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# THE ANISOTROPIC CELL MODEL IN THE COLLOIDAL PLASMAS

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**Abstract**—The anisotropic spherical Wigner-Seitz (WS) cell model — introduced to describe colloidal plasmas — is investigated using the linearized Poisson-Boltzmann (PB) equation. As an approximation, the surface potential of the spherical macroparicle expanded in terms of the monopole ($q$) and the dipole ($p$) is considered as an anisotropic boundary condition of the linear PB equation. Here, the "apparent" moments $q$ and $p$ are the moments 'seen' in the microion cloud, respectively. Based on a new physical concept, the momentneutrality, the potential around the macroparticle can be solvable analytically if the relationship between the actual moment and the "apparent" moment can be obtained according to the momentneutrality condition in addition to the usual electroneutrality. The calculated results of the potential show that there is an attractive region in the vicinity of macroparticle when the corresponding dipole part of the potential dominates over the monopole part, and there is an attractive region and a repulsive region at the same time, i.e., a potential well, when the corresponding dipole part of the potential just comes into play. It provides the possibility and the conditions of the appearance of periodic structure of the colloidal plasmas, although it is a result of a simple theoretical model.

## 1. INTRODUCTION

Colloidal plasmas (CP) are plasma containing a large number of charged solid or liquid particles. The well-known examples of CP are charged colloidal suspensions and dusty plasmas. The macroparticles in the colloidal suspensions include macroions (synthetic and biological) and charged latex particles. The macroparticles in

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the dusty plasmas are solid particles. The common feature of these colloidal plasmas makes it possible to adopt the same model: Charged macroparticles and microion clouds (sheath or double layer) around macroparticles. The Derjaguin-Landau-Verwey-Overbeek theory predicts that an isolated pair of highly charged macroparticles will experience a purely repulsive screened Coulomb interaction. This reasonable prediction was contradicted by the experimental observation [1, 2]. Optical tweezer measurements subsequently demonstrated that anomalous attractions appear only when charged spheres are confined to a plane by charged surfaces [3, 4]. A long-lived controversy was ignited by the suggestion [5] that like-charged macroparticles need not repel each other as predicted by Poisson-Boltzmann (PB) mean field theory [6–13]. In the area of dust plasmas, one can observe transitions from a disordered gaseous-like phase to a liquid-like phase and the formation of ordered structures of particles-plasma crystals [14–17]. The wake effect has been proposed as the most promising candidate for the formation of the dust-plasma crystals [18, 19], and it was experimentally confirmed to be responsible for the attraction of two macroparticles by optical manipulations using radiation pressure from laser light [20, 21]. Nevertheless, it is found that both the point charge and the dipole moment can be responsible for the wake potential [22]. The importance of the dipole interaction has become increasingly recognized. In addition, the nonuniform distribution of surface charge of macroparticles [23–27] has been proposed to be responsible for the attractive interaction between two like–charged macroparticles in colloidal suspensions. In a sense, it also leads to the dipole interaction.

To our knowledge, there are three kinds of physical mechanisms proposed to explain the formation of a dipole: (a) Field-induced dipole which is derived from dielectric polarization produced by the nearby charged object and the external field [28–37]; (b) compound-dipole which is caused by displacement of the centers of macroparticle charge and microion clouds (deformed sheath or double layer), it means that the macroparticle charge and the microion cloud charge form two poles of dipole [38–43]; (c) charging-resulted dipole which is caused by the nonuniform distribution of surface charge of macroparticles [44–48]. Whatever the reason for the formation of the dipole, the dipole or the macroparticle is in anisotropic microion cloud. In these dipole models, most of the studies do not attempt the difficult analytical calculation of the dipole moment. Some treated the dipole moment as a parameter, some gave the order of the dipole moment, and some gave the dipole moment according to the simple approximate expression, $4\pi\varepsilon_e a^3 E_0$. Such a dipole has not been investigated thoroughly.

On the other hand, it is very common to adopt a spherical Wigner-Seitz (WS) cell to investigate the physical properties of colloidal plasmas with a finite density of macroparticles. Given proper boundary conditions, the WS cell reduces the initial many-particle system to the much simpler problem of a single macroparticle [49–55]. It is most noticeable that all the theories mentioned above assumed that the WS cells with spherical macroparticle are isotropic, i.e., the potential distribution is symmetrical ball. Actually, such a model is questionable due to the presence of dielectric polarization of macroparticles, nonuniform distribution of surface charge of macroparticles, microion clouds inhomogeneity as well as a finite microion flows.

Considering that only the numerical methods, such as Finite Element Method, molecular dynamics method and Monte Carlo simulation [56] can be used to solve the nonlinear Poisson-Boltzmann (PB) equation, we will investigate an anisotropic spherical WS cell model using the linearized PB approximation for the analytical solution in this paper. As an approximation, the surface potential of the macroparticle expanded in terms of the monopole ($q$) and the dipole ($p$) is considered as an anisotropic boundary condition of the WS cell. By introducing a new physical concept in our previous study [57], momentneutrality (WS cell possess no moments, including electroneutrality), the relationship between the actual moment ($q_0$ or $p_0$) and the "apparent" moment ($q$ or $p$) can be obtained, and then the potential around a macroparticle is solvable analytically. Different from the previous paper we published, this paper focuses on the case of a finite density of macroparticles, rather than a single macroparticle in an infinite microion cloud. The remainder of this paper will be organized as follows. In Section 2 we introduce the known cell model for a charged macroparticle in an isotropic microion cloud. In Section 3, we report a theoretical study of the cell mode for a dipole in an anisotropic microion cloud according to the momentneutrality ruleand then a charged macroparticle in anisotropic microion cloud. In the following Section 4, the calculated results are discussed. Finally, Section 5 summarizes our results.

## 2. THE CELL MODEL FOR A CHARGED MACROPARTICLE IN AN ISOTROPIC MICROION CLOUD

First, let us consider the cell model for a charged spherical macroparticle, $q_0$, immersed in an isotropic microion cloud. The radius of the macroparticle is $a$ and the cell radius is $b$, as shown in Figure 1. It also means that the distance between two macroparticles is $2b$.

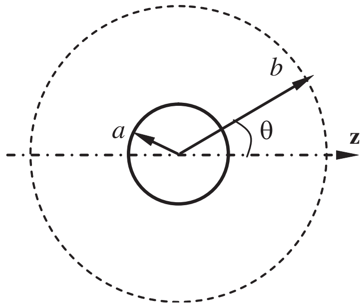


**Figure 1.** One cell of the colloidal plasmas.

The macroparticle, whose hard cores occupy a volume fraction $f$, is treated within the WS cell model. We will restrict ourselves to the case of spherical cell of radius $b = a/f^{1/3}$. If collisions are sufficiently frequent, it may be assumed that the isotropic microion cloud is in thermal equilibrium locally, and the local densities are then given by the Boltzmann distribution, $n_{\pm} = n_0 \exp(\mp e\varphi_q/k_BT)$. Although it is not a good assumption, especially for the case of dusty plasmas because of the plasma absorption on the macroparticle, it has some reasonable components if we think that it is just the most basic model. For convenience, we assume that the density at the cell boundary is $n_{cs}$, i.e., $n_0 = n_{cs}$, it also means that the potential at $r = b$ is zero. If the potential energy of ions due to its nearest neighbor is much smaller than its kinetic energy ($e\varphi \ll k_BT$), the ion density may be linearized with respect to $\varphi_q$, $\rho \approx -2n_{cs}e^2\varphi_q/k_BT = -\varepsilon_0\kappa^2\varphi_q$, where $1/\kappa$ is the screening length (Debye length), with $\kappa^2 = 2n_{cs}e^2/\varepsilon_0 k_BT$, $k_B$ is Boltzmann's constant, $T$ is the absolute temperature. The linear PB equation is $\nabla^2\varphi_q = \kappa^2\varphi_q$.

If $q_0$ is the actual charge on the macroparticle, we assume that $q$ is the charge 'seen' in the microion cloud. It means that the potential on the surface of macroparticle caused by the actual macroparticle charge $q_0$ and the microion cloud charge $q_c$ is $\varphi_q(a) = q/4\pi\varepsilon_e a$. In the following, this charge $q$ will be called the 'apparent charge'. Now the boundary question is

$$\left\{ \begin{array}{l} \nabla^2\varphi_q = \kappa^2\varphi_q \\ \varphi_q\,(a) = q/4\pi\varepsilon_0 a \\ \varphi_q(b) = 0 \end{array} \right. , \tag{1}$$

Solving this equation results in the potential distribution:

$$\varphi_q = A\frac{\exp\left(-\kappa r\right)}{r} + B\frac{\exp\left(\kappa r\right)}{r}. \tag{2}$$

According to the boundary conditions, we can get

$$A = -\frac{q \exp(\kappa b)}{8\pi\varepsilon_0 \sinh(\kappa(a-b))}, \quad B = \frac{q \exp(-\kappa b)}{8\pi\varepsilon_0 \sinh(\kappa(a-b))}. \tag{3}$$

The charge distribution around the macroparticle is $\rho = -\varepsilon_0 \kappa^2 \varphi_q$, and the total microion cloud charge is

$$q_c = \int_{sheath} \rho dV = \int_0^\pi \int_a^b \rho 2\pi r^2 \sin\theta \mathrm{d}\theta \mathrm{d}r = -\frac{q}{2\sinh(\kappa(a-b))} g \tag{4}$$

Here,

$$g = (1-\kappa a)\exp(-\kappa(b-a)) - (1+\kappa a)\exp(\kappa(b-a)) + 2\kappa b. \tag{5}$$

Considering that the colloidal plasma has roughly equal values of macroparticle charge and microion cloud charge, that is the electroneutrality relation

$$q_c + q_0 = 0, \tag{6}$$

Solving this equation results in

$$q = \frac{2\sinh(\kappa(a-b))\, q_0}{g}. \tag{7}$$

Substituting into Eqs. (3) and (2), we can get the known potential distribution which can be obtained from the other method, that is

$$\varphi_q = -\frac{q_0 \exp(-\kappa(r-b))}{4\pi\varepsilon_0 g r} + \frac{q_0 \exp(\kappa(r-b))}{4\pi\varepsilon_0 g r}. \tag{8}$$

It is worth noting that we do not choose the electroneutrality boundary condition $\partial\varphi_q(b)/\partial r = 0$. The reason is that the WS cell maybe anisotropic (see the next part).

## 3. THE CELL MODE FOR A DIPOLE IN AN ANISOTROPIC MICROION CLOUD

Now, let us consider the cell model for a dipole, $p_0$, immersed in an anisotropic microion cloud, and do not consider the mechanism for macroparticle polarization that could arise due to the presence of dielectric polarization of macroparticles, nonuniform distribution of surface charge of macroparticles, inhomogeneity of microion clouds as well as a finite microion flows. Note that the $z$ direction is taken as the direction of the dipole $p_0$. Similarly, we introduced the concept of 'apparent dipole' seen in the microion cloud. It means that the potential on the surface of macroparticle caused by the actual dipole $p_0$ and the microion cloud dipole $p_c$ is the dipole potential caused by the "apparent" dipole $p$. Considering the assumption of the same densities $n_{cs}$ at the cell boundary, we assume that the potential at the cell boundary is $\varphi_p(b) = 0$. The boundary value problem is

$$\begin{cases} \nabla^2 \varphi_p = \kappa^2 \varphi_p \\ \varphi_p(a) = p\cos\theta / 4\pi\varepsilon_0 a^2 \\ \varphi_p(b) = 0 \end{cases} \tag{9}$$

Solving this equation results in the potential distribution:

$$\varphi_p = \left( C\frac{(1+\kappa r)\exp(-\kappa r)}{r^2} + D\frac{(1-\kappa r)\exp(\kappa r)}{r^2} \right)\cos\theta \tag{10}$$

Considering the boundary conditions, we can get

$$\begin{aligned} C &= \frac{p}{4\pi\varepsilon_0}\frac{(1-\kappa b)\exp(\kappa b)}{(1+\kappa a)(1-\kappa b)\exp(\kappa(b-a)) - (1-\kappa a)(1+\kappa b)\exp(-\kappa(b-a))}, \\ D &= -\frac{p}{4\pi\varepsilon_0}\frac{(1+\kappa b)\exp(-\kappa b)}{(1+\kappa a)(1-\kappa b)\exp(\kappa(b-a)) - (1-\kappa a)(1+\kappa b)\exp(-\kappa(b-a))} \end{aligned} \tag{11}$$

The charge distribution around the dipole is $\rho = -\varepsilon_0 \kappa^2 \varphi_p$, and the microion cloud dipole moment is

$$\begin{aligned} p_c &= \int_{sheath} z\rho dV = \int_0^\pi \int_a^b r\cos\theta\rho 2\pi r^2 \sin\theta \mathrm{d}\theta \mathrm{d}r \\ &= -\frac{p}{3[(1+\kappa a)(1-\kappa b)\exp(\kappa(b-a)) - (1-\kappa a)(1+\kappa b)\exp(-\kappa(b-a))]} h \end{aligned} \tag{12}$$

Here,

$$\begin{aligned} h = {} & (1-\kappa b)\left(\kappa^2 a^2 + 3\kappa a + 3\right)\exp(\kappa(b-a)) \\ & - (1+\kappa b)\left(\kappa^2 a^2 - 3\kappa a + 3\right)\exp(-\kappa(b-a)) + 2\kappa^3 b^3 \end{aligned} \tag{13}$$

We assume that one cell of the colloidal plasma has roughly equal moments of macroparticle dipoles and microion cloud dipoles. It means that the macroparticle and microion cloud system possess no dipole moment, that is the 'momentneutrality' relation

$$p_c + p_0 = 0 \tag{14}$$

Now, we can get the relation between the actual dipole moment and the "apparent" dipole moment

$$p = \frac{3[(1+\kappa a)(1-\kappa b)\exp(\kappa(b-a)) - (1-\kappa a)(1+\kappa b)\exp(-\kappa(b-a))]}{h} p_0 \tag{15}$$

Substituting into Eqs. (10) and (11), we can get

$$\varphi_p = \left(\frac{3p_0\left(1-\kappa b\right)\left(1+\kappa r\right)\exp\left(-\kappa\left(r-b\right)\right)}{4\pi\varepsilon_0 hr^2} - \frac{3p_0\left(1+\kappa b\right)\left(1-\kappa r\right)\exp\left(\kappa\left(r-b\right)\right)}{4\pi\varepsilon_0 hr^2}\right)\cos\theta \tag{16}$$

Here, we give some explanation of the means and the validity for the momentneutrality. First, the condition of the electroneutrality arises from the charging process and results in no electrical characteristic of the plasmas as a single entity or a cell. Based on the similar thoughts, we assume that the momentneutrality is the intrinsic characteristic of colloidal plasmas in addition to the electroneutrality. Second, we can also get the analytical solution of the linear PB equation without the momentneutrality, but the apparent moment $p$ is an unknown quantity. It is the relation between the actual dipole moment $p_0$ and the apparent moment $p$ that results in a closed- form analytical model. The ratios of the apparent dipole moment to the actual dipole moment as a function of $\kappa a$ are shown in Figure 2. As can be seen from the data, we can find that the ratio increases with

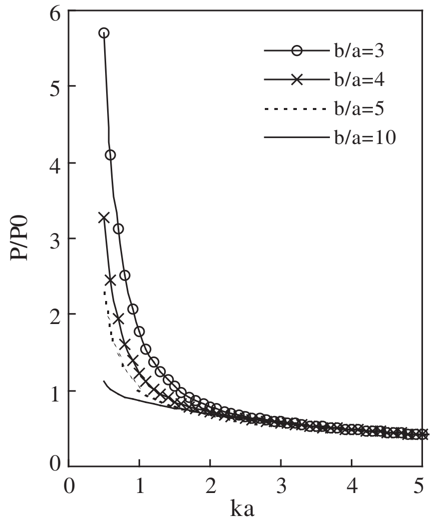


**Figure 2.** The ratio of the apparent dipole moment to the actual dipole moment as a function of $\kappa a$.

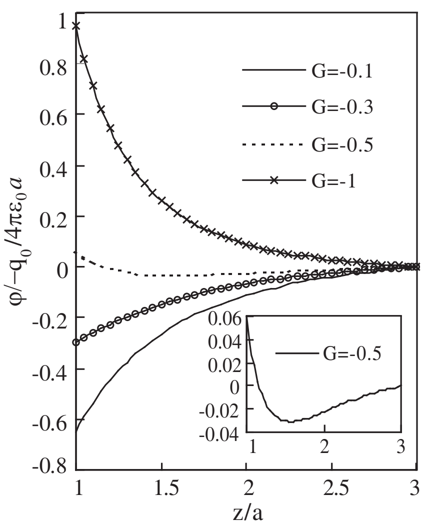


**Figure 3.** The normalized potential along the $z$ axis, if $\kappa a = 1$, $b/a = 3$ and $\theta = 0°$. In the inset, the same results for $G = -0.5$ are shown at small scales.

decrease in $\kappa a$. On the other hand, there are different methods to calculate the concrete values of the actual moment $p_0$ for different polarization mechanism. For the case of dielectric polarization, we can assume $p_0 = 4\pi\varepsilon_e a^3 \beta E_0$, here $\beta = (\varepsilon_i - \varepsilon_e)/(\varepsilon_i + 2\varepsilon_e)$, $\varepsilon_i$ is the permittivity of the macroparticle and $\varepsilon_e$ is the permittivity of the environment media [57]. In a word, the momentneutrality condition is needed although it deserves some further consideration.

Now we study the case of a charged macroparticle in anisotropic microion cloud, and do not consider the polarization mechanism. We assume that the actual charge is $q_0$, thus the "apparent charge" is $q$. On the other hand, we assume that the actual moment of the dipole is $p_0$, thus the "apparent" dipole is $p$. As an approximation, the surface potential of the spherical macroparticle expanded in terms of the monopole ($q$) and the dipole ($p$) is considered as an anisotropic boundary condition of the linear PB equation. The boundary value problem is

$$\begin{cases} \nabla^2\varphi = \kappa^2\varphi \\ \varphi|_{r=a} = \frac{q}{4\pi\varepsilon_0 a} + \frac{p}{4\pi\varepsilon_0 a^2}\cos\theta \\ \varphi|_{r=b} = 0 \end{cases} . \tag{17}$$

The solution is given by

$$\varphi = \varphi_q + \varphi_p \tag{18}$$

Here, $\varphi_q$ and $\varphi_p$ can be obtained according to Eqs. (8) and (16), respectively.

## 4. RESULTS AND DISCUSSION

If we assume $r' = r/a$, $b' = b/a$ and $G = p_0/q_0 a$, we can get the normalized potential, i.e., the ratio of $\varphi$ to $-q_0/4\pi\varepsilon_e a$ ($q_0 < 0$):

$$\varphi_q' = \frac{\varphi_q}{-q_0/4\pi\varepsilon_0 a} = \frac{\exp\left(-\kappa a\left(r'-b'\right)\right)}{gr'} - \frac{\exp\left(\kappa a\left(r'-b'\right)\right)}{gr'} \tag{19}$$

$$\begin{aligned} \varphi_p' &= \frac{\varphi_p}{-q_0/4\pi\varepsilon_0 a} \\ &= \left(-\frac{3G\left(1-\kappa ab'\right)\left(1+\kappa ar'\right)\exp\left(-\kappa a\left(r'-b'\right)\right)}{hr'^2}\right. \\ &\quad \left.+\frac{3G\left(1+\kappa ab'\right)\left(1-\kappa ar'\right)\exp\left(\kappa a\left(r'-b'\right)\right)}{hr'^2}\right)\cos\theta \end{aligned} \tag{20}$$

$$\varphi' = \varphi_q' + \varphi_p' \tag{21}$$

The calculated results of the potential distribution along the $z$ axis are shown in Figures 3–7. As can be seen from the data, we can get some results as follows:

(1) With an increase in $z/a$, the potential increase gradually when $|G| < 0.5$ and decreases gradually when $|G| > 0.5$ if $\kappa a = 1$, $b/a = 3$ and $\theta = 0°$, as shown in Figure 3. It means that the field begins to have a reversed direction when $|G| > 0.5$, i.e., there is an attractive region when the corresponding dipole part of the potential dominates over the monopole part. In the inset, the same results for $G = -0.5$ are shown at small scales. The most important thing to note here is that there is a potential well behind the particle. It means that there is an attractive region and a repulsive region at the same time when $G = -0.5$. It also implies that microion sign reversal occurs. However, the potential well will disappear when $|G| > 0.5$, i.e., the dipole part of the potential actually dominates over the monopole part. On the direction reversal along the $z$ axis ($\theta = 180°$), however, this kind of situation will not appear, i.e., there is no attractive region, as shown in Figure 4.

(2) With a decrease in $b/a$, the field strength increases gradually and there is no attractive region when $\kappa a = 1$ and $\theta = 0°$ if $G = -0.1$, as shown in Figure 5. But there is an attractive region and a potential

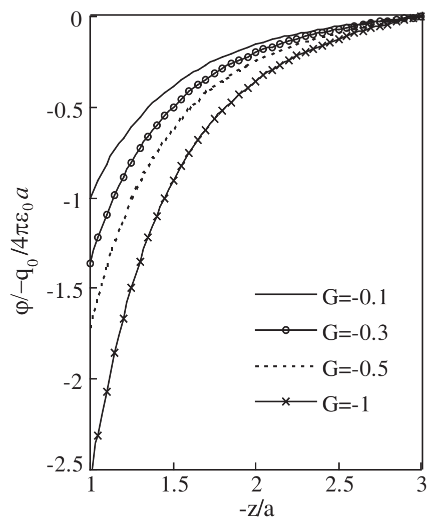


**Figure 4.** The normalized potential along the $z$ axis, if $\kappa a = 1$, $b/a = 3$ and $\theta = 180°$.

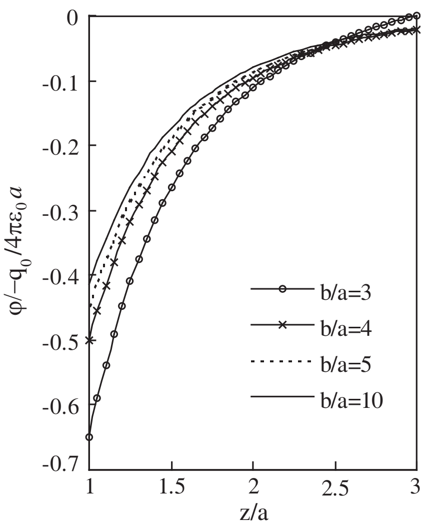


**Figure 5.** The normalized potential along the $z$ axis, if $\kappa a = 1$, $G = -0.1$ and $\theta = 0°$.

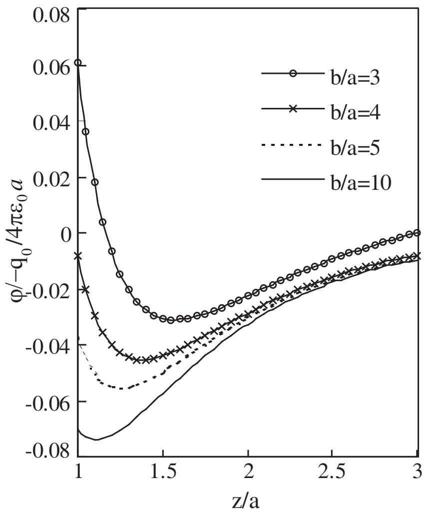


**Figure 6.** The normalized potential along the $z$ axis, if $\kappa a = 1$, $G = -0.5$ and $\theta = 0°$.

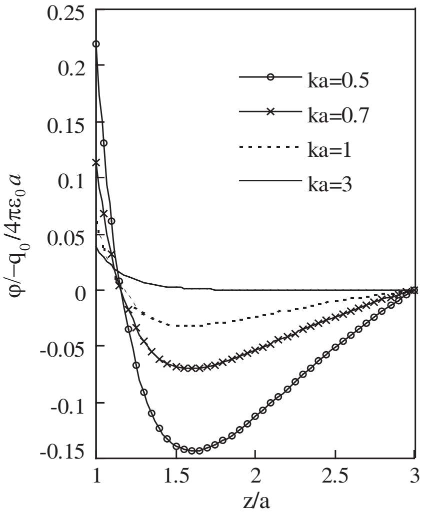


**Figure 7.** The normalized potential along the $z$ axis, if $b/a = 3$, $G = -0.5$ and $\theta = 0°$.

well if $G = -0.5$, as shown in Figure 6. With an increase in $b/a$, the attractive region will disappear gradually. It also means that the well occurs only for very small distances away from the macroparticle surface when b/a tends infinity [57].

(3) With an increase in $\kappa a$, the potential well will disappear gradually when $b/a = 3$, $G = -0.5$ and $\theta = 0°$, as shown in figure (7). There is an attractive region but no potential well when $\kappa a > 1$ and an attractive region and a potential well when $\kappa a \leq 1$.

Using the molecular dynamics method, Messina, Holm and Kremer reported a mechanism that can lead to long-rang attraction between like-charged spherical macroions, stemming from the existence of metastable ionized states. They demonstrated that, in the region of strong Coulomb coupling, the counterion clouds are very likely to be unevenly distributed [10]. Carbajal-Tinoco and Gonzale-Mozuelos applied the Ornsein-Zernike equation to determine the microstructure of a colloidal suspension at finite concentrations. It also shown that the charge inversion generated by the surrounding microion cloud induces the fairly long-ranged attraction [12]. Actually, Sogami and Ise also thought that Coulombic intermacroion attraction is through the intermediary of counterions [5]. Velegol and Thwar have developed an analytical model for randomly charged particle surfaces [23]. The resulting spherically systemetric potential depends on the values of the average surface potential ($\zeta$-potential) and on the standard deviation of the surface potential among different regions on the particle surface. Based on the model proposed by Velegol and Thwar, Cametti et al. investigate the aggregation kinetics of polyion-induced colloidal complexes through Monte Carol simulation [27]. The results reveal that the final size of the aggregates grows on increasing the standard deviation and on decreasing the $\zeta$-potential because of the attractive interaction. Although the microion clouds inhomogeneity is neglected in this model, the result confirms that the higher the standard deviation of the surface potential, the higher the degree of non-uniform of the surface potential and then the stronger was the attractive force. It is worth noting that the non-uniformity in our model would be higher when the corresponding dipole part of the potential dominates over the monopole part. In the dusty plasmas, the wake effect was experimentally confirmed to be responsible for the attraction of two macroparticles by optical manipulations using radiation pressure from laser light [20, 21]. In a sense, the wake means that the microion cloud is unevenly distributed even if it is derived from the dynamic effects. Takahashi et al. found that the upper particles could cause an attractive force on the lower ones ($\theta = 0°$) and the lower ones could not cause that on the upper ones ($\theta = 180°$). Hence, the interaction between the macroparticles is clearly nonreciprocal or asymmetric [20]. The results of our study have provided support to these finds in different areas. The well whether it results in the appearance of periodic structure of the colloidal plasmas is worthy of further study.

## 5. CONCLUSION

Because of the presence of dielectric polarization of macroparticles, nonuniform distribution of surface charge of macroparticles, inhomogeneity of microion clouds as well as a finite microion flows, the potential distribution around the spherical macroparticle should be anisotropic. The potential of the particle surface can be expanded in terms of a multipolelike expansion. If just neglecting higher-order terms, i.e., the particle replaced by a charge $q_0$ and a dipole $p_0$, and not modifying the moment of the monopole and the dipole, the approximation method is not sufficient for describing the actual potential distribution. On the other hand, given proper boundary conditions, the WS cell reduces the initial many-particle system to the much simpler problem of a single particle. By introducing the concepts of the "apparent" charge $q$ and "apparent" dipole $p$, we modify the moments according to the momentneutrality condition in addition to the usual electroneutrality in one cell, which are assumed as the intrinsic characteristics of colloidal plasma. Based on the rules, the relationship between the actual moment and the "apparent" moment can be acquired, and the potential around the macroparticle can be calculated analytically.

The calculated results of the potential show that there is an attractive region when the corresponding dipole part of the potential comes into play and there is a potential well behind the particle when $|p_0/q_0a| \approx 0.5$, $b/a < 10$ and $\kappa a \leq 1$. It provides the possibility and the conditions of the appearance of periodic structure of the colloidal plasmas, although it is a result of a simple theoretical model.

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