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Computation of Shear Viscosity: A Systems Approach

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Abstract— **Macroscopic material transport properties such as viscosity, diffusivity, conductivity, etc., may be computed by using molecular level simulation such as molecular dynamics or Monte Carlo methods. This computation is time consuming since simulations over sufficiently long times are needed to ensure that the assumed statistical properties are satisfied. As a result, such tools are useful in gaining insight and understanding of the underlying mechanisms behind observed physical phenomena, but are not amenable to material property design or material process control. In this paper, we focus on the computation of shear viscosity of a fluid-like material. We take a systems approach by regarding viscosity as a scalar input/output map from shear stress to shear strain rate. Linearizing this map about an equilibrated trajectory results in a linear time varying system. By freezing the time along the equilibrated trajectory, we obtain a set of linear time invariant systems. These systems are usually unstable, but may be transformed to stable systems by weighing all signals with sufficiently fast decaying exponential functions. Viscosity is then estimated directly from the frequency responses of these systems. Model reduction such as approximate balanced truncation may be applied to further reduce model complexity and computation load. This approach has a potential computation advantage since extensive simulation runs using high order molecular dynamics model are not required. Our long term objective is to develop efficient computation methods to facilitate rapid material and process design iterations. To illustrate the approach described in this paper and compare it with the traditional molecular dynamics methods, we have included the simulation results involving a simple Leonard-Jones fluid.**

I. Introduction

Molecular simulation has long been used to gain understanding of phenomena observed in physical systems [1]. In particular, macroscopic transport properties may be computed by using molecular simulation methods such as the molecular dynamics or Monte Carlo methods. There are two types of molecular dynamics approaches: equilibrium molecular dynamics (EMD) and non-equilibrium molecular dynamics (NEMD).

In the EMD approach, transport properties are obtained based on the unforced response of a collection of energy conserving molecules by using the so-called Green-Kubo relation [2], [3]. The basis of which is the observation that the molecular response due to a weak external excitation decays in the same way as a spontaneous fluctuation at an equilibrium [4]. The drawback of this method is that simulations must be run for very long times with many particles to get a reasonable approximation of the ensemble average.

NEMD is a more direct approach that applies a perturbation to the collection of molecules and calculates the transport coefficients from the resulting response. In the case of shear viscosity, the perturbation may be an imposed shear strain rate and the response is shear stress [5]; or the perturbation could be an imposed shear stress with shear strain rate as the response [6].

As these simulation-based molecular dynamics approaches are generally time consuming, they are useful in gaining insight and understanding of the underlying mechanisms behind observed physical phenomena, but are not amenable to material property design or material process control. The goal of this paper is to present an alternate approach to calculate shear viscosity based on a systems perspective of the NEMD model. Our ultimate goal is to develop an efficient means to calculate transport coefficients, and to use it as a tool for material design optimization and process control. We regard viscosity as a scalar input/output map from shear stress to shear strain rate. Linearizing this map about an equilibrated trajectory results in a linear time varying (LTV) system. We fix the times along the equilibrated trajectory to obtain a set of linear time invariant (LTI) systems. However, these systems are usually unstable. By multiplying all signals with a sufficiently fast decaying exponential, we transform these systems to stable systems. Matching with the corresponding response of the Navier-Stokes equation, we compute the viscosity directly from the DC (i.e., steady state) gain of these LTI systems. This approach has a potential computation advantage since extensive simulation runs are not required and well developed model reduction tools for LTI systems can also be applied.

To illustrate the approach described in this paper, we have included the simulation results based on a simple Leonard-Jones (LJ) fluid, and compare them with the results using the traditional EMD and NEMD methods.

Notation: The coordinate of particle i is denoted by $r_i \in \mathbb{R}^3$. The coordinates of a system of n particles are combined into a vector $r \in \mathbb{R}^{3n}$. The corresponding velocity and acceleration are denoted by $\dot{r}$ and $\ddot{r}$, respectively.

II. Viscosity Estimation Methods

A. Equilibrium Molecular Dynamics

The EMD approach considers a collection of energy-conserving particles with potential energy $P(r)$ and normalized kinetic energy $K = \frac{1}{2}\|\dot{r}\|^2$. The equation of motion is

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given by the Newton's Second Law:

$$\ddot{r} + \frac{\partial P}{\partial r} = 0. \tag{1}$$

Transport properties may be obtained through the EMD simulation by using the Green-Kubo relations.

Specifically for the shear viscosity, the Green-Kubo relation is given by:

$$\eta^{xy} = \frac{1}{Vk_BT}\int_0^\infty \langle \sigma^{xy}(0)\sigma^{xy}(t)\rangle \, dt \tag{2}$$

where $\langle\cdot\rangle$ denotes ensemble average but computed using time average (based on the ergodicity assumption), V is the volume, k_B the Boltzmann constant, T the temperature, and σ^{xy} the shear stress given by:

$$\sigma^{xy} = \sum_{i=1}^{N}\left(v_i^x v_i^y + \frac{1}{2}\sum_{j\neq i}^{N}(x_i - x_j) f_y(r_i - r_j)\right)$$

where x_i is the x component of r_i, and (v_i^x, v_i^y) are the (x, y) components of $\dot{r}_i$.

B. Non-Equilibrium Molecular Dynamics

The NEMD approach directly imposes an external perturbation. In the case of shear viscosity, one approach applies a shear force and extracts the corresponding steady state shear strain rate response [6]. The second approach imposes a shear strain rate and computes the corresponding shear stress [5].

In this paper, we consider the formulation in [6]. The basic fluid behavior can be described by the following equations:

$$\nabla \cdot \mathbf{J} = -(\rho/m)\mathbf{F}(\mathbf{r}, t) + \rho\dot{\mathbf{u}} \tag{3}$$

$$\mathbf{J} = -p\mathbf{I} - \eta_B(tr\mathbf{E})\mathbf{I} + 2\eta\mathbf{E} \tag{4}$$

$$\mathbf{E} = 1/2\,(\nabla\mathbf{u} + (\nabla\mathbf{u})^*) \tag{5}$$

Equation (3) is the equilibrium equation of motion, (4) the constitutive relation, and (5) the fluid kinematics. Collectively these equations are known as the Navier-Stokes equations. $\mathbf{J}$ is the Newtonian stress tensor, ρ is the mass density, m is the mass, $\mathbf{F}(\mathbf{r}, t)$ is an external force field, $\dot{\mathbf{u}}$ is the material derivative of the velocity field, p is the pressure, η_B is the bulk viscosity, and $\mathbf{E}$ is the strain rate tensor. If the applied force force $\mathbf{F}(\mathbf{r}, t)$ is in the x direction only and varies only with z, i.e.,

$$\mathbf{F} = \begin{bmatrix} F_x(t, z) & 0 & 0 \end{bmatrix}^T$$

then the resulting steady state velocity will also have the same property, i.e., only in the x direction and dependent only on z. Then (4) and (5) reduce to:

$$\mathbf{J} = \begin{bmatrix} 0 & 0 & \eta\frac{\partial u_x}{\partial z} \\ 0 & 0 & 0 \\ \eta\frac{\partial u_x}{\partial z} & 0 & 0 \end{bmatrix} - p\mathbf{I}$$

Substitution into (3) yields the Laplace's equation:

$$\rho\frac{\partial u_x}{\partial t} = \eta\frac{\partial^2 u_x}{\partial z^2} + \frac{\rho}{m}F_x(t, z). \tag{6}$$

We choose the forcing function profile to be:

$$F_x(t, z) = F_0(t)\sin(2\pi i z/L) \tag{7}$$

where i is any positive integer, L is the boundary of the periodic box, and F_0 is the magnitude of the input force field. Assume the fluid is initially at rest. Because the external force is zero at the boundary, u_x is assumed to be also zero at the boundaries:

$$u_x(t, 0) = u_x(t, L) = 0. \tag{8}$$

The operator $\frac{\eta}{\rho}\frac{\partial^2}{\partial z^2}$ is self-adjoint and negative definite. Therefore, its spectrum consists of only negative eigenvalues λ, and orthonormal eigenfunctions ϕ:

$$\lambda_\ell = -\frac{\eta 4\pi^2\ell^2}{\rho L^2},\ \ell = 1, 2, \ldots \tag{9}$$

$$\phi_\ell = \sqrt{2/L}\sin(2\pi\ell z/L). \tag{10}$$

The solution to (6), $u_x(t, z)$, can now be modally decomposed as:

$$u_x(t, z) = \sum_{\ell=1}^{\infty} q_\ell(t)\phi_\ell(z) \tag{11}$$

where the dynamics of the modal amplitude q_ℓ are decoupled and satisfy:

$$\dot{q}_\ell = -\frac{\eta 4\pi^2\ell^2}{\rho L^2}q_\ell + \frac{\langle F_x, \phi_\ell\rangle}{m}. \tag{12}$$

With $F_x(t, z)$ given by (7), the forcing terms are zero for all ℓ except when $\ell = i$. In this case, the dynamics is completely given by the first order ordinary differential equation (ODE):

$$\dot{q}_i = -\frac{\eta 4\pi^2 i^2}{\rho L^2}q_i + \sqrt{\frac{L}{2}}\frac{F_0}{m}, \tag{13}$$

and the velocity field is given by (11):

$$u_x(t, z) = \sqrt{\frac{2}{L}}q_i(t)\sin(2\pi i z/L). \tag{14}$$

The steady state q_i is

$$q_{i_{ss}} = \sqrt{\frac{L}{2}}\frac{\rho L^2}{\eta 4\pi^2 i^2}\frac{F_0}{m}. \tag{15}$$

Denote the amplitude of the steady state velocity field by

$$u_0 := \sqrt{\frac{2}{L}}q_{i_{ss}}.$$

Combining with (15), the viscosity η may be estimated based on F_0 and u_0. We choose $i = 1$, then the η estimate becomes:

$$\eta = \underbrace{\frac{\rho L^2}{4\pi^2 m}}_{:=k}\frac{F_0}{u_0}. \tag{16}$$

To obtain the viscosity estimate from an NEMD simulation, we impose the sinusoidal force profile described above and measure the resulting steady-state velocity profile $u_{x_{ss}}(z)$.

The magnitude of the applied force F_0 should be large enough to obtain a well defined drift velocity profile and small enough for linearization to hold. We choose F_0 to be the average magnitude of the molecular force when the input is zero (i.e., an EMD simulation). Since the applied force is only in the x-direction, we divide this average force by 3.

III. Viscosity based on Linearization of NEMD

A. Equation of Motion

The equation of motion of the NEMD model in Section II-B may be written as:

$$\ddot{r} + \frac{\partial P(r)}{\partial r} = g(r)F \tag{17}$$

where $F = F_0$ and $g(r)$ provides the desired force profile:

$$g(r) = \left[\begin{array}{ccccccc} \sin(2\pi z_1/L) & 0 & 0 & \sin(2\pi z_2/L) & \dots \end{array} \right]^T \tag{18}$$

From (14), the expected solution to Navier-Stokes equation is $\dot{r} = g(r)y$, where $y = u_0$. Therefore,

$$y = \frac{g^T(r)}{g^T(r)g(r)}\dot{r}, \tag{19}$$

which means that y is the least square fit of the measured velocity profile to the expected velocity profile. Following the NEMD approach, the viscosity may be estimated to be a constant multiplied by the ratio between the steady state input and output:

$$\eta = k\frac{F}{y}. \tag{20}$$

B. Addition of Damping

The forced system (17) has no damping which does not agree with the viscous behavior of the physical fluid. The NEMD approach fixes the problem by scaling the particle velocities to maintain a constant temperature (effective damping). This motivates us to also add a simple linear damping term to the NEMD model (17):

$$\ddot{r} + \alpha\dot{r} + \frac{\partial P(r)}{\partial r} = g(r)F. \tag{21}$$

The kinetic energy now evolves according to

$$\frac{d}{dt}\left(\frac{1}{2}\dot{r}^T\dot{r}\right) = \dot{r}^T\left(-\alpha\dot{r} - \frac{\partial P}{\partial r} + g(r)F\right).$$

To ensure that the temperature (and hence kinetic energy) remains constant, we need to set α to

$$\alpha = \left(\frac{\dot{r}^T g(r)F - \dot{r}^T\frac{\partial P}{\partial r}}{\|\dot{r}\|^2}\right). \tag{22}$$

An NEMD simulation after equilibration would provide an estimate of α. Notice this α depends on the magnitude of F.

C. Linearization and State Space Model

Since the transport property of interest (viscosity) is based on the linear response theory (due to small external perturbations), it is natural to apply linearization to the nonlinear state space model (21) and (19). Let $(r^*, \dot{r}^*)$ be the unforced ($F = 0$) response of (17) and (19) (obtained from an EMD simulation). A small external perturbation F will result in $(r, \dot{r})$ that approximately satisfies the linearized system:

$$\begin{aligned}(\ddot{r} - \ddot{r}^*) + \alpha(\dot{r} - \dot{r}^*) + \frac{\partial^2 P(r^*)}{\partial r^2}(r - r^*) &= g(r^*)F - \alpha\dot{r}^* \\ y - y^* &= \underbrace{\left(\frac{\dot{r}^{*T}\frac{\partial g(r^*)}{\partial r}}{\|g(r^*)\|^2} - 2\frac{g^T(r^*)\dot{r}^* g^T(r^*)\frac{\partial g(r^*)}{\partial r}}{\|g(r^*)\|^4}\right)}_{C_1}(r - r^*) \\ &\quad + \underbrace{\frac{g^T(r^*)}{\|g(r^*)\|^2}}_{C_2}(\dot{r} - \dot{r}^*).\end{aligned} \tag{23}$$

Define the state as

$$x := \left[\begin{array}{c} r - r^* \\ \dot{r} - \dot{r}^* \end{array} \right].$$

We then obtain the linearized state space model with a scalar input and a scalar output:

$$\begin{aligned}\dot{x} &= Ax + BF + B_0\dot{r}^* \\ y - y^* &= Cx + Du\end{aligned} \tag{24}$$

where

$$\begin{aligned}A &= \left[\begin{array}{cc} 0 & I \\ -\frac{\partial^2 P(r^*)}{\partial r^2} & -\alpha I \end{array} \right],\ B = \left[\begin{array}{c} 0 \\ g(r^*) \end{array} \right] \\ B_0 &= \left[\begin{array}{c} 0 \\ -\alpha I \end{array} \right],\ C = \left[\begin{array}{cc} C_1 & C_2 \end{array} \right],\ D = 0.\end{aligned}$$

This system is time varying in general since $(r^*, \dot{r}^*)$ is time varying. If the time variation is sufficiently slow, we can instead consider a series of "frozen" (in time) systems, where $(r, \dot{r})$ is in the neighborhood of $(r^*(t), \dot{r}^*(t))$ for some t. However, these systems are usually unstable, preventing any steady and frequency response analysis. To address this problem, we apply the following time-dependent coordinate transformation:

$$r_1 = e^{-\beta t}(r - r^*), \tag{25}$$

for some $\beta > 0$. Multiplying both sides of the linearized dynamics (23) by $e^{-\beta t}$ and simplifying we get:

$$\begin{aligned}&\ddot{r}_1 + (2\beta + \alpha)\dot{r}_1 + (P''(r^*) + (\alpha + \beta)\beta I)\, r_1 \\ &\quad = g(r^*)e^{-\beta t}F - \alpha e^{-\beta t}\dot{r}^* \\ &e^{-\beta t}(y - y^*) = (C_1 + \beta C_2)r_1 + C_2\dot{r}_1.\end{aligned} \tag{26}$$

If β is chosen sufficiently large, so that $P''(r^*) + (\alpha + \beta)\beta I$ is positive definite, the system is stable. In this case, the frequency response from $e^{-\beta t}F$ to $e^{-\beta t}y$ is

$$\begin{aligned}&H(j\omega) = ((C_1 + \beta C_2) + C_2 j\omega) \\ &\left[-\omega^2 I + (2\beta + \alpha)j\omega I + (P''(r^*) + (\alpha + \beta)\beta I)\right]^{-1} g(r^*).\end{aligned} \tag{27}$$

Note that terms corresponding to $e^{-\beta t}\dot{r}^*$ and $e^{-\beta t}y^*$ drop out since they vanish as $t \to \infty$.

To estimate the viscosity, we return to the Navier-Stokes equation (6), but with $e^{-\beta t}$ multiplied to both sides:

$$e^{-\beta t}\rho\frac{\partial u_x}{\partial t} = e^{-\beta t}\eta\frac{\partial^2 u_x}{\partial z^2} + e^{-\beta t}\frac{\rho}{m}F_x(t, z). \tag{28}$$

It follows that

$$e^{-\beta t}\frac{\partial u_x}{\partial t} = \frac{\partial \hat{u}_x}{\partial t} + \beta\hat{u}_x, \quad \text{where } \hat{u}_x = e^{-\beta t}u_x.$$

The Navier-Stokes equation then becomes

$$\rho\frac{\partial \hat{u}_x}{\partial t} = -\rho\beta\hat{u}_x + \frac{\partial^2 \hat{u}_x}{\partial z^2} + e^{-\beta t}\frac{\rho}{m}F_x(t, z). \tag{29}$$

D. Viscosity Estimation

If we choose the input force to be

$$e^{-\beta t}F_x(t,z) = F_o \sin(2\pi z/L),$$

then at steady state, the forced solution is of the form

$$\hat{u}_x = u_o \sin(2\pi z/L)$$

and

$$\rho\beta u_o = -\frac{\eta 4\pi^2}{L^2}u_o + \frac{\rho}{m}F_o,$$

which can be used to solve for η:

$$\eta = k\left(\frac{F_o}{u_o} - m\beta\right) \tag{30}$$

where $\frac{F_o}{u_o}$ is simply the inverse of the DC response, i.e.,

$$\eta = k\left(H(0)^{-1} - m\beta\right). \tag{31}$$

To summarize, our approach to estimating the shear viscosity involves the following steps:

1) Run an equilibrated EMD simulation to obtain $(r^*, \dot{r}^*)$.
2) Estimate α (using (22)) from an NEMD simulation with $\frac{1}{3}$ of the average molecular force magnitude from the EMD run above. The simulation only needs to be long enough to ensure a consistent value of α is obtained.
3) Choose β to obtain stable LTI systems and use the DC gain of the frequency response (27) to estimate the viscosity.

Though EMD and NEMD simulations are still used, we only need short simulation runs to obtain $(r^*, \dot{r}^*)$ and α.

E. Reduced Order Model

The state space model for the linearized system (26) is given by $(A, B, C, 0)$ where

$$A = \begin{bmatrix} 0 & I \\ -(P'' + (\alpha+\beta)\beta I) & -(2\beta+\alpha)I \end{bmatrix}$$

$$B = \begin{bmatrix} 0 \\ g(r^*) \end{bmatrix},\ C = \begin{bmatrix} C_1 + \beta C_2 & C_2 \end{bmatrix}.$$

The size of the state space is $6n$, where n is the number of particles. We may apply standard LTI model reduction techniques to first obtain a reduced order model, and then use it to compute the complex modulus. For high order systems, these standard model reduction tools are time consuming and plagued by numerical inaccuracy. Fortunately, for balanced truncation, reliable approximate methods have been proposed and implemented [7]–[9]. By choosing a cut-off of the (approximate) Hankel singular values corresponding to $(A, B, C, 0)$, we can find matrices S_B and S_C (with the number of columns corresponding to the number of retained Hankel singular values) to approximate the state as

$$z = S_c^T x, \quad x = S_B z, \tag{32}$$

where z is of much lower dimension than x. The linearized equation (26) may then be approximated by:

$$\begin{aligned} \dot{z} &= S_c^T A S_B z + S_c^T B \hat{u} \\ \hat{y} &= C S_B^T \end{aligned}$$

where $(\hat{u}, \hat{y})$ are the appropriate input and output signals. The DC gain $H(0)$ in the viscosity estimation (31) may now be approximated by

$$\hat{H}(0) = -C S_B^T (S_C^T A S_B)^{-1} S_C^T B. \tag{33}$$

If the dimension of z is much smaller than the original state dimension, the computation cost would be much lower since only a small matrix inversion is required.

IV. Simulation Experiments

For a comparison between EMD, NEMD, and the linearization method described in this paper, we consider a simple LJ fluid. The potential energy is given by:

$$\begin{aligned} P(r) &= \sum_{i=1}^{n}\sum_{j=1}^{n} p(r_{ij}),\ r_{ij} := r_i - r_j \\ p(r) &:= \begin{cases} 4\epsilon\left(\frac{\sigma^{12}}{\|r\|^{12}} - \frac{\sigma^6}{\|r\|^6}\right) & \|r\| < L/2 \\ 0 & \text{otherwise} \end{cases} \end{aligned} \tag{34}$$

We use the normalized model (in reduced units), so $\epsilon = \sigma = 1$. Periodic boundary conditions are used, therefore, $r = \texttt{mod}(r, L)$ to ensure all particles remain in the simulation box. The minimum image criterion is also used, i.e., if the particle separation in any one dimension is greater than half of the box dimension, $\left|r_{ij}^k\right| > L/2$, $k = x, y, z$, then the distance from a replicated image of the particle is used, $r_{ij}^k = r_{ij}^k - \text{sgn}(r_{ij}^k)L$.

A. EMD and NEMD Methods

EMD simulations are performed with the velocity Verlet algorithm. The reduced time step is chosen to be 0.005. During the equilibration phase, the velocity is rescaling at every time step to maintain the correct equilibrium temperature. Production runs are then performed without the velocity scaling. Each NEMD run uses the same equilibration phase as the EMD, but during the production run an additional forcing term is added. The viscosity is then calculated from the steady state velocity field. The results match reasonably well with the EMD results as the system size gets larger. Both our EMD and NEMD results match well with the results published in [10] and [6].

B. Linearization Method

For the linearization approach described in this paper, we use $N = 108$, $T = 1$, and $\rho = .8279$. The first step in our approach is to obtain $(r^*, \dot{r}^*)$ and the average molecular force from the EMD simulation. We use 5000 steps for equilibration and 500 steps for the production run. The average force magnitude divided by 3 (over the production run) is 8.34 with standard deviation of 0.75.

The next step is to estimate the linear damping α corresponding to $F_0 = 8.34$. The simulation with velocity scaling (to maintain constant temperature) is shown in Fig. 1. The α estimated using (22) has average value $\alpha = 2.57$ with standard deviation 0.53. To check the validity of this estimate, we run the NEMD simulation again including the damping term but without the velocity scaling. As can be seen in Fig. 2, even though the kinetic energy fluctuates more, it remains approximately around the required mean of 1.5.

The final step is to choose β so that (26) is stable. The shear viscosity can then be found from (31). With $\beta = 15$, the viscosity values for the entire r^* trajectory over 5000 time steps are shown in Fig. 3. The few spurious data points correspond to where the system become unstable (i.e., β not large enough). The overall average value is 1.93 with standard deviation 0.17, which is on par with the EMD and NEMD estimation ranges. The results for different β values (again over 5000 simulation steps) are shown in Table I. The estimated η values are reasonably close to the NEMD and EMD predictions (which range from 2.1 to 2.5, and 2.7 to 3.2, respectively). More numerical tests are being conducted for further validation of our procedure.

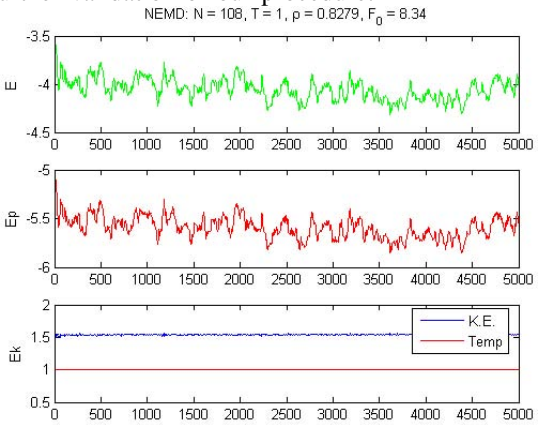

Fig. 1. Energy evolution in NEMD simulation with velocity scaling

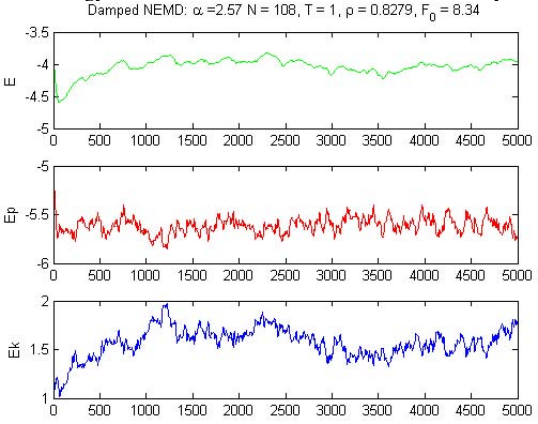

Fig. 2. Energy evolution in damped NEMD simulation (without velocity scaling)

β	η mean	η standard dev.
12	1.82	0.31
15	1.93	0.17
20	1.95	0.10
25	1.92	0.08
30	1.89	0.07

TABLE I

VISCOSITY ESTIMATES FOR DIFFERENT CHOICES OF β. THE MINIMUM CHOICE OF β IS AROUND 11.8 TO ENSURE STABILITY OF THE MODEL.

C. Model reduction

We have applied two approximate balanced truncation methods, low rank square root method (LRSRM) and dominant subspace projection model reduction (DSPMR) [8] to reduce the state dimension which is 648 in this

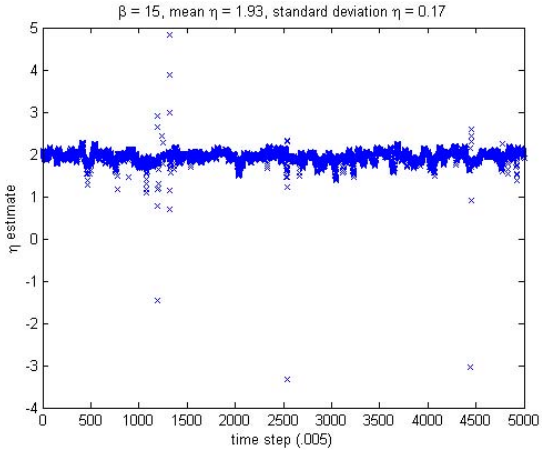

Fig. 3. Estimates of viscosity over 5000 time steps of $r^*(t)$

case. We found that for all the LTI systems, there is only *one* significant Hankel singular value (at least 2 orders of magnitude larger than all other Hankel singular values). The largest 2 Hankel singular values over 1800 time steps are shown in Fig. 4. The fact that there is a single dominant Hankel singular value means that we can approximate the original 648th order system by a first order system! The estimated shear viscosities based on the first order approximate systems are close to the full order estimate. For example, for $\beta = 15$, the average viscosity estimates based on the two model reduction methods are $\eta_{\text{LRSRM}} = 2.0$ and $\eta_{\text{DSPMR}} = 2.17$. The viscosity estimates for LRSRM over 1800 time steps is shown in Fig. 5 (DSPMR results are similar). Both model reduction schemes show consistent estimates of the viscosity in time. Therefore, a random $(r^*(t), \dot{r}^*(t))$ may be chosen for linearization. Fig. 6 shows the Bode plot comparison between the full order, LRSRM, and DSPRM model. All three cases have almost identical DC gains, but LRSRM matches the full order frequency response almost exactly while DSPRM shows considerable error in higher frequencies.

The reduced first order system is of the form

$$\hat{H}(s) = \frac{b}{s+a} \tag{35}$$

for some constants a and b. From (13) and (29), we know that the time constant is given by $-\frac{\eta 4\pi^2}{\rho L^2} - \beta$. Therefore, η may also be directly estimated from a:

$$\eta = \frac{\rho L^2}{4\pi^2}(a - \beta). \tag{36}$$

Viscosity estimates using this method together with the LRSRM model are shown in Fig. 7. Since the LRSRM model captures the true time constant well, the viscosity estimates are also very close to the expected (the average is 2.13).

The ability to reduce the high order molecular dynamics model to a first order model is not entirely surprising since we have already shown that the Navier-Stokes equation under the forcing function (7) is just a first order ODE given by (13). It is interesting that the NEMD model validates this property in the macroscopic model.

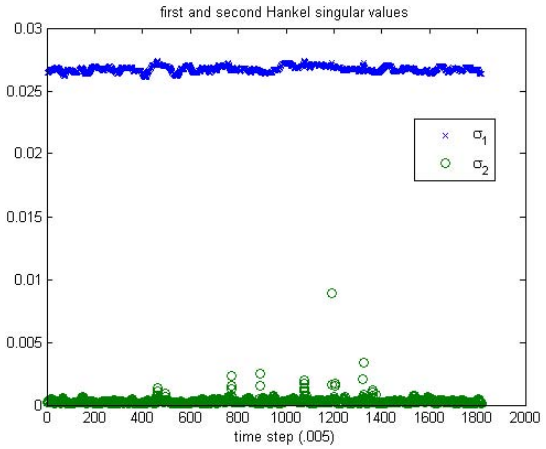

Fig. 4. Two largest Hankel singular values over 1800 time steps of $r^*(t)$

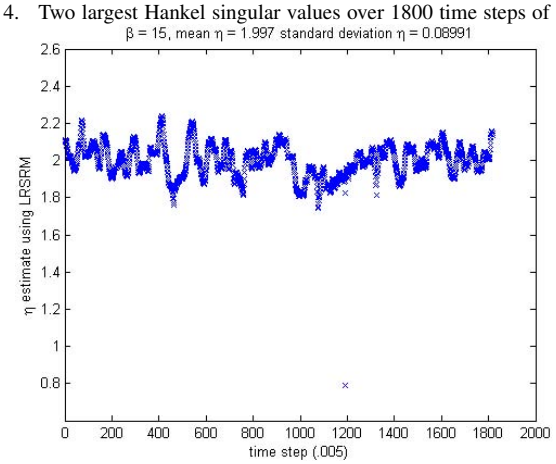

Fig. 5. Viscosity estimates based on the first order LRSRM model, over 1800 time steps of $r^*(t)$

V. Conclusion and Future Work

This paper considers the computation of shear viscosity based on a molecular dynamics model. We obtain a set of LTI systems by linearizing the NEMD equation about an EMD trajectory. Through an exponential transformation, the transformed LTI systems become stable, and the viscosity can be directly computed from the frequency responses. This approach has the potential of reducing the computation requirement needed in designing for specific material properties. Results from a simple LJ fluid confirms the consistency of the estimates between EMD, NEMD, and the linearization based method described here. Furthermore, the higher order NEMD model may be approximated by a first order model and still yields a good estimate for viscosity. Our next step is to apply the approach to polymeric systems with the finite extensible non-linear elastic (FENE) potential energy, and ultimately polymers with filler particles. Our long term goal is to develop a set of computationally efficient tools for transport properties that is suitable for material design optimization and process control.

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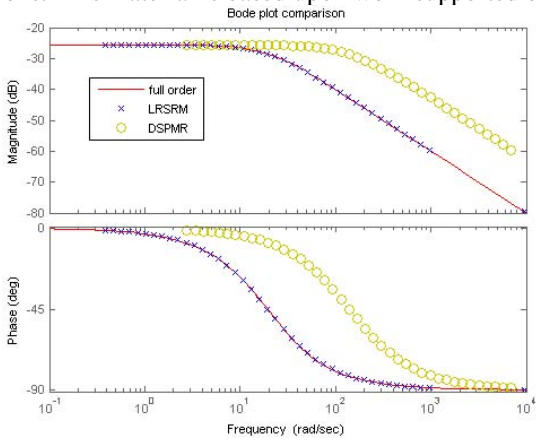

Fig. 6. Bode Plot comparison between full order, first order LRSRM, and first order DSPMR models

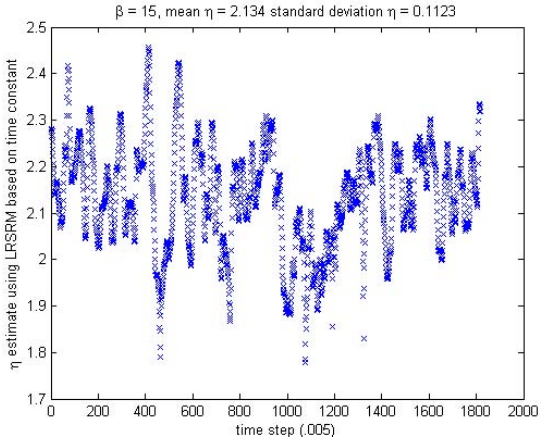

Fig. 7. Viscosity estimates using (36) and the first order LRSRM model, over 1800 time steps of $r^*(t)$

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