Waterlike anomalies in a 2D core-softened potential

José Rafael Bordin\textsuperscript{1,*} and Marcia C. Barbosa\textsuperscript{2,+}

\textsuperscript{1}Campus Caçapava do Sul, Universidade Federal do Pampa,
Av. Pedro Anunciação, 111, CEP 96570-000, Caçapava do Sul, RS, Brazil
\textsuperscript{2}Instituto de Física, Universidade Federal do Rio Grande do Sul
Caixa Postal 15051, CEP 91501-970, Porto Alegre, RS, Brazil

Abstract

We investigate the structural, thermodynamic and dynamic behavior of of a two dimensional core-corona system using Langevin Dynamics simulations. The particles are modeled using a core-softened potential that exhibits waterlike anomalies for the 3D case. For quasi-2D systems we have observed previously a new region of structural anomaly. Now, our results show that a new region of structural, density and diffusion anomalies arises for the 2D system. Our findings indicates that while the anomalous region at lower densities observed is due the competition between the two length scales in the potential, the traditional mechanism, the higher densities anomalous region is related to changes in the particles conformation and a melting region.

* josebordin@unipampa.edu.br
† marcia.barbosa@ufrgs.br
I. INTRODUCTION

Anomalous materials show characteristics which differ from the observed in most substances. For instance, it is expected that liquids contract upon cooling at constant pressure and diffuse slower upon compression. However, anomalous fluids expand as the temperature is decreased and move faster as the pressure grows. The most known anomalous system is water, more than 70 known anomalies [1], but there are another anomalous fluids. The maximum in the diffusion coefficient at constant temperature was observed not only for water [2] but also for silicon [3] and silica [4]. The maximum in the density well known in water [5] is also seen in silicon [4], silica [6], Te [7], Bi [8], Si [9], GeTe85 [10], liquid metals [11], graphite [12] and BeF2 [13].

Since the seminal work by Jagla [14–16] core-softened potentials have been widely used in the literature to study the behavior of anomalous fluids [17–24]. The origin of this behavior is associated with the existence of two characteristic length scales in the potential [25, 26]. The competition between the conformation at the first or the second length scale can be directly related to the anomalies [27].

Core-softened potentials have been also applied to study colloidal systems. Experimental works have shown that the effective interaction between colloids can be modeled by core-softened potentials [28, 29]. The origin of the two length scales goes as follows. The colloids are usually made of molecular subunits which form a central packed agglomeration and a less dense and more entropic periferical area. This core-corona structure can be described by a hard core and a soft corona. Then it becomes natural to model the system by a two length scales potential which leads to the self-assembled patterns observed in these colloidal systems [30–40].

Most of works have focused in the self-assembly and distinct patterns observed in these systems [34–39, 41, 42], with few works focusing on the fluid phase and the dynamics [31, 40, 43–45]. Therefore, a natural question that arises is how the fluid phase of the 2D core-corona system behaves for different pressures and temperatures and particularly when exposed to a solvent.

In order to address this question in this paper the interparticle colloid-colloid interaction has a repulsive core with a smooth shoulder. For a molecular system in 3D this potential both in the bulk [20, 46] and when confined in quasi-2D systems [47–49] shows waterlike anomalies.
Interesting, in the quasi-2D case a new region of structural anomaly was observed [50]. Now, we show that for 2D systems this same potential presents a second region of anomalies in the pressure versus temperature phase diagram. A mechanism for the appearance of this second anomalous region is proposed.

Our paper is organized as follows. In the Section II the model and the details about the simulation method are presented. In the Section III results are discussed. The conclusions are shown in Section IV.

II. THE MODEL AND THE SIMULATION DETAILS

![Figure 1](image.png)

**FIG. 1.** Core-softened interaction potential $U$ between two core-corona particles. Inset: schematic depiction of the particles, with the core (first length scale at $r_{ij} \equiv r_1 \approx 1.2\sigma$) and the soft corona (second length scale at $r_{ij} \equiv r_1 \approx 2.0\sigma$).

For simplicity, all the physical quantities are computed and displayed in the standard Lennard Jones (LJ) reduced units [51]. The system consists of $N = 2000$ disks with diameter $\sigma$ and mass $m$ with a potential interaction composed of a short-range attractive Lennard Jones potential and a Gaussian therm centered in $r_0$, with depth $u_0$ and width $c_0$,

$$U(r_{ij}) = 4\epsilon \left[ \left( \frac{\sigma}{r_{ij}} \right)^{12} - \left( \frac{\sigma}{r_{ij}} \right)^{6} \right] +$$

$$u_0 \exp \left[ -\frac{1}{c_0^2} \left( \frac{r_{ij} - r_0}{\sigma} \right)^2 \right], \quad (1)$$
where \( r_{ij} = |\vec{r}_i - \vec{r}_j| \) is the distance between two disks \( i \) and \( j \). This potential can be parametrized to have a ramp-like shape, and was extensively applied to study systems with water-like anomalies [20, 46]. The parameters used in this work are \( u_0 = 5.0, c = 1.0 \) and \( r_0/\sigma = 0.7 \). The interaction potential, showed in figure 1, has two length scales. The first scale is at \( r_{ij} \equiv r_1 \approx 1.2\sigma \), where the force has a local minimum, and the other scale at \( r_{ij} \equiv r_2 \approx 2\sigma \), where the fraction of imaginary modes of the instantaneous normal modes spectra has a local minimum [52]. The cutoff radius for the interaction is \( r_c = 3.5 \). The two length scales in the potential allow for representing hard core-soft shell colloids [35, 41].

In this work we use the Langevin thermostat [51] to mimic the solvent effects. Hydrodynamics interactions were neglected. Since the system is in equilibrium we do not expect that this will change the long-time behavior. The temperature was simulated in the interval between \( T = 0.01 \) and \( T = 0.40 \). The number density is defined as \( \rho = N/A \), where \( A = L^2 \) is the area and \( L \) the size of the simulation box in the \( x \)- and \( y \)-directions. \( \rho \) was varied from \( \rho = 0.05 \) up to \( \rho = 0.60 \), and the size of the simulation box was obtained via \( L = (N/\rho)^{1/2} \). For clarity, in the \( p \times T \) phase diagram the higher isochore shown is \( \rho = 0.525 \) since no anomalous behavior was observed above this density.

The time step used in the simulations was \( \delta t = 0.001 \), and periodic boundary conditions were applied in the two directions. We performed \( 3 \times 10^7 \) steps to equilibrate the system. These steps are then followed by \( 5 \times 10^7 \) steps for the results production stage. To ensure that the system was equilibrated, the pressure, kinetic and potential energy as function of time was analyzed. Snapshots of the system was also used to verify the equilibration. Also, two distinct initial configurations were used for each point: a random fluid-like configuration and a solid-like in a square lattice. The results showed independent from the initial configuration.

To study the dynamic anomaly the relation between the mean square displacement (MSD) with time was computed, namely

\[
\langle [\vec{r}(t) - \vec{r}(t_0)]^2 \rangle = \langle \Delta \vec{r}(t)^2 \rangle ,
\tag{2}
\]

where \( \vec{r}(t_0) = (x(t_0)^2 + y(t_0)^2) \) and \( \vec{r}(t) = (x(t)^2 + y(t)^2) \) denote the coordinate of the particle at a time \( t_0 \) and at a later time \( t \), respectively. The MSD is related to the diffusion coefficient \( D \) by

\[
D = \lim_{t \to \infty} \frac{\langle \Delta \vec{r}(t)^2 \rangle}{4t} .
\tag{3}
\]

The structure of the fluid was analyzed using the radial distribution function (RDF)
$g(r_{ij})$, and the pressure was evaluated with the virial expansion. Directly related to $g(r_{ij})$, we characterize the structural anomaly using the translational order parameter $\tau$, defined as [53]

$$\tau \equiv \int_0^{\xi_c} | g(\xi) - 1 | \, d\xi,$$

(4)

where $\xi = r\rho^{1/2}$ is the interparticle distance $r$ divided by the mean separation between pairs of particles $\rho^{1/2}$. $\xi_c$ is a cutoff distance, defined as $\xi_c = L\rho^{1/2}/2$. For an ideal gas (completely uncorrelated fluid), $g(\xi) = 1$ and $\tau$ vanishes. For crystal or ordered fluids a translational long order ($g(\xi) \neq 1$) persists over long distances, increasing the value of $\tau$.

In order to check if the system shows density anomaly the temperature of maximum density (TMD) was computed for different isochors as follows. Using thermodynamical relations, the TMD was characterized by the minimum in the pressure versus temperature diagram along isochores,

$$\left( \frac{\partial p}{\partial T} \right)_\rho = 0.$$

(5)

The separation between the fluid and amorphous solid phases was defined by the analysis of the total energy, RDF, MSD and system snapshots. When the particles have a well defined structure and have a very low or zero mobility the phase was defined as solid. When the system has nonzero mobility, it was considered to be in the fluid phase. These separations were confirmed by the evaluation of the heat capacity [51]. The results were supported by larger simulations, using using $N = 5000$ disks and $5 \times 10^9$ steps.

III. RESULTS AND DISCUSSION

For most fluids, the diffusion constant $D$ decreases with the density $\rho$. The reason for this behavior is that the particles become more structured as the density increases. Then the translational order parameter $\tau$, defined by the Eq. 4 grows with $\rho$ as follows. At low densities, $g(r) \approx 1$ and then $\tau \approx 0$. As the density increases, $g(r) \neq 1$ for many values of $r$ and then $\tau$ grows. Anomalous fluids show the opposite behavior. For these materials in a certain range of temperature and pressures, the anomalous region, the diffusion coefficient increases with density, and $\tau$ decreases with $\rho$. The figure 2(a) shows the dependence of the diffusion coefficient, $D$, with the density, $\rho$. As the density is increased from the gas phase, the diffusion coefficient decreases, reaches the first minimum in the density and increases
FIG. 2. (a) Diffusion constant $D$ and (b) translational order parameter $\tau$ as function of the system density. In both figures maxima and minima that characterize the anomalies are represented by a dashed red line. For the diffusion anomaly, the anomalous region at lower densities ranges from the isotherm $T = 0.07$ to $T = 0.24$, while the second anomalous region goes from $T = 0.07$ to $T = 0.15$. In the case of the structural anomaly, the first anomalous regions goes from the isotherm $T = 0.07$ to $T = 0.40$, and the anomalous region at higher densities is located between the temperatures $T = 0.07$ and $T = 0.24$. The errors bars in $D$ and $\tau$ are smaller than the data point.

reaching a the first maximum which characterizes the first anomalous region from isotherm $T = 0.07$ to $T = 0.24$. Then, as the density is increased even further, for isotherms between $T = 0.07$ and $T = 0.15$, a second minimum and a second maximum are observed.

The translational order parameter versus density shown in the figure 2(b)also a shows the existence of two anomalous regions. The first is located between the isotherms $T = 0.07$ and $T = 0.40$ and lower values of density, while the second occurs at higher densities and from the isotherm $T = 0.07$ to $T = 0.24$.

In the figure 3 the pressure versus temperature phase diagram is illustrated for the system. The isochores are the gray lines. The temperature of low density line, related to the density anomaly, is shown in green. The TMD anomaly is also present in both anomalous region.

For the 3D molecular liquid the TMD region in the pressure versus temperature phase diagram is located inside the diffusion maxima and minima regions which are inside the maxima and minima of the translational order parameter [20, 46] regions. This sequence
FIG. 3. $p \times T$ phase diagram of the colloidal system. The gray lines are the isochores. The dashed blue line delimits the structural anomaly regions, with the maximum and minimum values of $\tau$. The dotted-dashed red line delimits the diffusion anomaly regions, with the minimum and maximum values of $D$. The green line defines the density anomaly region and corresponds to the temperature of maximum density (TMD) line. The black stars are located over the isotherm $T = 0.12$ and correspond to the densities $\rho = 0.15$, $\rho = 0.225$, $\rho = 0.325$, $\rho = 0.35$, $\rho = 0.425$ and $\rho = 0.525$. The dotted black line delimits the fluid and amorphous solid region. The errors obtained for the mean value of $p$ and $T$ were smaller than $10^{-4}$ for all cases and the errors bars were omitted for simplicity.

is the same that observed in water, the so-called waterlike hierarchy. Here, unlike the 3D molecular system, the 2D Brownian system the hierarchy in the anomalies is distinct from the water hierarchy. This change in the hierarchy was already observed in others works, and it is attributed to the changes in the competition between the scales [21, 45, 54], to the formation of an ordering structure [55] or to the dimensional change from 3D to 2D [56].

In our case, the change in the hierarchy is due to the presence of solid-like (or pinning-like) structures and to the change in the dimensionality as it is shown next. In addition to the hierarchy, another question is also relevant: why there are two anomalous regions in this
system?

In order to understand the mechanism which generates the two anomalous regions, the behavior of the system at the isotherm \( T = 0.12 \), show as stars in the phase diagram, figure 3, is analyzed. In the case of the 3D system the mechanism which explains the existence of the waterlike anomalies is the competition between the two length scales [25]. This is observed in the radial distribution function of molecular systems as follows.

In the anomalous region the first peak of the RDF increases with the density while the second peak decreases [27]. This behavior is also observed in the figure 4(a) which corresponds to the low density and low pressure region of the figure 3. In this region as \( \rho \) increases, particles move from the second at \( \approx 2.0 \) to the first length scale at \( \approx 1.2 \). Therefore, the system has competition between the scales and, as consequence, waterlike anomalies.

The figure 4(b) shows the RDF for densities bellow, inside and above the second anomalous region. As the density is increased the peaks of the \( g(r_{ij}) \) related to the first and to the second length scales increase, reach a maximum, and then decrease as the density is increased. Therefore, even though no competition between the two length scales is observed, the system shows density, diffusion and structural anomaly. In order to reveal the origin of the anomalies, instead of looking to the two peaks it is necessary to exam the valley between them. As the density increases from \( \rho = 0.35 \) to \( \rho = 0.425 \) this valley goes down, becoming zero. These zero for the RDF suggests that as the density \( \rho = 0.425 \) is approached from lower densities, the system is becoming solid, or well structured. The MSD, illustrated in the figure 4(c), supports this result. The slope of the MSD decreases from \( \rho = 0.35 \) to \( \rho = 0.425 \) which indicates a decrease in the diffusion. This is reinforced by the snapshots shown in the figure 4(d) as an stripe phase. However, increasing the density of the system even further to \( \rho = 0.525 \), the disks becomes disordered. Despite the absence of competition, this behavior can also be understood based in the two length scales characteristics.

When the fluid is in the stripe structure the interparticle distance between disks in the same stripe is the first length scale, and the stripes are separated by the second length scale - this is why both peaks increase from \( \rho = 0.35 \) to \( \rho = 0.425 \). At \( \rho = 0.425 \) the particles have the minimum in the diffusion. Increasing the density, there is no more space for the stripes remain at the distance \( \approx 2.0 \), and they break into the disordered fluid. Essentially, the entalpic contribution to the free energy (second length scale) is overcomed by entropic
FIG. 4. Analysis of the isotherm $T = 0.12$ for the colloidal system. (a) Radial distribution function (RDF) $g(r_{ij})$ for densities inside the first anomalous region indicates that these anomalies are originated by the competition between the two length scales. The arrows shows how the peaks in the $g(r_{ij})$ moves. The black arrow shows the grow of the second peak for densities below $\rho = 0.225$, the red arrow the decrease in the second peak and the green arrow the increase in the first peak for densities between $\rho = 0.225$ and $\rho = 0.325$.  (b) Radial distribution function (RDF) $g(r_{ij})$ for densities inside the second anomalous region indicates that there is not a competition between the scales. Both peaks increase from $\rho = 0.35$ to $\rho = 0.425$, while the valley between them decreases. This is indicated by the red arrows. The green arrows shows that from $\rho = 0.425$ to $\rho = 0.525$ the peaks decrease and the valley increases. Therefore, the system becomes more structured and then more disordered, which explain the second structural anomaly. Related to this transition from disordered-ordered-disordered structured, the slope of the MSD curve decreases and then increases, as is shown in (c). The snapshots in (d) show the disks conformation, including a kagome lattice at $\rho = 0.60$. 
contribution (the first length scale) [57]. Then the system goes from a disordered fluid to a ordered fluid (similar to a liquid-crystal) with lower diffusion and then gets disordered again, diffusing faster. In this reentrant melting region we observe that $D$ increases with the density while $\tau$ decreases, leading to the second anomalous regions. As the density increases even more, the system goes to a solid phase with a kagome lattice, as the snapshot in the figure 4(d) shows.

Previous studies have shown that the existence of multiples competitive scales leads to multiples anomalous regions. In the work by Barbosa and co-workers [26] they have used a soft-core potential with three characteristic length scales and have found two TMD lines and transitions between three fluids phases. Also, we have observed two structural anomalous regions in quasi-2D systems, were the new anomalous regions can be related with the melting of the central layer between two walls [50]. This is similar to what we observed for the 2D system, were a reentrant melting region leads to the appearance of anomalies.

IV. CONCLUSION

Langevin Dynamics simulations of 2D core-softened disks were performed in order to analyze the system fluid phase for structural, thermodynamic and dynamic anomalous behavior.

The core-corona system shows the presence of two anomalous regions in the pressure versus temperature phase diagram. Also, a change in the waterlike hierarchy of anomalies was observed which can be associated with the change in the dimensionality.

The two distinct regions with anomalous behavior observed for the colloidal system arises due to two distinct mechanisms. The first region, at low densities, is associated with the competition between the two length scales in the interaction potential. This is the same mechanism observed in previous works and in the molecular system. We have shown that the second anomalous region is not related to the competition observed in the RDF, but to a reentrant fluid phase. This leads the fluid to suffer a transition from a disordered structure to a ordered structure and then back to a disordered structure, resulting in a increase in the diffusion as the density increases and a decrease of $\tau$ as $\rho$ increase - the anomalous behavior.

Nevertheless, this was not the first time that we have observed two anomalous regions for core-softened fluids. In a previous work, core-softened potentials with three scales had
lead to two regions of density anomaly [26]. As well, we have showed that fluids modeled by potential equation (1) confined between two flat walls have a second structural anomaly. This new anomaly was not related to the competition between the potential scales, but to changes in the number of fluid layers between the walls [50]. The change in the number of layers is a additional competition induced by the confinement. In this work, the competition was induced by the resulting fluid reentrant phase. Therefore, our main finding is that another mechanisms, despite the competition between the scales, can generate competitions in the system that lead to waterlike anomalies.

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