



Anomalous and non-Gaussian diffusion in Hertzian spheres

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HIGHLIGHTS

- Dynamic heterogeneity is increased with temperature.
- Decoupling between τ_α and τ_{max} .
- Anomalous and non-Gaussian diffusion is associated with weakly correlated mean-field behavior.
- At extremely high temperatures, $\tau_{max} \sim 1/\rho$.

ARTICLE INFO

Article history:

Received 1 December 2017

Received in revised form 16 March 2018

Available online 27 March 2018

Keywords:

Non-Gaussian diffusion

Simulation

Hertzian spheres

Ultrasoft particles

Dynamic heterogeneity

ABSTRACT

By means of molecular dynamics simulations, we study the non-Gaussian diffusion in the fluid of Hertzian spheres. The time dependent non-Gaussian parameter, as an indicator of the dynamic heterogeneity, is increased with the increasing of temperature. When the temperature is high enough, the dynamic heterogeneity becomes very significant, and it seems counterintuitive that the maximum of non-Gaussian parameter and the position of its peak decrease monotonically with the increasing of density. By fitting the curves of self intermediate scattering function, we find that the character relaxation time τ_α is surprisingly not coupled with the time τ_{max} where the non-Gaussian parameter reaches to a maximum. The intriguing features of non-Gaussian diffusion at high enough temperatures can be associated with the weakly correlated mean-field behavior of Hertzian spheres. Especially the time τ_{max} is nearly inversely proportional to the density at extremely high temperatures.

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1. Introduction

The diffusion is fundamental in many fields such as biology, physics, chemistry and material science. As an important process of fluids, the diffusion can be generally described using the macroscopic law known as Fick's law [1]. The displacements of particles follow random walks to find a Gaussian function for the self van Hove distribution. And the mean square displacement (MSD) is proportional to the time t which can be written as the well-known Einstein relation $\langle \Delta r^2(t) \rangle = 2dD_f t$, where d is the dimension and D_f is the self diffusion coefficient. However, a real diffusion process is not that ideal and previous observations on many systems including monodisperse and polydisperse fluids have shown that the self van Hove distribution often deviates from Gaussian shape [2–20]. Different from normal Brownian diffusion, there is a kind of so-called anomalous diffusion whose MSD exhibits a non-linear dependence on the time t [21]. Even when the

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diffusion in a few fluids is seeming Fickian where MSD is a linear function of the time t , yet the self van Hove distribution is ever observed to be not Gaussian unexpectedly [15–20].

Recently the ultrasoft particles, which include emulsions, soft colloids, microgels, many macromolecules and their self-assembled entities, have attracted a lot of interests. The repulsive interactions of ultrasoft particles are often very soft or core-softened. Among those models of ultrasoft particles, there are an extreme class of repulsive potentials that are bounded, i.e., they remain finite for the whole range of interparticle separations, even with full overlap between the particles. Many works have suggested that ultrasoft particles with bounded potential exhibit intriguing behaviors [22–36]. Specifically it has been proposed over last decades that the diffusion of fluids with bounded interactions have rather different characters. For instance, the self-diffusivity is shown to be anomalous and there is relevant scaling relation between dynamic and thermodynamic behaviors [25–29]. Despite a detailed knowledge of the dynamic and thermodynamic anomalies, there is lack of the studies on the non-Gaussian diffusion of fluids with bounded potential. Therefore the issues that whether a non-Gaussian diffusion exists in such ultrasoft particles and what its character looks like are still open up to now.

2. Model and simulation methodology

In this work we consider the ultrasoft particles interacted with Hertz potential which is written by

$$U(r_{ij}) = \begin{cases} \frac{\epsilon}{\alpha}(1 - r_{ij}/\sigma)^\alpha, & r_{ij} < \sigma \\ 0, & r_{ij} \geq \sigma \end{cases}, \quad (1)$$

where r_{ij} is the inter-particle separation between i th and j th particle, and the parameter ϵ and σ set the strength and maximum distance of the interaction. Here we will study the non-Gaussian diffusion of Hertzian spheres, so we take the parameter $\alpha = 2.5$. The model of Hertzian spheres, which is initially describes the change in the elastic energy of two deformable objects when subjected to an axial compression, has also been proposed to represent the interaction of deformable soft colloids in a number of experimental studies [37–40].

The method used in this work is molecular dynamics (MD) simulation. In reduced units, the parameters ϵ , σ and the mass of particle m are set as 1. The periodic boundary conditions are applied and the equation of motion is integrated using velocity Verlet algorithm [41,42] with the time step $\delta t = 0.01$. The ensemble for MD simulations is canonical ensemble where the number of particles $N = 1000$ and the constant temperature is controlled via Berendsen thermostat [43]. The behavior of diffusion is observed above the maximum freezing temperature $T_m = 3.536 \times 10^{-3}$ of Hertzian spheres [25]. Notice here that we have performed 1000 independent MD simulations starting from different initial conditions for each state point to get good statistics when calculating desired parameters.

3. Results and discussion

In order to investigate the influence of temperature on the non-Gaussian diffusion, we fix the number density ρ and increase the temperature from slightly above T_m to a very high value. The deviation of particle's motion from Gaussian behavior can be qualified by a time dependent non-Gaussian parameter which is calculated by

$$\alpha_2(t) = \frac{3\langle \Delta r^4(t) \rangle}{5\langle \Delta r^2(t) \rangle^2} - 1, \quad (2)$$

where $\Delta r(t) = |\mathbf{r}(t) - \mathbf{r}(0)|$ is the distance between the position of a particle at time t and its original position. Actually the non-Gaussian parameter has been suggested to be an indicator of the dynamic heterogeneity of fluids [44]. The top panel of Fig. 1 displays the curves of non-Gaussian parameter $\alpha_2(t)$ against time t at a set of temperatures for $\rho = 1.0$. Upon increasing the temperature, the maximum of $\alpha_2(t)$ becomes increasing to indicate a more and more significant non-Gaussian behavior (see the bottom panel of Fig. 1). So we should increase the temperature to a high enough value if we want to see an apparent dynamic heterogeneity of Hertzian spheres.

It is also fundamental to calculate the self intermediate scattering function for a good understanding of liquid dynamics. The self intermediate scattering function is given by

$$F_s(q, t) = \left\langle \frac{1}{N} \sum_i \exp(-i\mathbf{q} \cdot (\mathbf{r}_i(t) - \mathbf{r}_i(0))) \right\rangle, \quad (3)$$

where \mathbf{q} is the wave vector. For convenience, we take the modulus of the wave vector $q = 1.0$. Notice here that q value is just chosen arbitrarily. In principle q can also be other values that will lead to some deviations, but the conclusions do not change substantially. The lines in Fig. 2 show the decay of $F_s(q, t)$ which can be fitted using a stretched exponential function $F_s(q, t) = \exp(-(t/\tau_\alpha)^\beta)$. The fitting parameter τ_α corresponds to the character relaxation time of fluid. In a normal fluid, the peak of $\alpha_2(t)$ is correlated strongly to τ_α , i.e., the time τ_{max} where $\alpha_2(t)$ reaches a maximum is coupled to the character relaxation time τ_α . But for Hertzian spheres the case is rather different because τ_{max} increases while τ_α decreases with the increasing of temperature (see Fig. 3). Such an unexpected behavior of the decoupling between τ_{max} and τ_α is another interesting character of the fluid dynamics in ultrasoft particles, which is observed for the first time and needs a further study for the reason why it happens.

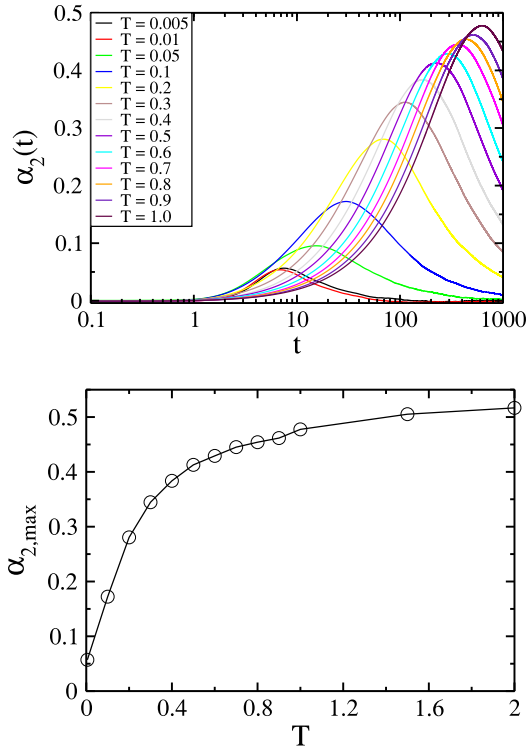


Fig. 1. Top: Curves of non-Gaussian parameter $\alpha_2(t)$ versus time t for the fixed density $\rho = 1.0$ and different temperatures. Bottom: Maximum of $\alpha_2(t)$ as a function of the temperature T at $\rho = 1.0$.

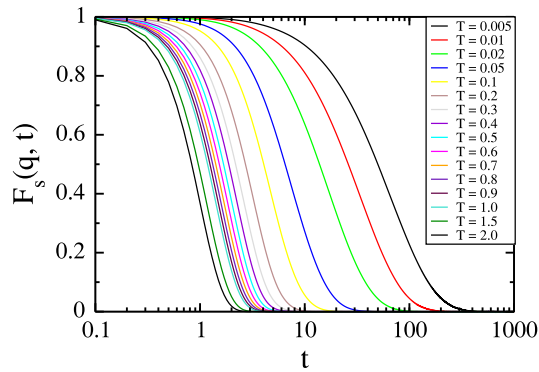


Fig. 2. Curves of self intermediate scattering function $F_s(q, t)$ for the fixed density $\rho = 1.0$ and different temperatures. The modulus of the wave vector is taken as $q = 1.0$.

Now let us see the temporal evolution of MSD during the motion of particles, which is defined by

$$\langle \Delta r^2(t) \rangle = \frac{1}{N} \sum_i (\mathbf{r}_i(t) - \mathbf{r}_i(0))^2 . \tag{4}$$

It can be assumed that $\langle \Delta r^2(t) \rangle \sim t^\nu$, so the effective exponent ν is calculated by

$$\nu = \frac{d(\log \langle \Delta r^2(t) \rangle)}{d(\log t)} . \tag{5}$$

As is known, $\nu = 1$ corresponds to a normal diffusive motion. For the anomalous diffusion of $\nu \neq 1$, the exponent ν determines whether the process belongs to subdiffusion ($0 < \nu < 1$) or superdiffusion ($1 < \nu \leq 2$) [21]. Specifically $\nu = 2$ corresponds to a ballistic motion. Seen from Fig. 4, there is no subdiffusive process like some glassformers (e.g., Ref. [33,44]) but apparently the crossover between ballistic limit and normal diffusive motion happens, which is hardly surprising

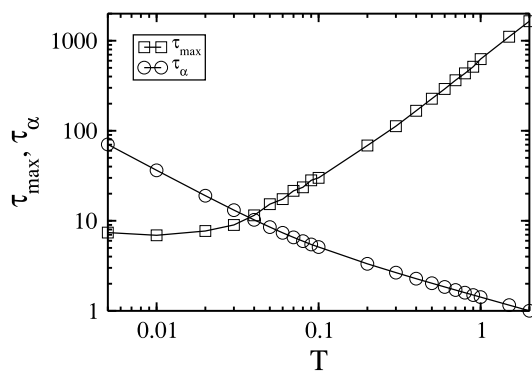


Fig. 3. Comparison between the curves of τ_{max} (the time when $\alpha_2(t)$ reaches a maximum) and τ_α (character relaxation time obtained from the fitting of self intermediate scattering function) versus the temperature T . The density is fixed $\rho = 1.0$. Squares and circles represent τ_{max} and τ_α respectively. Lines are guide to the eye.

because it has been commonly observed in many other kinds of fluids [4,6,15,17,24,33,44]. Therefore we can say that the dynamic heterogeneity appearing in Hertzian spheres is not due to the subdiffusion but other possible reasons related to the superdiffusion (see Fig. 4, $1 < \nu \leq 2$). In the fluid, particles diffuse via ballistic motions with few collisions and enter the diffusive regime with $MSD \sim t$ at long time scales. In the bottom panel of Fig. 4, we present the time dependence of ν at different temperatures. It appears to indicate that increasing temperature makes the crossover time from ballistic to diffusive motion shift to longer times. This is actually a natural result for the ultrasoft particles such as Hertzian spheres. As the temperature is increased, the thermal energy becomes more and more significant and can eventually dominate over the repulsive potential so that those particles can penetrate or even overlap each other. Thus for a single particle at high enough temperature, it seems to be able to almost freely slide in the fluid. When the temperature is low the thermal energy is not dominant over the repulsive potential any more, so the particle in fluid is influenced remarkably by the surrounding repulsions and especially at low enough densities Hertzian spheres bear some similarities of the fluids with harder repulsion.

As is mentioned above, the dynamic heterogeneity of Hertzian spheres becomes significant at high enough temperature. Next we set a temperature $T = 0.5$ where the non-Gaussian behavior is apparent (see Fig. 1) and present a further study on the effect of density. Fig. 5 displays the curves of non-Gaussian parameter $\alpha_2(t)$ against time t at several densities for $T = 0.5$. At such a temperatures, $\alpha_2(t)$ shows some subtle features. Firstly the value of non-Gaussian parameter $\alpha_2(t)$ is nearly zero in short time (typically $t < 1$), which is also investigated in Fig. 1 and other kinds of fluids [4,6,15,17,33]. Indeed it is natural for the system very close to ballistic regime (see also Fig. 4), because the velocity nearly has a Maxwell–Boltzmann distribution so as to make the self-part of the van Hove function be also of a Gaussian shape. Secondly, different from many other kinds of fluids (including both the monodisperse and polydisperse fluids), there is not a density independent step in $\alpha_2(t)$ before it approaches to the maximum [3,4,6,17,33]. Thirdly, the maximum value of $\alpha_2(t)$ decreases monotonically with increasing the density which is oppositely different from the non-Gaussian behavior of most of other kinds fluids where the maximum value of $\alpha_2(t)$ is an increasing function of the density [4,6,15]. Furthermore the position of the peak, i.e. the time τ_{max} where $\alpha_2(t)$ reaches its maximum, also shows a similarly monotonic dependence on the density. As is known, such a position of the peak is considered as the time scale where a single particle escapes from the cage and enters the diffusive regime. So it means that the particles on average need less and less time to enter the diffusive regime with the increasing of density. This seems counterintuitive because it is generally thought to be more difficult for a single particle in dens fluids to escape from its surrounding cage.

Similar to the things done under different temperatures (see Fig. 2), we also observe the curves of self intermediate scattering function for different densities with $T = 0.5$ to find all of the curves of $F_s(q, t)$ almost overlap each other which leads to the result that the character relaxation time τ_α for $T = 0.5$ is hardly dependent on the density (see Fig. 6). Although the decoupling between τ_{max} and τ_α is expected as the observations of Fig. 3, the independence of character relaxation time obtained on the density seems to be incredible at first glance. Nonetheless the behavior of τ_α , as a matter of fact, has given a hint of the characteristic structure of Hertzian spheres. Indeed the strange non-Gaussian behavior observed at high temperatures can be attributed to the fact that Hertzian spheres behave as a weakly correlated mean-field fluid. Such a weakly correlated mean-field behavior has been found in the fluids of ultrasoft particles, e.g. the Gaussian core model (GCM) [22,45] and harmonic spheres [29]. Here the decoupling between τ_{max} and τ_α can be explained as follows. The system of Hertzian spheres exhibits a character of normal fluid at low temperatures, but it approaches to a weakly correlated mean-field fluid with the increasing of temperature where τ_{max} behaves strangely (see Fig. 6). For τ_α , the case is different because the particles can move more and more freely in the fluid making τ_α decreased as the temperature is increased (see Fig. 3). When the fluid of ultrasoft particles exhibits a weakly correlated mean-field behavior, most particles are distributed uniformly acting as the ideal gas and a few small clusters are separated from each other resulting in an approximate independence of τ_α on the density.

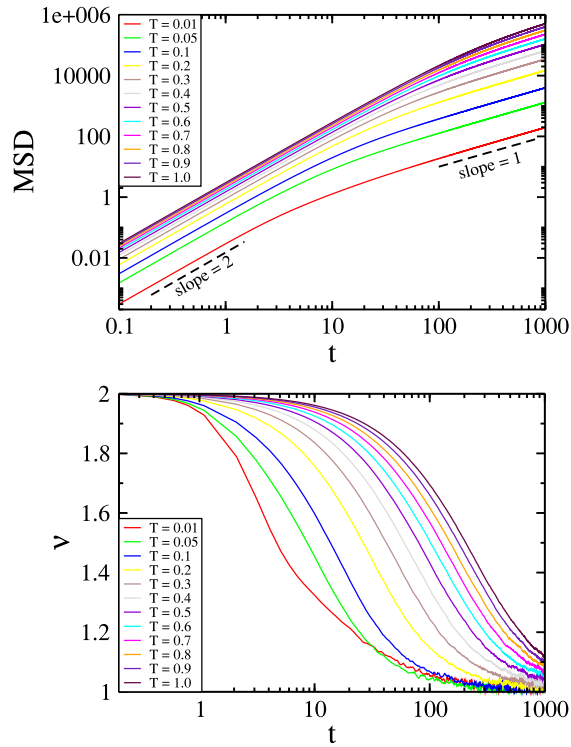


Fig. 4. Curves of MSD (top) and effective exponent ν (bottom) versus time t for the fixed $\rho = 1.0$ and different temperatures. Assume that the MSD $\langle \Delta r^2(t) \rangle \sim t^\nu$.

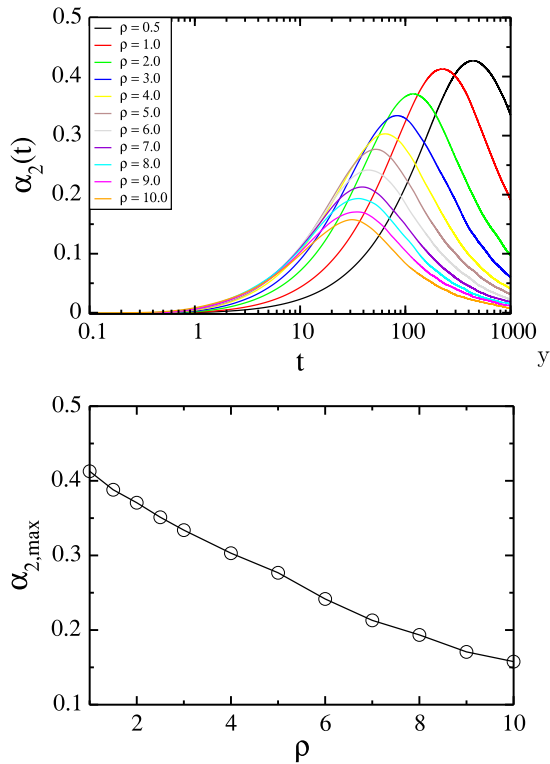


Fig. 5. Top: Non-Gaussian parameter $\alpha_2(t)$ versus time t . Bottom: Maximum of $\alpha_2(t)$ as a function of ρ . The temperature is taken as $T = 0.5$.

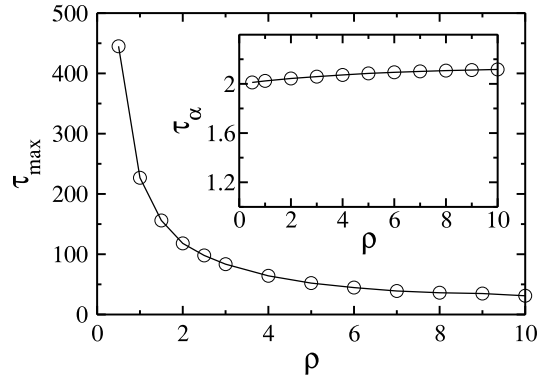


Fig. 6. Dependence of τ_{max} (the time when $\alpha_2(t)$ reaches a maximum) on the density ρ for $T = 0.5$. The inset shows the curve of τ_{α} (character relaxation time obtained from the fitting of self intermediate scattering function).

As the characteristic weakly correlated mean-field fluid consists of ideal-gas-like particles among which a particle can freely slide and a few separated clusters that might form cages, a single particle's dynamic is determined by the character of the clusters. Let us firstly make an inspection on the structure of each cluster. The particles in each cluster are close to or even sit on top of each other. Although the separation distance between clusters is nearly independent on the density [29], the correlation of particles inside the clusters may be different under different density. On the other hand, when the correlation of Hertzian spheres is strong, the sliding of a particle in the fluid is expected to be less smooth so that its superdiffusion behavior is weaker and will take more time to enter the diffusive regime. Seen from the radial distribution function shown in the top panel of Fig. 7, the “soft” correlation hole is gradually reduced as the density ρ increases. In the high density limit, it can be imagined that $g(r)$ approaches to 1 even for overlaps which corresponds to ideal-gas-like behavior. Using Hypernetted-chain equation, a closure relation for solving the Ornstein–Zernike equation, we can estimate the direct correlation function

$$c(r) = -\beta U(r) + g(r) - 1 - \ln(g(r)), \quad (6)$$

where $\beta = 1/k_B T$. Similar to the structure of Gaussian core fluid which has been shown to exhibit weakly correlated mean-field behavior over a wide density and temperature range [22], the direct correlation function of Hertzian spheres specifically for the particles in the clusters also decreases with the increasing of density (see the bottom panel of Fig. 7) so as to make a single particle under higher densities escape slightly easier from the clusters.

For a further study, we have investigated the non-Gaussian diffusion and meanwhile estimated the time τ_{max} where $\alpha_2(t)$ reaches to a maximum under different high enough temperatures. As is shown in Fig. 8, the curves of τ_{max} versus the density ρ are plotted for a set of temperatures. The system, as has been mentioned already, is considered to be weakly correlated mean-field fluid from $T = 0.2$ to $T = 1.0$. Seen from Fig. 8, the curves of τ_{max} versus ρ can be approximately fitted by a power-law function $\tau_{max} \sim \rho^\gamma$. For $T < 1.0$, the exponent γ is slightly larger than -1.0 as the correlation between particles has a little bit influence on the non-Gaussian diffusion. For $T \geq 1.0$, the exponent γ is nearly equal to -1.0 . Such an investigation of $\tau_{max} \sim 1/\rho$ is actually reasonable for a very weakly mean-field fluid at extremely high temperature where the effect of correlation between particles on τ_{max} can be neglected. As a single particle in the very weakly mean-field fluid can freely slide, the time τ_{max} is expected to be completely determined by the size of cages that is proportional to $1/\rho$.

4. Conclusion

In conclusion, we study the non-Gaussian behavior of monodisperse Hertzian spheres above its freezing point via molecular dynamics simulations. Hertzian spheres, as a kind of ultrasoft particles, have some similar characters of single particle dynamics to the other kinds of fluids with hard repulsion. For instance, the single particle dynamics also exhibits a crossover from ballistic ($\langle \Delta r^2(t) \rangle \sim t^2$) to diffusive behavior ($\langle \Delta r^2(t) \rangle \sim t$) after some time that is dependent on both the temperature and density. And the measure $\alpha_2(t)$ of non-Gaussian behavior is shown to be nearly zero at short time scale suggesting the self van Hove function is Gaussian shape. However, the most interesting finding of this work is that the fluid of Hertzian spheres has some intriguingly different dynamics behaviors, especially the impressive feature of its non-Gaussian diffusion. When the temperature is increased to high enough, the particle's diffusive motion becomes to deviate significantly from Gaussian behavior, and the peak in $\alpha_2(t)$ abnormally shows a monotonic decrease with the increasing of density. After observing the self intermediate scattering function, we should say that there is surprisingly no coupling between the character relaxation time τ_{α} and the time τ_{max} where $\alpha_2(t)$ reaches to a maximum. The non-Gaussian behaviors at high enough temperatures, which is rather different from many kinds of fluids, can be attributed to the fact that Hertzian spheres exhibit the character of weakly mean-field fluid. Specifically for extremely high temperatures where the influence of the correlation between particles is considered to be negligible, the time τ_{max} is nearly inversely proportional to the density.

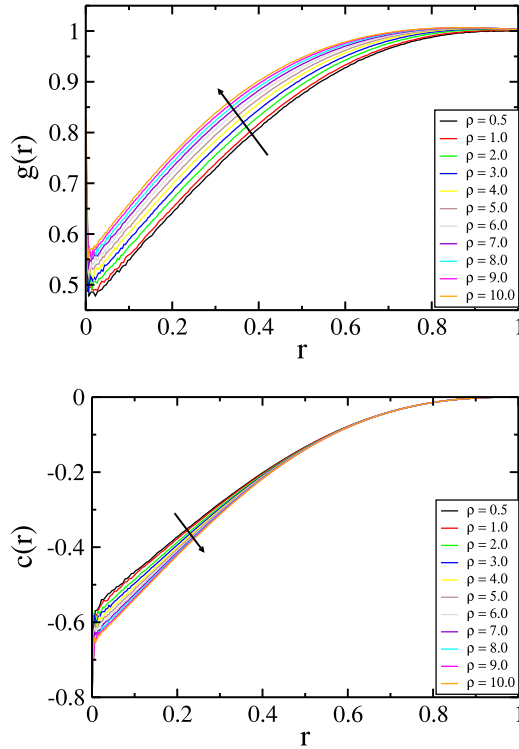


Fig. 7. Radial distribution function $g(r)$ (top) and direct correlation function $c(r)$ (bottom) for different densities at $T = 0.5$. The arrows indicate increasing the density ρ .

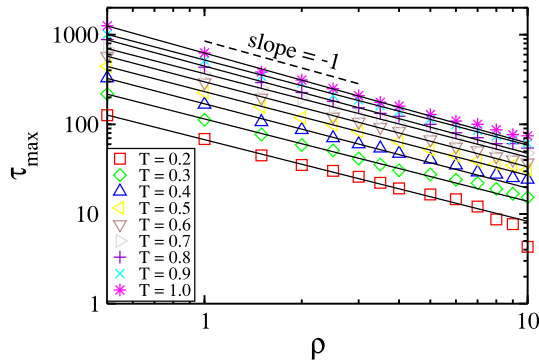


Fig. 8. Dependence of τ_{max} where the non-Gaussian parameter $\alpha_2(t)$ has a maximum value on the density ρ at a set of high enough temperatures. From the bottom to the top, $T = 0.2 - 1.0$. The solid lines represent the curves of fitting power-law function $\tau_{max} \sim \rho^\nu$. The dashed line is the guide line with scaling factor of -1 .

We would like to mention finally that the characters proposed above may probably apply to other core-softened fluids especially the system with generalized Hertz potential (set other exponential parameters α in Eq. (1)). For the GCM fluid that also exhibits such a mean-field behavior over a surprising wide density and temperature range [22,45], it is possible that a similar or even more significant non-Gaussian diffusion will be observed.

Acknowledgment

This work is supported by Grants Nos. 11572322 and 11172302 from the National Natural Science Foundation of China.

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