Supplementary Information: The stress deformation response influenced by the chain rigidity for mesostructures in diblock copolymers

Gaohang Chen,[†,‡] Hui Zhang,[‡] Teng Lu,[¶] and Ying Jiang[*,†,§]

†*School of Chemistry & Key Laboratory of Bio-Inspired Smart Interfacial Science and Technology of Ministry of Education & Center of Soft Matter Physics and Its Applications, Beihang University, Beijing 100191, China*

‡*School of Mathematical Sciences, Beijing Normal University, Beijing 100875, China*

¶*Computer Network Information Center of the Chinese Academy of Sciences, Beijing 100190, China*

§*Beijing Advanced Innovation Center for Biomedical Engineering, Beihang University, Beijing 100191, China*

E-mail: yjiang@buaa.edu.cn

Supporting Information Available

I. The derivation of MDE in the nonorthogonal coordinate system

In this section, we concentrate on the derivation of modified diffusion equation (MDE) for a wormlike chain in the nonorthogonal coordinates by means of the Chapman-Kolmogorov equation, which is commonly utilized to determine the probability density for the perspective of the stochastic process closely related to the Brownian motion.

Assuming the Markov property for the propagator in an external field $w(\mathbf{x}, s)$, we can write

$$\begin{aligned} & q(\mathbf{x}, \mathbf{u}, s + \Delta s) \\ =& e^{-\Delta s\, w(\mathbf{x},s)} \int \mathrm{d}(\Delta\mathbf{x}) \int \mathrm{d}(\Delta\mathbf{u})\, \Psi(\Delta\mathbf{x}, \Delta\mathbf{u}; \mathbf{x} - \Delta\mathbf{x}, \mathbf{u} - \Delta\mathbf{u})\, q(\mathbf{x} - \Delta\mathbf{x}, \mathbf{u} - \Delta\mathbf{u}, s), \end{aligned} \tag{1}$$

where $\Psi(\Delta\mathbf{x}, \Delta\mathbf{u}; \mathbf{x} - \Delta\mathbf{x}, \mathbf{u} - \Delta\mathbf{u})$ represents the conditional transition probability [1] that the added segment has the positional and orientational displacements $\Delta\mathbf{x}$ and $\Delta\mathbf{u}$ in the nonorthogonal coordinates, propagating from the position $\mathbf{x} - \Delta\mathbf{x}$ with the orientation $\mathbf{u} - \Delta\mathbf{u}$, due to an increment Δs of the contour variable. Considering the relation $\mathbf{r} = \mathbf{h} \cdot \mathbf{x}$ resulting from the coordinate transformation, we can convert a displacement $\Delta\mathbf{r}$ in the Cartesian coordinate to its counterpart $\Delta\mathbf{x}$ in the nonorthogonal coordinates and have the form

$$\Delta\mathbf{x} = \mathbf{h}^{-1}\Delta\mathbf{r} = \mathbf{h}^{-1} \int_s^{s+\Delta s} \mathrm{d}s\, \mathbf{u}(s) = (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s + O(\Delta s^2). \tag{2}$$

which explicitly restricts the positional displacement $\Delta\mathbf{x}$ by Δs and $\mathbf{u}$. Followed by a shift $\mathbf{x} \rightarrow \mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s$, then, the formula (1) can be simplified further into

$$q\Big(\mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s, \mathbf{u}, s + \Delta s\Big) e^{\Delta s\, w(\mathbf{x},s)} = \int \mathrm{d}(\Delta\mathbf{u})\, \Phi(\Delta\mathbf{u}; \mathbf{x}, \mathbf{u} - \Delta\mathbf{u})\, q(\mathbf{x}, \mathbf{u} - \Delta\mathbf{u}, s), \tag{3}$$

where the normalized probability $\Phi(\Delta\mathbf{u}; \mathbf{x}, \mathbf{u})$ regulated only by $\Delta\mathbf{u}$ due to a contour step size Δs is introduced as

$$\Psi(\Delta\mathbf{x}, \Delta\mathbf{u}; \mathbf{x}, \mathbf{u}) = \Phi(\Delta\mathbf{u}; \mathbf{x}, \mathbf{u})\, \delta\left(\Delta\mathbf{x} - (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s\right). \tag{4}$$

Expanding the modified Chapman-Kolmogorov equation Eq. (3) in terms of Δs and $\Delta\mathbf{u}$, we arrive at

$$\begin{aligned}
&[1 + w(\mathbf{x}, s)]\, e^{\Delta s\, w(\mathbf{x},s)}\, q(\mathbf{x}, \mathbf{u}, s) + e^{\Delta s\, w(\mathbf{x},s)} \left\{ \Delta s \nabla_{\mathbf{x}} q\left(\mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s, \mathbf{u}, s + \Delta s\right) \cdot \mathbf{G} \cdot (\mathbf{h}^{-1} \cdot \mathbf{u}) \right. \\
&\left. + \Delta s \frac{\partial q\left(\mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s, \mathbf{u}, s + \Delta s\right)}{\partial s} + O(\Delta s^2) \right\} \\
=& q(\mathbf{x}, \mathbf{u}, s) - \nabla_{\mathbf{u}} \cdot [\langle \Delta\mathbf{u} \rangle_{\Phi} q(\mathbf{x}, \mathbf{u}, s)] + \frac{1}{2!} \nabla_{\mathbf{u}} \nabla_{\mathbf{u}} : [\langle \Delta\mathbf{u}\Delta\mathbf{u} \rangle_{\Phi} q(\mathbf{x}, \mathbf{u}, s)] + O(\langle \Delta\mathbf{u}\Delta\mathbf{u}\Delta\mathbf{u} \rangle_{\Phi})
\end{aligned} \tag{5}$$

where $\nabla_{\mathbf{x}}$ and $\nabla_{\mathbf{u}}$ denotes the gradient with respect to variables $\mathbf{x}$ and $\mathbf{u}$, and the metric matrix $\mathbf{G} = \mathbf{h}^{\intercal}\mathbf{h}$ is introduced. $<>_{\Phi}$ represents the integral with respect to the transition probability density Φ. In the coordinate system described by the nonorthogonal basis $\{\mathbf{h}_1, \mathbf{h}_2, \mathbf{h}_3\}$, in particular, we need to carefully deal with $\nabla_{\mathbf{x}} f(\mathbf{x})$ as[2]

$$\nabla_{\mathbf{x}} f(\mathbf{x}) = \left(\frac{\partial}{\partial x_1}, \ \frac{\partial}{\partial x_2}, \ \frac{\partial}{\partial x_3} \right) f(\mathbf{x}) \cdot \mathbf{G}^{-1}. \tag{6}$$

Substituting the expression (6) into the left-hand side (LHS) of Eq. (5), with truncation error $O(\Delta s^2)$, we can have

$$\begin{aligned}
\text{LHS} =& e^{\Delta s\, w(\mathbf{x},s)} \left\{ 1 + \Delta s \left[\frac{\partial}{\partial s} + \mathbf{u} \cdot \left(\mathbf{h}^{-1}\right)^{\intercal} \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3} \right)^{\intercal} + w(\mathbf{x}, s) \right] \right\} \\
& q\left(\mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s, \mathbf{u}, s + \Delta s\right),
\end{aligned} \tag{7}$$

while the right-hand side of Eq. (5) straightforward reads[1]

$$\text{RHS} = \left[1 + \frac{L\Delta s}{a} \nabla_{\mathbf{u}}^2 \right] q\left(\mathbf{x} + (\mathbf{h}^{-1} \cdot \mathbf{u})\Delta s, \mathbf{u}, s + \Delta s\right). \tag{8}$$

Replacing both sides of Eq. (5) by Eqs. (7) and (8), we can have

$$\left\{e^{\Delta s\, w(\mathbf{x},s)}\left[\frac{\partial}{\partial s}+\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^{\mathsf{T}}\cdot\left(\frac{\partial}{\partial x_1},\ \frac{\partial}{\partial x_2},\ \frac{\partial}{\partial x_3}\right)^{\mathsf{T}}+w(\mathbf{x},s)\right]\right\}q\Big(\mathbf{x}+(\mathbf{h}^{-1}\cdot\mathbf{u})\Delta s,\mathbf{u},s+\Delta s\Big) \\ =\left[-\frac{(e^{\Delta s\, w(\mathbf{x},s)}-1)}{\Delta s}+\frac{L}{a}\nabla_{\mathbf{u}}^2\right]q\Big(\mathbf{x}+(\mathbf{h}^{-1}\cdot\mathbf{u})\Delta s,\mathbf{u},s+\Delta s\Big). \tag{9}$$

After taking the continuous limit $\Delta s \to 0$, we eventually obtain the Fokker-Planck-like MDE

$$\frac{\partial}{\partial s}q(\mathbf{x},\mathbf{u},s)=\left[\frac{L}{a}\nabla_{\mathbf{u}}^2-L\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^{\mathsf{T}}\cdot\left(\frac{\partial}{\partial x_1},\ \frac{\partial}{\partial x_2},\ \frac{\partial}{\partial x_3}\right)^{\mathsf{T}}-w(\mathbf{x},s)\right]q(\mathbf{x},\mathbf{u},s). \tag{10}$$

II. Armijo-Goldstein inexact line search algorithm for the optimization of simulation cell

In this section, we briefly introduce the Armijo-Goldstein inexact line search algorithms that is used to optimize the shape and size of the simulation cell. A similar algorithm was previously utilized by Jiang *et al.*.[3]

In our calculations, the steepest descent method[4] is adopted to optimize the simulation cell matrix i.e. **h**. Since the matrix **h** will be updated iteratively, we focus on the component h_{ij}, $(i, j = 1, 2, 3)$ and hold the rest part of **h** unchanged as

$$\frac{\partial h_{ij}}{\partial t} = -\eta\,(a\rho_0/L)\frac{\partial F}{\partial h_{ij}}, \tag{11}$$

where the search direction is defined as the opposite direction of the gradient $\frac{\partial F}{\partial h_{ij}}$. The analytical expression of $\frac{\partial F}{\partial h_{ij}}$ is intractable so far, so we intend to numerically compute this gradient as

$$\frac{\partial F}{\partial h_{ij}} \approx \frac{F(h_{ij} + \Delta h_{ij}) - F(h_{ij})}{\Delta h_{ij}}. \tag{12}$$

The relaxation step size η is calculated by Armijo-Goldstein inexact linear search algorithm.[5] The calculation can be regarded as a one-dimensional optimization problem, which is to determine a step size η under given starting position h_{ij} and search direction $-\frac{\partial F}{\partial h_{ij}}$, to adequately reduces the objective function, which is chosen as the single chain configuration part $-\ln Q$ rather than the whole free energy F, so the computational resource of SCFT iterations can be saved highly. However, it is usually not wise to find the precisely value of η corresponding to the minimum, since it requires the full derivative information of the objective function in the vicinity of h_{ij}. The Armijo-Goldstein condition is chose because it only needs the point-wise derivative in the starting position h_{ij}, by which η satisfies the conditions as

$$\ln Q\left(h_{ij} - \eta\,(a\rho_0/L)\frac{\partial F}{\partial h_{ij}}\right) \;\geq\; \ln Q(h_{ij}) - \eta\,\rho\,\frac{\partial \ln Q}{\partial h_{ij}}(a\rho_0/L)\frac{\partial F}{\partial h_{ij}}, \tag{13}$$

$$\ln Q\left(h_{ij} - \eta\,(a\rho_0/L)\frac{\partial F}{\partial h_{ij}}\right) \;\leq\; \ln Q(h_{ij}) - \eta\,(1-\rho)\,\frac{\partial \ln Q}{\partial h_{ij}}(a\rho_0/L)\frac{\partial F}{\partial h_{ij}}, \tag{14}$$

where the parameter $0 < \rho < 0.5$ controls the degree of how inexactly η is calculated, and the $\dfrac{\partial \ln Q}{\partial h_{ij}}$ is also calculated numerically. This method can be explained as the first condition (13) ensures the decrease of the objective function, while the second (14) prevents the η from being too small. By this method, the step size η can be adjusted automatically to improve the numerical convergence.

III. Recovery of the Gaussian chain model for modified diffusion equation

The main task in the self-consistent field theory is to solve the modified diffusion equation (MDE),

$$\frac{\partial}{\partial s}q(\mathbf{x},\mathbf{u},s) = \left[\frac{L}{a}\nabla^2_{\mathbf{u}} - L\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^{\intercal}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\intercal} - w(\mathbf{x},s)\right]q(\mathbf{x},\mathbf{u},s), \tag{15}$$

which offers the probability of finding any segment along the polymer chain in space. In this section, we analytically verifies that Eq. (15) derived from the wormlike chain model in the limit of $L/a \gg 1$ can be reduced into the Gaussian chain MDE, which was previously obtained by Barrat *et al*. in the nonorthogonal simulation cell.[6]

In the current nonorthogonal coordinate system, the segment direction vector $\mathbf{u}$ is also constrained to be $|\mathbf{u}| = 1$. Then, the spherical harmonics basis $Y_\ell^m(\mathbf{u})$ is used to expand the propagator as

$$q(\mathbf{x},\mathbf{u},s) = \sum_{\ell=0}^{\infty}\sum_{m=-\ell}^{\ell} q_\ell^m(\mathbf{x},s)Y_\ell^m(\mathbf{u}). \tag{16}$$

Here the expansion coefficient reads

$$q_\ell^m(\mathbf{x},s) = \frac{1}{4\pi}\int \mathrm{d}\mathbf{u}Y_\ell^{m*}(\mathbf{u})q(\mathbf{x},\mathbf{u},s), \tag{17}$$

which can be obtained by using the orthogonality condition

$$\frac{1}{4\pi}\int \mathrm{d}\mathbf{u}Y_\ell^{m*}(\mathbf{u})Y_{\ell'}^{m'}(\mathbf{u}) = \delta_m^{m'}\delta_\ell^{\ell'}, \tag{18}$$

where, $Y_\ell^{m*}(\mathbf{u})$ is the complex conjugate to $Y_\ell^m(\mathbf{u})$ and satisfies $Y_\ell^{m*}(\mathbf{u}) = (-1)^m Y_\ell^{-m}(\mathbf{u})$.

The fact that $Y_l^m(\mathbf{u})$ is the eigenfunction of the operator $\nabla^2_{\mathbf{u}}$, directly leads to

$$\nabla^2_{\mathbf{u}}Y_\ell^m(\mathbf{u}) = -\ell(\ell+1)Y_\ell^m(\mathbf{u}). \tag{19}$$

By virtue of Eqs. (16) and (18), Eq.(15) straightforward arrives at

$$
\begin{aligned}
\frac{\partial}{\partial s} q_\ell^m(\mathbf{x}, s) = & \left[-\ell(\ell-1)\frac{L}{a} - w(\mathbf{x}, s)\right] q_\ell^m(\mathbf{x}, s) \\
& - \frac{L}{4\pi}\int \mathrm{d}\mathbf{u} Y_\ell^{m*}(\mathbf{u}) \left[\sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'} Y_{\ell'}^{m'}(\mathbf{u})\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^\intercal \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3}\right)^\intercal q_{\ell'}^{m'}(\mathbf{x}, s)\right].
\end{aligned} \tag{20}
$$

For expressing the last term at the right-hand side of Eq. (20), we aim to simplify the formula $\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^\intercal \cdot (\partial/\partial x_1, \partial/\partial x_2, \partial/\partial x_3)^\intercal q_{\ell'}^{m'}(\mathbf{x}, s)$. The unit vector $\mathbf{u}$ can be rewritten in the nonorthogonal coordinates as

$$
\mathbf{u} = (\sin\theta\cos\phi,\ \sin\theta\sin\phi,\ \cos\theta)\cdot\left(\mathbf{h}^{-1}\right)^\intercal \cdot \begin{pmatrix}\mathbf{h}_1\\ \mathbf{h}_2\\ \mathbf{h}_3\end{pmatrix} \tag{21}
$$

Then, we have

$$
\begin{aligned}
& \mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^\intercal \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3}\right)^\intercal q_{\ell'}^{m'}(\mathbf{x}, s) \\
= & \left[(\sin\theta\cos\phi,\ \sin\theta\sin\phi,\ \cos\theta)\cdot\left(\mathbf{h}^{-1}\right)^\intercal \begin{pmatrix}\mathbf{h}_1\\ \mathbf{h}_2\\ \mathbf{h}_3\end{pmatrix}\right]\cdot\left[\left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3}\right) q_{\ell'}^{m'}(\mathbf{x}, s)\cdot\mathbf{G}^{-1}\cdot\begin{pmatrix}\mathbf{h}_1\\ \mathbf{h}_2\\ \mathbf{h}_3\end{pmatrix}\right]^\intercal \\
= & (\sin\theta\cos\phi,\ \sin\theta\sin\phi,\ \cos\theta)\cdot\left(\mathbf{h}^{-1}\right)^\intercal \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3}\right)^\intercal q_{\ell'}^{m'}(\mathbf{x}, s).
\end{aligned} \tag{22}
$$

where the definition of the metric tensor $\mathbf{G} \equiv \mathbf{h}^\intercal\mathbf{h}$ is introduced. Further, applying the the recurrence formulas[7]

$$
\begin{aligned}
Y_{\ell'}^{m'}(\theta,\phi)\cos\theta = & \sqrt{\frac{(\ell'-m'+1)(\ell'+m'+1)}{(2\ell'+1)(2\ell'+3)}} Y_{\ell'+1}^{m'}(\theta,\phi) + \sqrt{\frac{(\ell'-m')(\ell'+m')}{(2\ell'-1)(2\ell'+1)}} Y_{\ell'-1}^{m'}(\theta,\phi), \\
Y_{\ell'}^{m'}(\theta,\phi)\sin\theta e^{\pm i\phi} = & \mp\sqrt{\frac{(\ell'\pm m'+1)(\ell'\pm m'+2)}{(2\ell'+1)(2\ell'+3)}} Y_{\ell'+1}^{m'\pm 1}(\theta,\phi) \pm \sqrt{\frac{(\ell'\mp m')(\ell'\mp m'-1)}{(2\ell'-1)(2\ell'+1)}} Y_{\ell'-1}^{m'\pm 1}(\theta,\phi),
\end{aligned} \tag{23}
$$

the last term at the right-hand side of Eq. (20) can be explicitly expressed as

$$
\begin{aligned}
&-\frac{L}{4\pi}\int \mathrm{d}\mathbf{u} Y_{\ell}^{m*}(\mathbf{u})\left[\sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'} Y_{\ell'}^{m'}(\mathbf{u})\mathbf{u}\cdot\left(\mathbf{h}^{-1}\right)^{\mathsf{T}}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\mathsf{T}} q_{\ell'}^{m'}(\mathbf{x},s)\right]\\
&=\sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'}\Big(X_1(m',\ell'),X_2(m',\ell'),X_3(m',\ell')\Big)\cdot\left(\mathbf{h}^{-1}\right)^{\mathsf{T}}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\mathsf{T}} q_{\ell'}^{m'}(\mathbf{x},s),
\end{aligned}
\tag{24}
$$

where we have

$$
\begin{aligned}
X_1(m',\ell') &= \frac{L}{2}\Bigg[-\sqrt{\frac{(\ell+m+1)(\ell+m+2)}{(2\ell+1)(2\ell+3)}}\delta_{m-1}^{m'}\delta_{\ell-1}^{\ell'}+\sqrt{\frac{(\ell-m+1)(\ell-m+2)}{(2\ell+1)(2\ell+3)}}\delta_{m+1}^{m'}\delta_{\ell-1}^{\ell'}\\
&\qquad+\sqrt{\frac{(\ell-m)(\ell-m-1)}{(2\ell-1)(2\ell+1)}}\delta_{m-1}^{m'}\delta_{\ell+1}^{\ell'}-\sqrt{\frac{(\ell+m)(\ell+m-1)}{(2\ell-1)(2\ell+1)}}\delta_{m+1}^{m'}\delta_{\ell+1}^{\ell'}\Bigg],\\
X_2(m',\ell') &= \frac{L}{2i}\Bigg[-\sqrt{\frac{(\ell+m+1)(\ell+m+2)}{(2\ell+1)(2\ell+3)}}\delta_{m-1}^{m'}\delta_{\ell-1}^{\ell'}-\sqrt{\frac{(\ell-m+1)(\ell-m+2)}{(2\ell+1)(2\ell+3)}}\delta_{m+1}^{m'}\delta_{\ell-1}^{\ell'}\\
&\qquad+\sqrt{\frac{(\ell-m)(\ell-m-1)}{(2\ell-1)(2\ell+1)}}\delta_{m-1}^{m'}\delta_{\ell+1}^{\ell'}+\sqrt{\frac{(\ell+m)(\ell+m-1)}{(2\ell-1)(2\ell+1)}}\delta_{m+1}^{m'}\delta_{\ell+1}^{\ell'}\Bigg],\\
X_3(m',\ell') &= L\Bigg[\sqrt{\frac{(\ell-m+1)(\ell+m+1)}{(2\ell+1)(2\ell+3)}}\delta_{m}^{m'}\delta_{\ell-1}^{\ell'}+\sqrt{\frac{(\ell-m)(\ell+m)}{(2\ell-1)(2\ell+1)}}\delta_{m}^{m'}\delta_{\ell+1}^{\ell'}\Bigg]
\end{aligned}
$$

Substituting Eq.(24) into Eq.(20) and then comparing the terms related to the ℓ^{th} and $(\ell-1)^{\text{th}}$ ranks, we can straightforward draw a conclusion that the leading order of magnitude of $q_{\ell}^{m}(\mathbf{x},s)$ satisfies

$$
q_{\ell}^{m}(\mathbf{x},s)\propto O\left(\frac{a}{L}\right)^{\ell}
\tag{25}
$$

which indicates, in the limit of $L/a \gg 1$ we concern here, the propagator $q(\mathbf{x},\mathbf{u},s)$ in Eq. (16) exhibits a rapid decay on the orientational dependence for the high-rank expansion. In order to make a direct comparison to the modified diffusion equation based on the Gaussian chain model, in particular, we focus on the leading term with $\ell = 0$ in Eq. (20)

$$\frac{\partial}{\partial s} q_0^0(\mathbf{x}, s) = -w(\mathbf{x}, s) q_0^0(\mathbf{x}, s) - \sum_{\ell'=0}^{\infty} \sum_{m'=-\ell'}^{\ell'} \frac{L}{\sqrt{6}} \Big((\delta_1^{\ell'} \delta_{-1}^{m'} - \delta_1^{\ell'} \delta_1^{m'}),\ -i(\delta_1^{\ell'} \delta_{-1}^{m'} + \delta_1^{\ell'} \delta_1^{m'}),\ \sqrt{2} \delta_1^{\ell'} \delta_0^{m'} \Big) \cdot \left(\mathbf{h}^{-1} \right)^{\mathsf{T}} \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3} \right)^{\mathsf{T}} q_{\ell'}^{m'}(\mathbf{x}, s), \tag{26}$$

According to Eq.(25), the left-hand side has an order

$$\frac{\partial}{\partial s} q_0^0(\mathbf{x}, s) \propto O\left(\frac{a}{L}\right)^0. \tag{27}$$

This implies that the terms on the order $O(a/L)^0 \sim 1$ at the right-hand side of Eq. (26) will be kept, if one expects to recover the Gaussian-chain-based MDE. Correspondingly, it also turns out

$$w(\mathbf{x}, s) \propto O\left(\frac{a}{L}\right)^0. \tag{28}$$

To retain the terms with the same order from the right-hand side of Eq. (26), we need another equation set, taken from Eq. (20) with $\ell = 1$, to complete the derivation. For $m = -1, 0, 1$, we have

$$\begin{aligned} \frac{\partial}{\partial s} q_1^m(\mathbf{x}, s) = -\left(w(\mathbf{x}, s) + \frac{2L}{a} \right) q_1^m(\mathbf{x}, s) - L \sum_{\ell'=0}^{\infty} \sum_{m'=-\ell'}^{\ell'} \Big(Y_1(m', \ell'), Y_2(m', \ell'), Y_3(m', \ell') \Big) \cdot \\ \left(\mathbf{h}^{-1} \right)^{\mathsf{T}} \cdot \left(\frac{\partial}{\partial x_1}, \frac{\partial}{\partial x_2}, \frac{\partial}{\partial x_3} \right)^{\mathsf{T}} q_{\ell'}^{m'}(\mathbf{x}, s) + O\left(\frac{a}{L}\right)^2, \end{aligned} \tag{29}$$

where

$$\begin{aligned} Y_1(m', \ell') &= \frac{1}{2} \left[\sqrt{\frac{-m(1-m)}{3}} \delta_0^{\ell'} \delta_{m+1}^{m'} - \sqrt{\frac{m(1+m)}{3}} \delta_0^{\ell'} \delta_{m-1}^{m'} \right], \\ Y_2(m', \ell') &= \frac{1}{2i} \left[-\sqrt{\frac{-m(1-m)}{3}} \delta_0^{\ell'} \delta_{m+1}^{m'} - \sqrt{\frac{m(1+m)}{3}} \delta_0^{\ell'} \delta_{m-1}^{m'} \right], \\ Y_3(m', \ell') &= \sqrt{\frac{(1-m)(1+m)}{3}} \delta_0^{\ell'} \delta_m^{m'}. \end{aligned}$$

We notice that the term at the left-hand side of Eq. (29) has an order $O(a/L)^1$, beyond which the terms at the right-hand side will be discarded. More explicitly, the equation set reads

$$
\begin{aligned}
\frac{2}{a}q_1^0(\mathbf{x},s) &= -\sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'}\sqrt{\frac{1}{3}}\left(0,\,0,\,\delta_0^{\ell'}\delta_0^{m'}\right)\cdot\left(\mathbf{h}^{-1}\right)^{\intercal}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\intercal}q_{\ell'}^{m'}(\mathbf{x},s)+O\left(\frac{a}{L}\right)^2,\\
\frac{2}{a}q_1^1(\mathbf{x},s) &= \sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'}\sqrt{\frac{1}{6}}\left(\delta_0^{\ell'}\delta_0^{m'},\,-i\delta_0^{\ell'}\delta_0^{m'},\,0\right)\cdot\left(\mathbf{h}^{-1}\right)^{\intercal}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\intercal}q_{\ell'}^{m'}(\mathbf{x},s)+O\left(\frac{a}{L}\right)^2,\\
\frac{2}{a}q_1^{-1}(\mathbf{x},s) &= \sum_{\ell'=0}^{\infty}\sum_{m'=-\ell'}^{\ell'}\sqrt{\frac{1}{6}}\left(-\delta_0^{\ell'}\delta_0^{m'},\,-i\delta_0^{\ell'}\delta_0^{m'},\,0\right)\cdot\left(\mathbf{h}^{-1}\right)^{\intercal}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\intercal}q_{\ell'}^{m'}(\mathbf{x},s)+O\left(\frac{a}{L}\right)^2.
\end{aligned}
\tag{30}
$$

Substituting Eq. (30) into Eq. (26), we eventually obtain a closed equation for $\ell = 0$

$$
\begin{aligned}
\frac{\partial}{\partial s}q_0^0(\mathbf{x},s) =& -w(\mathbf{x},s)q_0^0(\mathbf{x},s)\\
&+\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)\cdot\mathbf{h}^{-1}\cdot\frac{La}{6}\cdot\left[\left(\mathbf{h}^{-1}\right)^{\intercal}\cdot\left(\frac{\partial}{\partial x_1},\frac{\partial}{\partial x_2},\frac{\partial}{\partial x_3}\right)^{\intercal}q_0^0(\mathbf{x},s)\right],
\end{aligned}
\tag{31}
$$

where we use the following relations to attain the $\dfrac{La}{6}$ as the common coefficient of $\mathbf{h}_1, \mathbf{h}_2$ and $\mathbf{h}_3$,

$$
\begin{aligned}
\frac{La}{6} &= -L\left(-\sqrt{\frac{1}{6}}\mathbf{h}_1 - i\sqrt{\frac{1}{6}}\mathbf{h}_2\right)\cdot\left(\sqrt{\frac{1}{6}}\mathbf{h}_1 - i\sqrt{\frac{1}{6}}\mathbf{h}_2\right)\cdot\frac{a}{2}\\
&= -L\left(\sqrt{\frac{1}{6}}\mathbf{h}_1 - i\sqrt{\frac{1}{6}}\mathbf{h}_2\right)\cdot\left(-\sqrt{\frac{1}{6}}\mathbf{h}_1 - i\sqrt{\frac{1}{6}}\mathbf{h}_2\right)\cdot\frac{a}{2}\\
&= -L\sqrt{\frac{1}{3}}\mathbf{h}_3\cdot\left(-\sqrt{\frac{1}{3}}\mathbf{h}_3\right)\cdot\frac{a}{2}
\end{aligned}
\tag{32}
$$

Eq. (31) can further be rewritten in the component-wise version as

$$
\frac{\partial}{\partial s}q_0^0(\mathbf{x},s) = -w(\mathbf{x},s)q_0^0(\mathbf{x},s) + \frac{La}{6}\sum_{i,j=1}^{3}\mathbf{G}_{ij}^{-1}\frac{\partial^2 q_0^0(\mathbf{x},s)}{\partial x_i\partial x_j}.
\tag{33}
$$

where the metric matrix $\mathbf{G} = \mathbf{h}^{\intercal}\mathbf{h}$ is used.

The diffusion-like equation Eq. (33) exactly recovers the Gaussian-chain-based MDE in the nonorthogonal coordinates, which was previously derived by Barrat *et al.*[6] Note, the prefactor

$La/6$ before the second term at the right-hand side of Eq. (33) is closely related to R_g^2, the square radius of gyration. This explicitly manifests that for the wormlike chain model, in the flexible limit $L/a \gg 1$, $\sqrt{La/6} \sim R_g$ inevitably becomes the characteristic length to measure the length scale of flexible polymer chain systems.

IV. The enhancement of the perpendicular alignment to the interface for the more rigid chains

For the lamellar phase under tensile deformation, the systems with a higher chain rigidity tends to enhance the orientational alignment perpendicular to the interface than the flexible chains, and in this section the numerical evidence is presented.

We can calculate the orientational probability distribution of the specific segment s^{int} at interface for lamellae as

$$P(s = s^{\text{int}},\, \cos\theta) = \int \mathrm{d}z\, q(z,\, \theta,\, s = s^{\text{int}}) q^{\dagger}(z,\, \theta,\, s = s^{\text{int}}) \tag{34}$$

which represents the probability of finding the joint segment for a given orientation θ in the whole space. As well known, owing to incompatibility between components A and B, diblock copolymers prefer to be aligned perpendicular to the interface, which directly indicates the probability $P(s = s^{\text{int}},\, \cos\theta = 1)$ is predominant. In order to evaluate the perpendicular orientation affected by the tensile stress, we define a ratio

$$\widetilde{P}(s = s^{\text{int}}, \cos\theta = 1) = \frac{P_{\text{Extension}}(s = s^{\text{int}}, \cos\theta = 1)}{P_{\text{Stress-Free}}(s = s^{\text{int}}, \cos\theta = 1)}. \tag{35}$$

As shown in Fig. S1, for any chain persistency L/a, $\widetilde{P}(s = s^{\text{int}}, \cos\theta = 1) > 1$ is always hold, indicating the tensile stress preferentially forces the orientational alignment of segments perpendicular to the interface. Further, a monotonic increase of $\widetilde{P}(s = s^{\text{int}}, \cos\theta = 1)$ with decreasing L/a reveals that more rigid chains profoundly tend to exhibit a more perpendicular alignment, in contrast to the flexible chains.

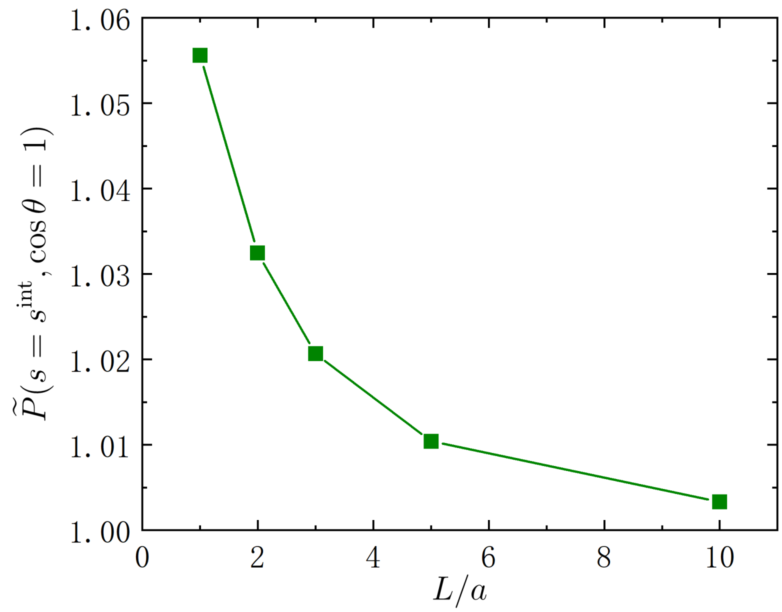

Figure S1: The probability ratio $\widetilde{P}(s = s^{\text{int}}, \cos\theta = 1)$ as the function of chain persistency L/a for a given tensile stress $V\sigma/(nk_BT) = 0.95$.

V. Crossover to the Gaussian-chain limit in shear stress-strain relations of cylindroids

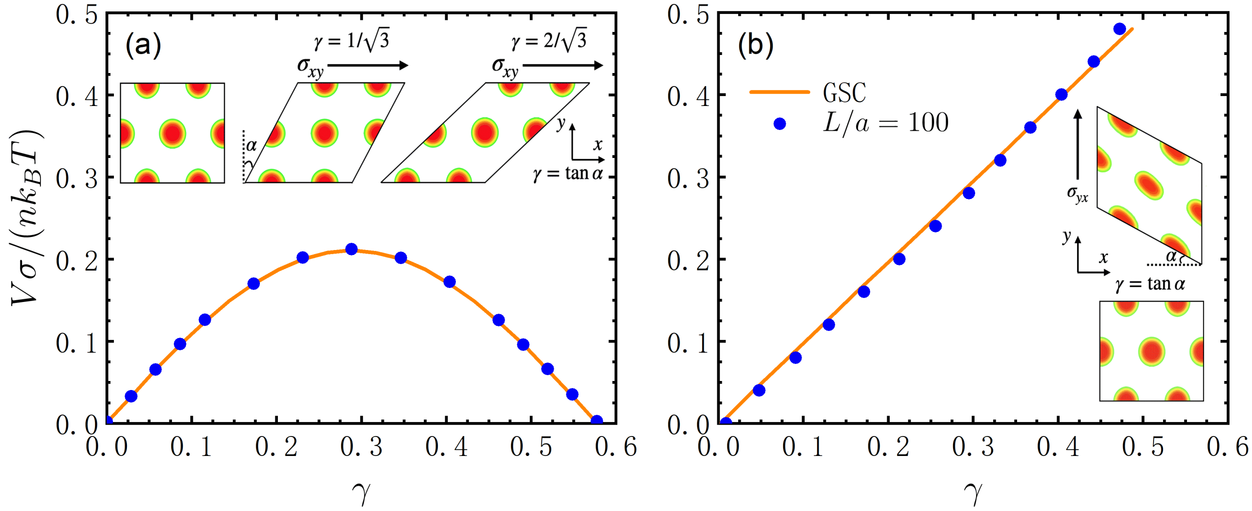

Figure S2: The stress-strain curve of cylinders applied by the shear stress along the directions (a) σ_{xy} and (b) σ_{yx} for the Gaussian chain (GSC) and the flexible wormlike chain (L/a = 100) with $\chi N = 23$, $f = 9/23$.

VI. The orientational angle of the cylindriods with the shear strain

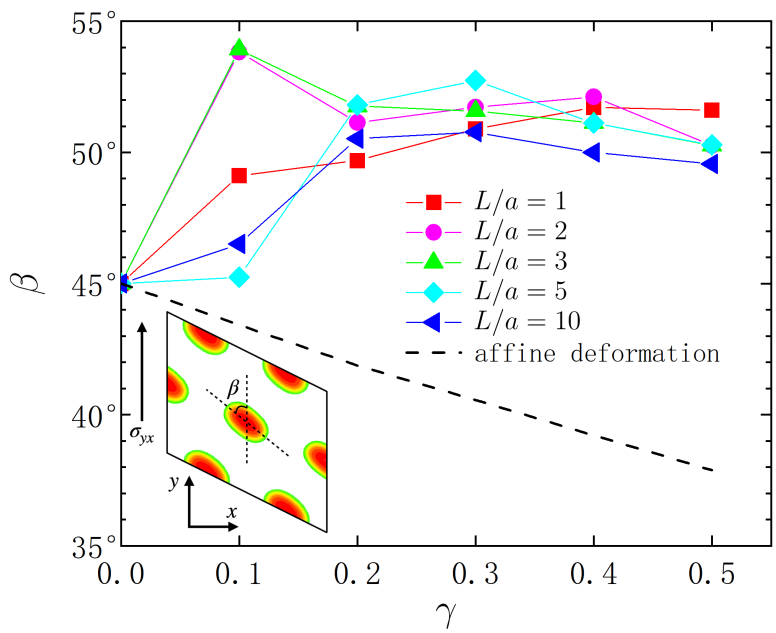

Figure S3: The orientational angle β of the cylindriods as the function of the shear strain γ for various value of chain persistency $L/a = 1, 2, 3, 5, 10$. The parameters used here are the same as ones in Fig. 5 of the main text. β is defined as the angle that the long axis of the cylindriods makes with the y-axis,[8] as shown in the inset. The $\beta \sim \gamma$ relation affected by the purely affine deformation is also shown as a dashed line in figure.

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