

Role of Hydrodynamic Interactions in the Deformation of Star Polymers in Poiseuille Flow

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Abstract Stretching polymer in fluid flow is a vital process for studying and utilizing the physical properties of these molecules, such as DNA linearization in nanofluidic channels. We studied the role of hydrodynamic interactions (HIs) in stretching a free star polymer in Poiseuille flow through a tube using mesoscale hydrodynamic simulations. As increasing the flow strength, star polymers migrate toward the centerline of tube due to HIs, whereas toward the tube wall in the absence of HIs. By analyzing the end monomer distribution and the perturbed flow around the star polymer, we found that the polymer acts like a shield against the flow, leading to additional hydrodynamic drag forces that compress the arm chains in the front of the star center toward the tube axis and lift the arm chains at the back toward the tube wall. The balanced hydrodynamic forces freeze the polymer into a trumpet structure, where the arm chains maintain a steady strongly stretched state at high flow strength. In contrast, the polymer displays remarkably large conformational change when switching off HIs. Our simulation results explained the coupling between HIs and the structure of star polymers in Poiseuille flow.

Keywords Hydrodynamic interactions; Star polymer; Poiseuille flow; Migration

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INTRODUCTION

The drastic conformational changes of soft mesoscopic particles induced by the external fields are of paramount importance from both a fundamental and an applied viewpoint.^[1,2] Quite rich intriguing dynamical behaviors, such as tank-treading, tumbling, etc., have been found for deformable soft objects, in particular for polymers.^[3–6] Such complex motions strongly perturb the fluid flow field through hydrodynamic interactions.^[7,8] Thus, there is a growing interest to elucidate the coupling between the deformable soft matters and the perturbed flow field. Star polymers present a special macromolecular architecture in which several linear polymers of identical length are linked together by one of their ends to a common center. This morphology is particularly fascinating because we can study the continuous change of properties from a linear polymer to a spherical colloidal particle by tuning the number of arms. The properties of star polymers in and near equilibrium have been studied intensively, from both theoretical and experimental aspects.^[9–13] However, in the nonequilibrium conditions, the coupling between the deformation of star polymers and hydrodynamic interactions within the polymer is still poorly

understood.

The motion of one part of the polymer modifies the hydrodynamic forces acting on the other part of the chain by perturbing the surrounding flow.^[14] If the polymer is near equilibrium, the hydrodynamic coupling results in the polymer dynamics like a solid object moving through the surrounding solvent and this chain is said to be “non-draining”. Zimm^[15] predicted that the total hydrodynamic force acting on the “non-draining” chain scales as $F_{\text{drag}} \sim L^{0.5}$, where L is the length of the polymer. If the polymer is strongly stretched, the chain is often assumed to be “free-draining”,^[16–20] acting on which the hydrodynamic drag F_{drag} scales linearly with L .^[21] In contrast to this simple picture, experimental studies of a tethered polymer in a uniform flow by fluorescence microscopy have revealed that the hydrodynamic coupling within the chain is not negligible even if it has been stretched as $\sim 0.8L$.^[22] Moreover, hydrodynamic simulations have shown that a highly stretched tethered polymer in shear flow reduces the local shear rate remarkably by sticking the surrounding solvent.^[23,24] These observations have challenged a fundamental hypothesis in all theories dealing with a polymer in flow that the presence of the polymer does not affect the solvent flow profile.

Most practical flows, such as shear flow, consist of a mixture of both rotational and extensional components.^[25] Unlike the pure elongational flow, in which a polymer can exist in a stable stretched state, this combination of stretching and

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rotation causes large conformational changes, in particular for a free polymer,^[26–33] and hence hydrodynamic interactions (HIs) between monomers in the chain fluctuate significantly. To calculate analytically, theoretical descriptions generally neglect such hydrodynamic fluctuations.^[34–36] Nevertheless, Winkler's observations of the deformation of a free semiflexible polymer in shear flow agreed well with those based on experiment and simulation over a wide range of shear rate, suggesting the weak dependence on HIs.^[36] Hydrodynamic simulations, however, have also revealed a non-monotonic stretching response at large shear rates, which is believed to be caused by HIs.^[37–40]

In the present work, we applied a hybrid mesoscopic hydrodynamic simulation approach to study the effect of HIs on the deformation of star polymers in Poiseuille flow over a wide range of flow strengths. The standard molecular dynamics simulation describes the polymers, and the solvent is treated explicitly by using the multi-particle collision dynamics (MPCD) method. In order to gain a deep understanding of HIs on polymer deformations, a comparison of distributions and conformational properties between the systems with and without HIs is made. We show that the star polymer migrates toward the centerline of the tube as flow strength increases, and performs as a stable trumpet structure at high flow strength. In contrast, the polymer migrates toward the tube wall with dramatic conformational changes in the absence of HIs.

The outline of the paper is as follows. At first, we briefly describe the employed model and simulation approach. Then, we systemically investigate the effects of HIs on the distribution, conformation, and dynamics across the tube of a free star polymer exposed to a tube flow, and provide new insight into the coupling between HIs and the deformation of star polymers. Finally, we summarize the findings and draw our conclusions.

MODEL AND SIMULATION METHOD

Star Polymer

In our model system, a star polymer is composed of n linear polymers with one end linked to a common center. Each linear polymer has N_f monomers of mass M each and thus the total number of monomers for a star polymer is $N = nN_f + 1$. In our simulations, we studied the deformation of the star polymer with the number of arms $n = 20$ and the arm length $N_f = 10$. Short arms can easily achieve a highly stretched state under flow and thus are convenient for studying the hydrodynamic coupling within the stretched polymer. Excluded-volume interactions between monomers are taken into account by a shifted and truncated Lennard-Jones (LJ) potential:^[41]

$$U_{\text{LJ}} = \begin{cases} 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] + \epsilon, & r \leq r_c \\ 0, & \text{else} \end{cases} \quad (1)$$

here, $r = |\mathbf{r}_i - \mathbf{r}_j|$ denotes the spatial distance between monomer i and j located at $\mathbf{r}_i$ and $\mathbf{r}_j$. σ and ϵ are taken as the unit length and energy, respectively. The short-range, repulsive interaction is achieved by using the cutoff radius $r_c = 2^{1/6}\sigma$.

The connectivity between adjective monomers is described by a finitely extensible nonlinear elastic (FENE) potential:^[42]

$$U_{\text{FENE}} = -\frac{\kappa}{2} R_0^2 \ln \left[1 - \left(\frac{r}{R_0} \right)^2 \right] \quad (2)$$

where R_0 is the maximum bond length and κ is the spring constant. $R_0 = 1.5\sigma$ and $\kappa = 30\epsilon/\sigma^2$ are chosen to avoid bond crossing.^[43] The velocity Verlet algorithm with time step h_p is used to integrate Newton's equations of motion of monomers.

The relaxation time of star polymer chain τ_p is determined by an exponential decay autocorrelation function of center-to-end vector,^[44] $C_{\mathbf{R}}(t)$:

$$C_{\mathbf{R}}(t) = C_0(t) \exp(-t/\tau_p) \\ = \langle (\mathbf{R}(t) \cdot \mathbf{R}(0)) - \langle \mathbf{R} \rangle^2 \rangle / (\langle \mathbf{R}^2 \rangle - \langle \mathbf{R} \rangle^2) \quad (3)$$

where $\mathbf{R}$ is the center-to-end vector for a single arm and $C_0(t)$ is the adjustable fitting parameter.

Hydrodynamic Solvent

The explicit solvents are modeled as point-like particles of mass m in MPCD.^[45–47] The MPCD algorithm consists of two alternating streaming and collision steps. In the streaming step, solvents move ballistically within the time interval h between collisions and the position of solvent i is updated according to

$$\mathbf{r}_i(t+h) = \mathbf{r}_i(t) + \mathbf{v}_i(t)h \quad (4)$$

where $\mathbf{r}_i(t)$ and $\mathbf{v}_i(t)$ denote the position and velocity of solvent particle i at time t , respectively. In the collision step, all the particles are sorted into cubic cells of edge length a and their relative velocities, with respect to the center-of-mass velocity of the cell, are rotated around a randomly oriented axis by a fixed angle α , i.e.:^[48]

$$\mathbf{v}_i(t+h) = \mathbf{v}_{\text{cm}}(t) + \mathbf{R}(\alpha) [\mathbf{v}_i(t) - \mathbf{v}_{\text{cm}}(t)] \quad (5)$$

where $\mathbf{v}_i(t)$ denotes the velocity of particle i at time t , and $\mathbf{R}(\alpha)$ is the rotation matrix.

$$\mathbf{v}_{\text{cm}} = \frac{\sum_{i=1}^{N_c^s} m\mathbf{v}_i(t) + \sum_{j=1}^{N_c^m} M\mathbf{v}_j(t)}{mN_c^s + MN_c^m} \quad (6)$$

is the center-of-mass velocity of all particles in the cell. N_c^s and N_c^m are the numbers of solvent and monomers in the collision cell, respectively. The mass, local momentum, and kinetic energy are conserved during the collision. To ensure the Galilean invariance, a random shift of the collision grid is performed at every collision step.^[49,50] A cell-level Maxwellian thermostat is used to maintain the temperature of the fluid at the desired value.^[51]

HIs between monomers can be switched off in MPCD, where each monomer independently performs stochastic collisions with a phantom particle which mimics a fluid element of size a^3 . The phantom particle momentum is taken from a Maxwell-Boltzmann distribution of variance $mpk_B T$ and a mean $\mathbf{P} = (mpv_x, 0, 0)$,^[31] where k_B is the Boltzmann constant and T is the temperature.

All simulations are performed with $\alpha = 130^\circ$, $\sigma = \epsilon = a = 1$, the average number of solvent particles per cell $\rho = 10$, $m = 1$, $M = \rho m$, the time unit $\tau_0 = \sqrt{m\sigma^2/k_B T}$, $h = 0.1\tau_0$, $h_p = 0.005\tau_0$, and $k_B T = 1$. With the selected parameters, the viscosity of the fluid $\eta = 8.7\sqrt{mk_B T/\sigma^2}$, and the Schmidt number $Sc = 16.9$, which have been shown to be reliable to account for the properties of liquid-like MPC fluid.^[52] For each simulation run,

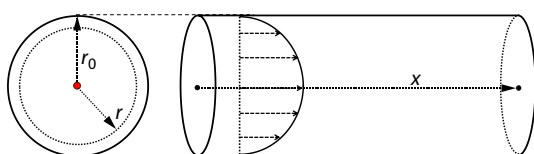
polymer conformational data are recorded after their averages reach a steady value, which is confirmed by initializing the run with different random chain conformations and enlarging the initializing run time at least ten times.

Poiseuille Flow

A Poiseuille flow is created by applying a body force,^[53–56] $F_g = mg$ where g denotes the acceleration parameter and characterizes the flow strength, on the solvent to drive them through a cylindrical pipe of radius r_0 along the flow direction (x) where periodic boundary conditions are imposed. The radius and length of the cylindrical channel are $r_0 = 13$ and $L_{\text{tube}} = 120\sigma$, respectively, as shown Scheme 1. The bounce-back interactions between solvent and tube wall are implemented where virtual particles in the wall are included to guarantee the non-slip boundary conditions.^[57,58] Solving the Navier-Stokes equation for the tube geometry gives a parabolic flow profile,

$$v_x(r) = \rho g (r_0^2 - r^2) / 4\eta \quad (7)$$

where r is the radial distance from the tube axis. The maximum fluid velocity $v_{\text{max}} = \rho g r_0^2 / 4\eta$ and the Reynolds number $\text{Re} = \rho r_0 v_{\text{max}} / \eta$.^[58,59] The simulations are implemented at the acceleration parameters g ranging from 0 to $0.02k_B T / m\sigma$. Thus, the maximum value of Reynolds number $\text{Re}_{\text{max}} = 14.5$, far below that of the turbulent flow, $\text{Re} = 1000$,^[52] illustrating that the fluid flow in the simulations is laminar. Fig. 1 shows the time-averaged solvent velocity profile of fluids flowing through a tube of radius $r_0 = 13\sigma$ in the absence of polymers. The simulation data are in quantitative agreement with those calculated by Eq. (7).



Scheme 1 Scheme of the Poiseuille flow in a cylindrical channel of radius r_0 along the flow direction (x). r is the radial distance from the center-of-mass of the star polymer to the centerline of the tube.

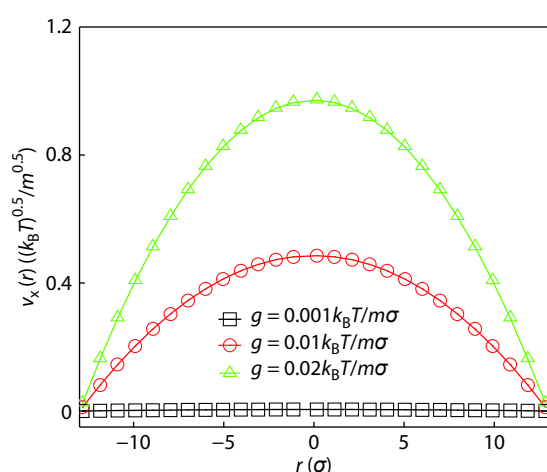


Fig. 1 Velocity profiles of the fluid along the flow direction (x axis) in the tube. The solid lines indicate the theoretical velocity profiles of the Poiseuille flow in the tube, obtained from Eq. (7).

RESULTS AND DISCUSSION

Radial Density Distribution

The imposed parabolic Poiseuille flow drives star polymers out of the equilibrium and determines its steady state. Such phenomenon can be characterized by the radial center-of-mass distribution function $P_{\text{cm}}(r)$,

$$1 = \int_0^{r_0} P_{\text{cm}}(r) 2\pi r dr \quad (8)$$

where r is the radial distance between the center-of-mass of star polymers and tube axis. Fig. 2 displays the radial center-of-mass distribution functions $P_{\text{cm}}(r)$ of star polymers for various flow strength g . At low flow strength ($g = 0.00001k_B T / m\sigma$), the system is close to the equilibrium state. The distribution is fairly uniform across the tube, and there is one depletion layer near the tube wall which extends approximately one radius of gyration ($R_g \approx 4.23\sigma$) into the tube. With increasing the flow strength, star polymers migrate toward the centerline induced by HIs, which is reflected by the fact that the star polymers are more depleted from the tube wall, and their concentration at the tube center increases. At high flow strength ($g = 0.02k_B T / m\sigma$), the concentration of star polymers in the center of the tube ($P_{\text{cm}}(0) \approx 0.88$) is more than one hundred times higher than that of the low flow strength case ($P_{\text{cm}}(0) \approx 0.0066$). Moreover, the widths of the distribution functions become narrower gradually, illustrating that the position of star polymers in the tube at high flow strength is stable. For the linear and ring chains, two main factors affect the distributions of polymers: the Brownian motion drives polymers toward the tube wall, and HIs drive polymers toward the tube center. The competition of the above two factors results in the bimodal structures of P_{cm} .^[26,27,34] However, the bimodal structure of P_{cm} is not observed for star polymers. We attribute this to the more symmetric architecture and weaker deformability of star polymers than linear and ring chains.

To reveal the effect of HIs on the migration behavior of star polymers, we perform simulations in which HIs are completely switched off, corresponding to a free-draining chain dragged by the shear gradient. In equilibrium, the distribu-

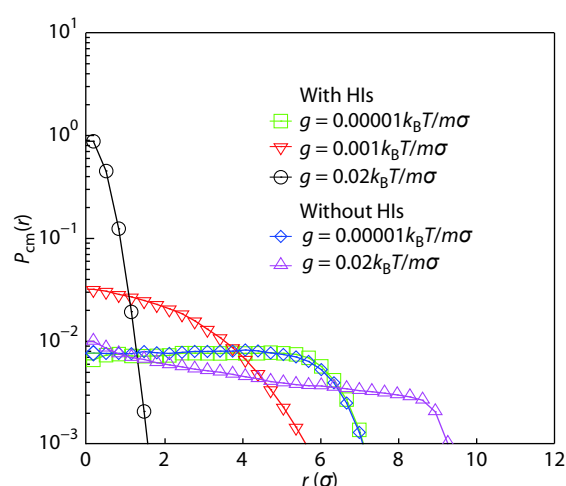


Fig. 2 Center-of-mass distributions of the star polymers with the number of arms $n = 20$ and the arm length $N_f = 10$ in the tube with and without hydrodynamic interactions.

tion profiles of star polymers with and without HIs present an identical platform structure away from the tube wall, so that HIs do not affect equilibrium distribution of star polymers. However, at high flow strength, HIs significantly affect the distribution functions of star polymers: when HIs are switched off, the distribution function of star polymers becomes wider, leading to a less pronounced depletion layer near the wall (the width of the depletion layer is about $0.5R_g$) compared with the equilibrium result. In contrast, when including HIs, the distribution function of star polymers presents a remarkable single-peak structure in the center of the tube with a much wider depletion layer (the width of the depletion layer is about $2.5R_g$).

Conformational Revolution

In Poiseuille flow, star polymers suffer the position-dependent shear flow and deform as a consequence. To evaluate the role of HIs on the deformation of star polymers, we calculate the time-fluctuating radial position $r(t)$ and the resulting instantaneous conformation of star polymers with HIs, and compare the results with those in the absence of HIs. The conformation properties of star polymer can be conveniently measured by the gyration tensor, $G_{\alpha\beta}(t)$, defined as:

$$G_{\alpha\beta}(t) = \frac{1}{N} \sum_{i=1}^N \Delta r_{i,\alpha}(t) \Delta r_{i,\beta}(t) \quad (9)$$

where $\Delta r_i(t)$ is the distance between monomer i and the center-of-mass of a star polymer at time t , and $\alpha, \beta \in (x, y, z)$ are the Cartesian components. The diagonal components $G_{\alpha\alpha}(t)$ are the squared radius of gyration in the α direction. The eigenvalues of gyration tensor $G_{\alpha\beta}$ give the principle axes of polymer chains which denote the shape of polymer chains. In the cylindrical tube, we apply $G_{xx}(t)$ and $G_{rr}(t) = G_{yy}(t) + G_{zz}(t)$ to represent the sizes of star polymer along the flow direction and along the radial direction, respectively.

Fig. 3 shows the sample time trajectories of the radial position of star polymers and the components of the radius of gyration tensor along the flow and radial directions. At low flow strength ($g = 0.00001k_B T/m\sigma$), star polymers undergo the Brownian motion in the radial direction with large position fluctuations. The sizes of star polymers fluctuate around the equilibrium values in both flow and radial directions, illustrating that star polymers have a coil conformation. With increasing the flow strength, star polymers migrate close to the centerline and the fluctuations of polymer position along the radial direction become smaller. Meanwhile, star polymers become more stretched in the flow direction and more shrunk in the radial direction. Furthermore, we compare the radial position and the size of star polymers in the systems with and without HIs at high flow strength ($g = 0.02k_B T/m\sigma$). When including HIs, the star polymer is stable near the centerline with a stretched conformation along the flow direction. In contrast, when switching off HIs, the polymer prefers to migrate toward the tube wall. Besides that, the stretching in the flow direction and shrinkage in the radial direction are more pronounced than those with HIs. We attribute this phenomenon to the fact that the motion of one part of the polymer affects the other part of the chain through HIs, thus the lower viscous drag force on the polymer chain, leading to

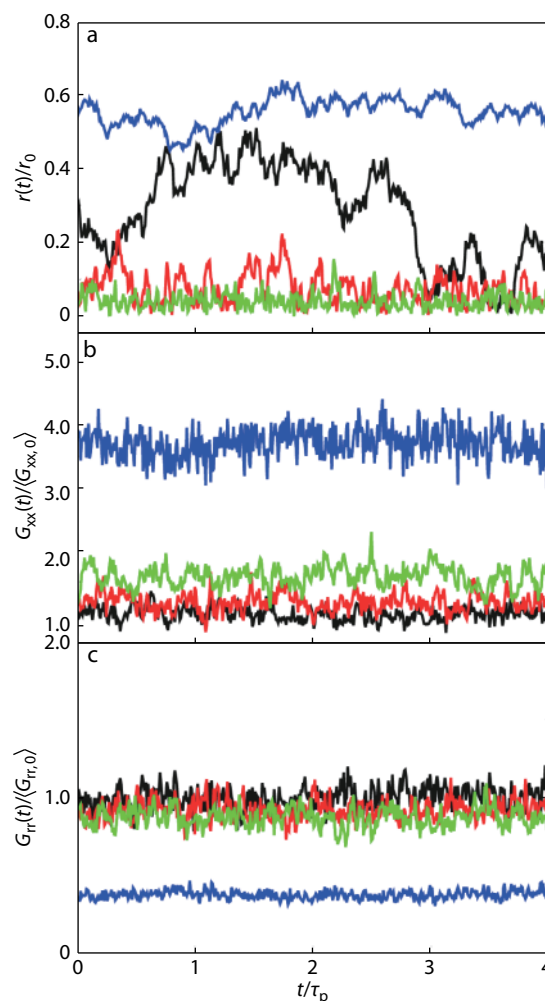


Fig. 3 (a) Normalized time trajectories of the radial distance $r(t)$ between the center-of-mass of star polymers and tube axis, and normalized components of the radius of gyration along (b) the flow direction $G_{xx}(t)$ and (c) the radial direction $G_{rr}(t)$. $\langle G_{xx,0} \rangle$ and $\langle G_{rr,0} \rangle$ are equilibrium components of radius of gyration. $\tau_p = 11905\tau_0$ is the longest relaxation time of star polymer. The colors of lines correspond to the flow strengths $g = 0.00001k_B T/m\sigma$ (black), $0.005k_B T/m\sigma$ (red), $0.02k_B T/m\sigma$ (green) with hydrodynamic interactions, and $0.02k_B T/m\sigma$ without hydrodynamic interactions (blue).

weaker stretching than that without HIs.

In order to clearly clarify the structure of the star polymer under Poiseuille flow, we calculate the end monomer distributions of star polymers along the radial and flow directions for various flow strengths g , and normalize them by the arm number $n = 20$, as shown in Fig. 4. At low flow strength, star polymers perform as a coil conformation in the tube. The distributions of end monomers along the radial direction perform an identical structure in both front and back of the center core of star polymers. In addition, the end monomers distribute symmetrically around the center of star polymers in the flow direction. The corresponding distribution function $N(x_e)$ exhibits a platform region near the center of polymers. As the flow strength increases, the end monomers in the front are more concentrated in the centerline. In contrast, $N_B(r_e)$ ex-

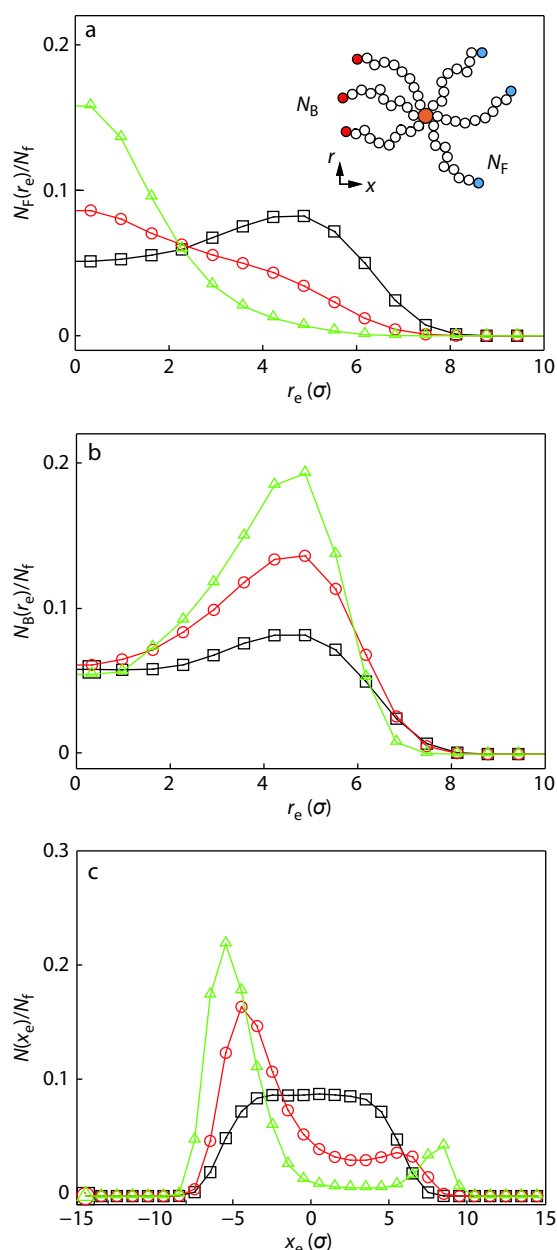


Fig. 4 Normalized end monomers distributions of the star polymer with $n = 20$ arms and length $N_f = 10$ in Poiseuille flow along radial direction (a) in the front and (b) in the back of the center of the polymers, and (c) along the flow direction. r_e is the radial distance between the end monomer and the centerline of the tube. x_e is the axial distance between the end monomer and the center core of the star polymer. The colors of lines correspond to the flow strengths $g = 0.0001 k_B T / m\sigma$ (black), $0.005 k_B T / m\sigma$ (red), and $0.02 k_B T / m\sigma$ (green). Inset: the illustration of end monomers in the front (blue) and in the back (red) of the polymer center core (orange).

hibits a pronounced off-center peak. In the flow direction, $N(x_e)$ becomes asymmetric as g increases, with fewer arms staying in the front of the polymer center and more arms in the back. At high flow strength ($g = 0.02 k_B T / m\sigma$), the front arms are concentrated near the centerline, whereas the back arms are concentrated at $r_e = 5\sigma$. Moreover, two distinct

peaks in $N(x_e)$ locate at $x = -6\sigma$ and $x = 8\sigma$. From the above, we could speculate the corresponding structure of star polymers at high flow strength performs as a trumpet structure composed by a rod part from the front arms and an empty shell part from the back arms.

The above considerations of polymer structures can be confirmed by the characteristic snapshots, which help us to visualize the conformation of star polymers in Poiseuille flow. When including HIs in MPCD, the snapshots of star polymers for various flow strengths g are shown in Fig. 5. At the low flow strength, star polymers perform as a coil conformation which continuously alternate between collapsed and stretched configurations. As g increases, this continuous conformational change is interrupted by the appearance of the trumpet structure. This trumpet structure becomes more stable with increasing g and eventually holds at large g . Such observation verifies the speculation from the results of end monomer distribution at the high flow strength: almost all arm chains maintain a steady stretched state in the stable trumpet structure, among which a small amount is in front of the center core but the others are at the back. The front arms are compressed around the tube axis and the back arms display a perfect axial symmetry shell. A Poiseuille flow consists of both rotational and extensional components, suggesting that the star polymer will not attain a stable, strongly stretched state as in a steady shear flow. The trumpet structure of the star polymer indicates that hydrodynamic drag forces exerted by the rotational component of the flow on the arm chains are obviously balanced by certain forces due to the perturbation of the star polymer.

When switching off HIs in MPCD, the star polymer becomes free-draining and exhibits completely different conformational change with increasing flow strength g . The simulation shows that the arm chains of the “free-draining” star polymer maintain in a highly stretched state (the snapshots are not shown) and display remarkably large conformational changes at high flow strength. These results suggest that HIs are crucial for the formation of the trumpet structure.

Perturbed Flow

To seek hydrodynamic coupling to stabilize star polymers in a stretched state, we analyse the flow fields perturbed by the star polymer in the trumpet shape. The perfect axial symmetry and center-anchored conformation of the star polymer allow us to directly describe the perturbed flow on the flow (x)-radial (r) plane of the star center reference frame. Fig. 6(a) shows the perturbed velocity field $\Delta \mathbf{v}(x, r)$ which is the velocity difference between the local fluid flow velocity $\tilde{\mathbf{v}}(x, r)$ obtained from simulation and the theoretical velocity $\mathbf{v}(x, r)$ at $g = 0.02 k_B T / m\sigma$. The star polymer acts like a shield against the flow, giving rise to a backflow. Fig. 6(b) shows that the flow surrounding the polymer center ($r = 1\sigma$) slows down, while the flow surrounding the back arms ($r = 5\sigma$) speeds up. The decrease in the velocity at the polymer center results in an extensional force that elongates the front arms along the tube axis. When the star polymer chain holds stationary with a fixed shape at tube axis in Poiseuille flow, namely its velocity along the radial direction $v_{r,p} = 0$, it requires a zero net external force acting on the polymer chain. In our system, the only external force along the radial direction is the

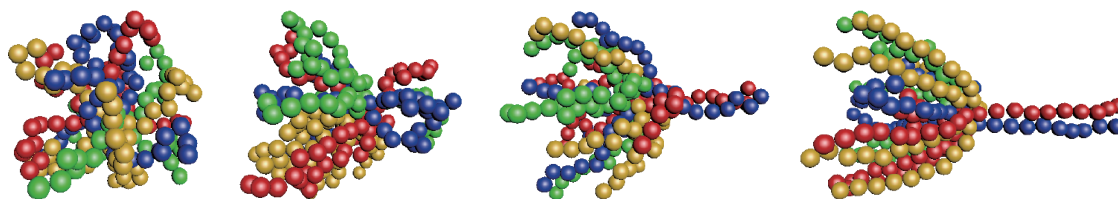


Fig. 5 Schematic diagram of the star polymer with $n = 20$ arms and length $N_f = 10$ in Poiseuille flow at various flow strengths g with hydrodynamic interactions. The images from left to right correspond to the flow strength $g = 0.00001 k_B T / m \sigma$, $0.005 k_B T / m \sigma$, $0.01 k_B T / m \sigma$, and $0.02 k_B T / m \sigma$. For illustration, arm chains are highlighted in various colors.

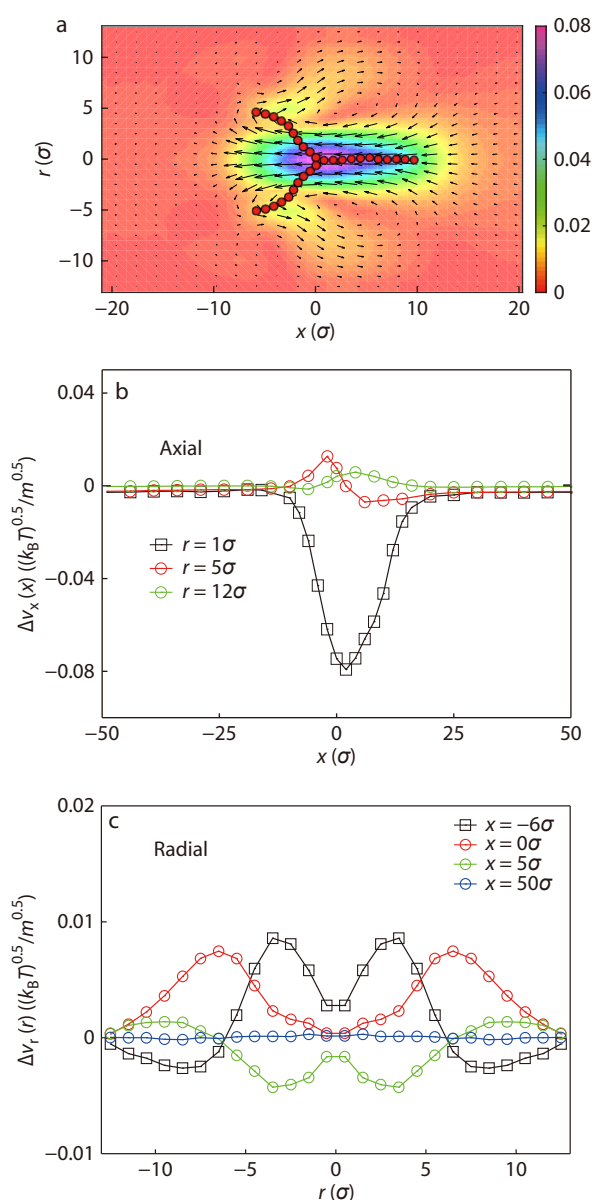


Fig. 6 Perturbed velocity fields of the liquids flowing through the tube of radius $r_0 = 13\sigma$ along the x direction at $g = 0.02 k_B T / m \sigma$ in the presence of the star polymer of $n = 20$ and $N_f = 10$. (a) The perturbed flow field $\Delta \mathbf{v}(x, r)$, represented by the vectors. Color bar denotes magnitudes of $\Delta \mathbf{v}(x, r)$. The arrows represent the direction of the perturbed flow. Inset: Schematic illustration of the cross-section view of the star polymer. (b) The component along the flow direction Δv_x and (c) the component along radial direction Δv_r of $\Delta \mathbf{v}(x, r)$.

hydrodynamic drag force by the solvent, which can be written as $f_{r,i} = \zeta(\tilde{v}_{i,r} - v_{r,p})$, where $\tilde{v}_{i,r}$ is the radial velocity of the solvent around bead i and ζ is the friction constant of solvent. Please note that the theoretical velocity of the flow along the radial direction is $v_r = 0$, and the perturbed velocity field can be written as $\Delta \mathbf{v}_r = \tilde{v}_r - v_r = \tilde{v}_r$. Thus, the hydrodynamic drag force $f_{r,i}$ is proportional to Δv_r . Strikingly, the radial component of the flow surrounding the arm chains is nonzero, as shown in Fig. 6(c). We find that Δv_r is positive in the region $-5\sigma < r < 5\sigma$ at $x = -6\sigma$, illustrating that the hydrodynamic drag force f_r is outward that lifts the back arms toward the tube wall. Similarly, Δv_r is negative in the region $-5\sigma < r < 5\sigma$ at $x = 5\sigma$, illustrating that the hydrodynamic drag force f_r is inward that compresses the front arms toward the tube axis. The two radial hydrodynamic forces represent the nature of the effects of HIs on polymer deformation and the crucial discrepancy between free-draining and non-draining systems. The regular Poiseuille flow is recovered far from the star polymer.

CONCLUSIONS

In conclusion, we studied the effects of HIs on the distribution and conformation of a free star polymer in Poiseuille flow through a tube. Near the equilibrium state, star polymers distribute uniformly with a coil conformation. When increasing the flow strength g , star polymers migrate close to the centerline of the tube due to HIs, and the fluctuations of polymer position become smaller along the radial direction. In contrast, when switching off HIs, star polymers migrate toward the tube wall, and behave like a free-draining chain with a large conformational change. We calculated the end monomer distribution and perturbation of the flow around the star polymer, which allowed us to analyse how hydrodynamic forces act on the polymer. We showed that the star polymer perturbs the surrounding flow at high flow strength, giving rise to a backflow which exerts additional hydrodynamic drag forces that compress the front arm chains toward the tube axis and lift the back arm chains toward the tube wall. The hydrodynamic forces are balanced and freeze the polymer into a trumpet structure, where the arm chains maintain a steady strongly stretched state at high flow strength. These results conflict with the predictions from theory and Brownian dynamic simulation that do not include the contribution of hydrodynamic coupling within the polymer. Our simulations comprehensively address the interaction of stretched polymers with the surrounding flow. These results shed new light on the microscopic understanding of the non-Newtonian rheology of polymer solutions.

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