Self-Diffusion in 2D Dusty Plasma Liquids: Numerical Simulation Results

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Abstract

We perform Brownian dynamics simulations for studying the self-diffusion in two-dimensional (2D) dusty plasma liquids, in terms of both mean-square displacement and velocity autocorrelation function (VAF). Super-diffusion of charged dust particles has been observed to be most significant at infinitely small damping rate $\gamma$ for intermediate coupling strength, where the long-time asymptotic behavior of VAF is found to be the product of $t^{-1}$ and $\exp (-\gamma t)$. The former represents the prediction of early theories in 2D simple liquids and the latter the VAF of a free Brownian particle. This leads to a smooth transition from super-diffusion to normal diffusion, and then to sub-diffusion with an increase of the damping rate. These results well explain the seemingly contradictory scattered in recent classical molecular dynamics simulations and experiments of dusty plasmas.

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Diffusion processes play an important role in determining dynamical properties of many physical, chemical and biological systems [1]. Diffusion in two-dimensional (2D) physical systems is of particular interest because of its significance in fundamental physics [2, 3, 4], as well as its applications in many 2D systems, such as strongly-correlated electrons on the liquid helium surface [5], colloidal suspensions [6], core-softened fluid [7], strongly coupled non-neutral plasmas [8] and, in particular, strongly coupled dusty plasmas (SCDPs) [9], which have been generally recognized as a promising model system to study many phenomena in solids, liquids and other strongly coupled Coulomb systems at the kinetic level and have been used frequently to study many physical processes in 2D systems, such as heat conduction [10], shear flow [11] and certainly diffusion of charged dust particles [12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22].

Diffusion is usually described by using particles’ mean-square displacement (MSD) and velocity autocorrelation function (VAF), defined by \( \text{MSD}(t) = \langle |\mathbf{r}_i(t) - \mathbf{r}_i(0)|^2 \rangle \) and \( Z(t) = \langle \mathbf{v}_i(t) \cdot \mathbf{v}_i(0) \rangle / \langle v_i^2(0) \rangle \), respectively. Here \( \mathbf{r}_i(t) \) and \( \mathbf{v}_i(t) \) are the position and velocity of \( i \)th particle at time \( t \), respectively, and the angular bracket denotes an ensemble average. Note that the (normal) diffusion coefficient \( D \) is determined by the MSD through the Einstein relation \( D = \lim_{t \to \infty} \text{MSD}(t)/(4t) \) and by VAF through Green-Kubo formula \( D = \int_0^\infty Z(t)dt/(2\Gamma) \). What makes things interesting in 2D systems probably lies in the fact that the diffusion processes could exhibit some anomalous behaviors. For the normal diffusion, one would expect \( \text{MSD}(t) \propto t \), and \( |Z(t)| \) decays faster than \( t^{-1} \), when \( t \to \infty \), in order to obtain a meaningful diffusion coefficient. Diffusion that do not comply with these characteristics are usually regarded as anomalous one, for example the so-called super-diffusion and sub-diffusion. In the former case (super-diffusion), one expects \( \text{MSD}(t) \propto t^{1+\alpha} \), where \( \alpha \) is positive, and \( |Z(t)| \propto t^{-1} \), when \( t \to \infty \) and in the latter (sub-diffusion), \( \alpha \) becomes negative, when \( t \to \infty \).

Early molecular dynamics (MD) simulations with 2D hard-disk liquids [2] observed an asymptotic decay of the VAF in the form that is \( \propto t^{-1} \), which led to the conclusion that particle motions in 2D hard-disk systems are non-diffusive (or super-diffusive). Later theories of 2D simple liquids [3, 4] came to the same conclusion, but with a wider argument saying that this asymptotic behavior should also exist in kinetic parts of correlation functions for the shear viscosity and heat conductivity, regardless of the liquid density and interaction force, and finally lead to the breakdown of the hydrodynamics in 2D systems.

The anomalous diffusion of dust particles in quasi-2D and 2D dusty plasma liquids (DPLs) has been frequently reported in many laboratory experiments [12, 13, 14, 15, 16]. In particular, the
super-diffusion of charged dust grains in 2D (monolayer) experiments has been recently observed by different groups \cite{13, 14, 15, 16}. Quinn and Goree \cite{13} were the first to report the observation of super-diffusion at long timescales in a 2D DPL experiment, followed by Ratynskaia et al. \cite{14, 15}, and more recently by Liu and Goree \cite{16, 17, 18}. These observations were supported by the early theoretical prediction \cite{2, 3, 4} and recent MD simulations with 2D and quasi-2D Yukawa liquids \cite{16, 17, 19, 20}.

However, seemingly contradictory observations exist in both experiments and simulations. Nunomura et al. \cite{21} investigated the self-diffusion in 2D DPLs for a wide range of the Coulomb coupling strength, and observed a normal diffusion process till the melting point, at which charged dust particles are frozen and therefore a sub-diffusion was observed. Their observation was supported by their own MD simulation results, and recent Brownian dynamics simulation\cite{22}, in which the neutral damping effect was considered and normal diffusion was observed in the whole liquid region. Yet, 2D simulations with softer interactions \cite{7} observed normal diffusive motion.

The present work is particularly motivated by these contrasting observations in recent 2D dusty plasma experiments and simulations, and is intended to clarify some of the above incompatibility, for example, the effects of the neutral gas damping and stiffness of the dust particle interaction on super-diffusion, by using extensive numerical simulations.

Our simulation is based on the Brownian dynamics (BD) method \cite{23}, which brings the simulation closer to real dusty plasma experiments, as the effects of the neutral gas damping and dust particle Brownian motions are included in a self-consistent manner. Most relevant experiments are those conducted in equilibrium conditions \cite{13, 14, 21}.

In our simulation, N = 20000 particles are placed in a square with periodical boundary conditions (PBC). A 5th-order Gear-like Predictor-Corrector algorithm \cite{24} was used to integrate the Langevin equations of every interacting Brownian dust particles. As usual, we assume here pairwise Yukawa interaction between charged dust particles: $\Phi(r) = (Q^2/r) \exp(-r/\lambda_D)$, where Q is the charge on a dust particle, r is the interparticle distance, and $\lambda_D$ is the Debye screening length. Such a system can be fully characterized by three parameters \cite{25}: the Coulomb coupling parameter $\Gamma = Q^2/(aT)$, the screening parameter $\kappa = a/\lambda$, and the damping rate $\gamma/\omega_{pd}$ due to the neutral gas, where $T$ is the system temperature (in energy units), $a = (\pi n)^{-1/2}$ the Wigner-Seitz radius, and $n$ the equilibrium dust number density. The dusty plasma frequency is $\omega_{pd} = [2Q^2/(ma^3)]^{1/2}$, where $m$ is the dust particle mass. The system size is approximately $250a \times 250a$, which together with PBC and the dust-acoustic speed \cite{26} decides our maximum independent observation time to
be $320\omega_{pd}^{-1}$.

Our analysis will be based on MSD and VAF. For a better visualization of different types of diffusion processes, we have used throughout this paper the quantity $\text{MSD}(t)/t$, rather than $\text{MSD}(t)$ itself. Let us firstly see the evolution of MSD in the zero damping limit. Figure 1 displays $\text{MSD}(t)/t$ for $\kappa = 1.0$, $\gamma/\omega_{pd} = 0.001$, but for different $\Gamma$, which covers from nonideal gaseous state ($\Gamma = 1$) to the melting point ($\Gamma = 180$) [26]. Shown together in this figure are linear fits (in log-log scale) of their asymptotic behaviors at short and long time limits, respectively. The former gives a constant value of $\alpha = 1$, indicating the ballistic motion, whereas the latter varies for different $\Gamma$, as depicted by the values above or below the corresponding dash lines. The variation of $\alpha$ with $\Gamma$ at long time limit shows clearly a hill shape, with a flat peak of $\alpha$ in the range of $\Gamma \in [50, 80]$ and a peak value of $\alpha \approx 0.18$. This behavior indicates a clear transition from normal diffusion $\rightarrow$ super-diffusion $\rightarrow$ normal diffusion, when $\Gamma$ goes from 1 to 180. Super-diffusion is most significant at the intermediate Coulomb coupling strength. The general tendency agrees qualitatively with previous MD simulations of Ott et al. (in the 2D limit)
FIG. 2: The velocity autocorrelation function for $\kappa = 1.0$, $\gamma/\omega_{pd} = 0.001$ and different $\Gamma$. The decay of $t^{-1}$ and $t^{-1.5}$ are also shown for comparison. Dash-line follows $t^{-1} \exp(-\gamma t)$.

[19] and Liu and Goree [18], except that in the latter the peak appears around $\Gamma = 20$ with a value of $\alpha \approx 0.3$. This discrepancy could have been caused by the limited observation timescale (which then was determined by the finite size of the system.) in Ref. [18]. Their latest simulations [16] with substantially more particles gave a peak value very close to ours. In addition, the diffusion coefficients calculated by using the Green-Kubo formula were also shown as circles for $\Gamma$ with a normal diffusive behavior. Their horizontal location indicates the integration limits. We found a very close agreement on the diffusion coefficient by using these two methods, which proves the consistence of our simulation.

Figure 2 depicts the VAFs (their absolutes) for different $\Gamma$ in the same conditions as Fig. 1. Shown together in this figure are different decay rates following $t^{-1.5}$, $t^{-1}$ and $t^{-1} \exp(-\gamma t)$, respectively. One observes a continuous transition from a fast decay at $\Gamma = 1$ (faster than $t^{-1.5}$) to a slow decay at $\Gamma = 50$ approximately following $t^{-1}$, indicating the occurrence of the super-diffusion. The last three tails all follow roughly $t^{-1}$ decay, however, they seem to be better fitted by $t^{-1} \exp(-\gamma t)$, as shown by dash lines in the figure, with the exponential part coming from the VAF of a free Brownian dust particle. Since the exponential factor obviously suppresses the super-diffusion, what we observe here is a super-diffusion in a transient timescale. In other words,
there should be no absolute super-diffusion (in a sense of infinitely long time limit) in a damped system in the equilibrium state. However, for this specific damping rate $\gamma/\omega_{pd} = 0.001$, its effect is negligibly small at our maximum timescale. One would need a timescale of order $10^3\omega_{pd}^{-1}$, which then require about one million dust particles, to observe a complete suppression of the super-diffusion in this case. Nevertheless, this tendency will be clearer next when we vary the damping rate.

Figure 3 displays the VAF for $\kappa = 1.0$ and $\Gamma = 10$ but different $\gamma$, as a typical example exhibiting the damping effect on the long time tails of the VAF. The long time decay of all VAFs can be well fitted in the form of $t^{-1}\exp(-\gamma t)$ up to about $\gamma/\omega_{pd} = 0.1$. For higher damping rates, the VAF reaches the noise level (about $10^{-3}$) too fast, so that no reliable fits can be applied. It should also be noted that a damping as small as $\gamma/\omega_{pd} = 0.01$ is sufficient to bring the decay close to $t^{-1.5}$ in our timescale and to suppress the super-diffusion. Since the typical value in similar dusty plasma experiments [14, 16, 21] is about a few percent of $\omega_{pd}$, we would expect that there should be no super-diffusion in the equilibrium state. This explains well the absence of the super-diffusion in the experiment of Nunomura et al. [21].

Figure 4 exhibits the variation of $\text{MSD}(t)/t$ for $\kappa = 1.0$ but for different $\Gamma$ and $\gamma$ values. When
FIG. 4: MSD(t)/t (normalized by \( \omega_{pd}a^2 \)) for \( \kappa = 1.0 \), but different \( \Gamma \) and damping rates (normalized by \( \omega_{pd} \)). Circles are corresponding to diffusion coefficients obtained through the Green-Kubo formula.

\( \gamma \) goes from infinitely small to a finite value, another transition from the super-diffusion \( \rightarrow \) normal diffusion \( \rightarrow \) sub-diffusion is clearly seen at long timescale especially from panels with higher \( \Gamma \), which implies an enhancement of the caging effect due to the neutral damping. This explains the sub-diffusion close to the melting point observed in the experiment of Nunomura et al. [21].

Circles in Fig. 4 correspond to the diffusion coefficients \( D \) obtained through the Green-Kubo formula, which agree with those from MSD (last points of MSD(t)/t) very well. Relative big discrepancies are found for the highest Coulomb coupling strength \( \Gamma = 150 \) and highest damping rates \( \gamma/\omega_{pd} = 0.5 \) and 1.0, which implies that our timescale is not long enough to reach the steady value.

One sees from both circles and long time behavior of MSD(t)/t that \( D \) decreases with the increase of the damping rate. This is physically clear because the damping effect retards the dynamics of charged dust particles. This tendency also agrees qualitatively with observations in 3D BD simulations [22, 27] and in experiments [25] of dusty plasma liquids, in which \( D \) was
FIG. 5: MSD(t)/t (normalized by $\omega_{pd} a^2$) for $\Gamma_{\text{eff}} = f(\kappa)\Gamma = 50$ ($f(\kappa)$ is given in Ref. [26]), $\gamma/\omega_{pd} = 0.001$ and different $\kappa$. The inserted figure shows exponential $\alpha$ obtained by fitting at long time limit for different $\Gamma_{\text{eff}}$, with other conditions the same. The uncertainties in the fits are $\pm 13\%$ of the shown values for $\Gamma_{\text{eff}} = 10$ and $\pm 8\%$ for $\Gamma_{\text{eff}} = 50$ and 100, respectively.

found to decay as $1/(1+\gamma/\omega_{pd})$ with the damping rate, in a certain range of the Coulomb coupling parameter [22, 25, 27].

Finally, we have investigated the effect of stiffness of dust particle interactions on the diffusion processes. Here we vary the screening parameter $\kappa$, while keeping the effective Coulomb coupling parameter $\Gamma_{\text{eff}} = f(\kappa)\Gamma$ constant, where the universal scaling function $f(\kappa)$ is obtained by fitting the MD simulation results [26]. Figure 5 shows the variations of MSD for $\Gamma_{\text{eff}} = 50$, $\gamma/\omega_{pd} = 0.001$, but for different $\kappa$ values, as examples. It is seen that the dust particle motion at this coupling strength (and timescale, to be more precisely) is super-diffusive. When $\kappa$ increases, the interaction becomes stiffer and stiffer and more and more like a hard-disk system. During this course, one observes that the migration of dust particles becomes weaker and weaker due to the decrease of the temperature, except at the highest $\kappa$, where the diffusion is enhanced shortly after the ballistic regime. The effect on the super-diffusion seems complicated, as is shown (the inserted figure) by the variation of exponential $\alpha$ at long time limit for different $\Gamma_{\text{eff}}$ values. Varying $\kappa$ has
little effect for $\Gamma_{\text{eff}} = 10$, as the $\alpha$ rises slightly from 0.13 at $\kappa = 0.5$ to 0.138 at $\kappa = 4.0$. For $\Gamma_{\text{eff}} = 50$ and 100, the largest value of $\alpha$ was observed at highest $\kappa$, indicating the most significant super-diffusion. The observation seems contradictory with the theoretical prediction of Ernst et al. [3], in which they predicted that the super-diffusion motion should exist in all 2D liquids regardless of the interaction force, but in accordance with a previous speculation based on many computer simulations that the super-diffusion might be more significant in systems with stiffer interactions. However, we did not observe a monotonous increase of $\alpha$ with an increase of $\kappa$ (hence stiffness of interactions), which indicates that dependence of the super-diffusion on the interaction force might be much more complicated than one would have thought.

In summary, we have studied the self-diffusion processes in 2D dusty plasma liquids by using the Brownian dynamics simulation. The latter shows, for the first time, the transition from super-diffusion to normal diffusion then to sub-diffusion with the increase of the damping rate. In particular, our observations suggest that there should be no real super-diffusion at long time limit in a 2D dusty plasma liquids in the equilibrium state, such as the experimental observations by Nunomura et al. [21]. Most of other experiments of super-diffusion were made in systems that were not in a strict equilibrium state and therefore collective excitations generally cannot be ruled out. This applies to the experiments of Ratynskaia et al. [14,15], where super-diffusion was observed in a viscoelastic vortical dust fluid, and to the experiments of Liu and Goree [16,17], which were made in a driven dissipative system. Our simulation results, therefore, may stimulate further experimental investigations.

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