

Superdiffusion in quasi-two-dimensional Yukawa liquids

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The emergence and vanishing of superdiffusion in quasi-two-dimensional Yukawa systems are investigated by molecular dynamics simulations. Using both the asymptotic behavior of the mean-squared displacement of the particles and the long-time tail of the velocity autocorrelation function as indicators of superdiffusion, we confirm the existence of a transition from normal diffusion to superdiffusion in systems changing from a three-dimensional to a two-dimensional character. A connection between superdiffusion and dimensionality is established by the behavior of the projected pair distribution function.

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I. INTRODUCTION

Diffusion is a fundamental transport mechanism which plays a dominant role in many physical, chemical, and biological systems [1,2]. It is not only of academic but also practical interest to study diffusion in two-dimensional systems since many real-world systems can be described as being two-dimensional or quasi-two-dimensional, including surfaces or layers of small width—e.g., quantum wells. Two-dimensional diffusion has long been known to exhibit anomalous behavior for a number of interactions and systems. One of the possible anomalies is the so-called superdiffusion: If the average mean-squared displacement $\langle |\vec{r}(t) - \vec{r}(t_0)|^2 \rangle$ of the particles grows faster than linearly with time, the system exhibits superdiffusive behavior [3]. In such a system, the diffusion proceeds faster than normal diffusion in the sense that a particle achieves a greater distance from its starting point than expected from Fick's law.

For three-dimensional simple systems in thermodynamical equilibrium, to the best of our knowledge, no superdiffusion has been observed to date. On the other hand, under nonequilibrium conditions, anomalous transport is well known—e.g., for chaotic systems [4], turbulent flows [5], or plasmas in turbulent magnetic fields [6]. These will not be considered here. For systems which are neither three-dimensional nor strictly two-dimensional, we expect a gradual transition from superdiffusive behavior to Fickian diffusion.

In two-dimensional systems of hard disks, a slow αt^{-1} decay of the long-time tail of the velocity autocorrelation function (VACF) was first observed by Alder and Wainwright [7]. Such a decay results in a divergent Green-Kubo integral. Superdiffusion has been observed experimentally by many researchers. In particular, quasi-two-dimensional complex plasmas [8–11], including driven-dissipative systems [12] and systems under laser-induced shear [13], were found to exhibit superdiffusive motion. No experimental evidence for superdiffusion was found by Nunomura *et al.* who examined an underdamped liquid complex plasma [14].

The intent of this study is to examine the superdiffusion in systems which are quasi-two-dimensional; i.e., the extension

in one spatial dimension of the system is much smaller than in the other two. This is often the situation in many experimental setups—for example, in dusty plasma experiments where the dust grains are levitated by an electrostatic force which is counteracted by gravity. It is clear that under such circumstances the particles are not rigorously restricted to a two-dimensional plane, but form a quasi-two-dimensional system. For example, Ref. [9] reports on experiments in which the dusty plasma under consideration consisted of two to three layers.

Here we consider a macroscopic system of charged particles interacting via a screened Coulomb (Yukawa) potential. It is of relevance to dusty plasmas [15], colloidal suspensions, electrolytes, and other systems. It is also an important theoretical tool since it allows one to tune the interparticle interaction (by varying the screening length of the Yukawa potential) from being very long ranged to almost contact interaction.

We report on molecular dynamics studies performed for quasi-two-dimensional systems. Our focus lies on the influence of the degree of quasi-two-dimensionality on the vanishing and emergence of superdiffusion.

II. MODEL AND SIMULATION TECHNIQUE

We study the system using equilibrium molecular dynamics simulations (e.g., [16]). The interaction of the particles is given by the Yukawa pair potential

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

Here, Q is the particles' charge and λ_D the screening length.

A two-dimensional Yukawa system is characterized by two parameters: the Coulomb coupling parameter $\Gamma = (Q^2/4\pi\epsilon_0)(1/ak_B T)$ and the screening parameter $\kappa = a_{ws}/\lambda_D$. T is the temperature and $a_{ws} = (n\pi)^{-1/2}$ is the Wigner-Seitz radius for two-dimensional systems, which depends on the areal density n . While our simulation box is periodic in the x and y directions, it is unbound in the z direction. The particles' perpendicular movement in the z

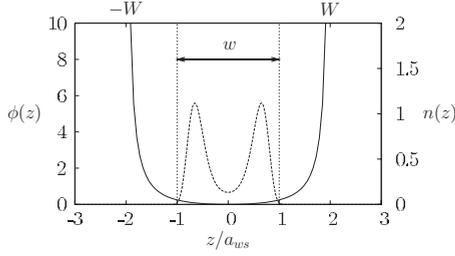


FIG. 1. Soft-box potential $\phi(z)$ and corresponding particle density $n(z)$. The trap is impenetrable at $\pm W$ and the particles can typically explore the trap width w (here, $w=2a_{ws}$).

direction is restricted by one of two confinement potentials

$$V^{\text{harm}}(z) = f \frac{Q^2}{4\pi\epsilon_0 a_{ws}^3} \frac{z^2}{2}, \quad (2)$$

$$V^{\text{box}}(z) = \frac{Q^2}{4\pi\epsilon_0} \left(\frac{e^{-(z+W)/\lambda_D}}{z+W} + \frac{e^{-(-z+W)/\lambda_D}}{-z+W} \right). \quad (3)$$

The confinement (2) is a simple harmonic trap where f denotes the trap amplitude [17].

The second model, Eq. (3), uses a box-shaped confinement with “soft” walls. It can be thought of as consisting of two rigid walls of immovable particles with the same interaction as the actual particles—i.e., a Yukawa interaction; see Fig. 1 for an illustration. Here, W is defined as $W = \frac{w}{2} + a_{ws}$, with w being the “width” of the box—i.e., twice the maximum displacement in the $+z$ or $-z$ direction. We use the width w as a third parameter for the soft-box confinement.

The choice of harmonic confinement allows us to reproduce typical experimental situations such as in dusty plasma experiments where the particles’ motion is often confined by a similar potential [18]. On the other hand, the confinement (3) gives us the possibility to study the effect of the dimensionality on superdiffusion while retaining comparable plasma conditions at all times. We achieve this by the following procedure: In the harmonic confinement, as the amplitude of the trap is decreased, the particle number is left unchanged and the particles can explore a wider vertical space. This results in an increased mean interparticle distance; i.e., the [three-dimensional (3D)] density is decreased. Contrarily, in the soft-box confinement, we chose $w = a_{ws}$ as a point to fix for the 3D density and scale the particle number as necessary to maintain that density for all values of w .

We performed simulations for a fixed coupling parameter $\Gamma = 200$. Prior to measurement, the particles’ velocities are rescaled at each time step to the desired temperature until a Maxwell distribution is well established. During the measurement, the velocities are not rescaled. Our simulations are carried out for $\kappa = 2.0$ and $\kappa = 3.0$. At these parameters, a 2D Yukawa system is well in the liquid phase, with the melting points being $\Gamma \approx 415$ and $\Gamma \approx 1210$ for $\kappa = 2.0$ and 3.0 , respectively [19].

The particle number in our simulations is $N = 6000$ for the harmonic confinement and $N = 4000 - 16\,000$ for the soft-box confinement with $N \geq 6000$ for $w \geq 1.5a_{ws}$.

In the following, time is given in units of the inverse plasma frequency $\omega_p = (Q^2/2\pi\epsilon_0 m a_{ws}^3)^{1/2}$, with m being the mass of the particles and lengths are measured in units of a_{ws} . We solve the equation of motion for each particle using the velocity Verlet algorithm [20].

To analyze diffusion properties we calculate the time dependence of the mean-squared displacement (MSD)

$$u_r(t) = \langle |\vec{r}(t) - \vec{r}(t_0)|^2 \rangle, \quad (4)$$

where $\langle \cdot \rangle$ denotes an ensemble average and $\vec{r} = (xy)$ is the position vector in the plane; the z component is bound from above due to the confining force and does not need to be taken into account.

The structure of the systems is characterized by measuring the density distribution $n(z)$ perpendicular to the confined direction and by the *projected pair distribution function* $g^*(r)$. The motion can be classified according to the time dependence $u_r(t) \sim t^\gamma$. Normal Fickian diffusion is characterized by a linear time dependence, $\gamma = 1$. If $\gamma > 1$ or $\gamma < 1$, motion is super- or subdiffusive, respectively. Ballistic—i.e., undisturbed—motion is trivially marked by $\gamma = 2$.

We have carried out calculations of the MSD for different trap amplitudes and box widths and determined the slope of the curve on a double-logarithmic plot between $t = 100\omega_p^{-1}$ and $t = 300\omega_p^{-1}$, which yields the diffusion exponent γ .

A more direct insight into a particle’s movement can be obtained from the decay of the VACF

$$Z(t) = \langle \vec{v}(t) \cdot \vec{v}(t_0) \rangle, \quad (5)$$

where $\vec{v} = (v_x, v_y)$. $Z(t)$ is a measure of the memory of the system. For uncorrelated binary collisions, $Z(t)$ is expected to decay exponentially. If $Z(t)$ decays algebraically, $Z(t) \sim t^{-\alpha}$, α needs to be larger than 1 for diffusion to be Fickian, because otherwise no valid diffusion coefficient can be calculated from the Green-Kubo formula. For 3D, the decay has been found to be algebraic with $\alpha = 1.5$ for a number of pair potentials in experiments [21–23], in simulations [7,24], and by theoretical models [25,26].

For 2D Yukawa systems exhibiting superdiffusion, Liu and Goree have found an algebraic decay with $\alpha \approx 1$ [27]. This is an indication of superdiffusive behavior.

In our simulations, we calculated the velocity autocorrelation for different trap amplitudes. To obtain accurate statistics, we performed between 10 and 50 runs of 700 000 time steps for each trap amplitude and again determined the slope of the curve in a double-logarithmic plot, this time between $t = 100\omega_p^{-1}$ and $t = 250\omega_p^{-1}$. Special attention must be paid to the error estimation. We used the jackknife method to evaluate our data, because standard error estimates may not be sufficient in this case [28,29].

III. RESULTS AND DISCUSSION

A. Harmonic confinement

We begin by noting that particles interacting via a Yukawa potential and confined by a harmonic trap support the formation of layers [17,30]. The number of layers formed depends on the temperature, the trap frequency, and the screening

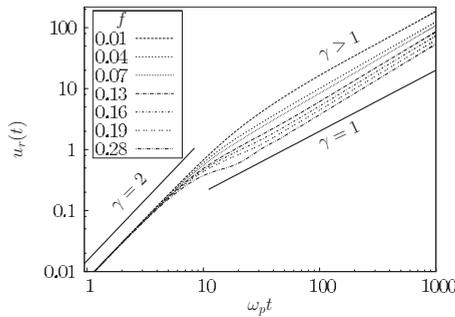


FIG. 2. The MSD over time for different trap amplitudes in the harmonic confinement and $\kappa=2.0$. The solid lines have slope 2.0 (ballistic regime) and 1.0 (normal diffusion), respectively. The slope of the MSD in this log-log plot is the diffusion exponent; see Fig. 3.

length. For our choice of parameters, we found that for $\kappa = 2.0$ a second layer builds up at $f=0.12$ while for $\kappa=3.0$ the system consists of a single layer for the whole range of f examined (cf. the insets above Figs. 3 and 4).

Typical results for the time dependence of $u_r(t)$ are shown in Fig. 2. In this double-logarithmic plot, the slope of the curves corresponds to the exponent of the algebraic behaviour. For $t < 10\omega_p^{-1}$, $u_r(t)$ grows quadratically with time, which corresponds to a ballistic motion of each individual particle. After a narrow transition region of about $10\omega_p^{-1}$, the particles' movement is dominated by diffusive processes. The slope in this region allows us to classify motion as superdiffusive, diffusive or subdiffusive; i.e., it is the diffusion exponent γ . In Fig. 2, $u_r(t)$ is depicted for different trap amplitudes f . For strong confinements, $u_r(t)$ deviates strongly from a purely diffusive behavior of $\gamma=1$. For increasingly more relaxed confinements, the slope of $u_r(t)$ tends more and more towards unity; that is, the migration becomes less superdiffusive.

The dependence of the diffusion exponent γ on the trap amplitude f is shown in Figs. 3 and 4. It is clear that the degree of superdiffusivity, as measured by the diffusion exponent γ , decreases with an increasing width of the system.

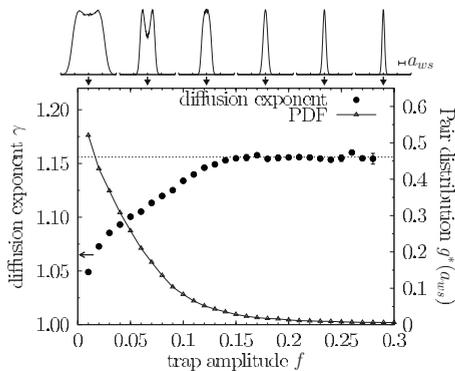


FIG. 3. The diffusion exponent for different trap amplitudes and $\kappa=2.0$ in the harmonic confinement. The straight line is a guide for the eyes. Also shown is the value of the (projected) pair correlation function at the distance $r=a_{ws}$. The top graphs show the density profile in the confined direction from $z=-4a_{ws}$ to $z=4a_{ws}$ at the trap amplitude indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude.

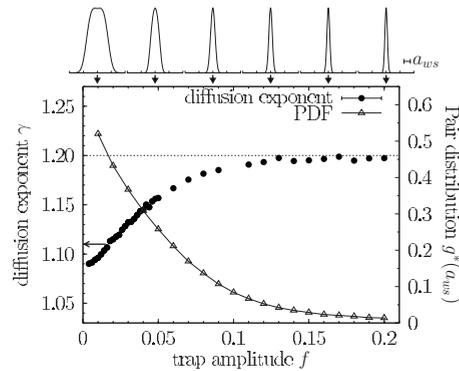


FIG. 4. The diffusion exponent for different trap amplitudes and $\kappa=3.0$ in the harmonic confinement. Also shown is the value of the (projected) pair correlation function at the distance $r=a_{ws}$. The top graphs show the density profile from $z=-4a_{ws}$ to $z=4a_{ws}$ in the confined direction at the trap amplitude indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude.

For $\kappa=2.0$, Fig. 3, the diffusion exponent is in the vicinity of $\gamma=1.16$ for strong confinements, which coincides with the value we obtained for strictly 2D systems. When the width of the particle distribution along the z axis exceeds $2a_{ws}$ (which is near the mean interparticle separation in the strictly 2D case), the diffusion exponent shows a marked drop and continues to fall for lower trap amplitudes f . The same behavior can be seen in Fig. 4 for $\kappa=3.0$. Here, γ saturates at ≈ 1.20 for strongly confined systems and again falls when the system width exceeds $2a_{ws}$. As noted before, the 3D density of the system differs for different trap amplitudes. To exclude the possibility that the vanishing of superdiffusion is only an effect of the density, we also simulated ideal 2D systems in which we changed the particle density until the first peak in the pair distribution function coincided with that of our quasi-2D systems. Our data (not shown) clearly indicate that a decreased density does not lower the diffusion exponent in our parameter range. In fact, for $\kappa=2.0$ the superdiffusion is stronger for systems of lower density.

To support the idea that the vanishing of superdiffusion is connected with the fact that particles can pass each other in the z direction, we analyze the projected pair distribution function $g^*(r)$. Its value at $r=a_{ws}$ is also shown in Figs. 3 and 4. For nearly 2D systems (high f), $g^*(a_{ws})$ is practically zero. The reason for this is that for strongly correlated liquids, the particles' kinetic energy is not sufficient for two particles to come together as close as one Wigner-Seitz radius. Instead, they are trapped in local potential minima from which they can escape only after some time. Figs. 3 and 4 show that $g^*(a_{ws})$ is nonzero for lower trap amplitudes $f \lesssim 0.1$. This is a result of the projection of all particles onto a two-dimensional plane. Individual particles are still separated by more than a_{ws} except now the finite width of the system allows the particles to go "over and under" each other in the z direction and the particles' projections can be close.

By inspection of Figs. 3 and 4, we see that the behavior of the dynamic quantity $\gamma(f)$ is closely mirrored by the static quantity $g^*(a_{ws})|_f$. Indeed, the dependence of γ on $g^*(a_{ws})$ is close to linear as shown in Fig. 5. This underlines the fact that superdiffusion is connected with the dimensionality of the system.

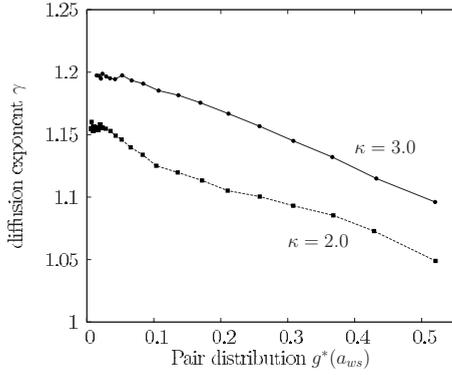


FIG. 5. The diffusion exponent γ as a function of the projected pair distribution $g^*(a_{ws})$ for $\kappa=2.0$ and $\kappa=3.0$ in the harmonic confinement.

Figure 6(a) depicts the asymptotic behavior of $Z(t)$ for different trap amplitudes f in a double-logarithmic plot. The results for the decay of the VACF are shown in Figs. 6(b) and 7 for $\kappa=2.0$ and $\kappa=3.0$.

The data shown in Fig. 6(a) validate our approach of modeling the asymptotic behavior of $Z(t)$ as an algebraic decay since $Z(t)$ closely follows a straight line in the log-log plot. The peak seen in Fig. 6(a) at long times for high f is due to our finite simulation box and is caused by sound

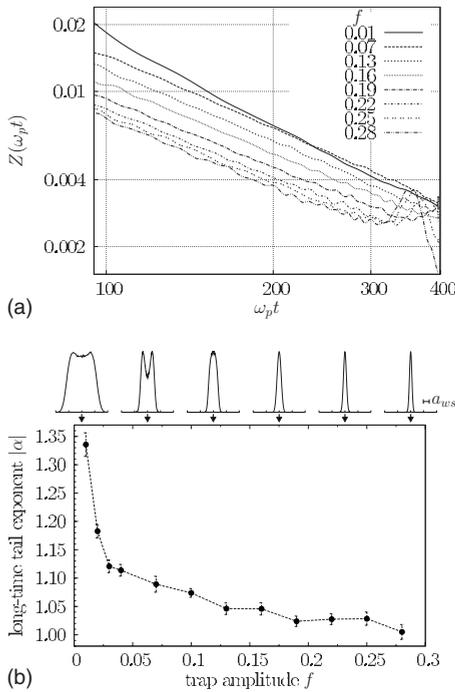


FIG. 6. (a) Double-logarithmic plot of the long-time tail of the VACF $Z(t)$ [$Z(0)=1$] from $f=0.01$ (top) to $f=0.28$ (bottom). (b) The exponent of the algebraic decay of the velocity autocorrelation function for $\kappa=2.0$ and different trap amplitudes in the harmonic confinement. Error bars denote standard errors from the jackknife estimator. The top graphs show the density profile in the confined direction from $z=-4a_{ws}$ to $z=4a_{ws}$ at the trap amplitude indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude.

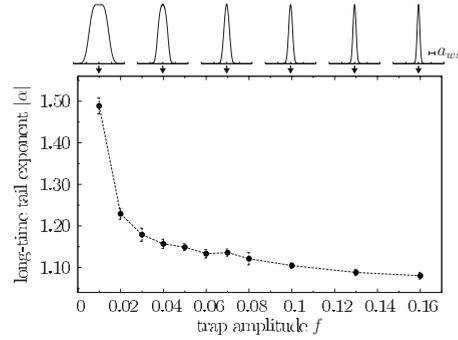


FIG. 7. The exponent of the algebraic decay of the velocity autocorrelation function for $\kappa=3.0$ and different trap amplitudes in the harmonic confinement. Error bars denote standard errors from the jackknife estimator. The top graphs show the density profile in the confined direction from $z=-4a_{ws}$ to $z=4a_{ws}$ at the trap amplitude indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude.

waves traveling through and reentering the system due to the periodic boundary conditions. Measurement is limited to times smaller than the time of the sound wave traversal. The dependence of α on the trap amplitude f , Fig. 6(b) and 7, indicates a vanishing of superdiffusion for broader systems. Starting close to the value $\alpha=1.0$ for narrow systems, we see an increase in α for broader systems. This corresponds to a faster decay, which is indicative of a loss of the particles memory; i.e., at subsequent times, a particle is less likely to travel in its original direction.

B. Soft-box confinement

We now turn our attention to the case of the soft-box confinement. Again, let us note that here, too, the system forms layers when given enough space in the confined direction. We find that the number of layers formed is higher than in the harmonic confinement, which we attribute to the con-

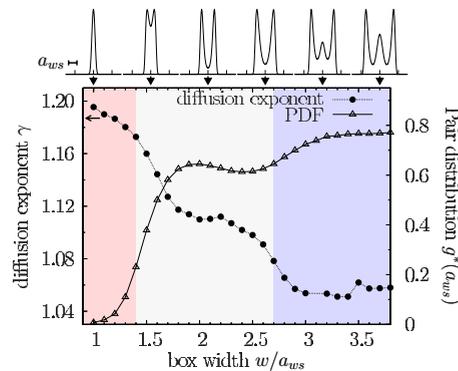


FIG. 8. (Color online) The diffusion exponent for different box widths and $\kappa=3.0$ in the soft-box confinement. Also shown is the value of the (projected) pair correlation function at the distance $r=a_{ws}$. The top graphs show the density profile in the confined direction from $z=-3a_{ws}$ to $z=3a_{ws}$ at the box width indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude. The background color separates regions of one, two, and three layers.

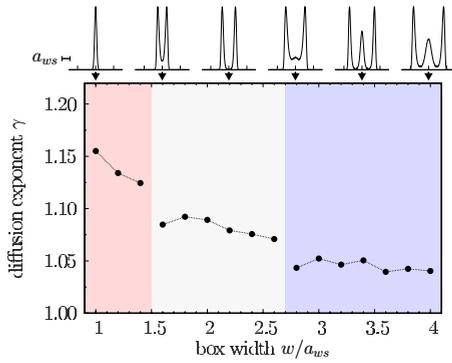


FIG. 9. (Color online) The diffusion exponent for different box widths and $\kappa=2.0$ in the soft-box confinement. The top graphs show the density profile in the confined direction from $z=-3a_{ws}$ to $z=3a_{ws}$ at the box width indicated by the arrows. The $n(z)$ distributions are normalized here to unit amplitude. The background color separates regions of one, two, and three layers.

stancy of the particle density. Recall that in this case we change the number of particles to maintain a constant 3D density.

The dependence of the diffusion on the width of the system, Figs. 8 and 9, is more involved than for the harmonic confinement. Again, the general trend is for superdiffusion to vanish for increasingly broader systems. But here, the vanishing happens in stages: After a first drop, the diffusion exponent reaches a plateau from which it drops to a second plateau. This behavior is connected to the formation of layers in the system as indicated by the different background colors in Figs. 8 and 9. (cf. top graphs in these figures). As another layer is formed in the system, the diffusion exponent experiences a drop. The nonmonotonicity of the curve in Figs. 8 and 9 is due to statistical errors.

We see that two to three layers are already sufficient to reduce the superdiffusive behavior substantially. In addition, we again notice that the value of the projected pair distribution function $g^*(a_{ws})$ is directly correlated with the diffusion exponent.

IV. SUMMARY

A study of superdiffusion in quasi-two-dimensional Yukawa liquids was performed by equilibrium molecular dy-

namics simulations. The two indicators for superdiffusion employed, the MSD and the VACF, both show sensitivity to the dimensionality of the system. For increasingly broader systems, superdiffusion gradually vanishes. The transition from superdiffusion to normal diffusion was tested for two representative values of κ and found to be qualitatively comparable. This leads us to the conclusion that the transition is universal for Yukawa systems in the fluid phase.

To ensure that the choice of confinement does not interfere with the change in dimensionality, we have used two different schemes to confine the system. The general trend of the vanishing of superdiffusion appears to be independent of the type of confinement. The finer details of how the vanishing happens depend on the choice of confinement and here especially on the formation of layers.

The strength of superdiffusion at zero width and the number of layers in the systems depend on the interparticle potential and the system parameters. At a fixed Coulomb coupling parameter $\Gamma=200$, superdiffusion is stronger for $\kappa=3.0$ and weaker for $\kappa=2.0$.

By inspection of the projected pair distribution function we have established a close connection between superdiffusion and the dimensionality of the system. The dependence of the diffusion exponent γ on the value of the projected pair distribution $g^*(a_{ws})$ was found to be approximately linear for the harmonic confinement (Fig. 5). For the soft-box confinement the general behavior is similar, although details are more complex due to the occurrence of plateaus in the curves; cf. Fig. 8.

It remains an interesting question what types of other pair potentials also support superdiffusion and how its strength depends on the interaction range. Finally, it will also be of high interest for future analysis to see how quantum effects influence superdiffusion. This could be done, e.g., by use of effective quantum pair potentials [31,32].

ACKNOWLEDGMENTS

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