Exact curvilinear diffusion coefficients in the repton model.

Arnaud Buhot

UMR 5819 (UJF, CNRS, CEA) SPrAM, Département de Recherche Fondamentale sur la Matière Condensée, CEA Grenoble, 17 rue des Martyrs, 38054 Grenoble cedex 9, France. e-mail: abuhot@cea.fr

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Abstract. The Rubinstein-Duke or repton model is one of the simplest lattice model of reptation for the diffusion of a polymer in a gel or a melt. Recently, a slightly modified model with hardcore interactions between the reptons has been introduced. The curvilinear diffusion coefficients of both models are exactly determined for all chain lengths. The case of periodic boundary conditions is also considered.

Key words. Repton model – Polymer reptation – Diffusion coefficient

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1 Introduction

The reptation of a polymer in an entangled melt was studied long ago by De Gennes [11] who predicted the polymer length dependences of the curvilinear and self diffusion coefficients as well as the viscosity and the relaxation time. Later, Rubinstein introduced a lattice model for the polymer motion incorporating most of De Gennes’ ideas of reptation [2]. The so called repton model was then generalized by Duke to take into account the case of charged polymers during gel electrophoresis allowing the determination of the drift velocity [3]. This repton model seems particularly well adapted for DNA gel electrophoresis when the pore size is comparable to the persistence length [14].

The theoretical prediction of the viscosity dependence with the polymer length is in apparent conflict with the experimental observations [4][7][8]. This discrepancy is also observed in numerical simulations of the repton model [20]. Furthermore, a lot of interest in the calculation of the self-diffusion in the repton model focussed on the long polymer limit. The next to leading order term of the self-diffusion was long debated due to discrepancy between analytical and numerical results [9][10][11][12][13].

At the same time, the diffusion of a polymer chain in small channels attracts an increasing interest since it applies to a great range of experimental situations. Brochard and De Gennes [14] considered the case of a flexible polymer in a channel large compared to the monomer size but small compared to the polymer length whereas Odijk [15] studied the case of stiff polymers with a persistence length larger than the channel width. The recent experimental access to nanometer scale channels allows to study the crossover behaviour between both regimes [16][17]. The transport of long flexible polymer chains through Carbon nanotubes bring the interest to channels of width comparable to the monomer size allowing the determination of the curvilinear diffusion coefficients [18]. Furthermore, the case of polymer diffusion in porous media with nanometer scale holes has been recently studied [14] with a slightly modified repton model [20]. This model presents identical dynamical rules for the curvilinear motion of the chain than the repton model and motivated the present interest on the analytical determination of the curvilinear diffusion coefficients as function of the chain length.

The paper is organized as follows. In section 2 the repton model is presented with the slight modifications introduced by Guidoni et al. [20]. In section 3, the main result of the paper, the exact calculation of the curvilinear diffusion coefficients, is presented and compared with previous results. Section 4 focuses on the repton model with periodic boundary conditions. Finally, in section 5, we discuss the next to leading order corrections of the exact curvilinear diffusion coefficients and we give some conclusions.

2 Description of the model

In the Rubinstein or repton model [2], the polymer chain contains $N$ beads or reptons. A configuration $C$ is characterized by $N-1$ variables $\tau_i$ corresponding to the existence $(\tau_i = 0)$ or not $(\tau_i = 1)$ of a stored length between the two reptons $i$ and $i + 1$ along the chain. The polymer diffusion in a melt or a gel is obtained by the motion of stored lengths along the chain. Duke [3] generalized the model incorporating the diffusion along a spatial direction subject to an electric field in order to model the electrophoresis of a polymer in a gel. The variables $\tau_i$ are then replaced by $N-1$ variables $\sigma_i$ with $\sigma_i = 0$ corresponding to the existence of a stored length and $\sigma_i = \pm 1$ to the direction of the chain along the field between the reptons $i$ and $i + 1$.
rates of the moves are proportional to the number of accessible cells occupied by a neighbor and reaches the cell of the other neighbor repton. Cell occupancies need to be consistent of its neighbor if both cells differ, and c) the motion of a gap between two reptons represents a hole. The possible curvilinear positions of the different reptons along the time, the evolution of the global variable of the chain may either reduce or increase the ratio \( p_b/p_c \). Thus, in \( d \) dimensions and with a cubic lattice, \( p_b = p_c \) and \( p_a = 2dp_b \). However, the existence of larger or smaller monomers at the two ends of the chain may either reduce or increase the ratio \( p_b/p_c \). Similarly, in case of a polymer embedded in a gel, the gel structure in which the polymer diffuses may affect the ratio \( p_a/p_b \). The later ratio is responsible as we shall see to the equilibrium properties and especially the curvilinear length of the polymer chain. The former ratio only affects the dynamics of the polymer.

Guidoni et al. introduced recently a slightly modified model with hardcore reptons in one dimension. The cells may only be occupied by a single repton and two neighbor reptons are either in neighbor cells (\( \tau_i = 0 \)) or in next neighbor cells (\( \tau_i = 1 \)). The description of the chain in terms of the variables \( \tau_i \) is thus identical to the repton model. \( \tau_i = 1 \) corresponds to a hole between reptons and \( \tau_i = 0 \) to the existence of a stored length. From the repton model, only the length of the chain is slightly modified to take into account the repton size. The polymer diffuses thanks to the motion of stored lengths or holes. Furthermore, as seen on Fig. 1 the dynamical properties of the curvilinear motion are identical for both models. As a consequence, the curvilinear diffusion coefficients are equal.

### 3 Exact curvilinear diffusion coefficients

The curvilinear positions of the different reptons along the chain are defined by \( s_i = s_1 + a \sum_{k<i} \tau_k \) with \( s_1 \) the position of the first repton and \( a \tau_i \) the length between the two reptons \( i \) and \( i + 1 \). In case of hardcore reptons, an extra term \((i - 1)a\) should be added to take into account the repton lengths. It is also interesting to define \( s_c = \sum_i s_i/N \) the curvilinear position of the center of mass as well as \( s_m = (s_1 + s_N)/2 \) the middle position of the polymer chain. This middle position differs from the center of mass one for a particular configuration \( C \):

\[
\delta s = s_c - s_m = a \sum_i (N - 2i) \tau_i / 2N.
\]

This difference leads to an internal force that drives the polymer as will be shown later.

Due to the \( N - 1 \) variables \( \tau_i \) and their two possible values, there exist \( 2^{N-1} \) internal configurations \( C = \{ \tau_i \}_{i=1,...,N-1} \) for the polymer chain. We define the probability \( P(C, s, t) \) for the configuration \( C \) to have a curvilinear center of mass position \( s_c = s \) at time \( t \). Due to the correlations between the internal configurations of the chain along the time, the evolution of the global variable \( s \) alone is not sufficient to determine the curvilinear diffusion coefficient. The initial condition \( P(C, s, t = 0) = P_{eq}(C) \delta(s) \) is assumed with \( \delta(s) \) the usual delta function and \( P_{eq}(C) \) the equilibrium probability of the configuration \( C \). From this initial condition, the averaged curvilinear position over the ensemble of configurations remains zero at all times. Furthermore, the averaged squared curvilinear position increases linearly with time proportionally to twice the curvilinear diffusion coefficient \( D_s(N) \) of the polymer chain with \( N \) reptons. Note that we may expect the average position for a single configuration \( C \) to shift a finite
value from zero due to the initial difference $\delta s$ between the middle and the center of mass of the chain. The knowledge of this shift in position is necessary to determine the curvilinear diffusion coefficients. In fact, it reflects the correlations inside the chain between the configurations [20].

In order to define the dynamical rules, we introduce the configurations $C_i$ for $i = 0$ to $N-1$ which only differ from the configuration $C$ by the fact that both variables $\tau_i$ and $\tau_{i+1}$ have changed to $1-\tau_i$ and $1-\tau_{i+1}$. The changes from $C$ to $C_i$ are the only allowed moves in the repton model and it corresponds to a stored length passing through the repton $i+1$ (see Fig 1). In order for the move to exist the variables $\tau_i$ and $\tau_{i+1}$ must differ. With these constraints in mind, the evolution equation for the probability $P(C, s, t)$ is:

$$\frac{\partial}{\partial t} P(C, s, t) = -\sum_{i=0}^{N-1} w(C \to C_i) P(C, s, t)$$

$$+ \sum_{i=0}^{N-1} w(C_i \to C) P(C_i, s + \Delta s(C), t)$$

where the rates $w$ are given in Table 1 for open boundary conditions and $\Delta s(C) = (\tau_{i+1} - \tau_i)/a/N$ corresponds to the curvilinear motion of the center of mass. The variables $\tau_0 = 1 - \tau_1$ and $\tau_N = 1 - \tau_{N-1}$ have been introduced for consistency. The rate $p_a$ corresponds to the terminal repton exploring a new cell, the rate $p_b$ to the entrance of the terminal repton into the cell of its neighbor and the rate $p_c$ to the motion of stored length inside the chain (see Fig 1). Those rates are identical to those of the model of Guidoni et al. [20].

The determination of the curvilinear diffusion coefficients depends on three steps: the determination of i) the equilibrium probability $P_{eq}(C)$, ii) the long time limit of the averaged curvilinear position of a configuration $C$ and iii) the long time limit of the derivative of the averaged squared curvilinear position (in the last case the average stands for the position as well as the configuration averages). The different moments of the curvilinear positions of the center of mass are defined by:

$$\int s^k P(C, s, t) ds \equiv \langle s^k \rangle(C, t) P(C, t)$$

where $P(C, t)$ is the probability to find the configuration $C$ at time $t$. The time evolution for this probability is given by Eq. 1 where $P(C, s, t)$ is replaced by $P(C, t)$. From our choice of the initial condition, $P(C, t) \equiv P_{eq}(C)$ at all times and satisfies Eq. 1 with $\partial P/\partial t = 0$. The equilibrium probability $P_{eq}(C)$ is the product of an identical probability $P(\tau)$ for all variables $\tau_i$:

$$P_{eq}(C) = \prod_{i=1}^{N-1} P(\tau_i).$$

The probability $P(\tau)$ may be deduced from the evolution rates of the terminal reptons $p_a$ and $p_b$ independently of $p_c$. The particular configuration with all $\tau_i = 0$ leads to $p_a P(0) = p_b P(1)$. Thus, we deduce $P(1) = p_a/(p_a + p_b)$ and $P(0) = p_b/(p_a + p_b)$. The curvilinear length of the chain $L = s_N - s_1 = (N-1)aP(1)$ where the overline stands for an average over the configurations with the equilibrium probability $P_{eq}(C)$. This result compares with the equilibrium length $Na + L$ in the model of Guidoni et al. [20] where the $Na$ difference comes from the length of the $N$ reptons.

From Eq. 1 we deduce a differential equation for the average curvilinear position of the center of mass:

$$\frac{\partial}{\partial t} \langle s \rangle(C, t) = -\sum_{i=0}^{N-1} w(C \to C_i) \langle s \rangle(C, t)$$

$$+ \sum_{i=0}^{N-1} w(C_i \to C) [\langle s \rangle(C_i, t) - \Delta s(C)] \frac{P_{eq}(C_i)}{P_{eq}(C)}$$

where we used the property that $\int s^k P(C, s + \Delta s, t) ds = \langle s - \Delta s \rangle^k(C, t) P(C, t)$ with $P(C, t) = P_{eq}(C)$. In the long time limit, the averaged curvilinear positions $\langle s \rangle(C, \infty)$ saturate and the left hand part of Eq. 4 vanishes. The same line of arguments on the averaged curvilinear squared positions leads to the determination of the curvilinear diffusion coefficient $D_c(N)$. The long time limit of $\partial \langle s^2 \rangle/\partial t$ equals:

$$2D_c(N) = 2 \sum_{i=1}^{N-1} w(C \to C_i) \Delta s(C) \langle s \rangle(C, \infty)$$

$$+ a^2 \sum_{i=1}^{N-1} w(C \to C_i).$$

After some transformations, the long time limit of the squared positions $\langle s^2 \rangle(C, \infty)$ cancelled out. Consequently, $D_c$ depends on the rates $w$ (Table 1) and the average curvilinear positions $\langle s \rangle(C, \infty)$. For the latter, we assume the following general expression:

$$\langle s \rangle(C, \infty) = a \sum_{i=1}^{N-1} f(i) \tau_i$$

where $f(i)$ is a function to be determined. Consider the configuration with all variables $\tau_i = 0$. Eq. 4 leads to $f(1) = -f(N-1)$. For the configurations $C_k$ with $\tau_k = 0$ except $\tau_k = 1$, if $2 \leq i \leq N-2$, we obtain $f(i+1) = 2f(i) -...$

| $i$ | $w(C_i \to C)$ |
|-----|----------------|
| 1 to $N-2$ | $p_a \tau_1 + p_b (1 - \tau_1)$ |
| $N-1$ | $p_a \tau_{N-1} + p_b (1 - \tau_{N-1})$ |
| $i$ | $w(C \to C_i)$ |
| 0 | $p_a (1 - \tau_1) + p_b \tau_1$ |
| 1 to $N-2$ | $p_a (1 - \tau_{i+1}) + p_b (1 - 2\tau_i) + p_c \tau_i$ |
| $N-1$ | $p_a (1 - \tau_{N-1}) + p_b \tau_{N-1}$ |

Table 1. Rates $w(C \to C)$ and $w(C \to C_i)$ in the repton model. The variables $\tau_i$ corresponds to the configuration $C_i$. 

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f(i − 1) and, for i = 1, f(2) = (Δ + 1)/f(1) + (Δ − 1)/N with Δ = (p_a + p_b)/p_c. Note the particular case Δ = 1 or p_a + p_b = p_c for which f(i) = 0 is a trivial solution. This particular case corresponds to the lack of correlations as discussed by Guidoni et al. [20]. In the case Δ ≠ 1, f(i) = A(Δ)i + B(Δ) with A(Δ) = −2B(Δ)/N and B(Δ) = (1 − Δ)/[(N − 2)Δ + 2]. From this expression, it is possible to deduce ⟨s⟩(C, ∞) = 2B(Δ)δs. It is easy to check that the expression for ⟨s⟩(C, ∞) given by Eq. 6 satisfies Eq. 4 for all configurations C.

Two limiting cases are of interest. The fast internal motion of reptons compared to the end-repton motion corresponds to Δ → ∞. In this case, ⟨s⟩(C, ∞) ≈ −2δs/(N − 2). In the case of slow internal motion of reptons, Δ → 0 and ⟨s⟩(C, ∞) ≈ δs. It is interesting to notice the change of sign in the prefactor between those two limiting cases implying a shift ⟨s⟩(C, ∞) in different directions for the same difference δs between the center of mass and middle position of the chain.

The exact curvilinear diffusion coefficient D_c(N) is deduced from Eq. 4 and:

$$\sum_{i=0}^{N-1} w(C \rightarrow C_i) = 2(N - 2 + 2\Delta)p_cP(0)P(1) \quad (7)$$

leading to:

$$D_c(N) = \frac{p_ap_b}{p_a + p_b} \left( \frac{a^2}{(N - 2)\Delta + 2} \right) = \frac{D_0P(0)P(1)}{N - 2 + 2\Delta^{-1}} \quad (10)$$

with $D_0 = p_a a^2$ the individual diffusion coefficient of a free internal repton. This result is exact for all polymer lengths $N \geq 2$ and for all sets of rates $p_a, p_b$ and $p_c$. Note the linear dependence of $D_c^{-1}$ with the number of reptons:

$$D_c^{-1}(N) = \frac{N - 2 + 2\Delta^{-1}}{D_0P(0)P(1)} \quad (11)$$

Fig. 2 illustrates this linear dependence for three different sets of parameters. The equality between the rates $p_a$ and $p_b$ implies identical equilibrium properties ($P(1) = P(0) = 1/2$). The individual diffusion coefficient $D_0 = 1$ is imposed by setting $a = 1$ and $p_c = 1$ leading to an identical slope for all sets of rates considered. The sets only differ by the value of $\Delta$, with $\Delta = 1/2, 1$ and 2 from top to bottom, leading to a shift of the different lines on Fig. 2. The numerical results are obtained from Monte-Carlo simulations. The average of the curvilinear squared position as function of time for $10^5$ chains with initial conditions distributed with the equilibrium probability $P_0(C)$ are determined for different chain lengths. Linear fits allow us to determine the curvilinear diffusion coefficients. The numerical errors are smaller than the symbols on Fig. 2.

It is possible to understand Eq. 11 as follows. There exists $N - 2$ equivalent internal reptons and 2 extremal reptons. The inverse diffusion coefficient reflects the contribution of the two kinds of reptons. The free internal reptons have a diffusion coefficient $D_0$. The probability for them to move inside the chain is $P(0)P(1)$, the probability that there exists a unique stored length on the sides of the repton. This leads to the contribution $(N - 2)/D_0P(0)P(1)$. The contribution of the two extremal reptons may be split in two parts: an extremal repton exploring a new cell (with a rate $p_a$ and a probability $P(0)$ to have a stored length on its side) and an extremal repton moving into the cell of its neighbor (with a rate $p_b$ and a probability $P(1)$ to have no stored length on its side). Both contributions are identical ($p_ap_b = p_bP(1)$) and add up to give the contribution $2\Delta^{-1}/D_0P(0)P(1)$ to the inverse diffusion coefficient $D_c^{-1}$ in Eq. 11.

In a d-dimensional square lattice, the choice $p_a = p_c = 1$ and $p_b = 2d$ is customary and reflects the 2d possible directions for an extremal repton to explore a new cell compared to the single cell possibility for the other moves. The curvilinear diffusion coefficient is then

$$D_c(N) = \frac{2da^2}{(2d + 1)[N(2d + 1) - 4d]} \quad (12)$$

Exact results already obtained by Guidoni et al. [20] are recovered: the uncorrelated case $\Delta = 1$ leads to $D_c(N) = D_0P(0)P(1)/N$ and the particular case $N = 3$ to $D_c(3) = D_0P(0)P(1)/(\Delta + 2)$. However, as can be seen from Fig. 3, the second order in 1/N differs from the one proposed in [20] due to the improper account of the correlations in their calculations. For example, the limit $p_c \rightarrow 0$ or $\Delta \rightarrow \infty$ leads to the consistent limit of the curvilinear diffusion coefficient $D_c(N) \approx a^2 P(0)P(1)p_c/(N - 2) \rightarrow 0$ in contrary to [20]. In contrary, the fast internal motion of reptons ($\Delta \rightarrow 0$ or $p_c \rightarrow \infty$) leads to a finite $D_c = P(0)P(1)a^2/2$ independent of $N$. 

![Fig. 2. Inverse of the curvilinear diffusion coefficient $D_c$ as function of the number of reptons $N$ for different sets of rates. The lines correspond to the exact analytical calculations and the symbols to numerical simulations with $(p_a, p_b, p_c) = (1/4, 1/4, 1)$ circles, (1/2, 1/2, 1) squares and (1, 1, 1) diamonds.](image-url)
and the symbols to numerical simulations with $P(1) = 1/2$ circles, $1/3$ squares, $1/4$ diamonds, $2/3$ up triangles and $3/4$ left triangles.

4 Periodic boundary conditions

In the following, we consider the case of periodic boundary conditions studied by van Leeuwen and Kooiman [21,22]. Those conditions correspond to introduce two new variables $\tau_N$ and $\tau_0$ with $\tau_N = \tau_0$. The rates $w(C \rightarrow C)$ and $w(C \rightarrow C_i)$ for $i = 0$ to $N - 1$ have the same expression given in Table 1 for $i = 1$ to $N - 2$. The number of independent variables $\tau_i$ in the periodic boundary case is increased to $N$ compared to the $N - 1$ variables in the open boundary case leading to an increased number of configurations by a factor 2.

In the case of periodic boundary conditions, the number $N_1$ of non-zero variables $\tau_i$ or $N_0 = N - N_1$ of stored lengths is conserved by the dynamics. In this respect, the dynamics is non-ergodic. All configurations $C$ with the same number of stored lengths have the same probability $P(N_0)$ and the equilibrium curvilinear length of the polymer with $N_0$ stored lengths is $L_{pb} = s_N - s_1 = (N - 1)\bar{a} = (N - 1)aN_1/N$ since only $N - 1$ variables $\tau_i$ are present in $s_N - s_1$. Furthermore,

$$\sum_{i=0}^{N-1} w(C \rightarrow C) = 2p_cN_1(N - N_1)/(N - 1)$$

(13)

$$\sum_{i=0}^{N-1} w(C \rightarrow C_i)(\tau_{i+1} - \tau_i) = p_c(\tau_N - \tau_0) = 0$$

(14)

lead to:

$$D_{pb}(N, N_1) = \frac{N^2p_cN_1(N - N_1)}{N^2(N - 1)} = \frac{D_0P(0)P(1)}{N - 1}$$

(15)

where we replaced $N_0/N$ by the probability $P(0)$ to have a stored length and $N_1/N$ by $P(1) = 1 - P(0)$ in analogy with the open boundary case. Note that the periodic and open boundary conditions present to the same leading behavior for the curvilinear diffusion coefficient $D_c \sim D_0P(0)P(1)/N$ in the long chain limit. This result is also obtained for the self-diffusion coefficient but with a different length dependence [21,22].

Fig. 4 illustrates the linear dependence of the inverse curvilinear diffusion coefficient for different probabilities $P(1)$ as function of the number of reptons $N$. The numerical results are obtained from Monte-Carlo simulations by linear fits of the average of the curvilinear squared position as function of time for $10^5$ chains with initial conditions comprising $N_1 = N P(1)$ variables $\tau_i = 1$. The numerical errors are smaller than the symbols.

5 Discussion and conclusion

In this paper, we have determined exactly the curvilinear diffusion coefficients $D_c(N)$ of the repton model. The cases of open and periodic boundary conditions were considered. The inverse curvilinear diffusion coefficients present a linear behavior with the number of reptons in both situations. The next to leading order term in the curvilinear diffusion coefficient $D_c$ shows interesting properties :

$$\frac{N D_c(N)}{D_0P(0)P(1)} - 1 \approx \frac{2(\Delta - 1)}{N \Delta}.$$  

(16)

This term is positive for $\Delta > 1$ and negative for $\Delta < 1$. The later case would correspond to a chain with large end-reptons limiting their motion compared to the internal reptons. On Fig. 5 $D_c(N)$ as function of $N$ on a log-log scale is presented for three different values of $\Delta$, from bottom to top $\Delta = 1/2, 1$ and 2. The change of concavity of the curves is representative of the sign change in Eq. 16.
A possible consequence of the next to leading order term concerns the determination of the effective exponent obtain by a linear fit of the diffusion coefficient in a log-log scale. Let us define a finite size effective exponent $\nu_c(N)$ for the curvilinear diffusion coefficient $D_c(N)$ as follows:

$$\nu_c = \frac{\partial \ln D_c}{\partial \ln N} = 1 + \frac{2(\Delta - 1)}{(N-2)\Delta + 2}$$  \hspace{1cm} (17)$$

Depending on $\Delta$, this exponent is either larger or smaller than the expected one $\nu_c = 1$ in the long chain limit except for the particular case $\Delta = 1$ where $\nu_c(N) = 1$ (see Fig.5). The non-monotonous behaviour observed for the self-diffusion $D_s$ is not present for the curvilinear diffusion $D_c$. This difference is due to the absence of an anomalous behavior for $D_c$ whereas $D_s$ presents corrections of order $1/N^{1/2}$ due to fluctuations in the chain length [23].

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