Structure of the Lennard-Jones liquid estimated from a single simulation

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(Dated: September 24, 2020)

Combining the recent Piskulich-Thompson approach [Z. A. Piskulich and W. H. Thompson, J. Chem. Phys. 152, 011102 (2020)] with isomorph theory, the structure of a single-component Lennard-Jones system (LJ) is obtained at an arbitrary state point in almost the whole liquid region of the temperature-density phase diagram from a single simulation. The LJ exhibits two temperature range where the van’t Hoff’s assumption that energetic and entropic forces are temperature independent is valid. A method to evaluate the structure at an arbitrary state point along an isochore from the knowledge of only structures at two temperatures on the isochore is also discussed. We argue that the structure of any R-simple system obeying van’t Hoff’s assumption in the whole range of temperatures can be determined in the whole liquid region of phase-diagram from only a single simulation.

I. INTRODUCTION

The structure of an equilibrium liquid is characterized by the radial distribution function \( g(r) \). This quantity can be obtained by light scattering experiments, simulation or liquid state theory\[1-3\]. This quantity provides not only an idea of the structure, but also facilitates in predicting the various thermodynamics quantities as \( g(r) \) is related to the latter through interparticle interactions\[4\]. The static structure factor, which is the Fourier transform of \( g(r) \), is an input of the mode-coupling theory (MCT), which yields the dynamical quantities such as mean-squared displacement (MSD) or intermediate scattering function\[5, 6\].

Experiments are tricky to perform for supercooled liquids, which have a strong tendency to crystallize. Simulations are equally difficult and need to perform for a very long time due to associated long relaxation time\[7\].

Theoretical study of the temperature dependence of the structure will facilitate the prediction of structure from limited experimental or simulation data; however, such studies are limited\[8\]. Piskulich and Thomson\[9\] have shown that the radial distribution function \( g(r) \) of TIP4P/2005 water\[10\] at several temperatures can be obtained from a single simulation. This theory is based on the van’t Hoff’s assumption\[11\] that the energetic and entropic forces are temperature independent.

In this paper, we have tested the van’t Hoff’s assumption for the single-component Lennard-Jones (LJ) system. This assumption is not valid for the whole range of temperatures, but it is valid for two ranges of temperatures separately. The Piskulich-Thompson theory, then, has been employed to LJ to predict the structure at other temperatures along the same isochore in each temperature range separately. We have also prescribed a method to predict the structure from knowledge of \( g(r) \) at two different temperatures along the same isochore, without performing any simulation or experiment. Along an isomorph\[12-15\] the structure and dynamics of many systems, known as the Roskilde or R-simple system, are invariant in the reduced unit\[16, 17\]. We have combined the Piskulich-Thompson\[9\] approach with isomorph theory to predict the structure of the LJ at an arbitrary state point in the liquid region of the temperature-density phase diagram.

We describe the Piskulich-Thompson theory in Sec. II. Sec. III describes the simulation method used. The results are given in Sec. IV. The Sec. V explains how the \( g(r) \) along an isochore can be obtained without any simulation if the radial distribution functions at two temperatures along the same isochore are known. The extension of the Piskulich-Thompson theory for R-simple liquids is described in Sec. VI. A summary and discussions are given in Sec. VII.

II. PISKULICH-THOMPSON THEORY

The radial distribution function is defined by\[18\]

\[
g(r) = \frac{V}{N^2} \left\langle \sum_i \sum_{j \neq i} \delta(r - r_{ij}) \right\rangle, \tag{1}
\]

where \( r_{ij} \) is the distance between particles \( i \) and \( j \), \( V \) and \( N \) are volume and the number of particles, respectively. The \( \left\langle \cdots \right\rangle \) represents the ensemble average. The temperature dependence of the radial distribution function \( g(r) \) is given by\[9\]

\[
\frac{\partial g(r)}{\partial \beta} = -\frac{V}{N^2} \left\langle \sum_i \delta H \sum_{j \neq i} \delta(r - r_{ij}) \right\rangle, \tag{2}
\]

where \( \delta H = H - \left\langle H \right\rangle \) is the fluctuation of Hamiltonian from its mean value \( \left\langle H \right\rangle \), and \( \beta = (k_B T)^{-1} \), \( k_B \) and \( T \) being the Boltzmann constant and temperature,
respectively. The fluctuation of kinetic energy in a constant temperature simulation is zero, and therefore, one can replace the Hamiltonian by the potential energy\cite{9}.

The Helmholtz free energy profile $\Delta A(r)$ can be written in terms of the radial distribution function $g(r)$ as\cite{9}

$$ \Delta A(r) = -k_B T \ln g(r) - k_B T \ln \nu(r), \quad (3) $$

where $\nu(r) = r^2$ is a geometric factor. Without the geometric factor, the free energy is simply the potential of mean force $PMF(r)$. The derivative of Helmholtz free energy with respect to $\beta$ is

$$ \frac{\partial A(r)}{\partial \beta} = k_B T \left[ \frac{g_H(r)}{g(r)} + k_B T \ln g(r) + k_B T \ln \nu(r) \right]. \quad (4) $$

$$ = k_B T \left[ \frac{g_H(r)}{g(r)} - \Delta A(r) \right], \quad (5) $$

where $g_H(r) = -\frac{\partial g(r)}{\partial \beta}$. The Helmholtz free energy $\Delta A(r)$ can be written in terms of internal energy and entropy as

$$ \Delta A(r) = \Delta U(r) - T \Delta S(r). \quad (6) $$

With the assumption that both $\Delta U(r)$ and $\Delta S(r)$ doesn’t depend on the temperature (van’t Hoffian assumption), comparison of equations (5) and (6) yields expression for the internal energy and the entropy as,

$$ \Delta U(r) = \frac{g_H(r)}{g(r)}, \quad (7) $$

and

$$ \Delta S(r) = \frac{1}{k_B T^2} \frac{\partial A(r)}{\partial \beta}. \quad (8) $$

While the $\Delta U(r)$ can be readily evaluated from equations (1) and (2), the entropy $\Delta S(r)$ can be determined from equations (4) and (1). Thus one can calculate the value of $\Delta U(r)$ and $\Delta S(r)$ from simulation data at a given temperature $T_0$. Now from equation (3), the radial distribution function at an arbitrary temperature $T$, same density, can be written as

$$ g(r; \beta) = \frac{1}{\nu(r)} e^{-\beta \Delta U(r) + \Delta S(r)/k_B}, \quad (9) $$

where $\Delta U(r)$ and $\Delta S(r)$ are evaluated at the temperature $T_0$. The above equation is van’t Hoff plot\cite{19} and $\Delta U(r)$ and $\Delta S(r)$ are assumed to be temperature independent.

Substituting the values of $\Delta U(r)$ from equation (7) and $\frac{\partial A(r)}{\partial \beta}$ from equation (4), the equation (9) becomes\cite{9}

$$ g(r; \beta) = g(r; \beta_0) e^{\Delta U(r)(\beta_0 - \beta)}. \quad (10) $$

This expression of $g(r; \beta)$ depends only on $\Delta U(r) \equiv g_H(r)/g(r)$. It must be noted that throughout the derivation the van’t Hoffian assumption will be assumed.

### III. SIMULATION DETAILS

We have performed a canonical ensemble molecular dynamics simulations (NVT) of the LJ system employing Nose-Hover thermostat with $N = 2000$ particles at various densities and temperatures. Employing a shifted-forces cutoff\cite{20} the LJ interaction potential between particle $i$ and $j$ is given as

$$ \phi(r_{ij}) = \begin{cases} \left( \frac{\sigma_{ij}}{r_{ij}} \right)^{12} - \left( \frac{\sigma_{ij}}{r_{ij}} \right)^6 + C_1 r_{ij} + C_2, & r_{ij} < 2.5 \sigma, \\ 0, & r_{ij} \geq 2.5 \sigma, \end{cases} \quad (11) $$

where $C_1$ and $C_2$ ensure that the $\phi(r)$ and its first derivative are continuous at the cut-off $r = 2.50 \sigma$. The simulations were performed using RUMD (Roskilde University Molecular Dynamics) software\cite{21} which is a GPU (graphical processing unit) code. All quantities reported in this paper are in LJ units: length, time and temperature are expressed in units of $\sigma$, $\sqrt{m \sigma^2/\epsilon}$ and $\epsilon/k_B$, respectively.

### IV. RESULTS

#### A. The validity of van’t Hoffian assumption for LJ

If the van’t Hoffian assumption is correct, then from equation (9) or (10), ln $g(r; \beta)$ vs. $\beta$ should be a straight line. We plot the $g(r)$ against $1/T$ in log-linear scale in figure 1(a) for LJ system at density $\rho = 0.80$ for a range of $r$ values. It shows that the ln $g(r)$ vs $1/T$ are not straight lines throughout the considered temperature range. It means that the van’t Hoffian assumption that the $\Delta U(r)$ and $\Delta S(r)$ are temperature independent is not correct. The van’t Hoffian assumption has been seen not to be valid in other liquids or liquid mixtures with covalent bonds\cite{22, 23} as well. Interestingly, the van’t Hoffian assumption is not valid for the LJ which is a very simple liquid without any covalent bonds. However, two temperature ranges can be assigned where ln $g(r)$ vs. $1/T$ plots are fairly straight lines, though with different slopes. The main variation of $g(r)$ with inverse temperature is seen near the first peak of the $g(r)$, i.e., near $r = 1.0$. Fig. 1(b) exhibits the $1/T$-dependence of $g(r)$ at $r = 1.0$. It shows a non-monotonic behavior with maximum near $T = 1.0$. On either side of the peak the plot is a straight line, and thus the van’t Hoffian assumption holds good in two temperature ranges, one at low $T$ and another at high $T$.

#### B. Thompson theory applied to LJ

We now apply the Piskulich-Thompson theory at two temperatures, one on each side of the peak in Fig. 1(b) where the van’t Hoffian assumption is approximately valid, in order to determine $g(r)$ at other temperatures on that side. Fig. 2(a) exhibits $g(r)$ at $T =$
0.80, 1.00, 1.20, 2.20, 2.60, 2.60, and 3.00 obtained by applying Piskulich-Thompson theory at reference temperature $T = 1.8$ and density $\rho = 0.80$. The radial distribution functions determined by employing Piskulich-Thompson theory (lines) have been compared with that of obtained from MD simulation (symbols). They are in good agreement. The data have been shifted upward for clarity. Fig. 2(b) shows the comparison of $g(r)$ at various temperatures between simulation and Piskulich-Thompson theory applied at the reference temperatures (a) $T = 1.8$ and (b) $T = 6.0$. Low $T$ and high $T$ region of the $g(r = 1.0)$ vs. $1/T$ peak in Fig. 1(b) are shown in (a) and (b), respectively. The density is given by $\rho = 0.80$.

V. THE STRUCTURE AT AN ARBITRARY TEMPERATURE ALONG AN ISOCHORE ESTIMATED FROM JUST TWO RADIAL DISTRIBUTION FUNCTIONS

Equation (10) can be re-written as

$$\Delta U(r) = \frac{T_0 T}{T - T_0} \ln \left[ \frac{g(r; \beta)}{g(r; \beta_0)} \right], \quad (12)$$

where $\beta_0 = \frac{1}{k_B T_0}$. Thus $\Delta U(r)$ can be evaluated from $g(r)$ at two different temperatures at a given density. The procedure for obtaining $g(r)$, whether in experiments or simulations, is irrelevant. These two temperatures can be anywhere on the isochore in question, as long as the van’t Hoffian assumption is valid. However, one has no prior knowledge of the temperature range where van’t Hoffian assumption is valid for the system under consideration. It is, therefore, intuitive to consider two temperatures that are not far away from each other and hence the van’t Hoff’s assumption is valid in that small tempera-
ture range. Once the $\Delta U(r)$ is determined, the $g(r)$ of liquids can be calculated at any temperature along the isochore from equation (10) without performing further simulation (or conducting more experiments). This is quite useful for a liquid which interparticle interaction is unknown, and hence in this sense, the method is superior to the liquid state theory, which requires the knowledge of the interactions between the particles.

Fig. 3(a) shows the comparison of $\Delta U(r)$ obtained by using Eq.(12) with that determined directly from MD simulations at $T = 30$, $T = 45$, and $T = 80$ at density $\rho = 2.0$. The $\Delta U(r)$ at $T = 30$ has been evaluated using $g(r)$ at temperatures $T = 30$ and $T = 32$. Similarly, the function $\Delta U(r)$ at $T = 45$ and $T = 80$ have been obtained from $g(r)$ at $T = 45$ & $T = 50$ and $T = 80$ & $T = 75$, respectively. The $\Delta U(r)$ at these three temperatures obtained by using Eq.(12) (lines) are in good agreement with that obtained from MD simulations (symbols) directly. Thus one doesn’t need the fluctuation of Hamiltonian $\delta H$ to evaluate $\Delta U(r)$, as assumed in the Piskulich-Thompson theory. Fig. 3(b) shows a comparison of $g(r)$ obtained from $\Delta U(r)$ (from $g(r)$ at two nearby temperatures) with the one determined from MD simulations (symbols) directly at $T = 30$, $T = 45$, and $T = 80$. They are in good agreement with one another. This method opens up the possibility of predicting the $g(r)$ along an isochore of a liquid of which interparticle interactions are unknown if its $g(r)$ at two nearby temperatures are available, say, from light scattering experiments. This method is robust for the temperature range where the van’t Hoffian assumption is valid.

VI. PISKULICH-THOMPSON+ISOMORPH THEORY

So far we have discussed the determination of $g(r)$ along an isochore either (i) directly using Piskulich-Thompson theory or (ii) by employing Eq.(10) and (12) where only $g(r)$ at two temperatures needed. Now we generalize this into a method to calculate the $g(r)$ at an arbitrary temperature in the $T - \rho$ phase-diagram from just one simulation at the reference state point ($\rho_0, T_0$). To achieve this we combine the Piskulich-Thompson theory with isomorph theory. First we determine the isomorph passing through the reference point ($\rho_0, T_0$). The equation for an isomorph of the LJ system is given by[24–26]

$$T(\rho) = \frac{T_0}{\rho_0} \left[ \frac{\rho}{\rho_0} - 1 \right] \left( \frac{\rho}{\rho_0} \right)^4 - \frac{\gamma_0}{2} \left( \frac{\rho}{\rho_0} \right)^2 \right) \quad (13)$$

where the so-called density-scaling exponent $\gamma_0$ is calculated from equilibrium fluctuations at the reference state point by means of

$$\gamma_0 = \frac{\langle \Delta U^2 \rangle}{\langle \Delta U \rangle^2}. \quad (14)$$

Here, $\Delta U$ and $\Delta W$ are fluctuations in potential energy and virial.

Fig. 4(a) exhibits $g(r)$ at various state points along the isomorph starting from the reference state point ($\rho_0, T_0$) = (1, 1) for the LJ system. The inset of Fig. 4(b) shows the isomorph (line) and the state points (red symbols) where MD simulations have been performed. The main panel of Fig. 4(b) shows the $g(r)$ in Fig. 4(a) in the reduced unit. The color scheme for both figures are the same.

As expected, the $g(r)$ is invariant in the reduced unit i.e. $g(\rho^{1/3}r) = \text{constant}$ along the isomorph (Fig. 4(b)). Hence, the $g(r)$ of the system at any point of the isomorph line, say, ($\rho_1, T_1$), can be obtained easily from the $g(r)$ of the reference point ($\rho_0, T_0$). Thereafter, the Piskulich-Thompson theory can be employed along the isochore at $\rho_1$. In order to apply the Piskulich-Thompson theory we require the potential energy of all the config-

\[
\begin{align*}
T(\rho) &= \frac{T_0}{\rho_0} \left[ \frac{\rho}{\rho_0} - 1 \right] \left( \frac{\rho}{\rho_0} \right)^4 - \frac{\gamma_0}{2} \left( \frac{\rho}{\rho_0} \right)^2 \right) \\
\gamma_0 &= \frac{\langle \Delta U^2 \rangle}{\langle \Delta U \rangle^2}.
\end{align*}
\]
show a comparison of potential energy at the reference point \((\rho_0, T_0)\). However, the potential energy at \((\rho_1, T_1)\) can also be obtained by scaling the same at the reference point \((\rho_0, T_0)\).

The scaled potential energy \(U\) of LJ at \((\rho_1, T_1)\) in terms of potential energy at the reference point \((\rho_0, T_0)\) is given by[27]

\[
U = \tilde{\rho}^{m/3}U_0^{m} + \tilde{\rho}^{n/3}U_0^{n},
\]

where \(\tilde{\rho} = \rho_1 / \rho_0\) and \(U^k \equiv \sum_{i<j} \rho^k(r_{ij})\). The \(U^m\) and \(U^n\) are repulsive and attractive parts of the LJ potential (and hence \(m = 12\) and \(n = 6\), respectively, implying that

\[
U = U^m + U^n,
\]

in which \(U_0^m\) and \(U_0^n\) are the values of \(U^m\) and \(U^n\) at the reference point \((\rho_0, T_0)\) on the isomorph. The \(U^m\) and \(U^n\) are given by[27]

\[
\begin{align*}
U^m &= 3W - mlU \overline{\rho}^m, \\
U^n &= -3W + mlU \overline{\rho}^n,
\end{align*}
\]

The above equation is based on the fact that \(U^k \overline{\rho}^k = \text{constant}\) (ignoring the linear term in the shifted-force cutoff LJ potential, see (11)).

Fig. 5 shows a comparison of \(-g_H(r)\) in the reduced unit. i.e. \(-g_H(\rho^{1/3}r)\) obtained from isomorph scaling and direct simulation at state points \((\rho = 1.2, T = 2.45)\), \((\rho = 1.5, T = 6.77)\), \((\rho = 2, T = 23.29)\) and \((\rho = 2.5, T = 59.01)\) on the isomorph line. The \(-g_H(\rho^{1/3}r)\) obtained from isomorph scaling (lines) described above are in good agreement with those obtained from MD simulations (open symbols) at these state points. Now we have \(g(r)\) as well as \(-g_H(r)\) (so \(\Delta U(r)\)) at the state point \((\rho_1, T_1)\), and therefore, the Piskulich-Thompson theory can be applied easily. Figures 6 (a)-(d) show comparisons of \(g(r)\) obtained by employing Piskulich-Thompson+isomorph theory with those from simulations. They are in good agreement when \(|T - T_1|\) is small. The discrepancy at large \(|T - T_1|\) is associated with following two facts: (i) the LJ obeys the van’t Hoff’s assumption applied to a good approximation only in a limited temperature range and (ii) the isomorph is incapable to capture the first peak correctly[16] One can observe the discrepancy between isomorph theory and simulation in \(-g_H(\rho^{1/3}r)\) near the first peak in Fig. 5, as well. To summarize, for a van’t Hoffian valid liquid, \(g(r)\) can be calculated at any arbitrary state point in the liquid region of the temperature-density phase diagram from a single simulation data.

VII. DISCUSSIONS AND SUMMARY

The van’t Hoffian assumption is that the energetic and entropic forces are temperature independent. We have shown that the LJ disobeys the van’t Hoff’s assumption when viewed over the entire temperature range studied. Unlike other non-van’t Hoff liquids[22, 23], the LJ does not have any covalent bond. The fact that the van’t Hoffian assumption breaks down might be due to different activation energy at low and high temperatures. While at very high temperature, the LJ is governed by entropic forces and energy play a little role; at low temperature the energy dominates, and some of the particles remain close to one another for quite a long time, behaving as a quasi-covalent bond.

While the van’t Hoffian assumption for the LJ is not valid for the whole range of temperatures studied, there are two distinct temperature ranges (see Fig. 1(b)) where van’t Hoffian assumption applies approximately. This is validated by the excellent agreement of the \(g(r)\) obtained from Piskulich-Thompson theory and simulation (see Fig. 2). The Piskulich-Thompson theory has been applied at one temperature in each low \(T\)-range \((T = 1.8)\) as well as high \(T\)-range \((T = 6)\) to determine \(g(r)\) at other temperatures in that range (see Fig. 2). For a van’t Hoff-
van’t Hoffian system, just a single simulation is required to determine \( g(r) \) at an arbitrary temperature at the same density employing Piskulich-Thompson theory. For a non-van’t Hoffian system, such as LJ, the range of temperatures where Piskulich-Thompson theory can be applied to determine \( g(r) \), is limited. Thus for such a system, one can only determine \( g(r) \) at the temperatures in the vicinity of the state point where simulation data is available. The simulations of supercooled liquids are challenging due to its long relaxation time and strong crystallization tendency. At such low temperatures, the van’t Hoffian assumption would probably be valid for all systems, and thus Piskulich-Thompson theory can be applied. In such a scenario, this theory could be helpful.

We have shown that the energetic force \( \Delta U(r) \) can be evaluated from a knowledge of \( g(r) \) at two temperatures, say \( T_0 \) and \( T \), in the van’t Hoffian region (see Eq. (12)). This method is particularly useful when the interatomic/intermolecular interactions are not known, forbidding computer simulations, or when simulations/experiments are extremely challenging. Since many systems may have multiple temperature ranges with valid van’t Hoffian assumption similar to the LJ, it is imperative to consider \( g(r) \) at two very close temperatures, where van’t Hoff’s assumption is bound to be valid. Once \( \Delta U(r) \) is determined, the \( g(r) \) can be predicted at various temperatures along the isochore.

Piskulich-Thompson theory works only along an isochore. We have extended this theory to calculate the \( g(r) \) at an arbitrary state point of the liquid region of the phase diagram from a single simulation at a reference state point \((\rho_0, T_0)\). For this, we have combined Piskulich-Thompson approach with the isomorph theory. The structure of a liquid along an isomorph is invariant in reduced units. It should be noted that not all systems have isomorphs (are R-simple) [28] – water is a striking counter example – and the current theory is of course limited to R-simple liquids. The isomorph theory is not valid in the gaseous region as well[16, 29], and hence, this theory can not be applied to determine the radial distribution function in the low-density region of the phase diagram.

In order to calculate \( g(r) \) at an arbitrary state point \((\rho, T)\), we first calculate \( g(r) \) at \((\rho, T_{iso})\), where \( T_{iso} \) is on the isomorph line. We here need to scale the potential energy from the reference point to the \((\rho, T_{iso})\) as well. For LJ, this is done as shown in Eq. (17-18) following Ref. 27. This expression is system dependent, and one needs to find the expression for other potentials as per the isomorph theory described in the Ref. [27]. Thus \( \Delta U(r) = g_H(r)/g(r) \) is known at \((\rho, T_{iso})\). Thereafter, the Piskulich-Thompson theory is employed along the isochore \( \rho = \rho \) to calculate \( g(r) \) at the designated state point \((\rho, T)\). Again, for a perfect van’t Hoffian system \( g(r) \) at every state point of the phase diagram (liquid region) can be obtained. On the other hand, if the system doesn’t show single temperature range where the van’t Hoffian assumption is valid; this theory cannot be used to evaluate \( g(r) \) in the whole liquid phase of the diagram from a single simulation. But if the information of different temperature ranges and one simulation data in each temperature ranges are available, one can calculate \( g(r) \) in the whole liquid phase part of the phase diagram for an R-simple system. We would again remind that not all systems exhibit isomorphic invariance[13, 28]. However, many systems are R-simple[13] and this Piskulich-Thompson+isomorph theory should apply to any such system.

All thermodynamic quantities are related to the radial distribution function \( g(r) \), and hence they can be evaluated in the liquid region of phase-diagram whenever this theory is applicable. But again, there are systems with three-body interactions such as silicon[30–32] where this theory will not be applicable. As far as dynamics is concerned, the MCT requires the structure factor (which is Fourier transform of \( g(r) \)) and interparticle interactions to provide the dynamics such as MSD and intermediate scattering function. Thus for an R-simple system, employing Piskulich-Thompson+isomorph theory along with MCT, one can calculate all thermodynamics as well as dynamical quantities.

In summary, we have shown that: (i) the LJ disobeys the van’t Hoff’s assumption that the energetic and entropic forces are temperature independent. However, we have identified two temperature ranges in which the van’t Hoffian assumption is valid to a good approximation. We validated this by comparing the \( g(r) \) determined by employing Piskulich-Thompson theory with that obtained from the simulation with excellent agreement. (ii) one can obtain the energetic force term \( \Delta U(r) \) without any simulation, and only \( g(r) \) at two temperatures in the temperature range where van’t Hoffian assumption is valid is required. Then \( g(r) \) along an isochore can be calculated from \( \Delta U(r) \) at all temperatures where van’t Hoff’s
assumption is valid. (iii) the $g(r)$ can be determined at an arbitrary state point in the liquid region of the phase diagram for an isomorphic invariant (R-simple) liquid from just a single simulation by employing Piskulich-Thompson+isomorph theory.

It would be interesting to investigate if the van’t Hoffian assumption is valid in the whole temperature range in other R-simple liquids, e.g. inverse-power law, Yukawa potential or Morse potential.

**ACKNOWLEDGEMENTS**

This work was supported by the VILLUM Foundation’s Matter grant (No. 16515).

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FIG. 6: The comparison of $g(\rho^{1/3} r)$ obtained by employing Piskulich-Thompson+isomorph theory with that of simulation at different points of (a) $\rho = 1.10$ (b) $\rho = 1.50$ (c) $\rho = 2.00$, and (d) $\rho = 2.50$. The base temperature and density for Piskulich-Thompson+isomorph theory is $(\rho_0 = 1.0, T_0 = 1.0)$. 