Charge inversion of colloids in an exactly solvable model

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PACS. 82.70.Dd – Colloids.
PACS. 61.20.Gy – Theory and models of liquid structure.
PACS. 02.30.Ik – Integrable systems.

Abstract. – We study a two-dimensional model for a long cylindrical stiff charged macroion immersed in a charge-asymmetric electrolyte with charge ratio $+2/-1$. The model is integrable and it allows an exact analytical determination of the effective charge of the macroion, which characterizes the electrostatic potential at large distances (compared to the screening length) from the macroion. At high coulombic coupling, this model predicts charge inversion: for a highly negatively charged macroion, the effective charge could become positive, indicating an overscreening of the macroion by the divalent counterions. By studying the behavior of the coions and counterions density profiles close to the macroion, we show that the counterion condensation threshold is shifted to a lower value in absolute value. This plays an important role in the charge inversion phenomenon.

The determination of the effective interactions between charged macroions immersed in electrolyte solutions is a central topic in colloidal science [1]. Based on the linear Debye–Hückel (DH) theory, for a very long and thin (zero radius) cylindrical macroion, with linear charge density $e/ℓ$, the reduced effective potential at a distance $r$ is $y(r) = 2λK_0(κr)$, where $λ = l_B/ℓ$, $K_0$ is the modified Bessel function of order 0, $y = eψ/(k_B T)$ with $ψ$ the electric potential, $e$ the elementary charge, $T$ the temperature and $k_B$ is Boltzmann constant. Here, $λ$ is the reduced linear charge density, expressed in units of the inverse of the Bjerrum length $l_B = e^2/(k_B T ε)$, where $ε$ is the electric permittivity of the solvent. The Debye screening length is $κ^{-1} = (4πl_B ∑ α n_a^b v^2_α)^{-1/2}$, where $n_a^b$ is the bulk density of the microions of the electrolyte of type $α$ and $v_α$ their valence. For highly charged macroions, the linear approach is inappropriate. A first improvement over the linear theory is to use the mean field nonlinear Poisson–Boltzmann (PB) equation. Under this approximation, the effective potential, at large distances, is again a screened potential: $y(r) ∼ 2λ_{eff} K_0(κr)$ for $κr ≫ 1$, but the prefactor $λ_{eff}$ is not anymore the bare charge $λ$ of the colloid, but it is known as the effective or renormalized charge [2,3].

PB approach is adequate [4,5] if the coulombic couplings between microions are small, for example for a two-component electrolyte: $v^2_1 Τ ≪ 1, v^2_2 Τ ≪ 1, |v_1 v_2| Τ ≪ 1$ where $Γ = 2l_B/a$, with $a$ the average distance between microions in the electrolyte. For large coupling, new phenomena can occur that cannot be explained by the mean field PB approach. The most striking one is the phenomenon of charge inversion: a large fraction of counterions can condense into the macroion and the resulting dressed macroion can have a charge of opposite sign than
its bare charge. Closely related to this phenomenon is the possibility of attraction between two like-charged macroions. Several experimental observations and studies of these phenomena have been reported [6–8] and some theoretical explanations have been put forward [9–14], for a review see [15].

In this work, we report exact results for the renormalized charge of a cylindrical macroion, valid beyond the mean field approximation, in a whole range where the coulombic coupling can be large: $0 \leq \Gamma < 2/3$. The model under consideration is an infinitely long charged cylindrical colloid, with radius zero, immersed in a charge-asymmetric electrolyte solution composed of microions with valences $v_1 = 2$ and $v_2 = -1$. Due to the translational symmetry along the direction of the colloid, we consider a two-dimensional (2D) model: we will be interested only in the microions density profiles and the electric potential along the radial direction from the colloid. Our model predicts the possibility of charge inversion: the effective charge and bare charge of the colloid have opposite signs when $\lambda < \Gamma - 1 < 0$ and $\Gamma > 1/2$.

Being two-dimensional, our model describes correctly a cylindrical macroion immersed in an electrolyte formed by cylindrical parallel coions and counterions, with linear charge densities $v_1, v_2 e/a$. As above, the coulombic coupling is $\Gamma = 2l_B/a$. Many like-rod colloids and polyelectrolytes can be described by the 2D model presented here. For example, synthetic polyelectrolytes such as poly(p-phenylene) and poly(styrenesulfonate) backbones used in water management have tunable linear charge density and a very large persistent length (20 nm) compared to other length scales involved in the problem. Of particular interest for biological and biophysics process, DNA (single and double stranded) and actin filaments are anionic cylindrical polymers that have also a large persistent length, and as such could be, in a first approach, described by the present model. With these cylindrical polyelectrolytes the situation described by this model could be reproduced experimentally. Experimental situations similar (although not exactly equal) to the one presented here are described in [7,16]. The experiments described in [7] and the possibility of charge inversion in those situations have important applications in gene therapy. Besides the possible applications to polyelectrolytes, it is well-known [17] that the 2D Coulomb gas is also a prototype model that can describe the physics of many interesting systems such as vortices in a superfluid, the XY model, and dislocations in a 2D crystal.

For a cylindrical macroion with point-like coions and counterions, one important ingredient in the theoretical explanations [10] for charge inversion is the formation of a 2D liquid of counterions in the vicinity of the surface of the macroion and the strong longitudinal correlations between them. Our model, being two-dimensional, does not take into account these longitudinal correlations, but nevertheless shows that charge inversion is possible, thus showing that the radial correlations are enough to drive the charge inversion phenomenon.

Our 2D model with zero radius charges is stable against the collapse of pairs of opposite charges provided that $0 < \Gamma < 1$. For technical reasons explained below our results are valid for $0 < \Gamma < 2/3$. To ensure the stability against collapse of counterions into the macroion, we require $-1/2 < \lambda < 1$. The study of the effective charge beyond those limits requires to consider a macroion with nonzero radius and it is beyond the scope of this Letter, see however [18,19]. Provided $-1/2 < \lambda < 1$, the introduction of a small hard core radius $R$ ($\kappa R \ll 1$) for the macroion is an irrelevant perturbation. These limits for $\lambda$ correspond to the Manning thresholds for counterion condensation [20] derived within a mean field approach. In all the theoretical approaches to charge inversion [9–11] the counterion condensation phenomenon is crucial. This is not an exception here. We will show that, due to the strong correlations between microions, the threshold for counterion condensation, in the case $\lambda < 0$, is changed to $(\Gamma - 1)/2$: it take place before than predicted by the traditional Manning theory.

Exact results for our model can be obtained due to the recent advances in the the-
ory of two-dimensional Coulomb systems: the exact bulk thermodynamics of the charge-symmetric [21] and the (+2/−1) charge-asymmetric [22] two-dimensional two-component plasma are known. This has been possible due to their relationship with the integrable sine-Gordon (sG) and complex Bullough-Dodd (cBD) models. Using the field theoretical tools from the sG model, exact results for the effective charge and other quantities for a cylindrical macroion in a charge-symmetric electrolyte have been obtained [19]. Here, we use the known expressions for the form-factors of exponential fields of the cBD model to obtain results for the charge-asymmetric case.

First, let us consider the electrolyte in the absence of any macroion. Carrying out a Hubbard-Stratonovich transformation, the grand canonical partition function of the electrolyte can be cast as the partition function of the cBD model, with action [22]

$$S = \int \left[ \frac{1}{16\pi} \left| \nabla \phi(\mathbf{r}) \right|^2 - z_1 e^{ib\phi(\mathbf{r})} - z_2 e^{-ib\phi(\mathbf{r})/2} \right] \, d\mathbf{r}$$

where $z_{1,2}$ are the fugacities of the microions and $b = \sqrt{\Gamma}$. To give a precise meaning to the fugacities in the cBD model the conformal normalization should be used: $\langle \phi(0) \phi(\mathbf{r}) \rangle_{z_1=z_2=0} = -\ln r$. In the following we shall denote $\langle \cdots \rangle$ an average with the cBD action (1). We now consider that a single macroion with charge $\lambda$ is immersed in the electrolyte at the origin. Let $Q = \lambda/\Gamma$. The density of positive and negative microions at a position $\mathbf{r}$ from the macroion are [19, 23, 24]

$$n_\pm(\mathbf{r}) = n^b_\pm \frac{\langle e^{ibQ\phi(0)} e^{ibqz\phi(\mathbf{r})} \rangle}{\langle e^{ibQ\phi(0)} \rangle}$$

with $q_+ = 1$ and $q_- = -1/2$ respectively and $n^b_\pm$ are the bulk densities, far from the colloid. The above correlation function can be expressed as a sum over all intermediate $N$-particle states of the cBD model as [22, 25]

$$\langle e^{ibQ\phi(0)} e^{ibqz\phi(\mathbf{r})} \rangle = \langle e^{ibQ\phi(0)} \rangle \langle e^{ibqz\phi(\mathbf{r})} \rangle + \sum_{N=1}^\infty \sum_{\epsilon_1, \ldots, \epsilon_N} \int_{\mathbb{R}^N} \frac{d\theta_1}{(2\pi)^N} F_Q(\theta_1, \epsilon_1; \ldots; \theta_N, \epsilon_N) F_{qz}(\theta_1, \epsilon_1; \ldots; \theta_N, \epsilon_N) e^{-r \sum_{i=1}^N m_{\epsilon_i} \cosh \theta_i}$$

where the $\epsilon_i$ labels the particle spectrum of the cBD model. The rapidity of the $i$-th particle, which is of the type $\epsilon_i$, is $\theta_i$ and has mass $m_{\epsilon_i}$. $F_Q$ and $F_{qz}$ are the form-factors of exponential fields in an $N$-particle state with particle spectrum $\epsilon_1, \ldots, \epsilon_N$. The particle spectrum of the cBD is studied in [26]. This expression is appropriate to find the large distance expansion of the correlation functions. The dominant term is obtained by considering the lightest particle in the spectrum of the cBD model. For $\Gamma$ small enough (see below) the lightest particle is the 1-breather. Its mass is known [27] and it can be expressed in terms of the Debye length as [22]

$$m = \kappa \left[ \frac{2\sqrt{3}}{\pi \xi} \sin \left( \frac{\pi \xi}{3} \right) \sin \left( \frac{\pi (1 + \xi)}{3} \right) \right]^{1/2}$$

where $\xi = \Gamma/(2 - \Gamma)$. The corresponding form-factor is [28, 29]

$$\frac{F_Q}{\langle e^{ibQ\phi} \rangle} = 4\rho \sin \left( \frac{2\pi Q\xi}{3} \right) \cos \left( \frac{\pi}{6} (1 + 2\xi - 4\xi Q) \right)$$

with

$$\rho = i \left[ \frac{\sin(\pi/3)}{\sin(2\pi\xi/3) \sin(2\pi(1 + \xi)/3)} \right]^{1/2} \exp(Ib/2)$$
and
\[ I_b = -4 \int_0^\infty \frac{\cosh\left(\frac{t}{2}\right) \sinh\left(\frac{3t}{2}\right) \sinh\left(\frac{1+\xi t}{2}\right)}{\sinh t \cosh(t/2)} \, dt. \] (7)

Notice that the form-factor diverges for \( \Gamma = 2/3 \). This indicates a change of behavior in the expansion (3): some subdominant terms (for \( \Gamma < 2/3 \)) become of the same order as the contribution of the 1-breather at \( \Gamma = 2/3 \) (see [24] for a similar situation in the short-distance expansion of correlation functions of the sG model). In the following, we restrict our analysis to \( \Gamma < 2/3 \).

Let us define the effective interaction energy (in units of \( k_B T \)) \( E_{\lambda,\Gamma q_\pm}(r) \) of a microion with charge \( 2q_\pm \) (+2, -1) respectively of the electrolyte with the macroion by the relation \( n_{\pm}(r) = n_0 \exp[-E_{\lambda,\Gamma q_\pm}(r)] \). At large distances, \( r \to \infty \), \( n_{\pm}(r) \approx n_0 \exp[(-E_{\lambda,\Gamma q_\pm}(r))/k_B T] \). Using the expansion (3) with only the dominant contribution from the 1-breather form-factor, we obtain, for \( mr > 1 \),
\[ E_{\lambda,+\Gamma}(r) = \frac{8\sqrt{3}}{\pi} \cos\left(\frac{\pi}{3}(1 - 2\xi)\right) e^{\xi/3} \sin\left(\frac{2\pi \lambda}{3(2 - \Gamma)}\right) \cos\left(\frac{2\pi \lambda}{3(2 - \Gamma)} - \frac{\pi}{6} - \frac{\pi \xi}{3}\right) K_0(mr). \] (8)
\[ E_{\lambda,-r/2}(r) = -\frac{8\sqrt{3}}{\pi} \cos\left(\frac{\pi}{3}(1 + 4\xi)\right) \sin\left(\frac{\pi}{3}\right) e^{\xi/3} \sin\left(\frac{2\pi \lambda}{3(2 - \Gamma)}\right) \cos\left(\frac{2\pi \lambda}{3(2 - \Gamma)} - \frac{\pi}{6} - \frac{\pi \xi}{3}\right) K_0(mr). \] (9)

Replacing these expressions into the density profiles and integrating Poisson equation, we find the effective electrostatic potential \( y(r) \) created at a distance \( r \gg m^{-1} \) from the macroion. At large distances, the electrostatic potential takes a similar form to the one predicted by the linear DH theory \( y(r) \sim 2\lambda_{\text{eff}} K_0(mr) \), supporting the hypothesis of the existence of an effective or renormalized charge \( \lambda_{\text{eff}} \). Notice however that there is also a “renormalization” of the screening length: it is \( m^{-1} \) given in (4), instead of the usual Debye length \( \kappa^{-1} \). For \( 0 < \Gamma < 2/3 \), the screening length \( m^{-1} \) is smaller than the Debye length \( \kappa^{-1} \). The effective charge reads
\[ \lambda_{\text{eff}} = \frac{\xi \sqrt{3} \sin(\pi \xi) e^{\xi/3} \sin\left(\frac{2\pi \lambda}{3(2 - \Gamma)}\right) \cos\left(\frac{2\pi \lambda}{3(2 - \Gamma)} - \frac{\pi}{6} - \frac{\pi \xi}{3}\right)}{3 \sin\left(\frac{2\pi \lambda}{3}(1 + \xi)\right) \sin\left(\frac{2\pi \lambda}{3}(1 + \xi)\right) \sin\left(\frac{\pi \xi}{3}\right)}. \] (10)

In the low coupling limit \( \Gamma \ll 1 \) we recover the results from PB theory [18, 30]: \( \lambda_{\text{eff}} = \sqrt{3}[2\sin(\frac{2\pi \lambda}{3} - \frac{\pi}{6}) + 1]/(2\pi) \) as expected.

Figure 1 shows the effective charge as a function of the bare charge. An important result is that the phenomenon of charge inversion is possible: when \( \Gamma > 1/2 \) the effective charge and the bare charge have opposite signs when \(-1/2 < \lambda < \Gamma - 1 < 0 \). The charge inversion occurs for a highly charged negative macroion which is overscreened by positive microions of valence +2 resulting in a positive effective charge. On the other hand, for a positive macroion, screened by microions of valence -1 the charge inversion does not occurs.

The mean field PB formalism does not predict the charge inversion phenomenon, thus the microions correlations are fundamental for this phenomenon to take place. The charge inversion has previously been predicted for a planar charged interface [12] and for spherical colloids [13] with divalent counterions and monovalent coions within the hypernetted chain approximation (HNC). Our model, which is exact (we have solved exactly, without any approximations, the statistical mechanics of the model), put on more firm ground the predictions of [12, 13] and thus suggests that the HNC approximation captures adequately the microions correlations responsible of the charge inversion.
The charge inversion can be actually very large: for example, for $\Gamma = 0.65$ and $\lambda = -0.48$ we have $\lambda_{\text{eff}} = 1.22$, thus a charge inversion ratio of more than 200%. This is similar to the “giant” charge inversion studied in [31], although here the effective charge characterizes the long distance signature of the potential, contrary to its short-distance behavior as considered in [31]. The charge asymmetry of the electrolyte seems to be fundamental for the charge inversion to take place. Exact results for a symmetric electrolyte show no charge inversion [19, 23].

The charge inversion is accompanied by a change of behavior in the large distance behavior of the effective interaction between the macroion and the microions. For $\lambda > \Gamma - 1$, at large distances, the effective interaction between the macroion and a counterion is attractive, whereas its interaction with a coion is repulsive. However, as it can be seen from (8-9), when $\Gamma > 1/2$ and $-1/2 < \lambda < \Gamma - 1 < 0$ the interaction of the macroion with a counterion is now repulsive and its interaction with a coion is attractive, provided $mr \gg 1$.

A similar change of behavior also occurs at short distances, but only for the interaction with a coion. We can obtain the short-distance behavior of the density profiles by using the operator product expansion (OPE) in the correlation function appearing in (2). The OPE for the cBD model has been developed in [32]. Using the OPE (see also [24, 33]) we find, for $-1/2 < \lambda < 0$ that $n_+(r) \propto r^{4\lambda}$ when $r \to 0$, yielding an attractive effective interaction $E_{\lambda,+\Gamma}(r) \sim -4\lambda \ln r$ of the macroion with a counterion (charge +2) at short distances, which is the expected behavior. On the other hand the density profile of the coions (charge −1) at short distances exhibits a change of behavior:

$$n_-(r) \propto \begin{cases} r^{-2\lambda} & \text{for } \Gamma - 1 < \lambda < 0 \\ r^{2(\lambda-\Gamma+1)} & \text{for } -\frac{1}{2} < \lambda < \frac{\Gamma-1}{2} < 0 \end{cases}$$

(11)
giving an effective potential at short distances, $mr \ll 1$,

$$E_{\lambda,-\Gamma/2}(r) \sim \begin{cases} 2\lambda \ln r & \text{for } \Gamma - 1 < \lambda < 0 \\ 2(\Gamma - 1 - \lambda) \ln r & \text{for } -\frac{1}{2} < \lambda < \frac{\Gamma-1}{2} < 0 \end{cases}$$

(12)
The change of behavior at $\lambda = (\Gamma - 1)/2$ can be understood as a first step in the counterion condensation [23, 24] for large coupling $\Gamma$. A fraction of the counterions are condensed into the
The strong coupling between the microions, which is part of the mechanism of charge inversion, macroions provided their linear charge density \( \lambda \)'s are solvable and predicts charge inversion at high coupling \( \Gamma > \lambda \). In PB theory, the effective interaction between a coion and the macroion behave at short-distances as the bare Coulomb potential \( 2\lambda \ln r \) down to \( \lambda = -1/2 \) \[34\].

Interestingly, as \( \lambda \) decreases beyond \( \lambda < (\Gamma - 1)/2 \) the effective interaction, at short distances, of the macroion with the coions becomes less and less repulsive. Paradoxically, it even becomes attractive when \( \lambda < \Gamma - 1 \), provided \( \Gamma > 1/2 \). The divalent counterions are strongly attracted to the macroion and since they attract the monovalent coions, there can be a net attraction of the coions to the macroion. This is accompanied, as we have seen before, with the charge inversion at large distances. The very different behavior, at high coupling, of the coion and counterion density profiles at short distances shows that it is difficult to define a "dressed" charge of the macroion which characterizes the potential at short distances. Indeed, when \( \lambda < (\Gamma - 1)/2 < 0 \), the coions see a dressed charge \( \Gamma - 1 - \lambda \) which can be even positive, whereas the counterions still see a charge \( \lambda < 0 \).

Returning to the charge inversion phenomenon, we should point out that it does not necessarily implies attraction between like-charge macroions. In particular for two identical macroions with charge \( \lambda < \Gamma - 1 < 0 \), as both macroions have an inverted effective charge, the net effect would still be a repulsion between the macroions. To study the possibility of like-charge attraction, let us consider two macroions with linear charge densities \( \lambda_1 \) and \( \lambda_2 \) immersed in the electrolyte at the origin and at \( r \) respectively. In the language of the cBD model, the effective interaction between the two macroions is given by \[19, 23, 24\]

\[
\exp [-E_{\lambda_1, \lambda_2}(r)] = \frac{\langle e^{ibQ_1\phi(0)}e^{ibQ_2\phi(r)} \rangle}{\langle e^{ibQ_2\phi(0)}e^{ibQ_2\phi(0)} \rangle} \tag{13}
\]

with \( Q_{1,2} = \lambda_{1,2}/\Gamma \). Using the form-factor theory explained above we find, for \( mr \gg 1 \), \[22\]

\[
E_{\lambda_1, \lambda_2}(r) \sim \frac{8\sqrt{3} e^{ib}\hat{\lambda}_1\hat{\lambda}_2 K_0(mr)}{\pi \sin \left( \frac{2\pi \xi}{3} \right) \sin \left( \frac{2\pi (1+\xi)}{3} \right)} \tag{14}
\]

with

\[
\hat{\lambda}_{1,2} = \sin \left( \frac{2\pi \lambda_{1,2}}{3(2 - \Gamma)} \right) \cos \left( \frac{2\pi \lambda_{1,2}}{3(2 - \Gamma)} - \frac{\pi}{6} - \frac{\pi \xi}{3} \right). \tag{15}
\]

The "charges" \( \hat{\lambda}_{1,2} \) exhibit the same changes of sign as the effective charge \( \langle 0 | \phi | 0 \rangle \) shown in Fig. 1. Thus, two negatively charged macroions can have an attractive effective interaction at large distances provided that \( \lambda_1 < \Gamma - 1 < 0 \) and \( \Gamma - 1 < \lambda_2 < 0 \) for \( \Gamma > 1/2 \). Also a positive macroion (say \( \lambda_2 > 0 \)) can have a repulsive effective interaction with a negative macroion (charge \( \lambda_1 \)) provided \( \lambda_1 < \Gamma - 1 < 0 \).

Summarizing, we have considered a model that describes the effective charge of cylindrical macroions immersed in a charge-asymmetric +2/-1 electrolyte solution. This model is exactly solvable and predicts charge inversion at high coupling \( \Gamma > 1/2 \) for negatively charged macroions provided their linear charge density \( \lambda < \Gamma - 1 < 0 \). Also, we have shown that a shift in the counterion condensation threshold from \( \lambda = -1/2 \) to \( \lambda = (\Gamma - 1)/2 \) occurs due to the strong coupling between the microions, which is part of the mechanism of charge inversion.
Financial support from ECOS-Nord/COLCIENCIAS, COLCIENCIAS (1204-05-13625) and Comité de Investigaciones, Facultad de Ciencias, Universidad de los Andes is acknowledged. The author thanks L. Šamaj for interesting discussions on the model, and M. Camargo for a useful remark.

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