# Appendix A
# Polymeric Thin Films

In Appendix A we collect basic information on the physical chemistry of the polymers that are used to make thin films, on the substrates and on the experimental methods.

**Polystryrene** The most commonly used polymer is *polystyrene*, PS, $[C_8H_8]_n$, see Fig. A.1. PS is one of the most widely used plastic materials. An important aspect for the polymer properties is the structural arrangement of the monomers. PS has different *isomers* which vary in their *tacticity*, the placement of the sidegroups. In isotactic polystyrene, all sidegroups lie on the same side, in syndiotactic polystyrene they change position periodically, while in atactic polystyrene, their positioning is random. Most experiments described in this book were done with atactic polystyrene.

Tacticity influences, e.g., the glass temperature $T_g$ of the polymer.

**PMMA/PDMS** Other polymers used in thin-film experiments are PMMA, Poly(methyl-methacrylate) $[C_5O_2H_8]$ and PDMS, polydimethylsiloxane, $[C_2H_6OSi]_n$ (see Fig. A.2).

The molecular weight $M_n$ of a single polymer chain is determined by the polymerisation degree $n$ and the molar mass of a monomer via

$$M_N = N M_{mono}. \tag{A.1}$$

For polystyrol, $M_{mono} = 104$ g/mol. If we pass from a single molecule to a distribution of chains, one can introduce the average molar mass

$$M_n = \sum_N n_N M_N \tag{A.2}$$

where $n_N$ is the number of molecules of polymerisation degree $N$. Further, one defines

$$M_N = \sum_N w_N M_N \tag{A.3}$$

where $w_N = n_N N_N / M_n$ is the mass proportion of molecules of degree $N$. The *polydispersity index* is then given by $M_w/M_n$.

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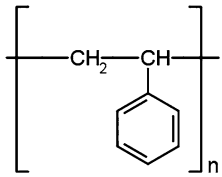


**Fig. A.1** Chemical formula of PS

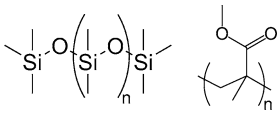


**Fig. A.2** PDMS and PMMA

## A.1 The Substrates and the Surface Coatings

As substrates, most often Si-wafers are used, usually coated with oxide layers of variable thickness. As discussed in the book, the thickness of the oxide layer is a means to influence the effective interface potential.

Of prime importance are the surface coatings on top of these wafers, since they affect the hydrodynamic boundary condition at the wall. Typically, the coatings are made with *self-assembled monolayers* based on *silanes* (Brzoska et al. 1994). These have an endgroup that covalently binds to the silicon oxide. If a sufficient chain length of the silane is available, brush-like surfaces can be generated. For this purpose, typically $n$-alkyltrichlosilanes with $n > 10$ are used. The most typical example is octodecyltrichlorosilane $CH_3(CH_2)_{17}SiCl_3$, OTS, which bears a chain composed of 18 carbon atoms. Dodecyltricholorsilane, DTS, bears 12 carbon atoms in the chain. PDMS can also be used to coat the substrate, in conjunction with an elastomer. The surface density $R$ of a coating is related to the average number of grafted chains per unit surface, $m$, through $m = R/a^2$, with $a$ the size of a monomer, $a = 0.5$ nm for PDMS.

## A.2 Preparation and Measurement of Thin Films

Polymer films are usually produced by *spin coating*. In this procedure, a droplet of the polymer solution (polymer and solvent, the latter of which evaporates) is put on a rotating surface. This procedure works if the solvent wets the substrate; if it does not, the film needs to be spin coated on a wettable substrate and brought onto the unfavorable substrate e.g. by *floating*. In this process, the coated solid film is brought onto a water interface and transferred to the wafer.

Measurement of thin films are done with *ellipsometry* and, most importantly, with *atomic force microscopy*, AFM. The first technique relies on refraction of light at the interfaces, while the second brings a atomic-scale probe into interaction with the substrate.

# Appendix B
# Minkowski Measures

In the analysis of the dewetting images we have made use of the *Minkowski measures* from the field of *integral geometry* (Mecke and Stoyan 2000). Their use is based on *Hadwiger's theorem* which classifies the isotropic measures on compact convex sets in $d$-dimensional Euclidean space. Every measure can be expressed as a linear combination of $d+1$ fundamental measures. In two dimensions, there are three possible measures of this type, one corresponding to area, a second corresponding to perimeter, and a third corresponding to the *Euler characteristic*.

In our application, we have to consider two-dimensional height surface in three dimensions, the film height $h(x,t)$. Of course, one could analyse the patterns arising in a rupturing thin film also by Fourier analysis, but this would only give information on the second-order moments. It is therefore advantageous to define contour lines, just as in a geographic map

$$\mathcal{C}_h = \big\{x \in \mathbb{R}^2 \mid h(x,t) = h\big\}. \tag{B.1}$$

$\mathcal{C}_h$ consists of closed planar lines, or loops, which can be parametrised by a continuous variable $s$. The Minkowski measures, or more precisely in our context, *Minkowski functionals* are defined as integrals of the profile $h(x,t)$. As stated above, the first of them is the area $F$ (note that we drop the time-dependence in the following)

$$F(h) \equiv \int_A d^2x \Theta\big(h(x) - h\big) \tag{B.2}$$

where

$$\frac{\partial F}{\partial h} = -\int_A d^2x \frac{1}{|\nabla h(x)|}\delta\big(|x - \mathcal{C}_h|\big) = -\int_A d^2x \delta\big(h(x) - h\big). \tag{B.3}$$

The second is the boundary length

$$U(h) = \int_A d^2x \delta\big(|x - \mathcal{C}_h|\big) = \int_A d^2x \big|\nabla h(x)\big|\delta\big(h(x) - h\big), \tag{B.4}$$

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and finally the Euler characteristic

$$\chi(h) = \int_A d^2x \kappa(x) \delta\big(|x - C_h|\big) = \int_A d^2x \kappa(x) \big|\nabla h(x)\big| \delta\big(h(x) - h\big) \tag{B.5}$$

where $\kappa(x)$ is the curvature of the contour line at point $x$.

In our application, we have normalized the functionals in a different manner. This is guided by an application to *Gaussian random fields*. For these, one has

$$F(h) = F_0\left(1 - \frac{1}{\sqrt{2\pi\sigma}} \int_{-\infty}^{h} dx e^{-\frac{(s-h_0)^2}{2\sigma^2}}\right), \tag{B.6}$$

$$U(h) = U_0 e^{-\frac{(s-h_0)^2}{2\sigma^2}}, \tag{B.7}$$

$$\chi(h) = \chi_0 h e^{-\frac{(s-h_0)^2}{2\sigma^2}}. \tag{B.8}$$

If one now normalizes according to

$$s(h) = -\frac{\partial F}{\partial h} \frac{1}{U(h)}, \tag{B.9}$$

$$u(h) = \ln U(h), \tag{B.10}$$

and

$$\kappa(h) = \frac{\chi(h)}{U(h)} \tag{B.11}$$

one ends up with the simple algebraic expressions

$$s(h) = s_0 \tag{B.12}$$

$$u(h) = u_0 - h_1(h - h_0)^2 \tag{B.13}$$

and

$$\kappa(h) = \kappa_1(h - h_0)^2. \tag{B.14}$$

# Appendix C
# Numerics of the Thin-Film Equation

In this appendix we will give, for completeness, a schematic description of the algorithm developed to solve the thin-film equation numerically which was used for the results in Part I. We summarize the essentials of the work by Grün and Rumpf (2000) and Becker and Grün (2005), and refer for a full description to these papers.

**The Problem** We consider the discrete solution to the problem

$$h_t - \nabla\big(m(h)\nabla p\big) = 0, \quad \Omega \times (0,T), \tag{C.1}$$

where

$$p = -\Delta h + \Phi'(h). \tag{C.2}$$

The boundary conditions are given by the *no-flux conditions*

$$\partial_\nu h = \partial_\nu p = 0, \quad \text{on } \partial\Omega \times (0,T) \tag{C.3}$$

and the initial data

$$h(\cdot,0) = h_0(\cdot), \quad \text{on } \Omega. \tag{C.4}$$

The mobility is given by the function

$$m(s) = s^n \tag{C.5}$$

whereby, in general, $n \in (0,\infty)$ is allowed. Further, the interface potential $\Phi(h)$ is assumed to be bounded from below and supposed to be a composed as

$$\Phi(h) = \Phi_+(h) + \Phi_-(h) \tag{C.6}$$

where $\Phi_+$ is a convex, $\Phi_-$ a concave function, $C^1$ over $\mathbb{R}$.

**The Triangulation** The domain $\Omega \in \mathbb{R}^N$ for $N = 1, 2$ needs to be triangulated with simplicial elements, which for $N = 2$ are supposed to be rectangular. One then defines by $V^l$ the space of continuous linear functions on each element $E$ of a triangulation $\mathcal{T}^l$. A function $V \in V^l$ is then uniquely defined by its values on the

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nodes of the triangulation $\mathcal{T}^l$. A set of basis functions dual to the set of nodal points is given by $\phi_j \in V^l$ with $\phi_j(x_i) = \delta_{ij}$. Finally one defines a scalar product

$$(\Theta, \Psi)_l \equiv \int_\Omega \mathcal{I}_l(\Theta\Psi) \tag{C.7}$$

with $\mathcal{I}_l : C^0(\Omega) \to V^l$ as an interpolation operator

$$\mathcal{I}_l = \sum_{j=1}^{\dim V^l} u(x_j)\phi_j. \tag{C.8}$$

**Time Discretization** The time interval $I \equiv [0, T]$ is subdivided in intervals $(t_k, t_{k+1}]$ with $t_{k+1} = t_k + \tau_k$ for increments $\tau_k > 0$ and $k = 0, 1, \ldots, K-1$. An implicit backward Euler scheme for Eqs. (C.1), (C.2) is given as follows. Writing the initial condition in triangulated form $H^0 = \mathcal{I}_l h_0 \in V^l$, functions $H^{k+1} \in V^l$ and $P^{k+1} \in V^l$, for all $k$, need to be determined which fulfill the discrete equations

$$\left(H^{k+1} - H^k, \Theta\right)_l + \tau_k\left(M_\sigma\left(H^{k+1}\right)\nabla P^{k+1}, \nabla\Theta\right) = 0, \tag{C.9}$$

$$\left(\nabla H^{k+1}, \nabla\Psi\right) + \left(\Phi'_+\left(H^{k+1}\right), \Psi\right)_l + \left(\Phi'_-\left(H^k\right), \Psi\right)_l = \left(P^{k+1}, \Psi\right)_l \tag{C.10}$$

for all $\Theta, \Psi \in V^l$. The destabilizing term $\Phi'_-$ is discretized explicitly in time, whereas the stabilizing term $\Phi'_+$ is discretized implicitly; this is needed in order to bound the energy at time $T$ by the energy of the initial data. We now close by sketching the construction of the discrete mobility, $M_\sigma$, and of $\nabla H$.

**Constructing the Mobility** The discrete mobility $M_\sigma$ is a continuous mapping

$$M_\sigma : V^l \to \bigotimes_{k=1}^{|\mathcal{T}_l|} \mathbb{R}^{N\times N} \tag{C.11}$$

from a function $H \in V^l$ to an associated field of $(N \times N)$-matrices (symmetric and positive definite), each of which is constant on a triangulation element. Further,

$$\nabla H = M_\sigma(H)\nabla\mathcal{I}_l G'_\sigma(H), \tag{C.12}$$

where

$$G_\sigma(s) \equiv \int_\Lambda^s dr\, g_\sigma(r) \tag{C.13}$$

with

$$g_\sigma(s) = \int_\Lambda^s dr\left[m_\sigma(r)\right]^{-1}. \tag{C.14}$$

$G_\sigma$ is by construction non-negative and convex, and $m_\sigma$ is an approximation to the continuous mobility $m$, e.g., $m_\sigma \equiv m(\max(\sigma, h))$ with $\sigma \ll 1$.

The matrix $M_\sigma$ is calculated, for $N = 2$, by mapping it with the help of an affine mapping $A$ to a reference triangle $\widehat{E}$ with nodes at $(0, \alpha e_1, \alpha e_2)$. The $2 \times 2$-matrix is then given by

$$\widehat{M} = \left(\delta_{ij}\varrho\left(H(0), H(\alpha_i e_i)\right)\right)_{i,j=1,2} \tag{C.15}$$

with

$$\varrho(x,y) \equiv \begin{cases} \left(\frac{1}{y-x}\int_x^y \frac{ds}{m_\sigma(s)}\right)^{-1}, & x \neq y \\ m_\sigma(x), & x = y \end{cases} \tag{C.16}$$

Afterwards, the matrix is transformed back from $\widehat{E}$ to $E$ via $M \equiv A\widehat{M}A^{-1}$. The discretization procedure is then closed by expressing the term

$$M_\sigma\left(U^{k+1}\nabla P^{k+1}, \nabla\Theta\right) \tag{C.17}$$

with respect to the base functions $\phi_i$ in the form

$$L_l^M(H) \equiv \left(\left(M_\sigma(H)\nabla\phi_i, \nabla\phi_j\right)\right)_{i,j=1}^{\dim V^l} \tag{C.18}$$

so that finally the equation to solve can be written as

$$\begin{aligned} H^{k+1} - H^k + \tau_k M_l^{-1} L_l^M\left(H^{k+1}\right)\left[M_l^{-1} L_l H^{k+1} + \mathcal{T}_l \Phi'_+\left(H^{k+1}\right)\right. \\ \left. + \mathcal{T}_l \Phi'_-\left(H^k\right)\right] = 0, \end{aligned} \tag{C.19}$$

with the base scalars

$$M_h \equiv \left((\phi_i, \phi_j)_l\right)_{i,j=1}^{\dim V^l} \tag{C.20}$$

and

$$L_h \equiv \left((\nabla\phi_i, \nabla\phi_j)_l\right)_{i,j=1}^{\dim V^l}. \tag{C.21}$$

The final step then is to set up a *Newton method* to solve the fixed-point problem

$$B\left(H_{i+1}^{k+1}\right) = 0 \tag{C.22}$$

for

$$\begin{aligned} B\left(H_{i+1}^{k+1}\right) \equiv H_{i+1}^{k+1} - H^k + \tau_k M_l^{-1} L_l^M\left(H_i^{k+1}\right)\left[M_l^{-1} L_l H_{i+1}^{k+1}\right. \\ \left. + \mathcal{T}_l \Phi'_+\left(H_{i+1}^{k+1}\right) + \mathcal{T}_l \Phi'_-\left(H^k\right)\right], \end{aligned} \tag{C.23}$$

setting $H_{i+1,0}^{k+1} = H_i^{k+1}$.

# Appendix D
# Towards 'Better' Theories of Viscoelastic Thin Films

In this final appendix we close the book with a look further on to better models in the hydrodynamic description for non-Newtonian fluids. All our discussion was based on a phenomenological approach by employing the *Jeffreys model*. We have seen that a more 'microscopic' knowledge is required for a comparison with experiment. In this appendix we ask whether we can we make our models more realistic for the problems under study, or at least justify them better.

We go into two aspects of this question, both of which are related to better descriptions of the fluid's stress tensor, as it enters into momentum equation, the Navier-Stokes equation. The first tries to go more fundamental in a macroscopic hydrodynamic description in the sense that it tries to exploit basic (symmetry) principles, while the second employs a kinetic approach in which one tries to build continuum models from more microscopic theories.

## D.1 A Nonlinear Fluid Dynamics of Non-Newtonian Fluids

Pleiner et al. (2000, 2004), Temmen et al. (2000) have developed a nonlinear dynamic theory for non-Newtonian fluids based on hydrodynamic principles. If one starts out as usual from the momentum equation, the Navier-Stokes equation in the form

$$\varrho \frac{Dv_i}{Dt} + \nabla_i p + \nabla_j \tau_{ij} = 0 \tag{D.1}$$

where $p$ is the isotropic pressure, one can write the stress tensor $\tau_{ij}$ as

$$\tau_{ij} = -\Psi_{ij} + \Psi_{ki} U_{jk} + \Psi_{kj} U_{ik} + \tau_{ij}^{ph} \tag{D.2}$$

where $\Psi_{ij}$ is the *elastic stress tensor*, the thermodynamic conjugate to the strain tensor. Here,

$$U_{ij} = \frac{1}{2}\left[\nabla_j u_i + \nabla_i u_j - (\nabla_i u_k)(\nabla_j u_k)\right] \tag{D.3}$$

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is the *Eulerian strain tensor*, where $u_i(r)$ is the displacement field. It fulfills the dynamic equation

$$\frac{D}{Dt}U_{ij} - A_{ij} + U_{ki}\nabla_j v_k + U_{kj}\nabla_i v_k = X_{ij}^{ph} \tag{D.4}$$

with

$$A_{ij} = \frac{1}{2}[\nabla_i v_j + \nabla_j v_i]. \tag{D.5}$$

Further, $X_{ij}^{ph}$ is a phenomenological (quasi-)current containing relaxational processes, e.g., diffusions.

Returning to Eq. (D.2), in this construction, the three terms involving $\Psi$, both linear and nonlinear, are counterterms to the flows in Eq. (D.4), needed to avoid entropy productions due to reversibility. In this formulation these terms are thus generic; by contrast, the last term is *phenomenological* and reflects material properties. Thus, material properties require to express explicit models for

$$X_{ij}^{ph} = -\alpha_{ijkl}\Psi_{kl} \tag{D.6}$$

and

$$\tau_{ij}^{ph} = -\nu_{ijkl}A_{kl} \tag{D.7}$$

describing strain relaxation and viscosity, respectively, which also admit further generalizations via cross-couplings. Further, one has the relation between strain and elastic stress

$$\Psi_{ij} = K_{ijkl}U_{kl} \tag{D.8}$$

with the elastic tensor $K_{ij}$.

The tensors $\widehat{\alpha}$ and $\widehat{\nu}$ can also be expanded in the strain tensor $\widehat{U}$, and symmetries exploited for these expressions, a point we do not pursue here any further in all generality. Instead, we briefly show how the expressions can be based on the stress tensor and its derivatives, rather than by strain. This can be done by the corresponding expansions in $\widehat{U}$, but taken only to second order. The dynamic strain equation can then be written as

$$\frac{D}{Dt}U_{ij} - A_{ij} + U_{ki}\nabla v_k + U_{kj}\nabla_i v_k = -\frac{1}{\tau_1}U_{ij} - \frac{1}{\tau_2}U_{ik}U_{jk} \tag{D.9}$$

where $\tau_1^{-1} = \alpha_1 K_1$ and $\tau_2^{-1} = 2\alpha_1 K_2 + 2\alpha_2 K_1$, with elastic moduli $K_1$, $K_2$, and the other four parameters deriving from the expansions of the tensors $\widehat{\alpha}$ and $\widehat{\nu}$. The stress tensor $\tau_{ij}$ in the Navier-Stokes equation is given by, following Eq. (D.2) and the expansions

$$\tau_{ij} = -K_1 U_{ij} + K_2' U_{ik}U_{jk} - \nu_1 A_{ij} - \nu_2(U_{ik}A_{jk} + U_{jk}A_{ik}) \tag{D.10}$$

with $K_2' = 2(K_1 - K_2)$. Taking the derivative $D/Dt$ of the stress tensor yields

$$\frac{D}{Dt}\tau_{ij} = -F\left[U_{ij}, A_{ij}, \frac{D}{Dt}A_{ij}, \Omega_{ij}\right] \tag{D.11}$$

where $\Omega_{ij}$ is the vorticity which we encountered earlier already,

$$\Omega_{ij} = \frac{1}{2}[\nabla_j v_i - \nabla_i v_j]. \tag{D.12}$$

To end up with an equation for $\tau_{ij}$, one needs to invert Eq. (D.10), which can only be done approximately by expanding in $U_{ij}$, leading to terms linear and quadratic in $A_{ij}$. This leads to the expressions

$$\begin{aligned}\tau_1 \frac{D_a}{Dt}\tau_{ij} + \tau_{ij} = &-\nu_\infty A_{ij} - \nu_1 \tau_1 \frac{D_b}{Dt} A_{ij} + \frac{r}{K_1}\tau_{ik}\tau_{jk} \\ &+ \frac{\nu_1 \nu_2}{K_1}\left([\tau_{jk} + \nu_1 A_{jk}]\frac{\partial}{\partial t}A_{ij} + [\tau_{ik} + \nu_1 A_{ik}]\frac{\partial}{\partial t}A_{jk}\right) \\ &+ \mathcal{O}(3),\end{aligned} \tag{D.13}$$

where $\nu_\infty = \nu_1 + \tau_1 K_1$ and $r = \tau_1/\tau_2 - K_2'/K_1$ and the derivative is defined by its action on any tensor $T_{ij}$ to be

$$\frac{D_a}{Dt}T_{ij} \equiv \frac{D}{Dt}T_{ij} - a(T_{ik}A_{jk} + T_{jk}A_{ik}) - (T_{ik}\Omega_{jk} + T_{jk}\Omega_{ik}). \tag{D.14}$$

For $a = -1$ ($a = +1$), $D_a/Dt$ is the lower (upper) convected derivative. If $a = 0$, we have the *Jaumann* or *corotational derivative* which we encountered before. For general $a$ and $b$ one has the material-dependent behaviours

$$a = -1 + \frac{\nu_1}{K_1\tau_2} - \frac{K_2'}{K_1^2}\frac{\nu_\infty}{\tau_1} \tag{D.15}$$

and

$$b = -1 + \frac{\nu_1}{2K_1\tau_2} - \frac{K_2'}{2K_1^2}\frac{\nu_\infty}{2K_1} - \frac{K_2'}{2K_1} - \frac{\nu_2}{\nu_1} \tag{D.16}$$

*A final Task. Identify the Maxwell and Jeffreys models from this more general description.*

## D.2 The Rolie-Poly Equation

In the second approach, one starts out from a microscopic level in order to arrive at macroscopic quantities, like the stress tensor (Byron Bird et al. 1987b). The well-accepted starting point in this context is the *Doi-Edwards tube model*. The basic quantity here is a *Langevin equation* for the motion of a single primitive chain whose motion is confined to a tube. The primitive chain is composed of effective segments, representing sections of a real chain, whereby each tube segment is understood as being the distance between entanglement points, and sufficiently large to have properties of a Gaussian coil.

In this theory, different relaxation mechanism have been built into. The fastest relaxation mode is *chain retraction*, which occurs on the Rouse time, $\tau_R$. It corresponds to a return of the chain to equilibrium under flow deformations of the tube.

*Reptation*, on the other hand, is the slowest mode, a one-dimensional Brownian motion along the tube, with a timescale $\tau_d$. It dominates in linear chains at very small strain rates, $\dot{\gamma} < \tau_d^{-1}$. Contour length fluctuations of the tube are stochastic, breathing-mode retractions of the chain ends. *Constraint release* is a relaxation mode of the tube due to disappearance of entanglement points when the chain relax out of their tube by reptation of contour length fluctuations. A particular such mode is convective constraint release (CCR), which occurs at shear strain rates larger than the reptation rate, $\dot{\gamma} > \tau_d^{-1}$, when the entanglement are removed with the convective flow of the surrounding chains.

In order to model such effects, and in particular to arrive at a stress tensor description, the stochastic equation of the tube reads as (Likhtman and Graham 2003)

$$\mathbf{R}(s,t+\Delta t) = \mathbf{R}(s+\Delta\xi,t) + \Delta t\left(\kappa_v\cdot\mathbf{R} + \frac{3}{2}\frac{v}{|R'|}\mathbf{R}'' + \mathbf{g}(s,t) + \frac{1}{2\pi^2\tau_R}\frac{(\mathbf{R}''\cdot\mathbf{R}')\mathbf{R}'}{\mathbf{R}'^2}\right). \tag{D.17}$$

Here, $\mathbf{R}(s,t)$ is a stochastic vector for the position of the tube segment; $s$ labels the monomers in the tube, measured in entanglement segments from one end of the chain, $s = 0,\ldots,Z$, with $Z = N/N_e$. The first term in Eq. (D.17) describes reptation with a noise with variance

$$\left\langle \Delta\xi(t)\Delta\xi\left(t'\right)\right\rangle = 2D_c\delta\left(t-t'\right). \tag{D.18}$$

The second term in Eq. (D.17) describes deformation by flow with $\kappa_v$ as the velocity gradient tensor, the third and forth describe convective constraint release, and the last retraction along the tube contour due to stretch relaxation.

The term $\mathbf{g}$ is stochastic with variance

$$\left\langle \mathbf{g}(s,t)\mathbf{g}\left(s',t'\right)\right\rangle = \frac{\mathbf{I}}{|R'|}va^2\delta\left(s-s'\right)\delta\left(t-t'\right) \tag{D.19}$$

where $\mathbf{I} = \delta_{ij}$. $v$ is the frequency of constraint release. Further, $\tau_R$ is the *Rouse relaxation time* of one entanglement segment, and $D_c = 1/(3\pi^2 Z\tau_R)$ the reptation diffusion constant.

From this equation, a partial differential equation for the tangent correlation function

$$f_{ij}\left(s,s',t\right) \equiv \left\langle \frac{\partial R_i}{\partial s}\frac{\partial R_j}{\partial s'}\right\rangle \tag{D.20}$$

can be obtained. It allows to derive the polymeric contribution to the stress tensor

$$\tau_{ij}^{poly} = \frac{3G_e}{Z}\int_0^Z ds f_{ij}(s,s). \tag{D.21}$$

The equation can be simplified in order to generate a so-called 'single-mode' equation, which runs under the name *ROuse LInear Entangled POLYmers*—Roly-Poly.

Its form is in an appropriate approximation for the non-stretching case

$$\frac{d}{dt}\tau = \kappa_v^T \cdot \tau + \tau \cdot \kappa_a^T - \frac{1}{\tau_d}(\tau - \mathbf{I}) - \frac{2}{3} tr(\kappa_v^T \cdot \tau)\big(\tau + \beta(\tau - \mathbf{I})\big) \tag{D.22}$$

where $\beta$ is a CCR-coefficient. The Rolie-Poly approach has proved successful in the description of macroscopic flows and there are also indications towards the importance of CCR in thin films, see Roth et al. (2005).

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