1 Introduction

It has been known that most liquids become denser upon cooling and more viscous upon compression. However, water and some other liquids with tetrahedrality such as silica, silicon, carbon, and phosphorus have been found to exhibit anomalous behavior close to their freezing lines, i.e., their phase diagrams have regions where the thermal expansion coefficient is negative, self-diffusivity increases upon compression, and the structural order of the system decreases with an increase in temperature. Jagla showed in the pioneering paper that these anomalies are true because the ultrasoft particles tend to avoid overlapping and behave like effective hard spheres. Interestingly, the plots of $D$ versus two-body excess entropy $S_2$ approximately collapse onto a single master curve despite the structural and dynamical anomalies, which has also been observed in other core-softened fluids with bounded potential.

The interactions between particles of simple atomic and molecular liquids are generally steeply repulsive when the particles get more and more close to each other. But soft particles, such as macromolecules and their self-assembled entities, emulsions, soft colloids, grains etc., have softer effective pair interactions. As the shape of the pair potential often determines the phase behavior of the system, one can expect that the phase behavior of soft particles will have a wide difference from that of hard ones. Some types of soft particles may elastically deform and even mutually interpenetrate when colliding with each other, so their phase behaviors become more complex. Actually, it does not necessarily require a singular repulsion to make interparticle separations vanish in some systems with core-softened particles. In this respect, it is interesting to consider an extreme class of repulsive potentials that are bounded, i.e., they remain finite for the whole range of interparticle separations, even at a full overlap between the particles. Many previous studies have shown that the systems with bounded potential have rich and complex phase behaviors. Among those phase behaviors, the anomalies of the systems with core-softened particles have attracted much interest in recent years.

To fully characterize the thermodynamic and dynamic behavior among the studies of liquids, the connection between the diffusion coefficient $D$ and the excess entropy $S_{ex}$ is of fundamental interest. It not only provides a clue to the long-standing puzzle concerning how the structural and thermodynamic properties correlate with the dynamics of fluids, but also has practical consequences. For instance, if the diffusion coefficient can be approximately represented as a single-valued function of the excess entropy, then the knowledge of the latter allows indirect prediction of the former. In 1977, Rosenfeld first proposed that the reduced diffusion coefficient can be connected to the excess entropy of the system through the formula:

$$D_R = A \exp(B S_{ex}),$$

where $A$ and $B$ are the constants which interestingly show an extremely weak dependence on the material and can be considered universal. The excess entropy $S_{ex}$ which refers to the difference between the entropy per particle of the fluid and that of an ideal gas of particles with the same number density $\rho$, is a negative quantity. The diffusion coefficient is expressed in...
reduced units based on the mean length related to the density of the system \( \rho^{-1/3} \) and the thermal velocity \((k_B T/m)^{1/2}\), so that \( D_{\text{R}} = D_T \rho^{1/3} T^{-1/2} \). Afterwards, Rosenfeld further extended the quasi-universal excess-entropy scaling to the regime of dilute gas, where \(-S_{\text{ex}}\) is low and the power-law form follows from Enskog theory for \( D_T^{26} \)

\[
D_{\text{R}} = z(-S_{\text{ex}}) \beta, 
\]

where \( z \) and \( \beta \) are constants too. Although the original idea is to associate the system under investigation to the hard-sphere system, it has been demonstrated that the entropy scaling relation developed by Rosenfeld is valid in various model fluids.26–28 Here one may ask the question whether the Rosenfeld scaling relation is still valid in the fluids with ultrasoft particles which are essentially different from the hard spheres. Krekelberg et al. studied the anomalous behaviors of Gaussian-core (GC) fluids, which belong to a class of bounded potentials, showing that the Rosenfeld diffusivity \( D_{\text{R}} \) is a function of two-body excess entropy \( S_2 \) alone and that a different combination \( D_T = D_T^{1/2} \) collapses as a function of \( S_{\text{ex}} \).4 Fomin and coworkers observed that the Rosenfeld scaling formula for the fluids of GC and Hertzian spheres is valid only in the infinite-temperature limit where there are no anomalies.6 Until now, whether these observations will generally hold for other fluids with bounded potential is still an open issue and it needs a more detailed study.

In this paper, we present a further study on the anomalous behaviors of another type of core-softened fluid with bounded potential. The fluid consists of soft spheres where the harmonic repulsion between two approaching particles starts when their distance is smaller than the sum of their radii. This model, originally proposed to describe wet foams,29 already finds experimental realization in emulsions, soft colloids, and grains.30,31 Although many results regarding its phase behaviors have been presented in theories and simulations over the last few decades,13,16–19,32 its anomalous behaviors and the relevant scaling relation between thermodynamic and dynamic behavior are not known yet in detail. Furthermore, we should stress that a clear explanation of why the fluids with soft particles are different from ‘simple’ hard-sphere like fluids is still lacking until now. In order to address this point, we carry out molecular dynamics (MD) simulations in this study to investigate the structural and dynamical anomalies, as well as to analyze the entropic scaling for the system. We show that the soft particles behave like effective hard spheres only at sufficiently low temperatures and densities so as to make the Rosenfeld entropic scaling hold true. Especially the core-softened fluid exhibits an unusual configuration as the temperature is increased high enough, i.e., the behavior of the system is like a weakly correlated mean-field fluid. Due to such a weakly correlated mean-field behavior, a scaling relation between temperature scaled diffusion \( D_T \) and \( S_{\text{ex}} \) holds, and \( D_T \) is approximately inversely proportional to \(-S_{\text{ex}}\) at least for soft particles interacting through harmonic repulsions. Therefore, we believe that our original findings above may be valuable and contribute insightfully to the subject of anomalous behaviors in the fluids with ultrasoft interactions.

### 2 Model and simulation methodology

The soft particles interact through the harmonic repulsion, which is defined as

\[
U(r_{ij}) = \begin{cases} \frac{\varepsilon}{2}(1 - r_{ij}/\sigma)^2, & r_{ij} < \sigma, \\ 0, & r_{ij} \geq \sigma, \end{cases} 
\]

where \( r_{ij} \) is the pair distance between the \( i \)th particle and the \( j \)th particle. The parameters \( \varepsilon \) and \( \sigma \) govern the strength and the maximum distance of the interaction, respectively. For the soft spherical particles, the maximum distance of the interaction \( \sigma \) is equal to their diameters.

To explore the dynamic properties of the systems, we perform MD simulations in a canonical ensemble, i.e., the total number of particles \( N \), the temperature \( T \), and the volume of the simulation cell \( V \) are kept constant. For convenience, the reduced units are used in the simulations. The basic units are chosen as follows: energy unit \( \varepsilon \), length unit \( \sigma \) and the mass of particle \( m \). The periodic boundary conditions are applied. The equation of motion is integrated using the velocity Verlet algorithm33,34 with the time step \( \delta t = 0.01 \), and the constant temperature is controlled via a Berendsen thermostat.35

The diffusion coefficient \( D \) can be calculated by fitting the long time average mean-squared displacement of particles \( \langle \delta r^2 \rangle \) according to the Einstein relation

\[
6D_T = \langle \delta r^2 \rangle. 
\]

In order to investigate the relationship between the anomalous dynamic behavior and the structural order, we need to calculate the value of the two-body excess entropy defined as

\[
S_2 = -2\pi k_B \rho \int_0^{\infty} \{g(r) \ln[g(r)] - [g(r) - 1]\} r^2 dr, 
\]

where \( g(r) \) is the pair correlation function (PCF) and \( \rho \) is the number density. For a completely uncorrelated system, \( g(r) = 1 \) and \( S_2 = 0 \). For a system with a long-range order, spatial correlations persist over long distances and \( -S_{\text{ex}} \) becomes very large. Thus, \( S_2 \) can be taken as an indicator of structural order. This parameter yields information about the average relative spacing of the particles, i.e., it describes the tendency of particle pairs to adopt preferential separations.

Although the calculation of \( S_2 \) is very useful and the computation is easy, we also need to study the scaling relation between the diffusion coefficient \( D \) and the full excess entropy \( S_{\text{ex}} \) for the systems with soft particles, the difference between \( S_{\text{ex}} \) and \( S_2 \) is large, especially when the density of the particles is high where three- and higher-body terms contribute significantly to the excess entropy. Below, we describe an approach for evaluating the full excess entropy.34 Firstly the excess free energy \( F_{\text{ex}} \) is determined by integrating the pressure \( P(\rho) \) along an isotherm starting at low densities where the fluid behaves effectively as an ideal gas,

\[
F_{\text{ex}} = N \int_0^{\infty} \frac{P(\rho) - k_B T \rho}{\rho^2} d\rho. 
\]
Then the excess entropy $S_{ex}$ is obtained from the relation

$$S_{ex} = \frac{U - F_{ex}}{Nk_B T},$$

(7)

where $U$ is the internal potential energy of the system. In principle, thermodynamic integration (eqn (6)) could also be done along the temperature to get $F_{ex}$, with the advantage that using five points (i.e., five simulations at different temperatures) already provides with rather accurate results have been obtained. Here the simulations are carried out for the isotherms $T = 0.01$–0.1 with the step of 0.01 and the isotherms $T = 0.2$–1.0 with the step of 0.1. In making the thermodynamic integration, we used the simple trapezoidal rule which works rather accurately in this case. The integration is done over two density regions, from $\rho = 0.01$ to $\rho = 2.0$ with an interval of $d\rho = 0.01$ and from $\rho = 2.0$ to $\rho = 10.0$ with an interval of $d\rho = 0.04$, to ensure that the accuracy of the thermodynamic integration in both of the density regions is satisfactory.

### 3 Results and discussion

According to the phase diagram constructed via thermodynamic integration, the maximum melting temperature of the system is $T_c \approx 6.35 \times 10^{-3}$. We perform all the simulations above the melting temperature to ensure that the system is always in a fluid state. The number of particles is chosen as $N = 1000$, and the temperature is set from $T = 0.01$ to $T = 1.0$. Each simulation in the $NVT$ ensemble is carried out separately. For a typical simulation, the particles are put in the cubic box randomly and their velocities satisfy the Maxwell–Boltzmann distribution. After the system is relaxed to attain equilibrium, another run is done to calculate the average values of some parameters needed. The equilibration run is performed in $10^5$ time steps and the production run in $10^6$ time steps for accuracy.

Firstly, we investigate the dependence of self-diffusivity on density along a set of isotherms (see Fig. 1), to study the dynamic behavior of the fluid. The diffusion coefficient, which decreases monotonically with an increase in the density in normal liquids, shows a clear nonmonotonic behavior at low temperature. Compressing the low-density fluid initially decreases $D$, but the compression can eventually lead to an anomalous increase in $D$ at sufficiently high density. The density $\rho \approx 2.0$ at which the minimum in $D$ occurs as a boundary between the regions of normal and anomalous dynamic behavior can be seen in the top panel of Fig. 1. Such dynamical anomalies have also been observed in other fluids with soft particles. When the temperature is increased high enough, the diffusion coefficient starts to decay upon increasing the density and reaches an almost constant value finally (see the bottom panel of Fig. 1), indicating that the dynamical anomaly disappears.

As is already mentioned, the quantity $S_2$ is the two-body contribution to the excess entropy and effectively characterizes the degree of structural order present in the fluid. It can be conveniently computed and analyzed given its straightforward connection to PCF (see eqn (5)). Fig. 2 displays the density dependence of $S_2$ calculated from MD simulations along a series of isotherms in a density range up to $\rho = 10.0$. Like the diffusion coefficient, the two-body excess entropy $S_2$ has a minimum at low temperatures but it becomes a monotonically decreasing function of the density upon increasing the temperature high enough. Compared with the case of dynamical anomaly, the temperature where the region of structural anomaly disappears seems to be higher. Moreover, the curve obtained at $T = 0.01$ (lowest temperature) not only goes through a minimum around $\rho = 2$, but then through a maximum around $\rho = 5.5$. This behavior suggests the presence of a second minimum above $\rho = 10$, so one may assume to get back the same large-$\rho$ behavior as those obtained at all the other temperatures studied.

The quantity $S_2$ is connected with PCF, so the structural order is reflected by the formation of distinct first, second, and more distant coordination shells in $g(r)$. In Fig. 3, the PCF curves obtained for several density ranges at $T = 0.01$ are shown. Compression of the fluid initially leads to an increased height of the first peak in PCF, but further compression after a maximum makes the height of the first peak in PCF decrease. This investigation is in accord with the inspection of $S_2$ in Fig. 2. Moreover, we should mention here that such a result is also consistent with the theoretical calculation of the system’s structural anomaly via hyper-netted chain approximation of liquid state theory, in which the theory predicts an anomalous...
(nonmonotonic) evolution of the intensity of the first peak when the density is increased at constant temperature. In fact, the structural anomaly is physically a direct consequence of particle softness, which can be attributed to a competition between the energy and the entropy. At sufficiently low densities and low temperatures, the effective repulsive “core” of the soft potential is like the hard-sphere interaction to make the system behave as an effective hard-sphere fluid, so it can easily minimize energy by avoiding particle overlaps. In this regime, the structural order is increased with the increase of the density as it does in a hard-sphere fluid. Further compression of the fluid makes it harder and thus entropically less favorable to avoid particle overlaps because the repulsion between particles is too soft and the system must find more efficient ways to pack the particles. In other words, the system of soft particles pays only a finite potential-energy penalty for each overlap, while avoiding overlaps carries a substantial entropic penalty, so the system tends to evolve toward a more uniform average structure at sufficiently high density. As a result, the structural order is decreased with the compression of the system in this regime.

It is useful to locate the boundaries of structural and dynamical anomalies as the curves of $D$ and $S_2$ have been obtained. Fig. 4 shows the estimated locations of the extrema of $D$ and $S_2$ in the $T$–$\rho$ plane. Obviously, the state points which exhibit the dynamical anomaly are a subset of those linked with the structural anomaly. Such a phenomenon qualitatively agrees with the findings reported by Krekelberg et al. concerning the anomalous behaviors of Gaussian-core (GC) fluids. This qualitative agreement is not surprising because both GC and harmonic particles belong to the core-softened particles with bounded potential. However, the densities of the boundaries of structural and dynamical anomalies in the fluid with harmonic particles are much larger than those in the GC fluid, which is probably due to the different softness of particles between the two kinds of fluids. Compared with GC particles, the harmonic particles have harder repulsions so as to be able to “return” to hard spheres at a larger density.

Next, we consider the Rosenfeld scaling relation between the full excess entropy $S_{ex}$ and the diffusion coefficient $D$ for the fluid. In simple atomic fluids, $S_2$ is known to closely approximate $S_{ex}$ at low to intermediate densities. However, the fluid with soft particles has a structural behavior which is very different from that of simple atomic fluids. In fact, there are...
often significant differences between \( S_2 \) and \( S_{\text{ex}} \) for the fluids with soft particles\(^6\) due to the three- and higher-body contributions. Before the study of the Rosenfeld scaling relation between \( S_{\text{ex}} \) and the diffusivity, it would be useful to see the results of \( S_{\text{ex}} \). Fig. 5 presents the curves of \( S_{\text{ex}} \) as a function of density \( \rho \). Similar to the case of \( S_2 \), \( S_{\text{ex}} \) also shows a nonmonotonic density dependence along the isotherm at low temperatures, and it becomes a monotonically decreasing function of the density at high enough temperatures.

In Fig. 6, the dependence of the Rosenfeld reduced diffusion co-efficient \( D_R \) on the excess entropy along some isotherms is shown. Looking at the curves obtained at low temperatures \( T = 0.01-0.1 \) (see the top panel of Fig. 6), we can divide these curves into two distinct regions with different slopes which are denoted 1 and 2. The density increases in the right-to-left direction along a curve, i.e., region 2 has higher densities than region 1. Also from Fig. 5, it can be seen that \( S_{\text{ex}} \) decreases with the increase of density in the region of normal behavior; \( S_{\text{ex}} \) increases with the increase of density in the region of anomalous behavior. With the increase of temperature, region 2 becomes shorter and shorter. Already at \( T = 0.2 \), region 2 is negligibly small. As is seen from the plots at different temperatures and densities, the data for \( D_R \) versus \( -S_{\text{ex}} \) apparently do not collapse onto a single curve. However, the plots of region 1 at some low temperatures (up to \( T = 0.05 \)) seem to collapse onto the same curve (see Fig. 7). For comparison, eqn (1) and (2) with parameters that fit the simulation data for a variety of model systems \( A = 0.58, B = 0.78 \) (at high \( -S_{\text{ex}} \)) and \( \alpha = 0.37, \beta = 2/3 \) (at low \( S_{\text{ex}} \)) are also shown in Fig. 7. Apparently, the form of the relationship between the Rosenfeld reduced diffusion co-efficient and the excess entropy is virtually the same, whether one considers the soft spheres with harmonic repulsions or a model system for hard-sphere and simple atomic fluids. Notice here the volume fraction \( \phi = \frac{\pi}{6} \rho \sigma^3 < 1 \) in region 1. Therefore, such observations can be explained in the following: the fluids act like hard-sphere and simple atomic fluids at rather low temperatures and densities where both structural and dynamical behaviors are normal, so as to make the Rosenfeld scaling relation valid for the system. Furthermore, while the structural anomaly becomes less and less marked upon increasing the temperature resulting in the vanishing of region 2, the curves of \( D_R \) versus \( S_{\text{ex}} \) along different isotherms deviate significantly. In a word, the general scaling relation between \( D_R \) and \( S_{\text{ex}} \) does not work in the regions of structural anomaly and at higher temperatures, which is just like the behaviors of other fluids with soft particles.\(^{4,6,14,15}\)

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**Fig. 5** Dependence of excess entropy \( S_{\text{ex}} \) on density \( \rho \) along a set of isotherms. Top: \( T = 0.01, 0.02, 0.03, 0.04, 0.05 \). Bottom: \( T = 0.1, 0.2, 0.3, 0.4, 0.5 \). The arrows indicate an increase in temperature \( T \).

**Fig. 6** The Rosenfeld scaled diffusivity \( D_R = D_r^{1/3}T^{-1/2} \) versus the excess entropy \( -S_{\text{ex}} \) along a set of isotherms. Top: low-temperature behaviors from \( T = 0.01 \) to \( T = 0.1 \). Bottom: high-temperature behaviors from \( T = 0.2 \) to \( T = 1.0 \).
Another scaling formula for the systems with soft particles, which looks like the Rosenfeld one with density omitted, has been proposed. For instance, it was ever reported that the plots of

\[ D_T = D_T^0 C_0^{1/2} \]

versus \( S_{ex} \) approximately collapsed onto a single master curve in GC fluids. Here we also plot the data for \( D_T \) versus \( S_{ex} \) for all the temperatures and densities studied, but do not see the plots collapse onto the same curve. Nevertheless, to further analyze the data, we find that the plots of \( D_T \) versus \( S_{ex} \) eventually collapse onto a single master curve when the temperature is increased high enough (\( T \approx 0.2 \), see Fig. 8). It is still not known exactly why this alternative scaling holds for the core-softened fluids at present. Here we show that the fluid exhibits an unusual configuration. In the top panel of Fig. 9, the PCF curves obtained for some densities at rather high temperature \( T = 0.5 \) are displayed. It can be seen that there are fairly a few particles that overlapped with each other, and that the particles become uncorrelated for a long range (\( r > 1.0 \)) as \( g(r) \) approaches unity. Next, we may consider the number of particles whose centers are, on average, within a distance \( r_c \) from a given particle in the fluid. The number of particles \( n_c \) can be calculated using

\[ n_c = 1 + 4\pi\rho \int_0^{r_c} g(r)^2 dr. \]  

(8)

In the bottom panel of Fig. 9, the curves of quantity \( 4\pi g(r)^2 \) within the distance \( r = 1.0 \) for several densities at \( T = 0.5 \) are shown. There is little deviation between the curves of different densities, which suggests that \( n_c \) is nearly a linear function of \( \rho \) and the particles are distributed almost uniformly. Therefore the system behaves as a weakly correlated mean-field fluid looking like an ideal gas. However, such a mean-field fluid is somewhat different from the usual ideal gases. With the increase of temperature, the particles can easily penetrate each other because the thermal energy becomes more and more significant and may eventually dominate over the bounded potential. Actually, the system can be considered as a fluid consisting of most particles distributed uniformly which act as the ideal gas and a few small clusters which are separated from each other. The self-diffusion coefficient is dominated by

\[ n_c = 1 + 4\pi\rho \int_0^{r_c} g(r)^2 dr. \]

(8)
‘jumps’ of particles in between clusters. According to eqn (8), one can expect that the average number of particles in each cluster (especially the particles sitting on top of each other) also linearly increases with the increase of the macroscopic density \( \rho \), indicating that the local density inside the clusters will not be affected strongly by the macroscopic density. In other words, even if the average number of particles per cluster increases, the typical spacing between clusters remains constant. Hence the density does not influence the single-particle dynamics, making the density to fall out of the scaling relation between the excess entropy and the diffusion coefficient.

For a further study of the scaling relation between \( D_T \) and \( S_{ex} \), we have fitted the data for \( D_T \) versus \(-S_{ex} \) in the regime of mean-field fluid behavior. We find that the temperature scaled diffusivity \( D_T \) is almost inversely proportional to \(-S_{ex} \) (see the dashed line in Fig. 8), and the fitting formula can be given by

\[
D_T \approx -0.22/S_{ex}. \tag{9}
\]

At the moment, such a scaling formula between \( D_T \) and \( S_{ex} \) is only proved to be valid in the fluid with harmonically repulsive particles. And whether this kind of scaling formula can also be used in other fluids of bounded interaction needs a confirmation. Nevertheless, we can at least say that the alternative scaling relation \( (D_T \text{ versus } S_{ex}) \) seems to be relevant to the mean-field fluid behavior of the system. For instance, the system of Hertzian spheres is expected to exhibit a mean-field behavior and to have a scaling relation similar to eqn (9) at high enough temperatures as Hertzian potential only has a rather minor modification in the exponent (from 2 to 2.5). To investigate the characteristics of the weakly correlated mean-field fluid, we have studied the compressibility factor \( Z = \beta P/\rho \) and the excess free energy per particle \( F_{ex}/N \) in the regime where the alternative scaling relation \( D_T \text{ versus } S_{ex} \) appears to hold. As can be seen in Fig. 10, both the compressibility factor \( Z \) and the excess free energy per particle \( F_{ex}/N \) are approximately linear functions of density \( \rho \). This is rather like the equation of state reported in GC fluids.\(^{41,42} \) Indeed, the plots of \( D_T \) versus \( S_{ex} \) collapse onto a single master curve for all of the densities and temperatures studied in GC fluids,\(^4 \) which is probably due to the fact that the GC fluid exhibits such a mean-field fluid behavior over a surprisingly wide range of densities and temperatures.\(^{41,42} \) And the different regime of mean-field behavior between harmonic spheres and GC fluids is possibly caused by the harshness of the potential, i.e., the GC interaction is softer.

It is also interesting to study the dependence of the Rosenfeld reduced diffusion co-efficient \( D_R \) on the two-body excess entropy \( S_2 \). Previous researches on some other kinds of fluids with bounded potential, e.g. Hertzian spheres and GC fluids, have shown that the plots of the Rosenfeld self-diffusivity \( D_R \) versus \( S_2 \) fall on the same master curve, despite the anomalous dependency of \( D \) and \( S_2 \) upon \( \rho \). Here we have studied the scaling relation between \( D_R \) and \( S_2 \) for the system with harmonic spheres. In Fig. 11, we show the curves of the Rosenfeld reduced diffusion co-efficient \( D_R \) on the two-body excess entropy \( S_2 \) along different isochors for a convenient comparison with previous results of other systems with soft particles. Apparently all of the plots at different densities approximately fall on the same master curve too, like the behaviors of other core-softened fluids.\(^4,6 \) Until now, such a scaling relation has been shown to hold for several systems with soft particles, so it can be possibly expected to describe the behaviors of all the fluids with bounded potential. Nevertheless, further studies in different systems need to be done to explore the issue whether
this scaling relation between $D_R$ and $S_2$ is generally valid in the fluids with bounded potential.

4 Conclusion

We present a detailed study on the behaviors of the fluid with harmonic repulsions via molecular dynamics simulation. Like the other systems with core-softened particles, the fluid exhibits both structural and dynamical anomalies especially at low temperatures, i.e., the dependence of self-diffusivity $D$, two-body excess entropy $S_2$ and full excess entropy $S_{ex}$ on density $\rho$ is nonmonotonic. The Rosenfeld entropy scaling ($D_1$ versus $S_{ex}$) of the system holds true only in the region of normal behavior at sufficiently low temperatures ($T < 0.05$), where the soft particles may tend to avoid overlapping and behave like hard spheres or simple atomic particles. Furthermore, the Rosenfeld scaled diffusivity $D_T$ follows a scaling relation with the two-body excess entropy $S_2$, which is the same as what has been observed in other fluids with bounded potential. Such a coincidence is possibly general to the systems of bounded potential, but the conformation needs more careful studies in different systems. When the temperature is increased high enough, the plots of the temperature scaled diffusion $D_T$ versus $S_{ex}$ collapse onto the same curve ($D_T$ is inversely proportional to $S_{ex}$ at least for this fluid with harmonic spheres), which is relevant to the fact that the system acts as a weakly correlated mean-field fluid. We should mention here that the findings in this paper, especially the structural characteristic that distinguishes the different dynamical regimes, are not only limited to the fluid with harmonic spheres, but also apply to GC fluids, other core-softened fluids (e.g., ref. 20) that exhibit weakly correlated mean-field behavior should also have a scaling relation between $D_T$ and $S_{ex}$ which makes further studies desirable.

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