

Application of the uniform source-and-sink scheme to molecular dynamics calculation of the self-diffusion coefficient of fluids

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SUMMARY

A uniform source-and-sink scheme was developed by Cao and Li [*J. Chem. Phys.* **133** (2010), 024106] to calculate the thermal conductivity of solid argon. Now, we aim to apply this scheme to the calculation of the self-diffusion coefficient. We divide the particles into equal halves, and label them with A or B. By exchanging the labels of individual atoms from the right and left half systems, we can produce an internal matter flux, and hence the internal matter source and sink can be realized. The density profile is piecewise quadratic throughout the system and the self-diffusion coefficient can be easily extracted from the mean densities of the right and left half systems rather than by fitting the density profile. In particular, this is a nonequilibrium molecular dynamics method but established on an equilibrium system. The scheme is applied to calculate the self-diffusion coefficient by taking a Lennard–Jones fluid as a case, examining its homogeneity, convergence, label exchange interval, and dependence on density, temperature, and system size. The uniform source-and-sink scheme is demonstrated to be able to give accurate results with fast convergence. Copyright © 2012 John Wiley & Sons, Ltd.

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KEY WORDS: self-diffusion coefficient; nonequilibrium molecular dynamics; Lennard–Jones; uniform source-and-sink scheme

1. INTRODUCTION

The self-diffusion coefficient is a key parameter for measuring a diffusion process of particles in a pure fluid, which is widely involved in engineering transport processes, such as separation, tertiary-oil recovery, and extraction of essential oils. Also, the self-diffusion coefficients set time and spatial scales of various transport processes in fluids such as in the Stokes–Einstein relation [1]. Thus, it is highly desired to study the self-diffusion coefficient. Unfortunately, experimental studies on the self-diffusion coefficient are scarce because of the difficulty in measuring the diffusion of an individual particle while the surroundings are the same species [2, 3]. As a result, the possible solution points to the ‘computer experiment’, that is, molecular dynamics (MD) simulation.

The molecular dynamics method is a powerful tool that is widely used in physics, chemistry, and material sciences, such as simulations of the structure, mechanical and transport properties of carbon nanotubes [4, 5], or nanoscale fluid flows [6–8]. The MD methods for calculating the transport properties can be classified as equilibrium molecular dynamics (EMD) and nonequilibrium molecular dynamics (NEMD) schemes [9–11]. The EMD methods can either use the Green–Kubo integral formula to calculate the time correlation function or use the Einstein relation, also known as the

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mean squared displacement method [12]. These two schemes have been proven to be equivalent. In the case of one-dimensional self-diffusion, the Green–Kubo relation can be expressed as [13]

$$D = \frac{1}{3N} \sum_{i=1}^N \int_0^{\infty} \langle v_i(t) \cdot v_i(t_0) \rangle dt, \quad (1)$$

and the Einstein relation is in the form of

$$D = \lim_{t \rightarrow \infty} \frac{1}{6N} \sum_{i=1}^N \frac{d}{dt} \langle [r_i(t) - r_i(t_0)]^2 \rangle. \quad (2)$$

In these equations, $\langle \dots \rangle$ means the average over particles and time origins. Previous work has shown the slow decay of the time correlation functions termed as the ‘long time tail’, which may lead to a very long time convergence [11]. Then, obtaining the precise evaluation of the transport properties can be rather tough. Therefore, in the process of seeking an optional method, the NEMD schemes, which converge much faster, are put forward and have been widely applied in recent years.

A good NEMD algorithm should exhibit some features such as homogeneity, fast convergence, periodic boundary conditions, small temperature gradient, and Hamiltonian [14]. A classical way is to introduce a perturbation that can generate the same response as experiments, and then through the resulted flux the transport coefficients can be calculated. An opposite way is to reverse the cause and effect by imposing a flux that can either be energy, momentum, or matter corresponding to the calculation of the thermal conductivity, shear viscosity, or diffusion coefficient, respectively [15]. In the process of calculating these transport coefficients, researchers have reported temperature [16], velocity [15], and number density [17] jumps at the boundaries, which are quite harmful to acquiring reliable simulation results. Reference [18] developed a scheme to eliminate the temperature jump at the boundary by shifting the perturbation into the simulation system, which resulted in a fast excited vibration relaxation. Furthermore, a uniform heat source and sink based on heating and cooling individual atoms, which greatly enhanced the relaxation rate, was used in Reference [19].

Thus, a new NEMD method called uniform source-and-sink (USS) scheme, which combines the ideas of References [14, 18, 19], was developed by Cao and Li [20] and was first used to calculate the thermal conductivity of solid argon. It shares all the good features mentioned in Reference [14] and also solves the problem of temperature jump by enhancing the excited phonon modes through internal heat source and sink. Their work illustrates a small system size effect, fast convergence, and accurate results of the USS method, which are clearly stated in Reference [20]. Now, we attempt to extend this scheme from the calculation of thermal conductivity to the self-diffusion coefficient to further check its validity.

In this paper the USS scheme is realized by exchanging labels of individual particles to calculate the self-diffusion coefficient of a Lennard–Jones (LJ) fluid. We divide the particles into equal halves labeled as A and B. Every time we perform a label exchange between two particles with different labels from the left and right half systems and create a source and a sink for particles A and B, respectively. We further make the source and sink uniform, so that a periodically quadratic density profile can be produced and the self-diffusion coefficient can be extracted from the mean densities of the left and right half systems. In Section 2, we will describe the theory and methodology in detail. Simulation results are presented in Section 3 with discussion included. Finally, this paper concludes in Section 4.

2. THEORY AND METHODOLOGY

For the case of heat conduction, energy is transported, which is driven by the temperature differences and obeys Fourier’s law. Similarly, diffusion occurs because there are density (concentration) differences, which lead to matter transport following Fick’s law. Therefore, the USS method, which establishes a heat flux, can also be used to construct a matter flux. For one dimensional self-diffusion,

Fick's law is in the form of

$$m_x = -D \frac{\partial \rho}{\partial x}, \quad (3)$$

in which the matter flux is proportional to the gradient of density.

An N particles system is considered and the particles are divided into equal halves labeled as A or B. The simulation box is divided into slabs along one direction, say the x direction. The matter flux is constructed in this way: A slab in the left half system is first selected and we pick up one particle labeled as A. Then another slab from the right half is chosen and a particle labeled as B is identified. By exchanging the labels of these two particles, we successfully create a matter flux from the left to the right (for the A particles) and another flux from the right to the left (for the B particles). Figure 1 exhibits the label exchange process as the particles labeled as A are shown with open circles while B are shown with solid circles. Then the left half system has a sink for the A particles and a source for the B particles, while the right half system is just the opposite. We define an exchange interval W , and every W time steps, the exchange is performed. The slabs picked up from the two halves are selected to have minimum exchange rates, respectively, in the left and right half systems, which will ensure that our matter source and sink are uniform. This is realized by recording the total times of label exchange of every slab over the past simulation steps. Then in the next step, the slab _{i} and slab _{j} with the least times of the label exchange are picked out. Because the label exchange is equivalent to matter exchange, the matter densities of these two slabs will approach those of the others. It should be noted that the driving force of the matter transport is the density differences of A (B) particles throughout the system. Because these particles are only distinguished from the labels while their atomic mass and diameters are the same, the density of particles A + B is identical in the whole system. Therefore, in the simulations of the self-diffusion coefficient, the USS scheme is used as an NEMD idea but established on an equilibrium system, because no perturbation is introduced over the simulation process.

The source or sink in our USS scheme particularly refers to one labeled particles, say particles A with the source in the right and the sink in the left. For convenience in the next sections of this paper, we will only focus on particles A, because the label exchange we perform is symmetrical. Therefore, we can write out the matter source/sink density as (with a negative value meaning the matter removal rate)

$$m_v = 2 \frac{\sum_{\text{transfers}} m}{t L_x L_y L_z}, \quad (4)$$

in which L_x , L_y , L_z are the box lengths in x , y , z directions, respectively, t is the simulation time and m is the mass of an atom. As discussed above, for every W time steps, a label exchange is

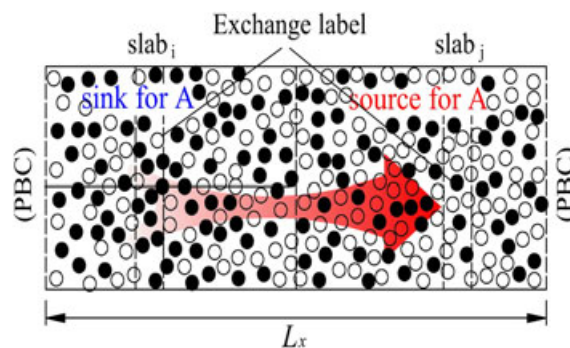


Figure 1. Schematic diagram of the NEMD simulations system applying the USS scheme with open circles representing the A particles and solid circles representing the B particles.

performed. Then for our one-dimensional case and steady state, the density profile is in the form of

$$\rho = \begin{cases} \frac{m_v}{2D}(x + \frac{L_x}{4})^2 - \frac{m_v L_x^2}{32D} + \rho_0, & (-\frac{L_x}{2} \leq x < 0) \\ \frac{-m_v}{2D}(x - \frac{L_x}{4})^2 + \frac{m_v L_x^2}{32D} + \rho_0, & (0 \leq x \leq \frac{L_x}{2}) \end{cases} \quad (5)$$

where ρ_0 is the mean density of the entire system and D is the self-diffusion coefficient. This piecewise quadratic profile results from the combination of the periodic boundary conditions and the label exchange from one half to the other, that is, the matter source and sink. Then the mean densities of particles A in the left and right halves of the system can be derived from Equation (5),

$$\bar{\rho}_L = \rho_0 - \frac{2}{L_x} \int_{-L_x/2}^0 \rho(x) dx = \rho_0 - \frac{m_v L_x^2}{48D} \quad (6a)$$

$$\bar{\rho}_R = \rho_0 + \frac{2}{L_x} \int_0^{L_x/2} \rho(x) dx = \rho_0 + \frac{m_v L_x^2}{48D}. \quad (6b)$$

Then we can obtain the average density difference between the left and right mean densities, that is, $\bar{\rho}_L$ and $\bar{\rho}_R$, and the system mean density

$$\Delta\bar{\rho} = \frac{(\rho_0 - \bar{\rho}_L) + (\bar{\rho}_R - \rho_0)}{2} = \frac{m_v L_x^2}{48D}, \quad (7)$$

and hence the self-diffusion coefficient can be given as

$$D = \frac{m_v L_x^2}{48\Delta\bar{\rho}}. \quad (8)$$

3. SIMULATION RESULTS AND DISCUSSION

We calculate the self-diffusion coefficient of a Lennard–Jones fluid to verify the USS scheme. The Lennard–Jones interaction potential is in the form of

$$\varphi(r) = 4\varepsilon \left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^6 \right], \quad (9)$$

where r is the intermolecular distance, σ is the molecular diameter and ε is the potential well depth [21]. The whole simulation box is divided into 20 slabs along the x direction. As discussed above, the atoms will be divided into equal halves with different labels as A or B. We use a face-centered cubic configuration as the initial state for the displacement of the atoms in our simulation box. The interaction is truncated at an intermolecular separation of $r_{\text{cut}} = 2.0\sigma$ and a leap frog Verlet algorithm [22] with a time step of 0.005τ is used to integrate the equations of motion for the molecules. We use the canonical ensemble through the Nose–Hoover thermostat method to maintain a constant temperature. Also, the relaxation time is $t_{\text{relax}} = 0.05\tau$. Then, through Equations (4)–(8), the self-diffusion coefficient could be extracted.

For convenience, rescaled units are introduced to exhibit the simulation results indicated by the superscript ‘*’, for example, the temperature $T^* = Tk_B/\varepsilon$, the density $\rho^* = \rho\sigma^3/m$, the velocity $v^* = v\sqrt{m/\varepsilon}$, the time $t^* = t\sqrt{\varepsilon/m}/\sigma = t/\tau$, the diameter $r^* = r/\sigma$, the self-diffusion coefficient $D^* = D\sqrt{m/\varepsilon}/\sigma$ [2], and the matter source/sink density $m_v^* = m_v(\tau\sigma^3)/m$.

To illustrate our theories above, we first need to have a thorough examination of the inner features of the USS scheme. For the Lennard–Jones fluid, we choose a state point of $\rho^* = 0.6$ and $T^* = 1.81$ in the liquid state region [23]. The simulated system contains 1000 atoms and the length of the simulation cell in the x direction is 18.8σ . The box sizes along the three directions satisfy $L_x : L_y : L_z = 2 : 1 : 1$. Figure 2(a) shows the density profiles of particles A, which validate our previous theory that the label exchange along with the periodic boundary conditions indeed creates

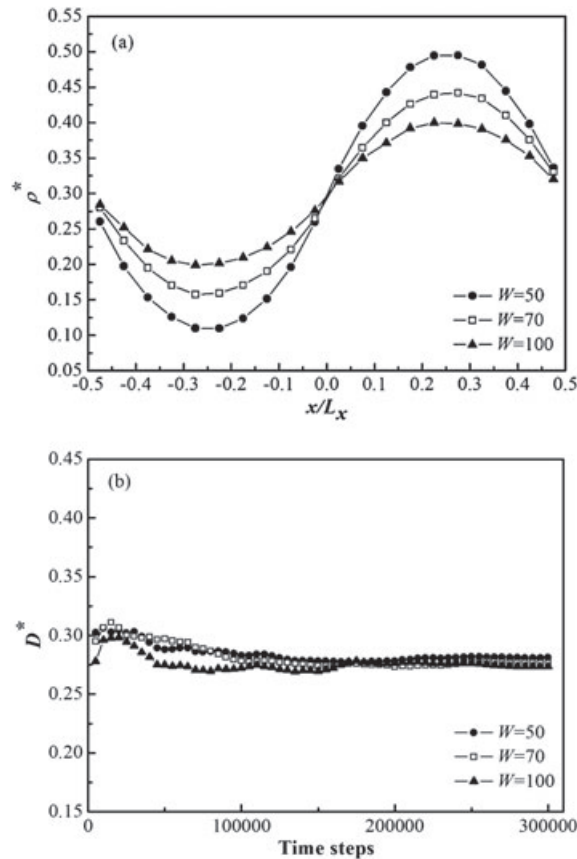


Figure 2. (a) Density profile and (b) time averaged self-diffusion coefficient for exchange interval $W = 50$, 70, and 100.

a piecewise quadratic distribution of the densities in the slabs. The smaller the exchange interval, the stronger the perturbation and it will lead to a more obvious deviation from the left and right mean densities. From Equation (4) we can also conclude that the source and sink matter density will decrease with the increase of W . Figure 2(b) exhibits the time averaged self-diffusion coefficient from 1500τ to 3000τ . The first 1500τ is used for the system to reach a steady state. The fluctuation is large at first but then quickly converges.

The dependence of the self-diffusion coefficient on the matter exchange interval W is shown in Figure 3. We record a series of the self-diffusion coefficients along with time under conditions of different matter exchange intervals. From the statistical error of the fluctuation we can acquire the uncertainty of the results. There are several points that should be mentioned. First, we can see an almost monotonic increase of the relative uncertainty with changing exchange interval W . The least relative uncertainty is at $W = 10$ and the value is only $\pm 0.7\%$, while the largest is at $W = 300$ and the value is $\pm 11.5\%$. Also, we can tell that when exchange interval W is below 100, the relative uncertainty is rather small compared with the situation when W is above 100. Second, when exchange interval W comes between 10 and 100, we have some relatively similar values of the self-diffusion coefficients, which also agree well with the reference data [21, 24]. Third, the simulation indicates that for a weak perturbation, that is, a relatively large matter exchange interval W , the time we need to establish a steady state is also relatively long. As a result, the preferable exchange interval should not be too large. However, it may be worth mentioning that a too small exchange interval will cause the A particles in the left selected slab or the B particles in the right selected slab to be used up. Then the actual exchange interval is larger than that we have settled initially. Fortunately, this will not affect the accuracy of the results. Then, for the case of 1000 atoms, we recommend the exchange interval W chosen to be less than 100.

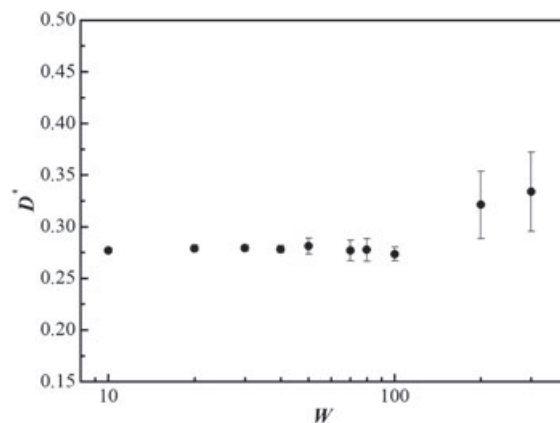


Figure 3. Dependence of the calculated self-diffusion coefficient on the label exchange interval. Data are collected from a simulation case with $N = 1000$.

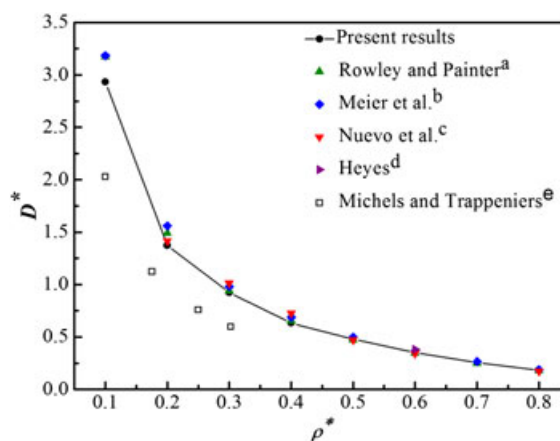


Figure 4. Self-diffusion coefficient D^* on the isotherm $T^* = 2.5$ as a function of density. ^aReference [12], ^bReference [2], ^cReference [21], ^dReference [24], and ^eReference [25].

The dependence of the calculated self-diffusion coefficient on temperature and density is shown in Figures 4 and 5. Comparisons with the reference data are also made. Figure 4 is under the supercritical isotherm $T^* = 2.5$ with the densities ranging from 0.1 to 0.8. Figure 5 is under the isochore $\rho^* = 0.6$ with a temperature span of $T^* = 0.71 \sim 4.45$. All these results are simulated with 1000 particles, and it determines the length of the simulation cell in the x direction. First, the self-diffusion coefficient decreases with a density increase or with a temperature decrease, which is really the physics of self-diffusion. Second, our USS scheme can give very close results compared with the previous works. Third, that the results from Michels and Trappeniers [25] have a big deviation from others' works may be due to the system size effect because their simulations were conducted with only 125 particles.

To check the convergence of the present USS scheme, we perform a direct comparison with Reference [2] in which the Einstein relation method was used. The simulation parameters agreeing with Reference [2] are chosen as follows: number of molecules $N = 1372$, time step $dt = 0.003\tau$, cutoff radius $r_{\text{cut}} = 5.5\sigma$, temperature $T^* = 3.0$, density $\rho^* = 0.2, 0.4, 0.6$, and 0.8 . Specifically, for our USS scheme the exchange interval W equals 10 and we use 10^5 time steps for the system to reach steady state and another 10^5 time steps to gather statistics. The calculated self-diffusion coefficients with comparison to those from Reference [2] are shown in Table I. Apparently, the relative errors for these four data points are all below 6%, which indicates a good agreement between

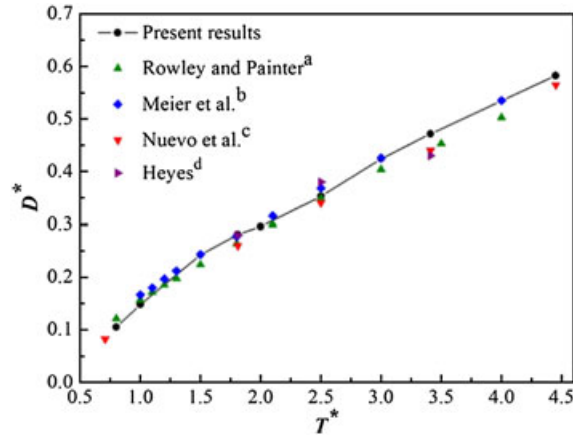


Figure 5. Self-diffusion coefficient D^* on the isochore $\rho^* = 0.6$ as a function of temperature. ^aReference [12], ^bReference [2], ^cReference [21], and ^dReference [24].

Table I. Self-diffusion coefficient obtained from Reference [2] and the present USS scheme under $T^* = 3.0$ for various densities. The statistical uncertainties are shown in the brackets.

| | $\rho^* = 0.2$ | $\rho^* = 0.4$ | $\rho^* = 0.6$ | $\rho^* = 0.8$ |
|---------------------------|----------------|----------------|----------------|----------------|
| D^*_{EMD} Reference [2] | 1.765 (~1.0%) | 0.771 (~0.5%) | 0.419 (~0.5%) | 0.217 (~0.5%) |
| D^*_{USS} | 1.872 (0.8%) | 0.746 (1.2%) | 0.395 (0.7%) | 0.205 (1.4%) |
| Relative error | 6.0% | 3.2% | 5.7% | 5.5% |

these two methods. In Reference [2], the authors claimed that the statistical uncertainty was estimated to be 0.5% at densities larger than $\rho^* = 0.2$, and 1% at lower densities. Also, the statistical uncertainties are gathered in our USS method shown in the brackets of Table I with similar value compared with the referenced data. Because of the different ensembles and integration algorithms of Reference [2] and the present work, the relative error between the two methods is larger in all cases than the corresponding statistical uncertainty. However, considering the small relative error of less than 6%, we can tell that the results are close enough, indicating the applicability of the USS method. It should be noted that the total simulation length in Reference [2] is $4500 \sim 6000\tau$ and we perform the USS simulation within 600τ . It shows that this USS method can converge faster than the EMD method without hurting the accuracy of the results.

For the system size dependence of the self-diffusion coefficient, lots of articles have carried out investigations into this problem. Erpenbeck [11] did a comparison between the Green–Kubo and NEMD self-diffusion constants on this topic and found that the results exhibited some clear conflicts between these two methods. Meier *et al.* [2] discussed this issue years later, and pointed out that the system size dependence was different at various densities for the EMD method. Almost at the same time, Yeh and Hummer [26] showed that the self-diffusion coefficient increased with the system size growing for EMD and even developed an equation for correction

$$D_{\infty}^* = D^* + \frac{T^* \xi}{6\pi \eta^* L^*}, \quad (10)$$

where η^* is the shear viscosity, L^* is the system length along the x direction, D^* is the simulated self-diffusion coefficient, and ξ is a parameter that approximately equals 2.837. Then from Equation (10) we can obtain the approximate self-diffusion coefficient for infinite system size.

Now, following the path of the predecessors, we will take a look at the system size dependence of the self-diffusion for the purpose of checking the USS scheme. We choose the state point of $\rho^* = 0.7$ and $T^* = 2.75$, just the same as Reference [26]. The exchange interval W for $N = 500, 1000, 1500$ is 50, 100 for $N = 2000$ and 200 for $N = 4000, 6000$. The time we use to reach steady state for

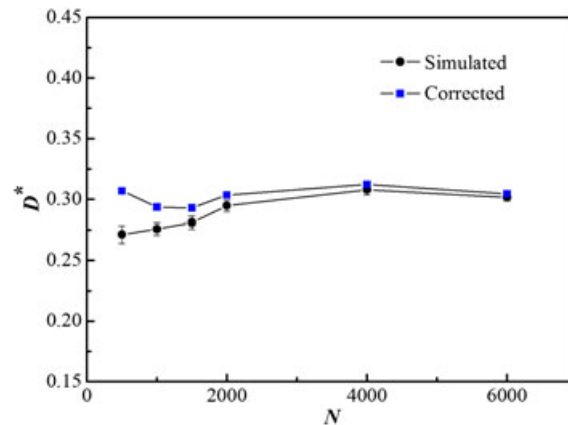


Figure 6. Dependence of the simulated and corrected self-diffusion coefficients on the system size.

the case of $N = 500 \sim 2000$ is 1500τ , 2500τ for $N = 4000$, and 5000τ for $N = 6000$. The length of the system in the x direction ranges from 8.9 to 107.3. The results are shown in Figure 6 with the corrected results using Equation (10) also included. The shear viscosity used in Equation (10) equals 1.31 [26]. First, the mean self-diffusion coefficient is 0.2886 by the six data points and the maximum relative error is only 6.6%. The mean results after correction is 0.3021 and the maximum relative error is 3.3%. Also, the relative differences between these two series of results decrease from 11.5% to 0.9% as the system grows larger. Therefore, from the above mentioned points we can conclude that the system size effect is very small for the scheme. Second, we extend the system size to a very large one ($N = 6000$) and previous articles seldom covered that region. Reasonably, the result with $N = 6000$ should be quite close to that of the real infinite size system and according to Reference [26] the corrected result is 0.3194. It indicates that our simulations are accurate. Third, with the increase of the system size, the self-diffusion coefficient slightly increases except the last point. However, keeping in mind that the last two points are both similar to the infinite case and considering the systematic error of the NEMD method, we can accept this small decline. In a word, the present USS scheme can give accurate results with fast convergence.

4. CONCLUSIONS

We extend the new NEMD approach called uniform source and sink scheme developed by Cao and Li from simulation of thermal conductivity to the self-diffusion coefficient. We recommend this NEMD method because it reveals some good features that can benefit the simulation process and results. First, in the simulation process of the self-diffusion coefficient, no density gradient is introduced, and it guarantees the system is homogeneous. Second, periodic boundary conditions and the USS scheme lead to a piecewise quadratic density profile. The self-diffusion coefficient can be extracted directly from the mean densities of the right and left half systems rather than by fitting the density gradient. Third, for every exchange interval, only two out of many particles in two slabs are involved in the exchange event. Compared with the whole size of the system, it is the slightest perturbation, and hence greatly shortens the time to relax. Fourth, because so many works have reported boundary jumps of some quantities (e.g., density, temperature, velocity) for the NEMD methods, which harm the efforts for MD simulations, the source and sink slabs are shifted away from boundaries and thus the boundary jump is completely eliminated. Apart from all these good features, this method is very simple to carry out because the label exchange is brief but reflects the physics of the self-diffusion process. This method yields reliable results compared with the reference data. Finally, we should point out that in the case of self-diffusion, the present USS scheme is an NEMD method but established on an equilibrium system.

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