

Dynamics of polymeric manifolds in melts: the Hartree approximation

 V.G. Rostiashvili^{1,2,a}, M. Rehkopf¹, and T.A. Vilgis¹
¹ Max-Planck-Institut für Polymerforschung, Postfach 3148, 55021 Mainz, Germany

² Chemical Physics, Russian Academy of Science, 142432, Chernogolovka, Moscow region, Russia

Received: 22 May 1998 / Received in final form: 31 August 1998 / Accepted: 8 September 1998

Abstract. The Martin-Siggia-Rose functional technique and the selfconsistent Hartree approximation is applied to the dynamics of a D -dimensional manifold in a melt of similar manifolds. The generalized Rouse equation is derived and its static and dynamic properties are studied. The static upper critical dimension, $d_{uc} = 2D/(2 - D)$, discriminates between Gaussian (or screened) and non-Gaussian regimes, whereas its dynamical counterpart, $\tilde{d}_{uc} = 2d_{uc}$, discriminates between Rouse- and renormalized-Rouse behavior. The Rouse modes correlation function in a stretched exponential form and the dynamical exponents are calculated explicitly. The special case of linear chains $D = 1$ shows agreement with Monte-Carlo simulations.

PACS. 05.20.-y Statistical mechanics – 83.10.Nn Polymer dynamics – 02.40.Vh Global analysis and analysis on manifolds

Much attention has been paid recently to the theory of the dynamical behavior of polymers (or generally speaking polymeric manifolds) and flux-lines in a quenched disordered random medium [1] or in a melt [2–4]. Technically either the projection formalism and the mode-coupling approximation [2,3] or the selfconsistent Hartree approximation (HA) [1,4] have been used for the derivation of the equations of motion.

In this paper, the HA is used to investigate the static and mainly the dynamic properties of a polymeric D -dimensional manifold (or a fractal) in the melt of the same manifolds. The generalized Rouse equation (GRE), which we derived, reproduce in the static limit the screening and saturation of D -dimensional manifolds [5] in a different way. The whole dynamical consideration results in a subdiffusive behavior and exponents, which are confirmed for the 3-dimensional melt by MC and MD simulations [6–8]. We should stress, that the manifolds in our consideration are crossable, so that entanglements cannot occur and reptation dynamics is not considered. The dynamics considered here corresponds to chains below the critical molecular weight: entanglements are not of importance for the dynamic behavior [9]. We describe the manifolds below only in terms of connectivity and excluded volume. The connectivity defines the D -dimensional subspace which is embedded in the Euclidean space of d dimension. The model we have chosen allows the interpolation between linear polymer chains, which correspond to $D = 1$, and tethered membranes ($D = 2$). By analytic continuation to

rational numbers of the spectral dimension statements on polymeric fractals can be made. In a series of papers [5], we have considered the different regimes in static scaling. In the present letter we place emphasis on the dynamics. We will show below, that a new dynamical regime for the motion of the manifold segments appears.

Let us start with a melt of D -dimensional manifolds in a d -dimensional space. The test manifold is represented by the d -dimensional vector $\mathbf{R}(\mathbf{x}, t)$ with the D -dimensional vector \mathbf{x} of the internal coordinates. In the same way the manifolds of the surrounding matrix are specified by $\mathbf{r}^{(p)}(\mathbf{x}, t)$ ($p = 1, 2, \dots, M$). We have chosen the notation in such a way, that the boldfaced characters describe the external degrees of freedom in Euclidian D -dimensional space, whereas the arrow hatted vectors correspond to the internal D -dimensional space. The model of the melt of M (monodisperse) tethered manifolds used in the following is based on the generalized Edwards Hamiltonian,

$$\mathcal{H} = \frac{Td}{2l^2} \sum_{p=1}^M \int d^D x \left(\nabla_{\mathbf{x}} \mathbf{r}^{(p)}(\mathbf{x}) \right)^2 + \frac{1}{2} \sum_{p,p'=1}^M \int d^D x \int d^D x' V \left(\mathbf{r}^{(p)}(\mathbf{x}) - \mathbf{r}^{(p')}(\mathbf{x}') \right). \quad (1)$$

In this melt an additional (test) manifold is immersed which is described by the variables $\mathbf{R}(\mathbf{x})$. The number of monomers along one side of the manifold is N and limits the \mathbf{x} -integration.

^a e-mail: rostiash@mpip-main.mpg.de

The corresponding Langevin equations in Cartesian components j for the test chain has the form

$$\begin{aligned} \xi_0 \frac{\partial}{\partial t} R_j(\mathbf{x}, t) - \epsilon \Delta_x R_j(\mathbf{x}, t) \\ + \frac{\delta}{\delta R_j(\mathbf{x}, t)} \int d^D x' V [\mathbf{R}(\mathbf{x}, t) - \mathbf{R}(\mathbf{x}', t)] \\ + \frac{\delta}{\delta R_j(\mathbf{x}, t)} \sum_{p=1}^M \int d^D x' V [\mathbf{R}(\mathbf{x}, t) - \mathbf{r}^{(p)}(\mathbf{x}', t)] \\ = f_j(\mathbf{x}, t) \end{aligned} \quad (2)$$

and similarly for all other polymers in the melt

$$\begin{aligned} \xi_0 \frac{\partial}{\partial t} r_j^{(p)}(\mathbf{x}, t) - \epsilon \Delta_x r_j^{(p)}(\mathbf{x}, t) \\ + \frac{\delta}{\delta r_j^{(p)}(\mathbf{x}, t)} \int d^D x' V [\mathbf{r}^{(p)}(\mathbf{x}, t) - \mathbf{R}(\mathbf{x}', t)] \\ + \frac{\delta}{\delta r_j^{(p)}(\mathbf{x}, t)} \sum_{m=1}^M \int d^D x' V [\mathbf{r}^{(p)}(\mathbf{x}, t) - \mathbf{r}^{(m)}(\mathbf{x}', t)] \\ = \tilde{f}_j(\mathbf{x}, t) \end{aligned} \quad (3)$$

where ξ_0 is the bare friction coefficient, $\epsilon = Td/l^2$ the elastic modulus with the Kuhn segment length l , $V(\dots)$ the excluded volume interaction function, Δ_x denotes a D -dimensional Laplacian and the random forces f_j and \tilde{f}_j have the standard Gaussian distribution.

We find it more convenient to reformulate the Langevin problem (2,3) in the MSR-functional integral representation [4]. This representation is especially useful for performing transformations to collective variables or integration over a subset of variables. In our case we introduce the matrix density $\rho(\mathbf{r}, t)$ and the response field density $\pi(\mathbf{r}, t)$

$$\rho(\mathbf{r}, t) = \sum_{p=1}^M \int d^D x \delta(\mathbf{r} - \mathbf{r}^{(p)}(\mathbf{x}, t)) \quad (4)$$

$$\pi(\mathbf{r}, t) = \sum_{p=1}^M \sum_{j=1}^d \int d^D x i\hat{r}_j^{(p)}(\mathbf{x}, t) \nabla_j \delta(\mathbf{r} - \mathbf{r}^{(p)}(\mathbf{x}, t)). \quad (5)$$

In reference [10] the first systematic expansion of the effective action in the MSR-functional integral in terms of ρ and π was given.

The aim now is to integrate over the matrix variables (4,5). To do this, we make the expansion of the effective Action up to the 2nd order with respect to ρ and π , which corresponds to the random phase approximation (RPA). After performing the (Gaussian) functional integration all information about the matrix is comprised into the RPA correlation $S_{00}(\mathbf{k}, t)$ and response $S_{01}(\mathbf{k}, t)$ functions [10].

The resulting Action includes still the test manifold variables in a highly non-linear way. In order to handle it we use the Hartree-type approximation and also take into account the fluctuation-dissipation theorem for both,

the test manifold and the matrix variables. This strategy leads (see for details in [4]) to the following GRE

$$\begin{aligned} \xi_0 \frac{\partial}{\partial t} R_j(\mathbf{x}, t) \\ + \int d^D x' \int_0^t dt' \Gamma(\mathbf{x}, \mathbf{x}'; t-t') \frac{\partial}{\partial t'} R_j(\mathbf{x}', t') \\ - \int d^D x' \Omega(\mathbf{x}, \mathbf{x}') R_j(\mathbf{x}', t) = \mathcal{F}_j(\mathbf{x}, t) \end{aligned} \quad (6)$$

with the memory function

$$\begin{aligned} \Gamma(\mathbf{x}, \mathbf{x}'; t) = \frac{1}{T} \int \frac{d^d k}{(2\pi)^d} k^2 |V(\mathbf{k})|^2 \\ \times F(\mathbf{k}; \mathbf{x}, \mathbf{x}'; t) S_{00}(\mathbf{k}, t) \end{aligned} \quad (7)$$

(where $F(\mathbf{k}; \mathbf{x}, \mathbf{x}'; t)$ is the test manifold density-density correlator), the effective static elastic susceptibility

$$\begin{aligned} \Omega(\mathbf{x}, \mathbf{x}') = \epsilon \delta(\mathbf{x} - \mathbf{x}') \Delta_x - \int \frac{d^d k}{(2\pi)^d} k^2 |\mathcal{V}(\mathbf{k})|^2 \\ \times \left[F_{st}(\mathbf{k}; \mathbf{x}, \mathbf{x}') - \delta(\mathbf{x} - \mathbf{x}') \int d^D x'' F_{st}(\mathbf{k}; \mathbf{x}, \mathbf{x}'') \right] \end{aligned} \quad (8)$$

and the random force has the correlator

$$\begin{aligned} \langle \mathcal{F}_i(\mathbf{x}, t) \mathcal{F}_j(\mathbf{x}', t') \rangle = 2T \delta_{ij} \left[\xi_0 \delta(\mathbf{x} - \mathbf{x}') \delta(t - t') \right. \\ \left. + \theta(t - t') \Gamma(\mathbf{x}, \mathbf{x}'; t - t') \right]. \end{aligned} \quad (9)$$

In equation (8) the effective potential

$$\mathcal{V}(\mathbf{k}) = V(\mathbf{k}) [1 - V(\mathbf{k}) S_{st}(\mathbf{k})/T] \quad (10)$$

gains the standard screened form [9]

$$\mathcal{V}(\mathbf{k}) = V(\mathbf{k}) \left[1 + V(\mathbf{k}) F_{st}^{(0)}(\mathbf{k})/T \right]^{-1} \quad (11)$$

(where $F_{st}^{(0)}(\mathbf{k})$ is the free system correlator) if the standard RPA-result is used for the static correlator $S_{st}(\mathbf{k})$.

It is an important point that we treat on an equal footing both, the static and dynamic parts of the GRE (6). Let us start from the static behavior of equations (6–11). The static limit of these equations for the Rouse mode correlator, $C(\mathbf{p}) = \langle \mathbf{R}(\mathbf{p}) \cdot \mathbf{R}(-\mathbf{p}) \rangle$, yields the Dyson-like form

$$C(\mathbf{p}) = \frac{d}{\mathcal{N} \left[\frac{d}{l^2} \left(\frac{2\pi\mathbf{p}}{N} \right)^2 + \Sigma(\mathbf{p}) \right]} \quad (12)$$

where $\mathcal{N} = N^D$ is the total number of monomers in the manifold and the “self-energy” is given by

$$\begin{aligned} \Sigma(\mathbf{p}) = \int \frac{d^d k}{(2\pi)^d} k^2 \frac{V(\mathbf{k})/T}{1 + V(\mathbf{k}) F_{st}^{(0)}(\mathbf{k})/T} \\ \times \mathcal{N} [F_{st}(\mathbf{k}; \mathbf{p}) - F_{st}(\mathbf{k}; \mathbf{p} = \mathbf{0})]. \end{aligned} \quad (13)$$

The test manifold static correlator in equation (12) is parameterized by the wandering exponent ζ in such a way

$$F_{st}(\mathbf{k}; \mathbf{p}) = \frac{1}{\mathcal{N}} \int d^D x \exp \left(-\frac{k^2 l^2}{2d} x^{2\zeta} - i \frac{2\pi}{N} \mathbf{x} \cdot \mathbf{p} \right). \quad (14)$$

In its turn the exponent ζ is determined by the correlator $C(\mathbf{p})$:

$$Q_{st}(\mathbf{x}) = \int d^D p \left[1 - e^{-i \frac{2\pi}{N} \mathbf{p} \cdot \mathbf{x}} \right] C(\mathbf{p}) \propto x^{2\zeta}. \quad (15)$$

The system of equations (12–15) should be analyzed self-consistently. Straightforward calculations yield

$$\Sigma(\mathbf{p}) = -c_1 \left(\frac{\pi|\mathbf{p}|}{N} \right)^{\zeta(d+2)} - c_2 \left(\frac{\pi|\mathbf{p}|}{N} \right)^{\zeta(d+d_f^0+2)-D} \quad (16)$$

with the Gaussian fractal dimension $d_f^0 = 2D/(2-D)$ [5,12] and c_1, c_2 are some constants depending on d and D . Physically, the condition for the exponent ζ comes from the balance between the entropic and the interaction terms in the denominator of equation (12). The analysis shows that the only way to satisfy the equations (12–15) is to impose the exponents in equation (16) the condition: $\zeta(d+2) = \zeta(d+d_f^0+2) - D \geq 2$. This holds at $d \geq d_{uc} = 2D/(2-D)$ and the only solution is the Gaussian one with $\zeta = \zeta_0 = (2-D)/2$. Besides that, the necessary condition $d_f^0 < d$ immediately implies $D < D_s = 2d/(2+d)$. The *upper critical dimension* d_{uc} in a melt and the *spectral critical dimension* D_s was discussed first in [5]. At $d < d_{uc}$ the interaction term in equation (12) overwhelms, $|\Sigma(\mathbf{p})| \gg (2\pi\mathbf{p}/N)^2$, and the system become unstable. The manifold is saturated in a melt, *i.e.* it loses its fractal nature and becomes compact [5].

We now consider the dynamics at $d \geq d_{uc}$. There are two dynamic exponents, z and w . The exponent z measures the time dependence of a monomer displacement, *i.e.*

$$Q(t) = \int d^D p \int_{a-i\infty}^{a+i\infty} \frac{ds}{2\pi i} [1 - e^{st}] C(\mathbf{p}, s) \propto t^{2z} \quad (17)$$

and the exponent w measures the same for the center of mass

$$Q_{cm}(t) = \lim_{\mathbf{p} \rightarrow 0} \int_{a-i\infty}^{a+i\infty} \frac{ds}{2\pi i} [1 - e^{st}] C(\mathbf{p}, s) \propto t^w. \quad (18)$$

In equations (17, 18) $C(\mathbf{p}, s) = \langle |\mathbf{R}(\mathbf{p}, s)|^2 \rangle$ is the Rouse-Laplace component of the correlator $C(\mathbf{x}, t)$. The formal solution of equations (6–8) for $C(\mathbf{p}, s)$ is given by

$$C(\mathbf{p}, s) = \frac{C_{st}(\mathbf{p})}{s + \frac{\epsilon \left(\frac{2\pi\mathbf{p}}{N} \right)^2}{\xi_0 + \mathcal{N} \Gamma(\mathbf{p}, s)}} \quad (19)$$

where $C_{st}(\mathbf{p}) = l^2(N/2\pi\mathbf{p})^2/\mathcal{N}$ and $\Gamma(\mathbf{p}, s)$ is the Rouse-Laplace transformation of the memory function (7).

In the RPA the matrix density correlator $S_{00}(\mathbf{k}, t)$ is well approximated by

$$S_{00}(\mathbf{k}, t) = S_{st}(\mathbf{k}) \times \begin{cases} \exp \left\{ -k^2 D_{coop}(\mathbf{k}) t \right\}, (kl)^{d_f^0} \mathcal{N} \ll 1 \\ \exp \left\{ -\frac{k^2 l^2}{2d} \left(\frac{t}{\tau_0} \right)^{2z_0} \right\}, (kl)^{d_f^0} \mathcal{N} \gg 1 \end{cases} \quad (20)$$

where the cooperative diffusion coefficient $D_{coop}(\mathbf{k}) = \rho V(\mathbf{k})/\xi_0$, $\tau_0 = \xi_0 l^2/Td$ and $z_0 = (2-D)/4$ is the Gaussian z -exponent. The corresponding Ansatz for the test manifold yields

$$F(\mathbf{k}; \mathbf{x}; t) = F_{st}(\mathbf{k}, \mathbf{x}) \times \begin{cases} \exp \left\{ -k^2 D_G t \right\}, (kl)^{d_f^0} \mathcal{N} \ll 1 \\ \exp \left\{ -\frac{k^2 l^2}{2d} \left(\frac{t}{\tau_0} \right)^{2z} \right\}, (kl)^{d_f^0} \mathcal{N} \gg 1 \end{cases} \quad (21)$$

where D_G is the self-diffusion coefficient.

(i) By making use of equations (20, 21) and equation (7) in the limit $(kl)^{d_f^0} \mathcal{N} \gg 1$ one can derive the result

$$\mathcal{N} \Gamma(\mathbf{p}, s) = const. \left(\frac{1}{\tau_0 s} \right)^{1-\beta} \quad (22)$$

with

$$\beta = z_0(d - d_{uc} + 2). \quad (23)$$

In the derivation of equation (22) we also used the assumption $l^2 \ll Q(t) \ll l^2 \mathcal{N}^{2/d_f^0}$. The condition $\beta < 1$ immediately defines the *dynamical upper critical dimension*

$$\tilde{d}_{uc} = \frac{4D}{2-D} = 2d_{uc} \quad (24)$$

i.e. the dimension above which the manifold has the simple Rouse behavior, at $d = \tilde{d}_{uc}$ one can call it the marginal Rouse behavior and only at $d < \tilde{d}_{uc}$ the dynamic exponents z and w are renormalized.

At $d < \tilde{d}_{uc}$ equation (19), after inverse Laplace transformation, yields

$$C(\mathbf{p}, t) = C_{st}(\mathbf{p}) \times \sum_{m=0}^{\infty} \left[-\epsilon A \left(\frac{2\pi\mathbf{p}}{N} \right)^2 \left(\frac{t}{\tau_0} \right)^\beta \right]^m / \Gamma(m\beta + 1) \quad (25)$$

where $A = [\beta |V(k=0)|^2 S_{st}(k=0)/l^{d+2}]^{-1}$ and $\Gamma(x)$ is the gamma-function. The equation (25) is very close to the stretched exponential form found by the MC- and MD-simulations [6]. The equation (25) was actually calculated in the limit $p \rightarrow 0$, and we can use it

first of all to comparison with simulation results on the center of mass mean square displacement. By using equation (25) in equation (18) we obtain

$$Q_{cm}(t) = \frac{\mathcal{D}_0}{\mathcal{N}} \left(\frac{t}{\tau_0} \right)^w \quad (26)$$

where $\mathcal{D}_0 = l^2 \epsilon A / \Gamma(\beta + 1)$ and

$$w = \beta = z_0(d - d_{uc} + 2). \quad (27)$$

By the same way from equation (17) we can derive:

$$z = z_0 \beta = z_0^2(d - d_{uc} + 2). \quad (28)$$

In reference [3] this renormalized dynamics was formally used as a projected dynamics for the polymer mode coupling approximation (PMCA) and eventually leads to the GRE which, as the authors claim, can describe the entangled dynamics. We will argue in an extended paper [11] that this is a result of misinterpretation of the GRE.

(ii) If we assume that the main contribution to the integral (7) comes from the small wave vectors, $(kl)^{d_f^0} \mathcal{N} \ll 1$, then we arrive at

$$\mathcal{N}\Gamma(\mathbf{p}, s) \propto s^{(d-d_{uc})/2}. \quad (29)$$

Since $d > d_{uc}$, the simple Rouse behavior in the small wave vector regime does not change.

At the large displacement regime, $R_G^2 \ll Q_{cm}(t)$, one should expect a simple diffusive behavior:

$$Q_{cm}(t) = dD_G t \quad (30)$$

with

$$D_G = T/\mathcal{N}[\xi_0 + \mathcal{N}\Gamma(p=0; s=0)]. \quad (31)$$

Now the problem is how D_G depends from \mathcal{N} ? One can assume that for this case only the small wave vectors, $(kl)^{d_f^0} \mathcal{N} \ll 1$, are relevant, *i.e.* the dynamics of the matrix is driven by the cooperative diffusion coefficient D_{coop} and the dynamics of the test manifold by the self-diffusion one D_G (see Eqs. (20, 21)). Since in any way $D_{coop} \gg D_G$, the calculation yields

$$\mathcal{N}\Gamma(p=0; s=0) \propto [D_{coop}]^{-1} \mathcal{N}^{(1-\frac{d}{d_f^0})}. \quad (32)$$

But $D_{coop} = \mathcal{O}(N^0)$ and $d_f^0 < d$, then $\Gamma(p=0; s=0) \rightarrow 0$ at $\mathcal{N} \rightarrow \infty$. As a result $D_G = T/\mathcal{N}\xi_0$, *i.e.* the simple Rouse result does not change. In Figure 1 we have summarized the overall schematic behavior for $Q_{cm}(t)$. At the relatively short times, $\tau_0 < t \ll \tau_R$, and displacements, $l^2 < Q_{cm}(t) \ll R_G^2$, the test chain dynamics is mainly determined by the fluctuations from the interval $(kl)^{d_f^0} \mathcal{N} \gg 1$. As a result the renormalized Rouseian behavior dominates and *e.g.* for the melt of polymer chains ($D = 1$) in the 3-dimensionnal space the exponent $w = 3/4 = 0.75$. In the opposite limit, $\tau_R \ll t$ and $R_G^2 \ll Q_{cm}(t)$, the long wavelength fluctuations $(kl)^{d_f^0} \mathcal{N} \ll 1$ are relevant and the

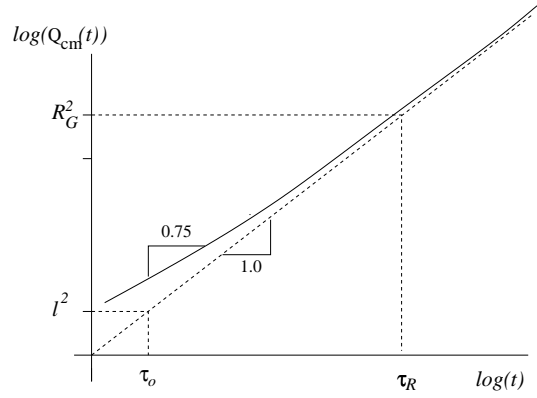


Fig. 1. A schematic plot of $Q_{cm}(t)$ for the simple Rouse (dashed line) and the renormalized Rouse (solid line) dynamics.

melt almost does not influence the test chain: the simple Rouse regime is recovered.

MC-simulations of the bond-fluctuation model [6] as well as the MD-simulations [7] of the athermal melt have been undertaken. Recently also the static and dynamic properties of a realistic polyethylene melt have been studied [8]. Both in MC and MD simulations a slowed down motion at intermediate times for the center of mass is clearly observable. It was found *e.g.* that for the chain length $N = 200$ at the relatively short time $Q_{cm}(t) \propto t^w$ with $w = 0.8$ (instead of $w = 1$) in [6] and $w = 0.71$ in [7]. This deviation from the simple Rouse regime also occurs for short chains ($N < N_e$) which clearly are not entangled [8].

The best test of the renormalized Rouse dynamics predictions would be the simulation of rather long crossable (to avoid reptation) chains but still with an excluded volume interaction. In a recent MC-simulation [13] the statics and dynamics of such melts have been studied. Unfortunately in [13] the plot $Q_{cm}(t)$ is not given explicitly, *i.e.* it stays unclear from this simulation how the mode $p \rightarrow 0$ is renormalized.

In summary, we have shown that by using MSR-functional technique and the Hartree approximation the GRE for a D -dimensional manifold in the melt of similar manifolds can be derived. In this equation the static and dynamic parts are treated on an equal footing. Besides the static upper critical dimension, $d_{uc} = 2D/(2-D)$, its dynamical counterpart, $\tilde{d}_{uc} = 2d_{uc}$ was found (such that at $d_{uc} < d < \tilde{d}_{uc}$ the manifold is Gaussian but renormalized-Rouseian). We have calculated the dynamical exponents, w and z , and have explained some novel computer simulation findings.

We have benefited from discussions with J. Baschnagel, K. Binder, K. Kremer, W. Paul and T.B. Liverpool. We are grateful to the Sonderforschungsbereich 262 and the Bundesminister für Bildung und Forschung for financial support.

References

1. H. Kinzelbach, H. Horner, *J. Phys. I France* **3**, 1329 (1993); D. Cule, Y. Shapir, *Phys. Rev. E* **53**, 1553 (1996).
2. W. Hess, *Macromol.* **21**, 2620 (1988).
3. K.S. Schweizer, *J. Chem. Phys.* **91**, 5802, 5822 (1989); M. Fuchs, K.S. Schweizer, *J. Chem. Phys.* **106**, 347 (1997).
4. M. Rehkopf, V.G. Rostiashvili, T.A. Vilgis, *J. Phys. II France* **7**, 1469 (1997).
5. T.A. Vilgis, *Phys. Rev. A* **36**, 1506 (1987); *Physica A* **153**, 341 (1988); *J. Phys. II France* **2**, 1961 (1992).
6. W. Paul, K. Binder, D.W. Heermann, K. Kremer, *J. Chem. Phys.* **95**, 7726 (1991).
7. S.W. Smith, C.K. Hall, B.D. Freeman, *J. Chem. Phys.* **104**, 5616 (1996).
8. W. Paul *et al.*, *Phys. Rev. Lett.* **80**, 2346 (1998); W. Paul, G.D. Smith, D.Y. Yoon, *Macromol.* **30**, 7772 (1997).
9. M. Doi, S.F. Edwards, *The Theory of Polymer Dynamics* (Clarendon Press, Oxford, 1986).
10. V.G. Rostiashvili, M. Rehkopf, T.A. Vilgis, *cond-mat./9710156*.
11. V.G. Rostiashvili, M. Rehkopf, T.A. Vilgis, *J. Chem. Phys.* to be published.
12. M.E. Cates, *J. Phys. France* **46**, 1059 (1985).
13. J.S. Shaffer, *J. Chem. Phys.* **101**, 4205 (1994); J.S. Shaffer, *J. Chem. Phys.* **103**, 761 (1995).