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1987 J. Phys. A: Math. Gen. 20 5071

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Transport properties for simple ring polymers using the renormalisation group

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Received 5 January 1987, in final form 31 March 1987

Abstract. Transport properties for simple ring polymers are derived using renormalisation group techniques. The translational diffusion constant and the relaxational spectrum in the presence of hydrodynamic interactions are calculated to $O(\epsilon)$ ($\epsilon = 4 - d$, d being the spatial dimensionality). Explicitly time-dependent correlations are presented in universal functional form to $O(\epsilon)$.

Universal properties of long flexible polymer chains have been systematically studied with the use of renormalisation group (RG) techniques. Whereas static properties of both dilute and semidilute solutions have been explored in great detail [1–4], transport properties like the diffusion constant, relaxational spectrum and intrinsic viscosity have not been investigated to the same extent, even for dilute polymer solutions. The minimal model (defined below) which has been studied within the RG framework gives, to lowest order in $\epsilon = 4 - d$ (d being the spatial dimensionality), many universal predictions which can be compared with experiment. These predictions have mainly been derived for flexible linear chains. There is, however, another class of experimentally interesting systems to which RG techniques can be applied: flexible, simple (single) ring polymers. Although ring structures are less frequent in macromolecular substances than are linear or branched structures, molecules of some DNA exist in the form of a single or multiple ring. It is therefore natural to ask how ring formation will affect universal (critical) properties of flexible (Gaussian) polymer chains. An attempt to answer this question has been made in [5], where the conformation space RG technique has been used to derive the distribution function for an internal vector of a ring polymer in the presence of self-avoiding interactions. However, the transport properties of ring polymers have only been studied using techniques different from RG. The friction coefficient has been studied in [6] with the aid of the Kirkwood equation [7] and the intrinsic viscosity in [8] in the presence of self-avoiding and hydrodynamic interactions. Generally speaking, it has been found that the friction or sedimentation coefficient is less sensitive to ring formation than is the intrinsic viscosity.

In this paper we are interested in comparing transport properties of linear and ring polymers and we will consider in detail how ring polymers are affected by the

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long-range hydrodynamic interactions. The simplest time-dependent correlation function from which the translational diffusion constant D_R and the relaxation spectrum can be extracted is the correlation $\langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle$ [9]. Here $\{\mathbf{c}(\tau, t)\}_{\tau=0}^{N_0}$ describes the conformation of a polymer with bare chain length N_0 parametrised by a contour variable τ at time t . According to the Green-Kubo formula we have

$$D = \lim_{t \rightarrow \infty} \frac{1}{2t} \langle [\mathbf{c}(\tau, t) - \mathbf{c}(0, 0)]^2 \rangle. \quad (1)$$

For simplicity we assume $\mathbf{c}(0, 0) = 0$. The minimal model referred to above, within which our calculation is performed, is defined by the following set of Langevin equations describing coupled chain-solvent dynamics [3, 10]:

$$\frac{\partial \mathbf{c}}{\partial t} = -\frac{1}{\zeta_0} \frac{\delta H_E}{\delta \mathbf{c}(\tau, t)} + \mathbf{g}_0 \mathbf{u}(\mathbf{c}(\tau, t), t) + \Theta(\tau, t) \quad (2a)$$

$$\frac{\partial \mathbf{u}}{\partial t} = \eta_0 \Delta \mathbf{u}(\mathbf{x}, t) - \mathbf{g}_0 \int_0^{N_0} d\tau \frac{\delta H_E}{\delta \mathbf{c}(\tau, t)} \delta(\mathbf{x} - \mathbf{c}(\tau, t)) - \nabla p + \mathbf{f}(\mathbf{x}, t) \quad (2b)$$

together with the incompressibility condition $\nabla \cdot \mathbf{u} = 0$. In (2a) and (2b) $\mathbf{u}(\mathbf{x}, t)$ describes the solvent velocity field, η_0 is the bare solvent viscosity, Δ is the Laplacian, p denotes the pressure, $\zeta_0 \equiv \lambda_0^{-1}$ is the bare translational friction constant and \mathbf{g}_0 is the strength of the hydrodynamic interaction. H_E is the Edwards Hamiltonian [11]

$$H_E = \frac{1}{2} \int_0^{N_0} d\tau \left(\frac{\partial \mathbf{c}}{\partial \tau} \right)^2 + \frac{1}{2} v_0 \int_0^{N_0} d\tau_1 \int_0^{N_0} d\tau_2 \delta(\mathbf{c}(\tau_1) - \mathbf{c}(\tau_2)) \quad (3)$$

with v_0 being the bare excluded volume parameter and Θ, \mathbf{f} Gaussian random processes with zero mean and covariance given by

$$\langle \Theta(\tau, t) \Theta(\sigma, s) \rangle = 2\zeta_0^{-1} \delta(\tau - \sigma) \delta(t - s) I \quad (4)$$

$$\langle \mathbf{f}(\mathbf{x}, t) \mathbf{f}(\mathbf{x}, s) \rangle = -2\Delta \delta(\mathbf{x} - \mathbf{x}) \delta(t - s) I \quad (5)$$

where I is the $d \times d$ unit matrix. We have recently derived [12] an effective Lagrangian describing the system of Langevin equations (2a) and (2b). Introducing the two (imaginary) fields, $\tilde{\mathbf{c}}(\tau, t)$, a conformation response field, and $\tilde{\mathbf{u}}(\mathbf{x}, t)$ a solvent velocity response field, this formalism is suitable for the use of field-theoretic RG techniques. In particular, as the solvent velocity field $\mathbf{u}(\mathbf{x}, t)$ appears explicitly in any order of the perturbation expansion, this method allows for calculations of dynamical quantities to orders higher than $O(\varepsilon)$ where the Markov approximation cannot be used [12]. Utilising this formalism, we obtain

$$\langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle = \int d\{\tilde{\mathbf{c}}, \mathbf{c}\} d\{\tilde{\mathbf{u}}, \mathbf{u}\} \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \exp J \quad (6)$$

where the effective Lagrangian $J = J_0 + J_1$ can be decomposed into a free part J_0 and an interaction part J_1 with

$$J_0^{(1)} = \int_0^{N_0} d\tau \int dt (\tilde{\mathbf{c}}(\tau, t) \lambda_0 \tilde{\mathbf{c}}(\tau, t) - \tilde{\mathbf{c}}(\tau, t) \partial_t \mathbf{c}(\tau, t) + \tilde{\mathbf{c}}(\tau, t) \lambda_0 \partial_\tau^2 \mathbf{c}(\tau, t)) \quad (7)$$

describing the conformation field and

$$J_0^{(2)} = \int d^d \mathbf{x} \int dt (\tilde{\mathbf{u}}(\mathbf{x}, t) \eta_0 (i\nabla)^2 \tilde{\mathbf{u}}(\mathbf{x}, t) - \tilde{\mathbf{u}}(\mathbf{x}, t) \partial_t \mathbf{u}_\perp(\mathbf{x}, t) - \tilde{\mathbf{u}}(\mathbf{x}, t) \eta_0 (i\nabla)^2 \mathbf{u}_\perp(\mathbf{x}, t)) \quad (8)$$

describing the solvent velocity field. In (8) \perp denotes the transverse part. The hydrodynamic interaction is written

$$J_1^{(2)} = -g_0 \int_0^{N_0} d\tau \int dt \tilde{\mathbf{c}}(\tau, t) \mathbf{u}_\perp(\mathbf{c}(\tau, t), t) - g_0 \int_0^{N_0} d\tau \int dt \tilde{\mathbf{u}}_\perp(\mathbf{c}(\tau, t), t) \frac{\delta H_E}{\delta \mathbf{c}(\tau, t)}. \quad (9)$$

Introducing normal coordinates into (7) which, in order to account for the periodic boundary conditions of a ring, are chosen in the form

$$\mathbf{c}(\tau, t) = \sum_{k=0}^{\infty} Q_{\tau k}^{(1)} \boldsymbol{\xi}_k^{(1)} + \sum_{k=0}^{\infty} Q_{\tau k}^{(2)} \boldsymbol{\xi}_k^{(2)} \quad (10a)$$

$$\tilde{\mathbf{c}}(\tau, t) = \sum_{k=0}^{\infty} Q_{\tau k}^{(1)} \tilde{\boldsymbol{\xi}}_k^{(1)} + \sum_{k=0}^{\infty} Q_{\tau k}^{(2)} \tilde{\boldsymbol{\xi}}_k^{(2)} \quad (10b)$$

with

$$Q_{\tau k}^{(1)} = \left(\frac{2}{N_0}\right)^{1/2} \cos\left(\frac{2\pi k\tau}{N_0}\right) \quad k = 1, 2, \dots \quad (11a)$$

$$Q_{\tau k}^{(2)} = \left(\frac{2}{N_0}\right)^{1/2} \sin\left(\frac{2\pi k\tau}{N_0}\right) \quad k = 1, 2, \dots \quad (11b)$$

$$Q_{\tau k}^{(1)} = Q_{\tau k}^{(2)} = \left(\frac{1}{N_0}\right)^{1/2} \quad k = 0 \quad (11c)$$

we obtain for the free response propagator of the conformation field of a simple polymer ring

$$\langle \tilde{\mathbf{c}}(\tau, t) \mathbf{c}(\sigma, t') \rangle_0 = \Theta(t' - t) G_0(\tau, \sigma | t' - t) \quad (12)$$

where

$$G_0(\tau, \sigma | t) = \frac{1}{N_0} \left(1 + 2 \sum_{k=1}^{\infty} \cos 2\hat{k}_0(\tau - \sigma) \exp(-\lambda_{i(k)} t) \right) \quad (13)$$

is the Green function matrix, $\lambda_{i(k)} = \lambda_0(2\pi k/N_0)^2$, $\hat{k}_0 = \pi k/N_0$ and Θ is the step function. From (12) we determine the static Green function $G_0^*(\tau, \sigma) = \langle \mathbf{c}^*(\tau) \mathbf{c}^*(\sigma) \rangle_0$ for a polymer ring in the centre of mass system using

$$G_0^*(\tau, \sigma) = \lambda_0 \int_0^x dt G_0^*(\tau, \sigma | t) \quad (14)$$

where we have excluded the $k=0$ mode in $G_0(\tau, \sigma | t)$. Performing the sum over k we find

$$\langle \mathbf{c}^*(\tau) \cdot \mathbf{c}^*(\tau') \rangle_0 = \frac{dN_0}{12} - \frac{d}{2} |\tau - \tau'| + \frac{d}{2N_0} (\tau - \tau')^2 \quad (15)$$

and

$$\langle \mathbf{c}(\tau) \cdot \mathbf{c}(\tau') \rangle_0 = d \min(\tau, \tau') - (d/N_0) \tau \tau'. \quad (16)$$

According to (16) the free correlations of a Gaussian ring polymer are smaller than those of a linear chain because of the second term in (16). Introducing Fourier transforms for the solvent velocity and the solvent velocity response fields in equation (8), we determine the free solvent velocity response function

$$\langle \tilde{\mathbf{u}}(\mathbf{k}, t) \mathbf{u}_\perp(\mathbf{k}', t') \rangle_0 = \Theta(t' - t) \delta(\mathbf{k} + \mathbf{k}') P \exp[-\eta_0 k^2 (t' - t)] \quad (17)$$

with $P = (I - (\mathbf{k}\mathbf{k})/k^2)$. Using (12) and (17) we obtain the correlation function (6), to order g_0^2 ,

$$\begin{aligned} \langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle &= \langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle_0 + 2g_0^2 \eta_0^{-1} \int_0^{N_0} d\alpha \int_0^{N_0} d\alpha' \int_0^t ds G_0(\tau, \alpha | t-s) \\ &\quad \times G_0(\tau, \alpha' | t-s) \langle \text{Tr } T(\mathbf{c}(\alpha, s) - \mathbf{c}(\alpha', s)) \rangle_0 \\ &\quad + 2g_0^2 \eta_0^{-1} \int_0^{N_0} d\alpha \int_0^{N_0} d\alpha' \int_0^t ds \\ &\quad \times G_0(\tau, \alpha | t-s) \left\langle \mathbf{c}(\tau, t) T(\mathbf{c}(\alpha, s) - \mathbf{c}(\alpha', s)) \frac{\partial^2}{\partial \alpha'^2} \mathbf{c}(\alpha', s) \right\rangle_0 \end{aligned} \tag{18}$$

where Tr denotes the trace. The remaining averages in (18), denoted by $\langle \rangle_0$, are performed with respect to the functional J_0 . In (18) $T(\mathbf{c}(\alpha, s) - \mathbf{c}(\alpha', s))$ is the Oseen tensor

$$T(\mathbf{c}(\alpha, s) - \mathbf{c}(\alpha', s)) = \int_k \frac{1}{k^2} P \exp[i\mathbf{k} \cdot (\mathbf{c}(\alpha, s) - \mathbf{c}(\alpha', s))] \tag{19}$$

which occurs because we have invoked the Markov approximation, an approximation correct only to lowest order ($O(\epsilon)$). The free correlation function $\langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle_0 / dN_0 = Q(\tau, t | \tau, t)$ can be written as a sum of two contributions:

$$Q_0^1 = \text{Tr} \int_0^{N_0} d\gamma \int_0^{N_0} d\gamma' G_0(\tau, \gamma | t) G_0(\tau, \gamma' | t) \langle \mathbf{c}(\gamma) \cdot \mathbf{c}(\gamma') \rangle_0 / dN_0 \tag{20}$$

$$Q_0^2 = 2\lambda_0 \text{Tr} \int_0^{N_0} d\gamma \int_0^t ds G_0(\tau, \gamma | t-s) G_0(\tau, \gamma | t-s) / N_0 \tag{21}$$

which, on performing the remaining integrals and using (13) and (16), leads to

$$Q(\tau, t | \tau, t) = 2\lambda_0 t / N_0^2 + \frac{1}{6} - \sum_{p=1}^{\infty} \frac{1}{(\pi p)^2} \cos 2\hat{p}_0 \tau \exp(-\lambda_{(p)} t). \tag{22}$$

Evaluating the remaining averages involving the Oseen tensor in the one-loop terms in equation (18), performing the momentum integrations and combining with (22), we secure

$$\begin{aligned} \langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle &= \frac{dN_0}{6} + \frac{2\lambda_0 dt}{N_0} - dN_0 \sum_{p=1}^{\infty} \frac{1}{(\pi p)^2} \cos 2\hat{p}_0 \tau \exp(-\lambda_{(p)} t) \\ &\quad + 2g_0^2 \eta_0^{-1} \left(\frac{d-1}{d-2} \right) (2\pi)^{-d/2} N_0^{-2} \left[\frac{4t}{\epsilon} N_0^{1+\epsilon/2} \right. \\ &\quad \left. + 4N_0 t \sum_{p=1}^{\infty} \exp(-\lambda_{(p)} t) \cos 2\hat{p}_0 \tau \left(\frac{2}{\epsilon} N_0^{\epsilon/2} + \text{ci}(2\pi p) - \hat{\gamma} - \ln 2\pi p \right) \right] \end{aligned} \tag{23}$$

where

$$\text{ci}(x) = \ln x + \int_0^x \frac{dt}{t} (\cos t - 1) + \hat{\gamma}$$

with $\hat{\gamma}$ being Euler's constant.

According to equation (1) the translational diffusion constant D_R is then given by

$$D_R = \frac{1}{\zeta_0 N_0} \left[1 + g_0^2 \eta_0^{-1} \zeta_0 (2\pi)^{-2} (2\pi N_0)^{\epsilon/2} \left(\frac{3}{2\epsilon} + \frac{5}{8} \right) \right]. \quad (24)$$

The pole term in (24) is the same as the pole term for a linear chain, an observation which confirms the expectation that the critical properties of linear chains and rings can be described by the same renormalisation constants and therefore the same critical exponents to lowest order [5]. Renormalisation is performed with

$$N = Z_N N_0 \quad (25a)$$

$$\zeta = Z_\zeta \zeta_0 \quad (25b)$$

$$\xi = (\zeta / \eta_0) L^{\epsilon/2} \quad (25c)$$

where L is a length scale. Setting $g_0^2 = \eta_0 = 1$ (as both couplings are not renormalised), we obtain at the non-draining Gaussian fixed point, $\xi^* = 8\pi^2/3\epsilon$,

$$D_R = \frac{3}{4\pi\epsilon} (2\pi N)^{(2-d)\nu} \exp\left(\frac{5}{12}\epsilon\right). \quad (26)$$

where $\nu = \frac{1}{2}$ is the Flory exponent. For a linear chain, D_L has been determined as [13]

$$D_L = \frac{3}{4\pi\epsilon} (2\pi N)^{(2-d)\nu} \exp\left(-\frac{1}{12}\epsilon\right). \quad (27)$$

Therefore we find a universal ratio

$$D_R / D_L = e^{1/2} \quad (28)$$

in $d = 3$, a result which should be possible to investigate experimentally. The fact that $D_R > D_L$ has its origin in the Gaussian (equal time) correlation function

$$\langle [c(\alpha, s) - c(\alpha', s)]^2 \rangle_0 = d \left(|\alpha - \alpha'| - \frac{1}{N_0} (\alpha - \alpha')^2 \right) \quad (29)$$

which enters into the expression for the Oseen tensor.

Renormalising the one-loop terms in (23) in the same way and exponentiating the obtained form so as to give the correct results for arbitrary times in conformity with singular perturbation theory [14], we find the effective eigenvalue $\lambda_{(p)}$ describing the relaxation spectrum to $O(\epsilon)$:

$$\lambda_{(p)}^R = \frac{3}{2\epsilon} \frac{1}{(2\pi)^{2-\epsilon/2}} \left(\frac{2\pi p}{N} \right)^{2-\epsilon/2} \exp\left\{ \epsilon \left[\frac{5}{12} + \frac{1}{2} ci(2\pi p) - \frac{1}{2} \hat{\gamma} \right] \right\} \quad (30)$$

which compares with the result for a linear chain [15]:

$$\lambda_{(p)}^L = \frac{3}{2\epsilon} \frac{1}{(2\pi)^{2-\epsilon/2}} \left(\frac{\pi p}{N} \right)^{2-\epsilon/2} \exp\left\{ \epsilon \left[\frac{5}{12} + \frac{1}{2} ci(\pi p) - \frac{1}{2} \hat{\gamma} - (1/2\pi p) \left(\frac{1}{2}\pi + si(\pi p) \right) \right] \right\}. \quad (31)$$

Asymptotically $\lambda_{(p)}^R, \lambda_{(p)}^L$ behave as $\lambda_{(p)}^{R,L} \sim p^{2-\epsilon/2}$ ($= p^{uz} = p^{d/2}$) with z being the dynamical exponent $z = d$. Finally, introducing new units $\bar{t} = t \lambda_{(1)}^R, \bar{\lambda}_{(p)}^R = \lambda_{(p)}^R / \lambda_{(1)}^R, \bar{\tau} = \tau / N$, we write our result for $\langle c(\tau, t) \cdot c(\tau, t) \rangle$ in universal form:

$$\langle c(\bar{\tau}, \bar{t}) \cdot c(\bar{\tau}, \bar{t}) \rangle / dN = \frac{1}{6} + \frac{2\bar{t} \exp(-\epsilon C_1)}{(2\pi)^{2-\epsilon/2}} - \sum_{p=1}^{\infty} \frac{\cos 2\pi p \bar{\tau}}{(\pi p)^2} \exp(-\bar{\lambda}_{(p)}^R \bar{t}) \quad (32)$$

with

$$C_1 = \frac{1}{2} ci(2\pi) - \frac{1}{2} \hat{\gamma}.$$

The fact that our result, equation (32), is so simple (there are no terms to $O(\epsilon)$ which depend on $\bar{\tau}, \bar{t}$) has its origin in the periodic boundary conditions for the ring. The renormalisation result (32) is basically of the same form as the free correlation function (22) with the effective eigenvalue modified.

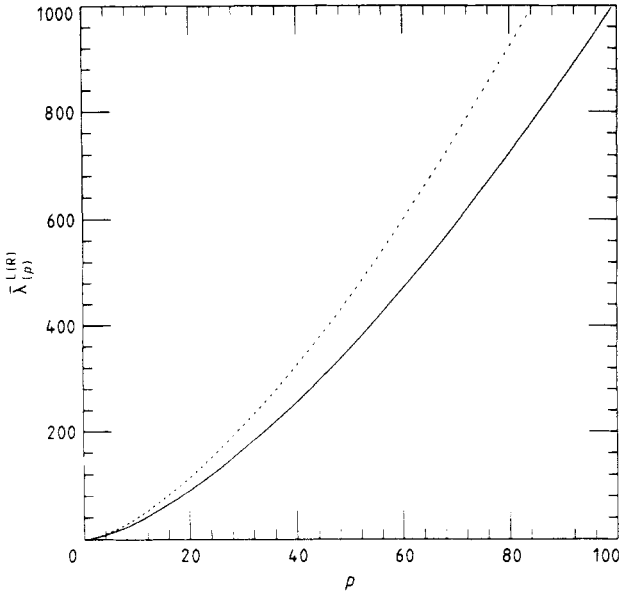


Figure 1. The normalised eigenvalues $\bar{\lambda}_p^R$ (for the ring; full curve) and $\bar{\lambda}_p^L$ (for the linear chain; broken curve) plotted as a function of p .

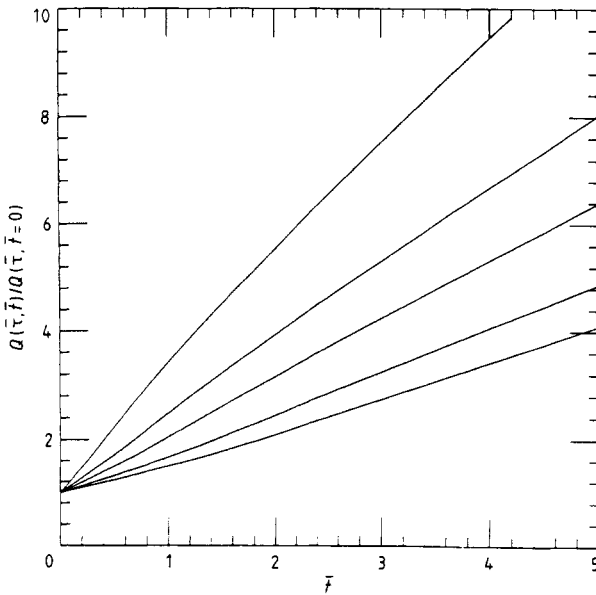


Figure 2. The correlation function $Q(\bar{\tau}, \bar{t})/Q(\bar{\tau}, \bar{t}=0)$ presented as a function of time \bar{t} in a universal plot. The parameter values (from top to bottom) are $\bar{\tau}=0.1$ (0.9), $\bar{\tau}=0.15$, $\bar{\tau}=0.2$ (0.8), $\bar{\tau}=0.3$ (0.7) and $\bar{\tau}=0.5$.

In figure 1 we have plotted $\bar{\lambda}_p^R$ and $\bar{\lambda}_p^L$ for the ring (R) and for the linear chain (L) as a function of p . In figure 2 we have plotted the correlation function $Q(\bar{\tau}, \bar{t})/Q(\bar{\tau}, \bar{t}=0)$ as a function of the new time unit \bar{t} . Note that figure 2 represents a universal plot. As expected, for large \bar{t} the translational diffusion of the centre of mass dominates, so $Q(\bar{\tau}, \bar{t}) \sim \bar{t}$. For shorter times the internal chain motion becomes important. In the $\bar{t} \rightarrow 0$ limit (but $t \gg 1$) we find $Q(\bar{\tau}=0, \bar{t}) \sim \bar{t}^{2/z}$, a result in agreement with scaling arguments. It can be obtained by replacing the sum over p in (32) by an integral and putting $\bar{\tau}=0$. The crossover between these two limiting behaviours is shown in figure 3.

We would like to comment on the inclusion of self-avoiding interactions. It is possible [16] to show that the translational diffusion constant for a polymer ring is given by

$$D_R = \frac{1}{\pi \epsilon} L^{1-d/2} \exp\left(\frac{5}{16}\epsilon\right) \left(\frac{2\pi N}{L}\right)^{(2-d)\nu} \tag{33}$$

with $\nu = \frac{1}{2} + \frac{1}{16}\epsilon$, which gives, comparing with the result for a linear chain [13], a universal ratio in $d = 3$:

$$D_R/D_L = e^{3/8}. \tag{34}$$

Finally, we would like to point out that because we describe the polymer dynamics using the Edwards model, the ring structure which is originally a circle could change into a knotted ring. This can only be prevented by putting a topological constraint (allowing winding number zero only) in our starting path integral. However, such a constraint would have to be treated non-perturbatively and we do not know how to perform the RG analysis in this case. We do expect, however, that the presence of a topological constraint will tend to make the ring more swollen [17] (compared to a linear chain) even in the absence of self-avoiding interactions. Therefore the ratios

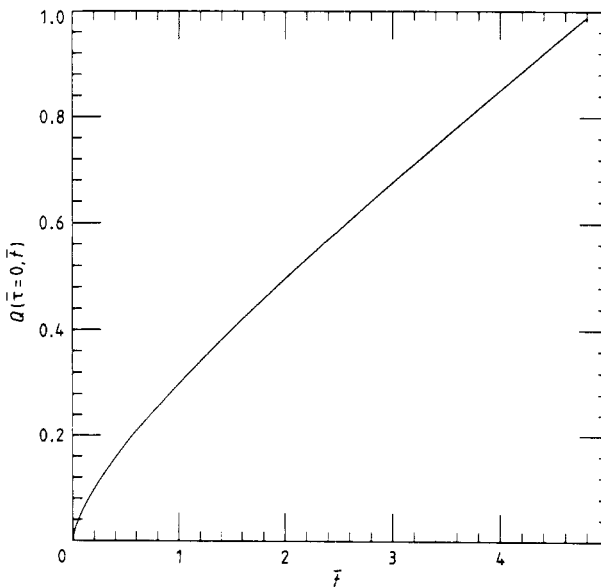


Figure 3. The crossover between the long-time, purely diffusive and the short-time behaviour as a function of \bar{t} .

(28) and (34) can only give a lower bound, and the exponent ν_R may in fact be larger than $\frac{1}{2}$ or $\frac{1}{2} + \frac{1}{16}\varepsilon$.

To summarise, we have determined the time-dependent correlations $\langle \mathbf{c}(\tau, t) \cdot \mathbf{c}(\tau, t) \rangle$ for a simple ring polymer in the presence of hydrodynamic interactions, and we have extracted the translational diffusion constant D_R and the relaxational spectrum to $O(\varepsilon)$. The universal ratios derived from our calculations should be possible to check experimentally. The next step in a systematic investigation of the transport properties for simple ring polymers should be the calculation of the intrinsic viscosity in order to obtain more information about the influence of ring formation on critical properties.

Acknowledgments

This work is supported by Defense Advanced Research Projects Agency contract N00014-86-C-2326, La Jolla Institute Independent Research and Development funds and National Science Foundation Grant NSF/DMR82-12570.

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