Like-charge attraction at the nanoscale: 
ground-state correlations and water destructuring

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(Dated: March 22, 2022)

Like-charge attraction, driven by ionic correlations, challenges our understanding of electrostatics 
both in soft and hard matter. For two charged planar surfaces confining counterions and water, we 
prove that even at relatively low correlation strength, the relevant physics is the ground-state one, 
oblivious of fluctuations. Based on this, we derive a simple and accurate interaction pressure, that 
fulfills known exact requirements and can be used as an effective potential. We test this equation 
against implicit-solvent Monte Carlo simulations and against explicit-solvent simulations of cement 
and several types of clays. We argue that water destructuring under nanometric confinement dras-
tically reduces dielectric screening, enhancing ionic correlations. Our equation of state at reduced 
permittivity therefore explains the exotic attractive regime reported for these materials, even in 
absence of multivalent counterions.

INTRODUCTION

When two identical charged colloids are immersed in 
a solvent, their electrostatic interaction is mediated by 
fluctuating smaller species, such as microions [1–6]. Pi-
oneered by Gouy [7] and Chapman [8], the statistical 
treatment of this phenomenon, accounting for thermal 
fluctuations, is a cornerstone of colloid science and goes 
by the name of Poisson-Boltzmann theory [3, 9]. Within 
such theory, macromolecules with a bear charge of the 
same sign invariably experience a repulsive force, which 
provides the Coulombic contribution to the DLVO theory 
[3, 6]. In this framework, electrostatic interactions be-
tween similar bodies, of arbitrary geometry, are necessar-
ily repulsive [10]. However, as initially shown by Monte 
Carlo simulations [11] and integral equations studies [12], 
like-charge macromolecules in solution can attract. This 
counterintuitive phenomenon is the hallmark of electro-
static correlations between ions [13]. Experiments and 
system-specific simulations proved it to be of paramount 
importance to explain cement cohesion [14] [16], docking 
of vesicles [17] [18], DNA condensation in viruses or cells 
[19], as well as the behavior of like-charged mica sur-
faces [20], polyelectrolytes [21], lamellar systems [22], and 
lipid bilayers [23] [24]. A time-honored rule of thumb is 
that like-charge attraction requires multivalent counteri-
ons [2] [9].

From a theoretical standpoint, like-charge attraction 
provides a complex many-body problem [25] [27]. Analy-
tical progress is solely possible in the case where counteri-
ons are the only small species present (no added salt) and 
for simple geometries, e.g. where point ions are confined 
in water between two planar charged surfaces. These 
simplifications maintain physical relevance: on the one 
hand, confinement often leads to coion exclusion and no-
salt conditions [38] [40]; on the other hand, effective in-
teractions for more complex geometries can be obtained 
by means of a Derjaguin approximation, once the planar 
geometry has been solved [41].

We shall address the problem of understanding the 
equation of state of a correlated salt-free system, i.e. how 
the force between charged plates changes with their dis-
tance. The system is represented in the insets of Fig. 1, 
while the rest of the figure shows a sketch of its equation 
of state, which is non-monotonic. We argue below that 
the challenge is to understand the increasing \( W \) branch, 
while the short distance \( IG \) regime follows from a simple 
ideal gas argument. More importantly, it is the \( W \) branch 
that is relevant for a number of applications. While much 
analytical and computational effort has been invested in 
this very question and the role of ionic correlations em-
phasized [13] [25] [27] [30] [37] [22] [23], most theories fail at 
accounting for the \( W \) branch.

Our motivation is threefold. First, we show that the \( W \) 
branch is more universal than previously thought and is 
closely related to the zero-temperature equation of state. 
Second, elaborating on a number of exact results, we de-
rive a versatile and accurate equation for the pressure,
that covers not only the W branch, but the whole distance range. This equation of state passes the tests of exact known limiting behaviors whilst remaining simple, at variance with previous attempts. The third objective of this paper is to investigate a situation where our slab setting is of much relevance, that is under extreme confinement (small interplate distance $d$, on the order of the size of a water molecule), where the molecular nature of the solvent cannot be disregarded. In line with a number of recent works [23, 46–50], it then becomes essential to account for the destructuring of the water network in the slab, which leads to a dramatic decrease of screening, and a concomitant increase not only of correlations, but also of the attractive force between the surfaces. We show that our equation of state applies here in a “vacuum” formulation that we coin the “locked water picture”, and that our equation of state applies here in a “vacuum” regime. The so-called coupling parameter, quantifying the importance of ionic correlations, follows as

$$\Xi = \frac{q^2 l_B}{\mu} = 2\pi l_B^2 q^3 \sigma. \quad (1)$$

Comparing electrostatic and thermal energies, $\Xi$ can be viewed as a dimensionless measure of surface charge, inverse permittivity or, more formally, inverse temperature. $\Xi < 1$ is the regime where Poisson-Boltzmann theory prevails and the two plates repel, while for $\Xi > 12$ like-charge attraction sets in [31]. Given that most charged natural or synthetic surfaces have $\sigma < 1$ nm$^{-2}$ and that $l_B \approx 0.7$ nm in bulk water, monovalent counterions ($q = 1$) lead to small couplings ($\Xi < 4$) and fall under the Poisson-Boltzmann repulsive phenomenology. This explains the rule of thumb alluded to above: attraction, if any, requires $q \geq 2$ and is not possible with monovalent ions. Because of its cubic dependence on valence $q$, $\Xi$ can reach or significantly exceed a few tens in a wealth of experiments. This is the strong coupling regime we are interested in. Two additional lengths need to be introduced. First, $a = \sqrt{q/\sigma}$ defines the Wigner lattice spacing in the ground state [33–35], i.e. the distance between neighbour ions when $\Xi \to \infty$ and ions crystallize on each plane. This quantity, which loses relevance in the Poisson-Boltzmann regime, is indicated in Fig. 1. It remains essential whenever $\Xi$ is not small. The last player here is the distance $\ell$ that discriminates between the two regions, IG and W in Fig. 1. We will see that $\ell \propto \sqrt{\alpha \mu}$. For large $\Xi$, the four lengths are in the order $\mu < \ell < a < l_B$ and their ratios only depend on $\Xi$, as indicated by the axes in Fig. 1.

The two branches in Fig. 1 correspond to opposite limiting situations. The left branch has ideal gas nature [33] and is simple to explain: the two plates are so close that all microions lie in the same plane, and the inter-ions electric field vanishes by symmetry (as a consequence, the potential is quadratic with lateral displacement, a result used below). Since the electric field due to the equal plates also vanishes in the slab, microions become homogeneously distributed along the z coordinate perpendicular to the plate (see Fig. 1); their number density reads $n(z) = 2\sigma/(qd)$ by electroneutrality. We then in-

**RESULTS AND DISCUSSION**

**Relevance of ground-state physics.** We start by a length scale analysis [36, 37], that provides a fresh outlook. Until stated otherwise, the solvent is consid-
This yields a rescaled pressure covering both IG and W sectors. Besides ionic positions, the Hamiltonian of the system depends only on the single parameter $\Xi$, so that the rescaled pressure has two arguments: $d$ and $\Xi$. We consider two ways of taking the $\Xi \to \infty$ limit: by keeping $d/\mu$ constant or by keeping $d/a$ constant. Since $\ell/\mu \propto \Xi^{1/4} \to \infty$, the former magnifies the IG branch; conversely, since $\ell/a \propto \Xi^{-1/4} \to 0$, the latter magnifies the W branch. We can then write the exact relations:

$$
\lim_{\Xi \to \infty} \tilde{P}(d, \Xi) \bigg| _{d} = \frac{2\mu}{d} - 1; \quad (4)
$$

$$
\lim_{\Xi \to \infty} \tilde{P}(d, \Xi) \bigg| _{d} = \tilde{P}_{gs}(d); \quad (5)
$$

where $\tilde{P}_{gs}$ varies over distances of scale $a$, as shown in Fig. 2. In Eq. (4), the large $\Xi$ limit is taken at fixed $d/\mu$ while it is the ratio $d/a$ that is held fixed in Eq. (5).

Statement (4) indicates a flat, ideal gas profile on a scale $a$.

FIG. 2. Rescaled pressure versus distance. Monte Carlo simulations from $\Xi_{gs}$ (symbols) are compared to the ground-state pressure $\tilde{P}_{gs}$ ($\Xi \to \infty$, continuous black line) and to the analytical equation of state [2, 5] (dashed lines). The inset shows $\kappa$ versus $d/a$: the solid line is the exact ground-state branch, while the dashed line is the analytical approximation [8].

By the contact theorem [3, 11, 56], a general and exact result relating pressure $P$ to ion density at contact $n(0)$:

$$
P = kT (n(0) - 2\pi l_0 \sigma^2). \quad (2)
$$

This yields a rescaled pressure

$$
\tilde{P} = \frac{P}{2\pi l_0 \sigma^2 kT} = \frac{2\mu}{d} - 1. \quad (3)
$$

This ideal gas equation of state (dashed gray line in Fig. 1) is in good agreement with the pressure measured in the decreasing IG regime.

The complementary W branch is more subtle, and fundamentally many-body. We plot in Fig. 2 the dimensionless pressure $P_{gs}$ as a function of $d/a$ for various couplings $\Xi$ (symbols). This reveals a remarkable collapse: $\tilde{P}_{gs}$, that should converge towards the ground state pressure $P_{gs}$ (black line, worked out in [53]) only as $\Xi \to \infty$, remains very close to this limiting curve down to unexpectedly small $\Xi$. Even at low coupling, attraction must then stem from the staggering of ions on opposite plates, an ion facing a hole, as in the zero-temperature crystal phase [53, 55]. Increasing $d$, one transitions rather abruptly from the ideal gas branch where interactions are immaterial, to the W branch that is truly many body.

The minimum of the pressure curve, giving the crossover IG $\leftrightarrow$ W at $d = \ell$, is found by equating both limiting results, Eq. (4) and $P_{gs}$ (we use $P_{gs} + 1 \propto d/a$ at small $d$, as per Fig. 2). This yields $\ell \propto a \Xi^{-1/4} \propto \mu \Xi^{1/4}$. This exact result, while confirming findings from other methods [32], disproves the so-called Rouzina-Bloomfield criterion [25, 40] that had $\ell \propto a$.

**Analytical equation of state.** We turn to the second objective of finding an accurate equation of state covering both IG and W sectors. Besides ionic positions, the Hamiltonian of the system depends only on the single parameter $\Xi$, so that the rescaled pressure has two arguments: $d$ and $\Xi$. We consider two ways of taking the $\Xi \to \infty$ limit: by keeping $d/\mu$ constant or by keeping $d/a$ constant. Since $\ell/\mu \propto \Xi^{1/4} \to \infty$, the former magnifies the IG branch; conversely, since $\ell/a \propto \Xi^{-1/4} \to 0$, the latter magnifies the W branch. We can then write the exact relations:

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where $\tilde{P}_{gs}$ varies over distances of scale $a$, as shown in Fig. 2. In Eq. (4), the large $\Xi$ limit is taken at fixed $d/\mu$ while it is the ratio $d/a$ that is held fixed in Eq. (5).

Statement (4) indicates a flat, ideal gas profile on a scale $a$.
at $T = 0$. Exact in the ground state, we will show it is an excellent approximation even at finite $\Xi$.

Since the ionic layer thickness is always much smaller than the inter-ion distance $a$ [43], the ionic profile follows from a single particle argument in the effective potential $\kappa q e E_0 z = kT \kappa z / \mu$; thus $n(z) \propto e^{-\kappa z / \mu}$. We normalize $n$ by imposing electroneutrality (i.e. $\int_0^d n(z) dz = 2\sigma / q$) and, using once more the contact theorem [2], we obtain the equation of state

$$\tilde{P} = \kappa \frac{1 + e^{-\kappa d / \mu}}{1 - e^{-\kappa d / \mu}} - 1,$$

(7)

which exhibits a dependence on the two length scales $\mu$ and $a$ (through $\kappa$). Such functional form coincides with the leading order of a large-$\Xi$ expansion in the Wigner Strong Coupling approach [43]. What remains is to find an expression for $\kappa = 1 + \tilde{P}_{gs}$. It was shown that $\tilde{P}_{gs} \sim -3 \exp(-a d / \alpha)$ for $d \gg a$, with $\alpha = 4\pi/(3^{1/4}\sqrt{2}) \approx 6.75$ [55]. Given that $\kappa \to 0$ for $d \ll a$, the simplest form compatible with the two limits is

$$\kappa = 1 - \frac{3}{2 + e^{\alpha d / a}}.$$

(8)

This is compared with Eq. (6) in the inset of Fig. 2.

Equation of state (7), supplemented with (8), is our jackknife pressure. It is not exact, but it is the only available simple pressure compatible with exact limiting results, such as (4) and (5). It complies with the energetic attraction/entropic repulsion phenomenology put forward in earlier works, e.g. [15]. Fig. 2 (dashed lines) illustrates its good accuracy down to $\Xi = 50$ when compared to Monte Carlo results (symbols), thus confirming the relevance of ground-state physics at surprisingly low $\Xi$.

**Water destructuring under confinement.** The previous discussion holds within the (solvent implicit) primitive model, where water enters the description only through its permittivity $\varepsilon_r$. It is customary to take $\varepsilon_r = 80$, the bulk value. Yet, in situations of strong confinement, when $d$ becomes comparable to the size of water molecules, this choice is questionable, if not misguided. Recent works indeed point to the fact that water organization is strongly affected at small $d$, with a freezing of orientational degrees of freedom: this decreases its effective $\varepsilon_r$ [23] [46] [51]. One might believe that these considerations ruin implicit-water approaches, and in particular the primitive model. We show evidence that this is not the case.

Recently, pure water confined between neutral surfaces below 2 nm was shown to exhibit a relative permittivity between 1 and 4, depending on $d$ [47]. This came as a confirmation of theoretical results that anticipated a drastic decrease in the component of the dielectric tensor perpendicular to the confining surfaces [46] [49]. A complete understanding of how confinement and environment affects $\varepsilon_r$ is still lacking, but we speculate that for strongly charged surfaces and in presence of ions, water mobility is suppressed even more strongly, resulting in a permittivity close to that of vacuum. We argue that decreasing $d$, the number of counterions in the slab remains fixed by electroneutrality, while water content diminishes. Water is then dominantly immobilized in hydration layers around counterions, and cannot screen Coulombic interactions like it does in the bulk. This behaviour was confirmed in simulations [50], where qualitative measurements of both components of the dielectric tensor showed a drastic decrease. As we seek to understand attraction from first principles only, we make the assumption that water molecules in the slab are so few that none of them is free: this leads to the crude approximation $\varepsilon_r \to 1$, for $d$ smaller than a couple nanometres. We coin the resulting suppressed-permittivity primitive model “locked water” model [50]: in it, $l_B$ is renormalized, substituted by $l_B^{\text{locked}} = 80 l_B$ in all expressions. Besides dampening van der Waals interactions [23] [57], this enormously increases coupling to $\Xi^{\text{locked}} = 80^2 \Xi$ and drives a massive increase in attraction: this is consistent with what proposed in Ref. [43], where pressure between confined decanol bilayers was shown to be described by a larger coupling than expected. A consequence is that for small enough $d$, a very small surface charge $\sigma$ becomes sufficient to lead to attraction even with monovalent ions ($q = 1$), unlike previously thought: the condition for attraction mentioned above, now $\Xi^{\text{locked}} > 12$, is met for $\sigma q^3$ just above 0.03 nm$^{-2}$, at room temperature.

The equation of state (7)-(8) within the “locked water” primitive model can be tested against water-explicit simulations. We take as sample systems cement and clays, whose effective interactions at the nanoscale have puzz...
FIG. 4. Pressure predictions for clays. Above, montmorillonite (MMT, $\sigma = 0.75\text{ nm}^{-2}$); below, vermiculite (VMT, $\sigma = 1.5\text{ nm}^{-2}$). Valence $q$ increases columnwise, from 1 to 3. Coupling $\Xi_{\text{locked}}$ varies accordingly, from 15000 (top left) to 820000 (bottom right). Circles show dipolar model MD simulations from [62], while black lines are from Eq. (7)-(8) at $\varepsilon_r = 1$. A shift accounts, as in Fig. 3, for finite ion and wall sizes. The “locked water” model predicts a pressure between the two black curves.

zled scientists for decades. Fig. 3 compares different models for calcium silicate hydrate (C-S-H), the main binding agent of cement [14]; these include fully atomistic simulations [51] and two coarse-grained models [50], where particles are Lennard-Jones spheres and the dielectric properties of a water molecule emerge either from a point dipole (dipolar model, DM) or from point partial charges (SPC/E model [58]). Analogously, Fig. 4 compares dipolar model simulations of clays from [62] with Eq. (7)-(8) for “locked water”. In both figures, curves are shifted horizontally to account for different ion sizes and different descriptions of the walls. This shift is not the result of a fit and is determined by parameters used in simulations for wall thickness or ion diameter. Ambiguities resulting from surface roughness or soft potentials are small and have been resolved by checking for the positions of the two furthermost peaks of the counterion density. In addition, while in simulations of cement the high electric fields involved, at any $d$, squeeze counterions against the walls and expel half of their solvation shell [50], the lower surface charges of clays are not always sufficient to quench hydration: for clays, the shape of the solvation shells is known to drastically depend on distance, incidentally causing swelling [59]. This is why in Fig. 4 we include two analytical curves, shifted two water molecule diameters from one another. The theoretical prediction lies between the two curves.

Charge regulation and surface charge heterogeneity are not included explicitly in our model. These aspects might play a role in determining the strength of attraction. For instance, pH is known to drastically regulate charge in cement, as the hydration reaction proceeds [50], [61]. A relevant observation is that, in the ground-state picture that inspires our equation of state, ions are immobilised on the surface, as also observed in simulations [50]. This is effectively equivalent to having counterions chemically bound to the surface, assuming that they produce a localised excess positive charge on the negative plate. In sum, the only thing that matters to have correlation-induced attraction is a (staggered) non-uniform charge distribution on the plates, be it from localised chemically bound charges or from ions dwelling at the interfaces. Consistently with this scenario, charge regulation with discrete titration sites has been recently suggested to even increase attraction at the nanoscale [63].

In both Figs. 3 and 4, the dramatic increase from the bulk $\Xi$ to $\Xi_{\text{locked}}$ decreases the crossover length $\ell$ by a factor $\sqrt{80} \approx 9$. This extends the relevance of the $W$ branch and contributes to masking the $\Xi G$ regime. In simulations, the steep rise at small distances is indeed not of electrostatic origin, but due to steric ion-wall repulsion. In general, in real systems, the $\Xi G$ branch and possibly the initial part of the $W$ branch will be masked by other additive components of the pressure, such as hydration or surface-surface contact forces. These short-range forces, typically described at a phenomenological level [41], are not accounted for in our first-principles model. However, the many experimental and numerical observations of like-charge attraction under confinement, as well as Fig. 3 and 4, suggest that they are unlikely to screen the longer-range attractive branch of the electrostatic correlation pressure.
CONCLUSIONS

We have derived a robust, simple and accurate equation of state for strongly charged plates with ions and water in between. This jackknife pressure, Eq. (7)-(8), satisfies exact requirements, both at the scale of the Gouy-Chapman length $\mu$, Eq. [4], and at that of the Wigner spacing $\alpha$, Eq. [5]. This clarifies the rather elusive primitive model phenomenology, showing that ground state physics, a basic ingredient of our equation of state, is unexpectedly relevant even at moderate coupling. The origin of attraction must then lie in the fact that ions dwelling next to one plane anticorrelate with those next to the other plane, reminiscent of the staggered lattice. This “locked water” view predicts in particular the possibility of attractive effects with monovalent counterions, at variance with common belief, but explaining the behaviour of clays and lipid bilayers [23]. Using the present jackknife equation of state (1)-(8) as an effective potential to upscale coarse-grained simulations is a promising avenue.

ACKNOWLEDGEMENTS

We thank Martin Trulsson for useful discussions and for providing us with simulation data. This work has received funding from the European Union’s Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement 674979-NANOTRANS. The support received from VEGA Grant No. 2/0092/21 is acknowledged.

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