

Unexpected secondary flows in reverse nonequilibrium shear flow simulations

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We simulated two particle-based fluid models, namely multiparticle collision dynamics and dissipative particle dynamics, under shear using reverse nonequilibrium simulations (RNES). In cubic periodic simulation boxes, the expected shear flow profile for a Newtonian fluid developed, consistent with the fluid viscosities. However, unexpected secondary flows along the shear gradient formed when the simulation box was elongated in the flow direction. The standard shear flow profile was obtained when the simulation box was longer in the shear-gradient dimension than the flow dimension, while the secondary flows were always present when the flow dimension was at least 25% larger than the shear-gradient dimension. The secondary flows satisfy the boundary conditions imposed by the RNES and give a total flow field with a lower rate of viscous dissipation than the corresponding unidirectional flows. This work highlights a previously unappreciated limitation of RNES for generating shear flow in simulation boxes that are elongated in the flow dimension, an important consideration when applying RNES to complex fluids like polymer solutions.

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I. INTRODUCTION

The reverse nonequilibrium simulation method (RNES) developed by Müller-Plathe [1,2] is a well-established technique for computing transport coefficients of particle-based fluid models. It was initially proposed as a method to determine thermal conductivity [1] and was later extended to the shear viscosity [2]. RNES has been used to calculate the thermal conductivity of simple liquids [1,3], salts [4], carbon nanotubes [5,6], and silicon [7]; to study the Ludwig-Soret effect [3,8]; and to measure the shear viscosities of simple fluids [9], polymer solutions and melts [10–12], ionic liquids [13], alcohols [14], and water [15]. RNES has also been successfully applied to more complex systems under shear, including colloidal suspensions of nanoparticles [16–20], surfactant solutions [21], and asphalt [22]. More recently, RNES was used not only to compute transport properties but also to investigate dynamic phenomena like the shear-induced reorientation of diblock-copolymer lamellae [23] and the aggregation of patchy particles in flow [24].

The underlying idea of RNES is to impose an “effect” on a system in an unphysical way and measure the “cause.” In many nonequilibrium simulation techniques, a gradient (cause) is imposed and a flux (effect) is measured [25], but RNES reverses this picture. For example, in RNES, stress can be generated by an unphysical transfer of momentum between particles [2], driving the system out of equilibrium and resulting in a physical momentum flux. The stress gives rise to a corresponding flow profile that can be measured. Given an imposed flux and measured gradient,

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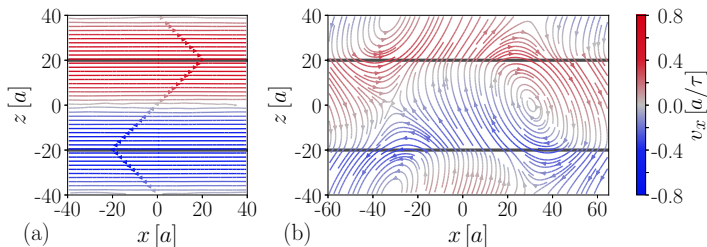


FIG. 1. Streamlines in the xz plane for the flow induced by RNES in (a) a cubic box ($80a \times 80a \times 80a$) and (b) an orthorhombic box ($125a \times 80a \times 80a$). Color indicates the x component of the velocity, v_x . Gray horizontal lines mark the exchange slabs of width $w = 1a$ at $z = \pm 20a$.

the relevant transport coefficient (e.g., shear viscosity) can be extracted within the linear-response regime.

RNES possesses many desirable properties of a nonequilibrium method [1,26]. It is quickly convergent compared to equilibrium methods, such as the Green-Kubo relations [27], to determine transport coefficients. RNES is compatible with a multitude of different particle-based models. It also does not require a specific simulation box geometry and accommodates periodic boundary conditions, which eliminates artificial wall effects that could cause measured fluid properties to differ from the bulk in small, wall-bounded simulation boxes [28]. Additionally, RNES can be made to conserve momentum and energy and acts as its own thermostat [26], avoiding the challenges of applying an external thermostat out of equilibrium [29]. Because of its straightforward implementation and flexibility, RNES is widely used and is implemented in many simulation packages, including LAMMPS [30], HOOMD-BLUE [31], OPENMD [32], and ESPRESSO [33].

Despite these many positive attributes, we have uncovered a previously unappreciated limitation of RNES for simulating shear flow. We applied RNES to two different model fluids in various simulation box geometries. Unexpectedly, we obtained not only standard shear flow [Fig. 1(a)], but also more complicated flow patterns [Fig. 1(b)] in certain noncubic boxes with periodic boundary conditions. In those cases, the flow had significant secondary components along the shear gradient, and the shear stress did not have constant magnitude throughout the system. Such flows are undesirable because they severely complicate the calculation of the shear viscosity using RNES.

In this article, we systematically interrogate the flow field, stress, and viscous dissipation in RNES as functions of the simulation box geometry and the shear rate to determine *why* and *when* RNES does not generate the expected shear flow. We show that the secondary flows emerge due to the periodic boundary conditions and are not specific to the fluid model or the RNES algorithm, suggesting a hydrodynamic instability inherent to the flow and geometry. The rest of the article is organized as follows. The RNES algorithm and its known properties and features are explained in Sec. II. The simulation details and fluid models are described in Sec. III. Our results are presented and discussed in Sec. IV and are followed by our conclusions in Sec. V.

II. RNES ALGORITHM

We first summarize details of the RNES algorithm for simulating shear flow and its known features and limitations. The simulation box is defined to be of size $L_x \times L_y \times L_z$, with x being the direction of flow and z being the direction of the shear gradient (Fig. 2). The box is periodic in all three dimensions. In the RNES algorithm [2], two exchange slabs of width w are constructed at $z = \pm L_z/4$. At regular intervals Δt during the simulation, pairs of particles are selected from the slabs and their momenta are swapped. The particle with the most negative momentum in the flow direction, p_x^- , is selected from the upper slab, while the particle with the most positive x momentum, p_x^+ , is chosen from the lower slab. An unphysical swap move exchanges p_x^- and p_x^+ ,

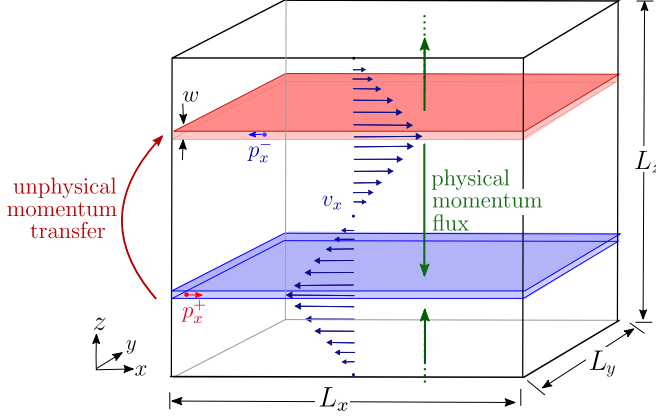


FIG. 2. Sketch of the simulation box of dimensions $L_x \times L_y \times L_z$, where x is the flow direction and z is the gradient direction. The two exchange slabs of width w are located at $-L_z/4$ (bottom, blue) and $+L_z/4$ (top, red). Unphysical momentum swaps between pairs occur in the top and bottom slabs, driving a shear-induced physical momentum flux back (green). The expected velocity field given by Eq. (4), $v_x(z)$, is sketched in blue.

resulting in a transfer of momentum $\Delta p_x = p_x^+ - p_x^-$. The exchange of momentum generates a physical momentum flux with a corresponding shear stress, τ_{zx} , at steady state [2]:

$$\tau_{zx} = \frac{\langle \Delta p_x \rangle}{2L_x L_y \Delta t}. \quad (1)$$

Here, $\langle \Delta p_x \rangle$ denotes the average amount of momentum exchanged during the interval Δt , and the factor of 2 is due to the periodic boundary conditions. If both particles in the swapped pair have the same mass, the total momentum and kinetic energy of the system are conserved during the momentum transfer.

The imposed stress is expected to generate a flow field \mathbf{v} . Assuming the steady, incompressible flow of a Newtonian fluid, the standard continuity equation and momentum balances governing the flow are [34]

$$\nabla \cdot \mathbf{v} = 0, \quad (2)$$

$$\rho \mathbf{v} \cdot \nabla \mathbf{v} = -\nabla p + \nabla \cdot \boldsymbol{\tau}, \quad (3)$$

where ρ is the density, p is the pressure, and $\boldsymbol{\tau} = \mu[\nabla \mathbf{v} + (\nabla \mathbf{v})^T]$ is the viscous stress tensor with μ being the shear viscosity. We approximate the exchange procedure by a constant shear stress τ_{zx} at the center of the exchange regions ($z = \pm L_z/4$) and a uniform pressure, which implies that there is no flow in y or z . Applying the periodic boundary conditions of the simulation box gives the steady-state shear flow field, $v_x(z)$,

$$v_x(z) = \begin{cases} \dot{\gamma}(-L_z/2 - z), & z < -L_z/4 \\ \dot{\gamma}z, & |z| < L_z/4, \\ \dot{\gamma}(L_z/2 - z), & z > L_z/4 \end{cases} \quad (4)$$

where $\dot{\gamma} = \tau_{zx}/\mu$ is the shear rate in this geometry. This flow field is illustrated in Fig. 2. Note that due to the periodic boundary conditions, the standard Couette flow ($|z| < L_z/4$) is extended outside the exchange regions, resulting in an overall triangular profile. In RNES, the width of the exchange regions w should be as small as possible to minimally disturb this expected flow field [23], while still keeping the regions large enough to contain a sufficient number of swapping pairs.

The imposed shear stress can be used to determine the shear viscosity μ from the measured flow field within the linear-response regime,

$$\tau_{zx} = \mu \left\langle \frac{\partial v_x}{\partial z} \right\rangle = \begin{cases} \mu \dot{\gamma}, & |z| < L_z/4 \\ -\mu \dot{\gamma}, & |z| > L_z/4 \end{cases}. \quad (5)$$

The shear rate $\dot{\gamma}$ can be varied by tuning the momentum exchange rate to change τ_{zx} , and μ is usually extracted from a series of measurements at different $\dot{\gamma}$. RNES has technical limitations at both very low and very high shear rates. For low shear rates, infrequent momentum swaps may not lead to a steady flow profile [35,36]. Analogous problems have been reported for the determination of thermal coefficients using RNES [7]. As suggested in Ref. [36], this issue can be partially alleviated with a weaker, more frequent exchange of momentum, e.g., by choosing pairs with momenta close to a target value instead of the maximum [35]. This modification allows the flow rate to be tuned more precisely [35,37], and also leads to a weaker system-size dependence and better convergence of the measured viscosity [37]. For high shear rates, the velocities in the exchange regions deviate from the expected Boltzmann distributions [35]. The velocity distribution in the lower slab develops a shoulder toward lower values, whereas the distribution in the upper slab develops a shoulder toward higher values because the imposed momentum transfer exceeds the system's ability to thermalize. In this case, RNES significantly underpredicts viscosities relative to other computational methods [35]. The numerical bounds on the accessible shear rates depend on the fluid model and must be determined carefully by trial and error.

The RNES algorithm does not specify a box size or shape, and different geometries have been chosen by different authors. For example, Müller-Plate [1] originally used a box that was three times longer in the gradient direction than in the flow direction ($L_z = 3L_x$). Nikoubashman and Howard chose $L_z = L_x$ to simulate polymer solutions in shear [11], while Schneider *et al.* utilized boxes with L_z ranging from $2L_x$ to $4.6L_x$ to study reorientation of diblock copolymers melts [23]. Generally, and as is standard in molecular simulations [38], the simulation box must be large enough in all dimensions to avoid unphysical self-interactions through periodic boundaries, particularly between macromolecules like polymers. Since many macromolecules align and stretch with flow [39], the simulation box can similarly be expanded along the flow dimension to match the expected deformation ($L_x > L_z$). This geometry reduces the overall computational cost of the simulation compared to expanding the box in all dimensions. However, as we will show in Sec. IV, elongating the box in the flow dimension has previously unknown and undesirable consequences for RNES.

III. SIMULATION MODELS

We applied RNES to simulate shear flow for two fluids, one modeled using multiparticle collision dynamics (MPCD) [40,41] and the other using dissipative particle dynamics (DPD) [42–44]. Both MPCD and DPD are particle-based mesoscale models that faithfully resolve hydrodynamic interactions and incorporate the effects of thermal fluctuations [45]. These models were chosen for their computational efficiency, but we were able to obtain similar results with a more computationally demanding Lennard-Jones fluid model using RNES with conventional molecular dynamics simulations [2]. In this article, we will describe the model parameters and our results using a as the unit of length, ε as the unit of energy, and m as the unit of mass. In this system of units, $\tau = \sqrt{ma^2/\varepsilon}$ is the unit of time.

The MPCD fluid consisted of pointlike particles of mass m . The particle positions and velocities were propagated in alternating streaming and collision steps [40,41]. Particles moved ballistically during the streaming step and were subsequently binned into cubic cells of size a . The particle coordinates were collectively shifted by a random value between $\pm a/2$ during binning to ensure Galilean invariance [46,47]. In the ensuing collision step, the particle velocities relative to the cell-average velocity were rotated by a fixed angle α around an axis randomly chosen from the unit

sphere for each cell. This collision procedure, also called stochastic rotation dynamics, conserves linear momentum in each cell and so also globally.

The properties of the MPCD fluid are controlled by the particle number density, the rotation angle, the temperature, and the time between collisions [40,41,48]. We chose the density as $\rho = 5 a^{-3}$ and the rotation angle as $\alpha = 130^\circ$, and performed a collision every 0.1τ . We additionally applied a Maxwell-Boltzmann rescaling thermostat [49,50] to each cell to maintain a constant temperature $T = 1.0 \varepsilon/k_B$ throughout the fluid, where k_B is Boltzmann's constant. Since a thermostat is not required in RNES, we confirmed that removing the thermostat did not qualitatively change our results. The viscosity μ can be estimated for the MPCD fluid using kinetic theory [51], giving $\mu = 3.96 \varepsilon \tau/a^3$ for our parameters.

The DPD fluid [42,52] was also represented by particles of mass m . A conservative force, a dissipative force, and a random force acted between pairs of particles within a distance r_c of each other. The dissipative and random forces were consistent with the fluctuation-dissipation theorem and conserved local momentum. Details of the functional forms of the DPD interactions can be found in Ref. [44]. We employed standard parameters: a maximum conservative force of $25 \varepsilon/a$, a drag coefficient of $4.5 m/\tau$ for the dissipative force, a cutoff of $r_c = 1 a$, and an integration time step of 0.01τ [44]. The particle number density was $\rho = 3 a^{-3}$ [44,53], and the temperature was $T = 1.0 \varepsilon/k_B$.

We determined the viscosity of the DPD fluid for our parameters using RNES in cubic boxes with edge lengths ranging from $25 a$ to $65 a$, finding $\mu = 0.87 \varepsilon \tau/a^3$. We independently validated the RNES measurement using the periodic Poiseuille flow method [54]. We divided the simulation box into two domains with $z < 0$ and $z \geq 0$ and applied a constant body force with equal magnitude but opposite direction ($\pm x$) between the regions. We extracted the viscosity by fitting the velocity profile to the expected parabolic form [54]. The obtained value of $\mu = 0.87 \varepsilon \tau/a^3$ was in good agreement with the RNES measurement and values reported elsewhere [55].

All simulations were performed on graphics processing units using HOOMD-BLUE (modified version 2.3.0) [31,56–58] with our own implementation of RNES [59]. We additionally reproduced selected results for the MPCD fluid using another implementation [60] and for the DPD fluid using LAMMPS (11 Aug 2017) [30,61]. An example LAMMPS script for simulating the DPD fluid is included as Supplemental Material [62].

IV. RESULTS AND DISCUSSION

We first simulated the MPCD fluid in two boxes: a cubic box of size $80 a \times 80 a \times 80 a$ and an orthorhombic box, elongated along the flow direction, of size $125 a \times 80 a \times 80 a$. The slab exchange width was $w = 1 a$, and the swapped particles were chosen to have momenta closest to a target value of $\pm 0.5 ma/\tau$. We swapped 202 pairs in the cubic box and 316 pairs in the orthorhombic box (0.6% of particles in the exchange regions) every step, resulting in the same imposed stress ($\tau_{zx} = 0.158 \varepsilon/a^3$) for both boxes. The cubic box developed the expected triangular velocity profile [Eq. (4)] with extrema in the exchange regions [Fig. 1(a)]. No flows occurred in either of the other directions ($v_y = v_z = 0$). We extracted a viscosity of $\mu = 3.95 \varepsilon \tau/a^3$ from the measured flow field using Eq. (5), in quantitative agreement with the viscosity estimated from kinetic theory.

We expected that applying the RNES method in the orthorhombic box should result in the same velocity profile because the same stress was applied; surprisingly, a markedly different flow field developed [Fig. 1(b)]. The average velocity in the exchange regions was still along x as expected, but substantial flows along the shear gradient (z) were also obtained, giving an overall two-dimensional flow field (v_y remained zero). A movie of the three-dimensional streamlines as they develop from a quiescent fluid can be found in the Supplemental Material [62]. The resulting streamlines exhibited two vortices and two stagnation points, with one of each between the exchange slabs. The vortices and stagnation points were stationary during the accessible simulation time. The DPD fluid under shear had the same qualitative behavior as the MPCD fluid, indicating that this surprising flow field is not specific to the MPCD model. We have extensively tested and verified that the momentum

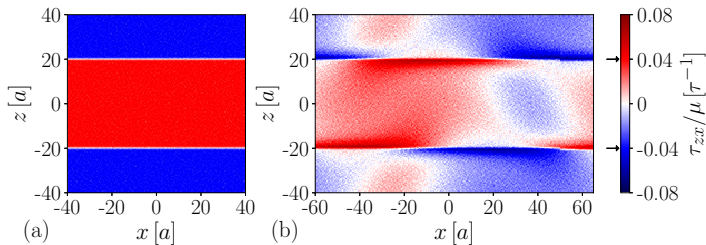


FIG. 3. Local shear stress τ_{zx}/μ in the fluid in (a) a cubic box and (b) an orthorhombic box for the flow fields of Fig. 1. The expected values based on Eq. (5), $\tau_{zx}/\mu = \pm 0.04 \tau^{-1}$, are indicated by arrows on the color bar.

exchanged between the slabs is in agreement with the expected value in all simulated systems and that all flow fields satisfy the continuity equation [Eq. (2)]. An example of this calculation can be found in Fig. S1 [62].

One possible explanation for the presence of vortices in the flow could be the emergence of turbulence in the Couette flow. To estimate this effect, we defined a Reynolds number $\text{Re} = \rho U L_z / 4\mu$ based on the maximum velocity in the exchange slabs U and the half-width between the exchange slabs, $L_z/4$. It has been shown that the transition from laminar to turbulent flow in planar Couette flow can occur for Reynolds numbers as low as $\text{Re} \approx 300$ [63–65]. For Fig. 1(a), the Reynolds number is $\text{Re} \approx 20$, which is well below the laminar-turbulent transition for the classical Couette flow. Expanding the box along x does not change Re , and so turbulence based on the flow is not expected for the orthorhombic box either. To exclude possible finite-size effects, we performed a simulation in a box of size $125a \times 80a \times 125a$ at $\text{Re} = 20$ as well as at $\text{Re} = 50$ and obtained only flows like Fig. 1(a).

As an additional stability test, we introduced two vortices and two stagnation points into a cubic box ($80a \times 80a \times 80a$) and followed the evolution of the flow. The vortices dissipated completely after $70\,000 \tau$ and the flow returned to the expected triangular flow profile. This shows that the expected shear flow is not metastable with respect to the vortices for the cubic box. Likewise, we initialized the expected shear flow in an orthorhombic box ($125a \times 80a \times 80a$) and observed the emergence of vortices from the triangular flow on a similar timescale. The final steady state was independent of how the fluid was initialized for both box shapes.

One of the benefits of using the RNES method is the ability to readily compute the shear viscosity from simulations from a simple shear flow profile [Eq. (5)]. The secondary flows in the orthorhombic box effectively prevent this calculation and are undesirable for simulating simple shear. In the next sections, we investigate the local shear stress (Sec. IV A), the effect of boundary conditions (Sec. IV B), and the viscous dissipation in the fluid (Sec. IV C) to understand when and why the secondary flows appear so that they can be avoided.

A. Shear stress

In deriving the expected shear flow field [Eq. (4)], we assumed that the shear stress in the exchange regions was constant and equal to the value imposed by RNES. In order to test this assumption, we computed the viscous stress tensor τ from the measured flow fields. Figure 3 shows the shear stress, τ_{zx} , corresponding to the flows in Fig. 1. The cubic box [Fig. 3(a)] exhibited the expected step function behavior [Eq. (5)], $\tau_{zx}/\mu = \pm 0.04 \tau^{-1}$. Only the exchange regions of width $w = 1a$ at $z = \pm 20a$ deviated from the expected values, showing a shear stress close to zero. This is not surprising because the stress in the fluid should vary continuously. In contrast, the shear stress in the orthorhombic box [Fig. 3(b)] was strongly position dependent. The shear stress was lower near the vortices and stagnation points, but exhibited localized areas of significantly higher stress close to the exchange regions.

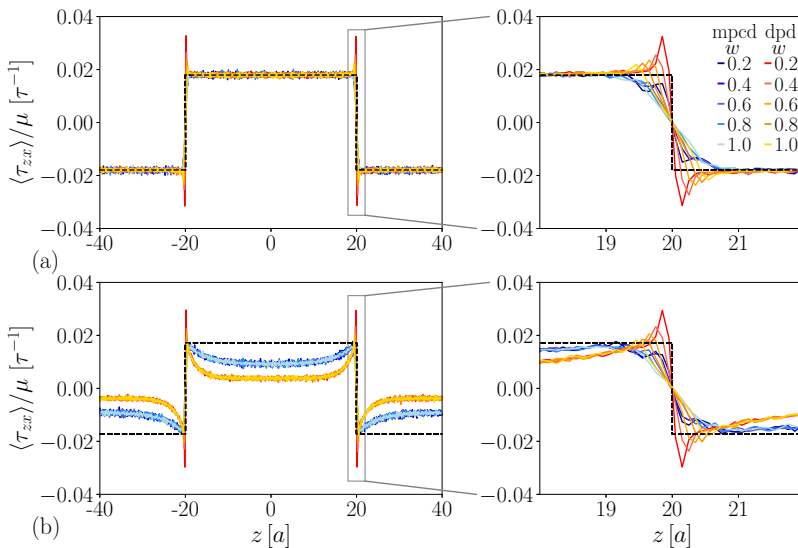


FIG. 4. Average shear stress $\langle \tau_{zx} \rangle$ in (a) a cubic box ($80a \times 80a \times 80a$) and (b) an orthorhombic box ($125a \times 80a \times 80a$) for the MPCD and DPD fluids. The width w of the exchange region was varied from $0.2a$ to $1.0a$ as indicated in the legend. The expected step function, $\tau_{zx}/\mu = \pm 0.0172 \tau^{-1}$, is shown as a dashed line.

In order to confirm that the width of the exchange region w did not significantly affect the viscous stress, we systematically decreased w for both the MPCD and DPD fluids. Since the number of possible pairs to swap decreased with w , we chose a lower target shear stress than in Fig. 3, using $\tau_{zx} = 0.068 \varepsilon/a^3$ for MPCD and $\tau_{zx} = 0.015 \varepsilon/a^3$ for DPD, which gives an expected shear rate of $\dot{\gamma} = 0.0172 \tau^{-1}$ for both fluids. The computed shear stress was averaged over x to give the average stress, $\langle \tau_{zx} \rangle$, as a function of z .

For the cubic box [Fig. 4(a)], $\langle \tau_{zx} \rangle$ was again a step function for both fluids outside the exchange regions, as expected, regardless of the width, but some deviations were observed near the exchange regions. The stress profile for the MPCD fluid was rounded and increased in steepness with decreasing w , whereas the shear stress in the DPD fluid overshoot the expected value close to the exchange region by an amount that increased with decreasing w . The overshoot in shear stress corresponds to the velocity being larger than expected near the exchange region, which is consistent with previous simulations of DPD models [23]. The rounded stress profile for the MPCD fluid is similarly consistent with rounding of the velocity profile in the exchange region that has been reported elsewhere [19].

For the orthorhombic box [Fig. 4(b)], both fluids showed a significant reduction in $\langle \tau_{zx} \rangle$ outside the exchange regions compared to the cubic box. As for the cubic box, the exchange region width again did not influence the value of the stress in these regions. Also consistent with the cubic box, the DPD fluid had stress overshoot in the exchange region, whereas the MPCD fluid had a rounded profile. Most significantly, we note that $\langle \tau_{zx} \rangle$ in the exchange regions was essentially indistinguishable between the cubic and orthorhombic boxes. Although the shear stress in the exchange regions of the orthorhombic box was not *constant*, the RNES algorithm still imposed the same *average* stress in the exchange regions for both simulation boxes.

B. Boundary conditions

Our shear stress measurements suggest that the RNES method imposes boundary conditions that are inconsistent with the assumptions used to derive Eq. (4): namely, a constant shear stress τ_{zx} at

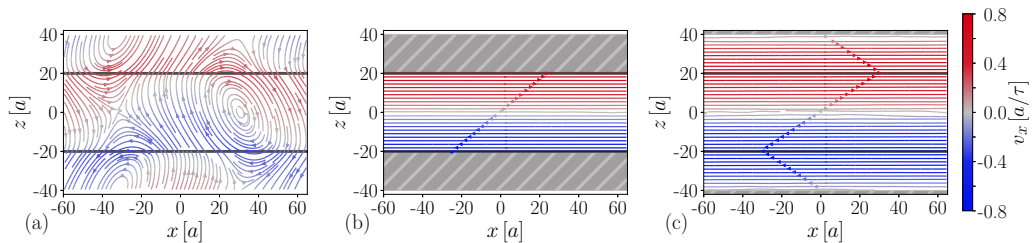


FIG. 5. Streamlines in the xz plane for the flow induced by RNES in (a) an orthorhombic box ($125a \times 80a \times 80a$) with periodic boundary conditions in all dimensions, (b) in the same box with slip walls placed at $z = \pm 20.5a$, and (c) in the same box with no-slip walls placed at $z = \pm 40a$. Color indicates the x component of the velocity, v_x . Gray horizontal lines mark the exchange slabs of width $w = 1a$ at $z = \pm 20a$.

$z = \pm L_z/4$ and a unidirectional flow ($v_y = v_z = 0$). In the previous section, we showed that RNES imposes the average shear stress in the exchange regions. This is a weaker boundary condition than a constant stress and does not enforce that $v_z = 0$. Clearly, $v_z \neq 0$ for the orthorhombic box [Fig. 1(b)].

To test the influence of including the condition that $v_z = 0$ in RNES, we inserted two smooth, hard walls into the orthorhombic box just outside the exchange regions, effectively simulating half of the system ($|z| < L_z/4$). We performed this test only for the MPCD fluid, which does not exhibit local structuring near a wall. The MPCD particles were reflected from the hard wall using bounce-back reflections for a slip boundary condition. The walls enforce that $v_z = 0$ since the fluid cannot penetrate the hard surface, but the slip boundary condition does not modify the flow tangent to the surface. We then applied the RNES momentum exchange procedure to the fluid. We found that only the expected shear flow, $v_x(z)$, formed between the exchange regions, even in boxes with $L_z > L_x$, as shown in Fig. 5(b). From this test, we conclude that secondary flows only form in RNES for fully periodic boxes that are subject to an average stress boundary condition but do not constrain $v_z = 0$.

Because the previous test restricted the flow field to only one half of the RNES with periodic boundary conditions, we also inserted two smooth, hard, no-slip walls at $z = \pm L_z/2$, effectively replacing the periodic boundary condition in one dimension while keeping the rest of the system unchanged. We included virtual MPCD particles [66,67] at random positions in the walls with velocities drawn from the Maxwell-Boltzmann distribution to help enforce the no-slip condition. In this case, the entire flow field given by Eq. (4) is recovered, as shown in Fig. 5(c). To test possible finite-size effects, we repeated the simulation in a box twice as big in the x and z dimensions and did not observe a difference in flow behavior.

Because the RNES algorithm chooses particles to swap based on their momenta in the flow direction, the distribution of the positions of the selected particles in the slabs was not uniform once the secondary flows developed. To test the extent to which the secondary flows depend on this RNES-specific momentum exchange procedure, we considered an alternative nonequilibrium scheme for generating shear flow. We applied constant, opposite body forces to all particles in the exchange regions instead of the usual RNES momentum swaps. (Each particle in the upper slab experienced a force $+\Delta f$ in the x direction, while particles in the lower slab had a force $-\Delta f$ in the x direction.) Although the method of generating the flow was different, the resulting flow profiles and stress distributions were identical to the ones obtained by the RNES swaps when the average stress was matched. Together, these tests reveal that the emergence of the secondary flow is not unique to the reverse nonequilibrium method by which the flow was created, but rather appears to be an effect of the periodic boundary conditions with the targeted flow field.

C. Viscous dissipation

We have demonstrated that the unexpected secondary flows generated by RNES for simulation boxes that are elongated in the flow dimension are consistent with the continuity equation and

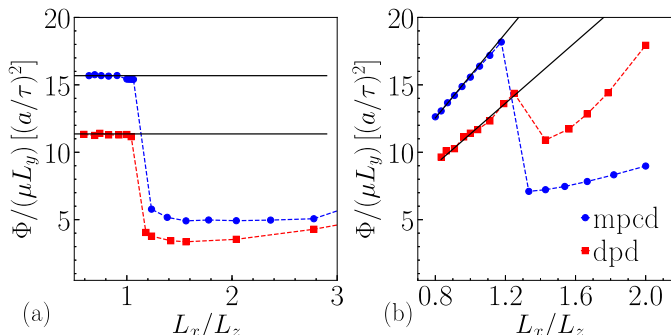


FIG. 6. (a) Total viscous dissipation Φ as function of box shape for fixed average shear stress of $\tau_{zx}/\mu = 0.04 \tau^{-1}$ for the MPCD fluid and $\tau_{zx}/\mu = 0.0688 \tau^{-1}$ for the DPD fluid. (b) Total viscous dissipation for fixed maximum velocity in the exchange regions. Note that dissipation in the RNES exchange regions has been excluded from Φ in both the numerical and theoretical calculations.

average momentum flux imposed by RNES and appear to form due to the periodic boundary conditions in certain boxes. However, it is still unclear *when* and *why* a certain flow field is realized for a given simulation box. We use the total rate of viscous dissipation, Φ , [34]

$$\Phi[\mathbf{v}(\mathbf{r})] = \int d\mathbf{r} \tau[\mathbf{v}(\mathbf{r})] : \nabla \mathbf{v}(\mathbf{r}), \quad (6)$$

as an order parameter to detect the transition from the expected shear flow to the regime containing secondary flows. (Other choices like the maximum value of v_z are possible if the direction of the secondary flow is known in advance.) The rate of viscous dissipation is directly proportional to the rate of entropy production in an isothermal, incompressible flow [34]. The expected viscous dissipation for the flow field given by Eq. (4), Φ_0 , can be computed analytically as $\Phi_0/V = \mu \dot{\gamma}^2$, where $V = L_x L_y L_z$ is the volume of the simulation cell.

To test if the rate of viscous dissipation detects the different flow profiles, we simulated the MPCD and DPD fluids in rectangular boxes with aspect ratios varying from $L_x/L_z = 0.5$ to 2.7 . We performed the simulations at both a fixed shear stress and a fixed maximum velocity according to Eq. (4). For the fixed shear stress simulations for the MPCD fluid, we varied $L_y = L_z$ from $80a$ to $125a$ and kept the area $A = L_x L_y$ of the exchange region constant at $A = 100^2 a^2$. The DPD fluid simulations at fixed shear stress were performed in boxes with an exchange slab area of $A = 50^2 a^2$ and varied L_z from $30a$ to $65a$. For the fixed maximum velocity simulations, we picked boxes with $L_x = L_y = 100a$ and varied L_z from $50a$ to $125a$ for the MPCD fluid. Similarly, the DPD fluid simulations were performed in boxes with $L_x = L_y = 50a$ and L_z from $28a$ to $60a$. Averaged velocity profiles $\langle v_x \rangle$ and $\langle v_z \rangle$ can be found in Figs. S2 and S3 [62].

We then computed the total viscous dissipation Φ from the measured flow fields using Eq. (6). As demonstrated in Sec. IV A, the simulated flow field deviated from the theoretically expected profile near the exchange slabs even when no secondary flows occurred. To better facilitate comparison between the simulations and theory, we excluded the exchange slabs of size $w = 1a$ from both the theoretical and numerical calculations of Φ . We confirmed that neglecting these regions did not qualitatively change our findings, although it did lead to a small, constant shift of at most 5% in the reported values of Φ .

We calculated the total viscous dissipation Φ for both the MPCD and DPD fluids for various different box aspect ratios L_x/L_z at a constant average shear stress [Fig. 6(a)] and a constant maximum velocity, resulting in a varying shear stress [Fig. 6(b)]. The viscous dissipation measured from the velocity gradients agreed with the theoretical predictions for all cases where the box aspect ratio was smaller than approximately $L_x/L_z < 1.05$. Above a critical value of approximately

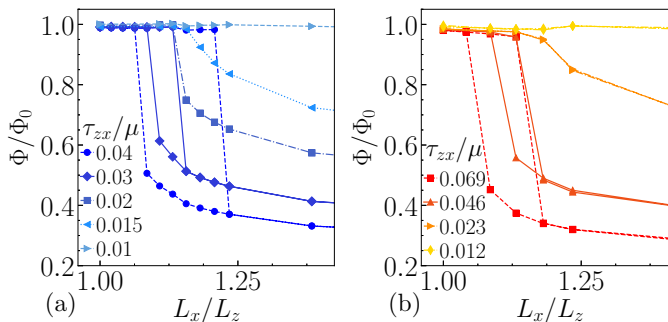


FIG. 7. Ratio of measured to expected viscous dissipation, Φ/Φ_0 , for the (a) MPCD fluid and (b) DPD fluid in simulation boxes of varied aspect ratio, L_x/L_z , and expected shear stress, τ_{zx}/μ . The shear stresses are indicated in the legends.

$L_x/L_z > 1.25$, the total viscous dissipation dropped significantly, concomitant with the emergence of secondary flows. The flow field that is realized in the simulation appears to minimize the total viscous dissipation. The maximum value of v_z showed a similar steplike behavior, increasing with respect to L_x/L_z , indicating that it is also a viable order parameter when the secondary flows in z are known to exist.

We subsequently investigated the shear rate dependence of the secondary flows using Φ as a parameter to identify the two different flow field types. We simulated both fluids at different momentum exchange rates in various box sizes and calculated Φ (Fig. 7). We found that the system showed significant hysteresis effects for box aspect ratios L_x/L_z between 1.05 and 1.25. We initialized two simulations for each box geometry and shear rate: one with a flow field given by Eq. (4) and the other with two vortices superimposed by an additional component $v_z(x) \sim \cos(2\pi x/L_x)$. For lower shear rates, the two different flows quickly converged to the same viscous dissipation and the same flow profile. For the higher shear rates, the two systems did not converge during the length of the simulation run of 600 000 τ for MPCD and 70 000 τ for DPD. We therefore report both measured values of the viscous dissipation in those cases.

For both fluids and all shear rates, secondary flows formed when $L_x/L_z > 1.25$. The drop in the viscous dissipation was most pronounced for the biggest perturbations, even when scaled relative to Φ_0 . The viscous dissipation varied only weakly for the lowest perturbation, but weak secondary flows still developed in the elongated boxes, as shown in Fig. S4 [62]. We were unable to detect a lower critical shear rate to avoid secondary flow within the simulation accuracy, although the secondary flows must vanish, along with the expected shear flow, when τ_{zx}/μ goes to zero.

Because of the hysteresis, it was difficult to estimate a precise box aspect ratio for the emergence of the secondary flows. As visible in Figs. 6 and 7, boxes which were cubic or elongated in the gradient dimension ($L_x \leq L_z$) always developed the expected shear flow. In contrast, simulations in boxes with $L_x/L_z > 1.25$ always converged to a flow profile containing secondary flows. Since it can be challenging to detect the secondary flows without careful sampling of the three-dimensional velocity field, we advocate applying the RNES method for generating shear flow only when $L_x \leq L_z$.

V. CONCLUSIONS

In this work, we showed that the RNES method for simulating shear flow can generate unexpected secondary flows, which do not allow a reliable calculation of the shear viscosity, in simulation boxes that are elongated in the flow dimension. We demonstrated these flows occurred over a range of box sizes and shear stresses for two different fluid models, revealing that the effect is general. Indeed, we were also able to reproduce the same trends for a Lennard-Jones fluid using RNES with classical molecular dynamics simulations, which we have omitted from this article for

brevity. Although it was challenging to precisely identify the box geometries leading to secondary flows due to hysteresis effects, we showed that no secondary flows formed during the accessible simulation time for aspect ratios, L_x/L_z , below 1.05, while they always occurred for ratios above 1.25. Weak secondary flows were obtained even for very low shear rates, indicating that there was no detectable lower threshold within the accuracy of the simulations.

The flow fields in both the cubic and elongated boxes were shown to be consistent with the same boundary condition on the average stress imposed by RNES. However, this boundary condition did not constrain the velocity along the shear gradient, v_z . When the RNES boundary conditions were augmented by hard walls to obligate $v_z = 0$, the secondary flows were suppressed and the expected Couette flow profile was recovered. This test demonstrated that the periodic boundary conditions of the simulation box were needed to obtain the secondary flows. For elongated simulation boxes, the fact that the RNES algorithm is compatible with periodic boundary conditions turns out to not be a feature but rather the origin of the undesired behavior.

We measured the velocity fields to calculate the stresses and viscous dissipation. We showed that the viscous dissipation is a good predictor for the emergence of secondary flows, with the total viscous dissipation dropping significantly compared to the theoretically expected value when the secondary flows occurred. The amount of reduction in viscous dissipation depended on the shear rate, with a greater reduction relative to the expected value at higher shear rates. We speculate this behavior may be due to an underlying hydrodynamic instability with respect to the boundary conditions in this particular geometry and flow. The stability analysis of the Navier-Stokes equations for RNES is left as an intriguing subject of future work.

This study reveals a previously unappreciated limitation of the RNES method. We found that only boxes which are approximately cubic or elongated in the shear-gradient dimension can be reliably used to simulate standard shear flow, especially for higher shear rates. This has important implications for recent applications of RNES to study flow behavior of complex fluids like colloids [19,20,24], nanoparticles [16–18], and polymers [11,21,23]. Both of the fluids tested here are frequently used as background solvents in these simulations. Although it may be tempting to extend the simulation box in the flow dimension for computational efficiency, we recommend using only boxes with $L_x \leq L_z$ to reliably generate the expected shear flow.

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