Molecular Dynamics Simulations for Transport Coefficients of Liquid Argon : New Approaches

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The stress and the heat-flux auto-correlation functions in the Green-Kubo formulas for shear viscosity and thermal conductivity have non-decaying long-time tails. This problem can be overcome by improving the statistical accuracy by N (number of particles) times, considering the stress and the heat-flux of the system as properties of each particle. The mean square stress and the heat-flux displacements in the Einstein formulas for shear viscosity and thermal conductivity are non linear functions of time since the quantities in the mean square stress and the heat-flux displacements are not continuous under periodic boundary conditions. An alternative to these quantities is to integrate the stress and the heat-flux with respect to time, but the resulting mean square stress and heat-flux displacements are still not linear versus time. This problem can be also overcome by improving the statistical accuracy. The results for transport coefficients of liquid argon obtained are discussed.

Key Words : MD simulation. Diffusion. Shear viscosity. Thermal conductivity. Liquid argon

Introduction

Transport coefficients - self-diffusion coefficient, shear viscosity, and thermal conductivity - are defined in terms of the response of a system to a perturbation. For example, the diffusion coefficient relates the particle flux to a concentration gradient, whereas the shear viscosity is a measure of the shear stress induced by an applied velocity gradient. By introducing such perturbations into the Hamiltonian, or directly into the equations of motion, their effect on the distribution function may be calculated. By retaining the linear terms in the perturbation, comparing the equation for the response with a macroscopic transport equation, we can identify the transport coefficient. This is usually the infinite time integral of an equilibrium correlation function of the form known as the Green-Kubo formula:

$$\gamma = \int_0^\infty dt \langle \dot{A}(0) \cdot \dot{A}(t) \rangle . \tag{1}$$

where γ is the transport coefficient and A is a variable appearing in the perturbation term in the Hamiltonian. Associated with any expression of this kind, there is also the Einstein formula:

$$\gamma = \frac{1}{2} \lim_{t \to \infty} \frac{d}{dt} \langle [A(t) - A(0)]^2 \rangle . \tag{2}$$

which holds at large t (compared with the correlation time of A).

In recent years, non-equilibrium molecular dynamics (NEMD) simulations have emerged as a powerful tool for the study of transport coefficients of both simple and molecular fluids.[1-20] The general principle of the NEMD method[21] is to introduce a (possibly fictitious) external field X into the equations of motion of the system, which derives

the corresponding thermodynamic flux J. The first requirement for this applied field is that it should be consistent with the periodic boundary conditions to ensure that the simulation box remains homogeneous. The second requirement is that the transport coefficient γ of interest can be calculated from the constitutive relation:

$$\gamma = \lim_{X \to 0} \left[\lim_{t \to \infty} \frac{\langle J(t) \rangle}{X} \right]. \tag{3}$$

The formal proof that an algorithm satisfies these two requirements is given by linear response theory.[15,22,23]

Eq. (3) indicates an extrapolation method - applying 10-20 values of the external field, obtaining the thermodynamic flux, and extrapolating to zero external field. This kind of calculation is fairly expensive. Especially, the averaging of the thermodynamic flux at small external fields requires very long time simulations. Another drawback of NEMD compared with equilibrium molecular dynamics (EMD) simulation is to provide only one fundamental transport coefficient, while EMD produces an entire range of correlation functions and transport coefficients from the output of a single equilibrium run by Green-Kubo and Einstein formulas. The problem in the calculation of transport properties, using the EMD simulation is a non-decaying long-time tail of the time correlation functions in Eq. (1) or a non-linear slope in the mean square displacement of the variables in Eq. (2).

In the present paper, we examine the Green-Kubo and Einstein formulas for the calculation of the transport properties of liquid argon, using the EMD simulation. The primary goal of this study is to seek an alternate route for the non-decaying long-time tail of the time correlation functions in Eq. (1) or the non-linear slope in the mean square displacement of the variables in Eq. (2). In the following section, we describe the technical details of MD simulation. We present some theoretical aspects in Section III, our results in Section IV, and concluding remarks in Section V.

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Molecular Dynamics Simulation Details

The usual Lennard-Jones (LJ) 12-6 potential for the interaction between atoms was used with LJ parameters. $\sigma = 0.34$ nm and $\varepsilon/k = 120$ K. where k is the Boltzmann constant. The inter-atomic potential was truncated at 0.85 nm. which is the cut-off distance used in many other simulations. Long-range corrections were applied to the energy, pressure. etc. due to the potential truncation.[24] The preliminary canonical ensemble (NVT fixed) MD simulation of 1728 argon atoms was started in the cubic box of length L = 4.3696 nm. of which the density is equal to 1.374 g/cm³ at 94.4 K and 1 atm. First. the equations of motion were solved using the leap-frog algorithm[25] with a time step of 10^{-14} second. but later it was switched to a velocity Verlet algorithm[26] for NVT EMD simulation and a fifth-order predictor-corrector Gear integration[27] for NpT EMD simulation. Both systems were fully equilibrated and the equilibrium properties were averaged over five blocks of 100.000 time steps. The brief summary of thermodynamic averages is given in Table 1. The configurations of argon atoms were stored every time step for further analysis.

Theoretical Backgrounds

Self-diffusion coefficient. The Green-Kubo formula for self-diffusion coefficient D_s in three dimensions is given by

$$D_s = \frac{1}{3} \int_0^\infty dt \langle v_i(0) \cdot v_i(t) \rangle, \tag{4}$$

while the Einstein formula for self-diffusion coefficient is

$$D_s = \frac{1}{6} \lim_{t \to \infty} \frac{d}{dt} \langle |r_i(t) - r_i(0)| \rangle^2. \tag{5}$$

These averages are computed for each of the N particles in the simulation. the results were added together. and divided by N. to improve statistical accuracy.

Shear viscosity. The Green-Kubo formula for shear viscosity η is given by

$$\eta = \frac{V}{kT} \int_0^\infty dt \langle P_{\alpha\beta}(0) \cdot P_{\alpha\beta}(t) \rangle, \tag{6}$$

where

$$P_{\alpha\beta}(t) = \frac{1}{V} \left[\sum_i m v_{i\alpha}(t) \cdot v_{i\beta}(t) + \sum_i \sum_{j>i} r_{ij\alpha}(t) \cdot f_{ij\beta}(t) \right] \tag{7}$$

or $P_{\alpha\beta}(t) = \frac{1}{V} \left[\sum_i m v_{i\alpha}(t) \cdot v_{i\beta}(t) + \sum_i r_{i\alpha}(t) \cdot f_{i\beta}(t) \right]$ (8)

and $\alpha\beta = xy$. xz. yx. yz. zx. and zy. The equality of these two

Table 1. Liquid argon systems at T = 94.4 K

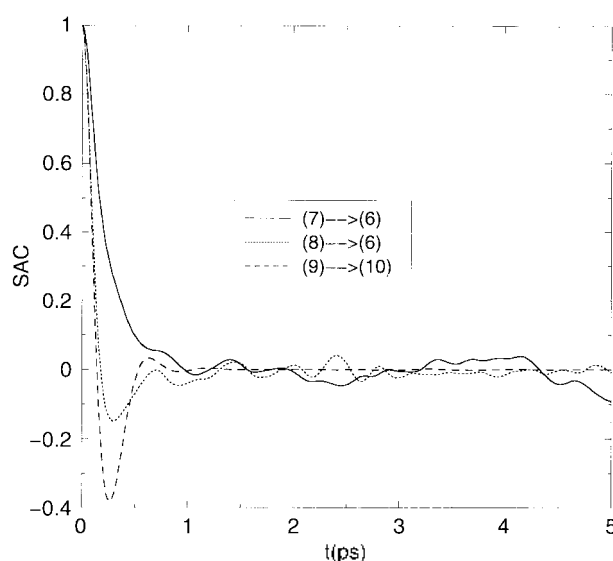| Methods	ρ (g/cm³)	p (atm)	-Etotal (kJ/mol)
Verlet NVT EMD	1.374	38	4.641
Gear NpT EMD	1.361	1.0	4.592

Figure 1. Normalized stress auto-correlation (SAC) functions of liquid argon in Eq. (6) using the stress tensors defined by Eqs. (7) and (8), and that in Eq. (10) using Eq. (9), obtained Verlet NVT EMD simulations.

stress tensors without potential truncation is fully discussed[28] and it is recommended to use the former form in a simulation that employs periodic boundary conditions. The stress autocorrelation (SAC) functions. the integrand of Eq. (6). obtained from Eqs. (7) and (8) do not decay to zero in the long time as shown in Figure 1 and the resulting shear viscosity is not well-defined.

Though the stress is a property of the system. it can be considered as a property of each particle i. writing $P_{i\alpha\beta}$ as

$$P_{i\alpha\beta}(t) = \frac{1}{V} [m v_{i\alpha}(t) \cdot v_{i\beta}(t) + r_{i\alpha}(t) \cdot f_{i\beta}(t)]. \tag{9}$$

With this expression. η in Eq. (6) may be rewritten as

$$\eta = \frac{V}{kT} \int_0^\infty dt \sum_i \langle P_{i\alpha\beta}(t) \cdot P_{i\alpha\beta}(t) \rangle. \tag{10}$$

Then the statistical accuracy is improved. being N (number of particles) as large as that of using Eq. (7) or (8). Figure 1 shows a perfect decay of the SAC. the integrand of Eq. (10). This kind of treatment in calculating shear viscosity has already appeared in solute molecular dynamics in a mesoscale solvent.[29] Since there is no interaction between solvent particles in the mesoscale solvent. the second term of Eq. (9) vanishes and $v_{i\beta}$ is treated as cell-jump velocity.[29]

The Einstein formula for shear viscosity is

$$\eta = \frac{1}{2} \frac{V}{kT} \lim_{t \to \infty} \frac{d}{dt} \langle [Q_{\alpha\beta}(t) - Q_{\alpha\beta}(0)]^2 \rangle. \tag{11}$$

where

$$Q_{\alpha\beta}(t) = \frac{1}{V} \sum_i r_{i\alpha}(t) \cdot p_{i\beta}(t). \tag{12}$$

In the previous section time differentiating of r_i in Eq. (5) gives v_i in Eq. (4) and the same relationship stands for $Q_{\alpha\beta}$ in Eq. (12) and $P_{\alpha\beta}$ in Eq. (8) - time differentiating of $Q_{\alpha\beta}$ gives $P_{\alpha\beta}$. Since $Q_{\alpha\beta}$ is not continuous under periodic boundary
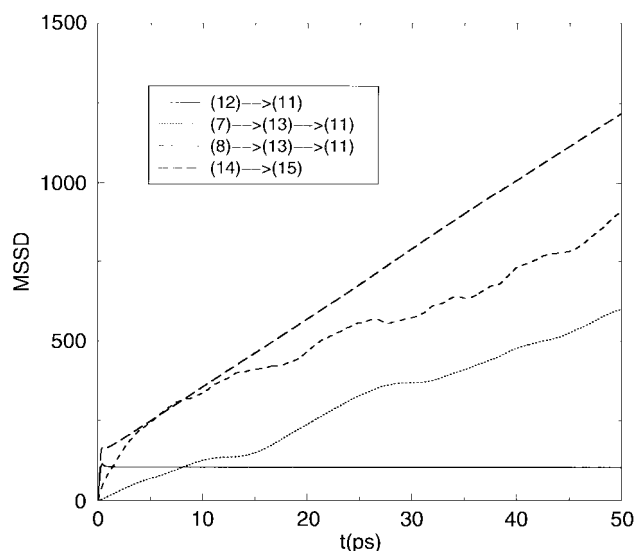
Figure 2. Mean square stress displacement (MSSD) of liquid argon in Eq. (11) using $Q_{\alpha\beta}$ defined by Eq. (12), those in Eq. (11) using $Q_{\alpha\beta}$ defined by Eq. (13) through $P_{\alpha\beta}$ of Eqs. (7) and (8), and that in Eq. (15) using Eq. (14), obtained Verlet NVT EMD simulations. The units are in $(g/(mol \cdot ps \cdot nm))^2$.

conditions.[30] the calculated mean square stress displacement (MSSD), using Eq. (12) is not a linear function of time as shown Figure 2. An attractive alternative for $Q_{\alpha\beta}$ is to integrate $P_{\alpha\beta}$ with respect to time:

$$Q_{\alpha\beta}(t) - Q_{\alpha\beta}(0) = \int_0^t d\tau \, P_{\alpha\beta}(\tau) . \tag{13}$$

There are two $P_{\alpha\beta}$ in Eqs. (7) and (8) for Eq. (13) but both calculated MSSD of $Q_{\alpha\beta}$ are also non linear functions of time as shown Figure 2.

To improve the statistical accuracy, we define $Q_{i\alpha\beta}$ as a property of each particle i from Eq. (9):

$$Q_{i\alpha\beta}(t) - Q_{i\alpha\beta}(0) = \int_0^t d\tau \, P_{i\alpha\beta}(\tau) , \tag{14}$$

and the shear viscosity is defined as

$$\eta = \frac{1}{2kT} \frac{V}{T} \lim_{t \to \infty} \frac{d}{dt} \sum_i \langle [Q_{i\alpha\beta}(t) - Q_{i\alpha\beta}(0)]^2 \rangle . \tag{15}$$

The calculated MSSD of $Q_{i\alpha\beta}$ gives a perfect linear function of time as shown Figure 2.

Thermal conductivity. The discussion here is exactly the same as the previous section. First, the Green-Kubo formula for thermal conductivity λ is given by

$$\lambda = \frac{V}{kT^2} \int_0^\infty dt \langle J_{Q\alpha}(0) \cdot J_{Q\alpha}(t) \rangle . \tag{16}$$

where

$$J_{Q\alpha}(t) = \frac{1}{V} \left[\sum_i \varepsilon_i(t) \cdot v_{i\alpha}(t) + \sum_i \sum_{j>i} r_{ij\alpha}(t) \cdot [v_{ij}(t) \cdot f_{ij}(t)] \right] \tag{17}$$

or

$$J_{Q\alpha}(t) = \frac{1}{V} \left[\sum_i \varepsilon_i(t) \cdot v_{i\alpha}(t) + \sum_i r_{i\alpha}(t) \cdot [v_i(t) \cdot f_i(t)] \right] . \tag{18}$$
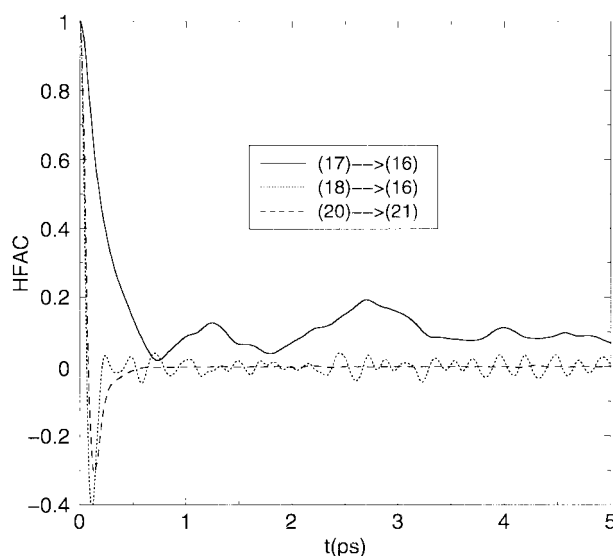

Figure 3. Normalized heat-flux auto-correlation (HFAC) functions of liquid argon in Eq. (16) using the hea flux defined by Eqs. (17) and (18), and that in Eq. (21) using Eq. (20), obtained Verlet NVT EMD simulations.

$$\varepsilon_i(t) = \frac{p_i(t)^2}{2m_i} + \frac{1}{2} \sum_{j \neq i} \phi[r_{ij}(t)] . \tag{19}$$

and $\alpha = x, y,$ and $z.$ $\phi[r_{ij}(t)]$ is the potential energy between particles i and j at time $t.$ The heat-flux auto-correlation (HFAC) functions, the integrand of Eq. (16), obtained from Eqs. (17) and (18) do not decay to zero as shown in Figure 3 and the resulting thermal conductivity is also not well-defined.

Again the heat flux is considered as a property of each particle i, writing $J_{Q i\alpha}$ as

$$J_{Q i\alpha}(t) = \frac{1}{V} [\varepsilon_i(t) \cdot v_{i\alpha}(t) + r_{i\alpha}(t) \cdot [v_i(t) \cdot f_i(t)]] \tag{20}$$

With this expression, λ in Eq. (16) may be rewritten as

$$\lambda = \frac{V}{kT^2} \int_0^\infty dt \sum_i \langle J_{Q i\alpha}(0) \cdot J_{Q i\alpha}(t) \rangle . \tag{21}$$

Here again the statistical accuracy is improved, being N (number of particle) as large as that of using Eq. (17) or (18). Figure 3 shows a perfect decay of the HFAC, the integrand of Eq. (21).

The Einstein formula for thermal conductivity is

$$\lambda = \frac{1}{2kT^2} \frac{V}{\lim_{t \to \infty}} \frac{d}{dt} \langle [\varepsilon_\alpha(t) - \varepsilon_\alpha(0)]^2 \rangle . \tag{22}$$

where

$$\varepsilon_\alpha(t) = \frac{1}{V} \sum_i r_{i\alpha}(t) \cdot \varepsilon_i(t) . \tag{23}$$

or sometimes

$$\varepsilon_\alpha(t) = \frac{1}{V} \sum_i r_{i\alpha}(t) \cdot [\varepsilon_i(t) - \langle \varepsilon_i \rangle] . \tag{24}$$

Here again time differentiating of ε_α in Eq. (23) gives $J_{Q\alpha}$ in

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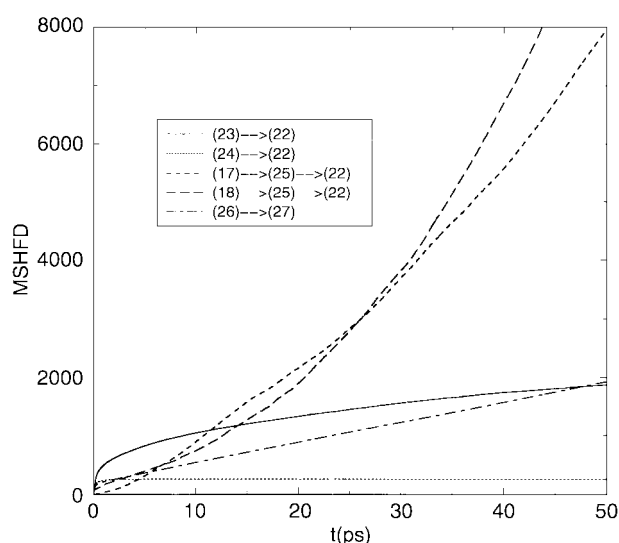*Bull. Korean Chem. Soc.* **2003**, Vol. 24, No. 2   181

Figure 4. Mean square heat-flux displacements (MSHFD) of liquid argon in Eq. (22) using ε_α defined by Eqs. (23) and (24), those in Eq.(22) using ε_α defined by Eq. (25) through $J_{Q\alpha}$ of Eqs. (17) and (18), and that in Eq. (26) using Eq. (27), obtained Verlet NVT EMD simulations. Here MSHFD is multiplied by V and the units are in $(g/(molp\cdot s^2))^2 nm^3$.

Eq. (18). Since ε_α is not continuous under periodic boundary conditions,[29] the calculated mean square heat-flux displacements (MSHFD) using Eqs. (23) and (24) are non linear functions of time as shown Figure 4. An attractive alternative for ε_α is to integrate $J_{Q\alpha}$ with respect to time:

$$\varepsilon_\alpha(t) - \varepsilon_\alpha(0) = \int_0^t d\tau\, J_{Q\alpha}(\tau).\qquad(25)$$

There are two $J_{Q\alpha}$ Eqs. (17) and (18), for Eq. (25) and both calculated MSHFD's are also non linear functions of time as shown Figure 4.

Again, to improve the statistical accuracy, we define $J_{Q i\alpha}$ as a property of each particle i from Eq. (20):

$$\varepsilon_{i\alpha}(t) - \varepsilon_{i\alpha}(0) = \int_0^t d\tau\, J_{Q i\alpha}(\tau).\qquad(26)$$

and the thermal conductivity is defined as

$$\lambda = \frac{1}{2}\frac{V}{kT^2}\lim_{t\to\infty}\frac{d}{dt}\sum_i \left\langle [\varepsilon_{i\alpha}(t)-\varepsilon_{i\alpha}(0)]^2\right\rangle.\qquad(27)$$

The calculated MSHFD gives a perfect linear function of time as shown Figure 4.

Results and Discussion

The calculated velocity auto-correlation (VAC) function, the integrand of Eq. (4), decays to zero quickly(not shown) and the resulting self-diffusion coefficient is well-defined. Also, the calculated mean square displacement (MSD) is perfectly linear (not shown) and one-sixth of the slope gives the self-diffusion coefficient. The self-diffusion coefficients obtained from VAC (Eq. (4)) and MSD (Eq. (5)) are listed in Table 2 and the agreement with the experimental results[31] is quite good. But non-equilibrium molecular dynamics (NEMD)

Table 2. Comparison of self-diffusion coefficients (D_s, 10^{-5} cm²/ sec), shear viscosities (η, millipoise), and thermal conductivities (λ, 10^{-4} cal/K·cm·sec) of liquid argon at 94.4 K obtained from the Green-Kubo [Eq. (4), (10), and (21)] and Einstein formulas [Eq. (5), (15), and (27)], with the experimental measures and NEMD results[32]

Transport properties	Verlet NVT EMD	Gear NpT EMD	Experimental	NEMD
Ds (Eq.4)	2.48 ± 0.07	2.56 ± 0.04	2.83[a]	4.02
Ds (Eq.5)	2.47 ± 0.07	2.54 ± 0.05	2.43[b]	
η (Eq.10)	3.08 ± 0.19	3.14 ± 0.22	1.97[a]	2.13
η (Eq.15)	3.15 ± 0.23	3.01 ± 0.19		
λ (Eq.21)	3.05 ± 0.22	3.54 ± 0.18	2.74[a]	2.31
λ (Eq.27)	3.07 ± 0.24	3.37 ± 0.21		

[a]Obtained from Lagrange interpolation of experimental results[31] at 94.4 K. [b]At 90 K and 1.374 g/cm³.

result[32] overestimates the experimental results.[31]

Figure 5 shows the shear viscosities of liquid argon, time-integrated from the stress auto-correlation functions(SAC), Figure 1, as a function of time, and only the last result gives a constant value in the long time, which is listed in Table 2. The mean square stress displacements(MSSD) of liquid argon are shown in Figure 2. The only linear curve among them is the last one and the obtained shear viscosity is listed in Table 2. The obtained shear viscosities from the Green-Kubo formulas, using $P_{i\alpha\beta}$ in Eqs. (9) and (10) and the Einstein formulas, using $Q_{i\alpha\beta}$ in Eqs. (14) and (15) in velocity Verlet algorithm[26] NVT and fifth-order predictor-corrector Gear integration[27] NpT EMD simulations are generally in good agreement but overestimate the experimental result.[31] However, NEMD result[32] gives a better agreement with the experimental result[31] than EMD results.

The results for the thermal conductivity of liquid argon have a very similar situation to those for the shear viscosity.
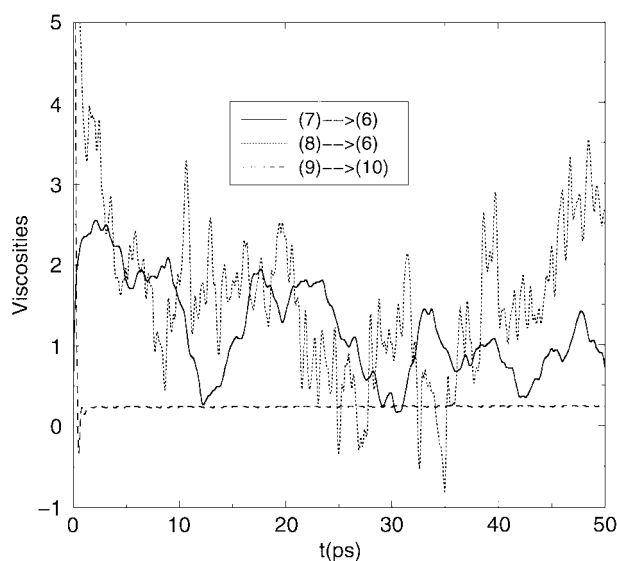

Figure 5. The shear viscosities (arbitrary units) of liquid argon, time-integrated from the stress auto-correlation functions (SAC), Figure 1, as a function of time.
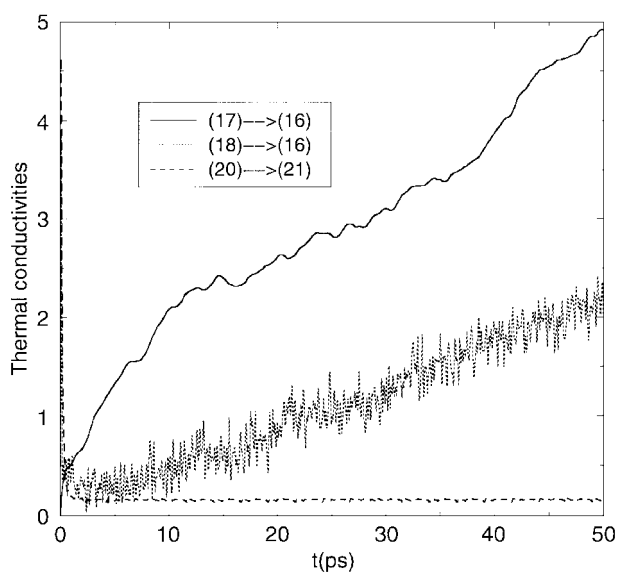
Figure 6. The thermal conductivities (arbitrary units) of liquid argon, time-integrated from the heat-flux auto-correlation functions (HFAC), Figure 3, as a function of time.

In Figure 6, we show the thermal conductivities of liquid argon, time-integrated from the heat-flux auto-correlation functions (HFAC). Figure 3, as a function of time, and only the last result, gives a constant value in the long time, which is listed in Table 2. The mean square heat-flux displacements (MSHFD) of liquid argon are shown in Figure 4. The only linear curve among them is the last one and the obtained thermal conductivity is listed in Table 2. The thermal conductivities obtained from the Green-Kubo formulas, using $J_{Qi\alpha}$ in Eqs. (20) and (21) and the Einstein formulas, using $\varepsilon_{i\alpha}$ in Eqs. (26) and (27) in the velocity Verlet algorithm[26] NVT EMD are slightly lower than that obtained from those from the fifth-order predictor-corrector Gear integration[27] NpT EMD simulations but also overestimate the experimental result.[31] Unlike the viscosity result, the EMD result gives a slightly better agreement with the experimental result[31] than NEMD result.[32]

Concluding Remarks

In the present paper, we examine the Green-Kubo and Einstein formulas for the calculation of the transport properties of liquid argon, using the equilibrium molecular dynamics (EMD) simulation. For self-diffusion coefficient, there is no problem since the velocity auto-correlation (VAC) function decays to zero quickly and the mean square displacement (MSD) gives a linear line. This is because the velocity and position of each particle are properties of particle. However, for shear viscosity and thermal conductivity, the stress (SAC) and the heat-flux auto-correlation (HFAC) functions in the Green-Kubo formulas have non-decaying long-time tails, and the mean square stress (MSSD) and heat-flux displacements (MSHFD) in the Einstein formulas are non linear functions of time. This is because the stress and the heat flux are not properties of particle but of system. By

considering the stress and the heat-flux of the system as properties of each particle, this problem can be overcome by improving the statistical accuracy by N (number of particles) times. The obtained results for shear viscosities and thermal conductivities of liquid argon from the Green-Kubo and Einstein formulas using velocity Verlet algorithm NVT EMD and fifth-order predictor-corrector Gear integration NpT EMD simulations give a reasonable agreement with the experimental results and NEMD results. The applications of this method for molecular liquids, such as H_2O, CH_4, and C_4H_{10} are presently under study.

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