated:

1. The ion density was taken to be a Boltzmann function of Φ for $\Phi \geq \Phi_0$, and to have the collisionless form of Equation (9) for $\Phi < \Phi_0$, where Φ_0 is an arbitrarily chosen threshold usually taken to be -0.1 to -0.2 V.
2. $n_i(\Phi) = n_0 e^{-q_e \Phi/kT}$, $\Phi \geq \Phi_0$
 $n_i(\Phi) = n_0 e^{-q_e (2\Phi_0 - \Phi)/kT}$, $\Phi < \Phi_0$.

Here Φ_0 is an arbitrarily chosen threshold potential, usually taken to be about $-5kT_i$. This form has the property that it concentrates the positive ion density in the region where the potential is Φ_0 .

3.
$$n_i(\Phi) = n_0 \frac{\Phi_0^2 + \delta^2}{(\Phi - \Phi_0)^2 + \delta^2},$$

where Φ_0 and δ are arbitrarily chosen constants, typically taken to be about $V_0/5$ and $\Phi_0/10$, respectively. This functional form is similar to that of the previous case, in that it tends to concentrate ion density in regions where $\Phi = \Phi_0$.

The modified Boltzmann and the Lorentzian functional forms (items 2 and 3 above) were con-

structed in order to place the positive ion density where it would be most effective in producing an electrostatic attraction. In all cases we investigated, however, the force was repulsive, and monotonically-decreasing in magnitude for separations between about $0.5\lambda_D$ and $5\lambda_D$. Based on this observation, we speculate that there is no local functional dependence of ρ on Φ which will lead to a net attraction between the two particles.

Except in thermodynamic equilibrium, there is no physical reason for the charge density being a local function of the potential. Indeed, such models ignore an important feature of the physics — the loss of ions at the particle surface due to recombination. Perhaps a fluid model might describe the situation more accurately. In this model, the density is typically not a local function of Φ . Although we have not done so, it would be interesting to investigate the predictions of such models for this system.

Another approach, is to determine the ion density using a Monte Carlo approach. Choi and Kushner [18] report the results of such a calculation, although the assumed conditions were rather different than those common for Coulomb crystal formation. They calculate the interparticle electrostatic force, and find it everywhere repulsive.

5. Conclusion

Recent reports (based on the Debye–Hückel approximation) of an electrostatic attraction between negative particles imbedded in a plasma are incorrect because they neglect the exchange of energy between the positive ions and the external plasma. Correctly done, the interparticle force is repulsive for all particle separations under the Debye–Hückel approximation. A difficult point in any calculation of the interaction between particles in a plasma is the determination of the positive ion density. We have directly calculated the force between two particles under several approximations besides Debye–Hückel relating the positive ion density to the local potential. In no case do we find an attractive force, and we speculate that for any local relationship the force will be repulsive. We have not, however, investigated non-local relationships, such as would be produced by the fluid model.

Acknowledgements

We have benefited from conversations with M. J. Kushner, M. E. Riley, and E. E. Kunhardt. This work was supported by the State of Nebraska.

References

1. H. Thomas, G. E. Morfill, V. Demmel, J. Goree, B. Feuerbacher, and D. Möhlmann *Phys. Rev. Lett.* **73** (1994), p. 652.
2. C. A. Murray and D. H. V. Winkle *Phys. Rev. Lett.* **58** (1987), p. 1200.
3. V. E. Fortov, A. P. Nefedov, V. M. Torchinskii, V. I. Molotkov, A. G. Khrapak, O. F. Petrov, and K. F. Volykhin *Pis'ma Zh. Eksp. Teor. Fiz.* **64** (1996), p. 86.
4. I. Waki, S. Kassner, G. Birkel, and H. Walther *Phys. Rev. Lett.* **68** (1992), p. 2007.
5. G. E. Morfill and H. Thomas *J. Vac. Sci. Technol. A* **14** (1996), p. 490.
6. Y.-P. Chen, H. Luo, M.-F. Ye, and M. Y. Yu *Phys. Plasmas* **6** (1999), p. 699.
7. Y. Hayashi and K. Tachibana *Jpn. J. Appl. Phys.* **33** (1994), p. L804.
8. J. B. Pieper, J. Goree, and R. A. Quinn *J. Vac. Sci. Technol. A* **14** (1996), p. 519.
9. A. Melzer, V. A. Schweigert, and A. Piel *Phys. Rev. Lett.* **83** (1999), p. 3194.
10. U. Konopka, G. E. Morfill, and L. Ratke *Phys. Rev. Lett.* **84** (2000), p. 891.
11. S. V. Vladimirov and M. Nambu *Phys. Rev. E* **52** (1995), p. R2172.
12. V. A. Schweigert, I. V. Schweigert, A. Melzer, A. Homann, and A. Piel *Phys. Rev. E* **54** (1996), p. 4155.
13. O. M. Belotserkovskii, I. E. Zakharov, A. P. Nefedov, V. S. Filinov, V. E. Fortov, and O.A. Sinkevich *Zh. Eksp. Teor. Fiz.* **115** (1999), p. 819.
14. D. P. Resendes, J. T. Mendonça and P. K. Shukla *Phys. Lett. A* **239** (1998), p. 181.
15. G. E. Astrakharchik, A. I. Belousov, and Y. E. Lozovik *Phys. Lett. A* **258** (1999), p. 123.
16. P. F. Williams, A. Belolipetski, A. Goussev, and M. E. Markes, Coulomb crystals and glasses for self-assembly of nanoparticles. In: M. Cahay, S. Bandyopadhyay, D. J. Lockwood, N. Koshida, J. P. Leburton, M. Meyyappan, and T. Sakamoto, Editors, *Advanced Luminescent Materials and Quantum Confinement*, The Electrochemical Society, Pennington, NJ (1999), p. 189.
17. M. E. Riley, R. J. Buss, The interaction of Debye-Shielded particles, Sandia Report SAND99-1017, 1999.
18. S. J. Choi and M. J. Kushner *J. Appl. Phys.* **75** (1994), p. 3351.